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Effective thermal conductivity of submicron powders: A numerical study

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Abstract: Effective thermal conductivity is an important property of granular materials in engineering applications and industrial processes, including the blending and mixing of powders, sintering of ceramics and refractory metals, and electrochemical interactions in fuel cells and Li-ion batteries. The thermo-mechanical properties of granular materials with macroscopic particle sizes (above 1 mm) have been investigated experimentally and theoretically, but knowledge remains limited for materials consisting of micro/nano-sized grains. In this work we study the effective thermal conductivity of micro/nano powders under varying conditions of mechanical stress and gas pressure via the discrete thermal resistance method. In this proposed method, a unit cell of contact structure is regarded as one thermal resistor. Thermal transport between two contacting particles and through the gas phase (including conduction in the gas phase and heat transfer of solid-gas interfaces) are the main mechanisms. Due to the small size of particles, the gas phase is limited to a small volume and a simplified gas heat transfer model is applied considering the Knudsen number. During loading, changes in the gas volume and the contact area between particles are simulated by the finite element method. The thermal resistance of one contact unit is calculated through the combination of the heat transfer mechanisms. A simplified relationship between effective thermal conductivity and loading pressure can be obtained by integrating the contact units of the compacted powders.

Introduction

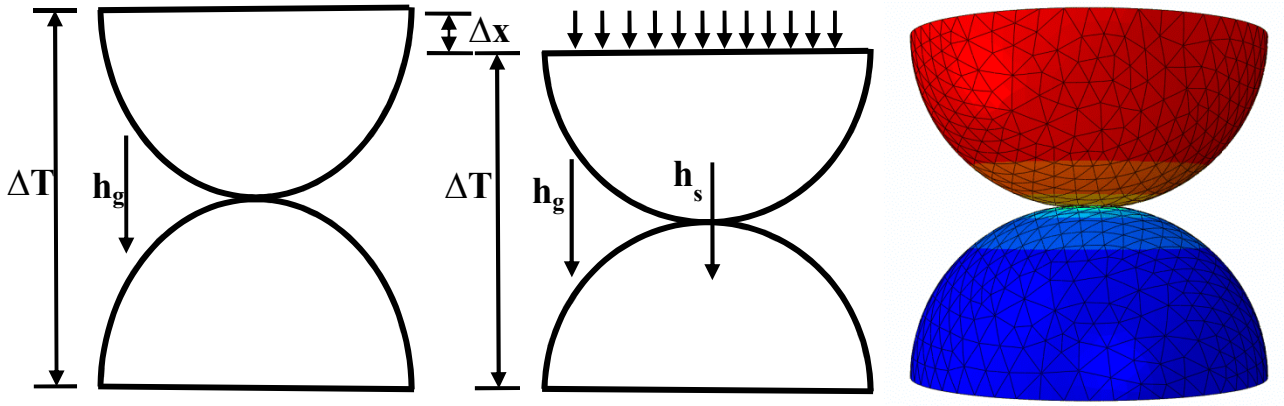
In energy systems, granular media are commonly used to store, convert, capture and produce energy, including lithium-ion batteries, solid oxide fuel cells, and thermal storage systems. Heat transport in these energy systems is a key issue necessitating extensive research to 1) establish a fundamental understanding of the thermo-mechanical properties and 2) provide useful knowledge for process optimisation [1-3]. Due to heterogeneous material properties, complex packing structures and various inter-particle interactions, different theoretical methods have been developed to study the heat transport mechanisms and the influencing external factors [4, 5].

