

Radiation conductivity for a random void-solid medium with diffusely reflecting surfaces

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Abstract—A variational principle is formulated for radiant heat transfer through the void spaces and conduction in the solid, of an arbitrary void-solid system with diffusely reflecting surfaces. Variational upper bounds on the effective conductivity of a void-solid suspension are expressed in terms of certain averages characterizing the random geometry. A radiation conductivity upper bound, calculated for a model porous medium generated by randomly placed, overlapping spheres, is compared with well-known results from kinetic theory and radiant heat transport. The variational principle provides a means to obtain useful estimates and upper bounds on the thermal radiation conductivity, that include multiple and anisotropic diffusive scattering in a bed of large particles.

INTRODUCTION

RADIATION heat transport through the open spaces of a void-solid system is generally important either at high temperatures [1] or at low pressures [2]. Beek [3] and Argo and Smith [4] have included radiant heat transport in an analysis of the radial heat transport within a packed bed heat exchanger or chemical reactor. Whitaker [5] using volume averaging has obtained an equation for the heat flux through a diffusely reflecting, opaque solid-void system, which when linearized in the temperature gradient and evaluated for an isotropic, thick bed, consists of a direct sum of solid conduction and void radiation terms. Beek [3] has pointed out that there is at present no experimental basis for including the surface emissivity or assigning the length variable in the expression for the thermal radiation conductivity. Vortmeyer [6] has reviewed the various structural models [7-11] for radiation transport in packed solid, and in this context stated that the long range effects of scattering from a void region to nearby void regions have not been included in any theory of radiation transport. A rigorous scattering theory will permit both a derivation of the bed surface emissivity dependence, and proper transport path length variable for the void radiation conductivity.

In this paper, equations for the radiation heat transport through the open spaces of a void-solid suspension of arbitrary geometry with simultaneous thermal conduction in the solid is formulated. The solid is opaque with gray emitting and diffusive reflecting surfaces. Any particle dimensions are presumed to be much larger than the wavelength of the

thermal radiation. A complete solution of the radiation heat transfer problem for complex bed geometries, e.g. fluidized or packed beds, is hampered in practice both by the difficulty of the equations, and often also by an incomplete knowledge of the void-solid structure. A variational upper bound on the effective bed conductivity and the void radiation conductivity is derived for an arbitrary void-solid geometry. The variational conductivity expression is applied to randomly dispersed solids and written in terms of appropriate statistical functions of the structure.

As it does for the simpler flat plate problems, the radiosity formulation of diffusive scattering developed in the paper avoids the sum over successive multiple surface scatterings, but now for arbitrary solid surfaces. If instead of the radiosity, the equations were cast only in terms of the temperature, the radiant thermal conductivity expression becomes an infinite sum over terms $j = 0, 1, 2, \dots$. Each term contains the surface emissivity product $\varepsilon^2(1-\varepsilon)^j$; a product of view factors, one view factor every straight line segment of the path the photons travel, diffusely scattered at j successive surface collisions between emission and absorption; and a surface temperature dependence. Then in addition, this combination is integrated over all possible paths. This type of expanded form of the radiant conductivity was obtained by Whitaker [5] as a formal result, though no calculations were performed for explicit model structures. A variational principle can be written that parallels Whitaker's result, however, because of the difficult necessary integrations over the complex paths and the j summation, it is a formidable task to evaluate. The radiosity approach avoids these difficulties.

An illustration is presented using straightforward trial functions, a linear temperature profile across the

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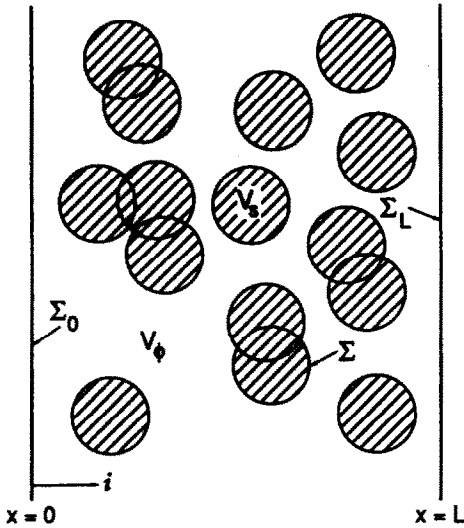


FIG. 1. Randomly overlapping solid spheres.

phase function constant, the linear anisotropic scattering and the variational radiant conductivity expressions are the same. The selected value of the phase function constant implies a very strong anisotropic backscatter. The radiant conductivity expression is a rigorous upper bound for any emissivity, that reduces to the exact physical forms in both the extremes $\epsilon = 0$ and 1 , and these features suggest that it might be useful as an estimate of the radiant conductivity for a dilute sphere bed. For the identical problem of Knudsen void gas conduction dilute sphere bed kinetic theory results [16, 17] again establish that the variational conductivity is an exact solution in the limits of thermal accommodation coefficients of zero and unity. The variational principle provides a means to obtain useful estimates and upper bounds on the thermal radiation conductivity, that rigorously include both multiple and anisotropic diffusive scattering in a bed of large particles.

FUNDAMENTAL EQUATIONS

Suppose just outside the ends of a large porous slab (Fig. 1) of total volume V , solid plane surfaces Σ_0 and Σ_L are positioned at $x = 0$ and L , respectively, and that i is a unit vector pointing across the slab in the positive x -direction. The total volume V is divided into two subregions, a region of solid V_s and a void region V_ϕ . The interface Σ between these two regions makes up the void-solid interface. We assume Σ_0 , Σ , and Σ_L are opaque gray surfaces, the radiation of which is emitted and reflected diffusely according to Lambert's cosine law [12]. The emitted flux depends on the absolute temperature T of the surface, the surface emissivity ϵ and the Stefan-Boltzmann constant σ , in the combination $\epsilon\sigma T^4$. Kirchhoff's law states that the same surface element will absorb only a fraction ϵ of the incident radiation, reflecting the

fraction $(1 - \epsilon)$. If H represents the radiant flux incident on a unit surface, then for a diffusely reflecting surface the radiosity B , the radiation diffusely leaving a unit surface, is given by

$$B = \epsilon\sigma T^4 + (1 - \epsilon)H \quad (\mathbf{r} \text{ on } \Sigma). \quad (1)$$

