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Methodology development for through-plane thermal conductivity prediction of composites

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Abstract

The prediction and tailoring of thermal conductivity of two-phase composites is essential. In this work a new semi-empirical model was developed, which was derived from the rule of mixtures. Furthermore, a new methodology was developed to determine the thermal conductivity of the fillers and the maximum achievable filler content. To validate the new model, polypropylene-based composites were prepared with different fillers, such as talc, boron-nitride and graphite, with 20, 40 and 60 vol% filler content. The results obtained from the proposed model are in good agreement with the experimental data. Various other theoretical models were also introduced and compared to the experiments, but in most cases those underestimate or overestimate the thermal conductivity of composites.

Keywords: Effective thermal conductivity; Polymer-matrix composites; Analytical modeling; Particle reinforced polymer; Compression molding; Rule of mixtures;

1. Introduction

It is well-known that polymeric materials have low thermal conductivity ($\lambda=0.1-0.5$ W/mK). Accordingly, foams produced from polymers are widely used by the building industry [1, 2]. However, the thermal conductivity of polymers can be improved significantly by compounding them with conductive fillers, such as aluminum nitride, boron nitride, graphite,

carbon black, copper powder or some other good thermally conductive material [1, 3-8]. Thermally conductive polymer composites have several advantages compared to conventionally used metals, such as low density, good corrosion resistance and low processing costs. These properties generated widespread research and development in this area, and these new materials receive more and more attention in industrial applications [6-10].

To keep the prices of polymer composites as low as possible, it is important that their properties can be tailored to needs. Hence it is important that the composite can be designed using the proper type and ratio of the matrix and fillers [11, 12]. However, not only mechanical, but also thermal properties, such as thermal conductivity should be predictable. The thermal conductivity of composite materials is influenced by several factors, such as filler concentration, particle size and shape, filler dispersion and distribution in the matrix, the thermal conductivity of the components, the contact between the particles and the contact surface resistance between the matrix and the filler [1, 11]. Although numerous empirical, semi-empirical and theoretical models have been developed for the prediction of thermal conductivity of two- or multiphase polymer composites, its reliable and precise prediction still remains a challenge. The three basic models are the rule of mixtures (parallel model), the inverse rule of mixtures (series model) and the geometric mean model. In the rule of mixtures it is assumed that the components contribute to the thermal conductivity of the composite proportionally. It generally overestimates the experimental values and provides an upper bound for conductivity. This model assumes the existence of a percolation network of the filler in the matrix and perfect contact between the filler particles. On the other hand, the inverse rule of mixtures assumes that there is no contact between the particles, thus it underestimates the experimental values and provides a lower bound for conductivity. The geometric mean model (Equation (1)) is an empirical method for the prediction of the thermal conductivity of composites. It provides better results than the rule of mixtures and inverse rule of mixtures [1, 13, 14].

$$\lambda_c = \lambda_f^\varphi \cdot \lambda_m^{(1-\varphi)} \quad (1)$$

Besides these basic models, many advanced models have been developed. The most important theoretical equations are the Maxwell [15], Bruggeman [13], Cheng-Vachon [16], Hamilton-Crosser [17] and Meredith-Tobias [18] model. On the other hand, there are numerous second-order empirical and semi-empirical models that contain experimental factors for thermal conductivity and for the volume fraction of the components. Agari and Uno [19] and Lewis

and Nielsen [20] developed such models, for example. These models also show good correlation with the experiments up to 30 vol% filler content. Only the Lewis-Nielsen model gives a better fit above 30 vol%, thanks to the introduction of the maximum volume fraction of fillers in the equation [1, 13].

