INJECTION MOULDING OF THERMALLY CONDUCTING POLYMERS FOR MECHATRONIC APPLICATIONS

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The use of highly filled thermal-conductive thermoplastics is an innovative approach to directly adjust the thermal conductivity of plastic parts for heating and cooling systems. Compared to standard resins thermal conductive thermoplastics show a higher thermal conductivity in the range of 2 to 20 W/m/K. The filler-content and the high thermal conductivity affect directly the flow- and cooling-conditions during injection moulding. Therefore, the manufacture of injection-moulded parts requires adjusted processing strategies.

In this paper properties of thermal conductive thermoplastics relevant to the design of injection-moulded parts as well as approaches to intelligent processing strategies (mould concepts, process control) are introduced.

Introduction

There is a growing need for innovative concepts in the thermal management of technical applications due to increasing device complexity (electronification), constraints on construction method (miniaturization) and altered thermal framework conditions. The input and dissipation of heat into and out of individual components and functional modules is therefore an important conceptual task. An efficient approach is to take existing components and generate further functions via these. For example, plastic housings could take on thermal functions in addition to mechanical ones /1, 2/.

The relevant characteristic for technical applications is thermal conductivity. Thermal conductivities of 2-15 W/m/K, as opposed to 0.15-0.5 W/m/K for commodity plastics, are sufficient, and can be obtained by adding metallic and ceramic fillers in portions of 40 to 60 vol. % /3/. The high filler fraction and the thermal conductivity, which is orders of magnitude higher, lead to changes in process conditions, relative to standard injection moulding, especially as regards temperature and pressure conditions during formation of the moulded part/4/.

For moulded part and tool design, it is therefore necessary to describe the processing behaviour of thermally conducting polymers by means of both experiment and simulation. Especially, moulded-part filling needs to be examined by comparing practical studies with filling simulations.

Basics

Types of heat transfer

In thermodynamics, heat transfer in technical processes is described by defining heat as a form of energy that is transferred across system borders only because of a temperature difference /5/. Heat transfer is possible by three different mechanisms:

- convection (free and forced),
- radiation, and
- conduction.

Heat transfer through walls is a common process and involves all three mechanisms. In theory, increasing the thermal conductivity leads to a linear improvement in heat transport. For the linear case, the relationship reduces to:

$$\dot{Q} = -\lambda \cdot A \cdot \frac{\partial T}{\partial x}$$

where $\dot{Q}$ is the heat flow, λ is the specific thermal conductivity, A is the transducer surface, T is the temperature and x is the path coordinates in the direction of view.

Determination of thermal conductivity

Non-steady-state methods are increasingly finding application in polymers because they allow the thermal characteristics of specimens with realistic geometry and morphologies, i.e. thin wall thicknesses, as well as directly on the products, e.g. films, to be determined. Measuring times are much shorter and the manufacturers claim that measurements can be made over a wide range from 0.01 to 400 W/m/K /6/.

Hot-wire methods (e.g. hot-plane method, Fig. 1) work by incorporating a quantity of heat appropriate to the thermal conductivity of the specimen by means of an electrically heated wire (hot wire), which may be designed as a strip (hot strip) or a circular foil (hot disk). The resulting temperature in the specimen is measured as the heat is being incorporated and is plotted against time. Each specimen yields a characteristic thermal conductivity curve, from which the thermal conductivity can be computed.

Pulse methods (laser flash method, Fig. 1) work by inputting energy into one side of the specimen and at the same time measuring the temperature rise on the other. The curve of the temperature response can be easily used to derive the thermal conductivity via the density and heat capacity of the specimen.

