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Modeling the effective thermal conductivity of random packing of spheres through densification

C. ARGENTO† and D. BOUVARD‡

Laboratoire Sols, Solides, Structures, UJF/INPG/CNRS, BP53X, 38041 Grenoble Cédex, France

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Abstract—The thermal conductivity of densified random packing of spheres is studied. First, the resistance to heat flow of two touching deformed spheres is determined by the finite element method. A discrete model is proposed to extend the single contact results to random packings of spheres. The packings are densified by expanding the particle radius. Finally a continuous analytical expression is proposed to describe the relation between the effective thermal conductivity and the relative density of the packing.

INTRODUCTION

The determination of the conductivity of packings of spheres has long been a subject of great importance. Much of the work done in this domain relates to applications such as cryogenic insulation, boilers and heat exchangers, chemical catalysts and powder metallurgy. Industrial processes such as the hot forming of metal and ceramic powders involve particulate materials at high temperatures. Knowing the thermal properties of these materials and its evolution throughout the densification is essential to allow the correct simulation of any hot forming process.

The formal problem is to determine the effective conductivity of a random packing of spheres and its change during the densification of the packing. This conductivity, which shall be used in the thermal diffusion equation, should take into account all phenomena occurring, i.e. pure conduction through the solid phase, conduction and convection through the void phase and radiation.

The effective conductivity can be obtained experimentally (Agapiou and DeVries [1]). The required experiments are delicate and often imprecise. Numerical simulation of the phenomena can therefore be very useful, not only to understand the involved mechanisms but also to estimate the effective conductivity through densification.

Even though, in some cases, the radiation and convection occurring through the void phases can be important (Tien and Vafai [2]), the conduction through the solid phase is often the dominant contribution. We concentrate here on this mechanism. Other mechanisms are supposed to be largely dominated by solid phase conduction. This assumption implies that the effective conductivity depends on the conductivity of the solid material, the geometry of the contact and the geometry of the packing. Solutions for this problem have been obtained by several authors, in particular Batchelor and O'Brien [3] for regular packings. Jagota deduced the effective conductivity of a random packing of spheres through either a continuum calculation (Jagota and Hui [4]) or a discrete model (Jagota [5]).

In this work, a medium composed of equal sized spheres in a random packed bed is considered. This packing is put to densify and the relation between the effective thermal conductivity and the density of the medium is obtained. The 'relative density' is defined as the volumetric fraction of the particles.

We begin this work by studying the thermal resistance of two touching spheres with different contact radii. Next, a discrete model is presented to deduce the macroscopic thermal conductivity of the medium through densification, and finally, we propose a relation describing the effective thermal conductivity through the whole range of densities with a smooth transition from a typical particulate material behavior to a Maxwell upper-bound material behavior. This relation is especially useful for the simulation of forming processes like hot isostatic pressing of metal powders.

CONDUCTION THROUGH A SINGLE CONTACT

Consider the following problem: in a particulate medium, composed of monosized spheres, we want to determine the resistance to the heat transfer by solid phase conduction of two touching spheres. The one-dimensional thermal resistance of a solid volume is defined as

† Current address: E. I. Dupont de Nemours and Co. Inc., P.O. Box 0356, Wilmington, DE 19880-0356, U.S.A.

‡ Current address: GPM2/ENSPG, BP 46, 38402 St. Martin d'Heres Cedex, France.

NOMENCLATURE

A_t	cylinder cross-section area
J	heat flow
k	thermal conductivity
n_c	mean coordination number
R	particle radius
R'	particle radius after expansion
r_c	contact radius
R_T	one-dimensional thermal resistance
R_T^c	thermal resistance of a cylinder
t	final slope of relation (11)
T	temperature
z	coordinate in the direction of heat flow.

Greek symbols

ρ	packing relative density.

