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EFFECTIVE HEAT CONDUCTIVITY OF POLYMERIC COMPOSITE MATERIALS: THE INFLUENCE OF COMPONENT PROPERTIES

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The goal of this study is to increase the effective heat conductivity (EHC) of the polymer composite materials. We have carried out numerical simulation of the polymer composite EHC when making microsized high-thermal conductivity fillers of various shape a component of the composite, and its volume fraction being varied as well. The influence of particles' thermal conductivity on the polymer composite materials' EHC was analyzed. Recommended practice for the polymer composite EHC increasing was suggested. Shape changing, volume fraction optimization, the filler particles', and the matrix' thermal conductivity increasing was proposed. The calculation results were in good agreement with our experimental data and published in scientific literature. This study gave an insight into the function of high-thermal conductivity fillers on the EHC of the polymer composite materials.

Keywords: numerical simulation, composite material, effective thermal conductivity, high-thermal conductivity filler

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ВЛИЯНИЕ СВОЙСТВ КОМПОНЕНТОВ НА ЭФФЕКТИВНУЮ ТЕПЛОПРОВОДНОСТЬ ПОЛИМЕРНЫХ КОМПОЗИТНЫХ МАТЕРИАЛОВ

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Целью данного исследования является повышение эффективной теплопроводности полимерных композитных материалов. В связи с этим проведено численное моделирование теплопроводности при введении в полимерную матрицу композита высокотеплопроводных микрочастиц различной формы. При этом варьировалась объемная концентрация этих микрочастиц. Проанализировано влияние теплопроводности частиц наполнителя на эффективную теплопроводность полимерных композитных материалов. Предложены конкретные рекомендации по повышению эффективной теплопроводности путем изменения формы, концентрации, теплопроводности частиц наполнителя и повышения теплопроводности полимерной матрицы.

Результаты расчетов хорошо согласуются с экспериментальными и опубликованными данными. Проведенное исследование позволило глубже понять действие высокотеплопроводных наполнителей на эффективную теплопроводность полимерных композитов.

Ключевые слова: численное моделирование, композитный материал, эффективная теплопроводность, высокотеплопроводный наполнитель

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Introduction

Polymer composite materials (PCM) are structures consisting of two or more components with different physical properties and a clear interface between them, currently finding diverse applications in different fields of science and technology.

Improving heat conductivity while maintaining high dielectric characteristics are some of the key issues in using PCM in electrical insulation systems for high-voltage electrical equipment. The polymer matrix typically has low thermally conductive and high dielectric properties. Heat transfer in PCM can be improved by introducing micron-sized dielectric fillers with high thermophysical characteristics into the polymer matrix [1 - 13].

This technique might reduce the electrical insulating properties of PCM, whose nature depends on the electrophysical properties of particles and their concentrations in PCM [9, 13–15].

The efficiency of heat conductivity in PCM can be increased by correctly selecting the properties, sizes and concentrations of filler particles in the polymer matrix. Since the PCM are two-component systems, analytical models should be used for solving thermophysical problems. The analytical models that have proved the most popular for approximate calculations of effective heat conductivity of composites are given in [1–4, 10]. However, analytical models cannot provide sufficiently reliable results for running calculations with the option of widely varying the concentrations, shapes or sizes of filler particles [10].

Development of numerical model

Modern computing tools allow to perform direct simulation of thermal conductivity of media with complex structures. Standard packages such as ANSYS, FLUENT, etc., can be used for these purposes.

To date, the ANSYS simulation environment, used for solving a wide range of engineering problems, including those related to thermal physics, has proven its efficiency, flexibility and versatility. These features of the package allow to further improve the computational models, for example, by adding a module for calculation of electrical processes in APDL (ANSYS Parametric Design Language).

Uniform distribution of particles was considered at the first stage of simulation. The volume of the PCM matrix can be represented as a set of unit cells, each containing one filler particle with the given shape (Fig. 1). Calculations were carried out for particles of different shapes and orientations.

