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A review of models for effective thermal conductivity of composite materials

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Abstract

The solutions of Maxwell and Rayleigh were the first of many attempts to determine the effective thermal conductivity of heterogeneous material. Early models assumed that no thermal resistance exists between the phases in heterogeneous material. Later studies on solid-liquid and solid-solid boundaries revealed that a temperature drop occurs when heat flows through a boundary between two phases and, as a consequence, the interfacial thermal resistance should be included in the heat transfer model. This paper is a review of the most popular expressions for predicting the effective thermal conductivity of composite materials using the properties and volume fractions of constituent phases. Subject to review were empirical, analytical and numerical models, among others.

Keywords: thermal conductivity, interfacial thermal resistance, composite materials

1. Introduction

The problem of heat conduction in heterogenic materials is mathematically analogous to the problems of electrical conductivity, permittivity and magnetic permeability of such materials. Study of these topics dates back to early works of Maxwell and Lord Rayleigh [1, 2]. Since the late 19^{th} century, many models allowing the prediction of effective thermal conductivity of various types of composite materials were proposed. In the majority of older models the interfacial thermal resistance (ITR) between matrix and filler is not taken into account. However, later studies show that this type of thermal resistance may have a relatively large influence on the value of effective thermal conductivity [3, 4]. First expressions for effective thermal conductivity of composite materials, which included the influence of ITR, were derived by Hasselman and Johnson [5]. in the 1980s by modifying the original Maxwell model. In this paper, models of effective thermal conductivity of composite materials most frequently appearing in the literature are presented and compared, both including and not including the influence of ITR.

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2. On the nature of Interfacial Thermal Resistance

When heat flows through an interface between the constituents of a composite, a temperature drop occurs at the interface. This disturbance of heat flow can be described by means of thermal resistance. It is known as interfacial thermal resistance (ITR) and refers to the combined effect of two thermal resistances. The first is referred to as thermal contact resistance (TCR) and is caused by poor mechanical and chemical bonding between constituent phases. The second is thermal boundary resistance (TBR), which occurs due to differences in the physical properties of constituent materials. The latter is also known as Kapitza resistance in memory of P. Kapitza who was the first to observe a temperature drop at a boundary between liquid helium and metals [6]. The ITR is defined as the ratio of temperature discontinuity $\triangle T$ occurring at the interface to the heat rate $\dot{Q}$ per unit area A flowing across the interface between two phases in contact, according to the equation:

$$R_{int} = \frac{\triangle T}{\dot{Q}/A} \tag{1}$$

where R_{int} is the interfacial thermal resistance [7].

Lets us explain the nature of the thermal boundary resistance component. Even assuming perfect contact at the

Table 1: List of symbols

α	particle size and interfacial thermal resistance coefficient;
β	particle shape and orientation-dependent coefficient;
γ	newline effective phase contrast;
ϕ	volume fraction;
ϕ_m	max. filler volume fraction;
ψ	Lewis-Nielsen model auxiliary coefficient;
a	filler particle radius;
a_k	Kapitza radius;
A	particle shape coefficient;
B	Lewis-Nielsen model auxiliary coefficient;
Bi	Biot number;
C_1, C_2	auxiliary constants;
h_c	thermal boundary conductivity;
k_1	thermal conductivity of the filler;
k_m	thermal conductivity of the matrix;
k_{eff}	effective thermal conductivity of the composite;
$\dot{Q}$	heat rate;
R_{int}	interfacial thermal resistance;
$\triangle T$	temperature drop at the interface;

atomic level, the boundary between two solid phases is resistive to heat flow. This is due to differences in vibrational and electronic properties in different materials. The heat carrier (phonon or electron) arriving at the interface will undergo scattering, and the probability of transmission after scattering will depend on available energy states on both sides of the interface [7]. The acoustic and diffuse mismatch models (AMM and DMM; see [7]) describing interfacial heat transfer at the solid-solid boundary allow one to calculate the value of TBR, but their predictions are accurate only for temperatures below 40 K. For higher temperatures the scattering mediated mismatch model (SMAMM) developed by Prasher and Phelan [8] gives accurate results, but requires estimation of additional parameters which depends on material properties, and thus introduces an unwanted uncertainty to the calculations [4].

