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Change in Particle Size Distribution of Fractal Agglomerates during Brownian Coagulation in the Free-Molecule Regime

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Time evolution of particle size distribution of fractal agglomerates undergoing Brownian coagulation in the free-molecule regime was investigated. A simple analytical solution for the size distribution change was obtained by using the assumption that the size distribution during the coagulation process can be represented by a time-dependent log-normal function. The derived solution consists of three parameters of the log-normal distribution function. This study is believed to provide the first analytical solution for all the parameters of the log-normal distribution of fractal agglomerates undergoing coagulation in the free-molecule regime. To validate the derived solution, numerical computations were performed. The results were compared with the analytical solution and good agreement was obtained.

INTRODUCTION

Aerosol agglomerates generated from industrial and commercial combustion processes are frequently observed as irregular fractal-like structures. The mass of the fractal agglomerates is conventionally related to their size by a power law exponent, which is called the mass fractal dimension, varying between 1 and 3 (1). Extensive results from computer simulations as well as experiments with colloidal and aerosol systems show that D_f is typically found in the range from 1.7 to 2.5, depending on the growth mechanism (2–7). The restructuring processes in which pairs of rigid clusters can readjust their positions with respect to each other after collision are often sufficient to increase the fractal dimensionality (8).

Aerosol agglomerates generated in flames often lie in the free-molecule regime because of both the small particle size and the increased mean free path length of the gas due to high temperature. Therefore, understanding the free-molecular coagulation of fractal agglomerates is important for prediction of their size distribution change. Previous work on Brownian coagulation of fractal agglomerates in the free-molecule regime includes Mountain *et al.* (9), Mulholland *et al.* (5), Kaplan and Gentry (10), Matsoukas and Friedlander (11), Wu and Friedlander (12), and Vemury and Pratsinis (13). Most of these studies, however, were based on numerical methods because of the complexity of the integro-differential coagulation equation. Matsoukas and Friedlander (11) presented an analytical solution for the growth rate of the mean particle volume and thus also the number concentration. Later, this solution was improved by Lehtinen *et al.* (14) to find the dependence of the growth rate on the fractal dimension. These solutions, however, were based on the self-preserving assumption. Thus, they could not provide the time evolution of particle size distributions before the self-preserving state was achieved.

The only analytical solution to the free-molecular coagulation problem for the entire coagulation time was given by Lee *et al.* (15) but it is applicable only for the spherical particles. Jain and Kodas (16) analytically investigated the Brownian coagulation of fractal agglomerates both in the continuum regime and in the free-molecule regime. However, they were successful only in deriving an analytical solution for the asymptotic geometric standard deviation value. They could not provide the time evolution of the entire particle size distribution.

Recently, we derived an analytical solution that resolves the size distribution of a coagulating agglomerates system in the continuum regime (17). Time-dependent size distributions of fractal agglomerates undergoing Brownian coagulation were assumed to preserve a log-normal function. As a result of this approach it was possible to examine quantitatively the effects of mass fractal dimension on the number concentration decay, on the increase of the mean particle size, and on the change in the standard deviation.

The purpose of this study is to extend the study of Lee *et al.* (15) and Park *et al.* (17) to develop an analytical solution which gives the size distribution change of coagulating agglomerates in the free-molecule regime for the entire coagulation time.

DERIVATION OF ANALYTICAL SOLUTION AND RESULTS

A complete size distribution of agglomerates undergoing coagulation is governed by the following nonlinear integro-differential equation if particle size is taken to be continuous

