

# INTRODUCTION

This introduction:

- Lists the properties of materials which are influenced by fillers
- Lists typical properties of fillers
- Provides definition of the term “filler”
- Defines how fillers function in various applications
- Suggests how fillers may be classified
- Discusses the markets for fillers and the emerging trends in filler use

The introduction will define the scope of the book and provide a brief overview of each chapter.

It is our intention to show how an understanding of the diverse functions of fillers in materials can lead to a well designed material formulation.

## 1.1 EXPECTATIONS FROM FILLERS

What caused fillers to be added to materials in the first place was probably the quest for lower costs. Fillers were inexpensive, thus using them would make the material cheaper. We do not know who the inventor of the idea was but it was probably not one, but many people in many different places. However, as the following discussion shows, cost reduction is no longer the only, or even the most important, consideration for using fillers in formulating composite materials.

These examples which follow list attributes of materials to the formulator's various objectives.

*Cost reduction*<sup>1</sup>

Cost reduction depends on the relative cost of the polymer and the filler. Polymer prices in 1996-7 were approximately:

	US\$/kg	US\$/l
ABS	1.98	2.05
PE	0.77	0.70
PET	1.65	2.67
PP	0.88	0.79
PS	0.79	0.84
PVC	0.66	0.92

Filler prices depend greatly on the particle size. In the list below, fillers are divided into *large* particle size materials (up to 100 μm; e.g., ground CaCO<sub>3</sub>), *medium* particle size (around 10 μm; e.g., clay), *small* particle size (around 1 μm; e.g., TiO<sub>2</sub> or precipitated CaCO<sub>3</sub>), and *very small* particle size (below 0.1 μm; e.g., fumed silica). These are approximate prices:

	US\$/kg	US\$/l
Large	0.05	0.14
Medium	0.31	0.81
Small	1.00	2.80
Very small	6.60	14.50

If we consider only cost, it is the cost per unit volume that must be compared. The table shows that only the use of large particle size fillers (very crude products) can potentially contribute to savings in the manufactured cost of materials made from commodity polymers. At the same time, fillers decrease many mechanical properties of the material so cost reduction is achieved at the expense of performance. Medium particle sized fillers are less attractive economically because costs of processing, inventory and transportation will increase and must be added to the total. This shows that there must be other motives to compound polymers with fillers. These follow.

*Material density*<sup>2</sup>

Fillers can be used either to increase or to decrease the density of a product. Because the density of a filler can be as high as 10 g/cm<sup>3</sup> or as low as 0.03 g/cm<sup>3</sup>, there may be a large difference between the density of the filler and the polymer. Thus a broad range of product densities can be obtained. There are high density products (above 3 g/cm<sup>3</sup>) such as materials used in appliances or casings for electronic devices. More common are densities below 2 g/cm<sup>3</sup>, glass fiber filled composites being a typical example. The effective density of the polymer can be decreased by filling a foam with hollow polymer spheres. In this

example, the density of a material can be lower than  $0.1 \text{ g/cm}^3$ .

### *Optical properties*<sup>3-6</sup>

Optical properties of compounded materials depend on the physical characteristics of the filler and the other major ingredients including the polymer. Most important is the relative refractive index of the two ingredients. Depending on how they match, one can obtain clear or opaque materials. Light absorption by the non-polymer ingredients is essential in preventing UV degradation. Some fillers (e.g.,  $\text{TiO}_2$ ,  $\text{ZnO}$  or talc) effectively absorb light. Aluminum trihydroxide in UV curable polyurethanes is noteworthy in that it accelerates the curing process because it is transparent to UV light. Calcinated clay as a filler in greenhouse film at a 10% level drastically reduces infrared absorption during the day and heat loss during the night. This application of physical principles has been an important factor in increasing the productivity of greenhouses.

### *Color*

Fillers frequently cause problems in color matching and must be accounted for in product color design. Many fillers have a distinctive color which is useful in material coloring. Recently metal powders have been used in combination with pigments to make the composite appear metal like.

