MATERIAL PROPERTIES OF THERMAL CONDUCTIVE THERMOPLASTICS FOR MOLD DESIGN AND PROCESSING STRATEGIES

Abstract

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 - 20 W/mK. The filler-content and the high thermal conductivity affect directly the flow- and coolingconditions during injection molding. Therefore, the manufacture of injection-molded parts requires adjusted processing strategies. In this paper properties of thermal conductive thermoplastics relevant to the design of injectionmolded parts as well as the effect to relevant processing parameters are introduced.

Introduction / Motivation

Modern composite matrials have substantial advantages concerning material specific properties [1,2]. New mechatronic applications result from compounding simple plastics, often thermoplastics, with metallic, ceramic or organic fillers to functional compounds, e.g. multifunctional housings. In spite of the high amout of filler content up to 65 Vol.-%, the molding is still possible using standard injection molding machines. Additional functions can be integrated into the plastic material, depending on the filler:

- Improved thermal conductivity and/or
- · Specific electrical conductivity or
- magnetic functions.

Changes to different physical parameters, as the thermal conductivity, heat capacity and viscosity, which influence next to the wanted change of the part functionality also the processability of such materials significantly, are due to the high filler content [1].

The knowledge of these parameters is very important for the practical part- and mold-design, e.g. also for simulation software, so that the thermal and thermodynamic processes during flow and cooling in the mold of the injection molding process can be better described.

Basics

The substantial mechanism for the heat transfer inside compact solids is the heat conductivity in the solid or in the polymer melt, respectively, as well as the heat transition to the surrounding media. The thermal conductivity can be handled on two different ways: first, absolutly phenomecally and second atomically because of the scattering of acustical phonons at the atoms and molecules of the concerning materials and among each other [1,2]. The first mentioned approach leads over an experience depending definition of the thermal conductivity to the common differential equation of thermal conductivity.

The determination of the thermal conductivity in stationary, one-dimensional systems, for which the heat flow through the specimen is constant over time, is directly possible applying energy equation. For this purpose the so called plate-process after DIN 52612, which is connected to numerous testing constructions in the field of isolating technology, is practically used for large area and surface specimen. Concerning the describtion or characterisation of building materials and components in the field of environmental engineering, the so called k-value is often specified, equation 1. This value is related to the quotient of the thermal conductivity λ and the part thickness d. Therefore, the classification of a material by the k-value includes the specific dimensions [1,3,4].

$$k - value = \frac{\lambda}{d} \tag{1}$$

The thermal conductivity *a* is a physical parameter, which describes how fast a material is reacting on temperature changes from the surrounding environment. This term is often described as thermal diffusivity or temperature conductance. A correlation between temperature and thermal conductivity exsits over the density ρ and the already discussed heat capacity c_p , equation 2 [1,2,4].

$$\lambda = a \cdot \rho \cdot c_n \tag{2}$$

Important for the evaluation of the injection molding process and its modelling, e.g. FE-simulation, is the dependance of temperature and pressure on the parameters [5-7].

Experiments

In order to evaluate the potential of thermalconductive thermoplastics, the properties relevant for the part functionality were measured on injection molded parts. Furthermore, the processability, e.g. flow and cooling conditions, of thermal-conductive thermoplastics were investigated with regard to the possibilities in part design and processing with such materials.

Compounds with polyamid 6 as matrix and aluminumoxid, strontiumferrit and copperplates as ceramic and metallic fillers, respectively, were used in a filler content range of 5 to 60 Vol.-% (Table1). The compounds were produced on a twin-srew extruder (Leistritz ZSE 27 HP 40 D). The evaluation of the thermal or thermodynamic and rheological parameters were done through a temperature dependent characterisation of the produced compounds. The investigations were done in the solid state as well as in the molten state of the compounds. Instationary heatingwire methods (Hot-Disk, SWO) were applied to measure the thermal conductivity. The analysis of the rheological behavior of the different compounds were carried out through viscosity measurements in a high-pressure capillary rheometer at different temperatures and various noozle geometries. As a further important property for the methodical design of injection molded parts, the correlation between the expansion behavior or the specific volume, the pressure and temperature was analysed through pVTmeasurements.

During the second part of the investigations, injection molding tests were done using a standard injection molding machine (Demag ergotech 250-80) at different processing paramters, whereas characteristical processing paramters were recorded. A simple plate with the geometry of 50 x 50 x 2 mm was used as a specimen (Table 1). The plates were injection molded in a modular mold with a 1 mm film gate. Next to the machine paramters, the internal mold pressure inside the cavity was recorded at the beginning of the flow (near the gate) and at the end of flow (afar the gate). The pressure conditions during injection molding were analysed with this data.

Results

As a requirement for the production of injection molded parts made of thermal-conductive thermoplastics, the properties for the evaluation of the flow and cooling behavior during processing are discussed as follows. Furthermore, the results of study of the filling behavior as well as of the cooling process in the real process in dependence of material and processing are presented.

Thermal Conductivity and Heat Capacity

The comparison of the thermal conductivities of the used compounds measured with the Hot-Disk-test shows that the thermal conductivity improves significantly adding well thermal conductive thermoplastics (Fig.1). The dependence of the type of filler is small in the range of low filler contents. In this case, esspecially the polymer properties seem to be the main factor. In contrast to this, the well conductive fillers as copper show a stronger increase of the thermal conducticity than fillers with lower selfconductivity for compounds with high filler contents.

