

Xiao Lin, Xue Wang, Gangfeng Liu, and
Guobin Zhang

Recycling of Power Lithium-Ion Batteries

Technology, Equipment, and Policies



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Preface

The global new energy vehicle industry has been developed rapidly and the global sales volume of new energy vehicles is increasing year by year. Since 2016, the new energy revolution has swept the world. By 2020, there are more than 10 million electric cars cumulatively in the market. As the most crucial technology, power lithium-ion batteries have achieved explosive growth in the production and sales with the gradual electrification of transportation. The installed capacity of power lithium-ion batteries in 2021 was about 300 GWh with a year-on-year increase of 115%. As the first batch of new energy vehicles have been used for eight years, the small peak of power lithium-ion battery retirement with cumulative capacity of more than 300 000 tons (35 GWh) in 2021 has arrived. Therefore, the recycling of spent power lithium-ion batteries will play a significant role in the new energy industry chain.

Recycling of power lithium-ion batteries has multiple attributes of safety, environment, resources, and regionality. From the perspective of safety, improper disposal of waste power batteries has potential dangers such as electric shock, explosion, and hydrogen fluoride corrosion. From the environmental perspective, there are heavy metals pollution caused by nickel, cobalt, copper, and manganese as well as organic pollution caused by electrolyte and binders. Besides, dust, waste gas, wastewater as well as waste residues in the recycling process can also endanger the environment. From the resource perspective, it contains key resources such as lithium, nickel, cobalt, manganese, etc. Finally, from a geographical point of view, there are great differences in environmental protection policies, recycling channel systems, battery types and stock sizes as well as the recycling technologies for various regions.

Based on the industrial status and development of power lithium-ion batteries, the current book mainly focuses on the recycling technology and equipment, typical cases in industry, whole life cycle analysis, recycling regulations, new application scenarios, etc., to show the current situation and future development of global recycling industry of spent power lithium-ion battery. This book consists of seven chapters. Chapter 1 starts with the market dynamics of new energy vehicles and power lithium-ion batteries, as well as the status and development trend of key materials for power batteries. Based on the global distribution of raw materials containing critical metals for power lithium-ion batteries, the supply and demand analysis of critical metals for power batteries are displayed with and without considering

recycling. Chapter 2 mainly introduces the research and technology of battery recycling including pretreatment, hydrometallurgy, pyrometallurgy, and direct recycling technologies. Besides, the typical equipment associated with the recycling process is also introduced in detail. Followed by some typical industrial cases in China, Europe, and the United States, which are overviewed in Chapter 3. In Chapter 4, the carbon emissions caused by the pyrometallurgical process, hydrometallurgical process, and direct recycling process are compared via life cycle assessment, with the aim to propose the best recycling technology of power lithium-ion batteries. In Chapter 5, the relevant management regulations, technical standards, and support policies of various countries for the battery recycling industry are summarized. Except for the major application in electric vehicles, Chapter 6 illuminates new application scenarios of power lithium-ion batteries, such as two-wheeled electric bicycles, electric boats, and energy storage. With the development of battery swapping, platform-based business model, and considering consumers' differences in usage frequency and daily management of batteries, power lithium-ion batteries will be expanded in more detailed and dispersed application scenarios. As a result, a more diversified technical route is required in the future. Chapter 7 tries to balance the environmental friendliness and economy from the perspective of life cycle assessment, and promising green battery design for recycling are recommended. The book aims to form a roadmap for the development of power lithium-ion battery recycling, in order to provide support for the sustainable development of the new energy vehicle industry.

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1

Status and Development of Power Lithium-Ion Battery and Its Key Materials

1.1 Market Status of Power Lithium-Ion Battery

Lithium-ion batteries (LIBs) were invented by Sony in 1990 and brought to the market for commercialization in 1991, which kicked off the rapid development of LIBs. Initially (before 2000), most of the world's lithium batteries were produced in Japan [1–3]. However, by the year of 1997, South Korea's lithium secondary battery market began to rise, and it once surpassed Japan in the field of portable mobile electronics. In 1996, China Electronics Technology Group successfully developed 18650 batteries that can be mass-produced, marking the start of China's lithium batteries industry. At present, as an advanced energy storage technology, power LIBs have been widely used in new energy electric vehicles, providing critical support for the current greenhouse gas emission reduction.

According to the International Energy Agency (IEA), there was 16.5 million electric vehicles in the world by the end of 2021. The electric car registrations and sales share in major countries and regions in the world from 2016 to 2021 were shown in Figure 1.1. It is estimated that the total number of electric vehicles in the world will reach about 200 million by 2030, accounting for 20% of the total number of vehicles worldwide. The booming development of electric vehicles will lead to an exponential increase in the demand for power LIBs.

According to SNE Research data, the global installed capacity of power LIBs will be approximately 296.8 GWh in 2021 with a yearly increase of 115%. In view of the huge market prospects of the lithium-ion power battery industry [4], the relevant companies from different countries have deployed the power battery industry development plans. Among the world's top 10 battery companies (Figure 1.2), five of them are from China, namely Contemporary Amperex Technology Co., Ltd. (CATL), Build Your Dreams (BYD), China Aviation Lithium Battery (CALB), Guoxuan High-tech (GOTION), and Envision AESC with a total market share of 47.6%; LG Chem (LGC), Samsung SDI, and SKI, located in Korea, have a total market share of 30.4%; and Panasonic's (Japan) global market share is 12.2%.

Based on the ground of national level, the current lithium-ion power battery industry has basically developed into “three-legged (China, Japan, and South Korea)

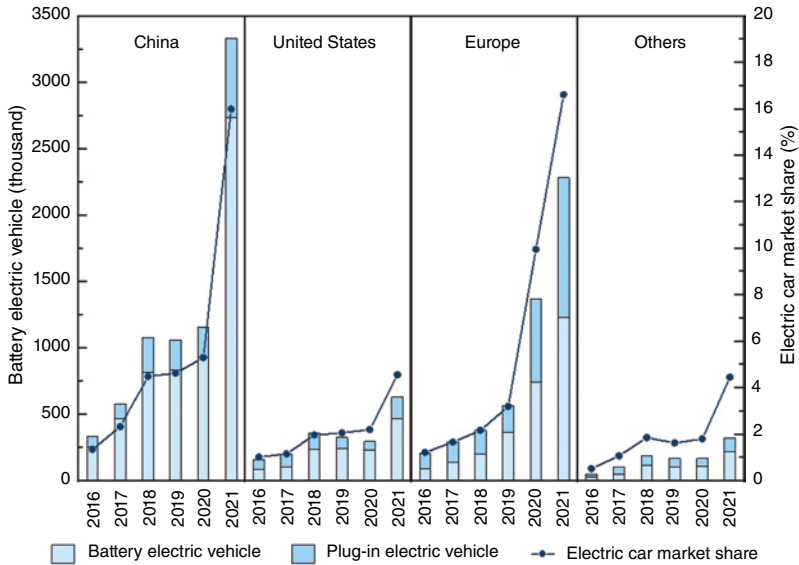


Figure 1.1 Registration volume and market share of electric cars in major countries and regions in the world, 2016-2021. *Source:* IEA.

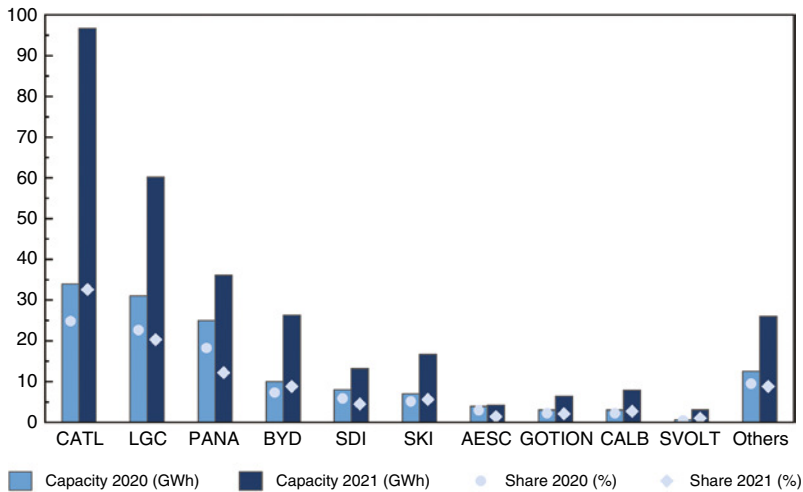


Figure 1.2 Top 10 global lithium-ion power battery companies and their installed capacity.

stand-up” pattern, and each of them has a leading enterprise in the industry. The Chinese company CATL has become not only the leader of China’s lithium battery industry but also the world’s largest supplier of power LIBs since 2017 [5] and was the first company to realize the mass production of NCM811 square batteries, which were successfully selected by GAC and BMW. From the technical route point of view, CATL also successfully realized the transition from NCM523 to NCM811.

Table 1.1 The main material production capacity of the world's major producers of lithium batteries.

	Cathode material	Anode material	Electrolyte	Separator
Country	3000 kilotons	1200 kilotons	339 kilotons	1987 million m ²
China	42%	65%	65%	43%
Japan	33%	19%	12%	21%
Korea	15%	6%	4%	28%
US	—	10%	2%	6%
Other	10%	—	17%	2%

Source: Data from Eddy et al. [7].

For South Korea's companies, LGC began to study lithium batteries in 1996 [6], and in 2020, it became the sole supplier of GM Chevrolet Volt electric vehicles. LGC's advantage is its advanced theoretical technology on soft pack batteries. It is also the first company in the world to be proficient in laminated-stacked soft packs. However, in the application of NCM811, it is behind the CATL. The Japanese company Panasonic began to develop the lithium batteries as early as 1994 and was supported by the Sumitomo Consortium. In 2008, it began to cooperate with Tesla, the world's largest electric vehicle company and built a super battery factory in 2014. Panasonic is the first company worldwide to realize the mass production of NCA18650 + silicon carbon anode cylindrical batteries, and it is also in a leading position in terms of the electrochemical system, production yield, and consistency.

In addition, China has already set up a strong leading position in the manufacturing of key materials for power LIBs (Table 1.1). According to the report "The metal mining constraints on the electric mobility horizon," China's production capacity in cathode materials, anode materials, electrolytes, and separators is 42%, 65%, 65%, and 43% of the world's total production capacity, respectively, far ahead of other countries and regions. Japan, by contrast, has a minor advantage in manufacturing cathode materials, whereas South Korea has an advantage in manufacturing diaphragms. In addition, other countries and regions such as the United States and the European Union have a relatively small market share in the production and manufacturing of materials for powered lithium batteries. The report shows that the market space for battery production and manufacturing in Western countries still has the great potential.

1.2 Key Materials and Development of Power Battery

Power LIBs are mainly composed of cathode materials, anode materials, separators, electrolyte binders, and current collectors. The cathode materials account for more than 40% of the total cost of lithium batteries, whose properties directly affect a variety of performance indicators of lithium batteries. Therefore, the cathode

materials take a core position in lithium batteries industry [7, 8]. Currently, commercialized cathode materials for lithium batteries include lithium cobalt oxide (LCO), lithium manganese oxide (LMO), lithium iron phosphate (LFP), and ternary materials. The proportion of LFP and ternary materials is as high as 90% in all the cathode materials.

1.2.1 Dominant Cathode Materials

1.2.1.1 Lithium Nickel Cobalt Manganese Oxide

The molecular formula of the lithium nickel cobalt manganese oxide (NCM) oxide ternary cathode material is $\text{LiNi}_a\text{Co}_b\text{Mn}_c\text{O}_2$, where $a + b + c = 1$. The naming rule of specific materials is usually based on the relative content of the three elements, for example, $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ is referred to as NCM811 for short. The different proportions of the three elements endow the NCM cathode material with different properties, which can meet the needs of diversified applications. NCM material combines the advantages of three types of materials as follows: LCO, lithium nickel oxide (LNO), and LMO. By adjusting the ratio of transition metal elements, the performance of the cathode material can be effectively regulated, and the cost of the cathode material can be reduced. Among them, the Ni element is beneficial to the increase of the specific capacity of the cathode material, which, however, is harmful to its thermal stability [9]; the Co element is beneficial to improve the electrical conductivity and rate performance of the material, but it is expensive; the presence of Mn plays a role in stabilizing the crystal structure of the polycrystalline, but it will also reduce the specific capacity of the cathode material when an excessive content was used.

The main preparation methods of NCM cathode materials include high-temperature solid-phase, sol-gel, co-precipitation, hydrothermal synthesis, and other methods. At present, commercial NCM materials were generally prepared with NCM hydroxide by precipitation method. The NCM precursor is mixed with a lithium source and calcined to prepare a finished NCM cathode material. The production of NCM precursor generally adopts the hydroxide co-precipitation method, that is, the mixed salt solution of nickel, cobalt, and manganese, precipitating agent, complexing agent, etc., are added to the reactor at the same time, and the NCM precursor is synthesized under specific conditions. The internal structure of the reactor and the control of the synthesis process are very demanding. Therefore, it takes an important position in the industry chain and has a high technical barrier. However, it has an important impact on the quality of the NCM cathode material. From the practical application point of view, the characteristics of the material particles, such as the morphology, particle size distribution, specific surface area, and tap density, have a great influence on the processing performance of battery electrode and the core electrochemical performance of lithium battery, such as energy density, rate performance, and cycle life. Therefore, the spherical NCM cathode material with high density and uniform particle size distribution has become the current pursuit goal of industry.

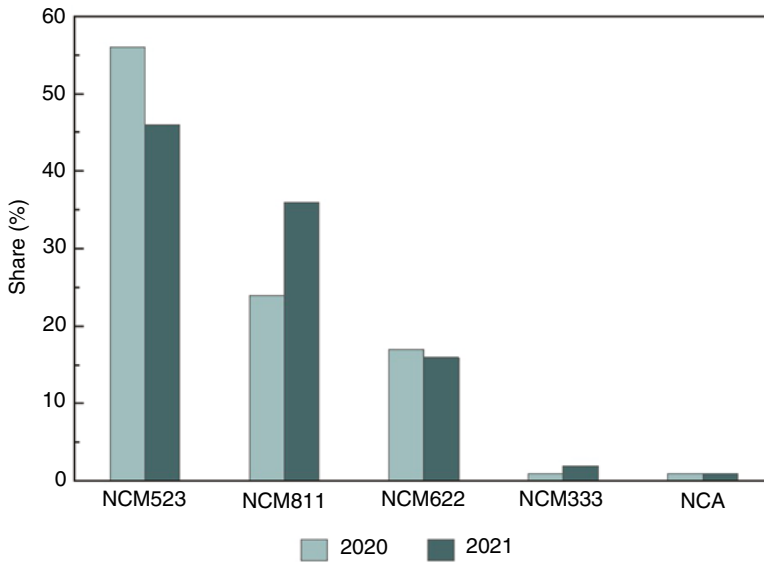


Figure 1.3 Comparison of the product structure of China's NCM materials in 2020 and 2021. *Source: SMM.*

Shanghai Metals Market (SMM) analysis of the results based on the output structure shows that, although the NCM material is still mainly NCM523, the trend of using low cobalt and high nickel is very clear. The proportion of NCM811 increased from 24% to 36% in 2021 (Figure 1.3). According to China Powder Network, GEM has developed an NCM precursor material with a molar ratio of nickel metal higher than 90% and has built an NCM precursor production capacity of 100 000 tons/yr. Sales volume of the high nickel and single crystals account for more than 80% of total sales. SKI, another power battery giant in South Korea, is earlier in the layout of ultra-high-nickel batteries and plans to launch a new NCM battery before the end of 2021. The battery has a nickel content of 88% and a cobalt content of 6%. Another South Korean company, LGC, plans to release lithium nickel cobalt manganese aluminum oxide (NCMA) batteries in 2022, with the 90% nickel content, 5% cobalt, and 1–2% manganese and aluminum.

1.2.1.2 Lithium Nickel Cobalt Aluminum Oxide

The lithium nickel cobalt aluminum oxide (NCA) material is composed of three main elements of nickel, cobalt, and aluminum, and the molar ratio is usually 8 : 1.5 : 0.5. By combining LiNiO_2 and LiCoO_2 , not only the reversible capacity becomes high, but the cost also becomes low. Currently, doping Al into metal oxide and replacing manganese with Al (transition metal) are one of the most popular directions in commercial cathode materials. At present, high-nickel materials can be divided into two categories: NCM811 and NCA materials. The reversible capacity of both materials can reach about 190–200 mAh/g. However, because Al is an

amphoteric metal, it is not easy to precipitate, and the conventional precipitation method cannot be used to prepare the NCA precursor. The NCA sintering process requires a pure oxygen atmosphere, which requires not only high tightness of the production equipment but also the oxidation resistance of the internal components of the kiln equipment. Due to the above-mentioned requirements of mass production, there are certain thresholds in the manufacturing process of NCA materials. In terms of route selection, Japan is mainly based on NCA routes, while South Korea tries to let NCM and NCA routes go hand in hand. China's current NCA output is relatively small, and NCM routes are the mainstay.

At present, the most mainstream preparation method is as follows: the precipitation of nickel, cobalt, and aluminum hydroxide is first prepared with metal sulfate as the raw material and sodium hydroxide or a complexing agent as the precipitation agent. The precipitate formed is then mixed with lithium hydroxide and then calcined into an oxide product. The advantages of this process are low production cost, simple process, and suitable for large-scale production. For example, Japan's Sumitomo and Japan's Toda have entered the mass production stage. Internationally, the upstream and downstream of NCA have formed a complementary industrial chain and a relatively stable and mature supply chain. However, China's domestic market is still at the initial stage of development.

1.2.1.3 Lithium Iron Phosphate

The chemical formula of LFP is LiFePO_4 , and its theoretical specific capacity is 170 mAh/g. The actual specific capacity of the product can exceed 160 mAh/g (0.2 C, about 25 °C, the voltage platform is 3.2–3.5 V, and the tap density is 1.2 g/cm³). In the LiFePO_4 structure, there is a strong covalent bond between O and P to form a tetrahedral $(\text{PO}_4)^{3-}$ polyanion; hence, O is difficult to deintercalated, and no oxygen escapes after overcharge. When LiFePO_4 is used as a cathode electrode material, battery safety can be promised.

Solid-phase synthesis is the most widely used and most mature synthetic method for LFP production. The iron source used in this method is generally ferrous oxalate, iron oxide, iron phosphate, etc.; the lithium source is generally lithium carbonate, lithium hydroxide, lithium acetate, etc.; and the phosphorus source is generally ammonium dihydrogen phosphate and diammonium hydrogen phosphate. The disadvantage of the solid-phase synthesis method is that it is easy to produce ammonia that pollutes the environment. Representative manufacturers include A123 systems, Tianjin Strand, Hunan Rui Xiang, Peking University First, Defang Nano, etc. At present, most of the synthesis method in our country adopts the ferrophosphorus process. This process generally uses iron phosphate as the iron source, which is first mixed with the lithium source at the nanoscale. After that, the particles can be converted into high-quality LFP materials with controllable particle size by spray drying and calcination in nitrogen atmosphere. Representative manufacturers include Phostech in Canada, Shandong Feng Yuan Lineng Technology Co., Ltd., and Sichuan Yuneng New Energy Battery Materials Co., Ltd.

The emergence of LFP is a breakthrough in LIB cathode materials. The following advantages of LFP, such as low price, environmental friendliness, high

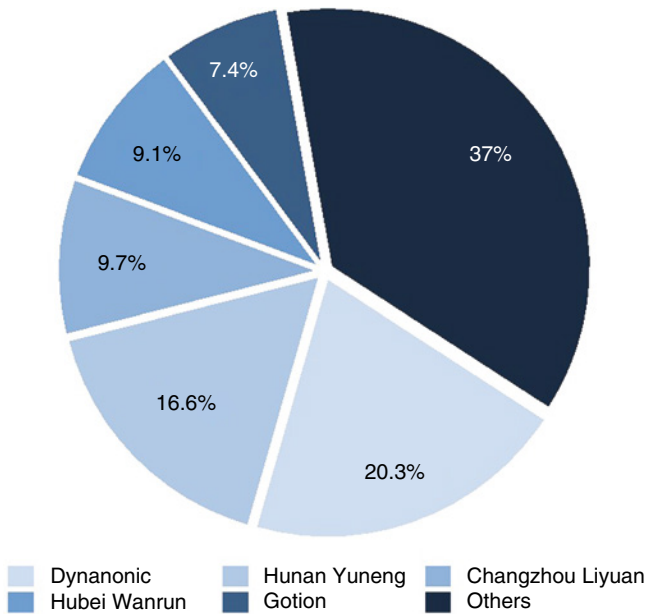


Figure 1.4 Market share of LFP manufacturers in 2021. *Source:* GGII.

safety performance, better structural stability, and cycle performance, have prompted it to have a wider market application field: Energy storage equipment, power tools, light electric vehicles, large electric vehicles, small equipment, and mobile power sources, among which, the LFP for new energy electric vehicles accounts for about 45% of its total production. The market share of LFP in China is displayed in Figure 1.4. Due to the structural reorganization of LFP batteries, the increase in energy density, the decline of new energy vehicle subsidies, the popularization of charging piles, the price advantage, and high safety advantages of LFP batteries, the installed capacity of LFP batteries in China has rapidly increased. Thus, the shipment of LFP cathode materials in China has been 470 000 tons by 2021, accounting for a year-on-year increase of 277%, according to the survey data of the Advanced Industrial Research Institute of Lithium Battery Research (hereinafter referred to as GGII).

1.2.1.4 Lithium Nickel Manganese Oxide

The molecular formula of lithium nickel manganese oxide (LNM) is $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$, which belongs to the spinel structure. The voltage platform is about 4.7 V, the theoretical specific capacity is 146.7 mAh/g, and the actual specific capacity is about 130 mAh/g; LNM has a high working voltage, high energy density, and low production cost. It combines the advantages of NCM materials and LFP materials, which is the goal of a new generation of cathode materials.

There are many preparation methods for LNM, including solid phase, co-precipitation, sol-gel, solution combustion synthesis, hydrothermal, solvothermal, and spray deposition methods. Public information shows that Honeycomb Energy mainly improves the performance of LNM through three methods: cation doping, single crystal technology, and nano-network coating. Doping technology involves the use of cations with high chemical bond energy with oxygen to dope into the crystal structure, which is conducive to the stability of the structure after de-lithiation under high voltage. Single crystal technology has a higher particle strength than traditional spherical polycrystalline particles, which is beneficial to improve safety and cycling performance. The use of a nano-network coating can make the material more uniform and reduce the side reaction with the electrolyte to improve the cycle life.

Reducing the cobalt content has become the primary measure for reducing the cost of NCA or NCM cathode materials, and the development of high-nickel and cobalt-free materials has become an inevitable trend. Tesla has always used the ternary battery (NCA) provided by Panasonic, in which the Co content is less than 3%, and the next generation of products can reduce the content of Co to zero. Since then, cobalt-free batteries and cobalt-free materials were born. According to Gaogong Lithium Grid, Honeycomb Energy's Changzhou plant has officially mass-produced cobalt-free materials, with an annual output of up to 5000 tons. Mine Road Network speculates that, by 2025, the global production of LNM oxide materials will reach 85 000 tons, and the demand will reach 100 000 tons. If LNM solves the problems of large-scale production and high-potential electrolyte tolerance, it will surely become the next generation of mainstream cathode materials.

1.2.2 Anode Materials

The negative electrode material is the main body that stores lithium when the power battery is charged, accounting for about 10% of the battery cost. Anode materials can generally be divided into two categories: carbon materials and noncarbon materials. Carbon materials include artificial graphite, natural graphite, mesophase carbon microspheres, petroleum coke, carbon fiber, and pyrolytic resin carbon. Noncarbon materials include titanium-based materials, silicon-based materials, tin-based materials, and nitrides.

1.2.2.1 Graphite

Since graphite materials have the advantages of high electronic conductivity, high specific capacity, stable structure, and low cost, they have become the most widely used and mature anode materials, and they are the absolute mainstream route in the anode industry, accounting for 95% of the total amount. Graphite can be divided into natural graphite and artificial graphite. Due to raw material and process characteristics, the internal structure of artificial graphite anode materials is more stable than natural graphite products.

Artificial graphite is generally divided into four major processes, namely crushing, granulation, graphitization, and sieving. Among them, the technical threshold

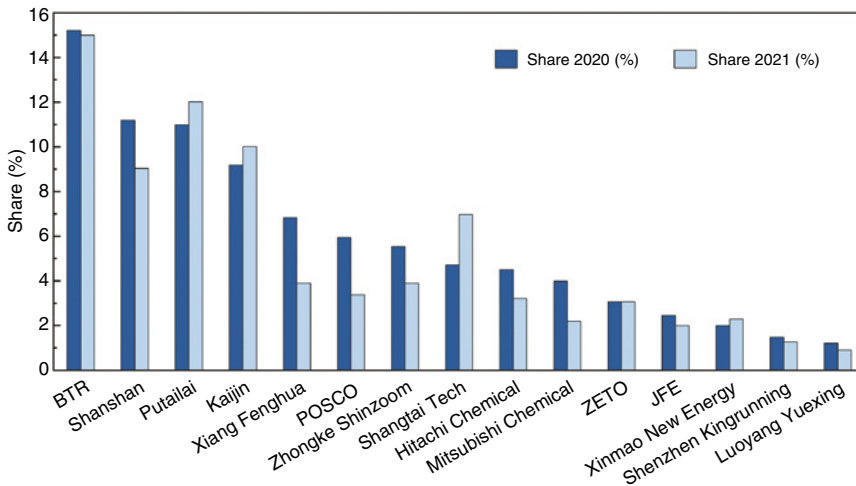


Figure 1.5 Market share of global anode material manufacturers in 2020–2021.

Source: White Paper on global lithium-ion battery industry, Starting Point Research.

and production level of the negative electrode industry are mainly reflected in the two links of granulation and graphitization. Granulation needs to control the particle size, particle size distribution, and morphology of graphite, and these physical parameters directly affect the performance indicators of the anode material. For example, the smaller the particles, the better are the rate performance and cycle life, but the worse are the first-time efficiency and compaction density; therefore, a reasonable particle size distribution is required.

From a global perspective, companies from Japan and China have always dominated the market of anode materials (Figure 1.5). The top five Chinese companies, including Berterry (BTR), Shanshan, Putailai (Jiangxi Zichen), Kaijin, and Xiang Fenghua, account for more than half of the global market. According to the data from EVTank, due to the significant increase in demand for LIBs in various fields around the world, global shipments of anode materials reached 905 000 tons in 2021 with a year-on-year increase of 68.2%. From the perspective of negative electrode product structure, the proportion of artificial graphite products will further increase (Figure 1.6). Other anode materials represented by silicon-based anodes failed to achieve the expected growth due to the switch of the main shipment models of Chinese cylindrical battery products and the delay in the upgrade of the high-nickel system of square power LIBs, and their market share declined dramatically.

1.2.2.2 Lithium Titanate

The chemical formula of lithium titanate is $\text{Li}_4\text{Ti}_5\text{O}_{12}$, referred to as LTO, which has been studied by many energy workers because of its “zero strain” advantage. “Zero strain” means that the volume and lattice constant of the lithium titanate material change very little during the phase change. Therefore, the main advantages of lithium titanate are good cycle performance. The cycle life of lithium titanate energy storage batteries is more than 15 000 times, with the good charging rate in the range

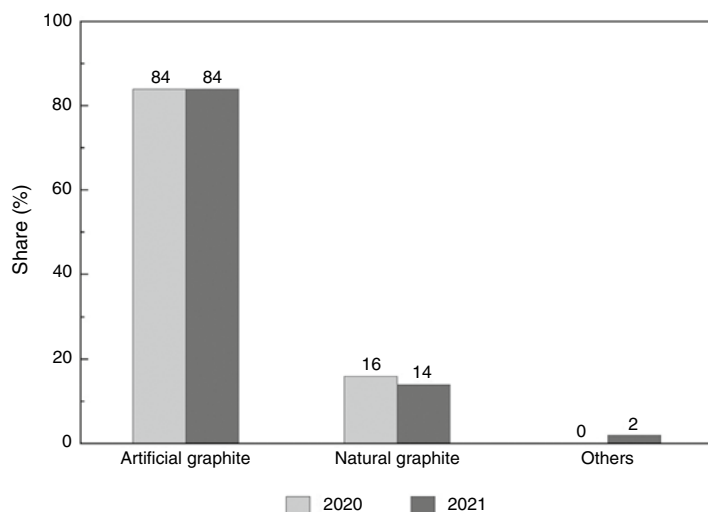


Figure 1.6 In 2020–2021, anode material industry structure. *Source:* GGII.

of 10–20 °C. Lithium titanate also has a good low-temperature performance and still has 70% capacity in the extremely cold environment of -50°C . Lithium titanate material has a low energy density, and its theoretical capacity is 175 mAh/g. At present, commercial lithium titanate has been developed to 170 mAh/g, and the first effect can be as high as 99.5%.

The industrial production method of lithium titanate still adopts the high-temperature solid-phase method. Usually, battery-grade TiO_2 and lithium salt $\text{LiOH}\cdot\text{H}_2\text{O}$ or Li_2CO_3 are dispersed in water or organic solvent according to a specific stoichiometric ratio and mixed uniformly. After high-energy ball milling to nano-scale, spray-drying granulation, and high-temperature calcination, the final product lithium titanate is obtained.

In terms of industrialization, the leading companies include the American Ao Titanium Nano Technology Company (now acquired by Yinlong New Energy), Japan's Ishihara Industry Co., Ltd., and the British Johnson Matthey Corporation. There are Sichuan Xingneng New Material Co., Ltd. and Huzhou Weihong Power Co., Ltd. in China. Dong Mingzhu's perseverance to "Yinlong Titanium" made Yinlong's lithium titanate battery famous within a short period. With its fast-charging, low-temperature properties and safety characteristics, lithium titanate is widely used in public transportation buses and logistics vehicles that do not require high mileage. The market size of lithium titanate batteries is worth the wait.

1.2.2.3 Silicon Carbon

Silicon is the anode material with the highest theoretical capacity found so far. The theoretical specific capacity is 4200 mAh/g, which is 10 times more than that of graphite. Mixing silicon and graphite to form a silicon–carbon-based composite

anode material not only combines the characteristics of good carbon conductivity and high silicon specific capacity but also can effectively buffer the expansion of silicon, which can significantly improve the overall electrochemical cycle performance of the material. From the commercialization path, the silicon-carbon material can be divided into silicon-carbon anode and oxygen-silicon-carbon anode materials.

The silicon-carbon anode material is first prepared by forming a precursor of nano-silicon and the matrix-graphite material through a granulation process, and the precursor is then prepared by surface treatment, sintering, pulverization, screening, demagnetization, and other processes. At present, the commercial application capacity of the silicon-carbon anode is about 450 mAh/g.

The silicon-oxygen negative electrode material is made by synthesizing pure silicon and silicon dioxide into silicon monoxide to form a silicon-oxygen negative electrode material precursor, and the precursor is then prepared into a modified SiO_x/C through the processes of pulverization, classification, surface treatment, sintering, sieving, demagnetization, etc. The modified SiO_x/C is then mixed with graphite according to the required capacity of the negative electrode, and a new type of oxygen-silicon graphite negative electrode material can be obtained; the capacities of the new oxygen-silicon graphite anode materials are mainly concentrated in two types: 420 and 450 mAh/g, with a small amount of 500 and 600 mAh/g. According to the China Powder Network, BTR's silicon-based anode products are at the leading position in the industry. It has made new breakthroughs in the development of silicon-carbon anode materials, and its specific capacity has been increased to 1500 mAh/g; BTR has completed a variety of technical development and mass production of sub-silicon products, and the specific capacity of some products reaches more than 1600 mAh/g. Snow's SiO product has a gram capacity of more than 500 mAh/g and the first coulombic efficiency is more than 89%, and it can achieve an 800-week cycle retention rate of more than 80%. It has completed the small-scale development and is in the stage of preparing for the pilot and mass production.

The industrialization of silicon-based anode batteries by global battery manufacturers is steadily advancing. As early as 2012, Japan's Panasonic has applied silicon carbon anodes to lithium batteries, while Hitachi Chemical is the largest supplier of silicon-carbon materials in the world as well as Japan's Shin-Etsu, Wu Yu Chemical, and American Ampris, all of which have deployed silicon carbon anode materials. The industrial application of silicon carbon anodes in our country is still at the initial stage. At present, CATL, BYD, GOTION, BAK, and Tianjin Lishen are all making efforts in the field of silicon-carbon materials, and BTR has achieved mass production. The high-tech lithium power grid predicts that the market demand for silicon-based anodes will exceed 30 000 tons in 2022, and the market size is expected to exceed 3.5 billion in the future.

1.2.3 Electrolyte

The electrolyte plays a role in transferring charge between the cathode and anode electrodes. According to the physical form of the electrolyte, it can be divided into

liquid electrolyte, solid electrolyte, and solid–liquid composite electrolyte. Batteries that use solid electrolytes are called solid-state batteries.

1.2.3.1 Liquid Electrolyte

The function of the electrolyte is to transfer lithium ions between the anode and the cathodes, and the electrons are not transferred, which ensures the smooth progress of charging and discharging. Electrolyte is very important to the performance of lithium batteries and is known as the “blood” of LIBs. According to statistics from the Huajing Industrial Research Institute, electrolytes generally account for about 7–12% of battery costs. The general composition is a solvent, lithium salt, and additives. The most frequently used lithium salt is lithium hexafluorophosphate. The solvents are mainly carbonates, ethers, and carboxylates. According to their functions, additives can be divided into film additives, conductive additives, and flame retardant additives.

The preparation of the electrolyte is mainly divided into three sections: solvent synthesis, material mixing, and post-processing. Among them, the technical barrier is the composition of the formula at the material mixing stage. The electrolyte composition of different battery types is slightly different. The formula is basically dominated by downstream lithium battery companies, while the electrolytes of some subdivided into high-end consumer products or high-nickel power products are generally jointly developed by lithium battery companies and electrolyte companies. As the core key material of the electrolyte, lithium hexafluorophosphate is very deliquescent and needs to be synthesized in nonaqueous solvents such as anhydrous hydrogen fluoride and low alkyl ether, and the production conditions are extremely harsh.

Before 2010, the production capacity of lithium hexafluorophosphate was mainly concentrated in Japanese companies such as Stella, Kanto Denka, and Morita Chemical and the South Korean company Hosei. In 2011, Chinese companies’ hexafluoro carbon production capacity accounted for less than 15%. After 2011, companies such as Tianci High-tech (TINCI), Xinzhoubang, and other companies have achieved technological breakthroughs and began to expand their production significantly. In 2021, the market share of Chinese enterprises increased to more than 80% (Figure 1.7). According to the “White Paper on the Development of China’s Lithium-ion Battery Electrolyte Industry (2022)” released by EVTank, China’s electrolyte market will ship 507000 tons in 2021 with a year-on-year increase of 88.5%. According to the forecast of Guohai Securities, the global demand for lithium batteries will reach about 1200 GWh by 2025, corresponding to an electrolyte demand of about 1.32 million tons, with a compound annual growth rate of 35% during this period. The electrolyte market has a huge space.

1.2.3.2 Solid Electrolyte

Liquid electrolyte has hidden dangers such as volatile organic solvents and high-temperature flammability, and its development in the field of lithium batteries is very limited. The solid electrolyte is not only difficult to decompose at high temperatures, but it can also inhibit the generation of lithium dendrites, effectively alleviating the safety and durability of lithium batteries. Solid-state electrolyte is the core component

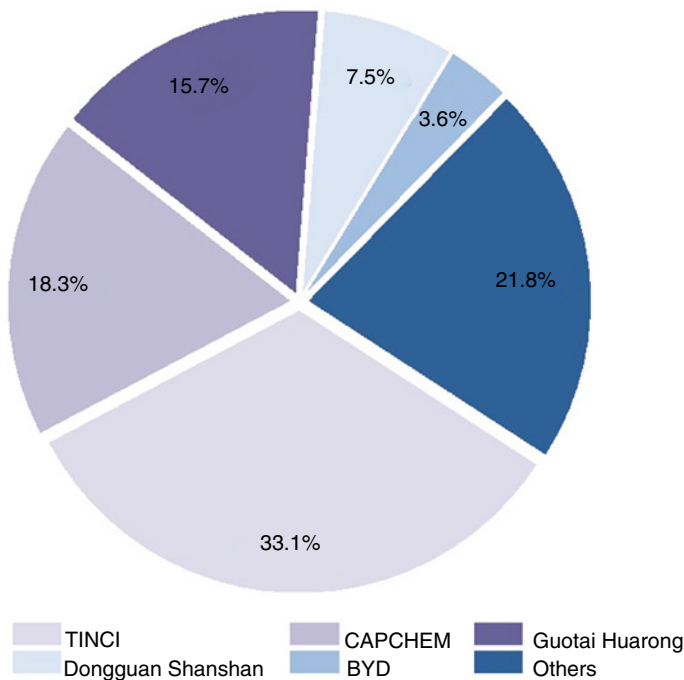


Figure 1.7 Market share of electrolyte in China in 2021. *Source:* Xinzhu Information, Huajing Industry Research Institute.

of solid-state lithium batteries and the technical focus of solid-state battery development. At present, solid electrolytes are mainly divided into three categories: polymer solid electrolytes, oxide solid electrolytes, and sulfide solid electrolytes.

The polymer solid electrolyte is complexed by a polymer matrix and a lithium salt. Commonly used lithium salts include LiPF_6 , LiClO_4 , and LiAsF_6 . The matrix includes polypropylene oxide (PPO), polyvinylidene chloride (PVDC), polyethylene oxide (PEO), polyacrylonitrile (PAN), and polyvinylidene fluoride (PVDF). Representative types of oxide electrolytes include lithium lanthanum zirconium oxide (LLZO), lithium aluminum germanium phosphate (LAGP), and lithium aluminum titanium phosphate (LATP). The general process is to mix the desired oxidizing material with lithium source ball milling, press into tablets, and then calcine to increase the density. The sulfide solid electrolyte evolved on the basis of the oxide electrolyte, but the sulfide is really easy to react with water that generates the toxic H_2S gas. The production conditions are also harsh. At present, methods such as halide doping or introduction of new elements are used to improve its chemical stability in water (Table 1.2).

From the perspective of technology route of enterprise, Japanese and Korean enterprises mostly adopt sulfide solid electrolyte technology [10], Chinese enterprises mostly use oxide electrolytes, while European and American enterprises choose polymer, oxide, and sulfide routes. Solid electrolytes still have problems such

Table 1.2 Comparison of three solid electrolytes.

Classification	Subdivision	Representative enterprise	Advantages	Shortcomings
Organic electrolyte	Polymer	Bolloré, Solid Energy	Low-density, good viscoelasticity, and the most mature technology, taking the lead in small-scale mass production	Room temperature ionic conductivity is low Low theoretical energy density upper limit
Inorganic electrolyte	Oxide	Sakti3, ProLogium, QingTao Development, Beijing WeLion, Ganfeng Lithium Group	Excellent battery rate and cycle performance, commercial products are already available	Mechanically hard, the solid–solid interface is not in good contact, and the mass production cost is high
	Sulfide	Toyota, Samsung, Panasonic, CALT	The highest ionic conductivity, most likely to be used in batteries on a large scale	Unstable, high requirements on the production environment, difficult to develop

Source: Adapted from Evergrande Research Institute and Zhongqi Consulting.

as low interface stability, large-scale grain boundaries, vacancies, and local electronic conductance. Solid-state batteries have not yet formed an industrial chain. The “New Energy Automobile Industry Development Plan (2021–2035)” puts forward the requirements for strengthening the R&D and industrialization of solid-state batteries. For the first time, solid-state batteries have been raised to the national level, and solid electrolytes and solid-state batteries are expected to improve dramatically. According to industry chain research, solid-state batteries will be gradually commercialized in 2025 and become the main technical route for power batteries in 2030. According to Bank of China International Securities, the global demand for solid-state lithium batteries is expected to reach 494.9 GWh by 2030, with a market space of more than 150 billion yuan.

1.2.4 Separator

Separator is a key material to ensure the safety of battery system and affect its performance. It is placed between the cathode and anode as a device to isolate the electrodes. Therefore, the separator must have good insulation to prevent short circuiting of the cathode and anode electrodes or short circuits caused by burrs, particles, and dendrites. In addition, separator is a microporous channel with charging and discharging functions and rate performance. Therefore, the separator needs to have a certain tensile strength, puncture strength, high porosity, and uniform distribution.

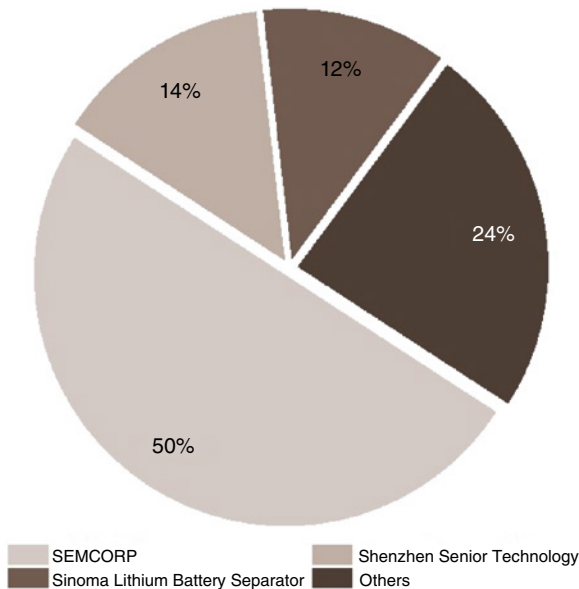


Figure 1.8 Pattern of separator for wet process in 2021. *Source:* Intelligence Research Group.

The pore size of separator is generally 0.03–0.05 or 0.09–0.12 μm ; the thickness is generally less than 25 μm . By ensuring the mechanical strength, the thinner the separator, the better is the performance of the battery. The current mainstream wet base film products in the power market are mainly concentrated in the 9 and 12 μm fields. The 5 μm separator is mainly used in the field of consumer batteries. The mainstream materials for the separator are polypropylene (PP) microporous film and polyethylene (PE) microporous film. Separator is the material with the highest technical barrier among lithium battery materials. Microporous preparation technology is the core of the separator preparation process. Therefore, the separator process can be divided into dry and wet methods. The raw material of the dry process is generally PP; the process is simple, and the cost is low. The raw material of the wet process is generally PE, which has more advantages in high-power batteries and has relatively higher strength.

In recent years, the output and performance of Chinese wet process separator manufacturers have come closer to those of foreign companies; the production of separator has basically achieved localization, and their market share has continued to rise (Figure 1.8). However, there are still gaps in indicators, production technology, and equipment technology when compared with Japanese and American companies such as Asahi Kasei and Tonen. The separator company W-Scope estimates that the global lithium battery market will have 20 billion square meters of separator demand by 2025 and close to 32 billion square meters by 2030, which is 5.3 times that of 2020.

1.2.5 Binder

Binders are indispensable in LIBs, and their amount is very small, accounting for about 1–10% of the active material components. The main function of the binder is

to bind the various components of the pole piece (active material and conductive agent) and the current collector together to form a stable pole piece structure, relieve the volume change of the electrode material during the charging and discharging process, and regulate dispersion effect of the slurry. Therefore, the binder must be able to withstand the swelling and corrosion of the electrolyte and the electrochemical effects of the charge and discharge process.

The current commercial binders include oil-based binder systems: mainly PVDF and PAN; aqueous binder system: sodium carboxymethyl cellulose (CMC), styrene butadiene rubber (SBR), sodium alginate (ALG), polyacrylic acid (PAA), and polyvinyl alcohol (PVA). At present, the cathode electrode of the battery is mainly PVDF binder, and *N*-methylpyrrolidone (NMP) is used as the solvent, accounting for up to 90%. The negative electrode material usually uses SBR as the binder, CMC as the thickener, and water as the solvent.

Solvay Group in Belgium, Kureha Co., Ltd. in Japan, Arkema Group in France, and Shanghai Sanaifu New Materials, Shandong Huaxia Shenzhou New Materials, Sinochem Lantian, etc., in China share the global lithium battery cathode binder PVDF market. Japan's Zeon Co., Ltd., Japan Paper, and China's Blue Ocean Blackstone Technology occupy the field of water-based adhesives. According to the statistical data, the output of lithium battery binders in China has increased annually in the past five years. As of 2021, it has reached 48 000 tons with a yearly increase of 34.5%. Benefiting from the rising demand for lithium batteries, China's lithium battery binders will also maintain rapid growth in the next five years. It is estimated that by 2025, China's lithium battery binder market is expected to approach 10 billion yuan.

1.2.6 Current Collector

The current collector is a component that is bonded to the outside of the cathode and anode electrodes in the battery and collects the current generated by the active material of the battery. It is one of the indispensable components in LIBs. Materials that can be used as current collectors for lithium batteries are metal conductor materials such as aluminum, copper, nickel, and stainless steel. With the continuous development of lithium battery technology, the trend of current collectors is to reduce the thickness and weight by ensuring new energy.

1.2.6.1 Copper Foil

Copper has many advantages such as excellent electrical conductivity, good ductility, and abundant resources. It is easily oxidized at a higher potential and is often used as a current collector for negative electrode active materials such as graphite, silicon, tin, and cobalt-tin alloy. The quality and cost of a copper foil account for about 13% and 8% of the total mass and total cost of a typical lithium battery, respectively [11]. There are mainly two types of copper foil: rolled and electrolytic ones. Rolled copper foil is an original foil made by repeatedly rolling a copper plate several times, and roughening treatment is carried out according to the requirements. Electrolytic copper foil is made by electrodepositing copper solution in a special

dissolving vessel under the action of direct current with copper sulfate electrolyte and then undergoing a series of surface treatments to obtain copper foil with smooth and rough surfaces. When copper foil is used as a current collector, the thickness is reduced from 12 to 10 μm and then to 8 μm . Up to now, most battery manufacturers use 6 μm for mass production.

According to the statistics of the Huajing Industry Research Institute, the global lithium battery copper foil production capacity will be a total of 435 000 tons in 2020. China and South Korea are the main lithium battery copper foil production countries; the top three companies in terms of production capacity are Nordisk, Lingbao Huaxin, and Jiujiang De Blessing. Affected by the increase in demand for upstream power batteries, copper foil companies of lithium battery have actively expanded their production. According to Gaogong Lithium battery, the global demand of copper foil for lithium batteries in 2021 is 380 000 tons with a yearly increase of 52%, of which the demand for power batteries is 240 000 tons (an annual increase of 75%); the total global demand of copper foil for lithium batteries is expected to be 100 000 tons in 2025, and the demand of copper foil for lithium battery will expand three times in the next five years.

1.2.6.2 Aluminum Foil

Aluminum foil is currently the main cathode current collector for LIBs. It has the advantage of good conductivity, light weight, and low cost, and the passivation layer on its surface can avoid electrolyte corrosion during the charging and discharging process. According to the composition and impurities, aluminum foil can be divided into 1 series, 3 series, and 8 series, corresponding to pure aluminum, aluminum manganese series, and other aluminum alloy series, respectively. Aluminum foil has been reduced from 16 to 14 μm and then to 12 μm in past years. Presently, many battery manufacturers have mass-produced aluminum foil of 10 μm and even 8 μm . The production of aluminum foil is to roll the aluminum foil blanks to the required thickness through multiple rolling and heat treatments. After two processes of rough rolling and finishing rolling, the aluminum foil is surface-treated, and finally, the aluminum foil is slit into the width and length required by the lithium battery manufacturer. Recently, more studies have been done on carbon-coated aluminum foil, where a thin layer of conductive carbon is coated on the surface of a normal aluminum foil to optimize the performance of battery.

Aluminum foil of lithium batteries can be divided into power battery foil, consumer battery foil, and energy storage battery foil. Among them, power battery foil is currently in the largest demand, accounting for more than 50%. Foreign suppliers of aluminum foil for lithium batteries are mainly concentrated in Japan, such as Toyo Aluminum and Hitachi Metals. Domestically, there are Dingsheng New Materials, Wanshun New Materials, Huaxi Aluminum, Nannan Aluminum, Sifangda, etc. Benefiting from the high growth in demand for power batteries and energy storage batteries, leading lithium batteries have expanded their production. According to Huaan Securities, based on the consumption of 400–600 tons of aluminum foil per GWh lithium battery, it is estimated that the global lithium battery

aluminum foil is expected to reach 454–681 000 tons in 2025. The Na-ion press conference of CATL is expected to realize the industrial chain in 2023. The anode and cathode of sodium-ion batteries are made of aluminum foil. By then, the market space of lithium-ion aluminum foil will expand.

1.2.6.3 Others

Nickel is relatively inexpensive, has a good electrical conductivity, and is relatively stable in acid and alkaline solutions. Nickel can be used as both a cathode electrode current collector and a negative electrode current collector. Nickel is matched with both cathode active material LFP and negative active materials such as nickel oxide, sulfur, and carbon–silicon composite materials. The shape of the nickel current collector usually has two types: foamed nickel and nickel foil. Due to the well-developed pores of the foamed nickel, the contact area with the active material is large, thereby reducing the contact resistance between the active material and the current collector. However, when nickel foil is used as an electrode current collector, as the number of charging/discharging process increases, the active material is likely to fall off, which greatly affects the performance of the battery.

Stainless steel refers to alloy steel containing nickel, molybdenum, titanium, silver, copper, iron, and other elements, which also has good electrical conductivity. It is stable enough to resistant to corrosive media such as air, steam, water, acid, alkali, and salt. The surface of stainless steel is also easy to form a passive film, which can protect its surface from corrosion. At the same time, stainless steel can be processed thinner than copper, which has the advantages of low cost, simple process, and large-scale production.

Carbon nanotubes (CNTs) are a new type of one-dimensional nanomaterials. The unique graphitized structure and ultra-high aspect ratio of CNTs result in its excellent electrical properties; at the same time, the density of CNTs is extremely low compared to that of metals. Therefore, the thin film prepared with CNTs is expected to replace aluminum and copper foil and become a new-generation LIB collector.

1.3 Development and Trends in Power Lithium-Ion Battery

1.3.1 The Layout of Lithium-Ion Battery Production Capacity

In 2025, the global power battery demand will exceed 1 TWh. Driven by the high demand, power battery companies are vigorously expanding their production capacity. According to incomplete statistics of the starting point lithium battery, as of August 2021, 20 Asian companies including CATL, LG New Energy, CALB, Yiwei Lithium, SKI, and BYD, have a planned production capacity of more than 3000 GWh. In the past two years, European and American companies are accelerating the deployment of power LIBs. A report published in June by EU NGO Transport and Environment (T&E) put the total number of Gigafactory's built or under construction in existing projects in Europe to 38, with a combined expected annual output of

1000GWh. Across Europe, several local battery companies, including Sweden's Northvolt, France's Verkor, Britain's Britishvolt, Norway's Freyr, and Slovakia's InoBat Auto have been established, and large-scale battery production plans have been announced. In addition, Germany plans to invest 1 billion euros to support German lithium-ion power battery production by 2021. With the successive completion of 38 super factories, the production of electric vehicle batteries in Europe will also increase significantly. It is expected to produce 460GWh in 2025 and 1140GWh in 2030, which is 13 times of the expected supply this year (87GWh). In addition, to ensure the supply of batteries, major car companies have begun to build their own lithium-ion power battery factories to achieve cost control and control the initiative of lithium-ion power battery supply. Among them, Tesla plans to build the future Gigafactory near Berlin into one of the largest factories in the world, with an expected production capacity of 250GWh in 2030. Volkswagen Group plans to join hands with its partners to build six battery factories in Europe. Overall, Europe is expected to become the world's second largest supplier of power LIBs for electric vehicles in the near future, which will bring huge challenges to the Asian lithium-ion power battery market.

1.3.2 The Changing Trend of Lithium-Ion Battery Material Types

Statistics from SMM show that the total installed capacity of power LIBs in China continues to grow from 16GWh in 2015 to 154GWh in 2021 (Figure 1.9). In terms of battery types, NCM batteries and LFP batteries dominate the lithium-ion power

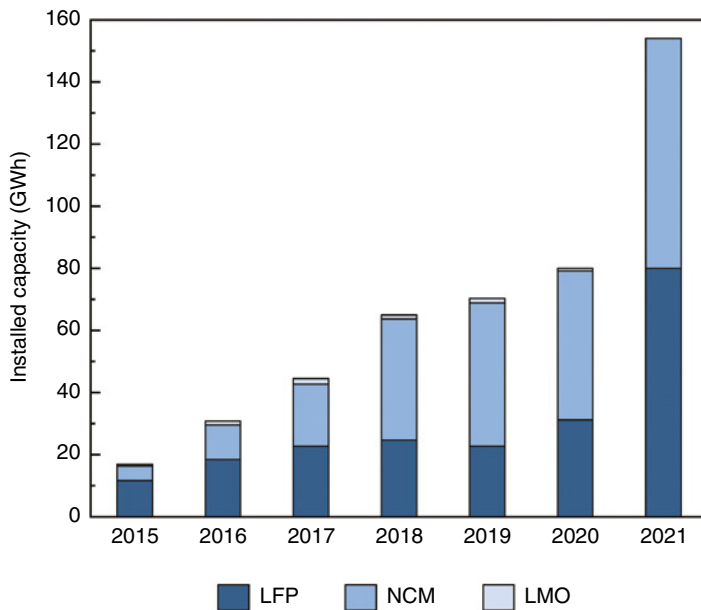


Figure 1.9 Main power batteries by material types and their installed capacities in China from 2015 to 2021. *Source:* SMM.

battery market. The high energy density of NCM batteries is gradually favored by the passenger vehicle market, reaching more than 60% in 2019. LFP batteries are mainly used in commercial vehicles and pure electric passenger vehicles. With the introduction of BYD's blade battery technology in 2020 and the advancement of battery assembly technology, low-cost LFP batteries will achieve breakthroughs in energy density. It will lead to an increase in the sales of LFP batteries in the short term; thus, the market share will rebound in 2020–2021.

1.3.3 Development Goals and Plans in Various Regions of the World

Since the commercial use of LIBs in 1991, the battery system with the LCO/LMO/LFP as the cathode electrode and the graphite as the anode electrode has basically been inherited. In recent years, due to requirements for the higher energy and safety of power LIBs, its technological development was thus boosted. Figure 1.10 shows the development history, status, and future trends of lithium batteries around the world [12]. It is worth noting that the energy density of the Panasonic 18650 battery has only approximately tripled between 1990 and 2015. At present, lithium batteries with an energy density of 240 Wh/kg have achieved mass production, and lithium batteries with an energy density of 300 Wh/kg or even 400 Wh/kg are still in development. As a result, countries worldwide are developing and planning to achieve electric vehicle range (>500 km, charging time <20 minutes) and cycle life (>3000 cycles).

In China, the power battery development plan at various stages is very clear. The “Technical Roadmap for Energy-Saving and New Energy Vehicles” has detailed the corresponding requirements for each stage of China's power LIBs and new type batteries, which are mainly divided into three stages:

- (1) In 2020, LIBs should meet the needs of pure electric vehicles over 300 km, that is, the energy density of a single unit will reach 300 Wh/kg and 600 Wh/L,

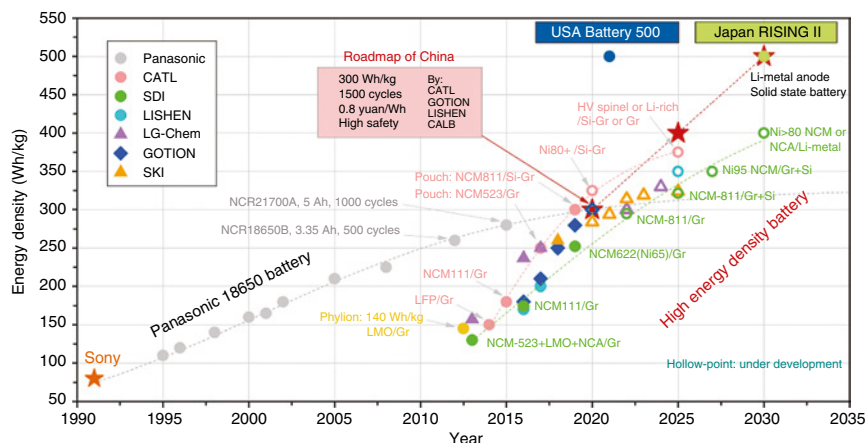


Figure 1.10 The history, current state, and development route of LIBs.
Source: Lu et al. [12] / with permission of Elsevier.

- the unit cost will be reduced to 0.8yuan/Wh, and the cycle life will be 1500 times;
- (2) In 2025, LIBs should meet the needs of pure electric vehicles over 400 km, that is, the energy density of a single unit will reach 400 Wh/kg and 800 Wh/L, the unit cost will be reduced to 0.5 yuan/Wh, and the cycle life will be 2000 times;
 - (3) In 2030, LIBs should meet the needs of pure electric vehicles over 500 km, that is, the energy density of a single unit will reach 500 Wh/kg and 1000 Wh/L, the unit cost will be reduced to 0.4 yuan/Wh, and the cycle life will be 3000 times.

At present, against the goal of 300 Wh/kg, the research on power LIB technology mainly focuses on the development of Li-rich ternary cathode materials, silicon-carbon anodes, and electrolytes with wide voltage windows.

For example, Li-rich layered oxides (LLOs) deliver extraordinary capacity exceeding 250 mAh/g have greatly attracted research interests to further contribute the energy density. Despite the high capacity, LLOs suffer from poor rate performance and voltage fading as well as capacity decay during cycling. Guo et al. [13] from the Ningbo Institute of Materials Technology and Engineering (NIMTE)-CAS constructed abundant nanoscale defects via chemical delithiation and built dual Al₂O₃ layer via hierarchical surface configuration to suppress surface lattice oxygen release. Besides, they also designed 3D porous LLO and created oxygen vacancies through gas-solid interface reaction, facilitating the ionic diffusion to enable excellent rate capability [14]. Based on the scientific achievements, a 300-ton pilot production line of LLO cathode material has been established. By 2030, all-solid-state (ASS) batteries are expected to achieve large-scale commercialization, which will further promote the application of metallic lithium anodes to meet the energy density requirement of 500 Wh/kg.

In Japan, New Energy and Industrial Technology Development Organization (NEDO) released “Research and Development Initiative for Scientific Innovation of New Generation Battery” (RISING II) project in 2018. In terms of power batteries, they also focus on the research and development of ASS batteries. By 2025, the first generation of ASS batteries will be popularized, and its energy density will reach 300 Wh/kg. By 2030, the second generation will be popularized and is expected to realize 500 Wh/kg. In addition, Japan is also striving for developing new types of batteries such as sulfide-based ASS batteries and zinc-based batteries.

In South Korea, the Korea Battery Industry Association set the power electricity roadmap and four key materials roadmaps in 2018. The specific energy density of power battery will reach 330 Wh/kg and 800 Wh/l in 2025, and the cycle life of power battery will reach 15 years with 1000 cycles.

Europe: LIBs are part of the EU’s efforts to develop a decarbonized and renewable energy society. The “Horizon 2020” plan, the latest framework of the EU’s 10-year economic development plan, proposes to invest a total of 77.028 billion euros in industry, scientific research, business, and other fields from 2014 to 2020, of which a total of 114 million euros are earmarked for batteries, including lithium battery materials and transmission models and research and innovation of advanced lithium batteries. The energy density of power LIBs will reach 250 Wh/kg in 2025 and will continue to increase to 500 Wh/kg by 2030. In addition, the energy technology

strategic development plan “Battery 2030+” proposes to invest more than 100 billion euros to promote the comprehensive development of the lithium-ion power battery industry chain, covering the entire process from raw materials to battery recycling.

The United States: The US Department of Energy (DOE) established the Battery 500 program in 2021. The total investment in the next five years is more than US\$ 50 million, and the goal is to develop lithium metal batteries and replace the existing graphite anode with metal lithium, so that the energy density can reach 500 Wh/kg and the number of cycles will be 1000 times.

1.3.4 Critical Challenges for the Future Lithium-Ion Power Battery Industry

1.3.4.1 Reducing the Cost of Lithium-Ion Power Battery

The cost of raw materials accounts for more than 60%, which is the key way to reduce the overall cost of power battery [14]. Among them, the cost reduction of cathode materials lies in the metal resource price of upstream Li, Co, and Ni and the manufacturing process; the cost reduction of anode material lies in the raw material purchase of needle coke and graphite processing technology; the cost reduction of separator material lies in the improvement of the yield of the production process and the improvement of equipment; and the cost reduction of electrolyte is limited, and its large-scale production may help to reduce the price. However, under the existing technology, the cost reduction space for raw materials is very limited. The focus of cost reduction will be at the module and pack level, such as BYD’s introduction of blade technology, which will reduce battery system costs by 20–30%.

1.3.4.2 Improving the Energy Density of Power Battery

The path of energy density improvement mainly includes two aspects: core density and system density. In terms of cells, new technological breakthroughs such as ASS and ternary Li-rich batteries need to be achieved, and technologies such as changing the ratio of cathode and anode electrodes electrode materials need to be optimized. In terms of battery packs, the modular cell-to-pack (CTP) scheme is designed to improve the efficiency of battery pack grouping, optimize the layout structure, and use low-density materials. For example, CATL’s CTP technology, based on a NCM material battery technology architecture, can increase space volume utilization by 15–20%, reduce the number of internal components by 40% and indirectly increase system energy density by 10–15%. In addition, the blade battery designed by BYD based on LFP battery technology architecture can increase the volume energy density by 50%.

1.3.4.3 Improving Safety of Power Battery

Fire concerns of power battery generally includes charging spontaneous combustion (charging current is large), collision combustion (battery damage, internal short circuit), driving spontaneous combustion (battery damage, internal short circuit), and wading spontaneous combustion (battery sealing is insufficient, liquid

causes external short circuit). These problems can cause the whole battery pack temperature to rise sharply, thermal runaway, and then spontaneous combustion explosion. Battery thermal runaway can be controlled by battery design or supporting facilities. For example, it can prevent the cathode electrode from releasing oxygen; suppressing the flammability of electrolyte; improving the sealing, heat insulation, and impact resistance of battery pack structure; and optimizing the battery thermal runaway management system.

1.3.4.4 Recycling Power Battery

The full life cycle of power battery involves many subjects and links. Only some enterprises master the life cycle data of their products, and the fragmentation of information restricts the recycling and reuse of power LIBs. In addition, retired batteries lack testing standards, and battery residual value assessment technology as well as the talent reserves are insufficient. In the process of battery design and manufacture, the manufacturers did not consider recycling and disposal factors.

1.4 Analysis of the Supply and Demand of Critical Metal Raw Material Resources for Power Lithium-Ion Batteries

LIBs play a crucial role in global electrification and help to tackle climate change. Global battery demand is expected to scale up 19 times compared to the current level by the end of 2030 [15]. This can be hardly achieved if relying on the current way of how the materials are sourced, produced, and used. The challenge can only be overcome with the collaborative efforts through the entire value chain. Among all the challenges, the supply of critical metal raw materials, i.e. lithium, nickel, and cobalt, are one of the most tough ones. Material production of lithium battery is very dependent on these critical metals. A recent report by the IEA suggests a typical electric car requires six times the mineral inputs of a conventional car [16], and currently, they are predominantly extracted from minerals on earth. These minerals are unevenly distributed geographically, with lithium raw material mainly produced in Australia, nickel in ASEAN countries (Indonesia and Philippines), and cobalt in the Democratic Republic of Congo (DRC). The scale-up in mineral sourcing might lead to negative social, environmental, and integrity impacts in these regions, especially for cobalt. The DRC is one of the world's least developed countries, whose economy hugely relies on cobalt. A total of 10–12 million people depend directly or indirectly on mining, and 80% of exports are mining products. However, severe social risks have been well documented in the DRC's artisanal mining industry, which include hazardous working conditions, deaths due to poorly secured tunnels, potentially various forms of forced labor, the worst forms of child labor, and exposure to fine dusts and particulates as well as the DNA-damaging toxicity [15]. All these issues have resulted in a rather fragile front-end LIB supply chain.

At the same time, driven by the strong demand in the field of lithium batteries, the price of these key metal materials has shown a rapid upward trend since the beginning of 2021, showing supply shortages of varying degrees.

To underscore the importance and potential risks associated with these critical metal resources, the White House's 100-day review under Executive Order 14017, issued in June 2021, dedicated an entire chapter to review the topic and concluded that reliable, secure, and resilient supplies of key strategic and critical materials are essential to the US economy and national defense.

In the successive part of this chapter, the geographical distribution of lithium, nickel, and cobalt and their production status have been introduced, followed by a discussion on the supply and demand outlook.

1.4.1 Geographical Distribution of Critical Metal Raw Materials and Their Production Status

1.4.1.1 Lithium

Lithium is a relatively rare element on earth, whose abundance in the earth's crust is 0.0065%, ranking 27th. Although seawater contains rich lithium resources, around 260 billion tons, currently it is not commercially available as the content is low, only 0.17 mg/l.

According to the latest USGS report [17], in 2020, the global lithium mine reserves were approximately 21.06 million tons. Among them, Chile has the most reserves, about 9.2 million tons, accounting for about 43.7% of global reservation. Australia ranks second in reserves, with reserves of about 4.7 million tons in 2020, accounting for 22.3% of global reserves. Argentina ranks the third in resource reserves. The detailed data of global lithium reserves in 2020 are listed in Table 1.3.

The global lithium resources are mainly divided into two types, namely rock minerals and brine minerals, of which closed basin brine accounts for 58%; pegmatite (including lithium-rich granite) accounts for 26%; hectorite clay accounts for 7%; and oilfield brine, geothermal brine, and lithium borosilicate ore each account for 3% [18].

The world's brine lithium resources are mainly distributed in the "Lithium Triangle" plateau areas of Chile, Argentina, and Bolivia in South America; western United States; and western China. The world's rock lithium resources are mainly distributed in Australia, China, Zimbabwe, Portugal, Brazil, Canada, Russia, and other countries.

Approximately 82 200 metric tons of lithium ore were mined in 2020 globally. Australia is the main contributor, with 40 000 metric tons lithium yield, accounting for almost half of the world's production, followed by Chile and China, with 21.9% and 17.0% contributions, respectively. The detailed data of global lithium mine yield in 2020 are listed in Table 1.4.

1.4.1.2 Nickel

According to the latest USGS report [17], in 2020, the global nickel mine reserves were approximately 94 million tons. Among them, Indonesia has the most reserves, about 21 million tons, accounting for about 22.4% of world total. Australia ranks

Table 1.3 Global lithium mine reserves in 2020.

Country	Lithium mine reserves (metric tons)	Percentage (%)
United States	750 000	3.6
Argentina	1 900 000	9.0
Australia	4 700 000	22.3
Brazil	95 000	0.5
Canada	530 000	2.5
Chile	9 200 000	43.7
China	1 500 000	7.1
Portugal	60 000	0.3
Zimbabwe	220 000	1.0
Rest of world	2 100 000	10.0
World total	21 055 000	100

Source: Data from USGS [17].

Table 1.4 Global lithium mine yield in 2020.

Country	Lithium mine yield (metric tons)	Percentage (%)
United States	—	—
Argentina	6 200	7.5
Australia	40 000	48.7
Brazil	1 900	2.3
Canada	—	—
Chile	18 000	21.9
China	14 000	17.0
Portugal	900	1.1
Zimbabwe	1 200	1.5
Rest of world	—	—
World total	82 200	100

Source: Data from USGS [17].

second in reserves, with the amount of about 20 million tons in 2020, accounting for 21.3% of global reserves. Brazil ranks third in resource reserves. The detailed data of global nickel reserves in 2020 are listed in Table 1.5.

The global nickel resources are mainly divided into two types: laterite nickel and nickel sulfide ore, which account for 60% and 40% of total reserves, respectively. Laterite nickel mines are mainly distributed in countries within the Tropic of Cancer, including Australia, New Caledonia, Indonesia, Brazil, and Cuba, while

Table 1.5 Global nickel mine reserves in 2020.

Country	Nickel mine reserves (metric tons)	Percentage (%)
United States	100 000	0.1
Australia	20 000 000	21.3
Brazil	16 000 000	17.0
Canada	2 800 000	3.0
China	2 800 000	3.0
Cuba	5 500 000	5.9
Dominican Republic	—	—
Indonesia	21 000 000	22.4
New Caledonia	—	—
Philippines	4 800 000	5.1
Russia	6 900 000	7.3
Rest of World	14 000 000	14.9
World total	93 900 000	100

Source: Data from USGS [17].

nickel sulfide mines are mainly distributed in Russia, Canada, Australia, South Africa, and China.

Laterite nickel mines are mostly open-pit mines, which are convenient for mining, but the processing technology is more complicated due to their low grade. With the recovery of nickel prices and progress in the refining technology, the proportion of primary nickel produced from laterite ore has steadily increased. The proportion of nickel supply from laterite ore increased from 51% in 2017 to 62% in 2019, which has completely changed the previous dominated industry pattern of sulfide ore.

Approximately 2.5 million metric tons of nickel ore were mined globally in 2020. The main contribution comes from the ASEAN region, where Indonesia produced 0.76 million tons (accounting for 30.7% of world's production), and Philippines produced 0.32 million tons with 12.9% worldwide share. Russia ranks third, with a yield of 0.28 million tons similar to that of Philippines, accounting for 11.3% of the global total. The detailed data of global nickel mine yield in 2020 are listed in Table 1.6.

1.4.1.3 Cobalt

According to the latest USGS report [17], in 2020, the global cobalt mine reserves were approximately 7.1 million tons. Among them, Congo has the most reserves, about 3.6 million tons, accounting for about 50.5% of the world's total. Australia ranks second in reserves, with reserves of about 1.4 million tons in 2020, accounting for 19.6% of global reserves. Cuba ranks the third in resource reserves. The detailed data of global cobalt reserves in 2020 are listed in Table 1.7.

Table 1.6 Global nickel mine yield in 2020.

Country	Nickel mine yield (metric tons)	Percentage (%)
United States	16 000	0.6
Australia	170 000	6.9
Brazil	73 000	2.9
Canada	150 000	6.1
China	120 000	4.8
Cuba	49 000	2.0
Dominican Republic	47 000	1.9
Indonesia	760 000	30.7
New Caledonia	200 000	8.1
Philippines	320 000	12.9
Russia	280 000	11.3
Rest of World	290 000	11.7
World total	2 475 000	100

Source: Data from USGS [17].

Table 1.7 Global cobalt mine reserves in 2020.

Country	Cobalt mine reserves (metric tons)	Percentage (%)
United States	53 000	0.7
Australia	1 400 000	19.6
Canada	220 000	3.1
China	80 000	1.1
Congo	3 600 000	50.5
Cuba	500 000	7.0
Madagascar	100 000	1.4
Morocco	14 000	0.2
Papua New Guinea	51 000	0.7
Philippines	260 000	3.6
Russia	250 000	3.5
South Africa	40 000	0.6
Rest of World	560 000	7.9
World total	7 128 000	100

Source: Data from USGS [17].

Table 1.8 Global cobalt mine yield in 2020.

Country	Cobalt mine yield (metric tons)	Percentage (%)
United States	600	0.4
Australia	5 700	4.2
Canada	3 200	2.4
China	2 300	1.7
Congo	95 000	70.4
Cuba	3 600	2.7
Madagascar	700	0.5
Morocco	1 900	1.4
Papua New Guinea	2 800	2.1
Philippines	4 700	3.5
Russia	6 300	4.7
South Africa	1 800	1.3
Rest of World	6 400	4.7
World total	135 000	100

Source: Data from USGS [17].

Cobalt rarely forms separate ores and is mostly associated with copper, nickel, manganese, iron, arsenic, lead, and other deposits. Cobalt resources usually exist in the following areas [17]: sediment-hosted stratiform copper deposits in Congo and Zambia; nickel-bearing laterite deposits in Australia and its nearby island countries, i.e. Indonesia, Philippines, New Caledonia, and Cuba; magmatic nickel-copper sulfide deposits hosted in mafic and ultramafic rocks in Australia, Canada, Russia, and the United States; manganese nodules and crusts on the floor of Atlantic, Indian, and Pacific Oceans

The total yield of cobalt mine was 135 000 metric tons in 2020, predominantly from Congo. Congo alone has produced 95 000 tons, contributing to over 70% of the global total. Russia and Australia ranked second and third, accounting for 4.7% and 4.2%, respectively. The detailed data of global nickel mine yield in 2020 are listed in Table 1.8. Compared to lithium (Table 1.4) and nickel (Table 1.6), cobalt presents the most unbalanced global supply, which is extremely dependent on a single country, i.e. Congo.