### *Surface properties*<sup>7-10</sup>

For hundreds of years sticky surfaces have been dusted with powder (e.g., talc) to keep them separated. Talc is broadly used in cable and profile extrusion to obtain a smooth surface. Similarly, in injection molding, the application of aluminum trihydroxide gives a better surface finish. Talc,  $\text{CaCO}_3$ , and diatomite provide anti-blocking properties. Graphite and other fillers decrease the coefficient of friction of materials. PTFE, graphite and  $\text{MoS}_2$  allow the production of self-lubricating parts. Here, PTFE, a polymer in powder form, acts as a filler in other polymers. Matte surfaced paint is obtained by the addition of silica fillers.

*Product shape*<sup>6,11,12</sup>

Fillers reduce shrinkage of polymer foams. Mica and glass fiber reduce warpage and increase the heat distortion temperature. Intumescent fillers increase in volume rapidly as they degrade thermally expanding the material and blocking fire spread.

*Thermal properties*<sup>13</sup>

Fillers may decrease thermal conductivity. The best insulation properties of composites are obtained with hollow spherical particles as a filler. Conversely, metal powders and other thermally conductive materials substantially increase the dissipation of thermal energy.

*Electrical properties*<sup>14</sup>

Volume resistivity, static dissipation and other electrical properties can be influenced by the choice of filler. Conductive fillers in powder or fiber form, metal coated plastics and metal coated ceramics will increase the conductivity. Many fillers increase the electric resistivity. These are used in electric cable insulations. Ionic conductivity can be modified by silica fillers.

*Magnetic properties*<sup>15</sup>

Ferrites induce ferromagnetic properties and are used to make plastic magnets.

*Permeability*<sup>7</sup>

Gas and liquid permeability are influenced by the choice of filler. The platelet structure of mica or talc as a filler in paints and plastics decreases the transmission of gases and liquids.

*Mechanical properties*<sup>1,16,17</sup>

All mechanical properties are affected by fillers. Filler combinations may be selected to optimize a variety of mechanical properties. Fillers reinforce and provide abrasion resistance.

*Chemical reactivity*<sup>11,18</sup>

Many fillers can be used to influence chemical reactions occurring in their presence. The reaction rate can be decreased or increased. Fillers such as ZnO will react with UV degradation products in PE to limit damage. The pot-life of curing mixtures can be increased. Cure rates can be slowed, exothermic effects can be controlled, incompatible polymers can be blended and molecular mobility reduced.

*Rheology*<sup>7,10</sup>

The rheology of many industrial products depends on the filler addition. Examples include sealants, tooth pastes, cosmetics, hotmelts,

papers, paints, etc. Normally, additions of fillers increase the viscosity and contribute to non-Newtonian flow characteristics, but there are also combinations such as filler mixtures and specially designed glass beads which either reduce the viscosity or do not affect it.

### *Morphology*<sup>11,19</sup>

Polymer crystallization and structure are affected by fillers. They may increase or decrease the nucleation rate (and thus the crystallization rate). An increase in the nucleation rate is observed in PET in the presence of mica. Fillers, especially fibers, may also decrease the mechanical properties of filled materials because of their effect on transcrystallinity. The polymer structure at the interface with fillers is different than in the bulk.

### *Material durability*<sup>3,18,6,12,20-22</sup>

Fillers which screen radiation and react with degrading molecules contribute to material durability. The opposite effects may also occur where fillers participate in photochemical reactions which decrease photostability. Some fillers are used for their absorption of highly penetrating radiation such as nuclear radiation or filler use in neutron shielding. Thermal degradation can be either decreased and increased by the presence of fillers. Fillers such as borates and montmorillonite also protect materials from biodegradation. The addition of starch generates numerous mechanisms which increase biodegradability by supplying nutrients and also participate to initiate thermal and UV degradation which reduces chain length and allows biological conversions.

