Condensation-driven aggregation in one dimension

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We propose a model for aggregation where particles are continuously growing by heterogeneous condensation in one dimension, and solve it exactly. We show that the particle size spectra exhibit a transition to dynamic scaling $c(x,t) \sim t^{-\beta} \phi(x/t^z)$. The exponents β and z satisfy a generalized scaling relation $\beta = (1+q)z$ where the value of q is fixed by a nontrivial conservation law. We show that the value of 1+q is always less than the value 2 for aggregation without condensation.

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I. INTRODUCTION

The formation of clusters by aggregation of particles is a characteristic feature of many seemingly different processes in physics, chemistry, biology, and engineering. Examples include aggregation of colloidal or aerosol particles suspended in liquid or gas [1-3], polymerization [4], antigenantibody aggregation [5], and cluster formation in galaxies [6]. This wide variety of applications has resulted in numerous studies which reveal that, when chemically identical particles aggregate, almost always scale-invariant clusters emerge. Note that due to the nonequilibrium nature of the aggregation process the standard theoretical framework developed for equilibrium statistical physics is found redundant. However, the application of stochastic theory is found to be increasingly useful in capturing a wide class of non-equilibrium phenomena.

Typically, nonequilibrium systems are described by the rate equation approach, having the form of a master equation which is often governed by some conservation principle. The Smoluchowski equation for the kinetics of irreversible aggregation is one such example, where the distribution function c(x,t) of a particle of size x at time t evolves according to the following integro-differential equation [7,8]:

$$\frac{\partial c(x,t)}{\partial t} = -c(x,t) \int_0^\infty K(x,y)c(y,t)dy + \frac{1}{2} \int_0^x dy \ K(y,x-y)c(y,t)c(x-y,t).$$
(1)

Here, the kernel K(x,y) is symmetric with respect to its argument and it determines the collision time in which a particle of size *x* collides with another particle of any size *y* and they merge into a particle (aggregate) of size (x+y). The first term on the right-hand side of Eq. (1) describes the loss of a particle of size *x* due to merging of particles of size *x* with other particles of any size, while the second term describes the gain of *x* due to merging of particles of size x-y with *y*. The Smoluchowski equation has been studied extensively for a large class of kernels satisfying $K(bx,by)=b^{\lambda}K(x,y)$. The homogeneity exponent λ is shown to play a crucial role in classifying gelling and nongelling models. For instance, λ < 1 describes the nongelling model whose dynamics is governed by the conservation of mass principle, and $\lambda > 1$ describes the gelation transition accompanied by the violation of the mass conservation law [9,10]. Note, however, that, despite the seemingly simple structure of Eq. (1), it is solved exactly for the nongelling model only for a constant kernel λ =0, and that solution was given more than 100 years ago by Smoluchowski himself. Finding another exact analytical solution of Eq. (1) for the nongelling model (λ < 1) therefore still remains an open challenge.

One of the reasons that the Smoluchowski equation was so successful is that it has provided much of our theoretical understanding about dynamic scaling and associated exponents which are in agreement, at least qualitatively, with those extracted from real experiments and numerical simulations [11]. For instance, for $\lambda < 1$ it was shown that the distribution function c(x,t) exhibits dynamic scaling,

$$c(x,t) \sim t^{-\beta} \phi(x/t^z) \quad \text{with } z > 0, \tag{2}$$

in the long-time $(t \rightarrow \infty)$ large-size $(x \rightarrow \infty)$ limit where $\phi(\xi)$ is a scaling function whose argument $\xi = x/t^z$ is a dimensionless quantity.

The exponents β and z satisfy a scaling relation $\beta = \theta z$ with $\theta = 2$, which follows from the conservation of mass principle. A scaling form like Eq. (2) is shared by an extraordinarily diverse range of other phenomena, not just aggregation, e.g., systems exhibiting self-organized criticality, cluster growth in driven diffusive systems, fragmentation processes, etc. [12,13]. The ubiquity of this scaling form suggests the existence of a common underlying mechanism which makes such seemingly disparate systems behave in a remarkably similar fashion.