1.4.2 Supply and Demand Outlook of Critical Metal Raw Materials

1.4.2.1 Lithium

The Lithium Carbonate Equivalent (LCE) in 2020 is around 369 000 tons, among which, LIBs account for 59%, while the rest 41% comes from the industrial fields. The demand domain in LIB can be further divided into consumer electronics, electric vehicles, electric mobilities, e.g. scooters, electric bikes, and energy storage.

While electric vehicles have only begun to grow in the last few years, their share is increasing very fast, already consuming 35% of global lithium demand. Energy storage is another fast-growing area, benefiting from energy transition to solar and wind, although it contributed only 3% in 2020.

In the industrial field, lithium is normally used as a raw material for glass and ceramics, grease, flux, and polymers, and it can also be used in solid fuels, aluminum smelting, and other fields. Lithium demand in the industrial field is rather stable, and its share in total lithium demand will be significantly reduced with the massive adoption of electric vehicles and energy storage (Figure 1.11).

1.4.2.2 Nickel

The total nickel demand in 2020 is around 2.39 million tons [18]. As the main characteristics of nickel are high hardness and oxidation resistance, most of the world's finished nickel (near 70%) has been used in the production of stainless steel. The major applications for nickel and their corresponding demand share are illustrated in Figure 1.12.

The contribution from the battery sector only account for 6% of the overall nickel consumption in 2020, which is dramatically different from that of lithium (Figure 1.11) and cobalt (Figure 1.13), where the contribution from the battery sector is greater than 50%. However, with the continuous advancement of electric vehicles, the demand for nickel in the battery industry is expected to grow rapidly in the next few years. Nickel sulfate is a key raw material for LIBs, mainly used in the production of NCM and NCA battery precursors/cathode materials. The market

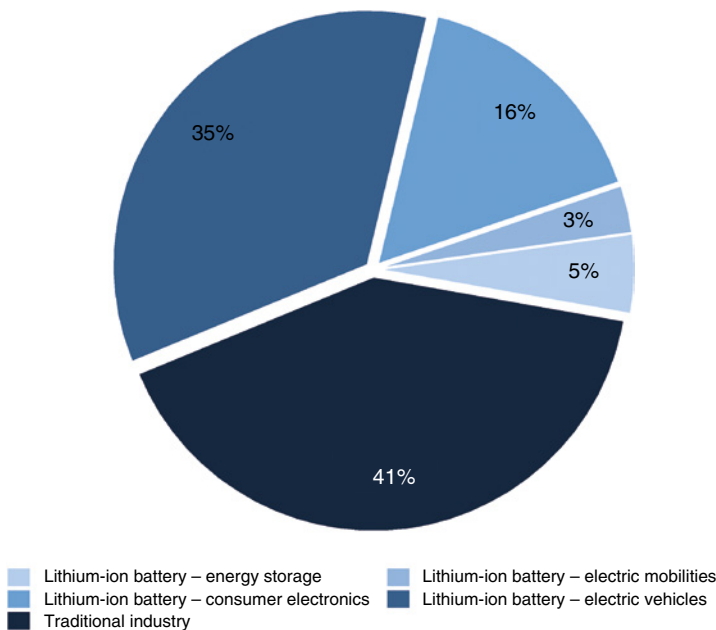


Figure 1.11 Lithium demand by application in 2020. *Source:* Data from Antaika [18].

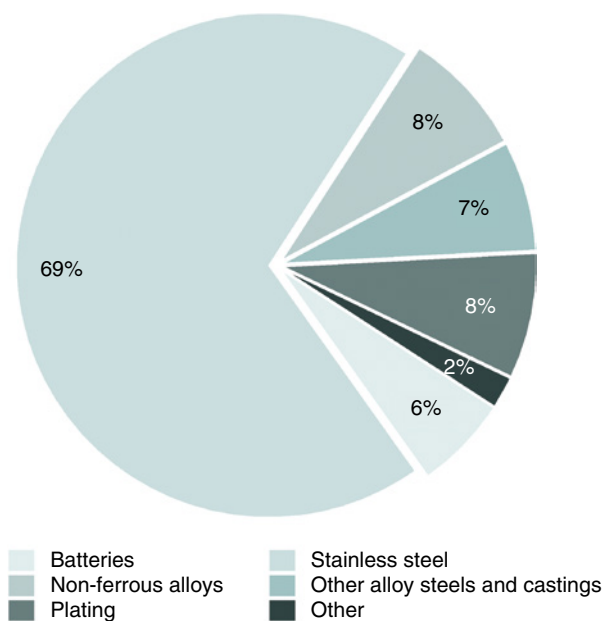


Figure 1.12 Nickel demand by application in 2020.
Source: Adapted from Fraser et al. [19].

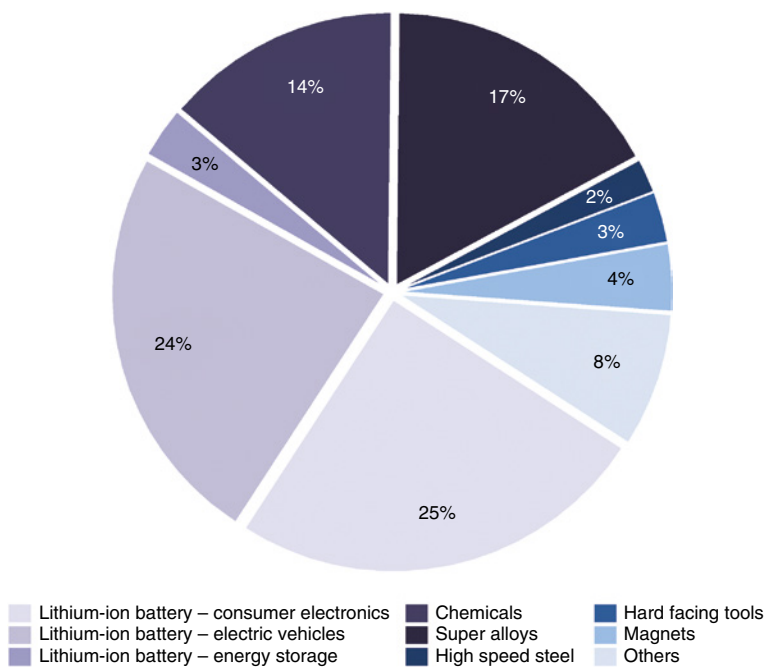


Figure 1.13 Cobalt demand by application in 2020. Source: Cobalt Institute [20].

presently is moving toward NCM ternary batteries of high nickel content because of the pursue for higher battery energy density and continued cost reduction (to replace cobalt with nickel to reduce total raw material cost), and the considerations of supply chain security, thereby further stimulating the demand for nickel in batteries.

1.4.2.3 Cobalt

The total cobalt demand in 2020 is around 140 000 tons [18], where metallurgical application accounts for 48% and LIBs for the rest 52%. The details can be found in Figure 1.13. For metallurgical application, cobalt's strength and resistance nature at high temperature make it an ideal choice to produce high-temperature alloys in power plants, high-speed steel drill bits, and blades as well as for use in hard face, carbide, and diamond tools for cutting applications and magnets. In addition, cobalt can also be used to make catalysts and desiccants. As for the LIB, cobalt is an important raw material for its manufacturing. Cobalt tetraoxide is used for LCO battery cathode, whereas cobalt sulfate is used for cathode material of ternary NCM and NCA battery. In the past 10 years, the growth in demand for cobalt has mainly come from the development of 3C batteries, such as use in smartphones, tablets, laptops, notebooks, computers, etc. Due to the rapid adoption of electric vehicles, the increase in cobalt demand is mainly driven by the battery sector in these two years, therefore, adding up the proportion of cobalt used in LIBs.

1.4.3 Scenario Without Recycling

Various research organizations [15, 21, 22] have predicted that due to energy transition toward renewable energy, especially in the electric vehicle and energy storage sectors, the demand for lithium, nickel, and cobalt will experience a huge increase in this decade and onward. Global battery alliance (GBA) [15, 21] predicts 6.4 times increase of lithium, 2.1 times increase of cobalt, and 24 times increase of class I nickel in 2030 as compared to that in 2018 [15]. The prediction from IEA is even more aggressive; according to their sustainable development scenario (SDS), the demand of lithium will grow by 43 times, nickel by 41 times, and cobalt by 21 times in 2040 when compared with that in 2020 [21].

At present, the supply of either lithium, nickel, or cobalt mainly comes from primary mineral resources. From the initial mine exploration to a new mine that can be officially put into operation is a long process, which usually takes up more than 10 years. Because of this, it is rather difficult to fulfill such tremendous growth; lithium, nickel, and cobalt minerals all have their key challenges [22], which are summarized below in Table 1.9, and all of them will experience shortage in this decade.

IEA predicts that the deficit in lithium supply will start from 2023 onward accompanied by a rapid gap increase, though the supply and demand unbalance may even begin in 2021. As for cobalt, the supply shortage will occur from 2024 and experience a similar trend as lithium. Nickel's supply is the most secure among the three, where the unbalance only starts from 2028 (Figure 1.14).

Table 1.9 Lithium, nickel, and cobalt minerals key challenges.

Mineral	Key challenges
Lithium	<ul style="list-style-type: none">● Possible bottleneck in lithium chemical production as many smaller producers are financially constrained after years of depressed prices● Lithium chemical production is highly concentrated in a small number of regions, with China accounting for 60% of global production (over 80% for lithium hydroxide)● Mines in South America and Australia are exposed to high levels of climate and water stress
Nickel	<ul style="list-style-type: none">● Possible tightening of battery-grade Class 1 supply, with high reliance on the success of HAPL projects in Indonesia; HAPL projects have track records of delays and cost overruns● Alternative Class 1 supply options (e.g. conversion of NPI to nickel matte) are either cost-prohibitive or emissions-intensive● Growing environmental concerns around higher CO₂ emissions and tailings disposal
Cobalt	<ul style="list-style-type: none">● High reliance on the DRC for production and China for refining (both around 70%) set to persist, as only a few projects are under development outside these countries● Significance on artisanal small-scale mining makes the supply vulnerable to social pressures● New supply is subject to developments in nickel and copper markets as some 90% of cobalt is produced as a by-product of these minerals

Source: Data from IEA [22].

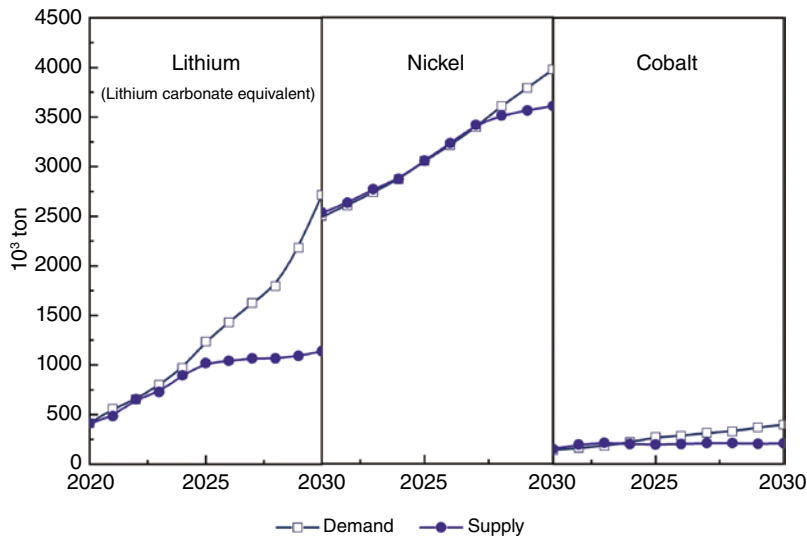


Figure 1.14 Supply and demand forecast for lithium, nickel, and cobalt.
Source: Data from IEA [22].

1.4.4 Scenario with Recycling

Metal recycling has the potential to be a significant resource of secondary supply, although it comes with its own set of challenges. Recycling comprises physical collection and metallurgical processing. Potential sources for recycling include tailings from process scrap used during manufacturing and scrap from end-of-life products.

All the LIBs installed today will eventually reach the end of life depending on the applications. The normal service life for a battery is ~5 years for consumer electronics, ~6 years for small electric mobilities, ~8–10 years for electric vehicles, and ~10 years for energy storage. When these batteries are retired, they can become valuable resources containing considerable amount of lithium, nickel, and cobalt. Especially those LIBs installed in electric vehicles, whose capacities are usually higher than 50 kWh, will cause serious safety and environmental problems if they are not properly disposed. According to the study from Circular Energy Storage (CES) [23], the expected EV battery that will reach the end of life is expected to be 174 GWh worldwide in 2030. This number is even larger than the total installed electric vehicle battery capacity in 2020, which is 136.3 GWh based on GGII data [24]. The expected retired battery amount is listed in Figure 1.15.

While not many electric vehicle batteries have reached the end of their normal life, even fewer of them will eventually be recycled. As more such batteries are retired after mid-century, along with more regulated recycling channels and strict recycling policies and regulations, recycling will be an important addition to the main sources of lithium, nickel, and cobalt. Recycling batteries will help to curb volatility in the supply chain and prices of raw materials or battery manufacturing

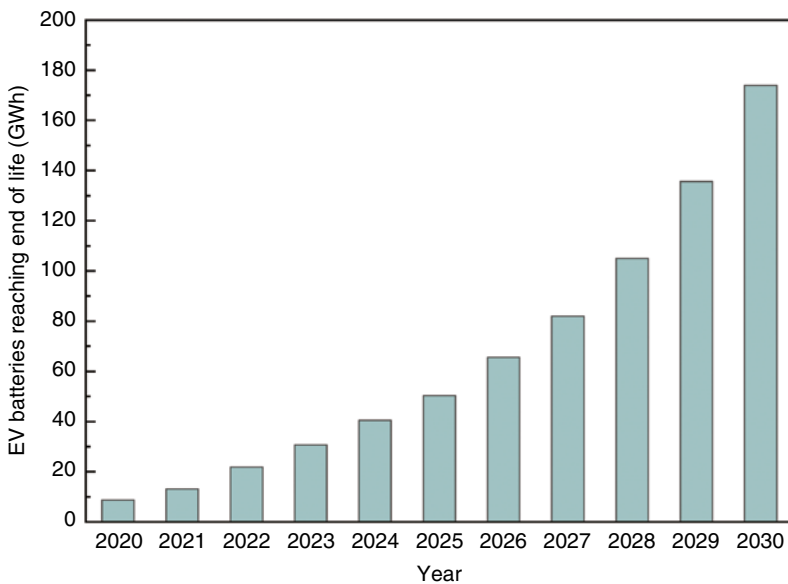


Figure 1.15 EV battery reaching end of life.

and can therefore play a key role in alleviating energy security concerns in countries that rely heavily on imports of these minerals. Based on the recent IEA report, in their SDS scenario, recycling and reuse of EV and storage batteries can reduce the primary supply requirement for minerals by 5% for lithium, 8% for nickel, and 12% for cobalt in 2040 [22].

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2

Battery Recycling Technologies and Equipment

2.1 Brief Introduction of Lithium-Ion Battery Recycling

Generally, when the battery capacity decays to less than 80% of the initial capacity, it meets the conditions of electric vehicle (EV) power battery retirement. Most batteries are still in good working conditions, thus they can continue to be used in the application scenarios with lower performance requirements for EV batteries cascade utilization. Such EV batteries cascade utilization includes energy storage systems, two- or three-wheeled EVs, electric forklifts, and low-speed vehicles. Battery cascade utilization seems to extend the battery's service life. With the current rapid progress in battery technology and the ever-increasing battery energy density, it is clear that prolonging the battery's life is equal to the inefficient use of battery key metal resources. By contrast, it is the best way to maximize the use efficiency of metal resources by efficient recycling of critical metals in the spent battery and remanufacturing of next generation of high energy density battery. In addition, we compared the carbon forecast model in the whole life cycle of the raw, regenerated, and cascade utilization NCM and LFP batteries in the application scenarios of two-wheeled EV (Figure 2.1). Then we found that the cascade utilization battery does have lower carbon emissions than raw and regenerated batteries at the early use stage. However, such an advantage is gradually offset by more carbon emissions with increased mileage, thus resulting in the highest carbon emissions over the life cycle.

On the contrary, the regenerated battery achieves the lowest carbon emissions over the life cycle. More importantly, current industry standards for cascade utilization of spent batteries are not mature enough and lack scientific detection methods; however, many small workshop processing enterprises emerge, resulting in numerous security risks. Therefore, we believe that efficient recycling is a more reasonable technical route to deal with retired power batteries than the cascade utilization of batteries.

The recycling of spent power batteries, as one of the critical links in the new energy vehicle industry chain, has the highest environmental risk and needs



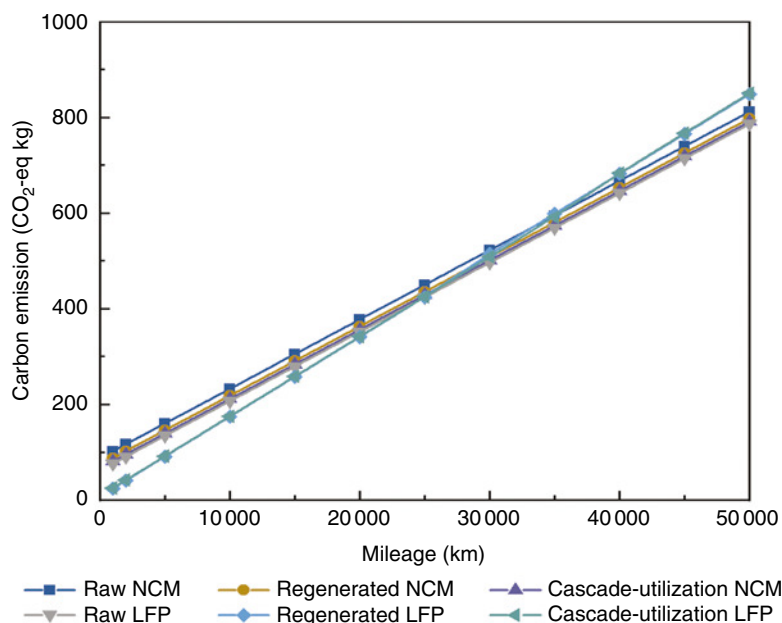


Figure 2.1 The carbon emission in the whole life cycle of the raw, regenerated, and cascade utilization NCM and LFP batteries in the application scenarios of two-wheeled EV.

technological progress urgently. Starting from 2018 to 2019, there will be many power batteries entering the decommissioning period in China. The residual energy detection, dismantling, cascade utilization of spent batteries, fine sieving of materials, and in-depth utilization of resources will form an independent recycling industry chain, which is also different from the traditional industrial system. In terms of cascade utilization, several problems should be solved. Firstly, the compatible reorganization of heterogeneous spent power batteries should be achieved. The rapid detection of residual energy and intelligent screening of hybrid state batteries should be performed. The safe/automated dismantling still requires technological advancement and industrial support.

In terms of recycling, it requires substantial improvements in technology and equipment. There are several challenges that should be figured out. For instance, how to achieve rapid discharge or process safety; how to improve the separation efficiency of black mass powder/copper/aluminum/organic materials; how to improve the degree of fine separation of materials; how to improve the level of process pollution control, and how to achieve the selective recovery of target metals; how to attain short-process material regeneration; etc. This chapter will analyze the current technology status of the recycling of spent power batteries, aiming to compare the characteristics and differences of different technologies and the degree of integration with the industry and the application situation, as well as to analyze their advantages and existing problems. Finally, the development perspective of the technology is outlooked.



2.2 Introduction of the Battery Recycling Process

2.2.1 Conventional Process of Cell Disassembly

There are two recycling methods for retired power batteries: cascade utilization and disassembly recycling. Cascade utilization is mainly for those batteries whose capacity was reduced, but not scrapped, which affects the regular operation of EVs. After retesting, screening, assembly, etc., those batteries can be used for low-speed EVs, energy storage power stations, etc., which do not require high energy density. Dismantling and recycling is the process of direct discharge, disassembly, sieving, and purification of spent power batteries to extract valuable metals such as cobalt, nickel, and lithium. Theoretically, when the battery's actual capacity decays from 80% to 60% of the rated capacity, it can be used for cascade utilization. The battery can be disassembled and recycled if the capacity is less than 60% of the rated capacity.

Related theoretical research and demonstration projects for battery cascade utilization started late in China, and large-scale commercial operations have not yet begun. Even at industry conferences, experts often discuss whether the cascade utilization of power batteries is a false proposition. The reasons are as follows:

- (1) Spent batteries enter the market again, which causes safety risks.
- (2) It is costly to test, screen, and assemble those spent batteries before being used in cascade utilization.
- (3) The production capacity of new batteries continues to rise, and the prices continue to fall.
- (4) The cascade utilization market for energy storage and low-speed EVs has not yet been released on a large scale.

Although cascade utilization is an ideal solution for battery recycling, the implementation has excellent resistance, so disassembly recycling is the current mainstream method of battery recycling in China. According to the different back-end metallurgical technology, the disassembly recycling process can be divided into hydrometallurgy recycling and pyrometallurgy recycling (Table 2.1).

The purpose of the dismantling is to sort resources such as separators, shells, copper, and aluminum particles and to collect the positive and negative electrode materials into the hydrometallurgy process while avoiding electrolyte pollution. Generally, the disassembly process can be divided into five stages according to different disposal purposes: discharge and crushing, electrolyte disposal, separator recovery, positive and negative electrode powder preparation, and copper–aluminum separation (Figure 2.2).

Discharge crushing: The battery cell is crushed under gas protection after the discharge process. Gas protection ensures no explosion and combustion during the battery crushing process. The crushed product enters the low-temperature drying device to volatilize electrolyte. At present, many battery recycling manufacturers



Table 2.1 Summary of battery dismantling and recycling technologies.

Classification	Process composition	Specific method	Advantages and disadvantages
Hydrometallurgy recycling	Disassembly	Discharging, crushing, sieving, and separating the outer casing and powder of the battery, and processing the waste liquid and waste gas generated during the dismantling process	Advantages: high recovery rate of valuable metals, low processing cost, and good process stability
	Hydrometallurgy	Sieving and recovering valuable metals such as nickel, cobalt, manganese, and lithium in the form of the element, compound, or mixture through various methods such as dissolution, extraction, precipitation, electrolysis, etc.	Disadvantages: the little amount of processing, a large amount of wastewater, complex process flow
Pyrometallurgy recycling	Disassembly	Discharging, crushing, sieving, and separating the outer casing and powder of the battery, and processing the waste liquid and gas generated during the dismantling process	Advantages: simple process flow and wide application range Disadvantages: expensive
	Pyrometallurgy	The battery cell is reduced and roasted, and the cobalt and nickel metals are reduced in the form of iron alloy, and aluminum, fluorine, chlorine, etc. become part of the residue, and then the alloy and the residue are separated	equipment, high energy consumption, low purity of valuable metals, and may produce highly toxic gases

in China are attempting to crush batteries without discharging, but a slightly larger processing capacity (more than 1 ton/h) will cause explosion and combustion.

Electrolyte disposal: The bag dust collection device collects the powder of the crushing process and volatilization of electrolyte, then the electrolyte is condensed, the solidified organic solvent is burned, and the unsolidified gas is sprayed with the lye to remove F and P. In recent years, governments of various countries have attached great importance to environmental protection, requiring that waste gas, wastewater, and solid waste must be strictly controlled in the production process of enterprises.

Separator, metal, and other substances recovery: The separator is recovered by winnowing. Then the iron and nickel are separated by magnetic separation, and the remaining material enters the friction powder process.

Pulverization and separation of copper and aluminum: The positive and negative electrodes are powdered by high-speed friction, and then the remaining



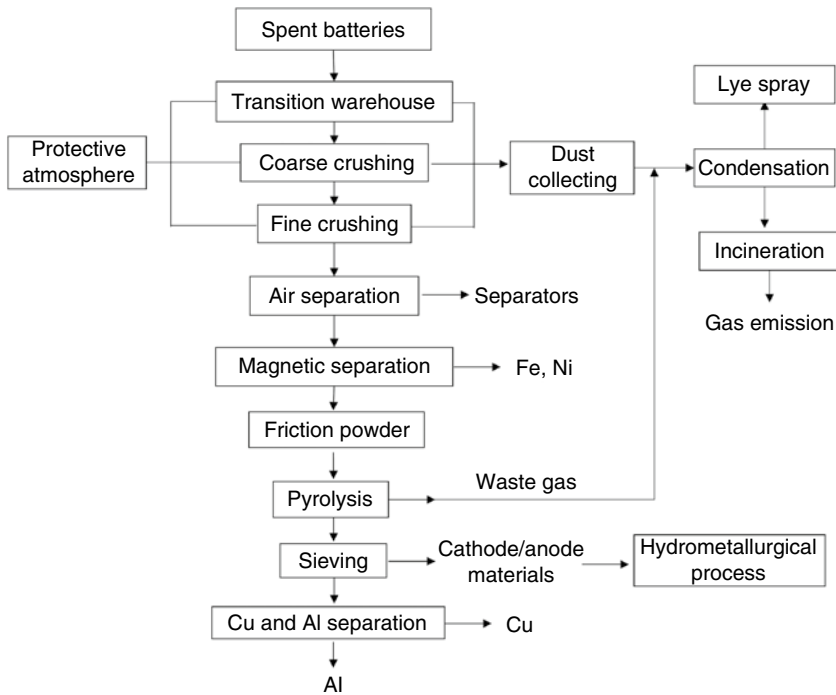


Figure 2.2 The conventional process of dismantling a single battery.

electrolyte and binder are cracked at high temperatures. The positive and negative powders are collected under the sieve and enter the back-end metallurgical process, and the copper and aluminum are sifted and separated by specific gravity sieving.

Hydrometallurgy is the most widely used process method for recycling spent batteries. Usually, the cathode and anode powders are acid leached (if the powder contains fine aluminum particles, the aluminum is removed by alkaline leaching first), and high value-added metals such as lithium, nickel, and cobalt are transferred to the solution. After the leachate is purified and impurity removed, the high value-added metal elements are separated by chemical precipitation, extraction, and other methods to obtain corresponding high value-added products. Commonly used leaching agents include inorganic acids, organic acids, and alkaline solutions. Taking the spent LFP battery as an example, the disassembled LFP positive and negative electrode powders are treated by acid leaching–impurity removal–lithium precipitation–evaporation process to recover battery-grade lithium carbonate and sodium sulfate (Figure 2.3).

Acid leaching process: The enriched LFP positive and negative powders are slurried, and concentrated H_2SO_4 is added to react with LiFePO_4 to produce Li_2SO_4 and FePO_4 . Lithium enters the leaching solution in the form of ions, and the negative graphite powder will be precipitated in the form of phosphorus iron slag



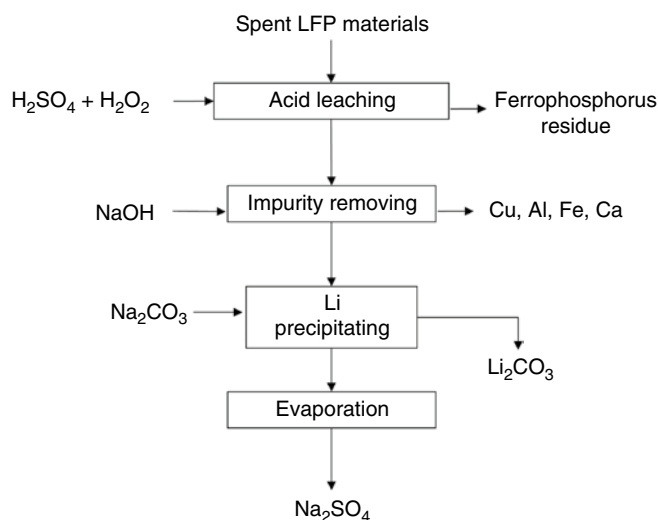


Figure 2.3 Hydrometallurgical process of spent LFP battery. *Source:* Adapted from Li [1].

and separated by filtration. Add hydrogen peroxide to change the divalent iron into trivalent iron to prepare for the subsequent impurity removal process.

Impurity removal process: The disposal process depends on the composition of impurity elements in the leaching solution. Usually, iron powder, NaOH , Na_2CO_3 , etc. are added to react with Cu, Al, Fe, and Ca in the solution to form a precipitate, and the filtrate is suctioned and filtered to enter the lithium precipitation process.

Lithium precipitation process: Use a certain amount of Na_2CO_3 solution as the bottom liquid; add the impurity removal liquid through a separatory funnel; stir and react at a specific temperature to produce Li_2CO_3 and lithium precipitation mother liquor; the lithium precipitation mother liquor enters the evaporation process, and the Li_2CO_3 is washed for several times to get the final product.

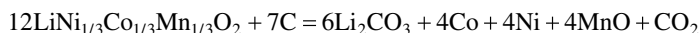
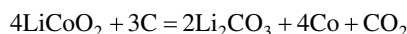
Evaporation process: H_2SO_4 is added to the lithium precipitation mother liquor to adjust the pH, and the mother liquor is evaporated in a rotary evaporator to obtain sodium sulfate.

The pyrometallurgical process is the high-temperature smelting of positive and negative anode powder in a reducing atmosphere, which needs a certain reducing agent, the reduction and decomposition of the battery cathode material, to obtain high value-added metal elements. The pyrometallurgical process finally gets alloy products to achieve the separation and recovery of high value-added metal in the retired lithium battery. At present, many industrial recoveries and decommissioning lithium battery processes are based on pyrometallurgical technology. There are two main reasons: first, high-temperature reaction chemical conversion rate is fast, the process is short, and the material is adaptable; second, the recycling industry of retired lithium batteries has just started, and many technological processes are still in progress. Therefore, it is easier to realize industrial applications by fully using existing metallurgical technology and equipment. However, retired lithium battery recycling is a complicated system engineering, and pyrometallurgy technology is



just one step in recycling. After high-temperature treatment, retired lithium batteries still need to adopt condensation, sieving, magnetic separation, and leaching to classify physical and chemical methods, such as processing, which eventually recycle to develop metal elements.

The powder is mixed with a reducing agent (usually limestone and coke) evenly and roasted at a high temperature above 600 °C so that high-priced metal ions such as Co^{3+} , Ni^{3+} , and Mn^{4+} in the powder are reduced to low-priced compounds or metallic elements and Li exists in the form of Li_2CO_3 . The main reaction mechanism is as follows:



The metal element or compound obtained after reduction is subjected to water immersion, acid leaching, purification, and other technologies to realize the separate recovery of metal Li, Co, Ni, and Mn. The main process flow is shown in Figure 2.4 [2, 3].

The advantages of the pyrometallurgical recovery process are strong processing capacity, simple process, and handling a wide variety of batteries. Still, the disadvantages are the high energy consumption of the equipment; it needs to be combined with the hydrometallurgical process, and a large number of the harmful gas will be generated during the process.

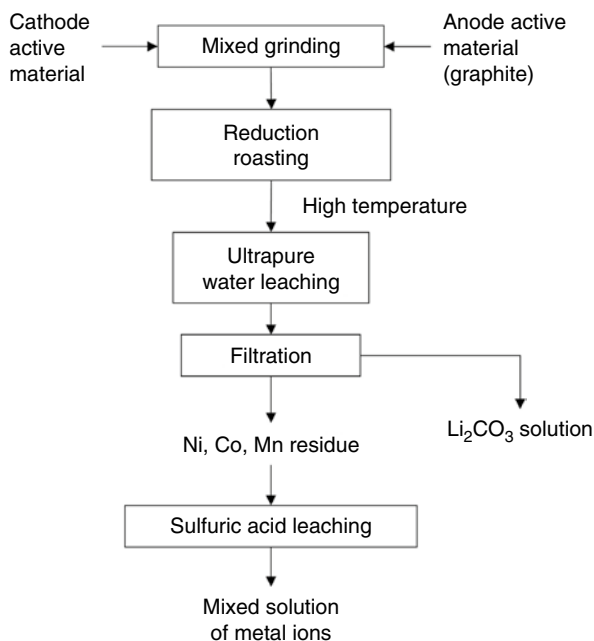


Figure 2.4 Pyrometallurgical process of scrapped LIBs.



2.2.2 Future Development Direction of Cell Recycling Process

- (1) Mechanical disassembly device for battery packs to avoid manual disassembly.
- (2) Live crushing, or find a fast, low-cost, nonpolluting discharge method to reduce the detection process and cost before the battery is crushed.
- (3) Electrolyte disposal: The electrolyte lithium hexafluorophosphate is easily volatile during the crushing process and is converted into hydrogen fluoride. At the same time, organic solvents are also hazardous waste. It is necessary to find a rapid collection and treatment device for the electrolyte.
- (4) Separate the cathode and anode plates in advance and make powder separately to ensure the good shape of the copper and aluminum plates.
- (5) Short-process recovery of LFP and NCM batteries, priority extraction of lithium from cathode and anode electrode powder, and preparation of iron phosphate.

2.3 Pretreatment Technology for Battery Recycling

2.3.1 Cell Discharge

In the comprehensive recovery of spent power LIBs, generating a large amount of heat is easy due to battery short circuit in the subsequent disassembly and crushing process due to residual battery voltage. Even dangerous conditions such as explosions may occur, causing accidents. To achieve safe dismantling, discharge treatment should be carried out before decommissioning retired power lithium-ion batteries.

There are three current discharge methods. One is chemical discharge, in which the battery is immersed in a salt solution (NaCl , Na_2SO_4 , CuSO_4 , ZnSO_4 , FeSO_4 , MnSO_4) and the residual electricity in the battery is consumed through the electrolysis process. The electrolysis reaction and the reaction between the electrolyte solution and the battery's positive electrode structure cause the battery's structure to be destroyed, the electrode material loses activity, and a micro short circuit or charge transfer occurs inside the battery, which causes the battery to fail. The advantage of the immersion method is that it can completely release the remaining electric energy in the battery; at the same time, the battery will not overheat during the discharge process. The disadvantage is that it takes a long time to soak, and the saltwater is difficult to handle.

Another method is physical discharge, which uses wires, loads, and batteries to form a series of discharges. In addition to wires and loads, materials such as graphite powder and conductive glue are also commonly used as circuit media. The advantage is that the discharge is fast and low cost. The disadvantage is that a large amount of heat will accumulate in a short time, which may cause the battery to explode.



The last method uses the discharge cabinet, which can monitor the remaining voltage of the used batteries. Still, the equipment and operating costs are higher, the production efficiency is lower, and the actual production is less used.

2.3.2 Mechanical Separation

2.3.2.1 Crushing

The rigid structure in the cell mainly includes the outer shell and the positive and negative electrodes. The main constituent metals are iron, aluminum, copper, etc. These metals have good flexibility and toughness, and conventional crushing methods cannot break them, so the crushing machine should be a shear-type toothed roller crusher based on shearing force. This crusher type is usually divided into single shaft, double shaft, and four shaft. The double-shaft toothed roller crusher has the characteristics of large torque and strong material adaptability. It is suitable for the crushing of battery materials. Still, battery crushing usually needs to reduce its particle to a very small size, so the crushing ratio is relatively large. It is difficult for a single double-shaft toothed roller crusher to achieve a crushing ratio. Therefore, a two-stage double-shaft toothed roller crusher is used in series, or a four-shaft crusher is used to ensure sufficient crushing. At the same time, it can ensure the uniform discharge than the demand. In addition, there are also applications of hammer crushers and shredders in lithium battery recycling.

2.3.2.1.1 Double-Shaft Shearing Gear Roller Crushing

The double-shaft shearing gear roller crushing uses the relative rotating cutter between the two rollers to cut and shred to reduce the particle size of the material. Driven by dual motors + dual planetary reducers or hydraulic motors, its power is strong, and its operation is stable. It is often used in various environmental protection fields, such as waste incineration pretreatment, bulky waste treatment, decoration waste treatment, industrial waste treatment, resource regeneration, utilization, etc.

The action process of the crushing tooth on the material is mainly divided into the following three stages:

(1) Sieving Stage

When all the mixed materials are fed into the crusher, the materials with suitable size will be discharged directly through the crushing toothed bar along with the space between the teeth and the side clearance of the teeth, as well as the gap between the toothed roller and the side comb plate. The rotating screening and grading effects of the roller screen are similar. At the same time, the material with a larger particle size between the crushing rollers and comb plates on both sides will be drawn into the crushing cavity between the two crushing rollers for the next crushing step.



(2) Bulk Bite Stage

When encountering larger particle sizes than required materials, the teeth will puncture, shear, tear, etc. to break the materials. In this process, when the materials enter the relative motion of the teeth, they will first be pierced and sheared by the staggered tooth. If the bulk material is not crushed at this time, it will be further torn. The crushed material will be bitten in by the teeth and be crushed in three stages. If the material is still not crushed, the teeth will forcibly slide along the material's surface. The spiral distribution of the teeth will turn the material over, and the next pair of teeth will continue to align the material until the material is crushed to be bitten in.

(3) Deep Crushing Stage

In the second stage of crushing, if the material has been initially crushed and successfully engaged by the toothed rollers, the third stage of deep crushing will be carried out. In this stage, the material is crushed by the shearing and squeezing action of the front edge of the current pair of teeth and the thick edge of the opposite pair of teeth. Until the toothed rollers are disengaged, the cross section of a couple of teeth is changed from large to minimum. In this process, the crusher discharges while crushing, and large-size materials will be destroyed due to the gradual reduction of the contained volume. Forcibly crushing, the crushed material will be squeezed out and leak out from the tooth side gap. When the first pair of teeth start to disengage, the cross-sectional area between the teeth gradually increases from the smallest, and the broken material through the third section will drain and discharge with the separation of the two pairs of teeth.

At this point, the crushing stroke of a pair of teeth is over. Therefore, when the toothed roller runs for a week, how many pairs of teeth are on the toothed roller, how many times will this process be repeated, and the feeding material will be repeatedly screened and carried out. Puncture, shear, tear, bite, shear, and squeeze break the braking effect.

The grading crusher adopts a fixed rack (also known as a splitting rod) to increase the crushing ratio of a single device. The advantage of this structure is that it increases the single-stage crushing ratio without increasing the height of the equipment. The disadvantage is that it is crushed by squeezing, which is serious. The wear of the teeth is fast, the power consumption is significant, and it is not easy to maintain.

2.3.2.1.2 Single-Shaft Shearing Gear Roller Crushing

Single-shaft shearing tooth roller crushing is usually used for the secondary crushing of lithium batteries. The output particle size is 10–120 mm, and the particle size is relatively uniform. Then, the materials can be dried or rinsed to remove the separator and adhesive (polyvinylidene fluoride [PVDF]).

The single-shaft shearing toothed roller crusher is also called a fine crusher and single-shaft shredder. It uses movable or rotatable cutters (from now on referred to as movable knives) and fixed cutters (referred to as fixed knives) to interact to shear the material. The material can be processed to smaller particle sizes by shredding, squeezing, shearing, etc., and it is widely used in the fine crushing of various solid



wastes. It has a small discharge size, replaceable screen, wide material applicability, and high efficiency.

After entering the single-shaft crusher, the hydraulic cylinder's battery material is driven to the cutter shaft by the push plate. The motor's power is conveyed to the reducer by the transmission of the belt. The operation of the reducer makes the cutter shaft rotate, and the material is cut and crushed by the fixed knife and the movable knife. The finished product conforming to the screen size falls through the screen, and the screened material returns rebroken.

The movable knife is fixed on the knife seat of the knife shaft by bolts. The material is shredded when the equipment is running by cutting and crushing the movable knife and the fixed knife. The bolt can adjust the gap between the movable knife and the fixed knife. The shredded material particles are discharged through the screen, and the screen holes determine the output size.

2.3.2.1.3 Four-Shaft Shearing Gear Roller Crushing

The four-shaft shearing gear roller crusher is also called the four-axis shredder, which uses the rotation between the cutters to produce mutual shearing, tearing, and squeezing to process materials. It is used for the broken of various solid wastes. The crushing process can process the material to a smaller particle size at one time. So the crushing process is often used in environmental protection fields such as municipal solid waste (MSW) disposal, resource regeneration, waste incineration pretreatment, and cement kiln collaborative pretreatment. The equipment adopts a low-speed, high-torque design and has the characteristics of large shear force, stable equipment, and uniform discharge.

Four hydraulic presses or motors, respectively, drive the four cutter shafts to rotate forward and backward. The upper row of cutter shafts and the lower row of cutter shafts cooperate to perform the initial breaking of materials and have the functions of material shifting and feeding. The lower row of knife shafts mainly performs the secondary crushing to complete the processes of shearing, squeezing, tearing, etc. The size of the discharged product is mainly determined by the thickness of the blade installed on the knife shaft and the opening size of the screen.

The cutting principle is as follows:

- ① Main cutting method: Formed between the cutting edges of adjacent main cutting tool groups.
- ② Secondary cutting method: Relative cutting between the cutting edges of the adjacent main cutting tool group and the second cutting tool group.

The screen controls the discharge size of the four-shaft crusher. After the material is cut once, the particle size smaller than the screen mesh will be discharged from the mesh. The material size larger than the mesh will pass through the main cutter and auxiliary by the guiding action, return to the crushing box along the inner surface of the screen for secondary cutting, and so on until the material can be discharged from the screen mesh.



2.3.2.1.4 Hammer Crushing

Hammer crushing is to crush materials through impact. There are two types of hammer crushing methods: single rotor and double rotor. This crushing method has relatively large and suitable for the coarse crushing of large materials. The maximum feed size can reach 600–1800 mm, and the output size ≤ 25 mm. It is widely used in cement, chemical, electric power, metallurgy, and other industries. The hammer crusher is suitable for materials with medium hardness. The compressive strength of the crushed materials does not exceed 150 MPa, such as limestone, slag, coke, and other materials.

When the hammer crusher is working, the driving device drives the rotor to rotate at high speed. After the material enters the crusher, the high-speed rotating hammer impacts, shears, and tears the material. At the same time, the material will be punched from the high-speed rotating hammer to the baffle and sieve bar due to gravity. The material larger than the sieve hole remains on the sieve plate and continues to be hit and ground by the hammer until it is crushed to the required particle size and finally discharged through the sieve plate.

The advantages of hammer crushing are large crushing ratio (generally 10–25 high up to 50), high production capacity, uniform products, less over-powder phenomenon, low energy consumption per unit product, simple structure, light equipment, easy operation and maintenance, etc. It can crush raw materials of different sizes into uniform particles to facilitate processing in the next process, with reliable mechanical structure, high production efficiency, and good applicability.

The disadvantages of hammer crushing are that the hammerhead and the grate bar screen wear quickly and the maintenance and balancing time are long. When crushing hard materials, the wear is faster, and the consumption of metal materials is much more; it is easy to block the grate bar screen gap when crushing sticky materials. For this reason, it is easy to cause downtime (the water content of the material should not exceed 10%), resulting in a decrease in production capacity.

2.3.2.1.5 Smash

Smash is to pulverize solid raw materials of smaller size to materials of required good size. Crushing is usually composed of coarse crushing, fine crushing, wind conveying, and other devices, with a high-speed impact on smash materials, mainly used in mining, building, and other industries. In the battery recycling industry, crushing is mainly used in the fine crushing process after coarse crushing. With the different elasticity and flexibility of the current collector and electrode powder, it is crushed into different particles by impact force, rolling pressure, and shear force to obtain different grade products to be separated by sieving.

Smashers mainly include ball mills, rod mills, and stirring mills in the production process. The ball mill and the rod mill are composed of a horizontal cylinder, a hollow shaft for feeding and discharging materials, and grinding media. The cylinder is a long cylinder with grinding media inside. The cylinder is usually made of steel plates. The grinding media is generally steel balls or rods. The balls are usually of different diameters and are packed in a particular proportion to meet a specific gradation.



In contrast, the rods are packed with a certain number of steel or iron rods of different lengths and thicknesses. When the cylinder starts to rotate, the grinding medium is attached to the cylinder and taken away by the cylinder due to inertia, centrifugal force, and friction. When it comes to a certain height, the falling grinding medium crushes the material in the cylinder like a projectile due to gravity.

When the stirring mill is working, the grinding medium is driven by the stirring rotor to make a rotary movement, including revolution and rotation. After the material enters the stirring barrel, it collides, squeezes, and makes friction with the grinding medium in the stirring. Shearing force and squeezing force exist everywhere in the whole barrel. This phenomenon is observed because, along the radial direction, the moving speed of the material and the medium is different. Along the axial direction, the moving rate of the material and the medium between the layers is not equal, so the speed gradient exists. The crushing efficiency of materials near the stirring rotor is high. In addition to circular motion, the media and materials also have different degrees of up and down movement. Some grinding balls collide and friction with the agitator, and there is still a specific impact near the agitator.

2.3.2.2 Sieving

Sieving is a method of separating the particle group according to the different powder properties of each particle. Usually, sieving is carried out according to the difference in particle size. The essence is the process of grading materials according to the particle size. It is often coordinated with smashing to make the particle size of the smashed material more uniform to ensure that it meets specific requirements or avoids the phenomenon of over smashing. In the field of battery sieving, the crushed or smashed materials need to be classified with the aid of a screening device before they can enter the next sieving operation. Commonly used screening equipment mainly includes vibrating screens and relaxation screens.

2.3.2.2.1 Vibration Sieving

Vibrating sieving is when granular materials are passed through one or several layers of screens to divide them into several granularity levels according to their size. It is widely used in many industrial sectors. For the particles to have a good chance of penetrating through the sieve, the maximum size of the particles should be smaller than the mesh size. However, when the particle size is too small, the following situations may occur:

- (1) Walk along the screen surface at a breakneck speed.
- (2) Collide on the screen surface when falling.
- (3) Extrude between particles and form an arch on the screen.

It prevents the particles from penetrating through the mesh, making it difficult to penetrate the sieve even when the sieve hole is six times larger than the particles.

The vibrating screen has a rectangular screen box with a flat-screen surface. The screen box is supported (or hung) on the frame with elastic elements and is excited by a vibration exciter. Therefore, it vibrates elastically, and the amplitude is affected by feeding and other dynamic factors to be changed. Vibrating screens mostly use



high-frequency and low-amplitude methods to make the material jump on the screen surface, and the processing capacity and screening efficiency are high.

The following formula can calculate the screening efficiency:

$$F = \frac{a-b}{a(1-b)}$$

In the formula, a is the content of the under-sieve in the material, %, and b is the content of under-sieve in above-sieve material, %.

2.3.2.2.2 Relaxation Sieving

The screen of the relaxation sieving is made of flexible polyurethane rubber material. When it is working, the screen is alternately tensioned and loosened to make the material bounce, preventing the material from adhering to the screen and blocking the screen holes. At the same time, due to the flexible sieve plate, the projecting acceleration of the material reaches 30–50 times the acceleration of gravity. Therefore, the screening efficiency is improved, while clogging the sieve holes is avoided, and the processing capacity is increased. It effectively solves the plugging of sticky and wet materials in practical applications and simplifies the process flow. The commonly used relaxation sieving in China is mainly mechanical and vibrating.

The mechanical relaxation sieving is also called the crank connecting rod relaxation sieving. The Lowell relaxation sieving mainly represents it, composed of two screen frames, I and II. Screen frame II is placed in screen frame I. II is supported on the frame with vibration isolation springs, and there is a hanging device between them. The screen frame I is hung on the screen frame II in parallel. At the same time, the hanging device can be used as a guide to making the screen frames I and II work relative parallel movement. Each screen frame is composed of two side plates connected by cross beams, and the two ends of the polyurethane screen plate are, respectively, fixed to the cross beams of screen frames I and II. The drive of the screen frame is that the motor drives the eccentric shaft to rotate through the belt pulley. The eccentric shaft will continuously push and pull the screen frame I through the connecting rod, causing screen frame I and screen frame II to move toward and away from each other alternately because the masses of the two screen frames are similar. When screen frame II pushes and pulls screen frame I, the latter also pushes and pulls screen frame II. Assuming that the absolute amplitude of each screen frame is A , then the relative amplitude of the two screen frames is $\pm A$. The beams installed on the screen frame drive the flexible screen plates to be alternately tensioned and loosened. In this way, the material on the screen surface will undergo a “bounce–fall–bounce” cycle process. The working frequency of the mechanical relaxation screen is 500–600 r/min.

The vibrating relaxation sieving is developed based on the traditional circular or linear vibrating screen. The single drive produces double vibration. Two vibrations are provided by one exciter or group of exciters. The rotation of the eccentric block makes the active screen frame produce circular or linear vibration as the basic vibration; the additional vibration is done by shearing spring to make the generated elliptical vibration of the floating screen frame. When the main floating screen frame



produces relative movement, the polyurethane screen panels installed on the fixed and floating beams at both ends continue to expand and contract. The flexible screen panels produce large deflection deformation, thereby causing the material particles on the screen a very high acceleration, up to 50g or more. The sieve holes are constantly stretched and deformed, which can effectively prevent the sieve holes from clogging and improve the efficiency of material screening. The working frequency of the flaccid vibrating sieve is mainly about 800 r/min.

2.3.2.3 Airflow Sieving

Airflow sieving refers to the technology that uses air as the sieving medium to separate particles according to density or size under airflow action. The basic principle of airflow sieving is that the airflow will carry the “lighter” particles upward or horizontally to a farther place within a certain range of wind speed. In contrast, the “heavier” particles are settled because the updraft cannot offset the influence of gravity, or because the particles have enough inertia, their main movement direction cannot be changed by the horizontal airflow and settle through the airflow. The light materials carried away by the airflow enter the cyclone separator for gas–solid separation and dust removal and are then discharged.

Specific to the disassembly of lithium batteries, the winnowing machine is mainly used to remove the light separator or heavy shell in the battery and to separate the current collector in the state of the large pole piece. Airflow sieving can be divided into constant airflow sieving and pulsating airflow sieving according to the air movement mode of the sieving section, and pulsating airflow sieving can be divided into aortic and arterial airflow sieving.

2.3.2.3.1 Constant Airflow Sieving

Constant airflow sieving can be divided into two types: vertical and horizontal. It mainly separates the materials by passing airflow into the sieving machine. It is mainly used in areas where the density difference is relatively large, and the sieving accuracy is not high. Its advantages are large processing capacity, simple structure, low energy consumption, low pollution, and no need for any media.

Vertical constant airflow sieving is also known as traditional airflow sieving. It can also be considered as the first generation of airflow sieving. A blower is used to pass a constant airflow into the straight cylinder sieving column to realize the separation of materials. But it is only suitable for materials with large differences in density, slight differences in particle size and shape, or large differences in particle size, and slight differences in thickness and shape. For materials with large differences in shape or with a certain difference in particle size and density, its sieving accuracy is difficult to guarantee, especially when the thickness of solid particles is much greater than the difference in air density. Therefore, as time goes by, vertical constant airflow sieving is gradually fading out of the stage of history.

Horizontal constant airflow sieving is mainly used in common equipment for household garbage mechanical sieving. By adjusting the angle of the fan and the air inlet speed, the quantitatively fed materials are blown away by the airflow sent



during the descending process, and various components fall into different collection tanks according to different motion trajectories to achieve the purpose of sieving.

The material enters the horizontal air sieving machine for free fall motion. At this time, under the action of the horizontal airflow, the light material will move a long distance in the horizontal direction. It will be discharged from the light product discharge port. In contrast, the heavy product will fall into the collection tank through the horizontal airflow due to its inertia to separate light and heavy products. The advantages of horizontal air sieving are large processing capacity, simple structure, large sieving particle size, and multicomponent sieving (more than two kinds), but the sieving accuracy is not high. It is only suitable for materials sieving with very different densities. Therefore, it is widely used in the pre-sieving of waste sieving.

2.3.2.3.2 Aortic Flow Sieving

The aortic flow sieving is a device that strengthens the separation of the materials to be sorted according to the density by passing a periodically changing airflow to the device. The low-density particles in the sieving area can obtain a more significant acceleration effect than the high-density particles. To a certain extent, it overcomes the problem of the isotherm phenomenon of particles in the air movement in the traditional air sieving, resulting in poor sieving efficiency, and the material is sorted by density to the greatest extent.

The pulsating wind generated by the aortic airflow sieving can be divided into two types: full pulsation and partial pulsation. Full pulsation means that all the air flows during the airflow sieving process is pulsating airflow, which only needs an active fan and a pulsation valve. That is, the specific calculation formula is as follows:

$$v = v_0 - v_0 \sin(\omega t + \varphi)$$

where

v is the pulsating wind speed, m/s

v_0 is the wind speed of the active fan, m/s

v_1 is the pulsating fan wind speed, m/s, usually 5–20% of the wind speed of the active fan

ω is the angular frequency, s^{-1} with a period of $2\pi/\omega$

φ is the initial phase

Full pulsating airflow sieving is more suitable for material sieving with a small difference in particle density. The air velocity changes more, and the acceleration effect generated by different particles is also greater. It is easier to achieve separation through a certain period of periodic oscillation. Still, the large air velocity changes will cause light materials to stay longer in the separation column and reduce the separation efficiency. Some pulsating air separation is more suitable for separating materials with large particle densities because a small airflow fluctuation will separate the material with relatively different densities. At the same time, because the active wind machine provides a relatively large constant airflow, light materials can be discharged quickly, thereby improving the separation efficiency.



The pulsating frequency of the pulsating airflow has different acceleration effects on materials of different densities and directly affects the final separation effect of winnowing. The best pulsating frequency needs to be based on the different components of the material, and the corresponding theoretical analysis and experimental verification are obtained. Pulsating frequency is suitable for lithium battery recycling.

2.3.2.3.3 Passive Pulsating Airflow Sieving Technology

Like the aortic airflow sieving technology, the passive pulsating airflow sieving technology is also used to increase the difference in acceleration effects of different particles through the airflow pulsation, thereby strengthening the density sieving of materials and improving the sieving accuracy. The difference is that the passive pulsating airflow sieving depends on the structure of the device itself to approximate the pulsating airflow. The actual airflow is a constant flow. When the diameter of the sieving machine changes or the irregular structure increases, the airflow flow rate will be changed. Change occurs, resulting in a pulsating effect. Passive pulsating airflow sieving can use the form of full induced air to generate a constant airflow. Compared with the blast, the advantage of induced air is that it can better distribute the airflow evenly in the sieving machine.

There are three main types of separation by passive pulsating flow: turning type (Z-shaped), damping type, and variable diameter type.

The turning pulsating airflow sieving changes the sieving section into a Z-shaped structure. When the airflow passes by, it will change the original laminar flow state and generate turbulence, improving the airflow sieving effect. On the other hand, due to the addition of the turning structure, the speed between the two plates of the turning structure will show the characteristics of high center and low sidewalls, which makes low-density materials tend to move to the center. In contrast, high-density materials tend to move toward the edges. The lower the gas velocity near the sidewall, the more conducive to the downward discharge of heavy products. This phenomenon can overcome the influence of shape and particle size on the material sieving process to a certain extent so that materials can be better classified by density, thereby improving the separation efficiency of airflow separation.

The main factors affecting the separation efficiency of the turning airflow sieving are the angle of the turning plate, the distance between the plates, and the number of turns. Compared with the vertical direction, the larger the angle of the turning plate is, the more favorable it is for the heavy product to fall, and the few mismatches in the light product. However, an excessive turning angle will make it difficult to discharge the light product. The smaller the distance between the plates, the faster the center to sidewall gas velocity changes, the more conducive to separating light, and heavy products will reduce the processing capacity and be prone to clogging. The more the number of turns, the longer the effective separation time, and the higher separation accuracy will increase separation. The height of the selection machine reduces the processing efficiency. Therefore, only by adjusting the angle of the turning plate, the distance between the plates, and the number of turns can increase and maximize the efficiency of turning airflow sieving.



Damped pulsating airflow sieving is to add a damping block to the sieving section of the traditional air sieving machine so that the airflow will have an acceleration or deceleration effect when passing through the sieving section, thereby generating a pulsating airflow and realizing the separation of strengthened materials according to the density. The variable diameter pulsating air sieving machine directly transforms the straight tube structure of the sieving section into a variable diameter structure. The variable diameter structure is mostly a tapered section and a tapered section-generating pulsating airflow and pulsating damping airflow. The sieving machine is similar in that the flow rate of the passing airflow is changed by changing the diameter of the sieving section to realize the generation of pulsating airflow.

2.3.2.4 Flotation Separation

As an essential method for separating fine-particle minerals, flotation separation technology also plays an important role in recycling and utilizing spent batteries. After the spent lithium battery materials are disassembled and crushed, the large-particle pole pieces are separated by winnowing and reselection to realize the separation of the positive and negative electrode materials and the foil. The fine-particle products with a particle size of 10–50 μm produced by the crushing mainly contain positive electrode active material and negative electrode graphite powder. This part of waste also has a certain recycling value. Based on the density difference between the positive electrode active material and graphite, air separation and reselection are often used for separation. Still, the particle size of this part of the mixture is relatively fine, and the density difference can no longer help its separation. The positive electrode active material and graphite have different crystal types. Their hydrophilic and hydrophobic properties are different, resulting in a large difference in the wettability of their surface energy. The flotation separation technology is based on the difference in hydrophilicity and hydrophobicity caused by the surface properties of the mixture to achieve the separation of the two. As far as battery materials are concerned, the most positive electrode active materials are hydrophilic materials. At the same time, graphite has strong hydrophobicity, so the flotation separation of fine particle positive electrode active materials and graphite mixed materials has almost become an inevitable choice. The common flotation process is shown in Figure 2.5. The mixed electrode material generally enters the flotation system after surface modification. The flotation process can be increased or reduced according to product quality requirements.

Since the positive electrode active material and graphite have opposite surface hydrophobic properties, flotation separation is ideal for recovering the positive electrode material and graphite separately. However, because the organic binder covers the surface of the positive electrode material and graphite, surface properties are reduced. It is necessary to reasonably modify the electrode material's surface to achieve flotation separation. Because of this key technical difficulty, there are currently three main treatment methods: high-temperature roasting, mechanical grinding, and organic oxidation dissolution. The He Yaquin team conducted laboratory research on these three methods. In terms of high-temperature pyrolysis [4], they believe that the organic binder and residual electrolyte on the surface of the



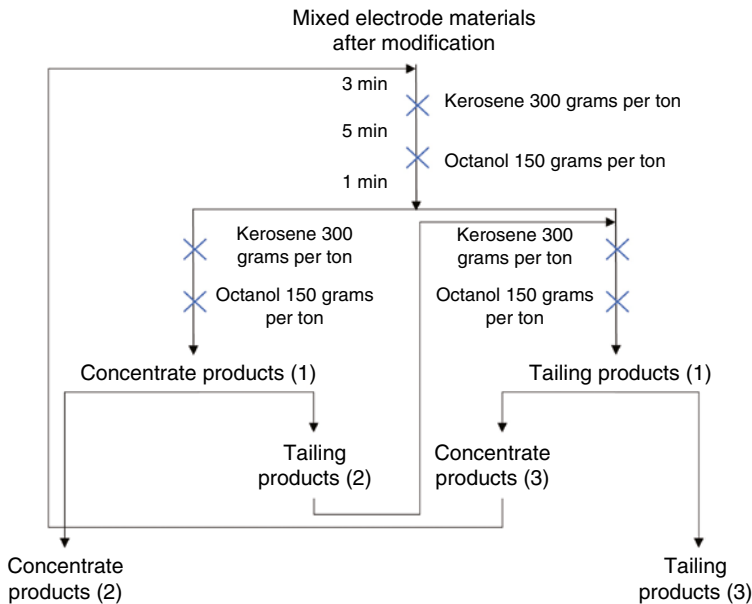


Figure 2.5 Schematic diagram of the flotation process. *Source:* Zhang [4].

electrode material can be removed by pyrolysis. So high-temperature pyrolysis can realize the mutual dissociation of the positive and negative material particles, thereby making the initial hydrophilic and hydrophobic properties of the surface of the exposed and negative materials. The organic binder on the graphite surface of the negative electrode is reduced. The hydrophobicity and the adsorption capacity of the collector are enhanced; the surface of the positive electrode material increases the affinity with water due to the increase of the hydrophilic factor. The pyrolysis treatment can strengthen the difference between the hydrophilic and hydrophobic properties of the surface of the positive and negative electrode material particles. The flotation separation efficiency of the two electrode materials is improved. At present, the best pyrolysis temperature they have obtained is 550 °C. After pyrolysis followed by a combination of the two-stage flotation process, the grade of the cathode material can reach 98.00%. In terms of mechanical grinding [5], they believe that the positive and negative plates of LIBs are glued on the current collector material in a laminated structure with electrode material particles, conductive agents, and other additives through an organic binder. The binder on the surface of the battery material becomes brittle at a low temperature, so the low-temperature grinding method can make the binder be abraded and peeled off, thereby exposing the hydrophilic, hydrophobic surface of the electrode material particles. At the same time, the layered structure of graphite is damaged by low-temperature grinding.

When it is destroyed, more lamellar new surfaces are produced, the difference in properties between the positive electrode material and the graphite surface is more and more exposed, and the efficiency of flotation separation is also improved. In terms of organic oxidative dissolution [6, 7], they believe that the Fenton reagent



can effectively oxidatively degrade the organic carbonate in the passivation film. At the same time, the polymer PVDF will be decomposed into small molecules such as $\text{—CF}_2\text{CF}_2\text{—}$ and $\text{—CF}_2\text{CH}_3$. The bonding effect subsequently fails, the passivation film is disintegrated, and the crushed cathode product's surface is activated and modified. Fenton oxidation is relatively milder. The degradation of the passivation film on the particle surface can only affect the particle's outer surface, and the part that is blocked by the particle cannot react deeply. Therefore, the phenomenon of particle agglomeration will still occur after modification. However, this method can prevent the overoxidation of graphite and maintain the original crystal form of graphite, thereby improving the efficiency of flotation separation.

As far as industrial applications are concerned, none of these three methods have been substantively industrialized, but they provide a theoretical basis and technical guidance for the subsequent flotation process. At present, the flotation process is mostly used in the coal preparation industry. It has not been promoted and applied in the battery recycling industry, but this is still a key technology that can be considered in the pretreatment process of battery recycling. The key point of flotation technology lies in the separation efficiency of positive and negative black mass and product grade. By adjusting flotation reagents, flotation process optimization, flotation equipment selection, high-efficiency separation, and high-grade product output are achieved, which reduces the difficulty of subsequent leaching processes.

2.3.3 Heat Treatment

In the LIB recycling process, pyrolysis equipment is mainly used to decompose and remove organic matter. It is usually used to remove organic binders in electrode materials and the pyrolysis of electrolytes and separators.

2.3.3.1 Rotary Kiln

A rotary kiln refers to a rotary calcining kiln, divided into cement kiln, metallurgical chemical kiln, and lime kiln according to different materials. Because the rotary kiln can be pyrolyzed in an inert gas atmosphere in a closed state, the lithium battery recycling process is mainly used to remove the organic binder of the electrode sheet and the pyrolysis of the electrolyte. The removal temperature of the organic binder is 450–600 °C, the time is one to two hours, and the pyrolysis temperature of the electrolyte is 180 °C, so the organic binder and electrolyte can be simultaneously pyrolyzed and removed in a negative pressure inert gas environment.

A rotary kiln is composed of gas flow, fuel combustion, heat transfer, and material movement. The rotary kiln is how to make the fuel fully burned, and the heat of fuel combustion can be effectively transferred to the material. After the material receives the heat, a series of physical and chemical changes occur, and, finally, the product is formed.

2.3.3.2 Microwave Steel Belt Kiln

The microwave steel belt kiln is mainly used to roast large nonmetallic materials in an air atmosphere. The equipment has the characteristics of high energy utilization,



a short firing cycle, high production efficiency, large daily output, uniform product heating, good product quality, high degree of automation, safety and reliability, a small footprint, and no environmental pollution. It is suitable for air sintering, synthesis, decomposition, and discharging nonmetallic materials under atmosphere and oxygen-rich conditions. A microwave steel belt kiln can be used to remove the separator.

2.3.3.3 Microwave Roller Kiln

A microwave roller kiln is a kind of continuous firing kiln by the rotation of sticks. The pyrolysis tunnel kiln is a continuous firing kiln. Its heating method is microwave heating, called a microwave roller kiln. It is mainly used for the carbothermal reduction of metal oxide ore, such as drying, pyrolysis, roasting, calcination, and sintering of minerals, and the desulfurization of metal sulfide ore. In the field of lithium batteries, it can be used for drying and synthesizing battery cathode materials. It can also remove binders for lithium battery electrode materials in the recycling process.

During the working process, the material can be placed directly on the roller, or the material can be placed on the roller through the hot plate. The continuous rotation of the roller can make the material advance in order. There is a sprocket at the end of each roller, and the chain can drive it to rotate. When running, the chain is divided into several groups to drive once to ensure smooth and safe transmission during the working process. The low-temperature rollers are made of heat-resistant nickel-chromium alloy steel, and the high-temperature-resistant high-alumina ceramics such as corundum porcelain are used as the rollers. The microwave roller kiln's upper and lower heating chambers adopt microwave heating to heat, dry, pyrolyze, roast, and sinter the indoor materials. Between the microwave inlet and the roller table, there is a refractory material to isolate, and it does not directly contact the product being fired.

The microwave roller kiln adopts a high-stability, long-life, continuous industrial-grade microwave source to ensure continuous and stable long-term operation of the equipment, with huge production capacity. The microwave power and kiln length can be set on-demand, the cross section of the kiln cavity is large, and the material is large. The same applies; the microwave power distribution is scientific, and the temperature control is accurate and suitable for various atmospheres such as air, oxygen, nitrogen, weak reduction, etc. It can be switched at any time, accurately controlled, the temperature field is uniform and controllable, the process controllability is more energy-saving, and the degree of automation is high.

2.3.3.4 Carbonization Machine

The carbonization machine consists of four systems: gasification system, flue gas purification system, carbonization system, and cooling system. All internal parts are sturdy and durable. Special high-temperature-resistant precious metal steel is not deformed, is nonoxidized, has good insulation performance, and is simple to operate, safe, and reliable. Because it is carried out in a closed environment, it can be applied to the removal of organic binders in the field of lithium battery recycling.



The carbonization machine first burns the material through the gasification system to produce flue gas. After filtering by the flue gas purification system, the flue gas is transmitted into the carbonization system for combustion. When a certain temperature is reached, the carbonized material is added and transmitted through the pipeline to get the material burned in the carbonization machine. The organic matter combustion needs to meet three points: heat, oxygen, and organic matter. The oxygen demand cannot be met because the carbonization machine is almost closed. The material will pass through the carbonization machine at 800° inside the carbonization machine at a high temperature. The adjustment of the speed of the internal conveying device will not burn into ash, but only burn into charcoal. In the lithium battery recovery process, the carbonization temperature can be adjusted to remove the binder under the premise of ensuring that the electrode powder is not carbonized. The flue gas purification process processes the flue gas generated by the material burned in the carbonization machine. After that, it returns to the carbonization machine for combustion so that the thermal energy of the machine runs continuously, achieving a smokeless, environmentally friendly, and continuous effect. It can be seen that the organic waste gas generated in this process can be reused on the carbonization machine. The final cooling system can quickly drop the temperature of the discharged materials to 50–80°C, which greatly improves the processing efficiency.

2.3.4 Solvent Dissolution

The main high-value resources for LIB recycling and utilization mainly concentrated on the cathode material. The existing methods for separating various cathode materials and current collectors include crushing, acid–base leaching, and high-temperature calcination. There are disadvantages such as high energy and material consumption, incomplete separation of cathode material and current collector, inability to effectively recover aluminum elements, and inability to realize the recycling of production raw materials. The positive electrode material is fixed on the aluminum foil by the binder in the positive electrode sheet. The existing treatment process transfers the positive electrode material elements, such as nickel, cobalt, manganese, lithium, and other elements, into the solution through acid–base leaching or calcination leaching methods using the extraction method to extract these elements. In this process, aluminum easily enters the leaching system along with the positive element, which is difficult to recover and increases the difficulty of the extraction and recovery process of the positive element. If the aluminum foil can be separated from the active material without changing the shape, not only the active material can be completely separated, but the aluminum sheet can be directly recycled. Therefore, as long as the bonding effect of the binder is destroyed, the overall separation of the active material powder and the aluminum foil can be achieved. The key to the problem lies in removing organic binders, and the removal method can also be achieved by organic dissolution and heating, and volatilization.

Solvent dissolution [8] is a method to remove organic binders based on the principle of “similar phase dissolution.” For two different molecules, solute and solvent,



Table 2.2 The interaction between solute and solvent molecules.

Solute A	Solute B	Interaction			Solubility
		A–A	B–B	A–B	
Polarity	Polarity	Strong	Strong	Strong	Maybe higher
Polarity	Nonpolarity	Strong	Weak	Weak	Maybe lower
Nonpolarity	Polarity	Weak	Strong	Weak	Maybe lower
Nonpolarity	Nonpolarity	Weak	Weak	Weak	Maybe higher

when the attraction between solute and solvent molecule is stronger than solute or intermolecular attraction within solvent molecule itself, solute will dissolve in solvent, thus forming a solution. The attraction between solute and solvent molecules is related to their polarity. As shown in Table 2.2, the interaction between molecules with different polarities varies greatly. Therefore, it is necessary to select an appropriate solvent for the organic binder in the battery according to its characteristics to achieve effective removal.

At present, the commercially available lithium battery binder materials are mainly divided into water based and oil based. The water-based binders mainly include sodium carboxymethyl cellulose (CMC), styrene butadiene rubber (SBR), LA132, etc. The oil-based binders are mainly PVDF resin, vinylidene fluoride (VDF) homopolymer, a copolymer of VDF, and a small amount of other fluorine-containing vinyl monomers. Its repeating unit is $\text{—CH}_2\text{—CF}_2\text{—}$. For water-based organic binders, the polarity is relatively weak. Choose a solvent similar to water, such as some organic acids, to effectively separate the active material from the current collector. A suitable solvent should be a polar organic solvent for PVDF with strong polarity. Common PVDF organic solvents include *N*-methyl pyrrolidone (NMP), dimethylacetamide (DMAC), dimethylformamide (DMF), acetone, etc. These molecular structures all contain at least one carbonyl group. There is a strong polarization effect, which allows these organic solvents to have strong polarity, and they have similar compatibility with PVDF. However, the viscosity of the organic solvent is relatively large, and the active particles obtained after dissolution are relatively fine, which causes the problem that the solid and liquid are difficult to completely separate. Incomplete solid–liquid separation makes the subsequent recovery of organic solvents more difficult. At the same time, the cost of organic solvents is relatively high, and the amount used is large. The establishment of an industrial recycling system requires a large investment of capital. Some organic solvents have potential harm to the ecological environment and workers' health (Table 2.3).

2.3.5 Alkaline Dissolution

The cathode material of LIBs is coated on aluminum foil. Aluminum is a kind of amphoteric metal. Using the principle that aluminum can react with an alkaline



Table 2.3 Laboratory parameters of electrode materials dissolved in common organic solvents.

Solvent	Test conditions	Advantages and disadvantages of testing
NMP	Cut into small pole piece electrode material, 900 °C, 5 min LIB, 100 °C, 1 h Production waste in LIBs, 80 °C, 15 min	1) Subsequent acid separation 2) Need NaOH to remove the aluminum foil 3) General dissolving effect
DMAC	Lithium-ion button battery, 120 °C, 12 h	Longer time
DMF	Fragment of LCO electrode, 60 °C, 1 h	Slower dissolution rate
Dimethyl sulfoxide (DMSO)	Dissolve PVDF, stir at 60 °C, 30 min	Solvent is toxic
Tetrahydrofuran (THF)	Dissolve PVDF, stir at 60 °C, 30 min	Solvent is toxic

Source: Adapted from Li et al. [8].

solution, in contrast, lithium cobaltate does not react with an alkaline solution; the material obtained by disassembled batteries can be immersed in the alkaline solution. The aluminum foil will dissolve sodium metal aluminate and enter the solution. Insoluble substances, including cathode materials, binders, acetylene black, etc., enter the slag to achieve the effect of separation. This method is easy to operate, simple, and can realize industrialized large-scale production. Figure 2.6 shows that after the disassembled battery positive electrode is broken [5, 6], the aluminum foil is removed by the NaOH soaking method. The aluminum is dissolved in the lye in the form of NaAlO_2 . The follow-up processing of the remaining residue is carried out to realize the separation of aluminum from cobalt and lithium. Finally, the pH value of the alkaline leaching solution is adjusted with the sulfuric acid solution, and the aluminum element is precipitated and recovered in the form of $\text{Al}(\text{OH})_3$. However, due to the positive active material coated on the pole piece, the alkaline solution cannot effectively contact the aluminum foil of the base, which will inevitably affect the reaction between the alkaline solution and aluminum. Therefore, the pole piece needs to be crushed to facilitate the dissolution and separation of aluminum.