### *Environmental impact*<sup>23-26</sup>

Fillers contribute to fire retardancy by suppressing fire, increasing autoignition temperature, decreasing smoke formation, increasing char formation, reducing heat transmission rate, preventing dripping, etc. Fillers are used in combinations to balance properties. For example, antimony trioxide increases smoke whereas  $\text{Al}(\text{OH})_3$  and  $\text{Mg}(\text{OH})_2$  reduce it. In combination, this allows a balance of properties. It is possible to make paper fire retardant through

the proper selection of fillers. Plastics recycling can be improved by incorporating fillers which reduce thermodegradation (stabilize some polymers) complex mixtures of polymer waste are more easily blended if compounded with fillers.

*Performance of other additives* Fillers are instrumental in improving the performance of other additives. Antistatics, blowing agents, catalysts, compatibilizers, coupling agents, organic flame retardants, impact modifiers, rheology modifiers, thermal and UV stabilizers are all influenced by a filler's presence.

*Health & safety* Fillers are probably the least hazardous component among additives. The major exception here is asbestos which is seldom, if ever, used now. Other fillers which may be hazardous are being carefully investigated although disputes still occur when data is incomplete or questionable.

Fillers produced today are manufactured by sophisticated processes. There are numerous examples of surface modification which changes a filler's properties. There are fewer methods of filler synthesis. Preparation of materials for specific medical applications can be carried out using template polymerization.<sup>27</sup> This has become a well established discipline which has contributed to the understanding of polymer structure. Here, the polymer is produced on organic and inorganic (e.g., fillers) templates. By choosing the template structure, polymer properties can be tailored to requirements. Natural biological materials are formed in this manner and synthetic materials can be formed in a similar manner. Filler properties can also be tailored by synthesizing fillers in the presence of other materials. This is used in medical applications where the filler becomes compatible with its surroundings as it forms in body fluids. Artificial bone materials can thus be formed with surface characteristics acceptable to (compatible with) the body's environment. These techniques are at the most advanced levels of engineering and design in filler synthesis.

In summary,

- Fillers usually do not reduce the cost of material manufacturing
- Fillers are not inert materials added to fill space (if they are used in this way, they likely degrade properties of the material)
- Fillers can be modified and tailored to any application
- Fillers modify practically all properties of the material and influence the design, manufacture, and use

- Plastics performance and the performance of other materials are highly influenced by fillers
- The plastics applications base has expanded greatly as the use of fillers has increased

## 1.2 TYPICAL FILLER PROPERTIES

We have outlined the product performance characteristics of fillers. This leads us to an identification of filler properties which allow different fillers to be compared and evaluated. When we go on to develop a definition of fillers in Section 1.3, this list will help to make the definition inclusive yet precise. It will also assist in the classification of fillers discussed in Section 1.4.

<i>Physical state</i>	All materials discussed are solids but they might be available in a pre-dispersed state
<i>Chemical composition</i>	May be inorganic or organic and of an established chemical composition. May also be a single element, natural products, mixtures of different materials in unknown proportions (waste and recycled materials), or materials of a proprietary composition
<i>Particle shape</i> <sup>28</sup>	Spherical, cubical, irregular, block, plate, flake, fiber, mixtures of different shapes
<i>Particle size</i>	Range from a few nanometers to tens of millimeters (nanocomposites to pavements or textured coatings)
<i>Aspect ratio</i> <sup>28</sup>	1 (spherical or cubical) to 1,600 (fibers)
<i>Particle size distribution</i>	Monodisperse, designed mixture of sizes, Gaussian distribution, irregular distribution
<i>Particle surface area</i> <sup>29</sup>	From 10 to over 400 m <sup>2</sup> /g. Depending on the specific surface area particles have different levels of porosity from completely non-porous and smooth to very porous with a range of pore sizes
<i>Particle internal structure</i>	Hollow to porous to void free solid
<i>Particle-particle association</i>	Singular, agglomerates, aggregates, flocculated materials
<i>Density</i>	From 0.03 g/cm <sup>3</sup> (expanded polymer beads) to 18.88 g/cm <sup>3</sup> (gold)
<i>Refractive index</i>	Typical range from 1 (air) to 3.2 (iron oxide)
<i>Color</i>	Full range of colors from colorless and transparent, with increasing opacity through white to black
<i>pH</i>	From 2 (carbon black) to 12 (calcium hydroxide)