Subscripts

0	initial
s	solid
eff	effective.

$$R_T = \frac{T_1 - T_2}{J}, \tag{1}$$

where T_1 and T_2 are the imposed temperatures at the extremities of the volume and J is the resulting heat flow. In the case of a plain cylinder in a uniform heat flow parallel to its axis, the thermal resistance is easily deduced from Fourier's diffusion equation

$$R_T^c = \frac{\Delta z}{k_s A_t}, \tag{2}$$

where Δz is cylinder height, k_s is material conductivity and A_t is the cross-section area of the cylinder.

Let us consider now the case of two touching spheres and assume that there is a uniform flow parallel to the center-to-center axis. If the temperatures on the median plane of each sphere, defined as the plane perpendicular to the center-to-center axis containing the center of the sphere, is uniform, a one-dimensional thermal resistance element can be defined as in equation (1).

There have been numerous works devoted to the determination of the thermal resistance of two touching spheres. The analytical solution for this geometry, when the contact radius is small compared to the particle radius, can be obtained by modeling the heat flow from a finite circular area through a semi-infinite half space. The solution to this problem can be found in Carslaw and Jaeger [6]

$$\frac{R_T}{R_T^c} = \frac{\pi}{4}\left(\frac{r_c}{R}\right)^{-1}, \tag{3}$$

where r_c is the contact radius and R is the particle radius.

Other authors obtained slightly different results for slightly different boundary conditions (Chan and Tien [7], Yovanovich [8]). All these solutions are somewhat constrained to the small contact radius domain which limits their applicability to low density conditions. As far as we know, there is no model calculating the conduction through the contact between largely deformed particles. In order to avoid this limitation, we decided to calculate the thermal resistance using the finite element method.

NUMERICAL SOLUTION

The calculations have been performed for a uniform, linear row of spheres with two contacts per sphere. The geometry of the contact was obtained by viscoplastic deformation of the spheres under a normal contact force up to various strain ratios (Dellis *et al.* [9]). Because of periodicity, only two touching half-spheres have been considered. A thermal flux per unit surface is imposed on the median plane of one sphere and a constant temperature is imposed on the median plane of the other sphere. This set of boundary conditions was chosen for convenience sake, the thermal flux being immediately available from the imposed flux face. Some previous tests have shown that the solution for the thermal resistance was almost independent of the particular set of boundary conditions chosen. The steady state solution has been obtained with the thermal module of LAGAMINE finite element code from the University of Liège. Figure 1 shows three different meshes used. The evolution of the inverse of the thermal resistance—normalized with regard to the thermal resistance of a cylinder of radius R and height $2R$—as a function of the contact radius—normalized with regard to the sphere radius—is shown in Fig. 2. The thermal resistance derived from numerical calculations reasonably agrees with the analytical solution up to a contact radius of about one third of the particle radius.

The following fit can be used to represent these results:

$$\frac{R_T}{R_T^c} = 0.899\left(\frac{r_c}{R}\right)^{-1} \tag{4}$$

EXTENSION TO A RANDOM PACKING OF SPHERES

Some methods have been proposed to predict the effective thermal conductivity of a random packing of spheres. Using a mean field assumption, Jagota and Hui [4] showed that the effective thermal conductivity is closely related to the fabric tensor of granular materials (Oda *et al.* [10]). The authors used the

