Determination of thermal conductivity of composites with dispersed spherical inclusions

A A Chernykh¹, A I Sharapov¹ and A G Arzamastsev¹

¹Lipetsk State Technical University, Lipetsk, Russia

Abstract. The existing shortcomings of the models for determining the thermal conductivity of composite materials with dispersed spherical inclusions raise the issue of creating more advanced calculation methods. This paper proposes a model for determining the thermal conductivity of composite materials, taking into account influence of thermal resistance at the material-inclusion boundary. The analysis of numerical and theoretical calculations showed that the analytical dependence is consistent with the results of numerical modelling for relatively small values of thermal conductivity of the filler and inclusion diameter. This paper considers several models that have been recently developed and that allow calculating thermal conductivity of such materials. They are compared with the model that takes into account influence of thermal resistance of the phase boundary. Applicability intervals of the considered model for various ratios of the thermal conductivity of inclusion material and matrix material are given.

1. Introduction

For many industries, it is of interest to study materials that contain various kinds of dispersed inclusions. This interest is dictated by a number of issues in the field of determining thermal conductivity and in calculation methods that would take into account all factors: size of dispersion inclusions, their shape, thermal resistance at the boundary of two phases, etc. [1–7]. When determining effective characteristics of composite materials, data of thermophysical measurements or analytical dependencies are usually used. For the first time, analytical formulas for calculating effective thermal conductivity coefficients at various concentrations of inclusions and thermal conductivity ratios were presented by Maxwell [1], Meredith [2–4], Rayleigh [3] and Bruggeman [3]. It is worth noting that Maxwell was the first to give analytical expressions for effective conductivity of the heterogeneous medium in his famous paper on electricity and magnetism [1]. He considered the problem of thermal conductivity of inclusions embedded in a continuous matrix. Maxwell’s model suggests lack of thermal interaction between dispersed inclusions, where thermal interaction between filler particles is neglected.

In recent years, there has been significant progress in the analytical description of such materials [4–11], which made it possible to take into account different forms of inclusions and their orientation. However, it should be noted that any of these formulas uses a relatively crude model of the heterogeneous medium, which affects determination accuracy of effective coefficients. Therefore, it is necessary to propose a model that will take into account the size of dispersion inclusions, their shape, thermal resistance at the boundary of two phases [12,13]. Experimental methods for determining
Effective thermal conductivity coefficients also have limitations [3,4,6,12,14-17] (test sample size, contrast of matrix-inclusion properties, etc.). When heat passes through the interface between composite components, the temperature drop occurs at the interface. This heat flux disturbance can be described by thermal resistance, which consists of: interfacial thermal resistance and thermal boundary resistance, which occurs due to differences in physical properties of constituent materials. If thermal resistance is not taken into account, it is possible to obtain approximate ratios for calculating thermal conductivity of composites [18]. For effective design and use of composites, it is necessary to understand and be able to predict their effective thermal conductivity. Numerous studies have focused on effective properties of composite materials based on numerical modelling [13, 15-17]. Molecular and dynamic modelling was used to predict effective thermal conductivities of composites with nanoscale inhomogeneities. Validation of any given model for calculating the thermal conductivity coefficient is based on experimental data. However, the experiment is an expensive research method. In addition, there is a small number of experiments in the papers on this subject. This is mainly due to the fact that measuring of the model parameters becomes labour- and cost-intensive process at sufficiently high values of thermal conductivity and inclusions’ diameter. The main results on thermotechnical properties of materials can be found, for example, in papers [2,3].

To calculate the thermal conductivity of materials with dispersed inclusions, one should use Maxwell formula [5], which has the form:

$$\frac{\lambda}{\lambda_1} = \frac{2 + \frac{\lambda_2}{\lambda_1} - 2\varphi \left(1 - \frac{\lambda_2}{\lambda_1}\right)}{2 + \frac{\lambda_2}{\lambda_1} - \varphi \left(1 - \frac{\lambda_2}{\lambda_1}\right)},$$

(1)

Where $\lambda_2$ – thermal conductivity coefficient of dispersed inclusions, $\lambda_1$ - matrices, $\varphi$ – volume content of dispersed inclusions.

For spherical inclusions, V. Odelevsky proposed the following formula for calculating the effective coefficient of thermal conductivity:

$$\frac{\lambda}{\lambda_1} = 1 - \frac{\varphi}{1 - \frac{\lambda_2}{\lambda_1} - \frac{3}{3}},$$

(2)

Let us compare the results of all the above dependences with the analytical dependence proposed in this paper, which we will indicate below, in Fig. 1. The figure also shows the processed numerical data.

