

# A new structural model of effective thermal conductivity for heterogeneous materials with co-continuous phases

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## Abstract

A new structural model for a heterogeneous material with multiple continuous phases is proposed. The corresponding equation for effective thermal conductivity was derived using three methods. The new model is substantially different from the conventional five fundamental structural models (Series, Parallel, two forms of Maxwell–Eucken, Effective Medium Theory). The model has two applications. First, as a new fundamental structural model to produce composite models using the combinatory rules previously proposed by J.F. Wang, J.K. Carson, M.F. North, D. J. Cleland, A new approach to modelling the effective thermal conductivity of heterogeneous materials, *International Journal of Heat and Mass Transfer*, 49 (17–18) (2006) 3075–3083. Second, to narrow the bounds of the effective thermal conductivity for heterogeneous materials where the physical structure can be characterised into general classes.

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## 1. Introduction

Heterogeneous or composite solid materials are widely used in heat transfer processes and thermal management equipment. Effective thermal conductivity is one of the key thermophysical properties used for quantifying the thermal behaviour of heterogeneous materials. Carson [1,2] reviewed the relevant modelling approaches. A heterogeneous material's effective thermal conductivity is strongly affected by its composition and structure. For materials with simple physical structures, the effective thermal conductivity can be modelled using existing fundamental structural models like those given in Table 1 (for two-phase materials). However, for some materials with complicated physical structures, these basic models are not appropriate. An alternative method is to use empirical

models which are generally obtained by modifying simple models such as those listed in Table 1. Another common way of estimating effective thermal conductivity for heterogeneous materials with known microstructures is to make rigorous numerical simulations using the finite difference method, the finite element method, or other numerical techniques [3–5]. However, analytical models are preferred over numerical models in many applications due to their physical basis, rapid and low cost of calculation, and reasonable accuracy even when microstructure is uncertain. Recently, Wang et al. [6] proposed a new procedure for modelling complex materials as composites of five basic structural models (Series, Parallel, two forms of Maxwell–Eucken, Effective Medium Theory) using simple combinatory rules. The advantage of this method is that 26 new ‘composite’ models can be produced from these five basic models and each has a distinct physical basis. But this method is limited to the number of the basic structural models used for combination. In other words, the basic models adopted must include all of the heterogeneous

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## Nomenclature

A,B,C	different phases/components constituting a material	PS	polystyrene
BN	boron nitride	$q$	heat flux ( $\text{W m}^{-2}$ )
$c_1, c_2$	constants in Eq. (19)	$r$	radial distance (m)
CC	co-continuous model	$R$	radius of sphere (m)
EG	ethylene glycol	SS	stainless steel
EMT	effective medium theory	$v$	volume fraction of a phase
$f$	volume fraction of a phase	<i>Greek symbols</i>	
$H$	temperature gradient ( $\text{K m}^{-1}$ )	$\phi$	elevation direction
$k$	thermal conductivity of a phase ( $\text{W m}^{-1} \text{K}^{-1}$ )	$\theta$	azimuthal direction
$K$	effective thermal conductivity of a heterogeneous material ( $\text{W m}^{-1} \text{K}^{-1}$ )	<i>Subscripts</i>	
ME1	Maxwell–Eucken model with the first phase as the continuous phase	c	continuous phase
ME2	Maxwell–Eucken model with the second phase as the continuous phase	e	effective
$N$	number of phases	$i$	$i$ th phase
$P$	depolarisation factor	$j$	$j$ th principal axis
PB	polybenzoxazine	p	parallel model
		s	series model

material's basic structures that can contribute to the overall physical structure and behaviour of the material.

Both the authors' previous work [1,2,6] and several other studies [11–14] on effective thermal conductivity have demonstrated the importance of these five fundamental structural models on theoretical analyses and for developing more complex theoretical and empirical models. In particular, an important analysis is to define thermal conductivity bounds for certain classes of physical structure. Of the five fundamental structural models shown in Table 1, the series and parallel models represent a laminate (layered) structure of the phases. The other three models, two forms of ME (Maxwell–Eucken) [7,8] and EMT (Effective Media Theory)[9,10], are based on the phases being continuous and/or dispersed: the ME model represents one continuous phase and one or more dispersed phases, and EMT by all phases being mutually dispersed. The authors conjecture the existence of a new fundamental model that is characterised by all phases being continuous. This paper will derive a co-continuous structural model using three techniques, and will show its application for improving the prediction of effective thermal conductivity of heterogeneous materials.