We made certain assumptions to simplify the computational model.

1. Problems are regarded as steady-state, since temperature equalization occurs much faster in the heat-conducting volume than the change in external conditions.
2. The materials making up the composite are regarded as isotropic, with their thermal conductivity independent of temperature.
3. The orientation of cubic and cylindrical filler particles coincides with the direction of the coordinate axes.
4. The sizes of filler particles are assumed to be the same (of the order of microns); the fractional composition of particles is not taken into account.
5. Thermal contact between the particles and the polymer is assumed to be perfect (this assumption entails an increase in thermal conductivity of the composite due to zero thermal resistance at the polymer/particle interface).
6. The presence of microdefects is not taken into account.

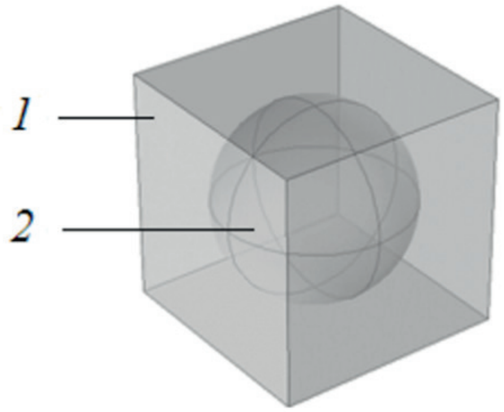

Fig. 1. Model representation of uniform distribution of filler particles: elementary volume of PCM matrix cell (*1*) with spherical filler (*2*)

Parameterization of the numerical model allows to vary the properties of the system, in this case, the particle sizes, their properties, and the properties of the PCM matrix.

According to developed model, the relationship between the length L of the cube edge (it determines the size of the computational domain containing one particle) and the volume of the particle V_p is given by the formula

$$L = \left(\frac{V_P}{N_V} \right)^{\frac{1}{3}},$$

where N_V is the volume concentration of particles.

The effective thermal conductivity λ_{eff} of the PCM was calculated by the formula:

$$\lambda_{eff} = q_m \cdot \frac{L}{\Delta T},$$

where q_m is the mean heat flux through the cube face with the particle, ΔT is the temperature difference at the cube faces;

$$\Delta T = T_0 - T_L.$$

A three-dimensional steady-state thermal conductivity problem is numerically solved in order to determine the value of q_m:

$$\frac{\partial}{\partial x}\left(\lambda \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y}\left(\lambda \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z}\left(\lambda \frac{\partial T}{\partial z} \right) = 0.$$

The thermal conductivity in this equation depends on the coordinates: $\lambda = \lambda\,(x, y, z)$.

Boundary conditions of the first kind are imposed on two faces of the cube, while thermal insulation conditions are imposed on the other four.

To simplify calculations and analysis of the results, dimensionless variables are introduced, taking the following form:

$$\Theta = \frac{T - T_L}{\Delta T},\ \bar{x} = \frac{x}{L},\ \bar{y} = \frac{y}{L},\ \bar{z} = \frac{z}{L},$$
$$\bar{\lambda} = \frac{\lambda}{\lambda_M},\ \bar{q} = \frac{q \cdot L}{\lambda_M \cdot \Delta T} \qquad (1)$$

The quantity λ_M in the last two expressions is the thermal conductivity of the matrix.

The problem takes the following form in terms of variables (1):

$$\frac{\partial}{\partial \bar{x}}\left(\bar{\lambda} \frac{\partial \Theta}{\partial \bar{x}} \right) + \frac{\partial}{\partial \bar{y}}\left(\bar{\lambda} \frac{\partial \Theta}{\partial \bar{y}} \right) + \frac{\partial}{\partial \bar{z}}\left(\bar{\lambda} \frac{\partial \Theta}{\partial \bar{z}} \right) = 0,$$

and the boundary conditions and the thermal conductivity take the form

$$\bar{x} = 0,\ \Theta = 1;\ \bar{x} = 1,\ \Theta = 0;\ \bar{y} = 0,\ \bar{y} = 1,$$
$$\frac{\partial \Theta}{\partial \bar{y}} = 0;\ \bar{z} = 0,\ \bar{z} = 1,\ \frac{\partial \Theta}{\partial \bar{z}} = 0$$

in this case $\bar{\lambda} = 1$ if the coordinates of the point belong to the matrix, and $\lambda = \lambda_p / \lambda_M$ if the coordinates of the point belong to a particle with thermal conductivity λ_p.