<i>Moisture</i>	Traces to 10+%
<i>Oil absorption</i>	From a few grams to over 1000 g/100 g of filler
<i>Thermal properties</i>	Thermal expansion coefficient and thermal conductivities vary widely
<i>Electric and magnetic properties</i>	Wide variations are possible between non-conductive and conductive and between magnetic and non-magnetic

These and other properties of fillers are used to describe individual products. The potential applications of a filler are determined by its set of properties listed above but, often, other characteristics must be known to select the optimum filler or fillers for specific application. Additional properties are discussed in Chapters 5 to 12.

### 1.3 DEFINITIONS

These different sources define fillers in different ways

<i>Dictionary</i>	A material used to fill a cavity or increase bulk of something
<i>Technical dictionary</i> <sup>30</sup>	A material added to a polymer in order to reduce compound cost and/or, to improve processing behavior and/or, to modify product properties
<i>Encyclopedia</i> <sup>31,32</sup>	Fillers, or extenders as they are called in the coatings industry, are finely divided solids added to polymer systems to improve properties and reduce cost
<i>Handbook</i> <sup>33</sup>	Fillers are solid additives, different from plastics matrices in composition and structure, which are added to polymers to increase bulk or improve properties
<i>ASTM C 709-91</i> <sup>34</sup>	In manufactured carbon and graphite product technology, carbonaceous particles comprising the base aggregate in an unbaked green-mix formulation
<i>ASTM C 859</i> <sup>35</sup>	A general term for a material that is inert under the conditions of use and serves to occupy space and may improve physical properties
<i>ASTM D 123-96</i> <sup>36</sup>	A relatively inert material added to a plastic to modify its strength, permanence, working properties or other qualities, or to lower costs
<i>ASTM D 1566-95a</i> <sup>37</sup>	A solid compounding material, usually in a finely divided form, which may be added in relatively large proportions to a polymer for technical or economic reasons
<i>ASTM D 1968-96a</i> <sup>38</sup>	A material, generally non-fibrous and inorganic, added to the fiber furnish
<i>ASTM D 3878-95c</i> <sup>39</sup>	A primarily inert solid constituent added to the matrix to modify the composite properties or to lower cost



These definitions fail in some ways:

- In many instances the filler is regarded as an inert solid used for cost reduction
- Some exclude fibers, some accept fibers as fillers
- None describe conditions under which the filler lowers the cost and/or affects other properties

Although not crucial to the technology itself a more rigorous definition will serve to set boundaries and include all that is vital to filler technology.

The word fill is synonymous with the action of filling, cluttering or dumping as these are very common human activities. It also means saturate, penetrate, infiltrate, impregnate, pack, quench all of which are consistent with what fillers are designed to do. They saturate and pack spaces depending on their shape, particle size distribution, etc. Fillers penetrate and infiltrate materials. But, there are hardly any cases in which the surrounding material penetrates the filler's outside boundary. Their impregnating and quenching activity can be translated into their ability to react or interact with the surrounding material.

Thus, the word “filler” adequately describes the “filler's” potential to perform in multicomponent systems. To follow this simple lead this definition provides the simplicity and precision needed:

“Filler is a solid material capable of changing the physical and chemical properties of materials by surface interaction or its lack thereof and by its own physical characteristics.”

If one compares this definition with the other above, the noticeable differences are as follows:

- It does not attempt to provide an incomplete list of properties. It suggests that a broad scope of properties can be influenced by fillers
- This definition implies the existence of two ways in which a filler performs in a system – through its own properties (e.g., hardness, particle size, particle shape, etc.) and through interactions with the material (the extent of which can vary from strong chemical/physical interaction to almost no interaction at all). This allows us to include all existing fillers (even the degrading fillers which have too large a particle size and too small an interaction to combine with the material in an economical manner)
- The definition does not exclude a material because of its shape, particle size or chemical composition.

