

Heat Conduction in Granular Materials

Watson L. Vargas and J. J. McCarthy

Dept. of Chemical and Petroleum Engineering, University of Pittsburgh, Pittsburgh, PA 15261

Heat transfer in particulate systems is important to a vast array of industries, yet is poorly understood even in the simplest case—conduction through the solid phase. This is due in part to the stress and contact heterogeneities inherent to these systems. Heat conduction in a packet bed of cylinders is investigated both experimentally and computationally. A novel model is developed based on the Discrete Element Method, which not only sheds light on fundamental issues in heat conduction in particles, but also provides a valuable test bed for existing theories. By explicitly modeling individual particles within the bulk material, bed heterogeneities are directly included, and dynamic temperature distributions are obtained at the particle level. Comparison with experiments shows that this model yields a quantitatively accurate temperature field without the need for adjustable parameters or detailed microstructural information. This simple system may also provide insight into such phenomena as reactor hot spot formation and spontaneous combustion of bulk reactive materials.

Introduction

Granular materials exhibit a vast array of unusual phenomena which has sparked considerable recent interest (Bocquet et al., 1998; Hornbaker et al., 1997; Liu et al., 1995; Metcalfe et al., 1995; Patton et al., 1986; Hunt, 1997; Masoudi and Phuoc, 1999). A primary cause of much of this behavior can be traced to the unique nature of particle-particle interactions. Unlike molecular interactions—in the companion cases of fluids or “continuous” solids—particle interactions often introduce new length scales which may create difficulty for continuum modeling of these systems (Haff, 1983). In particular, particle contact inhomogeneities caused by stress chains (Liu et al., 1995; Thorton and Barnes, 1986; Jaeger et al., 1996) (see Figure 1) have been shown to have a profound effect on the pressure distribution in stacked grains (Wittmer et al., 1996), the propagation of sound in granular materials (Liu and Nagel, 1993), and agglomerate breakage (Thorton et al., 1996). Stress chains occur in all but the most perfect of crystalline particle packings and often span many particle diameters (even orders of magnitude larger distances than the particle diameter, in certain instances (Howell and Behringer, 1999)).

Traditional research in the area of granular heat transfer, using the effective medium assumption (EMA), generally provides accurate solutions of steady, average temperature profiles using detailed characterization of the microstructure (such as for composites/porous materials (Quintanilla and Torquato, 1997; Torquato, 1987); for granular materials (Oda, 1974; Zhuang et al., 1995)) or restrictive simplifying assumptions (that the material is statistically homogeneous, for example). However, even the simplest case—conduction through the solid phase—presents problems under transient conditions (Sahimi and Tsotsis, 1997; Siu and Lee, 1999; Goddard, 1992) and for the determination of proper boundary conditions u (Bauer and Schlünder, 1978; Cheng et al., 1999; Logtenberg and Dixon, 1998; Gunn et al., 1987). Moreover, it is difficult to discern how or if these methods can be made amenable to situations where the microstructure may change with time in a way which is not known *a priori*—that is, in a granular flow.

In this article, a discrete simulation method for granular heat transfer, the Thermal Particle Dynamics (TPD) method, is developed and compared to experiment. The next section describes the development of TPD and its relationship to a traditional Particle Dynamics (PD) Simulation. Results from a simple, model system are compared to experiment, and extensions and limitations of the present model are outlined.

Correspondence concerning this article should be addressed to J. J. McCarthy.