Apart from acoustic mismatch, there are other phenomena responsible for the increase in thermal resistance between constituent materials of a composite. Localized atomic disorder, close to the interface, may lead to enhanced phonon and photon scattering. Lattice distortions caused by thermal stress are also common in this region. These crystal imperfections also act as phonon and photon scatterers. ITR may be linked to the presence, at the interface, of thin layers of material with different properties than matrix and filler. Such layers may result from interdiffusion or corrosion of composite components, particle coating, particle electrochemical treatment, and moisture absorption. Poor adhesion causes imperfect mechanical contact and also increases the value of ITR by increasing the TCR component. Thermal expansion mismatch between components may lead to the formation of gas-filled gaps in the interfacial region. Heat transfer only by radiation and gas conduction is permitted in such a gap, therefore it acts as thermal resistance [9].

Experimental study of particulate composites shows that adding conductive particles to the metal matrix enhances the effective thermal conductivity of the material only in the case of relatively large particle size [3, 4]. With decreasing average particle radius, the effective thermal conductivity decreases, and the area of interfacial contact per unit volume increases. Apparently in such cases, the ITR begins to play a significant role in overall heat transfer, and therefore it is very important not to omit its influence in the composite design process.

3. Models for effective thermal conductivity of composites

The most important models, from the historical and practical point of view, are presented in this section. Numerous models have been proposed to predict macroscopic properties of the heterogenic medium, knowing the properties and volume fractions of the constituents. These are known as effective medium theories (EMT) or effective medium approximations (EMA) and belong to the class of mean-field theories. In these models, heterogenic materials are considered as being macroscopically homogenic. Due to their nature, effective medium approximations are unable to accurately predict the properties of heterogenic material beyond the percolation threshold. Only recently have methods appeared that take percolation into consideration [4, 10–12]. The phenomenon of percolation and its modeling is discussed in section 3.7.

3.1. Maxwell model

Maxwell was the first to give analytical expressions for effective conductivity of heterogenic medium in his famous work on electricity and magnetism [2]. He considered the problem of dilute dispersion of spherical particles of conductivity k_1 embedded in a continuous matrix of

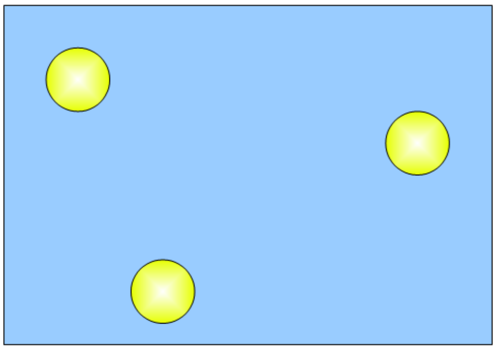

Figure 1: Dilute concentration of spherical particles embedded in a continuous matrix. The Maxwell model assumes a lack of thermal interaction between the embedded spheres.

conductivity k_m, where thermal interactions between filler particles were ignored [13]. The schematic of such material is shown in Fig 1.

Maxwell's expression is as follows:

$$\frac{k_{eff}}{k_m} = 1 + \frac{3\phi}{\left(\frac{k_1+2k_m}{k_1-k_m}\right) - \phi} \tag{2}$$

where ϕ is the volume fraction of the filler. Maxwell's formula was found to be valid only in the case of low ϕ (under about 25%). Many researchers modified Maxwell's model to include various effects. Eucken [14] extended it to allow calculation for multiple different phases of filler particles in one continuous matrix phase. Burger [15] and Hamilton and Crosser [16] included effects of different particle shapes.

3.2. Rayleigh models

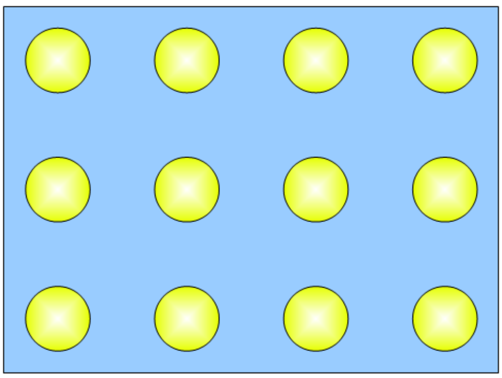

Figure 2: Spherical particles arranged in simple cubic array embedded in a continuous matrix, as considered by Rayleigh.

Rayleigh [1] considered material in the form of spherical inclusions arranged in a simple cubic array, embedded in a continuous matrix (See Fig. 2).

