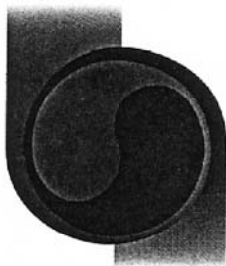


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ANISOTROPIC THERMAL CONDUCTANCE IN THIN LAYERS OF DISORDERED PACKED SPHERES

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ABSTRACT

The thermal conductance of packed spheres has been extensively investigated in the past, but the effect of packed-layer thickness on thermal conductivity and the physical mechanism of this effect have never been studied before. This work analyzes the effect of thickness on the thermal conductivities of packed-sphere layers based on statistical distributions of unit packing cells of spheres. Percolation and effective-medium approximation approaches are used to calculate the thermal conductivities in the directions perpendicular and parallel to the packed-layers. As in the case of thin solid films, the thin packed-sphere layers exhibit anisotropic thermal conductivity. The effect of packed-layer thickness on the variation of the thermal conductivity, however, has a different trend than that of thin solid films. With a reduction in thickness, the perpendicular thermal conductivity increases while the parallel conductivity decreases. In contrast to the thin solid film case, the packed-sphere thickness effect is due to the random distribution of the packed spheres, not to a limitation of the mean free path of the main heat carriers. The results indicate that the thickness effect becomes appreciable when the packed-layer thickness is less than 10 times the sphere diameter. A dramatic thickness effect is anticipated in disordered composites with very different component conductivities.

A. INTRODUCTION

There exist in the literature numerous studies on the thermal conductance of stagnant packed spheres due to the excellent thermal insulation characteristics of such materials (Kaviany, 1995; Nayak & Tien, 1978; Tien & Vafai, 1979). In applications, these materials are often in the forms of thin layers or films. The thickness effect on their thermal conductivities, however, is unknown.

The thickness effect on electrical conductivity has already been experimentally observed (Maarof & Evans, 1994; Ottavi

et al., 1978). Ottavi et al. (1978) observed an increasing slope of resistance versus height for packed mixtures of conducting and non-conducting spheres, indicating an increase of the electrical conductivity with decreasing thickness. Maarof and Evans (1994) measured the parallel resistance during the growth of Pt and Ni films. In the early stages of growth, small clusters nucleate on the substrate surface and grow into islands of the condensed phase. With continued deposition the islands grow until they come into contact, eventually creating a metallic network which then in-fills to form the continuous film. Maarof and Evans (1994) observed a decrease of the film resistance with increasing deposition thickness. The variations of the conductivity with thickness in such materials arise from the random distribution of clusters, not from the confinement of the mean free path of the heat carriers by boundaries. The conductivity of each component in the composites is assumed to be independent of the particle size.

For electrical conductivities, the attention is usually placed on the conductance parallel to the film surface. For heat transfer, variations of the conductivity in the perpendicular direction are generally more important. The minimization of electronic and mechanical devices causes the thickness effect to become important in applications such as layers/films of thermal insulation, conducting paste and micro-sensors. The thickness effect is expected in both the perpendicular and parallel directions (Neimark, 1990; Phelan & Niemann, 1997; Zhang & Stroud, 1995).

This work analyzes the thermal conductance of packed spheres in thin layers in the perpendicular and parallel directions. In section B, the thickness effect on perpendicular conductivity is investigated from the point of view of percolation and statistical configurations of unit packing cells of spheres. Section C studies the thickness effect on parallel conductivity by the effective-medium approximation of Kirkpatrick (1971). The discussion and conclusions follow in section D.