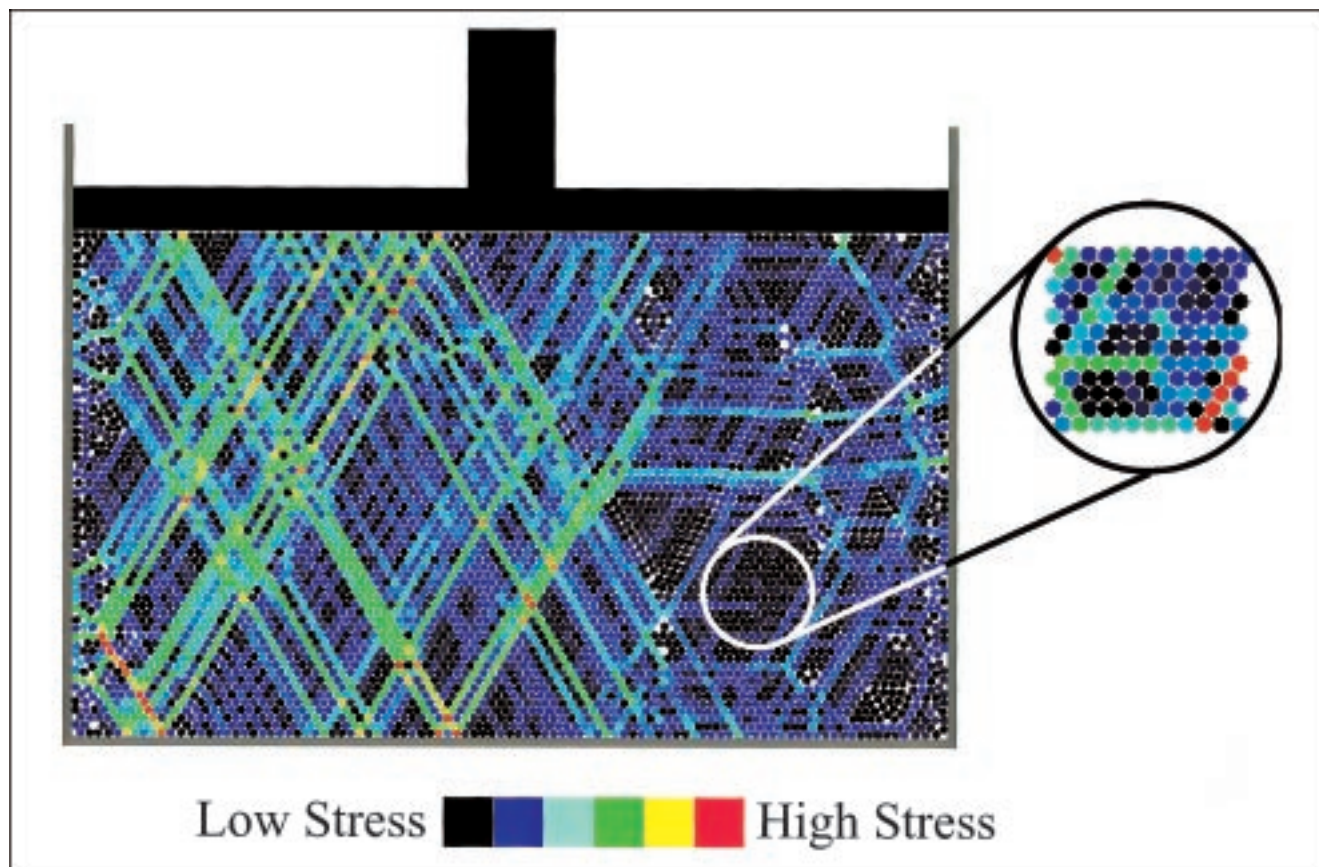


Figure 1. Stress chains in a particle bed under loading.

In all but a perfect lattice, forces follow preferred paths in a granular material—"stress chains." As the contact conductance (H) is proportional to the normal force (F_n) between the particles ($H \propto F_n^{1/3}$ for smooth, elastic particles; $H \propto F_n^{1.0}$ for rough contacts), stress inhomogeneities can be expected to play an important role in granular heat conduction. (Note that inhomogeneities are evident even in the (rescaled) blown up region.)

Discrete Modeling

Discrete modeling of granular materials has recently gained widespread acceptance as a research tool—often replacing/augmenting experimental studies of granular flows (Thorton, 2000). The predominant method of discrete simulation is still fashioned after Cundall and Strack's (1979) pioneering work, the Discrete Element Method (DEM). Relatively recently, investigators have expanded this model to include fluid drag effects (Tsuji et al., 1992; Hoomans et al., 2000) and complex force schemes such as van der Waals forces (Yen and Chaki, 1992), interstitial liquid effects (Thorton et al., 1996; McCarthy et al., 2000; Nase et al., 2001), and even solid-bridge formation (sintering) (Kuwagi et al., 2000).