The relative effective thermal conductivity (relative to the thermal conductivity of the matrix) is determined by integrating the dimensionless heat flux over the dimensionless unit surface of the cube face:

$$\overline{\lambda_{eff}} = \frac{\lambda_{eff}}{\lambda_M} = \int_{\bar{S}} \bar{q} \cdot d\bar{s}.$$

Calculations of effective thermal conductivity in PCM

The distribution of heat fluxes for the given particles in the cross-section of the computational domain is shown in Fig. 2. The heat flux fields shown correspond to a volume particle concentration N_V = 10% and exhibit considerable differences depending on the shape and position (for a cylindrical particle) of the particles.

The calculated results of the relative effective thermal conductivity of PCM are shown in Figs. 3 and 4. The volume concentration of particles is limited to N_V = 30% for all calculation variants to maintain the permissible electrical properties of the PCM.

In addition, Figs. 3 and 4 show a comparison of the results obtained in this study with the experimental data from [4, 5, 8, 10]. The experimental values of the effective thermal conductivity were obtained for hexagonal particles whose thermal conductivity varied in the range λ_p = 40–120 W/(m·K) introduced into the matrix (with the thermal conductivity λ_M = 0.28 W/(m·K)) [10]. We took the thermal conductivity of the matrix to be equal to λ_M = 0.28 W/(m·K), and the thermal conductivity of the particles to be equal to λ_p = 40 W/(m·K) in numerical simulation.

The effective thermal conductivity practically does not depend on the shape of the particle within the given model (Fig. 3, curves *1–3*). A more complex model taking into account polydisperse particles, the structure of their surface, their random distribution in the volume and the random formation of thermal bridges (structures formed by a group of particles) can yield a different result, i.e., a pronounced dependence of effective thermal conductivity on particle shape.

The calculated results generally agree with the experimental data. Using analytical models allows to estimate the effective thermal conductivity for low concentrations of the filler. Using Maxwell's model yields the best results (Fig. 3).

Calculations also showed that an increase in the thermal conductivity of the matrix leads to a directly proportional increase in the effective thermal conductivity of PCM. This conclusion is valid for low filler concentrations (up to 30%).

A strong dependence of the effective thermal conductivity on both particle concentration and orientation relative to the heat flux (along or across it) is observed for elongated cylindrical microparticles (see Fig. 4).

Thus, elongated microparticles should make a greater contribution to increasing the effective thermal conductivity, compared to the other types of particles. . The effect should be more pronounced for particle orientation along the heat flux.

Fig. 5 shows the results of the study considering how the length of cylindrical filler microparticles (with a constant volume) affected the effective thermal conductivity of PCM.