## 2. Derivations of co-continuous structural model

### 2.1. Mathematical deduction

Brailsford and Major [12] proposed a procedure for mathematically deriving the EMT model by using the ME model for an isotropic material with two dispersed phases and one continuous phase. Taking a similar approach, we assume an isotropic material with two con-

tinuous phases A and B and one dispersed phase C. As shown in Fig. 1, the dispersed phase C is assumed to be split between the two continuous phases with a volume fraction  $f_c$  dispersed in phase A and a volume fraction  $(1 - f_c)$  dispersed in phase B. Using the ME model, the effective thermal conductivity of the two sub-assemblies can be expressed by:

$$K_{e1} = \frac{k_A v_A + k_c f_c v_c \frac{3k_A}{2k_A + k_c}}{v_A + f_c v_c \frac{3k_A}{2k_A + k_c}} \quad (1)$$

$$K_{e2} = \frac{k_B v_B + k_c (1 - f_c) v_c \frac{3k_B}{2k_B + k_c}}{v_B + (1 - f_c) v_c \frac{3k_B}{2k_B + k_c}} \quad (2)$$

Assuming a value of  $f_c$  so that the effective thermal conductivity of the overall material and the sub-assemblies are identical means that:

$$K_e = K_{e1} = K_{e2} \quad (3)$$

Also, the summation of the volume fractions for the three phases is:

$$v_A + v_B + v_C = 1 \quad (4)$$

Solving Eqs. (1)–(4) gives:

$$K_e = \frac{k_A v_A \frac{3k_B}{2k_B + k_c} + k_B v_B \frac{3k_A}{2k_A + k_c} + k_C v_C \frac{3k_A}{2k_A + k_c} \frac{3k_B}{2k_B + k_c}}{v_A \frac{3k_B}{2k_B + k_c} + v_B \frac{3k_A}{2k_A + k_c} + v_C \frac{3k_A}{2k_A + k_c} \frac{3k_B}{2k_B + k_c}} \quad (5)$$

which can be rewritten as:

$$K_e = \frac{k_A v_A \frac{2k_B + k_C}{3k_A} + k_B v_B \frac{2k_A + k_C}{3k_B} + k_C v_C}{v_A \frac{2k_A + k_C}{3k_A} + v_B \frac{2k_B + k_C}{3k_B} + v_C} \quad (6)$$

Eqs. (5) or (6) gives the effective thermal conductivity of a three-phase material assuming phase C is dispersed. Phase

Table 1  
The five fundamental structural effective thermal conductivity models for two phase materials

Model	Structure schematic	Effective thermal conductivity equation	Ref.
Parallel model		$K = v_1k_1 + v_2k_2$	
Maxwell–Eucken 1 (ME1) ( $k_1 = \text{continuous phase}, k_2 = \text{dispersed phase}$ )		$K = \frac{k_1v_1 + k_2v_2 \frac{3k_1}{2k_1 + k_2}}{v_1 + v_2 \frac{3k_1}{2k_1 + k_2}}$	[7,8]
EMT model		$v_1 \frac{k_1 - K}{k_1 + 2K} + v_2 \frac{k_2 - K}{k_2 + 2K} = 0$	[9,10]
Maxwell–Eucken 2 (ME2) ( $k_1 = \text{dispersed phase}, k_2 = \text{continuous phase}$ )		$K = \frac{k_2v_2 + k_1v_1 \frac{3k_2}{2k_2 + k_1}}{v_2 + v_1 \frac{3k_2}{2k_2 + k_1}}$	[7,8]
Series model		$K = \frac{1}{v_1/k_1 + v_2/k_2}$	

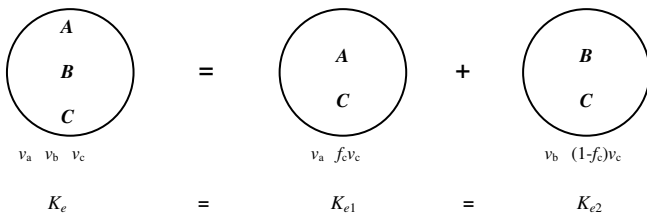


Fig. 1. A structural model with two continuous phases (A and B) and one dispersed phase (C).

C can be eliminated leaving a material comprising co-continuous phases A and B by assuming that phase C has a thermal conductivity equal to the average value of the conductivity of the two-phase structure comprising A and B only. Mathematically, this is achieved by setting  $k_c = K_e$  and  $v_c = 0$  in Eq. (6) and solving for  $K_e$  giving:

$$v_A \frac{(k_A - K_e)(2k_A + K_e)}{k_A} + v_B \frac{(k_B - K_e)(2k_B + K_e)}{k_B} = 0 \quad (7)$$

For a material with  $N$  co-continuous phases, a similar process can be used to derive the general model expression:

$$\sum_{i=1}^N v_i \frac{(k_i - K_e)(2k_i + K_e)}{k_i} = 0 \quad (8)$$

Eq. (8) can be further rewritten using the series and parallel models for a  $N$ -phase material giving:

$$K_e = \frac{K_s}{2} \left( \sqrt{1 + 8K_p/K_s} - 1 \right) \quad (9)$$

where:

$$K_s = \frac{1}{\sum_{i=1}^N \frac{v_i}{k_i}} \quad (10)$$

$$K_p = \sum_{i=1}^N k_i v_i \quad (11)$$

From Eq. (9), we can see that the effective thermal conductivity of a material with co-continuous phases has a simple model solution dependent only on the series and parallel model values.

