

Filled polypropylene: a cost-performance comparison of common fillers

Composite materials such as carbon fibre-filled epoxy find use in such applications as tennis rackets, golf clubs and military aircraft. However, these high performance materials only serve certain markets. Mineral-filled plastic compounds are more commonly found and are widely used in cars, appliances, wiring, furniture and many other applications. It is estimated that the global consumption of fillers in plastics is over six million tonnes per year and this figure has been growing steadily for the past several years at the rate of 5-8% per year. Dr. Chris DeArmitt of Electrolux Core Technology & Innovation describes the results of a study to compare the cost and performance of some common fillers in polypropylene compounds.

The reason for the popularity of composites and filled plastics compounds is their ability to combine some of the best properties from each material. For example, by blending a mineral filler into polypropylene (PP) it is possible to combine the processability and impact strength of the PP with some of the stiffness of the mineral component.

There are a number of reasons for adding a filler to a polymer. These include:

- Raising heat resistance (heat deflection temperature)
- Reducing cost (in some cases)
- Increasing stiffness
- Reducing shrinkage
- Increasing electrical conductivity
- Reducing flammability (for example using ATH or $Mg(OH)_2$)
- Improving impact strength
- Improving dimensional stability
- Improving thermal conductivity (reduce injection moulding cycle time)

There is a wide range of fillers that can be used to achieve processing and mechanical advantages or to impart special properties like electrical conductivity or shielding from electromagnetic radiation. Likewise, there is a wide range of polymers that are used. Common examples include PE, PP, PVC and nylons. This article focuses on PP compounds although the effect of the different fillers in PP are expected to be similar to those for other semi-crystalline thermoplastics such as polyethylene and nylon. The data presented was sourced from www.matweb.com - a free resource for materials data - with mineral fibre data from Lapinus Fibres BV and wood fibre data from American Wood Fibers. All data is for injection moulding grade PP homopolymer.

Fillers

It is often believed that fillers are used primarily to reduce cost. Obviously, cost is an important factor, however, there may be no overall cost reduction. Fillers are

often less expensive than polymers on a weight basis. However, in order to make a part of a certain size it is necessary to think in volume terms, particularly if existing moulds are to be used. Mineral fillers typically have a density in the range $2.5-2.8 \text{ gcm}^{-3}$, while common polymers are in the range $0.9-1.4 \text{ gcm}^{-3}$. Therefore, on a volume basis, the filler is usually not cheaper than the polymer. In addition, there is a processing cost for compounding the filler into the polymer, and this must also be considered in addition to the raw materials cost. However, common mineral fillers have ten times the thermal conductivity of many polymers and this aids cooling in the mould. Cooling time reductions of 30% are not uncommon and the resultant increase in productivity can be particularly advantageous. Where fillers are concerned, characteristics like particle size distribution, colour, hardness, density, aspect ratio and the presence or absence of surface treatment must be considered. There are several common fillers used in thermoplastics. By far the

most common is calcium carbonate, which is inexpensive, widely available, roughly spherical, soft and can be obtained with a high whiteness. Talc is more expensive than calcium carbonate but is platy and therefore can give some reinforcement. In addition, talc is a well known nucleating agent for PP - helping to reduce injection moulding cycle times. Glass fibres are colourless and far more expensive than the other common fillers for PP, but they provide the greatest reinforcement. These three fillers are common and have the advantage of giving white or colourless compounds. This allows for easy colour matching, which is essential for aesthetic (visible) parts. In applications where appearance is less important, coloured fillers such as wood fibres (light brown, low aspect ratio), mica (brown, platy), wollastonite (grey fibres aspect ratio of 1:5 to 1:20) and mineral fibres (green) should also be considered.