In his calculation, thermal interaction between particles had been taken into consideration, and thus one may expect it to give better results for higher filler fractions than Maxwell's expression. In fact, results from both formulae are very similar, and far from reality for higher ϕ Rayleigh's formula is [13]:

$$\frac{k_{eff}}{k_m} = 1 + \frac{3\phi}{\left(\frac{k_1-2k_m}{k_1-k_m}\right) - \phi + 1.569\left(\frac{k_1-k_m}{3k_1-4k_m}\right)\phi^{\frac{10}{3}} + \ldots} \tag{3}$$

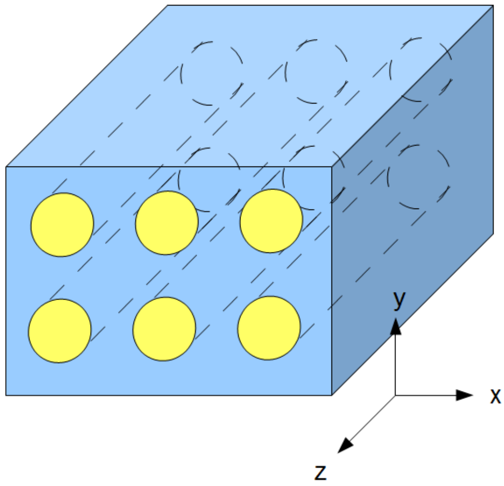

Figure 3: The schematic of the second type composite medium considered by Rayleigh, consisting of parallel cylinders embedded in a continuous matrix.

We can see an infinite series in the denominator in which higher order components are ignored. When ignoring all of them, equation (3) reduces to Maxwell's result (2). Nevertheless, Rayleigh's work is important as it includes analytical expressions for effective thermal conductivity of another type of composite – a continuous matrix reinforced with parallel cylindrical fibers arranged in uniaxial simple cubic array (See Fig. 3).

Thermal conductivity of such material is direction-dependent. Assuming z is the axis parallel to the fibers, Rayleigh's formulae are [13]:

$$\frac{k_{eff,ZZ}}{k_m} = 1 + \left(\frac{k_1 - k_m}{k_m}\right)\phi \tag{4}$$

$$\frac{k_{eff,XX}}{k_m} = \frac{k_{eff,yy}}{k_m} = 1+ \\ + \frac{2\phi}{C_1 - \phi + C_2\left(0.30584\phi^4 + 0.013363\phi^8 + \ldots\right)} \tag{5}$$

where $C_1 = \frac{k_1+k_m}{k_1-k_m}$ and $C_2 = \frac{k_1-k_m}{k_1+k_m}$

3.3. Hasselman-Johnson model

Hasselman and Johnson 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. Moreover, they examined the influence of interfacial gaps between filler and matrix on the thermal diffusivity and conductivity of Ni-glass composites [17]. They connected these observations with the presence of interfacial thermal resistance. Shortly after, they proposed simple modification of the original Maxwell and Rayleigh models to derive first expressions for effective thermal conductivity of composite materials with nonzero interfacial thermal resistance [5].

The novelty in the Hasselman-Johnson formulae is the dependence of the effective thermal conductivity on the particle radius a, and the boundary conductivity h_c [W/(m^{2}K] which is the reciprocal of interfacial thermal resistance [7]. The authors derived expressions for a continuous matrix phase with dilute concentrations of dispersions with spherical (6), cylindrical (7) and flat plate geometry (8). Expressions (6) and (7) are equivalents of Maxwell's result for spheres (2) and Rayleigh's result for cylinders perpendicular to the heat flow (5). Expression (8) is for flat plate dispersions oriented perpendicular to the heat flow [5]. Independently, Benveniste and Miloh obtained similar expressions for composites with ITR using micromechanical analysis [18, 19].

$$k_{eff} = k_m \frac{\left[2\left(\frac{k_1}{k_m} - \frac{k_1}{ah_c} - 1\right)\phi + \frac{k_1}{k_m} + \frac{2k_1}{ah_c} + 2\right]}{\left[\left(1 - \frac{k_1}{k_m} + \frac{k_1}{ah_c}\right)\phi + \frac{k_1}{k_m} + \frac{2k_1}{ah_c} + 2\right]} \tag{6}$$

$$k_{eff} = k_m \frac{\left[\left(\frac{k_1}{k_m} - \frac{k_1}{ah_c} - 1\right)\phi + \left(1 + \frac{k_1}{k_m} + \frac{k_1}{ah_c}\right)\right]}{\left[\left(1 - \frac{k_1}{k_m} + \frac{k_1}{ah_c}\right)\phi + \left(1 - \frac{k_1}{k_m} + \frac{k_1}{ah_c}\right)\right]} \tag{7}$$

$$k_{eff} = \frac{k_1}{\left[\left(1 - \frac{k_1}{k_m} + \frac{2k_1}{ah_c}\right)\phi + \frac{k_1}{k_m}\right]} \tag{8}$$

3.4. Bruggeman model

Mathematical formalism proposed by Bruggeman [20] and refined by Landauer [21] allows one to estimate many effective properties of heterogeneous materials, for example electrical and thermal conductivities, thermal diffusivity, magnetic permeability or electric permittivity. The main approach of this theory assumes that a composite material may be constructed incrementally by introducing infinitesimal changes to an already existing material. This approach leads to differential equations, and therefore it is called the differential effective medium theory or differential effective medium scheme (DEM).