One of the theoretical models for the calculation of the thermal conductivity of two-phased systems is the Maxwell model (Equation (2)). Maxwell supposed that spherical filler particles are randomly distributed in the matrix and there is no interaction between them. This model describes the thermal conductivity of composites with a low volume fraction of fillers well, but as filler content is increased, the particles start to develop interactions between each other and form conductive chains [13, 15, 21, 22].

$$\lambda_c = \lambda_m \cdot \frac{[2 \cdot \lambda_m + \lambda_f + 2 \cdot \varphi \cdot (\lambda_f - \lambda_m)]}{[2 \cdot \lambda_m + \lambda_f - \varphi \cdot (\lambda_f - \lambda_m)]}, \quad (2)$$

where λ_c is the thermal conductivity of the composite, λ_m and λ_f are the thermal conductivity of the matrix and the filler, and φ is the filler fraction. Bruggeman developed another theoretical model. This implicit relation (Equation (3)) also supposes that the spherical, non-interacting particles are homogeneously dispersed in the continuous matrix [11, 13, 21, 22].

$$1 - \varphi = \frac{(\lambda_f - \lambda_c) \cdot (\lambda_m - \lambda_c)^{1/3}}{(\lambda_f - \lambda_m)}. \quad (3)$$

Cheng and Vachon developed another theoretical model (Equation (4) and (5)) for two-phase composite materials. This equation assumes that the discontinuous phase has a normal distribution in the continuous matrix. The parabolic distribution constant was introduced and related to the volume fraction of the filler [13, 16, 23, 24].

$$\frac{1}{\lambda_c} = \frac{1}{\sqrt{C_{cv}(\lambda_m - \lambda_f) \cdot (\lambda_m + B_{cv}(\lambda_f - \lambda_m))}} \cdot \ln \frac{\sqrt{\lambda_m + B_{cv}(\lambda_f - \lambda_m)} + B_{cv}/2 \cdot \sqrt{C_{cv}(\lambda_m - \lambda_f)}}{\sqrt{\lambda_m + B_{cv}(\lambda_f - \lambda_m)} - B_{cv}/2 \cdot \sqrt{C_{cv}(\lambda_m - \lambda_f)}} + \frac{1 - B_{cv}}{\lambda_m}, \quad (4)$$

$$B_{cv} = \sqrt{\frac{3 \cdot \varphi}{2}}, \quad C_{cv} = -4 \cdot \sqrt{\frac{2}{3 \cdot \varphi}}, \quad (5)$$

From the literature survey it is obvious that the exact prediction of thermal conductivity for highly filled composites still poses difficulties. The theoretical models often underestimate the results and can be used only up to 30 vol% filler content [2, 21, 22, 25-27]. The semi-

empirical models give better correlation with the experiments, but they need more experimental parameters. Our goal was to establish a mathematical model that can predict the thermal conductivity of composites up to the maximum achievable filler content. In the literature various thermal conductivity values can be found for different fillers. To characterize the thermal conductivity and the maximum packing of these fillers a new methodology was developed.

2. Materials and methods

2.1 Materials

H145 F homo-polypropylene was purchased from Tisza Chemical Group Public Limited Company (Hungary). As fillers to enhance thermal conductivity talc, hexagonal boron-nitride (BN) and graphite were used. The properties of the fillers are summarized in Table 1.

2.2 The preparation of the samples

PP matrix compounds were prepared with 20, 40, and 60 vol% fillers. The components were mixed at 230°C for 20 minutes at a rotational speed of 25 1/min to achieve the desired homogeneity. Then 10 mm thick cylindrical specimens of a diameter of 30 mm were compression molded from the mixture (Collin Teach-Line Platen Press 200E) at 230°C for 15 minutes. Five specimens were produced from each series for the thermal conductivity measurements.

2.3 The measurement of thermal conductivity

A thermal conductivity meter was designed and built, based on the comparative longitudinal heat flow method [29, 30]. In this method the unknown sample is compressed between the known reference samples and heat flux passes through the measurement unit as a temperature difference is created between the two sides of the unit. The thermal conductivities of the sample and the reference sample are inversely proportional to their thermal gradients. The apparatus developed (Fig. 1) contains two C10 steel (55 W/mK) cylinders with a diameter of 30 mm; and a length of 30 mm. A specimen of a diameter of 30 mm and a thickness of 10 mm is placed between the steel cylinders. On the contact surface thermal grease was applied to decrease heat resistance. 3 thermocouples were inserted in each cylinder (with thermal grease) to detect temperature: one 3 mm below the top, one in the middle and one 3 mm above the bottom ($T_{m1,2,3,4,5,6}$ /Fig. 1).

