

State of the Art

Materials for increasing the thermal conductivity of plastics

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1 Introduction

The present report on the state of the art with regard to materials for increasing thermal conductivity of plastics was compiled within the framework of the PolyMetal project in the **Interreg V-A Slovenia-Austria 2014-2020** funding line and serves as a basis for decision-making on the most promising procedure for the prototypes to be processed in the project in order to achieve the project objective.

In technical applications, metallic surfaces, handles, coverings etc. are perceived as "more valuable" than comparable parts made of plastic. This is mainly due to the optical and haptic properties of the different materials. The so-called "cool-touch effect", achievable by the use of metals as opposed to plastics, plays a central role in this. The decisive difference between the two materials lies in the heat conductivities of metals and plastics. As shown in Table 1, the thermal conductivity of metals is 100 to 1000 times greater, depending on the material used, than that of polymeric materials.

Table 1: Comparison of the thermal conductivities of metals [1] and plastics [2].

Metal	Thermal conductivity (Wm ⁻¹ K ⁻¹)	Plastic	Thermal conductivity (Wm ⁻¹ K ⁻¹)
Steel	approx. 11 - 60	LDPE	0.33
Lead	34-35	HDPE	0.45 - 0.52
Iron	68-80	PP	0.14
Chrome	87-91	PS	0.04 - 0.14
Aluminum	221-247	PC	0.19
Gold	292-316	PA 66	0.26
Titan	15-22	PET	0.29
Silver	414-493	PMMA	0.15 - 0.25

Another criterion that makes metal surfaces appear more high-quality is the typical gloss. However, this is a sensory impression created by a number of optical phenomena such as refraction, diffraction and reflection and is therefore difficult to

measure. Various approaches for this have been proposed and are being studied at PCCL, but there are no comparative numerical values for the gloss of different materials to be found in the relevant literature that would permit a direct comparison. This is due, inter alia, to the fact that these effects do not depend solely on the materials used, but also, to a considerable extent, on the surface condition and surface geometry of the component in question.

There are several reasons why efforts are being made to replace metals with plastics in numerous applications. On the one hand, the injection moulding process as well as other production methods used in plastics processing enable a comparatively cost-effective, fast and design-flexible production of components. As a rule, little or no post-processing will be required for parts that are produced by means of the injection moulding process. Furthermore, the substitution of metal parts by plastics permits both a reduction of weight and cost savings.

In order to be able to obtain metal-like properties in the case of plastic components, the polymers used must be enhanced using additives. The choice of additives will depend on the exact requirement profile identified for the application and may vary widely.

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2 Methods of increasing the thermal conductivity of thermoplastics

To increase the thermal conductivity of plastics, thermally conductive additives must be added in a compounding step. There are many different materials to choose from. A selection of such materials may be found in Table 2. The thermal conductivities of metals, which, in powdered form, may also be used as fillers for plastics, are shown in Table 1.

Table 2: Thermal conductivity of plastic additives [2]

Filler	Thermal conductivity ($\text{Wm}^{-1}\text{K}^{-1}$)
Silicon oxide (SiO_2)	1.5 – 1.6
Silica (crystalline silicic acid)	3
Aluminum oxide (Al_2O_3)	38 - 42
Beryllium oxide (BeO)*	300
Zinc oxide (ZnO)	60
Boron nitride (BN)	29 - 300
Silicon carbide (SiC)	85
Diamond	2000

* Highly toxic

The various fillers differ greatly in their thermal conductivity and also with regard to their other properties, such as particle shape and size [3, 4], processability, potential dangers (beryllium oxide, very fine powders, etc.), possible electrical conductivity and of course their price. For example, diamond has excellent thermal conductivity, but the use of diamond powder as an additive in plastics processing is out of the question for economic reasons.

The particle shape of the various additives has a huge impact, as shown by the example of platelet-shaped boron nitride (the most important supplier of which is 3M Technical Chemicals, Kempten, Germany). The thermal properties of compounds containing such particles are highly anisotropic since they depend on particle orientation within the component. Conductivity through the component (normal to particle orientation) is significantly lower than that in the direction of particle orientation. Particle size and filler content also exert considerable influence on subsequent component properties, but at the same time also influence processability and thus the possibility of producing homogeneous compounds. In high concentrations, small particles and especially nanoparticles tend to form agglomerations. As a result, they lose the advantage of their large specific surface area and produce compounds with erratic properties. Furthermore, due to a possible development of dust in the dosing system (dust explosion) and possible health hazards to the machine operator (by inhalation), the processing of nanoparticles may not be entirely safe [5]. A similar hazard potential is also associated with very fine metal powders.