As indicated in [2], the formula (2) is applicable for calculating the thermal conductivity at relatively small values $\frac{d}{\alpha}$ and $\frac{\lambda_2}{\lambda_1}$, but at high values, the results of numerical modelling do not fit the general picture (Fig. 1) of the dependence (2). Since there are no data on composite materials with spherical inclusions at sufficiently high values $\frac{d}{\alpha} > 0.5$ and $\frac{\lambda_2}{\lambda_1} > 10$, it is necessary to develop a method that takes into account change in relative parameters $\frac{d}{\alpha}$ and $\frac{\lambda_2}{\lambda_1}$ at sufficiently high values.
At $d/a < 0.5$ and $\frac{\lambda_2}{\lambda_1} \gg 1$, there is general discrepancy between the results of Odelevsky formula and both numerical and Maxwell dependences. It is also worth noting that small values $d/a$ and $\frac{\lambda_2}{\lambda_1}$ are used in industry, therefore, the methodology for calculating a thermal conductivity coefficient proposed in this paper can be applied in practical calculations.

**Thermal Conductivity Calculation Model**

The method for calculating thermal conductivity coefficients described below is based on the approaches published earlier in papers [5,13]. As the studies have shown, it is necessary to develop a model that takes into account effect of changes in the thermal resistance of a sample at the material-inclusion boundary [5-7,9-11].

To determine thermal conductivity of materials with various spherical inclusions (Fig. 2a), we use the method that is based on geometric characteristics of the model.

Let us consider a prismatic sample, the spherical inclusions of which are uniformly distributed and are located at predetermined distance from each other. Next, we select an elementary arbitrary volume in the form of a cube with edge $a$ (Fig. 2a and b). Suppose that total heat flux $Q$ is known, and the specific heat flux is not changing $q = \text{const}$. The temperatures of the left and right lateral planes (Fig. 2b) are equal respectively to $T_1$ and $T_2$ and remain constant.
Figure 2. Model for calculating the thermal conductivity of a cubic sample with dispersed spherical inclusion: prismatic sample with spherical inclusions (a); cubic sample with spherical inclusion (b); prismatic surface element with disk inclusion, top view (c); elementary volume singled out from the previous plate (g)

In fig. 2c, we single out another surface element, which forms thin prismatic volume with the spherical inclusion part. Then the energy conservation law for elementary prismatic (Fig. 2d) volume is written in the form:

$$dT = q dR_t,$$

Where $R_t$ – thermal resistance of the sample. Let us integrate the ratio within:

$$\int_{T_a}^{T_b} dT = q \int_{R_{t1}}^{R_{t2}} dR_t.$$

The total thermal resistance for elementary prismatic volume at the phase boundary (in Fig. 2d) is then written in the form:

$$R_t = R_{t1} + R_{t2} + R_{t3} = \frac{2\sqrt{R^2 - x^2}}{\lambda_2} + \frac{a - h - 2\sqrt{R^2 - x^2}}{\lambda_1} + \frac{h}{\lambda_1}.$$

We integrate relation (3) with the conditions indicated above:

$$T_1 - T_2 = q \int_{0}^{\Delta y} dR_t.$$
Then reducing the terms in the total thermal resistance, we get that the specific heat flux for the elementary element is equal to:

\[ q = \frac{T_1 - T_2}{T_2 - x^2 + \frac{\lambda_1}{\lambda_2}}. \]  

(4)

To calculate the heat flux of the entire three-dimensional model \( Q \), we multiply (4) by \( q \delta y \cdot dx \), since this is nothing but the area of the lateral surface elements whose temperatures are equal to \( T_1 \) and \( T_2 \), respectively, and the product itself is equal to the total heat flux:

\[ dQ = q \delta y \cdot dx. \]

As a result, the integral relation (4) becomes the differential equation having the form

\[ dQ = \frac{\delta y}{2\sqrt{R^2 - x^2}} \frac{(T_1 - T_2)}{\lambda_2} dx. \]

Integrating it, we obtain the formula at constant known plate thickness \( \Delta y = \delta \) (Fig. 2b):

\[ Q = 2 \int_0^R \frac{\delta y}{2\sqrt{R^2 - x^2}} \frac{(T_1 - T_2)}{\lambda_2} dx. \]