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(18):

$$\frac{\partial n(v,t)}{\partial t} = \frac{1}{2}\int_0^v \beta(v-\bar{v},\bar{v})n(v-\bar{v},t)n(\bar{v},t)\,\mathrm{d}\bar{v} - n(v,t)\int_0^\infty \beta(v,\bar{v})n(\bar{v},t)\,\mathrm{d}\bar{v}, \quad [1]$$

where $n(v, t)$ is the particle size distribution function at time t, and $\beta(v, \bar{v})$ is the collision kernel for two particles of volume v and $\bar{v}$. For the case of coagulation of fractal agglomerates due to Brownian motion in the free-molecule regime, the collision kernel $\beta(v, \bar{v})$ is given by (11)

$$\beta(v,\bar{v}) = K\left(v^{1/D_f} + \bar{v}^{1/D_f}\right)^2 \cdot \left(\frac{1}{v} + \frac{1}{\bar{v}}\right)^{1/2}, \quad [2]$$

where K is the collision coefficient $[=(3/4\pi)^{2/D_f-1/2} \times (6k_BT/\rho)^{1/2}r_p^{2-6/D_f}]$, D_f is the mass fractal dimension, k_B is the Boltzmann constant, T is the absolute temperature, ρ is the particle density, and r_p is the primary particle radius. The above collision kernel is one of the so-called homogeneous kernels. The homogeneous kernels have a property expressed as $\beta(av, a\bar{v}) = a^\gamma\beta(v, \bar{v})$, where γ is called the degree of homogeneity. Therefore, in Eq. [2], the degree of homogeneity is $2/D_f - \frac{1}{2}$ and dependent on the mass fractal dimension. It is assumed in this study that the entire growth process can be described by a constant mass fractal dimension. Many numerical simulations and experimental studies of coagulation processes have shown that fractal dimension does not vary significantly with cluster size (6, 11, 19).

Mulholland *et al.* (5) pointed out that for $D_f < 2$ Eq. [2] may overestimates the collision frequency when one particle is much larger than the other one, whereas for equal-sized particles the collision frequency can be predicted well by Eq. [2]. They suggested an alternative collision kernel for $D_f < 2$ as follows:

$$\beta(v,\bar{v}) = \bar{K}\left(v^{1/D_f} + \bar{v}^{1/D_f}\right)^{D_f} \cdot \left(\frac{1}{v} + \frac{1}{\bar{v}}\right)^{1/2}, \quad [3]$$

where $\bar{K} = K_{D_f=2}$. For equal-sized particles, Eq. [3] may underestimate the collision frequency, whereas it provides accurate predictions for highly unequal-sized particles. Thus, the correct collision kernel should lie between Eqs. [2] and [3] for $D_f < 2$. The discrepancy between Eqs. [2] and [3] increases with decreasing D_f. Because the fractal dimension is close to 2 in most cases, the error arising from use of Eq. [2] is expected to be small, particularly for narrow size distribution. Therefore, we use Eq. [2] for the free-molecular collision kernel.

In this study the size distribution of fractal agglomerates undergoing coagulation is assumed to be represented by a time-dependent log-normal function. Many experimental results and numerical calculations indicate that particle size distributions during coagulation fit approximately a log-normal function (10, 20–22). The log-normal size distribution function is written as

$$n(v,t) = \frac{1}{3v}\frac{N(t)}{\sqrt{2\pi}\ln\sigma(t)}\exp\left[\frac{-\ln^2\{v/v_g(t)\}}{18\ln^2\sigma(t)}\right], \quad [4]$$

where $N(t)$ is the total number concentration of particles, $\sigma(t)$ is the geometric standard deviation, and $v_g(t)$ is the geometric number mean particle volume. The kth moment of the particle size distribution is written as

$$M_k = \int_0^\infty v^k n(v,t)\,\mathrm{d}v, \quad [5]$$

where k is an arbitrary real number.

By multiplying Eq. [1] by v^k and integrating from 0 to ∞, we have

$$\frac{\mathrm{d}M_k}{\mathrm{d}t} = \frac{1}{2}\int_0^\infty\int_0^\infty \{(v+\bar{v})^k - v^k - \bar{v}^k\} \times \beta(v,\bar{v})n(v,t)n(\bar{v},t)\,\mathrm{d}v\,\mathrm{d}\bar{v}. \quad [6]$$