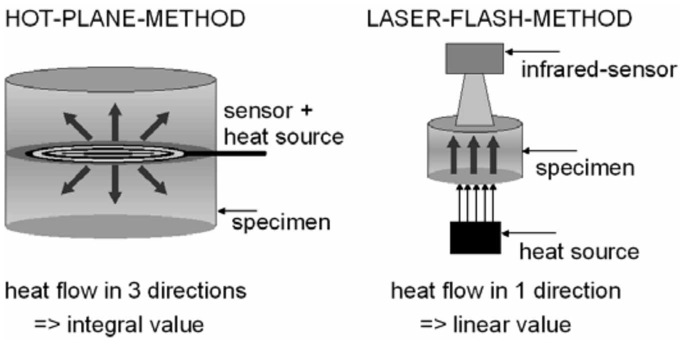


Figure 1: Methods for measuring the thermal conductivity of polymer

Significance of thermal conductivity in polymer engineering

There is a fundamental distinction between thermal conduction by polymers in the solid state (the service phase) and in the molten state (processing phase) /7/.

The first step in investigating the potential of using thermally conducting polymers in mechatronic components as housings, heat spreaders, coils, etc, is to maximize thermal conductivity. Intelligent concepts for removing heat include direct, controlled heat transmission, rapid heat distribution as well as short-term heat storage until further heat emission (Fig. 2) /1-3/.

The level of thermal conductivity which can be needed for technical applications and which thermally conducting polymers can reach is 1-20 W/m/K. In some cases, because considerable quantities of special metallic or ceramic fillers and additives are necessary, there are concomitant changes in other typical characteristics of polymers. Elongation at break is usually dramatically reduced while rigidity is increased. The change in strength is a function of the filler and its attachment to the matrix. Improvements can be made in the mechanical properties by adding special adhesion promoters.

The processing behaviour of polymers during injection moulding is described and influenced by two critical parameters. The first is the cooling behaviour of the polymer melt in the cavity and the second is the hydrodynamic behaviour of the melt, i.e. the pressure loss. A polymer melt of high thermal conductivity is characterized by markedly accelerated melt cooling.

The reduction in fluidity that accompanies a rising filler content is likewise counteracted with extra additives e.g. waxes. Filled polymers essentially have a rheology comparable to that of concentrated suspensions containing a high-viscosity suspension or matrix liquids. As with all suspensions, the flow is crucially determined by the concentration, the size and shape of the filler particles as well as the flow function of the matrix liquid (polymer melt) /8/.

As thermally conducting thermoplastics have thermal conductivities which are orders of magnitude higher than those of commodity thermoplastics, they may be expected to cool rapidly across their flow cross-section (Fig. 2). The extent to which the rapid growth of the solidifying edge zone and thus the decrease in the flow cross-section affect mould filling and the possibilities of reducing this are the subject of current research.

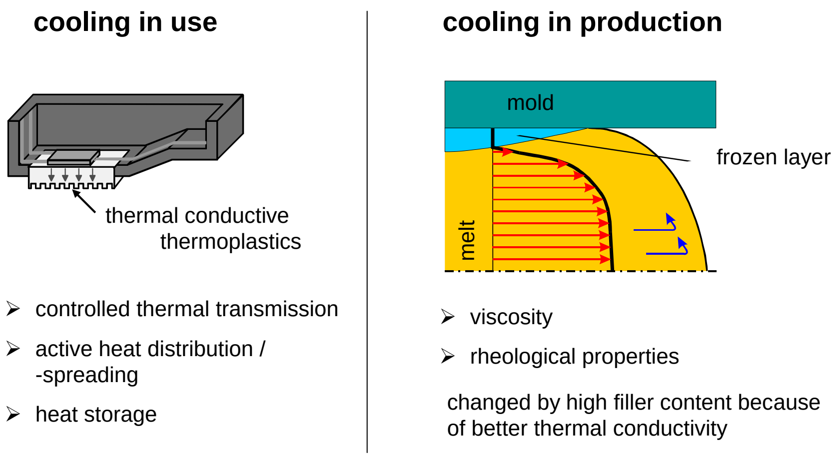
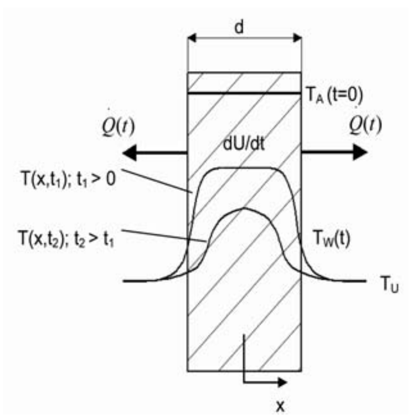


