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Thermal conductivity of metal powder-polymer feedstock for powder injection moulding

L. KOWALSKI, J. DUSZCZYK, L. KATGERMAN
Laboratory for Materials Science, Delft University of Technology, Rotterdamseweg 137, 2628 AL Delft, The Netherlands

Thermal conductivity of a powder injection moulding feedstock (mixture of metal powders and polymers) in solid and molten states has been measured by using the laser flash method. The filler material was 316L stainless steel powder and its content in the mixture amounted 60% by volume. An attempt has been made to employ two most promising existing mathematical models (theoretical Maxwell- and semi-theoretical Lewis & Nielsen model) to calculate the thermal conductivity of the mixture. Comparison of the experimental and calculated results has revealed that the Lewis & Nielsen model predicts better than Maxwell model the thermal conductivity of the feedstock. As the difference between the calculated (Maxwell model) and the measured results amounts to 15–85%, it is suggested that it can only be used for preliminary assessment of the thermal conductivity of so highly filled composite material. If accurate thermal conductivity data are required (as in case of numerical simulation of the powder injection moulding process), measurement of this property has to be performed if meaningful simulation results are to be expected.

1. Introduction

Injection moulding of plastics is a widely known process for the production of many engineering products and consumer goods. Injection moulding of polymers filled with dispersed metallic or ceramic particles is a recognized method for improving the mechanical-, physical- or thermal properties of the final moulded part. Through maximising the content of solid particles this process has evolved into the powder injection moulding (PIM) technique. Primarily due to substantial differences in material properties, PIM is more difficult to control and optimize. It is also inherently more susceptible to defects which can be introduced at the onset and become visible only at the later stages of the process. Therefore there is a growing interest in numerical simulation of the process to ease the tasks of tool and process engineers. Computer simulation of traditional injection moulding of plastics is already frequently used in the production practice. Some attempts have also been made to simulate injection moulding of metal/ceramic powders [1–2].

In order to perform a reliable simulation, extended input data (required for the numerical model of the process) have to be provided.

Rheological-, p-V-T-, specific heat-, thermal conductivity-, density data (in the solid and in the molten state) are only examples of those required as input for the simulation. Up till now such data do not exist (or are widely scattered) in the open literature. Some of the properties like thermal conductivity of the PIM feedstock are difficult to measure and are rare in the literature.

The present paper presents some of the results related to an investigation of the thermal conductivity of the commercially available 316L BASF powder injection moulding feedstock.

2. Results and discussion

Powder injection moulding feedstock is a mixture of a metal/ceramic powder and a binder system. BASF has developed and commercialized a ready-to-mould PIM feedstock based on polyacetal (POM) resin. The available metal feedstocks have a solid loading ranging between 57–64 vol %. The major goal of the binder system is to provide the necessary flowability and enable filling of the cavity during the injection moulding process. The binder is subsequently removed from the mixture during the debinding- and the first stage of the sintering processes. The volumetric percentage of powder in the mixture is termed “solid loading” of the feedstock. The value of the solid loading has large effects on virtually all properties of the feedstock (among others on the thermal conductivity).

Thermal conductivity is one of the very important properties of the material to be injected and one of the input parameters for numerical simulation of the PIM process. It determines the heat dissipation rate during the whole injection moulding process. Hence, thermal conductivity is an important heat transport property from both the theoretical and practical points of view. Thermal conductivity of the PIM feedstock is a parameter that does not easily lend itself to measurement. Therefore it is frequently suggested to use the