Selecting the right material

Each application has its own special set of requirements. The challenge is to identify the requirements and then to find the cheapest material that meets those requirements. The aim of this article is to show the effect of adding some common fillers to PP. The most important mechanical data is shown graphically for different loadings of each filler. It can be seen that the same or similar mechanical performance can be achieved in many ways. Where this is the case then cost will become the deciding factor in choosing the best overall solution. Therefore, the data of mechanical performance is compared with raw materials cost.

Mechanical properties

The data presented are intended to give an overview of properties. However, these must be viewed with some caution because they are generic data from several suppliers. Nevertheless, these generic data give similar trends compared to those from carefully controlled studies. The three most important mechanical properties are the modulus, yield strength

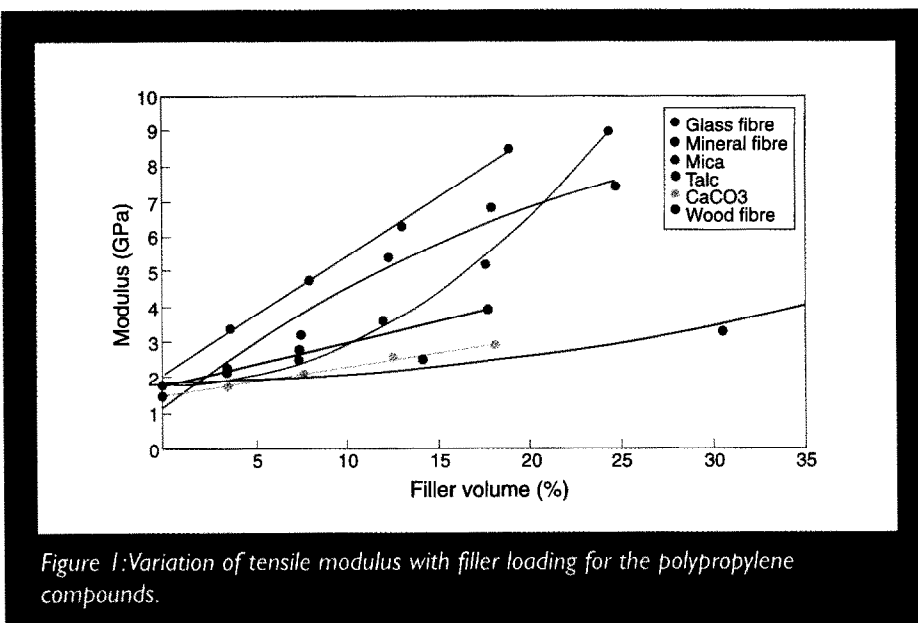


Figure 1: Variation of tensile modulus with filler loading for the polypropylene compounds.

and impact strength. These properties are usually plotted versus the weight percentage of filler. However, that approach should be avoided for several reasons. The mechanical, rheological, electrical and thermal properties of filled plastic compounds all depend on the volume percentage of each component, not the weight percentage. Furthermore, different fillers have different densities and this must be accounted for when comparing the effect of different filler types on the properties. Therefore, all properties need to be compared with the volume percentage of filler to allow a direct comparison of the effect of each filler.

In addition, the viscosity is also important because it is related to the throughput of the extruder. A drop in extruder throughput can cause a dramatic cost increase in the final material, which should not be overlooked. When the filler loading is varied, the density of the filled polymer melt is also changed. This means that MFI data cannot be compared directly for samples containing different fillers at different loadings. Sometimes it has been claimed that adding filler has improved the flow (increased the MFI) of the polymer. Such an increase in MFI is simply due to the higher density of the filled polymer melt. Instead of MFI, the volume MFI (VMFI or MVR) must be used. The VMFI is the MFI data corrected for

the density of the filled polymer melt. It represents the actual viscosity of the melt and has units of ml per 10 minutes.