The advantage of this scheme is that it covers a wide spectrum of materials, e.g. composites, nanofluids, porous materials, aerosols, space dust etc. It also gives formulae for multi-component systems, in addition to the classical case of two components. Among classical EMTs, the Bruggeman scheme is considered the most accurate for high filler volume fractions. Using Bruggeman's approach, Every and Tzou [3] obtained an expression for effective thermal conductivity of particulate composites by modifying Benveniste's result [19]. Their formula is:

$$(1-\phi)^3 = \left(\frac{k_m}{k_{eff}}\right)^{(1+2\alpha)/(1-\alpha)} \left(\frac{k_{eff} - k_1(1-\alpha)}{k_m - k_1(1-\alpha)}\right)^{3/(1-\alpha)} \tag{9}$$

where α is a dimensionless parameter depending on ITR between filler and matrix. It is defined as $\alpha = a_k/a$, where a is the particle radius, and a_k is the Kapitza radius ($a_k = R_{int}k_m$).

The formula has been verified by experimental measurements on ZnS/diamond composites with two particle sizes and varying percentages of filler. Assuming that filler conductivity is much greater than that of the matrix, which is true for ZnS/diamond composite, expression (9) may be simplified to:

$$\frac{k_1}{k_m} = \frac{1}{(1-\phi)^{3(1-\alpha)/(1+2\alpha)}} \tag{10}$$

A comparison of the theoretical results given by equation (10) and experimental results is presented in Fig 4.

Fig. 4. The ratio of ZnS/diamond composite thermal conductivity to the conductivity of pure ZnS matrix as a function of diamond filler volume fraction ϕ, where: dashed line – measurement results for two sizes of filler particles, solid line – theoretical predictions based on (10), a – average particle radius. Adopted from [3].

As seen in Fig. 4., for particles with greater average radius $a = 2\mu m$, the value of parameter α was estimated to be equal to 0.75. For smaller particle sizes, the experimental results lay below the limit of infinite α. This inconsistency may be caused by the nonspherical shape of diamond particles while the formula is for spheres [3]. Despite this discrepancy, the model proposed by Every and Tzou was a big step forward in understanding the influence of microstructure on the properties of particulate composites.

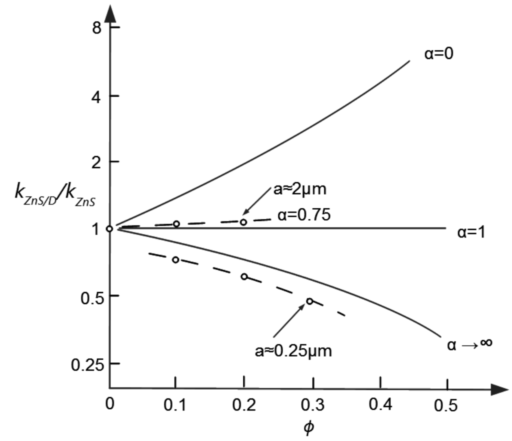

Figure 4: The ratio of ZnS/diamond composite thermal conductivity to the conductivity of pure ZnS matrix as a function of diamond filler volume fraction ϕ, where: dashed line – measurement results for two sizes of filler particles, solid line – theoretical predictions based on (10), a – average particle radius. Adopted from [3].

Also other authors, like Tavangar et al. [22], used DEM to obtain new expressions for effective thermal conductivity of composite material. The formula for particulate composites, derived by Tavangar et al. may be written as:

$$(1-\phi) = \frac{(k_m)^{1/3}\left(k_1 k_{eff} R_{int} + a k_{eff} - a k_1\right)}{\left(k_{eff}\right)^{1/3}\left(k_1 k_m R_{int} + a k_m - a k_1\right)} \tag{11}$$

Tavangar and his co-authors confirmed, by comparing with experimental results, that the differential effective medium scheme is a better tool for estimating effective thermal conductivity than the Maxwell-Eucken approach. They advised caution when using Maxwell-based schemes, including the popular Hasselmann-Johnson model, for estimating the effective thermal conductivity of composite, especially in the case of a high effective phase contrast between composite constituents. The effective phase contrast is defined as:

$$\gamma = \frac{k_{eff}}{\left(1 + \frac{k_1 R_{int}}{a}\right) k_m} \tag{12}$$

Their study shows that the results obtained with DEM were close to experimental in the whole examined range of γ (2 to 8), whereas the Hasselmann-Johnson model failed above $\gamma = 4$.