The results of the thermal-conductivity measurements starting in the molten state and ending in the solid state (cooling curve) are presented for different filler contents in Fig. 2. Basically, the expected increase of the conductivity at rising filler contents is described for aluminumoxid as well as for the copper-compound, whereas the PA6-coppercompound showed clearly higher thermal-conductivitycoeffitients in the molten state as well as in the solid state. Remarkable is a strong increase of the thermal conductivity at the transition state from liquid to solid. The 40 Vol.-% aluminumoxid filled Polyamid 6 measures from almost 2 W/mK to 3 W/mK an increase of more than 50 %, which has to be considered for the process evaluation, e.g. in simulations. The reason for this increase is the development of cristalline structures, which occur also for unfilled thermoplastics as shown in literature. Due to the high level increase, it seems that this fact can not be the only reason, so that additionally a strong influence of interactions between filler and matrix can be supposed.

The measured heat capacity correlates with the change of the thermal conductivity (Fig.1). A linear increase of the heat capacity can be recognized in the solid state, which is greater than in the molten state. The exothermal melting of the cristalline structures at the transition from solid to liquid can also be detected (Fig.3).

Viscosity

The shear velocity dependent viscosities measured in the high-pressure capillary rheometer show a significant influence of the filler content on the flowability for the aluminumoxid compounds as well as for the copper compounds (Fig.4). In the case of the aluminumoxid-compound with 40 Vol.-% filler content, the run of the viscosity curve for different noozle geometries can be confirmed, so that the flow-behavior can be assumed to be similar as it is for unfilled materials.

The rise of temperature increases the flowability measurably, whereas the effects are clearly lower than the ones for the investigated filler contents (Fig.5).

The measured viscosities of the aluminumoxidcompound with a relevant filler content for technical applications of 40-60 Vol.-% are about comparable with the measured viscosities of the copper-compounds with a filler content of 30-50 Vol.-%.

Filling Analysis for Injection Molding

The filling studies done on a plate specimen show that the flow resistance for a complete filling of the mold cavity increase significantly at a filler content of 40 Vol.-% (Fig.6). For both filler-matrix-systems, the cavity can be filled with almost no flow resistance, if the filler content is below 40 Vol.-%. In other words, the flow distance of 50 mm can be reached. Starting at a filler content of 40 Vol.-%, the injection pressure measures over 900 bar, so that clearly higher injection pressures are required to overcome the required flow resistances.

The comparison of the measured internal moldpressures near the gate with the pressures afar the gate (at the end of flow) show that the copper-compounds with a filler content of 30 and 40 Vol.-% have a high pressure drop along the flow path at the end of the melt injection process. Finally, the measurements of the specific volume show that the low shinkage of the filled materials has to be considered during mold design, whereas the low shrinkage values are advantagous for electronic applications (Fig.7).

Conclusion

The direct removal of lost heat from thermal loaded regions of thermal-conductive modified housings or components becomes more and more important. Different applications as e.g. transformer housings or electronic parts show this development. The potential of thermalconductive thermoplastics depends apparently on the processability in rational processes like injection molding and on the finally existing part properties.

The addition of ceramic and metallic fillers with a filler content of 30-60 Vol.-% enables the increase of the thermal conductivity into technically interesting ranges of 1 W/mK and clearly higher. The flow and cooling conditions are decisive for the evaluation of the processing and the part design.

These investigations could show reasonable dependancies of the filler content and the physical state during processing on the flow and cooling conditions of relevant material properties, as e.g. thermal conductivity and viscosity. Concerning calculating models or part- and mold-design, the temperature dependence of the thermal conductivity,

Filler Content Matrix Filler [Vol.-%] Polyamid 6 Aluminumoxid 5, 20. 40, 60 (ALCOA CL 4400) (Ultramid B3) Polyamid 6 Copper 10, 30, 50 (Cubrotec 5000) (Ultramid B3) Polyamid 6 Strontiumferrit 5, 10, 20, 30, 40, 50, 60 (Ultramid B3S)

near the gate





Fig. 1: Thermal conductivity and heat capacity of the compounds depending on filler content at room temperature.

esspecially the difference between solid and liquid sate, is of essential importance.

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Fig. 2: Temperature-dependent thermal conductivity of different matrix-filler-systems. left: Polyamid6-Copper-Compounds right: Polyamid6-Aluminumoxid-Compounds



 Fig. 3:
 Temperature-dependent heat capacity of different matrix-filler-systems left:
 Polyamid6-Copper-Compounds

 right:
 Polyamid6-Aluminumoxid-Compounds



Fig. 4: Viscosity curves at elevated shear rates depending on the filler content. left: Polyamid6-Aluminumoxid-Compounds and pure Polyamid6 right: Polyamid6-Copper-Compounds and pure Polyamid6



Fig. 5:Viscosity curves at elevated shear rates depending on the temperature.
left:
right:Polyamid6-Aluminumoxid-Compound with 40 Vol.%-Aluminumoxid and pure Polyamid6
Polyamid6-Copper-Compound with 30 Vol.-% Copper and pure Polyamid6



 Fig. 6:
 Reached flow length with injection molding during injection with limited injection pressure maximum.

 left:
 Polyamid6-Copper-Compounds

 right:
 Polyamid6-Aluminumoxid-Compounds



Fig. 7: Specific volume depending on temperature and pressure for Polyamid6-Aluminumoxid-Compounds.

Key Words

Thermal conductivity; highly filled plastics; mold design