Modulus

The modulus (stiffness) is presented for several common fillers versus increasing filler loading (Figure 1). The data presented are for the tensile modulus only, because the flexural modulus showed exactly the same trends, and even the numerical values were similar. All fillers increase the modulus, although different fillers act in different ways. The more spherical fillers, such as calcium carbonate, give moderate improvements in modulus. Platy fillers, such as mica and talc, give intermediate improvements, while the fibrous fillers give the greatest modulus increase. An exception is the wood fibre, which gives a low modulus improvement compared to the other fibrous fillers, due to the comparatively low modulus of wood. The trends seen are exactly as expected from the theory (Equation 1) and are in agreement with other studies. Equation 1 is based on a linear rule of mixtures approach, which assumes no bonding between filler and matrix and does not account for the effect of filler shape. The equation predicts a straight line, which fits the data presented quite well.

Fillers

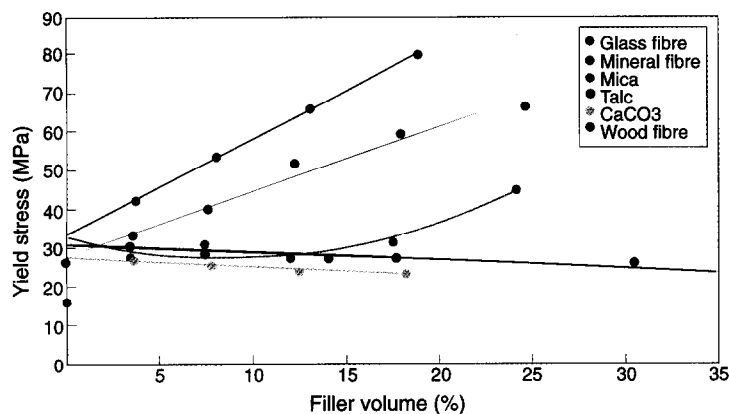


Figure 2: Variation of tensile yield stress with filler loading for the polypropylene compounds.

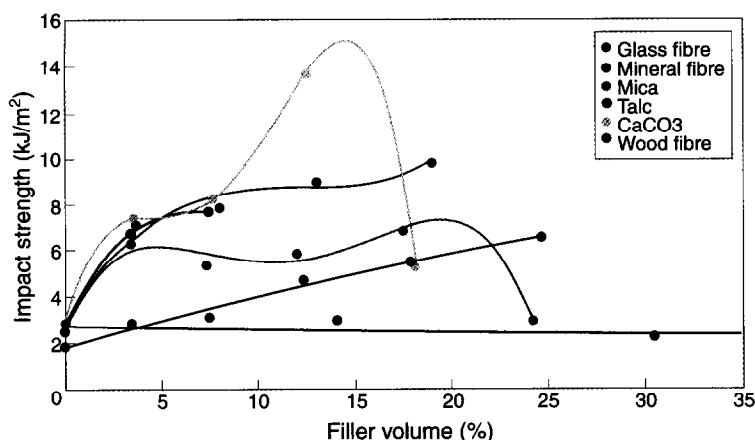


Figure 3: Variation of impact strength (notched Izod A) with filler loading for the polypropylene compounds.

$$E_c = (1 - \phi)E_m + \phi E_f \quad \text{Equation 1}$$

where E_c , E_m and E_f are the moduli of the compound, matrix and filler respectively and ϕ is the volume fraction of filler.

Yield strength

Like the modulus, the yield strength is also sensitive to the shape (degree of anisotropy) of the filler (Figure 2). Isotropic (roughly spherical) fillers like

calcium carbonate result in a slight drop in yield strength. Therefore, these are classified as non-reinforcing fillers. The platy fillers talc and mica show some increase in yield strength (reinforcement). Finally, the fibrous fillers provide the highest level of reinforcement. An exception is the wood fibre, which gives no increase in yield strength, probably due to their relatively low aspect ratio (1:3-4).