Figure 2: Importance of thermal conductivity as service indicator and processing indicator

Heat transfer during injection moulding

The cooling behaviour of the polymer melt in a mould cavity can be assessed from the thermal equation for the boundary conditions of constant melt and mould temperature and compounds of different thermal conductivity. For an infinite plate and a low ambient temperature, heat flow to the environment occurs from time t=0 (homogeneous plate temperature) (Fig. 3) /5, 9/.



temperature sequence during the cooling process in a plate

$$\frac{\partial T}{\partial t} = a \cdot \frac{\partial^2 T}{\partial x^2}$$

Figure 3: Schematic diagram of cooling (temperature curve) of a warm plate in a cold environment

The temperature distribution resulting from the heat flow at different times is shown in Fig. 4. After two seconds, the unfilled polyamide (with a thermal conductivity of approx. 0.3 W/m/K) has a pronounced temperature gradient ranging from the mould temperature on the outside to the melt temperature just 20 °C lower in the middle of the plate. By contrast, the temperature in the middle of the PA6 compound plate is estimated to be barely above 150 °C after the same amount of time has elapsed. In this case, all areas are already below the described no-flow temperature. This effect stems from the greater thermal conductivity. From this, it may be inferred that the period of time during which flow is possible in highly heat-conducting polymers is significantly reduced.

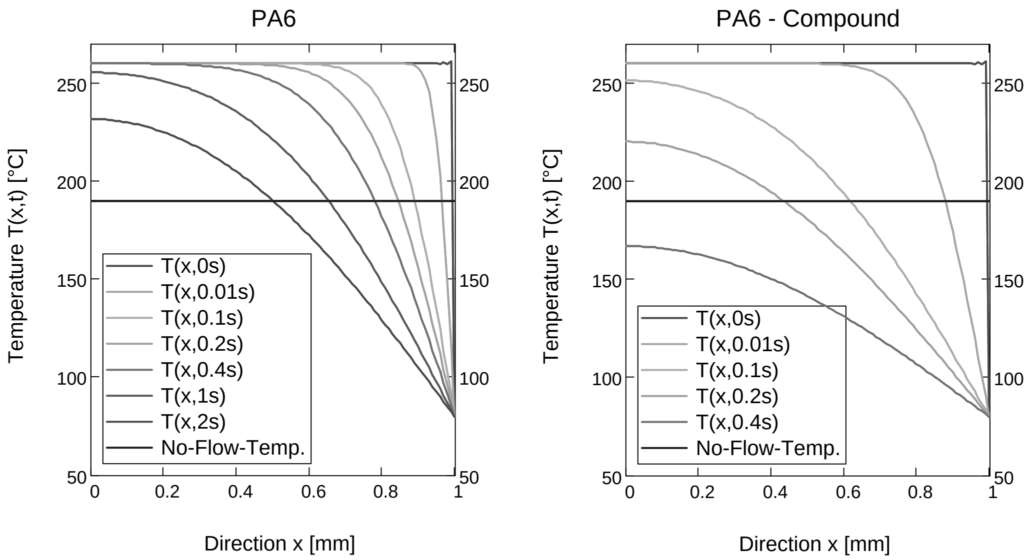


Figure 4: Place- (x) and time (t) - dependent temperature distributions on flat plates during cooling in a cold environment

Figure 5 summarizes the times needed for the temperature in the plate middle to reach the no-flow temperature, in unfilled PA6 and different PA compounds. With increase in thermal conductivity, of the level typically observed in plastic compounds, the cooling time falls to way below one second, as opposed to the 3.5 seconds needed in unfilled polymers.

