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Effective conductivity estimation of binary metallic mixtures

P. Karthikeyan, K.S. Reddy *

Heat Transfer and Thermal Power Laboratory, Department of Mechanical Engineering, Indian Institute of Technology Madras, Chennai – 600 036, India

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Abstract

The effective conductivity of binary metallic mixtures depends upon the concentration (c) of metallic mixtures and conductivity ratio (α). The binary metallic mixtures exist in a variety of shapes and sizes. No single theory can be expected to cover all the concentration and conductivity ranges. In the present work, a model has been proposed including both constant isotherms based unit cell approach and Bessel function based semi-empirical field solution approach. The non-dimensional effective conductivity (K) of macroscopically heterogeneous and anisotropic mixtures has been investigated. Another important aspect of the model is that it covers all ranges of concentration and conductivity ratio. The effect of concentration (c) on variable height of inclusion (h) has been studied. A comparison of the model has been made with two extreme bounds (parallel and series) and other well-known models, which gives a reasonable agreement. The model has also been compared with experimental data of various binary metallic mixtures such as Bi–Bi_2Pb, Bismuth–Tin, Mg_2Pb–Pb, Cadmium–Lead, Copper–Ferrous, Cu_2Sb–Sb, and Antimony–Lead. The conductivity estimated by the model for binary metallic mixtures is within 8% deviation from the experimental values.

Keywords: Effective conductivity; Unit-cell approach; Field solution approach; Macroscopically heterogeneous; Anisotropic; Variable height of inclusion in the unit cell

1. Introduction

The problem of estimating the conductivity of binary mixtures has attracted attention for almost a century. Only a few theories have been suggested and out of these only few have been compared with experiments. The Maxwell solution [1] is the starting point to find out the effective conductivity of the mixtures, but it is only valid for vanishingly small concentration of the dispersed phase. Lord Rayleigh [2] analyzed a cubical lattice with equal sized spheres at the corners. Bruggeman [3] proposed a model, to consider the properties of a composite medium with concentration greater than zero. Hashin and Shtrikman [4] proposed the most restrictive bounds for the value of effective conductivity. Zehner and Schlunder [5] proposed another model with particles in contact with each other and considering the effect of secondary parameters. An important failing in the Schlunder approach is that the deformation is taken only as a function of concentration, not as a function of the conductivity ratio. Series [6,7] and asymptotic [8] approaches for several regular arrays of spheres have been proposed. Raghavan and Martin [9] proposed a unit cell model that agreed exactly with field solutions and provided the basis for a fundamentally correct approach in the modeling of conductivity. The thermal conductivity of particles wetted with a binary mixture and packed beds of such particles has been investigated both theoretically and experimentally [10]. Hsiau [11] developed a correlation based on the kinetic theory of gases to estimate the effective thermal conductivity of binary mixture of granular materials. On the basis of the mean approximation method, the effective thermal conductivity of fibrous composite materials has been carried out by Barta [12]. Turian [13] developed a correlation based on the extrapolation and least square fitting to find out the thermal conductivity of coal-water mixtures. A two-dimensional steady state model to simulate the thermal behavior of copper and super alloy matrix composites has been carried out by Alcaraz Moreno [14].

The primary parameters influencing the effective conductivity of the binary metallic mixtures are conductivity ratio (α) and concentration (c). At present, there is no satisfactory solution

* Corresponding author. Tel.: +91 44 22574702; fax: +91 44 22570509, +91 44 22574652.
E-mail address: ksreddy@iitm.ac.in (K.S. Reddy).

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Nomenclature

c	volume concentration of dispersed phase
h	variable height of inclusion in the unit cell
K	non-dimensional effective conductivity $= k_{\text{eff}}/k_{\text{con}}$
k_{eff}	effective conductivity of binary system .. W/mK
k_{con}	conductivity of continuous phase ..... W/mK
k_{dis}	conductivity of dispersed phase ..... W/mK
K_{exp}	experimental non-dimensional conductivity
K_{max}	maximum non-dimensional conductivity
K_{min}	minimum non-dimensional conductivity
K_{pre}	predicted non-dimensional conductivity
K_{RM}	non-dimensional conductivity based on Raghavan–Martin model
K_{ZBS}	non-dimensional conductivity based on Zehner–Schlunder model
K_{Brug}	non-dimensional conductivity based on Bruggeman model
m, f, x	parametric constants