In addition to growth by aggregation, there exists a host of other mechanisms (e.g., condensation, deposition, and accretion) whereby particles can grow continuously between aggregations [14–17]. For instance, aerosol or colloidal particles are often not stable but evolve via aggregation and condensation, leading to gas-to-particle conversion. However, when the concentration of particles present is high and the supersaturation is low, the condensation is heterogeneous in nature since, in this case, condensation takes place only on the existing particles without forming new nuclei [1,16]. Otherwise, the system may have sufficient number of impurities, such as dirt or mist particles, which usually serve as potential nucleation sites on which condensation takes place, and the resulting process is known as homogeneous conden-



FIG. 1. Schematic representation of the model for $\alpha = 1$.

sation. In the latter case, the gas starts condensing on such nuclei and thereafter it is counted as a new particle, which then may take part in aggregation processes with other particles in the system. However, in the present work we will consider only the former case—heterogeneous condensation. Such condensation-driven aggregation does play an important role in the formation of the size spectra that ultimately determine the various physical properties of the aggregates. Motivated by this, we here propose a simple model which is defined in the next section.

The rest of the paper is organized as follows. In Sec. II, we present the definition of our model including its algorithm. Interesting results from numerical simulation based on the algorithm are presented in Sec. III. In Sec. IV, we propose a generalized version of the Smoluchowski equation and the appropriate parameters to describe the evolution of the distribution function c(x,t) following the rules set in the definition of the model. In Sec. V we give an explicit time dependent solution for c(x,t) and in Sec. VI we give its scaling description to obtain various scaling exponents and the solution for the scaling function. Finally, in Sec. VII, we discuss and summarize the work.

II. MODEL

We assume that initially the system consists of a large number of equal-sized chemically identical particles which are assumed to be immersed in the gas phase. These particles, while in Brownian motion, are continuously growing by heterogeneous condensation, leading to the gas-to-particle conversion, and merge irreversibly with other similarly growing particles upon encounter. To further specify our model, we assume that the amount of net growth by condensation of a given particle between collisions, in the most generic case, is directly proportional to its own size. The algorithm for one time unit of the model can then be defined as follows.

(i) Two particles are picked randomly from the system which mimics random collision via Brownian motion in one dimension.

(ii) The sizes of the two particles are increased by a fraction α of their respective sizes.

(iii) Their sizes are combined to form one particle.

(iv) The steps (i)-(iii) are repeated ad infinitum.

While the model is rather simple to define, the results it offers are far from simple. In order to illustrate how the rules

of the model work for monodisperse initial conditions, we give a simple example in Fig. 1 using an evolutionary-treebased approach. The state of the system at a given time is fully described by the numbers in the corresponding box. The evolution of these numbers is then described by a set of such boxes at successive times along the possible trajectory, e.g., $1 \rightarrow 3 \rightarrow 6 \rightarrow$ and so on. The first salient feature of this model is that the sum of all the numbers *L* in the successive boxes keeps increasing continuously with *t*, revealing that the conservation of mass law is clearly violated. Second, the numbers in the different boxes represent the size or mass of the aggregate and hence a given box can be characterized by a distribution function c(x,t) of particles of linear size *x* at time *t*.