2.3.6 Ultrasound Strengthens the Separation

With the rapid development of science and technology, the application of ultrasonic technology has become more and more extensive. Since the twentieth century, the fields of metallurgical engineering, materials engineering, bioengineering, etc. have begun to use ultrasonic technology to strengthen separation and leaching widely.



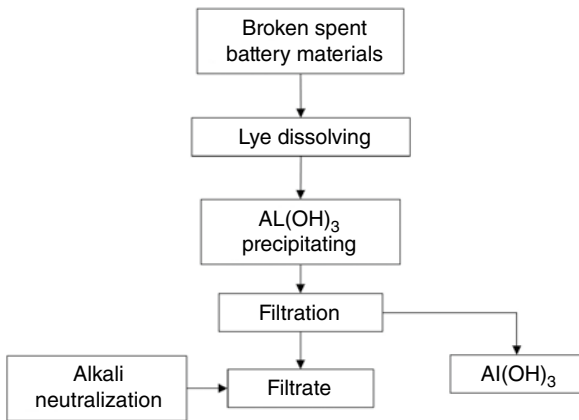


Figure 2.6 Schematic diagram of the alkali dissolution process. *Source:* Adapted from Jha et al. [9].

The method is mainly divided into the ultrasonic layered enhancement of active material and current collector separation and ultrasonic enhanced leaching in battery recycling.

Ultrasonic stratification can provide a rapid and sustainable method of separating two substances. This technology is not only significantly more efficient and environmentally friendly than hydrometallurgy and pyrometallurgy, but it can also produce higher purity materials. Some scholars [10] proposed a new recycling mode to use high-intensity ultrasound to recycle LIBs. Figure 2.7 shows that cavitation bubbles filled with steam are randomly formed at a certain ultrasonic frequency and power. After a lot of vibration and expansion, they burst on the surface of the active ingredient. The bursting force of the cavitation bubbles is greater than that of the adhesive. The force is stronger so that the active material and the current collector are completely layered. This method does not destroy the structure of the active material, and it is expected that the recovered active material will be directly sent back to the battery production line. Ultrasonic stratification will be a breakthrough in battery recycling technology. However, the effectiveness of ultrasonic delamination is limited by the type and molecular weight of the binder. At the same time, due

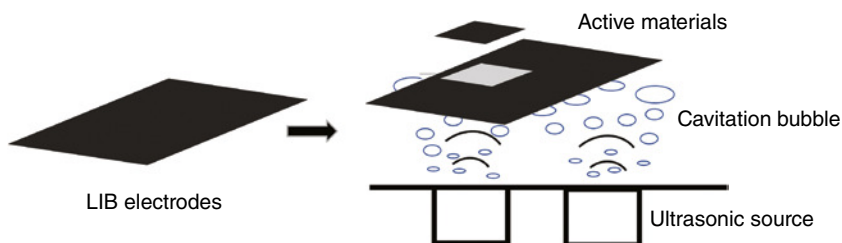


Figure 2.7 Schematic diagram of ultrasonic layering.



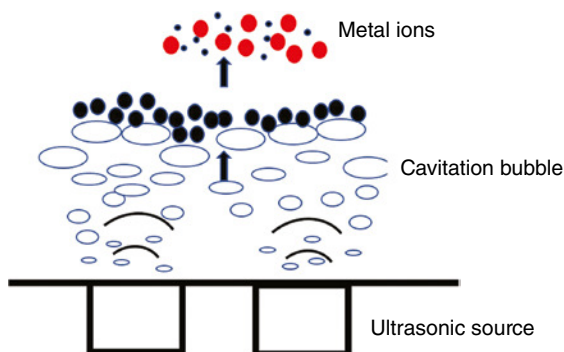


Figure 2.8 Schematic diagram of ultrasonic enhanced leaching.

to the presence of black carbon additives, the finer particles cannot be filtered and recovered. The corresponding wastewater treatment is also a potential problem.

Ultrasonic enhanced leaching [11, 12] is mainly through the cavitation effect of ultrasonic waves in the liquid. When ultrasonic waves irradiate the leaching surface, a large number of micro gas core cavitation bubbles are generated, along with a series of dynamic processes such as bubbling vibration, growth, shrinkage, rupture, etc., to enhance the mass transfer process, as shown in Figure 2.8. Ultrasound mainly promotes the convection movement to increase the solid–liquid contact area, thereby accelerating the leaching efficiency, and also provides a large amount of energy, which is conducive to the dissolution of waste. In the negative pressure stage, millions of tiny vacuum holes are formed in the liquid medium, and the gas dissolved in the solution enters these holes and generates a large number of bubbles. This process is called “cavitation.” In the positive pressure stage, cavitation bubbles are crushed during adiabatic compression, and huge energy is released when the bubbles burst. Therefore, the broken bubbles generate a very high pressure (more than tens of thousands of pounds per square inch) at the solid–liquid interface around the bubbles, which helps to improve the leaching efficiency. The cavitation of ultrasound greatly accelerates the dissociation of acid and the chelation process with metal elements.

2.3.7 Mechanical Chemistry Strengthens the Separation

Mechanochemistry is an interdisciplinary subject that has emerged in high-tech fields such as environmental protection, metallurgy, and material manufacturing. The mechanical force acts on the condensed matter so that part of the grinding material is broken and refined. This part of mechanical force is converted into mechanical energy with intuitive changes. At the same time, the other part is stored inside the particle system. This part of the energy will cause the distortion and plasma state of its surface structure, surface properties, composition, and lattice structure. The process of using mechanical forces such as friction, collision, impact, and shearing to cause materials to break, deform, and collapse, resulting in an increase in the internal energy of the material and an increase in reaction activity, is



called mechanical activation. The reaction material is subjected to strong mechanical force. The crystal grain size decreases, the specific area increases, the surface continuously collapses, the surface bond can change, and the structure tends to an amorphous state. As the time of mechanical force increases, the amorphous layer on the surface of the material crystal becomes thicker, and the crystal lattice produces dislocations, distortions, etc. The crystal structure becomes amorphous and crystalline changes. As the crystal structure changes significantly under the action of mechanical force, its physical and chemical properties also change accordingly. These changes include reduced density, increased solubility, lower melting point, improved electrical conductivity, increased surface energy, and increased surface adsorption and reactivity. Many factors affect mechanochemical reactions, which have different characteristics from conventional chemistry. For example, it can induce some chemical reactions that are difficult to achieve conventionally, and it can change the thermodynamic properties of the reactants so that certain mechanochemical reactions can follow. Under normal conditions, thermodynamics is impossible.

In the application of waste battery recycling, the substance's chemical properties can be activated through mechanical force, and the reaction can be carried out at a lower temperature, such as room temperature, without other harsh reaction conditions. Fane et al. [13] used the method of immersing used batteries in NaCl solution to achieve complete battery discharge. The recovered LiFePO_4 was subjected to a high temperature of 700°C for five hours to remove organic impurities. Oxalic acid is used as a grinding aid, mixed with recycled materials, and mechanically activated by a planetary ball mill. The mechanical activation process mainly includes three steps: particle size reduction, chemical bond breaking, and new chemical bond generation, as shown in Figure 2.9. After the grinding, mechanical activation, the mixed raw materials, and zirconia beads were rinsed with deionized water and soaked for 30 minutes. The filtrate was stirred and evaporated at 90°C until the concentration of Li^+ was greater than 5 g/l. Then the pH of the filtrate was adjusted with 1 mol/l NaOH solution to 4. The filtrate was continued to stir for more than two hours until the concentration of Fe^{2+} is less than 4 mg/l, then high-purity filtrate was obtained. After filtration, the purified lithium solution was adjusted to pH 8 and stirred at 90°C for two hours. The precipitate was collected and dried at 60°C for 24 hours to obtain an Li recovery product. The recovery rate of Li can reach 99%, and Fe is recovered in the form of $\text{FeC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$, and the recovery rate reaches 94%.

As shown in Figure 2.10, the common processes of mechanical activation in battery recycling for waste batteries included salt solution discharge, dismantling and separation, alkali dissolution, filtration and drying, mechanical activation, calcination, and then entering the physical separation system or entering the wet process of leaching system for further extract high-value metal elements. Mechanical activation technology has its unique advantages in battery recycling. It can promote the separation of battery materials and the recovery of elements by changing the surface properties of materials through simple physical and mechanical forces. The equipment is simple, the process flow is short, and there is no obvious environmental pollution. The method is expected to achieve widespread application in the battery recycling industry.



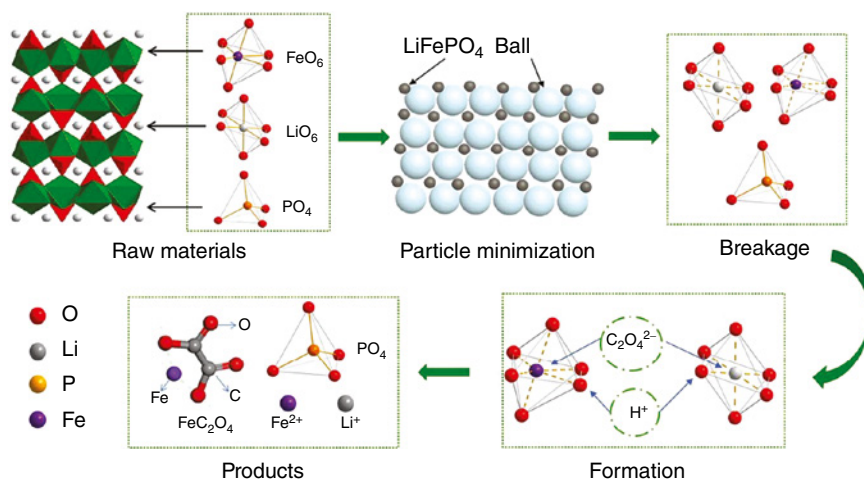


Figure 2.9 Schematic diagram of the mechanism of mechanical activation process.
Source: Fane et al. [13] / with permission of American Chemical Society.

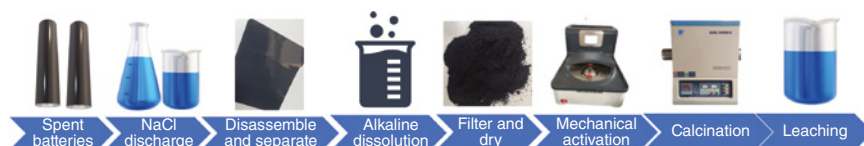


Figure 2.10 Schematic diagram of the mechanical activation process.

2.4 Hydrometallurgy

2.4.1 Metal Leaching

Leaching is the first step in the entire hydrometallurgical recovery of valuable metals from spent battery cathode materials. Leaching is converting the metal in the positive electrode active material into the metal salt form in the solution, which can then be separated and recovered by various chemical methods such as precipitation, extraction, and electrolysis. The commonly used leaching agent is an inorganic acid, organic acid, lye, or bacteria liquid. To improve the leaching efficiency, ultrasonic and mechanochemical methods can be used to assist. According to the different selectivity of leaching agents to metal, leaching can be divided into total metal leaching and selective leaching.

2.4.1.1 Full Leaching

2.4.1.1.1 Inorganic Acid Leaching

The inorganic acids of hydrochloric acid (HCl), sulfuric acid (H_2SO_4), and nitric acid (HNO_3) [14–16] have a wide source and low cost. They are often leaching agents to leach metals from spent power batteries. However, the Co and Mn in the



battery are insoluble Co^{3+} and Mn^{4+} . To improve the leaching efficiency, adding hydrogen peroxide (H_2O_2), sodium bisulfite, or glucose as a reducing agent is necessary to reduce the Co and Mn in the battery to Co^{2+} and Mn^{2+} , which are easier to be dissolved.

Joulie et al. [17] studied and compared the leaching effects of three inorganic acids of H_2SO_4 , HNO_3 , and HCl on the cathode materials of NCA ternary batteries and optimized the leaching temperature, acidity, reaction time, leaching agent concentration, solid–liquid ratio, and reducing agent concentration. The results show that when no reducing agent is added, the type of acid has a greater impact on the metal leaching rate. Since chloride ions in HCl can promote the reduction and dissolution of Co^{3+} , because there is no reducing agent in sulfuric acid and nitric acid, Co and Mn exist in high valence states of Co^{3+} and Mn^{4+} ; it is difficult to dissolve in the acid solution. Therefore, the leaching rate of HCl is the highest. Xu et al. [18] leached LiCoO_2 batteries with hydrochloric acid as the leaching agent and reached a similar conclusion that the presence of chloride ions promoted the dissolution of Co. The specific mechanism is as follows:



As shown in the previously presented formula, HCl can be used as both a bleaching agent and a reducing agent to promote Co's efficient reduction and leaching. But at the same time, HCl will be oxidized to produce Cl_2 , which is harmful to the environment. Therefore, sulfuric acid or nitric acid is usually used as the leaching agent in industrial use. To increase the leaching rate of Co and Mn during inorganic acid leaching, reducing agents such as $\text{Na}_2\text{S}_2\text{O}_5$, Na_2SO_3 , and H_2O_2 are usually added for reductive acid leaching to reduce Co^{3+} and Mn^{4+} in the positive electrode material to Co^{2+} and Mn^{2+} with better solubility. To avoid the introduction of impurity ions, H_2O_2 is often used as a reducing agent.

Lee and Rhee [16] found that when LiCoO_2 was leached with HNO_3 alone, the leaching rates of Li and Co were only 75% and 40%, respectively, under the optimal parameters. But after adding 1.7%(v/v) H_2O_2 , because Co^{3+} can be reduced to Co^{2+} , the leaching rate of Co and Li can be increased to more than 99%. The results of Chen et al. [19] showed similar conclusions. When H_2SO_4 is used as the leaching agent and H_2O_2 is used as the reducing agent, the leaching rates of Co and Li in lithium batteries can reach 95% and 96%, respectively.

Inorganic acid leaching is a relatively mature process technology. Many in-depth studies have been conducted on the concentration of the bleaching agent, the amount of reducing agent, the leaching temperature, time, and solid-to-liquid ratio. Whether HCl , H_2SO_4 , or HNO_3 , Ni, the leaching rate of Co, Mn, Li, etc., can be greater than 90%, and the mainstream leaching technology for waste lithium batteries is inorganic acid leaching.

2.4.1.1.2 Organic Acid Leaching

The leaching system used in industry today is mainly inorganic acid, which has high leaching efficiency, and the leaching rate of valuable metals can reach more than 90%. However, there are still some unavoidable problems in industrial applications.



First, when using inorganic acid leaching, harmful gases such as Cl_2 , SO_3 , NO_x , etc. will be released, which seriously threaten the environment and human health. Secondly, when using inorganic acid leaching, the pH of the leaching solution is low, and it is difficult for the valuable metals in the leaching solution to be directly recovered. To remove impurities such as Al, Cu, and Fe or recover valuable metals such as Ni, Co, and Mn requires a large amount of lye to neutralize the excess acid. Finally, much acidic wastewater will be produced after the leaching is completed, requiring subsequent treatment and operating costs.

Compared with inorganic acids, organic acids have the advantages of easy degradation, good circulation, less secondary pollution, and sufficient acidity to leach cathode materials, which is expected to become a substitute for inorganic acid leaching systems. Research has found that citric acid, oxalic acid, lactic acid, malic acid, trichloroacetic acid, aspartic acid, tartaric acid, and other organic acids can be leached agents for the recycling of spent batteries. H_2O_2 (1–6%) is often used as a reducing agent to improve the leaching efficiency. Zheng et al. [20] found that when 1% (v/v) H_2O_2 was added as a reducing agent, the leaching efficiency of Co in LiCoO_2 by citric acid could reach 99.07%. Zhang et al. [21] used trichloroethylene as the leaching agent and H_2O_2 as the reducing agent. Under the optimal leaching conditions, the leaching rates of Co, Ni, Mn, and Li could reach 91.8%, 93.0%, 89.8%, and 99.7%, respectively.

So far, many research reports on organic acid leaching have been carried out, but there are still many problems that need to be solved before it is used in industrialization. First, the price of organic acid is higher than that of inorganic acid, which leads to an increased recovery cost. Secondly, the leaching rate of organic acid is slow, and the consuming time is longer. Finally, the most suitable solid-to-liquid ratio for organic acid leaching is lower than that of inorganic acid, resulting in the capability of leaching cathode materials in a unit volume of organic acids being weaker than inorganic acids.

2.4.1.1.3 Bioleaching

Compared with inorganic and organic acids, bioleaching has been widely used in the leaching process of low-grade minerals, waste catalysts, and fly ash due to its environmental friendliness, low cost, and low demand for industrial applications. However, it is still in the laboratory research stage to leach spent power battery cathode materials.

Xin et al. [22] cultured S-oxidizing and Fe-oxidizing bacteria with a mixture of three energy sources, elemental S, pyrite (FeS_2), and S + FeS_2 mixture, and investigated their leaching performance on cathode materials. In the S energy source system, the pH of the leaching solution was the lowest, and the leaching rate of Li was the highest. For Li bioleaching, the most dominant mechanism was acid solubilization. The highest leaching rate for Co occurs in the S + FeS_2 mixed energy source system, indicating that the leaching of Co is not only influenced by acid solubilization but also by the catalytic reduction ability of Fe^{2+} , and Co^{3+} needs to be reduced to Co^{2+} by Fe^{2+} to be better dissolved in the acid solution.



The most obvious drawback of bioleaching is the low leaching rate, limiting its industrial application. To improve the leaching rate, Zeng et al. [23] found that copper ions could accelerate the leaching rate of Co. In the absence of copper ions, the leaching rate of Co was only 43.1% after 10 days of leaching, but with the addition of 0.75 g/L copper ions, the leaching rate of Co could reach 99.9% after six days of leaching. Similarly, Zeng et al. [24] found that the leaching efficiency of Co could be improved when a certain amount of silver ions was added as catalyst. The leaching rate of Co could be increased from 43.1%, which is the result of no addition of silver ions, to 98.4% after seven days of leaching (with 0.02 g/l silver ions). Compared with bacteria, fungi have stronger toxicity tolerance, shorter retardation period, and faster leaching rate and are widely used to recover heavy metals from a variety of solid wastes. During the leaching process, fungi secrete various organic acids such as malic, gluconic, oxalic, and citric acids, which can effectively leach heavy metals, with leaching rates of up to 100%, 95%, 70%, 65%, 45%, and 38% for Cu, Li, Mn, Al, Co, and Ni, respectively, at a slurry density of 1% [25].

Bioleaching has attracted extensive research due to its low cost and environmental friendliness, but some unsolved problems restrict its industrialization process. On the one hand, the bioleaching rate is slow, and the required microorganisms are difficult to cultivate effectively, so the bioleaching cycle is long even if a catalyst is introduced. On the other hand, high concentrations of metal ions are toxic to microorganisms, and the concentrations of metal ions in bioleaching are low, and the high posttreatment costs. For example, after increasing the slurry density from 2% to 4%, the leaching rates for Co and Li decreased from 89% and 72% to 10% and 37%, respectively [26]. The low concentration of the leachate undoubtedly increases the difficulty of the subsequent treatment.

2.4.1.2 Selective Leaching

Since the cathode material contains a variety of metals, to obtain valuable metals, a multistep process such as dehybridization and separation is usually required, and the recovery process is long. A selective leaching method can be used to simplify the separation and recovery process.

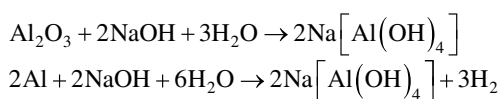
Higuchi et al. [27] studied a method for selective recovery of Li: adding $\text{Na}_2\text{S}_2\text{O}_8$ as an oxidant to the sulfuric acid leaching system can oxidize Li to water-soluble Li_2SO_4 , Mn, Co, and Ni to insoluble MnO_2 , Co_3O_4 , and NiOOH , respectively, and subsequent selective leaching of Li can be achieved by water leaching. Meshram proposed a method for selective leaching of Li and Co: firstly, Li and Co were converted to $\text{LiCo}(\text{SO}_4)_2$, LiMnO_3 , and Co_3O_4 by roasting at 300 °C for 30 minutes in a sulfuric acid atmosphere, and then 78.6% Li and 80.4% Co were leached by water leaching. At the same time, the leaching rate of Ni and Mn was less than 15%. Zhu et al. [28] used sulfuric acid as a bleaching agent, and suitable molar ratios selectively leached Li and Co in lithium cobaltate and LFP batteries. The leaching rates of both Co and Li were above 96%. The concentrations of cobalt and lithium in the leach solution were 64.41 and 17.23 g/L, respectively, and the contents of iron and phosphorus impurities were only less than 0.02 g/L; the leaching residue was mainly



iron phosphate and carbon powder, and the phosphate could be recovered by calcination or acid leaching iron.

Since oxalate of Ni, Co, and Mn are precipitated, while oxalate of Li is soluble, Li can be selectively leached by oxalic acid. Zeng et al. [29] used oxalic acid to leach LiCoO_2 . 98% Li can be selectively leached, while Co exists in the form of $\text{CoC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ precipitate in the slag. Zhang et al. [30] studied the leaching effect of oxalic acid on LNCM cathode material, and the recovery of Li was 81%. Zhang investigated the effect of oxalic acid on the leaching of LNCM cathode materials, and the recovery of Li was 81%, and the purity could reach 97%. While Ni, Co, and Mn are still present in the solid as oxalate precipitates. Phosphoric acid can also be used for selective extraction of Li. Chen et al. [31] studied the leaching effect of the phosphoric acid leaching system on LiCoO_2 and the addition of 4 vol% H_2O_2 as reducing agent to 0.7 M phosphoric acid solution could convert 99% of Co and Li into $\text{Co}_3(\text{PO}_4)_2$ precipitate and LiH_2PO_4 leachate, and thus the separation of Li and Co could be achieved. Bian et al. [32] investigated the leaching effect of phosphoric acid on LiFePO_4 . During the leaching process, LiFePO_4 was first dissolved into Li^+ , Fe^{3+} , and PO_4^{3-} , and then Fe^{3+} could be converted into FePO_4 precipitate at 85°C , while Li was stably present in the leachate to achieve the separation of Li and Fe.

The metal enrichment process generally uses acid to leach the metal in the cathode material, whether inorganic acid or organic acid; usually all kinds of metal in the cathode material can be leached, such as Cu, Fe, Al, Ni, Co, and Mn; the metal leaching is not selective. Since aluminum is an amphoteric metal, it can be dissolved in acid and alkali solutions, and none of the metals in the electrode material can react with alkali. Therefore, the alkali leaching method can dissolve the aluminum foil before acid leaching. It can avoid the dissolution of aluminum during acid leaching and affect the recovery of nickel, cobalt, and manganese. Nan et al. [33] found that when 10 wt% NaOH, 100 g/L solid–liquid ratios were used, the leaching rate of aluminum could reach 98% after five hours reaction at room temperature, and cobalt and lithium were not leached. Chen et al. [19] found that the electrode material was treated with 5% NaOH for four hours after grinding, and the leaching rate of aluminum could reach 99.9%.



This method is a simple process with high aluminum removal efficiency. Still, it is rarely used in industry because of the difficulty of recovering aluminum in the leach solution due to its special ionic form. And there is alkaline wastewater generated, which requires subsequent treatment.

Ammonia leaching has a selective leaching effect between target metals (Li, Ni, and Co) and nontarget metals (Fe, Mg, Al, and Mn). Therefore, selective leaching of Ni, Co, and Li can be achieved by ammonia leaching, avoiding the influence of impurities such as Fe, Mg, and Al. Wang et al. [34] studied the leaching behavior differences of valent metal elements in waste NCM battery cathode materials under



different reducing agents and buffer solutions and analyzed the ammonia leaching process in detail. It was found that the reductant promoted the leaching of metal elements in the NCM material without the additional buffer solution. Still, the dissolution of aluminum elements was inhibited when the ammonium sulfite was added as the reductant. The buffer solution could effectively inhibit the dissolution of aluminum elements and increase the dissolution efficiency of other metal elements besides aluminum elements. By studying the $\text{NH}_3\text{--}(\text{NH}_4)_2\text{CO}_3\text{--}\text{Na}_2\text{SO}_3$ selective ammonia leaching system, it was found that the single-stage leaching could achieve the efficiency selective leaching of 79.1% of lithium, 86.4% of cobalt, and 85.3% of nickel, and only 1.45% of manganese entered the solution. The multistage leaching could efficiently leach valuable metals (98.4% of lithium, 99.4% of cobalt, and 97.3% of nickel could be dissolved). Ku et al. [35] studied the $\text{NH}_3\text{--}(\text{NH}_4)_2\text{CO}_3\text{--}\text{Na}_2\text{SO}$ selective ammonia leaching system, in which ammonium sulfite reduced Ni and Co to more soluble divalent ions under the condition that ammonium carbonate was used as a buffer to ensure pH stability, which promoted Ni and Co to complex with NH_3 more easily so that Co and Ni could be leached selectively, but Mn and Al could not be leached. Thereby, leaching can achieve the separation of Ni, Co, Mn, and Al.

2.4.1.3 Enhanced Leaching

Both acid and alkali leaching generally require high-temperature treatment and long leaching times. To improve the leaching efficiency, some auxiliary methods can usually be used.

Ultrasonic waves are a common auxiliary enhancement method. It has been proved that ultrasound helps increase the leaching rate of valuable metals from various materials. On the one hand, ultrasound promotes the convective movement of the material and the exchange between solids and liquid during the leaching process. On the other hand, due to the cavitation effect, a large amount of energy is released at the solid–liquid interface, promoting the accelerated leaching rate of metal elements. Li et al. [8] investigated the ultrasound-assisted leaching of active cathode materials for lithium batteries. The results showed that the recovery of cobalt was 96.13%, and lithium was 98.4% under the dual action of 90 W ultrasonic power and acid and hydrogen peroxide. Zhu et al. [36] showed that ultrasound could provide hot cavity pressure with very high temperatures. The hot cavity leads to free radical reactions and induces the production of H_2O_2 , which is beneficial to improve the leaching efficiency. The results showed that the leaching efficiency of cobalt and lithium in the leach solution was significantly increased by ultrasonic-assisted leaching at low H_2SO_4 concentrations.

Mechanical activation can induce changes in minerals' crystal structure and physicochemical properties by mechanical force. Part of the mechanical energy is transformed into the internal energy of the material, which accelerates the rate of leaching reaction and achieves the purpose of enhancing leaching. Guan et al. [37] found that after mechanical grinding and activation, the particle size of the material is reduced, the specific surface area is increased, and the crystal structure is



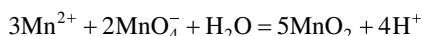
transformed, which can significantly improve the leaching efficiency of cobalt and lithium. The leaching efficiency of cobalt can be increased from 23% to 91%.

The leaching efficiency of cathode materials can also be enhanced in extreme environments. Bertuol et al. [38] used hydrogen peroxide and sulfuric acid as cosolvents to selectively leach Co from spent lithium batteries using supercritical CO₂ extraction. The recovery of cobalt could reach 95.5% within a five-minute reaction time. However, a reaction time of 60 minutes was required when only hydrogen peroxide and sulfuric acid were used to achieve the same effect.

2.4.1.4 Chemical Precipitation

Chemical precipitation methods are generally used for leachate removal or product preparation [39]. Chemical precipitation is based on the different solubility of metal compounds at a certain acidity and alkalinity to achieve separation. Waste LIB leachate usually contains valuable metal ions such as Li, Ni²⁺, Co²⁺, and Mn²⁺, while the liquid contains impurity metal ions such as Al³⁺, Fe³⁺, and Cu²⁺. Generally speaking, the solubility of transition metal hydroxides and oxalates is much lower than that of the corresponding lithium compounds. Also impurity metal ions such as Fe³⁺, Al³⁺, and Cu²⁺ usually precipitate at relatively low pH [19, 40, 41]. Therefore, impurity ions should be removed first to avoid coprecipitation in the subsequent separation process, followed by precipitation of transition metal ions, and finally recovery of the remaining lithium ions in solution. The precipitants usually used in the chemical precipitation process are sodium hydroxide, sodium carbonate, ammonium oxalate, potassium permanganate, etc. Therefore, to achieve the separation and recovery of metal components, the selection of precipitant and precipitation conditions are the key to chemical precipitation.

Based on the chemical precipitation method, Chinese researchers used KMnO₄ solution to selectively separate and precipitate Mn²⁺, and about 99.2% of Mn²⁺ was removed and precipitated in the form of MnO₂ and Mn₂O₃. Then the Ni-loaded Mextral® 272P was used as a new extractant to separate and recover the Co²⁺ from the leachate. Finally, the remaining Ni²⁺ and Li⁺ in the leachate were precipitated successively using NaOH and Na₃PO₃ solutions, respectively, and after filtration and drying, Ni²⁺ and Li⁺ were recovered as Ni(OH)₂ and Li₃PO₄, respectively. The recovery efficiencies of Cu, Mn, Co, Ni, and Li can reach 100%, 99.2%, 97.8%, 99.1%, and 95.8%, respectively, under the respective optimal experimental conditions.



The advantages of the chemical precipitation method are simple operation, good separation effect, and low equipment requirements. Still, its requirements for process parameters are more stringent. In contrast, the precipitation process may lead to the entrapment and adsorption of metal ions, and the purity of recovered products is low, and the metal loss rate is high.



2.4.2 Metal Extraction Separation

The solution after leaching contains valuable metals such as cobalt, lithium, nickel, and impurities such as copper, iron, aluminum, and manganese. The most studied methods are mainly solvent extraction and chemical precipitation.

2.4.2.1 Solvent Extraction Method

The solvent extraction method is widely used to separate and recover waste lithium batteries' metal elements. Its principle is to use organic solvents and leaching solutions in the target ions to form stable complexes. Then the appropriate organic solvent separates them and extracts the target metals and compounds. Commonly used extractants are (2,4,4-trimethylpentyl)phosphonic acid (Cyanex272), (2-ethylhexylphosphonic acid-mono-2-ethylhexyl)lipid (PC88A), (2-ethylhexyl phosphoric acid mono-2-ethyl)hexyl lipid (P507), tricine (TOA), and di-(2-ethylhexyl) phosphoric acid (D2EHPA). At present, the main process is the waste lithium-ion cathode material leachate in turn with a precipitating agent to remove copper, sodium carbonate to adjust the pH of the leachate to remove iron and aluminum impurities, and D2EHPA purification to remove impurities, D2EHPA extraction of manganese, and P507 step-by-step extraction of cobalt and nickel (Figure 2.11) [39]. But P507 separation coefficient is limited, so there are difficulties for high nickel-type cathode waste leachate separation. The separation coefficient of nickel and cobalt of Cyanex272 is greater than that of P507, and it can be used to extract high nickel and low cobalt solutions. However, this method also has shortcomings; separating nickel, manganese, and cobalt one by one is easy to cause the loss of nickel and lithium valuable metals, resulting in the overall low recovery of metal ions. On this basis, the synergistic extraction of multiple extractants can increase the selectivity between ions. At present, a new extraction agent is developed for the simultaneous extraction of transition metals: nickel, cobalt, and manganese. The mixed solution of manganese, cobalt, and nickel obtained by extraction separation can be directly used as the raw material liquid of the precursor system of NCM cathode material. The separation of transition metal and lithium components by this one-step method is a new trend in the research of metal ion extraction from the leaching solution of waste lithium batteries. It has been reported [19] in the literature that BC196 extractant can extract the metal nickel, cobalt, and manganese simultaneously, and it has a good separation effect with impurity ions (Figure 2.12).

2.4.2.2 Chemical Precipitation Method

The chemical precipitation method adds a specific precipitant to the leaching solution to precipitate out the metal ions in the metal leaching solution and get the corresponding metal compound products. The core of the chemical precipitation method is to control the pH of the solution and precipitate the corresponding metal ions at different pH. Commonly used precipitants are sodium hydroxide (NaOH), potassium permanganate (KMnO₄), butanedione oxime (C₄H₈N₂O₂), sodium carbonate, etc. [40, 41].



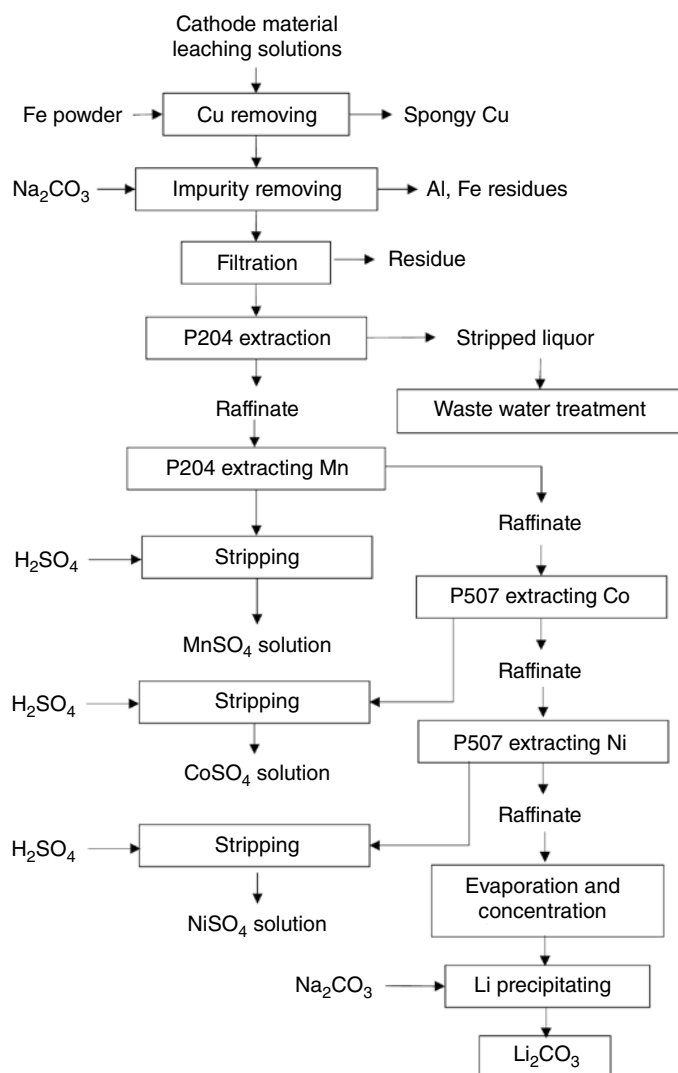


Figure 2.11 The process of combining solvent extraction and precipitation to recover valuable metal ions in the leachate. *Source:* Adapted from Li et al. [39].

In addition to the previously mentioned leach solution of iron and aluminum impurities removal using precipitation method, the chemical precipitation method is mainly used to sink Li^+ by regulating the solution pH, then adding saturated sodium carbonate solution to get Li_2CO_3 precipitation; after washing with acid back solution, the counter solution again sinks lithium to get high-purity lithium carbonate. Nayl et al. [42] regulated the leach solution pH with NaOH and added saturated Na_2CO_3 solution step by step to sink Mn^{2+} , Ni^{2+} , Co^{2+} , and Li^+ . At a leachate pH of 7.5, Mn^{2+} could be precipitated as MnCO_3 , and Ni^{2+} could be precipitated as NiCO_3 by adjusting the pH to 9. Co^{2+} was precipitated by adjusting the leachate pH to 11–12 to obtain Co(OH)_2 . Finally, Li^+ was precipitated as Li_2CO_3 in the residual solution, and the



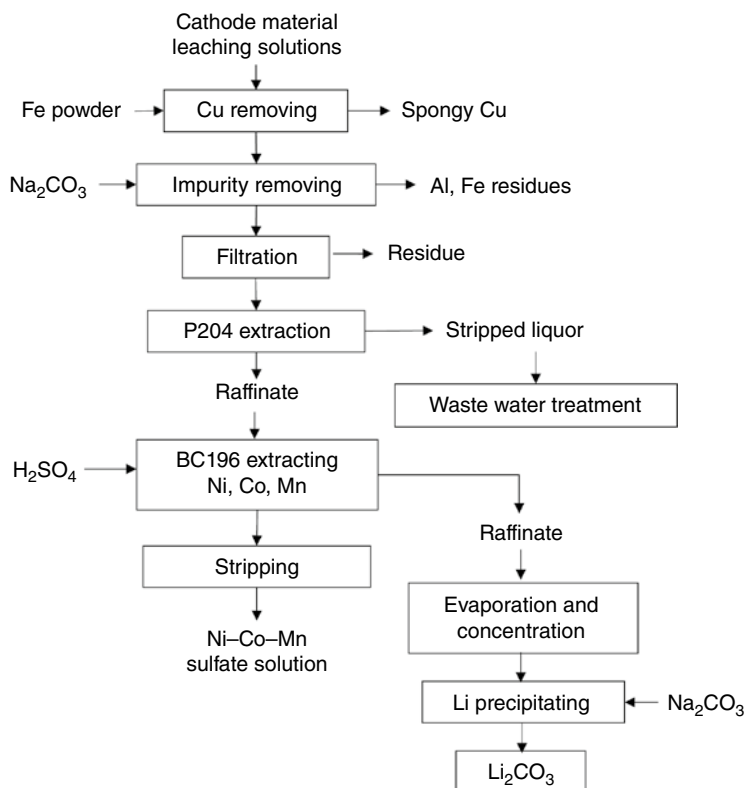


Figure 2.12 The novel process of the combination of solvent extraction and precipitation to recover valuable metal ions in the leachate. *Source:* Adapted from Chen et al. [19].

precipitation rates of Mn, Ni, Co, and Li in this method were 94%, 91%, 95%, and 90%, respectively. Sattar et al. [43] controlled the pH of the bleach solution to 2.5, added KMnO_4 to oxidize Mn^{2+} to MnO_2 for removal selectively, then controlled the solution pH to 5, and added butanedione oxime to precipitate Ni^{2+} . Co^{2+} was extracted by Cyanex272, H_2SO_4 reverse extraction of the loaded organic to obtain CoSO_4 solution, and finally adjusted the extracted residual solution pH 12 and added Na_2CO_3 to precipitate Li^+ , and got Li_2CO_3 precipitation. The recovery rates of this method of Li, Ni, Mn, and Co recovery were 99%, >99%, >98%, and 99.9%, respectively.

The chemical precipitation method only needs to control the pH of the solution and add specific precipitant, which has higher recovery and lower cost and is easy to realize industrial production. However, as the leaching solution contains a variety of metal ions, metal entrapment is inevitable during the precipitation process, which leads to impurities and low purity in the final precipitation product obtained.

2.4.2.3 Electrochemical Method and Other Methods

The electrochemical method, also known as the electrodeposition method, can dispose of spent lithium cobalt acid batteries. The Co^{3+} in the leaching solution is converted into Co^{2+} by electrochemical reduction technology and finally deposited in



$\text{Co}(\text{OH})_2$ at the anode. This method does not require the addition of other substances, is not easy to introduce impurities, and can obtain very pure cobalt compounds, which are directly used to prepare electrode materials. Still, the disadvantage is that it consumes a lot of electricity.

The ion exchange method uses the difference in adsorption capacity of different metal ion complexes such as Co and Ni on the ion exchange resin to achieve the separation and extraction of metals. The method is selective for target ions, simple, and easy to operate. It provides a new way to extract and recover valuable metals from spent lithium batteries, but the high cost limits the industrial application.

2.5 Pyrometallurgy

Pyrometallurgy technology is a kind of metal melting method. The raw materials will undergo a series of physical and chemical change processes at high temperature (using the latent heat of raw materials themselves, some chemical reaction heat, fuel combustion, or electricity generated heat). Then the metal and other impurities are separated. Because it has the advantages of simple operation and large processing capacity, it is the most widely used metal separation technology. It has been applied to recover valuable metals such as nickel and copper from secondary resources such as waste circuit boards. Taking the research results of Xiao Songwen et al. [44] as an example, the disposal of LIBs containing manganese, nickel, or cobalt is currently realized, and the main disposal process flow is detailed in Figure 2.13.

For spent LIBs, due to the existence of electrolytes, organic components, and electricity, and other energy sources, the latent heat contained in the battery itself can be used to carry out self-heating prereduction roasting in a rotary kiln. The roasting process makes full use of the battery's energy and can realize a self-heating reaction. In the roasting process, the battery can be roasted with electricity. At the same time, the

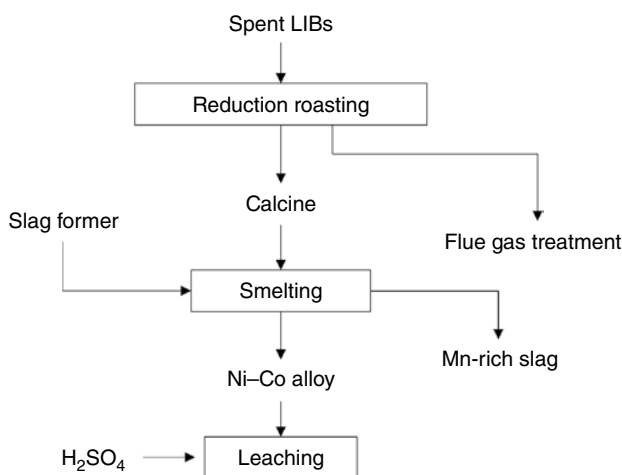


Figure 2.13 Pyrometallurgical process of LIBs.



residual electricity is fully utilized to fully burn the separator and another organic, and the heat generated is used as the heat source and the negative electrode as the reducing agent to realize the prereduction process of nickel/cobalt/manganese in the positive electrode. The flue gas from the prereduction roasting contains many small organic molecules, so a secondary combustion chamber needs to be set up to fully combust the small organic molecules. The heat generated will prepare low-pressure steam for the subsequent wet process section. This process is a completely self-heating reaction while providing a steam heat source for the subsequent wet process separation. The hydrometallurgy process uses the battery's latent heat and saves energy consumption while saving the discharge and the "three wastes" treatment brought by the discharge.

After the prereduction of the battery into the electric furnace melting, valuable metals such as nickel, cobalt, and copper enter the metallic phase by reducing melting. They are produced in the form of alloy and prepared into alloy powder by spray powder making when discharging. Aluminum foil precipitates into the slag through oxidation, and to reduce energy consumption, the lower value-added manganese slag is added into the melting slag; manganese-rich slag is produced. In this process, although the enrichment of valuable metals such as nickel, cobalt, and copper is efficiently achieved, it causes the loss of aluminum foil. Lithium enters the slag phase or dust collection ash.

Manganese and lithium enter the manganese-rich slag, from which manganese and lithium can be recovered. After sulfation roasting neutral leaching, manganese and lithium from the manganese-rich slag enter the solution as manganese sulfate and lithium sulfate, which are then recovered separately by the hydrometallurgy process. Because of the low value of manganese, the traditional hydrometallurgy extraction separation process is costly, and cannot support the recovery of manganese. The manganese-rich slag obtained by reduction melting by fire is very stable. It can be stockpiled for a long time to achieve harmless disposal and abate hazardous solid waste relative to the hydrometallurgy method.

The copper, nickel, and cobalt entering the metallic phase by melting reduction are highly pure and can be fully soluble when leaching. Then the corresponding metal salt products can be prepared separately by hydrometallurgy separation technology.

2.6 Direct Recycling Technology

2.6.1 Direct Recycling Process

Although hydrometallurgy technologies can effectively extract precious metal elements from the waste LIBs, there are still some challenges in such a long recovery process, including high reagent consumption and a large amount of waste residue, waste liquid, and waste gas. This costly recycling technology does not apply to cathode materials with little value. Figure 2.14 compares the value of battery-grade cathode material to its constituent elements. For high-cobalt cathodes (e.g. LCO), the fundamental value is similar to that of the intact cathode. Still, for low- or no-cobalt cathodes, specifically for LMO and LFP cathodes, the value that can be recovered from the elements is almost negligible, and would not compensate for the cost



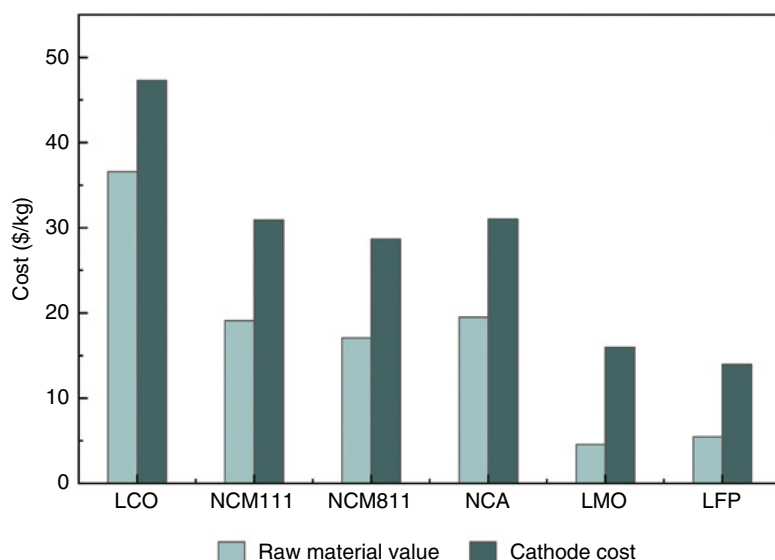


Figure 2.14 Estimated value of battery cathode materials and their components.
Source: Gaines et al. [45]/MDPI/CC BY 4.0.

of the recycling process. Therefore, it is important to develop a low-cost and short process to recover LIB cathode materials.

The loss of Li and the irreversible changes of phase structure is generally believed to be one of the main reasons for the capacity attenuation of spent LIB materials. As for those waste LIB materials with low impurity content or small structural changes, it is possible to regenerate the material components directly without destroying the chemical structure and causing secondary pollution [46]. Such a green and short recycling process is defined as direct recycling technology, which shows great potential in recovering low value-added cathode materials because several energy-intensive and costly processing steps can be avoided.

The direct recycling process involves four main parts, as shown in Figure 2.15. Disassembly is the first step to disassemble the spent lithium battery into pieces of suitable size and shape to enter the subsequent processing step. The most valuable components of lithium batteries mainly include black mass (cathode material and graphite), electrolyte, copper foil, and aluminum foil. The direct recycling technology requires a low content of impurities in the cathode material powder, so the separation process of the cathode material from the other components in the second step is crucial, which largely determines the material repair effect. Most separation



Figure 2.15 The process of direct recycling of spent LIB materials.



processes are based on different material properties, such as density, solubility, hydrophobicity, magnetization, etc. The separated cathode material will next enter the restoration regeneration process. Since LiFePO_4 maintains good structural stability during charge/discharge cycles, direct recycling techniques for this type of material are more feasible and thus have attracted extensive attention from researchers.

2.6.2 Direct Regeneration of Cathode Material from Spent LFP Batteries

Direct regeneration is to obtain the cathode material from the electrode, and after proper treatment, repair the cathode material structure and reapply it to the LIB cathode material. In this process, the LFP cathode powder is usually stripped from the aluminum foil collector using NMP solvent, alkaline solution, and heat treatment. The solid-phase lithium replenishment roasting is used to repair the surface and bulk phase defects [47]. Various lithiation processes [48] are summarized in Table 2.4, along with several novel methods that have been proven to be effective in recovering cathode materials. Finally, product validation is a key aspect of direct recycling technology. The chemical composition and crystal structure should be verified by X-ray diffraction (XRD), inductively coupled plasma (ICP), or glow-discharge optical emission spectroscopy (GDOES), and the cell capacity and cycle life must meet the industry standards.

The direct regeneration process can repair the structure of the cathode material. The capacity degradation of spent Li-ion batteries is related to the loss of lithium

Table 2.4 Comparison of lithiation process characteristics.

Process type	Lithium source	Conditions	Unique features
Thermal	LiOH with knowledge of lithium vacancy concentration	Heating in two stages	—
Hydrothermal	LiOH/KOH solution	Low temperature hydrothermal reaction followed by high temperature anneal	—
Redox mediator	Anode of electrochemical cell	Room temperature	Reaction facilitated by mediator
Ionothermal	Li salt in ionic liquid	Low temperature ionothermal reaction followed by high temperature anneal	Takes place in ionic liquid
Electrochemical	Anode of electrochemical cell	Room temperature	Roll-to-roll reactor under development



and side reactions. The loss of lithium is due to the thickening of the solid electrolyte interface (SEI) and the irreversible phase change of the material [49]. Wang et al. [50] separated the LFP active material from aluminum foil by density difference and directly regenerated the repaired LFP material by high-temperature solid-phase reaction, which showed excellent electrochemical performance in lithium batteries. Xu et al. [51] reported an efficient and environmentally friendly method for regeneration of LIBs based on defect-targeted healing. Specifically, the repair regeneration of used LFP cathode materials was successfully achieved by combining low-temperature aqueous solution reduction and rapid high-temperature annealing. Song et al. [52] stripped the powder from LiFePO_4 spent battery cathode sheet and added commercial LiFePO_4/C powder to conduct solid-phase roasting. When the ratio of commercial LiFePO_4/C to used LiFePO_4 powder is 3 : 7, the temperature is 700°C ; the first discharge capacity of 0.1 C was 144 mAh/g (commercial 150 mAh/g). The lost Li was replenished during the solid-phase roasting process by adding Li_2CO_3 . The regenerated LiFePO_4 cathode material at 650°C had a discharge capacity of 140.4 mAh/g after 100 cycles at 0.2 C, with a capacity retention rate of 95.32% (commercial LFP requirement: >92.43%), further improving the electrochemical performance of the recovered LiFePO_4 [53]. The performance of LiFePO_4 is susceptible to the content of impurity metals (aluminum and copper) [54], and the pretreatment process needs to control the content of metal impurities in the cathode powder. Since HF produced during the heating decomposition of the binder PVDF is a good fluorinating agent, it is easy to form fluoride with metal elements [55], and the residual binder content in the cathode material needs to be controlled in the direct regeneration process.

Regeneration of LFP cathode materials can also be achieved using ion doping technology. Ion doping is a means to improve the electrochemical performance of LiFePO_4 materials [56]. As shown in the process flow in Figure 2.16, Xu et al. [57] mechanically activated the spent LiFePO_4 powder, $\text{NH}_4\text{H}_2\text{PO}_4$, Li_2CO_3 , and V_2O_5 in proportion to the chemical formula $(1-x)\text{LiFePO}_4 \cdot x\text{Li}_3\text{V}_2(\text{PO}_4)_3$ (where $x = 0, 0.005, 0.01, 0.03$, and 0.1) and then mechanically activated them under argon atmosphere at 450°C for four hours, and the LiFePO_4 cathode material was regenerated by solid-phase roasting at 700°C for six hours. When $x < 0.01$, the V^{5+} doping was in the Fe^{2+} site, and when $x \geq 0.03$, the V^{5+} doping and $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ coexisted. $x = 0.01$, the structure of the regenerated material was $0.99\text{LiFePO}_4 \cdot 0.01\text{Li}_3\text{V}_2(\text{PO}_4)_3$, and the first discharge capacities were 154.3 and 142.6 mAh/g at 0.1 and 1 C, respectively. mAh/g and the capacity retention rate was close to 100% after 100 cycles at 1 C.

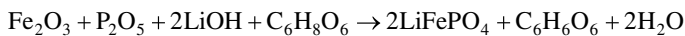
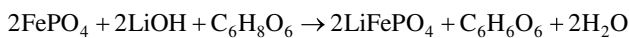
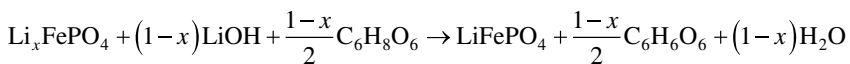
The surface coating of conductive substances is also a means to improve the electrochemical performance of LiFePO_4 materials [58], and the presence of C, N, and P atoms on the surface of the substances is beneficial for accelerating electron transfer and improving lithium storage efficiency [59]. Zhu et al. [60] coated C, N, and P atoms on the surface of LiFePO_4 cathode waste via mechanochemical activation based on the rich C, N, and P elements in lecithin. When 15% lecithin was added, the first discharge capacity of the regenerated LiFePO_4 cathode material was 164.9 mAh/g at 0.2 C. The first discharge capacity was 120 mAh/g at 5 C, and the





Figure 2.16 Recovery process of spent LiFePO_4 . Source: [57] Xu, B. et al., (2019), Elsevier.

capacity retention rate was 93% after 100 cycles, and the first charge/discharge capacity could reach 100.7 mAh/g at 20 °C, which was 41% higher than that of the uncoated LiFePO_4 scrap. Graphene is a good conductive material [61]. Song et al. [62] disassembled LiFePO_4 spent batteries and obtained positive and negative waste powders, respectively. The positive electrode was treated at 500 °C air atmosphere for three hours to remove the binder and conductive carbon black. The negative electrode graphite was exfoliated by a modified Hummers method to obtain graphene [63]. The treated cathode scrap is hydrothermally reacted with the treated negative electrode to regenerate LiFePO_4 , the cathode material, and the process flow is shown in Figure 2.17. With the addition of 5% GO (graphene oxide), hydrothermal reaction temperature of 180 °C and time of six hours, the regenerated LiFePO_4 cathode active material had a first discharge capacity of 153.9 mAh/g at 0.2 C and 150.4 mAh/g at 0.5 C, and the capacity retention rate was close to 100% after 300 cycles. The mechanism of its regeneration process is as follows:



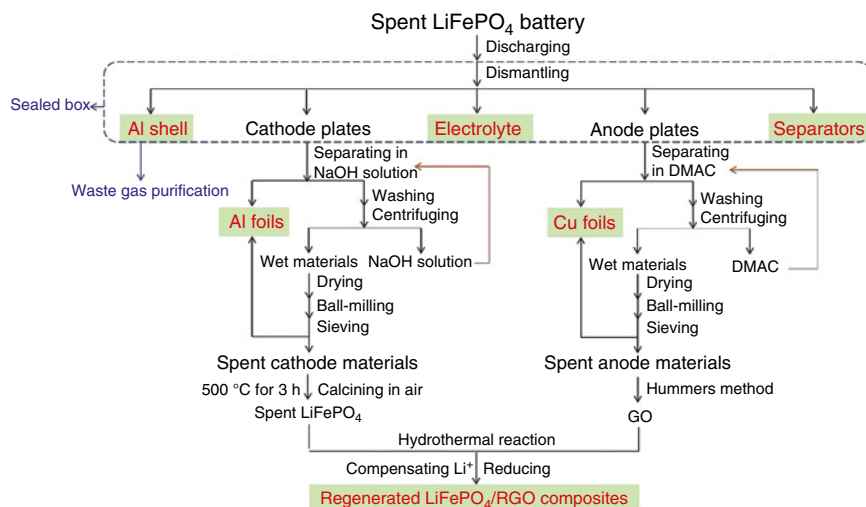


Figure 2.17 The regeneration process of spent LFP batteries. *Source:* Song et al. [62] / with permission of Elsevier.

In addition to solid sintering, ion doping, and surface coating, electrochemical recrystallization can be used to recover the loss of lithium from cathode powder [63]. Or the cathode powder can be immersed in a highly concentrated lithium salt solution, thus regenerating the LFP cathode material. Therefore, electrochemistry and chemical lithium are important methods for regenerating LFP.

2.6.3 Economic Analysis of LFP Cathode Material Recycling

LFP cathode materials are mainly recovered by hydrometallurgy recycling and direct regeneration processes, and the economics of the two processes are analyzed as follows.

The prices of chemical reagents involved in treating 1 ton of waste LiFePO_4 battery using the recycling process proposed by Sun and coworkers [64] are shown in Table 2.5. 260.7 kg FePO_4 , 56.1 kg aluminum foil, and 49.5 kg Li_2CO_3 can be recovered from 1 ton of waste LiFePO_4 battery, and the total cost of raw materials involved is 19379.68 yuan; considering only the cost of reagents and product prices, the profit from treating 1 ton of spent LiFePO_4 battery is about 11090.21 yuan. In addition, the depreciation cost of equipment, equipment maintenance cost, water consumption, and labor and energy costs are considered. The recycling process in this study can be divided into five parts: NaCl discharge and disassembly, leaching, filtration, drying and sieving, removal of impurities, and precipitation. The cost of discharging and disassembling, leaching, filtering, drying and sieving, removing impurities, and battery precipitation was calculated using the current recycling process to treat 1 ton of spent LiFePO_4 batteries; the cost of these processes was 2467.29 yuan.

The energy consumption analysis of treating 1 ton of spent LiFePO_4 battery is shown in Table 2.6. The energy consumption is mainly in the form of electrical energy, and based on the price of electricity, the cost of energy consumption can be calculated. The current recycling process's total energy consumption and total



Table 2.5 Prices of chemicals involved in treating 1 ton of spent LiFePO_4 cells using the recovery process of Sun and coworkers.

Substance	Price (yuan/kg)	Quantity (kg)	Cost (yuan)
Spent LFP batteries	18.50	1 000.00	−18 500.00
CH_3COOH	3.60	132.00	−475.20
35 wt% H_2O_2	0.95	184.80	−175.56
Na_2CO_3	3.10	72.60	−225.06
NaOH	4.20	0.92	−3.86
Waste Al foil	13.30	56.10	746.13
FePO_4	24.30	260.70	6 335.01
Li_2CO_3 (>99.5%)	472.50	49.50	23 388.75
Total			11 090.21

Source: Adapted from Yang et al. [64].

Note: The material price data in the table comes from Shanghai Metals Market (SMM), Fubao New Energy Lithium Power (<http://battery.f139.com/>), Business Society Network (<http://www.100ppi.com/>). Data updated on August 1, 2022.

Table 2.6 Energy consumption involved in the treatment of 1 ton of spent LiFePO_4 batteries using the hydrometallurgy recycling process of Sun and coworkers.

Unit operation	Energy consumption (kWh)	Price (yuan)
NaCl discharge and dismantling	207.5	290.5
Leaching	202.5	283.5
Filtration, drying, sieving	230	322
Impurity removal	120	168
Precipitation	100	140
Total	860	1204

Source: Adapted from Yang et al. [64].

energy cost are 860 kWh and 1204 yuan, respectively. In summary, a profit of 7418.92 yuan can be obtained by using the hydrometallurgy process to recycle 1 ton of spent LiFePO_4 batteries.

The solid-phase regeneration process uses the disassembly and separation of the waste LFP cathode powder. It adds lithium carbonate and ferrous oxalate to adjust the ratio of Li/Fe/P in the recovered LFP, to obtain regenerated LFP by solid-phase roasting. From the perspective of the cost of pharmaceuticals when using solid-phase regeneration method, processing 1 ton used LFP can make a profit of 10631.75 yuan (Table 2.7); the energy consumption and total energy cost of regenerating 1 ton used LFP is 3400 kWh and 2040 yuan. Comprehensively considering the cost of pharmaceuticals and energy consumption, regenerating 1 ton spent LFP can profit 8591.75 yuan (Table 2.8). Solid-phase regeneration profit depends largely on the energy density of regenerated LFP.



Table 2.7 Prices of chemicals involved in regenerating 1 ton of spent LiFePO_4 batteries.

Items	Content	Price (yuan/kg)	Quantity (kg)	Cost (yuan)
Raw materials	Spent LFP batteries	18.50	1000.00	−18500.00
Auxiliary materials	Lithium carbonate (battery grade)	472.50	4.5	−2126.25
	Ferrous oxalate	12.00	3.5	−42
Regenerated materials	LFP	156.50	200	31300
Total				10631.75

Table 2.8 Energy consumption involved in the treatment of 1 ton of spent LiFePO_4 batteries using the direct regeneration process.

Categories	Consumption (kWh)	Price (yuan)
Electricity consumption	3400	2040

2.6.4 Main Challenges for Direct Recycling and Regeneration

The main advantages of the direct regeneration recovery method include the following: (i) relatively simple process and high recovery economy, (ii) can be directly reused after regeneration, and (iii) compared with other recovery technologies, emissions and secondary pollution is significantly reduced. The main disadvantages of the direct regeneration recycling process include the following: (i) the performance of the directly regenerated cathode material is related to the health status of the spent battery; (ii) poor flexibility, which requires strict sieving/pretreatment based on precise active materials; (iii) it is a challenge to ensure high purity and consistency of the original crystal structure, which may not meet the stringent standards required by the battery industry; and (iv) it is not suitable for handling different types of mixed cathode scrap. However, in the short term, this technology is more likely to be used by battery manufacturers to recycle electrode production scrap with known chemical composition and low impurity content and has not undergone battery recycling.

The US Recell Center suggests four key core points to focus on indirect recycling technology as follows:

- i) Binder removal: Determine the best method to remove the binder and minimize the damage of the process to the cathode material.
- ii) Cathode material separation: Separate the cathode material from other components as much as possible to minimize the impurity content.
- iii) Cathode material regeneration: Develop a green and low energy regeneration process for the repair of a wide range of cathode materials (LCO, LMO, NCM, NCA, and their mixtures) in-growth.



- iv) Material upgrading and impurity impact: Upgrade materials with low market value to high-value materials through repair and regeneration, focusing on assessing the impact of impurities (e.g. Cu, Al, Fe, etc.) on material performance during the recycling process.

2.7 Equipment for Battery Recycling

2.7.1 Pretreatment Equipment

2.7.1.1 Crushing and Comminution Equipment

2.7.1.1.1 Shredder

Single battery cell mainly consists of shell, cathode and anode electrolyte, separator, and collector fluid. The shell is mainly metal steel or aluminum. The collector is mainly copper foil (anode), aluminum foil (cathode), and the cathode material includes lithium, nickel, cobalt, manganese, and other metals. The anode material is generally graphite. The cathode and anode materials are enriched in aluminum and copper foil and enveloped in each other. Due to the better ductility of copper and aluminum, the shredder is generally used in power battery recycling when shearing to tear the metal shell and cathode and anode grade pieces. Commonly used shredders are single-shaft shredder, two-shaft shredder, and four-axis shredder.

Single-Shaft Shredder

Single-shaft shredder uses movable knife particles and fixed knife interaction. Through the screen to control the discharge size, the material will be shredded, sheared, and extruded, and the material will be processed to a smaller particle size. It is often used for the fine crushing of various solid wastes. It can process the materials to small sizes at one time, which is widely used in resource recycling, refuse derived fuel (RDF) production, waste reduction, etc. It has the features of the small size of the discharge, replaceable screen, the wide applicability of materials, and high efficiency.

After the solid waste enters the single-shaft crusher through the hopper, the pushing disk pushes the solid waste to the knife shaft under the drive of the hydraulic cylinder. The motor rotates and transmits the power to the reducer through the belt drive, which drives the knife shaft to rotate and cut and crush through the fixed and movable knives, and the finished product that meets the screen size falls from the screen. The materials on the screen are returned to be crushed again.

Bolts fix the moving knife on the knife seat of the knife shaft. When the equipment is running, the incoming material will be shredded through the cutting and crushing of the moving knife and the fixed knife, and the adjusting bolt can adjust the gap between the moving knife and the fixed knife. After shredding, the material particles are extruded through the screen, and the hole of the screen determines the particle size of the discharge.



Equipment Composition The single-shaft shredder mainly consists of (i) knife shaft mechanism, (ii) screen mechanism, (iii) pushing mechanism, and (iv) driving system. In addition, there are feed hopper, frame body, discharge hopper, etc.

Knife shaft mechanism: It is required that the angle of the moving knife can be changed and used many times, the gap between it and the moving knife can be easily adjusted, and the material compatibility of the knife is good.

Screen mechanism: The screen can be changed quickly, and the screen holder can swing up and down (available hydraulic control).

Pushing mechanism: The stacking box has an adjustable guiding block to push the material to the cutting system for crushing.

Drive system: The drive system is equipped with frequency conversion control, and the rotor speed is from 80 to 240 r/min. It is possible to adjust the equipment according to the properties of the feed material (Figure 2.18).

Two-Shaft Shredder

Two-shaft shredder uses the principle of mutual shearing and tearing between two relatively rotating knives to crush the materials. It is driven by double motors + double planetary reducer (also driven by a hydraulic motor), with strong power and stable operation. It is often used in environmental protection such as municipal waste disposal, waste incineration pretreatment, bulky waste disposal, decoration waste disposal, industrial waste disposal, and resource recycling pre-crushing (Figure 2.19).

When working, the battery cell enters the crushing cavity from the inlet. The material is cut and torn into flakes by the relative rotating knives in the crushing cavity and discharged from the bottom discharge port. The size of the crushing

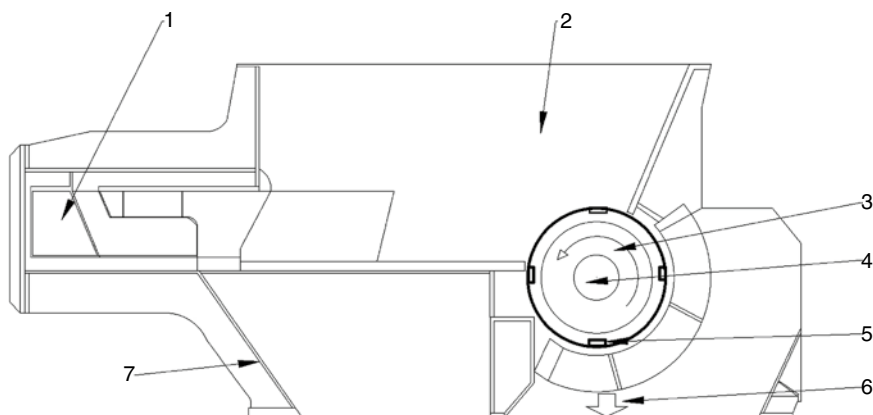


Figure 2.18 Single-shaft shredder. 1 – Pushing mechanism; 2 – feed hopper; 3 – cutter shaft mechanism; 4 – drive system; 5 – screen mechanism; 6 – discharge hopper; 7 – the frame body.



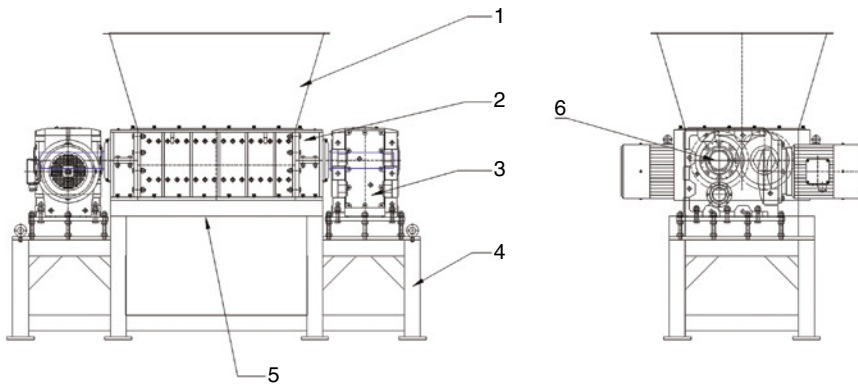


Figure 2.19 Two-shaft shredder. 1 – Feeding hopper; 2 – crushing chamber; 3 – driving device; 4 – frame body; 5 – discharge port; 6 – cutting tool.

chamber, the shape and size of the knives, and the rotational speed of the spindle are the key parameters that affect the shredder's performance.

Four-Shaft Shredder

Four-shaft shredder, which uses the working principle of mutual shearing, tearing, and extrusion between knives to process materials, is used for crushing various solid wastes, which can process materials to a smaller size at one time, and is often used in MSW disposal, resource recycling, waste incineration pretreatment, cement kiln co-processing, and other environmental protection fields. The equipment adopts low-speed and high torque design, with high shearing force, stable equipment, and uniform discharge.

The four hydraulic or electric motors drive the four knife shafts to rotate forward and backward. The upper row of knife shafts and the lower row of knife shafts cooperate in carrying out the primary breaking of materials and have the function of plucking and feeding. The secondary crushing is mainly done by the lower row of the rotor with shearing, squeezing, and tearing. The size of the discharged product is mainly determined by the thickness of the blades installed on the knife shafts and the size of the openings of the screen. The equipment can adjust the size of the discharged material by replacing the screen with different apertures (Figure 2.20).

2.7.1.1.2 Crusher

In the lithium battery pretreatment process, after the battery is coarsely broken, low-temperature cracking, and other processes, it needs to be further crushed to strip the black mass (cobalt, lithium, etc.) from the copper and aluminum collectors. The general size of the crushed material is about 30–50 mesh. Commonly used equipment is a horizontal hammer crusher and rotor centrifugal crusher.



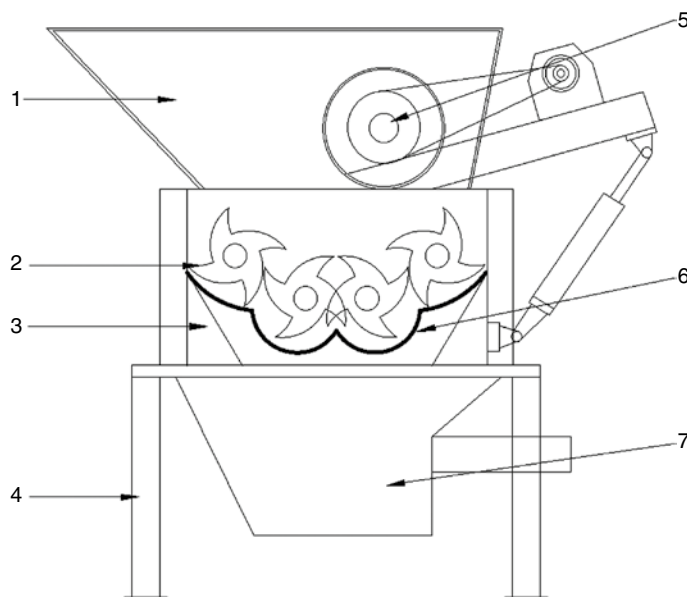


Figure 2.20 Four-shaft shredder. 1 – Feeding hopper; 2 – knife roll assembly; 3 – crushing chamber; 4 – the frame body; 5 – drive device; 6 – screen; 7 – the hopper.

Horizontal Hammer Mill

The hammer crusher mainly relies on the impact energy to complete the crushing operation of materials. When the hammer crusher is in operation, the motor drives the rotor to rotate at high speed, and the material enters the crusher cavity evenly. The high-speed rotating hammerhead impacts, shears, and tears the material to be crushed. Meanwhile, the gravity of the material itself causes the material to rush from the high-speed rotating hammerhead to the baffle plate and sieve bar inside the frame. The material larger than the sieve hole size remains on the sieve plate and continues to be struck and ground by the hammer until it is crushed to the required size. The material is discharged from the machine through the screen plate (Figure 2.21).

Rotor Centrifugal Crusher

The rotor centrifugal crusher comprises a rotor mounted on a vertical shaft rotating horizontally and a cylinder with a ring-shaped crushing chamber inside. The material is fed from above into the rotor rotating at high speed when it works. The material is subjected to centrifugal inertia force through the diversion plate redirected from vertical to horizontal spiral along with the runner plate. The outer circumference of the rotor exit is thrown and crushed in the crushing cavity. The material produces a series of energy exchanges between the chain reactions in the crushing cavity. It will form a sand spray phenomenon so that part of the material forms a



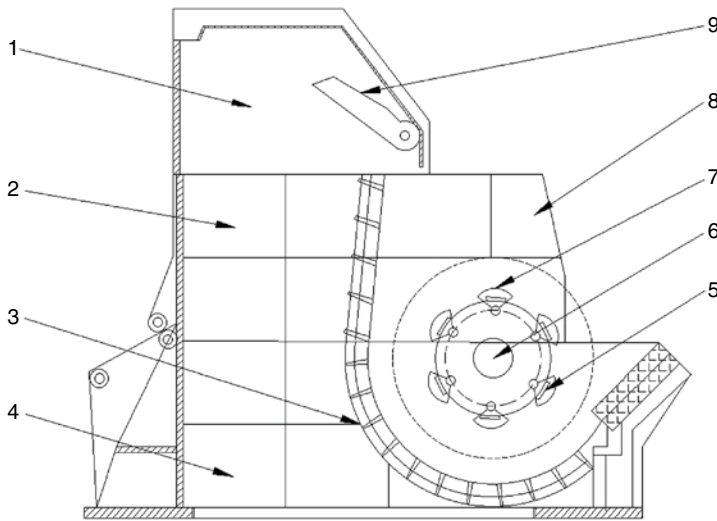


Figure 2.21 Horizontal hammer mill. 1 – Upper cover body; 2 – upper box; 3 – sieve plate; 4 – lower box body; 5 – anvil iron; 6 – spindle roller; 7 – hammer head; 8 – lining board; 9 – upper discharge door.

