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International Journal of Heat and Mass Transfer 47 (2004) 1015–1021
International Journal of HEAT and MASS TRANSFER
www.elsevier.com/locate/ijhmt

Analytical model for the thermal conductivity of a binary bed of packed spheroids in the presence of a static gas, with no adjustable parameters provided contact conductance is negligible

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Received 3 January 2003; received in revised form 1 August 2003

Abstract

A model is given for the thermal conductivity of uncompressed, binary, packed-pebble beds of small and large solid spheroids in a static gas. It requires no adjustable parameters if the conductance of the solid is much greater than that of the gas, and the heat flow through the physical contacts between spheres is negligible compared to that from either gaseous conduction or radiation. The model is in excellent agreement with experiment, provided one assumes there is an average gas gap next to the large spheres, of width about 0.7 of the radius of a small particle.

1. Introduction

The binary beds considered here contain two sizes of particles, with one type of particle having typically 10 times the radius of the other so that the tiny particles can easily enter the spaces between the larger ones. Therefore, the tiny spheroids can literally be poured into the bed of large spheroids to intermix well. Such beds are of particular interest in the proposed breeder blanket in a fusion reactor, where a binary bed of large and small beryllium spheroids may be used as a neutron multiplier [1]. The cost of making accurate measurements on test beds is high in both time and apparatus, so considerable effort has gone into developing models which can be used predictively to optimize bed performance. The ideal model is one which can predict the thermal conductivity directly from measurements on the constituent particles. The model discussed here is shown to be successful in doing this, as tested against the one set of experimental data for which the particle parameters have been adequately measured.

An exact analytical expression for the bed conductivity is not possible even for an idealized, perfectly regular packing of the spheres because the conductivity varies spatially in a complex fashion with temperature and pressure, and has different dependencies for the solid, gas, and radiative contributions. As a result, existing models are of two basic types. The first is the finite-difference numerical model which can treat the three-dimensionality of the problem by dividing the bed into a great many cells with the temperature and heat flow matched at their boundaries, but it can be difficult to extract the relative importance of different conduction pathways from such computer models. Examples of numerical models for binary beds are those of Ades and Peddicord [2] (the AP model), and of Adnani et al. [3] (the ACRA model). Both of these models first calculate the thermal conductivity of a bed of the small spheroids and then treat them as a homogenous material interspersed between the large spheroids. However, the AP model still requires an experimentally determined fitting

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doi:10.1016/j.ijheatmasstransfer.2003.09.004

Nomenclature

A_{cpl}	base area of the close-packed unit cell of the large spheroids
A_{vl}	base area of the "void" cell for the large spheroids
D	diameter of a gas molecule
G_c	conductance though the contact points between contacting spheroids
G_{cp}	conductance of the close-packed region of the unit cell
G_g	conductance through the gas layer of average width w between the surface of a large sphere and the adjacent packed tiny spheres
G_{gvt}	gas conductance across the void region of the tiny spheres
G_i	gas conductance through the "inner" gap between contacting spheres where $2\lambda/3 >$ gap
G_o	gas conductance through the "outer" gap between contacting spheres where $2\lambda/3 <$ gap
G_r	conductance between contacting spheres by radiation
G_{rvt}	conductance by radiation across the void region of the tiny spheres
G_s	conductance through a solid sphere
G_t	overall conductance through the tiny spheres
G_v	conductance though the "void" region in the unit cell
h	average height of the short-range surface roughness
j	temperature jump distance at surface
k	Boltzmann's constant
K_0	thermal conductivity of the gas
K_t	overall conductivity of a region occupied by tiny spheres
l	width of the gap between two surfaces confining the gas
L_l	height of the unit cell of large spheroids
L_o	distance between two contacting large spheres at the maximum ingression of small spheres, in the Ades and Peddicord model
n	molecular number density ($n = P/kT$)
N_c	number of effective contact points per unit cell
p_l	packing fraction for the large spheroids
P	gas pressure
r	average radius of a tiny spheroid
r_{tt}	radial distance from the point of contact of two large spheroids to the centre of the nearest tiny sphere
r_λ	radial distance to the point at which the separation of the large spheroids is $2\lambda/3$
R	average radius of a large spheroid
(s)	indicates two conductances in series
T	temperature in Kelvins
V_l	volume of the overall unit cell
w	average width of the gap between a large spheroid and the interspersed tiny ones

Greek symbols

δ	area of direct contact between adjacent spheroids
λ	molecular mean free path
θ	polar angle measured from the contact point
θ_{max}	maximum polar angle of integration about the contact point
θ_{tt}	polar angle to the centre of the nearest tiny sphere
$\theta_{\lambda l}$	polar angle at which the separation of the large spheroids is $2\lambda/3$

Subscripts

l	large sphere
t	tiny sphere

parameter from the single-particle beds. Unlike many other models, the ACRA model specifically includes the effect of surface roughness on the thermal conductivity. However, the roughness parameters used are also obtained by fitting to experimental data on the thermal conductivity of the bed.