It is noticed that the collision kernel $\beta(v, \bar{v})$ appearing in Eq. [2] is in a form that appears to be rather difficult to expand into a series with a manageable number of terms. Thus, we approximate $(1/v + 1/\bar{v})^{1/2}$ with $b_k(1/v^{1/2} + 1/\bar{v}^{1/2})$ following the approach of Lee *et al.* (23), where b_k is to be calculated by

$$b_k = \frac{\int_0^\infty\int_0^\infty v^{k/2}\bar{v}^{k/2}\left(v^{1/D_f} + \bar{v}^{1/D_f}\right)^2(1/v + 1/\bar{v})^{1/2}n(v,t)n(\bar{v},t)\,\mathrm{d}v\,\mathrm{d}\bar{v}}{\int_0^\infty\int_0^\infty v^{k/2}\bar{v}^{k/2}\left(v^{1/D_f} + \bar{v}^{1/D_f}\right)^2\left(1/v^{1/2} + 1/\bar{v}^{1/2}\right)n(v,t)n(\bar{v},t)\,\mathrm{d}v\,\mathrm{d}\bar{v}} \quad [7]$$

for $k = 0$ and 2. Calculations with Eq. [7] indicated that the values for b_k depend on the initial geometric standard deviation σ_o and on the mass fractal dimension D_f. In addition, it was confirmed from the calculations that b_0 and b_2 are always identical for given σ_o and D_f. Thus, the results of Jain and Kodas (16), which reported variable b_2/b_0 values, were erroneous. Numerical computations were performed using Mathematica. Table 1 shows the computed results. The values for $D_f = 3$ in Table 1 exactly agree with the results of Lee and Hwang (24) which were obtained for spherical particles. As shown in Table 1, the value for b ranges from $1/\sqrt{2}$ for $\sigma_o = 1.0$ to 1 for $\sigma_o = \infty$. For simplicity, b will be assumed constant and equal to the listed initial values. This type of simplification is regarded as being

TABLE 1
Values of *b*, Obtained by Eq. [7]

σ_o	$D_f = 1.5$	$D_f = 2.0$	$D_f = 2.5$	$D_f = 3.0$
1.0	0.7071	0.7071	0.7071	0.7071
1.5	0.7923	0.7767	0.7699	0.7664
2.0	0.9473	0.9059	0.8784	0.8617
2.5	0.9934	0.9770	0.9558	0.9366
3.0	0.9993	0.9954	0.9870	0.9756
∞	1	1	1	1

suitable considering that the assumption regarding the form of the size distribution is more critical (23).

Substituting Eq. [2] into Eq. [6] and writing the results for $k = 0$ and 2, we have

$$\frac{dM_0}{dt} = -bK\left(M_{2/D_f-1/2}M_0 + 2M_{1/D_f}M_{1/D_f-1/2} + M_{2/D_f}M_{-1/2}\right), \quad [8]$$

$$\frac{dM_2}{dt} = 2bK\left(M_{2/D_f+1/2}M_1 + 2M_{1+1/D_f}M_{1/D_f+1/2} + M_{1+2/D_f}M_{1/2}\right). \quad [9]$$

For $k = 1$, Eq. [6] becomes

$$M_1 = \text{const}, \quad [10]$$

which merely means that total volume of particles is conserved.

A log-normal function has the following property among its moments:

$$M_k = M_1 v_g^{k-1} \exp\left\{\frac{9}{2}(k^2-1)\ln^2\sigma\right\}. \quad [11]$$

Differentiating Eq. [11] with respect to t for $k = 0$ and 2, and using Eq. [10], we have

$$\frac{dM_0}{dt} = -M_1 v_g^{-1}\exp\left(-\frac{9}{2}\ln^2\sigma\right)\left\{\frac{d(\ln v_g)}{dt} + \frac{9}{2}\frac{d(\ln^2\sigma)}{dt}\right\}, \quad [12]$$

$$\frac{dM_2}{dt} = M_1 v_g \exp\left(\frac{27}{2}\ln^2\sigma\right)\left\{\frac{d(\ln v_g)}{dt} + \frac{27}{2}\frac{d(\ln^2\sigma)}{dt}\right\}. \quad [13]$$