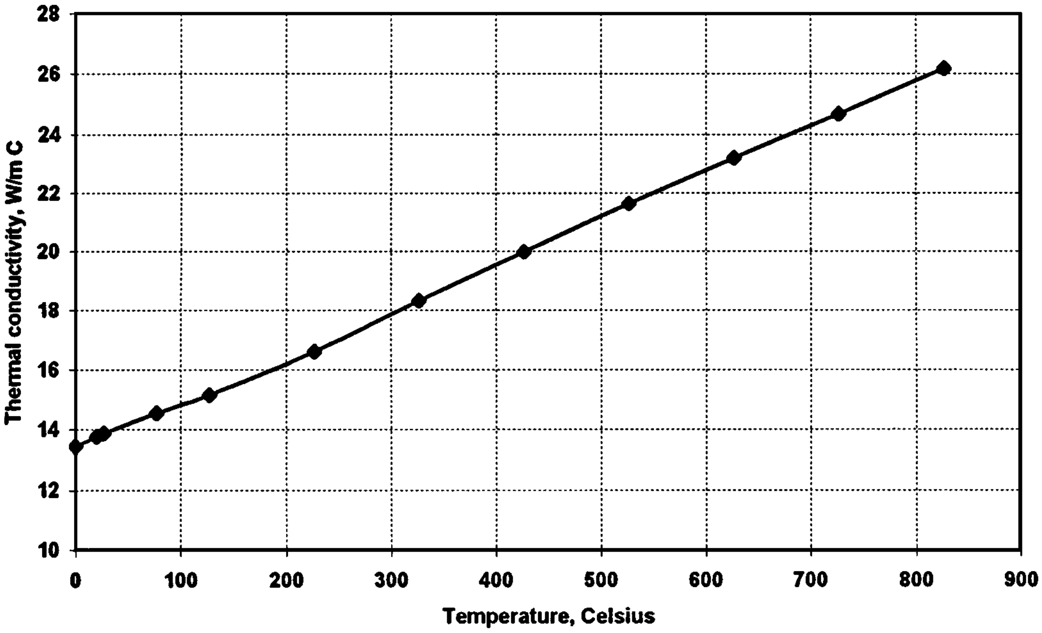

Figure 1 Thermal conductivity of the 316L steel [6].

rule of mixtures (ROM) theory to calculate the thermal conductivity of the plastic/metal- or ceramic powders mixtures [3]. According to this rule, the thermal conductivity of PIM feedstock depends only on the volume content and the inherent properties of the constituents.

There are different mathematical forms (from simple ones to more complex) of the "rule of mixtures" theory. In the present research, the theoretical Maxwell model [4] and the semitheoretical Lewis & Nielsen model [5] have been employed. The Maxwell model is believed to describe well the thermal conductivity of a composite comprising high conductivity spheres in a low conductivity matrix. The conductivity of such a composite is given by:

$$k = k_{\mathrm{m}}\left[\frac{k_{\mathrm{f}} + 2k_{\mathrm{m}} + 2V_{\mathrm{f}}\,(k_{\mathrm{f}} - k_{\mathrm{m}})}{k_{\mathrm{f}} + 2k_{\mathrm{m}} - V_{\mathrm{f}}\,(k_{\mathrm{f}} - k_{\mathrm{m}})}\right]$$

where "k_{f}" and "k_{m}" are the filler (316L powder) and matrix (POM based binder) thermal conductivities, respectively, and "V_{f}" is the filler volume fraction.

The Lewis & Nielsen model includes the effect of the shape of the particles and the orientation or type of packing for a two-phase composite system. The thermal conductivity of a composite is described by following formula:

$$k = k_{\mathrm{m}}\left[\frac{1 + ABV_{\mathrm{f}}}{1 - BV_{\mathrm{f}}\Phi}\right]$$

where

$$A = k_{\mathrm{E}} - 1$$

$$B = \frac{k_{\mathrm{f}}/k_{\mathrm{m}} - 1}{k_{\mathrm{f}}/k_{\mathrm{m}} + A}$$

$$\Phi = 1 + \left[\frac{1 - \Phi_{\mathrm{m}}}{\Phi_{\mathrm{m}}^2}\right]V_{\mathrm{f}}$$

"A" is related to the generalized Einstein coefficient "k_{E}" and includes the effect of the shape of the filler, "B" is related to the thermal conductivities of the components and "Φ_{m}" is the parameter including the effect of the maximum packing fraction of the filler. Values of "A" and "Φ_{m}" for different geometric shapes of the filler and type of packing are given in [5].

In the present research values of $A = 1.5$ (spherical shape of the filler), and $\Phi_{\mathrm{m}} = 0.65$ (random close packing of spheres) were used for calculation of the thermal conductivity of the feedstock.

Figs 1 and 2 present graphically the thermal conductivity as a function of temperature for the major constitutents of the PIM feedstock – 316L stainless steel [6] and binder system, respectively. Fig. 3 shows the calculated (Maxwell- and Lewis & Nielsen models) thermal conductivity of the 316L PIM feedstock. All calculations are based on 60 vol % powder loading. It is to be noted that the shape of the curves for the PIM feedstock is dominated by the thermal conductivity of the matrix material. The calculated values are based on the thermal conductivity of massive 316L steel as no results for powders could be obtained.