Most research in this field has focused on the effective thermal conductivity (ETC) of powder beds, employing three types of models to predict this parameter. Type I models are based on the materials' constitution and porosity and can be used to calculated the ETC by considering powder beds as dispersion media [4]. Type-II models regard powder beds as thermal circuits in order to obtain the effective thermal resistance through an analogy with electrical circuits [4, 6-9]. Type-III models discretise powder beds into many unit cells and integrate the cells' properties to derive the ETC [5, 10, 11]. The former two types of models can provide relatively simple equations to predict the ETC while the type-III models usually involve considerable numerical calculation. However, it is more convenient to study the effects of external factors on the ETC and to approximate the real situation in practical application by applying type-III models. To realize a useful type-III model, a proper description of the unit cell combing different heat transfer mechanism and external factors is required. An analytical solution for a contact unit involving two spheres was proposed by Batchelor and O'Brien in order to describe the relationship between ETC and contact area [10]. Baharami *et al*. employ the thermal contact resistance model to include the effects of rough contacts in the

spherical powder beds [7]. The particle size of the powder beds considered in these studies has typically been above 1 mm, thus the heat transfer through the interparticle gap is regarded as the same as through a continuum gas phase. Furthermore, Gusarov *et al.* considered the effect of the Knudsen number on the ETC of powder beds and the results shows that for millimetre- and micrometre-sized particles the increase of gas pressure increases the ETC with saturation occurring above a certain gas pressure [5]. However, these studies have not addressed the powder beds containing submicron or nano particles which are widely used in sintering process, thermal insulation, and solid oxide fuel cells. In this study, the finite element method (FEM) is utilised to calculate heat transport processes in the submicron units with different particle sizes. By altering the gas phase pressure and mechanical deformation, the relationship between the ETC and loading condition can be obtained.

Simulation Model

A two-hemisphere contact unit cell is used to evaluate the effective thermal conductivity of individual element as shown in Figure 1. The hemispheres, with constant material properties, form a contact along the axial direction concentrically. The gap between the hemispheres is filled with air, thus completing a cylindrical unit. In this simulation unit, a constant temperature difference between the top surface and bottom surface is applied, and uniform temperatures of the upper and lower hemispheres are established at the initial state as T_h and T_l, respectively. An axial compression is applied from the top surface and the bottom surface is fixed. In this study, the commercial finite element package ABAQUS is utilised to perform the FEM calculation. The temperature difference ΔT is maintained to be 1K and $T_h - T_l = \Delta T$ is created at initial state. The properties of the air between the gap of the two hemispheres are assumed to be independent of the temperature distribution of the air phase because ΔT is relatively small. Thus, in the numerical model, a distance-dependent the gap thermal conductance (*GAP CONDUCTANCE in ABAQUS) is used to approximate the thermal conductance via the gas gap.



(a) (b) (c)

Figure 1: Unit cell model: (a) schematic drawing of two spherical particles nearly in contact; (b) Two particles in contact; (c) The FEM model with temperature profile with an arbitrary unit. Here, ΔT, Δx, h_g and h_s denote the temperature difference, particle deformation, heat flux through the gas phase and through the solid contact, respectively.

Gas heat transfer at submicron-scale depends on the the ratio of the mean free path, l, of gas molecules to the gap distance, d as given by the Knudsen number, Kn. For the powder beds comprised of submicron particles, the gaps between particles become comparable to the mean free path of the gas molecules, leading to a transition of the heat transfer modes of gas phase. In other words, the equation $h_g = k_g/d$ to calculate the heat conductance of gas is invalid in this range, where k_g is the thermal conductivity of gas and d is the gap distance. An approximation based on

the Knudsen number is used to predict the gas phase heat conductance. The mean free path l is calculated based on the dynamic viscosity μ

$$l = \frac{\mu}{P}\sqrt{\frac{\pi RT}{2M}} \tag{1}$$

and the dynamic viscosity μ is calculated by

$$\mu = \mu_0 \frac{T_0+120}{T+120}\left(\frac{T}{T_0}\right)^{\frac{3}{2}} \tag{2}$$

where μ_0 is dynamic viscosity at T_0=296.15K. At a certain temperature T, the mean free path depends on gas pressure P and gas constitution, where M is the molar mass of the gas. When the Knudsen number Kn is very large $(Kn \gg 100)$, the heat conductance is approaching the free molecular limit [5]

$$h_0 = \frac{1}{4}\frac{\gamma+1}{\gamma-1} P \left(\frac{2R}{\pi MT}\right)^{\frac{1}{2}} \tag{3}$$