Greek symbols

α	ratio of conductivities $= k_{\text{dis}}/k_{\text{con}}$, $0 < \alpha < \infty$
β	transformed ratio of conductivities $= (\alpha - 1)/(\alpha + 2)$, $-0.5 < \beta < 1$
BesselM$[i, j]$	modified Bessel function of order i and argument j

for all ranges of α and c, because the neighbour interactions on the field produced higher order effects, which is difficult to model. In addition to the primary parameters there are secondary effects influencing the effective conductivity such as contact resistance, radiation, convection and Knudsen effect and also parameters like particle size, shape, location, size distribution, and orientation.

The above researchers have proposed models which do not take into account the wide range of α and c. Recognizing the need for an engineering solution and realizing that a solution may lie in isolating higher order interactions from the Maxwell interactions; modeling of Maxwell interactions in the simple way, a two-way approach to estimate the effective conductivity of binary mixtures is adopted here. In this paper a novel approach to predict the effective conductivity of binary mixtures based on the unit cell approach (constant isotherms) as well as on the semi-empirical field solution approach has been attempted. The model has been validated with experimental results for different binary mixtures and it predicts effective conductivity more accurately than the earlier proposed models.

2. Analytical model

The unit cell with a particle inclusion height 'h' as the equivalent representation of the binary mixtures is considered. It can be varying if the disperse medium concentration increases or decreases. The geometrical meaning of height of inclusion "h" is shown in Fig. 1. The earliest model of the unit cube described by Krischer [15]. The unit cell approach is based on the resistance approach (Ohms law model). The effective conductivity of binary metallic mixtures is determined by considering equivalent electrical resistances in parallel and series approach. It is assumed that the heat flow is one-dimensional and also the isotherms are straight and parallel to the direction of heat flow. The equivalent thermal network for binary mixtures based on constant isotherms is shown in Fig. 2.

The upper and lower limits to the conductivity of two-phase materials described by Wiener [16]. Raghavan and Martin [9] developed a unit cell model to calculate the conductivity based on parallel lines of heat flux. For systems, $\alpha < 20$,

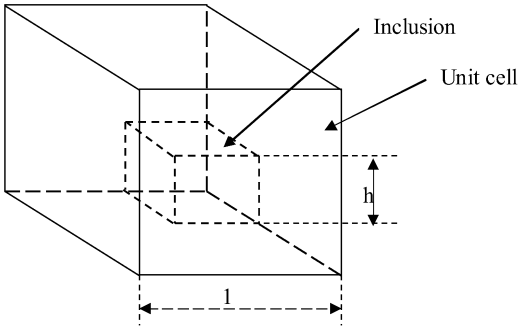

Fig. 1. Geometrical representation of height of inclusion in the unit cell.

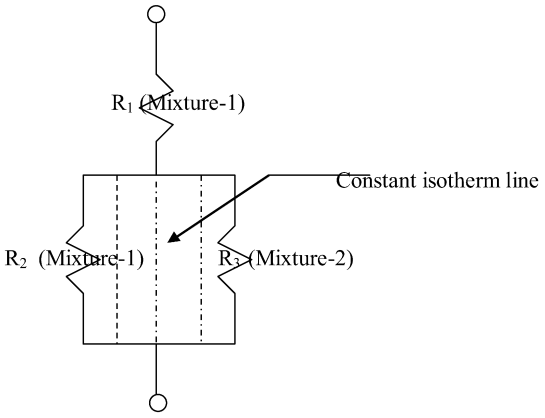

Fig. 2. Equivalent thermal network for binary mixtures.

$0.1 \leqslant c \leqslant 0.9$, based on the resistance approach, the effective conductivity under the conditions of constant isotherms is given by

$$K = 1 + \frac{3\beta c h}{h + \beta[3c(1-h) - h]} \tag{1}$$

The inclusion height 'h' may be expressed in terms of K, β and c

$$h = \frac{3\beta c(K-1)}{3\beta c - (1 - 3\beta c - \beta)(K-1)} \tag{2}$$

The 'h' is estimated by averaging it over the two most restrictive bounds proposed by Hashin and Shtrikman [4] and is given by

$$h = \frac{\int_{K_{\min}}^{K_{\max}} \frac{3\beta c(K-1)}{3\beta c-(1-3\beta c-\beta)(K-1)}\,\mathrm{d}K}{K_{\max}-K_{\min}} \tag{3}$$

where

$$K_{\max} = \frac{(1+2\beta)(1+2\beta c-\beta)}{(1-\beta)(1-\beta c+2\beta)} \quad \text{and} \quad K_{\min} = \frac{1+2\beta c}{1-\beta c}$$