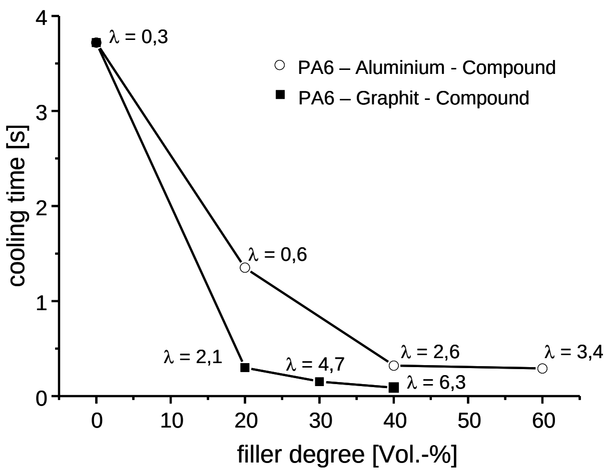


Figure 5: Estimated cooling time of compound melts in the middle of a 2-mm-thick, plate-like cavity, starting from a melt temperature of 260°C down to below the no-flow temperature

What this means in practice is that the possible injection times for highly heat-conducting polymers are substantially shortened, i.e. either the injection speeds must be high or only flow paths are possible.

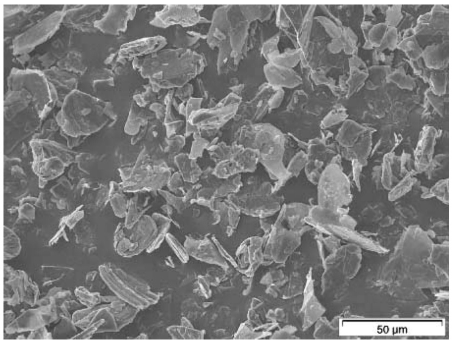
Experimental

Compound preparation

An unfilled PP containing 30 vol.% graphite powder and serving as thermally conducting compound was prepared in a Leistritz ZSE 27 HP-40 D twin screw extruder (Table 1).

Table 1: Raw materials used

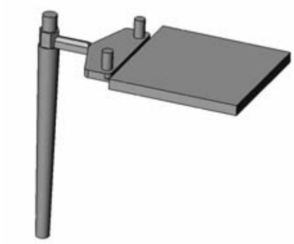
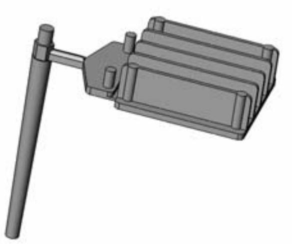
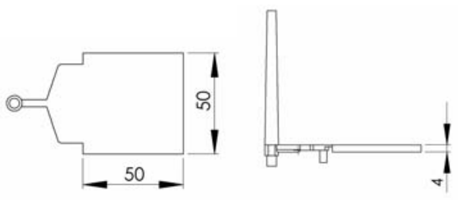
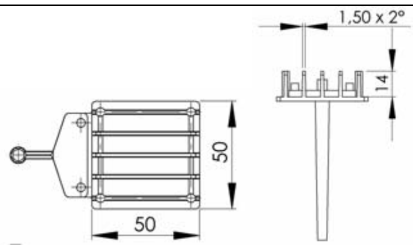
Polymer:	Polyamid 6
Type:	Ultramid B3
Company:	BASF AG, Ludwigshafen, Germany
Filler:	Graphite-Powder
Type	Timrex, KS 44
Company:	Timcal AG, Bodio, Switzerland



Production specimens

The compound was injection moulded on a standard Demag Ergotech 25/280-80 to yield simple specimen plates and a complex “heat sink” geometry by way of demonstrator. The geometry and respective processing parameters are shown in Table 2.