In this work, an extension of PD (here taken to be synonymous with DEM), which we call *Thermal Particle Dynamics* (TPD) is introduced. This technique captures both mechanical and transport properties of granular systems at multiple length scales by including contact conductance considerations into a tradition PD Simulation. This concept is similar, in spirit, to recent work by Hunt (1997) who incorporate gas-solid heat transfer into a hard-particle PD model, and Zhuang et al. (1995) who include electrical conductance models in a quasi-static model. In what follows, we present an investigation where this new technique is used to explore the connec-

tion between processing, microstructure, and the ultimate bulk properties of a model system: a uniaxially compressed bed of particles.

Particle dynamics

Particle dynamics, a discrete method of simulation, captures the macroscopic behavior of a particulate system via calculation of the trajectories of each of the individual particles within the mass; the time evolution of these trajectories then determines the global flow of the granular material. The particle trajectories are obtained via explicit solution of Newton's equations of motion for every particle (Cundall and Strack, 1979). The forces on the particles—aside from gravity—typically are determined from contact mechanics considerations (Johnson, 1987). In their simplest form, these relations include normal, Hertzian, repulsion and some approximation of tangential friction (due to Mindlin, 1994). A thorough description of possible interaction laws can be found in McCarthy and Ottino (1998); therefore, they will not be reviewed here.

The key feature of PD is that many simultaneous two-body interactions may be used to model a many-body system (Cundall and Strack, 1979). This idea works because the

time-step is chosen to be sufficiently small such that any disturbance (in this case a displacement-induced stress on a particle) does not propagate further than that particle's immediate neighbors within one time-step. Generally, this criterion is met by choosing a time-step which is smaller than r/λ , where r is the particle radius and λ represents the relevant disturbance wave speed (for example, dilational, distortional or Rayleigh waves (Thornton and Randall, 1988)).

In much the same way that contact mechanics for a two-body interaction is well understood (Johnson, 1987)—allowing PD simulations to accurately reflect particle mechanical properties—contact conductance models are also well established (Sridhar and Yovanovich, 1994; Fletcher, 1988; Lambert and Fletcher, 1997). It is appealing, therefore, to make a direct analogy with the PD's use of contact mechanics in the context of heat transfer (see the subsection on thermal particle dynamics).

Heat transfer between particles

In describing heat transfer within particle beds, the following mechanisms have been recognized (Borkink and Westert, 1992; Froment and Bischoff, 1990): thermal conduction through the solid; thermal conduction through the contact area between two particles; thermal conduction to/from the interstitial fluid; heat transfer by fluid convection; radiant heat transfer between the surfaces of particles; and radiant heat transfer between neighboring "voids."

This work is focused on the first two mechanisms which are expected to dominate when the interstitial medium is stagnant and composed of a material whose thermal conductivity is small compared to that of the particles. According to Batchelor and O'Brien (1977), this assumption is valid as long as

$$\frac{k_s a}{k_f r} \gg 1, \quad (1)$$

where a is the contact radius, r is the particle radius of curvature, k_s is the thermal conductivity of the solid material, and k_f denotes the fluid (interstitial medium) conductivity. This condition is identically true when $k_f \rightarrow 0$ (in vacuum), and approximately true for high conductivity ratios (k_s/k_f).

This work, therefore, is concerned primarily with contact conductance. Contact conductance refers to the ability of two touching materials to transmit heat across their mutual interface. While much of the early work in this area is devoted to the study of conductivity in microelectronic devices (Antonetti and Yovanovich, 1984), there has been considerable research directed towards applications as varied as composite materials (Batchelor and O'Brien, 1977), cryogenic super-insulators (Chan and Tien, 1973), and nuclear reactors (Lambert and Fletcher, 1997). A number of review articles aptly cover recent advances (Sridhar and Yovanovich, 1994; Fletcher, 1988; Lambert and Fletcher, 1997).

The most basic problem in contact conductance is that of conduction between two smooth, elastic particles under vacuum with small, but finite, area of contact. In this problem, the "resistance" to heat transfer is assumed to be solely due to the constriction of heat flow lines as they pass from one particle to the next.