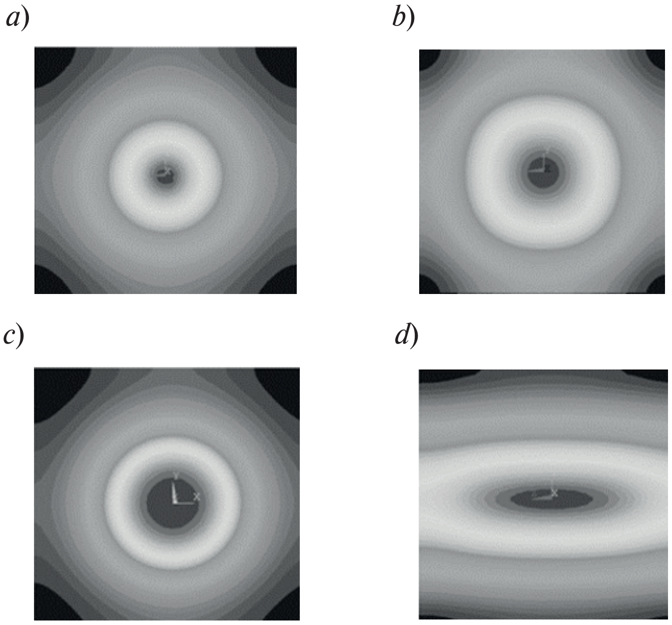

Fig. 2. Heat flux distribution in the cross-section of the computational domain for microparticles of different shapes: spherical (*a*), cubic (*b*) and cylindrical (*c*, *d*); the heat flux is directed along (*c*) and across (*d*) the particle; N_V= 10%

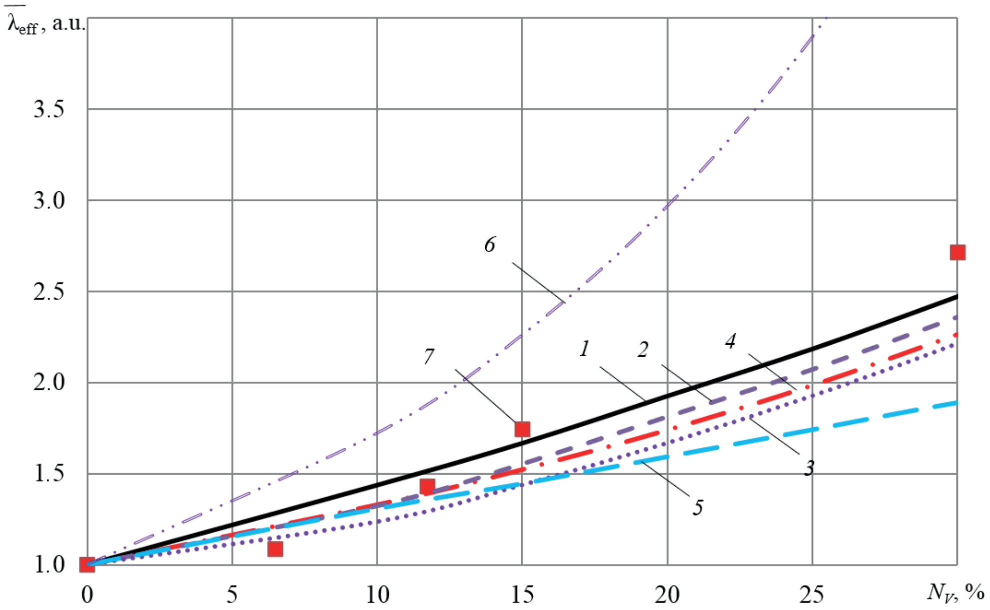

Fig. 3. Calculated (lines) and experimental [5] (symbols *7*) data on relative effective thermal conductivity of PCM with filler microparticles of different shapes: spherical (*1*, *7*), cubic (*2*) and cylindrical with $D = h$ (*3*). The calculated results were obtained using Maxwell's [4, 10] (*4*) and Timofeyeva's [4] (*5*) models, the geometric model [4, 10] (*6*) and the model in our study (*1–3*)