After substituting Eqs. [12] and [13] into Eqs. [8] and [9], and using Eq. [11], we have

$$\exp\left(\frac{27}{8}\ln^2\sigma\right)\left\{d(\ln v_g) + \frac{9}{2}d(\ln^2\sigma)\right\} = bK M_1 v_g^{\frac{4-3D_f}{2D_f}}\left[\exp\left\{9\left(\frac{2}{D_f^2} - \frac{1}{D_f}\right)\ln^2\sigma\right\} + 2\exp\left\{\frac{9}{2}\left(\frac{2}{D_f^2} - \frac{1}{D_f}\right)\ln^2\sigma\right\} + \exp\left(\frac{18}{D_f^2}\ln^2\sigma\right)\right]dt, \quad [14]$$

$$\exp\left(\frac{135}{8}\ln^2\sigma\right)\left\{d(\ln v_g) + \frac{27}{2}d(\ln^2\sigma)\right\} = 2bK M_1 v_g^{\frac{4-3D_f}{2D_f}}\left[\exp\left\{9\left(\frac{2}{D_f^2} + \frac{1}{D_f}\right)\ln^2\sigma\right\} + 2\exp\left\{\frac{9}{2}\left(\frac{2}{D_f^2} + \frac{3}{D_f}\right)\ln^2\sigma\right\} + \exp\left\{18\left(\frac{1}{D_f^2} + \frac{1}{D_f}\right)\ln^2\sigma\right\}\right]dt. \quad [15]$$

After eliminating dt, we have

$$d(\ln v_g) = \frac{\frac{27}{2}\exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma\right\} - 9}{2 - \exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma\right\}}\, d(\ln^2\sigma). \quad [16]$$

Integrating Eq. [16], we find the following relation between v_g and σ.

$$\frac{v_g}{v_{go}} = \exp\left\{\frac{9}{2}(\ln^2\sigma_o - \ln^2\sigma)\right\} \times \left[\frac{\exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma_o\right\} - 2}{\exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma\right\} - 2}\right]^{\frac{2D_f}{3D_f-4}}, \quad [17]$$

where v_{go} and σ_o are the initial values of v_g and σ, respectively. Substituting Eqs. [16] and [17] into Eq. [14] or [15], we have

$$\frac{9\exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma\right\}\left[2 - \exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma_o\right\}\right]d(\ln^2\sigma)}{v_{go}^{\frac{4-D_f}{2D_f}} H \exp\left\{\frac{9(4-D_f)}{4D_f}(\ln^2\sigma_o - \ln^2\sigma)\right\}\left[2 - \exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma\right\}\right]^2} = bK N_o\, dt, \quad [18]$$

where N_o is the initial value of $N[=M_0]$ and

$$H = \exp\left\{\frac{9}{2}\left(\frac{4}{D_f^2} - \frac{2}{D_f} + \frac{1}{4}\right)\ln^2\sigma\right\} + 2\exp\left\{\frac{9}{2}\left(\frac{2}{D_f^2} - \frac{1}{D_f} + \frac{1}{4}\right)\ln^2\sigma\right\} + \exp\left\{\frac{9}{2}\left(\frac{4}{D_f^2} + \frac{1}{4}\right)\ln^2\sigma\right\}.$$

One can note at this point that the other σ-containing quantities appearing in the denominator of the left-hand side of Eq. [18] would generally vary much less than $[2 - \exp\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma\}]^2$ for typical values of σ. Therefore, we approximate those quantities by setting $\sigma = \sigma_o$. This type approximation has proven to be quite reasonable (15, 17). As will be shown from the results, the fact that σ does not diverge but always converges to a certain value justifies this further. With this approximation, Eq. [18] is integrated to the following equation:

$$\ln^2\sigma = \frac{2D_f}{9(3D_f-4)}\ln\left[2 + \frac{\exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma_o\right\} - 2}{1 + \left(\frac{3D_f-4}{2D_f}\right)H_o bK\, v_{go}^{\frac{4-D_f}{2D_f}} N_o t}\right], \quad [19]$$

where H_o is the initial value of H. Substituting Eq. [19] into

Eq. [17], we further obtain v_g as a function of time:

$$\frac{v_g}{v_{go}} = \frac{\exp\left(\frac{9}{2}\ln^2\sigma_o\right)\left\{1+\left(\frac{3D_f-4}{2D_f}\right)H_o bK\, v_{go}^{\frac{4-D_f}{2D_f}} N_o t\right\}^{\frac{2D_f}{3D_f-4}}}{\left[2+\frac{\exp\left\{\frac{9(3D_f-4)}{2D_f}\ln^2\sigma_o\right\}-2}{1+\left(\frac{3D_f-4}{2D_f}\right)H_o bK\, v_{go}^{\frac{4-D_f}{2D_f}} N_o t}\right]^{\frac{D_f}{3D_f-4}}}. \quad [20]$$

Finally, we obtain the following equation for the particle number concentration decay by using Eqs. [11], [19], and [20]:

$$\frac{N}{N_o} = \frac{1}{\left\{1+\left(\frac{3D_f-4}{2D_f}\right)H_o bK\, v_{go}^{\frac{4-D_f}{2D_f}} N_o t\right\}^{\frac{2D_f}{3D_f-4}}}. \quad [21]$$

The entire particle size distribution for any time t can be constructed using Eq. [4] with the values of σ, v_g, and N given by Eqs. [19] through [21].

Figures 1 and 2 show changes in N and σ as functions of the dimensionless time $\tau[=\sqrt{6k_B Tr_p/\rho}(3/4\pi)^{1/6}N_o t]$ for different mass fractal dimensions and different σ_o values. It is shown in Fig. 1 that the number concentration decreases at a higher rate with a smaller D_f. This fact agrees well with the results of previous studies (10, 12, 13). Vemury and Pratsinis (1995) showed by numerical computations that the slope of the number decay curve is $-6/5$ for $D_f = 3$ and -2.0 for $D_f = 2$. In the present study, the slope proved to be $2D_f/(3D_f - 4)$ and it gives the same values as the results of Vemury and Pratsinis (13) for $D_f = 2$ and 3. Matsoukas and Friedlander (11) have derived a similar equation with Eq. [21] for self-preserving coagulation of fractal agglomerates. Especially, the exponent appearing in Eq. [21] is the same as that derived by Matsoukas and Friedlander (11). In addition, it is shown from Fig. 1 that the mass fractal dimension effects become more important as σ_o becomes larger. The same trend can be noticed also in Fig. 2. A more important

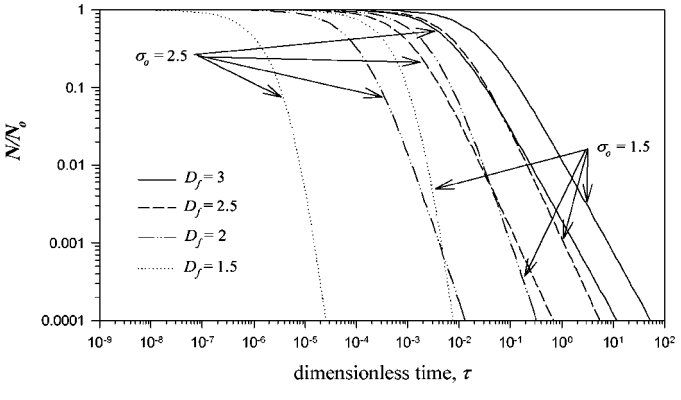

FIG. 1. Number concentration change as a function of σ_o and D_f.

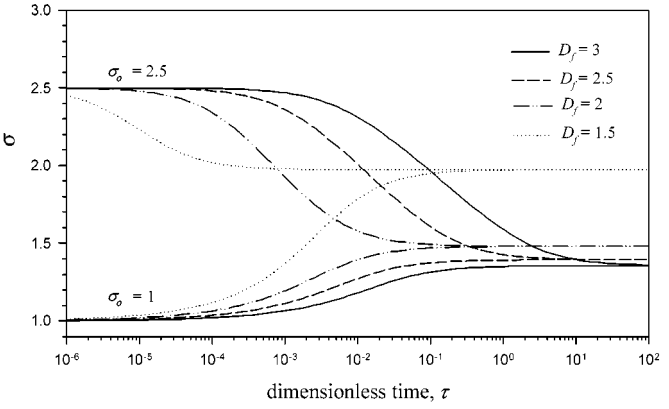

FIG. 2. Change in the geometric standard deviation as a function of σ_o and D_f.