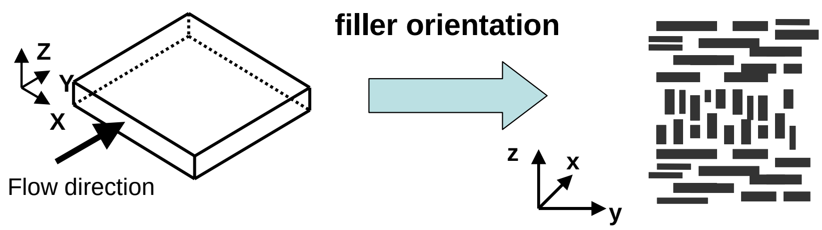
Table 2: Specimen and demonstrator geometry and injection moulding parameters

		Specimen plate 50 x 50 x 4 mm	Heat sink demonstrator Plate: 50 x 50 x 2.5 mm Ribs: 50 x 20 x 1 mm
			
			
Melt temperature	[°C]	260	260
Mould temperature	[°C]	80	80
Injection speed	[mm/s]	---	15 / 60

Results and Discussion

Determination of thermal conductivity

Whereas the specimen plates did not have to be machined prior to determination of thermal conductivity on the hot-disk instrument, the determination of thermal conductivity in the Nanoflash instrument required the machining of 13 mm x 13 mm specimens. The test specimens were inserted into the Nanoflash in accordance with the measuring direction. The density and heat capacity were determined in parallel.
The hot-disk results show that addition of 30 vol. % graphite increases the thermal conductivity by approx. 0.3 to 3.5 W/m/K. In view of flow-induced alignment of the fillers, a preferred direction for the thermal conductivity may be expected. For plates, this assumption proves to be significant in the Nanoflash measurement of the direction-dependent thermal conductivity. By analogy with the orientation of fibre-shaped fillers, an orientation model was derived for the graphite plates employed and was correlated with the thermal conductivity readings. In addition, it is assumed that the platelets in the individual layers are arranged like roofing tiles. It may be inferred from this that heat transport in any layer is favoured in both directions of this plane and hindered perpendicularly to it. Incipient formation of the edge zone and its effects are not considered. On the assumption that the layer has the structure shown in Fig. 6, it may be inferred that, in the x-direction, all three layers contribute to optimum transport, whereas, in the y-direction, only the upper and lower layer are “favourable”. In the z-direction, only the middle layer is optimally aligned. The measurement approves the difference between the z-direction and the x- / y-direction, but also in this case the conductivity in the y-direction is higher than in the x-direction. Maybe this is an effect of irregular orientation in the edge layer.



Method	thermal conductivity of PA6 + 30 Vol.-% Graphite [W/mK]		
Hot-Disk	*integral* 3,5		
Nanoflash	*X-direction* 4,4	*Y-direction* 6,8	*Z-direction* 1,8

Figure 6: Measured thermal conductivity of a PA6-graphite compound

Analysis of the processing behaviour

The processing behaviour was analysed by varying important process parameters, such as injection speed, for a heat sink demonstrator. Not only was the flow and filling behaviour studied, but a comparison was also made between experiment and simulation.

The pictures in Fig. 7 from the filling studies show the effects of the injection speed on multidimensional mouldings. While a fissured flow front is formed at the low injection speed shown left in the picture, an optically more defined front can develop at four-times that injection speed; however, this requires the injection pressure to be 30 - 50% higher.