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B. THE THICKNESS EFFECT ON THE PERPENDICULAR CONDUCTIVITY

The percolation behavior of packed spheres

Electrical percolation phenomena in packed spheres have been found experimentally (Lee et al., 1986; Ottavi, 1978). Percolation represents the simplest model of disordered systems (Bunde & Havlin, 1991). Many materials support percolation, such as porous silicon (Vezin et al., 1992), silica aerogel (Richter & Tien, 1994) and packed spheres (Ottavi et al., 1978). Consider a square percolation lattice with m layers (Fig. 1). Each site is occupied randomly with probability p or is empty with probability $(1-p)$. Occupied sites may stand for very different physical properties. For instance, suppose the occupied sites are conductors and the empty sites are insulators. Electric current or heat flow can occur only between neighbor conductor sites. The conductor sites are either isolated or form small clusters of nearest neighbor sites. A cluster is a set of connected conductors bounded by insulators. At a low concentration p , the mixture is an insulator since there is no conducting path connecting opposite edges of the lattice. All clusters are finite. At large p values, along with finite ones there is an infinite cluster, that is, conducting paths between opposite edges. The mixture is a conductor. At some concentration in between, there exists a threshold concentration p_c where for the first time the current can percolate from one edge to the other. This concentration, p_c , is called the percolation threshold or the critical concentration.

The correlation length, ξ , defined by the mean distance between two sites on the same finite cluster, characterizes the linear size of the finite clusters in a percolation system (Bunde & Havlin, 1991; Neimark, 1990). It depends on the concentration,

$$\xi = a|p - p_c|^{-\nu} \quad (1)$$

where ν is a universal exponent, depending only on dimensionality, and a is the size of basic cells of the conducting phase (see Fig. 1). For 3D systems, $\nu=0.875$. Beyond the correlation length, the system is macroscopically homogeneous. Below this length scale, the properties of the system depend on the size of the system.

Percolation theory is very often used to analyze electrical conductance (Lee et al., 1986; Neimark, 1990; Sahimi, 1994). The variation of perpendicular electrical conductivity can be

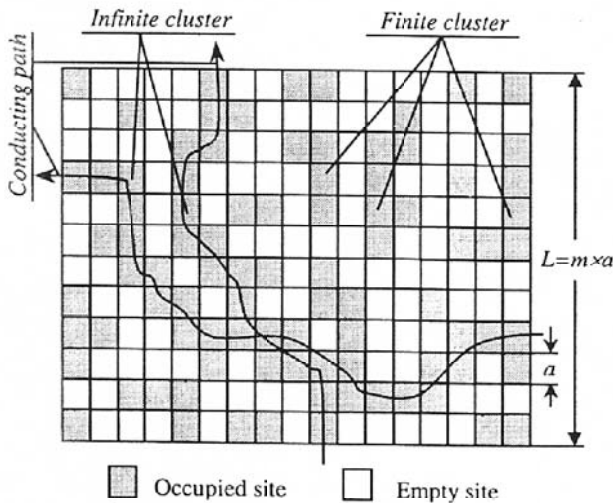


Figure 1: Illustration of a percolation system.

found from Neimark (1990),

$$\sigma_{\perp} = \sigma_0 [p_c + (p - p_c)(L/a)^{1/\nu}] (L/a)^{-\mu/\nu} \quad \text{for } \xi > L \quad (2)$$

where L is the thickness of the percolation system. The parameter σ_0 is obtainable from the bulk electrical conductivity around p_c (Bunde & Havlin, 1991; Lee et al., 1986; Neimark, 1990)

$$\sigma_{bulk} = \sigma_0 (p - p_c)^{\mu} \quad \text{for } p > p_c \text{ and } \xi < L \quad (3)$$

where μ is proposed to equal 2.0 for 3D systems (Sahimi, 1994). Percolation theory usually requires that p is in the vicinity of p_c . However, experiments have shown that the range of p where eq.(3) is applicable is quite broad (Deprez et al., 1989; Deptuk et al., 1985; Lee et al., 1986).