Here R is the gas constant and γ is the adiabatic exponent. For $0.01 < Kn < 100$, the heat conductance at different Knudsen number is approximated by

$$h = \frac{h_0}{2}\left(\frac{1}{1+1/(\chi * Kn)} + \frac{1}{\left(1+\sqrt{1/\chi * Kn}\right)^2}\right), \tag{4}$$

$$\chi = \frac{2\sqrt{\pi}}{3}\frac{9\gamma-5}{\gamma+1}. \tag{5}$$

It can be concluded that the gas heat conductance at $Kn \gg 100$ depends on not only the temperature but also the gas pressure while the heat conductance is not affected by the gas pressure for small values of Knudsen number.

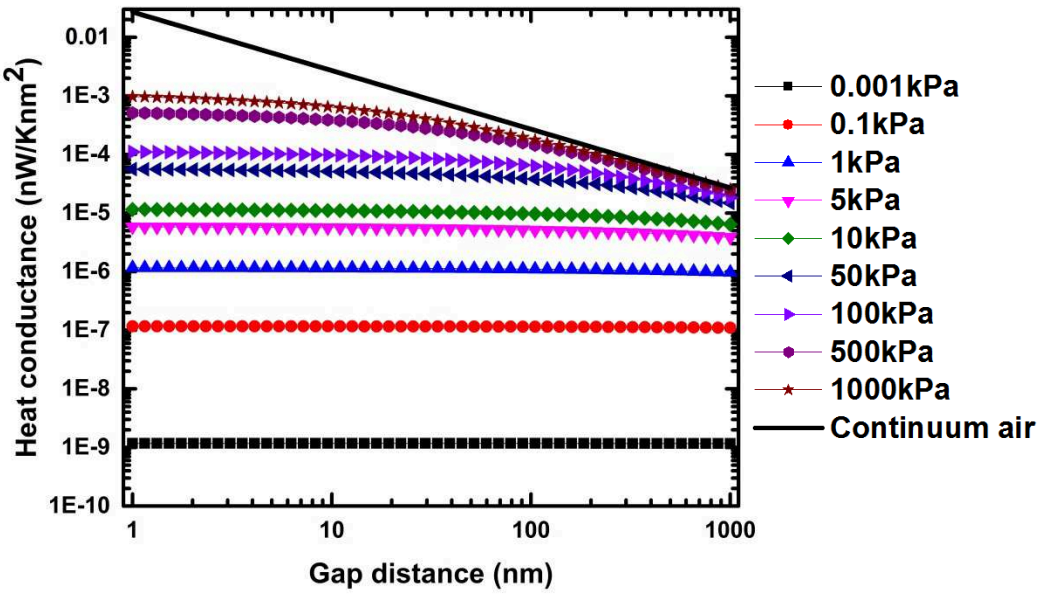


Figure 2: Air heat conductance as a function of gap distance at different gas pressure, at $T = 298K$. The solid line indicate the conductance calculated based on the continuum equation, $h_g = k_g/d$.

Further to the heat transfer of gas phase, the heat transfer through the contact region of two hemispheres in one unit cell needs to be defined. Thermal contact resistance is commonly used to describe the heat transfer across the solid contact between two relatively large hemispheres. In such situation, the contact region consists of a finite number of nanoscale contact spots. However, this contact model is not applicable for situation where the contact region itself falls into nanoscale. Due to the consistent material properties of the two contact hemispheres, the nano contact region is considered to have the same thermal properties as the bulk material. Thus the heat conductance of the contact region is defined as the heat conductance of 1nm thick material in this simulation.