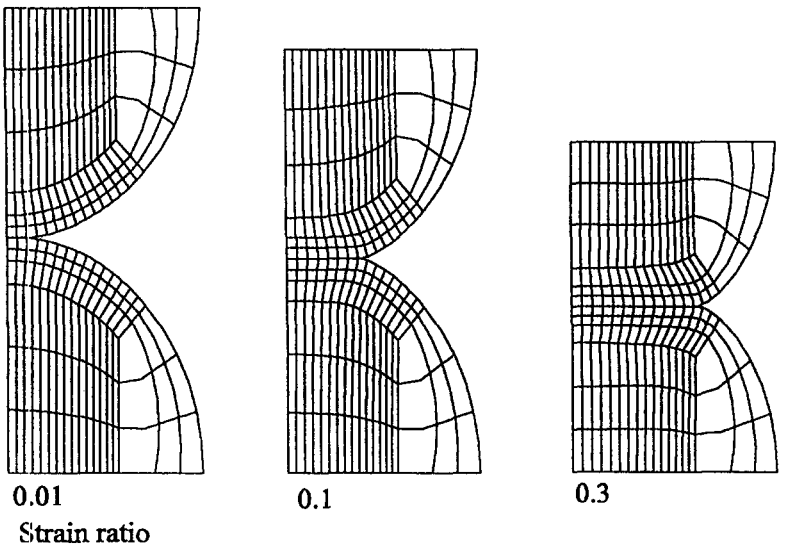

Fig. 1. Different meshes used for FEM calculations.

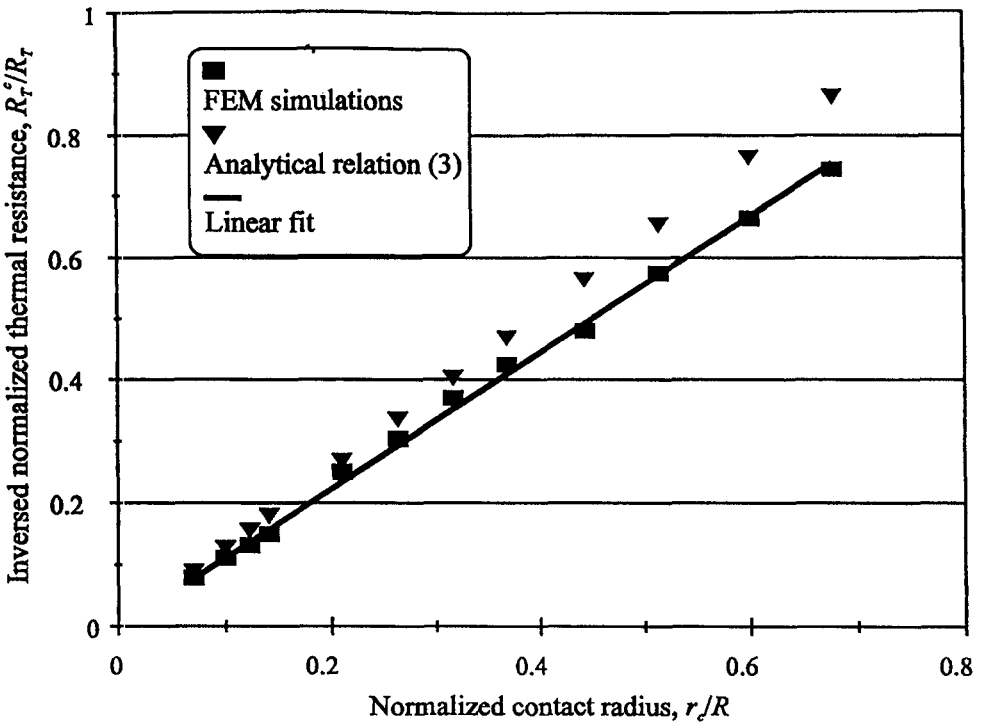

Fig. 2. Normalized thermal resistance as a function of normalized contact radius.

expression for the heat flow through a small contact derived by Carslaw and Jaeger [6]. In the isotropic case the effective conductivity reduced to

$$\frac{k_{\text{eff}}}{k_s} = \frac{1}{\pi R}\rho n_c r_c, \tag{5}$$

where n_c is the mean number of contacts per particle, r_c is the mean contact radius and ρ is the relative density of the packing. This model allows the determination of the effective conductivity at the beginning of the densification process, since it uses an expression for the heat flow valid only for small contacts.