### 2.2. Thermal field method

A more physically rigorous method to derive the co-continuous structural model is to use the thermal field method [15]. Consider an anisotropic spherical inclusion inserted into an isotropic material with effective thermal conductivity,  $K_e$ , subjected to a uniform temperature gradient  $\mathbf{H}$  as shown in Fig. 2a. The internal structure of the spherical inclusion is shown in Fig. 2b. It is composed of layers of the phases of the material in the inclusion. The laminate geometry means that each of the phases in the inclusion forms a co-continuous phase. The spherical shape also means that the effective thermal conductivities of the laminate spherical inclusion in the radial ( $r$ ), azimuthal ( $\theta$ )

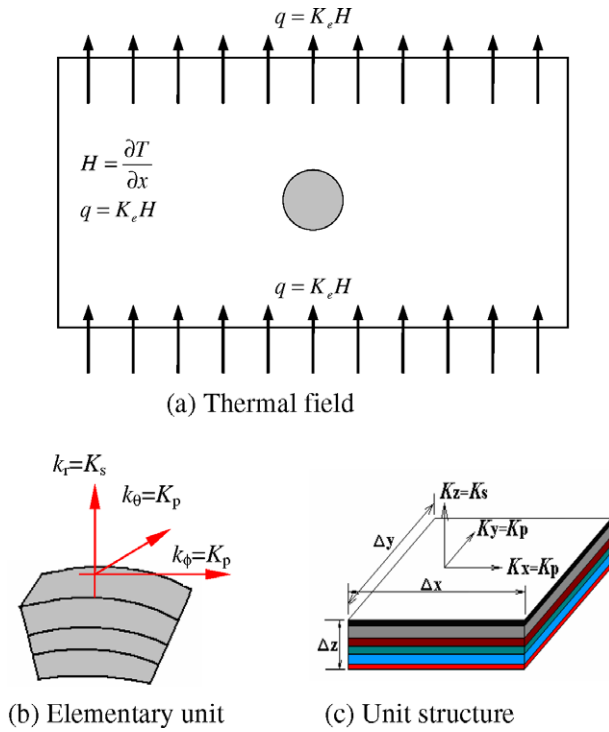


Fig. 2. A single spherical inclusion dispersed in an infinite medium with heat transfer in one direction: (a) thermal field, (b) elementary unit and (c) unit structure.

and elevation ( $\phi$ ) directions are  $K_s$ ,  $K_p$ ,  $K_p$  respectively (i.e. either series or parallel structures of the phases). In an isotropic material, the temperature is given by:

$$T = H \cdot r \cos \phi \quad (12)$$

To guarantee non-disturbance of the temperature field given in Eq. (12) by the spherical inclusion then:

$$T|_{r=R} = H \cdot R \cos \phi \quad (13)$$

$$q_r|_{r=R} = -K_s \frac{\partial T}{\partial r}|_{r=R} = -K_e H \cos \phi \quad (14)$$

Also for a steady state thermal field:

$$\nabla q = \text{grad}(q) = 0 \quad (15)$$

Heat fluxes in the radial and elevation directions are given by:

$$q_r = -K_s \frac{\partial T}{\partial r} \quad (16)$$

$$q_\phi = -\frac{K_p}{r} \frac{\partial T}{\partial \phi} \quad (17)$$

while heat flux in the azimuthal direction is identical to the elevation direction due to symmetry. Setting the solution of Eq. (15) to be of the form:

$$T = f(r) \cos \phi \quad (18)$$

where

$$f(r) = c_1 r^{\frac{-1+(1+8K_p/K_s)^{1/2}}{2}} + c_2 r^{\frac{-1-(1+8K_p/K_s)^{1/2}}{2}} \quad (19)$$

A finite solution for temperature at  $r=0$  implies that  $c_2 = 0$  in Eq. (19) and thus:

$$T = c_1 r^{\frac{-1+(1+8K_p/K_s)^{1/2}}{2}} \cos \phi \quad (20)$$

Using Eqs. (13) and (14), we can obtain the following expression for the effective thermal conductivity for the spherical inclusion in the uniform thermal field:

$$K_e = \frac{K_s}{2} \left( \sqrt{1 + 8K_p/K_s} - 1 \right) \quad (21)$$

Having a solution for one sphere, we can insert a second, third, and so on, and fill the space left between spheres with smaller and smaller spheres all constituted as the original sphere until the limit when the space is completely filled. Hence, Eq. (21) is the final result for the effective thermal conductivity of a material only comprising co-continuous phases of the spherical inclusion. The above derivation is based on the method proposed by Schulgasser [15] except that a co-continuous structure of the sphere is assumed.