The second type is the analytical model which breaks the problem into a relatively few distinct conduction paths (the contact between pebbles, the gas between them, radiation, etc.) and calculates the overall bed conductance as a series/parallel combination of the individual conductances for these paths. To make such models mathematically tractable, one normally treats the heat flow as being in straight lines within and between the spheres. The advantage to the analytical model, if successful, is that it enables one to evaluate easily the relative contributions of each pathway as a function of temperature, gas pressure, and particle size and roughness, and use this in the bed design. Either type of model obviously must involve averages over possible distributions of the particles. The appropriateness of the assumptions and approximations used in either type must be evaluated, in the end, from the agreement between the theoretical predictions and ac-

tual experimental data. If the agreement is good for beds of several different materials, gases, and particle size and roughness, then one has some confidence in using the model predictively in other situations.

Analytical models have been developed for binary beds, using the single-particle theories of Schlünder, Zehner, and Bauer (SZB) [4] or Hall and Martin [5] and including the binary nature by averaging [6,7] over cells of different pebble sizes and materials using a prescription similar to that by Okazaki et al. [8]. These models either weight each combination of particle–particle contact by the volume fraction of each particle, or treat the bed as consisting of single-material particles and average the properties of the solid components using the volume fractions. However, these weighting are more applicable to situations in which the two kinds of particles are uniformly interspersed as with particles of comparable size, rather than the present case in which the tiny particles segregate together in the gaps between the large particles. When used with particles of comparable size these models gave results which agreed with experiment within 30% [6] to 100% [7]. However, they all used experimental thermal conductivity data for single-particle beds to fit the value for the contact area between adjacent particles.

The present paper develops an analytical model for binary beds based on a recent model [9] for a bed of single-size particles (henceforth referred to as a "monobed"). The monobed model can predict the thermal conductivity of an uncompressed bed with an accuracy of about 15% over a wide range of temperature and pressure with no adjustable parameters, provided that two fairly common conditions are satisfied: (1) the conductance of a single solid particle is much greater than other conductances, and (2) the heat flow through the physical contacts between spheres is negligible compared to that from either gaseous conduction or radiation, as will generally be true of uncompressed beds except at very low gas pressures and low temperatures. (In fact, the model does provide for the contact conductance, but this requires knowledge of the contact area which is not generally known, although it may be approximated theoretically as discussed by Batchelor and O'Brien [10].)

The monobed model differs from most earlier ones in that it includes the effect of particle roughness explicitly. Moreover, an experimental procedure has been developed [9] for measuring the particle roughness which therefore eliminates the need for any adjustable parameters. Knowledge of the roughness is critical because it determines the minimum average gap between particles, which strongly affects the gaseous conductance. The monobed model gave results in good agreement, as discussed previously [9], with five different beds of alumina spheroids of different sizes measured by two different research groups, and for a bed of beryllium particles in two different gases measured by a third group. These results showed that, for many uncompressed beds, gaseous conduction between spheroids is the primary conduction mechanism except at high temperatures where radiation predominates, or when both the temperature and gas pressure are very low so conduction via direct contact predominates. This justifies the neglect of the contact conductance in such cases. The model could not be compared with other published experimental conductivity data because accurate values for the particle roughness have almost never been published.

The binary model is based on the monobed model, but also differs from the other analytical models discussed above by incorporating the binary nature of the bed by treating the small spheres between the large ones as a homogenous material as in the AP and ACRA numerical models, rather than averaging over the volume fractions of the two kinds of particles.

