Self-similarity theory of stationary coagulation

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A theory of stationary particle size distributions in coagulating systems with particle injection at small sizes is constructed. The size distributions have the form of power laws. Under rather general assumptions, the exponent in the power law is shown to depend only on the degree of homogeneity of the coagulation kernel. The results obtained depend on detailed and quite sensitive estimates of various integral quantities governing the overall kinetics. The theory provides a unifying framework for a number of isolated results reported previously in the literature. In particular, it provides a more rigorous foundation for the scaling arguments of Hunt, which were based purely on dimensional analysis.

1. INTRODUCTION

Aggregation phenomena are generally modeled using the kinetic equation first formulated by Smoluchowski in 1916 [1-2]:

$$\frac{dc_m}{dt} = \frac{1}{2}\sum_{n=1}^{m-1} K_{m-n,n}\, c_{m-n}\, c_n - c_m \sum_{n=1}^{\infty} K_{m,n}\, c_n. \tag{1}$$

Equation (1) has been used to model aggregating colloidal particles, coagulating drops in clouds, reacting polymers, growing gas bubbles in solids and liquids, fuel mixtures in engines, and star formation. For our purposes it will generally be more convenient to work with the continuous version of (1), due to Müller [3]:

$$\frac{\partial c(t,v)}{\partial t} = \frac{1}{2}\int_0^v K(v-u,u)\, c(t,v-u)\, c(t,u)\, du \, - \, c(t,v)\int_0^\infty K(v,u)\, c(t,u)\, du\,. \tag{2}$$

We shall refer to either (1) or (2) as the *Smoluchowski equation* (henceforth abbreviated SCE).

The equation is intended to describe an ensemble of particles, uniformly distributed in space, that remain uncorrelated for all time. The quantity $c_m(t)$ in (1) gives the number density of particles made up of m monomers. The quantity c(t,v) in (2) is the density at time t of particles of ‘size’ v, where ‘size’ may mean ‘mass’ or ‘volume’ or any other quantity conserved in the binary interactions. Henceforth, we shall refer to v simply as mass, but the broader interpretation of this quantity should be kept in mind since it is important for specific applications.

The key quantity identifying the type of coagulation process is the *coagulation kernel* or *collision frequency,* K_{mn} in the discrete case, and K(u,v) in the continuous case, respectively. This quantity models the physics of the coagulation process through its dependence on its arguments. Particles are implicitly assumed to move in some deterministic or stochastic way.

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When two come into contact, they coalesce into a single particle with a mass equal to the sum of the constituent masses. The density of particles is assumed to be sufficiently low that one may restrict attention to binary collisions. The first term on the right hand side of (2) gives the rate of change of particles of mass v due to particles of mass v – u and u coagulating. The second term counts the depletion of particles of mass v by those particles coagulating with particles of any other mass. The coagulation kernel is always assumed to be symmetric in its arguments (or indices). Because of its physical interpretation as a probability, it is non-negative.

There is an extensive literature on the different mechanisms that govern collisions of particles in various disperse systems, and on the derivation of the appropriate form of the coagulation kernel for each one. Kernels for coagulation via Brownian motion, coagulation of spherical particles in a laminar shear or pure straining flow, coagulation due to advection by a turbulent flow, coagulation in a turbulent flow taking account of particle inertia, coagulation due to differential sedimentation, and kernels representing yet other physical mechanisms have been derived [4-5].

In the discrete case one explicitly recognizes the existence of a smallest particle mass (a 'monomer'). In the continuous case we allow arbitrarily small particles, although we shall find it useful to consider (2) with a smallest particle size cutoff. It is clear from (2) that if at $t = 0$ the particle distribution is such that $c(0,v) = 0$ for all $v < v_0$, then $c(t,v)$ remains zero for $v < v_0$ for all time.

By choosing a particular mechanism, i.e., a certain coagulation kernel, and giving an initial distribution of particle sizes, various exact analytical solutions of SCE have been found [4], [6-7]. However, the study of an isolated case, by analysis or numerical simulation, cannot, of course, address the key question of how typical such a solution is or of how it is related to the evolution of real aggregating systems, where the kernel may not be precisely the one chosen for study. Our approach is aimed at these more global issues and so aims to work only with rather generic properties of the SCE, such as the degree of homogeneity of the coagulation kernel (discussed next), and the convergence of various integral quantities associated with SCE.

Many of the coagulation kernels proposed for various processes have the property that they are homogeneous functions of their arguments [4], i.e., that

$$K(\lambda u, \lambda v) = \lambda^{\alpha} K(u, v), \tag{3}$$

for any positive real number λ with a fixed exponent α. In particular, Smoluchowski studied the case of Brownian motion for which the kernel has $\alpha = 0$. For coagulation in a laminar flow $\alpha = 1$. For coagulation due to differential sedimentation $\alpha = 4/3$. And so on. Homogeneity of the coagulation kernel is the formal statement that the coagulation process does not have a characteristic scale, i.e., that aggregation of particles at different scales is assumed to happen similarly except for a possible change in the rate of the process.

In turn, this suggests either that asymptotic solutions of (1) or (2) should display a similarity form or that steady-state solutions – which would arise by, somehow, 'feeding' the coagulating mix so as to maintain the steady state – should be power-laws. That is, one is led either to the suggestion that the initial value problem has solutions of the form

$$c(t,v) = f(t)^{-2}\, \Psi(v/f(t)), \tag{4}$$

where $f(t)$ is to be determined. (The exponent in the pre-factor guarantees that the mean cluster size, $\langle v\rangle = \int vc(v,t)\, dv$, is constant.) Or one is led to suggest that for a 'forced' version of (2) the steady state solutions are of the form

$$c(v) = \text{const.} \times v^{-\tau}, \tag{5}$$

where τ is another exponent, appeared to us to be more analytically tractable. At issue, then, is the problem of ascertaining when (5) is, indeed, the steady-state solution (given some physically reasonable model of the 'forcing'), and how the exponent τ depends on the coagulation kernel, in particular through its homogeneity exponent, α (but, possibly, in other ways as well).