3.5. The Lewis-Nielsen model.

This empirical model is quite popular in the literature and gives relatively good results even though its equations do not include ITR. It was created for moderate filler volume fractions (up to 40%). For higher values it becomes unstable [10].

The advantages of the Lewis-Nielsen model are its simplicity and coverage of a wide range of particle shapes and patterns. The effective thermal conductivity of a composite according to the Lewis-Nielsen model is given as:

$$k_{eff} = \frac{1 + AB\phi}{1 - B\psi\phi} \tag{13}$$

where

$$B = \left(\frac{k_1/k_m - 1}{k_1/k_m + A}\right) \tag{14}$$

$$\psi = 1 + \left(\frac{1-\phi_m}{\phi_m^2}\right)\phi \tag{15}$$

In equations (13, 14, 15), k_m is the thermal conductivity of the matrix, k_1 the thermal conductivity of the filler, ϕ is filler volume fraction, ϕ_m is maximum filler volume fraction (see Table 2; for an explanation of packing types see [23]) and A is shape coefficient for the filler particles (see Table 3).

Table 2: Maximum packing fractions for different arrangements [24]

Shape of particles	Type of packing	ϕ_m
Spheres	Face-centered cubic	0.7405
	Hexagonal close	0.7405
	Body-centered cubic	0.6
	Simple cubic	0.524
	Random close	0.637
	Random loose	0.601
Rods or fibers	Uniaxial hexagonal close	0.907
	Uniaxial simple cubic	0.785
	Uniaxial random	0.82
	Three-dimensional Random	0.52

3.6. Percolation model

When increasing the amount of filler per unit volume, one eventually reaches a point at which particles of filler begin to contact. Assuming that the filler is highly conductive, heat transfer is easier between two contacting particles than between the particle and the matrix. With increasing filler fraction, chains of connected conductive particles begin to appear (see the schematic in Fig. 5).

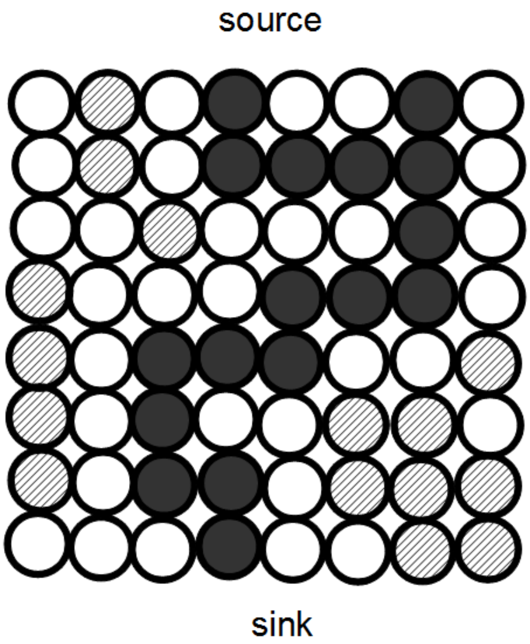

Figure 5: A scheme of heat transfer enhancement, in particulate composite material, due to percolation. High-conductivity particles, forming a continuous conductive chain from source to sink, are depicted as black. White is for low-conductivity particles, and hatching is for high-conductivity particles which do not form a continuous chain from source to sink.

These chains act as channels with increased heat conduction.

Table 3: Values of A for several Dispersed Types [24]

Shape	Aspect ratio of particles (length/diameter)	*A*
Spheres	1	1.5
Randomly oriented rods	2	1.58
	4	2.08
	6	2.8
	10	4.93
	15	8.38

Formation of such conductive channels causes a significant increase in the effective thermal conductivity of the material. This effect is visible as a shift from a flat to a steep slope of the effective thermal conductivity plotted versus filler volume fraction. The point or volume fraction at which this shift occurs is known as the percolation threshold [4].

The term percolation was initially used to describe the passing of a liquid through a porous substance or small holes, but then its meaning was expanded to describe the phenomenon of formation of conducting channels in many types of transport problems, e.g., electric circuits, public transport or spread of a disease. The example of percolation in composite material is schematically presented in Fig. 5, where a chain of highly conductive filler particles enhances heat transfer between heater and sink.

In general, effective medium approximations fail to predict the properties of a multiphase medium close to the percolation threshold. Efforts have been made to overcome this flaw. One of the most effective ways of percolation modeling is numerical simulation.