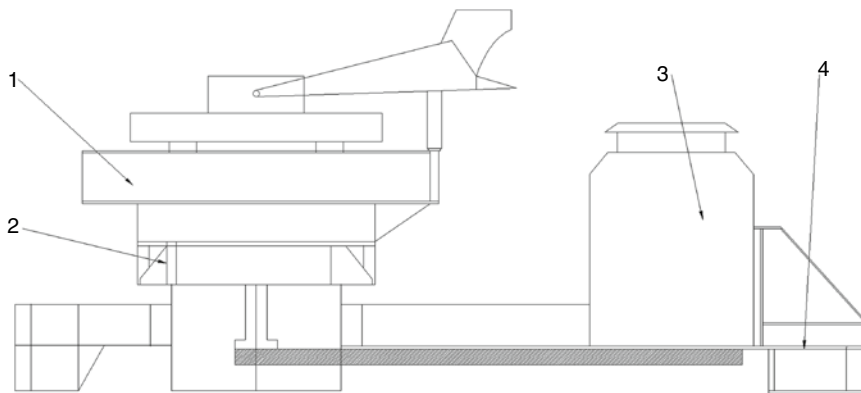


Figure 2.22 Rotor centrifugal pulverizer. 1 – Crushing chamber; 2 – rotor; 3 – driving system; 4 – frame body.

particle cloud, which flows around the crushing cavity until it loses enough speed and leaves it (Figure 2.22).

2.7.1.2 Nitrogen Protection System

Although the crushed power battery has been discharged in advance, some batteries are not completely discharged, and these batteries are prone to short circuits and fire or even explosions when crushed. For this reason, the crushing process must be carried out in a low-oxygen environment, and the common system is a nitrogen protection system (Figure 2.23).



The bottom of the battery feed hopper and the upper part of the discharge hopper is equipped with hydraulic gate valves. When crushing, the crushing chamber is filled with nitrogen gas in advance. The oxygen concentration is controlled below 8%; the hydraulic gate valve at the bottom of the feed hopper opens and closes intermittently to complete the material transportation. The hydraulic gate valve opens at the top of the discharge hopper to complete a crushing cycle after the material is crushed.

When crushing, the gas pressure sensor detects the pressure data in the crushing chamber in real time. It combines it with the oxygen concentration data to determine whether the crushing chamber fire and explosion occurs. When the explosion has been judged to have occurred, the control system, on the one hand, starts fire-fighting equipment to put out the fire and at the same time control the active explosion relief device to open, so that the high-temperature and high-pressure gas are generated to the designated safe location to release and to reduce the harm caused by the explosion on the crusher.

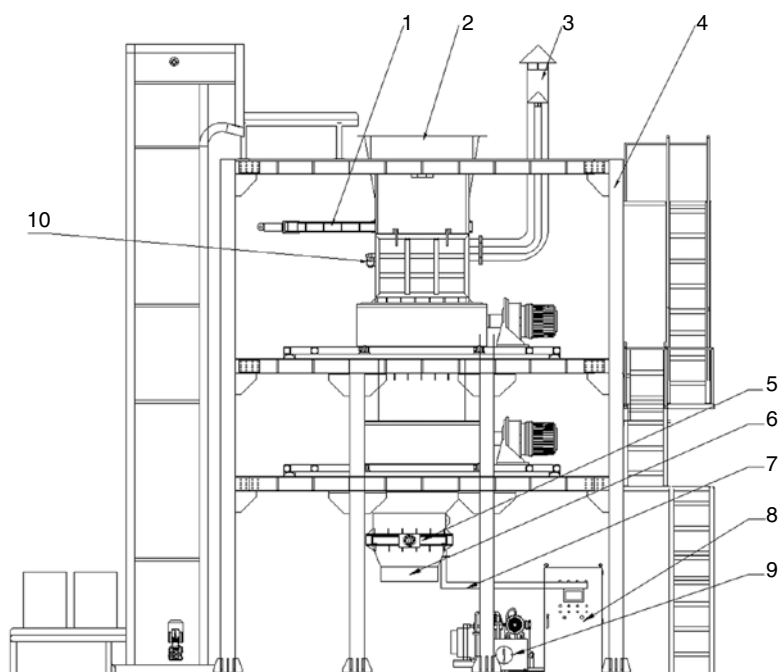


Figure 2.23 Nitrogen protection system. 1 – Fire gate valve 1; 2 – feeding bin; 3 – explosion relief valve; 4 – frame body; 5 – fire gate valve 2; 6 – discharge chute; 7 – fire extinguishing passage; 8 – control cabinet; 9 – hydraulic station; 10 – air pressure detection system.



2.7.1.3 Separation and Debris Removal Equipment

2.7.1.3.1 Wind Sieving Machine

A wind separator system (airflow sieving) separates particles by density or particle size with air as the sieving medium under airflow action. The basic principle of airflow sieving is that the airflow will take the lighter material upward or from the horizontal direction to a farther place. In contrast, the heavy material cannot support it due to upward airflow and sink or heavy material due to sufficient inertia. It cannot change the direction dramatically through the airflow to sink the light material taken away by the airflow and then further separated by the cyclone dust collector.

Specifically, to the lithium battery disassembly, a wind separator is mainly used to (i) remove the light separator in the battery, (2) separate battery shell and grade, and (iii) subsequently separate copper and aluminum in the state of the large pole piece. Commonly used wind sieving equipments are folding plate wind separators and pulsating airflow wind separator columns.

Folding Plate Wind Separator Folding plate-type wind separator (Z, the word) is a kind of vertical wind separator commonly used in solid waste treatment, processing particle size generally in 5–40 mm. Because of its high sieving efficiency, it is generally used for fine sieving or purification of solid waste treatment. It is also widely used in lithium battery recycling, such as BHS, etc.

When the folding plate wind separator works, the material (material A and B) generally enters the rotating feeder (star unloader). At this time, the centrifugal ventilator drums the airflow from the bottom of the wind separator. Materials in the process of falling, by the role of rising airflow, low-density materials (B), will move upward into the cyclone separator and be discharged from the bottom of the separator. High-density materials (A) will move downward by the action of gravity due to their own heavier and be discharged from the bottom of the wind separator to achieve material separation (Figure 2.24).

Pulsating Airflow Air Separation Column

Shown in Figure 2.25 is a pulsating airflow sieving column. When working, the material enters by the feeding device (generally a rotary feeder) 7. The airflow conveyed by the active fan 1 passes through the pulsation valve 4 to produce pulsating accelerated airflow. The pulsation frequency of the sieving airflow is controlled by the motor connected to the inverter. The pulsating valve is a butterfly valve, and the rotation of the valve spool realizes intermittent switching of the pipeline to produce pulsating airflow. The greater the acceleration of the pulsating airflow, the greater the pulsating acceleration of the particles to be sorted. The acceleration of the particles in the pulsating airflow and the density of the particles are inversely



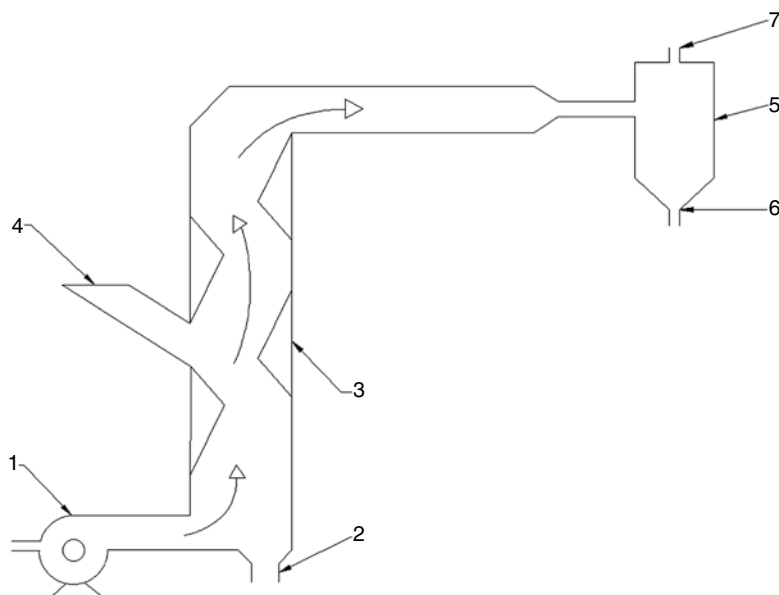


Figure 2.24 Folding plate wind separator. 1 – Centrifugal fan; 2 – discharge port 1; 3 – main body of air separator; 4 – feed inlet; 5 – cyclone separator; 6 – discharge port 2; 7 – the outlet.

proportional. That is, the greater the density of the particles, the smaller the pulsating acceleration, the particles tend to sink; the smaller the density of the particles, the greater the pulsating acceleration, the particles tend to rise.

Materials in the wind selection column 6 will be affected by the pulsating airflow generated by the active fan 1. The light material will move upward of the wind selection column 6 into the cyclone separator 9 by pulsating airflow and be discharged through the light material discharge 10. Heavy material in the role of gravity will be discharged by the heavy material discharge 11.

2.7.1.3.2 Rotary Kiln Furnace

The crushed primary battery is decomposed into the separator, shell, positive and negative grade pieces, etc. The positive and negative pieces are stained with binder PVDF. The rotary kiln can be used to remove the separator, binder PVDF and residual electrolyte, etc. to facilitate the subsequent sieving and crushing process.

The rotary kiln is a slightly inclined steel cylinder and lined with refractory bricks. When working, the crushed materials of the battery are fed into the kiln from the front end for burning, and the kiln rotates at a fixed speed to make the materials mix evenly and burn fully. The kiln is rotated at a fixed speed to mix the material evenly



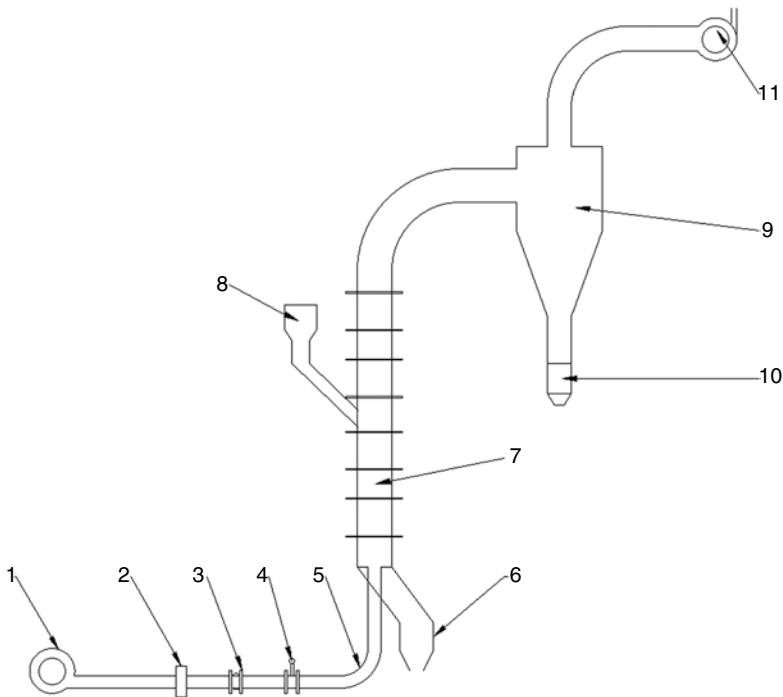


Figure 2.25 Pulsating airflow air separation column. 1 – Active fan; 2 – the valve; 3 – vortex flowmeter; 4 – pulsating valve; 5 – air inlet pipe; 6 – heavy material outlet; 7 – air separation column; 8 – feeding device; 9 – cyclone separator; 10 – light material outlet; 11 – induced draft fan.

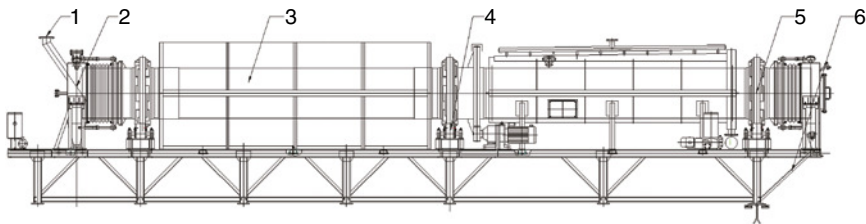


Figure 2.26 Rotary kiln. 1 – Feeding device; 2 – cylinder device; 3 – heating device; 4 – transmission device; 5 – discharge device; 6 – the frame body.

and burn it fully. The rotation should be kept at a proper angle to facilitate the material to slide. After sufficient burning, the material is cooled and discharged from the end of the rotary kiln to the next process. This process is generally carried out in an oxygen-free environment to prevent the oxidation of metals such as aluminum in the battery (Figure 2.26).



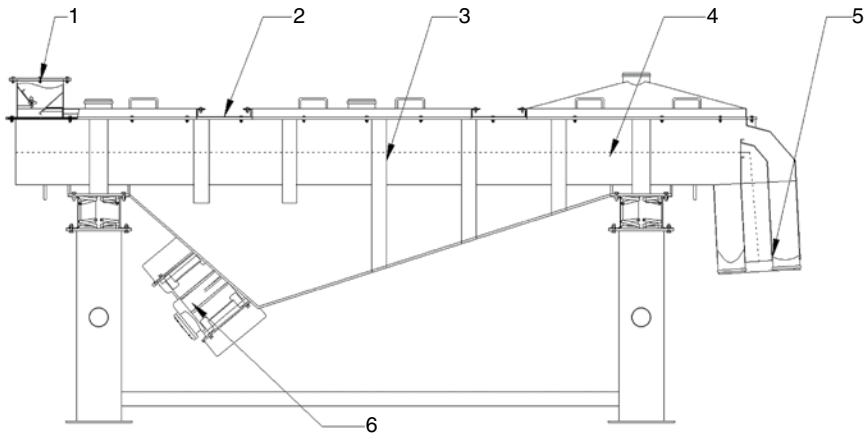


Figure 2.27 Straight line vibrating screen. 1 – Feed inlet; 2 – dust cover; 3 – screen box; 4 – screen; 5 – discharge port; 6 – drive device.

2.7.1.4 Sieving Equipment

Battery crushed materials are generally around 30–50 mesh. The crushed materials should be divided into different size classes to separate copper and aluminum powder from positive and negative powder – commonly used screening equipment for linear vibrating screens and circular vibrating screens.

2.7.1.4.1 Linear Vibrating Screen

Linear vibrating screen uses two eccentric vibrating motors or exciters as the power source. When the two exciters do synchronous, reverse rotation, the eccentric block generated by the excitation force in the direction parallel to the motor axis offsets each other, in the direction perpendicular to the motor axis superimposed as a combined force, so the screen trajectory moves in a straight line. The two motor axes have an inclination angle relative to the screen surface. Under the combined force of the excitation force and the material's gravity, the material is thrown forward in a straight line on the screen surface to achieve the purpose of screening and grading the material (Figure 2.27).

2.7.1.4.2 Round Shaking Sieve

A swing screen is a low-frequency rotary screen that directly imitates manual screening. Its motion trajectory combines radial displacement of this displacement's instantaneous and circular motion as the axis. The eccentric distance of the exciter can be adjusted to produce waste linear three-dimensional motion. The material moves on the screen surface in a way similar to manual screening to achieve the effect of screening materials. Swing screen is especially suitable for precise screening of cylindrical, sheet, and other irregular shape materials (Figure 2.28).



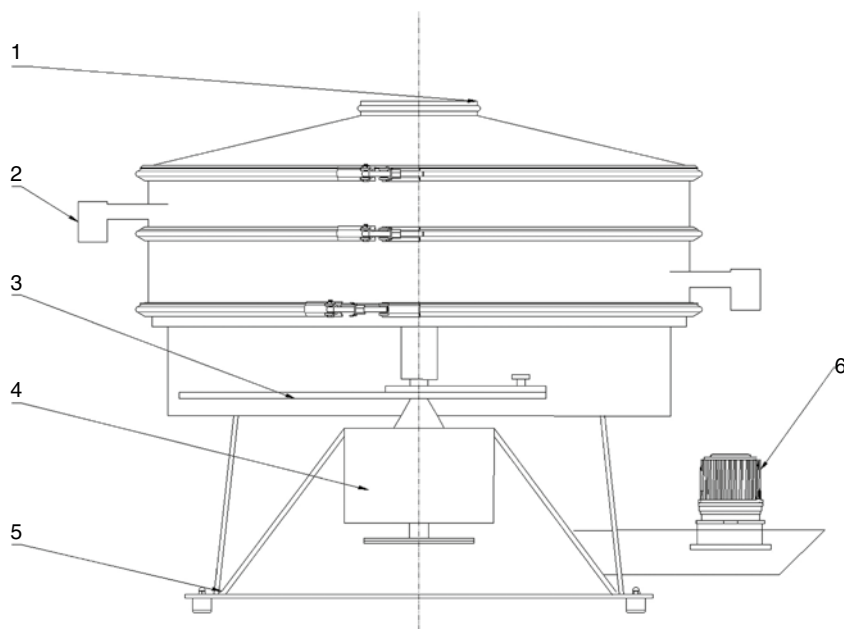


Figure 2.28 Round shaking screen. 1 – Feed inlet; 2 – discharge port; 3 – swing body; 4 – main pivot assembly; 5 – shelf; 6 – drive device.

2.7.2 Hydrometallurgy Equipment

2.7.2.1 Leaching Equipment

2.7.2.1.1 Mechanical Stirring Leaching Tank

Figure 2.29 shows the simple structure of a mechanical stirring leaching tank. The main components are as follows:

The material should have good corrosion resistance to the solution being treated.

Ordinary carbon steel can be used for an alkaline, neutral nonoxidizing medium.

For an acidic medium, enamel can be used. Still, under the condition of high temperature and concentrated hydrochloric acid, especially when the raw material contains fluoride, the service life of enamel is very short. Generally, the steel shell is lined with epoxy resin and then lined with graphite bricks or rubber lining. For HNO_3 medium, NH_4OH – $(\text{NH}_4)_2\text{SO}_4$ medium, stainless steel is available. A concentrated sulfuric acid system can be used in cast iron and carbon steel at room temperature, high-temperature application of ferrosilicon.

Heating system: In addition to the liner graphite or rubber, epoxy resin groove can be used in jacket or screw indirect steam heating. The trough lined with rubber or graphite anvil is generally heated directly by steam.

Stirring system: Mechanical agitator often turbine type, anchor type, screw type, frame type, rake type, and other types. The stirring speed and power are determined according to the size of the tank and the nature of the pretreated pulp.



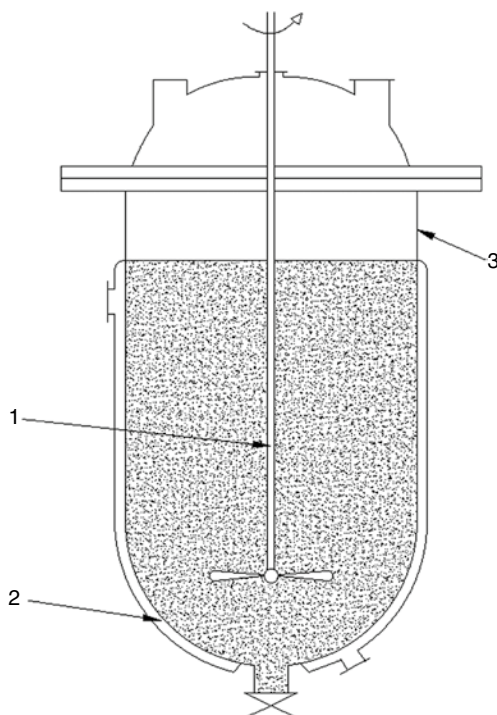


Figure 2.29 Schematic diagram of the mechanical stirring leaching tank structure.
1 – Stirrer; 2 – jacketed; 3 – tub.

2.7.2.1.2 Air Stirring Leaching Tank (Pachuca Tank)

The simple structure of the air stirring leaching tank (Pachuca tank) is shown in Figure 2.30. A central tube with openings at both ends is arranged in the groove, and compressed air is imported into the lower part of the central tube. In the process of bubbles rising along the tube, the pulp is sucked up and raised from the lower part of the tube. The bubbles flowed out from the upper end of the tube, then the liquid will extrude outside the tube. Compared with mechanical stirring leaching, the Pachuca tank is characterized by simple structure, easy maintenance, and operation, conducive to gas–liquid or gas–liquid–solid phase reaction, but its power consumption is about three times that of a mechanical stirring tank. The height–diameter ratio of the Pachuca groove is generally (2–3) : 1, and some are up to 5 : 1.

2.7.2.1.3 Fluidization Leaching Tower

The fluidization leaching tower structure is shown in Figure 2.31. The solid raw material is added into the leaching tower through the feeding port, and the leaching agent solution is continuously entered into the tower by the nozzle. In the tower, due to its linear velocity exceeding the critical velocity, the solid material is fluidized, and the fluidized bed is formed. Due to the good mass and heat transfer conditions between the two phases in the bed, various leaching reactions can be carried out quickly. When the leaching liquid flows to the expanded section, the flow rate



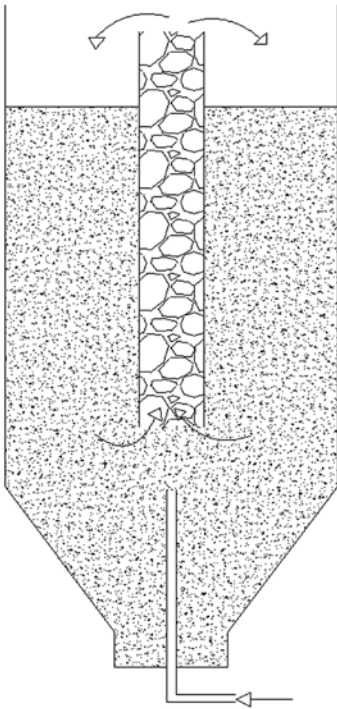


Figure 2.30 Air stirring leaching tank.

decreases below the critical speed, the solid particles settle, and the clear liquid flows out of the overflow port. To ensure the leaching temperature, the tank can be made into a jacket through steam heating and can also be heated by other heating methods.

In the fluidization leaching process, the linear velocity of the liquid phase in the column is an important parameter, and its value varies with the density and particle size of raw materials. The characteristics of fluidization leaching are as follows: the flow of solution in the column is similar to the piston flow, so it is easy to carry out solution conversion and multistage countercurrent leaching. Compared with mechanical stirring leaching, the particle grinding effect is small, so it is beneficial to maintain a certain particle size of the solid product after leaching. The fluidized bed has better mass and heat transfer conditions, so it has a faster reaction speed and a larger production capacity.

2.7.2.1.4 High-Pressure Leaching Kettle

The leaching rate generally increases with the increase of temperature, and some leaching processes need to be carried out above the boiling point of the solution. For some of the leaching processes with gas to participate in the reaction, the increase of pressure of the gas reaction agent is conducive to the leaching process, so it is carried out under high pressure. This leaching process is called high-pressure leaching or pressure leaching. High-pressure leaching is carried out in the autoclave. The working



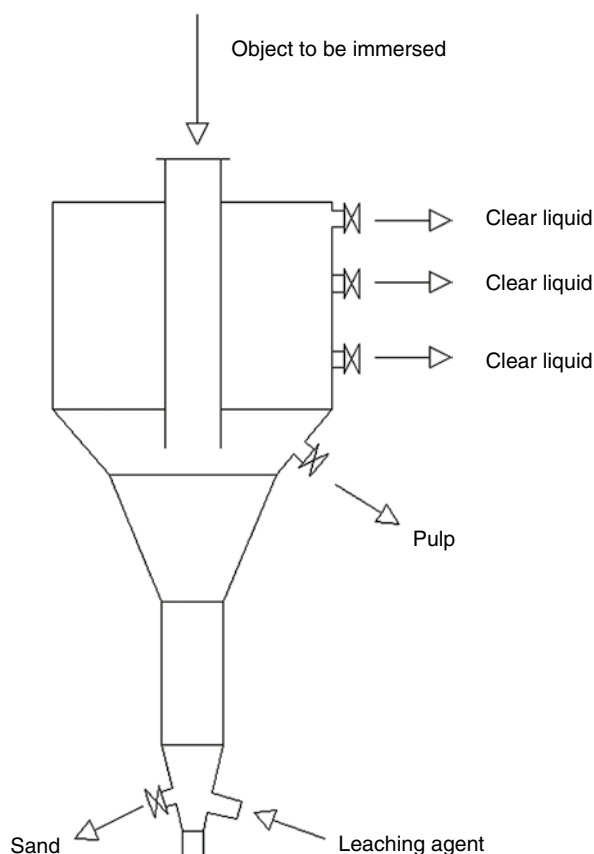


Figure 2.31 Schematic diagram of fluidization leaching tower structure.

principle and structure of the autoclave are similar to that of the mechanical stirring leaching tank, but it should withstand high pressure and be well sealed. It can be attributed to the mechanical stirring leaching if it is from the equipment. There are two types of autoclaves: vertical and horizontal. Figure 2.32 shows the structure of the horizontal autoclave. The material is similar to the mechanical stirring tank previously mentioned. Generally, the leaching tank is divided into several chambers, and the slurry flows continuously through each chamber, and each chamber has a separate agitator.

2.7.2.2 Extraction Equipment

The extraction equipment can realize the perfect separation of the components contained in the material liquid. The structure can be divided into mixing clarifier, extraction tower, and centrifugal extractor.

2.7.2.2.1 Mixed Clarifying Tank

The mixed clarifier is the earliest classical extraction equipment used in industrial production. It can be operated in a single stage or series, or parallel. The agitator is



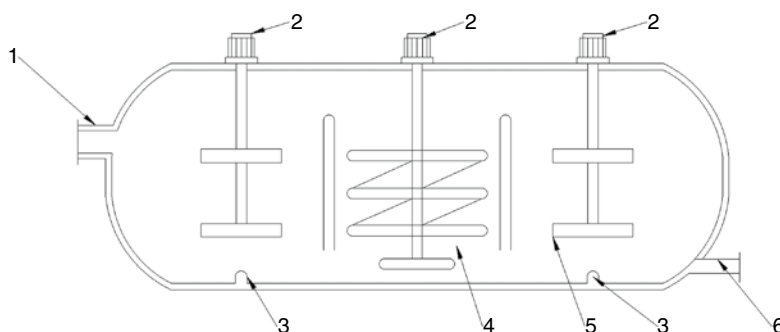


Figure 2.32 Structure of horizontal autoclave. 1 – Feed inlet; 2 – stirrer; 3 – oxygen inlet; 4 – cooling pipe; 5 – stirring paddle; 6 – discharge outlet.

often installed in the mixing tank to increase the contact area of the two phases and strengthen turbulent motion. A pulse or ejector can be used to achieve full mixing of the two phases. The role of the clarification tank is to separate the extractive phase and the raffinate phase, which are close to the equilibrium state. The mixture that is easy to clarify is generally stratified by the density difference between the two phases.

For the material system with a small density difference and interfacial tension between the two phases, a rotating liquid separator or disk centrifuge can be used to accelerate the separation of the two phases. In the operation process, the raw material liquid is first mixed evenly with the extraction agent, and one phase droplet is dispersed in the other continuous phase so that the material and the solvent close contact. To avoid the size of the clarification tank being too large, the dispersed droplets should not be too small, let alone emulsified.

Figure 2.33 shows the structure of a mixed clarification tank without a submersible chamber. Large diameter blades are usually used in the mixing and clarifying tank without a potential chamber. The blades rotate to form eddy currents to promote the oil and water two phases to enter the mixing chamber directly. After stirring and mixing, the fluid enters the mixing phase outlet into the clarification chamber for phase separation. The mixed-phase outlet is arranged far away from the two liquid phase inlets, and the two phases enter the next level of equipment after the completion of phase separation. There is no need to consider the constraints of the submersible chamber on the installation height and speed of the blade in the mixing and clarifying tank. The installation position of the stirring impeller can be selected, and the corresponding stirring speed can be set according to the needs so that the materials can be fully mixed without emulsification caused by excessive stirring. However, the materials in the mixing clarifier without a potential chamber may flow out of the mixing chamber without full mixing, resulting in a short fluid circuit. Meanwhile, the interstage flow capacity of the materials in the equipment is weak, and the processing capacity of the equipment with the same volume is smaller than that of the equipment with a potential chamber [65].



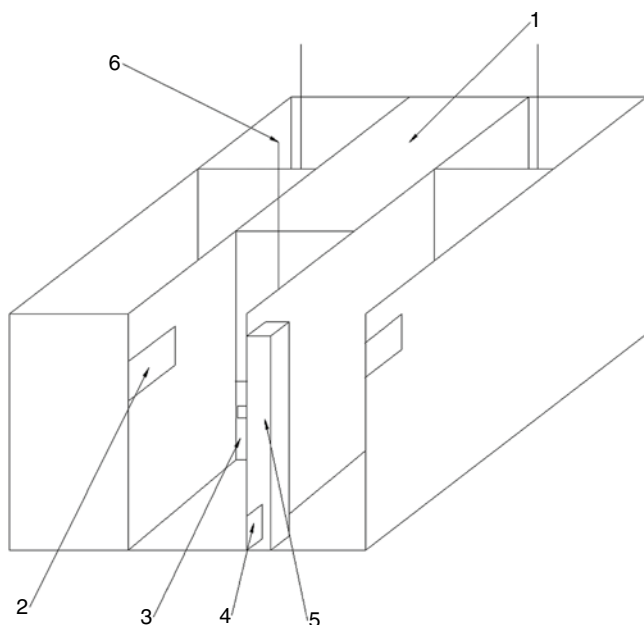


Figure 2.33 Structure drawing of mixed clarification tank without the submersible chamber. 1 – Clarification room; 2 – oil-phase weir; 3 – outlet of mixing chamber; 4 – water phase weir; 5 – water phase weir; 6 – impeller.

The mixed clarification tank has the following characteristics:

- (1) High efficiency: In each stage of the equipment, by adjusting the stirring and clarifying parameters, the extraction efficiency of the extractions can reach more than 90%.
- (2) Strong adaptability: Stable operation and efficient extraction can be achieved when the material's target solute concentration or ratio changes greatly.
- (3) Simple amplification: The volume of the mixed clarification tank can be gradually enlarged from small to cubic meter, and the principle of similar amplification is followed for equipment of different sizes.
- (4) Strong operability: When the fluid in the equipment has liquid flooding or emulsification and other production accidents, it can be solved by stopping and standing and quickly resuming operation after normal recovery.
- (5) Large floor area: The mixed clarification tank is usually operated in multistage series, and the whole extraction process occupies a large area when the extraction stage of the material is large.
- (6) Large material retention: In the multistage series operation mode, it is necessary to add sufficient material and liquid into the tank before starting operation. For the extraction process with large series, the amount of material and liquid in the equipment is huge, and the one-time investment cost of extraction and separation enterprises is high.



Although the mixed clarification tank appeared earlier, it is still widely used in petroleum, chemical, metallurgy, nuclear, and other fields. It is the most commonly used extraction equipment at present. Therefore, researchers at home and abroad continue to develop more efficient, energy-saving, simple forms of mixed clarification tanks to improve the comprehensive performance of the mixed clarification tank.

2.7.2.2.2 Extraction Tower

Extraction tower is also known as extraction column, a chemical industry, petroleum refining, environmental protection, and other industrial sectors commonly used liquid-liquid quality transfer equipment tower. Its internal structure is the use of gravity or mechanical action to break a liquid into droplets dispersed in another continuous liquid, liquid-liquid extraction.

Extraction tower has different structures and types: packed extraction tower, sieve plate extraction tower, rotary plate extraction tower, vibrating screen plate tower, multistage centrifugal extraction tower, and so on. Schematic diagram (Figure 2.34) of the rotary table extraction tower and packed extraction tower is as follows.

In the rotary disk extraction tower, a plurality of disks of the same size and spacing, called the turntable, is in the middle of the rotating shaft, which rotates uniformly

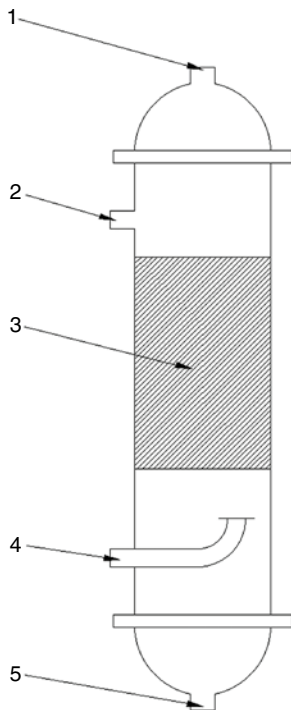


Figure 2.34 Packed extraction tower. 1 – Light liquid inlet; 2 – heavy liquid inlet; 3 – light liquid outlet; 4 – packing; 5 – heavy liquid outlet.

with the rotation of the shaft. The turntable is separated by circular disks of equal size and spacing fixed to the tower wall, known as the fixed plate. Smaller density of a solution from the bottom of the tower straight into the tower, due to the effect of buoyancy, flow to the top of the tower. In the process of upward flow, the smaller density solution was broken by continuous rotation of rotary centrifugation and scattered into droplets. Denser solvent straight into the top of the tower, due to the effect of gravity, flowing down the tower, and filled with the tower. The droplets dispersed in continuous solvent, through the contact between the droplet and the solvent, one or more components of the solution are selectively dissolved in a continuous solvent for mass transfer.

The solution crushing in the packed extraction column is through the action of the filler. The number, spacing, and other structures of the turntable, the size, and the height of the filler are calculated according to the performance of the material and the degree and purity of separation and other factors.

2.7.2.2.3 Centrifugal Extractor

Centrifugal extraction machine is a new, rapid, efficient liquid–liquid mixing separation equipment, high-speed rotating drum driven by centrifugal extraction machine using a motor; density is different. Each of two immiscible liquids in the drum or blades rotates under the action of shear force produced by the complete mixing and mass transfer and in the high-speed rotating drum under the action of centrifugal force produced by the rapid separation. So centrifugal extraction machine is widely used in fine chemical, pharmaceutical, hydrometallurgy, petrochemical, environmental protection, plant extraction, and other industries (Figure 2.35).

Centrifugal extractor mainly includes two processes, namely, mixing mass transfer and centrifugal separation. The centrifugal extractor can automatically and continuously complete the mixing and separation processes.

Mixed Mass Transfer After entering the centrifugal extractor, the water and organic phases are cut and dispersed into tiny droplets by the high-speed rotating drum or blade. The two phases fully contact to achieve the purpose of mass transfer.

Factors affecting mass transfer effect include the following:

- (1) **Mixing strength:** Within a certain range, the larger the mixing strength is, the smaller the dispersed particles of the two phases are, and the larger the contact area is, the more conducive to mass transfer, but the small droplet is not conducive to separation. The mixing strength can be increased by increasing the rotational speed or replacing the blade with a strong shear force.
- (2) **Contact time:** Increasing two-phase contact time is conducive to mass transfer. When the mass transfer process reaches equilibrium, the increase of contact time will not improve the mass transfer effect.



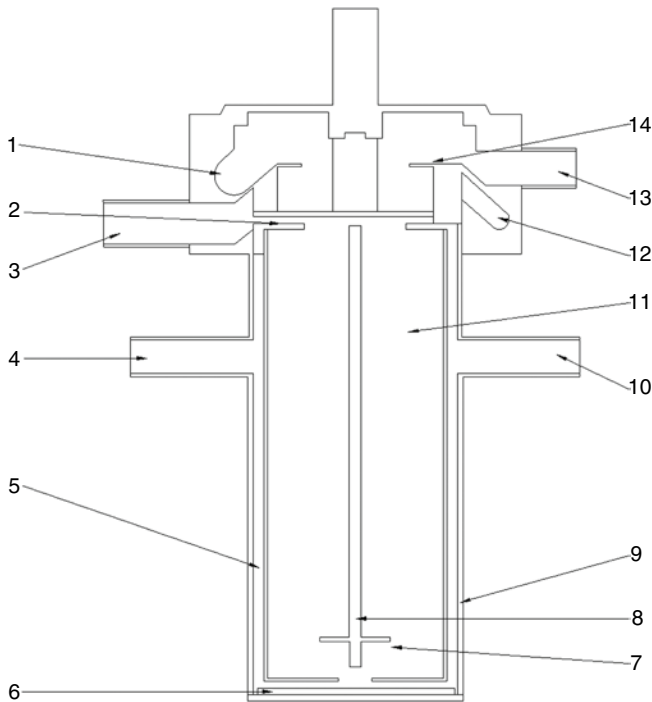


Figure 2.35 Schematic diagram of centrifugal extractor structure. 1 – Collection cavity; 2 – light phase weir plate; 3 – light phase inlet; 4 – light phase or mixed phase inlet; 5 – ring mixing zone; 6 – bottom impeller; 7 – dispersion plate; 8 – blade; 9 – shell; 10 – double or mixed phase inlet; 11 – separation zone; 12 – collection cavity; 13 – rephase outlet; 14 – adjustable rephase weir plate.

- (3) Temperature: The temperature will affect the mass transfer effect. The mass transfer effect of some systems will improve with the increase of temperature, and the mass transfer effect of some systems will decrease with the increase of temperature.
- (4) Material concentration is poor: The greater the difference in material concentration, the more conducive to mass transfer.

Centrifugal Separation Under the drive of the drum and its spoke plate, the mixed liquid and the drum rotate synchronously at high speed to produce centrifugal force. Under centrifugal force, the liquid with a higher density gradually moves away from the drum's center and toward the wall of the drum. The less dense liquid gradually moves away from the drum wall toward the center. Finally, the two-phase liquid is thrown into the collection chamber through the respective channel. The two phases flow out from the respective collection chamber, thus completing the two-phase separation process.



Factors affecting separation are as follows:

- (1) Speed: The faster the speed, the faster the separation of the two phases in the drum, the less entrainment of the two phases. At the same time, increasing the speed can also improve the processing capacity of the equipment.
- (2) Drum height: The mixture is slowly separated from the bottom to the top in the drum. The higher the drum, the better the separation effect.
- (3) Weir plate: The centrifugal extractor controls the outflow of the two-phase liquid through the adjustment of the wear plate on the top. The appropriate weir plate is an important guarantee for the complete separation of the two phases.
- (4) Material characteristics: The physical characteristics of the material itself, such as emulsification, foaming, density difference size, have a great impact on the two-phase separation.
- (5) Maximum separation amount of centrifugal extractor:

$$Q = 1.386 \times 10^{-4} \omega D_i^2 L$$

where ω is the speed of centrifugal extractor, D_i is the inner diameter of the drum, and L is the height of the drum.

2.7.2.3 Solid-Liquid Separation Equipment

The hydrometallurgy process is essentially a gradual separation of valuable metals in materials. The products are generally a mixture of solid and liquid, such as mineral raw materials (or secondary materials produced by metallurgy) through leaching treatment; the product is a mixture of solid and liquid – pulp. The pulp must be separated to achieve the final purpose of the process, even if the impurities are separated from the main metal. As its name implies, solid-liquid separation refers to the separation of the solid and liquid phases from a mixture. Solid-liquid separation is used in several processes to achieve the following objectives:

- (1) Recycle useful solid (waste liquid)
- (2) Recycle liquid (waste solid)
- (3) Recycle solid and liquid

There are many methods for solid-liquid separation in the actual production process, but according to their principles, they can be divided into two categories: concentration and filtration.

2.7.2.3.1 Enrichment Facilities

The main equipment for thickening is the thickening tank, also known as a thickener. The thickening tank is industrial equipment that can completely improve the concentration of mud and get the clarifying liquid by the sedimentation process. It is composed of a tank, rake arm, transmission device, lifting device, and other components. The concentrator is divided into the central transmission and peripheral according to the different transmission modes. The concentrator with a large diameter adopts peripheral transmission mode. According to the shape of the tank, the



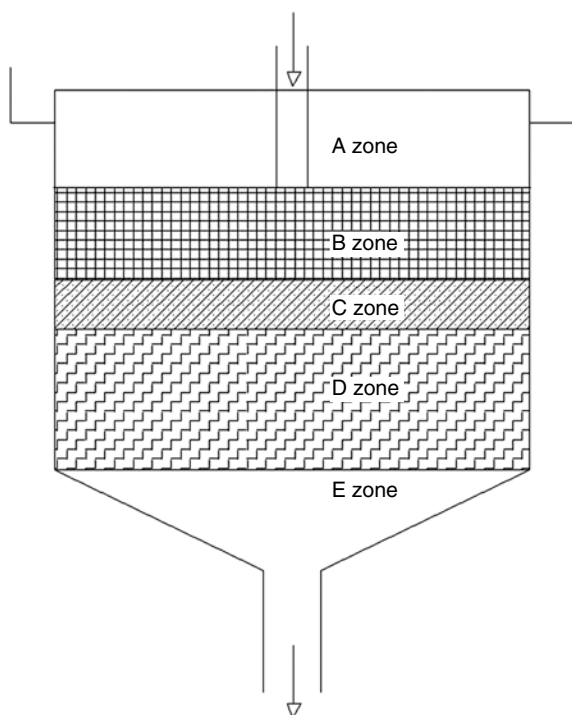


Figure 2.36 Process diagram of thickening tank. A – supernatant region; B – supernatant clarification area; C – mixed clarification region; D – mud clarification area; E – thick mud area.

concentration tank is divided into two: cone bottom and inclined bottom; the production process is the most used cone bottom concentration tank. The following describes the cone bottom enrichment tank for the center drive, as shown in Figure 2.36:

- (1) Tank body: The upper part of the concentration tank is cylindrical, with a common diameter of 10–18 m and a height of 3–7 m. The bottom of the tank is conical, and the cone angle is 160° , forming a funnel. Such a bottom can make the settled solid material move to the middle, and the thick mud is discharged from the bottom hole of the cone. The tank is made of reinforced concrete and lined with corrosion-resistant materials such as lead skin, antimony aluminum plate, epoxy glass cloth, and steel plate lined with corrosion-resistant materials.

A buffer cylinder is hung in the center of the tank, with a sieve plate at the bottom. The cylinder has a 1.5 m diameter and a height of 1.5 m, which is rolled from stainless steel plate. When the buffer cylinder is installed, the upper mouth should be higher than the liquid level. The cylinder separates the slurry to be concentrated into the concentration tank from the supernatant area to ensure the quality of the supernatant, and the sieve plate plays a buffer role. The leached pulp is fed into the feeding cylinder without mixing the clarification liquid. The clear



supernatant is released through a flow tank located at the upper edge of the concentration tank, and the concentrated mud in the middle is pumped out by a sand pump or otherwise discharged.

- (2) Rake arm: The thickening tank is equipped with a special mechanism consisting of a cross rake arm with rake teeth to stir the settling particles at the bottom of the tank to move the settling particles towards the center.
- (3) Transmission device: To ensure the movement of the whole cross rake arm with rake teeth, a set of the transmission devices is provided on the surface of the concentrator slot, which consists of a motor, a gear reducer, a worm gear reducer, and other parts. The central shaft is installed on the central hole of the worm wheel through the sliding key. The worm reducer is then driven to drive the central shaft when the motor drives the gear reducer. According to the diameter of the tank, the rotation speed of the central shaft can be controlled within the range of about one turn in 10 minutes.
- (4) Lifting device: A spiral lifting device is arranged on the top of the concentration tank, through which the central shaft and rake rod are lifted, to adjust the load and overhaul and maintenance of equipment.

2.7.2.3.2 Filtering Equipment

The basic principle of filtration is to use a porous material as a medium, creating a pressure difference between the two sides of the medium, creating a driving force so that the liquid through the small channels and suspended solids are trapped in the medium. Media types include woven fabrics, porous ceramics, porous metals, paper pulp, and asbestos. According to the pressure difference on both sides of the filter medium, the filter is divided into pressure filter (positive pressure) and vacuum filter (negative pressure).

Plate and Frame Filter Press Plate frame filter press is one of the most widely used intermittent filters. The general plate and frame filter press are composed of multiple filter plates, filter cloth, and filter frames arranged alternately. The filter plate, cloth, and frame used in each filter are arranged alternately, and then the head screw is turned to make the plate and frame tightly jointed. During the operation, the raw material liquid enters the filter frame through the hole on the filter frame under pressure, as shown in Figure 2.37. Under the action of pressure, the filtrate passes through the filter cloth attached to the filter plate and is discharged from the hole on the plate along the channel, and the resulting filtrate remains in the frame to form the filter cake. When the filter frame is filled with filter residue, relax the head screw, take out the filter frame, remove the filter cake, wash the filter frame and the filter cloth, and reinstall.

If the cake needs to be washed, the filter plate needs to have two kinds of structure. One kind of plate has a washing liquid inlet, called the washing plate; another has no washing liquid imports, called non-washing plate. Washing is carried out at the end of filtration, that is, when the filter frame is full of filter cake, the feed valve is closed, and the filtrate discharge valve under the washing plate is closed. Then



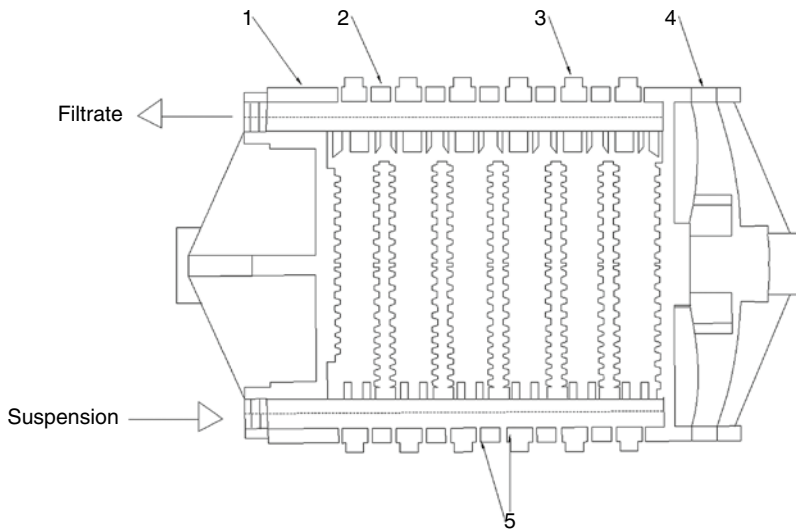


Figure 2.37 Structure of plate and frame filter press. 1 – Fixed head; 2 – plate; 3 – box; 4 – movable head; 5 – filter cloth.

the washing liquid is sent into the water under a certain pressure. The washing liquid enters from the washing plate and passes through the filter cloth, and the filter frame flows down to the outlet along with the opposite filter plate and is discharged.

Box Filter Press Figure 2.38 shows a box filter press. The ribbed surface of the filter plate is concave inward instead of the filter frame so that a separate filter box is formed between the adjacent filter plates. Figure 2.38a shows the opening condition, and Figure 2.38b shows the press drying condition of the filter cake.

Feed channels are usually different from plate and frame filter presses. The filter box is connected by a fairly large hole in the center of each plate, and the filter cloth is held in place by a screw joint with holes in the filter plate.

To press the cake dry, plastic bags that can be expanded are sandwiched between two filter plates. The cake is squeezed into expandable plastic bags when the filtration is complete to reduce the liquid content.

2.8 Global Industrial Participants and Their IP Layout

2.8.1 Introduction of Main LIB Recyclers

The major LIB recyclers are listed in Table 2.9. Most of them use the pyrometallurgical method for recycling since the general pyrometallurgical process is more adaptive for the inputs when compared with the hydrometallurgical process. However,



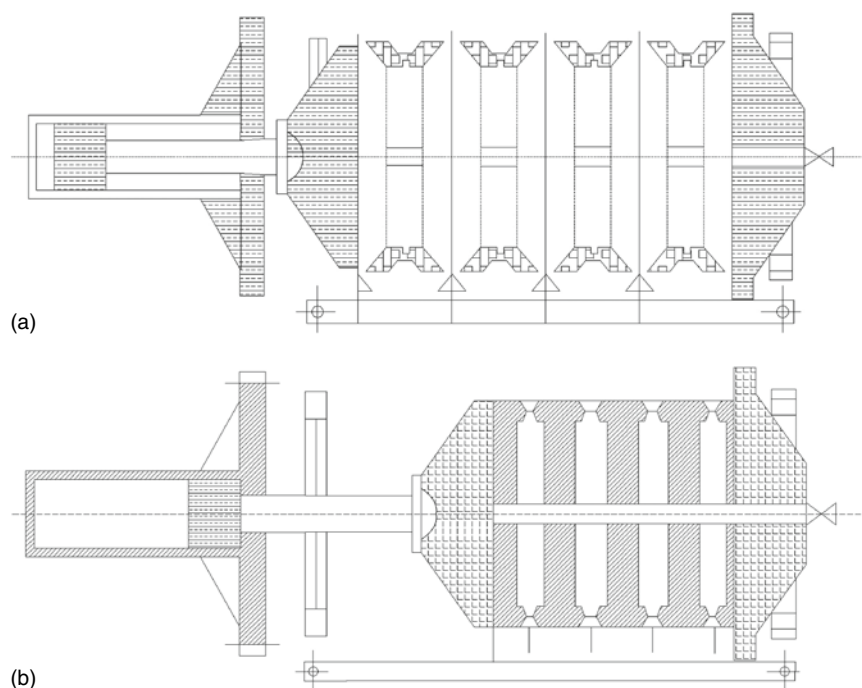


Figure 2.38 Schematic diagram of opening compression of box filter press. (a) Conditions of opening; (b) press dried filter cake.

this scenario is likely to change since the pyrometallurgical process is energy-intensive and will lead to high CO_2 emissions. It can be proven that the newly planned production line or built factories are mainly focused on the hydrometallurgical processes. The detailed recycling process route can be found in Chapter 3.

2.8.2 IP Layout and Development Trend Analysis

2.8.2.1 Patent Development Trend Analysis

Trend analysis reveals the trend of the number of patents applied by patent applicants each year to reflect the application time and technological development.

Figure 2.39 shows the change curve of patent applications (item) of decommissioned battery recovery technology over time since 2001. As shown from Figure 2.39, the number of annual applications reached 1065 in 2018 and 1053 in 2019. In 2013, the number of annual applications reached 419, one of the peaks because some applicant made 166 abnormal applications in 2013. If the influence is excluded, it can be found that from 2001 to now, the average annual growth rate of the number of patents for retired battery recycling technology has maintained about 30%, and the number of applications has been on the rise.

Further, after excluding the interference of 2013 data, the patent technology life cycle diagram is formed as shown in Figure 2.40 by analyzing the annual number of patents and the number of applicants. The abscissa is the number of patents, the



Table 2.9 List of global main LIB recycler.

Company	Main products	Technology	Location
Botree Cycling	Nickel/cobalt/manganese hydroxides, Li_2CO_3	Hydro-dominant	China
Brunp Recycling	Nickel/cobalt/manganese hydroxides, lithium salt	Hydro-dominant	China
Huayou Cobalt	Nickel/cobalt sulfate, Li_3PO_3	Hydro-dominant	China
GEM	Nickel/cobalt/manganese hydroxides	Hydro-dominant	China
ShunHua Lithium	Li_2CO_3 , FePO_4	Hydro-dominant	China
Ganpower	NiSO_4 , CoSO_4 , Li_2CO_3	Hydro-dominant	China
Qiantai Technology	Battery powder, Cu and Al metal	Physical	China
SDM	Shell, electrolyte, separator, cathode powder, anode powder	Physical	China
Umicore	Ni-Co alloy, NiCO_3 , NiSO_4 , CoCO_3 , CoSO_4	Pyro + hydro	Belgium
TES (Recupyl)	Li_2CO_3 , Co(OH)_3	Hydro-dominant	France
SNAM	NR	Pyro-dominant	France
ERAMET	Raw materials for special steel	Pyro-dominant	France
Accurec GmbH	Co alloy, Li_2CO_3	Pyro-dominant	Germany
BHS-Sonthofen GmbH	Battery powder, Cu and Al metal	Physical	Germany
Batrec AG	Battery scraps	Pyro-dominant	Switzerland
Glencore plc. (Xstrata)	Co alloy	Pyro-dominant	Switzerland
AkkuSer Oy	Metal powder	Pyro-dominant	Finland
Fortum	Co, Ni, Mn salt	Hydro-dominant	Finland
Li-Cycle	NiSO_4 , CoSO_4 , Li_2CO_3 , MnCO_3	Hydro-dominant	Canada
Inmetco	Co alloy	Pyro-dominant	USA
Retriev	Li_2CO_3 , mixed metal oxides	Hydro-dominant	USA, Canada

(continued)



Table 2.9 (Continued)

Company	Main products	Technology	Location
American Manganese	Ni cobalt hydroxide, Li_2CO_3	Hydro-dominant	USA
Redwood Materials	NiSO_4 , CoSO_4 , MnSO_4	Hydro-dominant	USA
Ascend Elements	NiSO_4 , CoSO_4 , MnSO_4	Hydro-dominant	USA
Nippon Recycle Center	Raw materials for special steel	Pyro-dominant	Japan
Sumitomo Metal Mining	Co alloy, Co metal	Pyro-dominant	Japan
GS Engineering & Construction Corp.	Ni, Co, Li, and Mn metal	Pyro-dominant	South Korea

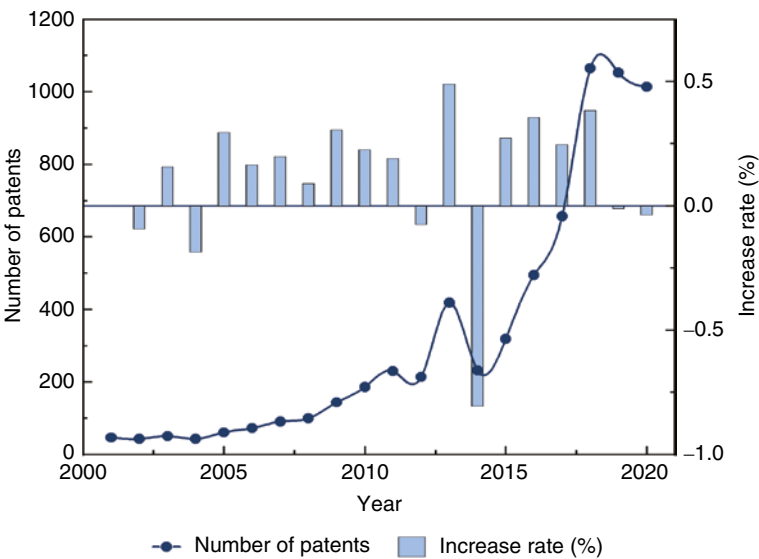


Figure 2.39 Global patent application trend of retired battery recovery technology.

ordinate is the number of applicants, and the data point is the year of application. So far, the number of applicants for related technologies has been growing gently, with research and development mainly concentrated on a small number of applicants. The number of patents has been increasing rapidly. The matched comparison chart (dotted line) shows that the patent technology has a high concentration embryonic stage. According to the increasing number of retired batteries in recent years, the theoretical number of retired power lithium-ion batteries will reach 94.48 GWh by 2025, with a compound annual growth rate of 47.16% [66]. In addition,



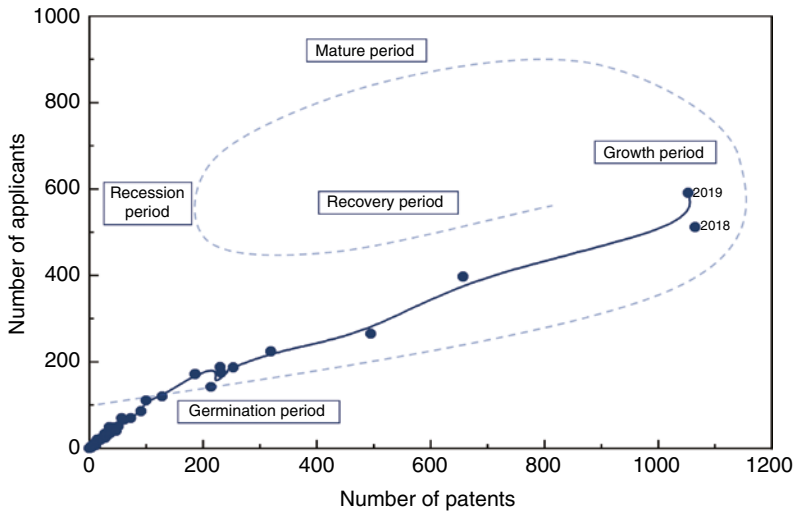


Figure 2.40 Patented technology life cycle diagram of retired battery recovery technology.

countries attach importance to power lithium-ion batteries and recycling technology, which is expected to enter a growth stage in recent years. In general, the future development prospect of decommissioned battery recovery technology is good, and more basic technology patents can be reserved for technological research and development at the present stage.

2.8.2.2 Patent Area Analysis

Based on the regional analysis, among the clearly advantaged countries, China's overall competitive situation is good. By market region, 56% of patents were distributed in China, followed by Japan (10%) and the United States (6%). In terms of technology origin, 55% of patents were filed by Chinese applicants, followed by Japan (15%) and the United States (5%).

They were combined with the market area and source technology, in the analysis of patent technology market to the layout, as shown in Figure 2.41. Compared with the technology market layout of China, the United States, Japan, Korea and Europe, applicants from various countries mainly home layout. Among them, there are more patent layouts for Chinese applicants, but most of them are concentrated in their own countries, with only 4 layouts in Europe, 13 layouts in Japan. However, the global layout of Japanese applicants is more reasonable. Although the total patents number of Japanese applicants is not as good as that of Chinese applicants' based on 808 domestic layouts, it has 129 pieces in the United States, 101 in China and 80 in Europe. It can be seen that, relatively speaking, foreign applicants have a global vision than Chinese applicants, and will become the main competitors of Chinese applicants' international layout, and related technologies may also become technical barriers for Chinese applicants to open up international markets. In the future, Chinese patentees should actively deploy in Japan, South Korea, Europe, the United States and other regions to strengthen the patent layout in the global market.



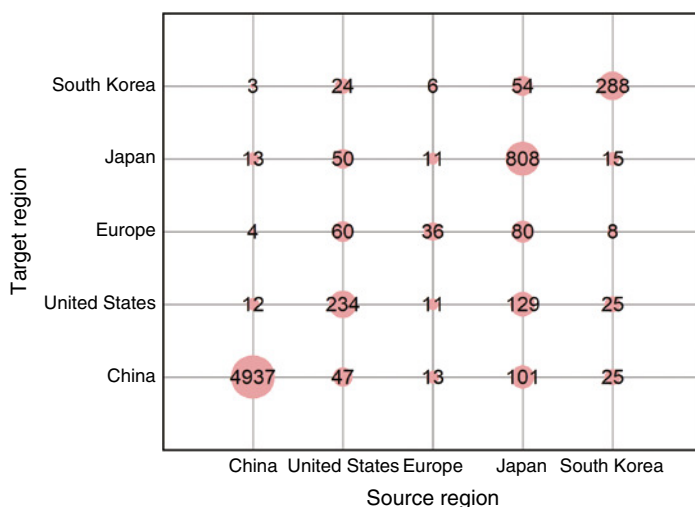


Figure 2.41 Market layout and direction of decommissioned battery recycling technology.

2.8.2.3 Analysis of Major Patent Applicants

As shown in Figure 2.42, the ranking of applicants according to the number of patents of the applicant (patentee) reflects the research status and technical level of different applicants on decommissioned battery recovery technology. It reflects the distribution of competitors, scientific research level, and technical expertise in the technical field.

The patent applicants who can find more innovation achievements are Sumitomo Metal, Central South University, BRUNP, GEM, GOTION, Institute of Process Sciences of Chinese Academy of Sciences, XJ Metal, etc. In addition, according to the countries and types of applicants, Sumitomo Metal, a Japanese company, ranked first with 157 patent applications, followed by XJ Metal and Toyota. Chinese universities

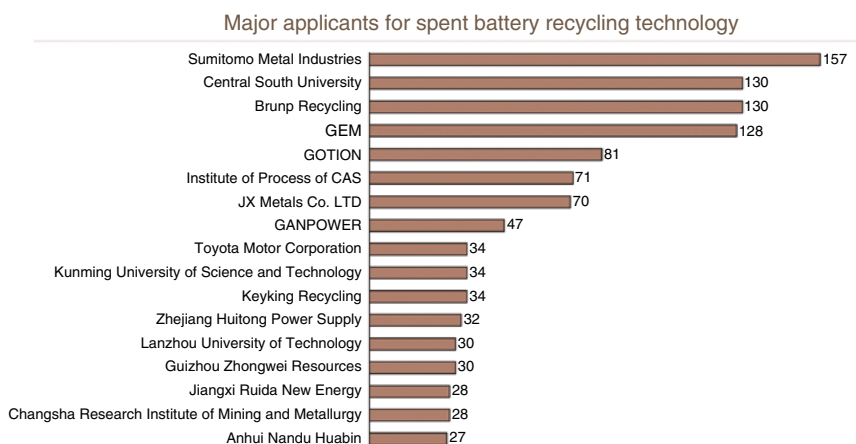


Figure 2.42 Ranking of major applicants for decommissioned battery recovery technology.



and research institutes include Central South University, Institute of Process Sciences, Kunming University of Science and Technology, Lanzhou University of Technology, etc. Chinese enterprises have BRUNP, GEM, GOTION, HIGHPOWER, etc. On the whole, Chinese enterprises, universities, and research institutes have strong strengths. Chinese universities and companies are in both the first and second tier of the ranking, indicating that Chinese R&D entities in retired battery recycling technology are evenly distributed. Chinese universities, institutes, and enterprises have theoretical research and practical application research, and the echelon of continuous research and development capacity is strong. However, there is still a gap between Chinese companies and Sumitomo Metal regarding the number of applications.

2.8.2.4 Branch Analysis of Patented Technology

By analyzing the patents of five technical branches of decommissioned battery recovery technology, such as discharge, disassembly, leaching, metal separation and extraction, and precursor synthesis, the innovation heat of each branch of decommissioned battery recovery technology was understood.

Figure 2.43 shows that the patent proportion and application trend of branch technology of decommissioning battery recycling are mainly dismantlement and metal separation and extraction technologies. The number of applications for dismantlement and metal separation and extraction technologies is 3374, accounting

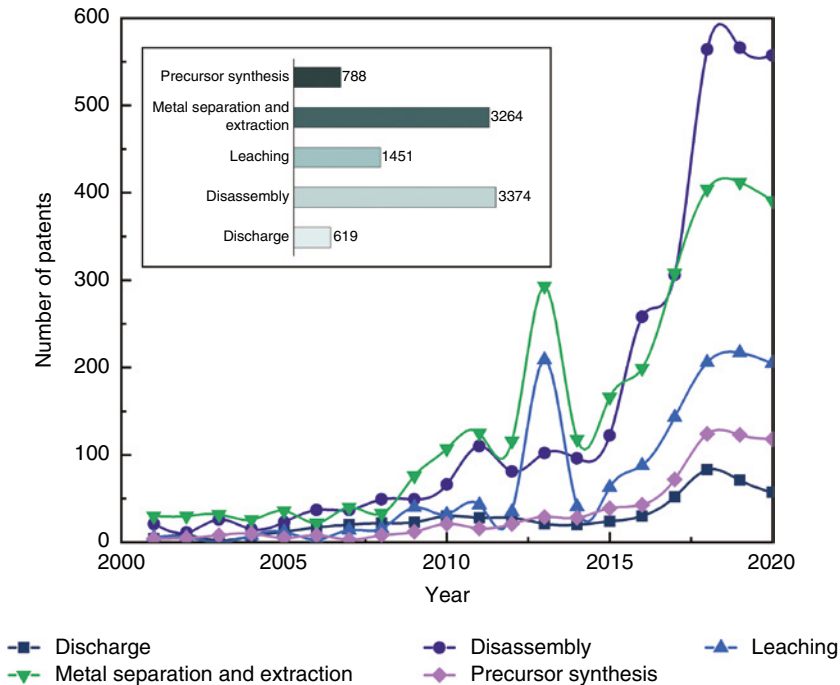


Figure 2.43 Patent application trend of decommissioned battery recovery branch technology.



for 47.12%, and the number of applications for metal separation and extraction technologies is 3264, accounting for 45.59%. This proportion is followed by leaching, precursor synthesis, and discharge technology. In terms of trend, except for Sichuan Normal University in 2013, the patents of the five technical branches showed an upward trend. Before 2008, the number of the five technical branches was flat. Since 2009, patent applications for disassembly and metal separation and extraction technology have increased rapidly. Since 2014, the number of patent applications for discharge, leaching, and precursor synthesis technology has increased significantly. From 2009 to 2015, the number of patent applications for metal separation and extraction was larger than that for disassembly patents year by year, while from 2016 to 2019, the number of patent applications for metal separation and extraction was larger than that for other technical branches in recent years. Due to the variety and specifications of the battery, the battery is composed of shell, positive electrode, negative electrode, electrolyte, diaphragm, etc., which is difficult to disassemble. To reduce environmental pollution and improve the comprehensive utilization level of renewable resources, dismantling is the basis of decommissioned battery recycling technology. And the dismantlement equipment is highly integrated, which was a key technology before the industrialization of decommissioned battery recycling.

Figure 2.44 shows the market layout of decommissioned battery recycling branch technology. The number of patents in the Chinese market is larger than in other countries in all technical branches, especially in dismantling and metal separation and extraction technology. The various technical branch ratios show that the Chinese

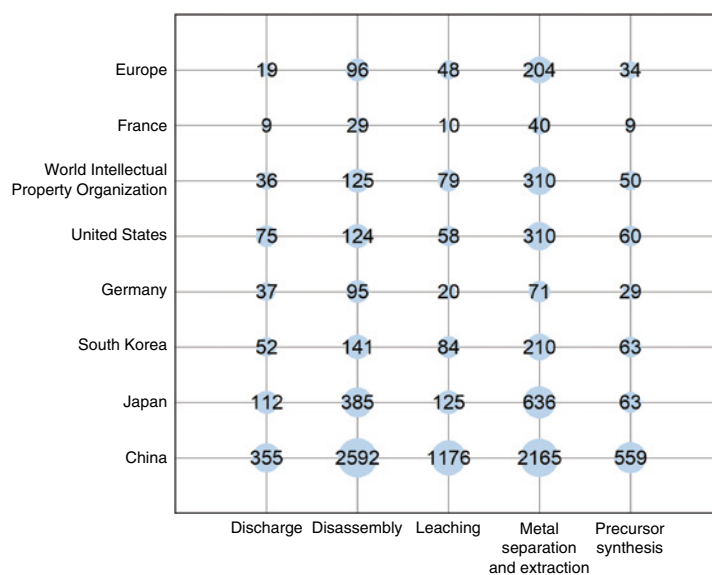


Figure 2.44 Market layout of decommissioned battery recycling branch technologies.



market and dismantling technology are greater than the metal extraction technology. In other countries and regions, such as Japan, the United States, South Korea, and the European Union, the world intellectual property organization, usually metal separation technology patent quantity is greater than the dismantling; branch of China is a retired battery recycling technology of main countries and market innovation. In recent years, the rapid development of the main technology focused on recycling technology, hot technology, disassembly, and metal separation and extraction, and constantly research and development to solve the problem of disassembly. At the same time, other countries and regions of metal separation and extraction technology belong to the main direction of research and development to improve metal recovery.

From the perspective of applicants in each technology branch, although the number of regional patents in China is far greater than that of other countries and regions in each technology field, however, Chinese applicants do not have absolute advantages in the main R&D subjects in each technology field. In the discharge technology, Toyota ranked the first; in dismantling technology, major Chinese enterprises such as GEM, BRUNP, and GOTION are in a leading position, while Japan's XJ Metal and Sumitomo Metal are in a leading position and also have a certain number of patents. In terms of leaching technology, China mainly concentrates on central South University, Institute of Process Sciences, and other universities and institutes, followed by BRUNP, XJ Metal, and Sumitomo Metal of Japan. Sumitomo Metal has an absolute advantage in metal separation and extraction technology, followed by Central South University and XJ Metal, and Chinese enterprises BRUNP and GEM also have a certain amount of patent layout. Chinese universities and institutes also have a certain patent layout in this field. In precursor synthesis, it is mainly Chinese enterprises and colleges and universities (Figure 2.45).

2.8.2.5 Patent Branch Analysis of Metal Separation and Extraction

By analyzing different battery types, metal separation and extraction can be divided into NCM battery, LFP battery, and lead-acid battery. According to the extraction of valuable metals, NCM batteries also include lithium cobalt acid batteries and lithium manganese acid batteries, including cobalt, manganese, nickel, and other metal batteries.

As shown in Figure 2.46, in terms of metal separation and extraction technology, NCM battery is the main component, accounting for 2825 items, accounting for 73.02%, followed by LFP battery and lead-acid battery. As the main lithium-ion power battery of existing EVs, it is also the main battery of battery retirement tide. Global technology is mainly focused on how to recycle lithium batteries. LFP battery life as the main bus, coach bus power lithium-ion batteries, is about eight years, and in the next few years, it will be retired explosive power lithium-ion batteries, the recovery value. Though less NCM batteries, the main value of lithium recovery, using high temperature to repair all components recycling technology, can realize high lithium, iron, and phosphorus recovery [67].



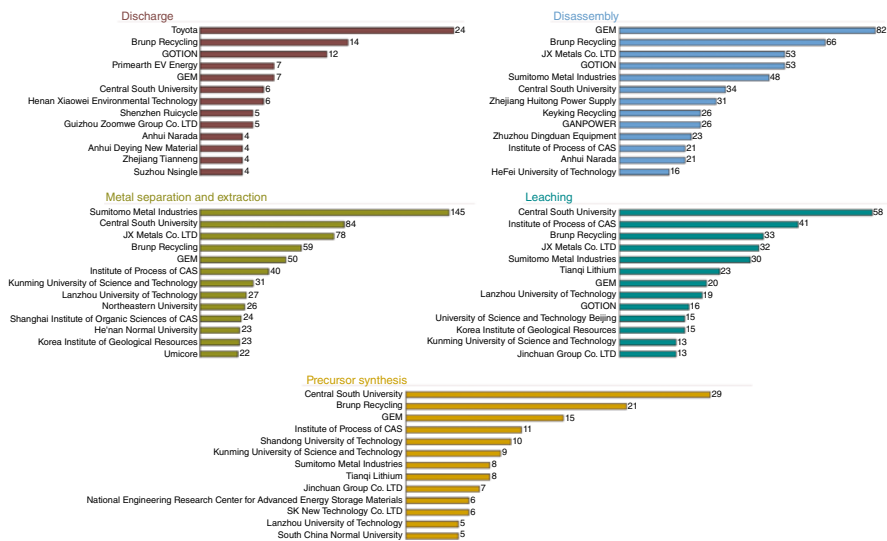


Figure 2.45 Applicants for the decommissioned battery recycling branch technology.



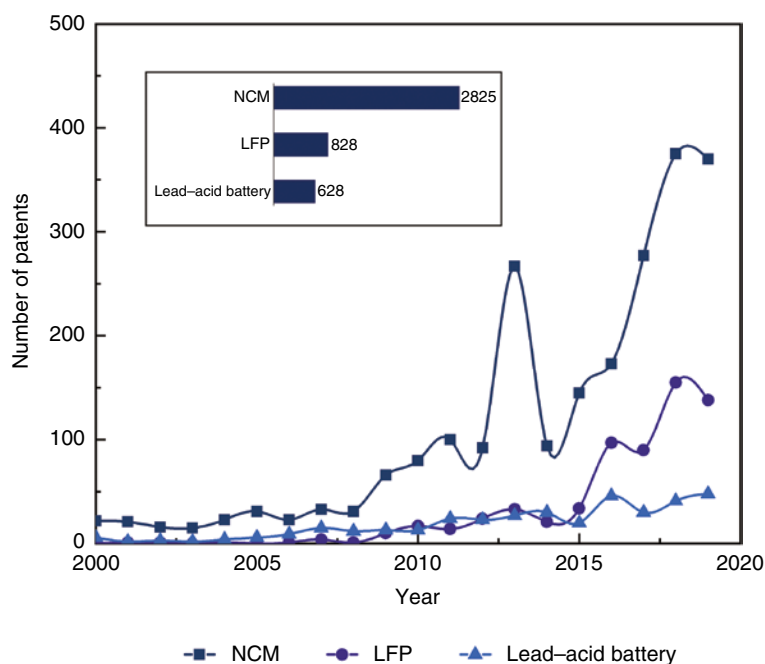


Figure 2.46 Trend of a patent application for a technical branch of metal separation and extraction.