The fraction $K(\mathbf{r}', \mathbf{r}) d^2\mathbf{r}'$ of radiation diffusely distributed, from a unit surface element located at \mathbf{r}' , that travels a straight line free path, and arrives at a second surface element $d^2\mathbf{r}$ located at \mathbf{r} , can be used to formulate the radiant exchange between surfaces. Since we are assuming diffuse scattering at the surfaces, K is given by the cosine law

$$K(\mathbf{r}', \mathbf{r}) = K(\mathbf{r}, \mathbf{r}') \quad (2a)$$

$$= -[\boldsymbol{\eta}(\mathbf{r}) \cdot \boldsymbol{\rho}][\boldsymbol{\eta}(\mathbf{r}') \cdot \boldsymbol{\rho}]/(\pi\rho^4) \quad (2b)$$

(if \mathbf{r}' can see \mathbf{r})

$$= 0 \text{ (otherwise)}. \quad (2c)$$

where $\boldsymbol{\eta}(\mathbf{r})$ and $\boldsymbol{\eta}(\mathbf{r}')$ are unit normals respectively at the points \mathbf{r} and \mathbf{r}' on the surfaces Σ_0 , Σ and Σ_L , pointing into the void, and $\boldsymbol{\rho} = (\mathbf{r}' - \mathbf{r})$. Of the diffuse radiation $B(\mathbf{r}') d^2\mathbf{r}'$ leaving $d^2\mathbf{r}'$ of Σ_0 , Σ and Σ_L , only the fraction $B(\mathbf{r}') d^2\mathbf{r}' K(\mathbf{r}', \mathbf{r})$ will arrive within a unit area at \mathbf{r} on Σ , then the total incident radiant flux at \mathbf{r} on Σ is

$$\int_{\Sigma_0 + \Sigma + \Sigma_L} K(\mathbf{r}', \mathbf{r}) B(\mathbf{r}') d^2\mathbf{r}' = H(\mathbf{r}) \quad (\mathbf{r} \text{ on } \Sigma). \quad (3)$$

When the total incident flux H from equation (1) is substituted into equation (3), and the radiosity B is subtracted from both sides, an integral equation in terms of the radiosity and temperature is obtained

$$\int_{\Sigma_0 + \Sigma + \Sigma_L} K(\mathbf{r}', \mathbf{r}) [B(\mathbf{r}') - B(\mathbf{r})] d^2\mathbf{r}' = \frac{\epsilon}{1 - \epsilon} [B(\mathbf{r}) - \sigma T^4(\mathbf{r})] \quad (\mathbf{r} \text{ on } \Sigma). \quad (4)$$

Note from its definition as a probability, and the symmetry property (2a), that the function $K(\mathbf{r}', \mathbf{r}) d^2\mathbf{r}'$ will sum to unity over the surfaces Σ_0 , Σ and Σ_L .

The steady state energy balance at a point \mathbf{r} within the solid V_s , and Fourier's law with solid conductivity λ , give

$$\nabla \cdot (\lambda \nabla T) = 0 \quad (\mathbf{r} \text{ in } V_s). \quad (5)$$

The thermal boundary condition equating the net radiative flux from the void-solid surface Σ at \mathbf{r} to the normal flux from the solid

$$-\lambda \boldsymbol{\eta} \cdot \nabla T = B - H \quad (\mathbf{r} \text{ on } \Sigma) \quad (6)$$

or from equation (1) in terms of the radiosity and temperature

$$\lambda \boldsymbol{\eta} \cdot \nabla T = \frac{\epsilon}{1 - \epsilon} [B(\mathbf{r}) - \sigma T^4(\mathbf{r})] \quad (\mathbf{r} \text{ on } \Sigma). \quad (7)$$

Equations (4), (5) and (7), together with the steady

surface conditions that the radiosity is assigned to be uniform at

$$B(\mathbf{r}) = B_0 \quad (\mathbf{r} \text{ on } \Sigma_0) \quad (8a)$$

and

$$B(\mathbf{r}) = B_L \quad (\mathbf{r} \text{ on } \Sigma_L) \quad (8b)$$

are in principle sufficient to determine B and T . In practice the complex geometry of the surface Σ and solid volume V_s prevent an outright solution.

To calculate the effective conductivity and the thermal radiation conductivity the fundamental equations must be linearized in the temperature. Steady, average surface temperatures T_0 and T_L , respectively, of the edge surfaces Σ_0 and Σ_L , and an average slab temperature $\bar{T} = (T_0 + T_L)/2$ are defined. The linearization presumes the temperature variation across the slab is small compared with the average slab temperature \bar{T} , i.e. $\Delta T/\bar{T} [= (T - \bar{T})/\bar{T}]$ is small. We retain in equations (4), (5) and (7) only terms zeroth and first order in the temperature variation about \bar{T} . As a consequence in both equations (4) and (7), σT^4 can be replaced by

$$\sigma T^4 = A + CT \quad (9a)$$

where

$$A = -3\sigma\bar{T}^4 \quad (9b)$$

and

$$C = 4\sigma\bar{T}^3. \quad (9c)$$

Also as a result of the linearization any temperature dependence of λ or ε can be neglected and the constants in equations (4), (5) and (7), evaluated at the average slab temperature. Equations (4), (7) and (9a)–(9c) are linear in temperature, and are the starting point of the variational analysis. Since the effective and thermal radiation conductivities do not depend explicitly on the temperature drop across the slab, no loss in the generality of the conductivity equations occurs when the problem is linearized. However, a direct application of the linearized equations and variational principle to obtain local temperature and radiation fluxes should be restricted to moderate temperature differences across the slab [18].