A numerical model allowing the prediction of effective thermal conductivity of a composite material including modeling of percolation and ITR was proposed by Devpura et al. [4, 10]. Their algorithm uses matrix algebra to represent the material and perform the calculations. The material is assumed to be made of cubic building blocks whose side is equal to the filler particle size. The block may be either matrix block or a filler particle block. The sphericity of the particles is introduced by means of an additional parameter. The conductivity of filler blocks is set to be much greater than the conductivity of the matrix. Interfacial thermal resistance is assigned to the walls for which the material on one side of the wall is different than the material on the other side. The value of ITR is estimated using AMM, therefore it includes only the TBR component.

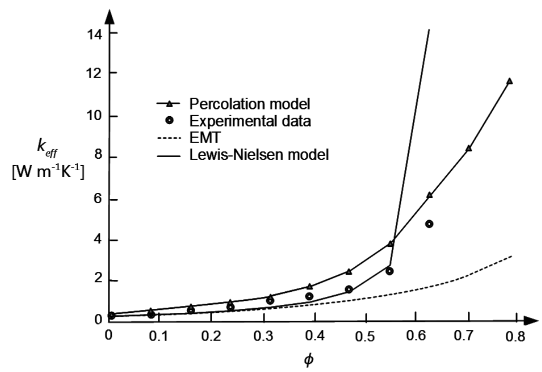

Figure 6: Effective thermal conductivity of bimodal distribution of Al_2O_3 filler (65:9 μm) in a polyethylene matrix as predicted by different models, compared with experimental results. ϕ – filler volume fraction Adopted from [10].

Despite the simplicity of the model, its predictions are in good agreement with experimental results. In Fig. 6, predictions of effective thermal conductivity of bimodal distribution of Al_2O_3 filler (65:9 μm) in a polyethylene matrix by various popular models are compared with experimental results.

As we can see, the Nielsen model [25] becomes unstable above a 40% filler volume fraction, whereas the EMT model [26] underpredicts the thermal conductivity. On the other side, the percolation model proposed by Devpura et al. [4] follows the trend of the experimental curve reasonably well, allowing one to predict the upper bounds of effective thermal conductivity for higher filler volume fractions.

Devpura et al. conducted insightful analysis of the influence of ITR and particle size on the effective thermal conductivity calculated by their percolation model. To characterize the value of ITR they utilized the Biot number, which, for a particle of spherical shape, is written as:

$$Bi = \frac{R_{int}k_m}{2a} \tag{16}$$

where a is the particle radius.

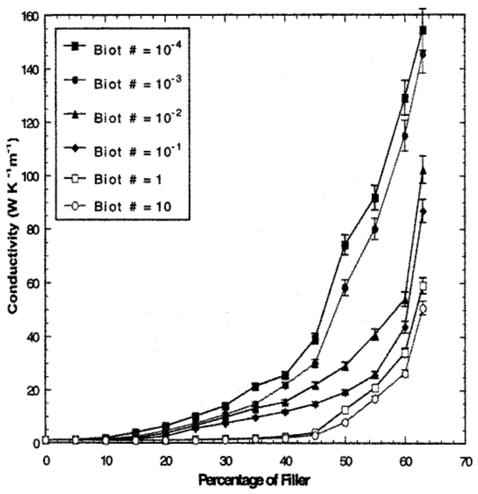

Figure 7: The effective thermal conductivity of alumina filler particles in the polyethylene matrix as a function of filler volume fraction for different Biot numbers[1] [4].

Fig. 7[1] and 8[2] present the results of conducted simulations. It is clear that a higher Biot number results in lower effective conductivity and also increases the percentage of filler required to achieve the percolation threshold. The Biot number may be increased either by increasing the value of ITR, or by decreasing the particle radius a. As observed from simulations, using filler with $Bi > 1$ causes a drop in the effective thermal conductivity of the composite to below the conductivity of the pure matrix, even though

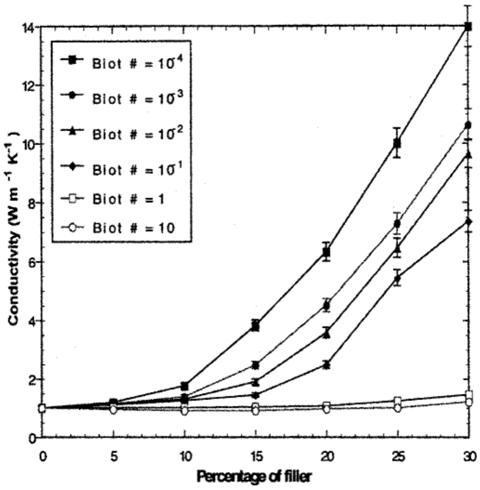

Figure 8: The effective thermal conductivity of alumina filler particles in the polyethylene matrix as a function of filler volume fraction for different Biot numbers. Blow-up for low volume fractions[2] [4].

the ratio of filler to matrix conductivities is 1000:1. Knowing the value of ITR, we are able to calculate the particle radius for which $Bi = 1$ (critical radius). Adding smaller particles to the matrix will cause reduction of the effective thermal conductivity instead of enhancement. The reviewed percolation model requires full numerical simulation, nevertheless its numerical cost is rather low and the predictions are accurate. The simplicity of the model is another advantage making it a useful and attractive tool for the estimations required in the process of composite material design.

