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Study of Heat Conduction between Fractal Aggregates and the Surrounding Gas in the Transition Regime Using the DSMC Method

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Heat conduction between fractal soot aggregates and the surrounding gas is one of the fundamental problems encountered in the theoretical analysis of soot particle size measurement using the laser-induced incandescence (LII) techniques. Improved the modelling capability of this fundamental phenomenon helps improve the accuracy of LII based techniques for soot particle sizing. Heat conduction rate between fractal aggregates and the surrounding monatomic gas in the transition regime was calculated using the direct simulation Monte Carlo (DSMC) method. The fractal aggregates were numerically generated using a combination of particle-cluster and cluster-cluster aggregation algorithms. The DSMC results were analyzed using the Fuchs approach for heat conduction between a spherical particle and the surrounding gas in the transition regime to arrive at the size of the heat transfer equivalent sphere for fractal aggregates. The heat transfer equivalent sphere radius for fractal aggregate is required in the formulation of aggregate based LII models. Such heat transfer equivalent sphere radius is recommended as the characteristic length scale of fractal aggregate heat conduction and is compared with the radius of gyration and the projected area based sphere radius.

I. Introduction

THE phenomenon of laser-induced incandescence (LII) has been utilized to develop powerful diagnostic techniques for spatially and temporally resolved quantitative measurements of soot volume fraction in many applications ranging from laminar flames, in-cylinder engine combustion, to engine exhaust [1 and references cited therein]. The principle of this technique is the rapid heating of soot particles to temperatures close to their sublimation temperatures (about 4000 K) using a high-energy short laser pulse of about 20 ns duration. During and after the laser heating, soot particles experience heat loss through sublimation and heat conduction. Sublimation rapidly lowers the soot temperature and ceases being important shortly after the laser pulse. Conduction takes place during the entire LII process and becomes the dominant particle cooling mechanism shortly after the laser pulse when sublimation essentially vanishes.

LII has also been demonstrated to be a simple method to infer the diameter of primary soot particles [2-6]. Different strategies have been proposed in the literature for the purpose of soot particle sizing using LII. Recent studies advocate the use of time-resolved incandescence intensity decay curve for the reasons that (1) it makes full use of available information, and (2) it carries information about the soot particle size distribution. One of the strategies for particle sizing using LII is to relate the temporal decay rate of soot particle temperature determined from the ratio of LII signals detected at two wavelengths in the visible to the primary particle size using a theoretical model for the particle cooling rate through conduction [3,6]. In fact, it has been shown by Liu et al. [6] that the initial soot temperature decay rate in low-fluence LII, where the peak soot temperature is sufficiently low that sublimation can be neglected, can be used to determine the Sauter mean of primary particle size.

Experimental evidence indicates that the primary soot particle diameters fall in the range between 10 and 60 nm from various combustion sources and under different combustion conditions. However, combustion-generated soot do not stay as isolated spherical particles but forms fractal-like aggregates containing nearly spherical primary particles as a result of particle aggregation [7,8]. Such fractal structure of soot particles may have significant implications for the heat and mass transfer processes experienced by soot during LII. To account for the effect of soot particle aggregation on its conduction heat loss, it is essential to formulate the LII model for a fractal aggregate, rather than an isolated primary particle [9,10].