The possibility (4) was pursued in the work of Friedlander [8]. To have a clear terminology we shall refer to this approach as the *self-preservation theory* and to the approach summarized by (5) as the *self-similarity theory*. The possibility of power-law solutions of the form (5) was raised by Hunt [9] in an important paper that, however, seems to have been somewhat overlooked in the literature on coagulation. Hunt patterned his reasoning on the Kolmogorov scaling theory for turbulent flow [10]. Thus, he enunciated four assumptions, similar to those made for the turbulent 'cascade', that allowed him to apply dimensional analysis arguments to the problem, and thus predict the exponent τ for various kernels. Because of the analogy to turbulence theory, we shall often refer to a power-law distribution for the mass density in coagulation as a 'mass spectrum'. The similarity solutions (4) have also been explored with the objective of identifying when this form will lead to power-law solutions asymptotically. For the most far-reaching work in this direction see the papers by van Dongen and Ernst [11-12].

There are important differences between the approaches summarized by Eqs.(4) and (5). A self-preserving distribution (4) presumes the existence of a single characteristic size in the system, which can be chosen equal to the average cluster size. Accordingly, the self-preserving distribution should have a shape with a single hump, similar to a log-normal distribution. The theory aims at the case when $\langle v\rangle$ is finite and so excludes what in polymer science is called the gelating case for which the average cluster size diverges after a finite time. The self-similarity theory explored in this paper, on the other hand, predicts scale-free power-law distributions that arise due to forcing. The average cluster size does not need to be finite. If it is not, the influx of mass into the system equals the mass flux to the infinite size cluster. There is no restriction to kernels that give finite average cluster size, i.e., both gelating and non-gelating cases are covered by the theory (modulo the restrictions identified later in the analysis).

Unfortunately, Hunt's assumptions [9] seem overly restrictive. For example, he assumed that collisions between particles of very different size would not contribute significantly to the flux of mass through the distribution and so could be ignored. This is similar to the assumption in turbulence theory that eddies very different in size do not contribute substantially to the flow of energy through the 'cascade', i.e., that the energy 'cascade' is 'local'. However, in the case of a constant kernel in (1), where the collision frequency is independent of particle size, one can solve for the steady-state mass spectrum analytically and one finds that it obeys Hunt's scaling predictions even though the key assumption of 'locality' underlying his analysis appears to be violated (see Appendix). This observation led us to re-examine the conditions under which (2) had steady-state solutions of the form (5). The main purpose of this paper is to report on the results of this re-examination. The remainder of the paper is thus set out as follows:

First, in Sec.II, we discuss what we mean by 'forced' Smoluchowski kinetics, and how such a notion of 'forcing' leads us to substitute for the initial-value problem for SCE a boundary-value problem for the 'forced' kinetics. It is this boundary value problem that has steady-state, power-law solutions of the form (5). In Sec.II we also introduce the mass flux through the 'spectrum' of coagulating particles.

Next, in Sec.III, we study the equations to be satisfied by a steady-state solution to the 'forced' problem, and we establish a very useful representation of these solutions which is the basis for our further analysis. A relationship, Eq.(20), between the power-law exponent, τ, in (5) and the homogeneity exponent, α, in (3) is found, but at this stage this relation contains an as yet undetermined, additive, 'anomalous' exponent θ.

In Sec.IV we introduce the additional assumption that K(u,v) becomes just a product of two powers when $u >> v$ or, because of the symmetry, when $u << v$. The exponents of these powers, which we call μ and ν, respectively, must, of course, add to α. One can view this extension of the homogeneity condition (3), an extension that is satisfied by many of the best known examples in applications, as our counterpart of Hunt's locality assumption. With it we can show that various inequalities must be obeyed by the various scaling exponents we have introduced. Establishing these relations by asymptotic analysis is the main subject of Sec.IV. The main results can be found in Eqs.(23) and (27).

In Sec.V we return to the equation for the mass flux from Sec.II. It turns out that the full integral expression can be substantially reduced in the case of a steady-state, power-law solution, and this reduction is important for further analytical progress.

Much of the work in Secs.IV-V is preparatory to Sec.VI where, having stripped down the expression for the mass flux, we are able to show, finally, that the anomalous scaling exponent, θ, must vanish. This leads to our main result stated in Eq.(37). Our concluding Sec.VII contains discussion of the results obtained.

The main results of this work were first reported at the annual meeting of the American Physical Society, Division of Fluid Dynamics in New Orleans, November 1999 [13]. While this paper was being prepared, we became aware of the work of Davies, King and Wattis [14-15] in which analytical results are obtained for coagulation kernels $K_{mn} = \frac{1}{2}(m^{\mu}n^{\nu} + m^{\nu}n^{\mu})$, with $\mu + \nu = \alpha$. Their exact results agree with key aspects of our more general arguments and thus provide important points of validation for the theoretical ideas advanced in this paper.

2. FORCED SMOLUCHOWSKI KINETICS

As indicated in Sec.I it is convenient to study (2) subject to the following modifications: (i) we assume there is a smallest particle mass v_0 in the system for all times, and (ii) we posit a 'forcing mechanism' that constantly replenishes particles. We may take this 'forcing' to be quite general, i.e., define a quantity j(t,v) that gives the influx of particles of mass v into the system at time t, and stipulate this function more or less freely. We shall focus on the case when j(t,v) is concentrated at the small particle end of the spectrum and acts to maintain the density of the smallest particles constant. In the discrete case we would simply stipulate that $c_1(t)$ be constant, but this is awkward in the continuous case, so we allow j(t,v) to be spread over a range of particles, say particles with mass $v_0 \le v \le 2v_0$, such that c(t,v) is maintained constant in this interval. The precise nature of the forcing is immaterial.

While these assumptions are most helpful to the analysis, we argue that they are also quite realistic physically in a variety of situations. Thus, the smallest particles in a chemical or combustion process, e.g., in a stirred tank reactor or in smoke, may be assumed to exist in a largely time-independent density. The counterpart to the notion that the initial-value problem has a similarity solution is then that the boundary-value problem has a steady-state solution that 'forgets' the smallest particle size, v_0, for $v >> v_0$.

We shall refer to (2) with the stipulations that c(t,v) = 0 for $v < v_0$ and a particle injection term j(t,v) on the right hand side as the *forced Smoluchowski equation,* henceforth abbreviated FSCE. Modifications to SCE wherein a mechanism for particle addition to the system is included have been considered previously by several authors, see [14] and references therein.