fact which should be noted in Fig. 2 is that regardless of the initial value of σ_o, all $\sigma(t)$s appear to approach a certain value during the coagulation process. Such an approach seems to be accelerated with decreasing mass fractal dimension. Further discussion on this behavior of $\sigma(t)$ will be presented in the next section.

DISCUSSION

Because the solution was given in an analytical form, we have examined a couple of limiting cases with the solution.

1. *Spherical Particle Case* ($D_f = 3$): This can be regarded as a special case of the solution derived in this study. If D_f is set to 3, Eqs. [21], [19], and [20] reduce, respectively, to

$$\frac{N}{N_o} = \frac{1}{\left(1+\frac{5}{6}H_o bK\, v_{go}^{1/6} N_o t\right)^{6/5}}, \quad [22]$$

$$\ln^2\sigma = \frac{2}{15}\ln\left\{2+\frac{\exp\left(\frac{15}{2}\ln^2\sigma_o\right)-2}{1+\frac{5}{6}H_o bK\, v_{go}^{1/6} N_o t}\right\}, \quad [23]$$

$$\frac{v_g}{v_{go}} = \frac{\exp\left(\frac{9}{2}\ln^2\sigma_o\right)\left(1+\frac{5}{6}H_o bK\, v_{go}^{1/6} N_o t\right)^{6/5}}{\left\{2+\frac{\exp\left(\frac{15}{2}\ln^2\sigma_o\right)-2}{1+\frac{5}{6}H_o bK\, v_{go}^{1/6} N_o t}\right\}^{3/5}}, \quad [24]$$

where H_o becomes $\exp(\ln^2\sigma_o/8) + 2\exp(5\ln^2\sigma_o/8) + \exp(25\ln^2\sigma_o/8)$. Obviously, Eqs. [22] through (24) represent the solution for the spherical particles that was previously given by Lee *et al.* (1990).

2. *Asymptotic Behavior* ($t \to \infty$): Considering the behavior after a sufficiently long time, Eq. [19] reduces to

$$\sigma_\infty = \exp\left(\sqrt{\frac{2D_f}{9(3D_f-4)}\ln 2}\right), \quad [25]$$

where σ_∞ is the asymptotic value of σ for $t \to \infty$. This equation

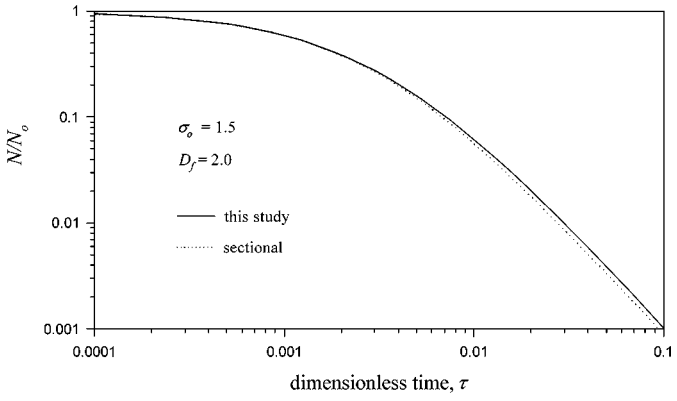

FIG. 3. Comparison with a numerical calculation for the number concentration decay.