To minimize or avoid soot mass loss due to sublimation for the purpose of soot particle sizing utilizing the temporal decay rate of soot temperature, it is highly desirable to conduct LII experiments in the low-fluence regime. On the other hand, it is in general also advantageous to use relatively high laser-fluences to gain adequate signal-to-noise ratios. Thus the laser fluence used in low-fluence LII experiments should be carefully selected. The upper limits of the laser fluence considered in the low-fluence LII regime can be estimated based on numerical modeling as demonstrated by Liu et al. [6]. In low-fluence LII, heat conduction between soot aggregates and the surrounding gas is the dominant soot particle heat loss mechanism at atmospheric and elevated pressures. This heat conduction process occurs in general in the transition regime between the free-molecular and the continuum regimes. Quantitative understanding of heat conduction between soot aggregates and the surrounding gas is crucial to interpret the experimentally measured time-resolved soot particle temperatures and to infer the primary particle diameter. Unfortunately, accurate calculations of heat conduction loss from fractal soot aggregates to the surrounding gas encounter two main challenges. First, the thermal accommodation coefficient of soot, which has significant impact on soot particle heat conduction loss rate in the free and near-free molecular regime, is not well established and subject to large uncertainties [11]. Secondly, there is currently lack of quantitative understanding on heat conduction between soot aggregates and the surrounding gas, especially in the transition regime. This study concerns only the latter. An interesting phenomenon arising from the problem of fractal aggregate heat conduction is the so-called ‘shielding effect’. This effect is caused by the fact that some primary particles in the interior of an aggregate are partially or even entirely blocked by other primary particles from collision with gas molecules. Therefore, the shielding effect reduces the accessible surface area of the aggregate by gas molecules and consequently the conduction heat loss of the aggregate.

The problem of heat conduction between a fractal aggregate and the surrounding gas is formidable and does not admit analytical solutions even in the limiting free-molecular or the continuum regime, let alone the intermediate transition regime. Numerical methods based on the simulation of the physics of the flow have to be used to gain quantitative understanding of such problem. In particular, the direct simulation Monte Carlo (DSMC) method developed by Bird [12] has been demonstrated to be a powerful tool to study particle heat conduction problems in the free-molecular and transition regimes [13-17]. Heat conduction between a spherical particle and the surrounding monatomic gas in the transition regime has been investigated using DSMC by Filippov and Rosner [13] and Yang et al. [15]. Liu et al. [17] later extended the work of Yang et al. [15] to a biatomic gas.

So far there have been only two studies devoted to gain quantitative understanding of the heat conduction rate between soot aggregates and the surrounding gas using the DSMC method [14,16]. Both studies only investigated this problem in the free-molecular regime. The scaling laws obtained by these researchers to quantify the shielding effect are very similar and these results have been implemented into a low-fluence LII model [18]. These studies are relevant to the interpretation of LII measurements conducted in low pressure problems or in flames at atmospheric pressure where the Knudsen numbers are expected to be sufficiently large so that the free-molecular regime condition is likely to be met even for relatively large aggregates. However, it remains uncertain on how to quantify the shielding effect in the transition regime, which is highly relevant to LII experiments conducted at high pressures or low ambient temperature and atmospheric pressure conditions. It is noticed that high pressure LII experiments are frequently encountered in many combustion related applications, such as in diesel engines and gas turbine combustors. Currently it is not clear on which length scale of the aggregate, such as the radius of gyration or the projected area based equivalent sphere radius [9], should be used as the characteristic length scale as far as heat conduction is concerned. An attempt was made in this study to establish the characteristic heat transfer length scale of fractal aggregates based on the DSMC results.

In this paper, the DSMC method was used to calculate the heat transfer rate between a hot fractal aggregate and the cooler surrounding gas in the transition regime. The fractal aggregates of different size were numerically generated using a combined particle-cluster aggregation (for small aggregates) and cluster-cluster aggregation (for larger aggregates) algorithms. This study was an extension of our previous work [16]. DSMC calculations were conducted for soot aggregates containing up to 100 primary particles. Our objectives are to gain quantitative understanding of fractal aggregate heat conduction in the transition regime and to establish the characteristic heat transfer length scale of fractal aggregates.

II. Theory

A. The DSMC Method

The DSMC method was originally developed by Bird [12]. It was first applied to the homogeneous gas relaxation problem and the shock structure. With more than forty-year development, it has become a powerful numerical tool to study a variety of rarefied gas dynamics problems. A general description of the DSMC method was given by Bird

[12]. Although the DSMC method does not explicitly solve the Boltzmann equation, it incorporates all the physical concepts of rarefied gas and physical assumptions used in the phenomenological derivation of the Boltzmann equation.