To describe the evolution of the effective conductivity through the densification, another approach is necessary. We consider the material to be composed of initially spherical particles in point contact. This packing is placed to densify under an isotropic pressure. To this densification will correspond an increase in the particle contact area. Consider inside this packing all particles located inside a cube away from the

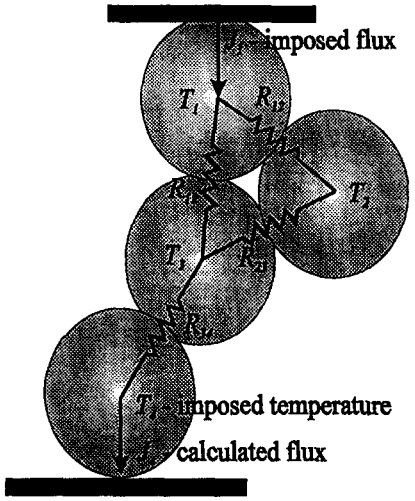

Fig. 3. Schematics of particle packing and equivalent thermal resistance network.

borders, to remove possible wall effects. A uniform heat flow is imposed on the center of all particles intersecting the upper side of the cube, whereas a uniform temperature is imposed on the center of the particles intersecting the lower side of the cube. The other faces are supposed to be adiabatic to reproduce a symmetry condition. Considering that all particles in the medium have a uniform temperature with localized disturbances around the contact, then the contact can be replaced by a one-dimensional thermal resistance element. Repeating the operation for all contacts, the particle packing is reduced to a three-dimensional network of thermal resistance elements, the nodes having the same locations as the particle centers (Fig. 3). A similar study has been done by Jagota [5] for slightly deformed particles.

For each node of the network, thermal equilibrium requires that

$$\sum_{i=1}^{n_c} J_i = 0, \tag{6}$$

where n_c is the number of contacts of the particle, i.e. the number of thermal resistance elements connected to the node. J_i, the heat flow through each element, is given by

$$J_i = \frac{\Delta T_i}{R_{Ti}}, \tag{7}$$

where ΔT_i is the temperature difference between the two touching particles and R_{Ti} is the thermal resistance of the element, given by equation (4). Applying relation (6) to all the particles in the packing yields a linear system of simultaneous equations where the temperatures of the particles are unknowns. The solution of the system gives the macroscopical temperature gradient and hence the effective conductivity.

CALCULATION OF THE EFFECTIVE CONDUCTIVITY OF RANDOM PACKINGS OF SPHERES DURING DENSIFICATION

Two numerically obtained packings of spheres were used for the calculation of the effective conductivity. The first one was obtained by Bouvard and Auvinet [11] with an algorithm proposed by Auvinet [12] that simulates the vertical deposition of the spheres inside a quasi-cubic box. This packing contains 420 rigid spheres in equilibrium under gravity. The average coordination number is six and the relative density is 0.57. The other packing used was obtained by Yen and Chaki [13] using a dynamic simulation of particle rearrangement. It contains 948 elastic spheres in equilibrium under gravity inside a cylinder. Its initial density was found to be 0.6.

To simulate an isotropic, homogeneous densification process the analogy proposed by Arzt [14] has been used. Instead of decreasing the volume of the packing, we assumed that every particle expanded from its initial radius R to a radius R' with its center remaining fixed. As the total volume of the packing is supposed to remain constant, the relative density ρ after expansion is calculated as

$$\frac{\rho}{\rho_0} = \left(\frac{R'}{R}\right)^3, \tag{8}$$

where ρ_0 is the initial density of the packing. The expansion process results in an increase of the average interparticle contact area and number (Fig. 4). The new contact radius is calculated by a simple geometry relation from the particle position and new particle radii, considering two overlapping spheres. Arzt [14] distributed the excess volume due to particle overlapping all around the free surfaces of the particle, which led to the creation of new contacts. As these contacts are tiny, they have been neglected in the present study.