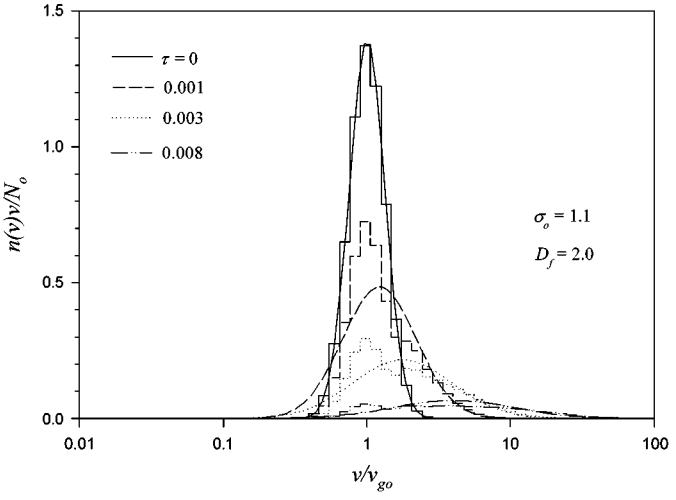

FIG. 5. Comparison with a numerical calculation for the particle size distribution change with $\sigma_o = 1.1$.

was previously presented by Jain and Kodas (16). It is noted from Eq. [25] that σ_∞ increases as D_f decreases in the free-molecule regime, whereas it does not depend upon D_f in the continuum regime (16, 17). This trend is in good agreement with previous numerical studies (12, 13). It is because in the free-molecule regime, the degree of homogeneity varies with D_f. Botet and Jullien (25) have pointed out that the asymptotic shape of the size distribution depends only on the degree of homogeneity of the collision kernel. In a recent study (26), we derived a general solution for σ_∞ as a function of the degree of homogeneity using an arbitrary homogeneous collision kernel. In that paper, Eq. [25] was shown to be a special case of the general solution.

To validate the analytical solution derived in this study, we performed numerical simulations and compared the results with the analytical solution. Figures 3 and 4 show the comparison results using an aerosol with $\sigma_o = 1.5$ and $D_f = 2.0$. In the numerical simulations the sectional method suggested by Landgrebe and Pratsinis (27), which did not assume any functional form for a particle size distribution, was used. For the section spacing factor 1.18 was used. It is seen in Figs. 3 and 4 that the analytical solution obtained in this study is in good agreement with the numerical simulation results. For a comparison for initially narrow size distribution, Fig. 5 shows the change in particle size distribution with $\sigma_o = 1.1$. One can note from the result of sectional method that a bimodal distribution occurs in the early stage of coagulation. The analytical solution cannot realize this phenomenon because of the log-normal assumption. As the time elapses, however, the smaller mode disappeared due to coagulation with larger particles and the difference between the results from the sectional and the analytical methods decreased.

For a further investigation of the self-preserving feature of coagulating agglomerates, the analytical solution has been compared with the previous numerical model of Vemury and

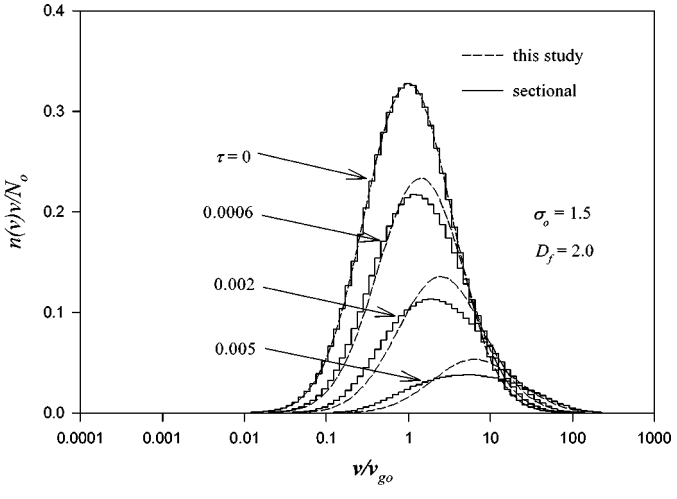

FIG. 4. Comparison with a numerical calculation for the particle size distribution change with $\sigma_o = 1.5$.

