COMPARATIVE FUNCTIONAL ANALYSIS FOR PARTICULATE COMPOSITES THERMAL CONDUCTIVITY

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Composites belong to those kinds of heterogeneous materials for which application in industry and everyday life is still expanding. The transient and steady-state temperature distribution in composite configurations consisting of several distinct thermally anisotropic subdomains have numerous applications to heat transfer problems in re-entry vehicles, air frames, nuclear reactors, etc. The increased use of composite materials in the automotive, aerospace, and construction industries has motivated research into experimental techniques and solution methods to determine the thermal properties of these materials. Thermal processes occur in composite materials during their production or application. Heat transfer in composites is mainly due to heat conduction. Other modes of heat transfer are of importance only in special cases. Studies carried out on composite media proved that effective properties of heat transfer in heterogeneous materials greatly depend on their microstructure. A large number of theoretical and numerical studies are devoted to obtaining a generalized formula for the thermal conductivity of composites, which would take into account the influence of geometric and thermophysical characteristics of inclusions on heat transfer processes.

Numerous models have been proposed to predict macroscopic properties of composite material, knowing the properties and volume fractions of the constituents. The Maxwell model (model 1 - models whose formulas will be used in this paper are numbered for convenience) should be attributed to the first attempts to analytically describe the thermal conductivity coefficient of two-component media. He considered the problem of dilute dispersion of spherical particles of conductivity κ_1 embedded in a continuous matrix of conductivity κ_m, where thermal interactions between filler particles were ignored.

Rayleigh (model 2) considered material in the form of spherical inclusions arranged in a simple cubic array, embedded in a continuous matrix. The thermal interaction between particles had been taken into consideration in his study.

Hasselman and Johnson (model 3) emphasized that for a composite with a given shape of inclusion, the effective thermal conductivity depends on not only the filler volume fraction, but particle size as well. The authors derived expressions for a continuous matrix phase with dilute concentrations of dispersions with spherical, cylindrical and flat plate geometry.

Bruggeman assumed that a composite material may be constructed incrementally by introducing infinitesimal changes to an already existing material. The advantage of his scheme is that it covers a large number of materials, e.g. composites, nanofluids, porous materials, aerosols etc. Using Bruggeman's approach, Every and Tzou obtained an expression for effective thermal conductivity of particulate composites. The formula has been verified by experimental measurements on *ZnS*/diamond composites with two particle sizes and varying percentages of filler. The advantages of Lewis-Nielsen empirical model (model 4) are its simplicity and coverage of a wide range of particle shapes and patterns.

Chen et al. [1] proposed a model for polymer composites containing aligned hexagonal boron nitride (h-BN) platelets. They derived the model based on the unit cell selection and calculation of anisotropic thermal conductivity. Xu et al. [2] reconstructed the Maxwell model using a continuum approach with contact resistance among particles. They also developed a statistical model using a circuit network approach.

Accounting for the effect of shape and size of fillers was the next obvious stage in the development of heat transfer models in composite materials, which contain an explicit analytical expression for effective heat conductivity. Chauhan et al. find out the effect of geometrical shapes such as spherical, elliptical and hexagonal fillers (graphite, copper and aluminum oxide) particles on the thermal conductivity of two-phase system [3].

Kumlutas and Tavman the numerical study of variation of effective thermal conductivity for spheres in cube and cubes in cube systems for different conductivity ratio of filler and matrix [4].

Most theoretical and experimental studies conclude that the effects of the shape and concentration of the filler on the effective thermal conductivity of the composites are decisive in comparison with the effects of the size of the filler particles on the processes of heat transfer in heterogeneous media. However, the experimentally discovered effects of contact heat transfer between the filler particles, especially for high concentrations indicate that the dispersion of the particle size of the filler should also be taken into account in analytical formulas for effective thermal conductivity.

The purpose of this work is a comparative analysis of characteristic analytical expressions for the effective thermal conductivity of particle-reinforced composites. In addition, the work plans to develop recommendations for the unification of formulas for thermal conductivity while taking into account primarily the geometric parameters of the filler.