The temperatures were registered with an Ahlborn Almemo 8990-6-V5 data acquisition module, whose resolution is 0.1°C. NiCr-Ni T190-0 type of thermowires (Ahlborn) were used

for the measurements. The diameter of a single wire was 0.5 mm, and the external diameter of the glass fiber coated thermocouple was 1.3 mm. The apparatus was clamped and the temperature difference maintained with a hot press (Collin Teach-Line Platen Press 200E). The assembled unit was insulated with 25 mm thick polyurethane foam hence the radial heat loss can be neglected. When the steady state is reached, the temperature slope is linear along the reference sample and the specimen thickness. Surface temperatures ($T_{1,2,3,4}$ /Fig. 1) can be calculated by extrapolation from the measured temperatures. Knowing the thermal conductivity of steel and the temperature difference between the surfaces, the heat flux of the hot and cold sides can be calculated with Fourier's law. From the average of the heat fluxes the thermal conductivity coefficient of the samples (λ_c) can be calculated with Equation (6).

$$\lambda_c = \lambda_r \frac{A_r \cdot \frac{1}{n} \sum_{i=1}^n \frac{\Delta T_i}{x_i}}{\frac{A_c}{x_c} \cdot \Delta T_c}, \quad (6)$$

where λ_r is the thermal conductivity and A_r is the cross-section of the reference steel cylinder, x_i is the distance between the sensors, ΔT_i is the temperature differences measured by the sensors, A_c and x_c are the cross-section and the thickness of the sample, and ΔT_c is the temperature drop on the sample. The temperature difference between the hot and cold sides was 30°C. The cold side was 50°C and hot side was 80°C, meaning that the average temperature was 65°C. This big difference in temperature was necessary to achieve a more precise result, because the thermal conductivity of the reference sample is significantly higher than the thermal conductivity of the sample. According to the tests the uncertainty of the thermal conductivity measurement was lower than 5% at any case.

3. The development of the new method

3.1 A novel thermal conductivity model

A new model, based on the rule of mixtures, was developed to predict the thermal conductivity of particle-filled composites. The relationship between the thermal conductivity and the filler fraction of the composites is non-linear, thus the rule of mixtures gives the upper limit. The following assumptions were made:

- the filler content varies between 0 and the maximum theoretically possible filler fraction (φ_{\max}),

- the thermal conductivity (TC) of the composite varies between the TC of the matrix and the bulk TC of the filler,
- the relationship between filler content and thermal conductivity is non-linear.

The rule of mixtures was modified according to these assumptions, yielding Equation (7).

$$\lambda_c = \lambda'_m \cdot \left[1 - \left(\frac{\varphi}{\varphi_{\max}} \right)^C \right] + \lambda'_f \cdot \left(\frac{\varphi}{\varphi_{\max}} \right)^C ; (0 \leq \varphi \leq \varphi_{\max}), \quad (7)$$

where λ_c is the thermal conductivity of the composite, λ'_m and λ'_f are the effective thermal conductivities of the matrix and the filler, φ and φ_{\max} are filler content and the maximum achievable filler content, and C is a constant describing the conductive chain formation capability and shape factor of the material. The model and its dependence on factor C are depicted in Fig 2. The dependence of the matrix $[1 - (\varphi/\varphi_{\max})^C]$ and filler $[(\varphi/\varphi_{\max})^C]$ thermal conductivity factor on exponent C is plotted in Fig 3. When the filler fraction is 0 vol%, the thermal conductivity is determined by the TC of matrix. Increasing the filler content, the effect of the filler will be more dominant. When the filler fraction is 100 vol%, the TC of the composite is determined by the filler. By increasing the C constant the effectiveness of the fillers will develop at higher filler fractions.

