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## HEAT TRANSFER CHARACTERISTICS OF A GAS-PARTICLE MIXTURE UNDER DIRECT RADIANT HEATING

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#### ABSTRACT

In this paper we first develop a new model for calculating the rate of heat transfer from a particle to a surrounding gas. The model, which assumes no relative motion between the particle and gas, is based on kinetic theory and is applicable at all Knudsen numbers. Then we use the result in a sample application in which a gas-particle suspension is radiantly heated. Both the temperature difference between the particles and gas and the rate of gas temperature increase are shown to have theoretical maximums. Using the optical and thermal properties of carbon, some numerical results for a carbon particle/air mixture are presented.

# Introduction

The rate of heat transfer from small particles suspended or immersed in a gas to the surrounding gas is an important quantity in many areas of research. It plays a role in determining ice formation in noctilucent clouds and in governing airborne aerosol equilibrium temperatures [1]. The aerosol particles may have a temperature that is significantly different than that of the atmosphere. This heat transfer rate is also important in combustion systems where soot or other particles are present [2]. A process in which knowledge of the particle-gas energy exchange rate is especially important is the use of small particles suspended in a gas to directly capture solar energy [3]. At Lawrence Berkeley Laboratory, this concept led to the development of SPHER (Small Particle Heat Exchange Receiver) [4]. It is this latter application that inspired this paper.

#### Heat Transfer Model

The heat transfer from a particle to a surrounding gas may be broken into three regimes depending on the value of the Knudsen number (Kn), defined as  $\lambda/d$ , where  $\lambda$  is the gas molecule mean free path and d is the characteristic dimension of the body in the gas (d is the diameter if the body is a sphere). For  $Kn < 10^{-3}$  the continuum approximation applies and different equations are available to calculate the heat transfer rate depending on whether the losses are purely conductive, or also convective, in nature  $[5]$ . For  $Kn > 10$  free molecular flow conditions prevail near the particle; again a fairly well recognized expression for the heat transfer based on molecular collisions exists [1]. In the transition region,  $10^{-3}$  <  $Kn$  < 10, analytical modeling of heat transfer is difficult because neither a continuum nor a kinetic theory approach is strictly correct. Currently, the standard approach to calculate heat transfer in this region is almost entirely empirical [6,7]. Three analytic treatments [8,9,10] of this problem are known to the authors, but their results do not appear to be in wide use. In the following, a simple treatment of particle-gas heat transfer is derived for arbitrary Kn; in particular, it applies in the transition region.

The model used is the following: a spherical particle with radius a is stationary in an infinite gaseous medium with temperature  $T_{\infty}$  as  $r \to \infty$  (r is the radial coordinate with origin at the center of the sphere as illustrated in Fig. 1). The region around the sphere is divided into two zones. Outside a sphere of radius  $\lambda + a$  continuum conduction is assumed to hold. Within one mean free path of the surface it is assumed that the gas molecules collide only with the particle and not with one another. The effect of convection is neglected. The molecules striking the particle are assumed to have a Maxweliian velocity distribution at temperature  $T_B$ , the zone boundary temperature. The particle is maintained at a fixed temperature  $T_p$ ; energy is supplied or removed by radiation or chemical reaction. Here the particle temperature is assumed to be greater than the gas temperature, but the same analysis applies in the other case.

First, the steady state conduction equation is solved in spherical coordinates for the temperature field outside a radius of  $a+\lambda$ . Using the boundary condition  $T = T_{\infty}$  as  $r \to \infty$  the solution is

$$
T = \frac{A}{r} + T_{\infty} \tag{1}
$$

with constant A to be determined.