Like most methods developed in computational fluid dynamics (CFD), the DSMC method also requires the computational domain to be divided into a grid of cells. The computational cells serve two purposes: to identify probable collision partners and to accumulate statistical information. The DSMC method is unsteady in nature. When applied to steady state problems, such as heat conduction from a fractal aggregate concerned in this study, the following procedure is followed. In the entire computational domain an arbitrary initial condition (velocity and position of molecules) of the simulated molecules is chosen randomly, corresponding to a Maxwellian equilibrium velocity distribution at the assumed initial gas temperature. Boundary conditions should also be provided at the particle surface and the outer computational boundary. With an adequately selected time step, velocity and position of molecules are stored and modified with time as the molecules undergo inter-molecular collisions and boundary interactions in the computational domain. This process is marched in time until a steady state is reached. Macroscopic quantities, such as temperatures (translational, internal, and overall) and heat fluxes, are accumulated for statistics after a prescribed time. In the DSMC method, the transport process of molecules is simulated by the motion and interaction of a large number of computational molecules, each of which represents a number of real molecules. The fundamental principle of the DSMC method is the splitting of continuous motion and collision of molecules at a time step into two sequential steps: ballistic molecule motion and inter-molecular collision. This approximation is valid only when a time step is shorter than the mean time between collisions. Another limitation to the time step is that it should be small enough so that the computational molecules do not cross more than one cell during one time step. The ballistic molecular motion is purely deterministic and is controlled by the velocity of the computational molecules. The inter-molecular collision, however, is stochastic with computational molecules colliding with other ones on a probabilistic basis based on their relative velocities. While the molecule size is required to calculate the collision rate, it does not affect the collision parameters. Collisions are binary and alter the velocities and the internal energies of the colliding molecules, but not their positions. The molecule/molecule collisions were considered by the well established simple hard sphere model [12], in which the cross-section is independent of the relative translational energy and the scattering is isotropic in the center of mass frame of reference. In this model, the energy of hard sphere molecules is connected only with their translation motion and their internal rotational and vibrational energy are neglected. Therefore, this model is applicable only to monatomic gas. The numerical approximations associated with the DSMC method include the ratio of the number of simulated molecules to the number of real molecules, the time step over which the molecular motion and collisions are uncoupled, and the finite cell and sub-cell sizes in the physical space.

To incorporate the effect of an arbitrary thermal accommodation coefficient of the particle, the molecule/particle collisions were simulated by using the Maxwell model at each collision: the molecule is either specularly reflected with a probability $(1-\alpha)$, or diffusively scattered with a probability α , where α is the thermal accommodation coefficient and is used to account for the energy accommodation of molecules on the particle surface. Numerically the Maxwell model is implemented as follows. Within a random portion of the time step dt used in the calculation, if a molecule collides with a primary soot particle, a random number R is generated. If R is greater than α , the molecule reflects specularly from the primary particle; otherwise, the molecule reflects diffusely. In the case of specular reflection, the velocity components of the molecule are conserved, i.e., no energy is transferred from the particle to the molecule. In the event of diffuse reflection, the normal and tangential components of the velocity of the reflected molecule are randomly sampled from the most probable molecular thermal speed at the particle temperature.

In DSMC method, the heat conduction between soot aggregates and the surrounding gas molecules was calculated based on the first principle, i.e., the collision of gas molecules with the primary particles in the aggregate and the energy transferred at each simulated molecule/particle collision was calculated from the difference between the translational energy of the molecule before and after collision. Therefore, it is crucial to correctly and fully account for the events of molecule/particle collision, especially for a fractal aggregate. As in previous study [14,16], the centers of the primary particles of the fractal aggregate and their radius are used in the DSMC code to identify the surface of the aggregate. The fractal aggregate was numerically generated using the approach described in the next section. An event of molecule/primary particle collision is considered to take place when the initial position of the molecule is outside the primary particle and the displacement of the molecule during a random portion of the time step dt causes it to intercept one of the primary particles in the aggregate. The procedure for ensuring that all the collisions between gas molecules and the fractal aggregate are correctly accounted for was detailed in our previous study [16].