The following explicit expressions for the effective thermal conductivity κ_e of composites are presented in the above models.

Model 1.

$$\kappa_e = \kappa_m + \frac{3\varphi\kappa_m}{\left(\dfrac{\kappa_f + 2\kappa_m}{\kappa_f - \kappa_m}\right) - \varphi}, \tag{1}$$

where κ_f is the thermal conductivity of the filler; κ_m is the thermal conductivity of matrix; φ is volume fraction of the filler.

Formula (1) was found to be valid only in the case of low φ.

Model 2.

$$\kappa_e = \kappa_m + \frac{3\varphi\kappa_m}{\left(\dfrac{\kappa_f - 2\kappa_m}{\kappa_f - \kappa_m}\right) - \varphi + 1.569\left(\dfrac{\kappa_f - \kappa_m}{3\kappa_f - 4\kappa_m}\right)\varphi^{10/3} + \ldots}, \tag{2}$$

Formula (2) includes an infinite series in the dominator in which higher order components are ignored. When ignoring all of them, equation (2) reduces to equation (1). Nevertheless, equation (2) is important as it includes analytical expressions of another type of composite – a continuous matrix reinforced with parallel cylindrical fibers arranged in uniaxial cubic array.

Model 3.

$$\kappa_e = \kappa_m \frac{2\left(\dfrac{\kappa_f}{\kappa_m} - \dfrac{\kappa_f}{a\kappa_c} - 1\right)\varphi + \dfrac{\kappa_f}{\kappa_m} + \dfrac{2\kappa_f}{a\kappa_c} + 2}{\left(1 - \dfrac{\kappa_f}{\kappa_m} + \dfrac{\kappa_f}{a\kappa_c}\right)\varphi + \dfrac{\kappa_f}{\kappa_m} + \dfrac{2\kappa_f}{a\kappa_c} + 2}, \tag{3}$$

$$\kappa_e = \kappa_m \frac{\left(\dfrac{\kappa_f}{\kappa_m} - \dfrac{\kappa_f}{a\kappa_c} - 1\right)\varphi + \left(1 + \dfrac{\kappa_f}{\kappa_m} + \dfrac{\kappa_f}{a\kappa_c}\right)}{\left(1 - \dfrac{\kappa_f}{\kappa_m} + \dfrac{\kappa_f}{a\kappa_c}\right)\varphi + \left(1 - \dfrac{\kappa_f}{\kappa_m} + \dfrac{\kappa_f}{a\kappa_c}\right)}, \tag{4}$$

$$\kappa_\mathrm{e} = \frac{\kappa_f}{\left(1 - \dfrac{\kappa_f}{\kappa_m} + \dfrac{2\kappa_f}{a\kappa_c}\right)\varphi + \dfrac{\kappa_f}{\kappa_m}}, \tag{5}$$

where a is filler particle radius; κ_c is thermal boundary conductivity.

Model 3 formulas assume the following types of geometries: spherical (3), cylindrical (4) and flat plate (5).

Model 4.

$$\kappa_e = \kappa_m + \frac{3\varphi\kappa_m\left[\kappa_f(1-\alpha)-\kappa_m\right]}{\kappa_f(1+2\alpha)+2\kappa_m}, \tag{6}$$

where $\alpha = r_K/a$; r_K is Kapitza radius.

A comparative analysis of the formulas for effective thermal conductivity was made using the following dimensionless values: κ_f/κ_m = 10, $\kappa_f/a\kappa_c$ = 5, α = 0.2. Therefore, it is necessary to take into account the geometric and thermophysical parameters associated with particle sizes, effects at the filler-matrix interface, and contact interparticle interaction when generalizing formulas 1-5 to the case of large values of filler concentrations.