Approximate analytical solutions have been proposed independently by Yovanovich (1967), Holm (1967), and Batchelor and O'Brien (1977). All of these models predict that the conductance from one particle centerline to the other is given by

$$\frac{H}{k_s} = 2 \left[\frac{3 F_n r}{4 E^*} \right]^{1/3} = 2 a \quad (2)$$

where H is the amount of heat which may be transported per unit temperature difference per unit time, E^* is the effective Young modulus for the two particles, and F_n is the normal force acting between the particle centers.

More sophisticated models have attempted to relax the assumptions of smoothness and/or elasticity. These models generally allow for more realistic contact (elasto-plastic (Sridhar and Yovanovich, 1996), for example) with varying degrees of roughness (Lambert and Fletcher, 1997; Majumdar and Tien, 1991). Some investigators also allow for the inclusion of oxide films (Greenwood and Tripp, 1967). In general, these more realistic models predict that the conductance goes as $H \propto F_n^N$, where N is greater than 1/3 (reported as high as 1.4 in one case (Greenwood and Tripp, 1967) and experimentally measured at 2.4 in Zhuang et al. (1995)). Experimental data for validating these models is available, but is relatively sparse (Zhuang et al., 1995; Peterson and Fletcher, 1990; Marotta and Fletcher, 1998). For the remainder of this work, the simple relation found in Eq. 2 is used.

Thermal particle dynamics

Incorporating the concept of contact conductance into a PD model can be accomplished as follows. Consider two particles (i and j) which are in contact and whose temperatures far from the contact points are given as T_i and T_j , respectively. The contact conductance theories above dictate H , whereby

$$Q_{ij} = H(T_j - T_i), \quad (3)$$

denotes the amount of heat which is transported across their mutual boundary per unit time. If only pair-wise contacts exist (all contacts are decoupled), the evolution of the temperature of particle i (in some average sense) may be given as

$$\frac{dT_i}{dt} = \frac{Q_i}{\rho_i c_i V_i}, \quad (4)$$

where T_i is the temperature of particle i , Q_i is the total amount of heat transported to particle i from its neighbor (particle j), and $\rho_i c_i V_i$ is the particle's "thermal capacity."

Scale-up from this equation to multibody contacts (as is required for TPD to work) is straightforward, but requires two caveats. The first is that each i - j particle contact "sees" the same temperature for particle i such that each heat contribution may be calculated from Eq. 3 and the total heat input Q_i in Eq. 4 may be approximated as the sum of the interactions of particle i with each of its neighbors

$$Q_i = \sum_{j=1}^N Q_{ij}. \quad (5)$$

While this approximation is not strictly correct—incorporating a contact “capacitance” is possible to make this formally true (Siu and Lee, 1999)—it is conceptually simple and is found to be accurate over the time-scales examined here. In order to assure that the temperature does not vary significantly from one contact point to another (that is, that each particle “sees” the same temperature for particle i), we must assume that the resistance to heat transfer inside the particle is significantly smaller than the resistance between the particles, that is

$$Bi = \frac{H}{k_s A/r} = \frac{H}{k_s \pi r} = \frac{2}{\pi} \left(\frac{a}{r} \right) \ll 1. \quad (6)$$

It is important to note that this first of our two validity criteria does not directly depend on material properties and, in fact, the condition that the contact radius a is small relative to the particle radius r is often required by the contact mechanics models in use.

Our second requirement is that the temperature of each particle changes slowly enough that thermal disturbances do not propagate further than its immediate neighbors during one time-step (in direct analogy to the quasi-static condition of PD). Mathematically, this quasi-steady temperature criterion can be shown to be met by choosing a time-step which satisfies

$$\frac{dT_i}{(T_j - T_i)} = \frac{H dt}{\rho_i c_i V_i} \ll 1, \quad (7)$$

where dT_i is the change in the temperature of particle i during the time-step, T_j is the temperature of one of particle i 's neighbors, H is the contact conductance, and dt is the time step. Expanding this expression in terms of the contact conductance of smooth elastic spheres (see Eq. 2) yields

$$\frac{2 k_s a dt}{\rho_i c_i V_i} \ll 1, \quad (8)$$

which can be satisfied by choosing a sufficiently small time-step dt . Finally, it should be noted that the condition of small dt for TPD is often orders of magnitude less restrictive than the analogous time-step restriction in PD, such that these methods may be combined with little additional computational overhead (over a traditional PD technique).