By analogy with the electrical conductivity, the perpendicular thermal conductivity λ_{\perp} becomes

$$\lambda_{\perp} = \lambda_0 [p_c + (p - p_c)(L/a)^{1/\nu}] (L/a)^{-\mu/\nu} \quad \text{for } \xi > L \quad (4)$$

$$\lambda_{bulk} = \lambda_0 (p - p_c)^{\mu} \quad \text{for } p > p_c \quad (5)$$

There is a lack of experimental data for thermal conductivities of packed spheres near the critical solid fraction under vacuum. The analytical results of thermal conductivities for unit packing cells of spheres, such as simple cubic (SC), body-centered cubic (BCC) and face-centered cubic (FCC) with lattice-vacancies (Nayak, 1976; Nayak & Tien, 1977) are used to determine λ_0^* in eq. (5) (see Table 1). For each packing structure, the value of the dimensionless effective thermal conductivity with the lowest solid fraction (closest to the critical solid fraction), λ^* , is listed in Table 1 and is assumed to be applicable to eq. (5). The critical concentrations p_c and solid fractions $\delta_{s,c}$ of these unit packing cells are from Scher & Zallen (1970), where $\delta_s = fp$. The concentration p is defined as the percentage of the sites occupied by spheres in each packing, and the filling factor f is defined as the maximum solid fraction occupied by solid spheres in each packing structure (corresponding to $p=1.0$). These values are listed in Table 1 together with the values of λ_0^* . The exponent μ is taken equal to 2.0, as proposed by Sahimi (1994). This value agrees with experiments on the electrical conductivity of silver-coated glass spheres and Teflon composites (Lee et al., 1986).

Fig. 2 illustrates the variation of the conductivity with thickness according to eqs.(4) and (5). The perpendicular thermal conductivity increases as the thickness decreases. The physical mechanism is simple for the increase of the conductivity. When the thickness is reduced, some finite size clusters begin to connect the opposite surfaces of the system and increase the number of conducting paths. Usually the thickness effect appears when there are fewer than four layers of packing cells. Since the conductivities in Table 1 are for unit packing

Table 1
Parameters for different packing structures

Packing	p_c^a	f^a	$\delta_{s,c}^a$	δ_s^b	λ^b	λ_0^{*c}
SC	0.310	0.524	0.162	0.458	1.21	3.79
BCC	0.240	0.680	0.163	0.392	0.748	6.60
FCC	0.195	0.740	0.144	0.555	1.81	5.87

^a Scher and Zallen, 1970

^b Specific cases with the lowest solid fractions in each packing structure from Nayak (1976), where λ^* is dimensionless thermal conductivity

^c Dimensionless factors obtained from δ_s and λ^* by eq. (5).

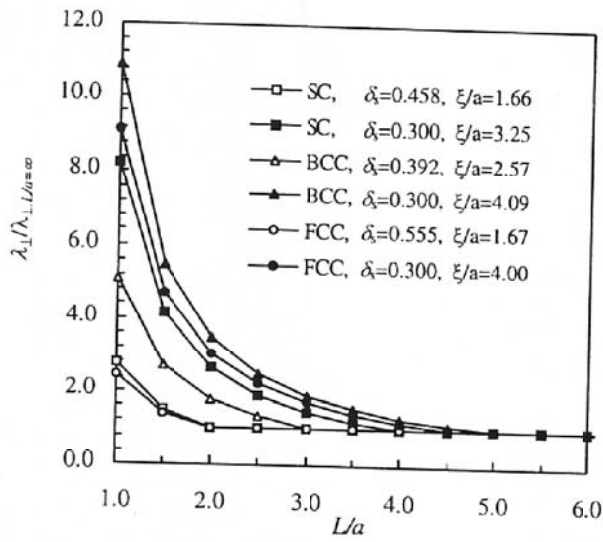


Figure 2: The variation of perpendicular conductivity with thickness based on percolation theory.

cells, parameter a should be the characteristic size of the unit packing cells, $a \approx 2d$, where d is the diameter of the spheres. For each packing structure, the thickness effect is more appreciable for those with smaller solid fractions. This can be seen from eq.(1). When the solid fraction approaches the threshold, the correlation length increases, leading to a more significant thickness effect on conductivity.

