Scaling in steady-state aggregation with injection

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A mean-field approach for steady-state aggregation with injection is presented. It is shown that for a wide variety of aggregation processes the resulting steady-size distribution obeys a power law $N(m) \sim m^{-\alpha}$ with $\alpha = (3+\beta)/2$ and β the degree of homogeneity of the coagulation kernel. The general conditions for this to happen are obtained. Some applications are studied. In particular, it predicts a potential behavior for coagulation in atmospheric aerosols with exponent $\alpha \approx 2$, in agreement with observations. The theoretical results also agree with some animal group-size distributions and with numerical simulations in fractal aggregates.

I. INTRODUCTION

Power-law distributions are widely observed in natural and social systems [1]. Understanding the origin of this abundance is a question of great interest that can provide valuable information about the origins of complexity. This question has deserved the attention of many researchers during the last decades and several mechanisms have been reported to provide scaling laws such as intermittency [2], coherent noise [3], self-organized criticality [4], or multiplicative processes arising from systems with interacting units with complex internal structure [5].

The type of processes we are dealing with in this paper belongs to the general class of cluster-cluster aggregation (CCA) where clusters as well as single particles diffuse and eventually get stick when they meet. These processes are widespread in physical and biological systems such as, for instance, coagulation of colloids and aerosols [6], polymerization [7], cluster formation of galaxies [8], red-blood-cell aggregation [9], or plankton ecology [10]. Much work has been done in the study of these aggregation processes. Basically, it has been addressed to two areas: the structure of the aggregates, which in many cases are fractals [11,12] and the kinetics of aggregation, experimentally and theoretically, either by calculating the explicit time-dependent distribution functions or mostly its scaling behavior both analytically and through numerical simulations [12,13].

Although many of these works correspond to nonstationary situations, stationary ones have also been extensively considered. On one hand, steady aggregation plus breakup, the so-called reversible aggregation, has been studied by means of numerical simulations and theoretical analysis [14,15] showing that for many coagulation and fragmentation kernels, the size distribution of clusters obeys a power law. In stationary aggregation with injection and sinks, some scaling laws relating the time to reach the stationary state and the total number of clusters have been derived theoretically [16] and confirmed by numerical simulations [17]. With respect to the steady-size distribution, however, the theoretical results have focused in uniform coagulation kernels, either considering spatial correlations or by using mean-field approaches [18], and kernels of the type $b(i,j) \sim i^\mu j^\nu + i^\nu j^\mu$ [19,20]. However, most physically relevant ones are more complex such as, for instance, $b(i,j) \sim (i^{-1/3}$

$+ j^{-1/3})(i^{1/3} + j^{1/3})$ for Brownian coagulation in a continuum regime [21].

Let us stress that stationary aggregation with injection bears a great fundamental importance since if these distributions are typically scaling laws, as some particular cases have shown [18,19], we would be dealing with a very general mechanism generating power-law distributions and it should be added to the ones listed above. In fact, these processes bear some resemblance with the ones leading to self-organized criticality: in both the cases, the system is set far from equilibrium by an injection mechanism; in ours, however, the dynamics lead the system to a stable nonequilibrium state and not to an unstable one.

Stationary coagulation with injection is also interesting for practical purposes and is expected to play a relevant role in ecological and social systems. Let us mention some examples where the steady-state coagulation plus injection may apply. One of special importance is the study of atmospheric aerosols: particles are continuously generated by natural or artificial means (like smokes ejected by cars, industries etc.), coagulate and big particles leave the system through sedimentation. A long standing problem is why size-distribution measurements provide potential laws with $\alpha \approx 2$ in continental air and urban polluted air [22] and also, in the study of stirred tank reactors for aerosols, used for modeling chemical reactors in industry (in this case, large particles are allowed to flow out from the chamber). Recently, a similar steady aggregation dynamics has been successfully applied to the description of the size distribution in plankton [10], and has also been introduced in the study of animal group-size distributions [23]. The three-dimensional turbulence has also been suggested to have a deep relation to the steady state of aggregation with injection [18].