The yield strength is expected to depend on the proportion of the cross-sectional area of the filler in the matrix and also on

the degree of bonding (B) between filler and matrix (Equation 2).

$$\sigma_c = \frac{(1 - \phi)}{(1 + 2.5\phi)} \sigma_m \exp(B\phi) \quad \text{Equation 2}$$

where B is an adhesion parameter ≥ 0 , ϕ is the volume fraction of filler and σ_c and σ_m are the yield strengths of the compound and matrix (polymer) respectively. For low values of B the equation predicts a decrease in yield strength (no reinforcement) as the filler loading increases, while higher values of B give an increase of yield strength (reinforcing effect).

Impact strength

Unlike the previous two properties, impact strength does not display such a simple behaviour. The impact strength is controlled by many factors and cannot be successfully predicted. It is well known that the presence of large particles and agglomerates (above 10-30 μm diameter) dramatically reduces the impact strength (particularly unnotched) of PP. These large particles act as flaws where cracks can initiate. In contrast, the inclusion of small, well dispersed particles can have a very positive influence on the impact strength of PP homopolymers (Figure 3). The graph shows that all fillers except for wood fibre had a very positive influence on the impact strength of PP homopolymer. The filler particles are poorly bonded to the PP and act as micro-voids that deflect the crack tip and they can even change the failure mechanism from brittle to ductile in some cases.

It is important to note that while particulate fillers can be very beneficial to the impact performance of PP homopolymers, the opposite is true for PP copolymers. This is because the filler particles can nullify the effect of the rubbery phase in impact modified PP and in PP copolymers.

Heat Deflection Temperature (HDT)

The HDT represents the temperature at which a specimen under a specified

flexural load deforms by a set amount. This is important if the material is to be used at elevated temperature, for example, in under-the-hood applications in the automotive industry.

The order in which fillers can improve HDT is (in ascending order) wood fibre, calcium carbonate, talc, mica, mineral fibre and glass fibre (Figure 4). This is the same order of efficacy as found for the modulus and yield strength improvement. It is expected that the HDT should be loosely related to the flexural modulus of the compound and this is seen to be the case.

However, the HDT improvement for the glass fibre is higher than expected on the basis of the modulus alone. It has been reported the glass fibre can promote crystal nucleation and transcrystallinity (epitaxial crystal growth from the fibre surface) in PP. This formation of crystals with a higher melting point would explain the high increase in HDT for the glass fibre filled PP.

Volume MFI (VMFI)

The VMFI is inversely proportional to the viscosity of the filled polymer melt. The measurement is done at low shear rates, while the melt experiences much higher shear under processing conditions.

Nevertheless, the VMFI may be used to get a qualitative indication of the relative ease of processing and throughput for each compound (Figure 5).

In general increased filler amount results in reduced VMFI (higher viscosity). This is in agreement with the equations for melt viscosity and with previous studies (Equation 3).

$$\log(\text{VMFI}_c) = \log(\text{VMFI}_m) - \frac{\beta\phi}{\beta f\phi + f^2}$$

Equation 3

where ϕ is the volume fraction of filler, f is the free volume of the polymer and β is the free volume loss due to addition of filler into the matrix.

Cost/performance

While there have been many studies in the scientific and trade literature regard-

ing the mechanical properties of composites and filled polymer compounds, there is a distinct lack of information on the relative raw material costs for different materials. In addition, the throughput of the extruder is almost never compared, although this is extremely important in determining the final cost of a material. There are many polymers and fillers on

the market. There are also many surface modifiers available that can improve the performance of the compound in different ways. It is essential to remember the primary aim when presented with the range of the choices of ingredients and mechanical data. The best plastic compound is the one that gives the required properties at the lowest price.