A general form of the FSCE is, then,

$$\frac{\partial c(t,v)}{\partial t} = j(t,v) - s(t,v) \tag{6a}$$

where j(t,v) is to be specified, and s(t,v) is the right hand side in (2) suitably modified to take account of the small-size cutoff. In particular, for $v_0 \le v \le 2v_0$:

$$s(t,v) = c(t,v)\int_{v_0}^{\infty} K(v,u)\, c(t,u)\, du\,, \tag{6b}$$

and for $2v_0 \le v$:

$$s(t,v) = -\frac{1}{2}\int_{v_0}^{v-v_0} du\, vK(v-u,u)\, c(t,v-u)\, c(t,u) + c(t,v)\int_{v_0}^{\infty} K(v,u)\, c(t,u)\, du\,. \tag{6c}$$

Just as the energy flux plays a key role in Kolmogorov's theory of turbulent flow, so does the flux of mass, E, play a key role in the self-similar solutions of coagulation kinetics. Indeed, these solutions are characterized by having a constant flux of mass through the spectrum of particle sizes. The rate of change of the total mass in the system is

$$\frac{dM}{dt} = \int_{v_0}^{\infty} \frac{\partial c(t,v)}{\partial t}\, dv = \int_{v_0}^{\infty} v\left(j(t,v) - s(t,v)\right) dv = J(t) - S(t), \tag{7a}$$

where J(t) is the total influx of mass,

$$J(t) = \int_{v_0}^{\infty} v\, j(t,v)\, dv\,, \tag{7b}$$

and S(t) is the total efflux of mass 'at infinity',

$$S(t) = \int_{v_0}^{\infty} v\, s(t,v)\, dv\,. \tag{7c}$$

We should think of these integrals initially as limits of integrals over a finite range of masses, $v_0 \le v \le V$, and then let $V \to \infty$. Since $j(t,v)$ is assumed to be concentrated at small v, the integral J(t) poses no convergence issues – its range could be truncated to $v_0 \le v \le 2v_0$. The integral S(t), however, merits closer examination. We have

$$\begin{aligned} S(t) = \lim_{V\to\infty} \int_{v_0}^{V} v\, s(t,v)\, dv \;=\; \lim_{V\to\infty} \Big[\int_{v_0}^{V} dv \int_{v_0}^{\infty} du\; vK(v,u)\, c(t,v)\, c(t,u) \\ - \tfrac{1}{2} \int_{2v_0}^{V} dv \int_{v_0}^{v-v_0} du\; vK(v-u,u)\, c(t,v-u)\, c(t,u) \Big]. \end{aligned} \tag{8}$$

In the second double-integral we write v as v – u + u. The integrand is then symmetric in the variables u and v – u. The integration domain is easily seen also to be symmetric in terms of these variables. Hence, the integral may be written as

$$- \int_{v_0}^{V-v_0} dv \int_{v_0}^{V-v} du\; v\, K(u,v)\, c(t,u)\, c(t,v)\,.$$

Taken together with the first integral we obtain

$$S(t) = \lim_{V\to\infty} \left[S_V^{(1)}(t) + S_V^{(2)}(t) \right], \tag{9a}$$

where

$$S_V^{(1)}(t) \;=\; \int_{v_0}^{V-v_0} dv \int_{V-v}^{\infty} du\; v\, K(u,v)\, c(t,u)\, c(t,v)\,, \tag{9b}$$

$$S_V^{(2)}(t) \;=\; \int_{V-v_0}^{V} dv \int_{v_0}^{\infty} du\; v\, K(u,v)\, c(t,u)\, c(t,v)\,. \tag{9c}$$

We shall see in Sec.V that for the solutions (5) of interest here, $S_V^{(2)}(t)$ will, not surprisingly, tend to zero as $V \to \infty$, but $S_V^{(1)}(t)$ will have a finite limit. Of course, for the steady-state solutions both integrals are time-independent.

We note that if the integral

$$\int_{v_0}^{\infty} dv \int_{v_0}^{\infty} du\, v\, K(u,v)\, c(t,u)\, c(t,v) \tag{10}$$

converged, then $S_V^{(1)}(t)$ and $S_V^{(2)}(t)$, and thus S(t), would vanish in the limit $V \to \infty$. However, to describe a stationary distribution sustained by a constant influx of mass, E, we have J(t) = E, and since the total mass of the system is to remain constant, we must have S(t) = J(t) = E according to (7a). Thus, assuming convergence of (10), which is sometimes done in analytical investigations of SCE, is an additional assumption that rules out the solutions we are after! In the literature on coagulation applied to polymers it is realized that (10) should diverge in certain cases, and this divergence is associated with the phenomenon of gelation [11].

3. STEADY-STATE SOLUTIONS OF FSCE

Consider a steady-state solution of (6a) for $v \geq 2v_0$ and assume the forcing is confined to smaller particles so that the balance of interest is

$$\frac{1}{2}\int_{v_0}^{v-v_0} du\, K(v-u,u)\, c(v-u)\, c(u) = c(v)\int_{v_0}^{\infty} K(v,u)\, c(u)\, du\,. \tag{11}$$

We have omitted the time dependence since we are seeking a steady-state solution. We introduce the quantities $k(u,v;v_0)$ by

$$k^2(u,v;v_0) = E^{-1}\, K(u,v)\, c(u)\, c(v)\, u^{3/2}\, v^{3/2}, \tag{12}$$

where E is the mass flux through the system. The 3/2 powers of u and v have been factored out for two reasons. First, this makes k dimensionless. Second, for u = v we have the representation

$$c(v) = \left[\frac{E}{K(v,v)}\right]^{1/2} v^{-3/2}\, k(v;v_0), \tag{13}$$

where the repeated argument in k has been dropped, and the spectrum $c(v) = \text{const.} \times v^{-3/2}$ turns out to be the solution for a constant kernel (see Appendix). Indeed, for this case (13) follows essentially by dimensional analysis. In general, Eq.(13) provides a representation of c(v) that consists of two factors, one involving the mass flux E, the other involving the small scale cut-off v_0.

So far we have accomplished nothing but to write one unknown quantity, c(v), in terms of another, $k(v;v_0)$. However, due to the scale invariance of the coagulation kernel, it turns out that $k(v;v_0)$ in (13) must, in fact, have the form $k(v/v_0)$.