Results

As a test of this model, consider an initially isothermal rectangular bed of particles under a uni-axial load (see Figure 1). If three side walls are insulated and the bottom wall is subject to a step change in temperature, the transient thermal response of the “slab” of particulate material can be evaluated. For a homogeneous system—or one for which a suitable effective medium assumption (EMA) may be made—this experiment represents an essentially one-dimensional heat-transfer problem (see Figure 2). In a (roughly) hexagonally packed system (like the one used in our experiments), the local void fraction is approximately constant so that effec-

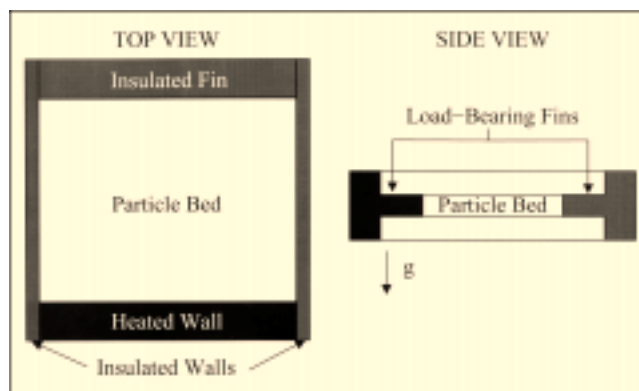


Figure 2. The quasi-2-D experimental/simulation apparatus.

A mono-layer of cylindrical particles are placed between the front and back plates (see side view). A uni-axial load is imposed by compressing the bed—fins transmit the force to the individual particles. Three walls are insulated while the bottom wall is heated.

tive properties may naively be expected to be essentially independent of space. In this case, the temperature front should propagate uniformly (across the width) through the bed.

Figure 3 shows a series of snapshots (essentially a contour plot, color-coded by temperature) of a TPD simulation, an experiment, and the corresponding one-dimensional, scalar EMA solution of this exercise. The experiments are carried out using a monolayer of cylindrical (304 stainless steel) particles in a quasi-two-dimensional, uni-axially loaded, heated bed (see Figure 2). The width of the device is roughly 200 particle diameters, and the height is 100 particle diameters. The three side walls (as well as the front and back plates) are insulated, while the other wall is heated. Temperature profiles within the bed are obtained using liquid-crystal thermography (Dabiri and Gharib, 1991). The parameters in the simulation are taken directly from the literature and consist solely of the material properties of 304 stainless steel (that is, there are no freely adjustable parameters in the model).

One can see in the case of both the TPD simulation and the experiment that the temperature front is, in fact, not uniform across the width of the container—a result which would be impossible in a one-dimensional, scalar EMA description. The presence of stress chains provides a simple explanation of this result. As the conductivity of each particle-particle junction is dependent on the imposed load, one would intuit that the conductivity along stress chains would be elevated. Therefore, even in a seemingly uniform bed, the temperature front will be jagged, with the vertical position of the front at a particular point along the container's width oscillating as stress chains converge and diverge along the bed height. However, for the particle beds examined here, a quantitative comparison of the width-averaged temperature as a function of bed height (see Figure 4) shows good agreement between both TPD simulation and the EMA model—using the effective thermal diffusivity as a fitting parameter—and our experiments. This suggests that macro-scale quantities may still be captured using an averaging technique (provided the distance between stress chains remains small compared to our averaging length).