λ'_m and λ'_f can be determined by direct measurements, or can be calculated from values in the literature with Equation (8) and (9).

$$\lambda'_m = C_m \cdot \lambda_m, \quad (8)$$

$$\lambda'_f = C_f \cdot \lambda_f, \quad (9)$$

where λ_m and λ_f are the thermal conductivities of the matrix and the filler from literature. C_m describes the morphology change in the matrix caused by the cooling rate, the processing technology and the nucleating efficiency of the fillers. C_f depends on the orientation of the filler, the number of contacts between the particles and the thermal contact resistance between them.

3.2 Maximum filler content

The maximum filler content was determined with the previously presented thermal conductivity meter. The powders were filled in a wooden frame and pressed with the steel reference cylinders. The displacement of the press plate was registered as a function of the compaction pressure. From the displacement (l), the inner diameter of the frame (d) and the mass of the measured filler (m) the apparent density (ρ_a) could be calculated (Equation (10)).

The Relative Density (RD) of the filler can be determined as a ratio of the apparent density and the real density of the filler (Equation (11)).

$$\rho_a(p) = \frac{m}{\frac{d^2 \cdot \pi}{4} \cdot l(p)}, \quad (10)$$

$$RD(p) = \frac{\rho_a(p)}{\rho}. \quad (11)$$

In the next step RD was plotted as a function of compaction pressure. To determine its limit, a saturation curve was fitted to the measured points. This saturation curve is based on a sigmoid curve (Equation (12)). In the equation c_1 is the minimum and c_2 is the range of the sigmoid function. The relative density at zero and infinite pressure can be obtained with the help of these constants, as shown in Equation (13) and (14). Combining Equation (12) with Equation (13) and (14), we get Equation (15), the final form of the saturation curve. By fitting the curve to the measured values, the parameters and the maximum of relative density can be determined.

$$RD_p = c_1 + \frac{c_2}{1 + e^{-a \cdot p^b}}, \quad (0 \leq p \leq \infty), \quad (12)$$

$$RD_0 = c_1 + \frac{c_2}{2} = \frac{\rho_a(p=0)}{\rho_{\text{particle}}}, \quad (13)$$

$$RD_\infty = c_1 + c_2, \quad (14)$$

$$RD_p = [2 \cdot RD_0 - RD_\infty] + 2 \cdot \left[\frac{RD_0 - RD_\infty}{1 + e^{-a \cdot p^b}} \right], \quad (0 \leq p \leq \infty). \quad (15)$$

3.3 The thermal conductivity of the filler

It is crucial to determine the exact bulk thermal conductivity values of the fillers for the modeling of the conductivity of the composite material. In the literature only approximate values are available; the TC of fillers depends to a great extent on their purity and structure. Moreover, the bulk thermal conductivity of a filler and the thermal conductivity of a single particle are very different, due to the thermal contact resistance between the particles. To determine the effective TC of the filler, the press was heated up to 50 and 80°C and the temperatures were shown as a function of the compacting pressure during the measurement of maximum filler content. Thermal conductivity was calculated from Fourier's law as a

function of the pressure. A sigmoid curve (Equation (16)) was fitted to the measured points to determine the theoretical maximum thermal conductivity of the compacted fillers.

$$\lambda'(p) = \lambda'_{\infty} \frac{1 - e^{-n \cdot p^m}}{1 + e^{-n \cdot p^m}}, \quad (0 \leq p \leq \infty), \quad (16)$$

where λ'_{∞} is the thermal conductivity at infinite pressure (p) and n and m are data fitting parameters.

4. Verification of the model

The new model and the methodology were verified with three types of fillers: talc, boron nitride and graphite. Consequently, the applicability of the model has been proved for a wider range of TC in the case of plate-like particles.

4.1. Maximum filler fraction

The maximum filler fractions (relative density limit) of talc, boron nitride and graphite were determined with the method developed (Fig. 4). To eliminate the uncertainties originating from the compression and thermal expansion of the reference steel cylinders, a baseline was determined as a function of the pressure and the measurements were compensated with it. The measured values show saturation characteristics as compaction pressure is increased. At zero pressure the relative densities of talc, BN and graphite were 28.1%, 8.6% and 6.8%, respectively. The relative density limits for each material (RD_{∞}) were determined by data fitting. The fitted curves have good correlation with the measurements, the R^2 value is over 0.94 in all cases. The relative density limits of talc, boron-nitride and graphite powder are 89.9%, 85.2% and 96.2% respectively.