To see this we substitute (13) into both sides of (11). We scale the variables u and v by v_0, introducing new variables $x = v/v_0$, and $y = u/v_0$. Then we use the homogeneity of the kernel, Eq.(3), to factor out v_0 as follows: $K(v - u, u) = K((x - y)v_0, yv_0) = v_0^{\alpha} K(x - y, y)$, and so on. In this way we obtain:

$$\frac{1}{2}\int_1^{x-1} dy \, \frac{K(x-y,y)}{\sqrt{K(x-y,x-y)\,K(y,y)}} \, (x-y)^{-3/2}\, y^{-3/2}\, k(x-y)\, k(y) = \int_1^{\infty} dy \, \frac{K(x,y)}{\sqrt{K(x,x)\,K(y,y)}} \, x^{-3/2}\, y^{-3/2}\, k(x)\, k(y) \tag{14}$$

where $k(x) = k(xv_0;v_0)$. Both E and, more remarkably, v_0 drop out of equation (14)! Setting

$$Q(x,y) = \frac{K(x,y)}{\sqrt{K(x,x)\,K(y,y)}} \tag{15}$$

we have the following integral equation for determining the function k:

$$\frac{1}{2}\int_1^{x-1} dy \; Q(x-y,\, y)\, (x-y)^{-3/2}\, y^{-3/2}\, k(x-y)\, k(y) = \int_1^{\infty} dy \; Q(x,\, y)\; x^{-3/2}\, y^{-3/2}\, k(x)\, k(y). \tag{16}$$

Assuming (16) has a solution, $k(x)$, we have $k(v;\, v_0) = k(xv_0;\, v_0) = k(v/v_0)$. We now have the more substantial version of (13) that

$$c(v) = \left[\frac{E}{K(v,v)}\right]^{1/2} v^{-3/2}\; k(v/v_0), \tag{17}$$

where $k(x)$ is a solution of (16).

We see that (16) will only determine k up to a multiplicative factor. This is consistent with (17) which must be augmented by the condition that E is, indeed, the mass flux. Recalling (9), and the definition (15), we have

$$\lim_{N\to\infty}\Big[\Big(\int_1^{N-1} dx \int_{N-x}^{\infty} dy \ + \int_{N-1}^{N} dx \int_1^{\infty} dy\Big) \frac{Q(x,y)}{\sqrt{x\, y^3}} \ k(x)\, k(y)\Big] = 1 \tag{18}$$

as the ‘normalization condition’ on the function k. Solutions of the pair of equations (16) and (18) produce steady-state solutions of the FSCE with kernel K(u,v) via (17). These solutions have a constant mass flux, E, which enters as a coefficient.

If c(v) in (17) is to behave as a power law when $v >> v_0$, as envisioned in Eq.(5), i.e., if we demand that $c(\lambda v) = \lambda^{-\tau} c(v)$, then we must have

$$c(\lambda v) = \left[\frac{E}{K(\lambda v,\lambda v)}\right]^{1/2} (\lambda v)^{-3/2}\, k(\lambda v/v_0) = \lambda^{-\tau} \left[\frac{E}{K(v,v)}\right]^{1/2} v^{-3/2}\, k(v/v_0)$$

or, since $K(\lambda v,\lambda v) = \lambda^{\alpha} K(v,v)$,

$$k(\lambda x) = \lambda^{-\tau + 3/2 + \alpha/2}\, k(x), \tag{19}$$

i.e., k must itself be a power, $k(x) = k_0\, x^{-\theta}$ (for x >> 1), where τ and θ are related by

$$\tau = \frac{3+\alpha}{2} + \theta. \tag{20}$$

If k goes to a constant for large arguments, i.e., if $\theta = 0$, then the system does, indeed, ‘forget’ the small size v_0, and scale invariance is fully restored at large particle masses. This is referred to as *similarity of the first kind* [16]. If, on the other hand, $\theta \neq 0$, we have *similarity of the second kind* [16], or in the language of critical phenomena, an *anomalous exponent*. The next section explores these issues further.

4. INEQUALITIES FOR SCALING EXPONENTS

We now augment the homogeneity condition (3) slightly by requiring, in addition, that

$$K(u,v) \approx u^{\mu}\, v^{\nu} \quad \text{for} \quad v >> u, \tag{21a}$$

where, of course, $\mu + \nu = \alpha$. Due to symmetry, (21a) also implies that $K(u,v) = K(v,u) \approx u^{\mu}\, v^{\nu}$ for v >> u, i.e., that

$$K(u,v) \approx u^{\nu}\, v^{\mu} \quad \text{for} \quad u >> v. \tag{21b}$$

This conditions (21) are satisfied by many of the kernels used in common applications of the SCE. Thus, for coagulation due to Brownian motion the kernel is

$$K_B(u,v) \propto (u^{1/3} + v^{1/3})^2 /(uv)^{1/3}, \tag{22a}$$

i.e., $\alpha = 0$, $\mu = -\nu = -1/3$. For coagulation due to laminar shear

$$K_{sh}(u,v) \propto (u^{1/3} + v^{1/3})^3, \tag{22b}$$

so $\alpha = \nu = 1$, $\mu = 0$,. And for coagulation due to differential sedimentation

$$K_{ds}(u,v) \propto (u^{1/3} + v^{1/3})^2 \, |u^{2/3} - v^{2/3}|, \tag{22c}$$

which gives $\alpha = \mu = 4/3$, $\nu = 0$.