Integrating Eq. (3), we obtain

$$h(\beta, c) = \frac{-1}{\zeta} + \frac{1}{\zeta^2\Delta}\log\left[\frac{1-\zeta(K_{\min}-1)}{1-\zeta(K_{\max}-1)}\right] \tag{4}$$

where $\zeta = (1-\beta-3\beta c)/(3\beta c)$ and $\Delta = K_{\max} - K_{\min}$.

The effective conductivity in terms of ζ and h is given by

$$K = 1 + \frac{h(\beta, c)}{1+\zeta h(\beta, c)} \tag{5}$$

The effective conductivity estimated from Eq. (5) shows large deviations from the actual values for higher values of α (> 20) because of higher order effects and higher distortions of the flow lines. We propose a new correlation based on the field solution approach for higher values of α (> 20). In the present work, a special case of binary metallic mixtures was considered. The composition of the mixtures has been assumed as one mixture being embedded in the other mixture in a spherical shape. Each sphere is in contact with six neighbours of mixture in the horizontal plane and the mixture above and below the vertical plane. It has been assumed that the conductivity of continuous medium is higher than the disperse medium conductivity of the mixture. So isotherms in the spheres intersect with the surface of the sphere at right angles. An approximate analytical solution to the steady state energy equation has been obtained for the idealized geometry.

The field solution approach is a result of Laplace's equations for the temperature field in the mixtures to obtain the effective conductivity. The energy balance can be applied to the differential volume between adjacent isotherms to yield an ordinary differential equation for the surface temperature and also obtained the expression for effective conductivity in terms of concentration (c) and conductivity ratio (α). The effective thermal conductivity of packed beds based on field solution approach has carried out by Dietz [17]. One of the major limitations of his expression was there is no dependency on the concentration of the dispersed phase. In the proposed correlation (modified field solution approach) the equation is based on concentration and conductivity ratio has been carried out [18]. A new function $G(f, \alpha)$ is defined for predicting the effective conductivity. The differential equation in terms of concentration and conductivity ratio has been formulated and solved by using mathematica software to obtain the correlation. Here Bessel $K[n, m]$ is the modified Bessel function of order 'n' and argument 'm'. In the proposed correlation, the function $G(f, \alpha)$ incorporates both concentration and conductivity ratio. The equation can be valid if the conductivity ratio $k_{\text{disp}}/k_{\text{con}} \gg 1$ then K is a function of $G(f, \alpha)$, If $k_{\text{disp}}/k_{\text{con}} = 1$ then K is a function of $G(f, 1)$.

The function $G(f, \alpha)$ is defined as:

$$\begin{aligned} G(f,\alpha) &= c^m\big\{f^x\alpha\big(\big(\sqrt{\tfrac{2}{f\alpha}}\,\mathrm{BesselK}\big[0,\sqrt{\tfrac{8}{f\alpha}}\big] \\ &\quad + \mathrm{BesselK}\big[2,\sqrt{\tfrac{8}{f\alpha}}\big]\big)\big/\,\mathrm{BesselK}\big[1,\sqrt{\tfrac{8}{f\alpha}}\big] - 1\big)\big\} \end{aligned} \tag{6}$$

For the parameters m, f and x, the respective expressions are given in following paragraphs. Also, a limiting condition is defined as:

$$y = \underset{c\to 1}{\mathrm{Limit}}\big[G(f,\alpha) - G(f,1)\big] \tag{7}$$

The effective conductivity for higher value of α is given by

$$K = 1 + \frac{\alpha}{y}\big[G(f,\alpha) - G(f,1)\big] \tag{8}$$

For the functions m, f and x, the best fitting parameters are used and expressions are assigned as follows:

For $20 \leqslant \alpha \leqslant 100$, $0.1 \leqslant c \leqslant 0.9$, we have $m = 2$, $x = 1.12$ and $f = \Gamma(1 + 2c/1 - c)$, where, $\Gamma = 1.5 - 0.03155\alpha + 0.000167\alpha^2$.

