Fractal dynamics in orthokinetic acoustic agglomeration processes

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We investigate the time evolution of an aggregate system undergoing orthokinetic acoustic agglomeration. The kernel function characterizing the dynamics of the process includes the fractal structure of the aggregates. We demonstrate that to leading order in average aggregate multiplicity $\langle k \rangle$, which is essentially the aggregate mass, the dynamic equation exhibits scaling properties with the scale function being proportional to $\langle k \rangle$. Numerical simulation carried out for fractal dimension $D>2$ confirms the approach of the asymptotic scaling solution. We demonstrate that starting from two very different initial distributions is the same final scaling distribution. This is in spite of the fact that the rates of their approach are sensitive to the initial distributions. [S1063-651X(96)03609-4]

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I. INTRODUCTION

Aerosols generated from combustion of fossil fuels and from processes used to produce industrial chemicals are typically in the submicrometer and micrometer size range. They are harmful to the human respiratory system and are significant contributors to air pollution [1]. The application of intense sonic fields to aerosol suspensions promotes agglomeration that systematically shifts the aerosols to larger sizes. The larger sized aggregates are less harmful and easier to remove. Reference [2] gives several references which will help the reader to trace the past important experimental and theoretical effort in acoustic agglomeration.

Acoustic agglomeration dynamics are based on the same theoretical framework used for more familiar coagulation processes. Consider the time evolution of an aggregate system and denote the number density of aggregates of multiplicity k (meaning there are k primary aggregate per aggregate with $k=1,2,\ldots$) by c_k. The Smoluchowski equation, given by

$$\frac{dc_k}{dt}=\frac{1}{2}\sum_{i,j} K_{ij}c_i c_j \delta_{k,i+j}-c_k\sum_{j=1}^{\infty} K_{kj}c_j, \qquad (1.1)$$

governs the acoustic agglomeration process. Here K is the kernel function that characterizes the dynamics of the process. In Ref. [2], Mednikov reviewed a range of possible mechanisms occurring in acoustic-aerosol interactions. He proposed that the orthokinetic interactions, caused by collisions due to acoustic-induced relative motions among aggregates of different sizes, are the dominant mechanism. Tiwary and Reethof showed that this mechanism is in qualitative agreement with the data [3]. A work by Song, Koopmann, and Hoffmann [4] discussed further refinements in the treatment of orthokinetic interactions, as well as the addition of the hydrodynamic interactions. Apparently the inclusion of both mechanisms gives better agreement with the data.

There are a number of theoretical works found in the literature that use various simple kernel functions to deduce ramifications in coagulation processes (see, for example, Ref. [5] and references cited therein). There are also works in the literature that concern the fractal structure of aggregates in the context of Brownian coagulation (see, for examples, [6–8] and references cited therein). Due to the relative complexity of the dynamics, research in the field of acoustic agglomeration remained restricted to numerical simulations. In this paper a partial analytic solution to the Smoluchowski equation for acoustic agglomeration is developed and then verified by numerical simulation.

II. ORTHOKINETIC KERNEL WITH FRACTAL STRUCTURE

This derivation neglects all interactions between aggregates besides collisions and concentrates on the motion of the aggregates induced by its relative velocity with the surrounding media. If the incident sound wave has a wavelength much greater than the aggregate fractal radius r_D and its angular frequency ω satisfies the condition $r_D^2\omega \ll \mu$, the hydrodynamic equations characterizing the interaction of the aggregate with the media reduce to the form of the equations in Ref. [6]. Here μ is the coefficient of viscosity of the media.

The analysis in Ref. [6] determines that the main force on the aggregate is the Stokes drag force with the normal radius of a solid sphere replaced by the fractal radius of the aggregate. We treat the agglomerate as spherically symmetric, which corresponds to an averaging over all possible aggregate orientations. We define the fractal radius r_D by its relation to the multiplicity

$$k=\left(\frac{r_D}{l}\right)^D \quad \text{or} \quad r_D=k^{1/D}l. \qquad (2.1)$$