In the context of low pressure cryogenic insulation, Barron [2] has pointed out the equivalence of Knudsen gas heat conduction and diffusely reflecting radiation for energy transport between parallel plates with uniform temperatures. It is interesting to note for the linearized forms (4), (5), (7) and (9), this equivalence extends to arbitrary solid geometries with complex surface temperature profiles, and thermal gradients in the solid. In low pressure, cryogenic insulation, the variable $\psi = n\bar{v}/4$, where n is the particle number density and \bar{v} the thermal speed, is a constant [19] in the absence of effusive flow. The equations for Knudsen gas conduction are obtained from equations (4), (5), (7) and (9), when for $\Sigma_0 + \Sigma + \Sigma_L$ we let $B = E_r$ represent the energy leaving the surface diffusely

(now not of photons, but of Knudsen gas molecules), replace the emissivity ε by the thermal energy accommodation coefficient α introduced by Knudsen [20], set $A = 0$ and write $2C = k\psi(\gamma + 1)/(\gamma - 1)$ in terms of the Boltzmann constant k and heat capacity ratio γ . A complete mathematical equivalence exists through to the final results. Linearization is not an issue in the Knudsen void gas thermal conductivity problem.

VARIATIONAL FORMULATION

In this section we will derive a variational upper bound on the total heat flux, the net rate heat passes through both the void and solid per unit total cross section of a slab with arbitrary pore geometry. The net heat flux J across the plane at $x = 0$ is just the difference of the flux in minus the flux out

$$J = (B_0 V)^{-1} L \int_{\Sigma_0} d^2\mathbf{r} \int_{\Sigma + \Sigma_L} d^2\mathbf{r}' K(\mathbf{r}, \mathbf{r}') B_0 \times [B_0 - B(\mathbf{r}')] \quad (10)$$

where V is the total volume of the slab and B_0 and B_L are the radiosity values on Σ_0 and Σ_L , respectively. The flux into the slab at $x = L$ is derived by a similar procedure. This flux can be obtained from equation (10) by interchanging the 0 and L subscripts; however, this gives the negative of J . Combining these two equations we find

$$-J \cdot \beta = V^{-1} \int_{\Sigma_0 + \Sigma_L} d^2\mathbf{r} \int_{\Sigma_0 + \Sigma + \Sigma_L} d^2\mathbf{r}' K(\mathbf{r}, \mathbf{r}') B(\mathbf{r}) \times [B(\mathbf{r}) - B(\mathbf{r}')] \quad (11)$$

where

$$\beta = (B_L - B_0)L^{-1}\mathbf{i}. \quad (12)$$

The upper bound on the heat flux is based on the variational functional

$$\begin{aligned} V\Gamma\{B^*, T^*\} = & \frac{1}{2} \int_{\Sigma_0 + \Sigma + \Sigma_L} d^2\mathbf{r} \int_{\Sigma_0 + \Sigma + \Sigma_L} d^2\mathbf{r}' K(\mathbf{r}, \mathbf{r}') \\ & \times [B^*(\mathbf{r}') - B^*(\mathbf{r})]^2 + \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2\mathbf{r} \\ & \times [B^*(\mathbf{r}) - A - CT^*]^2 + C \int_{V_s} d^3\mathbf{r} \lambda [\nabla T^*]^2 \quad (13) \end{aligned}$$

where the trial temperature T^* must be continuous and at least piecewise continuously differentiable in V_s , and the trial radiosity B^* must satisfy

$$B^*(\mathbf{r}) = \begin{cases} B_0 & (\mathbf{r} \text{ on } \Sigma_0) \\ B_L & (\mathbf{r} \text{ on } \Sigma_L). \end{cases} \quad (14)$$

The volume element $d^3\mathbf{r}$ is summed over the solid volume V_s . To show that the term $\delta\Gamma$ from equation (13), first order in the respective variations δB , δT of the trial functions about B , T , vanishes we write

$$\begin{aligned} \frac{1}{2} V \delta \Gamma = & \frac{1}{2} \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r} \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r}' K(\mathbf{r}, \mathbf{r}') \\ & \times [B(\mathbf{r}') - B(\mathbf{r})][\delta B(\mathbf{r}') - \delta B(\mathbf{r})] \\ & + \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2 \mathbf{r} [B(\mathbf{r}) - A - CT(\mathbf{r})][\delta B(\mathbf{r}) - C \delta T(\mathbf{r})] \\ & + C \int_{V_s} d^3 \mathbf{r} \lambda \nabla T \cdot \nabla (\delta T). \end{aligned} \quad (15)$$

Upon interchange of the integration variables \mathbf{r}, \mathbf{r}' and application of the symmetry condition (2a) of K in the $\delta B(\mathbf{r}')$ term of the first integral of equation (15), and integration by parts followed by the divergence theorem in the third integral, the first-order expression $\delta \Gamma$ becomes

$$\begin{aligned} \frac{1}{2} V \delta \Gamma = & \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r} \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r}' \delta B(\mathbf{r}) K(\mathbf{r}, \mathbf{r}') \\ & \times [B(\mathbf{r}) - B(\mathbf{r}')] + \int_{\Sigma} d^2 \mathbf{r} \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r}' \delta B(\mathbf{r}) \\ & \times K(\mathbf{r}, \mathbf{r}') [B(\mathbf{r}) - B(\mathbf{r}')] + \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2 \mathbf{r} \delta B(\mathbf{r}) \\ & \times [B(\mathbf{r}) - A - CT(\mathbf{r})] - \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2 \mathbf{r} C \delta T(\mathbf{r}) \\ & \times [B(\mathbf{r}) - A - CT(\mathbf{r})] + C \int_{\Sigma} d^2 \mathbf{r} \delta T(\mathbf{r}) \eta \cdot \lambda \nabla T(\mathbf{r}) \\ & - C \int_{V_s} d^3 \mathbf{r} \delta T(\mathbf{r}) \nabla \cdot [\lambda \nabla T(\mathbf{r})]. \end{aligned} \quad (16)$$