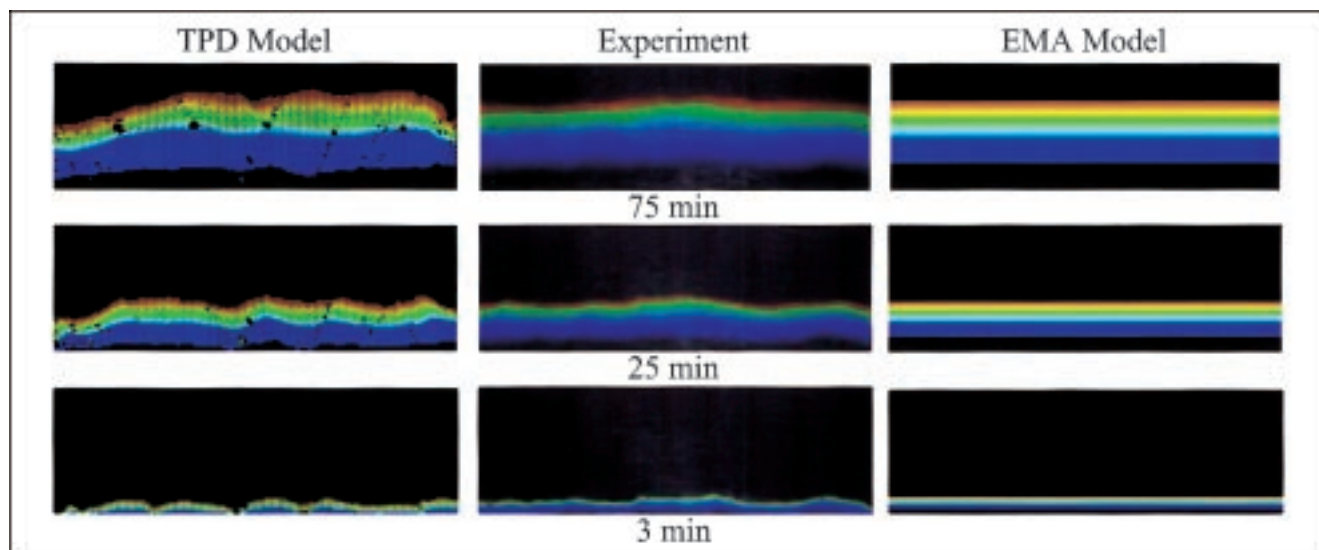


Figure 3. Thermal maps of a 2-D particle bed.

The temperature front in a transiently heated granular bed does not propagate uniformly, as may be expected using an effective medium approximation. Instead, the front oscillates as stress chains converge and diverge along the bed height. This figure shows snapshots of a TPD simulation, an experiment, and a one-dimensional EMA simulation of a heated particle bed at three different times.

Perhaps, a more striking result is seen when one examines both the stress distribution and the heat transfer together. Consider the granular bed shown in Figure 1. By simple measures of the local structure of the bed (void fraction, for example), the bed is approximately uniform; however, stress chains distribute the confining load nonuniformly. Superim-

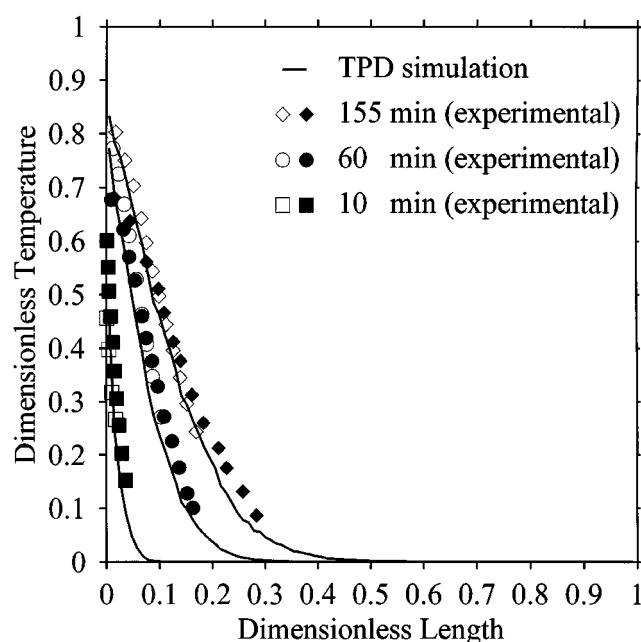


Figure 4. Quantitative comparison of TPD and experiment.