Here l is a length scale that makes the ratio r_D/l dimensionless. It is plausible that l should be of the order of the radius of the primary particle and in this work $l=a$, where a is the radius of the primary particle. The relaxation time is defined by

$$\tau=\frac{m}{6\pi\mu r_D}=\frac{k^{1-1/D}m_p}{6\pi\mu a}, \qquad (2.2)$$

where the aggregate and the primary particle masses are m and m_p, respectively, and $m=km_p$. The resulting equation of motion of the aggregate particle due to the Stokes drag force is $dv/dt=-v_{\rm rel}/\tau$. Here $v_{\rm rel}$ denotes the relative velocity between the aggregate and the media. For an acoustic wave, with angular frequency ω and maximum velocity u_0, traveling along the z direction, $v_{\rm rel}=v-u_0\cos(\omega t)$. Solving the dynamical equation gives an aggregate velocity

$$v=\eta\cos(\omega t-\phi)u_0$$
$$\text{with}\quad \eta=\frac{1}{\sqrt{1+p^4}},\quad \tan\phi=p^2,\quad p=\sqrt{\tau\omega}. \tag{2.3}$$

Now consider the relative motion of a pair of aggregates labeled by 1 and 2. Their relative velocity is along the z axis and is given by $v_{\rm rel}=v_2-v_1=u_0\eta_{12}\cos(\omega t-\alpha)$. Here the quantity η_{12} represents the relative entrainment between the aggregate pair, with $\eta_{12}=|p^2-q^2[(1+p^4)(1+q^4)]^{-1/2}$, where $p^2=\tau_1\omega$, $q^2=\tau_2\omega$, and α is a constant.

The collision volume is the volume surrounding the first aggregate, which the second aggregate must enter for the collision to occur. The rate of creation of collision volume for two aggregates is the kernel characterizing the collision process. Because the relative motion between the two aggregates is along the z axis, the rate of creation of collision volume can be calculated by multiplying the collision cross section, given in this case by $\pi(r_1+r_2)^2$, with the average relative distance one of the aggregates travels with respect to the other in a unit time. This is carried out by integrating the absolute value of the relative velocity over one acoustic period and then dividing by the period. The kernel becomes

$$K_{12}=2(r_1+r_2)^2\eta_{12}u_0. \tag{2.4}$$

Equation (1.1), together with this kernel, specifies the orthokinetic dynamics for the present analysis. Here the two radii are the fractal radii of the aggregates.

III. ASYMPTOTIC DYNAMICAL EQUATION

We found that despite the inhomogeneous form of the kernel, when either one or both of the colliding aggregates are small, an asymptotic scaling solution exists. We follow conventional methods [1,5] and introduce a scaling function s and a corresponding scaling variable $x=k/s$. We also make the standard assumption about the form of the scaling solution, i.e., it is of the form

$$c_k=\frac{N_p}{s^2}\,\rho\left(\frac{k}{s}\right)\to\frac{N_p}{s^2}\,\rho(x)\quad\text{with}\quad N_p=\sum_{k=1}^{\infty}kc_k, \tag{3.1}$$

where $\rho(x)$ is referred to as the reduced concentration and satisfies $\int_0^\infty dx\ x\ \rho(x)=1$.

We introduce two auxiliary quantities ζ_s and κ through the relations $p^2=\tau\omega\equiv k^{1-1/D}/\kappa=\zeta_s x^{1-1/D}$ with $\zeta_s\equiv(1/\kappa)s^{1-1/D}$ and $\kappa=6\pi\mu a/m_p\omega$. We also define an s-dependent reference point x_1 such that for large s,

$$\tau\omega=\zeta_s x_1^{1-1/D}\sim s^{\delta}\gg 1,\quad x_1\sim s^{[(\delta-1+1/D)/(1-1/D)]}\to 0, \tag{3.2}$$

where $0<\delta<1-1/D$ and D must satisfy $D>1$. We now divide the x integration into two regions: I, $0<x<x_1$ and II, $x>x_1$. As s goes to ∞, the contribution from region I becomes negligible. To evaluate the kernel of Eq. (2.4) in region II, we denote $p^2=\zeta_s y^{1-1/D}$, with $y=i/s$, and $q^2=\zeta_s z^{1-1/D}$, with $z=j/s$. This leads to

$$K_{ij}=\frac{2u_0a^2s^{2/D}}{\zeta_s}\,F(y,z)$$
$$\text{with}\quad F(y,z)=\frac{(y^{1/D}+z^{1/D})^2|y^{1-1/D}-z^{1-1/D}|}{(yz)^{1-1/D}}. \tag{3.3}$$