Measurements of thermal conductivity of the 316L PIM feedstock have been performed during a cooling scan using the laser flash method. The LFA 427 NETZSCH apparatus was employed for the measurements. It is estimated that the thermal conductivity data obtained are accurate to within 3–4% and 7–8% in the solid and molten states, respectively.

The laser flash (heat impulse) method is based on applying a high intensity and short duration heat pulse to one face of a parallel sided test piece and monitoring the temperature rise at the opposite face as a function of time. The thermal diffusivity "α" of the sample is then calculated according to the formula:

$$\alpha = \frac{1.37L^2}{\pi^2 t_{0.5}}$$

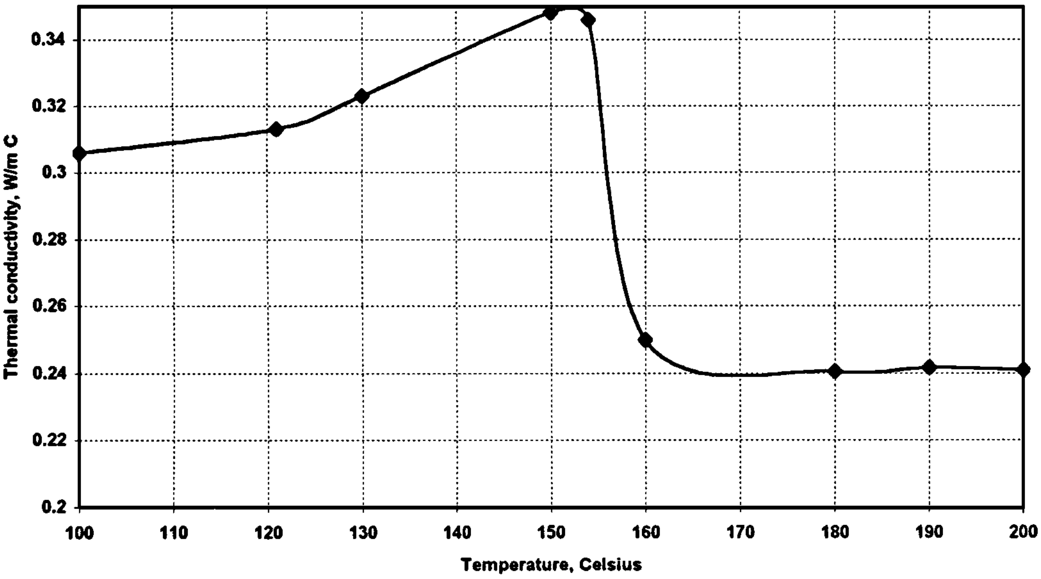

Figure 2 Thermal conductivity of the POM based binder system.