The experimental data performed by Tsekmes et al. [5] were analyzed in order to obtain an example of dependence $\kappa_e = \kappa_e(a)$. The authors investigated composites of epoxy resin with alumina (Al_2O_3) and silica (SiO_2) microfillers. In the case of high filler concentrations (φ > 0.3) composites exhibit much higher thermal conductivity compared to neat polymers. Two important parameters can be claimed that play a major role in determining the thermal conductivity of composites, i.e. the thermal conductivity of the fillers and interaction between them.

The results of the analysis led to the following analytical form of dependence $\kappa_e, W/(m \cdot K)$ on $a, \mu m$

$$\kappa_e = A_1 Ln(a) + A_2,\ A_1 = 0.0912,\ A_2 = -0.2102,\ R^2 = 0.6812. \tag{7}$$

Summarizing the above, it can be argued that the considered models 1–4 give similar expressions for the effective thermal conductivity of composites. To eliminate the discrepancies of the dependences 34 at high concentrations, it is necessary to take into account the nonlinear dependence of the effective thermal conductivity on the particle size of the filler.

Therefore, formulas (1) – (6) can be written in the following generalized polynomial-logarithmic form

$$\kappa_\mathrm{e} = \frac{\sum\limits_i \alpha_i\beta_i}{\sum\limits_j \gamma_j\beta_j},\quad \beta = \frac{\kappa_f}{\kappa_m},\quad \alpha,\gamma = \alpha,\gamma\left(Lna, \rho_\mathrm{m}, c_m, \upsilon_m, \varphi, \kappa_c\right), \tag{8}$$

where ρ_m is matrix density; c_m is the specific heat of matrix; υ_m is Debye velocity for matrix.

Conclusions. Analytical expressions for the effective thermal conductivity of composites are analyzed according to basic homogeneous theories. Maxwell's and Rayleigh's models indicate higher values of effective thermal conductivity compared to Hasselman-Johnson and Lewis-Nielsen models for high values of volume fraction of the filler. Discrepancies found in model predictions can be eliminated by taking into account the functional dependence of the thermal conductivity coefficient on the particle size of the filler. Approximate processing of experimental data indicates that such a dependence should be nonlinear. It is convenient to generalize the analyzed formulas for thermal conductivity in a polynomial-logarithmic form.

References

1. Lin Chen, Ying-Ying Sun, Hong-Fei Xu, Shao-Jian He, Gao-Sheng Wei, Xiao-Ze Du, Jun Lin. Analytic Modeling for the Anisotropic Thermal Conductivity of Polymer Composites Containing Aligned Hexagonal Boron Nitride. *Composites Science and Technology*. 2016. Vol. 122. P. 42–49. https://doi.org/10.1016/j.compscitech.2015.11.013.
2. Jinzao Xu, Benzheng Gao, Hongda Du, Feiyu Kang. A Reconstruction of Maxwell Model for Effective Thermal Conductivity of Composite Materials. *Applied Thermal Engineering*. 2016. Vol. 102. P. 972–979. https://doi.org/10.1016/j.applthermaleng.2016.03.155.
3. Deepti Chauhan, Nilima Singhvi and Ramvir Singh. Effect of Geometry of Filler Particles on the Effective Thermal Conductivity of Two-Phase Systems. *International Journal of Modern Nonlinear Theory and Applications*. 2012. Vol. 1. P. 40–46. https://doi.org/10.4236/ijmnta.2012.12005.
4. Dilek Kumlutas and Ismail Tavman. A Numerical and Experimental Study on Thermal Conductivity of Particle Filled Polymer Composites. *Journal of Thermoplastic Composite Materials*. 2006. Vol. 19. P. 441–455. https://doi.org/10.1177/0892705706062203.
5. I. A. Tsekmes, R. Kochetov, P. H. F. Morshuis, J. J. Smit. Thermal Conductivity of Polymeric Composites: A Review. International Conference on Solid Dielectrics, Bologna, Italy, June 30–July 4, 2013. P. 678–681. https://doi.org/10.1109/icsd.2013.6619698.