By using a mean-field approach, we find in this paper the conditions for steady aggregation with injection to yield power-law distributions for general homogeneous coagulation kernels and show that, if satisfied, the corresponding scaling exponent is $\alpha = (3+\beta)/2$ with β the homogeneity degree of the kernel. Since the critical dimension for these processes seems to be $d_c = 2$ [17,24], the results of our analysis may have a broad applicability.

The plan of the paper is as follows. In Sec. II the mean-field approach is shown, and Section III presents some applications.

II. MEAN-FIELD APPROACH

In order to evaluate the steady distribution function of an aggregation process with sources and sinks, one should, in principle, solve the Smoluchowski equation for coagulation in the steady state [25],

$$0 = a_k + \frac{1}{2} \sum_{i+j=k} b_{ij} N_i N_j - N_k \sum_j b_{kj} N_j - c_k N_k,$$

$$k = 1, 2, \ldots, \tag{1}$$

where N_k denotes the number of clusters of k particles, a_k is the injection rate of k clusters, b_{ij} the coagulation kernel, and c_k the removal probability of a cluster of size k. In the cases analytically solved, the sink term c_k often leads to an exponential decay at large k after a potential dependence at smaller ones [19,23]; it is also seen that the exponent in the potential subrange does not depend on c_k. In what follows, we will consider either that only clusters of size bigger than a critical one are eliminated or that no sink term exist. In the presence of more complicated c_k, we expect an exponential decay superimposed to the solutions found in its absence. For simplicity, we assume a monodisperse source of single particles.

Solving the Smoluchowski equation is not easy, and the attempts to do it have only succeded for particular coagulation kernels [19]. Instead, we take a different point of view and consider a continuum version of the aggregation process in which clusters grow when they meet smaller ones and ''die'' when the cluster they meet is a greater one. Both approaches should agree for large cluster sizes k. With this viewpoint, one may write the following balance equation under stationary situations:

$$\frac{d(N\dot{m})}{dm} = -N p_m \quad \text{for} \ m > m_0, \tag{2}$$

$N(m)$ being the continuous-size distribution, m_0 the mass of single particles, and $\dot{m}$ and p_m the growth and death rates due to coagulation, respectively (the generalization to non-stationary situations and the inclusion of sink and source terms is trivial). At $m = m_0$, one must add a source term in Eq. (2). The latter rates are expressed by

$$\dot{m} = \int_{m_0}^m dm_1 b(m,m_1) N(m_1) m_1, p_m$$

$$= \int_m^{m^*} dm_1 b(m,m_1) N(m_1)$$

with m^* being either the critical cluster mass over which clusters are removed or infinity if there are no sinks.

Let us consider that the coagulation kernel is a homogeneous function of degree β, i.e., $b(\lambda i, \lambda j) = \lambda^\beta b(i,j)$, as corresponds to most physical situations [19], and search for solutions of Eq. (2) of the type $N(m) = Cm^{-\alpha}$ with C a normalization factor. Straightforward calculations give

$$\dot{m} \simeq Cm^{2-\alpha+\beta} \int_0^1 dx b(1,x) x^{1-\alpha} \quad \text{for} \ m \gg m_0, \tag{3}$$

$$p_m \simeq Cm^{1-\alpha+\beta} \int_1^\infty dx b(1,x) x^{-\alpha} \quad \text{for} \ m \ll m^*. \tag{4}$$

The introduction of Eqs. (3) and (4) into Eq. (2) leads, after some direct calculations, to the following equation for α:

$$(2 - 2\alpha + \beta) \int_1^\infty dy b(1,y) y^{\alpha-\beta-3} = - \int_1^\infty dx b(1,x) x^{-\alpha}$$

whose solution is simply

$$\alpha = \frac{3+\beta}{2} \tag{5}$$

provided integral $\int_1^\infty dx b(1,x) x^{-\alpha}$ does exist. If $b(1,x)$ is a continuous function and $b(1,x) \sim x^\gamma$ for $x \to \infty$, as usually happens in physical situations, the convergence condition rewrites as $\alpha > 1 + \gamma$ or

$$\gamma < \frac{1+\beta}{2}. \tag{6}$$

Therefore, in aggregation processes with injection of single particles, if Eq. (6) satisfies, at stationary situations the size distribution is a power law with an exponent given by Eq. (5). One can confirm the validity of this result by comparison with observational data, numerical simulations, and solutions of the Smoluchowski equation.