For beds in which the particles are compressed together, as in constrained beds of relatively soft materials with substantial thermal expansion, either the assumption of only point contacts or the use of the measured roughness height before compression may not be valid. For example, it has been shown that the bed conductivity has a strong dependence on external pressure for beds of aluminum, beryllium, and lithium zirconate particles [11], and recent papers have shown a large hysteresis in the thermal conductivity due to differential expansion between the bed and its container [1,12]. Even in such cases, our monobed and binary bed models should still be useful as the starting points in developing models which do take bed compression into account, since the contact conductance and particle roughness are both included in our models. The theory of Batchelor and O'Brien [10] may be useful for addressing the contact conductance.

2. Theory

As in the monobed model, the deviation from close-packing of the spheroids, large or small, has been incorporated by separating the packing into two fractions: one where the spheres are close-packed and one where deviations from the close-packed structure have left regions void of any of these spheres. In the binary bed, of course, the "void" regions left among the large spheroids will actually be filled with tiny spheres, as will the gaps between contacting large spheres. In these regions, the conductivity will be considered as uniform and equal to the value obtained from the monobed model for the tiny spheres taken alone.

In what follows, the subscripts 'l' and 't' will refer to the large and tiny spheroids, respectively. The overall unit cell used in the monobed model consists of a close-packed cell in parallel with a void cell. For the large

spheres, the close-packed cell has area $A_{cpl} = 2\sqrt{3}R^2$ perpendicular to the direction of heat flow and height $L_l = \sqrt{8/3}R$, and contains one particle. The void cell has area A_{vl} and the same height L_l. The volume of the overall unit cell is $V_l = A_{cpl}L_l + A_{vl}L_l$. Knowledge of the packing fraction $p_l \equiv \frac{4}{3}\pi R^3/V_l$ gives $A_{vl} = [\sqrt{2/3}\pi/\ p_l - 2\sqrt{3}]R^2$. Similar expressions apply for the tiny spheres. We let G_{cpl} and G_{vl} be the conductances of the close-packed and "void" regions of the large spheres with their interspersed tiny spheres, to give an overall bed conductance $G = G_{cpl} + G_{vl}$.

The binary model is based on the geometry sketched in Fig. 1. The large and tiny spheres have average radii R and r, respectively. The short-range surface roughness, of average height h, decreases the gaseous conductivity between contacting spheres by increasing the average gap between them. Therefore, in the close-packed regions, the model treats the spheroids as being perfect spheres, separated at their contact points by a short cylinder of cross-sectional area δ and length $2h$. θ is the polar angle measured from the vertical, and $\theta_{\lambda l}$ is the polar angle for which the separation of the two spheroids is equal to $2/3\lambda$, where the molecular mean free path is λ. (The average distance that a gas molecule travels perpendicular to a surface before colliding with another molecule is $2/3\lambda$ [13, p. 264].) From [14, p. 178] one has

$$\lambda = \left(\sqrt{2}n\pi D^2\right)^{-1}, \tag{1}$$

with $n = P/kT$ being the molecular number density, D the molecular diameter, P the gas pressure, T the temperature in kelvins, and k Boltzmann's constant. Both h and λ will be assumed to be much less than R except for λ at pressures close to zero. Reference to Fig. 1 gives, for small $\theta_{\lambda l}$, $\theta_{\lambda l} = \sqrt{\left(\frac{2}{3}\lambda - 2h_l\right)/R}$ radians and $r_\lambda = \sqrt{\left(\frac{2}{3}\lambda - 2h_l\right)R}$ for $\frac{2}{3}\lambda > 2h_l$, but $\theta_{\lambda l} = 0$ and $r_\lambda = 0$ for $\frac{2}{3}\lambda \leqslant 2h_l$.

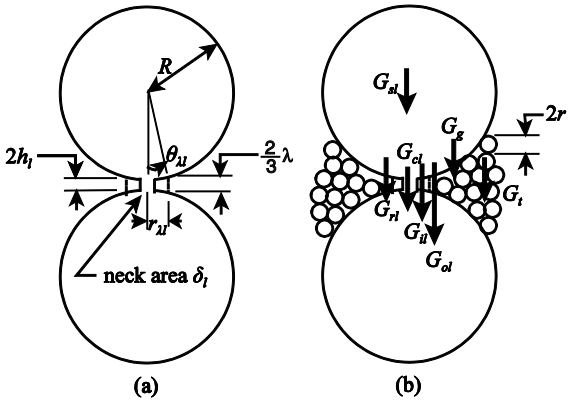

Fig. 1. Model parameters in the close-packed region: (a) geometrical parameters and (b) conductances.