2.9 Conclusion

In this chapter, through the patent retrieval and analysis method, on the battery recycling technology research, the results showed that (i) the existing battery recycling technology at the present stage is in the bud; high concentration, in recent years, is expected; overall, retired battery recycling technology has good prospects for future development, can research and develop the technology at the present stage, and can reserve more basic technology patents. (ii) The Chinese market and Chinese applicants are the main countries and applicants, respectively, of retired battery recycling technology, but Chinese applicants have a less international layout, and foreign applicants have more global vision, so there may be technical barriers to expand the international market in the future. In the future, Chinese patentees should actively distribute in Japan, South Korea, Europe, and the United States to strengthen their patent distribution in the global market. (iii) Chinese universities, research institutes, and enterprises have many patent applications and a balanced distribution of R&D subjects. They have theoretical research and practical application research and have a strong continuous R&D capability. However, there is still a gap between Chinese enterprises and Sumitomo Metal regarding the number of applications. (iv) In terms of the technical branch, dismantling technology has been the main application direction in recent years. The number of patents on metal separation and extraction technology in the Chinese market is more than that of metal separation and extraction technology. In comparison, the number of metal



separation and extraction technology patents in other countries and regions is more than that of metal separation and extraction technology. In discharge technology, Toyota ranked first, metal separation and extraction technology. Sumitomo Metal has an absolute advantage; Chinese enterprises still need to strengthen the patent layout of discharge and metal separation and extraction technology. (v) The metal separation and extraction technology subdivided mainly focuses on NCM battery technology. The patent layout of NCM and LFP batteries will be the main few years.

According to the industry's current situation, Chinese enterprises, universities, and institutes should further promote decommissioned battery recycling technology, strengthening the combination of industry, university, and research, at the same time, expand the international layout and the improvement of technical branches, promote the sustainable and healthy development of China's decommissioned lithium-ion power battery recycling technology, implement the "dual carbon" strategy, and speed up the energy transformation.

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3

Typical Cases in Industry

3.1 China

Lithium-ion battery (LIB) recycling industry started early in China, and hundreds of enterprises have been set up or planned to be set up; their recycling processes are mainly based on wet processes. Here, nine representative companies in China, including Btree Cycling, Brunp Recycling, Huayou Cobalt, GEM, Shunhua Lithium, Ganpower, Qiantai Tech, Hunan Jiangye, and Dingduan Equipment, are selected and introduced in detail.

3.1.1 Btree Cycling

Btree Cycling dedicates itself to providing a complete solution for rare-metal extraction and high-end metal materials recycling. By using innovative separation systems, the short-range closed-loop design of the process flow, and the development of intelligent equipment, Btree Cycling enables recycling enterprises, electric vehicle (EV) enterprises, battery manufacturers, and other entities responsible for recycling to realize a short-range, closed-loop, and sustainable recycling process of battery materials. The schematic illustration of this process is presented in Figure 3.1.

After discharging, the spent lithium power battery is lifted to the crushing section by the elevator. After single-stage coarse crushing, the crushed materials can be pyrolyzed in the pyrolysis furnace or cleaned and dried by an organic solvent, or directly dried to remove the organic components in the battery. After removing the organic components, the crude materials are treated by a magnetic separator to remove the iron sheet/shell. Then, the electrode powders and copper aluminum foils are separated by secondary crushing, vibration screen, and vibration sieving. After dust removal, secondary combustion, and cooling, the waste gas generated in the pretreatment process (crushing, pyrolysis, and sieving) goes to the waste gas treatment system. The electrode



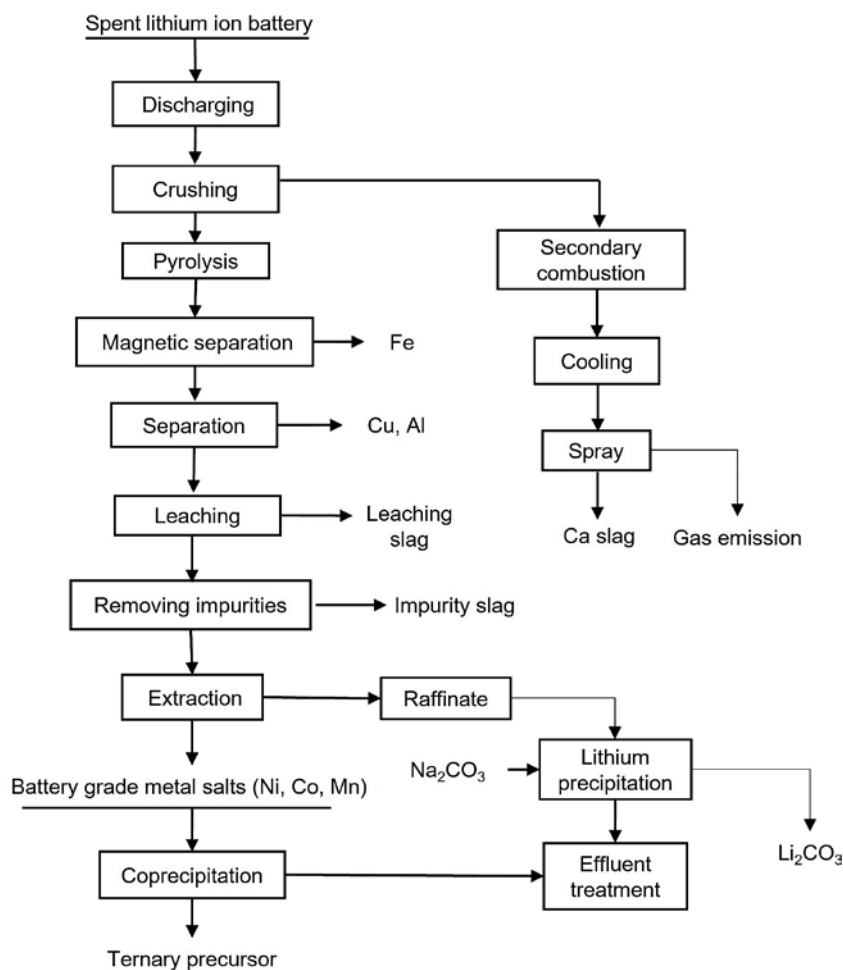


Figure 3.1 Spent lithium-ion battery recovery process of Botree Cycling.

powder is leached through the leaching kettle and then enters the extraction process to obtain battery-grade metal salts (Ni, Co, and Mn). The details of the organic component removal and nickel–cobalt–manganese co-extraction processes are provided below.

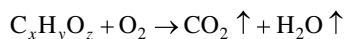
(1) There are mainly three ways to remove organic components, i.e. pyrolysis, organic solvent washing, and volatile collection.

(1) Pyrolysis

After crushing, the raw materials are fed into the pyrolysis furnace through the upper hopper by the belt conveyor, and the pyrolysis reaction is carried out at 350–600 °C to decompose and remove the binder and residual organic



matter in the battery powder. Pyrolysis flue gas is collected by the gas-collecting hood and enters the secondary combustion chamber. Combustion-supporting air is passed through the nozzle to make $C_xH_yO_z$ alkanes gas fully combusted and converted to H_2O and CO_2 .



The flue gas from the outlet of the secondary combustion chamber is first cooled down to 110–120 °C by the surface cooler and then enters the bag-house for dust collection. The flue gas from the baghouse outlet is then sent to the lye spray tower by the fan to remove fluorine gases such as HF and OPF_3 and finally discharged into the air when it meets the emission standards. The circulating liquid absorbent periodically reacts with calcium oxide, and the precipitates are separated to remove phosphate and fluoride ions from the solution.

(2) Organic solvent cleaning

The organic substance in the battery is dissolved and removed by a green organic solvent. Then, the dissolved organic mixture is separated by distillation, and the recovered organic solvent then returns to the cleaning process. The cleaned components enter the following process.

The features of the organic solvent cleaning process:

- (1) The organic solvent itself is green organics.
- (2) The organic solvent can be recycled and used repeatedly in the cleaning section, with almost no solvent consumption. The operating cost is low.

(3) Volatile collection

By making use of the volatile characteristics of organic substances in the battery, volatile collection operates at low-temperature heating conditions, and the organic components are collected by condensation.

Characteristics of volatile collection:

- (1) Low heating temperature and low energy consumption.
- (2) All the organics can be collected by condensation. Furthermore, there is almost no emission of organic substances.

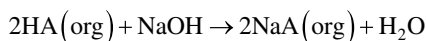
(2) Introduction of nickel–cobalt–manganese co-extraction.

After removing the impurity, the leaching solution is mainly the salts solution of nickel, cobalt, manganese, magnesium, and lithium. The traditional process extracts each metal step by step and prepares the ternary precursor by mixing. BC196 extractant independently developed by Botree Cycling can co-extract nickel–cobalt–manganese from solution in one step, and the stripping can directly produce ternary precursor materials.

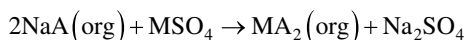
The co-extraction process has five parts: saponification, extraction, washing, stripping, and regeneration.



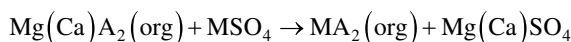
Saponification: It is a pre-balance process before the extraction section. The recycled organic phase is well mixed with basic compounds (sodium hydroxide and ammonia), and the saponified organics form is obtained as follows:



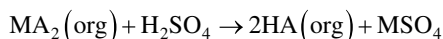
Extraction: Saponified organics and extractants are mixed with a set flow ratio, and the easy-to-extract metals such as nickel–cobalt–manganese are extracted into the organic phase to obtain the loaded organic phase, and the difficult-to-extract calcium and magnesium are left in the aqueous phase, known as the raffinate. However, a part of calcium and magnesium can still be extracted into the organic phase.



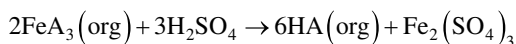
Washing: This process cleans out the calcium and magnesium impurities in the loaded organic phase. The calcium and magnesium impurities in the loaded organic phase come from the organic impurities and the entrained water phase. The washing process employs ion exchange, that is, detergent washes excess nickel–cobalt–manganese into the aqueous phase; nickel in the aqueous phase is exchanged with the loaded organic calcium and magnesium; and calcium and magnesium are exchanged into the washing solution. The loaded organic nickel–cobalt–manganese are purified.



Stripping: The extracted metals are extracted into the aqueous phase. The pH is adjusted to obtain the nickel–cobalt–manganese sulfate products. The organic extractant can be recycled.



Regeneration: To prevent the accumulation of elements such as iron, the organic phase needs to be washed with a stronger acid in the same way as in the washing stage.



Note: M is Ni^{2+} , Co^{2+} , Mn^{2+} , and other metals.

3.1.2 Brunp Recycling

Brunp Recycling is a national standardization pilot for recycling economy of spent batteries, Guangdong Province New Energy Vehicle Power Battery Recycling Pilot, National Waste Chemical Disposal Standardization Technical Committee Waste Battery Chemical Processing Working Group (SAC/TC294/WG1) Leader Unit,



National Automotive Standardization Technical Committee Vehicle Lithium-Ion Power Battery Recycling Standard Drafting Working Group Leader Unit, National Automotive Standardization Technical Committee Road Vehicle Recycling Working Group Member Unit, National Automotive Standardization Technical Committee Electric Vehicle Standards Research Working Group Member Unit, Member of heavy Metal Sub-committee of China Non-ferrous Metal Standardization Technical Committee, the Secretariat of the Technical Committee of Standardization Technical Committee for Product Recycling and Management of Guangdong Province, the Governing Unit of China Nonferrous Metals Industry Association, the Standing Director Unit of China Association for Comprehensive Utilization of Resources, and the Governing Unit of China Renewable Resources Industry Technology Innovation Strategic Alliance. Its main technical route of the lithium battery recycling business is shown in Figure 3.2.

Brup Recycling mainly recycles single batteries. Their recovery process involves the pretreatment process and the secondary treatment process. The pretreatment

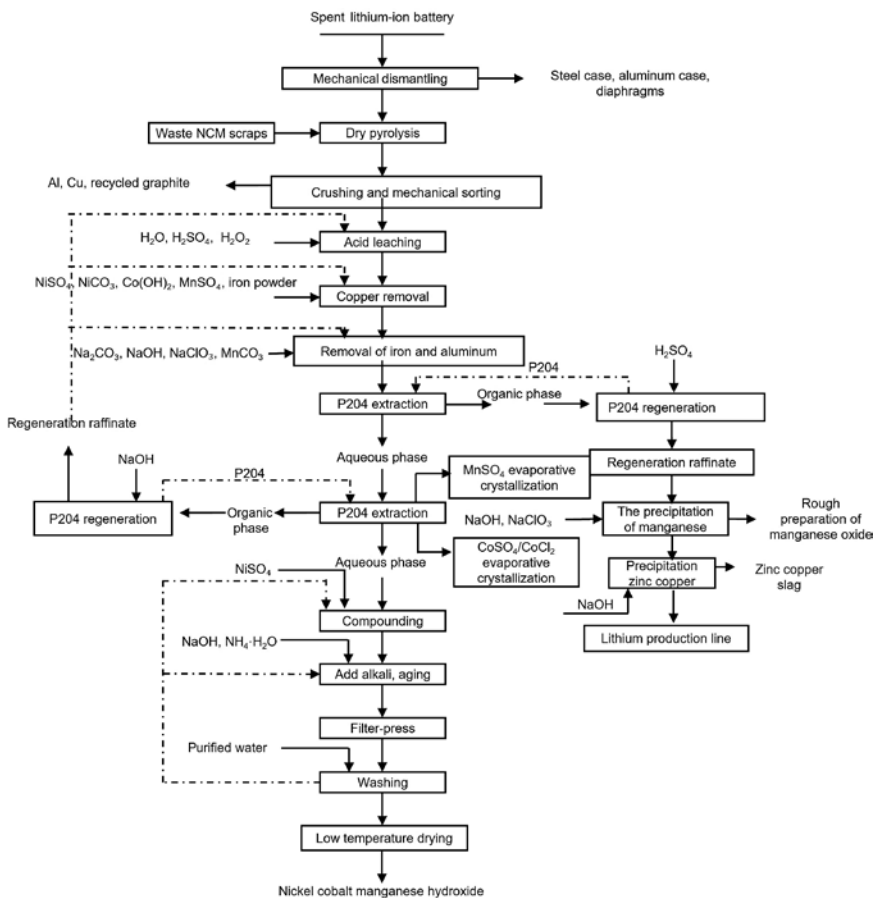


Figure 3.2 Brup Recycling spent lithium-ion battery recovery process. Source: Ref. [1].



process mainly includes disassembling, discharge, pyrolysis, crushing, and sieving, which eventually gives the copper–aluminum powder and black mass (electrode powder). The discharge process typically applies the shell set electrical discharge or 3C brine discharge methods. The pyrolysis process utilizes a rotary kiln, and pyrolysis flue gas is treated by secondary combustion + cyclone dust removal + alkali spray.

The secondary treatment process includes leaching; removal of copper; removal of iron and aluminum; extraction removal of impurity; co-extraction of nickel–cobalt–manganese ; and total precipitation. Finally, nickel–cobalt–manganese hydroxide precursors are generated. The specific process is that the temperature is stabilized at 40 °C by indirect steam heating, and the black mass is treated by acid leaching; the leaching agent is sulfuric acid and hydrogen peroxide. Nickel sulfate, nickel carbonate, and other materials are then added to stabilize the pH of the liquid between 1.5 and 1.7, and iron powder is then added to make the copper precipitate and separate through the displacement reaction. The obtained sponge copper can be sold as a by-product. After controlling the temperature at 90 °C and the pH between 1.5 and 1.7, sodium chlorate, sodium hydroxide solution, and manganese carbonate are added to the feed liquid to convert all the divalent iron into trivalent iron. A proper amount of manganese carbonate is used to adjust the proportion of nickel–cobalt–manganese . Then, the pH value is adjusted to 2.0–2.4 by sodium hydroxide solution to make the iron and aluminum ions produce iron–aluminum slag, which completes the process of removing iron and aluminum. The subsequent deep impurity removal and purification were carried out by extraction method, and the extractant is P204. Firstly, crude extraction is carried out at a certain temperature and pH to remove calcium, magnesium, and lithium impurities. The raffinate is continued to be refined. The temperature and pH of the sulfuric acid system are adjusted to co-extract and purify nickel–cobalt–manganese in the feed liquid. The acid mist absorption tower collects the sulfuric acid mist generated in the process, and the extractant after stripping can also be re-used. After the batching of nickel–cobalt–manganese sulfate, sodium hydroxide and ammonia are added for the alkalization treatment. The pH value should be adjusted to about 12.5. After holding for a certain time at 50 °C, the ternary precursor can be obtained by pressure filtration, washing, and drying. The ammonia after deamination treatment can also be re-used.

Brunp Recycling proposed a complete NCM battery technology route, including the whole process from the dismantling and crushing of spent lithium battery to the generation of nickel–cobalt–manganese hydroxide and a lithium salt, mainly by using P204 extractant for repeated extraction, to achieve the purpose of recycling lithium, nickel, cobalt, and manganese. The pretreatment process is relatively simple, whereas the subsequent processing process is relatively complex and lengthy; however, the recycled products are relatively pure.

3.1.3 Huayou Cobalt

Huayou Cobalt is a new high-tech enterprise specializing in research and development and manufacturing of new LIB materials; new cobalt-based materials; and the



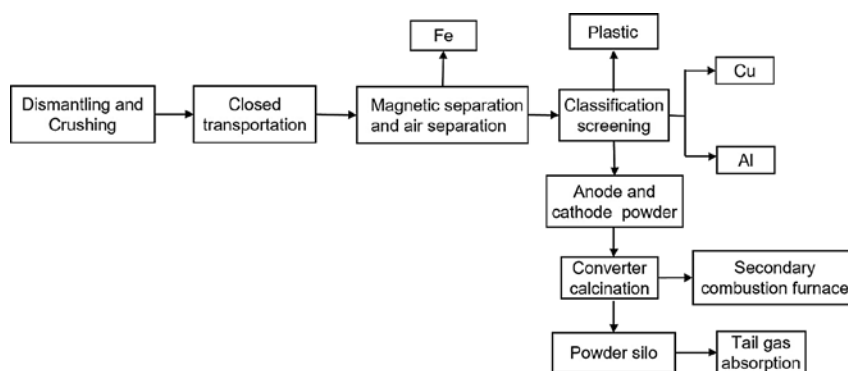


Figure 3.3 Pretreatment process of the spent lithium battery from Huayou Cobalt.
Source: Ref. [2].

mining, selection, and smelting of cobalt and copper metals. In 2017, Quzhou Huayou Resource Regeneration Technology Co., Ltd. was established by Huayou Cobalt, specializing in the spent lithium battery recycling business. The pretreatment stage of the spent lithium battery recycling process of Huayou Cobalt is crushing, sieving, and calcination to obtain black mass (electrode powder). The flowchart of the pretreatment process is shown in Figure 3.3.

The ternary spent battery is first crushed in the crushing chamber. The crushing process adopts the method of shear tearing. A high-speed knife roller is installed inside the chamber to break the battery into flake materials with specific particle sizes. Most of the plastic particles are large, which can be mostly eliminated by sieving in this process. The rest of the materials enter the magnetic separation process to separate iron particles. Then by one more time of crushing and sieving, the remaining plastics can be further screened out. Subsequently, the metal particles (copper and aluminum particles) can be separated by winnowing separation. Finally, the gained black mass (anode and cathode material powders) is calcinated in the converter and stored in the silo.

The subsequent treatment mainly focuses on the hydrometallurgy of the black mass, which will go through reduction leaching, manganese extraction, impurity extraction, cobalt extraction, nickel extraction, lithium enrichment, and finally, lithium precipitation by Na_3PO_3 . Specifically, the black mass is leached by sulfuric acid and hydrogen peroxide. During the leaching process, the pH is controlled at about 1.5, the temperature is controlled at 70°C , and the leaching time is six hours. nickel-cobalt-manganese leaching rates are all above 98%. Then, the leaching solution is extracted step by step according to the different solubility of different extractants to different metal ions. Firstly, extractant C272 is used to extract manganese. Extractant P204 is then applied to remove the raffinate's impurities (mainly calcium and magnesium). Then, P507 is used to extract cobalt and nickel. Finally, the raffinate contains relatively pure lithium, and Na_3PO_3 is added to precipitate lithium.



The pretreatment process provided by Huayou Cobalt is relatively complete, which is also the commonly used pretreatment process in the current market. It can realize the classified recovery of each component to gain the ternary anode/cathode materials ready for the subsequent process. The pretreatment process of Huayou Cobalt also contains acid leaching and extraction processes. However, its long extraction process, which applies three extractants C272, P204, and P507, leads to an increase in extraction cost and sewage treatment volume. But it is worth pointing out that this route can completely recover manganese in the extraction stage, making the recovered nickel and cobalt purer in the subsequent process.

3.1.4 GEM

GEM is a leading enterprise engaged in spent battery recycling in China. Through the multi-level recycling system of spent battery recycling boxes, electronic waste recycling supermarkets, and 3R recycling consumption community chain supermarket, GEM has built more than 20000 recycling branches. GEM recycles more than 10% of China's total electronic wastes, more than 10% of China's total spent batteries (lead-acid battery excluded), and more than 4% of China's total scrapped automobiles.

The spent lithium battery recycling process of GEM mainly consists of the crushing-separation process and the posttreatment process. The crushing-separation part mainly includes four modules of the brine discharge, dismantling, crushing, and separation, which is to remove the shells and collectors to obtain the electrode powder. The posttreatment process mainly adopts acid-leaching and extraction processes, which finally obtain the hydroxide of nickel-cobalt-manganese. Details are shown in Figure 3.4.

In the acid-leaching process, the temperature is controlled at 60 °C. After leaching, the pH value is adjusted by sodium hydroxide solution to precipitate the impurities such as iron and aluminum. The leaching solution is filtered to be ready for the extraction process. The extraction process applies extractant P204 to remove impurity and P507 to enrich the metals. After the counter-current extraction, a pure nickel-cobalt-manganese sulfate solution is collected. Subsequently, the precursor production process prepares the nickel-cobalt-manganese hydroxides. Sodium hydroxide solution and ammonia are mainly used in the synthetic process. After the reaction, the product needs to be aged at atmospheric pressure. The solids are separated by centrifugation, washed to remove the impurities, and dried. Finally, the particle size is controlled by screening, and iron particles are removed.

GEM's approach is also focused on NCM batteries, which can be used for the recovery of nickel-cobalt-manganate lithium batteries and nickel-cobalt-aluminate lithium batteries. A pure ternary battery precursor is obtained through a complete set of the recycling process. This relatively conventional process is the dominant recycling process in the current market. Meanwhile, P204 and P507 are the most widely used commercial extractants in lithium battery recycling.



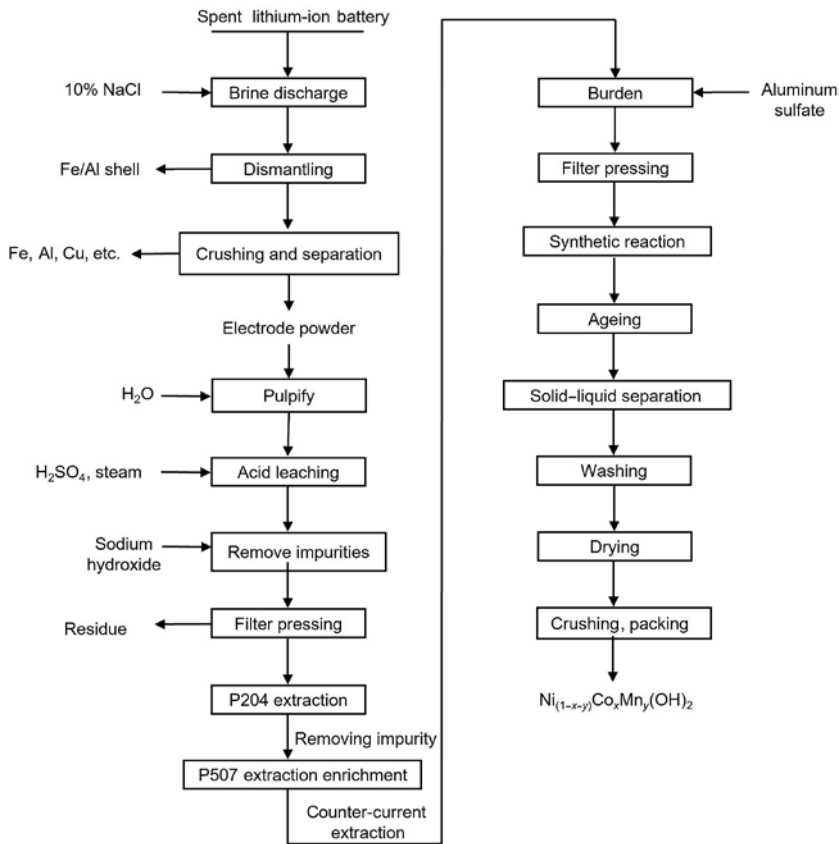


Figure 3.4 The process flow of GEM spent lithium battery recycling process.
Source: Ref. [3].

3.1.5 Shunhua Lithium

Shunhua Lithium is a technology-based startup that adheres to the purposes of high efficiency, green, energy saving, and environment friendly aiming to the safe and harmless recycling of spent LIBs. Shunhua Lithium is the pioneer of the “priority to lithium extraction” technology of spent lithium batteries. It runs the only production line of lithium iron phosphate (LFP) waste powder selective leaching to produce battery-grade lithium carbonate (1000 tons per year) in China. The main products include battery-grade lithium carbonate and anhydrous sodium sulfate. The main raw materials are the cathode scraps of LFP battery, and the main process is crushing – sieving – grinding – sieving – leaching – impurities removal – lithium precipitation – concentration – secondary precipitation of lithium, and finally obtaining battery-grade Li_2CO_3 and anhydrous sodium sulfate products. The process flow is shown in Figure 3.5.



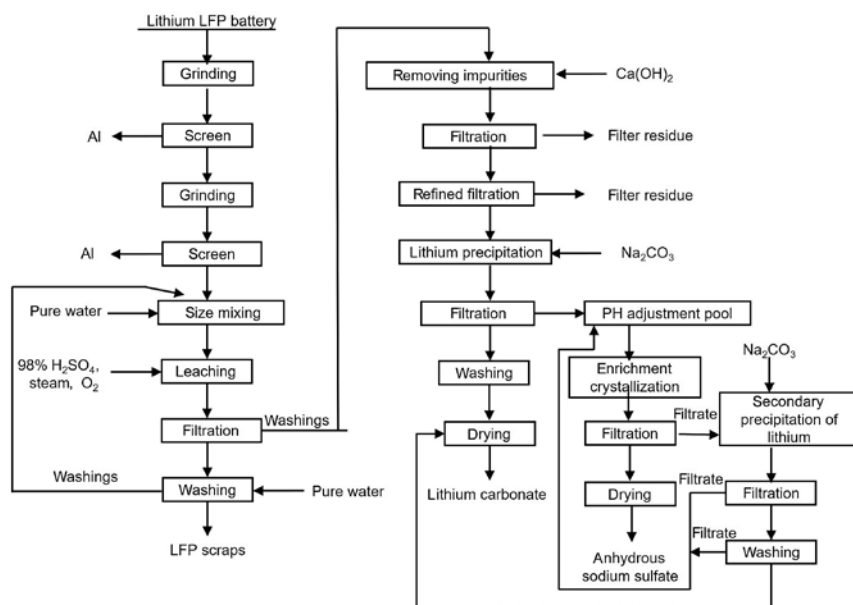


Figure 3.5 The process flow of Shunhua Lithium LFP battery recycling process.

Source: Ref. [4]/Hunan ShunHua Lithium Co., LTD.

The pretreatment process is to obtain raw LFP powder by crushing and grinding the anode scraps. Eighty mesh and 120 mesh screens are used for screening treatment, and the solid-to-liquid ratio of the recovered LFP powder is adjusted to about 1 : 3. Then, the acid leaching is conducted by adding concentrated sulfuric acid at about 80°C. The leaching suspension is filtrated. The residue is mostly iron phosphate, and it is disposed of after washing. The filtrate mainly contains Li_2SO_4 , which is purified by adding calcium hydroxide and fine filtered to be ready for lithium precipitation. Lithium precipitation reagent is sodium carbonate aqueous solution, and the sediment is lithium carbonate which is further washed and dried. The filtrate enters the pH adjustment pool for concentration and crystallization. After crystallization, anhydrous sodium sulfate is obtained by drying. Finally, there are still some lithium ions in the filtrate, which need to conduct the lithium precipitation process again to recover the remaining lithium carbonate.

Shunhua Lithium mainly focuses on the recycling of LFP batteries. The pretreatment process is relatively simple because it prioritizes recycling cathode materials. In the following process, due to the low price of LFP, Shunhua Lithium did not adopt an extraction process for recycling and removing impurity in LFP but adopted an acid-leaching process to recover the residue lithium in it. The recycling process produces anhydrous sodium sulfate as a by-product. The recycling process is relatively simple, but the recovered lithium carbonate has high purity and can reach battery grade, which can be directly used in battery production.



3.1.6 Ganpower

Ganpower is a high-tech enterprise specializing in spent battery recycling service, step spent EV power battery utilization, waste battery harmless treatment, and high purity metal salts production. The main products include cobalt salt, nickel salt, lithium salt, and cascade utilization series products. Ganpower focuses on the production of cobalt sulfate and nickel sulfate, and its raw materials are nickel-metal hydride batteries and LIBs. Through a process of punch discharge – dismantling – pyrolysis – crushing – magnetic separation, the iron–nickel oxide and nickel–cobalt scraps can be recovered. After alkali leaching to remove aluminum, the wet recovery process is applied. This process has an annual output of 2500 tons of highly pure nickel sulfate and 500 tons of highly pure cobalt sulfate. The process flow is shown in Figure 3.6.

The pretreatment process mainly involves putting the disassembled spent lithium batteries into the furnace for three to six hours at 350–900 °C. The pyrolyzed materials are crushed by a vertical high-speed rotary mill machine and screened by a vibrating screen machine. Then, the treated powders are screened by magnetic separation to obtain the magnetic materials (iron and nickel oxide, nickel and cobalt scraps) and non-magnetic materials.

The resulting nickel and cobalt scraps enter into the leaching process, and the metals are dissolved by inorganic acid. Then, iron and aluminum impurities in the solution can be removed by hydrolysis. After this chemical purification, the solution enters the extraction process and is successively extracted by P204 to remove impurity and P507 to separate cobalt and nickel, respectively, and finally obtaining the battery-grade nickel sulfate and cobalt sulfate solution.

Ganpower lithium spent battery recycling process also uses the acid-leaching and extraction processes. The P204 and the same P507 extractants are also used in the extraction process. In contrast, it takes advantage of the fact that P204 and P507 have different solubility characteristics for different ions. The separation and recovery of nickel and cobalt are achieved by adjusting the pH of the extraction process and controlling the ion species. This process can recover several metal compounds with high purity. But it applies three times of extraction, and the process is lengthy and complex, thus making this process costly and producing more wastewater and using more acid and alkali.

3.1.7 Qiantai Technology

Shenzhen Qiantai Technology Co., Ltd. was established in 2016 as a joint venture by a number of investment companies, including Shenzhen Qiantai Energy Renewable Technology Co., Ltd., Guangdong Wanzhonghui Investment Co., Ltd., Shenzhen Kunpeng Yichuang Strategic Emerging Industry Equity Investment Fund Partnership (Limited Partnership), and Shenzhen Expressway Infrastructure Environmental Protection Development Co., Ltd. Qiantai Technology's primary



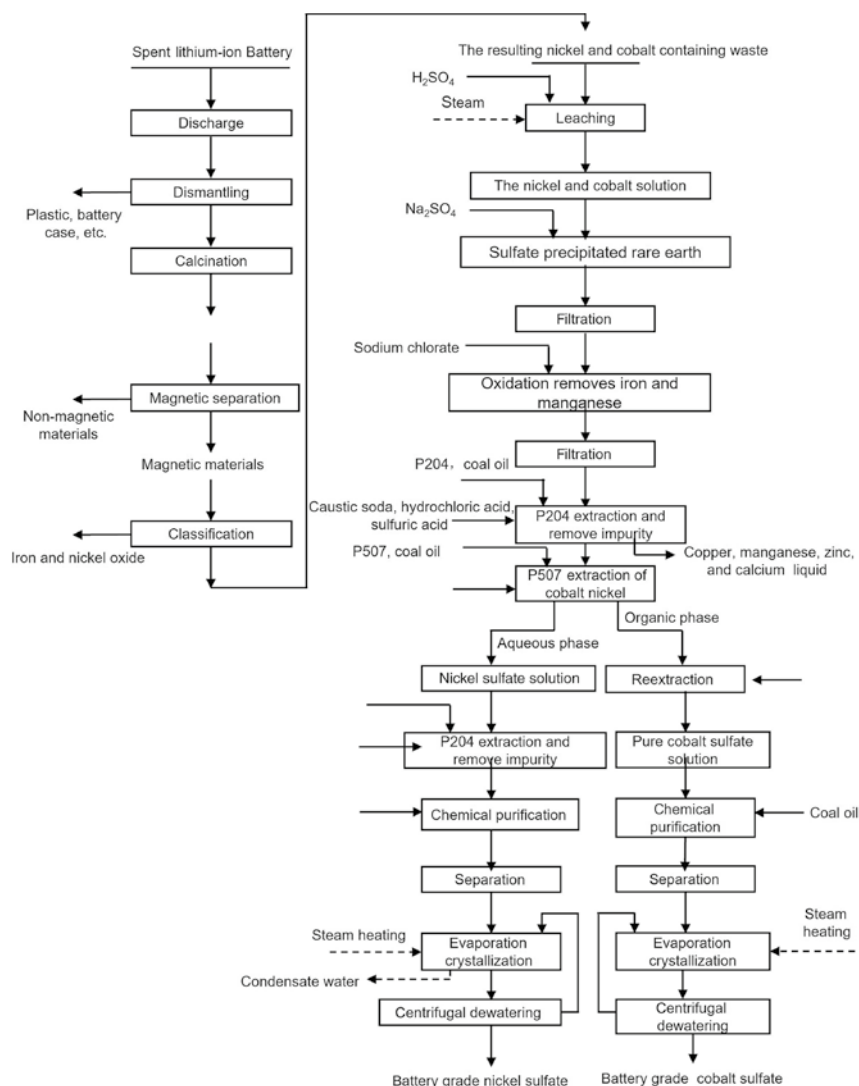


Figure 3.6 The process flow of Ganpower spent lithium battery recycling process.

Source: Ref. [5].

business scope includes power battery module recycling and cascade utilization, comprehensive utilization and recycling of spent battery, and scrap car recycling and disassembly. Qiantai Technology has been listed in the second batch of the allowed list of battery recycling companies by the Ministry of Industry and Information Technology of China, and the type is cascade utilization.

The battery recycling and dismantling line of Qiantai includes four processes: Code scanning, battery pack disassembly, battery discharge, and crushing sorting (Figure 3.7). Different discharge processes are used according to the different shapes and sizes of the batteries. The small cylindrical batteries that meet the testing



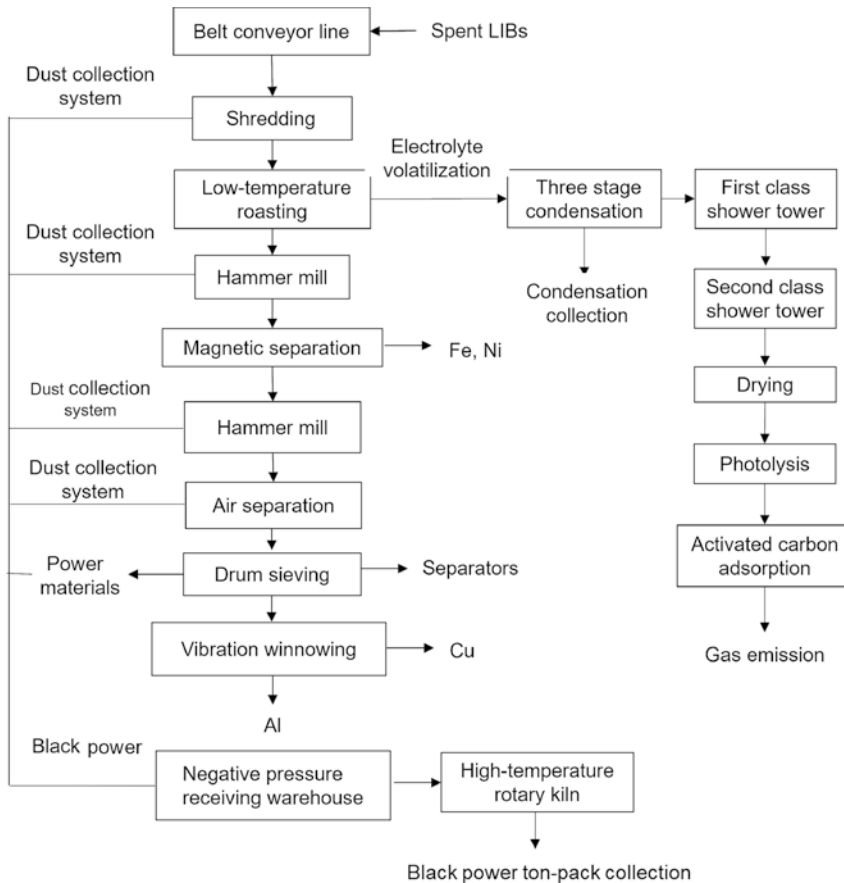


Figure 3.7 Flowchart of crushing and sieving of decommissioned lithium batteries.
Source: Ref. [6].

standards can be broken directly without discharging, and those that do not meet the standards are discharged first in a discharge tank and then broken; the soft-pack battery is discharged in a discharge cabinet, and the nickel and copper tabs are broken after taking out manually; the square battery is also discharged first, the shell is then cut, and the tabs are manually taken out and broken. The crushing and sieving process after battery discharge is exactly the same. The first shredding process is carried out under the protection of nitrogen. The entire dismantling line is produced under sealed negative pressure. The process is described in detail as follows:

QR Code scanning: The serial numbers of battery packs, modules, and cells are in one-to-one correspondence. The disassembly equipment is equipped with a QR code scanning system. Before disassembling, all QR codes must be scanned to track which battery pack the cells come from. After manually checking and revising, the obtained data will be uploaded to the lithium-ion power battery traceability



management system of the Ministry of Industry and Information Technology of China.

Battery pack disassembly: The voltage and remaining power are checked before unpacking the battery pack. If the voltage is greater than 30%, the battery pack is discharged below the battery management system (BMS) protection voltage. The battery pack is discharged in a discharge cabinet, and a negative voltage is applied for forced discharge. Battery packs with a capacity of less than 30% can be disassembled semi-automatically using manual + robotic arms. The pack is first disassembled to the module and then to the battery cell.

The battery pack's disassembly process is typical: (i) disassemble the upper cover; (ii) disassemble the tandem platform to reduce the voltage; (iii) disassemble the cables and sensors. These three steps are manual disassembly. (iv) Disassemble the BMS; (v) disassemble the module. After removing the module, the QR code is scanned to record and test the module's voltage, internal resistance, and power; (vi) the module is disassembled into battery cells. The disassembled cell also needs to undergo scanning of the code and testing for voltage, internal resistance, and power. During the disassembly, if it is welded, mechanical assistance is required to prevent the welded nickel sheet from being torn off in case it affects the secondary use of the battery. The small pack of the two-wheeled vehicle is basically disassembled manually, and the large battery pack needs to be hoisted by mechanical equipment. The whole set of equipment for dismantling the battery pack to the battery cell is more than 2 million yuan, including the discharge cabinet.

During the unpacking process, the operator manually divides the batteries into small cylindrical, soft-packed, and square-shell batteries according to the shape and size of the battery cells, and the batteries then enter the pretreatment process before crushing.

Battery cell discharge: All batteries are tested to determine the current and voltage values before entering the crushing and sorting process. If the voltage of the 18650 small cylindrical battery cell is below the cut-off voltage, it can be directly crushed without discharging. Soft-pack and square batteries need to be discharged before crushing. The discharge method used by Qiantai Technology is electronic load discharge. The voltage specifications of the discharge cabinet used are 300, 500, and 600V, and the current specifications are 300 and 500 A. It takes three to five minutes to discharge the battery with a 500 A type. Another way to collect residual energy after discharge is to re-use. However, the recovery cost currently is less than the equipment investment, which is not economically feasible.

Crushing and sorting process: The pretreatment is performed separately according to the battery shape and size. Small cylindrical batteries that meet the testing standards can be crushed directly without discharging, and those that do not meet the standards can be discharged with a discharge tank and then crushed; the soft-pack battery is discharged in a discharge cabinet, and the nickel and copper tabs are manually taken out and then crushed; the square battery is also first discharged, the shell is then cut, and the tabs are manually taken out and finally crushed. When switching between different types of batteries, the production line



should be idle running for 30 minutes to clean up the residual material inside the machine. The crushing and sorting process after pretreatment is exactly the same. The first crushing process is conducted under nitrogen protection, and the entire dismantling line is produced under sealed negative pressure. The process is described in detail as follows:

- Crushing without discharge:** The battery cell is crushed under the protection of nitrogen, all oxygen is replaced by the negative pressure of the isolation cabin, and the continuous feeding is completed. Then, the shredder shreds the battery into flakes, and the produced black mass is sucked into the storage bin through the integrated system. At this time, the material contains large amounts of electrolytes. The electrolyte is removed by low-temperature baking. The baking temperature does not exceed 200 °C, which promotes the volatilization of electrolytes, carbonates, and other organic solvents. Thereby, the electrolyte is removed.
- Electrolyte disposal:** The material dust and volatilized electrolyte gas produced during the crushing process are firstly condensed in three stages through the condensing device. The first stage uses ambient-temperature circulating water to condense the electrolyte and dust mixture from 200 to 40 °C; the second stage is the condensation of organic gases, which is cooled from 40 to 15 °C; the third stage is cooled from 15 to −5 °C. Through three-stage condensation, 95% of the mixture is condensed into liquid. The remaining gas passes through the primary and secondary washing towers to remove fluorine, phosphorus, and dust. After drying, it is photodegraded with a 40J light source. The organic matter is photodegraded into carbon dioxide and water. After absorbing the impurities and odors by activated carbon, the produced gas directly exhausts into the air.
- Recycling metals, separators, and other materials:** The materials after the volatilization of the electrolyte are subjected to a hammer mill and then undergo magnetic separation to remove 98–99% of iron–nickel and metal shells. The remaining material is subjected to second hammer milling to make the powder on the surface of the electrode fall off, and the powder and the separator are then stratified by combined winnowing; the powder and the separator are then separated by a cylindrical sieve.
- Pulverization and separation of copper and aluminum:** A total of 65–70% black mass can be obtained by cylindrical screening, and the content of copper and aluminum impurities is less than 2%. In addition, the entire crushing and separation process has a dust collection system to collect the powder; the black mass content is about 30%, and the overall powder recovery rate is >95%. After recovering the powder and separator, the copper and aluminum current collectors are separated by vibrating sieving and winnowing.
- Powder pretreatment:** All powders are treated in a rotary kiln to remove the binder and residual electrolyte, and the waste gas generated is connected to the electrolyte treatment system. The processed black mass is collected in a large powder bag for the subsequent hydrometallurgy process.

Qiantai Technology Co., Ltd. does not have a hydrometallurgical recycling production line for electrode materials.



3.2 Europe

Three typical industrial LIBs recycling processes from European companies are introduced in this chapter. Although the products might be similar, the processes differ a lot from different companies. Both Umicore Valéas[™] and Accurec processes use a combination of pyrometallurgy and hydrometallurgy, whose details are described in Sections 3.2.1 and 3.2.2, respectively. The Recupyl process focuses on the hydrometallurgical method introduced in Section 3.2.3.

3.2.1 Umicore

Umicore is one of the largest LIB recycling companies in Europe and has a battery-recycling plant with a capacity of 7000 tons located in Hoboken, Belgium. Umicore has independently developed the unique pyrometallurgical treatment and a state-of-the-art hydrometallurgical process – Valéas process, which can recycle all types and sizes of Li-ion (LFP, NCM, and lithium cobalt oxide (LCO)) and nickel metal hydride (Ni-MH) batteries. The schematic representation of this process is illustrated below in Figure 3.8.

The pyrometallurgical step employs Umicore's unique Ultra High Technology (UHT). It is designed to safely treat large volumes of different types of complex metal-containing waste streams and has the following advantages (<https://csm.umicore.com/en/battery-recycling/our-recycling-process/>): (i) a higher metal recovery rate compared to existing processes and the output of directly marketable products; (ii) direct feeding of the batteries, which avoids the need for any potentially hazardous pretreatment; (iii) the gas-cleaning system guarantees that all organic compounds are fully decomposed and that no harmful dioxins or volatile organic compounds (VOCs) are produced. Fluorine is safely captured in the flue dust; (iv) reducing the energy consumption and CO₂ emissions to a minimum by using the energy present inside the battery components (electrolytes, plastics, and metals); and (v) generating close to zero waste.

The Valéas process can be divided into three steps as given below:

In the first step, the battery, coke, reducing agent (Al and Zn), and slag-forming agent are mixed and fed into the vertical shaft furnace and preheated to 300 °C, where the electrolyte will slowly evaporate, which reduces the risk of explosion. Then, the materials are transferred to the plastic pyrolysis zone. The temperature increases to 700 °C to melt the plastic and decompose the binder into vapors. The pyrolysis gas and the electrolyte vapor burn in a high-oxygen atmosphere. Ca salt and Na salt are added to trap halide and prevent dioxin and furan formation.

In the second step, the preheated oxygen-enriched air stream is injected into the bottom of the furnace through the tuyere and reacts with the remaining raw materials. They are reduced and smelted in the smelting zone at the bottom of the furnace, where the temperature is between 1200 and 1450 °C. Li enters the furnace slag composed of Al and Si, while Cu, Co, Ni, and Fe enter the alloy phase. The alloy phase is further treated by the hydrometallurgical method to extract metals.



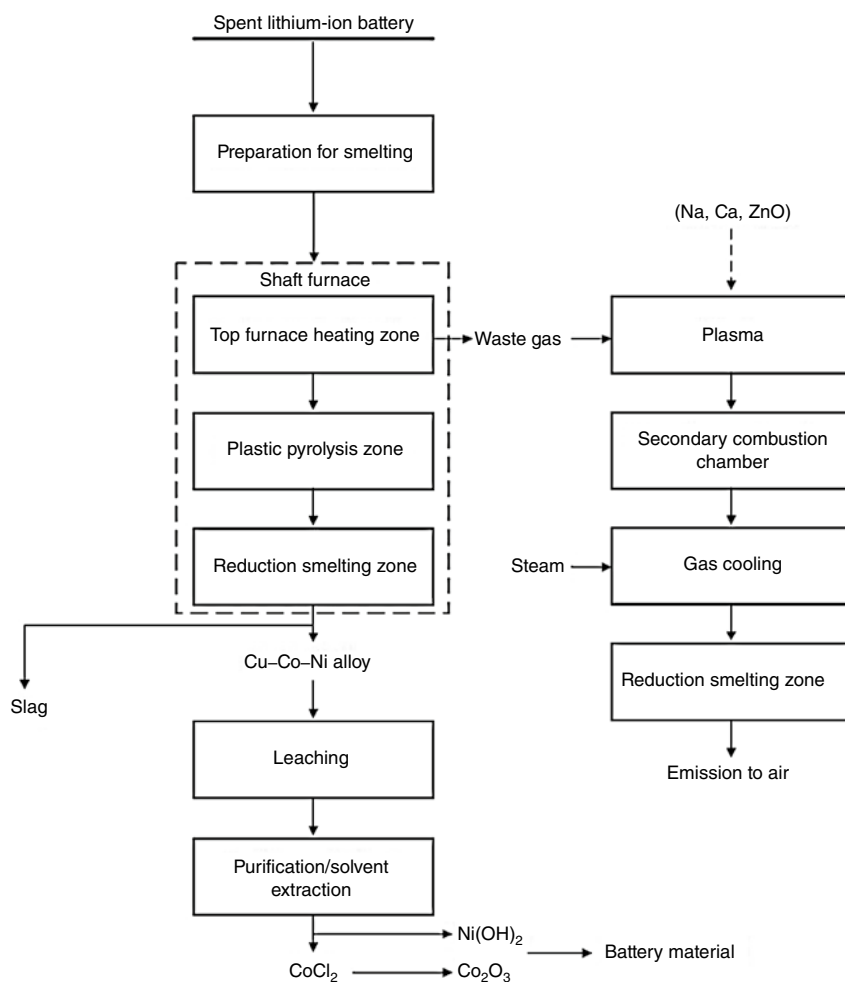


Figure 3.8 Schematic illustration of Umicore Valéas™ process. Source: Adapted from <https://csm.umicore.com/en/battery-recycling/our-recycling-process/>.

In the third step, the hydrometallurgical method is used for refining. The first stage of the refining process is to dissolve the alloy-containing Co, Ni, Cu, and Fe in sulfuric acid. Cu is removed by adding SO₂ to produce CuS and Cu₂S precipitation, and the Co and Ni are extracted and separated by using solvent extraction. The separated nickel-containing solution is treated by adding NaOH to increase the pH, thereby obtaining solid Ni(OH)₂ precipitation, which is further processed into new battery materials. After being stripped with the concentrated hydrochloric acid solution, the cobalt-containing organic phase is injected into the calciner and reacts with oxygen at a high temperature to convert to Co₂O₃.

The advantages of the Valéas process are as follows: (i) wide raw material adaptability and large system processing capacity; (ii) avoiding complicated mechanical



dismantling and physical sieving and realizing the mixed treatment of LIBs with different cathode/case materials; and (iii) making full use of the reducibility and energy contained in the materials such as aluminum, graphite carbon, and plastics, thus realizing the centralized and harmless disposal of toxic/hazardous substances and producing environment-friendly solid waste.

3.2.2 Accurec

The Accurec process was developed by the German company Accurec GmbH[®], which initially recycles Ni–Cd batteries and has extended its application to LIBs. The process is mainly composed of three parts, i.e. mechanical pretreatment for physical separation, the pyrometallurgical process for cobalt-based alloy production, and the hydrometallurgical process to produce Li_2CO_3 . The flow of this process is presented in Figure 3.9.

The process starts from manual sieving, cleaning, and dismantling into cells, where simultaneously plastics, electronics, and connecting parts are separated. The dismantled cell is then sent to the heat treatment under vacuum to remove electrolyte, solvent, and hydrocarbon, and the temperature is strictly controlled at 250 °C. After removing the organic components, the deactivated batteries are subjected to a series of mechanical treatments, e.g. milling and grinding, shredding into small pieces, vibrating screen for the separation of foil and powder, and magnetic separation for Fe. The Fe, Al, and Cu foils with relatively big sizes are separated from the cathode/anode powder with relatively small sizes.

The cathode/anode powder undergoes a spheroidization process with the help of binders and is ready for the pyrometallurgical process. The pyrolysis process is carried out in a rotary kiln at 800 °C, where cobalt alloy is produced through a carbothermal reduction reaction, while lithium is concentrated and goes into the slag phase.

In the last step, lithium-containing slag is treated by a hydrometallurgy process to produce Li_2CO_3 . The slag is broken down into small particles with sizes smaller than 100 μm and then leached out using H_2SO_4 . After solvent extraction, lithium is ultimately precipitated as Li_2CO_3 .

3.2.3 TES (Recupyl)

Recupyl, based in Grenoble, France, is an international company specializing in the recycling of waste batteries. In 2018, SINGAPORE-TES, the world's largest e-waste recycler, announced that it has acquired the assets of Recupyl SAS, which accelerates TES's move into the battery processing market in Europe. The Recupyl recovery process employs a purely hydrometallurgical method with low temperature and low energy. The schematic illustration of this process is presented in Figure 3.10.

The spent LIBs are firstly shredded by a low-speed rotary under the protection of a CO_2 and Ar mixed inert atmosphere. This atmosphere can ensure safety when there are charged batteries. The shredded parts will be further broken down into particles smaller than 3 mm by a mill machine. The waste gas generated during mechanical dismantling will be sent for further treatment before being exhausted



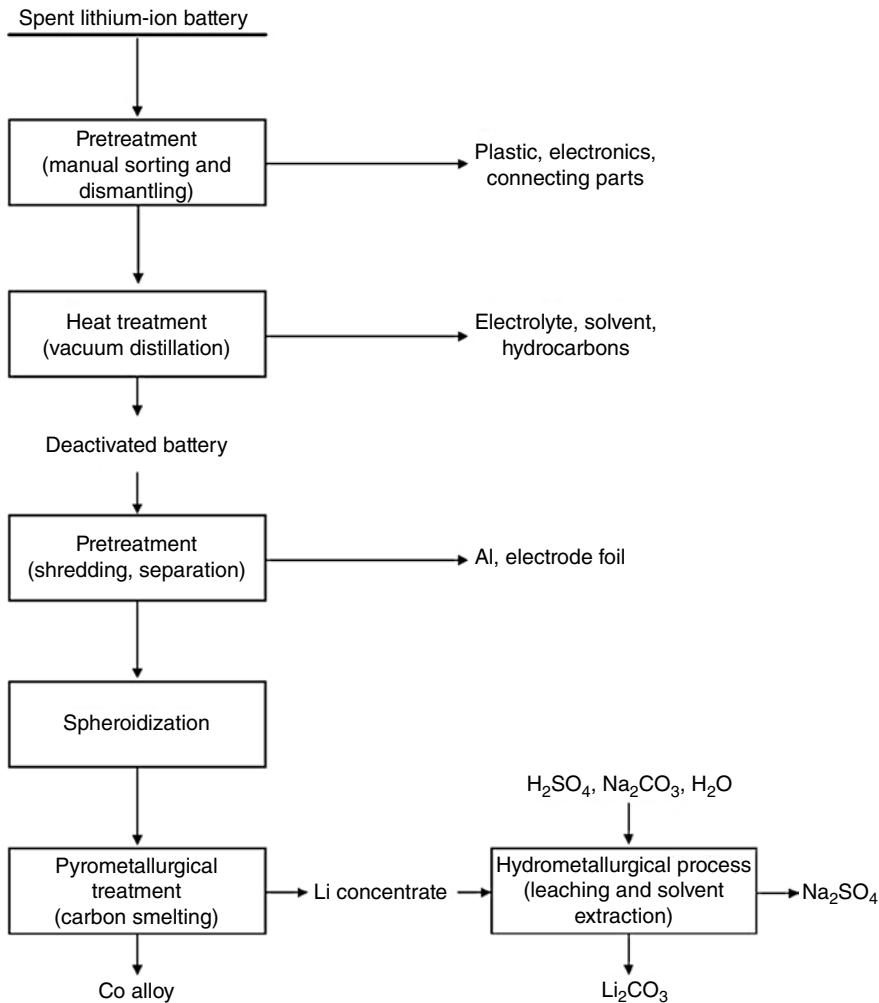


Figure 3.9 Schematic illustration of Accurec process. *Source:* Adapted from Ref. [7].

into the air. The shredded parts are separated with the help of a vibrating screen. The oversized particles are magnetically separated to remove the Fe particles, and the remaining particles are transported to the gravity separator, where the dense Cu/Al and light plastics/paper are separated.

The undersized particles pass through a finer sieve with a $500\mu\text{m}$ opening size to remove most of the remaining Cu particles, and the remaining fraction is mainly composed of active electrode materials and is sent for subsequent hydrometallurgical process.

The active electrode materials are firstly mixed with water, and the pH is adjusted to 12 by adding LiOH salt. H_2 is generated during this process because of the electrolysis reaction. Lithium is dissolved into the aqueous solution, while graphite will float on the liquid surface and can be separated by filtration. Lithium is precipitated



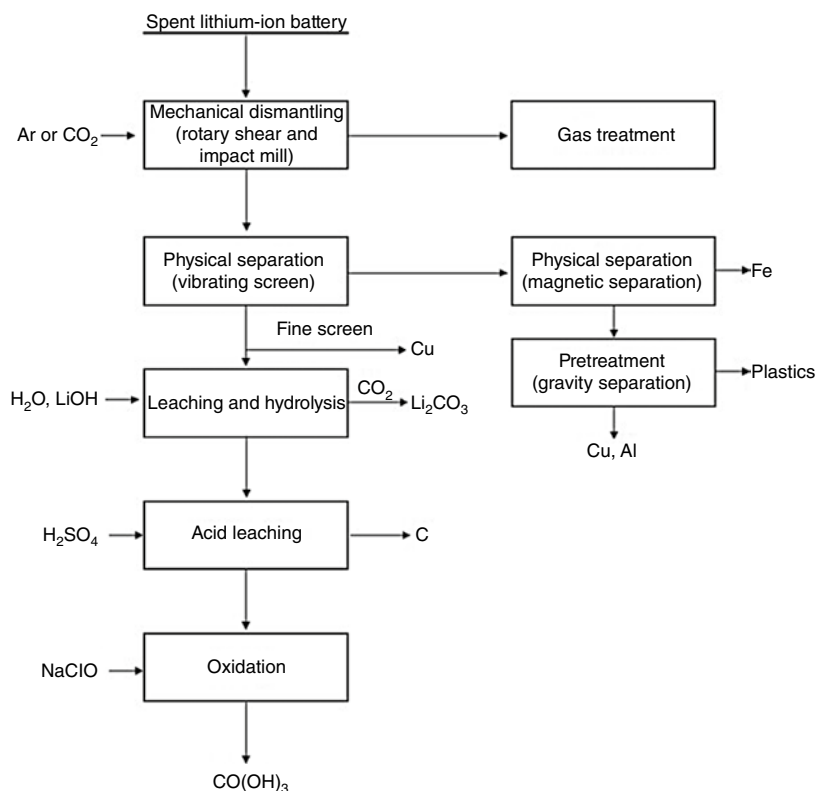


Figure 3.10 Schematic illustration of Recupyl process. *Source:* Adapted from Ref. [7].

as Li_2CO_3 by bubbling CO_2 gas into the solution. The remaining undissolved fraction is leached out by H_2SO_4 at 80°C , and carbon powder is filtered afterward. In the last step, NaClO is added into the solution to oxidize Co^{2+} to Co^{3+} precipitating as Co(OH)_3 .

3.3 North America

According to public media reports, the earlier established battery recycling companies in North America are mainly Inmetco and Retrie (formerly known as Toxco). Inmetco mainly uses collaborative smelting to recover valuable metals such as copper, nickel, and cobalt. Retrie focuses on lithium extraction. In the past two years, new companies have joined this field such as Li-Cycle, Redwood, and Battery Recycle.

3.3.1 Li-Cycle

Founded in 2016 and listed on New York Stock Exchange (NYSE) in 2021, Li-Cycle is on a mission to leverage its innovative Spoke & Hub Technologies™ to provide



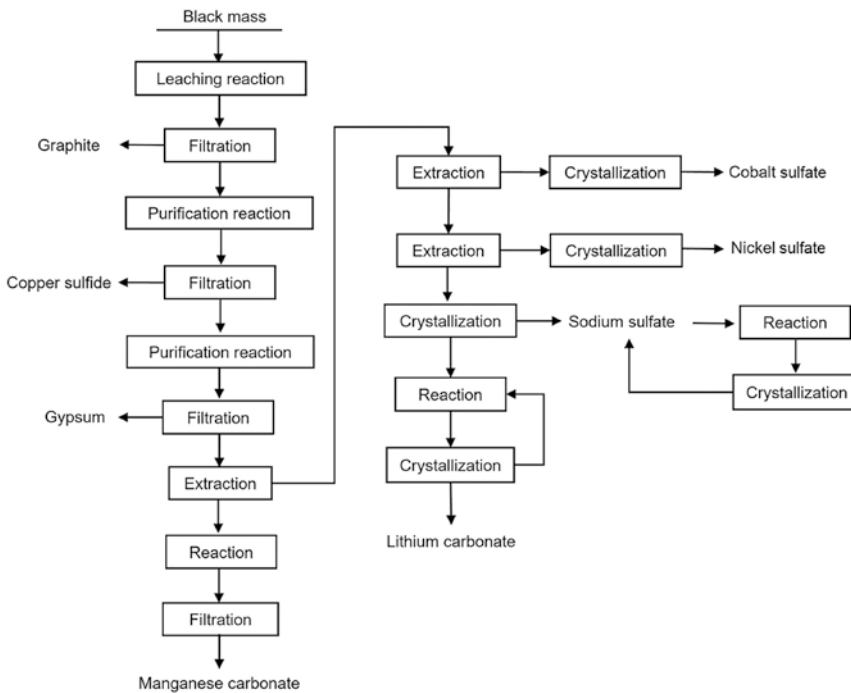


Figure 3.11 Schematic illustration of the Hub process. *Source:* Li-Cycle.

customer-centric, end-of-life solutions for LIBs, while creating a secondary supply of critical battery materials. Li-Cycle is working with major EV players such as GM, Daimler-Benz, Hyundai, and LG and is expanding in North America, Europe, and Asia Pacific region. Spoke and Hub battery process capacity will reach 65 000 and 90 000 tons per year, respectively, by 2025.

Their proprietary Spoke technology has the ability to accept all LIBs of full modules with full charge, and there is no need to separate different battery chemistries. Because of their patented Neutralizing Solution Shredder, the process realizes minimal clean water discharge, site-maximizing water recycling, and zero-impact air emissions. The black mass obtained from the Spoke process is further recovered through the Hub process (Figure 3.11). Li-Cycle announces their recovery rate up to 95% of the critical materials from LIBs of all chemistries and form factors, including lithium carbonate, cobalt sulfate, and nickel sulfate for battery use and manganese carbonate, copper sulfide, sodium sulfate, and gypsum for industry use.

3.3.2 Inmetco

Inmetco is an international metal recycling company located in Elwood, USA. As a leading environmental services provider to the US steel industry, Inmetco was founded in the 1970s with investment from INCO Canada. The company mainly uses a converter to process waste from stainless steel factories to produce direct



reduced iron (DRI) and recover valuable metals such as Ni and Cr. Its initial process was not designed to deal with spent lithium batteries. Because Co, Ni, and Fe contained in lithium batteries can be used in the production of iron-based alloys; Inmetco then decided to use the factory's converter to process spent lithium batteries. Its main technical route is shown in Figure 3.12.

The collected spent lithium batteries are pretreated by disassembling and crushing and mixed well with a carbon-based reducing agent. The gained powder materials then undergo a spheroidization process and are transferred into the converter for reduction melting at 1260 °C. The residence time of the material in the converter is about 20 minutes. After the reduction smelting process, Ni, Co, Mn, and other oxides in the battery are reduced to the metal state and enter the alloy phase, while lithium enters the slag phase. Smelting slag is sold as aggregate for building materials, and the alloy phase is refined in an electric furnace. After refining, the iron-base alloy-containing Ni, Co, Cr, and Mn are produced.

This process belongs to the co-smelting process, and the waste battery is used as one of the raw materials in the original main process system. After treatment, Li and Al in the battery enter the slag phase and cannot be recycled, while Ni, Co, Mn, and Cu enter the product and are recycled in the form of iron-base alloy.

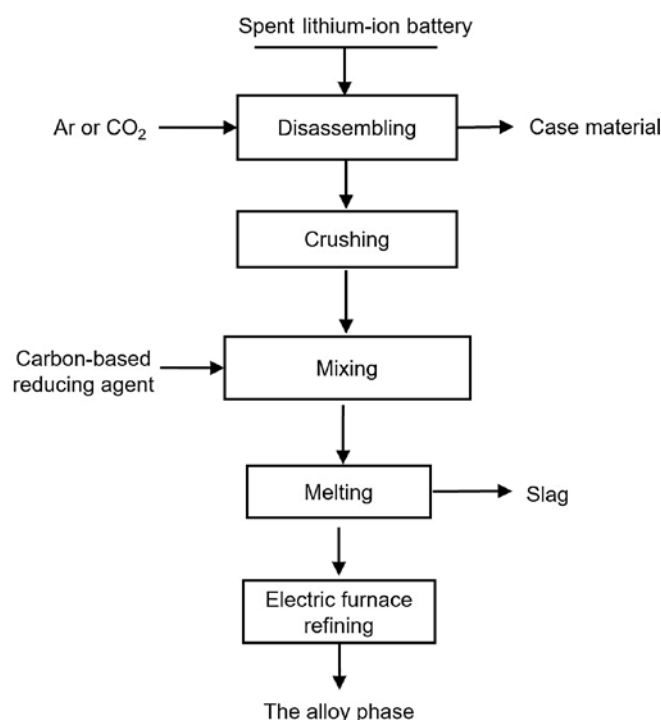


Figure 3.12 Inmetco spent lithium-ion battery recovery process. *Source:* Adapted from Immetco. <https://azr.com/about/inmetco>.



3.3.3 Retrieval

Retrieval Technologies (formerly Toxco, Inc.) has over 25 years of experience in battery recycling and management. Headquartered in Lancaster, Ohio, Retrieval has become one of the world's most diverse battery recycling companies and can recycle all types of batteries and battery chemistries. Its main process prioritizes lithium extraction and recovery. Other valuable metals are sold to professional companies for further recycling. The main flow of this process is presented in Figure 3.13.

The battery pack is manually disassembled to a single cell, and the single cell is cooled by liquid nitrogen to a temperature between -175 and -198°C . At this temperature, the battery does not have the risk of explosion. In addition, low temperatures make the battery's plastic shell brittle, making it easier to break and sort. After discharge, the battery can directly enter the crushing process. After removing the shell, the cells enter the wet milling process, and the solution is lithium salt solution. During the crushing process, lithium solution neutralizes the electrolyte, lithium ions are dissolved, and gas emission is prevented. After liquid–solid separation, the obtained Li-containing solution is purified to obtain LiOH solution and then precipitated to form Li_2CO_3 by adding CO_2 . The slag containing

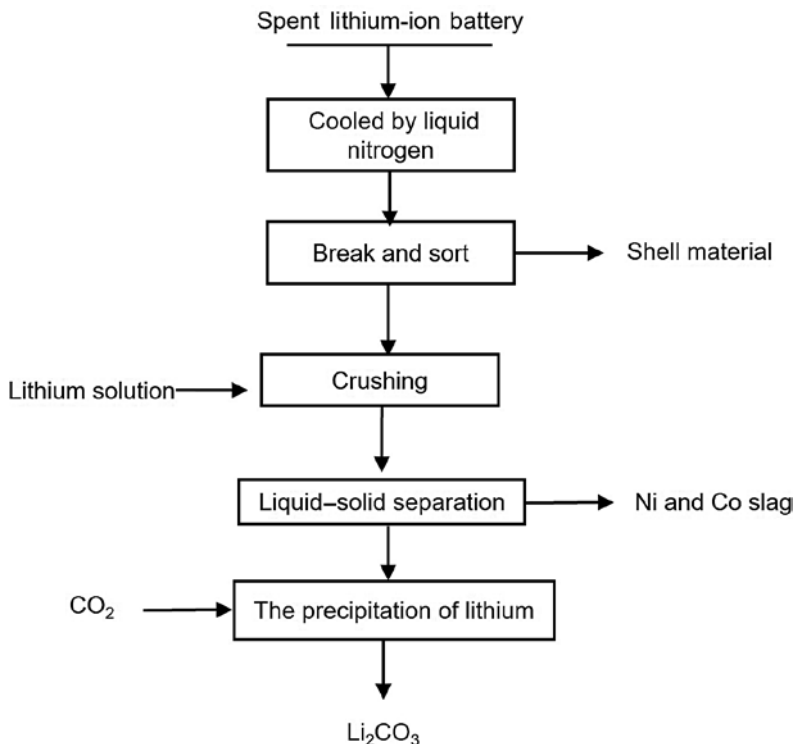


Figure 3.13 Toxco spent lithium-ion battery recovery process. Source: Adapted from Retrieval. <https://www.retrievaltech.com/company>.



valuable metals such as Ni and Co is sold to the downstream smelters for further processing.

Retriev uses a low-temperature wet ball milling process to recycle NCM batteries. Low-temperature crushing can solve the fire problem in the crushing process and improve the crushing efficiency. Lithium salt can neutralize the electrolytes and dissolve lithium in the crushing process, simultaneously achieving lithium, nickel, and cobalt metal separation and pollution reduction. CO₂ as the lithium precipitating agent is economical and environment friendly. However, low-temperature crushing needs to be carried out between −175 and −198 °C, which will undoubtedly increase the difficulty of equipment design, and the energy required to maintain such a low temperature for long term is also tremendous. Secondly, it adopts a very short process for recycling spent batteries, and its recovery efficiency and accuracy are difficult to guarantee.

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4

Current Status of the Carbon Footprint Life Cycle Analysis of Power Lithium-Ion Batteries and the Impact of Recycling on Them

4.1 Life Cycle Analysis of the Power Battery Manufacturing Process

4.1.1 Introduction to the Life Cycle Assessment Framework

Life Cycle Assessment (LCA) is not a buzzword; its history can be traced back to the end of the twentieth century when the methodology was named as Resource and Environment Profile Analysis (REPA). Coca-Cola conducted a study in 1969 to quantify the raw materials and fuels used as well as to determine the environmental impact of the manufacturing processes for their different beverage packaging bottles. The results of the study had even convinced Coca-Cola to shift their beverage packaging from glass bottles to plastic ones, as they have smaller overall environmental impact. This was the first well-known successful business case and treated as a milestone in the development of LCA; since then, more and more companies started to analyze their products in a similar fashion, although the standard of LCA has not been compiled for a long time. The concept of LCA was first introduced by the International Society for Environmental Toxicology and Chemistry (SETAC) at an international LCA seminar in 1991, and successively in 1993, they published the programmatic report, “Life-Cycle Assessment (LCA) Outline: A Practical Guide,” where the basic technical framework for the LCA method had been provided. Eventually, in 1997, International Organization for Standardization (ISO) officially published ISO 14040 standard: Environmental management – Life cycle assessment – Principles and framework, which is the international mainstream LCA framework that has been widely used nowadays. Since then, various standards focused on specific product footprint (e.g. ISO 14046 – Environmental management – Water footprint and ISO 14067 – Greenhouse gases – Carbon footprint of products – Requirements and guidelines for quantification) and specific product categories (Product Environmental Footprint Category Rules – PEFCR) have been gradually developed.

LCA is an assessment of the input, output, and potential environmental impact of the production system in the life cycle. It mainly includes four stages: goal and scope



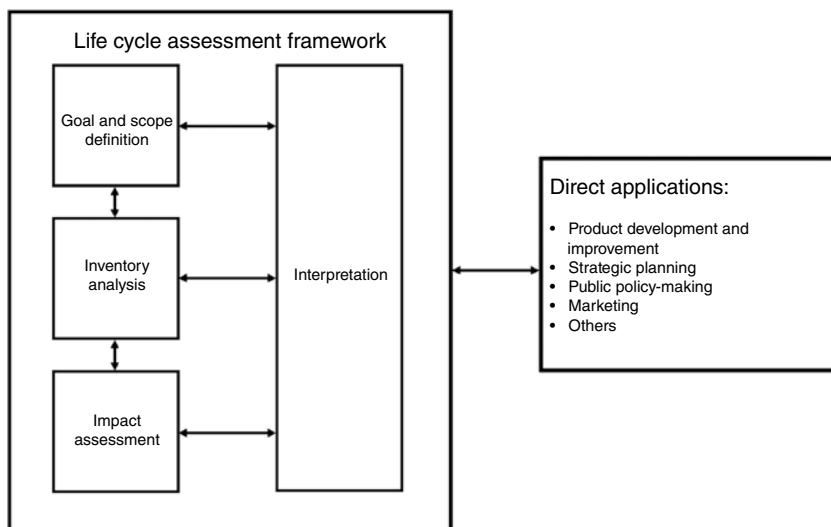


Figure 4.1 Main process of implementation of life cycle assessment. *Source:* ISO14040 [1].

definition, Life Cycle Inventory Analysis (LCI), Life Cycle Impact Assessment (LCIA), and life cycle interpretation, which are schematically illustrated in Figure 4.1.