In ABAQUS, the gap conductance is varied with Knudsen number Kn to simulate the gas heat transport at different gas pressure. Deformation of the unit cell can be achieved by applying displacement as the boundary condition. Table 1 summarizes the materials and model parameters used in the ABAQUS simulations. The hemispheres are assumed to be SiO_2 powders and the gas phase is air. The negative sign of axial deformation ratio means that the two hemispheres are separated and a finite gap is left between two hemispheres.

Table 1: Materials and model parameters used in ABAQUS simulation

Powder material: SiO_2 Thermal conductivity k_s=1.3 nW/nm·K, specific heat c=7*10^{11} nJ/kg·K, density ρ=2.65*10^{-24} kg/nm^3, Young's modulus=7*10^{-8} N/nm^2, Poisson's ratio=0.17.						
Gas: Air Molar mass M=0.028966 kg/mol, dynamic viscosity μ_0=1.827*10^{-5} Pa·s,						
Axial deformation ratio	-1%	0%	1%	2%	5%	
Particle diameter (nm)	100	200	500	1000		
Gas pressure (Pa)	1	100	10000	100000	1000000	1000000

Result and discussion

In these models, the reaction heat fluxes are calculated according to the temperature boundary conditions. At the equilibrium state, the effective thermal conductivity of individual unit cells can be calculated based on the Fourier's law

$$\phi = k_e \frac{\Delta T}{x} \tag{6}$$

where ϕ is the reaction heat flux, k_e denotes the effective thermal conductivity of unit cell, x is the axial -length of the unit cell and ΔT is the temperature difference.

Figure 3 presents the change of ETC of the unit cells versus the change of the gas pressure for different particle sizes. When the two hemispheres are not in contact (-1% deformation), the heat transfer inside the unit cell takes place entirely through the gas phase. It is clear that increasing the gas pressure continuously increases the ETC of the unit cells which is consistent with the trend in the heat conductance of air versus pressure in Figure 2. For conditions where the heat transfer occur solely through the gas phase, larger particle sizes result in a larger ETC of the unit cells. Note that the gap size is relative to the size of the unit cell, and in Figure 3(a) a displacement of -1% is considered. When the two hemispheres approach to each other to form a point contact, the effect of gas pressure diminishes at low gas pressure region. The gas pressure begins to affect ETC of unit cells at value above 10 kPa and this effect becomes more significant as the gas pressure increases. However, for conditions of particle contact the relationship between the particles size and ETC is opposite to the situation of separated hemispheres shown in Figure 3(a). Compared with the ETC of non-contact group, the ETC of this point contact group is much larger due to the presence of solid-solid contact conductance. The contact point provides a thermal shortcut for the heat transport because the heat conductance of the contact region is several orders (10^3~10^9) larger than the gas heat conductance.