4.2. Effective thermal conductivity of fillers

During the compaction of powders the temperatures of the reference cylinders were also registered. After reaching the steady state condition, the thermal conductivities were determined as a function of the compacting pressure (Fig. 5). The effective thermal conductivities of the talc, boron-nitride and graphite powder were determined as 2.2 W/mK, 8.8 W/mK and 20.7 W/mK respectively, based on the fitted sigmoid curves. Although the thermal conductivity of the powders at atmospheric pressure is not zero a simplification can be used in which the initial points of the sigmoid curves forced into the origin. It was proved that this simplification does not cause any significant changes in the thermal conductivity at infinite pressure (Equation (16)) of the fillers (λ'_{∞}).

4.3. The applicability of the model

The measured thermal conductivity of the polypropylene matrix is 0.36 W/mK, which is higher than the 0.11 W/mK [1] value found in the literature. The reason for the difference could be the difference in the preparation method of the samples, in the material composition and morphology and the measurement method. Hence the correction constant of the matrix is $C_{m,PP}=3.27$. The thermal conductivities of the fillers also differ considerably from the values in the literature. The correction constants for the talc, boron nitride and graphite are the following: $C_{f,talc}=0.224$, $C_{f,BN}=0.035$, $C_{f,graphite}=0.052$.

In Fig. 6 the measured thermal conductivities for the matrix, the composites and the fillers are represented in a graph and a non-linear relationship can be observed between thermal conductivity and filler volume fraction. With the newly developed conductivity model, the thermal conductivity of the composite materials can be described well as a function of the filler content; the correlation between the measurements and the model is higher than 0.95. In the models for polypropylene filled with plate-shaped graphite, boron-nitride or talc the chain and shape factors were 1.4 in all cases. It proves that the shape of the fillers determines the chain formation capability and contact between the particles.

4.4 Comparison to other models

Fig. 7 shows the measured thermal conductivities of polypropylene composites filled with talc, boron nitride and graphite. The measured values were compared to theoretical models developed by Maxwell, Bruggeman, Cheng and Vachon, to the geometric mean model and to our new model. In most cases, the theoretical models underestimate the measured values and as filler content increases, the differences also increase. The models show a good fit only in the case of talc up to 20 vol% and the Maxwell model describes the change of thermal conductivity as a function of talc content well. The geometric mean model gives a better estimation than the other models, but above 40 vol% it overestimates the thermal conductivity. Compared to the theoretical models, the newly proposed model gives by far the best fit to the measured values even above 40 vol% of fillers.

5. Conclusion

In this paper a new semi-empirical model was proposed to predict the thermal conductivity of two-phase composite materials. A new methodology was also introduced to determine the thermal conductivity of fillers and the theoretical maximum filler content, which were used as the input parameters of the model. In the first step the maximum filler content was determined

with a modified linear heat flow thermal conductivity meter. The particles were compressed between two steel cylinders, and the relative density of the sample was calculated from the displacement of the cylinders. The relative density was plotted as a function of the compacting pressure and a sigmoid-based saturation curve was fitted to the measurement points to determine the maximum filler content. According to the measurement values and the calculations, the theoretical maximum filler contents for talc, boron-nitride and graphite powder are 89.9%, 85.2% and 96.2% respectively. During compaction thermal conductivity was also shown as a function of compaction pressure. The calculated effective thermal conductivities of talc, boron-nitride and graphite powder are 2.2 W/mK, 8.8 W/mK and 20.7 W/mK respectively. To validate the model, composites were prepared by melt mixing and compression molding. The model was validated up to 60 vol% filler fraction. The thermal conductivities obtained from the proposed model were found to be in good agreement with the experimental results up to the investigated filler fraction. The constant in the model, which describes the conductive chain formation capability and shape of the fillers is 1.4 in all investigated cases. The new model was compared to various theoretical models, such as Maxwell, Bruggeman, Cheng and Vachon and the geometric mean model. It was found that in most cases these theoretical models under- or overestimate the results.