Shown is the width-averaged temperature of the particle bed from Figure 3 for both the TPD simulation (lines) and the experiment (symbols). Note that the agreement is quite good without requiring any adjustable parameters.

posing the stress-field onto a temperature contour obtained from TPD (see Figure 5a) shows that the propagation of heat is also nonuniform. The temperature front on the lefthand side has advanced considerably relative to that of the righthand side. Following our simple arguments above, this is certainly due, in part, to the high density of stress chains on the left. Additionally, however, the essentially *horizontal* stress chains on the right seem to act as barriers to heat transfer! These chains effectively channel heat away from the region until the lateral temperature gradient is so small as to prohibit further re-direction. As further evidence of this, Figure 5b shows a vector field of the heat flow through the bed. It is clear that, along the stress chains, heat conductivity is large and heat flow is rapid (large, red arrows), and that heat flow beyond the horizontal stress chains on the right of Figure 5b is severely hampered. Moreover, one can see that there are regions within the bed, perhaps isolated from their neighbors by similarly *insulating* stress chains, that receive little or no heat flow (open circles).

In order to examine the effective thermal conductivity as a function of external load several simulations and experiments are performed (see Figure 6). The conductivity in both cases is obtained by fitting the width-averaged data to a scalar EMA solution. Plotting the resultant values on a log-log scale, we see that the effective conductivity varies with load to the power ≈ 0.37 . This value is surprisingly close to the $1/3$ value that would be expected from the Hertzian deformation of *one* particle. This suggests that, as the total load is increased, the distribution of contact forces throughout the bed does not vary—in agreement with the findings of other authors (Liu et al., 1995; Howell and Behringer, 1999; Thorton, 1997).

Conclusions

The work presented here represents the TPD simulation in its simplest form—elastic, perfectly contacting particles un-

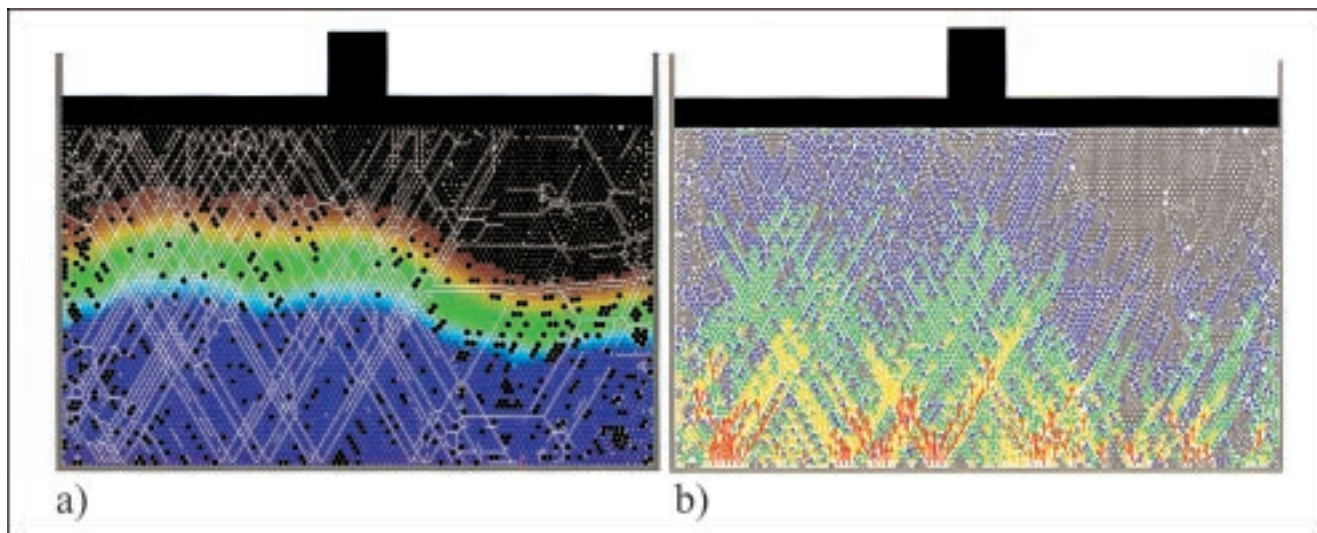


Figure 5. The effect of stress chains on heat transfer.