To find the energy being carried to the particle we first calculate its collision rate with the gas molecules. From kinetic theory, the molecular flux density crossing a plane in one direction  $(\Gamma)$  can be obtained, if a Maxwellian velocity distribution is assumed [11]. Applying this to the zone boundary, a total inward flow of  $4\pi(\lambda+a)^2\Gamma$  results. However, not all of these molecules strike the particle. If the molecules are randomly directed as they cross the boundary, then an analysis similar to that for obtaining shape factors in radiation heat transfer will show that only the fraction  $a^2/((a + \lambda)^2)$  will hit the particle  $[12]$ . Thus the particle suffers  $4\pi a^2\Gamma$  collisions per unit time.

From kinetic theory, the average energy of a molecule striking the particle surface is  $\epsilon_{in} + 2k_B T_B$ , where  $\epsilon_{in}$  is the internal energy if the molecule is not monatomic [11]. We note that  $2k_B T_B$ , and not  $(3/2)k_B T_B$ , appears because faster molecules hit the surface with greater frequency than do slower molecules and carry more kinetic energy. The average energy of the colliding molecule multiplied by the collision rate gives the total energy into the particle. To calculate the energy flowing away from the particle, the energy of a molecule as it leaves the particle must be determined. From kinetic theory, if any adsorption potential is neglected, it can be assumed that the molecules leave with a Maxwellian velocity distribution at a temperature between  $T_P$  and  $T_B$ , written as  $T_{\text{out}} = (T_P - T_B)\alpha + T_B$ , where  $\alpha$  is termed the accommodation coefficient [13]. If  $\alpha = 1$  (perfect accommodation) the molecules leave at  $T_P$ ; if  $\alpha = 0$  no energy is exchanged and they leave at  $T_P$ . In principle,  $\alpha$  may be different for translational energy exchange than for internal energy exchange; here an average  $\alpha$  is employed.





Nusselt number vs. Knudsen number

The average energy exchanged in a collision is  $(\epsilon_{in}(out) + 2k_B T_{out}) - (\epsilon_{in}(B) + 2k_B T_B)$ . If we are in a temperature range where the specific heat is roughly constant with temperature this can he simplified to  $\alpha(C_m - k_B/2)(T_P - T_B)$  where  $C_m - T$  has replaced  $\epsilon_{in} + 3/2k_B T$ , and  $C_m$  is the specific heat of the gas. Combining this with the collision rate gives

$$
Q_e = 4\pi a^2 n_g \alpha \left(\frac{k_B T_B}{2\pi m_g}\right)^k \left(C_s m_g + \frac{k_B}{2}\right) \left(T_P - T_B\right)
$$
 (2)

where  $Q_{\epsilon}$  is the rate of heat transfer from the particle to the gas; note that  $T_B$  is not yet known. In steady state the total outward heat flow  $Q<sub>c</sub>$  is independent of r. Using equation (2) and the heat flow in the continuum region (obtained from differentiating equation  $(1)$ ) A can be calculated. With A determined, the continuum temperature distribution (1) may be used to solve for  $T_B$ . The result is:

$$
T_B = \frac{T_{\infty} + \frac{\alpha \Phi}{Kn (2Kn + 1)\pi} T_P}{1 + \frac{\alpha \Phi}{Kn (2Kn + 1)\pi}}
$$
(3)

where  $Kn = \lambda/2a$  and  $\Phi = 32/75$  for a monatomic gas and 48/95 for a diatomic gas. Equation (5) gives the temperature one mean free path from the particle. In deriving equation (3) the thermal conductivity has been replaced by its kinetic theory value  $[13]$ :

$$
k = constant \times \pi C_{\bullet} m_{g} n_{g} \lambda \left( \frac{k_{B} T_{B}}{2 \pi m_{g}} \right)^{1/2}
$$
 (4)

where the constant is 25/16 for a monatomic gas and 19/16 for a diatomic gas. To obtain the constant the diatomic gas was assumed to have its rotational modes excited, but not its vibrational modes. Eucken's formula with  $\gamma = 7/5$  was used. If the molecule can also store energy in vibration (this depends on the gas and the temperature range), then the constant must be adjusted.