4.1 *The inequality* $\alpha - 2\nu + 2\theta + 1 > 0$

The conditions (21) give a nuance to the homogeneity condition (3) that allows us to obtain useful asymptotic estimates of various integrals and thus to write inequalities for the exponents we have introduced. As an easy example, from the discussion in Sec.II, particularly Eqs.(9), we see that

$$\int_{v_0}^{\infty} du \; v \, K(u,v) \, c(t,u) \, c(t,v)$$

must exist. Substituting (5) and (21b) we see that the integrand for large u varies as $u^{\nu-\tau}$. Thus, for convergence we must have $\nu - \tau < -1$ or

$$\tau - \nu - 1 > 0. \tag{23a}$$

or, using (20),

$$\alpha - 2\nu + 2\theta + 1 > 0. \tag{23b}$$

4.2 *The inequality* $\alpha - 2\mu + 2\theta + 1 > 0$

We turn next to Eq.(11) itself. We may reason as follows: Let v_c be such that with sufficient accuracy $c(v) = Av^{-\tau}$, with A a constant, for $v > v_c$. Split the integral on the left hand side of (11) into a sum of three integrals, the first from v_0 to v_c, the second from v_c to $v - v_c$, the third from $v - v_c$ to $v - v_0$. The first and third integral are identical as is seen by the substitution $u' = v - u$. Consider Eq.(11) for a large value of v, say $v >> 2v_c + v_0$. In an integral where $v_0 \le u \le v_c$, we see that $v - u >> v_c$. Hence, $c(v-u) = A(v-u)^{-\tau}$ with sufficient accuracy. In an integral where $v_c \le u \le v - v_c$ we have $c(u) = Au^{-\tau}$ but also $v - u \ge v_c$ so that $c(v-u) = A(v-u)^{-\tau}$. Thus, we get the asymptotic estimate

$$\frac{1}{2}\int_{v_0}^{v-v_0} du\, K(v-u,u)\, c(v-u)\, c(u) =$$

$$A\int_{v_0}^{v_c} du\;\; K(v-u,u)\; c(u)\; (v-u)^{-\tau} + \frac{1}{2}A^2 \int_{v_c}^{v-v_c} du\;\; K(v-u,u)\; u^{-\tau}\, (v-u)^{-\tau}$$

Now from (21′) use the estimate $K(v-u,u) = B\,(v-u)^{\nu}\, u^{\mu}$, where B is another constant, in the first of these integrals. As $v \to \infty$ we then have for this integral

$$AB\int_{v_0}^{v_c} du\;\; (v-u)^{\nu-\tau}\, u^{\mu}\;\, c(u) \approx \left[AB\int_{v_0}^{v_c} du\, u^{\mu}\;\, c(u)\right] v^{\,\nu-\tau}.$$

This is the mass influx due to that part of of the mass spectrum that has not achieved power-law form. If we look on the right hand side of (11), we see immediately that the integral from v0 to vc there, which describes the mass efflux due to the non-power law portion of the mass spectrum, will asymptotically exactly balance the influx!

We are left to consider the balance

$$\frac{1}{2}A^2 \int_{v_c}^{v-v_c} du\;\; K(v-u,u)\; u^{-\tau}\, (v-u)^{-\tau} = c(v)\int_{v_c}^{\infty} K(v,u)\, c(u)\, du \tag{24}$$

or, substituting in the asymptotic forms for the distributions

$$\frac{1}{2}\int_{v_c}^{v-v_c} du\;\; K(v-u,u)\; u^{-\tau}\, (v-u)^{-\tau} = v^{-\tau}\int_{v_c}^{\infty} du\;\; K(v,u)\; u^{-\tau}$$

We substitute $u = \xi\, v$ in the integrals and, using the homogeneity of the kernel, obtain

$$\frac{1}{2}\int_{\xi_c}^{1-\xi_c} d\xi\;\; K(1-\xi,\,\xi)\; \xi^{-\tau}\, (1-\xi)^{-\tau} = \int_{\xi_c}^{\infty} d\xi\;\; K(1,\,\xi)\; \xi^{-\tau}$$

where $\xi_c = v_c/v$. Because of the symmetry of the integrand on the left hand side, this balance equation may also be written

$$\int_{\xi_c}^{1/2} d\xi\;\; K(1-\xi,\,\xi)\; \xi^{-\tau}\, (1-\xi)^{-\tau} = \int_{\xi_c}^{\infty} d\xi\;\; K(1,\,\xi)\; \xi^{-\tau} \tag{25}$$

As $v \to \infty$, we have that $\xi_c \to 0$. Thus, close to the lower limit both integrands vary as $B\,\xi^{\mu-\tau}$, which diverges for $\tau \geq \mu + 1$ and converges for $\tau < \mu + 1$. Since the divergences are similar, and with the same coefficient, we obtain a balance in these cases to leading order. In the convergent case, however, we only obtain a balance if

$$\int_0^{1/2} d\xi \; K(1-\xi, \xi)\, \xi^{-\tau} (1-\xi)^{-\tau} = \int_0^{\infty} d\xi \; K(1, \xi)\, \xi^{-\tau}. \tag{26}$$

In general, this relation for K is not satisfied. Therefore, in the convergent case the necessary condition for a power-law spectrum is not satisfied (except possibly for exceptional cases).

We conclude from these considerations that in order to have a power-law steady-state distribution, we should insist that

$$\tau - \mu - 1 \geq 0. \tag{27a}$$

This relation looks deceptively similar to (23a), but the arguments given can leave no doubt that it is a deeper result. Note that equality is allowed in (27a), whereas (23a) is a strict inequality. As in (23) we may write (27a) in terms of the exponents α and θ:

$$\alpha - 2\mu + 2\theta + 1 > 0. \tag{27b}$$

Adding (23b) and (27b) we have the easy result that $\theta \geq -1/2$.

5. NORMALIZATION REVISITED

In this subsection we pursue estimates similar to those of Sec.IV for the normalization condition, Eq.(18). As a lead-in we show the result mentioned in Sec.II that for the steady-state, power-law solutions $S_V^{(2)}$, Eq.(9c), will tend to zero as $V \to \infty$, while $S_V^{(1)}$, Eq.(9b), will have a finite limit.

In the outer integral of

$$S_V^{(2)} = \int_{V-v_0}^{V} dv \int_{v_0}^{\infty} du \; v\, K(u,v)\, c(u)\, c(v)\,, \tag{9c}$$

we substitute $v = \xi V$ to obtain

$$S_V^{(2)} = V^2 \int_{1-v_0/V}^{1} d\xi \; \xi\, c(\xi V) \int_{v_0}^{\infty} du \; K(u, \xi V)\, c(u) \approx$$

$$V\, v_0\, c(V) \int_{v_0}^{\infty} du \; K(u, V)\, c(u) = A v_0 V^{1-\tau} \int_{v_0}^{\infty} du \; K(u, V)\, c(u). \tag{28}$$

The remaining integral is split into two, the first from v_0 to v_c, the second from v_c to ∞. In the first we can set $K(u,V) = Bu^{\mu} V^{\nu}$ according to (21a). It then varies asymptotically as V^{ν}. In the second we can set $c(u) = Au^{-\tau}$, and then

$$\int_{v_c}^{\infty} du\ K(u, V)\ Au^{-\tau} = AV^{1+\alpha-\tau}\int_{v_c/V}^{\infty} d\xi\ K(\xi, 1)\ \xi^{-\tau}.$$