(5)

Omitting rather cumbersome calculations, we obtain the thermal conductivity value for a thin plate with the disk inclusion in relative variables:

\[ \lambda = \left(1 - \frac{d}{a}\right) \lambda_1 + \frac{\lambda_1 \lambda_2}{2(\lambda_1 - \lambda_2)} \left[ \pi - \frac{4\lambda_2}{\sqrt{(\lambda_2^2 - \frac{1}{a})(\lambda_1 - \lambda_2)}} \arctg \sqrt{\frac{\lambda_2^2 - \frac{1}{a}(\lambda_1 - \lambda_2)}{2\lambda_2^2 - \frac{1}{a}}(\lambda_1 - \lambda_2)} \right]. \]

(6)

where \( \lambda_1, \lambda_2 \) – thermal conductivity coefficients of the sample material and dispersed phase material, \( W / m \cdot K). \)

For a three-dimensional sample (Fig. 2b) with spherical inclusion, after taking into account the heat content of the prismatic parts from the top and bottom model parts, we will get that the formula for calculating the thermal conductivity of a cubic sample with dispersed spherical inclusion will have the following form:

\[ \lambda = \left(1 - \frac{d}{a}\right) \lambda_1 + 2 \sum_0^R \frac{\delta(\frac{d}{a})\lambda_1}{\alpha} + 2 \sum_0^R \frac{\lambda_1 \lambda_2 \delta}{2\alpha(\lambda_1 - \lambda_2)} \left[ \pi - \frac{4\lambda_2}{\sqrt{(\lambda_2^2 - \frac{1}{a})(\lambda_1 - \lambda_2)}} \arctg \sqrt{\frac{\lambda_2^2 - \frac{1}{a}(\lambda_1 - \lambda_2)}{2\lambda_2^2 - \frac{1}{a}}(\lambda_1 - \lambda_2)} \right]. \]

(7)

Model calculation by numerical methods. Comparison of results

Using ANSYS Fluent software, we simulate the process of the heat flux propagation through cubic-type samples with spherical inclusion. Using processing of the data on changes in the specific heat flux over the entire height of a sample, one can obtain thermal conductivity values for prismatic samples with different inclusion radii using the averaging method. We build several models of cubes with inclusions \( \frac{d}{a} = 0.2, 0.5 \) and 0.75. Next, we set the temperature of the upper and lower faces to
1000 and 300 K. The lateral planes and faces are assumed to be adiabatic, i.e. isolated so as not to take into account losses to the environment. We obtain the distribution contours of the specific heat fluxes in Fig. 3 at different values \( \frac{\lambda_2}{\lambda_1} \).

**Figure 3.** The distribution contours of the specific heat flux over the entire volume of the cubic sample with the insulated side walls and computational grid, and with spherical inclusion with \( d = 0.5 \) m at given temperatures of 1000 and 300 K for the upper and lower faces, where \( \frac{\lambda_2}{\lambda_1} = 0,2 \) and 5, respectively.

Since use of the formula (7) is inconvenient, it is therefore necessary to replace it with some empirical dependence. Examples of changes in the specific heat flux in different cross sections of the sample are shown in Fig. 4. From the analysis of the data obtained, the most suitable for calculating is an empirical equation having the form:

\[
\lambda = \lambda_1 + 0.2 \cdot \lambda_1 \left( \frac{\lambda_2}{\lambda_1} - 1 \right) \left( \frac{d}{\alpha} \right)^3. \tag{8}
\]

This relationship is applicable when \( \frac{\lambda_2}{\lambda_1} > 1 \). For the case \( \frac{\lambda_2}{\lambda_1} < 1 \), it is necessary to apply the relationship:

\[
\lambda = \lambda_1 + \frac{\lambda_1}{\left( \frac{\lambda_2}{\lambda_1} - 1 \right) \left( \frac{d}{\alpha} \right)^3}. \tag{9}
\]
Figure 4. Distribution of the specific heat flux over the entire height of the sample in different cross sections with $d/a = 0.24$ and $0.72$ at $\lambda_2/\lambda_1 = 0.2 \pi 15$, respectively.

Next, we present comparison of the analytical model with the results obtained with numerical methods, Fig. 5. As a result, we get curves using the relation (8) for different ratios of thermal conductivity coefficients $\lambda_2/\lambda_1$.

\[
\lambda = \lambda_1 + 0.2 \cdot \lambda_1 \left( \frac{\lambda_2}{\lambda_1} - 1 \right) \left( \frac{d}{a} \right)^3
\]

Figure 5. Change in the thermal conductivity coefficient on the spherical inclusion radius for analytical dependencies with correction factors and empirical dependencies.