III. NUMERICAL SIMULATION

In order to extract a couple of basic features of the model, we have performed extensive numerical simulations based on the algorithm (i)-(iv). Perhaps it is worthwhile to recall the work of Falk and Thomas, who in 1974 obtained the molecular-size distribution by simulating the discrete version of Eq. (1) [18]. The most crucial aspect of the stochastic simulation is to find a way to define the time t. Note that the time necessary for a particle to come into contact with another particle should depend on the number of particles present in the system. Indeed, ultimately it is the number density of particles present in the system that should determine how fast or how slowly the process should proceed, provided the collisions for aggregation of particles are independent of their size, and hence we define the time t=1/N. In Fig. 2 we therefore have plotted $\ln[L(t)]$ against $\ln(t)$ for different α and found a straight line with slope equal to 2α . This implies that

$$L(t) \sim t^{2\alpha},\tag{3}$$

which immediately confirms that, for systems describing the condensation-driven aggregation, conservation of mass principle is always violated. In Fig. 3 we also have plotted $\ln[s(t)]$ where s(t) is the mean particle size defined as

$$s(t) = L(t)/N(t), \qquad (4)$$

against $\ln(t)$, and again found a straight line with slope equal to $1+2\alpha$ for all $\alpha > 0$. We can thus write the following growth law for the mean particles size:



FIG. 2. $\ln(L)$ vs $\ln(t)$ (where t=1/N since the number of particles present ultimately determines how fast or slowly the aggregation process should proceed) using simulation data from one realization. The solid lines represent theoretical predictions with gradient equal to 2α .

$$s(t) \sim t^{1+2\alpha}.\tag{5}$$

IV. ANALYTICAL MODEL

To solve the model analytically, we use the generalized Smoluchowski (GS) equation



FIG. 3. $\ln(s)$ vs $\ln(t)$ using data collected from numerical simulation. Data points (denoted by symbols) for a given curve represent one realization of our simulation. The solid lines represent theoretical predictions with gradient equal to $1+2\alpha$.

$$\left(\frac{\partial}{\partial t} + \frac{\partial}{\partial x}v(x,t)\right)c(x,t) = -c(x,t)\int_0^\infty K(x,y)c(y,t)dy + \frac{1}{2}\int_0^x dy \ K(y,x-y)c(y,t)c(x-y,t),$$
(6)

where v(x,t) is the velocity with which particles grow by condensation. In the absence of the second term on the lefthand side, Eq. (6) reduces to the classical Smoluchowski (CS) equation as given in Eq. (1) whose dynamics is governed by the conservation of mass law [7]. The GS equation does not automatically describe our model unless the expressions for the growth velocity v(x,t), the collision time τ , and the kernel K(x,y) are suitably specified. To obtain a suitable expression for v(x,t), it is worthwhile to recall the definition of the mean growth velocity,

average growth velocity =
$$\frac{\text{net growth size } \Delta x}{\text{elapsed time } \Delta t}$$
. (7)

According to rule (ii), the net growth of a particle of size *x* between collisions is $\Delta x = \alpha x$. On the other hand, a simple dimensional analysis of Eq. (6) reveals that the inverse of $\int_0^\infty K(x,y)c(y,t)dy$ is the collision time $\Delta t = \tau(x)$ during which the growth αx takes place. The mean growth velocity between collisions therefore is

$$v(x,t) = \frac{\alpha x}{\tau(x)} = \alpha x \int_0^\infty dy \ K(x,y) c(y,t). \tag{8}$$

On the other hand, rule (i) says that a particle collides with any particle in the system, irrespective of their size with an equal *a priori* probability. We therefore should choose the collision kernel independent of the size of the colliding particles, i.e., we choose a constant collision kernel or

$$K(x,y) = 2, \tag{9}$$

for convenience.

To check our results from the numerical simulation, we now incorporate the kth moment defined as

$$M_k(t) = \int_0^\infty x^k c(x,t) dx \quad \text{with } k \ge 0, \tag{10}$$

together with Eqs. (8) and (9) in Eq. (6) to write the rate equation for $M_k(t)$ in the closed form

$$\frac{dM_k}{dt} = \sum_{r=0}^k \binom{k}{r} M_r M_{k-r} + 2(\alpha k - 1) M_0 M_k, \qquad (11)$$

for integer k value. We can readily solve it for the first moment $M_1(t) \equiv L(t)$ to give

$$L(t) = [1 + N(0)t]^{2\alpha}.$$
 (12)