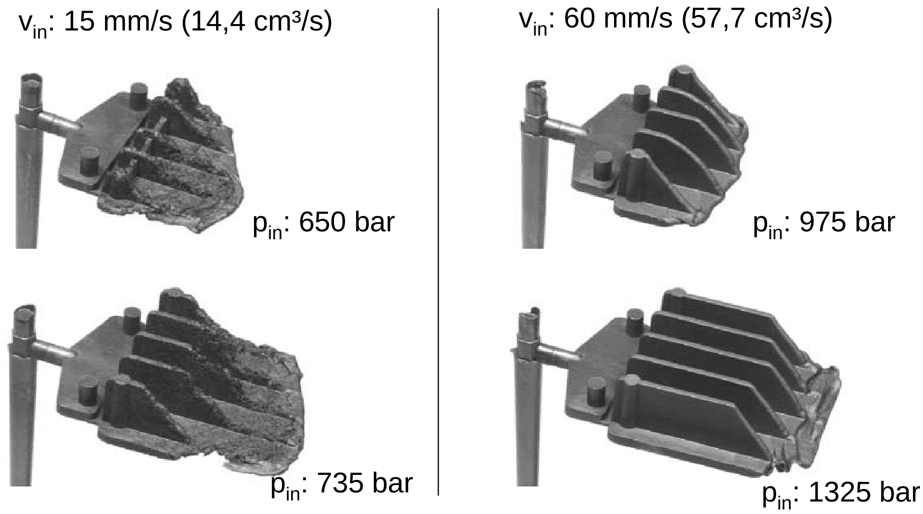


Figure 7: Filling study on the heat sink demonstrator with a PA6-graphite compound (PA6 + 30 vol. % graphite; T_{melt}: 260 °C; T_{mould}: 80 °C)

Comparative filling studies: Experiment versus simulation

The first step in comparing experimental mould filling with simulated filling was to determine and substitute the corresponding indicators into the Mould-Flow program:

- Thermal conductivity λ (T)
- Specific heat capacity c_p (T)
- Transition temperature T_t
- Thermal expansion as PVT behaviour
- Viscosity η

It was established that the experimental results cannot be simulated if isotropic thermal conductivity values are used. As shown in Fig. 8, the simulation results do not show proper mould filling. Under the same processing conditions, the pressure of 1800 bar needed in the simulation deviates significantly from the values of 700-800 bar needed in the experiment. In contrast, anisotropic thermal conductivity values produced very good simulation of experimental filling. Naturally, with regard to heat transfer towards the mould wall, which dominates in practice, the readings obtained perpendicularly to the direction of flow were employed. The necessary filling pressures were shown to be of similar magnitude.

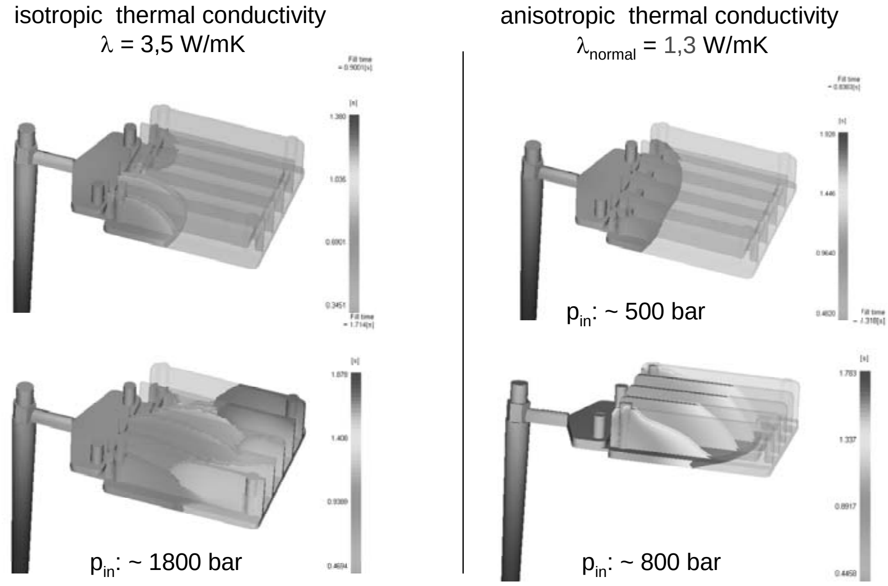


Figure 8: Influence of different thermal conductivities as inputs into simulation calculations (PA6 + 30 vol. % graphite; T_{melt}: 260 °C; T_{mould}: 80 °C)