4.1.1.1 Goal and Scope Definition

Being the first step of LCA, the goal and scope definition orient the whole work and specifically include the expected application of the results, the reason and background of the research, and the entity being informed of the results. Specifying the scope means identifying and specifying LCA research objects, that is, the product and other systems to be analyzed. This includes product system and function, functional unit, and reference flow. System boundary requirements include completeness requirements and selection criteria, types and sources of data information, quality requirements of LCI data, and types of environmental impacts covered by LCIA.

4.1.1.2 Life Cycle Inventory Analysis

The main work of this stage includes data collection and system simulation, collecting qualitative and quantitative data of each unit process within the system boundary. This includes process input and output flow, input and output factors, data calculation, and corresponding data sources and quality descriptions. At this stage, it should be noted that all data must be associated with unit processes and functional units, the flow of all unit processes should be associated with the reference flow, and all input and output data of the system are based on the functional unit. The result of LCI is the input of the subsequent LCIA stage, which is the main stage of data collection.



4.1.1.3 Life Cycle Impact Assessment

LCIA is based on the calculation index results of different environmental impact types by functional units, which converts the input and output of the benchmark flow in LCI into impact indicators related to human health, natural environment, and resource consumption. The evaluation process first divides the basic flow into different environmental impact types and multiplies each inventory data by the characterization factor to obtain the LCIA result. Then, there is the normalization process, where the LCIA result is divided by the total benchmark list to obtain the dimensionless LCIA result and finally weighted and summarized. This kind of normalization can directly compare the environmental impact caused by different objects and compare the different impact potentials based on the reference value of the region.

4.1.1.4 Interpretation

This stage mainly interprets the LCA results, including the analysis of the main problems identified; the inspection of the completeness, sensitivity, and consistency of the results; and finally, the project's conclusion and suggestions for improvement.

LCA helps to select products or processes with less environmental impact and identify the transformation of environmental impact from one medium to another, from one life cycle stage to another, from one substance to another, and from one country to another, which allows decision-makers to study the entire product system comprehensively and systematically.

4.1.2 Analysis of the Carbon Footprint and Energy Consumption of the Power Battery Manufacturing Process

Road transportation is one of the major Greenhouse Gases (GHGs) emissions contributors worldwide, which contributed 16% of the total GHG emissions in 2019 [2]. Most countries have reached a consensus that going electrification is the most effective and currently the best way to reduce the GHGs emissions in the transportation and mobility sectors. In 2021, a lot of countries, e.g. Italy, France, Spain, Norway, UK, and Sweden, and automotive Original Equipment Manufacturers (OEMs), e.g. Changan, BAIC group, Jaguar, Ford, VW, and Volvo, have set up a clear timetable for banning the sale of gasoline-powered vehicles in the world. The land transportation sector is experiencing a disruptive change where the new energy vehicles, including battery electric vehicles (BEV) and plug-in hybrid electric vehicle (PHEV), have rapidly taken up the market share. According to the latest data from Electric Vehicle (EV) volumes, the global EV market share has jumped to 8.3% in 2021 from 4.2% in 2020, and the total sales reached 6.75 million units [3].

Lithium-Ion Batteries (LIBs) have become the dominating battery technology for EVs, mainly because of their high-energy density and high charge-discharge efficiencies [4]. The typical life cycle of an EV power battery consists of mining, refining, cathode production, battery manufacturing, battery use, and end-of-life phase, which is schematically illustrated in Figure 4.2.



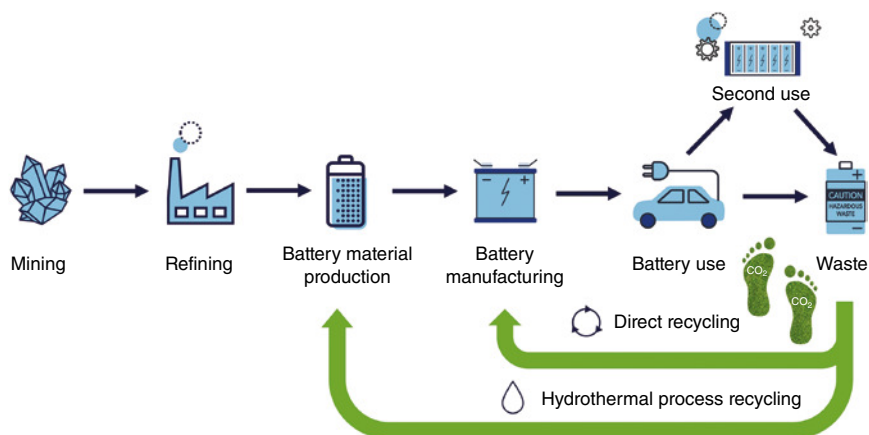


Figure 4.2 Life cycle of a typical EV battery. *Source:* Adapted from ReCell [5].

Although the use phase has contributed to the majority of carbon emissions due to the high intrinsic carbon nature from the electric grid, battery production, including the front-end mining, refining, and battery material production processes, also generates significant carbon emissions. Based on the study from Volvo, producing an XC40 EV (including battery production) will generate roughly 70% more carbon emissions than that of an XC40 internal combustion engine (ICE) [6]. With the increase of renewable energy share used for charging, the carbon emission during the use phase will gradually decrease; thus, the decarbonization of front-end production processes becomes more important.

Various researchers have extensively studied the life cycle environmental impacts of EV power batteries [7–12]; some of them can be even traced back to the early 2010s when EV was still at the infancy stage. Typically, various environmental impact categories, e.g. global warming, acidification, ozone depletion, photochemical smog, and eutrophication, were investigated in those LCA studies. Considering the current climate background, this chapter mainly focuses on Global Warming Potential (GWP) and Cumulative Energy Consumption (CES). In addition, since ternary lithium nickel cobalt manganese oxide (NCM) and lithium iron phosphate (LFP) monopolize the currently installed batteries in China's EV market, this chapter only compares and analyzes the carbon footprint of these two battery chemistries.

4.1.2.1 Carbon Footprint Analysis

Although it might differ in each study, the cathode material production process (take NCM811 as an example) and EV battery manufacturing process generally consist of similar major steps, as illustrated in Figures 4.3 and 4.4, respectively.

The carbon footprint of EV power battery is normally calculated based on 1 kWh battery capacity as the function unit and represented as kg CO₂-eq/kWh, whose values largely depend on the battery chemistry and the region where the batteries have been produced, as illustrated in Table 4.1.



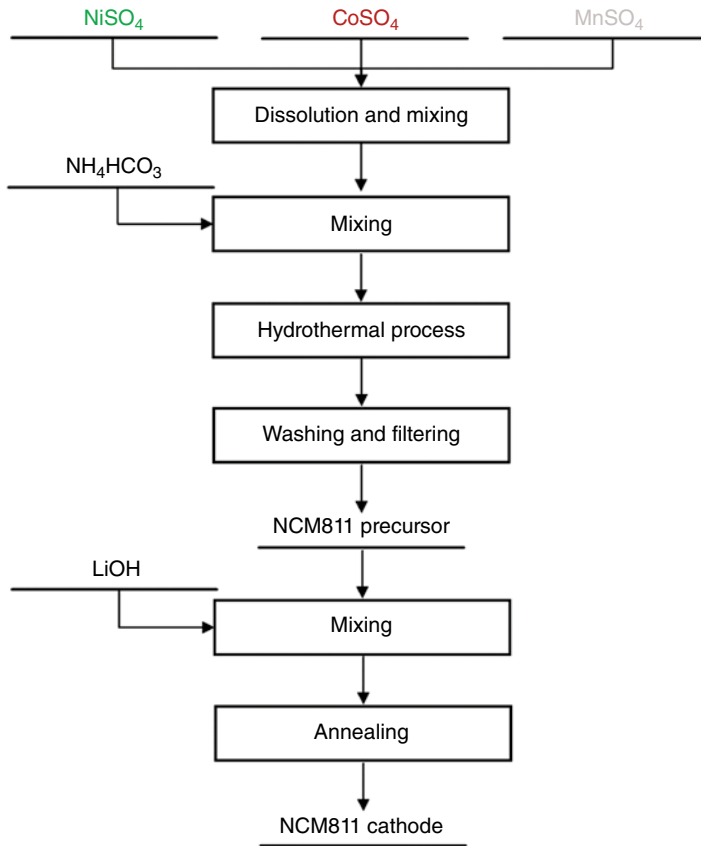


Figure 4.3 NCM811 battery cathode material production process. *Source:* Adapted from Widiyandari et al. [11].

In general, the carbon footprint of EV battery is the lowest in Europe regardless of the battery chemistry, followed by the United States, mainly because of their “greener” electric grid. LFP battery has a lower carbon footprint than that of NCM and lithium nickel cobalt aluminum oxide (NCA) batteries, though its energy density is normally smaller; this is because of the high carbon footprint generated from the NCM cathode materials and their front-end metal salts. The typical contributions of carbon emissions from different NCM EV battery components are illustrated in Figure 4.5.

4.1.2.2 Energy Consumption Analysis

The production and manufacturing process of products is a process of continuous accumulation of energy consumption. By demounting the product reversely, it can always be traced back to the original mineral/material on earth and the energy consumption in its subsequent production and processing. Therefore, the carbon footprint of a product is often positively correlated with the energy consumption used in its manufacturing process.





Figure 4.4 EV power battery production process. *Source:* Liu et al. [12] / with permission of Elsevier.

Table 4.1 Carbon footprint of EV power battery, kg CO₂-e/kWh of battery capacity.

	Europe	USA	China	South Korea	Japan
NCM111-C	56	60	77	69	73
NCM622-C	54	57	69	64	68
NCM811-C	53	55	68	63	67
NCA-C	57	59	72	67	70
LFP-C	34–39	37–42	51–56	46–50	50–55

Source: Gifford [13].

Dai et al. [14] from Argonne Laboratory split the whole process of 1 kWh NCM111 battery material production and battery production into electricity and fuel, as shown in Figure 4.6. As mentioned above, ternary cathode materials, cell production, and aluminum are the processes with the highest energy consumption and the highest carbon footprint input (Figure 4.5).

Therefore, if the same production process is used for a specific product, the final factor affecting the product's carbon footprint is the energy structure used in the processing of its raw materials and products.

Martin Beermann [15], from JOANNEUM Institute, compared the carbon footprint of cell production and battery pack assembly in China and Europe under the



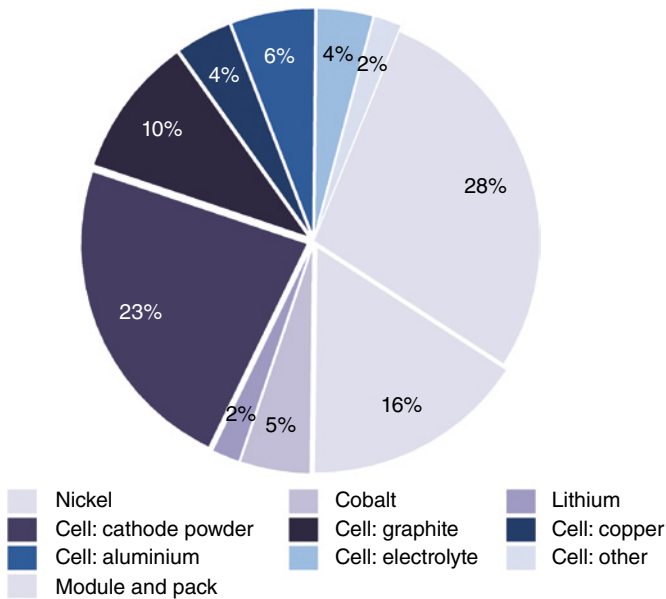


Figure 4.5 Carbon footprint of different EV battery components. *Source:* Adapted from Gifford [13].

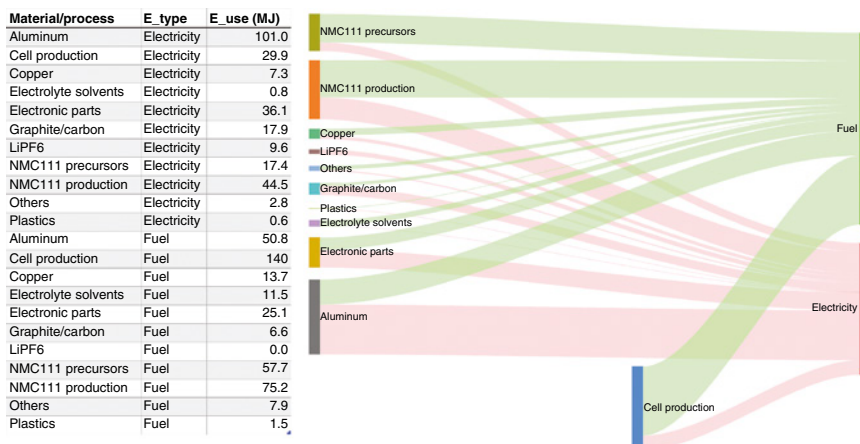


Figure 4.6 Energy consumption of 1 kWh NCM111 battery material and production process. *Source:* Dai et al. [14]/MDPI/CC BY 4.0.

same energy consumption of 55–65 kWh/kWh for 50 kWh NCM and LFP batteries, assuming 50% natural gas and 50% electricity consumption, as shown in Figure 4.7.

It can be clearly seen that the carbon footprint of cells and battery packs produced and assembled in Europe is only 2/3 of that in China, regardless of the battery chemistry. The main reason is that there is a big difference between China's and Europe's power grid structure. China's power grid in 2020 is mainly composed of thermal power (67.6%) and hydropower (17.8%), with a high carbon emission factor,



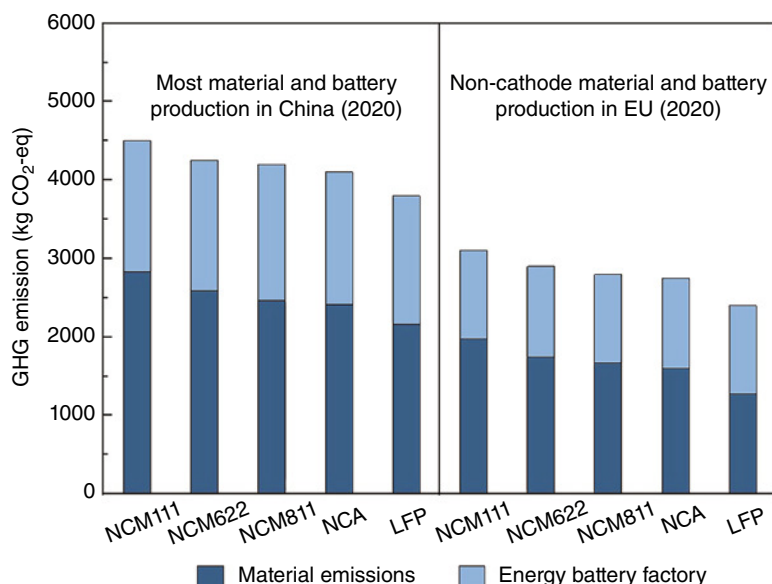


Figure 4.7 Comparison of the carbon footprint of 50 kWh cell production and battery pack assembly in China and Europe [15].

while Europe's power grid is mainly composed of a higher proportion of clean energy, such as nuclear energy (25.2%), scenery (14.4%), and biomass (5%). The proportion of thermal power is only 21.5%; therefore, the carbon emission factor is low.

Therefore, to reduce the carbon footprint of power batteries, one of the most critical factors is to reduce the energy consumption of their whole life cycle production and adopt greener and low-carbon electricity as much as possible.

With the rapid growth of power battery production capacity, it will generate considerable carbon emissions. Taking 80 kg CO₂-eq/kWh as the average value for worldwide cradle-to-gate battery production carbon emission and assuming that the global battery production capacity will reach 1 TWh in 2025, the production of those batteries will generate 80 million tons of CO₂-eq, which is approximately 0.25% of global GHG emissions (take 2020 data as reference). Recycling the spent batteries into battery raw materials is considered to have a high potential to greatly reduce carbon emission, as illustrated in Figure 4.8.

The LCA carbon footprint study of different power battery recycling technology has been detailed and discussed in Section 4.2.

4.2 Carbon Footprint of Different Power Battery Recycling Processes

The analysis and comparison in this section mainly focus on the standard pyrometallurgical recovery process and hydrometallurgical recovery process in industrial production.



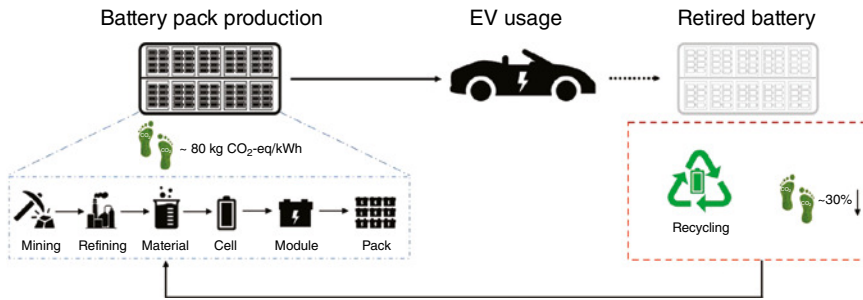


Figure 4.8 Role of battery recycling on the life cycle carbon footprint of power batteries.

4.2.1 Pyrometallurgical + Hydrometallurgical Method

Pyrometallurgy is widely used in metal smelting and metal recovery because of its short process and strong adaptability to raw materials. In lithium battery recycling, the typical representative of the pyrometallurgy process is Umicore in Belgium, whose process has been described thoroughly in Chapter 3. Therefore, it will not be described in detail here.

Although the pyrometallurgy process has its unique advantages, its high-temperature and high-energy consumption have brought more concerns considering today's climate-changing issues. In a recent paper, Mohammad Ali Rajaeifar et al. [16] compared the environmental impacts, particularly the carbon footprint of three pyrometallurgical and hydrometallurgical processes. Figures 4.9 and 4.10 have shown the flowchart of the pyrometallurgical + hydrometallurgical process and its corresponding carbon footprint analysis results. It can be clearly seen that except for the pretreatment + plasma melting process, other pyrometallurgical + hydrometallurgical processes will cause additional carbon emissions mainly resulting from the pyrometallurgical process with high energy consumptions.

4.2.2 Mechanical Pretreatment + Hydrometallurgical Method

Different from the high-temperature pyrometallurgical process, the low-temperature hydrometallurgical method is more widely used worldwide, especially in China. It generally consists of two major processes, i.e. a mechanical pretreatment process, followed by a hydrometallurgical process. The main purpose of the mechanical pretreatment is to dismantle battery packs into single battery cells and then further break down the cell into different components, e.g. shell, Al/Cu particles, diaphragm, and black mass (mixture of cathode and anode active materials), as schematically illustrated in Figure 4.11.

The most important output of the pretreatment process is the black mass powder, while the by-products, i.e. shell (steel or Al), Cu/Al particles can be directly sold to qualified suppliers as secondary materials and remanufactured into intermediate steel, aluminum, or copper products. Normally, no additional chemicals are needed in the pretreatment process; therefore, the carbon footprint mainly comes from the



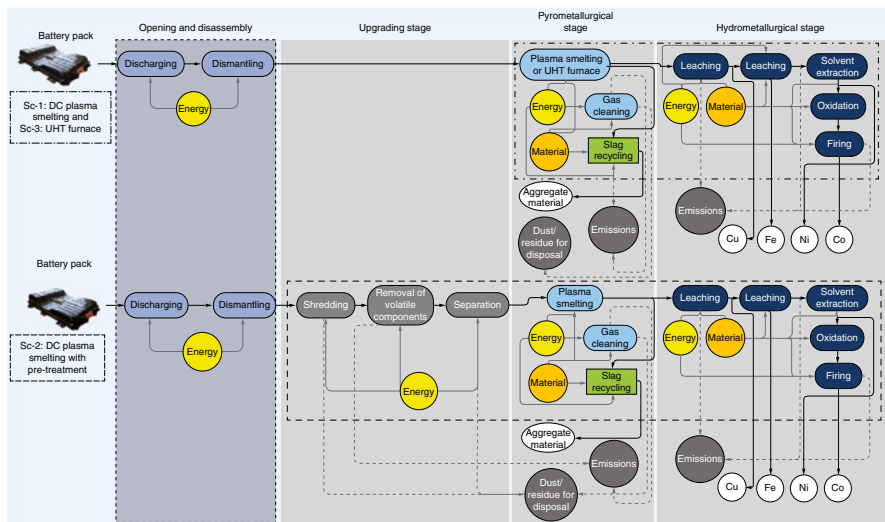


Figure 4.9 Three different pyrometallurgical+hydrometallurgical process flows of spent power battery recycling. Source: [16] Mohammad Ali Rajaeifar et al., (2021), John Wiley & Sons.



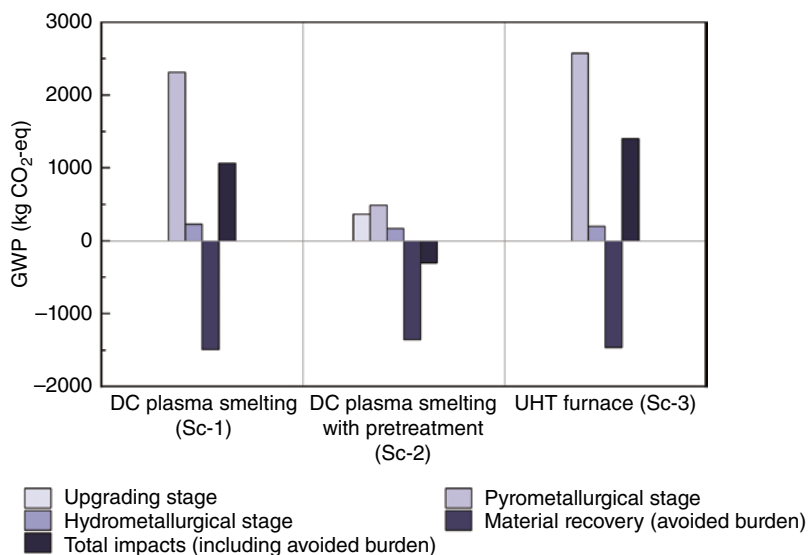


Figure 4.10 Carbon footprint analysis of three pyrometallurgical + hydrometallurgical spent power battery recycling processes. *Source:* Rajaeifar et al. [16]/John Wiley & Sons/CC BY 4.0.

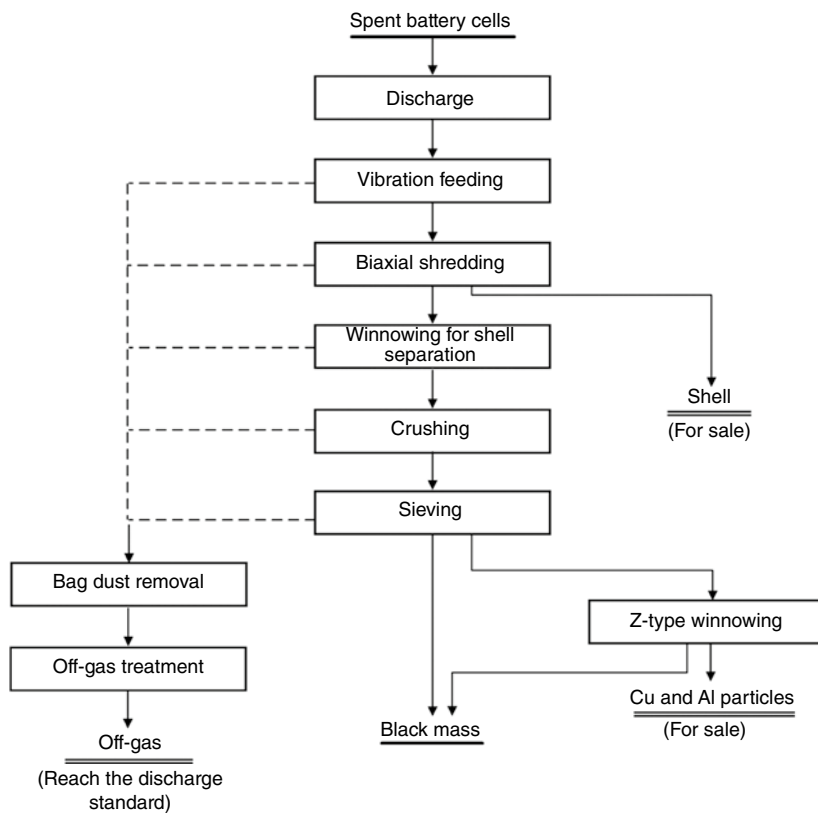


Figure 4.11 Process flow of battery mechanical pretreatment.



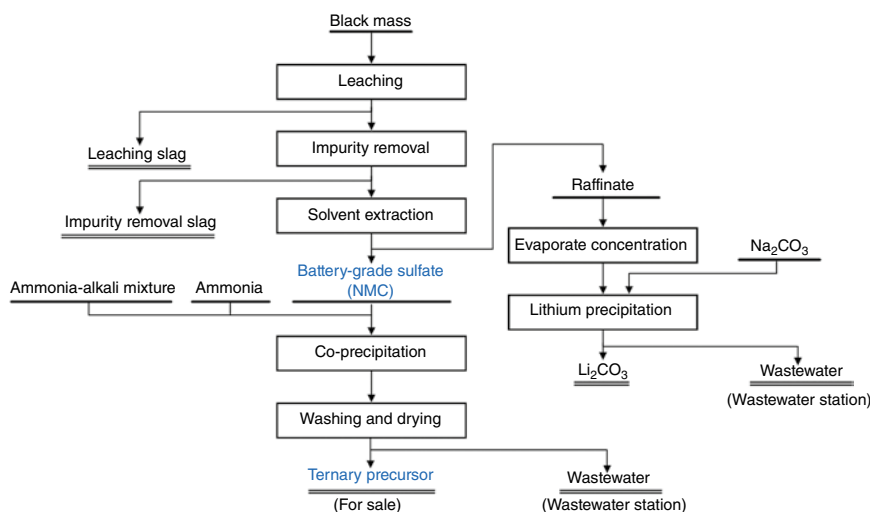


Figure 4.12 Process flow of black mass hydrometallurgical process.

energy consumption. The energy consumed in the pretreatment is mostly from the electricity that has been used, and sometimes natural gas as well if adopting a low-temperature pyrolysis process.

The black mass will be further treated in the successive hydrometallurgical process to extract the valuable metals, i.e. nickel, cobalt, lithium, and manganese. Depending on the capability of the factory, those valuable can be produced into metal sulfate or directly into ternary precursor, and the process is schematically illustrated in Figure 4.12.

Unlike the mechanical pretreatment, a considerable amount of chemicals is used during the hydrometallurgical process, especially acids and alkalis. Thus, the carbon footprint, in this case, comes both from the intrinsic carbon footprint of those chemicals and the energy consumption, i.e. the electricity and low/medium pressure steam.

Marit Mohr et al. [17] have carried out an LCA to evaluate the ecological benefits of LIB recycling processes on different battery chemistries, i.e. NCA, NCM, and LFP. In their study, they compared current hydrometallurgy and the advanced hydrometallurgy processes, where electrolyte and graphite are recycled in the later process. Their study has considered both GWP and abiotic resource depletion potential (ADP) (abiotic resource) perspectives, and their GWP results are shown in Figure 4.13.

As can be clearly seen from Figure 4.13, regardless of battery chemistries, both current hydrometallurgy and advanced hydrometallurgy processes can result in net GHG reduction when compared with production without the recycling process. The GHG reduction is far more significant for NCA and NCM battery chemistry, where the net reduction is around 20% for the current hydrometallurgy process and nearly 30% for the advanced hydrometallurgy process; meanwhile, it is only 4% and 12%, respectively, when considering LFP.



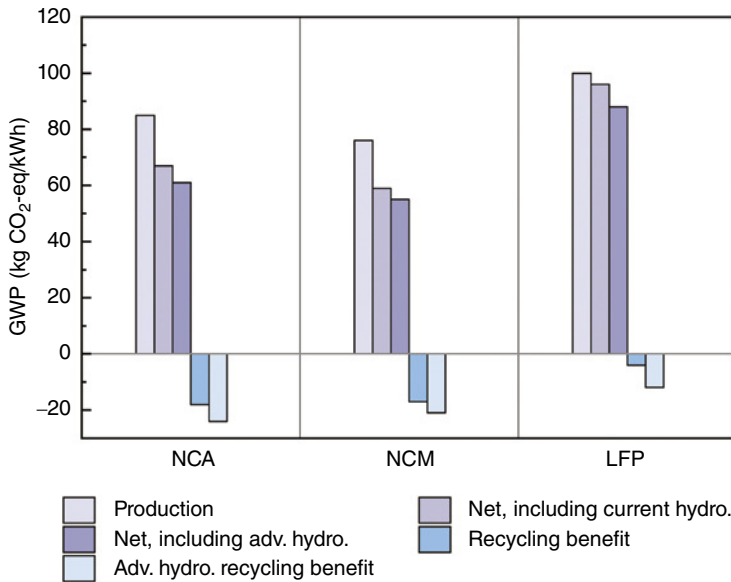


Figure 4.13 “Net” environmental impacts after subtracting recycling benefits.
Source: Adapted from Rajaeifar et al. [16].

The net GHG reduction mainly gains from the credit of those products generated during the recycling process, as illustrated in Figure 4.14.

As can be seen in Figure 4.14, the recycling of aluminum has generated the largest GHG benefit, followed by nickel, copper, and cobalt compounds. It makes sense since all those materials take up considerable mass fraction in the battery, and the production of those materials from virgin mineral ore usually experiences a long process and requires massive energy consumption. For NCA and NCM batteries, most of its valuable metals, i.e. nickel, cobalt, manganese, and lithium, have been recycled during the hydrometallurgy process, while for the LFP battery, only lithium has been recycled, thus generating less environmental benefits (Figure 4.13).

4.2.3 Direct Recycling Method

Direct recycling is a low-temperature and low-energy recycling process of spent lithium battery treatment using various physical and chemical methods to separate different cell constituents. Besides metal foil, almost all chemicals, active materials, and highly valuable metals can be recovered with this method. Qiang Dai (from the Argonne Laboratory) et al. [18] compared the GHG emission intensity generated by the three processes of hydrometallurgy, pyrometallurgy, and direct recovery. Studies have shown that the GHG emissions produced in the direct recovery process are significantly lower than those in the other two processes. This may be due to the simplification of the process flow, avoiding the input of more raw materials and more energy consumption.



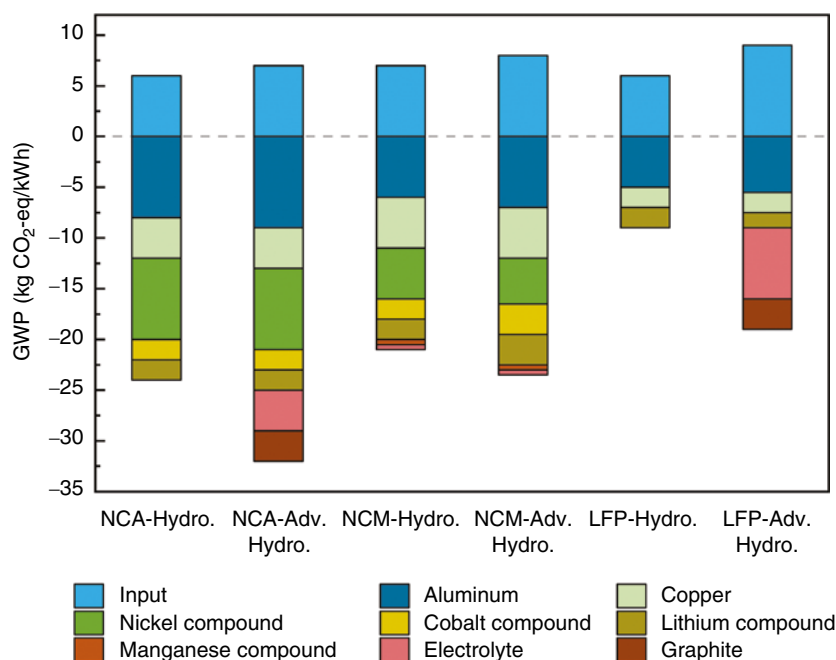


Figure 4.14 Environmental benefits, broken down to the contribution of each components. Source: Adapted from Rajaeifar et al. [16].

4.3 Best Power Battery Recycling Technology from a Life Cycle Carbon Footprint Perspective

Recycling will be an essential complement to the future raw material supply and significantly reduce power batteries' carbon footprint in EV. Nevertheless, current hydrometallurgical recycling technology, specifically the solvent extraction system, is not really developed and optimized for the high nickel content battery system, which often involves a long process, resulting in higher chemical and energy consumption.

To overcome the above issues and make the battery recycling process more efficient, Botree Cycling has developed an innovative and extremely short hydrometallurgical process to deal with the high nickel content battery system. The comparison of the main process flowchart between the market and the one developed by Botree Cycling is illustrated in Figure 4.15.

Botree employs a much shorter and more straightforward process flow when compared with the typical process in the market. The significant differences are summarized as follows:

- The typical process uses $\text{H}_2\text{SO}_4 + \text{H}_2\text{O}_2$ to dissolve all the cell components fully, while Botree employs the selective leaching method, which can achieve lithium pre-extraction.



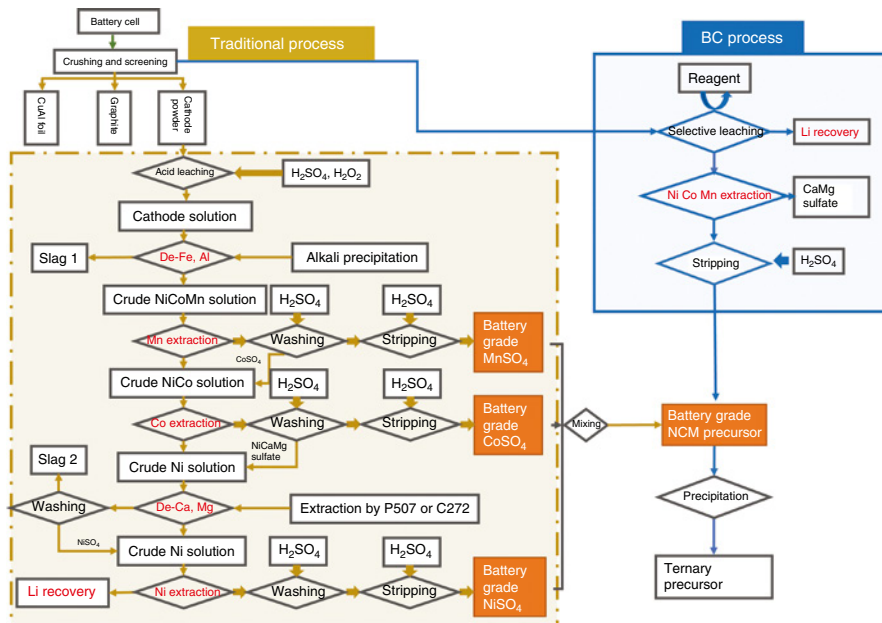


Figure 4.15 Comparison between traditional and Botree hydrometallurgical recycling processes.



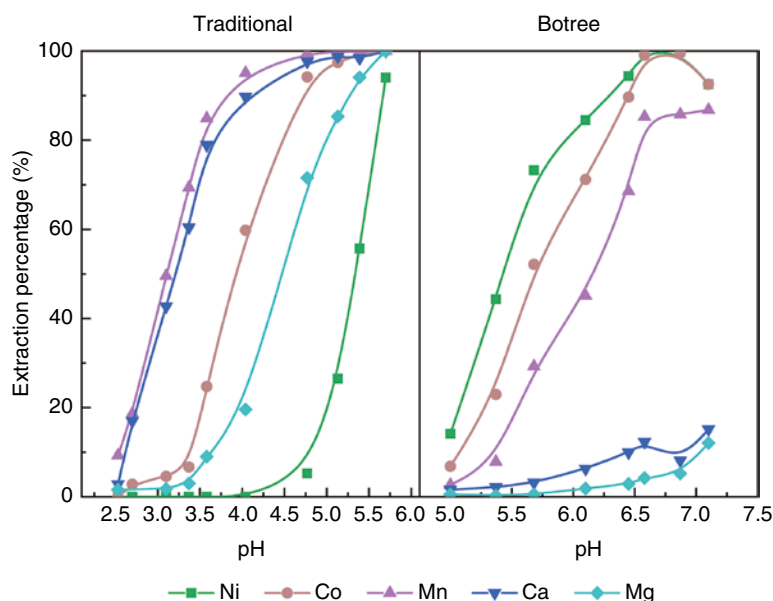


Figure 4.16 Selectivity comparison of the extractants from the traditional process and Botree Cycling.

- The typical process extracts Mn, Co, and Ni metals as sulfate salts at separate stages, while Botree employs a co-extraction method to extract Ni, Co, and Mn metals as metal sulfate salts at once.
- The typical process adds MnSO_4 , CoSO_4 , and NiSO_4 salts together to produce the NCM precursor, while in Botree's process, the NCM precursor can be directly produced from the Ni, Co, and Mn mixed solution.

The core of Botree's short processes is the newly developed extractant system. A traditional extractant has poor selectivity over Mn, Co, and Ni with impurity ions like Ca and Mg. Thus, additional impurity removal steps must be applied between each metal extraction process, resulting in a rather long flow. Instead, Botree's new extractant system has high selectivity over Ni, Co, and Mn with Ca and Mg impurity ions, as illustrated in Figure 4.16, which can achieve efficient Ni, Co, and Mn extraction, while leaving Ca and Mg in the aqueous solution.

Based on the pilot results, Botree's new process can reduce the overall energy and chemicals consumption by at least 10% and 15%, respectively. Since the consumed energy and chemicals mainly contribute to GHG during the recycling process, this new process will result in at least a 10% lower carbon footprint than the current ones, thus further increasing the carbon credit benefits.

As more companies like Botree Cycling have entered the power battery recycling field, more efficient, targeted, and intelligent solutions are expected to be developed in the near future, which helps build a greener and more sustainable power battery industry chain.



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5

Laws, Regulations, and Standards for Battery Recycling

5.1 Laws and Regulations Regarding Battery Recycling in Various Countries

The environmental impacts, recovery strategies, and recycling methods have been researched, and relevant laws and regulations have been formulated to promote the recovery and recycling of spent batteries in developed countries, especially in European countries.

In developed countries, the battery recycling is effectively supervised through a comprehensive legislative system and advanced recycling technology, and the recycling law system usually is built in the structure of “basic law – comprehensive law – special law” [1]. The “basic law” defines the guiding principles; the “comprehensive law” defines the links and frameworks of the recycling field; and the “special law” gives specific management measures and regulations.

5.1.1 The European Union

Currently, the European Union (EU) has established a recycling network of portable batteries and is exploring the feasibility of its application to the recycling of traction batteries and energy storage batteries for electric vehicles. EU has formulated a more comprehensive legal system in the field of power battery recycling, mainly including three categories: (i) battery directives; (ii) regulations for disposal of hazardous substances; and (iii) regulations for recycling of end-of-life vehicles. Among these directives and regulations, the Council Directive 2006/66/EC (transformed into the decree for each member state on 26 September 2009 and amended in December 2013), which is directly related to battery recycling, covers all types of batteries and requires the battery manufacturers to establish a recycling system of spent batteries for electric vehicles. Since 2008, the EU has taken mandatory measures to recycle spent batteries at the battery manufacturers’ costs (Table 5.1).

In addition to a large number of basic framework regulations for the treatment of waste, the EU has many extremely important waste disposal processes for these regulations. Batteries and accumulators are considered to be environmentally



Table 5.1 EU directives and regulations on the life cycle of batteries.

EU directive and regulation	Year	Description
Battery Directive 91/157/EEC	1991	EU Council Directive 91/157/EEC on batteries and accumulators containing certain dangerous substances
Battery Directive 93/86/EEC	1993	To adapt to technological progress, the Directive was developed to supplement the Battery Directive 91/157/EEC. The Directive specifies the labeling requirements for batteries. The labels shall give customers an accurate indication about the batteries isolated from household waste during recycling
Battery Directive 98/101/EG	1998	To adapt to technological progress, the Directive was developed to supplement the Battery Directive 91/157/EEC. The Directive reduces the limits of hazardous substances and draws up the draft of the subsequent directive
Battery Directive 493/2012	2014	This Directive shall apply to recycling processes of all types of spent batteries and accumulators. In addition, this Directive specifies that the producers of industrial batteries and accumulators or the third parties acting on their behalf can't refuse to take their responsibility for battery recycling for any reason; all spent batteries collected from end consumers must be recycled, and landfill is prohibited; the recycling efficiency of lead batteries and other batteries, including lithium-ion batteries, shall not be less than 65% and 50%, respectively; the battery system must be easily disassembled, and the chemical components and disassembly method of the battery must be marked in a prominent place of the battery system
Battery Directive 2006/66/EC	2006	The primary objective of this Directive is to minimize the environmental impact of batteries, accumulators, and spent batteries and accumulators and thus protect and improve the environment
Battery Regulation (EU) 2019/1020	2019	The Regulation (EU) 2019/1020 aims to improve the operation of the free movement of goods principle by strengthening the market surveillance of products covered by EU harmonized legislation. This will guarantee the health and safety of the people in general premises and workplaces and protect the consumers, environment, public security, and other public interests

relevant products; hence, the EU has issued the regulations for their production and treatment at their end of life. So far, the guidelines from the legal instruments of EU have essentially been used as a form of action. That also explains why spent batteries are recycled by EU member states in very different ways. In practice, it is preferred to issue a regulation rather than a directive when there is any doubt about the freedom of form. Since 1980, several EU countries have promulgated national regulations regarding dangerous substances such as mercury, cadmium, and lead. However, it shall be noted that the various regulations, including bans and labeling requirements, caused market distortion and hindered the development of the common market. As a consequence, the *Directive 91/157/EEC on Batteries and*



Accumulators Containing Dangerous Substances was issued to harmonize the laws of different countries. Later, to adapt to technological progress, the Directive was developed to supplement the Battery Directive 93/86/EEC. The Directive specifies the labeling requirements for batteries. The labels shall give customers an accurate indication about the batteries isolated from household waste during recycling. Subsequently, for the same purpose, the Battery Directive 98/101/EC was developed. This Directive reduces the limits of pollutants and draws up the draft of the subsequent directive. These directives on batteries and their disposal schemes are based on the Framework Directive issued in 1975 (Directive 75/442/EEC) and its amendment (Directive 91/156/EEC) issued in 1991 and Resolution 90/c122/02. This Framework Directive defines the terms “waste” and “waste treatment” and specifies that the member states have the obligation to avoid, recycle, and/or treat the waste, especially preferably avoid the waste. The various directives and regulations on batteries are explained.

5.1.1.1 Directive 91/157/EEC

The *Directive 91/157/EEC on Batteries and Accumulators Containing Dangerous Substances* promulgated on 18 March 1991 covers all the batteries containing more than 25 mg of mercury per cell and the alkaline-manganese batteries containing more than 0.025 wt% mercury, except for alkaline-manganese batteries containing more than 0.025 wt% cadmium and more than 0.4 wt% lead. The guideline prohibits certain batteries launched into the market. Furthermore, the guideline stipulates that the batteries and accumulators must be easily removed from the devices by the consumers at the end of their service life. To facilitate recycling, batteries shall be isolated from accumulators during recycling. In addition, the Directive gives some requirements for labeling and stipulates that the member states can organize a waste treatment system if necessary, thus saving raw materials and avoiding environmental pollution of dangerous substances such as mercury, cadmium, and lead.

5.1.1.2 Directive 93/86/EEC

To adapt to technical progress, the Directive 93/86/EEC was issued on 4 October 1993 to supplement the Directive 91/157/EEC. The Directive stipulates that in accordance with Article 4, Paragraph 2 of the Battery Directive 91/157/EEC, a detailed labeling system must be developed to record the battery labeling date, process, and person. The Directive also points out that the member states have to describe the information of the label to the public. The measures in the Directive are in line with the EU's initiative that its waste-related legislation shall adapt to technological progress.

5.1.1.3 Directive 98/101/EC

To adapt to technical progress, Directive 98/101/EC was issued on 22 December 1998 to supersede Directive 91/157/EEC. In the explanation of the Directive, it is clearly stated that it is necessary to make a reference to Article 10 of the Battery Directive 98/101/EC, which states that after a period of time, the EU Parliament must adjust certain elements in accordance with the prescribed procedure to adapt



to technological progress. Therefore, on the one hand, the Battery Directive 98/101/EC, which extends the scope of the Directive, reduces the limit of mercury content in batteries and accumulators, and takes the limit as the applicable criterion, came into effect from 1 January 1999. On the other hand, the Directive stipulates that the batteries and accumulators with the mercury content above the specified limit must not be launched into the market from 1 January 2000 latest taking the transitional period into account. In addition, the Directive stipulates that the batteries in devices are included in the scope to prevent avoidance or simplified treatment.

5.1.1.4 Council Directive 2006/66/EC

This Directive comprehensively reflects the EU legislation on spent batteries. It aims to protect, maintain, and improve the environment quality by minimizing the negative impact of batteries and accumulators and spent batteries and accumulators. It also ensures the smooth functioning of the internal market by harmonizing the requirements of each party for launching the batteries and accumulators to the market. With some exceptions, it applies to all batteries and accumulators, regardless of their chemistry, size, or design.

To achieve these objectives, the Directive prohibits the sales of the batteries containing certain hazardous substances, proposes the establishment of a high-level recycling system, and sets the targets for recycling activities. It also gives the requirements for battery labeling and easy removal of the batteries from their devices.

In addition, the Directive aims to improve the environmental impact of all participants in the life cycle of batteries and accumulators, including manufacturers, distributors, and end users, especially the operators treating and recycling the spent batteries and accumulators. The manufacturers and users of batteries and accumulators are responsible for the management of the disposal and recycling of the batteries and accumulators they have launched into the market.

5.1.1.5 Regulation (EU) 2019/1020

The regulation covers market surveillance of non-food products (“industrial products”) launched into the internal market and bound by the EU harmonized act. It lists a number of new provisions. For example, it:

- requires the manufacturers to designate an Authorized Representative within the Union to contact the market surveillance authorities more closely;
- specifies the cooperation between competing institutions;
- defines the member states’ obligations in the organization of market surveillance within their territory;
- provides a series of powers for market surveillance authorities to:
 - check the data and documents;
 - carry out field survey;
 - make a trial order;
 - give mystery visit and do shopping;
 - withdraw and destroy products, impose penalties, and recover the profits.



- points out that Union may designate the testing facilities;
- makes the procedure for mutual assistance in form of requests for information acquisition and enforcement and allows one member state for use of evidence obtained by another one;
- strengthens the customs controls on the products into the Union market;
- establishes the Union Product Compliance Network within the UC to coordinate the enforcement tasks between member states.

The purpose of the regulation is to require the companies not founded in EU countries to designate an economic operation representative within the EU to undertake the broad obligations to correct violations.

5.1.2 Typical European Countries

Germany has enacted some rules and regulations for spent batteries (Table 5.2)

5.1.2.1 Germany

Table 5.2 Rules and regulations in Germany for batteries.

Rules and regulations	Year	Description
KrW-/AbfG	1994	According to chapter 1 of the <i>Circular Economy Regulation</i> , the purpose of the law is to promote the circular economy to protect natural resources and ensure the environmental management of waste. As a basis for subsequent regulations, this regulation stipulates those specific types of waste shall be treated in cascade (Para.1, chapter 4 of <i>Circular Economy and Waste Regulation</i>)
Batterieverordnung	1998	Battery Ordinance aims to reduce the input of the batteries containing harmful substances to the market and specifies the responsibility for recycling to reduce the pollutant discharge from spent batteries
BattG	2021	The Battery Regulation aims to recycle more batteries and accumulators because they contain not only valuable materials, but also substances that are hazardous to environment and health

5.1.2.1.1 KrW-/AbfG

The *Circular Economy and Waste Regulation* (KrW-/AbfG) represents the implementation of Directive 91/156 and thus converts the recommendations of this Directive and the principles for waste treatment in Directive 75/442 into a binding national law. The regulation also provides a basis for the subsequent classification of waste in specific types of waste treatment regulations: The priority is to strive to avoidance of waste generation. If inapplicable, the resulting waste must be recycled or reused for energy recovery, i.e. to generate secondary materials or energy. Another key point is that the legislation also introduces the basic concept of “circular economy” in



Section 4. The regulation not only stipulates the use and recycling of waste, but also the production responsibility in Part III, and encourages the manufacturers to minimize waste as early as possible in the product design phase. Therefore, the manufacturers are obliged to take measures to prevent waste generation in the product development process. To implement these general provisions, Paragraph 1, chapter 24, states that the government will likely force the manufacturers to withdraw their products and take appropriate measures for standard compliance, such as a deposit system.

5.1.2.1.2 *Batterieverordnung*

One of these statutory ordinances to supplement the KrW-/AbfG is the Battery Ordinance, which expressly refers to the KrW-/AbfG. In addition to the implementation of the circular economy concept in the battery sector, the ordinance aims to generally prohibit the batteries containing pollutants and promote the production of recyclable batteries.

Batteries are divided into three classes in this ordinance:

- Batteries containing pollutants: This category includes the batteries with over-limit of mercury, cadmium, or lead content.
- Starter batteries: Refer to these accumulators which are “usually used in motor vehicles for starting, lighting and ignition.” A recycling system has been built for such batteries for a long time, so they aren’t bound by the provisions of this ordinance.
- Other batteries: All batteries that do not fall into the above categories.

5.1.2.1.3 *BattG*

The Battery Regulation (BattG) classifies the battery products into the following three categories:

- Industrial batteries (for commercial and agricultural purposes, and as traction batteries for electric and hybrid vehicles).
- Automotive batteries (for ignition, starting, and lighting of vehicles).
- Appliance batteries (portable encapsulated batteries).

Some of these battery products are nonrechargeable (primary batteries) and some are rechargeable (secondary batteries and accumulators); some can be installed and some can be used alone. In the BattG, a manufacturer is defined as any company that initially launched the batteries or the devices with built-in or enclosed batteries to German market for distribution, consumption or use, or that sells the batteries from unknown manufacturers in Germany. The BattG requires the manufacturers to register their batteries and accumulators before sales, label the batteries containing hazardous substances, report the volume of sales at a regular interval, and offer suitable facilities to recycle the batteries they have launched to the market in an environmentally acceptable way.



5.1.2.2 Norway

We listed some relevant Norwegian rules and regulations on battery recycling (Table 5.3).

Table 5.3 Rules and regulations in Norway for batteries.

Council directive and regulation	Year	Description
H.R.2853 – Battery Recycling and Research Act	1989	<i>Battery Recycling and Research Act</i> issued in 1989 amends the <i>Solid Waste Disposal Act</i> and points out that spent lead–acid batteries shall be disposed of only by the recycling method specified in the Act
H.R.1510 – Battery Recycling and Research Act	1991	Amends the H.R.2853 – <i>Battery Recycling and Research Act</i>
H.R.1808 – Lead Battery Recycling Incentives Act	1993	Amends the <i>Solid Waste Disposal Act</i> to direct the managers of the Norwegian Environmental Protection Agency (EPA) to issue the regulations for production, transportation, storage, recycling, and disposal of spent lead–acid batteries
H.R.1522 – Lead Battery Recycling Incentives Act	1995	Amends the H.R.1808 – <i>Lead Battery Recycling Incentives Act</i>
S.2157 – Lead–Acid Battery Recycling Act	1996	<i>Lead–Acid Battery Recycling Act</i> – Amends the <i>Solid Waste Disposal Act</i> to prohibit any individual to dispose of lead–acid batteries by landfill or incineration
S.3356 – Battery and Critical Mineral Recycling Act	2020	This bill generally stipulates the recycling of batteries, especially directs the Department of Energy to provide grants to (i) the entities for battery recycling research, development, and demonstration projects and (ii) state and local governments for battery collection, recycling, and reprocessing programs

5.1.2.3 Netherlands

The rules and regulations of Netherlands for batteries shown in Table 5.4.

Table 5.4 Rules and regulations of the Netherlands for batteries.

Council directive and regulation	Year	Description
MJZ2001120 768	2001	Resolution on managing the full life cycle of a vehicle
National waste management plan LAP3	2019	As an instrument for the circular economy; the NWMP3 consists of a policy framework and sector plans
Dutch Battery Management Decree (2008)	2008	This decree contains all the statutory provisions for the processing of batteries. The regulations apply to all batteries, including starter batteries and drive batteries from vehicles



5.1.2.4 The United States

The United States classifies the spent batteries into two categories: (i) hazardous batteries, including zinc–manganese batteries, lithium primary batteries, lithium-ion batteries (LIBs), and nickel–metal hydride batteries (not including California), and (ii) nonhazardous batteries, including button batteries, nickel–cadmium batteries, silver oxide batteries, enclosed lead–acid batteries, and automobile lead–acid batteries. The United States Environmental Protection Agency (USEPA) requires that hazardous batteries be collected according to the standardized procedures, and nonhazardous batteries can be disposed of as household garbage. Another country, Canada, manages the battery recycling at the provincial level. For instance, in British Columbia, both primary and secondary batteries are regarded as dangerous solid waste, and Call2Recycle, a battery recycling program organized by the Rechargeable Battery Recycling Corporation (RBRC), is regarded as an official program for the management of waste batteries; Cell2Recycle volunteers collect secondary batteries and Ontario’s environmental protection department collects primary batteries; in Quebec, the official program for management of spent batteries is Appel a Recycler MD.

The United States manages the recycling of spent batteries mainly by formulating the environmental regulations and enhancing the market supervision. For the recycling of spent batteries, federal governments, state governments, and local governments have established a comprehensive legal system of laws for nickel–cadmium batteries, small-sealed lead–acid batteries, mercury-containing batteries, and all other types of batteries in the United States.

In addition, the United States classifies the batteries into mercury-containing batteries, nickel–cadmium batteries, nickel–hydrogen batteries, lithium batteries, etc. based on the hazardous substances. In the United States, nickel–hydrogen batteries, LIBs, and polymer LIBs are generally considered harmless, although LIBs may be harmful before being fully discharged. Therefore, the above-mentioned types of batteries are not listed in the regulations for battery recycling in the United States (Table 5.5).

In addition to the Act regarding battery recycling stipulated by the federal government, the state governments shall issue the policies that are adaptable to the Resource Conservation and Recovery Act (RCRA) to ensure its implementation. The specific policies are as follows.

5.1.2.4.1 State Management Plans for Waste

The RCRA requires that the USEPA must issue basic requirements and guidelines for the state governments to formulate and implement their management plans for waste. The state governments shall prepare and submit the plans, which shall give the detailed measures and schedules to achieve the goals listed in the RCRA, to the EPA for approval. The plan shall have the effective period of at least five years and define the specific obligations for the state and local governments.

5.1.2.4.2 State Management Plans for Hazardous Waste Producers

According to the RCRA, the production of hazardous waste is the first link of the “from-cradle-to-grave” management system. All hazardous waste producers must



Table 5.5 Macropolicies regarding battery recycling in the United States.

Code	Year	Content
Resource Conservation and Recovery Act (RCRA)	1976	This Act specifies that spent nickel–cadmium, lead–acid, silver–oxide, and mercury-containing batteries are hazardous waste, and LIBs are potentially hazardous waste. The hazardous waste like lead–acid batteries is tracked and documented in the life cycle “from cradle to grave.” Enterprises must apply for a permit to recycle, store, transport, and dispose of hazardous waste. Moreover, the government monitors the recycling procedure of the enterprises by the permits and has the power to require the enterprises to gradually remove the previous pollution
Clean Air Act	2013	This Act lists lead as one of six standard pollutants and creates a set of standards for control and management of lead emissions
Clean Water Act	1977	This Act specifies all the indices of pollutants in the discharged water and states that only the discharge with a wastewater discharging license can discharge water into sewers or wastewater treatment plants
Mercury-Containing and Rechargeable Battery Management Act	1996	This Act mainly regulates the production, collection, transport, and storage of spent nickel–cadmium batteries, small sealed lead–acid batteries, and other rechargeable batteries. It also specifies that the batteries sold in the American market must meet the uniform labeling requirements so that consumers can assist in battery recycling. Moreover, this Act prohibits the sale of alkaline–manganese batteries containing mercury, zinc–carbon batteries (containing mercury that was intentionally introduced), and mercuric-oxide batteries
Batteries Act		Proposed and formulated by the Battery Council International (BCI), this Act expands the obligations of the manufacturers and requires the consumer who purchases a new one and does not return the spent automotive battery must pay a deposit

be obliged to determine whether the waste they produce is hazardous or not, and monitor the disposal of hazardous waste. Considering that different producers produce different amounts of waste and bring different levels of risk to the environment, the RCRA hierarchically manages the amounts of hazardous waste they produce. Moreover, hazardous waste producers cannot be exempted from the liabilities by signing agreements with third parties for transport or disposal. Even if the illegal disposal of waste is caused by the behaviors of a third party, the waste producer still undertakes joint responsibilities for the consequences caused by the illegal disposal.

5.1.2.4.3 State Management Plans for Hazardous Waste Transporters

The transporters of hazardous waste are supervised not only by the RCRA but also by the Hazardous Materials Transportation Act. The EPA requires that hazardous waste transporters must apply for an EPA ID code to monitor the transport of hazardous waste. Meanwhile, hazardous waste is transported with a transfer manifest.



To promote the recycling of resources, the transport of certain recycled hazardous waste may not be restricted by the requirements of transfer manifest.

5.1.2.4.4 Management Plans for Treatment, Storage, and Disposal Facilities of Hazardous Waste

The EPA formulates the requirements for the management of treatment, storage, and disposal facilities (TSDF), and requires that TSDF must bear permits. The permits are issued by the EPA and/or the EPA authorized state government. The recycling activities are exempted by the RCRA; hence, the recycling of hazardous waste is conducted without permit or isn't bound by the management requirements for TSDF.

5.1.3 Typical Asian Countries

5.1.3.1 China

Table 5.6 Policies and regulations regarding battery recycling in China.

Regulation	Department	Date	Content
Technical Policy for the Recovery of Automobile Products	National Development and Reform Commission, Ministry of Science and Technology of PRC, State Environmental Protection Administration	6 February 2006	Provides guidelines for establishing a disposal and recycling system of used automobile products and requires automobile manufacturers to take the obligations to recycle and dispose of batteries in the automobiles they sold
Energy-Saving and New Energy Automobile Development Plan (2012–2020)	State Council of PRC	28 June 2012	Establishes a management system of cascade utilization and recycling of power batteries, encourages to set up battery recycling enterprises, and formulates the admission criteria for battery recycling enterprises
Technical Policy for the Recovery of Automobile Power Accumulators (2015)	National Development and Reform Commission, Ministry of Industry and Information Technology of PRC, Ministry of Science and Technology of PRC, etc.	5 January 2016	Guides the enterprises to properly design, produce, and recycle the power batteries, establishes a battery recycling system for upstream and downstream enterprises, and expands the obligations of the manufacturers
Requirements of the Industry Standards for Vehicle Power Batteries (2017) (Draft for Comments)	Ministry of Industry and Information Technology of PRC	22 November 2016	Specifies that the enterprises shall meet the policies and regulations for recycling of power batteries formulated by the national and the local governments

(continued)



Table 5.6 (Continued)

Regulation	Department	Date	Content
Technical Policy for the Pollution Prevention of Waste Batteries	Ministry of Science and Technology of PRC	26 December 2016	Gradually establishes an IT monitoring system of collection, transport, storage, recycling, and disposal of spent power batteries in new-energy vehicles
Interim Measures for the Administration of Recycling Power Accumulators of New Energy Vehicles	Ministry of Industry and Information Technology of PRC and other six departments	28 January 2018	Specifies the obligations in the design, production, and recycling of power accumulators, and the requirements for setting up a recycling enterprise
Implement Plan for the Pilot Program of Recycling Power Accumulators of New Energy Vehicles	Ministry of Industry and Information Technology of PRC and other six departments	28 January 2018	Establishes a batch of demonstrative production lines of decommissioned power batteries, demonstrative projects of efficient battery recycling, demonstrative enterprises, key technologies, and technical standards, and publishes a set of measures for recycling of batteries and accumulators
Interim Provisions on the Management of Traceability of the Recycling and Utilization of Power Accumulators of New Energy Vehicles	Ministry of Industry and Information Technology of PRC	3 July 2018	Regulates the information collection in the production, sales, utilization, and recycling of power batteries and monitors the implementation of obligations in the whole process
Requirements for Recycling Vehicle Power Batteries (Draft for Comments)	National Technical Committee of Auto Standardization	27 July 2018	Defines specific terms, general requirements, technical standards, and pollution prevention principles for recycling of vehicle power batteries
Construction and Operation Guides for the Service Facilities Involved in Recycling Power Accumulators of New Energy Vehicles	Ministry of Industry and Information Technology of PRC	31 October 2019	Proposes the qualification, construction, operation, safety, and environment requirements for the recycling service network of waste power batteries of new-energy vehicles
Requirements of the Industry Standards for the Comprehensive Utilization of Waste Power Accumulators of New Energy Vehicles (2019)	Ministry of Industry and Information Technology of PRC	2 January 2020	Specifies the requirements for layout, location, technical equipment and process, resource utilization and energy consumption, and environmental protection of the new-energy vehicle battery manufacturers

(continued)



Table 5.6 (Continued)

Regulation	Department	Date	Content
Administrative Measures for the Cascade utilization of Power Batteries of New Energy Vehicles (Draft for Comments)	Ministry of Industry and Information Technology of PRC	10 October 2020	Enhances the cascade utilization of power accumulators of new-energy vehicles and improves the comprehensive utilization efficiency of resources

Three of the above policies for battery recycling are particularly important for the industry. The three policies are as follows (Table 5.6):

In June 2012, the State Council issued the Energy-Saving and New Energy Automobile Industry Development Plan (2012–2020), which requires to formulate the management measures for recycling of power battery, establish a cascade utilization and recycling system for power batteries, guide the power battery manufacturers to recycling the spent batteries, set up battery recycling enterprises, and set strict standards for setting up such enterprises. This Plan is a true beginning of China's battery recycling policies.

In January 2018, the Ministry of Industry and Information Technology of PRC and other six departments jointly issued the *Interim Measures for the Management of Recycling and Utilization of Power Batteries of New Energy Vehicles*, which clarifies the division of labor for the design, production, and recycling of power batteries, and further specifies the qualifications of enterprises and the obligations of government departments on the cascade utilization and recycling of batteries. The Measures summarize the policies of battery recycling in all aspects and lay the foundation for the formulation of subsequent battery recycling policies in China.

In October 2019, the Ministry of Industry and Information Technology issued the *Construction and Operation Guides for the Service Facilities Involved in Recycling Power Accumulators of New Energy Vehicles*, which specifies the construction, operation, safety, and environmental protection requirements of the recycling service network of spent power battery of new-energy vehicle. The Guideline provides a legal basis for the storage and operation of waste batteries in the industry.

Furthermore, local governments in China have thoroughly studied recycling of power batteries and thus have formulated a set of policies and subsidies measures to encourage the industrial development. Taking Jiangsu Province as an example, China has 4.92 million new-energy vehicles and about 200 000 tons of scrapped power batteries, and Jiangsu Province has 298 000 new-energy vehicles, accounting for 6% of China's new-energy vehicles by the end of 2020. A rough calculation shows that Jiangsu has 12 000 tons of waste batteries. Jiangsu Provincial Government has fully recognized the large potential of the battery recycling industry and the potential damage to the environment caused by lack of disposal facilities of waste batteries, and thus has intensively issued the policies to promote the development of the battery recycling industry (Table 5.7).



Table 5.7 Major policies and events regarding battery recycling in Jiangsu.

Policy/event	Department	Date	Content
Implement Plan of Promoting the Application of New Energy Vehicles in Jiangsu Province During the Thirteenth Five Years	Jiangsu People's Government	01 January 2017	The Plan requires vehicle manufacturers to establish a recycling system of vehicle power batteries and formulate the recycling and disposal regulations so as to promote the collection and recycling of spent power batteries
Jiangsu was listed as one of the pilot provinces for battery recycling	Ministry of Industry and Information Technology of PRC	30 July 2018	The Ministry of Industry and Information Technology of PRC, the Ministry of Science and Technology of PRC, and the Ministry of Ecology and Environment of PRC jointly implemented the pilot program on recycling power batteries and listed Jiangsu as one of the pilot provinces
The construction of Nanjing Energy Storage Station was launched	Jiangsu Power Co., Ltd.	19 March 2019	The Nanjing Energy Storage Station is the largest energy storage station for cascade utilization of vehicle power batteries, with a battery recycling capacity of 75 000 kWh, including 45 000 kWh of LFP batteries and 30 000 kWh of lead-acid batteries
The China Industry Alliance for the Recycling of Power Accumulator of New Energy Vehicles was founded	Jiangsu Provincial Department of Industry and Information Technology	30 May 2019	The Industry Alliance for the Recycling of Power Accumulator of New Energy Vehicles was founded by the Jiangsu Branch of China Tower Co., Ltd. and State Grid Jiangsu Power Co., Ltd., which were also selected as the chairmen, and Nanjing Guoxuan Battery Co., Ltd., Kaiwo New Energy Vehicle Co., Ltd., Zhejiang Huayou Recycling Technology Co., Ltd., and Jiangsu Jing Tai Clean Energy Co., Ltd., which were selected as vice-chairmen
Reply to the Proposal No. 0910 of the Second Session of the Twelfth Conference of Provincial Committee of the Chinese People's Political Consultative Conference (CPPCC)	Jiangsu Provincial Department of Industry and Information Technology	18 June 2019	Establishes a system of collection, cascade utilization, and recycling of vehicle power batteries across the province, in the principle of cascade utilization before recycling

(continued)



Table 5.7 (Continued)

Policy/event	Department	Date	Content
Notice on Constructing Regional Power Battery Recycling Centers	Jiangsu Provincial Department of Industry and Information Technology	10 March 2021	Starts the construction of regional centers for decommissioned batteries. A regional center is a large station for collection, storage, and transfer of decommissioned batteries. One center is set up in a district to recycle the decommissioned batteries, including low-speed vehicle batteries, disposable lithium batteries, and non-standard batteries, and encouraged for dismantling, testing, cascade utilization, and recycling of the batteries
The implementation meeting of the piloting of power battery recycling in Jiangsu province was held	Jiangsu Provincial Department of Industry and Information Technology and Other Departments	26 March 2021	Jiangsu Provincial Department of Industry and Information Technology, Jiangsu Provincial Development and Reform Commission, Jiangsu Provincial Department of Science and Technology, Jiangsu Provincial Department of Commerce, Jiangsu Provincial Department of Ecology and Environment, Jiangsu Provincial Department of Transport, and Jiangsu Provincial Administration of Market Regulation jointly proposed to formulate a 5-yr plan for battery recycling in Jiangsu Province

Hu Zhengxin, deputy director of the Energy Conservation and Comprehensive Utilization Division of the Jiangsu Provincial Department of Industry and Information Technology, said that Jiangsu has basically established a power battery recycling network covering the whole province, which is dominated by the automobile 4S (sale, spare part, service, and survey) stores and maintenance companies. Moreover, 79 new-energy vehicle manufacturers and other battery recycling companies have set up 907 sites for battery recycling across the province [2]. In the section of cascade utilization of decommissioned batteries, Jiangsu Branch of China Tower Co. Ltd., State Grid Jiangsu Integrated Energy Service Co., Ltd., and Jiangsu Huizhi Energy Engineering Technology Innovation Research Institute Co., Ltd. have become the leading forces. In the section of battery recycling, GEM (Taixing) Co., Ltd. and Nantong Beixin New Energy Technology Co., Ltd. have emerged as major players.

Necessity of R&D for the development of the battery recycling industry has become the consensus of local governments. Therefore, besides Jiangsu, other provinces in China have introduced policies to encourage the development of the battery recycling industry (Table 5.8).



Table 5.8 Major policies regarding battery recycling in other provinces and cities.

Policy	Province/city	Date	Content
Interim Measures of Shanghai Municipality on Encouraging the Purchase and Use of New Energy Vehicles	Shanghai	20 May 2014	Provides vehicle manufacturers with the subsidy of 1000 yuan per recycled power battery
Interim Measures of Shanghai Municipality on Encouraging the Purchase and Use of New Energy Vehicles (Revised in 2016)		04 March 2016	Specifies that vehicle manufacturers shall recycle spent power batteries of vehicles and have the capacity of collection, recycling, and disposal of batteries whose amount is equivalent to the total amount of new-energy vehicle sold by the enterprises
Notice of the Guangzhou Municipal People's Government on Printing and Distributing the Interim Measures of Guangzhou on Promoting the Use of New Energy Vehicles	Guangdong	28 November 2014	Points out that Guangzhou shall establish the channels for recycling of vehicle power batteries to recycle and dispose of power batteries as specified
Implementation Opinions of the Guangzhou Municipal People's Government on Promoting the Use of New Energy Vehicles		28 March 2016	Explores to promote the recycling of spent power batteries by introducing a fund system, a deposit system, and mandatory measures
Implementation Opinions of Guangzhou Municipal People's Government on the Pilot Program for Recycling Power Accumulators of New Energy Vehicles		17 September 2018	Builds a batch of demonstration programs, cooperation models, association standards, leading enterprises, and key technologies to basically improve the recycling policies
Notice of the Shenzhen Municipal People's Government on Printing and Distributing the Several Policies and Measures on Promoting the Use of New Energy Vehicles in Shenzhen	Shenzhen	08 January 2015	Specifies that vehicle manufacturers are responsible for the mandatory recycling of power batteries, with a special provision of 20 yuan/kWh as recycling funds. The local government provides subsidies of no more than 50% of the audited collective funds

(continued)

Table 5.8 (Continued)

Policy	Province/city	Date	Content
Financial Policies of Shenzhen on Supporting the Promotion of New Energy Vehicles (2016)		02 September 2016	Specifies that vehicle enterprises are responsible for the recycling of power batteries, with a special provision of 20 yuan/kWh as recycling funds. The local government provides subsidies of no more than 50% of the audited collective funds and ensures the proper use of special subsidies
Plan of Shenzhen City on the Pilot Program for the Construction of the Monitoring System of the Recycling of Power Accumulators of New Energy Vehicles (2018–2020)		02 April 2018	Monitors the power batteries of new-energy vehicles that are included in the scope of subsidy policies
Administrative Measures of Beijing Municipality on Promoting the Use of New Energy Commercial Vehicles	Beijing	24 July 2017	Proposes to build a sound system of the recycling of spent power batteries and formulates feasible plans for the recycling of spent power batteries
Administrative Measures of Beijing Municipality on Promoting the Use of New Energy Commercial Vehicles		24 February 2018	Specifies that vehicle manufacturers shall take the main responsibility for recycling of spent batteries
Notice of Adjusting the Policies on Promoting the Use of New Energy Commercial Vehicles	Hefei	09 May 2017	Provides the awards of 10 yuan/kWh for vehicle and battery manufacturers who have established and operated a system of power battery recycling
Action Plan of Tianjin City on the Development of New Energy Industry (2018–2020)	Tianjin	30 October 2018	Promotes the construction of the technology development and recycling network for power batteries to take the lead in building a network of the recycling, trading, dismantling, and cascade utilization of power batteries across the city
Implement Plan for the Pilot Program of Recycling Power Accumulators in New Energy Vehicles in the Beijing–Tianjin–Hebei Region		19 December 2018	This Plan was formulated based on the requirements in the <i>Implement Plan for the Pilot Program of the Recycling of Power Accumulators of New Energy Vehicles</i> , with a combination of the status quo of the new-energy vehicle and power battery recycling industries in the Beijing–Tianjin–Hebei region

(continued)



Table 5.8 (Continued)

Policy	Province/city	Date	Content
Implement Plan of Sichuan Province for the Pilot Program of Recycling Power Accumulators of New Energy Vehicles	Sichuan	18 March 2019	Aims to increase the output value of the cascade utilization of power batteries to 500 million yuan and the output value of material recycling to 3 billion by 2020, and plans to build three demonstration bases, two demonstration projects, and three benchmark enterprises by 2020
Implement Plan of Hunan Province for the Pilot Program of Recycling Power Accumulators of New Energy Vehicles	Hunan	16 April 2019	Specifies that vehicle manufacturers shall take the main responsibility for the recycling of power batteries and build a shared recycling network, and introduces more than 80% of decommissioned and scrapped batteries of new-energy vehicles into the network

Table 5.8 shows that with the steady growth of the number of battery recycling enterprises in the recent years, the local governments have gradually reduced the financial subsidies and formulated the policies to guide and regulate the development of the battery recycling industry instead.