B. Boundary Condition at the outer Computational Domain

The main challenge to extend our previous study [16] from the free-molecular regime to the transition regime lies in the treatment of the boundary condition at the boundaries of the computational domains. As in our previous study [16], the computational domain is a cube and the mass centre of the aggregate is located at the centre of the cube. While it is acceptable to assume that the temperature at all the surfaces of the cube is the same as the surrounding gas temperature when heat conduction is assumed to take place in the free-molecular regime [16], this assumption is no longer valid when the heat conduction between a fractal aggregate and the surrounding gas occurs in the transition regime, where the surface temperatures of the computational domain are higher than the gas temperature under the present conditions. Unlike the case of heat conduction of a single spherical particle in the transition regime where the outer boundary temperature can be updated unambiguously based on energy balance in spherical coordinates [15,17], the problem of fractal aggregate heat conduction encounters the uncertainty of how to update the temperatures of the domain boundaries due to lack of symmetry. This problem is schematically illustrated in Fig. 1. It is evident that the temperatures at the outer domain surfaces are in general non-uniform and there is lack of fundamental principle on how to make a reasonable assumption about such temperature distribution. Here we made an attempt to propose a simplified treatment to the boundary temperature at the outer computational boundaries.

Figure 1. Schematic of the cubic DSMC computational domain with a fractal aggregate placed at the centre.

Energy balance requires that the conduction heat loss rate at the surface of the aggregate must be equal to the heat loss rate of the domain surface to the surrounding gas. Throughout this work, the temperatures of the aggregate and the surrounding gas were assumed to be constant at T_p and T_g , respectively. If the computational cube is sufficiently large compared to the largest length of the aggregate, it can be assumed that the temperatures on the outer surfaces do not vary significantly and can be assumed uniform. To arrive at an approximate expression for the temperature at the outer surfaces, we first introduce an equivalent sphere for heat conduction between the solution domain (cube) and the surrounding gas. Here we chose the equivalent sphere whose centre is located at that of the cube and whose diameter satisfies $\pi D_{eq}^2 = 6L^2$ (L is the length of the cube). In other words, the equivalent sphere has the same surface area as the cubic computational domain. The cube should be chosen to be large enough to minimize the effect of the simplified outer boundary condition and to ensure that the heat conduction between the equivalent sphere and the surrounding gas is in the continuum regime, but should be as small as possible to reduce the computing time. Under these conditions the energy conservation of the system can be expressed as

$$4\pi R_{eq} \int_{T_g}^{T_b} k dT = Q_c \quad (1)$$

where $R_{eq} = 0.5D_{eq}$ is the radius of the equivalent sphere for heat conduction between the solution domain and the surrounding gas, k is the temperature dependent thermal conductivity of the surrounding gas, and Q_c is the total heat transfer rate at the aggregate surface, which is obtained from summing the molecule translational energy difference before and after each molecule/aggregate collision over all the collision events per unit time. In formulating Eq. (1) the heat conduction loss rate of a spherical particle in the continuum regime provided by Filippov and Rosner [13] was used. The thermal conductivity of the surrounding monatomic gas can be expressed as

$$k = k_g (T / T_g)^\omega \quad (2)$$

where subscript g denotes values of the surrounding gas and ω is the temperature-thermal conductivity exponent and is assigned a value of 0.5 for monatomic gas considered in this study. Substitution of Eq. (2) in to Eq. (1) leads to the outer boundary temperature

$$T_b = T_g \left(1 + \frac{(\omega+1)Q_c}{4\pi R_{eq} k_g T_g} \right)^{1/(\omega+1)} \quad (3)$$

The outer boundary temperature is updated at each time step according to Eq. (3). At the boundaries of the solution domain (six surfaces of the cube), molecules undergo diffuse reflection with their velocity components sampled from the most probable molecular thermal speed at the temperature determined from Eq. (3).