Statistical Configurations of Unit Packing Cells of Spheres

By applying the conductivities obtained for SC, FCC and BCC unit packing cells of spheres with (Nayak, 1976; Nayak & Tien, 1977) and without (Chan & Tien, 1973) lattice vacancies, Tien and Vafai (1979) showed that the thermal conductivity of packed spheres is not a unique function of the solid fraction. They accommodated the random arrangement of solid spheres by assuming that the packed bed was composed of different unit packing cells, randomly dispersed throughout the bed. This section will use this approach and set up two configurations according to the statistical rules.

Series-parallel connection. For a cubic lattice consisting of two components, component 1 has a concentration P with conductivity λ_1 and component 2 has a concentration $(1-P)$ with conductivity λ_2 ($\lambda_1 > \lambda_2$). The probability of a site's being occupied by component 1 is P . Otherwise the site is occupied by component 2 with a probability $(1-P)$. Suppose all sites in one column (in the perpendicular direction) are in series connection and all the columns are in parallel connection. Then the probability of all the sites in one column being occupied by component 1 is p^m , where m is the number of layers. The probability that component 1 occupies $(m-1)$ sites is $p^{m-1}(1-p)m$. In general, if component 1 occupies $(m-n)$ sites and component 2 occupies n sites in a single column, the probability of such an occupation in the lattice is $p^{m-n}(1-p)^n C_n^m$, where $C_n^m = m!/[n!(m-n)!]$. If the number of sites occupied by each component in each column is the same, the conductance of each column is the same regardless of the distribution of each component and equals

$$\left(\frac{m-n}{\lambda_1} + \frac{n}{\lambda_2} \right)^{-1}$$

Then the effective conductivity of the lattice is

$$\lambda_{\perp, s-p}(m) = \sum_{n=0}^m p^{m-n}(1-p)^n C_n^m \left(\frac{m-n}{\lambda_1} + \frac{n}{\lambda_2} \right)^{-1} \quad (6)$$

If $m=1$, $\lambda_{\perp, s-p}$ is the result of parallel connection of the two components. When $m=\infty$, eq.(6) represents the series connection of the two components. Eq. (6) is a lower bound of conductivity.

Parallel-series connection. In this configuration, P fraction of the total area in the first layer is occupied by component 1 and $(1-P)$ by component 2, dividing the cubic lattice into two columns (see Fig. 3a). The area in the Layer 2 which is directly above component 1 of the Layer 1 is occupied by P fraction of component 1 and $1-P$ fraction of component 2. The area in the Layer 2 which is directly above component 2 of the Layer 1 is divided in exactly the same way. Every upper layer is sub-divided into doubled P and $(1-P)$ fractions, as illustrated in Fig. 3a. For the m th layer, there are $2^{m-1}P$ fractions occupied by component 1 and $2^{m-1}(1-P)$ fractions occupied by component 2. Suppose each pair of sub-divided P and $(1-P)$ fractions in the same layer is in parallel connection and is connected in series with the fraction from which it branched. Fig. 3b shows the equivalent circuit of the connection. The effective conductivity for this connection is

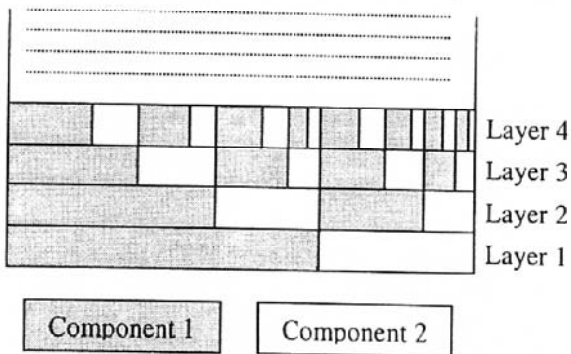


Figure 3a: Parallel-series connection.