For $100 \leqslant \alpha \leqslant 1000$, $0.1 \leqslant c \leqslant 0.9$, we have, $m = 2$, $x = 1 + 2^\kappa/(13+\kappa)\kappa$ and $\Gamma = 100/\alpha^2 5^{(\kappa-1)}$, where, $\kappa = \alpha/100$. For $\alpha \geqslant 1000$ and moderate concentrations of the dispersed phase, we have, $m \to 0$, $x \to 1$, $f \to 0.141663$ and $K \to G(f, \alpha)$.

The proposed correlation for the effective conductivity of binary mixtures incorporates the Maxwell and the phase-inverted Maxwell solution in the concentration regions $0 \leqslant c \leqslant 0.1$ and $0.9 \leqslant c \leqslant 1.0$ respectively. So, for any value of α, the correlations for the effective conductivity for the given concentration regions are expressed as:

$$\text{for } 0 \leqslant c \leqslant 0.1, \quad K = \frac{1+2\beta c}{1-\beta c} \tag{9}$$

$$\text{for } 0.9 \leqslant c \leqslant 1.0, \quad K = \frac{(1+2\beta)(1-\beta+2\beta c)}{(1-\beta)(1+2\beta-\beta c)} \tag{10}$$

The effect of concentration (c) on variable height of inclusion (h) has been studied. The quantity h has a physical significance that when it increases, the spacing between the particles in adjacent unit cells in the direction of the heat flow becomes smaller and the resistance to the heat flow decreases; so the conductivity of mixtures increases. The conductivity ratio (α) varying from 0 to infinity, if α tends to infinity or $\beta = 1$ (infinitely conducting inclusions), the model is same as the parallel approach. If α tends to 0 or $\beta = -0.5$ (non-conducting inclusions), the model will become the series approach. For zero distortions of flux lines, the concentration tends to zero then inclusion height in the unit cell (h) tends to zero and β tends to zero then the effect of variable height of inclusion in the unit cell (h) approach to h_{Maxwell}.

3. Results and discussion

The effective conductivity of various types of binary metallic mixtures has been estimated using the proposed model.

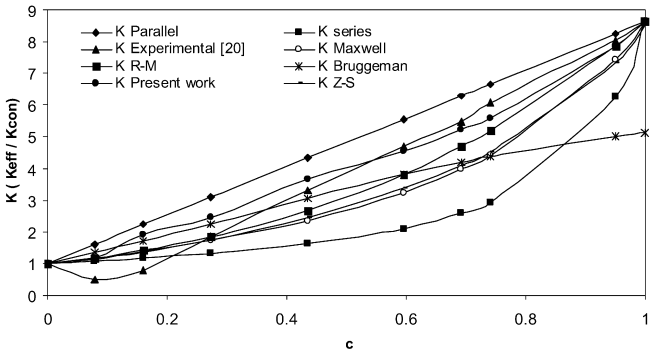

Fig. 3. Non-dimensional effective conductivity as a function of concentration of dispersed. phase for Bismuth–Tin system ($\alpha = 8.64$).

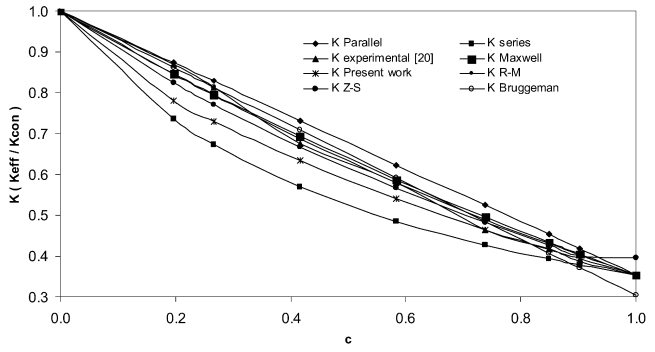

Fig. 4. Non-dimensional effective conductivity as a function of concentration of dispersed phase for Cadmium–Lead system ($\alpha = 0.3542$).