Figure 5 shows the node temperature distribution inside the densest packing, with a relative density of 0.607, which corresponds to a particle expansion of 1.005. This distribution is clearly linear and the effec-

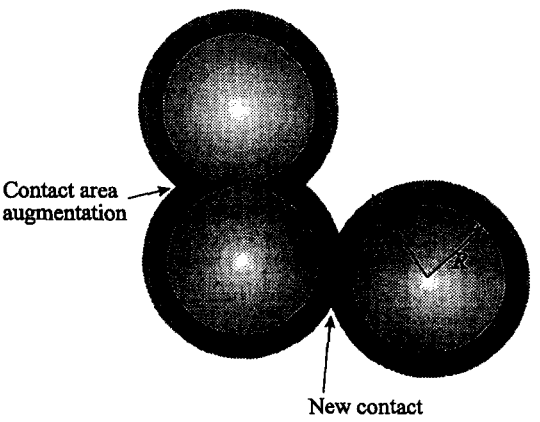

Fig. 4. Arzt [15] analogy for densification.

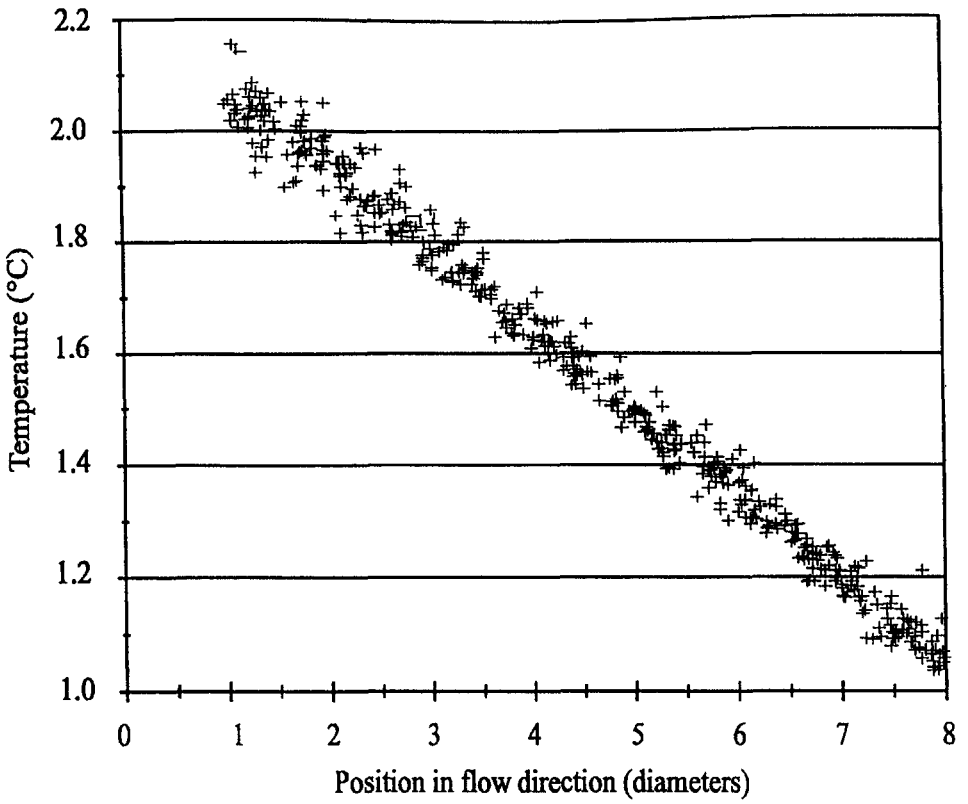

Fig. 5. Node (particle center) temperature distribution inside the packing as a function of position in the flow direction normalized with regard to particle diameter.

tive conductivity is then the slope of the plot. Calculating the effective conductivity for each packing at different particle expansions yields its relation with the relative density. Figure 6 shows the evolution of the normalized conductivity vs the density for both packings. It is clear that they follow parallel paths, shifted by the difference in the initial density. At densities approaching 1, this model is expected to fail since the notion of touching particles is no longer valid. In fact, relation (8) yields a density higher than 1.0 if applied to an expanded particle radius corresponding to a contact radius $r_c/R = 0.64$, which is still in the validity domain of relation (4). It is important to notice that, due to the construction algorithms used, both packings described are anisotropic as a result of the preferred direction of deposition. All the results shown were obtained with the heat flow in this direction. The other directions yield an effective conductivity approximately 5% lower.