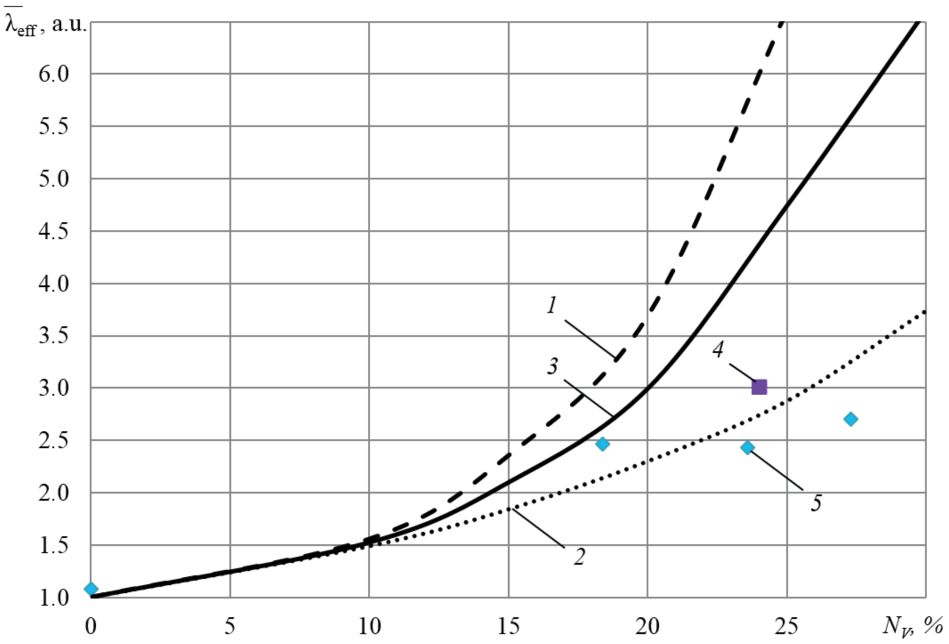

Fig. 4. Calculated (lines *1–3*) and experimental [8, 10] (symbols *4* and *5*, respectively) data for concentration dependences of relative effective thermal conductivity of PCM with filler microparticles of different shapes: cylindrical with $D/h = 2/3$ (*1–3*) and hexagonal (*4*, *5*).
Mean values of the effective thermal conductivity (*3*) and its values for heat fluxes directed along (*1*) and across (*2*) the microparticles were obtained

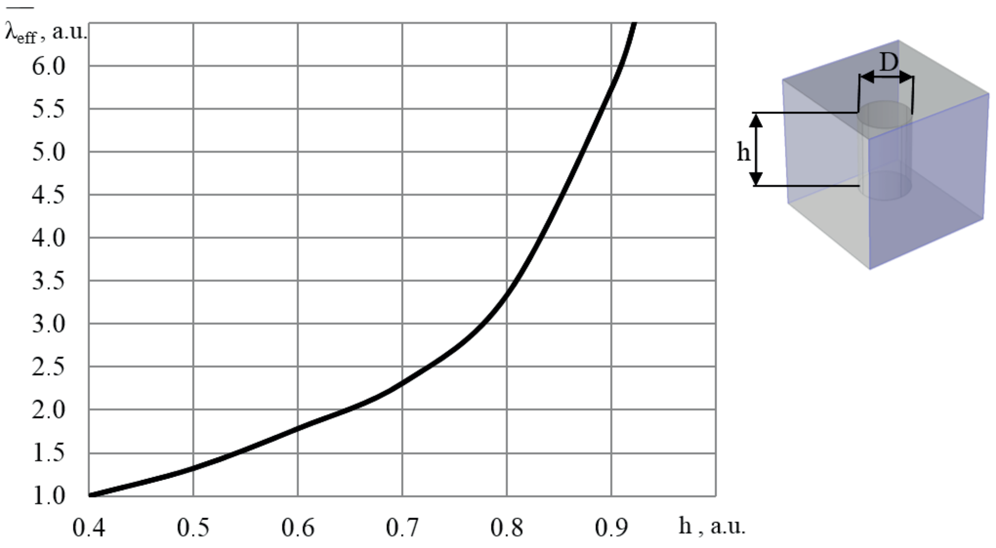

Fig. 5. Calculated dependence of effective thermal conductivity of PCM on relative length of the cylindrical filler microparticle;
N_V = 30%. The elementary cubic volume (1 × 1 × 1) of the PCM matrix cell with a cylindrical filler microparticle is shown on the right.

Simulation confirmed that an increase in the length of microparticles is accompanied by an increase in thermal conductivity of the PCM.

Fig. 6 shows the dependences of effective thermal conductivity of PCM on the thermal conductivity of particles of different shapes for two volume concentrations: 10% and 30%.