As sketched in Fig. 1(b), the heat flow will be represented by a number of conductances in series and parallel. The conductances considered for either the large or tiny spheroids are the following, where the 'l' and 't' subscripts have been dropped for simplicity. G_s is through the solid spheroid, and G_r is by direct radiation between the spheroids. For gaseous conduction, two regions exist between adjacent spheroids: an "inner" region of radius r_λ about the contact point, for which the gap $2h$ between the two spheroids is less than $\frac{2}{3}\lambda$, with a conductance G_i; and an "outer" region for which the gap is greater than $\frac{2}{3}\lambda$, with conductance G_o. The gas conductivity has different values in these two regions [9]. The conductance through the physical contact of area δ is G_c. Following the monobed work, it is assumed that G_c is negligible compared to the other conductances with which it is in parallel, so it will not be considered further. It is a simple matter to include it theoretically when this assumption is not justified, but it is not simple to obtain a value for it without experiment. Additional conductances must be included in the void fraction of the tiny spheres: G_{rvt} by radiation and G_{gvt} by gaseous conduction across the void. These conductances are included in the calculation of K_t, the total effective thermal conductivity for the regions filled by tiny spheres as a function of temperature and pressure. K_t is determined exactly as for the monobed in the earlier model [9]; therefore the details of K_t will not be repeated here. The full expressions for all the conductances involved are given in the monobed paper.

The conductances between adjacent large spheres are modelled as in Fig. 2, which shows the region near the contact point of two large spheroids. The region assumed to be occupied by the tiny spheres is shown cross-hatched, with the dashed circle representing the closest position of a tiny sphere to the point of contact of the large spheres. The parameter r_{tt} is the distance from the point of contact to the centre of the nearest small sphere, and the corresponding polar angle is $\theta_{tt} = \arcsin(r_{tt}/R)$. The conductance through the region filled with tiny spheres is modelled as a conductance G_g through a gas

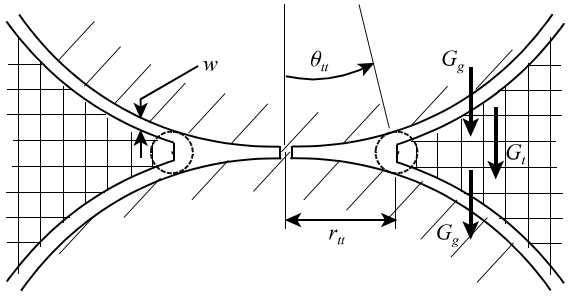

Fig. 2. Geometrical parameters and conductances near the point of contact between two large spheroids.

layer of average width w between the large spheres and the tiny spheres, in series with the conductance G_t through the tiny spheres. The conductance G_g occurs at both sides of the small sphere region. The value of w in the model is an average over the actual width that will vary from zero for actual contact between the tiny and large spheres up to an extreme maximum of R in the occasional case of an arching of the tiny spheres to leave a void between the large spheres. The presence of such a gas gap with width of order r is seen when tiny spheroids are poured between large ones placed between two glass plates, even after vibrating to help the small spheroids settle as much as possible. Moreover, the inclusion of this gas gap in the model was necessary to obtain satisfactory agreement with the experimental results. In fact, the series conductance of two gas gaps of width w is the same as the conductance of a single gap of width $2w$, so the inclusion of this gap allows for differences between the gaps at the upper and lower large spheres. Assuming that temperature differences within a given large sphere are much smaller than the temperature difference between two adjacent large spheres, which will be true if the conductance across a large sphere is much larger than the conductance between two spheres as is the case when K_s is large, G_t can be calculated as

$$G_t = \int_{\theta_{tt}}^{\theta_{max}} K_t \frac{dA}{l}, \tag{2}$$

where

$$dA/l = 2\pi R \sin\theta \cos\theta R\, d\theta/(2R - 2R\cos\theta + 2h - 2w),$$

which gives

$$\begin{aligned} G_t = K_t\pi\{&R[\cos(\theta_{max}) - \cos(\theta_{tt})] + (R + h_l) \\ &\times [\ln(R - R\cos(\theta_{max}) + h_l - w)] \\ &- [\ln(R - R\cos(\theta_{tt}) + h_l - w)]\}. \end{aligned} \tag{3}$$

Here dA is the incremental area perpendicular to the direction of heat flow and l is the distance over which the heat is flowing through dA. The upper limit of integration of $\theta_{max} = 60°$ from the contact point is chosen as being about the angle between contact points, but its exact value is not important because most of the conduction occurs near the contact point where l is shortest as discussed previously [9].