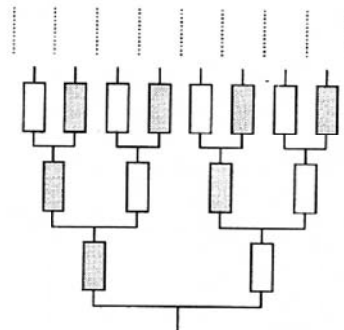


Figure 3b: Equivalent circuit of Fig. 3a.

$$\begin{aligned}\lambda_{\perp, p-s}(m) &= m\sigma_m \\ &= m \left\{ [\sigma_a^{-1} + (P\sigma_{m-1})^{-1}]^{-1} + [\sigma_b^{-1} + (1-P)^{-1}\sigma_{m-1}^{-1}]^{-1} \right\} \\ &\quad \text{for } m > 1\end{aligned}\quad (7)$$

where

$$\begin{aligned}\sigma_a &= P\lambda_1 \\ \sigma_b &= (1-P)\lambda_2 \\ \sigma_1 &= \sigma_a + \sigma_b \quad \text{for } m = 1\end{aligned}$$

Suppose the packed-sphere bed is composed of two kinds of unit packing cells. The solid fraction of the packed bed is (Tien and Vafai, 1979)

$$\delta_s = P\delta_{s,1} + (1-P)\delta_{s,2} \quad (8)$$

where $\delta_{s,1}$ and $\delta_{s,2}$ are the solid fractions of the unit packing cells of components 1 and 2, respectively. Here the packing structures with the highest solid fraction (FCC, $\delta_{s,1}=0.74$, $\lambda_1^*=2.89$) and the lowest solid fraction (BCC with lattice vacancy, $\delta_{s,2}=0.392$, $\lambda_2^*=0.748$, see Nayak, 1976) are substituted into eqs.(6) and (7) as components 1 and 2 to observe the thickness effect on conductivity. The results are shown in Fig. 4.

When there is only one layer, $\lambda_{\perp, s-p}$ and $\lambda_{\perp, p-s}$ have the same value. Components are connected in parallel. As the thickness increases, both $\lambda_{\perp, s-p}$ and $\lambda_{\perp, p-s}$ decrease and eq. (6) gives a value ($\lambda_{\perp, s-p}$) smaller than eq. (7) ($\lambda_{\perp, p-s}$) and converges more quickly. In the series connection, the component with lower conductivity dominates and eq.(6) approaches its limit in about 15 layers. The limit for eq.(6) as $m \rightarrow \infty$ is the series connection of the two components. The limit for eq.(7) can be easily proved to be the same. The parallel connection in eq.(7) retards the speed of convergence, resulting in higher conductivities. In reality, the thermal conductivity of the packed-sphere bed is in-between the values of series and parallel connections of the components. If the statistical bounds (eq. 4, Tien & Vafai, 1979) are used as the limits of eqs.(6) and (7) for $m \rightarrow \infty$, the effect of thickness occurs

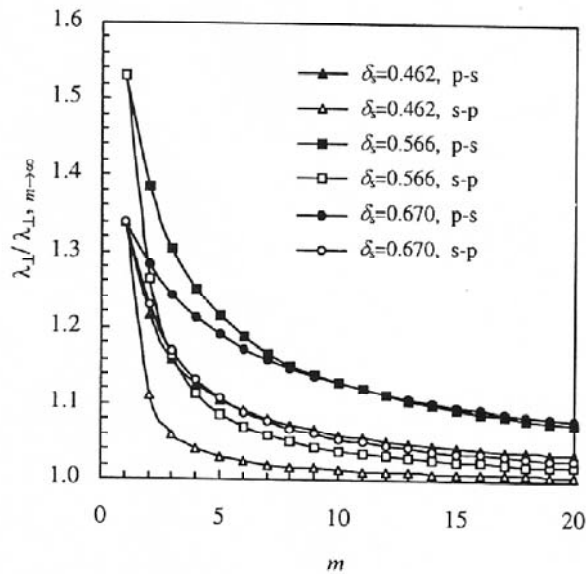


Figure 4: The variation of perpendicular conductivity with number of layers based on statistical configurations. p-s: parallel-series connection; s-p: series-parallel connection.

only when the number of layers is less than five, which is about 10 times the sphere diameter. This value agrees with the prediction of percolation theory.