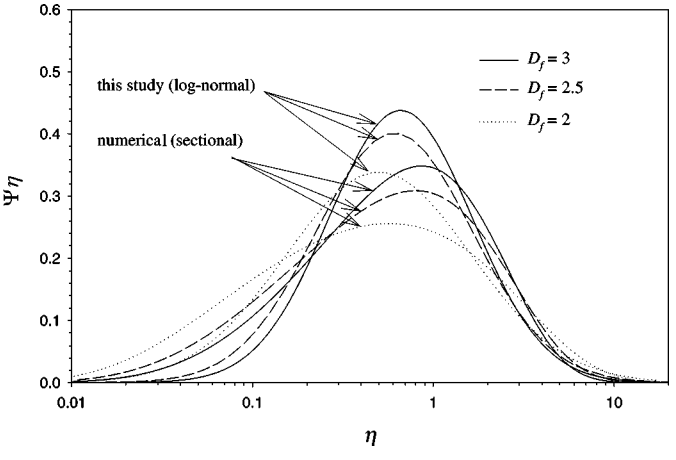

FIG. 6. Comparison with a numerical calculation for the self-preserving particle size distribution.

TABLE 2
Values of t_∞ for Various σ_o and D_f

σ_o	$D_f = 1.5$	$D_f = 2.0$	$D_f = 2.5$	$D_f = 3.0$
1.0	0.058	0.061	0.14	0.37
2.5	0.00001	0.056	2.1	30

Pratsinis (13) in which the sectional code was used to obtain the self-preserving size distribution. Conventionally, in the self-preserving formulation, the dimensionless particle volume and the dimensionless size distribution density function are defined as Eqs. [26] and [27], respectively:

$$\eta = v/\tilde{v}, \quad [26]$$

$$\Psi = n\tilde{v}/N, \quad [27]$$

where $\tilde{v}[=M_1/N]$ is the arithmetic mean particle volume.

Figure 6 shows the comparison result. It is noted in this figure that although the analytical solution and the numerical results show similar changes as the mass fractal dimension decreases, the analytical model of this study always gives a narrower distribution than the numerical results. The difference between the analytical and the numerical models results from the symmetrical assumption of the log-normal size distribution (28). Numerically predicted asymptotic σ values for $D_f = 3.0$, 2.5, and 2.0 were 1.462, 1.519, and 1.610, respectively, while the analytical solution obtained in this study gives the values of 1.355, 1.393, and 1.481, respectively.

Another thing of interest for many applications is the time it takes for a distribution to reach the asymptotic state. To derive an expression for this time, let us assume σ has reached σ_∞ if $1.9 < \exp[\{9(3D_f - 4)/(2D_f)\} \ln^2 \sigma] < 2.1$ compared with Eq. [25]. With this criterion, the following expression is derived from Eq. [19] for the required time to approach the asymptotic distribution, t_∞,

$$t_\infty = \frac{\frac{2D_f}{bH_o(3D_f-4)}\left[\pm 10\left\{\exp\left[\frac{9(3D_f-4)}{2D_f}\ln^2\sigma_o\right]-2\right\}-1\right]}{K v_{go}^{(4-D_f)/(2D_f)} N_o}, \quad [28]$$

where the sign convention may be used for $\sigma_o > \sigma_\infty$ and $\sigma_o < \sigma_\infty$ in that order. Table 2 shows the time required to reach such an asymptotic state for various σ_os and D_fs according to Eq. [28]. It is noted in this table that t_∞ decreases with decreasing D_f. This fact agrees with the result of the previous numerical study by Vemury and Pratsinis (13). The trend can also be found in Fig. 2.

CONCLUSIONS

An analytical solution to the Brownian coagulation of fractal agglomerates in the free-molecule regime was derived. The size distribution of coagulating agglomerates was obtained as a function of time. It was assumed that the particle size distribution can be represented by a time-dependent log-normal function during the coagulation process. The effect of the mass fractal dimension on the particle size distribution change was quantitatively analyzed from the results. It is believed that this study represents the first analytical solution to the Brownian coagulation of fractal agglomerates in the free-molecule regime for the entire coagulation time.

The sectional code of Landgrebe and Pratsinis (27) was used to numerically calculate the change in particle size distribution and to compare the results with the analytical solution derived in this study. The comparisons showed good agreement between the two models.

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