### 2.3. Average field approximation

The co-continuous model can also be derived using the AFA (Average Field Approximation) method [16]. The AFA is widely used for calculating the electrical conductance or resistance in electrical fields. In the method, the heterogeneous material is regarded as consisting of anisotropic grains surrounded by a homogenous medium and the effective conductivity  $K_e$  is determined self-consistently using:

$$\sum_j P_j \frac{k_j - K_e}{k_j + 2K_e} = 0 \quad (22)$$

The principal axes of the grain ( $x$ ,  $y$ ,  $z$ ) are isotropically distributed in space. For a sphere or cube, the depolarization factors  $P_x$ ,  $P_y$ ,  $P_z$  all take the value of 1/3. If the conductivities in the principal axes of the grain,  $k_x$ ,  $k_y$ , and  $k_z$  are set to be  $K_p$ ,  $K_p$  and  $K_s$ , respectively, as shown in Fig. 2c and Fig. 3a and the homogeneous medium is assumed to be continuous then:

$$\frac{1}{3} \frac{K_p - K_e}{K_p + 2K_e} + \frac{1}{3} \frac{K_p - K_e}{K_p + 2K_e} + \frac{1}{3} \frac{K_s - K_e}{K_s + 2K_e} = 0 \quad (23)$$

Solving for  $K_e$ , gives:

$$K_e = \frac{K_p}{4} \left( \sqrt{1 + 8K_s/K_p} + 1 \right) \quad (24)$$

Eq. (22) was first obtained by Helsing and Helte [16], and Eq. (24) is a specific solution to Eq. (22) with the structural model shown in Fig. 3a.

However, the structure of the phases in the grain shown in Fig. 3a is not co-continuous. If all of the rectangular anisotropic grains are closely connected with the identical adjacent sides of other grains, the interconnected grains will become a laminated belt-shaped continuous phase with the structure shown in Fig. 3c from the overall point of view. This structure can be treated as if it formed an isotro-

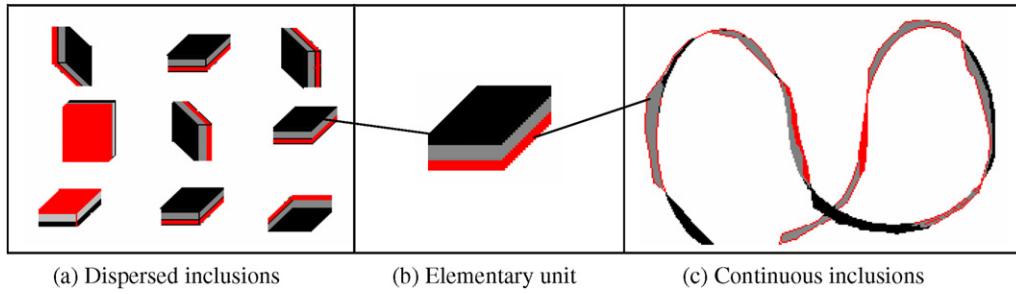


Fig. 3. Two structural models with anisotropic inclusions: (a) dispersed inclusions, (b) elementary unit and (c) continuous inclusions.

pic medium with conductivity  $K_e$  to be determined self-consistently if the principal axis conductivities are treated as being continuous. Again, taking  $K_x = K_y = K_p$ , and  $K_z = K_s$  in Eq. (22):

$$\frac{1}{3} \frac{K_p - K_e}{K_e + 2K_p} + \frac{1}{3} \frac{K_p - K_e}{K_e + 2K_p} + \frac{1}{3} \frac{K_s - K_e}{K_e + 2K_s} = 0 \quad (25)$$

Solving Eq. (25) gives the same earlier result for the co-continuous structure:

$$K_e = \frac{K_s}{2} \left( \sqrt{1 + 8K_p/K_s} - 1 \right) \quad (26)$$

Therefore, as shown in Fig. 3b, the structure of co-continuous model is anisotropic only on the micro-scale, and is effectively isotropic on the macro-scale. In addition, the co-continuous model is independent of the parallel or series model in structure although its model equation can be expressed in terms of the parallel and series model equations.

### 3. A new fundamental model?

The following features of the co-continuous model mean that could reasonably be classed as a ‘fundamental’ structural model:

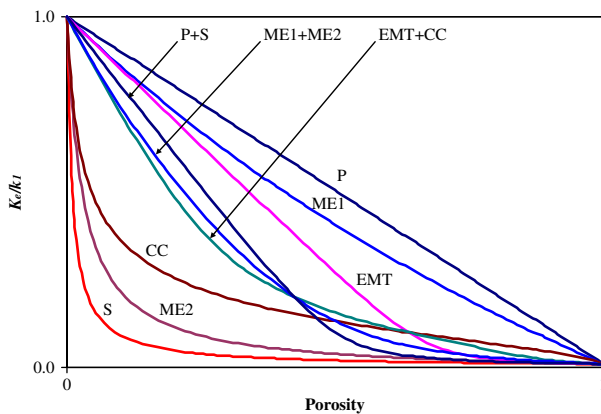


Fig. 4. Comparison of the co-continuous (CC) model with other five basic structural models and three composite models for a two-phase material with a conductivity ratio ( $k_1/k_2$ ) of 100 (see [6] for the detailed calculation method for the composite models).