Thus to leading order in s, the kernel function reduces to the standard form of Ref. [5]. Using Eqs. (3.1) and (3.3) we rewrite Eq. (1.1) as

$$-\frac{W}{x}\frac{d}{dx}[x^2\rho(x)]=\lim_{\varepsilon\to 0}H(x,\varepsilon), \tag{3.4}$$

with

$$H(x,\varepsilon)=\tfrac{1}{2}\int_{\varepsilon x}^{(1-\varepsilon)x}dy\ F(y,x-y)\rho(y)\rho(x-y)$$
$$-\rho(x)\int_{\varepsilon x}^{\infty}dy\ F(x,y)\rho(y).$$

Here

$$W=\frac{s^{1-3/D}\dot{s}}{C}$$
$$\text{with}\quad C=2u_0a^2\kappa N_p=12\pi u_0a^3\,\frac{\mu N_p}{\omega m_p},\quad \varepsilon\sim s^{-\delta}. \tag{3.5}$$

Notice that W is a function of s and ds/dt. *A priori*, one might think that W should depend on time. However, to be consistent with Eq. (3.4), W must be a constant. This allows the Smoluchowski equation to be decoupled into two sets of equations. To leading order in s, Eq. (3.4) constitutes the scaling equation for $\rho(x)$, while Eq. (3.5) specifies how the scale function s evolves in time.

With W being constant from Eq. (3.5), we continue to solve for s. For $1.5<D\leqslant 3$,

$$s=s_0\left(1+\frac{t}{\gamma t_1}\right)^{\gamma}, \tag{3.6}$$

with $t_1=s_0^{1/\gamma}/WC$ and $\gamma=D/(2D-3)$. Denote the ''scaling solution extrapolation'' of the average number multiplicity at $t=0$ as $\langle k_0\rangle$. From (3.6) it follows that

$$\langle k\rangle=\langle k_0\rangle\left(1+\frac{t}{\gamma t_1}\right)^{\gamma}. \tag{3.7}$$

Notice that $\langle k\rangle$ is essentially the average mass, since $k=m/m_p$. For $D\to D_c=3/2$, $\gamma\to\infty$,

$$\langle k\rangle=\langle k_0\rangle\exp(t/t_1)=\langle k_0\rangle\exp(CWt), \tag{3.8}$$

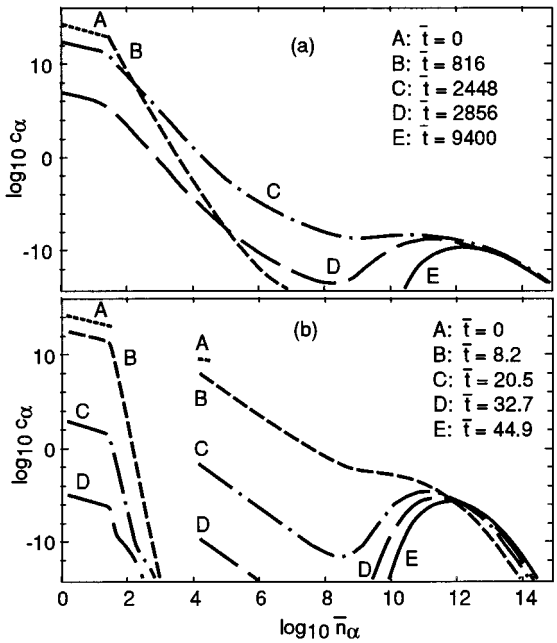

FIG. 1. Time evolution of the size distribution with $D=2.5$ for the (a) the mono-initial-interval and (b) the di-initial-interval distributions, at various reduced time defined as $\bar{t}=t/t_{\text{scale}}$, where $t_{\text{scale}}=1/(2u_0a^2N_0)$.

and for $D<D_c$, $\gamma<0$,

$$\langle k\rangle=\langle k_0\rangle\left(1-\frac{t}{t_c}\right)^{-|\gamma|}, \quad t_c=|\gamma|t_1. \tag{3.9}$$

Equations (3.7)–(3.9) are reminiscent of the nongelling, critical, and gelling cases discussed in the literature [5,9,10]. Following the approach of Ref. [5], the analytic solutions for the present case for $D>D_c$ in the small x and the large x regions are, respectively,

$$\rho\sim e^{-1/x^{1-1/D}}, \quad \rho\sim x^{1-3/D}e^{-cx}, \tag{3.10}$$

where c is a parameter.