The above analyses are also consistent in trend with the numerical results of Phelan & Niemann (1997). They calculated the perpendicular effective thermal conductivity of a thin, randomly oriented composite material in a 2D system and found an increase of the thermal conductivity with decreasing thickness. The effect of thickness is more profound in a 2D system than that in a 3D system. One can find such a dimensional effect in Neimark (1990). The physical reason is very simple. A 3D system has more conducting paths than a 2D system and the variation of the number of the conducting paths with thickness in 3D system is not so apparent as that in a 2D system.

A dramatic thickness effect on the overall thermal conductivities is anticipated for composite materials whose components have very different conductivities.

C. THE THICKNESS EFFECT ON THE PARALLEL CONDUCTIVITY

The conductivity in the direction parallel to the thin layers will decrease when the thickness is reduced. A reduction of thickness means that fewer layers are available to act as conduction pathways. To compensate for the lost pathways through other layers as thickness is reduced to a single layer (2D case), additional conducting sites must be added in the layer for conduction to occur. This is manifested in the increase of critical concentration from 3D to 2D systems (Sahimi, 1994). In addition, the average coordination number, or the number of contacting neighbors to a given particle, will decrease with the reduction of the thickness. The upper and lower surface layers have lower coordination numbers because they have no neighbors on one side. These surface layers affect the average coordination number in thin layers appreciably, resulting in the reduction of the average coordination number. Usually, a smaller coordination number corresponds to smaller conductivity (Nayak & Tien, 1978).

The thickness effect can be well accounted for (Zhang & Stroud, 1995) by interpolating the simple effective-medium approximation (EMA). For a binary mixture with λ_1 and λ_2 as the conductivities of components, the effective conductivity is (Kirkpatrick, 1971)

$$\lambda_{\parallel}(m) = \{ B + [B^2 + 2(z-2)\lambda_1\lambda_2]^{1/2} \} / (z-2) \quad (9)$$

where

$$B = (\frac{1}{2}zP - 1)\lambda_1 + [\frac{1}{2}z(1-P) - 1]\lambda_2$$

and z is the coordination number. For a cubic cell in a 3D lattice, $z=6$, and $z=4$ for a square in a 2D lattice. For a cubic cell which has a free facet at the surface, $z=5$. In the extreme case where there is only one layer, $z=4$. The system becomes 2D. Thus, the average coordination number is obtainable by interpolating between 2D and 3D (Zhang & Stroud, 1995)

$$z = [4 + 6(m-1)]/m \quad (10)$$

Again, the cubic unit packing cells of spheres are applied in eq.(9), taking $\lambda_1^*=2.89$ and $\lambda_2^*=0.748$. The variation of normalized conductivity with thickness is depicted in Fig. 5. For the parameters given, the thickness effect is apparent at 3-4 layers and there is a decrease of conductivity of about 8%. If λ_1

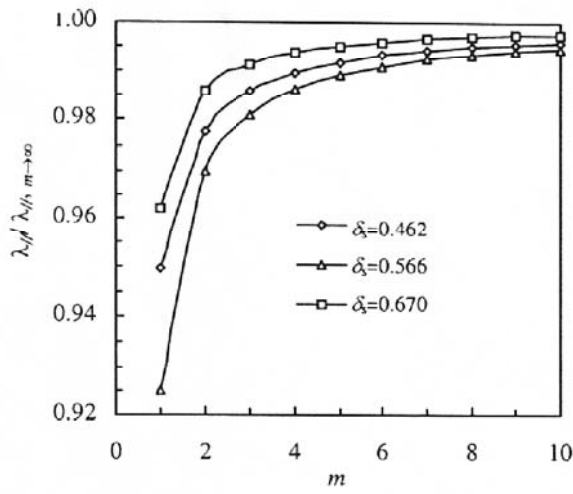


Figure 5: The variation of parallel conductivity with number of layers.

is two orders magnitude greater than λ_2 , however, a decrease up to 60% in conductivity could exist in about 10 layers.

D. DISCUSSION AND CONCLUSION

There are no experimental data for the thickness effect on the thermal conductivity of packed layers. Yet, due to the similarity between the thermal conductivity and electrical conductivity, comparisons between the present analysis and the experiments on electrical conductivities are feasible.