At the large-ξ limit the integral converges because $K(\xi,1)$ varies as $B\xi^{\nu}$ by (21a) and we have inequality (23a). At the small-ξ limit $K(\xi,1)$ varies as $B\xi^{\mu}$ by (21b) and the leading order term is of order $V^{1+\alpha-\tau}(v_c/V)^{\mu-\tau+1}$ or $V^{\nu}(v_c)^{\mu-\tau+1}$, i.e., of the same order as the first integral, V^{ν}, as $V \to \infty$. Multiplying both these asymptotic results by $V^{1-\tau}$, as in (28), we see that

$$S_V^{(2)} \propto V^{\nu-\tau+1} \to 0 \quad \text{as} \quad V \to \infty \tag{29}$$

because of (23a).

As anticipated, we are therefore left with $S_V^{(1)}$ in (18). But this statement may be refined further. Indeed, we will now show that only the integral over the mass range where both $c(u)$ and $c(v)$ can be adequately approximated by power-law forms contributes to $S_V^{(1)}$ in the large-V limit.

We start from

$$S_V^{(1)} = \int_{v_0}^{V-v_0} dv \int_{V-v}^{\infty} du\ v\ K(u,v)\ c(u)\ c(v) \tag{9b}$$

and split the outer integral into three, the first from v_0 to v_c, the second from v_c to $V - v_c$, and the third from $V - v_c$ to $V - v_0$. In the first integral, then, $c(u) = Au^{-\tau}$ to sufficient accuracy, and $K(u,v) = Bu^{\nu} v^{\mu}$ by (21b). In the third $c(v) = Av^{-\tau}$. In the second, which describes the contribution of the self-similar part of the distribution, both $c(u) = Au^{-\tau}$ and $c(v) = Av^{-\tau}$. to sufficient accuracy. Now we have an easy order of magnitude estimates for the first integral:

$$\int_{v_0}^{v_c} dv \int_{V-v}^{\infty} du\ v\ K(u,v)\ c(u)\ c(v) \approx AB\int_{v_0}^{v_c} dv\ v^{\mu+1}\ c(v) \int_{V-v}^{\infty} du\, u^{\nu-\tau}$$

$$= \frac{AB}{\nu-\tau+1}\int_{v_0}^{v_c} dv\ v^{\mu+1}\,(V-v)^{\nu-\tau+1}\ c(v) \ \propto\ V^{\nu-\tau+1} \tag{30}$$

so that it vanishes in the $V \to \infty$ limit.

For the third integral we reason as follows:

$$\int_{V-v_c}^{V-v_0} dv \int_{V-v}^{\infty} du\;\; v\, K(u,v)\, c(u)\, c(v) \approx A \int_{V-v_c}^{V-v_0} dv\;\; v^{\,1-\tau} \int_{V-v}^{\infty} du\;\; K(u,v)\, c(u)$$

$$= A \int_{v_0}^{v_c} dw\;\; (V-w)^{\,1-\tau} \int_{w}^{\infty} du\; K(u, V-w)\, c(u)$$

$$= A \int_{v_0}^{v_c} dw\;\; (V-w)^{\,1-\tau} \left[\int_{w}^{v_c} du + \int_{v_c}^{\infty} du \right] K(u, V-w)\, c(u)$$

In the first u-integral we can set $K(u, V-w) = Bu^{\mu}\ (V-w)^{\nu}$ by (21a) and it then becomes

$$AB \int_{v_0}^{v_c} dw\;\; (V-w)^{\,\nu-\tau+1} \int_{w}^{v_c} du\;\; u^{\mu}\;\; c(u) \;\propto\; V^{\nu-\tau+1}. \tag{31}$$

The second u-integral requires further work:

$$A \int_{v_0}^{v_c} dw\;\; (V-w)^{\,1-\tau} \int_{v_c}^{\infty} du\; K(u, V-w)\, c(u) \approx$$

$$A^2\, V^{\alpha-\tau+1} \int_{v_0}^{v_c} dw\;\; (V-w)^{\,1-\tau} \int_{v_c/V}^{\infty} dz\;\; K(z, 1-w/V)\; z^{-\tau}$$

The inner integral converges at the upper limit because of (21b) and (23a). From the lower limit using (21a) we obtain the leading order term in V as

$$A^2\, B\, V^{\mu-\tau+1} \int_{v_0}^{v_c} dw\;\; (V-w)^{\,\nu-\tau+1} \int_{v_c/V}^{\infty} dz\;\; z^{\,\mu-\tau}. \tag{32a}$$

For $\tau > \mu + 1$ this varies as $(v_0 - v_c)\ v_c^{\mu-\tau+1}\ V^{\,\nu-\tau+1}$ and, thus, also vanishes in the limit $V \to \infty$. It is interesting to note that even if v_c decreases, i.e., the distribution becomes self-similar at a small value of the mass, the integral increases! Thus, a short range of masses before the distribution becomes self-similar does not imply that this range makes a negligible contribution to the mass flux E.

For $\tau = \mu + 1$ we get

$$-\,A^2\, B\; V^{\,\nu-\mu} \log(v_c/V) \int_{v_0}^{v_c} dw\;\; (1-w/V)^{\,\nu-\mu}. \tag{32b}$$

Since by (23a) $\nu < \tau - 1 = \mu$, this expression will also tend to zero in the $V \to \infty$ limit.

In summary, assuming we have a power-law distribution (and the coagulation kernel satisfies

(21)), the mass flux must satisfy

$$A^2 \lim_{V \to \infty} \int_{v_c}^{V - v_c} dv \int_{V - v}^{\infty} du \; K(u,v)\, u^{-\tau} v^{1-\tau} = E \tag{33}$$

where we have omitted terms that can be shown to vanish independently in the large-V limit.