The Chinese government thinks much of the new-energy industry for the following four reasons:

- (1) China is poor in oil and thus can reduce its dependency on oil import by developing new energy such as battery and hydrogen energy, which is especially crucial in the context of Sino-US trade frictions;
- (2) Developing new energy can reduce environmental pollution and thus help China to fulfill its commitment to “peak carbon dioxide emissions by 2030, and reach carbon neutrality by 2060”;
- (3) Developing the new-energy industry can create jobs and tax revenue for the Chinese government. For instance, a plant with the capacity of 10000 tons per year recycled battery can create 100 jobs and annual tax revenue of 12 million yuan;
- (4) Especially in the sector of new-energy vehicles, developing new energy can facilitate the electrification of vehicles, which is a basis of intelligence, and provide opportunities for the Chinese automobile industry to overtake Europe, the United States, and Japan.

5.1.3.2 Japan

Japan started the recycling of spent batteries early and has formulated a relatively comprehensive legal system of battery recycling. Although there is no special



legislation on battery recycling, Japan has established a legal system for circular economy, consisting of basic laws, general laws, and special laws. The basic law is the *Basic Law for Establishing a Sound Material-Cycle Society*; general laws are the *Law for the Promotion of Effective Utilization of Resources* and the *Waste Management and Public Cleansing Law*; and special laws include the *Law for the Recycling of End-of-Life Vehicles*, the *Small Home Appliance Recycling Law*, and the *High-Pressure Gas Safety Act*. Among them, all general laws contain regulations regarding battery recycling and thus can be regarded as special laws at a higher level (Table 5.9).

In 1956, the industrial wastewater released from a chemical factory in Minamata, Kumamoto Prefecture, Japan, contained mercury and other hazardous substances, so the surrounding residents suffered from a neurological disease (thereafter named Minamata disease). Subsequently, all walks of life in Japan began to pay attention to the threat brought by mercury in waste batteries. After that, the incineration of batteries caused pollution to the atmosphere, further driving the Japanese government to discuss the management of waste batteries.

In 1978, the Battery Association of Japan, with the aim of controlling hazardous substances, including cadmium, established a recycling route of nickel–cadmium batteries for disaster prevention by cooperating with appliance stores and expanded the scope to other types of batteries. The *Law for the Promotion of Effective Utilization of Recyclable Resources*, which came into effect in 1991, specifies compact rechargeable batteries as reusable and recyclable products and the installation of battery recycling boxes. The *Law for the Promotion of Effective Utilization of Recyclable Resources*, which came into effect in 1993, clarifies three recycling channels for waste cadmium–nickel: nickel–hydrogen, lithium ion, and compact lead–acid batteries. In 2001, the *Law for the Promotion of Effective Utilization of Recyclable Resources* was renamed the *Law for the Promotion of Effective Utilization of Resources*, which specifies that for the manufacturers and sellers of batteries, the voluntary recycling of used compact rechargeable batteries is changed to mandatory recycling.

In addition, Japan has been promoting the collection and recycling of waste appliances and electrical products to properly recycle resources and reduce the burden on processing facilities. In the subsequent *Small Home Appliance Recycling Law* and *Law for the Recycling of End-of-Life Vehicles*, there are still regulations concerning battery recycling, which require that the batteries must be taken out before the recycling of appliances and vehicles to reduce resource consumption.

5.1.4 Comparison of Battery Recycling Laws in Different Countries

By comparing the laws regarding batteries recycling in different countries, we found that in most cases, European and American countries regulate the battery recycling in battery directives, not in the regulations for battery recycling. This reflects that the battery recycling in European and American countries has not promoted the improvement of laws and regulations yet. Especially in Europe, only several



Table 5.9 Macropolicies regarding battery recycling in Japan.

La ^{ws} and regulation	Year	Content
Basic Act for Promoting a Circular Society	2000	Focuses on the waste management, and aims to inhibit the consumption of natural resources and reduce environmental load while striving for cyclical use of circulative resources. Clarifies the responsibilities of the state, local governments, business operators, and citizens and aims to promote the policies for the establishment of a Sound Material-Cycle Society comprehensively and systematically
Law for the Promotion of Effective Utilization of Recyclable Resources (renamed as Law for the Promotion of Effective Utilization of Resources in 2001)	1991	Specifies compact rechargeable batteries as reusable and recyclable products. Launches to install the battery collection boxes and collects the batteries in a reversed route
	June 1993	Clarifies the three channels for the recycling of cadmium–nickel batteries and dry batteries from consumers to recyclers: (i) collected and collectively delivered by the local government; (ii) delivered by the distributor or manufacturer of batteries; (iii) delivered by major distributors and service centers of appliances, thus improving the recycling channel Devices powered with nickel–cadmium batteries are specified as Category I products (which are removable) and Category II products (with recycling label)
	April 2001	For the manufacturers and sales companies of batteries, the voluntary recycling of the used small secondary batteries is changed to mandatory recycling. They mainly collect rare metals such as nickel, cobalt, and lead that are used in compact rechargeable batteries
Waste Management and Public Cleansing Law	1971	Clarifies the concept of waste classification, storage, collection, and disposal and enhances public awareness
Law for the Recycling of End-of-Life Vehicles	February 2008	Adds the regulations on the recycling and storage of lithium batteries and nickel–hydrogen batteries to the part regarding the collection and recycling of products by disassembling the electric vehicles and hybrid electric vehicles. Moreover, the Law specifies that scrap dealers shall dismantle the batteries
High-Pressure Gas Safety Act	March 2019	In response to the popularization of hydrogen fuel cell electric vehicles and the advancement of clean energy technology, this Act was formulated to inspect the technical standards of compressed hydrogen refueling stations where safety issues are found in the review for fuel cell electric vehicles

(continued)



Table 5.9 (Continued)

Laws and regulation	Year	Content
Small Home Appliance Recycling Law (aims to promote the recycling of small home appliances)	March 2021	Formulated in April 2001 and amended in March 2021, this Law aims to recycle 140 000 tons of small waste electrical devices annually and specifies that local governments shall participate in the recycling of small waste electrical devices and proper recycling and disposal of waste products regarding lithium accumulators and clarify the proper way of classification and the locations of collection stations to the residents

countries where the battery recycling industry has been developing rapidly formulated more specific regulations on battery recycling, while most countries just adopted the general directives issued by EU. The United States, which take the regulations regarding battery recycling as part of the macropolicy, RCRA, has established a comprehensive legal system of battery recycling, which is amended every year and supplemented by other laws and regulations.

The countries in Asia have more specific and comprehensive laws, regulations, and policies regarding battery recycling than those in Europe and the United States. For example, China's macropolicies regarding battery recycling are not only stated in a battery or environment law but also issued by a battery recycling law. Moreover, all the provinces and cities in China have published local laws that are more stringent and detailed. Take Japan, for instance. Japan has built a sound legal order for the recycling, use, and disposal of spent batteries by formulating specific regulations on the whole life cycle of batteries, including design, production, sales, use, collection, and recycling, and clarification of the rights and obligations of all parties concerned.

Generally speaking, the laws regarding battery recycling in Asian countries are more comprehensive than those in European countries and the United States, and can better meet the production needs of enterprises.

5.2 Management Norms Regarding Battery Recycling

5.2.1 Management Norms Regarding Battery Recycling in Various Countries

5.2.1.1 The United States

Regarding battery management, collection, and recycling, Japan has issued the supporting laws and regulations to facilitate the recycling of resources, consisting of requirements for production, consumption, use, recycling, and disposal of products, in addition to the *Basic Act for Promoting a Circular Society* and the *Law for the Promotion of Effective Utilization of Resources* (Table 5.10).



Table 5.10 Management norms in the United States.

Code	Year	Content
Battery Act proposed by BCI (adopted by the vast majority of states)	1993	This Act stipulates that the lead–acid batteries shall not be disposed of by consumers and shall be sent to retailer and distributor or lead smelter. Battery retailers shall charge consumers a deposit of \$10 (or even more) when they purchase a new battery; the deposit will be reimbursed if they return the same type of a used battery within 30 d; otherwise, the deposit will belong to battery retailers. All batteries returned shall be delivered to wholesalers or lead smelters. The battery wholesalers offering batteries shall require consumers to return the waste batteries with the amount no less than that of new batteries they purchase. Local governments are obligated to monitor the performance of battery retailers and wholesalers and impose fines or other penalties in the case of violations of this Act
NYC Garbage Classification and Recycling Act	1989	Requires that citizens shall return spent batteries and tires to the recycling facilities, send the spent vehicle accumulators to specified recycling units and never mix them with other waste or dispose of them improperly; vehicle battery retailers shall recycle the used batteries without charge, and charge consumers an additional fee of \$5 for future recycling

5.2.1.2 China

In 2021, the *Technical Specification for the Recycling of spent Batteries* (GB/T 39224-2020) has been officially implemented. This Specification summarizes the specifications for spent battery recycling industry; covers technical requirements for battery collection, storage, and transport; and provides an overall and convenient way to go through industrial norms and regulations. This Specification refers to the following documents: the *Safety Signs and Guideline for the Use* (GB 2894), the *Requirements for the Placement of Fire Safety Signs* (GB 15630), the *Standards for Pollution Control on the Storage and Disposal Site for General Industrial Solid Waste* (GB 18599), *Quality Management System – Requirements* (GB/T 19001), *Environmental Management Systems – Requirements with Guidance for Use* (GB/T 24001), the *Store and Transport Regulation of Battery Scraps* (GB/T 26493), *One-Off Battery Scraps* (GB/T 26724), *Rechargeable Battery Scraps* (GB/T 26932), the *Classification and Code of Spent Batteries* (GB/T 36576), *Occupational Health and Safety Management Systems – Requirements with Guidance of Use* (GB/T 45001), the *Code for Fire Protection Design of Buildings* (GB 50016), the *Code for Design of Extinguisher Distribution in Buildings* (GB 50140), the *Technical Specification for Collection, Storage, Transport of Hazardous Waste* (HJ 2025), and the *Specification for Construction of Recyclable Resources Recycling System* (SB/T 10719) (Table 5.11).



Table 5.11 Management norms in China.

Specification	Year	Content
Technical Specification for the Recycling of Waste Batteries (GB/T 39224-2020)	2021	Lays down overall requirements for collection, classification, transport, and storage of spent batteries. Applicable for the whole process of the recycling of spent batteries, except for those that are hazardous waste
Technical Specification for the Collection, Storage, and Transport of Hazardous Waste (HJ 2025-2012)	2012	Specifies the norms and requirements for collection, storage, and transport of hazardous waste
Safety Signs and Guideline for the Use	2008	Clarifies the scope of safety information signs that are applicable for public places, industrial enterprises, and other places where people need to be reminded of safety
Administrative Measures for the Licensing of Hazardous Waste Operations	2004	Provides the proper measures for the operation and management of dangerous waste
Specifications on the Storage and Transport of spent Batteries		
Administrative Specification on the Construction of Resource Recycling Stations	2012	Applicable for the construction and operation of resource recycling stations, not including the recycling stations for recyclable resources that are specified by China's laws, regulations, and rules, such as imported solid waste as raw materials, hazardous waste, and scrapped vehicles
Interim Measures on the Administration of the Recycling and Utilization of Power Accumulators of New Energy Vehicles	2018	Specifies that vehicle manufacturers shall take the main responsibility for power accumulator recycling, and other related companies take the respective responsibilities in different stages of power accumulator recycling. Aims to ensure the effective utilization and environmentally acceptable disposal of power accumulations

5.2.1.3 Japan

Japan's legal system of battery recycling consists of three aspects (Table 5.12):

- (1) Guidance for citizens: Japan has set 11 November as the Battery Day and 12 December as the Accumulator Day, on which the government donates batteries and promotional materials to main communities, social welfare institutions, and the disabled to educate the whole society about the knowledge of batteries and environmental protection. In addition, spent batteries were clearly defined as hazardous substances in garbage classification in 1984 and recycled by citizens in 1985.



Table 5.12 Specifications in Japan.

Code	Year	Content
Asahikawa City in Japan formulated regulations on the recycling of waste batteries	April 1984	Requires that residents shall throw spent batteries into hazardous waste bins and classify the garbage, which is collected and bagged by the Environmental Sanitation Agency weekly
Akita City	June 1895	Launched the classification and recycling of dry batteries
Japanese Ministry of Health, Labour, and Welfare issued guiding opinions	1985	<p>Aims to produce the mercury-free batteries by 1990 and provides guiding suggestions:</p> <ul style="list-style-type: none"> a) Due to strict requirements for waste disposal facilities in Japan, spent batteries can be disposed of together with household garbage, causing no environmental issue and striving to reduce the mercury content in batteries and facilitate the recycling of mercury oxide button batteries to meet the needs of environmental protection. b) To meet the social and environmental requirements, all parties concerned shall jointly take measures to reduce the mercury content in batteries; c) The municipalities can determine whether to recycle spent batteries based on their own needs.
Japanese Dry Cell Industrial Association	1986	In 1986, Japanese Dry Cell Industrial Association took a series of measures to require that the battery manufacturers shall reduce the production of mercury-containing batteries and the mercury content of primary batteries to (i) facilitate the recycling of mercury batteries; (ii) encourage the use of zinc-air batteries instead of mercury batteries used in hearing aids; (iii) strive to reduce the mercury content in alkaline batteries to 1/6 of the current level by 1987; and (iv) label the batteries by 1987
Japanese Ministry of International Trade and Industry	1993	Requires that local governments shall launch the pilot programs on the classification and recycling of dry batteries so as to meet the demands of recycling units.
Japanese Ministry of International Trades and Industrial Production	1995	Specifies that since the end of 1995, the production of mercury oxide batteries shall be completely prohibited. 80% of mercury oxide batteries are used in hearing aids. By 1995, mercury-free zinc-manganese batteries and alkaline batteries were produced in Japan

(continued)



Table 5.12 (Continued)

Code	Year	Content
Japanese Ministry of Economy, Trade, and Industry	2001	Proposed a policy for recycling of compact rechargeable batteries. The policy emphasizes the recycling of rechargeable batteries and points out that the recycling of primary dry batteries shall be carefully explored due to the lack of economical and effective recycling technology in the world. The policy stipulates that the recycling of rechargeable spent batteries shall be organized by the Japanese Dry Cell Industrial Association and relevant groups. It also requires that by 2005, the recycling rate of waste copper-nickel batteries shall be increased from 45% in 1999 to 78%, hydrogen oxide batteries from 20% to 35%, lithium batteries from 20% to 40%, and small lead-acid batteries from 55% to 80%.
The Japanese government proposed the 3R Initiatives	2000	Aims to transform “mass production, mass consumption, and mass disposal” into 3R (reduce, reuse, and recycle)
The Japanese Ministry of Economy, Trade, and Industry issued the Laws on Home Appliance Recycling and Other Recycling Activities	April 2009	As stipulated, the appliance manufacturers, sellers, and consumers in Japan shall take different obligations. Appliance manufacturers shall recycle waste appliances by building or renting recycling factories, appliance sellers shall collect and transport waste appliances to the manufacturers, and consumers shall bear the above costs
Urayasu City launched the 4R program	2019	4R means reduce, reuse, recycle, and refuse. Add “refuse” to the 3R program
The Japanese Ministry of Economy, Trade, and Industry published the Research and Development Plan for the Next Generation of Accumulators Used in Electric Vehicles	August 2021	The Plan points out that in the next 10yr, the production technologies of high-performance accumulators such as all-solid-state batteries with low greenhouse gas emissions will be developed, with the carbon dioxide emissions decreased to 16% of global total carbon dioxide emissions. It is estimated that by 2040, the application of such technologies will reduce 260 million tons of carbon dioxide emissions globally each year and will raise up the market size of new vehicles to 6.2 trillion yen

- (2) Requirements for battery manufacturers: In 1985, battery manufacturers in Japan were required to gradually produce mercury-free batteries by 1990. As for the goal, the Japanese government issued the guiding opinions to stipulate the measures, which shall be taken by all parties, to (i) reduce the mercury content in dry batteries and facilitate the recycling of mercury batteries; (ii) regionally



recycle and dispose of spent alkaline batteries; (iii) regionally establish the recycling and disposing system of spent alkaline batteries; and (iv) build the organizations engaged in promoting the battery recycling with the active assistance of battery manufacturers.

- (3) Regulations on battery recycling enterprises: In 1985, the Japanese government proposed to build organizations engaged in promoting the battery recycling. In 2001, the Japanese Ministry of Economy, Trade, and Industry stipulated that the recycling of rechargeable spent batteries shall be organized by the Japanese Dry Cell Industrial Association and relevant groups. In Japan, generally, spent batteries are recycled by the factories with metallurgical capabilities, not by battery manufacturers. The spent batteries are always collected by associations, battery manufacturers (from recycling bins in shopping malls and public places), and local municipal departments. Most of the batteries collected by local municipal departments separately (without being mixed with other garbage) are delivered to specific battery recycling enterprises such as Nomura Kohsan Co., Ltd. and Toho Zinc Co., Ltd. for further recycling and reuse, while the spent batteries, not collected separately, are properly disposed of as non-burnable garbage.

5.2.2 Comparison of Management Norms in Different Countries

According to the extended producer responsibility policy, the laws for battery collection and recycling are formulated in the developed countries to require that all battery manufacturers and importers shall collect and recycle spent batteries. Moreover, batteries and secondary batteries containing hazardous substances are mainly recycled, while primary batteries are not mandatorily recycled. In general, more battery collecting sites can significantly raise the public awareness, and more educational activities facilitate battery recycling.

In the United States, most management specifications are issued by BCI, while some are separately issued by the states where the battery recycling industry has rapidly developed. In China, management specifications for each battery recycling process are uniformly formulated and published by the state so that each process can be supervised by law. In Japan, the legislation of battery recycling started early and a comprehensive legal system consisting of basic laws, general laws, special laws, and supplementary policies and requirements at the level of municipalities has been established.

In conclusion, not every country has separately stipulated the specifications for every single process of battery recycling. Most of the relevant contents are included in the macropolicies and Acts in the European countries and the United States. The details of the specifications can also reflect the development of the battery recycling industry and the policy-making farsightedness in different countries.



5.3 Technical Norms Regarding Battery Recycling

5.3.1 Technical Norms Regarding Battery Recycling in Various Countries

The spent battery enterprises in Japan shall comply with the ISO 14001 standard. Currently, the enterprises that have been certified to ISO 14001 include J&T Environmental Co., Ltd. certified in 1999 and Nomura Kosan Co., Ltd. certified in 2001. Established in December 1973, Nomura Kosan Co., Ltd. is a waste disposal company engaged in the disposal of spent disposable batteries and waste fluorescent lamps. This company was designed as a wide-area center for the recycling and disposal of waste batteries in 1986. During the early years, the Japanese government provided 80 yen/kg of spent batteries for recycling plants. In 2019, technical standards for battery recycling proposed by 4R Energy Corp. have been certified to UL 1974 (Tables 5.13–5.17).

Japan adopts the scheme of “ultimate recycler and dismantler” to establish and implement its battery recycling system. To recycle the portable batteries, the Japan Portable Rechargeable Battery Recycling Center (JBRC), which is a nonprofit producer responsibility organization, plays a role as ultimate recycler and dismantler.

Table 5.13 European norms (EN).

Code	Year	Content
EN 60622	1995	Covers technical standards for sealed NiCd square rechargeable monobloc batteries
EN 61429	1997	Labels the secondary cells and batteries according to international recycling symbol ISO 7000-1135 and applies to lead-acid batteries (Pb) and nickel-cadmium batteries (Ni-Cd)
EN 60623	2001	Secondary cells and batteries containing alkaline or other non-acid electrolytes – Vented nickel-cadmium prismatic rechargeable single cells (IEC 60623: 2001)
GENELEC EN 61960	2003	IEC 61960, Ed. 1: Secondary cells and batteries containing alkaline or other non-acid electrolytes – Secondary lithium cells and batteries for portable applications
EN 60622	2003	Secondary cells and batteries containing alkaline or other non-acid electrolytes – Sealed nickel-cadmium prismatic rechargeable single cells IEC 60622:2002
CENELEC EN 61960	2004	Secondary cells and batteries containing alkaline or other non-acid electrolytes. Secondary lithium cells and batteries for portable applications IEC 61960: 2003; Partially Supersedes EN 61960-1: 2001& EN 61960-2: 2001
EN 61960	2011	Describes the performance tests, designations, markings, dimensions, and other requirements for secondary lithium single cells and batteries for portable applications

Table 5.14 Germany industry norm (DIN).

Code	Year	Content
DIN EN 61429	1997	Label the secondary cells and batteries according to international recycling symbol ISO 7000-1135 (IEC 61429: 1995); German version EN 61429:1996 gives the labeling requirement for rechargeable batteries
DIN EN 60622	1997	Sealed nickel–cadmium prismatic rechargeable single cells (IEC 60622:1988 + a2:1992 + corrigendum 1992); German version EN 60622:1995
DIN EN 61960	2001	Secondary lithium cells and batteries for portable applications – Part 1: Secondary lithium cells
DIN IEC 61960	2002	Secondary cells and batteries containing alkaline or other non-acid electrolytes – Secondary lithium cells and batteries for portable applications (IEC 21A/340/CD:2001). Part 2: Secondary lithium batteries
DIN EN 60623	2002	Secondary cells and batteries containing alkaline or other non-acid electrolytes – Vented nickel–cadmium prismatic rechargeable single cells (IEC 60623:2001); German version EN 60623:2001
DIN EN 61960	2004	This international standard specifies performance tests, designations, markings, dimensions, and other requirements for secondary lithium single cells and batteries for portable applications. The objective of this standard is to provide the purchasers and users of secondary lithium cells and batteries with a series of performance evaluation criteria for secondary lithium cells and batteries made by different manufacturers
DIN IEC 61960	2008	Secondary cells and batteries containing alkaline or other non-acid electrolytes – Secondary lithium cells and batteries for portable applications (IEC 21A/445/CD:2008)
DIN EN 61960	2012	Secondary cells and batteries containing alkaline or other non-acid electrolytes – Secondary lithium cells and batteries for portable applications (IEC 61960: 2011)

Spent portable batteries are collected in collection boxes from consumers first and then delivered to the JBRC. The JBRC classifies and transports these batteries to recyclers for treatment as recyclable resources. The qualified recyclers include Recycle21 Co., Ltd. under Fuyo Metals, Japan Recycling Center Co., Ltd., Nippon Magnetic Dressing Co., Ltd., and Japan Kyoei Steel Co., Ltd.

Similarly, to recycle the lithium batteries, the manufacturers of electric vehicles such as Toyota Motor Corporation and the manufacturers of LIBs such as Panasonic Corporation and Samsung Group play the role as ultimate recycler and dismantler. These manufacturers established their dismantling centers to collectively manage, transport, dismantle, and classify the recycled lithium batteries. The batteries that are worthless for reuse will be transported to recyclers for smelting and processing as scrap metal.

Table 5.15 Netherlands norm (NEN).

Code	Year	Content
NEN EN IEC 61429	1997	Label the secondary cells and batteries according to International Recycling Symbol ISO 7000-1135 and give indications to Directives 93/86/EEC and 91/157/EEC
NEN EN IEC 61960-1	2001	Specifies performance and safety tests, designation, markings, dimensions, and other requirements for secondary lithium single cells. Secondary lithium cells and batteries for portable applications – Part 1: Secondary lithium cells
NEN EN IEC 61960-2	2001	Secondary lithium cells and batteries for portable applications – Part 2: Secondary lithium batteries
NEN EN IEC 60623	2001	Lays down general requirements, characteristics, marking, dimensions, and electrical and mechanical test methods for vented prismatic rechargeable nickel–cadmium cells
NEN EN IEC 61960	2004	Defines performance tests, designations, markings, dimensions, and other requirements for secondary lithium single cells and batteries for portable applications. Provides the purchasers and users of secondary lithium cells and batteries with a series of performance evaluation criteria of secondary lithium cells and batteries made by different manufacturers
NEN EN IEC 61960	2011	Secondary batteries and battery packs containing alkaline or other non-acidic electrolytes; secondary lithium batteries and battery packs for portable equipment

Table 5.16 Norway standards (NS).

Code	Year	Content
NS 9431	2000	Classification of waste
NS 9430	2002	General contract conditions for collection and transport of Chinese waste
NS 9431	2011	Classification of water
NS 9430	2013	General contract conditions for regular collection and transport of waste

5.3.2 Comparison of Technical Norms in Different Countries

Technical standards are formulated and promulgated by the governments in Europe and China. However, technical standards are formulated by the designated enterprises and approved by the government and implemented by relevant enterprises during production and processing in Japan. The battery recycling enterprises in China and the United States follow the third-party's standards and compete effectively. Japanese enterprises play a leading role in building a battery recycling network through their existing marketing network and accordingly make industry standards.

The technical standards, which are formulated in different ways in different countries, have different details, but all meet the needs of the countries.

Table 5.17 China standards.

Standard	Year	Content
Standards for Pollution Control of General Industrial Solid Waste Storage and Disposal Sites	2020	This Standard specifies the environmental requirements for the siting, construction, operation, closure, and land reclamation of general industrial solid waste storage and disposal sites as well as pollution control and monitoring
Safety Signs and Guideline for the Use	2012	Specifies the norms and requirements for the collection, storage, and transport of hazardous waste
Specifications of the recycling and dismantling of vehicle power batteries	2017	Specifies general requirements, operating procedures, and storage and management requirements for disassembling of vehicle rechargeable battery packs and modules
Residual Capacity Testing in the Recycling of Vehicle Power Batteries	2017	Specifies that the recycling procedures of spent vehicle batteries shall be tested according to the performance test and safety requirements. Provides the requirements for residual capacity testing, including visual inspection, polarity detection, voltage identification, charging and discharging current detection, and residual capacity testing
Coding Regulation for Vehicle Power Batteries	2017	Specifies the basic coding principle, coding object, code structure, and data carrier of vehicle power batteries. With their traceability and uniqueness, the codes for batteries can help accurately determine the responsible body of power battery recycling during production, maintenance, key parameter monitoring, and recycling of power batteries
Product Specifications and Dimensions of Power Batteries of Electric Vehicles	2018	Stipulates the specifications and dimensions of power battery cells, modules, and standard boxes that are installed to electric vehicles, including LIBs and metal hydride–nickel batteries
Requirements for Material Recycling on the Recycling of Batteries of Electric Vehicles	2018	Specifies the requirements for material recycling enterprises' recycling and treatment process, personnel operation, siting, and processing technology of battery recycling and defines the recycling rate and its calculation method

5.4 Support Policies Regarding Battery Recycling

5.4.1 Support Policies Regarding Battery Recycling in Different Countries

In addition to the subsidy policies stipulated by the US government, state governments at all levels have formulated the subsidy policies for purchasing new-energy vehicles. For example, California implemented the Zero Emission Vehicle Program in 1990 and formulated the Zero Emission Vehicle Regulation (ZEV Regulation), which innovatively designs the merit points and merit point trading system for new-energy

vehicles and stipulates that the car brand manufacturers with a certain volume of sales in California must have certain new-energy points, which can be accumulated and traded, and those that cannot meet the conditions will be fined (Tables 5.18 and 5.19).

Another force of promoting the popularity of clean vehicles is the subsidy program for clean vehicles. This program expands its scope and applies to three types of fuel cell vehicles (a subsidy of \$5000 for each), 21 types of pure electric vehicles (\$2500), 17 types of plug-in hybrid vehicles (\$1500), and 13 types of pure electric motorcycles (\$900).

Table 5.18 Support policies in European countries.

Country	Subsidy and preferential policy	Adjusted subsidy and preferential policy
Germany	<p>Since 19 May 2016, the subsidies of 4000 and 3000 euros have been given to customers purchasing pure electric vehicles and customers purchasing plug-in hybrid vehicles in Germany, respectively, and borne by the government and car manufacturers, with the total sum of 1.2 billion euros. The subsidy policy came into effect in July 2015 until all the subsidies will be paid out</p> <ol style="list-style-type: none"> (1) An electric vehicle purchased between 2016 and 2020 can share a license plate with another car at home, and the car owner needs to provide only one insurance policy for the two cars (2) Pure electric vehicles, plug-in hybrid electric vehicles, and fuel cell vehicles will enjoy free parking and bus lanes and other privileges 	<p>The subsidy fee has been paid off; no introduction of new policy</p>
Norway	<p>Customers purchasing electric vehicles in Norway can be exempt from sales tax and 25% value-added tax as well as reducing annual license fees</p> <ol style="list-style-type: none"> (1) No registration, import value-added tax, and road tax are levied on pure electric vehicles (2) Electric vehicles can occupy bus lanes and enjoy free charging, free parking, and city tolls 	<ol style="list-style-type: none"> (1) VAT exemption policy canceled (2) Tariff-free policy for imported models canceled

(continued)

Table 5.18 (Continued)

Country	Subsidy and preferential policy	Adjusted subsidy and preferential policy
Netherlands	<p>(1) Registration tax: All electric vehicles and plug-in hybrids are exempt from this tax. Plug-in hybrid buyers will have to pay extra for the carbon dioxide emitted</p> <p>(2) Purchase tax: All-electric vehicles are exempt from this tax, while plug-in hybrids receive a 50% discount. Vehicles with high CO₂ emissions and service period of more than 12 years will be subject to an additional 15% tax from 2019</p> <p>(3) Official vehicle use tax: Pay only 4% for all-electric vehicles and 22% for plug-in hybrids and high CO₂ emission vehicles</p>	<p>The Netherlands will provide subsidies for private purchasers of electric vehicles from July 2020. This policy doesn't apply to the hybrid models and pure electric vehicles with the price above 45 000 euros but applies to the second-hand electric vehicles that meet the conditions. Customers purchasing pure electric vehicles with the price between 1200 and 45 000 euros and a minimum driving mileage of 120 km will receive the subsidy of 4000 euros, while second-hand electric vehicles that meet the conditions can receive the subsidy of 2000 euros</p>

Table 5.19 Support policies in the United States.

Code	Year	Content
Energy Improvement and Extension Act	2008	Section 30D in this Act specifies the special tax credit policies for new qualified plug-in electric drive motor vehicles
American Recovery and Reinvestment Act	2009	Since 31 December 2009, taxpayers will enjoy a corresponding tax refund for purchasing plug-in hybrid electric vehicles and pure electric vehicles that meet the conditions. Specifically, the tax refund is calculated based on battery capacity. The tax refund is \$2500 for a vehicle with the battery capacity of 5 kWh or below, and plus \$417 for each kilowatt hour of additional battery capacity. The upper limit of total tax refund is \$7500

Some other states such as Colorado, more simply and directly, provide a tax subsidy of up to \$5000 for purchasing electric vehicles and \$2500 for renting electric vehicles (Table 5.20).

China's subsidy policies in 2020 have the following differences from those in 2018 and 2019:

- (1) The overall subsidies have been cut, and the subsidy for vehicles with cruising range of less than 300 km is canceled.

Table 5.20 Adjusted subsidy policies for electric drive motor vehicles in China.

Cruising range (R , km)	Base amount in 2020 (yuan/ vehicle)	Base amount in 2019 (yuan/ vehicle)	Energy density (Wh/kg)	Subsidy coefficient in 2020	Subsidy coefficient in 2019
$300 \leq R < 400$	16 200	18 000	125–140	0.68	0.8
$R \geq 400$	22 500	25 000	140–160	0.765	0.9
—	—	—	≥ 160	0.85	1
Technical requirements in 2020	(1) The cruising range of pure electric vehicles shall not be fewer than 300 km (2) The subsidy coefficient is set according to the energy consumption level of pure electric vehicles. With different vehicle curb weights (m), the energy consumption per 100 km (Y) shall meet the following threshold conditions: when $m \leq 1000$, $Y = 0.0112 \times m + 0.4$; when $1000 < m \leq 1600$, $Y = 0.0078 \times m + 3.8$; when $m > 1600$, $Y = 0.0044 \times m + 9.24$. For the vehicles with $0\% \leq Y < 10\%$ above the threshold value, the coefficient is 0.8; for vehicles with $10\% \leq Y < 25\%$ above the threshold value, the coefficient is 1.1 (3) For the maximum speed of pure electric vehicles in 30 min and energy density of power batteries used in pure electric vehicles and energy consumption of plug-in hybrid electric drive motor vehicles and the corresponding subsidies, see the <i>Notice of Improving the Policies on Government Subsidies for Promotion and Application of New Energy Vehicles</i>				

- (2) Establishes a price threshold, i.e. the before-subsidy price of the vehicles is 300 000 yuan or below.
- (3) Adds a measure to encourage the development of battery-swapping technology and specifies that vehicles adopting battery-swapping technology enjoy the subsidies regardless of their prices.
- (4) As from 2020, vehicle manufacturers shall make a single application for subsidy settlement of 10 000 vehicles.
- (5) The nonprivate vehicles (operating vehicles) cannot enjoy a full subsidy (70%).
- (6) Sets 23 April 2020 to 22 July 2020 as the transitional period to adopt new policies.

In conclusion, China's support policies mainly aim to provide subsidies for the new-energy vehicle industry, but these subsidies have been cut at a steady intensity and pace, i.e. the subsidies have gradually declined from the previous year.

5.4.2 Comparison of Support Policies in Different Countries

All the support policies for new-energy vehicles in different countries give financial subsidies to vehicle consumers and companies. The differences include the amount of subsidy, the policy duration, and the calculation method. However, all countries

provide subsidies for consumers to stimulate the consumption of electric vehicles and facilitate the industrial development and subsidies for enterprises to encourage their investment in the research and development of new energy.

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6

New Application Scenarios for Power Lithium-Ion Batteries

6.1 The Existing Application Scenarios of Power Batteries

6.1.1 Two-Wheeled Electric Vehicles

6.1.1.1 Global Market Development Status

The number of two-wheeled electric vehicles (EVs) in the world has exceeded 300 million, and the Chinese market leads the world. China is currently the largest country of two-wheeled electric vehicle production, sales, and export in the world. After more than 20 years of development, two-wheeled electric vehicles have become an important means of transport for Chinese residents to solve the problem of the last mile. According to the China white paper on the quality and safety of electric bicycles released by the National Quality Supervision and Inspection Center of Electric Bicycles in 2017 and the statistical data from the Ministry of Industry and Information Technology [1, 2], as shown in Figures 6.1 and 6.2, the two-wheeled electric vehicle parc in China was more than 300 million vehicles, 90% of the world's total two-wheeled electric vehicle parc, in 2020. From January to May 2021, the output of two-wheeled electric vehicles in China reached 12.611 million, with an increase of 42% over the previous year. The annual output of two-wheeled electric vehicles in China has approached 30 million since 2018, with the penetration rate of nearly 70%. However, the domestic market tends to become saturated, and the growth slows down significantly because on the one hand, the export growth is driven by the increase in overseas market demand, and on the other hand, the implementation of the new national standard promotes the transformation from quantity to quality of two-wheeled electric vehicles, and the replacement of stock has become the biggest driving force to maintain the development of the industry.

The penetration rate of electric vehicles in overseas markets continues to rise. According to Electric Bikes Worldwide Report (EBWR) and the annual data from Statista, the Asia-Pacific region and Western Europe accounts for the vast majority of the global e-bike market. The Asia-Pacific region and Western Europe account

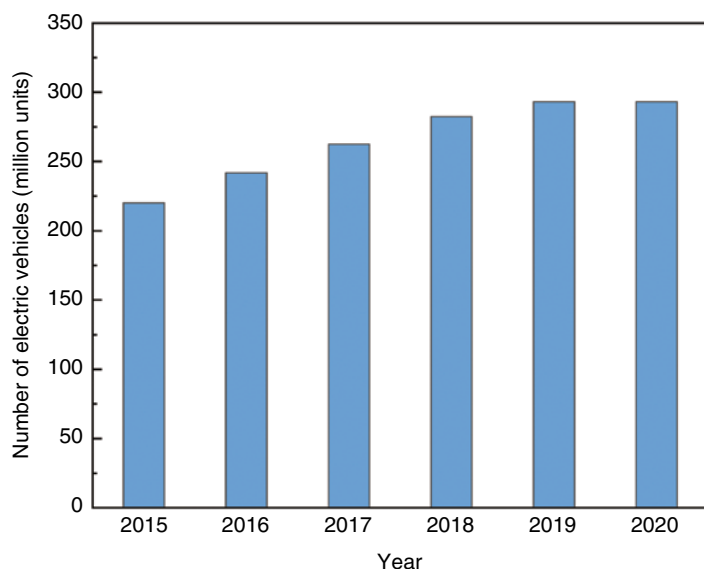


Figure 6.1 Electric vehicle parc in China [1].

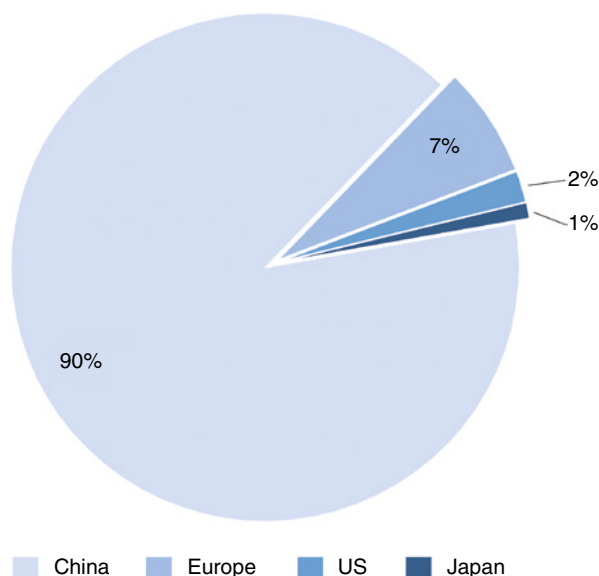


Figure 6.2 Percentage of two-wheeled electric vehicles. *Source:* Ministry of Industry and Information Technology of the People's Republic of China [2].

for 94.39% and 4.60% of the e-bike market, respectively [3]. Different from two-wheeled electric vehicles widely applied in China, the two-wheeled fuel motorcycles vehicles still dominate in other countries. The development of two-wheeled electric vehicles is very limited; hence, there is still a large space for growth. According to statistics from relevant institutions [4], the sales volume of two-wheeled electric vehicles in the United States and Europe reached 430 000 and

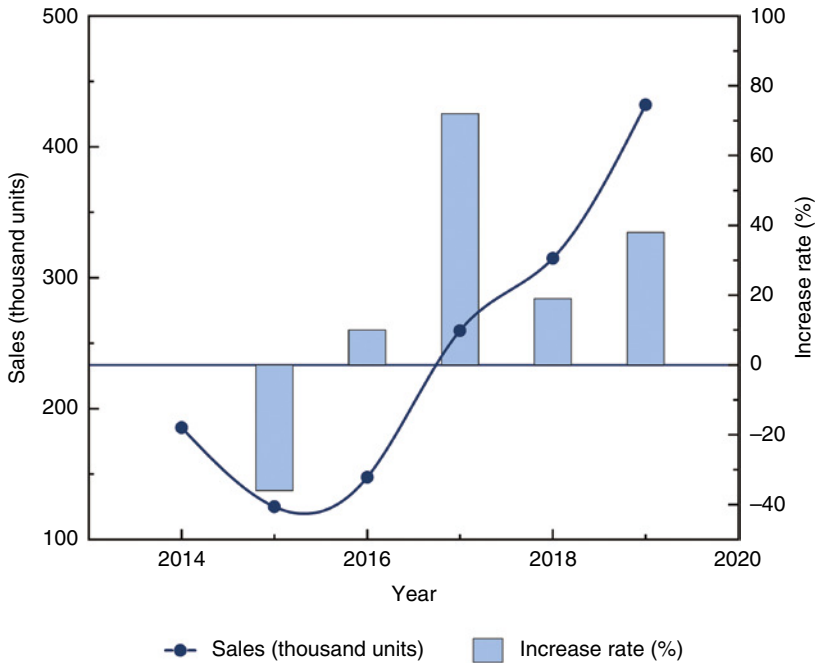


Figure 6.3 Sales of two-wheeled electric vehicles in the United States over the years.
Source: GF Securities Research and Development Center [4].

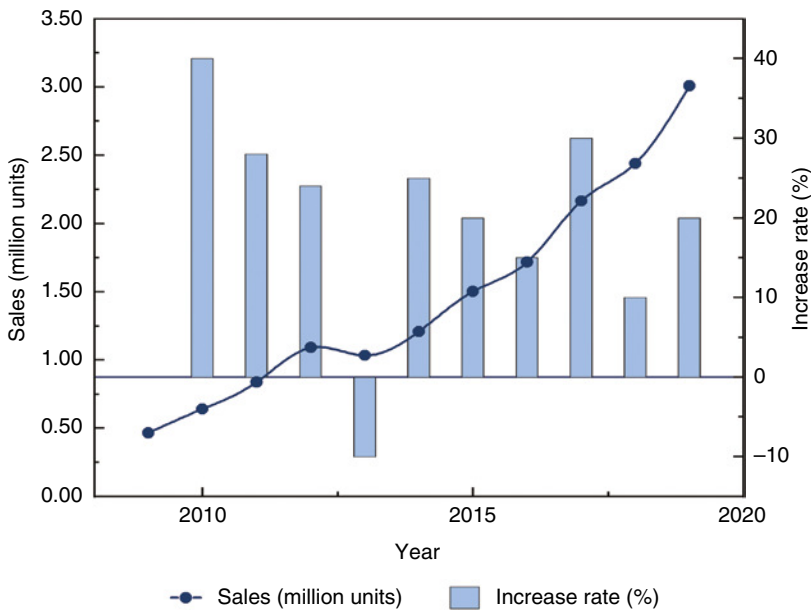


Figure 6.4 Sales of two-wheeled electric vehicles in Europe over the years.
Source: GF Securities Research and Development Center [4].

3 million in 2019, respectively, showing a significant annual growth (Figures 6.3 and 6.4). According to Technavio, the sales volume of two-wheeled electric vehicles in Europe is expected to grow by 18% on average until 2022; thus, the annual volume of sales in the European market will reach more than 4.5 million annually. As people are realizing that two-wheeled electric vehicles are very economical, environmentally friendly, and convenient means of transport, the demand in the United States and Europe will continue to grow, and the market penetration rate of two-wheeled electric vehicles will continue to increase, too.

6.1.1.2 Impact of National Policies

The loose constraint conditions of old national standards promote the rapid development of electric bicycles. Although a large number of electric bicycles are convenient for people's life, they bring about a series of problems such as too fast speed, unsafe structure, and quality noncompliance. To solve these problems, China implemented the new mandatory national standard GB 17761-2018, *Safety technical specification for electric bicycle* (Table 6.1), from 15 April 2019. The new national standard states that in accordance with the weight of the vehicle, power, etc., electric bicycles are classified into electric bikes, electric mopeds, and electric motorcycles.

Table 6.1 Technical specifications for safety of electric bicycles.

Category	Electric bicycle	Electric moped	Electric motorbike
Gross weight, kg	≤55	≥55	≥55
Maximum speed, km/h	≤25	≤50	>50
Battery voltage, V	≤48	—	—
Battery power, W	≤400	≤4000	>4000
Carrying capacity	Children under 12 allowed in some cities	0	One adult
Product nature	Non-motorized vehicle	Motor vehicle	Motor vehicle
Pedal	Mandatory	No pedal	No pedal
Product management	3C Certification	3C Certification and MIIT Catalog Announcement	3C Certification and MIIT Catalog Announcement
Standard	Mandatory standard, <i>Safety Technical Code for Electric Bicycles</i>	Recommended standard, <i>General Technical Conditions for Electric Bicycles</i> Recommended standard, <i>General Technical Conditions for Electric Motorcycles and Electric Mopeds</i>	Recommended standard, <i>General Technical Conditions for Electric Motorcycles and Electric Mopeds</i>

The electric bicycles not meeting the standard will be phased out. The new national standard specifies that the most widely used electric bike has the maximum speed limit of 25 km/h, gross weight (including the battery) of 55 kg, and motor power of 400 W. According to the data from the General Administration of Customs of China [5], about 19 million electric bicycles were exported in 2019 and 2020. It can be assumed that there was no vehicle meeting new national standard before 2019. Based on the sales volume of about 56 million in these two years, we can preliminarily estimate that about 88% of electric bicycles do not meet the new national standard. As the transitional period of the implementation of the new national standard comes to an end in all the parts of China, a huge number of electric bicycles not meeting the new national standard will exit the market and accordingly will be replaced. Thus, two-wheel electric vehicles are entering a new era.

Moreover, overseas countries are paying more and more attention to the e-bike market. This year, a number of European countries have introduced subsidies to encourage cycling. Among them, the Dutch government gives more than 30% of the e-bike price to the buyers as the subsidy for electric bicycle; the French government has set up a 20 million euros subsidy scheme and accordingly offers 400 euros to each employee who goes to work by e-bike. The city of Berlin in Germany is lacking electric bikes due to re-planning of new roads and widening of temporary bike lanes.

According to Navigant Consulting [6], the global electric bus and electric two-wheeler (electric motorcycle, electric bicycle, and electric scooter) industry will reach \$62.2 billion by 2025, with a compound annual growth rate of more than 10%.

6.1.1.3 Development Outlook of EV Battery for Two-Wheeled Electric Vehicle

The two-wheeled electric vehicles are obviously developing into the two-wheeled electric vehicles with lithium battery. With the decreasing cost of lithium battery, its comprehensive advantages have been highlighted. It is expected that lead-acid battery will be phased out. In China, about 260 million of the existing 300 million two-wheeled electric vehicles have conventional lead-acid batteries. As the transition period comes into an end in various regions, it is expected that the market demand will be at least 100 million lithium battery vehicles in the next three years. Based on the battery capacity of 0.6 kWh in each vehicle, the market demand for lithium batteries is 60 GWh in the next three years.

6.1.2 Electric Vehicles

6.1.2.1 Global Market Development Status

As energy sustainability and environmental friendliness are becoming the main development theme of transport vehicles, all the countries around the world have released the timetable to ban the sale of fuel vehicles, and the full use of electric vehicles has become a general trend. In addition, the countries have invested a lot of money in the research and development of lithium-ion power battery technology. Technological progress leads to a significant decrease of the cost, and the price of electric cars is more and more close to fuel cars. Although carbon dioxide is emitted

from the production and use of electric vehicles, electric vehicles will show more prominent advantage of low carbon with gradual reduction of carbon emission in the whole industrial chain.

Global Electric Vehicle Outlook 2021 released by the International Energy Agency (IEA) shows that after more than a decade of rapid growth, the world has the electric vehicle parc of 10 million by the end of 2020, 43% higher than 2019 [7]. Overall, the 2020 COVID-19 pandemic has significantly impacted the global markets of all types of vehicles. The number of newly registered electric vehicles in the first half of 2020 is about 1/3 smaller than that in the previous year, but this impact is partially offset by a strong trend in the second half of 2020, resulting in total decline of 16% in 2020. Notably, the global sales volume of electric vehicles increased by 70% in 2020 and reached a record 4.6% (of total sales volume of electric vehicles) in this context. A total of 3 million electric vehicles, including 1.4 million in Europe, 1.2 million in China, and 295 000 in the United States, were registered in 2020 (Figure 6.5).

China has become a leader in the development of electric vehicles. The data from the Traffic Administration of the Chinese Ministry of Public Security show that as of March 2021, China has 5.51 million new energy vehicles (including electric vehicles), including 4.49 million pure electric vehicles, with 81.5% of new energy vehicles [8]. The data of China Electric Charging Infrastructure Promotion Alliance show that by June 2021, China totally has 1.947 million charging stations, i.e. less than three electric vehicles share one charging station on average [9]. With the increasing number of electric vehicles and the continuous improvement of supporting infrastructure, China has established comprehensive advantages in terms of

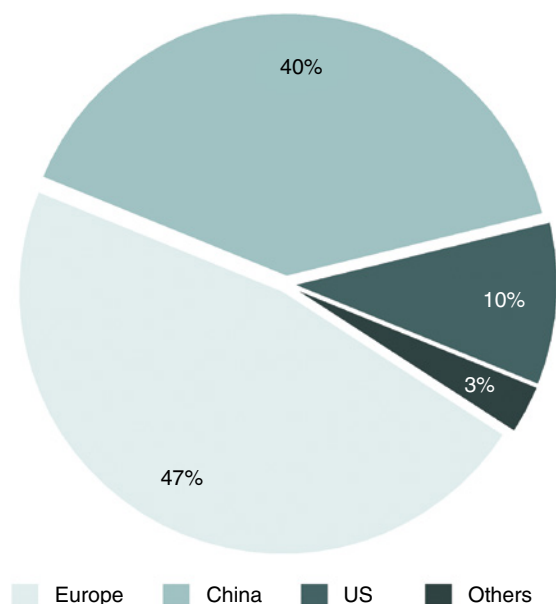


Figure 6.5 Number of newly registered EVs in 2020.

technology, cost, resources, and market. Therefore, China's development status of electric vehicles is the most representative and notable.

In terms of family cars, BYD Qin PLUS-DMI is a hybrid electric vehicle with total endurance mileage of more than 1200 km. Its starting price is 105 800 yuan, which is the same as that of ordinary fuel cars at the same level. BYD Han EV is a pure electric vehicle equipped with a blade battery. It is very competitive against the same class of fuel cars due to its endurance mileage of 605 km and starting price of 229 800 yuan [10].

In terms of commercial vehicles, electric buses are the key focus. According to the China Automobile Association [10, 11], leading companies such as Yutong, BYD, Nanjing Jinlong, and Hunan CRRC sold about 73 000 pure electric buses in 2020. At present, there are about 800 000 electric buses in China. The existing operation results show that electric buses have significant advantages over traditional fuel buses in energy saving, emission reduction, maintenance, and operation costs.

According to the statistics of The Ministry of Industry and Information Technology of China [2], electric vehicles rapidly grow in the context of the overall decline of automobile production and sales. From January to June 2021, the production and sales of electric vehicles reached 1.215 and 1.206 million respectively, twice more than the same period in the last year. According to the electric vehicle model, the production and sales of pure electric vehicles reached 1.022 and 1.005 million respectively, which was 2.3 and 2.2 times more than the same period in the last year, respectively. The production and sales of plug-in hybrid electric vehicles reached 192 000 and 200 000, respectively, which was 1 and 1.3 times more than the same period in the last year, respectively. The production and sales of fuel cell vehicles reached 632 and 479, respectively, which was 43.6% and 5.7% more than the same period in the last year, respectively.

It can be predicted that the penetration rate of electric vehicles in the Chinese market will continue to rise due to decreasing cost of electric vehicles, continuous improvement of charging infrastructure, and inherent advantage of low cost.

The European market is catching up with the Chinese market. The sales volume in Europe surpassed that in China for the first time in 2020; thus, Europe has become the world's largest market for electric vehicles. The development of electric vehicles in Norway as the most environmentally conscious country in Europe is attractive. According to the Norwegian Road Federation (OFV) [12] (Figure 6.6), electric vehicles accounted for 54.3% of all vehicles sold in 2020; thus, Norway has become the first country where more electric vehicles were sold than gasoline vehicles in 2020 in the world. The Norwegian government has introduced a series of regulations to encourage consumers and local governments to promote and use electric vehicles, including large tax reduction for the purchase or rental of electric vehicles and subsidies for the construction and maintenance of private and public charging facilities. In addition, electric vehicle owners can get a lot of convenience and rights in daily use. For example, they are not restricted by parking lots and expressway lanes, and they can enjoy fee discount when they use toll roads, car ferries, and parking lots. Norway has perfect associated facilities of electric vehicles, including charging stations at both sides of roads, hotels across the country, and

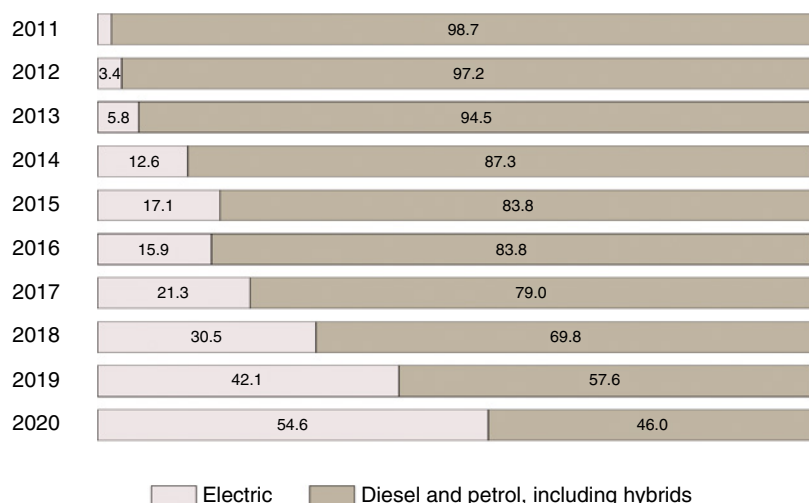


Figure 6.6 Market share of electric vehicles and fuel vehicles in Norway in recent 10 years. Source: Adapted from Sina Finance [12].

even B&B Hotels in remote areas. Moreover, Norway has set 2025 as the year for a complete ban of petrol cars and thus will play a demonstrative role in carbon emission reduction in the world.

Followed by China and Europe, the United States is an important growth pole. According to data from the IEA [8], the sales volume of EVs in the United States in 2020 was 296 000, lower than 328 000 in 2019, showing a downward trend. Although the United States lags behind Central Europe, its formidable automotive industry and policy determination cannot be underestimated. On 31 March, 2021, the Biden Administration released the *U.S. Infrastructure Plan* and proposed the investment of \$174 billion in the development of the US electric vehicle market, improvement of the domestic industrial chain, sales discounts and tax incentives, construction of 500 000 charging stations, electric school buses, federal fleets by 2030, etc. In a speech at a Ford plant in Michigan on 18 May, Joe Biden, US President, said that electric cars were “the future of the auto industry” and China was leading in the race; therefore, he called on the US to move quickly and regain its lead in the industry. According to the White House, Biden signed an executive order to set a new national goal for the auto industry, i.e. the sales volume of electric vehicles will be increased to 50% of all new vehicles sold in the United States by 2030. With strong technological strength and government support, the US market is expected to become an important growth pole for the global electric vehicle market.

6.1.2.2 Segmentation – Electric Passenger Vehicles

Electric passenger vehicles will be the most extensive application scenario of lithium-ion power battery in the automotive field in the future. The ordinary passenger vehicles account for the highest percentage in the automotive sector. According to the data of the IEA [8], a total of 77.97 million vehicles were sold worldwide in 2020,

including 55.6 million electric passenger vehicles, accounting for 71.3% of all vehicles. In spite of the largest market share in the automotive sector, the passenger vehicles are not the largest carbon emitter. The CO₂ test results of the European Environment Agency show that the carbon emission from passenger vehicles and light goods vehicles account for 13% of total emissions from all vehicles [13]. In 2019, China's energy sector consumed 4.7 billion tons of standard coal and 130 million tons of gasoline. Based on 200 million ordinary passenger vehicles with fuel consumption of 350 l and an average annual mileage of 50 000 km each, the estimated annual gasoline consumption of passenger vehicles is only 50 million tons. If 1 kg of standard coal produces 2.5 kg of CO₂ and 1 kg of gasoline produces 3.15 kg of CO₂, the carbon emissions from ordinary passenger vehicles account for less than 3% (Figure 6.7).

National policies encourage the development of electric passenger vehicles. China's *New Energy Vehicle Industry Development Plan (2021–2035)* clearly states that pure electric vehicles will become the mainstream of newly sold vehicles. In this context, the Chinese government has introduced specific incentives. In 2017, the Ministry of Industry and Information Technology issued the *Parallel Management Method of Average Fuel Consumption of Passenger Vehicle Enterprises and Points of New Energy Vehicles* (referred to as the double point policy); thus, the passenger vehicle enterprises are facing dual pressure. On the one hand, they must strive to reduce the fuel consumption of traditional fuel vehicles and then reduce the negative points. On the other hand, they must strengthen the development of electric vehicles so as to accumulate positive points. Furthermore, the policy allows

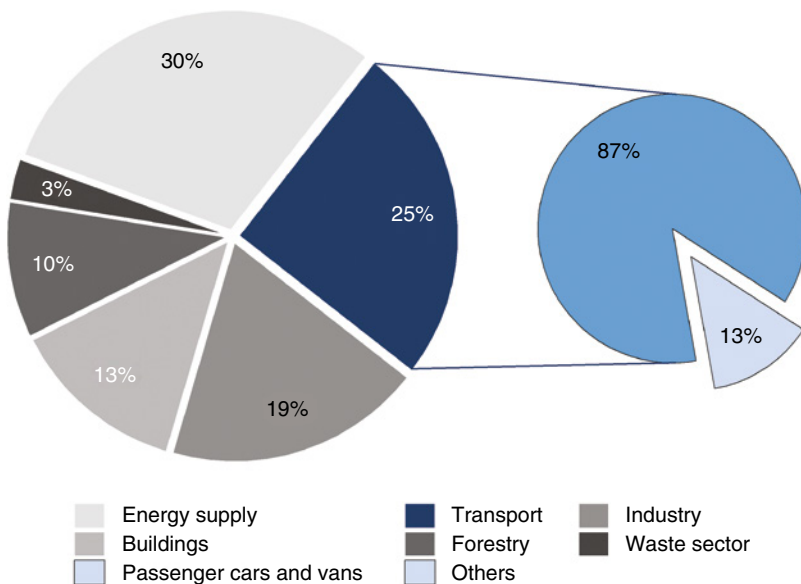


Figure 6.7 CO₂ emissions in different industries (including the proportion of passenger vehicle emissions in the transport sector). Source: European Environment Agency [13]/European Environment Agency/CC BY-4.0.

for the use of the positive points of electric vehicles to offset the negative points of traditional fuel vehicles, encouraging the passenger vehicle companies to produce more electric vehicles. The abolishment of the subsidy policy marks a new stage in the development of electric vehicles in China. To meet the new development requirements, China revised the “double point” policy in 2020 and optimized the method of points. The theme of China’s electric vehicle market has shifted from rapid development to healthy and rational development. In July 2021, the EU officially released a package of legislative proposals called “Fit for 55” to reduce the greenhouse gas emissions by at least 55% by 2030 (based on greenhouse gas emissions in 1990) and achieve carbon neutrality by 2050 through energy, transport, taxation, and other policies. The bill stipulates that the average emission from new additional passenger and commercial vehicles will decrease by 55% in 2030 and 100% in 2035, accelerating the transition to zero-emission vehicles. The proposed law basically predicts that after 2035, conventional fuel cars will completely exit from the European market.

6.1.2.3 Segmentation – Electric Buses

Electric bus is the most successful application scenario of lithium battery. In the initial stage of lithium battery application, people first turned their attention to the city bus field. Buses have the characteristics of fixed routes, short mileage, convenient infrastructure reform, and charging convenience at night. Cities also require the buses with zero emission and low noise during development. Thus, buses are one of the most suitable application scenarios of lithium-ion power battery. China is a global leader in electric buses. The statistical data from the IEA [8] show that by 2020, there were 515000 electric buses in the world, including 502000 in China, accounting for 97.5%. According to the official website of China’s Ministry of Ecology and Environment [14], the overall penetration rate of electric buses in China has reached 60%, and the buses in cities such as Shenzhen, Zhuhai, and Changsha are 100% electric buses. In recent years, the development of electric buses in some European countries has been significantly accelerated. The new energy buses sold (including electric buses) in Denmark, Luxembourg, and the Netherlands all account for more than 66%. For pure electric buses, battery decay is a serious problem. As public vehicles, electric buses are repeatedly charged and discharged and operated at high load throughout their whole life cycle, resulting in a greatly advanced decay of lithium-ion power batteries. As reported by Nanfang Metropolis in 2018, the batteries of 60 pure electric buses made by Shenzhen Bus Group Co., Ltd. generally deteriorate, although they were only five years old and had a minimum range of 50 km. Low endurance mileage leads to frequent charging and discharging and thus greatly reduces the service life of lithium-ion power batteries, i.e. the batteries expired earlier than passenger vehicles.

National policies vigorously promote the development of electric buses. According to China’s *New Energy Vehicle Industry Development Plan (2021–2035)*, electric vehicles in the public sector will be fully generalized in 2035. At the end of 2020, the Ministry of Finance of China issued a *Notice on Allocating the 2021 Energy Conservation and Emission Reduction Subsidy Budget in Advance* (the first batch),

which specifies that a total of 37.58 billion yuan of subsidies for new energy vehicles will be allocated in 2021, including 15.689 billion yuan for new energy buses, accounting for 41.74%. Thus, China will promote the application of new energy vehicles in the public sector in the future, conducive to the further improvement of the percentage of electric buses. Furthermore, European countries issued a series of important policies in 2020. *French Resilience and Recovery Plan* states that 30000 euros will be granted to the buyer of an electric bus. Germany announced financial subsidies for 80% of electric buses. The UK has released a *Ten-Point Plan for a Green Industrial Revolution*, which announces that 5 billion pounds will be invested to accelerate the application of electric buses and 120 million pounds will be invested to introduce 4000 home-made zero-emission buses by 2021. Poland has announced a 290 million euros subsidy for zero-carbon emission from all buses in the cities with the population of more than 100 000 by 2030.

Forecast of installed capacity of lithium-ion power battery of electric bus. According to the IEA [8], there will be 3.218 million electric buses by 2030 under current policies. Assuming that the sustainable development goal is achieved in a cost-effective manner, the number of electric buses is expected to reach 5.104 million. According to the average installed capacity of 200 kWh for each pure electric bus, the electric battery demand of pure electric buses will reach 643.6 GWh to 1 TWh by 2030.

6.1.2.4 Segmentation – Electric Heavy Trucks

Application of electric heavy trucks brings significant environmental benefits. In 2021, the Ministry of Ecology and Environment of China released the *China Mobile Source Environmental Management Annual Report (2021)* [15]. The annual report shows that in 2020, the total emission of four pollutants (carbon monoxide [CO], hydrocarbon [HC], nitrogen oxide [NO_x], and particulate matter [PM]) from motor vehicles in China reached 15.93 million tons; the pollutant emission from motor vehicles accounted for 93.3%; and the NO_x and PM emissions from heavy trucks accounted for 74% and 52.4% of those emissions from all vehicles, respectively (Figures 6.8 and 6.9).

According to the *2020 Statistical Bulletin on the Development of the Transport Industry* released by China's Ministry of Transport [16], China has 11.1 million trucks of all types. Statistics show that heavy trucks (heavy commercial vehicles with gross weight of more than 3.5 tons, including heavy trucks and heavy special purpose vehicles) account for less than 5% of the total vehicle parc, but they have the biggest pollutant emissions. The application of electric heavy trucks will boost energy saving and emission reduction in the field of road transport and achieve significant environmental benefits; thus, the electric heavy trucks are imperative.

Electric heavy trucks are supported by policies in various countries. In October 2020, China's Ministry of Industry and Information Technology issued a document in response to the proposal "on fully promoting the application of electric construction machinery and heavy trucks and other public service vehicles to win the battle of blue sky to form a global industrial highland" put forward by CATL Chairman Zeng Yuqun during the two sessions of that year. The Ministry of Industry and

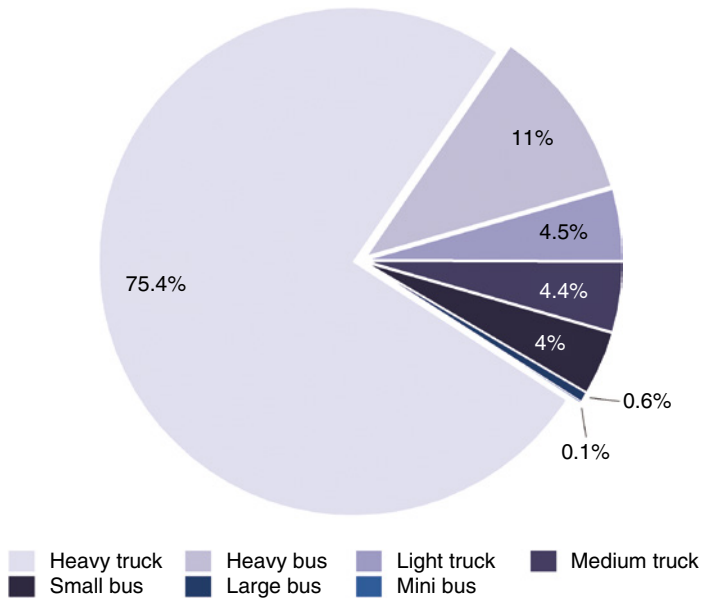


Figure 6.8 Percentage of NOx emissions from various types of vehicles. *Source:* Ministry of Ecology and Environment of the People's Republic of China [15].

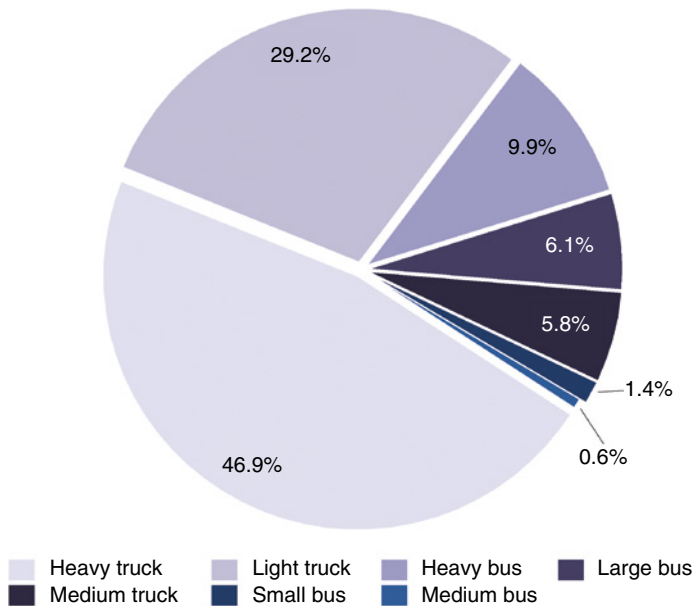


Figure 6.9 Percentage of particulate matter emissions from various types of vehicles. *Source:* Ministry of Ecology and Environment of the People's Republic of China [15].

Information Technology highly agreed with the development of electric heavy trucks and announced the implementation of the *Action Plan to Promote the Electrification of Public Sector Vehicles* in that month, with key deployment arrangements for the pilot application of heavy trucks, to promote the application of electric heavy trucks in short-distance transport, urban construction logistics, mining, and other special scenes. In July 2020, the California Air Resources Board announced a major policy which requires that automakers shall sell more zero-emission vehicles since 2024 and ensure that all large trucks they manufacture are electric trucks by 2045. Since then, 14 states have responded by pledging to increase the penetration rate of large electric vehicles and ensure zero emissions from all medium and heavy vehicles manufactured by 2050. According to the European Automobile Industry Association, the truck manufacturers in Europe agreed at the end of 2020 to achieve zero emissions from all heavy trucks sold by 2040 and full carbon neutrality by 2050. Due to a huge gap between reality and targets, the EU is working with national governments to accelerate the development of electric heavy truck charging network, while developing a coherent policy framework to support the industry.

The installed capacity of lithium-ion power batteries for electric heavy trucks is expected to grow rapidly. According to IEA, there are about 31 000 pure electric heavy trucks in the world by the end of 2020. In 2020, 7470 electric heavy trucks were sold worldwide, including 7409 pure electric trucks, accounting for 99.2%, which was 13.3% higher than that in the previous year. Furthermore, the IEA expects that the pure electric heavy truck parc is 860 000 from 2020 to 2030 under current national policies. Assuming that the sustainable development goal is achieved in a cost-effective manner, the number of pure electric heavy trucks is expected to reach 2 million by 2030. The number of worldwide heavy trucks has increased from 31 000 to 860 000, with the compound annual growth rate of 267% in 10 years. The average installed capacity of each heavy truck is about 300 kWh, the conservatively estimated and optimistically estimated installed capacity of lithium-ion power battery of pure electric heavy truck in the world will reach 258 and 600 GWh by 2030, respectively. China is a huge market for both upstream material manufacturers and battery manufacturers.

Cost, weight, and range are the three major indices of electric heavy trucks. In recent years, the prices of lithium iron phosphate (LFP) and NCM has dropped from about 1800 and 1700 yuan/kWh in 2017 to about 730 and 785 yuan/kWh in 2022, respectively. According to the above estimated installed capacity of lithium-ion power battery of 300 kWh per heavy truck, the battery cost per heavy truck will be reduced to 219,000 yuan on average. Nonetheless, the total price of an electric heavy truck is still high at present, over once higher than that of a fuel vehicle at the same level. In spite of low subsequent use cost, many people still hesitate to choose an electric heavy truck due to its high price. Furthermore, to provide long endurance mileage, vehicle manufacturers often install too many battery packs in electric heavy trucks, resulting in heavy weight. For example, the pure electric 6×4 tractor developed by Universiade and Wal-Mart has the net weight of 14 tons, and its photodynamic lithium battery has the weight of 7 tons, accounting for half of the gross weight. That not only reduces the efficiency of transport but also causes harm to road infrastructure. The short life of a battery is a common problem for electric

heavy trucks, and the electric heavy trucks with battery on the market have a range of 200–400 km. In addition, insufficient charging facilities limit the use of electric heavy trucks.

In a word, the application of electric heavy trucks is the general trend, and most countries have made various promotion plans. The urgent development requirements for the electric heavy trucks have been put forward in both the current policy and the goal of carbon emission reduction. Although it is a huge market, some major problems must be solved. Director General of the European Automobile Industry Association said that the development of electric heavy trucks depends not only on the technology but also on the deployment of proper infrastructure and the transfer of cost burden.

6.1.2.5 Development Outlook of Electric Vehicle EV Battery

In terms of battery materials, the energy density of LFP and NCM is increasing. On 8 January 2021, Gotion announced in Hefei that the energy density of its soft pack LFP cell reached 210 Wh/kg, comparable to the level of the ternary NCM5 system. On 20 June 2021, Penghui Energy Company announced that the energy density of the LFP battery also exceeded 200 Wh/kg. As the world's leading enterprise of lithium-ion power battery, CATL also plans to increase its energy density to 200–230 Wh/kg by designing and optimizing the LFP material system. In terms of NCM batteries, the energy density of the high-nickel chemical system as cell system developed by CATL reaches 215 Wh/kg. From the perspective of its material system, it is expected that the energy density of ultra-high nickel + silicon system can reach 400 Wh/kg in 2024. The first and second generations of sodium batteries have the energy density of 160 Wh/kg (comparable to the level of ordinary LFP batteries) and 200 Wh/kg, respectively.

In terms of structure, the leading enterprises in the industry begin to improve the efficiency of structural integration of systems by module-free technology (CTP, CELL TO PACK). BYD's lithium-iron phosphate blade battery has thin, long blade-shaped cells instead of a shell beam. These innovative structural changes not only improve safety but also the volume efficiency of the battery system from 40% to 60%. In March 2021, GAC released the “magazine battery.” It is said that such magazine battery uses a NCM battery and has characteristics such as no needle fire, ultra-high heat resistance, ultra-strong insulation, and rapid cooling. The magazine battery is very attractive now. It has a bit lower volume energy density than blade battery, but it is highly popular due to the safety of NCM battery. Blade battery and magazine battery have their own advantages, which reflect the continuous progress in the structure technology of lithium-ion power battery.

The battery market demand will grow in the next decade. The data from the IEA show that under current national policies, there will be 8 million pure electric passenger vehicles, 3.22 million electric buses, and 860 000 electric heavy trucks worldwide in 2030. The pure electric passenger vehicles, electric buses, and electric heavy trucks have the lithium-ion power battery capacity of 80, 300, and 300 kWh, respectively. Accordingly, the market demand for lithium-ion power battery will reach 5.8 TWh in the next decade. That is a huge market.