EFFECTIVE CONDUCTIVITY AT HIGH DENSITY

When deformations are too important, the model proposed above is no longer valid, because of interactions between the multiple contacts the particles experience, and the notion of touching particles needing to be replaced by a model of a dense matrix with distributed pores. It is clear from Fig. 6 that the discrete model overestimates the material conductivity for high density. Considering that the pores do not touch each other, the solution to this problem is known since Maxwell [15]. A more general solution was obtained by Bauer [16] for arbitrary pore shapes and concentrations. In the case of isolated spherical pores, Maxwell obtained the relation

$$\frac{k_{\text{eff}}}{k_s} = \frac{2\rho}{3-\rho} \tag{9}$$

while Bauer derived the relation

$$\frac{k_{\text{eff}}}{k_s} = \rho^{3/2}. \tag{10}$$

It is interesting to know that those expressions are almost identical above 0.85 of relative density and converge to density 1 with the same slope 3/2. This value of the final slope shall be used in our model.

AN ANALYTICAL CONTINUOUS RELATION FOR THE EFFECTIVE CONDUCTIVITY

It is useful to put together the results obtained via the discrete element model for low and intermediate densities, and the analytical models for high densities in a single continuous relation between the effective conductivity and the relative density. This relation

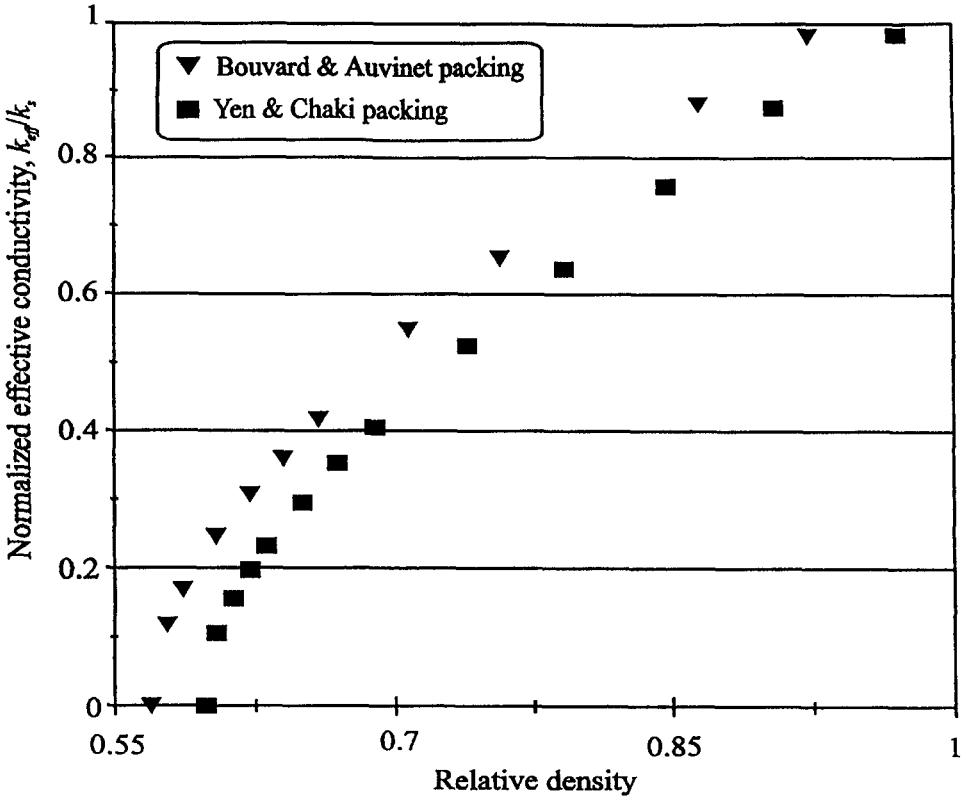

Fig. 6. Normalized effective conductivity for the two packings as a function of density.