C. Numerical Generation of Fractal Aggregates

Experimental evidence collected from flame-generated soot indicates that within any given aggregate, the sizes of primary particle have a narrow distribution and can be approximately treated as identical in diameter [7,8]. Although there is a small degree of overlapping or necking between two neighboring primary particles, it is reasonable to assume that primary particles are in point-contact [8]. The fractal aggregates investigated were generated numerically using a combined particle-cluster and cluster-cluster aggregation algorithms described by Filipov et al. [14] and Liu and Snelling [19] assuming all primary particles are identical and are in point-contact. All fractal aggregates satisfy exactly the following scaling law [20]

$$N = k_f \left(\frac{R_g}{a} \right)^{D_f} \quad (4)$$

It is noticed that we used particle radius a , instead of the particle diameter d_p , as the length scale in Eq.(4), where N is the number of primary particles within the given aggregate, k_f and D_f are the prefactor and fractal dimension, respectively, and R_g is the radius of gyration defined as [14]

$$R_g^2 = \frac{1}{N} \sum_{i=1}^N (\mathbf{r}_i - \mathbf{r}^0)^2 + a^2 \quad (5)$$

$$\mathbf{r}^0 = \frac{1}{N} \sum_{i=1}^N \mathbf{r}_i \quad (6)$$

where vectors $\mathbf{r}_i$ and $\mathbf{r}^0$ define the position of the i th primary particle centre and the centre of the aggregate, respectively. In this study, fractal aggregates were generated using typical fractal parameters of $k_f = 2.3$ and $D_f = 1.78$ for flame-generated soot with a fixed value of the primary particle radius of $a = 15$ nm. Three fractal aggregates of sizes $N = 10, 20$, and 100 are shown in Fig. 2.

Figure 2. Three fractal aggregates of different sizes of $N = 10, 20$, and 100 .

III. Results and Discussion

The computer code used in the present calculations was modified from a three-dimensional DSMC code described by Bird [12], which was also used in [16]. In our calculations air was chosen as the surrounding gas. For simplicity, air was assumed to be a simple gas of identical ‘averaged air’ molecules with all microscopic quantities of air obtained according to [12]. The computational domain was a cube of $1 \mu\text{m} \times 1 \mu\text{m} \times 1 \mu\text{m}$ for all the calculations. The domain was divided into $80 \times 80 \times 80$ uniform volumes. Real gas molecules in the domain were represented computationally by about 2.5 million simulated molecules in all cases. The time step was chosen to be less than 5% (4% in the present study) of the average time between intermolecular collisions which is the reciprocal of the mean collision rate. The mean collision rate ν is obtained by $\nu = \pi d_g^2 n_g c_r$, where d_g is the average molecular diameter of the gas, c_r is the mean magnitude of the relative velocity of colliding molecules and is $2^{0.5}$ times the mean molecular speed c , which is equal to $[8k_B T_g / (\pi m_g)]^{0.5}$. All calculations started from an initial state of isothermal gas. The numbers of time steps to reach convergence are about 2000 to 5000 for calculations at relatively low pressures (1 to 5 atm) and many more are required at higher pressures, due to smaller time step and increasingly importance of molecule/molecule collision. The code was first validated in the calculation of heat conduction rate between a single primary soot particle and the surrounding gas by comparing the DSMC results with the

Table 1 Physical parameters used in the DSMC calculations.

Physical Parameters	Values
Thermal accommodation coefficient α	0.4
Mean molecular diameter d_g	4.15×10^{-10} m
Temperature-thermal conductivity exponent ω	0.5
Mean molecular mass m_g	4.81×10^{-26} kg

known analytical solution of energy exchange rate for an isolated, motionless sphere in the free-molecular regime and those of the Fuchs method in the transition regime. The code was then applied to heat conduction of numerically generated fractal soot aggregates. All the calculations were conducted for a particle (single or aggregate) temperature of $T_p = 3000$ K and the gas temperature of $T_g = 300$ K. To vary the mean free path of the surrounding gas, calculations were carried out at six gas pressures of $p_g = 1, 2, 5, 10, 20,$ and 30 atm. Other physical properties used in the calculations are summarized in Table 1.