We may now judge the definition based on expectations developed in the discussion in Section 1.1. From a cost reduction analysis, it is evident that if a filler has a large particle size and no strong interaction with its surroundings it will decrease the intrinsic mechanical performance of the material. Such fillers are rightly called “degrading fillers”. The material density depends not only on the combined densities of the filler and the matrix but also on the interaction with and

wetting of (quality of mixing) the filler's surface by the matrix. Optical properties are affected in a similar manner. When transparency is needed, a proper match of refractive indices is required but also the filler must be incorporated with a minimum of voids through good mixing and wetting. Anti-blocking properties and lowering of the coefficient of friction are improved due to crystallization and orientation of the matrix on the filler's surface. Shape retention is affected by interactions on the filler's surface and in intumescent applications, the filler is not only responsible for producing volatiles to expand the material but it also retains the bubbles formed in the process. Thermal, magnetic, and electrical properties depend on both the filler and matrix but also on interactions between the filler and the matrix. Filler particles which are to influence permeability must have shape characteristics which permit close packing and a high affinity for each other and the matrix if permeability is to be maximized or, conversely, little affinity and minimum packing efficiency if minimum permeability is required. Many papers outline reasons for the improvement of mechanical properties. "Interaction" enters into most explanations along with properties such as surface, shape, rigidity, or strength. Chemical reactivity in the presence of a filler can change the probability of a reaction occurring often because the structure of the reactive molecule changes to make reactive groups more accessible. Each chemical reaction requires intimate contact between the chemical groups entering into the reaction. The durability and environmental impact conferred by fillers are caused by similar principles. The effects that fillers have on other components of the formulation are based on the ability of fillers to be UV absorbers, fire retardants, etc. or on mechanisms which cause the filler's surface to interact with additives (e.g., better retention due to absorption, reaction with adhesion promoters, slow release of catalyst, etc.). The rheological and morphological effects of fillers require interactions with surrounding materials.

To further test the definition we should verify that all materials known to be used as fillers can be included in the definition. Organic materials are of concern since other definitions seem to exclude them. This is a serious inconsistency given the fact that wood flour was one of the first fillers used in modern polymers. Today, when many recycled products are used as fillers, their exclusion does not serve any purpose since they do contribute to the improvement of the materials which will use them. They are included in our definition.

Also, fibers are controversial. In one currently used handbook,<sup>33</sup> natural, inorganic fibers such as wollastonite or asbestos have been included among fillers whereas other fibers were included in a separate group with only three materials: glass, aramid, and graphite. But, mixtures of fibrous and particulate materials are found in many composites today and various natural materials having fibrous structures are considered fillers in technical papers. Again our definition includes these examples.

Finally, carbon black and titanium dioxide are frequently classified as colorants, as opposed to fillers.<sup>33</sup> Tire customers “can choose any color as long as it is black”. This is economical technology due to the reinforcement and UV protection offered by carbon black. There are other instances in which carbon black is used for these two reasons. It seems wrong to classify it as a colorant. It is rather a filler which bestows several essential benefits due to its properties and its interaction with the matrix. In the paper industry,<sup>40</sup> titanium dioxide is qualified as a filler when it fills the space between fibers and pigments in the surface coating.  $\text{CaCO}_3$ , talc, clay, etc. are also considered pigments in the paper industry. There are very few reasons today to distinguish between fillers and pigments. In the past, it was perhaps simpler because any material which had a particle size below  $1\text{ }\mu\text{m}$  was considered a pigment. Today, the majority of fillers fit this criterion. We include titanium dioxide in this discussion because it has physical properties other than color (e.g., very high refractive index, photochemical activity, UV absorption, etc.) which contribute to the performance of the material in which it is compounded. Our definition also eliminates the exclusion, based on chemical composition or particle size, from the group and allows the inclusion of such materials as gold and nanoparticles.

## 1.4 CLASSIFICATION

In the first edition of this handbook,<sup>41</sup> fillers were assigned to groups according to their mineral origin and chemical composition (mineral, glass, carbon black, organic, metal). The group of mineral fillers was further divided according to geological classification. We now prefer not to use the physical origin or the chemical composition of the filler as a division.