- (1) The co-continuous model represents an isotropic material with a distinctive structure where all phases are continuous.
- (2) With the addition of the co-continuous model as a fundamental structural model, each basic model has a corresponding model with a complementary structure, i.e., the series and parallel models, the two

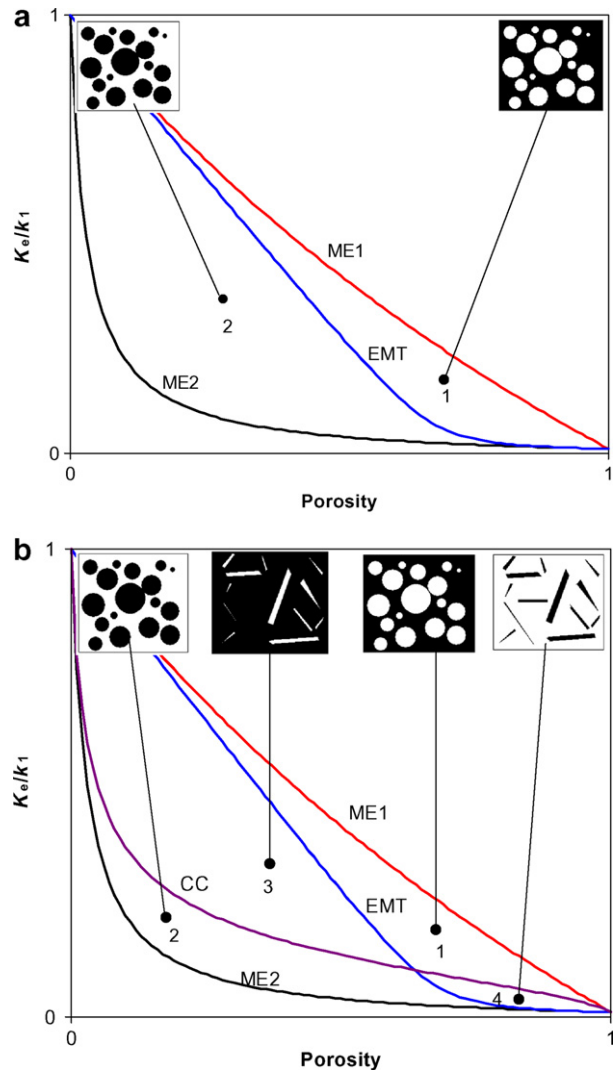


Fig. 5. Thermal conductivity bounds using the ME, EMT and CC models ( $k_1 > k_2$ ): (a) two zone bounds [14] and (b) four zone bounds using the CC model.

ME models, and the EMT and the CC (co-continuous) models. The EMT model is characterised by all of the phases being mutually dispersed, while the CC model is characterised by all of the phases being mutually continuous.

- (3) The CC model has a relation between effective conductivity and volume fraction that is very distinctive and different from the other fundamental models as shown in Fig. 4.

It should be noted that although the CC model is shown in Fig. 4 for the full range of volume fractions, not all the volume fractions will correspond to realistic physical structures. Just as the ME model, which is based on spheres dispersed randomly in a continuous matrix, does not have physical meaning above a dispersed phase volume fraction of 74% (the upper limit for the packing factor of identically sized spheres), likewise the CC structure will not necessarily have physical meaning for volume fractions close to 0 or 1.

**4. Combined structural models from co-continuous model**

Based on the volume fractions of each physical structure, Wang et al. [6] proposed a method to produce new

structural models using combinations of the five basic structural models shown in Table 1. The model combining the two ME models was proven to represent the Levy model which has been widely used for modelling the effective thermal conductivity of meats and other porous foods [17–19]. With the five basic models in Table 1, 26 new composite structural models were obtained.

If the CC model is added as another basic model, then 31 more composite structural models can be defined, i.e. 57 composite models plus six basic models. All 63 models are independent with different structures and different model values. As shown in Fig. 4, the three composite models, P + S, ME1 + ME2, and EMT + CC have different model values though each of the three composite models represents a type of intermediate structure between the two complementary structures.

The new composite models provide a new approach to modelling the thermal conductivity of some heterogeneous materials having known compositions and unknown but fixed microstructures. For example, most biological materials like bones have relatively fixed complex structures, and their thermal conductivity can be calculated with specific combined models. The application of the combined models