3.7. Dynamic methods

Most classical models assume steady conduction in the composite media. However, in many real-life applications, such as the aerospace and automotive industries, extremely high temperatures occur and a non-steady heat-flux is more common. Modeling such states is rather complex and therefore, to date, very little work involving non-steady effective thermal properties has been done. Nevertheless, a few approaches have appeared.

For example, Monde and Mitsutake [27] developed a method for determining the thermal diffusivity of solids using an analytical inverse solution for unsteady heat conduction. With the aid of modulated photothermal techniques, Salazar et al. [28] studied the effective thermal diffusivity of aligned circular cylinders embedded in a matrix made of different material. Fang [29] investigated the propagation of a thermal wave in matrix composite materials with high volume concentrations of particles by using a combination of quasicrystalline approximation and Percus–Yevick correlation function.

[1]Reprinted by permission of the publisher (Taylor & Francis Ltd, http://www.tandf.co.uk/journals) from: A. Devpura, P. E. Phelan, R. S. Prasher.: Size effects on the thermal conductivity of polymers laden with highly conductive filler particles, Microscale Thermophysical Engineering, vol. 5, p. 177, 2001.

[2]as before

The thermal wave method is frequently utilized. In 2009 Fang et al. [30] applied the thermal wave method to investigate the unsteady effective thermal conductivity of particular composites with a functionally graded interface. The analytical solution for the non-steady effective thermal conductivity of the composite with coated particles is presented in [31, 32]. The scattering of thermal waves in such material is investigated theoretically by the wave functions expansion method. Comparison with the steady effective thermal conductivity demonstrates the validity of the dynamical thermal model.

3.8. Modeling of porous media

Porous media, like silica aerogels, represent a different class than typical composite materials. One may consider the porous material as a composite system consisting of a solid matrix and gaseous inclusions. However, the heat transfer through the gas and the solid-gas boundary is significantly different than through a solid inclusion and the solid-solid boundary.

The total thermal conductivity of porous material arises from solid conduction, gas convection and conduction, and radiation [33]. One should note that gas convection is usually negligible because of the small pore sizes. Also, if the pore size is smaller than about 80 nm (the mean free path for air particles at ambient temperature and atmospheric pressure), the gas conduction is strongly suppressed [34]. In many cases, the solid fraction in such materials is very small, and the heat paths in the solid structure are insufficiently long and complex to assure good solid conduction. Because of these properties, porous materials are usually used for insulation. Among modern thermal insulations, silica aerogels possess extremely small total thermal conductivities, which are even lower than that of air under ambient conditions [35].

Due to the outstanding insulating properties of aerogels, there is a great technical interest and need for good models to predict their thermal conductivity. Early modeling approaches, like [34] utilize the fact that the effective thermal conductivity is an average quantity, and therefore some aspects of the microstructure of the material can be neglected in modeling. In these models, the random structure of an aerogel (See Fig. 9) is replaced by a regular structure (e.g. intersecting cylinders or rods arranged in a cubic array) and the effective thermal conductivity model including only gas and solid conduction is proposed.

In more recent modeling approaches there is a tendency to include more details of the material structure and heat transfer mechanisms. Zhao et al. [35, 36] proposed an analytical model that includes pressure-dependency of gaseous thermal conductivity and the secondary spherical porous particle aggregate structures (schematically shown in Fig. 9). The model also includes effects of particle size, pore and particle microstructures, and solid–gas coupling including quasi lattice vibrations for solid-like vibrating gas molecules in the gaps between adjacent secondary particles which were not included in previous models. For a better result, the authors developed a 3-D finite volume numerical model of the material to generate an approximately real silica aerogel structure [37] .