6.1.3 Electric Ships

6.1.3.1 Background

In 2018, the International Maritime Organization (IMO) released a preliminary strategy for reducing greenhouse gas emissions from shipping. The strategy states that the total greenhouse gas emissions from the global shipping industry will be reduced by at least 40% from the level in 2008 by 2030 and then by 70% by 2050, and finally to zero by the end of this century. With increasingly stringent requirements for energy saving and emission reduction in the shipping industry, many ports in the world have implemented strict standards for greenhouse gas emission from ships, thus forcing the ship manufacturing to shift to cleaner ships. Compared with the traditional marine power system, electric propulsion has the advantages of zero emissions, high technical added value, easy integration and standardization, low operating costs, and ecological acceptance and comprehensive benefits. In this context, electric ships have developed particularly significantly in recent years, and the market size is rapidly expanding (Figure 6.10).

6.1.3.2 National Support Policies

Norway is a global leader in electric ships. To enhance its competitiveness and core position in the shipping industry, Norway formulated the *Green Shipping Action Plan* in 2019 to reduce the emission from domestic shipping and fishery by a half by 2030. To promote the reduction of greenhouse gas emission in the ports, the Norwegian Government will work with municipalities and port authorities to achieve zero emissions in the ports by 2030. Through the implementation of the *Green Shipping Action Plan*, Norway is playing a leading role in the green transformation of international shipping. In the plan, different low-emission and zero-emission technologies are designed for all types of vessels. For example, pure battery power technology is suitable for ships sailing in short distances or conventional

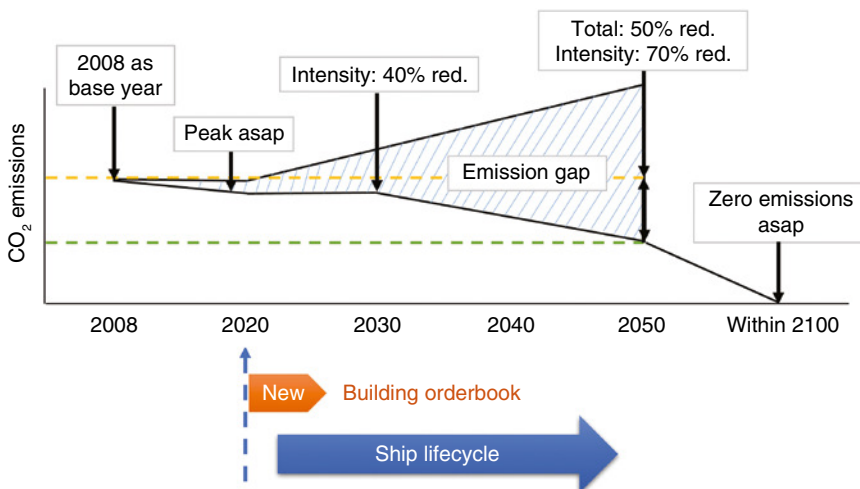


Figure 6.10 Preliminary shipping GHG mitigation strategy. Source: DNV GL [17].

routes, such as ferries and cruise ships, while hybrid power technology is suitable for ships sailing in long distances, such as international passenger ferries and cargo ships.

DNV GL has developed several tools to promote the application of battery and electric technology for ship propulsion, such as pilot classification specifications for battery power, guidelines for large marine battery systems, new tools for certification of battery systems, battery preparation services (technical, economic, and environmental performance analysis), battery estimation and optimization tools, and introductory courses for marine battery systems.

In early 2021, the South Korean government announced the *Mid and Long-Term Plan for Green Ships of South Korea*, known as the *2030 GreenShip-K Promotion Strategy*. This plan mainly promotes the development of advanced environmentally acceptable ship technology, improves the experimental basic conditions for the industrial application of new technology, and generalizes the Korean ship verification project, construction of fuel supply infrastructure and operation system, popularization of green ship, establishment of ecological system of green ship market, etc. To ensure that the green ship technology is worldwide leading in the future, South Korea supports the localization and upgrading of the associated core equipment such as LNG, power, and hybrid and supports and promotes the systematic and comprehensive development of low-carbon ship technology (including mixed fuel), carbon-free ship technology (including hydrogen and ammonia), etc. The plan requires that the greenhouse gas emissions from ships will be reduced by 20% by 2020, 40% by 2025, and eventually 70% by 2030.

With increasing emphasis on environment, the Chinese government has put forward new requirements for marine diesel engine exhaust emissions in recent years. In July 2018, the State Council issued the *Three-year Action Plan to Win the Battle against Blue Skies*, which proposes the upgrading of the standards for emissions from marine diesel engines and expansion of the emission control area. The Ministry of Transport issued the *Implementation Plan for Emission Control Zones for Ships*, which requires the expansion of emission control areas and gradual improvement of emission standards. In addition, the *Action Plan for Promoting green Development of Pearl River Water Transport* (2018–2020) was issued. However, these documents focus on the application of LNG power and shore power technology, and they play a limited role in the promotion of battery-powered ship technology. In October 2019, the National Development and Reform Commission issued the *Guidance Catalogue for Industrial Structure Adjustment (2019)*, in which “pure electric and natural gas vessels; alternative fuel, hybrid, plug-in hybrid engines, optimized power assembly system compatibility” are added as the national encouraged products. The catalog has been officially implemented since 1 January 2020.

6.1.3.3 Global Market Development Status

According to the statistics of Maritime Battery Forum, as of 21 July 2021, 490 electric ships are in operation and will be built worldwide, including 330 electric ships in operation and 160 electric ships to be built. From 2021 to 2027, the number of electric ships in operation has hardly changed, while the number of electric ships to

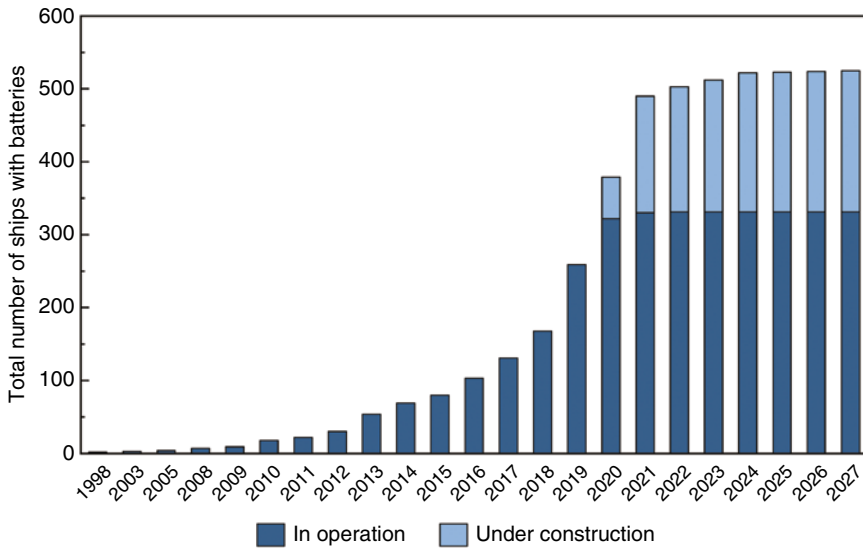


Figure 6.11 Trends in the number of the global electric ship market. *Source:* DNV website, Maritime Battery Forum 2021 [17].

be built has increased from 160 to 194 in 2021, showing a fast growth. According to a study from Markets and Markets in 2019 [17], the global electric ship market is expected to grow from \$5.2 billion in 2019 to \$15.6 billion in 2030, with a projected CAGR of 13.2% from 2025 to 2030 (Figure 6.11).

The first large pure electric propulsion ship “Ampere” [18] in the world is owned and operated by Norled. The integrated DNV GL class ferry designed and built by Fjellstrand is an pure aluminum-powered catamaran with an innovative propulsion system and an extremely efficient hull. Its commercial operation began in 2015. The 80 m-long vessel with 1040 kWh NCM battery pack can carry 120 vehicles and 360 passengers and sail across the Sonn Fjord between Lavik and Oppedal in Norway.

The vessel has a total of 34 sails, 20 minutes each. In the ports, it takes only 10 minutes to fully recharge the 1 MWh lithium-polymer battery pack in the vessel. The power required to charge the vessels exceeded the load of the power grids in Lavik and Oppedal; hence, buffer battery packs were installed in both ports. These buffer batteries can be continuously charged by the grids before they can quickly charge the ferry batteries.

“Ellen” – Denmark (2015): Ellen was built at the cost of 21.3 million euros in 2015 and operated in 2019. The ship is nearly 60 m long and 13 m wide, travels at speeds between 13 and 15.5 knots (knots/h) and can carry 198 passengers in summer and 147 in winter. The ferry’s open deck can also carry 31 cars or 5 trucks. The ferry has the battery capacity of 4.3 MWh. A fully charged battery has the endurance mileage of 22 nautical miles. The vessel sails in the 22-nautical mile waterway between Ayr and Fiingshaaf in Denmark, seven times as long as that of any other electric ferry previously in operation around the world.

To minimize its weight and electricity consumption of Ellen, the bridge is made of aluminum instead of steel, and the furniture is made of recycled paper instead of wood. Ellen's 4.3 MWh battery system is a high-energy G-NCM lithium battery (supplied by Leclanche) with unique safety features, including a double-layered design and ceramic separator. The battery system consists of 20 units, each of which is connected to an independent converter that controls the energy output. Ellen is the first electric ferry without an emergency generator in the world.

Three Gorges No. 1 – China [19]: The construction of the world's largest pure electric green commercial ship, Yangtze Three Gorges 1, which was jointly developed by CATL, China Changjiang Power Co., Ltd., and Hubei Yichang Transport Group Co., Ltd., began at the end of 2020. A total of 150 million yuan will be invested in this cruiser, which can accommodate 1300 seats. It is a pure electric passenger ship with the largest battery capacity, the most passenger seats, and the highest level of intelligence in the world. In terms of battery capacity, "Three Gorges No. 1" has made a breakthrough. The ship is equipped with the CATL LFP lithium-ion power battery, with the total power of 7.5 MWh, equivalent to the total battery capacity of more than 100 electric vehicles. After charged for six hours a night, it can sail 100 km.

Dianjing – China [20]: In July 2021, "Dianjing," a pure electric luxury cruise ship supported by AVIC Lithium, completed its first trial voyage, marking the further development of AVIC lithium in the electric ship market. "Dianjing" is 41.5 m in length, 9 m in width, and 2.3 m in depth. It is equipped with the standardized product of a lithium modular ship power supply system of AVIC, with the battery capacity of 1200 kWh, speed of 15.5 km/h, and full load capacity of 200 passengers. It is a pure electric tourist ship that can carry the most passengers in inland lake. "Dianjing" resists superior wind and wave and tilts very well. The ship has a pure electric propulsion system with advantages over the traditional fuel oil system, such as low noise and no pollution. It is an excellent "water bus" of Dian Lake, integrating the advantages of stability, comfort, and environmental acceptance. Passengers can enjoy the beautiful scenery and protect local environment as much as possible while traveling around Dian Lake on the "Dian Jing."

6.1.3.4 Battery Technology for Electric Boats

According to the data of the Maritime Battery Forum in 2018 [17], ternary NCM cells are most used in electric ships, in about 60% of electric ships, followed by LFP (LiFePO_4), in about 15% of electric ships. This is because electric ships developed early abroad, and the early LFP had low energy density; many countries were prone to use of ternary NCM lithium-ion power battery to meet the requirements for endurance mileage. Considering the potential safety risk of NCM cell batteries, most marine NCMs have low nickel content.

Different from the automotive lithium battery, the marine lithium battery has high requirements for safety. Due to the large passenger capacity of ships, especially sightseeing ships and barges, and the huge electric quantity of battery, it is difficult to escape in case of fire or explosion. Therefore, the ship batteries have very low

tolerance of accident risk. LFP batteries have inherent advantages over NCM batteries, i.e. higher safety, higher temperature resistance, lower cost, and longer life than NCM batteries. Therefore, China has chosen a completely different route from other countries, i.e. LFP is used in the lithium batteries of electric ships.

Recently, CATL officially released the first generation of sodium ion batteries with the energy density of 160 kWh/kg, very close to that of the LFP battery [21]. Furthermore, it has the characteristics of high integration efficiency, quick charging performance, and good performance at low temperature and thus eliminates the defects of LFP. CATL proposed the “integrated hybrid sharing” system scheme, that is, the sodium ion battery and lithium battery are proportionally connected in series and parallel in the battery module. This scheme may provide a new choice for technical route of lithium batteries for marine power.

6.1.3.5 Development Outlook of Electric Boat Power Battery

By 2018, China’s Yangtze River and Beijing-Hangzhou Grand Canal and other inland river basins have more than 60 000 ships with the total tonnage of 195 million tons. Based on the electric charge of 1.3 kWh per ton and rate of utilization in electric ships of 50%, the market demand for ship lithium battery in 2018 alone will exceed 100 GWh. However, the actual output of marine lithium battery in China in 2020 was only 75.6 MWh, with the market size of 95 million yuan [22].

Marine lithium battery has huge market demand and very low rate of lithium utilization because (i) electric ships have small endurance mileage and slow power supply and thus low operating efficiency; (ii) the relevant standards of electric ships are not perfect; (iii) there is no policies for electric ships; and (iv) a mature business model has not been established due to the high investment cost of the battery-swap station. At present, only some leading enterprises with certain technical and financial strength participate in the electric ship market. To revitalize the electric ship market in the future, the upstream and downstream enterprises of the industrial chain must work together to break through key technologies and improve the supporting facilities.

6.1.4 Energy Storage Device

6.1.4.1 Segmentation – V2G Technology

Vehicle to Grid (V2G) integrates lithium-ion power battery with power grid very strongly. Due to the rapid growth of new energy vehicles, especially electric vehicles, the huge demand for charging and energy storage brings great pressure to the power grid and provides opportunities. V2G technology can realize the interaction between the lithium-ion power battery and the power grid. The battery can be charged from the power grid in the off-peak stage of power grid and can be discharged to the power grid in the peak stage of power grid, with certain income. On the one hand, such two-way benign interaction improves the utilization efficiency of electric energy at the grid end, enhances the stability and security of the grid, and realizes “peak cut.” On the other hand, it can bring additional income to car owners, vehicle operators, charging and battery-swap stations, etc.

Nuvve Corporation, a green energy technology company of the United States, is a pioneer in V2G technology. This company has core patents and technologies, which connect several electric vehicles to virtual intermediate power plants and to the grid in a compliant and safe manner to achieve two-way charging and discharging. The company works together with Danish government in Paker project between 2016 and 2018 to build the world's first commercialized V2G center, which commercially operates V2G technology very well. The first batch of test vehicles includes 30 vehicles in the project. The owners of the vehicles will receive about \$10 000 at the end of the vehicle's life (about eight years) according to the agreement and market price.

The New Energy Vehicle Industry Development Plan (2021–2035) released by The General Office of the State Council of China in 2020 states that to promote the integrated development of new energy vehicles and energy, it is necessary to strengthen the interaction between new energy vehicles and the power grid (V2G) energy, strengthen the research of high-cycle life lithium-ion power battery technology, promote the application of small power DC technology, encourage local V2G application demonstration, collectively allocate new energy automobile charging and discharging and power, use the preferential policies for peak valley price and new energy car battery, efficiently interact between new energy vehicles and power grid energy, reduce the electricity costs of new energy vehicles, and increase the corresponding ability to control the power grid load and frequency.

In recent years, many provinces and cities in China have launched demand response to actively explore the interaction between vehicle-to-grid and energy. Among them, Shanghai is leading in the development of new energy vehicles; hence, its pilot projects are the most exemplarily significant. On 3 June 2020, The China Automobile 100 Association and the Natural Resources Defense Council jointly released a report entitled “*Business Prospects of Electric Vehicle-to-Power Grid Interaction – A Pilot Case of Demand Response in Shanghai*” [23]. In the pilot project, lithium-ion batteries will be connected to the grid through three bidirectional charging platforms: private charging stations, public charging stations, and battery-swap stations. The report shows that when the investment cost of charging stations of demand response load aggregators is fixed and the unit price of response compensation is fixed, the frequency of participation in demand response determines the income level. The estimates show that the average annual response frequency reaches 10, and the internal rate of return of private charging stations participating in demand response can reach 27%. The dedicated charging stations have much higher participation rate of demand response than private charging stations, and the average annual response frequency of 10 and the internal rate of return of 50%. Due to its strong charging time control capability, the battery-swap station has much higher economy of participating in demand response than private and dedicated charging stations. By 2019, Shanghai had 190 000 private charging stations, 50 000 public charging stations, and 40 000 dedicated charging stations. During the pilot test, the private charging stations, dedicated charging stations, and battery-swap stations have the response rate of only 5.3%, 75%, and 81.2%, respectively.

According to the report, it is technically feasible to change the lithium-ion power batteries into the distributed energy storage devices through V2G technology, with a

large market space. In the development of V2G, it is necessary to greatly improve the cycle life and energy density of lithium-ion power battery to explore a sustainable business model, gradually establish a stable market mechanism, design reasonable incentive policies, fully tap the potential of private charging stations, and build the industry standard system.

6.1.4.2 Segmentation – Cascade Utilization

Policies give the development opportunities to the cascade utilization of decommissioned lithium-ion power batteries. With the arrival of the first wave of decommissioned electric vehicle batteries, the government and enterprises have realized the importance of decommissioned lithium-ion power battery recycling. As early as 2012, The State Council of China issued the *Development Plan for energy-saving and New-energy Vehicle Industry (2012–2020)*. The plan states that the management methods for recycling and utilization of lithium-ion power batteries and a cascade utilization and recycling management system shall be established to encourage the development of professional battery recycling enterprises. Decommissioned batteries always have about 80% capacity and a long cycle life; thus, they can be used as storage devices. In this context, many enterprises have planned to build cascade energy storage power stations.

On 22 May 2018, the lithium battery cascade energy storage power station of Jiangsu Changneng New Energy Technology Co., Ltd. was completed and put into use in Wujin National High-tech Zone Innovation Industrial Park. The station uses the decommissioned power lithium-ion batteries and has a total capacity of 10 MW.

The energy center of an industrial park in Shanghai invested in the 50 kW/150 kWh storage containers, which were assembled and tested in the Great Wall Power Plant and shipped smoothly in June 2018. The 150 kWh lithium battery of this project is made from all decommissioned lithium-ion power batteries of electric vehicles, and the unpacked battery pack is directly utilized in the cascade.

On 1 September 2018, the Chinese industrial and commercial energy storage system project for 1 MW/7 MWh cascade utilization was successfully put into operation in Nantong Rudong, Jiangsu Province. The project was jointly undertaken by Xuda New Energy Co., Ltd. and Zhongheng Puri Co., Ltd., and accepted, metered by meters, and commercially operated by Nantong Power Supply Bureau. Relying on the advantages of energy storage converters and management system development, Xuda New Energy builds a cascade lithium electric energy storage system by combining the original group series energy storage converters with the decommissioned lithium-ion power batteries of electric vehicles.

In August 2019, the 2.15 MW/7.27 MWh cascade battery energy storage project jointly built by Shenzhen Bic Lithium-ion power battery Co., Ltd. and China Southern Power Integrated Energy Service Co., Ltd. was successfully put into operation [24]. As a user-side energy storage project, it will be applied to industrial and commercial parks. It mainly functions to cut the peak and provide auxiliary power services (Figure 6.12).

According to BloombergNEF [25], the installed capacity of the decommissioned lithium battery cascade utilization energy storage project has reached 5.5 GWh by

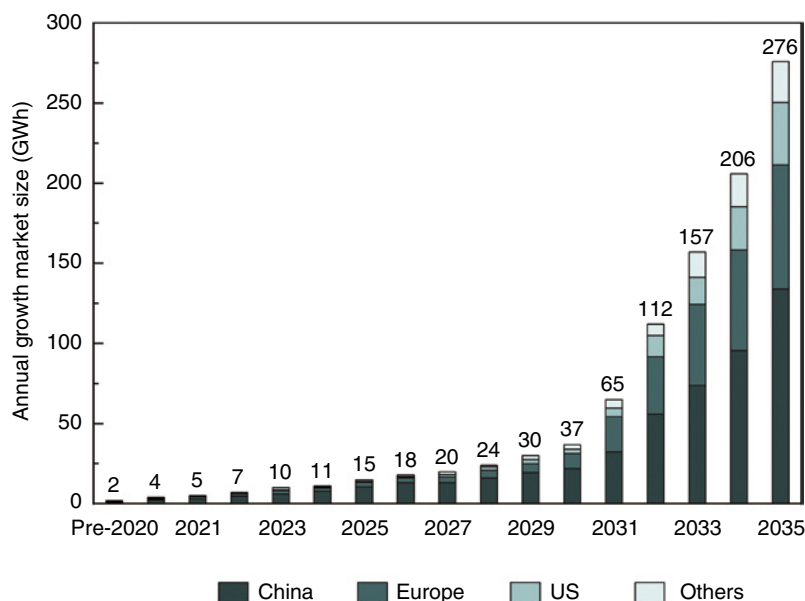


Figure 6.12 Annual growth market size of cascade utilization. *Source:* Bloomberg New Energy Finance [25].

May 2021. The scale of decommissioned lithium-ion batteries is expected to sharply rise after 2030, and the annual supply of cascade utilization products is expected to reach 276 GWh by 2035.

Safety accidents occur frequently; hence, the cascade utilization of decommissioned power lithium-ion batteries reaches a low point. In recent years, many safety accidents have occurred in the electrochemical energy storage power stations across the world. In April 2021, a fire broke out at an energy storage power station in Fengtai District, Beijing. Due to a sudden explosion during disposal, two firefighters died, another firefighter was injured, and an employee of power station went missing. In July 2021, a fire broke out at Victoria Battery project in Tesla, Australia. It took four days to control the fire. The safety accidents in energy storage power station led to the question about the safety of decommissioned lithium-ion power batteries.

On 22 June 2021, the Comprehensive Department of The National Energy Administration of China (NEA) issued the “*New Energy Storage Project Management Code (Interim) (Draft)*” (hereinafter referred to as the Code). The Code stresses that large-scale lithium battery cascade energy storage projects shall not be built in principle to avoid high safety risks. Before the decommissioned power lithium-ion batteries are recycled, their residual value can be fully utilized through cascade utilization, but they also face problems such as inconsistent quality, short cycle life, and thermal runaway risks. Different from the energy storage projects that use new batteries, the energy storage projects that use cascade utilization lithium-ion power batteries face higher safety risks. Therefore, when the relevant management

methods and technical conditions of decommissioned power lithium-ion batteries are not mature in the present stage, the large-scale lithium-ion power battery cascade utilization energy storage projects have very high safety risks, which are hardly controlled effectively. An effective measure to avoid safety risks is not to build large-scale lithium-ion power battery cascade utilization energy storage projects. The code requires that in principle, large-scale lithium-ion power battery cascade utilization energy storage projects shall not be built until the battery consistency management technology has been broken through and the performance monitoring and evaluation system of lithium-ion power battery is perfected. The battery performance shall be evaluated regularly, monitored, and supervised more frequently.

Safety first is the premise of decommissioned lithium-ion power battery cascade utilization in the future. According to incomplete statistics, 32 fires or explosions have occurred at the energy storage plants worldwide since 2011. All the batteries used in these storage stations are new batteries. It is conceivable that it is impossible to guarantee the safety of decommissioned power lithium-ion batteries under the existing conditions. On the one hand, the number of decommissioned power lithium-ion batteries sharply increases. On the other hand, more stringent requirements for safety are put forward. Therefore, it is extremely urgent to technically break through the consistent management of lithium-ion power batteries and establish and improve the performance monitoring and evaluation system of lithium-ion power batteries.

6.1.4.3 Development Outlook of Energy Storage Power Battery

In terms of battery materials, it is necessary to make a breakthrough in key technologies of lithium-ion power batteries, such as ultra-long life under high frequency charge and discharge. In the use stage, the lithium-ion power battery, as a mobile energy storage unit, is deeply integrated with the power generation terminal, conducive to healthy and orderly development of the power grid. It is expected to bring major development opportunities in the future. In terms of the market, new business models are needed to support the promotion and application of V2G technology so that most car owners can make profits and the vehicle-to-grid can interact and develop sustainably and healthily. In principle, the scrapped lithium-ion power batteries can be used in storage facilities on the premise of safety guarantee.

6.2 Development of Emerging Business Mode

6.2.1 Power Swap Mode of Two-Wheeled Vehicles

6.2.1.1 Industry Demand Drives the Development of Power Swap Mode

6.2.1.1.1 Instant Delivery (2B)

Two-wheeled electric vehicles are the most important means of transport for 13 million people employed in China's fast growing instant delivery industry, such as food delivery, express, and errands. According to statistics, full-time delivery workers generally have the cycling mileage of more than 100 km per day, while the current

general two-wheeled electric vehicles have endurance mileage of about 50 km. Many delivery workers have had to install larger and heavier batteries on their vehicles to improve their battery life, leading to safety concerns.

The power swap mode of two-wheel electric vehicle can solve this problem very well. Practices have proved that each deliveryman can run 10 more orders per day by saving charging time in the power swap mode. Based on the average delivery fee of 6 yuan per order, a deliveryman can earn additional 2000 yuan and pay the power swap service fee of only 300 yuan every month. Therefore, for terminal 2B users, the power swap mode means higher labor income. It is expected that the penetration rate of the power swap mode will reach 50% in the future.

6.2.1.1.2 Shared Travel (2B)

The development of sharing economy has also boosted the power swap mode of two-wheeled electric vehicles. The number of shared bikes in China reached 1 million by the end of 2019 and is expected to exceed 8 million by 2025 [26]. The most effective way to prolong the endurance mileage of shared motorcycles certainly is the power swap mode, which has the penetration rate of 100% in this scenario. Like the shared charging bank, the shared two-wheeled electric vehicles are picked up and returned at the fixed places. Operation and maintenance personnel know the remaining electric quantity of the vehicles through the background monitoring system, replace the batteries of the vehicles with low electric quantity, and put the replaced batteries into the power swapping cabinet for charging.

6.2.1.1.3 Individual Cycling (2C)

China has changed from bicycle kingdom to electric bicycle kingdom. In the large personal vehicle market, the electric power swap mode has very low penetration rate, and self-charging is still dominant at present mainly because the power swap mode has higher cost than the self-charging mode. A large amount of social capital is invested, leading to lower cost, more advantages, and bigger market space of the power swap mode. The preliminary estimate shows that the penetration rate of the power swap mode can reach 25% in the future (Figure 6.13).

6.2.1.2 New National Standard Boosts the Power Swap Mode

The new national standard mentioned above has significantly influenced the development of two-wheeled electric vehicles, indirectly promoted the rise of the power swap mode, and put forward more stringent requirements for vehicle safety. People begin to find the way to prolong the battery life under the premise that the vehicle body has lightweight and sufficient strength. The power swap mode can well meet this demand. One mobile phone and one APP can replace the battery in one minute and thus achieve an unlimited endurance mileage.

6.2.1.3 Battery Swapping Cabinet – A Huge Blue Ocean Market is Forming

Just like the development process of mobile phone shared charging bank in China, the market of power swap cabinet as the shared power package of two-wheeled electric vehicles is competitively laid out. A huge blue ocean market is forming.

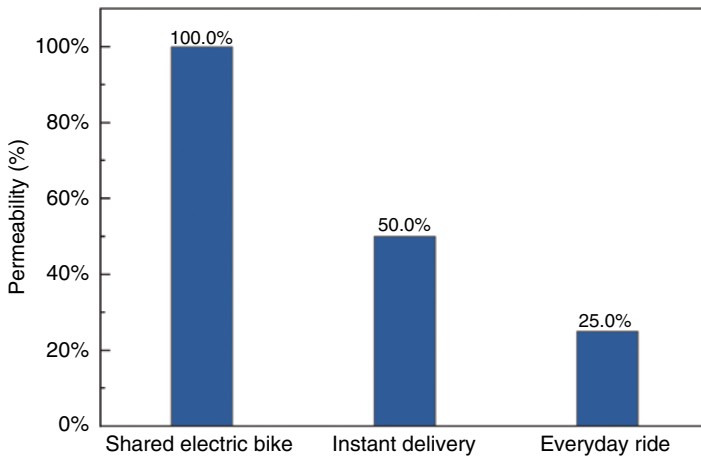


Figure 6.13 Estimated penetration rate of power swapping electric two-wheeled vehicles.

According to statistics, China's demand for battery swapping cabinets is expected to reach 1 million. Based on the unit price of 15000yuan, the corresponding market scale can be as high as 15billion yuan [27]. In terms of power swap operation, the volume of C-terminal is very large, reaching nearly 300million vehicles. Based on the daily charging frequency of 100million and annual charging frequency of 36.5billion and charging fee of 2–3yuan per charge, the estimated market scale is 70–100billion yuan.

At the end of 2018, over 300million yuan was financed in Round B for e-power swap, tens of millions of dollars were financed in Round B+ in the following May, and several was financed in Round C1 in 2020. By February 2021, E-power swap has been stationed in more than 60 cities in China, and nearly 10000 battery swapping cabinets have been installed, with 600000 power swaps every day on average [28].

In 2019, Hellobike, Ant Financial, and CATL jointly invested 1billion yuan to launch Hello power swap service, also known as Hello Power swap. In 2021, hundreds of millions of yuan were financed by Harbin Power Swap Business from the Greater Bay Area Fund, Pangu Capital, Muhua Capital, etc. By the end of June 2021, Hello power swap has distributed in more than 2000 stores in more than 200 cities across China [29].

In March 2019, 100million yuan was financed by EQI in Round B from Tencent, NIO, Kaihui and New Cognition United. At present, EQI has been deployed in nearly 10000 battery swapping cabinets in more than 30 cities [30]. Other power swap operators include Leifeng power swap, Fly Brother power swap, and Zhang Fei power swap, thus showing a prosperous scene.

6.2.1.4 Battery Technology for Power Swapping Two-Wheeled Electric Vehicles

At present, the two-wheel electric vehicles on the market generally use the lead–acid batteries with large volume and weight. Although lead–acid batteries dominate in the market at this stage, the lightweight lithium-ion power battery is an

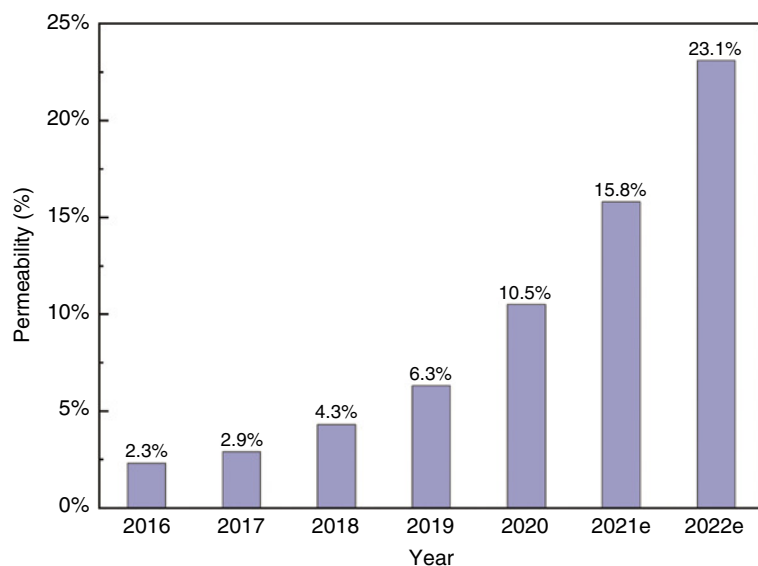


Figure 6.14 Predicted percentage of lithium batteries in the Chinese two-wheeled electric vehicles [31].

irresistible trend. Two-wheeled electric vehicles are gradually approaching to lithium batteries. Lithium batteries have many advantages over traditional lead-acid batteries, such as high energy density, long cycle life, small volume and weight, and applicability to power swap scenarios.

In recent years, the market penetration rate of lithium battery in Chinese two-wheeled electric vehicle continues to rise (Figure 6.14). Statistics [31] show that China had the total output of 48.34 million two-wheeled electric vehicles in 2020, with annual growth of 27.2%. China has the total output of 11.36 million lithium battery two-wheeled vehicles, with the penetration rate of 23.5% and annual growth of 84.7%. It is expected that the lithium two-wheeled electric vehicles will have the market share of over 23.1% by 2022. It is the general trend to replace lithium battery with lead-acid battery.

6.2.1.5 Advantages and Disadvantages of the Power Swap Mode

6.2.1.5.1 Advantage

(i) The power swap mode enhances the convenience and endurance mileage of the two-wheel electric vehicle. With the popularity of the battery swapping cabinet, people will never worry about the vehicle battery charging. With only one mobile phone, the “10-second battery swap” will become real and achieves an unlimited endurance mileage at any time anywhere; (ii) the power swap mode improves the safety of the two-wheeled electric vehicles; because most fire accidents occur during battery charging, the presence of battery swapping cabinets can reduce the risk of accidents. Currently, battery swapping cabinets are highly safe and have automatic fire extinguishing equipment, and thus they can control any fire inside them.

6.2.1.5.2 Disadvantage

(i) It is difficult to recover the cost in a short term due to the high construction cost and low utilization rate. There is still a long way to go to build sufficient battery swapping cabinets to support the market demand; and (ii) at present, the power swap mode is applicable to only 2B scenarios, and the daily use cost is too high for ordinary users of 2C terminal; hence, it is very difficult for users to form the use habit.

6.2.2 Power Swapping Electric Vehicles

6.2.2.1 History of Power Swapping Electric Vehicles

Vehicle–battery separation not only refers to the physical separation of battery and vehicle, but it also indicates the separation of battery ownership. Physical separation is to separate the battery from the vehicle body for power swap and quickly replace the lithium-ion power battery through a specific device in a short time. Ownership separation is the separation of ownership and use right of lithium-ion power batteries so that the lithium-ion power battery becomes a mobile service product owned by a battery asset management company and used by users at their rental costs.

6.2.2.1.1 Pathfinder: Better Place

Better Place of Israel tried to commercially operate vehicle–battery separation first in 2007, but the market conditions at that time were not enough to support the operation of the power swap mode. On the one hand, new energy vehicles were in the early stage of development, with a small market size. On the other hand, the construction of battery swapping station requires huge investment. Better Place, a pioneer of a vehicle–battery separation business model, declared its bankruptcy in May 2013.

6.2.2.1.2 Successor: Tesla

Although Better Place failed commercially, it left a relatively complete chassis power swap technology and operation experience to its successor. After that, Tesla, an automobile maker of the United States, developed a 90-second power swap technology based on Better Place's chassis idea. Later, however, Tesla found that power swap was no longer a technical problem, but it was difficult to solve the problems of inconsistent battery standards, high construction cost of a battery swapping station, low operation efficiency, and a long cost recovery cycle. Eventually, Tesla gave up the power swap and shifted to fast charging.

6.2.2.1.3 Promoters

Aulton and NIO (Chinese enterprises). Although technology model brings business model, if there is not enough market demand and capital investment, the business model cannot be implemented. Obviously, China is the most suitable country to carry forward the business model of vehicle-battery separation.

As China's largest power swap service provider, Aulton New Energy has explored in taxi, bus, and ride-hailing and logistics vehicles for 20 years. On 12 May 2021, CAI Dongqing of Aulton New Energy released its development plan for 2021–2025 to invest in the construction of 10 000 battery-swapping stations with the service capacity of more than 10 million vehicles within five years. That is undoubtedly the most ambitious power swap project in China now.

In August 2020, NIO officially launched the innovative business model “Battery as a Service” and vowed to solve the problems about electric vehicles for private users. On 9 July 2021, the first NIO Power Day was held in Shanghai. NIO shared the development history and core technologies of NIO Power on site and released the *NIO Power's Battery-Swap Station Layout Plan for 2025* [32]. To provide better charging service to more and more users, NIO will accelerate the construction of charging and power swapping network. NIO will increase the total number of battery-swap stations from 500 to over 700 in 2021. From 2022 to 2025, 600 new power stations will be built in the Chinese market every year. By the end of 2025, NIO will have more than 4000 battery-swap stations worldwide, including about 1000 battery-swap stations outside China. In addition, NIO announced that it would fully open NIO Power's charging and power swap system and BaaS service to the industry and share the battery-swap stations of NIO Power with the industry and smart electric vehicle users.

6.2.2.2 Policy Makes Power Swap Popular

Power swap vehicle users enjoy subsidy. After June 2019, new energy vehicles have entered the “post-subsidy era.” The gradual decline of subsidies has increased the purchase cost of consumers and weakened the market competitiveness of new energy vehicles. The power swap mode of “vehicle–battery separation” provides a new idea for “bare vehicle sales” to reduce the costs. In April 2020, the Ministry of Finance, the Ministry of Industry and Information Technology, the Ministry of Science and Technology, and the National Development and Reform Commission jointly issued the *Notice on Improving Fiscal Subsidy Policies for the Promotion and Application of New Energy Vehicles*, which defines that new energy passenger vehicles shall have the pre-subsidy price of 300 000 yuan or below, but power-swapping vehicles have any pre-subsidy price.

The battery-swap station is part of the new infrastructure. In May 2020, new infrastructure construction in the Governmental Work Report delivered at China's National People's Congress (NPC) and Political Consultation Congress (PCC) states that the phrase “build charging stations” is changed to “add charging stations, battery swapping stations, etc.” Battery-swap stations, 5G, artificial intelligence, and big data centers have become part of the new infrastructure. This reflects that the Chinese government has fully recognized the power swap mode and the vehicle–battery separation mode. In November 2020, The General Office of the State Council officially released the *Development Plan for The New Energy Vehicle Industry (2021–2035)*, which emphasizes the strengthening of the charging and power swap infrastructure and encouragement of the application of the power swap mode. In response to the national call, Beijing, Shanghai, Hainan, Jinan, and Tianjin have

issued a series of implementation policies and systems according to their local conditions, stepped up the construction of charging and power swap infrastructure, and carried out the pilot test of power swap mode.

6.2.2.3 Battery Asset Management Company

A battery asset management company is a battery bank. Such company, which was first launched by NIO, is part of BaaS service. All big auto companies have set up their own battery asset management companies to centrally manage the lithium-ion power batteries and obtain the assets operation value in whole life cycle as shown below. The management mainly includes battery rent, purchase, operation, energy storage – vehicle-to-grid, cascade utilization, dismantling, recycling, etc.

6.2.2.4 Advantages and Disadvantages

6.2.2.4.1 Advantage

(i) Power swap can greatly improve the speed of energy replenishment, realize charging like refueling, and greatly relieve the worry about endurance mileage; (ii) the power swap mode can reduce the purchase and use costs of electric vehicles, conducive to improving the sales of electric vehicles; (iii) power swap improves safety. Because electric vehicles always spontaneously burn during charging and the power swap operators can reduce the probability of risk through unified management and standardized operation; and (iv) the power swap mode unifies the recycling channel of lithium-ion power battery because the batteries are not owned by users to avoid entry of the decommissioned power lithium-ion batteries into unqualified private workshops.

6.2.2.4.2 Disadvantage

(i) There is no unified standard for batteries; thus, different automobile enterprises design different battery structures and interfaces, which restrict the promotion of the power swap mode; (ii) the battery swap stations not only have high costs of construction and operation, but they also cannot greatly relieve the worry about endurance mileage because they are insufficient, irrationally distributed, and managed in a non-normative manner.

6.2.2.5 Recycling Route of Decommissioned Lithium-Ion Power Batteries in the Power Swap Mode

The power swap mode is conducive to promoting the cascade utilization, regeneration and recycling of batteries. On the one hand, it is difficult to recover lithium-ion power batteries from a large number of consumers under the traditional business model. The power swap mode has such an inherent advantage that it can improve the recycling rate of decommissioned lithium-ion power batteries, introduce more decommissioned batteries, and form a closed loop of charging – battery swapping – energy storage by cascade utilization. On the other hand, the differences in performance and specification between lithium-ion power batteries lead to the inconsistent performance parameters of the decommissioned cell and thus affect the value of cascade utilization. For example, the differences in voltage and internal

resistance between battery packs reduce the actual available capacity of the system, lead to unstable current and voltage, and thus greatly reduce the battery reliability and safety during long-term operation. In the power swap mode, the lithium-ion power batteries can be evenly managed, charged and discharged scientifically, and effectively evaluated on their remaining capacity and remaining service life. In addition, the lithium-ion power batteries used in vehicle–battery separation mode have the relatively unified energy density, work intensity, and work scenarios; thus, the decommissioned batteries have consistent state and capacity, conducive to the cascade utilization of power lithium-ion batteries and automatic dismantling of battery packs. This will ensure the reliability and safety of the batteries during reuse and maximize their value.

6.2.3 Power Swapping Electric Trucks

6.2.3.1 Demand for Power Swapping Heavy Trucks

The current technology of lithium-ion power battery is not feasible for electric heavy truck to achieve the ideal endurance mileage by a single charging operation of passenger vehicles in a short term. To maximize the endurance mileage of electric heavy trucks, automobile manufacturers have to increase the number of batteries installed. At present, a General Electric heavy truck has the battery weight of 2–3 tons, which greatly reduces its payload capacity. Pure electric heavy trucks generally travel in fixed routes, short distance, and high frequency scenarios, such as urban muck transport, short-distance travel in mining area, factory area, and ports. To shorten the charging time and improve the operating efficiency of vehicles, various manufacturers have developed electric heavy trucks supporting the fast charging mode or even the super-fast charging mode. In June 2021, Daimler Trucks released its first mass-produced Mercedes-Benz eActros pure electric heavy truck, which can boost the electric quantity in a battery from 20% to 80% in a quick charging mode in about an hour [33]. In spite of oversized batteries and fast charging technology, the worry about the endurance mileage of electric heavy trucks has not been thoroughly eliminated. In addition, frequent rapid charging accelerates the performance degradation of power lithium-ion batteries and increases the risk of safety accidents. Most importantly, the electric heavy trucks have two to three times higher initial purchase cost than traditional fuel heavy trucks due to the existence of batteries. Thus, battery will undoubtedly become a major obstacle to promote and apply electric heavy trucks.

6.2.3.2 Economic Advantages of Power Swapping Heavy Trucks

The power swapping heavy trucks have much lower purchase price than self-charging heavy trucks. According to the data of the State Electricity Corporation in Table 6.2, the power swapping heavy trucks will have about 10% lower use costs than traditional fuel version 6×4 tractors due to their different vehicle bodies in five years [34].

Table 6.2 Comparison of purchase and use costs of 6 × 4 tractors and power swapping heavy truck in five years.

Project	Tanker	Power swapping heavy trucks	Notes
Purchase price (10K CNY)	36.00	40.00	—
Purchase tax (10K CNY)	3.19	0.00	Electric vehicles can be exempted from purchase tax
Engine maintenance for 5 yr (10K CNY)	2.40	0.00	According to the conventional engine maintenance of each bicycle every 20 000 km, 1200 yuan per maintenance, driving 80 000 km/yr, namely 4800 yuan/yr
Urea fees for 5 yr	2.50	0.00	According to the consumption of 10 kg urea per 800 km per cycle, the cost is 50 yuan, the annual driving 80 000 km, that is, 5000 yuan/yr
Total (10K CNY)	44.09	40.00	—
Cost savings per vehicle for 5 yr (10K CNY)	4.09		—
50 vehicles cost savings for 5 yr (10K CNY)	204.50		—
Proportion of savings	9.28%		—

Source: State Power Investment Group [34].

Based on the analysis of two typical use scenarios, electric heavy trucks have about 15% lower costs than fuel heavy trucks. In the whole life cycle of purchase and operation, the electric heavy trucks have 12–14% lower costs than traditional fuel vehicles. In general, the mode of “vehicle–battery separation” has significant economic benefits in the whole life cycle (Table 6.3).

6.2.3.3 Application Scenarios for Power Swapping Heavy Trucks

Because of the limited range and insufficient battery-swapping station, electric heavy trucks apply to only the transport at overload and low speed in a short distance, such as transport of coal ore, city clinker, domestic garbage, containers, and bulk cargo and internal transport in steel mills and mines. With increasing endurance mileage of batteries and construction of charging and battery swapping stations, the application scenarios of power swapping electric heavy trucks will be further expanded in the future.

6.2.4 Power Swapping Electric Ships

6.2.4.1 Exploration of Power Swapping Electric Ships

Due to their own characteristics, electric ships often carry a large number of lithium-ion power batteries. How to efficiently and safely charge the batteries has become a barrier for the development of electric ships. Due to the rapid development of the power swap mode in the field of two-wheeled vehicles and automobiles, the power swapping ships have become a concern.

In fact, as early as 2016, a Chinese company designed a patented ship [35] where the lithium-ion power battery pack can be replaced in the dynamic or static mode to solve the problem of power swap of electric ships in operation. This ship consists of a mother vessel and a lithium-ion power battery vessel. As an independent power system, the lithium-ion power battery leaves the mother vessel to the battery-swapping station on water for charging, while a new lithium-ion power battery combines with the mother vessel by remote control to complete the power swap. Although the technology has not been widely used, it provides an idea for the power swap mode of ships.

On 26 June 2021, China's first 64TEU pure lithium-ion power battery container ship Guochuang was officially launched [36]. It is reported that the ship is the special demonstration project of "Integrated Transport and Intelligent Transport" under the *National Key Research and Development Plan of 2018*. One of the key technologies is the battery swap by an integrated module mobile power supply system to support the battery swap at fixed docks. This technology is a breakthrough in the application of lithium-ion power battery in water transport equipment.

On 10 July 2021, China Shipbuilding of SDARI released a modular ship power system standardized product – S-CUBE, container Lithium-ion power battery Unit (S-CUBE, SDARI Containerized Utility Battery Module) at the Intelligent Ship Innovation Forum "AI Power Ocean" of the world Artificial Intelligence Conference 2021 [37]. S-CUBE uses a LFP battery approved by the China Classification Society (CCS) as well as 20-foot standard container as a carrier to form a universal modular power supply system. S-CUBE has the maximum capacity of 1540 kWh and needs only four containers to allow 3000 t cargo ship to sail 200 km. S-CUBE has automatic standard interfaces that can connect and disconnect with the power grid quickly and safely to replace the battery module by lifting and separate the ship from batteries. Standardized design concept and safe and reliable system design are the basis of ship–battery separation mode. The emergence of integrated and standardized power modules represented by S-CUBE will promote the construction of power swap system in the field of ships and improve the operation efficiency of ships.

6.2.4.2 LFP, The Mainstream of Shipboard Lithium-Ion Power Battery

There are stringent requirements of marine power for safety, service life, reliability, etc. LFP battery is the mainstream of marine lithium-ion power battery because it is

Table 6.3 Typical case studies.

<p>Case 1: A sand and gravel transportation project (regional short dumping) – 60 phase</p> <ul style="list-style-type: none"> • Per 50 Vehicles + 57 sets of 282kWh batteries, electricity price 0.4yuan/kWh; • The vehicle runs for 300d. The average daily minimum driving mileage per vehicle is 304km; • Including S06-2400KW battery bin fourth-generation standard changing station (the whole station is considered as an investment of 4 million yuan) 				<p>Case 2: A large steel mill (site overload steel short fall) – stage 60</p> <ul style="list-style-type: none"> • Per 50 Vehicles + 57 sets of 282kWh batteries, electricity price 0.4yuan/kWh; • The vehicle runs for 300d. The average daily minimum driving mileage per vehicle is 121.6km; • Including S06-2400KW battery bin fourth-generation standard changing station (the whole station is considered as an investment of 4 million yuan) 			
Comparison of fuel and electricity economy (CNY)				Comparison of fuel and electricity economy (CNY)			
Power swapping heavy trucks		Fuel heavy trucks		Power swapping heavy trucks		Fuel heavy trucks	
Basic electricity price (yuan/kWh)	0.40	Oil price (yuan/l)	6.00	Basic electricity price (yuan/kWh)	0.40	Oil price (yuan/l)	5.50
Equivalent to battery namely changing station rent (yuan/kWh)	0.75	Fuel consumption per km (l/km)	0.40	Equivalent to battery namely changing station rent (yuan/kWh)	0.75	Fuel consumption per km (l/km)	1.20
Battery discharge per km (kWh/km)	2.00	—	—	Battery discharge per km (kWh/km)	5.00	—	—
Integrated power consumption per km (kWh/km)	1.40	—	—	Integrated power consumption per km (kWh/km)	4.50	—	—
Energy expenditure measurement				Energy expenditure measurement			
Energy consumption expenditure per km (yuan/km)	2.06	Energy consumption expenditure per km (yuan/km)	2.4	Energy consumption expenditure per km (yuan/km)	5.55	Energy consumption expenditure per km (yuan/km)	6.60
Battery and changing station rental per km (yuan/km)	1.50			Battery and changing station rental per km (yuan/km)	3.75		
Basic electricity charge per km (yuan/km)	0.56			Basic electricity charge per km (yuan/km)	1.80		
Energy expenditure savings for 50 vehicles–60 phases (10k)		775.20		Energy expenditure savings for 50 vehicles–60 phases (10k)		957.60	
Fuel and electricity expenditure saving rate		14.17%		Fuel and electricity expenditure saving rate		15.91%	

Source: State Power Investment Group [34].

better than NCM battery in these aspects. In China, marine lithium batteries must be certified by the CSS. Currently, the CSS recognizes only square LFP batteries, which provide favorable conditions for the development of LFP in the shipbuilding field. At present, CATL, Yiwei Lithium Energy, Gotion, Penghui Energy, and Star Ying Technology have been certified by CSS.

According to GGII [38], the estimated market scale of lithium battery for electric ships will reach 35.41 GWh by 2025 based on the penetration rates of 0.035% in 2019, 0.55% in 2022, and 18.5% in 2025.

6.2.4.3 Suggestions

Under the new development opportunity, it is necessary to actively explore and promote the application of the power swap mode in the field of ships. The shipbuilding industry and lithium battery industry shall pay attention to the following development directions: (i) accurately grasp the demand characteristics of electric ships and promote the standardization of hull structure, interface, and safety of the electric ships; (ii) promote the standardization of lithium-ion power battery module structure, interface, safety, and performance and standardize the use of lithium-ion power batteries on ships; (iii) innovate in the operation mode of charging and power swap facilities in the ports, increase the investment, and strengthen the construction of battery swap infrastructure, especially combine offshore wind power generators, which not only serve as large-scale energy storage devices but also supply power to ships. In addition, national unified planning and guidance are needed to promote and guide the healthy development of electric ships by establishing the power swap mode demonstration routes.

6.3 Sub-summary

The rapid development of power battery technology has given rise to a variety of different application scenarios. However, the application scenarios mainly focus on the field of electric transport tools, including two-wheeled electric vehicles, electric vehicles, and electric ships. Power batteries have key technical bottlenecks in safety, endurance mileage, service life, etc. To solve the problems, the market demand has driven different business models. The vehicle–battery separation business model is beneficial to the recycling of decommissioned power batteries and solves the problems. In the future, power batteries will face bigger, more detailed and scattered application scenarios than today, inevitably leading to more diversified technical routes and business models. It will be an inevitable trend to standardize the batteries and centralize and standardize the recycling channels.

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7

Battery Recycling Technology Outlook

7.1 Advanced Battery Recycling System

The recycling of spent power lithium-ion batteries (LIBs) is a complex and constrained industry. Due to the lack of standardization in the production of power LIBs, an insufficient recycling system, and a lack of regulation over the recycling process, it is difficult to dismantle spent power LIBs, recycling technology is challenging to unify, and there is still a shortage of technology for large-scale battery production that is safe and environmentally friendly. In the face of the current small, chaotic, and miscellaneous recycling market and the expected generation of large quantities of end-of-life batteries, battery recycling technology urgently needs to be improved.

7.1.1 Economical Environmental Discharge Technology

Currently, most battery recycling companies use hydrochloric acid discharge method. Although this discharge technology has the economic advantage of low cost, it has drawbacks such as electrolyte leakage and costly wastewater treatment. In fact, considering the equipment investment and operation cost of the wastewater treatment section, the hydrochloric acid discharge technology does not offer high economic advantages. Battery discharge technologies capable of large fluxes remain a work in progress. Safe and environmentally friendly battery discharging technologies enable rapid discharge of individual cells to a safe level, which facilitates subsequent battery transportation, disassembly, and shredding.

7.1.2 High-Flux Battery Disassembly Equipment

The main purpose of disassembly is to physically separate the valuable components in the battery, such as electrode powders and copper and aluminum foils, and to dispose of the hazardous components in the battery, such as electrolytes and polyvinylidene fluoride (PVDF) binders. Studies on the conversion mechanism of battery components, physical dissociation and separation characteristics of each component in the battery disassembly process are extremely important. Scientific process flow

design, customized shredding and sorting equipment manufacturing, and efficient pollutant treatment solution planning are necessary for the development of high-flux, high-security, and environmentally friendly battery dismantle sets.

7.1.3 High-Efficiency Separation System for Valuable Metals

For ternary batteries containing specific valuable metals (nickel, cobalt, manganese, lithium), an unidentified composition ratio (LCO, LMO, NCA, NCM111, NCM523, NCM622, NCM811), and an unspecified impurity composition (calcium, iron, aluminum, magnesium, silicon, fluorine, organic matter, PVDF, etc.), developing adaptable hydrometallurgical processes to efficiently recover valuable metals from spent batteries continues to be a challenge. Based on the existing nickel–cobalt–lithium separation and purification process and equipment, the future recycling process should be an efficient separation process of valuable metals–impurities based on the composition of battery materials, especially the composition of impurity components. Through process optimization and characteristic separation process design, a highly adaptive and efficient metal separation process can be developed.

7.1.4 High-Value Utilization of Low-Value Batteries/Components

For low-value power LIBs such as LFP, LMO, and LTO, short process and high value-added process technologies need to be developed. On the basis of the current process focusing on lithium recovery, the development of comprehensive recovery technologies for iron, manganese, titanium, and other metals is worth further study. For the large amount of graphite materials produced in the recycling process, the development of an integrated process that collects pretreatment, impurity removal and reforming will facilitate the resource utilization of graphite.

7.1.5 Intrinsic Safety and Pollution Prevention

Spent lithium-ion power battery is a kind of hazardous waste. The existing discharge technology still cannot achieve complete discharge of the battery. During the pretreatment process of spent batteries, a large amount of graphite and metal dust will be released. Besides, the electrolyte contains a large and complex composition of organic substances as well as LiPF_6 and other hazardous substances. Therefore, spent lithium batteries need to be processed automatically and efficiently in an absolutely safe environment. Additionally, hydrometallurgy produces a lot of complex organics, promoting extra consideration of wastewater treatment and recycling.

7.2 Green Battery Design for Recycling

Establishing and enhancing carbon footprint traceability management throughout the life cycle of a lithium-ion power battery can assist in monitoring and tracking

carbon emissions across the entire process, from material production to assembly and manufacturing, battery use, and recycling. Through data analysis, high carbon emission links can be identified, and subsequently, the process can be optimized. Green battery design is critical for reducing carbon emissions of the whole life cycle of power LIB. The environmental characteristics of the battery (disassembly, recyclability, maintainability, reusability, etc.) are prioritized and taken as design objectives. While ensuring the basic performance, service life, and quality of the battery, the resource and energy utilization of the battery should be enhanced as much as possible. Reducing potential environmental pollution and pollution disposal costs can reduce the negative environmental impact of the battery during its entire life cycle.

The ecological challenges facing state-of-the-art lithium battery materials and components mainly include high energy consumption for material production (collector, graphite, and metal oxide), fluorinated and toxic compounds (electrolyte, cathode binder, and metal oxide), expensive disposal costs (organic solvents used for cathodes), and low recycling rate (Figure 7.1). Based on the mentioned issues, the ideal green battery design solution should consider lithium battery raw material selection, material production, battery manufacture, and recycling as well as the environmental benefits and economic constraints associated with green materials/processes.

7.2.1 Battery Raw Materials

Graphite is the most commonly used anode material in lithium batteries, which mainly consists of natural graphite and synthetic graphite. To improve the sustainability of synthetic graphite materials, the use of biomass or industrial wastes as precursors for the pyrolysis of carbon-based anode materials is gradually attracting attention. Wu et al. [2] conducted a comparative analysis of the environmental

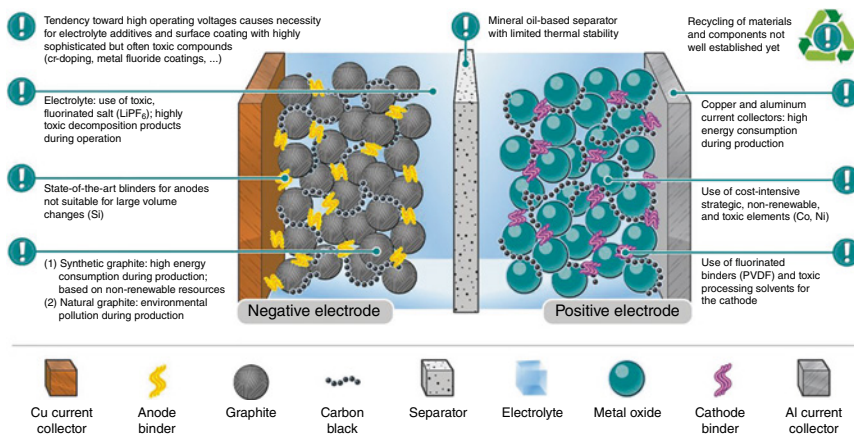


Figure 7.1 The main ecological challenges facing by LIB materials and its element.
Source: Dühnen et al. [1]/John Wiley & Sons/CC BY 4.0.

impact of three anode materials, graphite, silicon nanowires, and lithium metal, and found that a high specific capacity anode material would make the battery have a lower environmental impact. The results indicate that although the high specific capacity silicon anode reduces the negative environmental impact of the battery, its preparation process will result in extremely high carbon emissions. Therefore, the development of silicon cathodes based on biomass (e.g. rice husk) or decommissioned silicon wafers for the preparation of silicon anodes is also an important direction for future research [3]. In addition, lithium-metal cathodes have the least environmental impact compared to existing graphite and silicon-carbon cathodes and are highly promising next-generation environmentally friendly battery materials.

The carbonate organic electrolytes commonly used currently are flammable and volatile and have limited thermal stability, which makes them risky in use and low recyclability. Safety is an important aspect of electrolyte optimization design. During the charging and discharging process, factors such as resistance overheating, overcharging, and short circuit caused by lithium dendrites are easily to cause the internal temperature of the battery to rise sharply, resulting in thermal runaway. The electrolyte decomposes and releases gas at high temperatures, causing damage to the battery and even leading to fire or explosion. While adding chemicals and flame retardants to the electrolyte can enhance the operational safety of the battery, it also complicates the electrolyte composition, reducing recovery efficiency. Ionic liquid is a “green solvent.” Compared with organic solvents, ionic liquids are easier to recover, and are expected to improve the recovery rate of electrolytes in retired lithium batteries. Therefore, the exploration and development of low-cost ionic liquids suitable for lithium batteries is also a frontier research direction.

PVDF is widely used as a binder for lithium battery cathode materials because of its good electrochemical stability and binding properties. However, the drawback is that PVDF can only be dissolved within a few organic solvents. Currently, the commercial solvent used is NMP, which is expensive and toxic, resulting in higher production and recycling costs of the battery. Several water-soluble binders, including carboxymethyl cellulose (CMC) and styrene-butadiene rubber (SBR), have been investigated for use in the preparation process of anode materials, with the goal of reducing production costs and enhancing manufacturing and recycling safety. However, the use of water-soluble binders in cathode materials is very limited due to the water sensitivity of most cathode materials and the instability of water-soluble binders at high voltage. From a recycling point of view, there is a binary opposition between the stability of the binder during use and its solubility at the end of life. The selection of the binder not only affects the performance of the battery, but it is also directly related to the ease of separation and recycling of battery materials. As a result, the development of self-supporting electrodes without binders is gradually becoming a significant future trend.

At present, the ternary materials (NCM and NCA) have better overall performance than other materials, and they constitute the main commercial lithium battery cathode materials. Metals such as Li, Co, and Ni, the critical raw materials for lithium batteries, are closely related to the cathode material, accounting for one-third of the cost of lithium batteries. Notably, both Co and Ni are classified as

carcinogenic, reproductive toxicants. Moreover, Co is the dominant factor in battery production costs. In some parts of Central Africa, Co mining raises ethical and environmental concerns, such as substandard mining conditions and the use of child labor. Therefore, establishing a “sustainability” label (carbon footprint, ethical footprint, etc.) for the traceability of battery raw materials is currently a feasible approach. LFP and LMO materials are the most sustainable of the technically mature cathode materials. Although their energy density falls short of market demand, LFP and LMO batteries still have some room for growth in the future market from a life-cycle perspective. In addition, low cobalt and cobalt-free cathode materials such as LNMO and LMR-NCM are gradually attracting attention as potential future cathode materials [4].

“Post-lithium” battery technology based on inexpensive metal (sodium, potassium, calcium, magnesium, aluminum, zinc, etc.) or organic active materials has shown great potential to reduce the shortage of lithium resources. Innovations in battery technology will not only mitigate material supply issues but also have a significant impact on battery structure design. For example, a novel dual-ion battery [5] uses the same cathode and anode current collector and active material. This symmetrical cell structure will greatly simplify the cell manufacturing process and thus reduce the cell production and recycling costs.

7.2.2 Material Production

A large part of the cost of battery materials is determined by the material synthesis process. Currently, most cathode materials are prepared by high temperature solid-phase synthesis, but this method has high energy consumption and cost. Therefore, low temperature synthesis methods such as water (solvent) thermal synthesis, ultrasonic and microwave assisted synthesis, and ion thermal synthesis have been developed. Recent studies have shown that ion thermal methods can effectively modulate the morphology of electrode materials. For example, at a reaction temperature of nearly 200 °C, the phosphates and silicates obtained by ion thermal method exhibit specific structures with excellent morphology. Compared with high-temperature solid-phase synthesis, the reaction temperature of ion thermal method is reduced by about 500 °C [6]. In addition, some new lithium-based battery materials have been synthesized by ion thermal reactions, such as LiFeSO_4F , a novel material that can be synthesized by solid-phase reaction at 300 °C for only 20 minutes [7]. While the ion thermal method appears uneconomical due to the usage of an ionic liquid as the reaction medium, the approach is also environmentally friendly due to the simplicity with which ionic liquid may be recovered. Furthermore, emulating biological processes, researchers have investigated techniques to synthesize battery materials at near-room temperature. For instance, Belcher’s research team [8] used genetically engineered viruses as templates to rapidly prepare FePO_4 single-walled carbon nanotubes at room temperature. However, this method cannot be used for the synthesis of lithium-based battery materials at present. This method provides a new direction for the synthesis of low-cost lithium materials due to its high feasibility of scale up. In conclusion, the use of low-energy synthesis methods and avoiding the use of toxic solvents are effective ways to optimize the carbon footprint of the material production process [9, 10].

7.2.3 Battery Production

7.2.3.1 Single Battery

In terms of the preparation process of the battery cell, the optimized design is mainly focused on the improvement of the energy density and power density of the cell [11]. Based on the porous electrode model, Ramadesigan et al. [12] reduced the voltage loss of the electrode by differentiating the electrode void distribution. By adjusting the electrode porosity, it is plausible to reduce the ohmic resistance by 15–33% for a certain number of active materials, hence increasing the ability of electrodes to store and transmit energy. Golmon et al. [13] proposed a computational model of multi-scale lithium batteries with optimized electrode layout for enhancing the capacity of lithium batteries. By controlling the local porosity and particle radius of the electrode particles, the capacity of the lithium battery is maximized while limiting the electrode particle stress level. The optimization results indicated that the optimal functional gradient electrode could improve the performance of the battery compared to the situation of uniform porosity and particle radius distribution. Under the premise of satisfying specific power density requirements, Xue et al. [14] proposed a numerical framework for lithium battery design to achieve maximum battery energy density. Considering the irreversible capacity loss difference between cathode and anode, Bennett [15] proposed a single cell capacity matching method based on the experimental data of cathode and anode and predicted the discharge voltage of the resulting single cell after cathode and anode matching. Further, through the selection of different process methods, optimization of the process and control of toxic and hazardous substances, the environmental performance of the battery in the manufacturing process can be improved.

Recent studies have shown that efficient separation of electrode active materials can be realized by structuring the polymeric separators between the electrodes. Li et al. [16] designed the separator as a type of Z-fold with alternating windings between the cathode and anode. The cathode and negative anode pieces of the cell can be perfectly separated by a set of vacuum conveying devices equipped with a clamp and a series of skimmers (Figure 7.2).

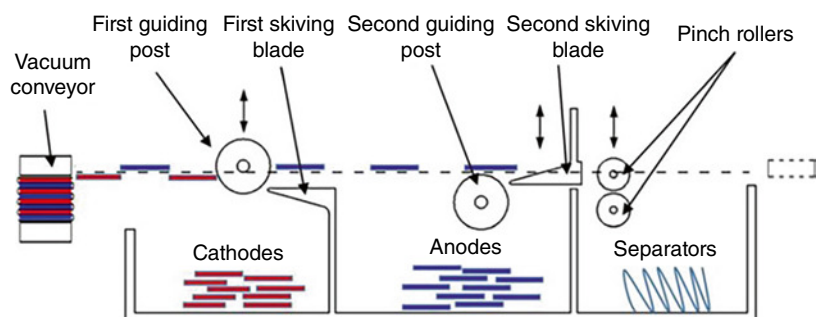


Figure 7.2 Device for separating cathode, anode, and separator from lithium battery diagram. Source: Li et al. [16]/With permission of Springer Nature.

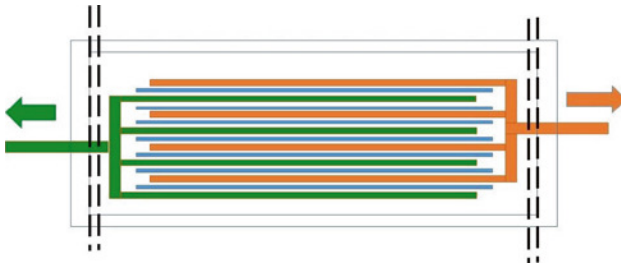


Figure 7.3 Schematic diagram of removing a battery (dotted line indicates the cutting point). *Source:* Thompson et al. [9]/Royal Society of Chemistry.

7.2.3.2 Battery Pack

At the structural level of the battery pack, the disassembly performance of the battery pack during in-service maintenance and end-of-life recycling can be improved by optimizing the geometry (topology, dimensions, layout, etc.) and using disassembly/assembly-oriented design methods and device parts [17]. Figure 7.3 shows a novel simplified battery pack structure [9]. By modifying the geometry of the electrode connector tabs, electrodes of the same polarity are connected together. In this way, when retired batteries are cut, the cathode and anode plates can be simply separated, and the polymer separator can be easily recycled. Chinese battery manufacturer BYD (BYD) has recently developed a long and thin blade battery [18]. This unique structure eliminates the need for individual modules and binders and gives the battery pack greater structural strength. The advent of this modular-free CTP (Cell to Pack) packaging technology makes battery disassembly easier and facilitates robotic automated assembly and disassembly line design.

The establishment of standardization of battery production is also an important step to enhance its recycling efficiency. In terms of the structure of battery cells, the structures commonly used in the market are cylindrical, square-shell, and soft pack. The unification of specifications is of great significance for efficient battery recycling. Moreover, the modular connection of battery packs and the standardization of package design also facilitate the use of uniform tools for battery disassembly [19]. In addition, the improvement of battery packaging labels is essential, including the display of information on the composition of cathode and anode materials, electrolyte, etc. Separating different types of batteries being disassembled before disassembly can avoid the risk of cross-contamination of different materials.

7.2.3.3 Battery Management System

At the battery system level, the main focus is on the optimization of the battery management system and the cooling system. Battery life depends on a wide range of factors related to battery chemistry, charge and discharge methods (maximum charge voltage, charge current, charge multiplier, etc.), operating temperature, and number of cycles [20]. The battery management system must include monitoring of battery charge and discharge parameters to provide information on battery capacity decay and should also have a fault diagnosis function to provide timely warnings of abnormal

battery performance. Based on experimental and actual test data, Dubarry et al. [21] developed a battery performance analysis model to achieve an accurate prediction of battery lifetime. The results showed that the cooling system of the battery can mitigate thermal runaway caused by extreme thermal conditions and battery performance degradation, although it usually increases the energy consumption of electric vehicles. Jarrett and Kim [22] studied the optimized design of the battery cooling plate. The optimized cooling plate can meet the requirements of refrigerant pressure drop, average cell temperature, and temperature uniformity of lithium-ion power battery pack.

7.2.4 Battery Passport

To implement better traceability, monitoring, and management of batteries, new EU draft battery regulation lays down labeling requirements to allow batteries to be placed on the market and put into service as well as for the collection, treatment, and recycling of waste batteries. It requires that all light transport batteries including electric vehicle batteries and rechargeable industrial batteries to be separately equipped with electronic documents, that is battery passport, in the form of QR code printed on the battery. Battery passport can be scanned by terminal devices according to device permissions, so as to obtain different level of contents, such as basic information like battery model and characteristics, as well as sensitive information like battery disposal methods and battery chemical composition.

Global Battery Alliance (GBA) proposes to establish the battery passport operating platform to carry out traceability management of battery life cycle in a visual way (Figure 7.4). The Oak Ridge National Laboratory recommends a battery passport for more efficient management during the recycling phase (Figure 7.5).

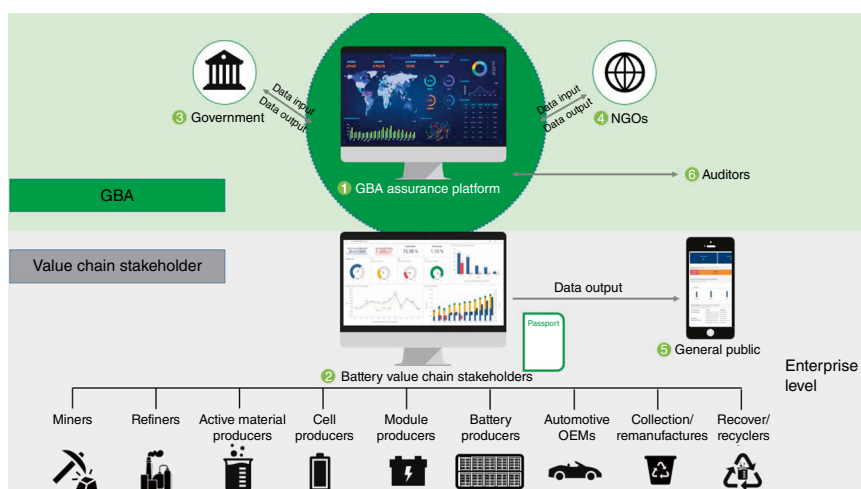


Figure 7.4 GBA platform vision for battery passport [23]. *Source:* jannoon028/Adobe Stock, burntime555/Adobe Stock.

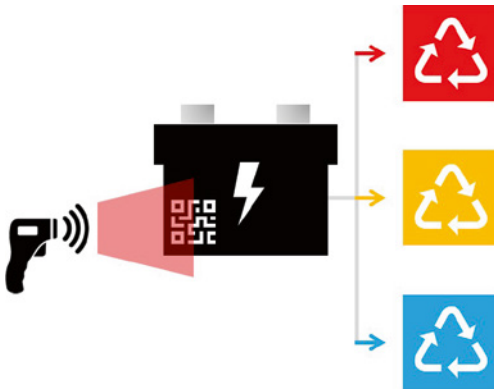


Figure 7.5 Oak Ridge Laboratory battery passport QR code for efficient processing in the recycling stage [24]. *Source:* Courtesy of Oak Ridge National Laboratory, U.S. Department of Energy.

The feasibility of battery cascade utilization is still controversial due to immature industry standards, lack of effective battery detection and sorting technology, and safety risks caused by insufficient market supervision. However, with the establishment and promotion of battery passport, any data in the whole life cycle of the battery can be traced back to the source, which provides a strong foundation for the standardized and large-scale market operation of the cascade utilization industry. In the future, cascade utilization technology will also become an important part of the recovery of spent power LIBs, and together with disassembly recycling technology, it will jointly contribute to the green, low-carbon, and sustainable development of the new energy vehicle industry chain.

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