A. Single Particle Heat Conduction

The purpose of conducting calculations of heat conduction of a single spherical particle in the transition regime is to validate the 3D DSMC code for this relatively simple problem. A similar validation of this code in the free-molecular regime was presented in [16]. Three recent studies of a single spherical particle heat conduction in the transition regime [13,15,17] showed that results of the Fuchs method [21] are in very good agreement with DSMC results calculated using 1D DSMC codes in the spherical coordinates system. Based on these findings, the present 3D DSMC results of the problem of a single particle heat conduction in the transition regime are compared with those of the Fuchs model. Details of the Fuchs can be found in [13,17,21] and are not given here.

Results of the 3D DSMC method are compared with those of the Fuchs method in Fig. 3, where the heat conduction rate is presented as the nondimensional Nusselt number Nu , which is defined as

$$Nu = \frac{Q_c}{2\pi a k_g (T_p - T_g)}, \quad (7)$$

as a function of the Knudsen number defined as the ratio of the mean free path λ to the particle radius a , i.e., λ/a . The mean free path λ is given as

$$\lambda = \frac{k_g}{f p_g} (\gamma - 1) \sqrt{\frac{\pi m_g T_g}{2 k_B}}, \quad (8)$$

where γ is the specific heat ratio ($5/3$ for monatomic gas), $f = (9\gamma - 5)/4$ is the Eucken factor. At $p_g = 1$ and 2 atm, which correspond to the two highest Kn numbers, both DSMC and Fuchs results are in good agreement with the analytical free-molecular solution, suggesting that the heat conduction occurs approximately in the free-molecular regime under these conditions. At higher pressures, however, the free-molecular solution starts to deviate from the DSMC and Fuchs results, implying that the free-molecular solution is no longer valid at pressures higher than 2 atm. Results of the DSMC and Fuchs methods are in good agreement up to $p_g = 10$ atm. At higher pressures, results of the DSMC method are somewhat higher than those of the Fuchs method. Although the DSMC method is based on the first principle and supposed to provide accurate heat conduction rates for this type of heat transfer problems, the Fuchs results are likely to be more reliable in this particular situation for the reasons given below. First, it becomes increasingly difficult to obtain accurate DSMC results as the pressure increases, especially in the present 3D calculations. This is because with increasing the pressure an increasingly smaller time step has to be used and a finer computational grid is required to resolve the larger temperature variation in the gas between the particle surface and the outer computational domain. Use of the finer grid makes the 3D DSMC calculation much more computationally demanding. It is possible that the grid resolution used in the present calculations may not be adequate at the two highest pressures considered. Secondly, previously studies [13,15,17] have demonstrated that results of the Fuchs method and the DSMC method implemented in the 1D spherical coordinates are in good agreement over the same range of Kn number. Nevertheless, the present 3D DSMC results of a single particle heat conduction problem in the transition regime are considered adequate at pressures less than about 10 atm.

Figure 3. Comparison of a single spherical particle heat conduction rates in the transition regime.

B. Fractal Aggregate Heat Conduction

It is expected that the total heat conduction rate increases with increasing the aggregate size due to larger available heat transfer area. To illustrate quantitatively the enhancement of heat conduction rate with the aggregate size, the nondimensional heat conduction rates are plotted as Nu against Kn in Fig. 4. Note that the Nusselt number Nu in Fig. 4 is calculated from its definition given in Eq. (7) using the primary particle radius a . Two observations can be made from Fig. 4. First, heat conduction rate increases with increasing the aggregate size, as expected. Secondly, the Nusselt number increases approximately linearly with decreasing the Knudsen for both single particle and fractal aggregates number under the conditions investigated here, which implies that the Nusselt number decreases exponentially with increasing the Knudsen number. It is interesting to notice that the slopes of the Nu - Kn curves of the four fractal aggregates are very similar; however, the curve of the single particle exhibits a larger slope.