6. ABSENCE OF ANOMALOUS SCALING

We now want to consider the limit in (33) more closely. We rescale u and v by setting u = yV, v = xV and obtain

$$A^2 \lim_{V \to \infty} \left[V^{3+\alpha-2\tau} \int_{v_c/V}^{1 - v_c/V} dx \int_{1-x}^{\infty} dy \; K(y,x)\, y^{-\tau} x^{1-\tau} \right] = E. \tag{34}$$

Recalling (20), we see that if $\theta \neq 0$, then the prefactor $V^{3+\alpha-2\tau}$ will either diverge (if $\theta < 0$) or go to zero (if $\theta > 0$). Let us pursue the latter case – the former case can be handled similarly. For (34) to hold, the double integral must then diverge, so that the product of the prefactor and the integral will have a finite limit. Writing V as $(1/V)^{-1}$ the limit

$$\lim_{1/V \to 0} \left[\frac{f(1/V)}{(1/V)^{3+\alpha-2\tau}} \right], \tag{35a}$$

where

$$f(1/V) = \int_{v_c/V}^{1 - v_c/V} dx \int_{1-x}^{\infty} dy \; K(y,x)\, y^{-\tau} x^{1-\tau}, \tag{35b}$$

yields an indeterminacy of the type that can be resolved by *L'Hôspital's rule* [19], i.e., we need the ratio of the derivatives with respect to 1/V of the integral (35b) and the denominator.

The derivative of the denominator is trivial. It scales as $(1/V)^{2+\alpha-2\tau}$. The derivative of the integral is

$$f'(1/V) = -v_c \left[\int_{v_c/V}^{\infty} dy \; K(y, 1 - v_c/V)\, y^{-\tau} (1 - v_c/V)^{1-\tau} + \int_{1 - v_c/V}^{\infty} dy \; K(y, v_c/V)\, y^{-\tau} (v_c/V)^{1-\tau} \right].$$

The two integrals on the right hand side converge at their upper limits because of (21b) and (23a). From the lower limits (and from the upper limit of the second integral) we get, using (21a), that the integrals scale as $(v_c/V)^{\mu-\tau+1}$.

L'Hôspital's rule, and our insistence on a finite limit, now shows that the assumption $\theta > 0$ implies the exponent relation

$$\mu - \tau + 1 = 2 + \alpha - 2\tau,$$

or, since $\alpha = \mu + \nu$,

$$\tau = \nu + 1.$$

But this contradicts (23a) according to which $\tau > \nu + 1$. A similar contradiction arises if we assume $\theta < 0$.

Thus, we conclude that the anomalous exponent θ must, in fact, vanish, and that the simple relation,

$$\tau = \frac{3+\alpha}{2}, \tag{36}$$

must hold for the steady-state, power-law solutions. The scaling function k(x) (see Sec.III) is a constant, and the final form of (17) is

$$c(v) = \left[\frac{E}{\kappa}\right]^{1/2} v^{-(3+\alpha)/2}, \tag{37a}$$

where α is the homogeneity index of the coagulation kernel, E is the mass flux (an independent parameter) and the constant κ arises from the normalization condition

$$\kappa = \int_0^1 dx \int_{1-x}^{\infty} dy \; K(x,y)\, x^{1-\tau} \; y^{-\tau}. \tag{37b}$$

Equations (37a-b) summarize our main result.

Let us also revisit (23) and (27) in light of the conclusion $\theta = 0$. These inequalities now provide necessary conditions for a power-law solution (37) to arise. Combining (36) with (23) we have

$$\tau = \frac{3+\alpha}{2} > \nu + 1,$$

or

$$\alpha - 2\nu + 1 > 0. \tag{38a}$$

Similarly from (27) and (36)

$$\alpha - 2\mu + 1 \geq 0. \tag{38b}$$

Since $\mu + \nu = \alpha$, we may also state these inequalities in the form

$$\frac{\alpha - 1}{2} \leq \nu < \frac{\alpha + 1}{2}, \tag{39a}$$

$$\frac{\alpha - 1}{2} < \mu \leq \frac{\alpha + 1}{2}. \tag{39b}$$

Any one of (38)-(39) is a necessary condition for the theory developed here to apply.

7. DISCUSSION

It will come as no surprise that at the level of Eqs.(36) and (37) our results reproduce those of Hunt [9]. Thus, for coagulation due to Brownian motion Hunt found $\tau = 3/2$ in accordance with (22a) which shows that $\alpha = 0$ for that process. Inequalities (39) are satisfied, since $\mu = -\nu = -1/3$ as noted already in Sec.IV. Note that $\tau = 3/2$ arises both for a constant kernel (see Appendix) and for the kernel (22a). Both kernels have $\alpha = 0$ but, of course, different values of μ and ν.

For coagulation in a laminar shear flow we find $\tau = 2$ since $\alpha = 1$. However, inequalities (39) are violated, albeit barely, since $\mu = 0$ and $\nu = 1$, so our theory does not apply. Coagulation due to differential sedimentation is ‘even worse’. The relation (36) gives $\tau = 13/6$ since $\alpha = 4/3$, and this spectrum was also obtained by Hunt who states that it is corroborated by observations [9]. However, inequalities (39) are now violated since $\mu = 4/3$ and $\nu = 0$. These results suggest that the present theory needs to be extended further to cover cases where an infinite flux of mass through the system is required. In reality, of course, the mass influx is always finite, but the system may be trying to approach solutions that arise analytically when E is infinite and another relation takes the place of our normalization condition (18). Thus, similarity solutions satisfying (36) where (39) are violated have been observed experimentally.

Having justified Hunt’s results, at least in part, our theory also shows that his assumptions are largely superfluous. Collisions between particles of very different sizes are, ordinarily, not to be considered improbable and they do contribute to the coagulation process. Indeed, for coagulation kernels with the properties assumed here, K(u,v), with u and v very different in size, varies as a product of powers of u and v, and both powers may be positive. ‘Locality’ of the flux is not a necessary condition for achieving self-similar coagulation spectra.

Hunt’s theory is very similar to theories of cluster-cluster aggregation as opposed to particle-particle aggregation. In other words, the assumptions of Hunt presume cluster-cluster aggregation to prevail over particle-particle aggregation. Such theories are known to provide a satisfactory description of aggregation kinetics in many cases. The present paper reveals strong reasons for this behavior.

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APPENDIX

In the body of the paper we have used the continuous formulation of SCE, Eq.(2). In this Appendix we collect various detailed results concerning the discrete SCE for the particular case of a coagulation kernel K_{mn} independent of its indices.