Classification by particle size is helpful in classification since particle size will affect performance but, by itself, falls short as a criterion when selecting fillers for applications which require certain levels of conductivity (thermal or electric) or of chemical interaction, etc. In one publication,<sup>33</sup> materials were divided into particulates, fibers, and colorants. These distinctions are not helpful for a material designer. For a classification to be useful in filler applications, it must include the most important properties of fillers which affect the resultant material. The eight most important are as follows:

- Particle size and distribution
- Aspect ratio
- Chemical composition of surface
- Mechanical properties of filler particles
- Electric and thermal conductivity
- Quantitative description of interactions
- Composition of admixtures
- Optical properties

The existing data may allow us to classify materials according to these properties, however, eight major denominators seem too complex to use to apply practically. Thus, we have decided that, of more than 70 groups of fillers in use today, each will be named based on its common use. These are derived from chemical composition (chemical name), method of filler preparation (precipitated, fumed, hydrated, etc.), mineral source, shape of particle, origin (e.g., original waste material from which ground product is manufactured, name of natural organic product, sand, etc), or material structure (e.g., metallized ceramics). This listing of products has some deficiencies but if presented in alphabetical order, fillers are easy to find. However, it is essential to think about fillers in terms of the eight major denominators or the full list of major properties listed in Section 1.1 or Chapter 5. This will provide the greatest benefit in selecting fillers for specific applications.

## 1.5 MARKETS AND TRENDS

Filler market in plastics alone totals over 10,000 tones per year.<sup>42</sup> Calcium carbonate takes about 2/3 of this market. The market is very large but a large segment of it consists of use in products which are not sophisticated technically. The main applications include:

- Plastics
- Construction
- Paper
- Paints and coatings
- Cosmetics and pharmaceuticals
- Fibers
- Food
- Friction materials
- Printing

These applications are covered in detail in Chapter 19.

Four polymers are the largest consumers of fillers (PVC, PP, PA, polyesters). The consumption of each polymer is immediately mirrored by the consumption of the fillers used in this polymer. The most recent changes in PVC consumption were reflected in the consumption of fillers. At the same time, future trends and developments in fillers are more related to the advances of plastics as they replace many traditional materials. For plastics to give the required performance, new filler technology was required. Current developments allow us to predict some future directions in filler markets. These technologies will become more important:

- Nanoparticles
- Conductive fillers
- Surface modification technology
- Filler mixtures
- Non-dusting fillers

- Morphology-specific fillers
- Compatibilizing fillers
- Low-cost reinforcing fillers

Many new applications of plastics (especially in the high technology sector) are becoming possible due to the advances in nanoparticulates and conductive filler technology. The studies in these areas remain closer to laboratory scale than to full production. Surface modification and filler mixtures will be driven by two expectations: increased mechanical properties and to use fillers more as rheology modifiers. Many new products are being tested in this area now and newer products will enter the market in the next few years.

Dust is one of the most troublesome hazards associated with fillers. Thus, compressed (pelletized) fillers will become more important and wetting technology will be more extensively used. New developments in medical applications require compatibility of medical devices with tissues and body liquids. Advances are expected from the synthesis of inorganic materials which will form artificial surfaces which are less intrusive and which meet performance requirements.

The current emphasis on material recycling requires materials to contain additives which will allow the processing of complex mixtures of polymers through compatibilization, increased thermal resistance during reprocessing, allow for filler recovery, and allow the use of ground waste as a filler. All these technologies have high growth potential because of social, regulatory, and economic pressures.

These current developments place an emphasis on the perfection of filler technology. This has resulted in the creation of many very high quality materials which are too expensive to use in most applications. There is a need to develop materials which are substantially more cost-effective but still allow the conservation of matrix materials. This will be driven by environmental concerns. Product life cycle evaluation, an emerging development, will have a strong impact on the choice of future technologies and fillers associated with these technologies.

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