Following Filippov et al. [14], the shielding effect can be quantified by the ratio of the total heat conduction rate of an aggregate Q_c^N to the product of the aggregate size N and heat conduction rate of the single primary particle Q_c^1 , i.e.,

$$\eta = \frac{Q_c^N}{NQ_c^1} \quad (9)$$

When there is no shielding effect, η is equal to 1. In general, η is less than unity as a result of the particle shielding. Variation of the shielding effect factor η with the primary particle Knudsen number for different aggregate sizes is shown in Fig. 5. It is evident that the shielding effect somewhat strongly depends on both the aggregate size N and the primary particle Knudsen number Kn . The occurrence of the plateau of the shielding effect at the highest pressure (smallest Knudsen number) considered is likely due to the unreliable 3D DSMC results, as shown in the single particle heat conduction case in Fig. 3. The dependence of the shielding effect on the aggregate size becomes increasingly weaker with increasing the aggregate size N , especially beyond $N = 50$, which is consistent with our previous findings based on a phenomenological model [9] and the 3D DSMC calculations in the free-molecular regime [16]. It is somewhat unexpected that the shielding effect is rather significantly enhanced with increasing the pressure (decreasing the Knudsen number), which is clearly associated with the larger slope of the single particle Nu - Kn curve shown in Fig. 4. Also plotted in Fig. 5 are the free-molecular solutions of the shielding effect factor η obtained in our previous study [16]. It is evident that the free-molecular assumption is not valid under the present conditions and this assumption results in significant underestimation of the shielding effect. Use of the heat transfer equivalent sphere size scaling based on our previous 3D DSMC calculations in the free-molecular regime [16] should be made with caution, especially in the interpretation of LII experiments conducted at elevated pressures.

Figure 4. 3D DSMC heat conduction rates of fractal aggregates of different sizes from 10 to 100.

Figure 5. Shielding effect as measured by the ratio of aggregate heat conduction rate to that of N isolated primary particles.

An issue we like to investigate is the effect of thermal accommodation coefficient. As pointed out earlier, all the DSMC calculations in this study were made for $\alpha = 0.4$, which is close to the recommended value of 0.37 for LII experiments in flames [9]. In the free-molecular regime, the thermal accommodation coefficient plays an important role in the particle heat conduction rate. In the continuum regime, however, it is expected that the thermal accommodation coefficient has no influence in the particle heat conduction process. A question of interest is that how important the thermal accommodation coefficient is under the conditions of this study. Due to the excessive computing time required by the 3D DSMC calculations, this question is addressed using the Fuchs method for single particle heat conduction and the results are shown in Fig. 6. It is clear that the heat conduction rate is significantly dependent on the thermal accommodation coefficient for the pressure range considered.

C. Heat Conduction Equivalent Sphere Size

One of the main interests of the 3D DSMC calculations of aggregate heat conduction in the transition regime is to arrive at the heat conduction equivalent sphere size which can be readily used in an aggregate based LII model [9,10]. To obtain the heat conduction equivalent sphere radius we used the Fuchs method inversely, i.e., we seek a spherical particle radius which yields the same total heat conduction rate as an aggregate of interest under the identical physical conditions. It must be emphasized that in doing so it has been implicitly assumed that the 3D DSMC results are sufficiently accurate for all the aggregate sizes and pressures studied. This assumption, however, was clearly not valid for the single particle case at pressures above 10 atm, let alone aggregates. Due to lack of other results of the aggregate heat conduction problems, the accuracy of the present 3D DSMC is uncertain and further study should be conducted to investigate the effects of grid resolution and the domain size on the results. Nevertheless, the heat conduction equivalent sphere radii are summarized in Table 2 as a first attempt to this formidable problem. For the smallest aggregate considered, $N = 10$, the heat conduction equivalent sphere radius remains almost constant with increasing the pressure up to 10 atm. At higher pressures, the equivalent sphere radius increases. For $N = 20$, the equivalent sphere size remains constant up to 5 atm, then starts to increase. For even larger aggregates, the equivalent sphere size increases rather significantly with increasing the pressure, especially for $N = 100$. Clearly, the results summarized in Table 2, which suggest the shielding effect is weakened with increasing the pressure, are in disagreement with those

Figure 6. Effect of α on the Nusselt number over the Knudsen number range considered.