(i) *Smoluchowski's solution*

We set the common value of all the K_{mn} equal to 2, which simply amounts to a rescaling of time, and are thus considering the equations

$$\frac{dc_m}{dt} = \sum_{n=1}^{m-1} c_{m-n}\, c_n - 2\, c_m \sum_{n=1}^{\infty} c_n \tag{A1}$$

in the unforced (initial value problem) case. Designating the total mass at time t = 0 by

$$\sum_{n=1}^{\infty} c_n(0) = c_0, \tag{A2}$$

and introducing the generating function

$$G(z,t) = \sum_{n=1}^{\infty} c_n(t)\, z^n, \tag{A3}$$

we have the following obvious formulae:

$$G(1,t) = \sum_{n=1}^{\infty} c_n(t); \tag{A4a}$$

$$G(1,0) = c_0; \tag{A4b}$$

$$\frac{\partial G(z,t)}{\partial t} = \sum_{n=1}^{\infty} \frac{dc_n}{dt}\, z^n; \tag{A4c}$$

$$G^2(z,t) = \sum_{m=2}^{\infty} z^m \sum_{n=1}^{m-1} c_{m-n}\, c_n. \tag{A4d}$$

Thus, multiplying (A1) by z^m and summing over m produces the following PDE for G(z,t):

$$\frac{\partial G(z,t)}{\partial t} = G^2(z,t) - 2G(z,t)G(1,t). \tag{A5}$$

To solve (A5) we first note that for z = 1 it reduces to

$$\frac{dG(1,t)}{dt} = - G^2(1,t), \tag{A6}$$

which, in view of (A4b) has the solution

$$G(1,t) = \frac{c_0}{1 + c_0 t}. \tag{A7}$$

When this is substituted into (A5), we find

$$\frac{\partial G(z,t)}{\partial t} = G^2(z,t) - \frac{2c_0}{1 + c_0 t} \; G(z,t),$$

or

$$\frac{\partial}{\partial t}\left(\frac{1}{G}\right) = - 1 + \frac{2c_0}{1 + c_0 t} \; \frac{1}{G}, \tag{A8}$$

a linear differential equation that can, in turn, be solved to give

$$G(z,t) = \frac{G(z,0)}{(1 + c_0 t)[1 + (c_0 - G(z,0))t]}. \tag{A9}$$

By expanding the right hand side in powers of z, individual $c_n(t)$ may be read off as coefficients of z^n. The initial distribution is embodied in G(z,0). Otherwise the solution depends only on c_0.

Smoluchowski considered the particular case $c_1(0) = c_0$, $c_n(0) = 0$ for $n \geq 2$, for which $G(z,0) = c_0 z$. The expansion in powers of z is straightforward and the result is that

$$c_n(t) = c_0 \, \frac{(c_0 t)^{n-1}}{(1 + c_0 t)^{n+1}}. \tag{A10}$$

For large t we have $c_n(t) = 1/c_0 t^2$ for all n.

(ii) *Forced Smoluchowski kinetics*

We now consider the discrete version of Eqs.(6), viz

$$\frac{dc_1}{dt} = 0, \tag{A11a}$$

$$\frac{dc_m}{dt} = \sum_{n=1}^{m-1} c_{m-n}\, c_n - 2\, c_m \sum_{n=1}^{\infty} c_n; \quad m \geq 2. \tag{A11b}$$

We assume that (A11a) is maintained by continuous injection of monomers. Thus, we set $c_1(t) = C$, a constant, and we assume $c_n(t) = 0$ for $n \geq 2$.

Introducing the generating function G(z,t) again, defined as in (A3), we now have

$$G(1,0) = C; \tag{A12a}$$

in place of (A4b) and

$$\frac{\partial G(z,t)}{\partial t} = \sum_{n=2}^{\infty} \frac{dc_n}{dt}\, z^n; \tag{A12b}$$

in place of (A4c). In place of (A5) we now obtain

$$\frac{\partial G(z,t)}{\partial t} = G^2(z,t) - 2[G(z,t) - Cz]G(1,t). \tag{A13}$$

Setting z = 1 we again obtain an ODE for G(1,t), the counterpart of (A6):

$$\frac{dG(1,t)}{dt} = -\, G^2(1,t) + 2CG(1,t). \tag{A14}$$

The solution is

$$G(1,t) = \frac{2C}{1 + e^{-2Ct}}. \tag{A15}$$

This leads to

$$\frac{\partial G(z,t)}{\partial t} = G^2(z,t) - 2[G(z,t) - Cz]\frac{2C}{1 + e^{-2Ct}} \tag{A16}$$

in place of (A13).

Solving (A16) is somewhat tedious. We substitute $G = -W_t/W$, where the subscript indicates partial differentiation with respect to time. We also introduce a new independent variable $\xi = -\exp(2Ct)$. These substitutions produce a version of Gauss' hypergeometric equation to be solved for W:

$$\xi(\xi - 1)W_{\xi\xi} - (\xi + 1)W_{\xi} + zW = 0. \tag{A17}$$

The solution

$$W(z,t) = F(-1 + \sqrt{1-z}\,, -1 - \sqrt{1-z}\,, 1, -\exp(2Ct)) \tag{A18}$$

then needs to be differentiated to produce $G = -W_t/W$, and the result expanded in powers of z to produce the individual $c_n(t)$!

All this, however, is unnecessary since we can go directly to the steady-state equation, which at the level of the generating function simply means finding $G(z,\infty)$. In turn, this function satisfies a simple algebraic equation, obtaining by setting the time derivative in (A16) to zero and replacing the decaying exponential by 0:

$$G^2(z,\infty) - 4C[G(z,\infty) - Cz] = 0 \tag{A19}$$

with the (physical) solution

$$G(z,\infty) = 2C(1 - \sqrt{1-z}\,). \tag{A20}$$

From the binomial formula we find the steady-state values of c_n as

$$c_n = -2C\,(-1)^n\binom{\frac{1}{2}}{n} = \frac{2C}{2n-1}\,\frac{(2n)!}{(2^n\,n!)^2}\,. \tag{A21}$$

Applying Stirling's formula to the factorials in this expression we find

$$c_n \approx \frac{C}{\sqrt{\pi}}\; n^{-3/2}. \tag{A22}$$

This is the –3/2 steady-state power law solution that is mentioned several times in the body of the paper.

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