Analyzing the given dependences, we established that an increase in the thermal conductivity of PCM can be achieved by increasing the thermal conductivity of the filler particles or by lengthening the particles with an increase in their

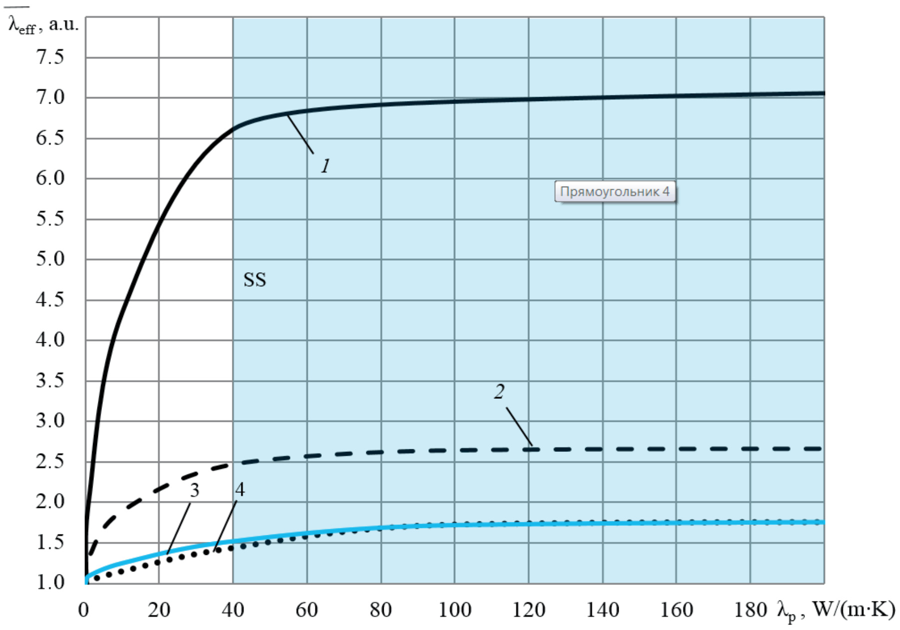

Fig. 6. Calculated dependence of effective thermal conductivity of PCM on thermal conductivity of filler microparticles of different shapes: cylindrical with D/h = 2/3 (*1*, *3*) and cubic (*2*, *4*); N_V = 30% (*1*, *2*) and 10% (*3*, *4*)
The shaded background corresponds to the saturated state (SS)

volume concentration in the polymer matrix. However, in practice, increasing the thermal conductivity of particles makes sense up to values of 40–100 W/(m·K), depending on the particles' shape and concentration.

A possible explanation for this is that the thermal resistance of microparticles becomes much less than the thermal resistance of the matrix and has almost no effect on the mean heat flux.

The trend of the dependences given in Fig. 6 is valid for particles of any shape.

Thus, the maximum effective thermal conductivity of the polymer composite material is determined by the thermal conductivities of both the polymer matrix and filler microparticles, as well as by the concentration of particles, their shape and orientation in the matrix relative to the heat flux direction.

Conclusion

Our study was carried out with the ANSYS package and consisted in direct simulation of heat conductivity of a polymer two-component medium with a complex structure.

We have solved the following problems through numerical simulation:

1. We have studied the effect of volume concentrations of cubic, spherical and cylindrical microparticles on the effective heat conductivity of PCM.
2. We have analyzed the effect of shape and position of cylindrical filler particles on the effective heat conductivity of PCM.
3. We have studied the effect of heat conductivity of the particles included in the polymer matrix on the effective heat conductivity.
4. We have established the effect of heat conductivity of the polymer matrix on the effective thermal conductivity.

Analysis of the obtained results lead us to conclude that the maximum value of the effective heat conductivity of PCM is determined by the thermal conductivities of both the polymer matrix and the filler, as well as by the concentration of particles, their shape and orientation in the matrix relative to the direction of the heat flux.

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