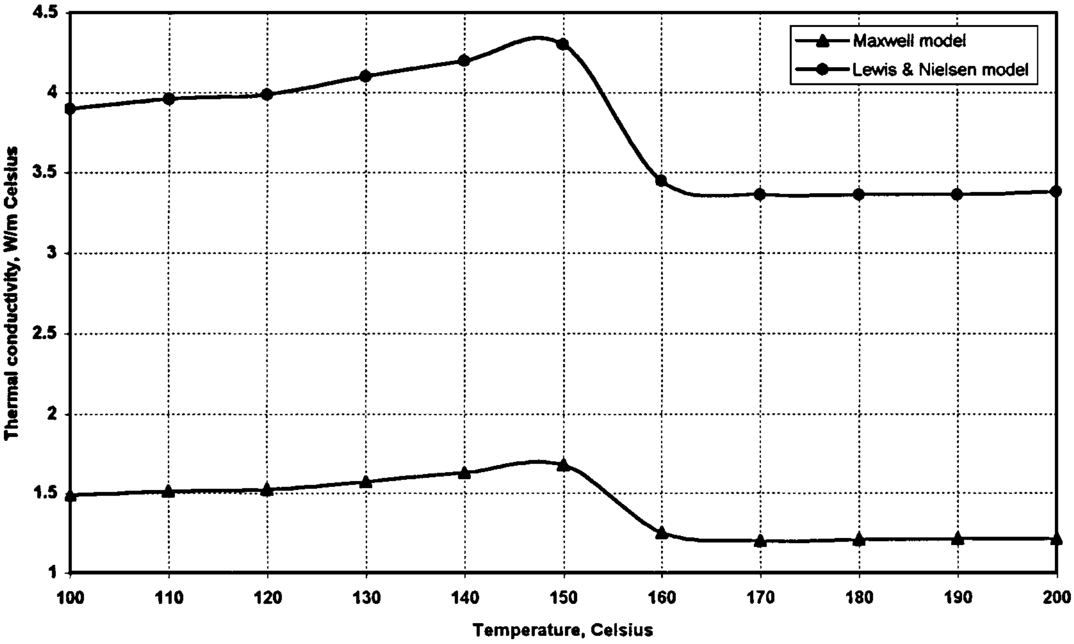

Figure 3 Calculated (Maxwell- and Lewis & Nielsen models) thermal conductivity of the 316L BASF feedstock – powder loading 60%.

where "L" is thickness of the specimen and "$t_{0.5}$" is the time from initiation of the pulse until the rear face of the test sample reaches one half of its maximum temperature.

The thermal conductivity "k" was thereafter calculated using the following relationship:

$$k = \rho C_\mathrm{p} \alpha$$

where "ρ" and "C_p" are the experimentally determined density and specific heat of the feedstock's sample.

The results from the experiments are presented in Fig. 4. It is to be seen that the measured values increase monotonically with decreasing temperature. The most distinct change of the thermal conductivity takes place in the 150–130 °C temperature range. As indicated in Fig. 3, both models used during calculation also predicted the rapid change of the thermal conductivity on cooling from the molten state. In this case however, the biggest change takes place in the region 160–150 °C and is purely related to the change of the thermal conductivity of the POM based binder system.

Comparison between experimental and calculated results indicates that the Maxwell model underestimates the measured values. Bearing in mind that during calculation, thermal conductivity values of the massive 316L steel were used (higher than the thermal conductivity of the powder), it seems that the Maxwell model substantially misjudges the thermal conductivity of the feedstock. In contrast, the Lewis & Nielsen model overestimates the measured values. It is believed, however, that part of the "overestimation" is related to the values

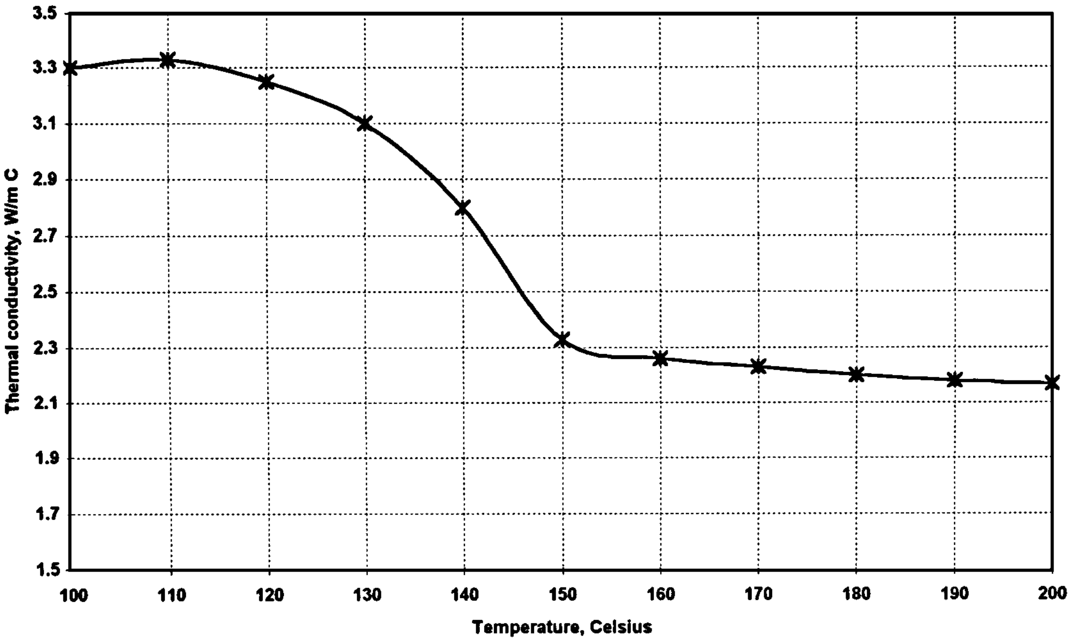

Figure 4 The measured thermal conductivity values of the BASF 316L powder injection moulding feedstock (over the processing temperature range).