(a) Here, the temperature field (color-coded) is superposed over a plot of the stress field within the material. The white lines denote particle contacts which experience larger-than-average stress. Note that the temperature front on the lefthand side has propagated further than the righthand side. (b) Examining the heat flow in the particle bed provides hints to the origin of these nonuniformities. Heat flow along stress chains is significantly enhanced (large, red arrows). Additionally, however, the *horizontal* stress chains on the right seem to act as barriers to heat transfer. Similar effects may explain the isolated regions of very low heat transfer (open circles), which are effectively insulated from the rest of the bed.

der vacuum; yet, it is capable of capturing details of particle-level temperature profiles which have not been previously reported, without requiring adjustable parameters. It is found that by matching the microstructure of an experimental system only qualitatively, quantitatively accurate estimates of ef-

fective properties are possible. In this sense, this technique is quite useful as a test-bed of new and existing theories of granular conductivity. In fact, even these simple results suggest that an important consideration has been missing from previous granular conduction studies—the stress distribution in the particle bed.

We find that stress and contact heterogeneities—due primarily to the existence of localized “chains” or particles which support the majority of an imposed load (stress chains)—may cause dramatic changes in the way that heat is transported by conduction. While these stress chains serve to augment heat flow along their axis, they effectively hamper perpendicular heat flow. This tends to create regions within a particle bed which are thermally isolated from their surroundings, possibly supporting previous claims that particle packing has a strong influence on the size and location of reactor hot spots (Balakotaiah et al., 1999). Moreover, nonuniformities in both temperature and heat flow (see Figure 1), similar to those found during combustion synthesis of powdered ceramics (Varma et al., 1998) or for electrical conduction during varistor failure (Vojta et al., 1996), occur over length scales which would be difficult, if not impossible, to capture in even the most rigorous of effective medium approximations (Zhuang et al., 1995). These nonuniformities could certainly have a profound effect on a variety of materials processing operations and require more investigation.

This work has considered only two of the different heat-transfer mechanisms within particle beds—thermal conduction through the solid and through the contact area between particles in contact. One advantage of the TPD technique, however, is that it is extensible in a variety of ways: incorporation of numerous contact mechanics/conductance models to account for particle roughness, plastic deformation, and so on; inclusion of interstitial fluid effects for either stagnant

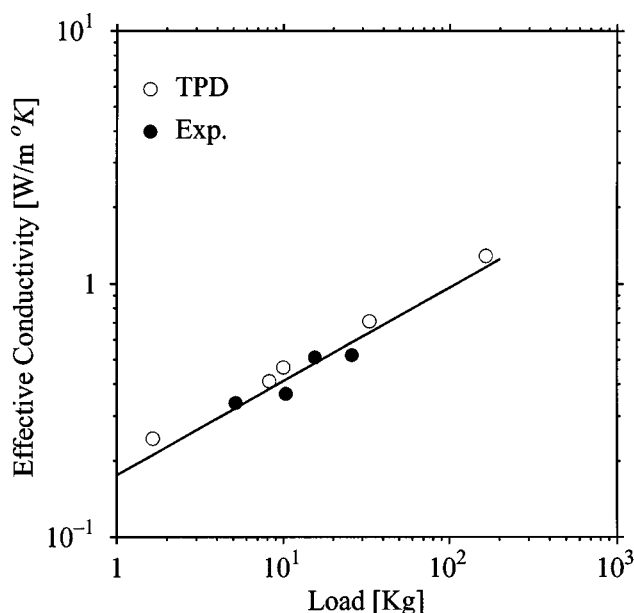


Figure 6. Effective conductivity as a function of load.

It is interesting to note that the slope for the solid line is 0.37, in close agreement with the theoretical value of $1/3$ which would be obtained for the Hertzian deformation of a single particle. A possible explanation of this result is that the distribution of contact forces is only a weak function of the overall contact force.

fluids (gases using the model proposed by Slavin et al. (2000) or liquids using Okaski's (1985) model) or flowing fluids (which would require a model of the fluid motion); and deformation/motion of the solid phase—a problem currently outside the reach of effective medium approaches.

Acknowledgments

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