Ottavi et al. (1978) measured the perpendicular electrical conductivity of a mixture of conducting and non-conducting spheres at different thicknesses. An increase of the electrical conductivities with decreasing thickness beginning at up to 30 layers of packed spheres was observed. This phenomenon agrees in trend with the analysis in section B. Although Ottavi et al. (1978) believe that this increase is due to the inhomogeneous distribution of the pressure in the containers, the thickness effect due to random distribution of spheres also plays an important role. Recently, Michels et al. (1998) applied thermal spray techniques to produce resistance-heating elements that provide very high heat fluxes to solid surfaces. They electrically insulated the surface to be heated by depositing an alumina layer on it, and on this layer, they deposited a thin metallic layer which served as an electrical heating element. The thickness of the insulating layer may in principle be determined by calculating the electrical resistance required to prevent significant current from conducting to the underlying copper layer to be heated, using the resistivity of sprayed alumina at an appropriate mean temperature. Their testing, however, showed that heaters deposited onto insulators having thickness in the predicted range (about 10 μm) had inadequate electrical insulation for air plasma spray films and high velocity oxygen fuel films. The reduction in the electrical insulation may also be caused from the thickness effect due to the disordered structure in the insulating layers.

The analysis of the thickness effect on the parallel thermal conductivity in section C is supported by the experiments of Maarouf & Evans (1994). They observed a decrease of the in-plane film resistance with the increase of deposition thickness for Pt and Ni films, indicating a decrease of the electrical conductivity with the reduction of thickness.

In section B, two methods are employed in analyzing the perpendicular conductivity, predicting the same trend but with different thickness scales. The percolation approach is based on

statistical theory, numerical simulation and experiments. It has been well developed and proved to be very effective in describing disordered systems (Sahimi, 1994). The two statistical configurations predict a more profound thickness effect on thermal conductivity than percolation, especially the parallel-series connection. Both configurations have the same limit, the series connection, as the thickness approaches infinity. The series connection is the lower bound of the effective thermal conductivity for composites. The upper bound is the simple parallel connection of components, where there is no thickness effect at all. The real case lies in between the series and simple parallel connections, and the thickness effect exists because the series connection exhibits the thickness effect. The series-parallel and parallel-series configurations in section B predict this thickness effect although they are not realistic and better configurations may exist. If the thermal conductivity predicted by Tien & Vafai (1979) is taken as the limit of eqs.(6) and (7) as thickness approaches infinity, the same magnitude of the thickness effect as that from percolation theory can be observed.

Through the above analysis, it is clear that the thin packed-sphere bed is anisotropic, but the variations of conductivities with thickness are different from those of thin solid films. With the reduction of the thickness the perpendicular conductivity of thin solid films decreases due to the limitation of the mean free path of heat carriers by boundaries, but the perpendicular conductivity of packed spheres increases due to the increase in conducting paths in the perpendicular direction. On the other hand, the parallel conductivity decreases with the reduction of thickness due to the reduction in the average coordination-number and the confinement of the conducting path into one plane. For packed-sphere thin layers, the thickness effect will be appreciable when the thickness is less than 10 times the sphere diameter. A dramatic thickness effect is expected for disordered composites with very different component conductivities. In two-dimensional disordered systems, such as parallel fibers, there are fewer conducting paths and lower coordination-numbers. The thickness effect will be even more significant in these cases. The thickness effect may also exist in thin porous films. Procedures similar to those in this paper are applicable to the analysis of the thickness effect in packed fibers, composite materials and multi-component materials.

ACKNOWLEDGEMENT

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NOMENCLATURE

a	size of the basic cubic cells
d	sphere diameter
f	filling factor
L	packed-layer thickness
m	number of layers
p	probability of a site occupied by conductors
P	concentration of component one
z	coordination number
δ	solid fraction
λ	thermal conductivity
σ	electrical conductivity
ξ	correlation length

subscripts

<i>bulk</i>	bulk properties
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c threshold
s solid

superscript

* dimensionless

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