In the long-time limit it grows following the same relation as in Eq. (3), which confirms a perfect matching with our numerical simulation. On the other hand, solving Eq. (11) with n=0, we find that

$$N(t) = \frac{N(0)}{1 + N(0)t},$$
(13)

and hence asymptotically the total number N decays following the same power law,

$$N(t) \sim t^{-1},\tag{14}$$

with the same exponent as that of the CS equation. This is consistent with the assumption that the new particles are not nucleated. A similar temporal behavior has also been observed in other theories and experiments [1,16]. One can use the two solutions for L(t) and N(t) in Eq. (4) to find the growth law for the mean particle size s(t),

$$s(t) \sim [1 + N(0)t]^{1+2\alpha}$$
. (15)

Therefore, one can immediately find that in the long-time limit the mean particle grows algebraically following the same relation as in Eq. (5), which further confirms that the GS equation together with the constant kernel and the growth velocity do describe the model in question.

V. EXACT SOLUTION

To solve the GS equation exactly, we define the Laplace transformation $\psi(p,t)$ of c(x,t) by

$$\psi(p,t) = \int_0^\infty dx \ e^{-px} c(x,t),$$
 (16)

and its inverse transform to obtain c(x,t) by

$$c(x,t) = \frac{1}{2\pi i} \int_{\gamma - i\infty}^{\gamma + i\infty} dp \ e^{px} \psi(p,t), \qquad (17)$$

with $\operatorname{Re}(p) > \gamma$. Differentiating Eq. (16) with respect to *t* and using Eq. (6), we find that $\psi(p,t)$ obeys the following non-linear partial differential equation:

$$\frac{\partial \psi(p,t)}{\partial t} + 2N(t) \left(1 - \alpha p \frac{\partial}{\partial p}\right) \psi(p,t) = \psi^2(p,t), \quad (18)$$

where, of course,

$$N(t) = \psi(0, t) = M_0(t).$$
(19)

Consequently, we need to solve Eq. (18) subject to the initial condition

$$\psi(p,0) = \int_0^\infty dx \ e^{-px} c(x,0) \equiv f(p).$$
 (20)

We incorporate the solution for N(t) from Eq. (13) in Eq. (18) and then rewrite it as follows:

$$\frac{\partial \psi(p,t)}{\partial t} - \left(\frac{2\alpha N(0)p}{1+N(0)t}\right) \frac{\partial \psi(p,t)}{\partial p} = \psi^2(p,t) - \frac{2N(0)}{1+N(0)t}\psi(p,t).$$
(21)

To solve this equation we use the method of characteristics, in which one usually writes

$$\frac{d\psi}{ds} = \frac{\partial\psi}{\partial t}\frac{\partial t}{\partial s} + \frac{\partial\psi}{\partial p}\frac{\partial p}{\partial s}.$$
(22)

Comparing the above two equations, we get

$$\frac{\partial t(s)}{\partial s} = 1,$$

$$\frac{\partial p(s)}{\partial s} = -\frac{2\alpha N(0)}{1+N(0)t}p(s).$$
(23)

The quantity ψ thus evolves following the ordinary nonlinear equation

$$\frac{d\psi}{ds} = \psi^2 - \frac{2N(0)}{1 + N(0)t(s)}\psi.$$
 (24)

Solving Eqs. (23) subject to initial data

$$t(s = 0) = 0,$$

 $p(s = 0) = p_0,$
 $\psi(s = 0) = f(p_0)$ (25)

yields

$$n = n [1 + N(0)s]^{2\alpha}$$
 (26)

$$p_0 - p[1 + N(0)s]$$
 (20)

We can transform Eq. (24) into a linear equation by setting

t = s.