As any trial radiosity must satisfy boundary conditions (8a) and (8b)

$$\delta B(\mathbf{r}) = 0 \quad \text{on } \Sigma_0 \text{ and } \Sigma_L \quad (17)$$

the first integral in equation (16) is zero. The second and third integrals together vanish due to equation (4) with equation (9a), the fourth and fifth terms combine to cancel from equation (7) with equation (9a), and the last term in equation (16) is zero by equation (5). From the interpretation of K as a probability, the integrals of equation (13) are clearly positive and the second-order term in the variation of Γ is also positive, hence

$$\Gamma\{B, T\} \leq \Gamma\{B^*, T^*\}. \quad (18)$$

To relate $\Gamma\{B, T\}$ to the heat flux J , the extremum form of equation (13) is rewritten, again using property (2a) of K , integration by parts, and the divergence theorem, much in the same manner as in the derivation of equation (16)

$$\begin{aligned} V \Gamma\{B, T\} = & \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r} \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r}' K(\mathbf{r}, \mathbf{r}') B(\mathbf{r}) \\ & \times [B(\mathbf{r}) - B(\mathbf{r}')] + \int_{\Sigma} d^2 \mathbf{r} \int_{\Sigma_0 + \Sigma_L} d^2 \mathbf{r}' K(\mathbf{r}, \mathbf{r}') \end{aligned}$$

$$\begin{aligned} & \times B(\mathbf{r}) [B(\mathbf{r}) - B(\mathbf{r}')] + \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2 \mathbf{r} B(\mathbf{r}) \\ & \times [B(\mathbf{r}) - A - CT(\mathbf{r})] - \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2 \mathbf{r} A \\ & \times [B(\mathbf{r}) - A - CT(\mathbf{r})] - \frac{\varepsilon}{1 - \varepsilon} \int_{\Sigma} d^2 \mathbf{r} CT(\mathbf{r}) \\ & \times [B(\mathbf{r}) - A - CT(\mathbf{r})] + C \int_{\Sigma} d^2 \mathbf{r} T(\mathbf{r}) \eta(\mathbf{r}) \\ & \cdot \lambda \nabla T(\mathbf{r}) - C \int_{V_s} d^3 \mathbf{r} T(\mathbf{r}) \nabla \cdot [\lambda \nabla T(\mathbf{r})]. \end{aligned} \quad (19)$$

The first integral in $V \Gamma\{B, T\}$ is identical to expression (11) for $-V \mathbf{J} \cdot \boldsymbol{\beta}$, the second and third integrals vanish from equations (4) and (9a), and the fifth and sixth integrals combine to zero from equations (7) and (9a). Upon substitution of equations (7) and (9a) into the fourth integral in equation (19) we note as there are no heat sinks or sources within the solid V_s , the fourth term sums over the void–solid interface to zero, and the last integral in equation (19) is zero from equation (5). The variational upper bound on the heat flux is

$$- \mathbf{J} \cdot \boldsymbol{\beta} = \Gamma\{B, T\} \leq \Gamma\{B^*, T^*\}. \quad (20)$$

With the same substitutions applied to equations (4), (5), (7) and (9), i.e. E_s for B , α for ε , $A = 0$, and $2C = k\psi(\gamma + 1)/(\gamma - 1)$, the radiation variational principle becomes a variational upper bound on the conductive thermal flux through a Knudsen void gas–dispersed solid system. Then an exact kinetic theory solution of the Knudsen void gas heat transport problem in a void–solid system is also a solution of the radiation transport problem.

TRIAL FUNCTIONS AND BED STATISTICS

The evaluation of the integrals in the upper bound (13) is considerably simplified if we pass to the limit of a very long slab (let L become large compared to typical bed dimensions, e.g. particle size and average pore diameter). Due to the angular distribution of the radiation diffusely emitted from Σ_0 and Σ_L , and blocking by the solid, radiation from the edges will penetrate only an infinitesimal distance across the slab before striking a surface. The contributions to the upper bound integrals of equation (13) from the end surfaces Σ_0 and Σ_L go to zero as L^{-1} . In a thick slab the radiation heat conductivity should not depend on the nature of the end surfaces. The end conditions in a thick slab are discussed briefly in the Appendix, where it is shown that the radiosity difference ($B_L - B_0$) can be replaced by the black body emission difference $\sigma(T_L^4 - T_0^4)$. Then with the linearization about the average slab temperature, $\boldsymbol{\beta}$ from equations (12) and (9a) becomes

$$\boldsymbol{\beta} = C(T_L - T_0)L^{-1} \mathbf{i} = C\boldsymbol{\theta} \quad (21)$$

with C given by equation (9c). For a thick slab the effective thermal conductivity $\lambda_e = |J/\theta|$, from equations (13), (20) and (21), is bounded above by the variational principle

$$C\theta^2\lambda_e \leq \frac{1}{2V} \int_{\Sigma} d^2\mathbf{r} \int_{\Sigma} d^2\mathbf{r}' K(\mathbf{r}, \mathbf{r}') \\ \times [B^*(\mathbf{r}') - B^*(\mathbf{r})]^2 + \frac{1}{V} \frac{\varepsilon}{1-\varepsilon} \int_{\Sigma} d^2\mathbf{r} \\ \times [B^*(\mathbf{r}) - A - CT^*(\mathbf{r})]^2 + \frac{C}{V} \int_{V_s} d^3\mathbf{r} \lambda [\nabla T^*(\mathbf{r})]^2. \quad (22)$$

A simple selection for the trial temperature is

$$T^*(\mathbf{r}) = T_0 + \theta \cdot \mathbf{r} \quad (23)$$

and for the trial radiosity

$$B^*(\mathbf{r}) = A + CT^* + \omega\theta \cdot \boldsymbol{\eta}(\mathbf{r}) \quad (24)$$

where ω is an adjustable parameter and $\boldsymbol{\eta}(\mathbf{r})$ the surface unit normal at \mathbf{r} on Σ pointing into the void. The linear part of equation (24) allows for a smooth variation across the bed, while the third term in B^* fluctuates (due to $\theta \cdot \boldsymbol{\eta}$) with the random local structure of the void–solid interface Σ .