Now that  $T_B$  has been evaluated, it can be used in the expression for the heat transfer  $Q_e$  giving:

$$
Q_c = \frac{4\alpha a k \Phi (T_P - T_\infty)}{Kn + \frac{\Phi \alpha}{(2Kn + 1)\pi}}
$$
(5)

As a check on the validity of equation (5), the limits as *Kn* approaches zero and infinity can be evaluated. As  $Kn \to 0$ ,  $Q_e \to 4\pi ak(T_p - T_\infty)$ . If  $Q_e$  is set equal to an equivalent convection loss  $4h ~\pi a^2(T_p - T_{\infty})$  then  $2ah / k = Nu = 2$ , a familiar result from continuum heat transfer for a motionless fluid.

As  $Kn \to \infty$ 

$$
Q_c = 4\alpha ak \frac{\Phi}{Kn}(T_p - T_\infty)
$$

where the fact that  $T_B \to T_\infty$  as  $Kn \to \infty$  has been used. When k is replaced by equation (6) this becomes the formula used by Fiocco and Grams [1] corresponding to a temperature jump at the particle surface.

The Nusselt number as a function of Kn can be calculated for the general case by setting  $Q_{\epsilon}$  in (5) equal to  $4h \pi a^2(T_p - T_\infty)$  and solving for  $Nu = 2ah/k$ . This result is plotted in Fig. 2. By finding the appropriate *Kn, Nu* can easily be determined from the graph. Also shown in Fig. 2 is a value of Nu

taken from Clift [6] for the transition region. Clift does not state the origin of this result, only that it matches the data of Takao [8]. One final point to note is that Nu decreases as Kn increases, and the heat flux per area from a particle is  $q = (Nuk/2a)(T_p - T_\infty)$ . So as the particle is made smaller, q increases as long as Nu does not decrease faster than  $a$  (which it does not). This means that it is increasingly ditflcult for a small particle to be at a different temperature than the surrounding gas as the particle size decreases. (We will see this also in the Application section.) Particles in the upper atmosphere may maintain different relative temperatures only because  $\lambda$  is so large, which makes  $N<sup>u</sup>$  small (see results of Fiocco and Grams).

### Application

One application of equation (5) is in an energy balance on a particle in a radiant field. Consider a gas-particle mixture irradiated by an energy beam as shown in Fig. 3. Assuming that the physical dimensions of the mixture is large compared to the particle radius, and utilizing symmetry, each particle can be considered as being surrounded by a unit cell of gas as shown in the same figure. The boundary of the cell is assumed to be adiabatic with respect to conduction heat transfer, but transparent to radiation. The energy balance of the particle and its surrounding gas within the cell can be written as

$$
\frac{4}{3}\pi a^3\rho_p C\frac{dT_p}{dt} = Q_e - Q_e - Q_e \qquad (6)
$$

$$
M_{g} C_{p} \frac{dT_{g}}{dt} = Q_{e}
$$
 (7)

where  $a$ ,  $\rho_p$ ,  $C$ , and  $T_p$  are the radius, density, specific heat, and temperature of the particle.  $M_q$ ,  $C_p$ , and  $T_q$  are the mass of gas within the unit cell, the specific heat at constant pressure of the gas, and its temperature.  $Q_e$  is the conduction heat transfer from the particle to the gas.  $Q_e$  is the absorption by the particle of the incident radiant flux.  $Q_e$  is the overall radiant heat transfer between the particle and its environment. The gas is assumed to be optically transparent in the development of equations (6) and (7). In terms of the initial mass loading of the mixture  $G$  (which is defined as the mass of solid per unit volume of the mixture),  $M_a$  can be written as

$$
M_g = \frac{4}{3} \pi a^3 \rho_{g0} \left( \frac{\rho_p}{G} - 1 \right) \tag{8}
$$

with  $\rho_{go}$  the initial density of the gas in the mixture. Since the gas is assumed to be heated at constant pressure, the volume of the unit cell increases with the gas temperature.