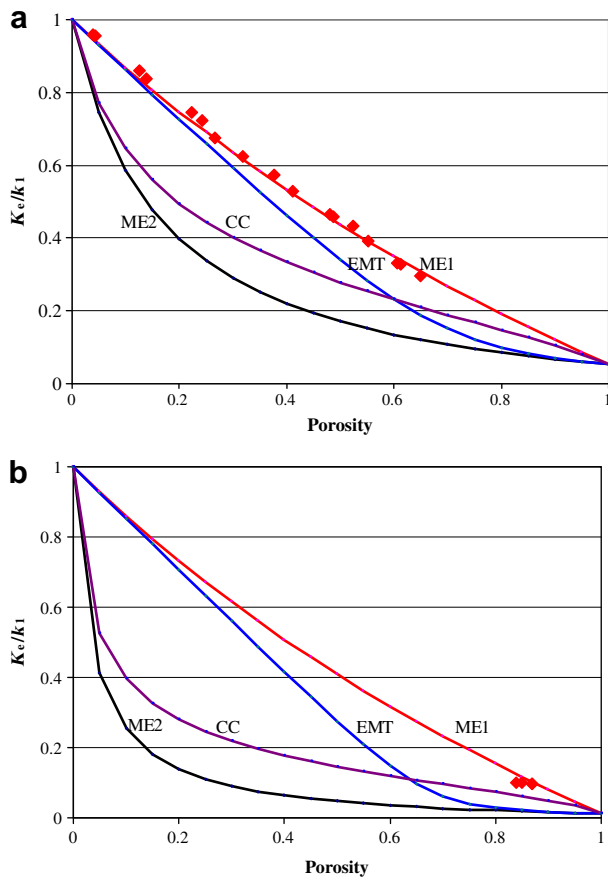


Fig. 6. Comparison between measured effective thermal conductivity and the predicted bounds given by the ME1, ME2, EMT and CC models for materials with structures in Zone 1: (a) food gel with PS balls[20] and (b) cellular ceramics with air pores [14].

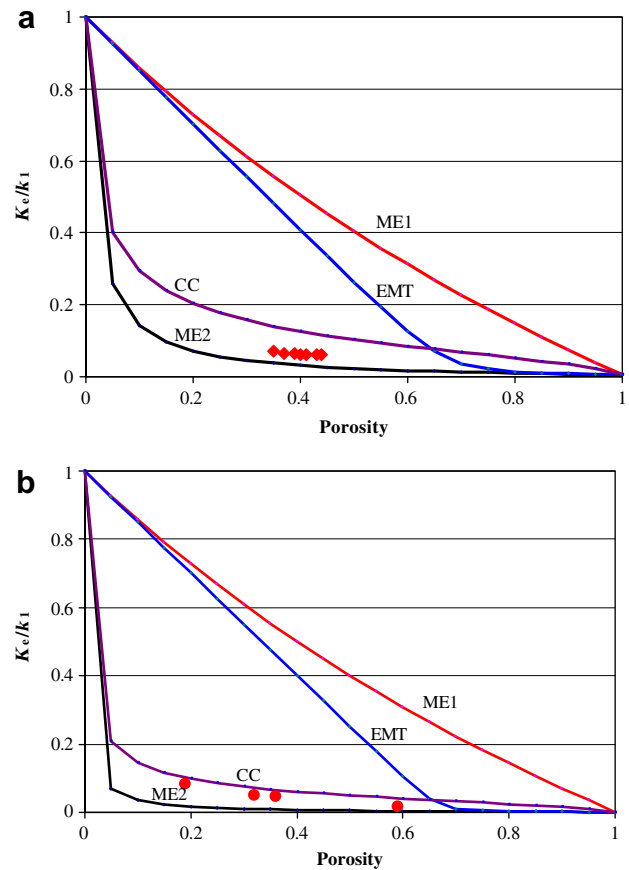


Fig. 7. Comparison between measured effective thermal conductivity [14] and the predicted bounds given by the ME1, ME2, EMT and CC models for materials with structures in Zone 2: (a) dry sand in air and (b) rock in air.



to the prediction of conductivity for a range of engineering materials and foods will be discussed in future work.

**5. New bounds using the co-continuous model**

For two-phase isotropic materials, the two ME models give the upper and lower bounds of the effective conductivity, known as the Hashin–Shtrikman (HS) bounds [11]. Carson et al. [14] introduced the EMT model into the HS bounds and obtained two zones with two different structures: internal porous structure and external porous structure, as shown in Fig. 5a. This approach effectively tightened the bounds for these structures to either between ME1 and EMT or between EMT and ME2.

With the addition of the CC model, as shown in Fig. 5b, the two zones within the HS bounds are divided into four zones. The different zones can be considered to comprise different phase structures and levels of connection and contact as follows.

(1) Zone 1: Internal porous structure. From the overall point of view, phase one is continuous and phase two is dispersed as separate near-spherical or cubic pores (inclusions) with some of the pores being con-

nected. Fig. 6 shows measured effective thermal conductivity data for a food gel filled with polystyrene (PS) balls and a cellular ceramics with air pores compared with predictions for the ME1, ME2, CC and EMT models. The thermal conductivity for these heterogeneous materials appears to fall in zone 1 consistent with their known physical structure.

(2) Zone 2: External porous structure (particle bed). From the overall point of view, phase two is continuous and phase one is dispersed as separate near-spherical or cubic particles (inclusions) with some of the particles being connected or contacted. Fig. 7 shows measured effective thermal conductivity data for two samples of dry sands compared with predictions for the ME1, ME2, CC and EMT models. Again, the thermal conductivity of the sand beds appears to fall into zone 2 which is consistent with their physical structure.