We assume the void–solid bed is statistically homogeneous and isotropic, define the void fraction ϕ , the void–solid interface area s per unit total volume, and an average pore diameter m , which is four times the void volume to void–solid interfacial area. In addition we define the mean surface area ξ that can be seen (i.e. reached by an unobstructed straight line) from a typical point on the void–solid interface, and the probability $u(\boldsymbol{\eta}) d^2\boldsymbol{\eta}$ that a point on the surface Σ has a surface unit normal $\boldsymbol{\eta}$ pointing into the void and falling within the element of solid angle $d^2\boldsymbol{\eta}$. Finally the probability $f(\boldsymbol{\rho}, \boldsymbol{\eta}, \boldsymbol{\eta}') d^3\rho d^2\boldsymbol{\eta} d^2\boldsymbol{\eta}'$, that two points on the void–solid interface Σ which can see one another, have a relative position vector lying in the volume element $d^3\rho$ about $\boldsymbol{\rho}$, and that the unit normal $\boldsymbol{\eta}$ falls within $d^2\boldsymbol{\eta}$ at the first point on Σ and within the solid angle element $d^2\boldsymbol{\eta}'$ about $\boldsymbol{\eta}'$ at the second, is defined. For a statistically homogeneous, isotropic void–solid bed the upper bound (22), with trial functions (23) and (24), and the form (2b) of K , can be expressed in terms of the five statistical quantities given above

$$C\theta^2\lambda_e \leq -2^{-1}s\xi \int d^2\boldsymbol{\eta} \int d^2\boldsymbol{\eta}' \int d^3\rho f(\boldsymbol{\rho}, \boldsymbol{\eta}, \boldsymbol{\eta}') \\ \times \frac{\boldsymbol{\eta} \cdot \boldsymbol{\rho} \boldsymbol{\eta}' \cdot \boldsymbol{\rho}}{\pi\rho^4} [C\theta \cdot \boldsymbol{\rho} + \omega\theta \cdot (\boldsymbol{\eta}' - \boldsymbol{\eta})]^2 \\ + \frac{\varepsilon}{1-\varepsilon} s \int d^2\boldsymbol{\eta} u(\boldsymbol{\eta}) [\omega\theta \cdot \boldsymbol{\eta}]^2 + C(1-\phi)\lambda\theta^2. \quad (25)$$

To determine the quantities u and f we must either make appropriate measurements in the void–solid system or generate a model material. One simple model

for a pore structure (Fig. 1) is obtained when $V\zeta$ spheres all of the same radius q are placed randomly in a volume V and allowed to freely overlap one another. All those points lying on a sphere surface but not on the interior of an overlapping sphere make up the pore wall surface Σ ; all those points within the interior of one or more spheres make up the solid volume V_s . The randomly overlapping sphere model has been discussed elsewhere [14, 21]. It suffices to point out that the probability P_v that a volume v is free of sphere centers is

$$P_v = \exp(-v\zeta). \quad (26)$$

That a point be in the void requires sphere centers be excluded from a spherical volume of radius q about the point and the probability of finding a point in the void is just the porosity

$$\phi = \exp(-4\pi q^3\zeta/3). \quad (27)$$

The total sphere area, overlapped or not, per unit total volume is $4\pi q^2\zeta$, and the exposed (not overlapped) sphere area per unit total volume is

$$s = 4\pi q^2\zeta\phi. \quad (28)$$

The average pore diameter (four times void volume to void–surface interface area) is

$$m = 4\phi/s = (\pi q^2\zeta)^{-1}. \quad (29)$$

If we consider a point on the exposed surface, all values of $\boldsymbol{\eta}$ are equally likely and the probability

$$u(\boldsymbol{\eta}) d^2\boldsymbol{\eta} = d^2\boldsymbol{\eta}/4\pi. \quad (30)$$

For two points exposed or overlapped lying on different spheres all values of $\boldsymbol{\rho}$, $\boldsymbol{\eta}$ and $\boldsymbol{\eta}'$ are equally likely and the probability of falling within the specified infinitesimals $d^3\rho$, $d^2\boldsymbol{\eta}$ and $d^2\boldsymbol{\eta}'$ is

$$\frac{d^3\rho d^2\boldsymbol{\eta} d^2\boldsymbol{\eta}'}{V 4\pi 4\pi}.$$

The probability $f(\boldsymbol{\rho}, \boldsymbol{\eta}, \boldsymbol{\eta}') d^3\rho d^2\boldsymbol{\eta} d^2\boldsymbol{\eta}'$ also requires the event that the two points are exposed and can see one another, i.e. the probability that no third sphere has its center within a right circular cylinder about $\boldsymbol{\rho} = \mathbf{r}' - \mathbf{r}$ capped at both ends with a hemisphere of radius q . Using equation (26) for the probability that sphere centers are excluded from this volume

$$f(\boldsymbol{\rho}, \boldsymbol{\eta}, \boldsymbol{\eta}') d^3\rho d^2\boldsymbol{\eta} d^2\boldsymbol{\eta}' = \frac{sV d^3\rho d^2\boldsymbol{\eta} d^2\boldsymbol{\eta}'}{\xi\phi^2 V 4\pi 4\pi} \\ \times \exp[-\rho\pi q^2\zeta - 4\pi q^3\zeta/3] \quad (31)$$

if $\boldsymbol{\rho} \cdot \boldsymbol{\eta} \geq 0$ and $\boldsymbol{\rho} \cdot \boldsymbol{\eta}' \leq 0$, and is zero otherwise. Normalization of f then leads to

$$\xi = 128\pi\phi^2/s^2. \quad (32)$$

RESULTS AND DISCUSSION

When the probability functions (27)–(31) for a random bed of overlapping solid spheres are substituted into inequality (25) and the integrals are evaluated, an upper bound on the effective thermal conductivity is obtained

$$\lambda_e \leq \frac{4}{3} \phi m C \left[1 - \frac{4\mu}{3} + \frac{13}{9} \mu^2 + \frac{\varepsilon}{1-\varepsilon} \mu^2 \right] + (1-\phi)\lambda. \quad (33)$$

The right-hand side of inequality (33) is minimized when

$$\mu = \frac{\omega}{mC} = \frac{6(1-\varepsilon)}{9+4(1-\varepsilon)} \quad (34)$$

and the best effective conductivity upper bound for trial functions of the type (23) and (24) is

$$\lambda_e \leq \phi \frac{12mC}{9+4(1-\varepsilon)} + (1-\phi)\lambda. \quad (35)$$