IV. DISCRETE-SECTIONAL MODEL AND NUMERICAL RESULTS

We used a numerical model to calculate the time evolution of the particle size distribution of an agglomerating system when driven by an acoustic source. The basic program was developed by Landgrebe and Pratsinis [11] and is based on a modified version of the discrete-sectional model of Wu and Flagan [12], evolved from the sectional model of Gelbard, Tambour, and Seinfeld [13]. In this program, the discrete-size spectra of the Smoluchowski equation are grouped into ''bins'' labeled by the index α. The maximum numbers of the discrete points and of the continuum sectors are M_d and M_s, respectively. For $\alpha\leqslant M_d$, the bins contain aggregates of a single multiplicity given by $n_\alpha=\alpha$ and for $\alpha>M_d$ the bins contain aggregates with the range of multiplicity $n_{\alpha-1}\leqslant i\leqslant n_\alpha$ for all $i\in\alpha$. In the sectional region, the multiplicity n_α is defined through the iterative relation

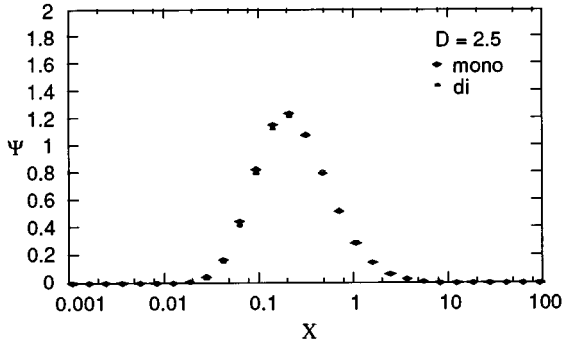

FIG. 2. Plot of Ψ vs X for distributions in the scaling region for the initial mono- and di-distributions with $D=2.5$. The reduced times are, respectively, $\bar{t}=94\,000$ for the mono-distribution and $\bar{t}=449$ for the di-distribution.

$n_\alpha=bn_{\alpha-1}$, where b is some parameter greater than 1 and $n_{Md+1}=M_d+0.5$.

Numerical calculations [14] determine the time evolution of the acoustic agglomeration process for two different initial distributions. Figure 1 shows plots of $\log_{10}c_\alpha$ vs $\log_{10}\bar{n}_\alpha$, with the fractal dimension $D=2.5$. Here c_α is the concentration of aggregates in the αth bin. The curves correspond to various reduced times indicated in the caption. Figure 1(a) corresponds to the case with an initial state populated by one interval of small size aggregates, referred to as the mono-initial-interval case. This system evolves slowly at first, building up larger sized aggregates through agglomeration between approximately the same sized aggregates. When enough large sized aggregates form such that the interaction between large and small particle sizes becomes appreciable, the agglomeration rate increases and the system begins forming an asymptotic distribution. Figure 1(b) shows the time evolution of the split-size initial distribution, or the di-initial-interval case. The interaction between the large sized aggregates and the smaller aggregates becomes the dominant interaction from the beginning. For small times, the agglomeration process continues at a much faster rate. This is a direct consequence of the dependence of the orthokinetic agglomeration kernel on the difference of aggregate mass. While both distributions undergo very different evolution for small times, *both develop into a similar distribution as the agglomeration process reaches the asymptotic scaling region*. As implied above, the only difference in the asymptotic distributions for each initial condition lies in the time interval needed to reach the asymptotic regime. The idea of introducing large-size catalytic aerosol has been explored quantitatively both theoretically, which is based on the orthokinetic and hydrodynamic mechanics, and experimentally [15]. Our results agree with the conclusion that the catalytic aerosols speed up the agglomeration process. Our main interest here is to demonstrate that beginning from two very different initial distributions is the same asymptotic scaling distribution. We now proceed to discuss the scaling property.

The scaling behavior or self-preserving behavior is usually displayed by the plot [1] of the dimensionless density function Ψ vs the dimensionless size variable X. Applied to the present case, with V_p being the hard-sphere volume of the primary particle,

$$X=\frac{N}{N_p}\,n_\alpha=\frac{n_\alpha}{\langle n_\alpha\rangle}\xrightarrow[\text{scaling limit}]{} x, \tag{4.1}$$

$$\Psi=\frac{V}{N^2}\,c_\alpha=\frac{\langle n_\alpha\rangle^2}{N_p}\,V_p c_\alpha\xrightarrow[\text{scaling limit}]{} V_p\rho(x). \tag{4.2}$$

Figure 2 is a plot of Ψ vs X for distributions in the scaling region for the mono-initial-interval case and the di-initial-interval case with $D=2.5$. These plots correspond to data taken at reduced times ten times longer than the reduced times of curves E in Fig. 1. This illustrates the approach to one common scaling distribution for a given fractal dimension. It turns out that as the fractal dimension decreases the width of the scaling distribution increases. Also our numerical results and the predictions of Eq. (3.10) in the large and small X regions are in reasonable agreement.