The use of anisotropic thermal conductivity values as well as the laboriously determined material characteristics of viscosity, heat capacity, etc, allows the influence of the processing parameters to be simulated in addition. By way of example, Fig. 9 shows the ease of simulation at different injection speeds.

For practical purposes, it may be noted that it is certainly possible to use simulation calculations to simulate mould filling, provided that a suitable data set is available and it is implemented and used in the program. This can be used in the development and design of parts moulded from thermally conducting polymers.

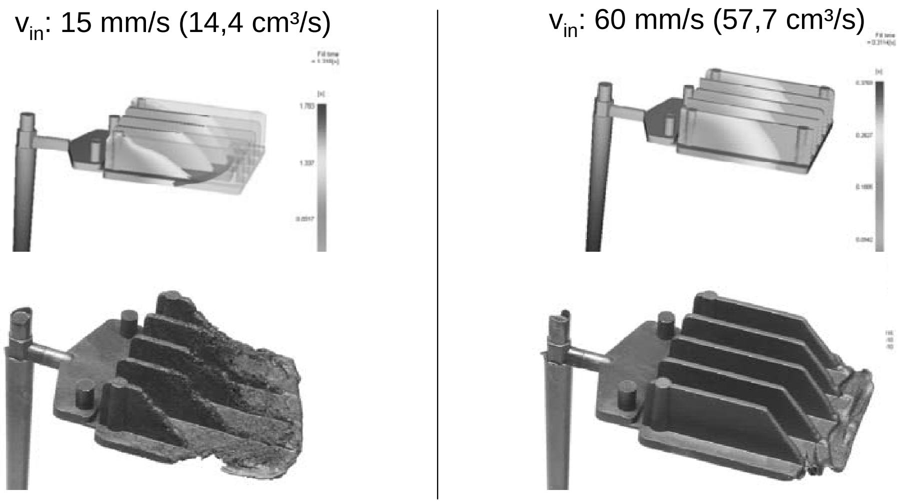


Figure 9: Influence of different thermal conductivities as inputs into simulation calculations (PA6 + 30 vol. % graphite; T_{melt}: 260 °C; T_{mould}: 80 °C)

Conclusion

Production of thermally conducting polymers requires that commodity thermoplastics be modified such that they contain high proportions of highly heat-conducting fillers. Since the high filler content modifies the flow and cooling conditions during processing, the generally established approaches to mould design cannot be used. In view of the rising numbers of inquiries as to the possibility of using such modified, thermally conducting polymers in the field of thermal management, e.g. complex electronic modules, it is necessary for design and processing guidelines to be determined for these new materials.

The theoretical analysis of the cooling behaviour of commodity thermoplastics and thermally conducting thermoplastics first of all underlines the fact that the filling times can be markedly reduced for highly heat-conducting polymers. Both the characteristic values of the polymers and the differences in the thermal characteristics of different tool steels must be taken into account.

A key insight arising from the processing of thermally conducting polymers was that the injection speed is particularly important for the pressure demand and the attainable flow distances. Comparisons between simulations and filling studies show that reliable simulation of the filling process with appropriate simulation software is only possible with suitable characteristic data, which are laborious to obtain. The cause of this is primarily the fact that thermal conductivity is anisotropic, especially in the case of fillers with a pronounced L/D ratio. A first material model for the unfolding three-dimensional filler orientation was derived.

Simulation thus offers developers and technical designers of injection moulded parts the possibility of arriving rapidly at a design which lends itself to the material and production, even where thermally conducting polymers are used.

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