A comparison has been made with two extreme bounds (parallel and series) [6,7], and also with the predictions of standard models like the Maxwell [1], Bruggeman [3], Zehner and Schlunder [5], and Raghavan–Martin [9]. The variation of non–dimensional conductivity, K with concentration for Bismuth–Tin system with $\alpha = 8.64$ is shown in Fig. 3. A binary metallic mixture, Cadmium–Lead system ($\alpha = 0.3542$) in which, disperse medium conductivity is poorer than the continuous medium for which the variation of K with the concentration is shown in Fig. 4. The K ranges from unity to infinity while the concentration ranging from zero and unity. It is evident that for $\alpha > 1$, a higher value of K has been predicted than Maxwell whereas a lower value than the Maxwell for $\alpha < 1$. The variation of h with c for transformed conductivity ratio, $\beta > 0$ (Bismuth–Tin) and for $\beta < 0$ (Cadmium–Lead) are shown in Figs. 5 and 6. The elongation or contraction of the unit cell is due to higher order interactions in the mixtures. The present work line decreases initially, because of use the Maxwell equation at lower concentration (Fig. 6).

The experimental data for various binary metallic mixtures have been reported in earlier published literatures [19–23] and consider for the comparison with the model. The electrical conductivity of binary metallic mixtures investigated by Landauer [24]. The analogy of electrical conductivity has been adopted to estimate the effective conductivity of binary metallic systems. The K has been estimated for binary metallic mixtures such as Bi–Bi_2Pb, Bismuth–Tin, Cadmium–Lead, Mg_2Pb–Pb, Copper–Ferrous, Cu_2Sb–Sb and Antimony–Lead using present

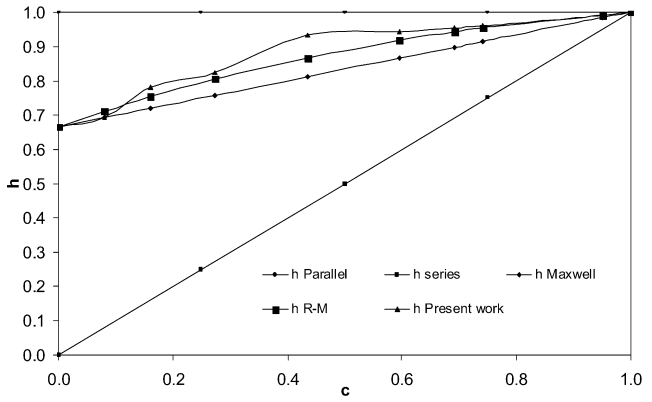

Fig. 5. Variable height of inclusion in the unit cell with concentration of dispersed phase for Bismuth–Tin system ($\alpha = 8.64$).

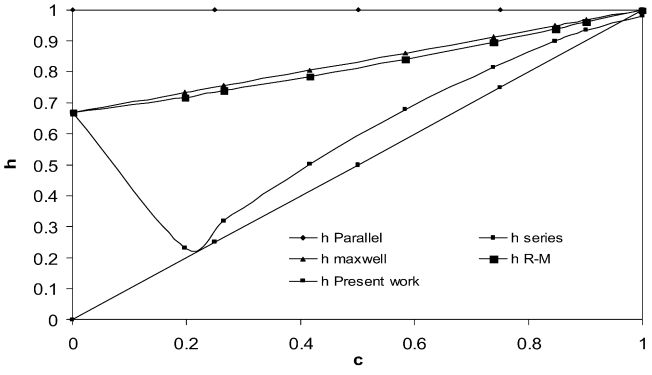

Fig. 6. Variable height of inclusion in the unit cell with concentration of dispersed phase for Cadmium–Lead system ($\alpha = 0.3542$).

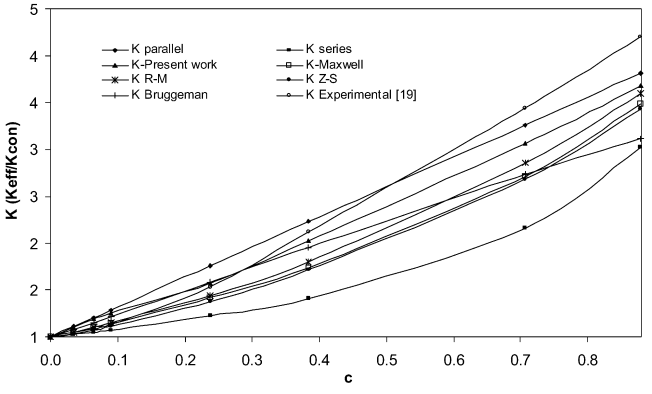

Fig. 7. Non-dimensional effective conductivity as a function of concentration of dispersed phase for Bi–Bi_2Pb system ($\alpha = 4.198$).