should have an infinity slope at the initial density in order to describe the finite increase of the contact radius (and so of the effective conductivity), due to an infinitesimal increase of the particle radii (and so of the relative density). The final slope should reproduce the theoretical high density model, equation (10). Between these values, it should reproduce the results obtained from the discrete element method. In order to respect these constraints, the following relation has been empirically chosen:

$$\frac{k_{\text{eff}}}{k_s} = \left(\frac{\rho-\rho_0}{1-\rho_0}\right)^{t(1-\rho_0)}, \qquad (11)$$

where t is the final slope of the relation and is obtained from the analytical models, $t = 3/2$. This relation satisfies all the conditions imposed. In Fig. 7, relation (11) is compared to the analytical relation (5) obtained by Jagota and Hui [5] and to the discrete model. Since the initial density of the highest density packing is closer to experimental values, this packing was chosen for the comparisons. The values of n_c and r_c used in the analytical relation (5) were, respectively, the average number of contacts per particle and the average contact radius, both measured from the packing as a function of the relative density. There is a very good agreement between the three models in the low density domain and between the discrete model and relation (11) in the intermediate density domain. In Fig. 8, relation (11) is compared to the values obtained from the discrete element model for the highest density packing and to different experimental results for densified spherical powders (refs. [1, 17] for 304L stainless steel powder and Abouaf [18] for a nickel based alloy powder). The agreement of the proposed relation with both the discrete model and the experimental results is very good.

CONCLUSION

We have first obtained a relation describing the thermal resistance of two largely deformed spheres in contact from numerical calculation of the steady state solution by the finite element method. Next, we proposed a discrete model to describe the particulate media as a three-dimensional network of one-dimensional thermal resistance elements, each element replacing a two particle contact. From this model we obtained the effective thermal conductivity in the low and intermediate density domain. Using the analytical description of the high density domain derived by Maxwell, we proposed a continuous relation for the effective thermal conductivity of a densifying random packing of spheres, evolving to a dense matrix with isolated spherical pores. This relation agrees well with other existing models, which are relevant either at low density or at high density, and with experimental data. It is very useful in the finite element simulation of coupled thermo-mechanical forming process as hot