We have performed numerical simulations to test the size range for which the scaling solution holds. In the simulations, we use cellular automata that emulate the aggregation process. Since they do not include space, they directly supply mean-field solutions. The results are obtained by adding the solutions every given time interval once the steady state has been reached. In Fig. 1, the size distributions for some aggregation processes are displayed. One observes that the results fit well with the potential laws from small to big sizes.

On the other hand, by solving the Smoluchowski equation for the kernel $b(i,j) \sim i^\mu j^\nu + i^\nu j^\mu$, Hayakawa found a power-law distribution for asymptotic large m (or k) with exponent $\alpha = (3+\mu+\nu)/2$, under condition $|\mu - \nu| < 1$ [19]. These are precisely the results predicted by Eqs. (5) and (6). Let us point out that the present method, apart from being mathematically simpler than Hayakawa's, does not need to deal separately with gelating and nongelating cases. Obviously, for the constant kernel, Eq. (5) supplies $\alpha = 3/2$, as obtained through different methods.

Finally, let us mention that Eq. (5) is also found by Cueille and Sire [20]. Curiously, in their derivation, completely different from ours, they do not include any restriction in the values of the parameters γ and β for the power solution to hold, restrictions that do appear in Hayakawa's deduction.
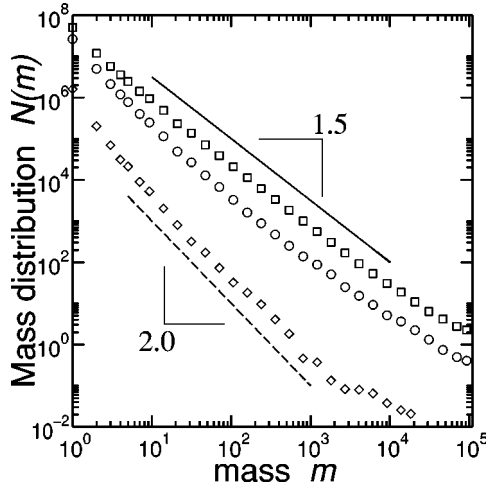
FIG. 1. Mass distribution of clusters $N(m)$ versus mass. Simulation results for some aggregation processes: Brownian coagulation, continuum regime (squares, $\alpha = 1.51$); Brownian coagulation, free-molecule regime (circles, $\alpha = 1.59$); and droplet coalescence in a convective cloud (diamonds, $\alpha = 1.99$). Straight lines are guides to the eye. One observes rather good potential laws along all the size spectrum with exponents in good agreement with expression (5)– see Table I. At very small sizes, the slope is slightly higher than the theoretical prediction and for sizes approaching the critical mass m^* over which clusters are removed, the slope decreases a bit, reaching values smaller than the theoretical ones. The latter behavior is more accused in the last two cases where the convergence condition (6) is not fulfilled. The values of the exponents given above are obtained from the regression in the interval between $m = 5$ and $m^*/10$.

III. APPLICATIONS

A. Compact aggregates

In Table I we apply our result (5) to several cases with special physical interest. Some of them are related to aerosol coagulation in the atmosphere, mainly, collection by gravitational settling and coagulation in shear flow. The first process provides $\alpha = 13/6$, very close to 2. The second one is in the limit of condition (6); however, numerical simulations show an approximately potential law with exponent 2, as predicted by expression (5). A third one, Brownian coagulation in a continuum regime, although maybe too simple to account for aggregation processes in the atmosphere since no

convection is considered, might also be included; it yields an exponent 1.5, smaller than 2 but close to some atmospheric observations. Therefore, one may conclude that the main coagulation processes in atmospheric aerosols yield potential laws with exponents $\alpha \approx 2$, in good agreement with observational evidence.

The Brownian kernel in a continuum regime may also give a plausible description of aggregation of some animals into groups. Observational data for free swimming tuna fish provide an exponent 1.49 before an exponential decay [23,27]. For sardinellas and African buffalos, the exponents found are, respectively, 0.95 and 1.15 [23], smaller than the mean-field value 1.5. The reason for this might be, as suggested by Bonabeau *et al.* that the aggregation process in these cases takes place in an effective dimension lower than two, below the critical dimension over which the mean-field theory gives right results. More work should be done to correctly predict those exponents.