(3) Zone 3: In this zone each phase is self-connected/contacted as an entity where the lumped phase 1 is surrounded by thin belt-shaped phase 2. Fig. 8 shows measured effective thermal conductivity data for solidified porous rock and sandstone, and sintered SS (stainless steel) powders and alumina powders compared with predictions for the ME1, ME2, CC

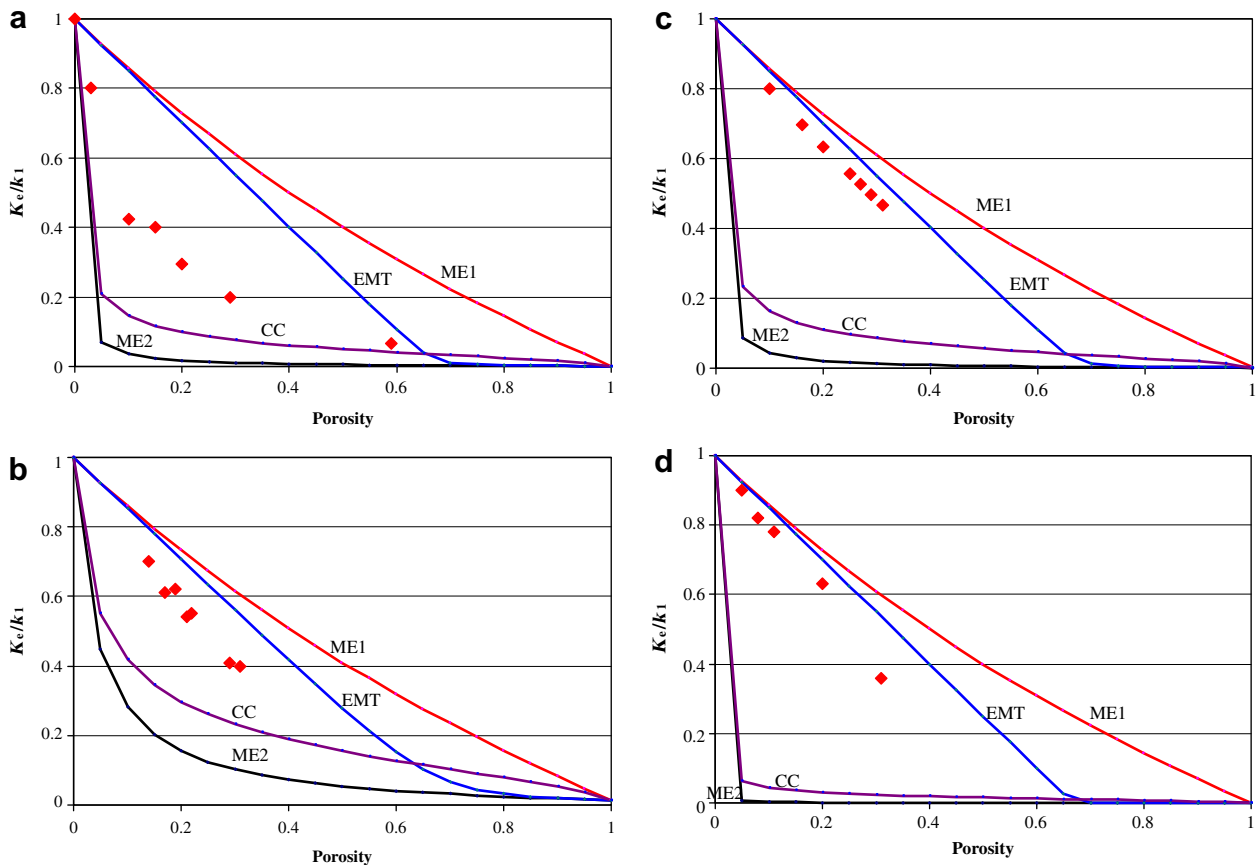


Fig. 8. Comparison between measured effective thermal conductivity and the predicted bounds given by the ME1, ME2, EMT and CC models for materials with structures in Zone 3: (a) solidified porous rock [14], (b) solidified porous sandstone [14], (c) sintered stainless steel (SS) powder in air [21] and (d) sintered aluminium powder in air [22].

and EMT models. Again, the zone 3 designation appears consistent with the structural characteristics for these materials.

- (4) Zone 4: In zone 4 each phase is self-connected/contacted as an entity where the lumped phase 2 is surrounded by thin belt-shaped phase 1. Fig. 9 shows measured effective thermal conductivity data for nanotubes in oil and nanoparticles with surfactant under Brownian motion in ethylene glycol (EG) compared with predictions for the ME1, ME2, CC and EMT models. Again the data appears to fall in zone 4 consistent with their structure.