model. The variation of non-dimensional conductivity and variable height of inclusion in the unit cell with the concentration for Bi–Bi_2Pb system are respectively shown in Figs. 7 and 8. For the system like Bi–Bi_2Pb [19], the composition of the Bismuth is negligibly different from pure Bismuth, so the Bismuth is saturated with Tin. The Tin conductivity is ten times more than Bismuth conductivity. So, Tin is acting as a conductor and the Bismuth is acting as an insulator. The data obtained from model have average deviation of about 6.86% from the experimental values. The average deviations

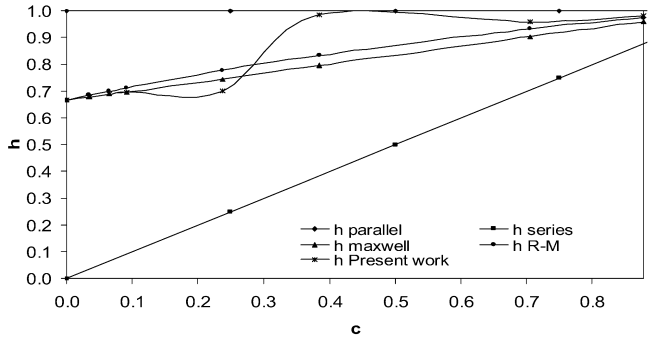

Fig. 8. Variable height of inclusion in the unit cell with concentration of dispersed phase for Bi–Bi_2Pb system ($\alpha = 4.198$).

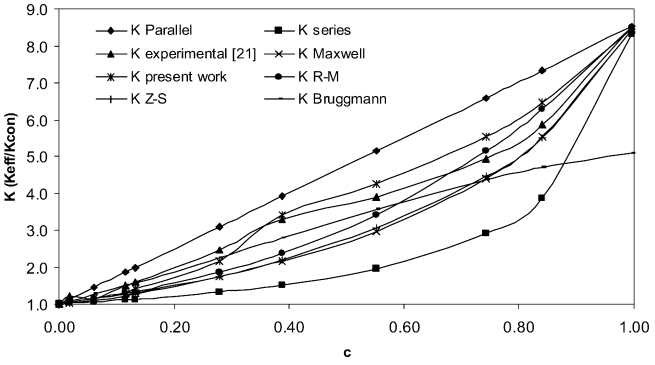

Fig. 9. Non-dimensional effective conductivity as a function of concentration of dispersed phase for Mg_2Pb–Pb system ($\alpha = 8.5358$).

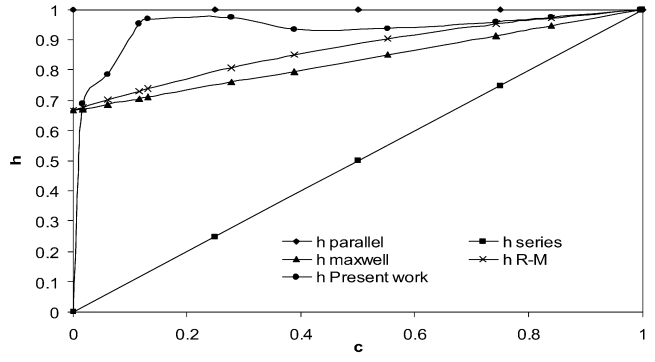

Fig. 10. Variable height of inclusion in the unit cell with concentration of dispersed phase for Mg_2Pb–Pb system ($\alpha = 8.5358$).

from the experimental data for Bruggeman, Raghavan and Martin and Zehner–Schlunder models have been predicted to be 9.08%, 6.88% and 9.48% respectively. A comparison of present model with the other models has been carried out for mixtures like Bismuth–Tin and Mg_2Pb–Pb (Figs. 3 and 9). The predicted values deviate from the experimental results about 7.53% for Bismuth–Tin system [20] and 8.05% for Mg_2Pb–Pb system [21]. The average deviation for Bruggeman, Raghavan–Martin and Zehner–Schlunder are calculated to be 22.27%, 9.02% and 15.42% respectively for Bismuth–Tin system and 11.48%, 11.00% and 14.45% respectively for Mg_2Pb–Pb system.