For situations in which all other energy exchange mechanisms, such as conduction, are negligible compared to radiation, Siegel and Howell [12] introduce the important case of thermal radiative equilibrium. For radiative void heat transport $C (= 4\sigma\bar{T}^3)$ is given by equation (9c) and ε is the sphere surface emissivity evaluated at \bar{T} . For thermal radiative equilibrium the solid conduction term in inequality (35) is neglected, and the variational result (35) gives a rigorous upper bound on the thermal radiation conductivity λ_r ,

$$\lambda_e = \lambda_r \leq \phi \left[\frac{48\sigma\bar{T}^3 m}{9+4(1-\varepsilon)} \right]. \quad (36)$$

The temperature trial function (23) when combined with equations (7) and (9a) generates the trial radiosity (24). Hence the variational expressions (35) and (36), for the effective and thermal radiation conductivities, are obtained from the linear temperature trial function. When a random two-phase suspension consists of two Fourier solids, the upper bound variational principle with a linear trial temperature (23) of the type used here gives the parallel bound [22]. It is interesting to note that inequality (35) is also a parallel bound, if we take the square bracketed quantity in expression (36) to be the void thermal radiation conductivity.

To interpret the physical significance of the variational expression (36) for the thermal radiation conductivity three cases are examined—a dilute bed of black spheres ($\phi \rightarrow 1, \varepsilon \rightarrow 1$), a dilute bed of totally reflecting spheres ($\phi \rightarrow 1, \varepsilon \rightarrow 0$), and linear anisotropic scattering. The thermal radiation conductivity upper bound (36), written for the dilute bed ($\phi \rightarrow 1$) black sphere ($\varepsilon \rightarrow 1$) limit, and expressed in terms of the sphere density ζ and sphere radius q from equation (29) for the average pore diameter m , has the form

$$\lambda_r \leq (16\sigma\bar{T}^3)/(3\pi q^2 \zeta), \quad (\phi \rightarrow 1, \varepsilon \rightarrow 1). \quad (37)$$

The absorption coefficient a can also be expressed in terms of ζ and q

$$a = \pi q^2 \zeta Q_a. \quad (38)$$

Tien and Drolen [23] have pointed out that the absorption efficiency Q_a for large particles can be set equal to the particle emissivity, which for black spheres is unity. The variational result (37) for a dilute bed of black spheres can be written in terms of the absorption coefficient (38)

$$\lambda_r \leq 16\sigma\bar{T}^3/(3a), \quad (\phi \rightarrow 1, \varepsilon \rightarrow 1) \quad (39)$$

and we obtain the well-known, rigorous form [12] of the thermal radiation conductivity for isotropic emission with no scattering. A single sphere, though large compared to the characteristic wavelength of thermal radiation λ_r , ($\pi d/\lambda_r > 100$), is still much smaller than the slab thickness L , and for the imposed temperature gradient θ across the slab, its temperature is very nearly uniform. If in addition, the sphere surface is black with no scattering, the radiosity is constant across its surface, the sphere does indeed emit isotropically, and equations (37) and (39) must be exact equalities. Note also that the optimized μ from (34) should be, and in fact is, zero for $\varepsilon = 1$, i.e. the scattering contribution does vanish in the optimized trial radiosity (21) on a black surface.

The thermal radiation conductivity upper bound (36) for a dilute bed ($\phi \rightarrow 1$) of totally reflecting spheres ($\varepsilon \rightarrow 0$)

$$\lambda_r \leq 48\sigma\bar{T}^3 m/13, \quad (\phi \rightarrow 1, \varepsilon \rightarrow 0) \quad (40)$$

is also a rigorous equality. Tien and Drolen [23] have pointed out, that there is a complete equivalence between Knudsen diffusion of a gas in a porous or packed solid and radiant heat transport with diffusive totally reflecting walls ($\varepsilon \rightarrow 0$), in the case of thermal radiative equilibrium. Abbasi and Evans [24] have used this equivalence in Monte Carlo simulations of radiant heat transport in packed beds and porous solids. In the context of Knudsen diffusion, Derjaguin [13] has proposed (40) as an equality, and Strieder and Prager [14] have shown that the right-hand side of (40) does approach the true transport coefficient, but only in the limit of a very dilute bed of spheres. There is always a local anisotropy for diffusive reflection off large spheres ($\pi d/\lambda_r > 100$), determined by the orientation of the sphere surface elements. As the 'average' photon approaches in the direction of the net flux, impacts, and scatters diffusely from the sphere surface, there are always some forward directions rendered inaccessible by the presence of the solid sphere; Derjaguin's coefficient (40) includes significant anisotropic backscatter.

Tien and Drolen [23] have stated that the semi-isotropic scattering, two-flux model is not an accurate approximation for the highly anisotropic scattering from the large particles considered here. For the same reasons, except near $\varepsilon = 1$, the isotropic scattering radiant thermal conductivity [12] should not work

very well either, but it is an upper bound as we shall show. The choice, $\mu = 0$, in inequality (33) is legitimate for *any* sphere emissivity, if in addition we consider a dilute sphere bed ($\phi \rightarrow 1$) with thermal radiative equilibrium, inequality (33) for $\mu = 0$ becomes

$$\lambda_r \leq 16\sigma\bar{T}^3/(3\pi q^2\zeta) \quad (41)$$

where $C (= 4\sigma\bar{T}^3)$ was given by equation (9c), and the average pore diameter was written in terms of the sphere density ζ and sphere radius q by equation (29). Tien and Drolen [23] have noted that the scattering efficiency Q_s for large particles can be taken as $(1 - Q_a)$, and the scattering coefficient σ_s ,

$$\sigma_s = \pi q^2\zeta Q_s = \pi q^2\zeta(1 - Q_a). \quad (42)$$

The extinction coefficient κ is by definition the sum of the absorption and scattering coefficients, then from equations (38) and (42)

$$\kappa = a + \sigma_s = \pi q^2\zeta \quad (43)$$

and with equation (43) the inequality (41) becomes

$$\lambda_r \leq 16\sigma\bar{T}^3/3\kappa. \quad (44)$$

The isotropic scattering case [12] is an upper bound on the thermal radiant conductivity for a dilute bed of diffusely scattering spheres. An optimized, non-zero selection of μ in inequality (33) will give anisotropic results.