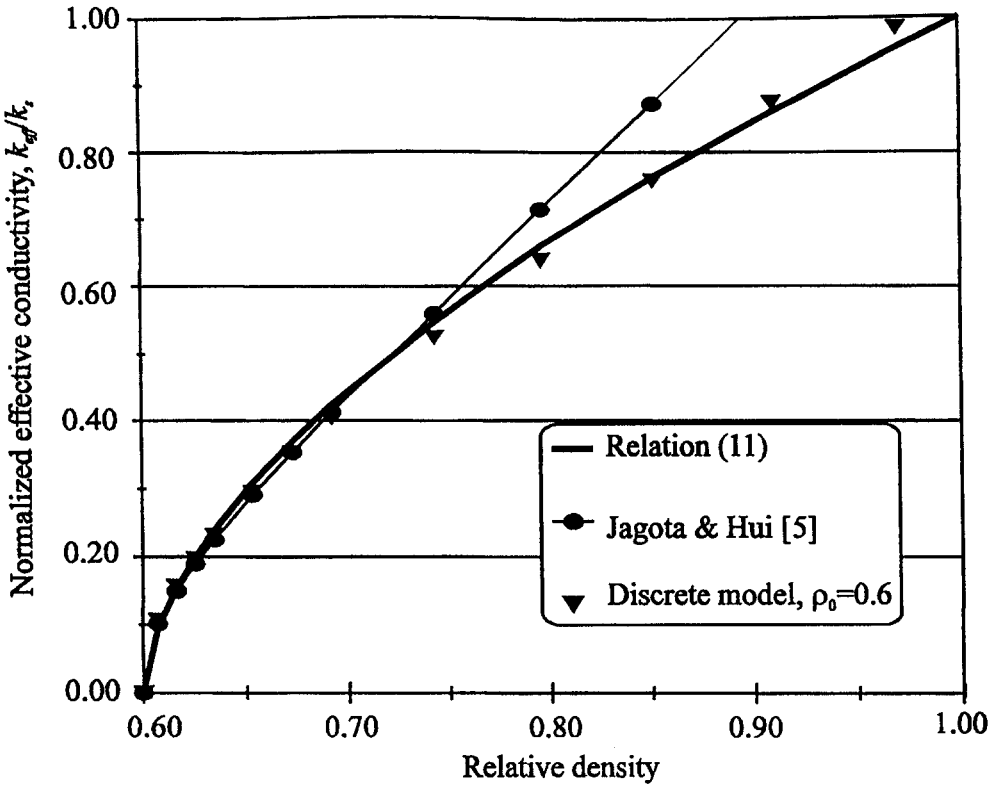

Fig. 7. Comparison between relation (11), results from the discrete model and Jagota and Hui [5] analytical model equation (10).

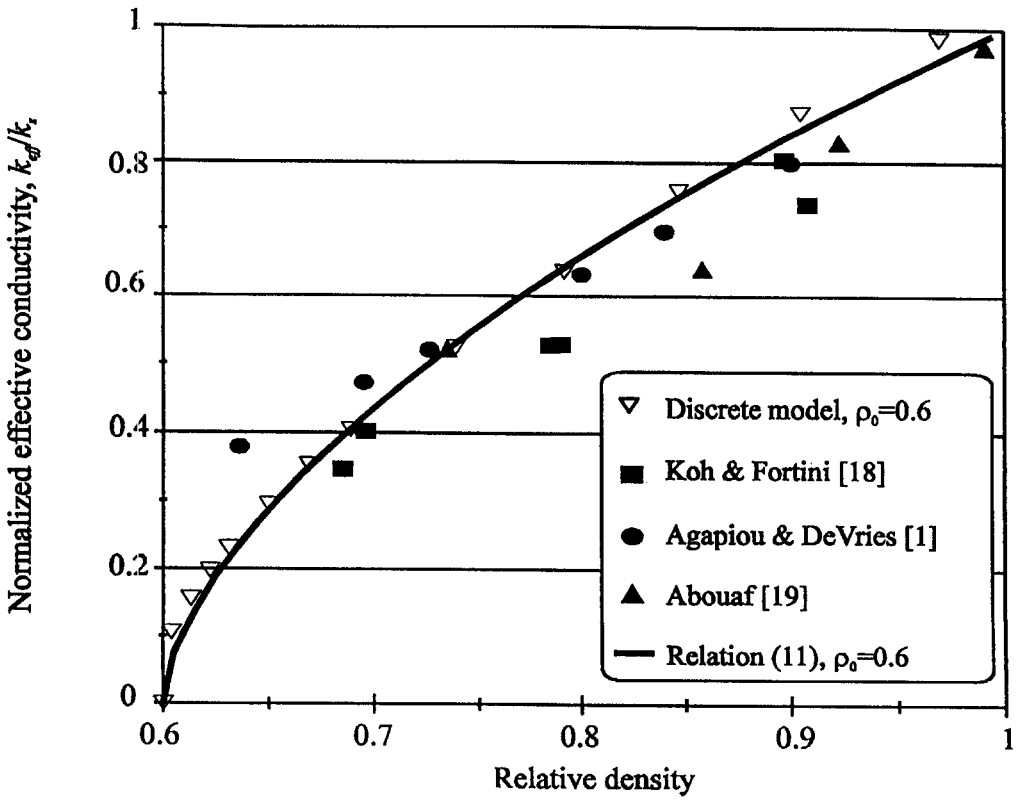

Fig. 8. Comparison between relation (11) and some experimental results.

isostatic pressing of metal powder. It has been successfully used for such simulations by Argento *et al.* [19].

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