The mixture like Cadmium–Lead (pure cadmium mixed with pure lead), the proposed model deviates with experimental

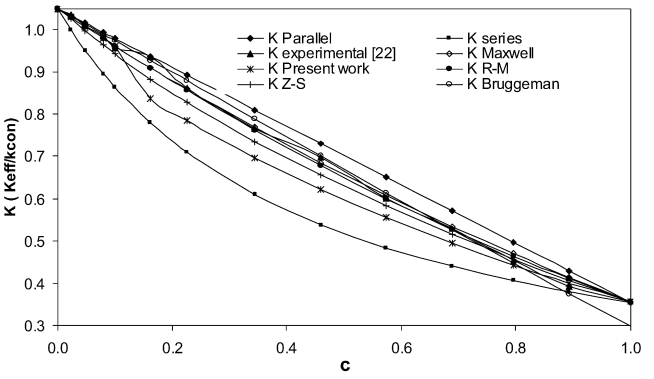

Fig. 11. Non-dimensional effective conductivity as a function of concentration of dispersed phase for Copper–Ferrous system ($\alpha = 0.3055$).

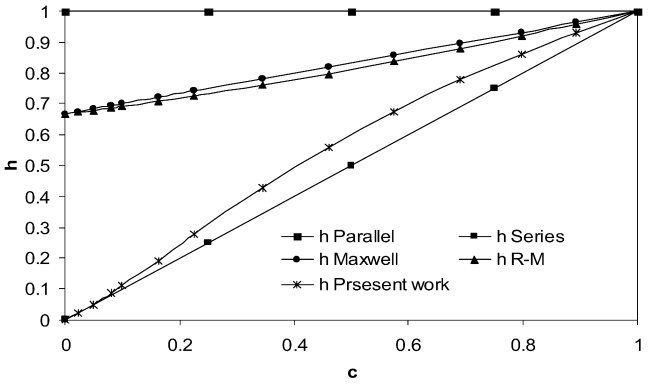

Fig. 12. Variable height of inclusion in the unit cell with concentration of dispersed phase for Copper–Ferrous system ($\alpha = 0.3055$).

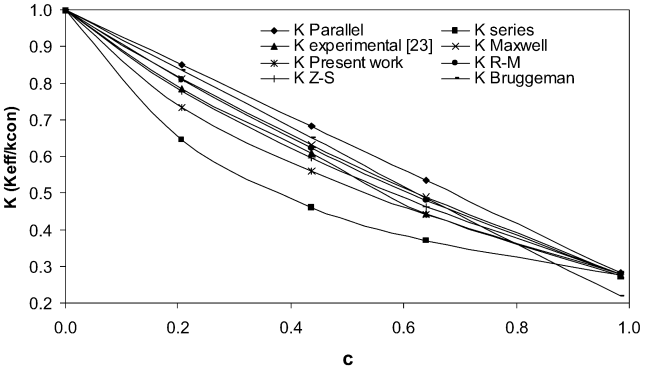

Fig. 13. Non-dimensional effective conductivity as a function of concentration of dispersed phase for Cu_2Sb–Sb system ($\alpha = 0.2727$).

values [20] with in the range of 3.96%. The average deviation for existing models [3,5,9] is 3.57%, 3.86% and 2.20% respectively. For the mixtures like Copper–Ferrous [22], Cu_2Sb–Sb [23] and Antimony–Lead [20], the proposed correlation for predicted K deviated from the experimental results with a deviation of 4.65%, 3.50% and 1.94% respectively (Figs. 11, 13 and 15). The average deviation for other models [3,5,9] are 2.71%, 2.74% and 1.30% for Copper–Ferrous system, 8.47%, 1.99% and 3.3% for Cu_2Sb–Sb system and 3.61%, 1.37% and 1.85% for Antimony–Lead system. The effect of concentration of the dispersed phase on variable height of inclusion in