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Figures:

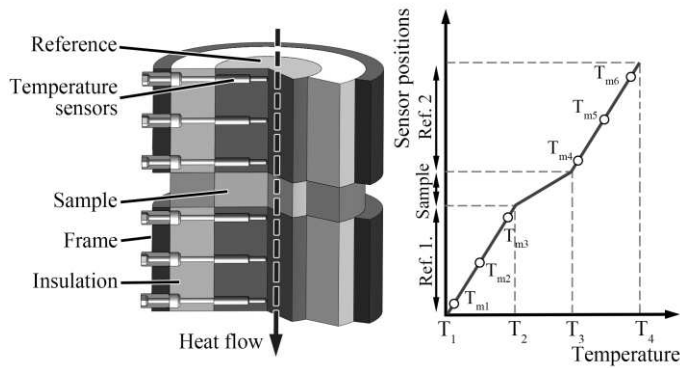


Fig. 1. Longitudinal heat flow measurement unit and its measurement principle

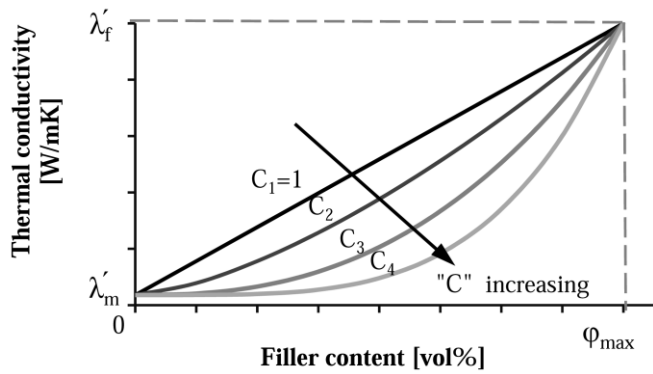


Fig. 2. Theory of the model for the calculation of the thermal conductivity of composites

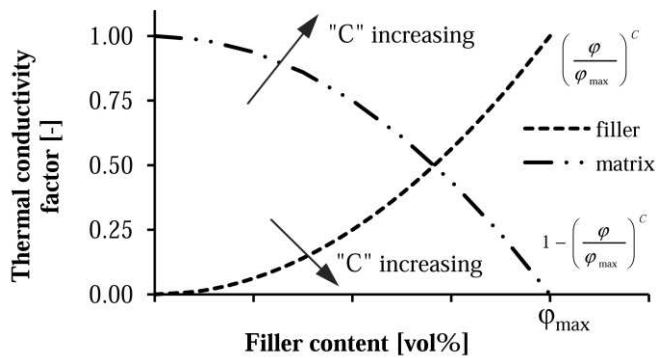


Fig. 3. Interpretation of the factor for the thermal conductivity model

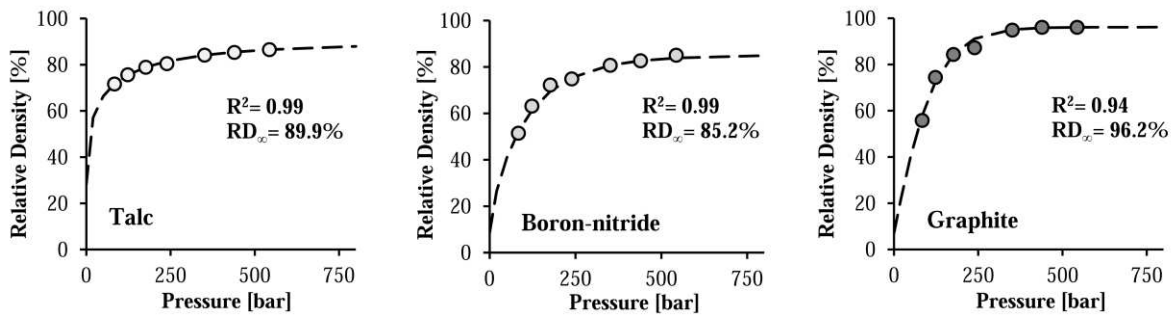


Fig. 4. Maximum relative density of talc, boron-nitride and graphite powders (the dots are the measured points and the dashed lines are the fitted functions)