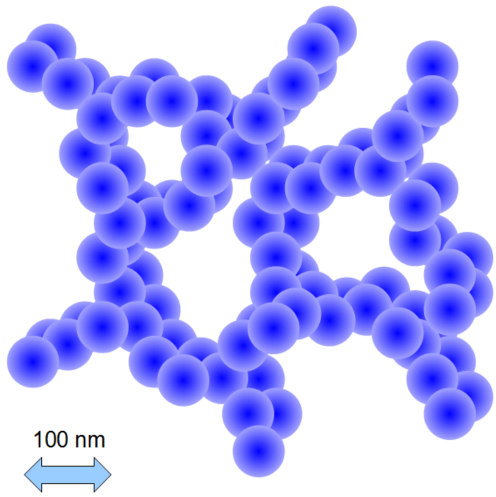

Figure 9: The schematic of an aerogel nanostructure. Silica aerogels obtained by sol–gel process and supercritical drying techniques are nanoporous, low-density, non-crystalline materials with large specific surface areas [35].

Porous insulation materials are usually brittle themselves but their poor mechanical properties can be enhanced by the addition of fiber reinforcement. The resulting material is multi-scale and multi-component. Effective properties of multi-component systems can be estimated by means of EMT, but for accurate results, complex modeling is required. Scale effects should be taken into account for thermal conduction through the nano-scale aerogel skeleton and pores, for which classical heat transfer theory cannot be applied. On the micron-scale, the addition of fibers and opacifiers improves the mechanical strength and reduces the infrared radiative transfer at high temperatures. On the other hand, they also increase thermal conduction since both have much higher solid conductivities than the pure aerogels [38]. A good example of a three-dimensional multi-scale model for the effective thermal conductivity of aeogel-based composite materials is given by Lu et al. [38] The model allows one to investigate the effects of non-ideal structures and high temperatures on the composite insulation performance.

3.9. Other methods

The finite element method (FEM) is frequently used to model heat transfer in composites as well as compute the effective thermal conductivity [26, 39–44]. It is often used as an additional method to verify the results obtained by other methods. The drawbacks of FEM are: long duration of complex geometry generation and meshing, requirement of high computing powers and time. On the other hand, modern commercial FEM software packages are readily available.

Apart from FEM, other numerical models had been proposed. Flaquer et al. [45] designed numerical algorithm to generate the geometry of synthetic diamond particles. They used the algorithm to build a model of a metal matrix composite with diamond filler. To compute the effective thermal conductivity, they proposed a line tracing method in which a set of lines, parallel to a specified axis, is conducted through a block of virtual composite material. For each line, the total lengths of segments within the matrix and segments within the filler are computed. Additionally, the number of filler-matrix interfaces crossed by the line is counted. On the basis of these parameters the total thermal resistance of the given material block is computed and finally its effective thermal conductivity is obtained.

The effective unit cell approach had been proposed by Ganapathy et al. [46]. Their method is characterized by low computing cost and can be applied to particle- and fiber-reinforced composites. The material is represented as a set of cubicoid building blocks. The filler particle may be represented by one or more blocks. Modeling of the particle's curvature is achieved by calculating the effective thermal conductivity of the particle from the effective medium theory. The method allows one to take into account the effects of percolation and ITR. The authors declared good agreement with experimental results (approx. ± 5% for a composite containing alumina fibers in a polyimide base). They also proved that their approach yields more accurate predictions than the Hasselman-Johnson model, especially close to and above the percolation threshold.

In the fields of nanocomposites and thin film technology, molecular dynamics simulations (MD) [47–50] are often used to determine conductive properties of materials and examine heat transfer at solid-solid interfaces. This type of modeling is quite useful for the study of nanostructured materials, but is not appropriate for greater scale composites where it is impossible to supply enough computational power to directly simulate large numbers of atoms.

4. Summary

Numerous analytical expressions for estimation of effective thermal conductivity of composite materials have been proposed since the 19^{th} century. Basic expressions apply to spherical filler particles [1, 2], but later models were developed to allow the inclusion of other particle shapes [2, 16, 25, 28, 51–53], the presence of particle coating [31, 32, 54–58] and the interfacial thermal resistance [3, 5, 18, 19, 59–61].

Models belonging to the class of effective medium approximations usually fail to predict the properties of a multiphase material close to and above the percolation threshold. It appears as a serious underestimation of effective thermal conductivity by these models (especially the popular Hasselman-Johnson model) in cases of higher filler volume fractions. Among EMTs, Bruggeman type approximations deal better with higher filler volume fractions than Maxwell-Eucken type approximations, but the latter are used more often due to their simplicity.

Numerical methods offer a simple way of modeling specific geometries (e.g. polyhedral particles) and phenomena which are difficult to include in classical mathematical analysis. The percolation model [4, 10] and effective unit cell approach [46] are good examples of intuitive methods with low numerical cost which can act as alternatives to the traditional finite element analysis and analytical approach.

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