If, as is frequently necessary e.g. in connection with electrical applications, electrically insulating materials offering an increased thermal conductivity are required, the range of selection of relevant additives is reduced to mineral fillers or metal oxides. However, the thermal conductivity of such materials, with the exception of boron nitride, which, at around 80 € / kg, is very expensive, usually falls within a medium range if the materials are pure. It must be remembered, though, that in a compound, these filler particles will be surrounded by the thermally insulating polymer matrix, which means that the values of the pure fillers can never be achieved in a compound. A realistic point of reference would be about 10% of the conductivity of the filler in cases where there is a high filler content. By increasing the filler content and obtaining microstructures (thermally conductive paths, see Figure 1) in the compound, it is possible to increase thermal conductivity.

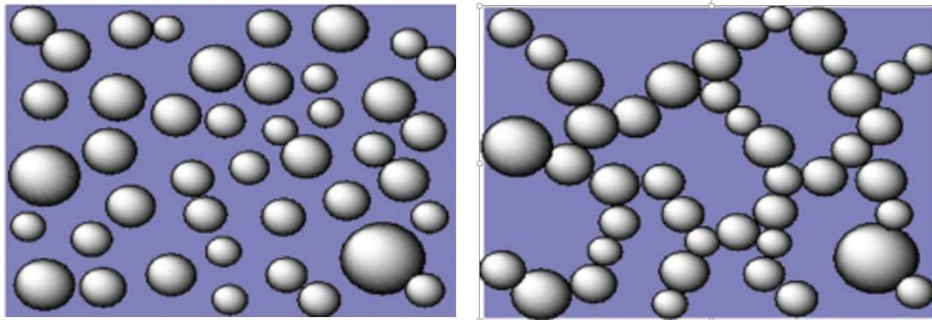


Figure 1: Schematic representation of a compound with the same filler content, but different microstructures. [2]

An increase in the proportion of the filler used, however, will not only affect the thermal conductivity of the compound, but will also have a serious impact on the mechanical properties of the material. As clearly shown in Figure 2, the compound's elongation at fracture decreases dramatically as the filler content increases. This means that the material becomes very brittle and that a component made of such material will break even at low mechanical stresses. Materials with extremely high filler content are also difficult to process and tend to form uneven and rough surfaces, which then require elaborate polishing in a subsequent step to make them visually appealing. In addition, highly filled plastics (depending on the filler) cause heavy wear on the machines and tools used during their processing.

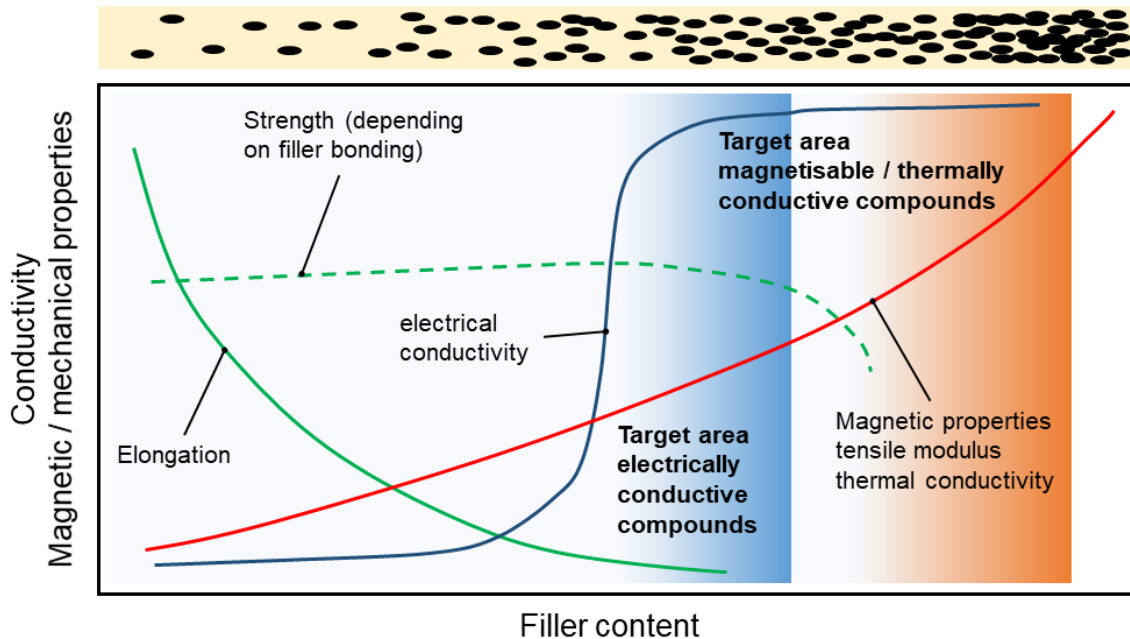


Figure 2: Impact of filler content on the properties of the compound. [6]