The variational result (36) can be related to the simple linear anisotropic scattering approximation [15]. For a dilute sphere bed ($\phi \rightarrow 1$), and with $(\pi q^2\zeta)^{-1}$ in place of the average sphere diameter m from equation (29), the variational upper bound (36) has the form

$$\lambda_r \leq 48\sigma\bar{T}^3/[9 + 4(1 - \varepsilon)]\pi q^2\zeta \quad (45)$$

then including the extinction coefficient from equation (43)

$$\lambda_r \leq 48\sigma\bar{T}^3/[9 + 4(1 - \varepsilon)]\kappa. \quad (46)$$

For an optically thick slab of large spherical particles ($\pi d/\lambda_v > 100$) and using the linear anisotropic scattering approximation, Dayan and Tien (equation (27) of ref. [15]) have derived the approximate equality

$$\lambda_r = 16\sigma\bar{T}^3/[3 - z(1 - \varepsilon)]\kappa, \quad (\text{L.A.S.}) \quad (47)$$

In equation (47), we have replaced Dayan and Tien's single scattering albedo ω_0 with its straightforward emissivity dependence [23] of $(1 - \varepsilon)$. For the simple linear anisotropic scattering approximation the phase function has been approximated by unity, plus a correction term linear in the cosine of the scattering angle. The coefficient z of the cosine term must be assigned a value. For $\phi \rightarrow 1$ and $\varepsilon \rightarrow 1$, λ_r from equation (47) already coincides with the exact result (39). As Derjaguin's coefficient is exact in the limit $\phi \rightarrow 1$, $\varepsilon \rightarrow 0$, we can require equation (47) in this limit to coincide with inequality (40), and this sets a value of z at

$$z = -4/3. \quad (48)$$

Dayan and Tien [15] note that $z \rightarrow 1$ represents strong forward scattering, while $z \rightarrow -1$ implies strong backward scattering. Strictly speaking the absolute value of z should be less than or equal to one to avoid negative values of the phase function, but Dayan and Tien [15] observe because of the approximate nature of the linear phase function, that the absolute value of z in many cases exceeds unity. In addition to very strong anisotropic backscattering, the variational upper bound result (46) establishes the simple linear anisotropic scattering approximation result (47) with $z = -4/3$ is a rigorous upper bound on the thermal radiation conductivity, that reduces to the exact physical forms in both of the extremes, $\varepsilon = 0$ and 1. These features suggest that inequality (46) might be useful as an estimate of the radiant conductivity.

As the void fraction is decreased from unity the bed of randomly placed, freely overlapping solid spheres resembles less a gray gas, and becomes more a packed solid or porous medium. Van Kreveld and Van den Hoed [25] have successfully used randomly overlapping spheres to model silica gels. Overlapping spheres should be appropriate as a model for sintered ceramic materials prepared by high temperature synthesis reactions [26]. To handle radiation in these structures, it would have been necessary to use one of the packed bed void thermal radiation conductivity equations from the literature [6–11]. Unfortunately, none of these void thermal radiation conductivity equations correctly include multiple scattering [6] from void to adjacent void regions. In the most appropriate model, Wakao and Kato [8] performed radiation transport calculations across the close packed layers of an orthorhombic lattice. Wakao and Kato treated the open areas between unit cells incorrectly, with the same emissivity and reflection laws as the solid sphere surface. As a result, Wakao and Kato's radiant thermal conductivity equation gives very poor results for surface emissivities much less than unity, and incorrectly vanishes as ε goes to zero. On the other hand, the variational equations apply for any void fraction and surface emissivity, and have the added advantage of properly including multiple scattering events from void to adjacent void regions within the void thermal radiation conductivity expression (square bracketed term in inequality (36)).

With the substitution of α for ε and $2C = 8\sigma\bar{T}^3 = k\psi(\gamma + 1)/(\gamma - 1)$, the variational upper bound expression (36) applies for void gas, Knudsen heat transport

$$\lambda_{\kappa n} \leq \phi \left[\frac{6mk\psi(\gamma + 1)/(\gamma - 1)}{9 + 4(1 - \alpha)} \right]. \quad (49)$$

Lassette [16, 17] has formulated rigorous Boltzmann type equations to predict Knudsen gas transport rates through a dilute bed of solid spheres ($\phi \rightarrow 1$). In particular in the limits of no accommodation ($\alpha \rightarrow 0$) and perfect accommodation ($\alpha \rightarrow 1$), Lassette has

obtained exact solutions that apply to Knudsen void gas heat transport rates, and that coincide with inequality (49) in both limits. Then for Knudsen void gas transport in the limits ($\phi \rightarrow 1$, $\alpha \rightarrow 1$) and ($\phi \rightarrow 1$, $\alpha \rightarrow 0$) the variational result is an exact equality, elsewhere it is an upper bound.

SUMMARY AND CONCLUSIONS

A variational upper bound principle has been formulated for the effective thermal conductivity and thermal radiation conductivity for a void-solid system with diffusely emitting and scattering surfaces. The equations are based on a radiosity formulation and rigorously include the effects of successive multiple scatterings down the bed. General forms of the variational expressions have been written in terms of appropriate probability functions for a random void-solid bed.

Explicit results have been calculated for a linear temperature trial function over a slab of randomly placed, freely overlapping solid spheres all of the same radius. In the context of such an explicit structure the issue arises of isotropic vs anisotropic scattering. The variational upper bound expression (36) for the thermal radiation conductivity for a dilute sphere bed ($\phi \rightarrow 1$) of black spheres ($\varepsilon \rightarrow 1$) has been shown to coincide with the well-known, exact gray gas result [12] for isotropic emission from the spheres. In the opposite limit of a dilute bed of spheres ($\phi \rightarrow 1$) with no absorption ($\varepsilon \rightarrow 0$), the variational result has been shown to reduce to Derjaguin's coefficient [13, 14], which is also known to be the exact solution for anisotropic scattering in a dilute totally reflecting sphere bed. Arguments were presented to demonstrate that the isotropic scattering form (44) is an upper bound on the radiant conductivity for any emissivity, though a good estimate only near $\varepsilon = 1$. Also for emissivities from zero to unity, the variational radiant conductivity (46) has been compared with linear anisotropic scattering theory [15]. For the proper selection of the phase function constant, the linear anisotropic scattering and the variational radiant conductivity expressions are the same. The selected value of the phase function constant implies a very strong anisotropic backscatter. The linear anisotropic scattering-variational radiation conductivity (46) is a rigorous upper bound, that reduces to the exact forms in both the limits $\varepsilon = 0$ and 1. This second upper bound (46) is a significant improvement over the previous isotropic bound (44), e.g. reduced by 31% for $\varepsilon = 0$. These features suggest that inequality (46) might be useful as an estimate of the radiant conductivity. For the identical problem of Knudsen void gas conduction, dilute sphere kinetic theory results [16, 17] establish that the variational void conductivity is an exact solution in both the limits of no accommodation and perfect accommodation at the spherical particle surface. The variational principle provides a means to obtain useful estimates and upper bounds on the