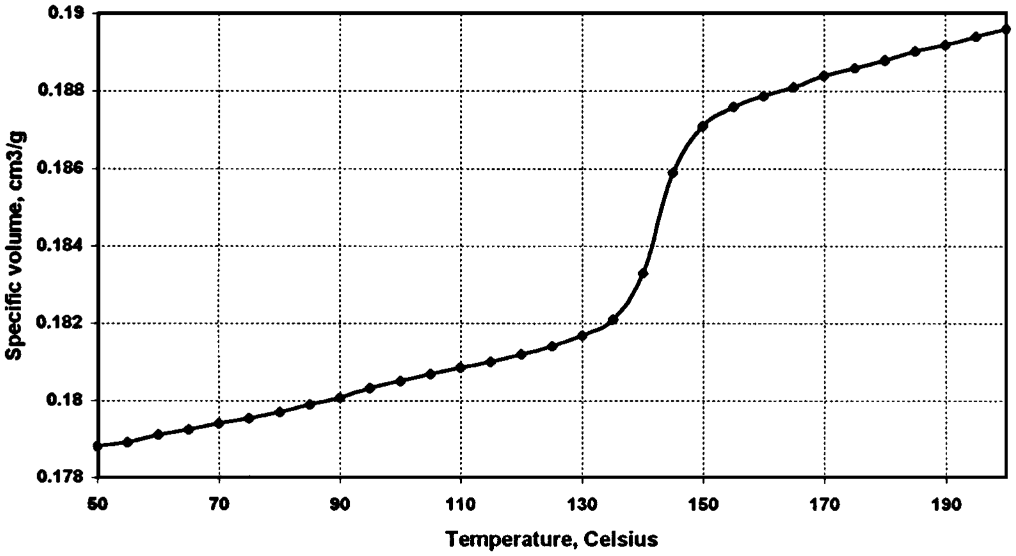

Figure 5 The measured specific volume change as a function of temperature for the 316L BASF feedstock – pressure 1bar.

of the thermal conductivity of the 316L steel. Therefore this model seems to be best suited for prediction of the thermal conductivity of the PIM feedstocks.

It is to be emphasized that no data related to the verification of the models employed for such high concentration of the filler ($V_f = 0.6$) were found in the literature. It is hypothesized that the change of the thermal conductivity of the PIM feedstock is related to two phenomena:

- change of the thermal conductivity of the components with temperature, and
- change of the distance between filler particles with temperature.

The experimental results presented in Fig. 4 show that the biggest increase of the thermal conductivity takes place in the temperature range 150–130 °C. This phenomenon corresponds well with the measured change of the specific volume of the feedstock (Fig. 5). The main effect, however, seems to be related to the change of the thermal conductivity of the matrix material (caused by the phase change). The other effects (of density change) are additional and small. The density (volume) change is only about 5%. The change of thickness of the matrix layers between particles is about 1.7%, which can not explain a conductivity change of 50%.

3. Conclusions

The simple rule of mixtures (series model) should not be used for the prediction of the thermal conductivity of PIM feedstocks – calculated results very much overestimate the measured ones. The Maxwell model substantially underestimates the measured values of the thermal conductivity. It seems that the Lewis & Nielsen's semi-theoretical model predicts better the thermal conductivity of the PIM feedstock. The difference however between the measured and the calculated values is still large (15–85%). None of the models fully take into account the phase change in the processing temperature range (specific volume change) of the matrix material and the change of the thermal conductivity of the composite material (feedstock) which is related to this phenomenon.

It is to be emphasized that most theoretical models were until now verified for much smaller filler concentrations (1–30 vol %). It is thus assumed that calculation of the thermal conductivity of the composite can only give reasonable results if a relatively thick layer of a matrix material separates all filler particles from each other. In the case of highly filled PIM feedstocks, it can not be precluded that the thickness of the matrix layer among some powder particles is close to zero. Therefore, employing of any kind of mathematical models for predicting the thermal conductivity of highly filled polymers can be burdened with substantial errors. If thermal conductivity data are to be used for the purpose of the numerical simulation of the powder injection moulding process, the accuracy of the calculated values is inadequate and experimental measurements of this property must be performed.

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