$$\psi = \frac{1}{\chi},\tag{27}$$

to obtain

$$\frac{d\chi}{ds} - \frac{2N(0)}{1+N(0)s}\chi = -1.$$
(28)

We solve it by using the integrating factor $I = [1 + N(0)s]^{-2}$ and find that

$$\chi(s, p_0) = [1 + N(0)s] \{ [1 + N(0)s]\chi(0, p_0) - s \}, \quad (29)$$

and hence using it back in Eq. (27) we get

$$\psi(p,t) = \frac{f(p[1+N(0)t]^{2\alpha})}{[1+N(0)t]^2 \left(1 - \frac{f(p[1+N(0)t]^{2\alpha})t}{1+N(0)t}\right)}.$$
(30)

Note that, according to the definition of our model, we are interested in systems containing a large number of chemically identical particles of unit size and hence without loss of generality we may choose the monodisperse initial condition

$$c(x,0) = \delta(x-1), \tag{31}$$

which gives

$$f(p) = e^{-p}$$
 and $N(0) = 1.$ (32)

To find c(x,t) we substitute Eq. (30) in the definition of the inverse Laplace transform [Eq. (17)] and then a short calculation yields

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$$c(x,t) = \frac{t^{(1+t)^{-2\alpha_{x-1}}}}{(1+t)^{2+2\alpha}(1+t)^{(1+t)^{-2\alpha_{x-1}}}}, \quad \forall t > 0.$$
(33)

We can readily see that in the limit $\alpha \rightarrow 0$ we get the well-known solution of the CS equation [7,8].

VI. SCALING DESCRIPTION

Finding scaling or self-similar solutions is, more often than not, of utmost importance. These are essentially the solutions in the long-time limit where the distribution function c(x,t) takes a simple universal form in the sense that it is independent of initial conditions. Most experimental systems evolve to the point where such behavior is reached. Taking the limit $t \rightarrow \infty$ and using the identity

$$\lim_{n \to \infty} \left(1 + \frac{1}{n} \right)^n = e \tag{34}$$

in Eq. (33), we get

$$c(x,t) \sim t^{-(2+2\alpha)} \exp\left(-\frac{x}{t^{1+2\alpha}}\right).$$
(35)

The structure of the solution given above is highly instructive as it satisfies

$$c(b^{(1+2\alpha)}x,bt) = g(b)c(x,t),$$
 (36)

where

$$g(b) = b^{-(2+2\alpha)}$$
. (37)

It implies that if we increase the units of measurement of x by a factor of $b^{(1+2\alpha)}$ and that of time by a factor of b, the numerical value of c(x,t) is decreased by a factor of g(b). The existence of scaling in the present case means that a plot of $c(x,t)/t^{-\beta}$ vs x/t^z collapses into one graph for all initial conditions. Mathematically, a solution of this kind is called scale invariant and it implies that the system lacks a characteristic length scale. A further testament to the fact that the solution given by Eq. (35) exhibits scaling is that it has exactly the same form as in Eq. (2), and hence by comparing the two we can extract the scaling exponents

$$\beta = 2 + 2\alpha, \quad z = 1 + 2\alpha, \tag{38}$$

and the scaling function

$$\phi(\xi) = e^{-\xi}.\tag{39}$$

We can now substitute Eq. (35) in Eq. (10) to obtain the asymptotic solution for the *k*th moment

$$M_k(t) \sim t^{-\gamma(k)},\tag{40}$$

where

$$\gamma(k) = 1 - (2\alpha + 1)k.$$
(41)

We thus see that the exponent $\gamma(k)$ is linear in k which means that there exists a constant gap between exponents of consecutive k, and hence we can define the mean cluster size PHYSICAL REVIEW E 77, 061404 (2008)

$$s(t) = \frac{M_k(t)}{M_{k-1}(t)} \quad \text{for integer } k \ge 1.$$
 (42)