The conductance G_g across a gap of width w is calculated as in Eq. (2) but with a constant value of $l = w + j$ where j is the temperature jump distance at a surface [9]. Integrating gives

$$G_g = K_0\pi R^2(\cos^2\theta_{tt} - \cos^2\theta_{max})/(w + j), \tag{4}$$

where K_0 is the conductivity of the gas [9].

The effective number of contact points between adjacent spheres in the unit cell is $N_c = 1.5$ as discussed previously [9]. We define the symbol (s) to mean the series combination of conductances G_1 and G_2 with $G_1(s)G_2 = G_1G_2/(G_1 + G_2)$. The overall conductance G of the bed is then calculated as follows (see Figs. 1 and 2).

$$G = G_{cpl} + G_{vl}, \tag{5}$$

where

$$G_{cpl} = N_c\{G_{sl}(s)[G_{rl} + G_{cl} + G_{il} + G_{ol} + (G_g(s)G_t(s)G_g)]\}$$

and

$$G_{vl} = K_tA_{vl}/L_l,$$

where K_t is the effective conductance of a bed of tiny spheres, from Eq. (4) of Ref. [9], and

$$G_t = G_{cpt} + G_{vt}.$$

Here,

$$G_{cpt} = \{G_{rt} + N_c[G_{st}(s)(G_{it} + G_{ot} + G_{ct})]\}$$

and

$$G_{vt} = G_{rvt} + G_{gvt}.$$

The last three equations are all from Eq. (5) of Ref. [9]. The individual expressions for the different conductances are given in Ref. [9] and so will not be repeated here. The one exception is G_{rl} which is the radiative conductance only through the area between the large spheres not occupied by tiny spheres, so is over an area of only πr_{tt}^2 rather than A_{cpl} as in [9]. One first calculates K_t for the relevant temperatures and gas pressures using the monobed equations for the tiny spheres, and then inserts its values into Eq. (5) above. The Sigma Plot code for both the monobed and binary beds is provided on the web [15].

3. Comparison with experiment

There is very little published experimental data with which to compare the current model, because it depends strongly on the roughness of the spheroids which has generally not been measured accurately enough to be useful, if at all. The only binary bed data, of which we are aware, for which this has been done is that of Enoeda et al. [16] with alumina spheres of diameter 0.3 and 3 mm, for which we have previously measured the roughness [9]. The experimental data for beds of each of these spheroids under 1 atm of He and from 400 to 800 K [16] has already been fitted successfully using the monobed model [9], as reproduced in Fig. 3 (open circles and dotted lines). This figure also shows the experimental data for the binary bed as solid circles, with the solid, short-dash and long-dash lines being theoretical calculations under different assumptions (the long-dash line coincides almost exactly with the solid line). As expected, the conductivity of the binary bed is substantially

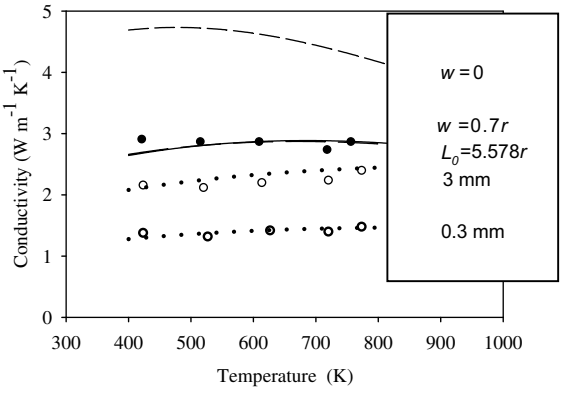

Fig. 3. Thermal conductivity for a binary bed of alumina spheroids in helium. Points: experimental data [16]; Lines: present theory. Closed points: binary bed. Solid line: theory with $w = 0.7r$; short-dash line: theory with $w = 0$; long-dash line: theory with $w = 0$ but with the Ades/Peddicord assumption for L_o. Open points and dotted lines: 3- and 0.3-mm monobeds [9,16].