thermal radiation conductivity, that rigorously includes both multiple and anisotropic scattering in a bed of large spheres.

Improvement of the variational upper bound should, of course, be possible, but the calculations involved increase rapidly with increasing sophistication of the trial function, even if we restrict ourselves to the case of randomly overlapping spheres. Efforts to improve the trial function will include a superposition [22] of the local heat fluxes around (insulating) or through (conducting) each of the dispersed solid particles. This trial function will take into account the fact that different spheres have different surroundings, and will provide the proper physical basis [27] to study the interaction of solid and void radiation modes of heat transport.

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APPENDIX

In engineering models for thermal conduction, the thermal energy flux J per unit total cross sectional area of a void-solid system is customarily related to the overall gradient $(T_L - T_0)/L$ across a slab with

$$J = -\lambda_e(T_L - T_0)/L. \quad (\text{A1})$$

On a physical basis we presume, because the slab is thick, that the effective conductivity λ_e does not depend on slab thickness L . If the conductivity in the solid $-\lambda\eta \cdot \nabla T$ is replaced by the thermal flux J , equation (7) can be written for the edge plates Σ_0 and Σ_L , respectively

$$B_0 = \sigma T_0^4 - (1 - \varepsilon_0)J/\varepsilon_0 \quad (\text{A2})$$

and

$$B_L = \sigma T_L^4 + (1 - \varepsilon_L)J/\varepsilon_L. \quad (\text{A3})$$

Equation (A2) is subtracted from equation (A3), the resulting equation is linearized in $\Delta T/\bar{T}$ and (A1) is substituted for the thermal flux, to give

$$B_L - B_0 = 4\sigma\bar{T}^3(T_L - T_0) - \{(1 - \varepsilon_L)/\varepsilon_L + (1 - \varepsilon_0)/\varepsilon_0\}\lambda_e(T_L - T_0)/L. \quad (\text{A4})$$

The second term on the right-hand side of equation (A4), of order L^{-1} , can be neglected when L is large, and from equations (12) and (9c) for a thick slab

$$\beta = C(T_L - T_0)i/L. \quad (\text{A5})$$

CONDUCTIVITE ET RAYONNEMENT POUR UN MILIEU VIDE-SOLIDE AVEC DES SURFACES REFLECHISSANTES

Résumé—Un principe variationnel est formulé pour le transfert radiatif à travers les espaces vides et la conduction dans le solide, le système vide-solide étant arbitraire et possédant des surfaces à réflexion diffuse. Les limites supérieures variationnelles de la conductivité effective d'une suspension vide-solide sont exprimées en fonction de certaines moyennes qui caractérisent la géométrie due au hasard. Une limite supérieure de la conductivité, calculée pour un milieu poreux constitué de sphères placées au hasard, est comparée avec des résultats bien connus de la théorie cinétique et du transfert de chaleur par rayonnement. Le principe variationnel fournit un moyen pour obtenir des estimations utiles et des limites supérieures de la conductivité thermique qui incluent la dispersion diffuse anisotrope dans un lit de grosses particules.

STRAHLUNGSTRANSPORT IN EINEM BELIEBIGEN PORÖSEN MEDIUM MIT DIFFUS REFLEKTIERENDEN OBERFLÄCHEN

Zusammenfassung—Für die Strahlungswärmeübertragung durch den Hohlraum und gleichzeitige Wärmeleitung im Festkörper eines beliebigen Hohlraum/Festkörper-Systems mit diffus reflektierenden Oberflächen wird ein Variationsprinzip formuliert. Die variablen Obergrenzen der effektiven Wärmeleitfähigkeit der Suspension werden durch geeignete Mittelwerte ausgedrückt, welche die zufällige Geometrie charakterisieren. Die Obergrenze der Strahlungswärmeübertragung in einem porösen Medium mit zufällig platzierten überlappenden Kugeln wird mit bekannten Ergebnissen aus der kinetischen Theorie und der Theorie der Wärmestrahlung verglichen. Das Variationsprinzip erweist sich als nützliches Werkzeug, um Abschätzungen und Obergrenzen für den Wärmetransport durch Wärmestrahlung bei mehrfacher und anisotrop diffuser Streuung in einer Schüttung großer Partikel zu erhalten.

РАДИАЦИОННАЯ ТЕПЛОПРОВОДНОСТЬ В ТВЕРДОМ ТЕЛЕ СО СЛУЧАЙНО РАСПРЕДЕЛЕННЫМИ ПУСТОТАМИ ПРИ ДИФФУЗНО ОТРАЖАЮЩИХ ПОВЕРХНОСТЯХ

Аннотация—Сформулирован вариационный принцип для радиационного теплопереноса через пустоты и теплопроводности в твердом теле со случайным распределением пустот с диффузно отражающими поверхностями. Верхняя граница теплопроводности такой системы выражена через некоторые средние значения, характеризующие случайное распределение. Верхняя граница радиационного теплопереноса, рассчитанного для модельной пористой среды, образованной хаотически расположенными перекрывающимися сферами, сравнивается с известными результатами из кинетической теории и радиационного теплопереноса. Вариационный принцип используется для получения полезных оценок и верхних границ радиационного теплопереноса, включая многократное и анизотропное диффузное рассеяние в слое крупных частиц.