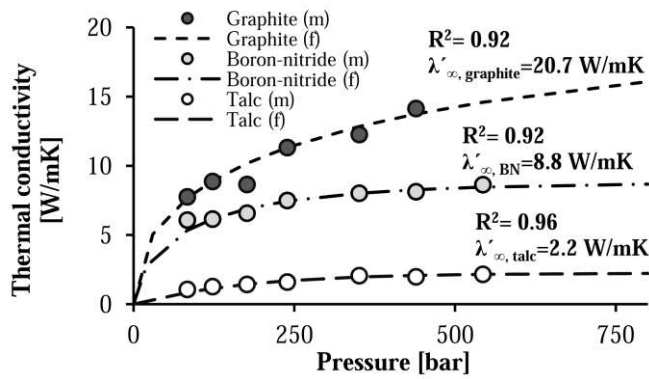


Fig. 5. Bulk thermal conductivity of the talc, boron-nitride and graphite powders (m=measured values, f=fitted values)

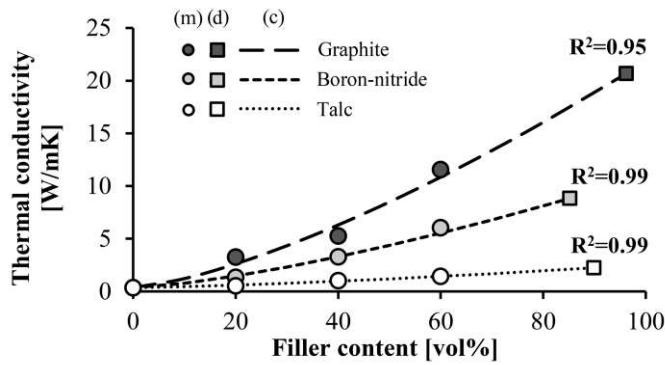


Fig. 6. Thermal conductivities of the compounds (m=measured values, d=derived from the maximum relative density and bulk thermal conductivity curves for the given fillers, c=calculated values with the proposed model)

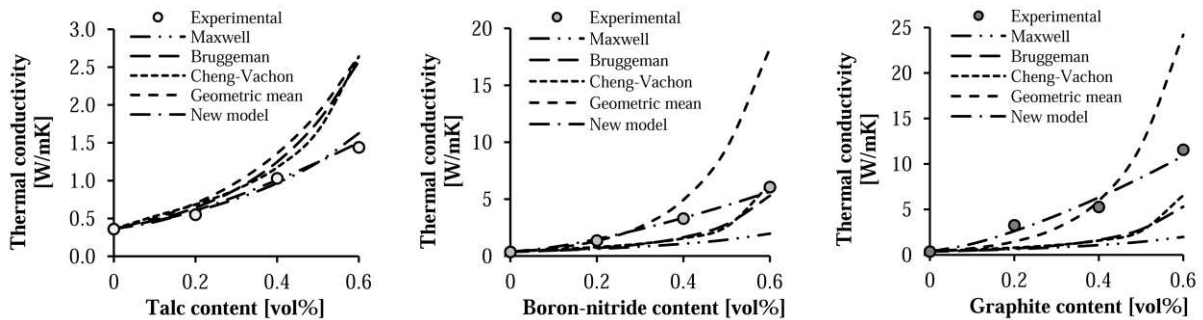


Fig. 7. Comparison of the new model to other theoretical models in the case of talc, boron nitride or graphite filled polypropylene

Table captions:

Name	Trade name	Manufacturer /distributor	Density [g/cm³]	Thermal conductivity (literature values) [W/mK]
Talc	Talc Powder 325 mesh, coating grade	QualChem Zrt.	2.7	2-10 [28]
Boron nitride	HeBoFill 482	Henze BNP GmbH	2.1	250-300 [1]
Graphite	Timrex C-Therm 011	Timcal Ltd./ HSH Chemie Ltd	2.2	100-400 [1]

Table 1. Properties of the investigated fillers