Table 2. Heat conduction equivalent sphere radius (nm) determined by the Fuchs method and the 3D DSMC heat conduction rates of fractal aggregates.

p_g , atm	$N = 10$	$N = 20$	$N = 50$	$N = 100$
1	42.2	59.7	94.27	136.85
2	42.0	59.47	95.28	140.95
5	41.78	59.75	97.74	146.95
10	42.48	61.24	101.76	153.72
20	45.16	66.21	113.05	170.9
30	48.64	72.55	127.4	195.8

Figure 7. Comparison of heat conduction equivalent sphere radii obtained from DSMC and Fuchs methods at $p_g = 1$ atm with the radius of gyration and that from the projected area based equivalent sphere.

shown in Fig. 5, where the shielding effect is demonstrated to be enhanced with increasing the pressure (decreasing the Knudsen number). In other words, it is expected from Fig. 5 that the heat conduction equivalent sphere size should decrease with increasing the pressure. This disagreement is caused by the overestimation of the heat conduction equivalent sphere size using the DSMC aggregate heat conduction rates, which are likely to be inaccurate at higher pressures and larger aggregates.

Although the present 3D DSMC results are inaccurate at high pressures, it seems reasonable to assume that they are adequately accurate at relatively low pressures, e.g., $p_g = 1$ atm. Therefore, the heat conduction equivalent sphere radii at this pressure shown in Table 1 are compared with other characteristic lengths of the aggregates, such as the radius of gyration, Eq. (4), and the projected area based equivalent sphere radius. The radius of the projected area based equivalent sphere was calculated using the expression $R_A = (N/f_a)^{1/2ca}$ given in [9]. These characteristic lengths in terms of the radius of the various equivalent spheres are compared in Fig. 7. Based on the limited data points from the DSMC-Fuchs methodology, it is shown in Fig. 7 that the radius of the heat conduction equivalent sphere is in good agreement with the radius of gyration up to $N = 100$. In other words, the radius of gyration of a fractal aggregate can be used to estimate the regime of heat conduction. However, this observation cannot be generalized for the following reasons. First, there are very limited data points available. Secondly, these data were obtained for a specific value of the thermal accommodation coefficient of $\alpha = 0.4$. In view of the significant effect of α on the heat conduction rate shown in Fig. 6, it is expected that α has significant influence on the radius of the heat conduction equivalent sphere derived from the DSMC-Fuchs methodology. It has been shown previously that in the free-molecular regime the shielding effect, which affects the heat conduction equivalent sphere size, depends on the thermal accommodation coefficient [16]. The present results shown in Fig. 5 indicate that the shielding effect is also dependent on the primary particle Knudsen number. Therefore, the shielding effect in the transition regime is much more difficult to quantify than that in the free-molecular regime. Due to very limited and uncertain results of the present 3D DSMC calculations, it is not possible to arrive at some scaling relationships for the heat conduction equivalent sphere size as functions of the aggregate size, the thermal accommodation coefficient, and the primary particle Knudsen number.

IV. Conclusion

Heat conduction between hot nano-sized fractal aggregates and the surrounding cooler monatomic gas in the transition regime was studied for the first time using the direct simulation Monte Carlo method. Due to excessive computing time required by DSMC in such calculations, the present results at high pressures and larger aggregates are not accurate likely due to inadequate grid resolution. Nevertheless, some interesting findings were made. First, the shielding effect is found to be enhanced with decreasing the primary particle Knudsen number. Secondly, the heat conduction equivalent sphere radius is close to the radius of gyration of the aggregate under the conditions investigated. Further DSMC calculations with finer grid are required to confirm the present findings and to establish the heat conduction equivalent sphere size as functions of the aggregate size, the thermal accommodation coefficient, and the primary particle Knudsen number.

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