Zones 3 and 4 are both physically interconnected structures, but the dominant lumped phase is different. There-

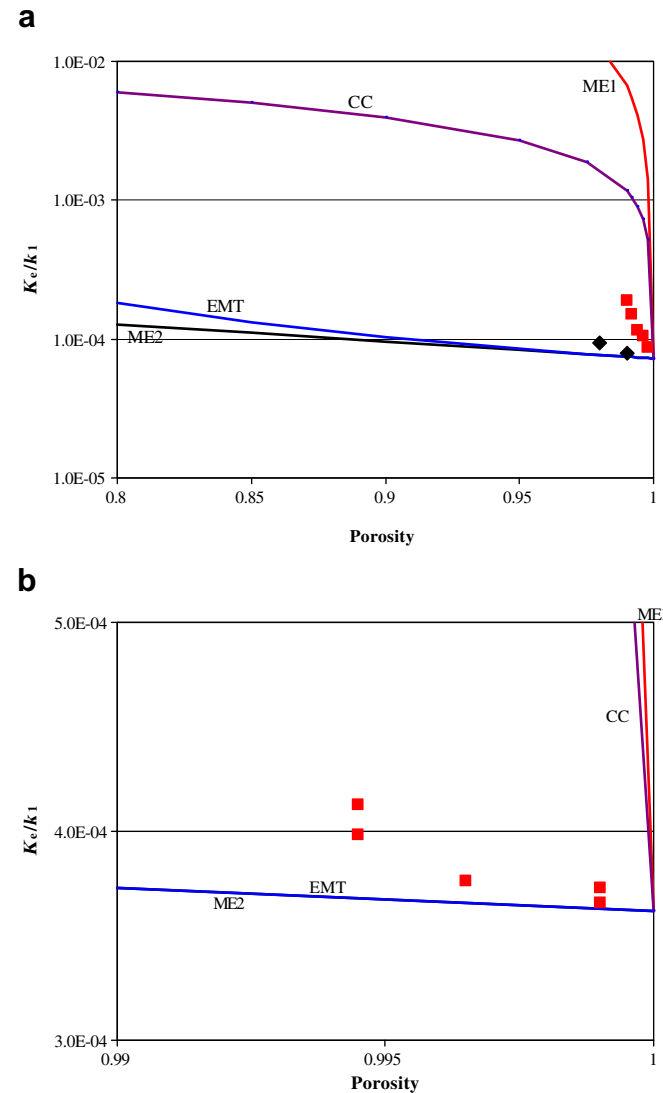


Fig. 9. Comparison between measured effective thermal conductivity and the predicted bounds given by the ME1, ME2, EMT and CC models for materials with structures in Zone 4: (a) nanotubes in oil (length/diameter > 2000, squares from [23], diamonds from [24]) (b). Nanoparticles with surfactant in ethylene glycol (EG) (Brownian motion and collision forming the connection between particles [26],  $d \approx 25$  nm)[25].

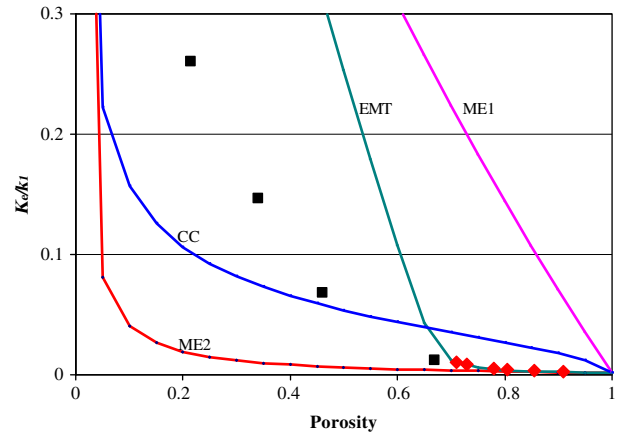


Fig. 10. Comparison between measured effective thermal conductivity for polybenzoxazine (PB) filled with platelet boron nitride (BN) (diamonds, [27]) and polystyrene (PS) filled with platelet BN (squares, [28]) and the predicted bounds given by the ME1, ME2, EMT and CC models.

fore, the CC model does seem to provide a way to narrow the thermal conductivity bounds based on physical structure considerations. It is interesting to see from Fig. 10 that different volume fractions of platelet boron nitride (BN) filled in PS and polybenzoxazine (PB) give effective thermal conductivities in zone 3 or zone 4 which is broadly consistent with which phase is likely to be dominant.

### 6. Conclusion

This paper has derived a new structural model of effective thermal conductivity for heterogeneous materials with multiple continuous phases by using mathematical deduction, a thermal field method, and an average field approximation. This new model has a distinctive structure which is substantially different from five conventional fundamental structural models (Series, Parallel, two forms of Maxwell–Eucken, Effective Medium Theory). The new model provides more structural models using the combinatory method proposed by the authors [6]. The model also provides narrower bounds of the effective thermal conductivity within the Hashin–Shtrikman bounds for heterogeneous materials where the physical structure can be characterised.

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