Assuming that the incident radiation comes from a blackbody of temperature *Ti* with and equivalent emissivity  $\epsilon_i$ ,  $Q_a$  can be written as

$$
Q_{\epsilon} = \pi a^2 \epsilon(T_i, a) Q_i \tag{9}
$$

where

$$
\epsilon(T,a) = \frac{1}{\sigma T^4} \int_0^{\infty} Q_{\text{obs}} e_{\lambda b}(T) d\lambda
$$
 (10)

**and** 

$$
Q_i = \epsilon_i \sigma T_i^4 \tag{11}
$$

with  $Q_{obs}$  being the absorption cross section of the particle, and  $e_{\lambda b} (T)$  the blackbody emissive power. The mathematical expression of  $Q_{obs}$ , which is a function of the particle radius and optical properties, can be obtained from standard Mie theory [14]. In equation (11),  $\epsilon_i$  includes the effect of geometry and also the "shadowing" effect of the neighboring particles. For an incoming solar beam focused with a concentrating factor  $\omega$ , and assuming that there is no shadowing from the neighboring particles,  $\epsilon_i$  can be written as  $\epsilon_i = 0.1245 \times 10^{-4} \omega$ . An atmospheric transmissivity of 0.6 has been assumed.



Particle in Flux and Unit Cell of Gas

Absorption Efficiency vs. Size Parameter for Carbon

In SPHER [3,4], a particle/air mass ratio of approximately  $10^{-4}$  (corresponding to a mass loading of  $G = 0.1$  g/m<sup>3</sup>) over an absorbing path length ranging from 0.6 to 0.9 m was observed to be sufficient for total absorption of the incoming solar radiation. In such applications, the ratio of the unit cell radius to the particle radius is typically greater than 100. The conduction heat transfer  $Q_c$  is thus equivalent to that between a particle and an infinite medium so that equation (5) applies.

The radiative exchange between the particle and its environment is written as

$$
Q_{\epsilon} = 4\pi a^2 [\epsilon (T_p, a) \sigma T_p^4 - \epsilon (T_{\infty}, a) \sigma T_p^4]
$$
 (12)

with  $T_{\infty}$  being an effective radiative temperature of the particle environment. Due to the uncertainty of parameters such as  $\epsilon_i$  and  $T_{\infty}$ , additional specification of the physical environment of the particle is clearly required before solutions to equations (6) and (7) can be obtained. However, interesting conclusions regarding the general behavior of  $T_p$  and  $T_q$  can be generated by considering the qualitative mathematical behavior of the heat transfer terms  $Q_{\epsilon}$ ,  $Q_{\epsilon}$ , and  $Q_{\epsilon}$ .

From thermodynamic considerations, it can be readily observed that  $dT_p/dt$ ,  $Q_e$ ,  $Q_e$ , and  $Q_e$  are all positive-definite quantities. Since  $Q_a$  is constant for a given particle size and geometry, and  $Q_e$ increases with the temperature difference  $\Delta T = T_p - T_g$ , the condition