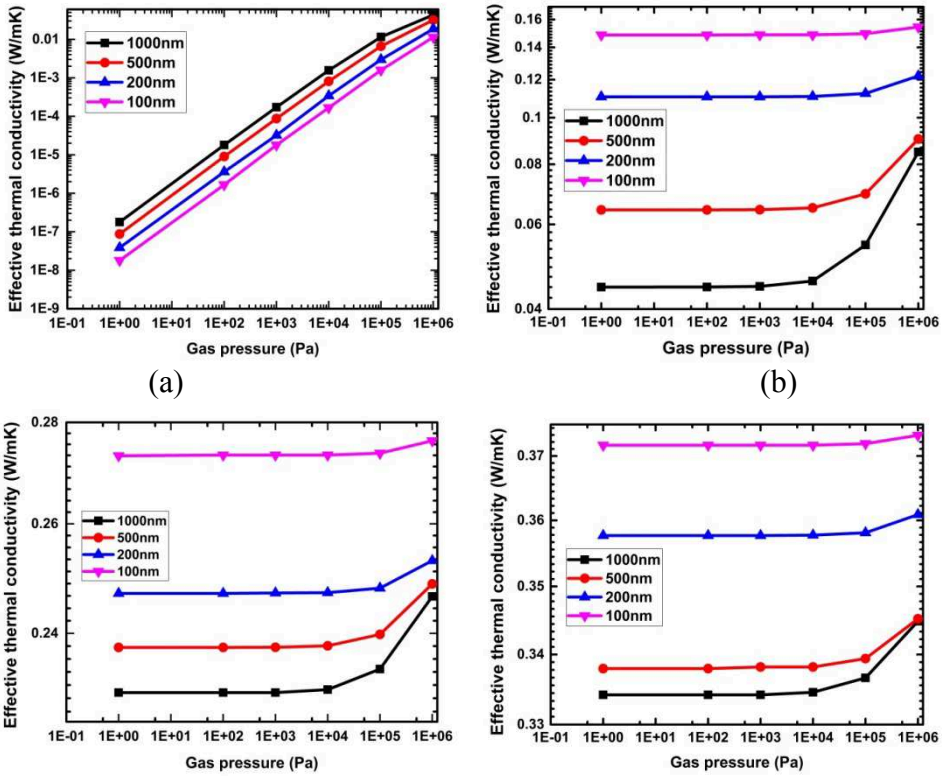


Figure 3: The effective thermal conductivity as a function of air pressure in different sized powders, at T=298K: (a) non contact, -1% deformation, (b) point contact, 0% deformation, (c) 2% deformation, and (d) 5% deformation

When the deformation increases as shown in Figure 3 (b-d), with an expanding contact region, the ETC of the unit cells rises and follows a similar pattern as a function of the gas pressure. The magnitude of the increase in the ETC due to the change in pressure in the three contact groups (0%, 2%, 5%) actually corresponds to the increment of ETC in the non-contact group (-1%). Only when the gas heat transfer is comparable to the heat conduction through the contact region, the gas pressure starts to significantly alter the ETC. This phenomenon indicates these two heat transport mechanisms, i.e., heat conduction by solid-solid contact and heat transfer through gas phase, are parallel, which is a general consideration in many other methods used to predict the ETC of powder beds. Figure 4 shows the effect of deformation on the ETC of unit cells. As the deformation increases, the ETC of unit cells of different converges. The reason is that the increasing contact area dominates the heat transport in the unit cell, and the gas gap becomes less significant.

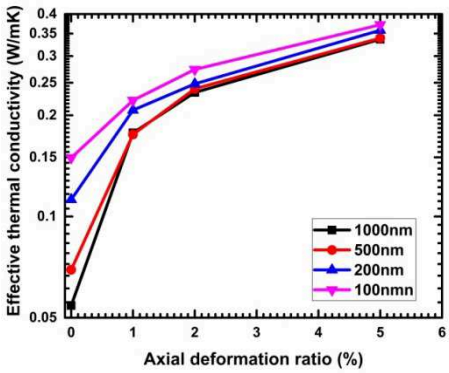


Figure 4: The effective thermal conductivity as a function of axial deformation in different sized powders, at P=100 kPa and T=298 K

Conclusion

The effective thermal conductivity of compacted submicron powders was studied for various loading conditions, including gas pressure, temperature and deformation. A microscopic unit cell was established to provide the basis for evaluating overall conductivity of the packed beds and was analysed using finite element method. The Knudsen number has been incorporated in the gas conduction model to include the influence of gap distance, gas pressure and temperature. The numerical result shows a clear dependency of the effective thermal conductivity on parameters of particle size, gas pressure and deformation. Increasing gas pressure tends to elevate the overall conductivity at high pressure. When interparticles contacts are form, smaller sizes lead to larger overall conductivity of unit cells. With increasing compaction, the overall conductivity increases and the solid-solid contact conduction dominates the heat transfer. This study provides a theoretical basis to consider the heat transfer in compacted beds with submicron-sized particles under a wide range of loading conditions.

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