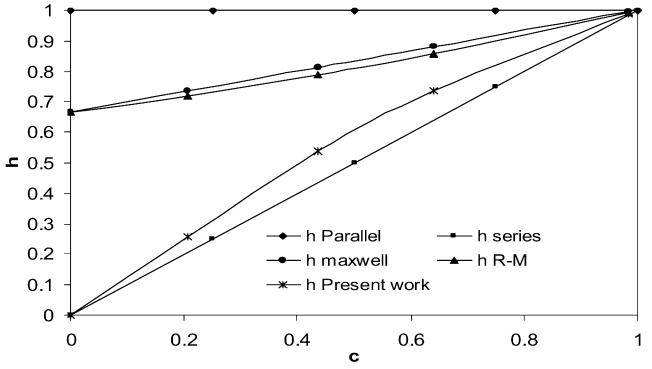

Fig. 14. Variable height of inclusion in the unit cell with concentration of dispersed phase for Cu_2Sb–Sb system ($\alpha = 0.2727$).

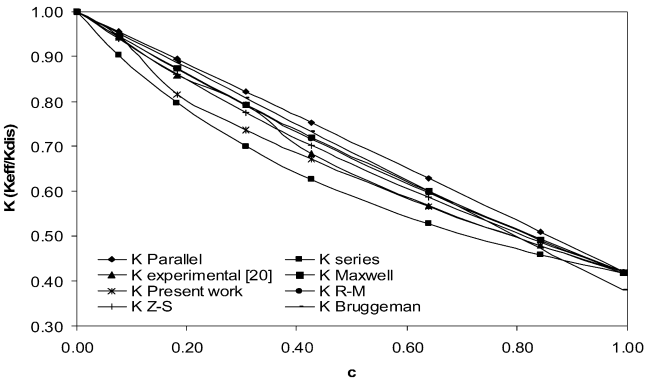

Fig. 15. Non-dimensional effective conductivity as a function of concentration of dispersed phase for Antimony–Lead system ($\alpha = 0.4168$).

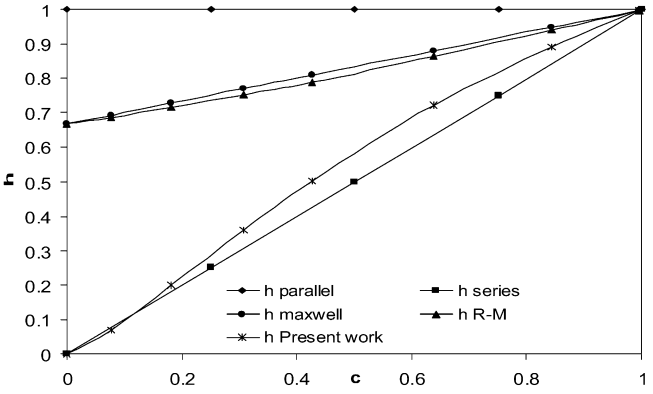

Fig. 16. Variable height of inclusion in the unit cell with concentration of dispersed phase for Antimony–Lead system ($\alpha = 0.4168$).

the unit cell has been studied for the mixtures like Bi–Bi_2Pb, Mg_2Pb–Pb, Copper–Ferrous, Cu_2Sb–Sb and Antimony–Lead (Figs. 8, 10, 12, 14 and 16). It is observed that with out any prediction, $\alpha > 1$ the present line is in between parallel and Maxwell line, similarly for $\alpha < 1$ the present line is in between Maxwell and series line. The results obtained from the model are in reasonable agreement with the other models and the deviation from the experimental values for various binary metallic mixtures is about 8%. The proposed model may effectively be used to predict all class of binary metallic mixtures.

4. Conclusions

For concentration varying from 0.10 to 0.9 and for lower α value ($\alpha \leqslant 20$), a model based on unit cell approach (for constant isotherm) has been proposed. The field solution approach is applicable for concentration varying from 0.1 to 0.9 for medium conductivity ratio ($\alpha > 20$). The relation between non-dimensional conductivity with the effect of concentration shows the physical corrections of the model and the effect of concentration with variable height of inclusion in the unit cell shows the transport properties of the mixtures. The model is applicable for various types of mixtures and it shows excellent agreement with the experimental results. The proposed model was compared with the standard models like Bruggeman, Zehner–Schlunder and Raghavan–Martin. For mixtures like Bi–Bi_2Pb, Bismuth–Tin and Mg_2Pb–Pb, the proposed model has shown better results than other models, for the remaining mixtures the proposed model is in fair agreement with the other models.

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