$$
Q_{\bullet} - Q_{\circ} \geq 0 \tag{13}
$$

yields a theoretical maximum temperature difference between the particle and gas for a given irradiation. For carbon particles, the absorption efficiency,  $\Psi_{obs}$ , as a function of the particle size d and the particle size parameter  $\pi d/\lambda$  is calculated from Mie theory and shown in Fig. 4. The dielectric functions available in the literature [15,16] are used in the Mie calculation. For each particle size  $\Psi_{obs}$  is calculated from a wavelength of 0.33  $\mu$ m to 12.4  $\mu$ m. Assuming that results in Fig. 4 are applicable over all temperatures, and that the accommodation coefficient in equation (5) is equal to l, the normalized theoretical maximum temperature difference  $(\Delta T_{\text{max}}/Q_i)$  between gas and particles can be found. The results are plotted as a function of particle diameter and gas temperature in Fig. 5, for solar irradiation with different concentration factors (note that the incoming radiant flux is 1  $kW/m<sup>2</sup>$  when the concentration is 1). It is interesting to note that both in the large and small particle limits, the maximum temperature difference is proportional to the particle diameter. The constants of proportionality, however, are different due to the different controlling heat transfer mechanisms (continuum conduction and geometric absorption in the large particle limit and free-molecular-flow conduction and Rayleigh absorption in the small particle limit). An increase in the gas temperature increases the maximum temperature difference for small particles but has the opposite effect for large particles. For an incident radiant flux of 100 kW/m<sup>2</sup> and particle size between 1 and 10  $\mu$ m, Fig. 5 indicates a maximum temperature difference of about 1 °K. This is consistent with experimental observations [3,4].

In many applications (including the design of SPHER) it is important to estimate the rate of temperature increase of the gas. Based on equation (7) and the fact that  $Q_e$  is a maximum and equal to  $Q_e$ at the maximum temperature difference, a normalized rate of gas temperature increase can be defined and expressed as

$$
\frac{1}{Q_i G} \left( \frac{dT_{\theta}}{dt} \right)_{\text{max}} = \frac{\epsilon (T_i, a)}{a} \left( \frac{1}{3} \pi C_p \rho_p \rho_{\theta \theta} \right)^{-1} \tag{14}
$$

The mass loading is assumed to be much smaller than the particle solid density in the development of the above equation. For a carbon/air mixture initially at STP, numerical values of the normalized rate of temperature increase are plotted in Fig. 6. In the small particle limit, the normalized heating rate is independent of particle radius due to the Rayleigh absorption effect. The gas heating rate generally decresses as the particle radius increases. For a mass loading of  $G = 1$  g/m<sup>3</sup> and an incident solar flux of 100 kW/m<sup>2</sup>, results in Fig. 6 can be converted to maximum heating rates for different particle sizes. Numerical values are shown on the vertical axis on the right of the same figure. For particle sizes in the range of 1 to 10  $\mu$ m, the maximum heating rate is on the order of 10 to 50 K/sec. However, since the heating rate decreases rapidly with increasing particle size (to  $0.5$  K/sec at 100  $\mu$ m, for example) the choice of particle size is critical to the effectiveness of a gas-particle suspension as a heat transfer fluid.



Normalized Temperature Difference vs. Particle Size Normalized Heating Rate vs. Particle Size

# Conclusion

In this paper we have developed a simple two zone model for heat transfer from a particle to a surrounding gas at arbitrary *Kn.* Convection was neglected, and a Maxwellian distribution for the gas was used, but the treatment is applicable to many cases of interest. The expression for the heat transfer rate depends on *Kn* and was shown to reach the appropriate limits as *Kn* goes to zero or infinity. The primary value of the equation, however, lies in its ability to predict the energy exchange at values of  $Kn \simeq$ I where a useful expression with an analytic basis was not previously in use. As an example of an application of this formula we have made estimates of the maximum temperature difference between a particle and gas in a suspension subject to radiant heating. These indicate that for particles under 10  $\mu$ m the temperature difference is less than 1 °K for incident fluxes up to 100 kW/m<sup>2</sup>. Calculations also show that the maximum heating rate for a gas- particle suspension is strongly dependent on particle size and mass loading.

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# Nomenclature



- $\lambda$  mean free path of gas molecules **(m)**
- Φ gas-dependent constant in eq. 6
- $\rho_g$  gas density  $(\text{kg/m}^3)$
- $\rho_{\rm s}$  particle density (kg/m<sup>3</sup>)
- Stefan-Boltzmann constant  $\sigma$
- Ŵ absorption efficiency
- concentration factor  $\omega$

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