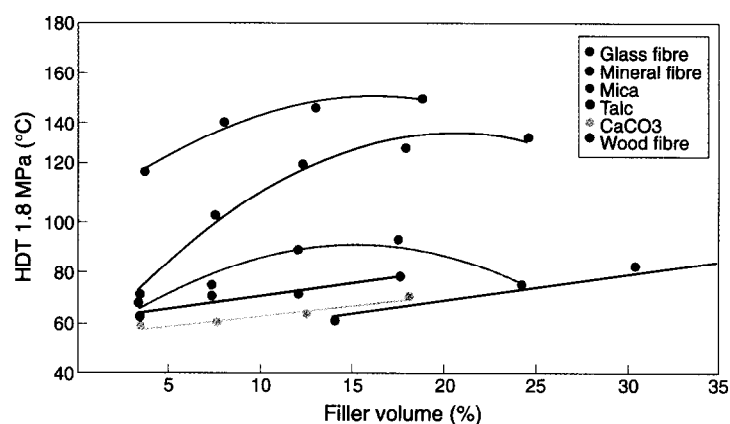


Figure 4: Variation of heat deflection temperature (HDT 1.8 MPa) with filler loading for the polypropylene compounds.

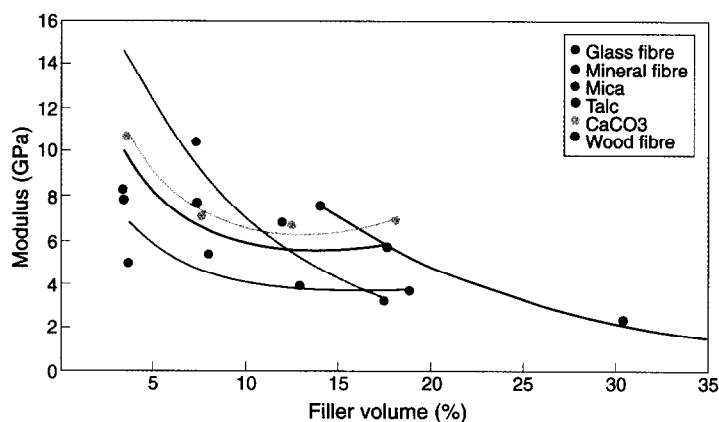


Figure 5: Variation of the volume melt flow index (VMFI) with filler loading for the polypropylene compounds.

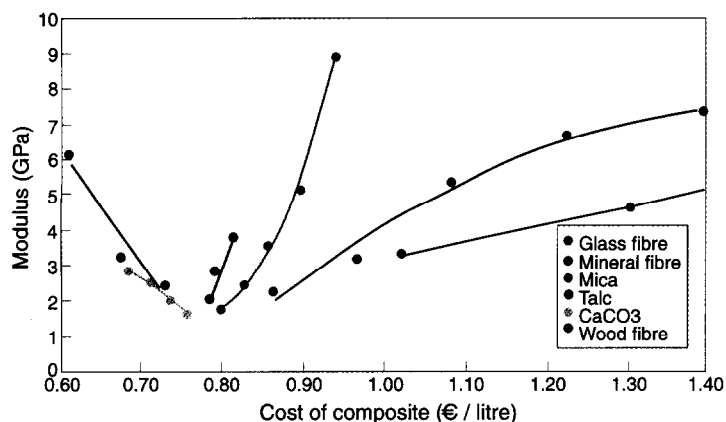


Figure 6: Tensile modulus versus cost for the polypropylene compounds.

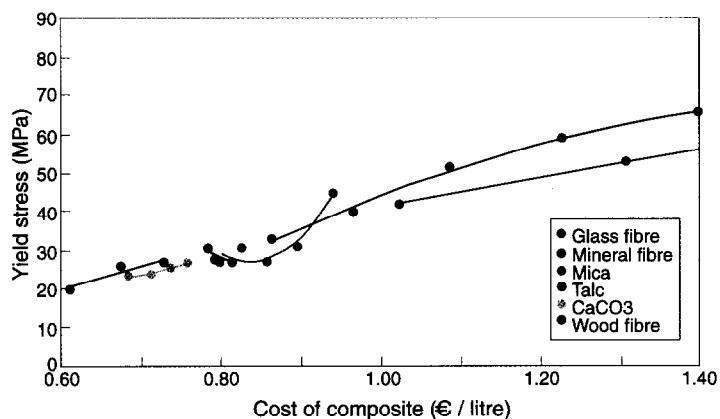


Figure 7: Yield stress versus cost for the polypropylene compounds.