above the monobed values because most of the void areas between the large spheres have been filled by the tiny spheres and alumina has a much higher thermal conductivity than does He gas. The short-dash line at the top gives the conductivity if there were no gas gap between the large spheres and the small ones ($w = 0$). It is seen that the calculated thermal conductivity is too large with this assumption. The solid line is the preferred model, for which a gas gap of width $w = 0.7r$ has been included at both surfaces of the contacting large spheroids. It is important to note that the value of $0.7r$ was a fitted parameter in the development of the model, but is not an adjustable parameter in the use of the model. This statement is supported by the good agreement that $w = 0.7r$ gives with results from measurements on a bed of $U_{0.85}Pu_{0.15}C$ pebbles as discussed below. Moreover, its value is reasonable: obviously $2r$ is an upper bound for this gap width since a larger value would allow more tiny spheres to move into the gap. In addition, the theoretical line gives good agreement with the form of the temperature dependence of the data.

The finite-element AP model [2] does not include a gas gap ($w = 0$), but does assume that the separation L_o between two contacting large spheres at the point of maximum ingression of tiny spheres in Fig. 2 is $L_o = 5.578r$, as they determined by fitting their conductivity model to experimental data. (Ades and Peddicord point out that physically the tiny spheres are often observed to be much closer to the large spheres than this.) The long-dash line in Fig. 3 (which coincides almost exactly with the solid line) uses our model but with these AP-model assumptions. This model fits the experimental data just as well as our model using $w = 0.7r$, showing that our introduction of a gas gap produces effectively the same reduction in the bed conductivity as the AP-model introduction of L_o. However, the use of a gas gap of width $0.7r$ next to the large spheroids is more consistent with observations of the actual particle packing. The good agreement between our model using the AP value of L_o, and using the value $w = 0.7r$, suggests that the fitted value of $w = 0.7r$ should apply generally to other beds, and so is not an adjustable parameter in the use of the model. Moreover, changing w by 30% to $0.5r$ changes the calculated value of the thermal conductivity by only 6%, which is less than the average 15% accuracy obtained with the monobed model [9] in any case, so the model is insensitive to the exact value of w.

Our model with $w = 0.7r$ fits the experimental alumina data essentially perfectly, compared to a 20% agreement of the finite-difference Ades and Peddicord model [8] with $U_{0.85}Pu_{0.15}C$ pebbles in helium gas. However, the Ades and Peddicord model relies on experimental measurements for the value of the bed accommodation coefficient whereas our model requires only experimental parameters of the individual particles as long as the contact conductance is negligible. The monobed model gave agreement with experiment to about 5% for the alumina spheroids considered here, as can be seen by the dotted curves in Fig. 3. This suggests that our binary bed model should work perform roughly as well as the monobed model does. Overall, the monobed model was generally good to about 15% [9], so one might expect similar results for the binary bed model.

The finite-difference ACRA model [3] does include the effect of roughness on the conductivity, modelling it by small cylinders sticking out of the particles which contact adjacent spheres. The dimensions of these cylinders were determined by fits to experimental data for beds of single-size aluminum spheres and for aluminum powder, and then these values were used in the model for a binary bed. The agreement with experiment was good, but again required fitting to experimental data to obtain some of the particle parameters. As discussed in the introduction, other analytical models have also been used to give reasonable agreement with experimental measurements on binary beds, but in all cases some experimental fitting parameters on actual beds were required unlike the present model which depends on measurements obtainable from the particles alone.

4. Conclusions

An analytical model has been developed for the thermal conduction of an uncompressed binary bed of small and large spheroids in the presence of a static gas. It agrees well with experiment in the single known case for which the particle roughness has been adequately

measured, but more testing is clearly necessary. The model uses the reasonable assumption that there is, on average, a narrow gas gap of 0.7 times the radius of a small particle at the interface between the small and large particles due to local irregularities in the packing, an assumption which is consistent with other published data on a different bed. Given this, the model has no experimental fitting parameters as long as the conductance of a solid spheroid is much greater than all other conductances in the model, and the conductance of the direct contact between spheroids is negligible which will often be true except at very low gas pressure and low temperature. It is important that future experimental studies of binary beds include careful measurements of particle roughness to allow testing of theoretical models.

Acknowledgements

The authors wish to acknowledge the financial support of the Natural Sciences and Engineering Research Council of Canada for this research and for a Summer Undergraduate Research Award for SP. Appreciation is expressed to A. De Monte and Fleming College for the loan of the profilometer, and P. Gierszewski for useful discussions.

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