Many of the processes in Table I satisfy the convergence condition but some do not (they are denoted by *). Even in these cases, numerical simulations of the aggregation process provide distributions that can be well approximated by potential laws with the exponents predicted by solution (5). For exemple, for Brownian coagulation in a free molecule regime, $\alpha \simeq 1.6$ and for $b(i,j) \sim (i+j)$, one has $\alpha \approx 2$ (see Fig. 1); both cases fall near the limit of the convergence condition. For $b(i,j) \sim (i^2 + j^2)$, however, the distribution is no longer a straight line in a log-log plot.

B. Fractal aggregates

In the cases dealt with in Table I, the aggregates are compact. One may wonder if our central result (5) is also valid if the aggregates have fractal structure. Vicsek *et al.* performed numerical simulations of diffusion-limited cluster-cluster aggregation with injection in steady-steate [17]. Curiously, although many scaling relations are discussed, nothing is said about the size-distribution plots, which clearly display scaling laws. For uniform diffusion constants, i.e., independent of size and a maximum cluster size, the exponent obtained is about 1.2 in one dimension and ≈ 1.8 in three-dimensional simulations, independently of the injection rate (Figs. 2 and 11 of Ref. [17], respectively). For constant elimination rates, the exponent is also ≈ 2 in three dimension before decaying, possibly in an exponential way.

TABLE I. Coagulation kernels for several aggregation processes and their corresponding coefficients (see text). The * denotes that the convergence condition (6) is not satisfied though numerical simulations show that the size-distribution exponent α obtained through Eq. (5) is reasonably valid.

Process [26]	$b(i,j) \sim$	β	γ	α
Brownian coagulation, continuum regime	$(i^{-1/3} + j^{-1/3})(i^{1/3} + j^{1/3})$	0	1/3	3/2
Brownian coagulation, free-molecule regime	$(i^{-1} + j^{-1})^{1/2}(i^{1/3} + j^{1/3})^2$	1/6	2/3	19/12*
Sintering-controlled catalyst aging	$(i^{2/3} + j^{2/3})$	2/3	2/3	11/6
Collection by differential gravitational settling	$i^{2/3}j^{2/3}[1 - \min(i^{2/3}j^{-2/3}, i^{-2/3}j^{2/3})]$	4/3	2/3	13/6
Coagulation in shear flow	$(i^{1/3} + j^{1/3})^3$	1	1	2*
Condensation polymerization, branched chains	$(i+2)(j+2)$	2	1	5/2
Droplet coalescence in a convective cloud	$(i+j)$	1	1	2*

In order to predict the exponent α in this case, one takes the coagulation kernel for constant diffusivity,

$$b(i,j) \sim (D_i + D_j)(R_i + R_j) \sim (i^{1/D} + j^{1/D})$$

with D the fractal dimension of the aggregates, which has been shown numerically and experimentally to be $D = 1.75$ in diffusion-limited CCA [11,12]. Then, $\beta = \gamma = 1/D$, so that Eq. (6) is satisfied and $\alpha = 1.79$, in excellent agreement with the simulations. Let us mention that our approach is also valid for reaction-limited CCA provided the appropriate kernel $b(i,j)$ is used.

IV. CONCLUSIONS

In summary, we have seen that, in steady conditions, aggregation with injection should be added to the list of generic mechanisms leading to potential size distributions. The sufficient condition for this to happen and the exponent of the scaling law have been obtained. One important application of these general analysis is that the main coagulation mechanisms in atmospheric aerosols lead to potential size distributions with exponent close to 2, in good agreement with observational data, and thus shedding some light to this long-standing question. Another interesting application is to animal group size distributions where a plausible Brownian kernel yields a potential distribution with exponent 1.5, in agreement to observations for free swimming tuna fish; it is, however, too high for some other animal groups possibly because the effective dimension of these systems are smaller than the critical one so that a mean-field description is not adequate. Finally, we have seen that our results are in good accord with simulations of coagulation of fractal aggregates.

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