It can be seen that the empirical relations are in good agreement with the numerical results. At the same time, for the analytical relationship itself there are correction factors which are introduced to take...
into account difference between numerical and analytical results. We emphasize that for \( \frac{d^2}{\lambda_1} < 1 \) the analytical dependence coincides with the empirical curve. At \( \frac{d^2}{\alpha} < 0.5 \) and at \( \frac{d^2}{\lambda_1} > 1 \), the results of the above dependences also agree well with each other.

**Conclusion**

When studying the prismatic model in the form of a cube with the spherical inclusion, the formulas for calculating a thermal conductivity coefficient, which take into account the ratio of thermal conductivity of the filler and inclusion, as well as the inclusion diameter, were obtained. In this case, the model was based on thermal resistance consideration. Taking into account the analysis of the obtained results, the empirical dependencies for more convenient use were introduced. As it was shown, the obtained analytical dependences of thermal conductivity can be used only for \( \frac{d^2}{\lambda_1} > 0.5 \), but for empirical dependences, the limit of use is wider, although it can be expected that, for sufficiently large \( \frac{d^2}{\lambda_1} \gg 1 \) or for sufficiently small \( \frac{d^2}{\lambda_1} \ll 1 \) values of the ratio of thermal conductivity coefficients of the binder and dispersed media, the theory will give a higher error. The methodology allows making necessary calculations, obtaining criteria for thermal conductivity of the material with small degree of the error from actual values for relatively small values of ratios \( \frac{d}{\alpha} \) and \( \frac{d^2}{\lambda_1} \).

**References**

[1] J. C. Maxwell. 1873. *Oxford University Press*. P. 500.
[2] Meredith R.E., Tobias C.W. 1961. *Journal of the Electrochemical Society*. Vol. 108, N. 3. P. 286-290.
[3] G. N. Dulnev, Yu. P. Zarichnyak. 1974. *L.: Energy*. P. 264.
[4] V. A. Mikheev, V. Sh. Sulaberidze, V. D. Mushenko. 2015. *Bulletin of Universities. Instrument making*. Vol. 58, No. 7. P. 167-172.
[5] Sharapov A.I., Korshikov V.D., Chernykh A.A., Peshkova A.V. 2020. *Journal of Chemical Technology and Metallurgy*. Vol. 55, Iss. 1. P. 148-155.
[6] Ngo I., Jeon S., Byon C. 2016. *Interactive J. Heat Mass Tran*. Vol. 98. P. 219–226.
[7] V.S. Zarubin, G.N. Kuvyrkin. 2013. *Thermophysics of high temperatures*. Vol. 51 No. 4. P. 578.
[8] Alshaer W. G., Nada S. A., Rady M. A., Del Barrio, E. P., Sommier A. 2015. *International Journal of Thermal Sciences*. Vol. 89. P. 79–86.
[9] Khedari, J., Suttisonk, B., Pratinthong, N. &Hirunlabh, J. 2001. *Cement and Concrete Composites*.
[10] Chen Y.-M., Ting J.-M. 2002. *Carbon*. Vol. 40. P. 359–362.
[11] Progelhot R.G., Throne L., Ructsch R.R. 1976. *Polymer Eng. Sci*. Vol. 16. P. 615.
[12] Weinan E., Enquist B., Li X., Ren W., Vanden/Eijnden E. 2007. *Comm. Comput. Phys*. Vol. 2. P. 367.
[13] A. A. Chernykh, A. M. Shmyrin. 2020. *Computational mechanics of continuous media*. Vol. 13, No. 1. P. 34-43.
[14] V. S. Zarubin, G. N. Kuvyrkin, I. Yu. Savelyeva. 2014. *Bulletin of Moscow State Technical University named after N.E. Bauman Series Natural Sciences*. No. 5(56). P. 94-108.
[15] Xu Y., Kinugawa J., Yagi K. 2003. *Mater. Trans*. V. 44, № 4. P. 629.
[16] Bensoussan A., Lions J./L., Papanicolau G. 2011. *American Mathem. Society*. P. 392.
[17] Bouguerra A., Laurent J., Goul M., Queneudec M. 1997. *J. Phys. D: Appl. Phys*. Vol. 30. P. 2900–2904.