To conclude, we have presented theoretical arguments to demonstrate both analytically and numerically the approach of the asymptotic scaling behavior in orthokinetic acoustic agglomeration process. With the inclusion of the fractal structure there is the critical dimension $D=\frac{3}{2}$, below which the average aggregate multiplicity will become infinite for some finite time.

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[1] S. K. Freilander, *Smoke, Dust and Haze* (Wiley, New York, 1977).

[2] For earlier work, see E. P. Mednikov, *Acoustic Coagulation and Precipitation of Aerosols* (USSR Academy Science of Moscow, Moscow, 1963). For recent work, see T. L. Hoffmann and G. H. Koopmann, J. Acoust. Soc. Am **99**, 2130 (1996); see also J. A. Gallego, E. Riera, G. Rodriguez, T. L. Hoffmann, J. C. Galvez, L. Elvira, F. Vazquez, F. Montoya, J. J. Rodriguez, F. J. Gomez, and M. Martin (unpublished).

[3] R. Tiwary and G. Reethof, J. Vib. Acoust. Stress, Rel. Design **109**, 185 (1987).

[4] Limin Song, Ph.D. thesis, Pennsylvania State University, 1990 (unpublished); see also L. Song, G. H. Koopmann, and T. L. Hoffmann, J. Vib. Acoust. **116**, 208 (1994).

[5] P. G. J. Van Dongen and M. H. Ernst, Phys. Rev. Lett. **54**, 1369 (1985).

[6] R. D. Mountain, G. W. Mulholland, and H. Baum, J. Colloid. Interface **114**, 67 (1986).

[7] T. Matsoukas and S. K. Friedlander, J. Colloid. Interface **146**, 495 (1991).

[8] S. Vemury and S. E. Pratsini, J. Aerosol Sci. Am. **26**, 175 (1995).

[9] F. Leyvraz and H. R. Tschudi, J. Phys. A **15**, 1951 (1982).

[10] E. M. Hendriks, M. H. Ernst, and R. M. Ziff, J. Stat. Phys. **31**, 519 (1983).

[11] J. D. Landgrebe and S. E. Pratsinis, J. Colloid. Interface **139**, 63 (1990).

[12] J. J. Wu and R. C. Flagan, J. Colloid. Interface Sci. **123**, 339 (1988).

[13] F. Gelbard, Y. Tambour, and J. H. Seinfeld, J. Colloid. Interface **76**, 541 (1980).

[14] The following set of parameter values was used for our numerical calculation. The radius, volume, and density of the primary aggregate are taken to be $a=10^{-8}$ m, $V_p=4.19\times10^{-24}$ m^3, and $\rho_p=1200$ kg/m^3, respectively. The thermodynamic properties of the fluid (air) media are temperature $T=300$ K, pressure $P=10^5$ Pa, and viscosity $\mu=1.81\times10^{-4}$ g/cm s. The frequency of the incident acoustic wave is 1 kHz and the sound pressure level is 154 dB, giving $u_0=(1/P)\sqrt{T/\gamma R}\,10^{(\mathrm{SPL}-94)/20}=5$ cm/s, where $\gamma=7/5$ and $R=8.31$ J/(K mol) were used. Following the approach of Ref. [11], we ''overload'' the system in order to arrive at a reasonable rate for the numerical calculation. We took $N_0=8\times10^{20}$ aggregates/m^3, which leads to $t_{\mathrm{scale}}=0.35$ ms. The asymptotic scaling region is reached for small-size case at about 0.1 s and for the split-size case at about 1 ms. In this work the default values are $M_d=20$, $M_s=100$, $b=1.5$, and $\zeta=2$, where ζ is a moment parameter; see Ref. [11] for details.

[15] T. L. Hoffmann, W. Chen, G. H. Koopmann, A. W. Scaroni, and L. Song, J. Vibration Acoust. **115**, 233 (1993).