Therefore, the key data has been re-plotted to show the properties versus raw materials cost.

Modulus

For some applications, the modulus is the most important mechanical property. Calcium carbonate has the best cost/performance ratio from 1.5 - 3 GPa. Wood fiber is best from 3 - 6 GPa,

while mica is best from 6 - 9 GPa. (Figure 6). These data are for tensile modulus but the values for flexural modulus are very similar. It is important to realize that the anisotropic fillers give compounds where the modulus varies depending upon the fibre orientation relative to the test direction. Therefore, with non-spherical fillers it is important to design the mould to optimize particle orientation.

Yield strength

The yield strength represents the maximum load that a material can take before it becomes permanently (inelastically) deformed. For polymers, it is commonly taken to be the maximum on the stress-strain curve and is a critical design parameter in many applications. Calcium carbonate, talc and wood fibre give lower yield strength while all other fillers result in an increase (Figure 7). In terms of cost/performance ratio, the best solution depends on whether a coloured filler is acceptable for the application. Where high whiteness and colour control are needed then calcium carbonate is best from 20 - 28 MPa and glass fibre is best from 30 MPa upwards. If colour is acceptable, then calcium carbonate remains best from 20 - 28 MPa, but mineral fibre is the best choice from 28 MPa and upwards.

A coupling agent was used for the mineral fibre filled PP, which explains the high yield strength. However, the cost of the coupling agent has already been added onto the filler and mineral fibre cost.

Heat Deflection Temperature (HDT)

HDT is important for high temperature applications. The cost/performance data can be summarized as follows: from 60 - 75°C calcium carbonate is most cost-effective; wood fibre is best from 75 - 100°C; mineral fibre is best from 100 - 120°C; while glass fibre is most cost-effective from 120 - 150°C (Figure 8).

Conclusions

Filled thermoplastics are an important class of material used in huge quantities and for many applications. Such compounds have the appeal that a wide range of properties can be attained through careful selection of polymer, filler type, filler loading and surface treatment of the filler. Filled PP was selected for study because it is an important material but the trends found are

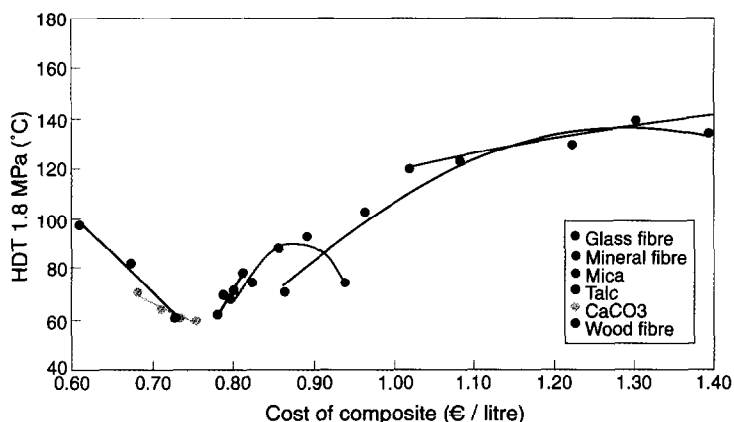


Figure 8: Heat deflection temperature (HDT 1.8 MPa) versus cost for the polypropylene compounds.

and then in terms of performance versus cost. Once the compounder has identified the requirements on the compound, the data can be used as a guide to determine what the most cost-effective solution is. However, other factors must also be considered such as colour, increased compounding cost, the cost of surface treatments and coupling agents, reduced cycle time and shrinkage.

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applicable to other filled thermoplastics. Calcium carbonate, wood fibre, talc, mica,

mineral fibre and glass fibre filled PP have all been compared in terms of performance

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