As the blue line in this figure shows, electrical conductivity will, up to a certain point - the so-called percolation point -, remain at a low level if the filler content remains low. In this way, electrically conductive additives can be used to increase thermal conductivity at low levels of filler content, without rendering the material electrically conductive.

When producing compounds aimed at increasing the thermal conductivity of polymers, therefore, a compromise must always be achieved between the thermal, mechanical, optical and electrical properties and the raw material costs and processability. In doing so, it will be necessary, first of all, to take a look at the proposed application and the given framework conditions and/or the amount of heat to be transported. If a component is to be used in a stationary device within a room and the heat is released into the environment only through free convection, thermal conductivities above approx. $5 \text{ Wm}^{-1}\text{K}^{-1}$ will not be advisable since it will be impossible to dissipate the heat without additional cooling.

The best results with thermally conductive, electrically insulating plastic compounds will be achieved using boron nitride. Since this is very expensive compared to the base polymer, its use will, in many cases, be prevented by economic considerations. Alternatively, some suppliers, such as Quarzwerke Frechen (Germany), offer aluminum

oxide and zinc oxide based fillers which, according to the manufacturer, offer thermal conductivities within the range of 14 to 30 $\text{Wm}^{-1}\text{K}^{-1}$ [7]. For better processability and an improved connection between the filler and the plastic, these fillers are offered with different coatings (epoxy, amino, metacysilane). In PA6, a thermal conductivity of 2.3 $\text{Wm}^{-1}\text{K}^{-1}$ or 1.3 $\text{Wm}^{-1}\text{K}^{-1}$ (depending on the direction of the measurement) can be achieved with a maximum filler content of 75% of Silatherm 1360.

A new option in the field of thermally conductive plastic additives is provided by bi- and trimodally distributed aluminum oxides (Al_2O_3) obtainable from Güpo GmbH (Kehl, Germany). These materials are produced by a calcination process of aluminum hydroxide. According to the manufacturer, the grain shape is designed so as to ensure that the wear during processing is minimal; the bi- and trimodal particle size distribution will, on the one hand, enable high filler content levels and, on the other hand, give rise to isotropic properties. In PA6 compounds, heat conductivities of 8.3 $\text{Wm}^{-1}\text{K}^{-1}$ can be achieved with maximum filler content levels of 65 vol% in Güpotim WP-02 (trimodal mixture).

To optimise the costs while ensuring the best possible thermal conductivity, some users opt for a hybrid filler made of boron nitride and other mineral fillers. With a suitable formulation (mixing ratio, particle sizes, coatings, etc.), it is possible, in this way, to produce comparatively cheaper materials with satisfactory thermal and mechanical properties.

Since the implementation of such tailor-made compounds with hybrid fillers is very demanding and involves complex material analyses and process and formulation optimisations, it may be expedient to resort to commercially available, thermally modified polymers. Different material manufacturers and compounders such as Lehmann and Voss, Sabic, Polyone, Ensinger or Lati Industria Termoplastici offer a wide range of thermally conductive materials using different carrier polymers. Depending on the base polymer and filler type and quantity, the manufacturers promise thermal conductivities of up to 10 $\text{Wm}^{-1}\text{K}^{-1}$ and in some cases even more. These data should, however, be viewed with caution, since there are different systems for measuring thermal conductivity (Hot Disk, Guarded Heat Flow Meter, Laser Flash),

which may sometimes produce significantly different results as regards thermal conductivity. This is partly due to the fact that some measuring systems take radial and axial heat conduction into account, while others only measure heat conduction through the test specimen. In addition, as already mentioned, particle distribution and orientation within the sample have a significant impact on thermal conductivity. This must be taken into account when producing injection-moulded parts, since different filler orientations may occur, depending on component geometry, tool design and also on the process conditions. These effects, as already mentioned, and as documented in numerous scientific papers, such as those by Grundler et al. [8], impact thermal conductivity in the component. This means that thermal conductivity values, as given in material data sheets, are not directly transferable to a component.

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3 Literature

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