The mean particle size s(t) therefore grows algebraically with an exponent equal to $\gamma(k) - \gamma(k-1) = 1 + 2\alpha$, which is exactly the same as in Eq. (5) obtained by numerical simulation. Using Eq. (5) in Eq. (2), we can therefore write yet another widely used form of the scaling ansatz,

$$c(x,t) \sim s(t)^{-\theta} \phi(x/s(t)), \qquad (43)$$

with the mass exponent $\theta = \beta/z$, and hence according to Eq. (38) we get

$$\theta = \frac{2+2\alpha}{1+2\alpha}.\tag{44}$$

The expression for the exponent $\gamma(q)$ reveals that the moment $M_a(t)$ becomes time independent if

$$q = \frac{1}{1+2\alpha} \quad \forall \quad \alpha > 0, \tag{45}$$

since $\gamma(q)=0$. Incorporating the value of q in Eq. (44) we immediately find that

$$\theta = 1 + q, \tag{46}$$

which is always less than 2 for all $\alpha > 0$. We can recover the classical value $\theta=2$ of aggregation without condensation by setting $\alpha=0$.

VII. DISCUSSION AND SUMMARY

One may solve the GS equation for other growth velocities following the same method. Cueille and Sire, in fact, solved the GS equation for v=1 and v=x exactly and found

$$c(x,t) \sim \frac{2}{t^2 \ln t} e^{-x/t \ln t}, \quad c(x,t) \sim \frac{4}{t^2 e^t} e^{-2x/t e^t},$$
 (47)

respectively, for constant kernel K(x, y) = 1 [15]. As we have the exact solutions for three different growth velocities, we find it worthwhile to compare them and see their dissimilarities. First of all, note that one cannot choose v=1 and v=xand describe them as the growth velocities between collisions because the elapsed time Δt during which the net growth occurs has not been chosen to be equal to the mean collision time $\tau = 1 / \int_0^\infty K(x, y) c(y, t) dy$. Second, neither of the two solutions given in Eq. (47) can be expressed in the form of Eq. (36) or in the form of Eq. (2) and hence they violate scaling. Third, one finds that the kth moment of both solutions (v=1 and v=x) no longer exhibits a power law against time t, which is a further testament to the violation of scaling. Finally, when we choose v=0 and $v=\alpha x/\tau$, the dynamics of the systems are governed by some conservation laws and the scaling exponents are in fact fixed by these laws. However, there is no such conservation law in the case of v=1 or v=x and the scaling is violated and hence exponents are nonexistent.

In this paper, we have presented an exactly solvable analytical model to study the kinetics of aggregation of particles growing by heterogeneous condensation with velocity $v = \alpha x/\tau$. As a result of the additional growth by condensation, we found an algebraic growth law for the mean particle size $s(t) \sim t^{1+2\alpha}$ instead of a linear growth $s(t) \sim t$ in the absence of growth by condensation. The size spectra of the aggregates formed by the condensation-driven aggregation is shown to exhibit universal scaling $c(x,t) \sim t^{-\theta z} \phi(x/t^z)$ with mass exponent $\theta = 1 + q$, which is always less than its classical counterpart $\theta = 2$, and kinetic exponent $z = 1 + 2\alpha$. We found the exact and explicit solution for the scaling function $\phi(\xi) \sim e^{-\xi}$. Interestingly, one obtains the same scaling function $e^{-\xi}$ for aggregation without condensation. The difference

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appears only in the z values of the scaling argument $\xi = x/t^z$

since the two systems have different z values. In this sense,

the form of the scaling function is universal in nature. We

have shown that the transition to such scaling is accompa-

nied by the emergence of a nontrivial conservation law, that

is, $M_a(t) \sim \text{const.}$ We believe that the present work will at-

tract a renewed interest in the subject. The ideas developed

in this paper could be taken further by investigating the same

model for a generalized homogeneous aggregation kernel

 $K(x,y) = (xy)^{\lambda/2}$ which we intend to do in our future endeav-

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