Kinetics of Aggregation-Annihilation Processes

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We investigate the kinetics of many-species systems with aggregation of similar species clusters and annihilation of opposite species clusters. We find that the interplay between aggregation and annihilation leads to rich kinetic behaviors and unusual conservation laws. On the mean-field level, an exact solution for the cluster-mass distribution is obtained. Asymptotically, this solution exhibits a novel scaling form if the initial species densities are the same while in the general case of unequal densities the process approaches single species aggregation. The theoretical predictions are compared with numerical simulations in 1D, 2D, and 3D. Nontrivial growth exponents characterize the mass distribution in one dimension.

Irreversible aggregation and annihilation processes occur in many natural phenomena [1,2]. The kinetics of each of these processes is well understood both on the mean-field level which provides an accurate description when fluctuations in reactant densities can be ignored [3,1,4] and in the fluctuation-dominated regime which occurs in low-dimensional systems, see [5], and references therein. It was found in particular that the cluster-mass distribution in aggregating systems typically approaches a universal scaling form [2,4].

In this work we investigate the competition between aggregation and annihilation, which gives rise to a surprisingly rich kinetic behavior. Our model is the simplest implementation of the two underlying processes, aggregation and annihilation: Similar species clusters combine to form larger clusters while dissimilar clusters combine to form an inert substitute. It is worth noting that when the number of species n diverges, $n \to \infty$, our model is equivalent to single-species annihilation, while in the other extreme, $n = 1$, single-species aggregation is recovered. Hence, the process is well suited for investigating the interplay between aggregation and annihilation. We find that the exponents characterizing the kinetics are nonuniversal in that they may depend on the reaction rates as well as the initial conditions. Still, there is a basic scaling form which describes the process in the long-time limit.

Although our goal in this study was to understand how the competition between aggregation and annihilation could change the kinetic behavior we want to stress that our model does mimic some natural processes. For example, in a two-component system with constituent monomers A and B, aggregation of similar species can produce *open* linear polymers, $A \cdots A$ and $B \cdots B$, while *closed* linear polymers can be energetically less favorable and hence never appear. On the other hand, when dissimilar polymers meet each other a ring copolymer is formed. While linear chains continue to participate in the reaction process, ring copolymers lose their reactive edge and stop participating in the reaction process.

We shall study our model both in the mean-field limit and in the diffusion-controlled limit for $d = 1, 2$ and 3. The mean-field approach to the binary reaction process assumes that the reaction proceeds with a rate proportional to the product of the reactants densities. Thus the mean-field approximation neglects spatial correlations and therefore typically holds in dimensions larger than some critical dimension d_c. For n-species pure annihilation processes, it was suggested [6] that $d_c = 4(n-1)/(2n-3)$, the result which turns into rigorously established values [7] of the critical dimension for two- and single-species annihilation, $d_c = 4$ and 2, respectively (the latter case corresponds to $n = \infty$). For pure aggregation processes, the critical dimension depends on the details of the reaction events and on the relation between the diffusion coefficient and the mass of the cluster [8]. Numerically, we investigate the particle coalescence model (PCM) in which clusters occupy single lattice sites and perform nearest neighbor hoping with the diffusion coefficient independent of the mass. For the PCM, it is known that $d_c = 2$ [9]. Since the aggregation-annihilation model interpolates between single-species aggregation and single-species annihilation processes, both of which belong to the same universality class, it is natural to expect that the critical dimension remains the same, $d_c = 2$. We confirm the mean-field predictions above this critical dimension numerically. In one dimension, spatial correlations are relevant asymptotically. A simple heuristic argument provides the exact asymptotic behavior of the concentration and a good approximation for the growth of the typical mass.

Following the above discussion, similar species aggregation is described by the binary reaction scheme

$$A_i + A_j \to A_{i+j}, \quad B_i + B_j \to B_{i+j}, \tag{1}$$

where A_i denotes a cluster consisting of i monomers of species A, and similarly for B_i. The two-species case contains the generic many-species behavior. Therefore, we shall focus on the two-species situation and cite the many-species results if appropriate. Annihilation between dissimilar clusters reads

$$A_i + B_j \to \text{inert}. \tag{2}$$

Thus, we assume that dissimilar clusters completely annihilate independent of their masses. In some situations it may be more reasonable to assume a partial annihilation, where the monomer difference number $i-j$ is conserved [10–12].

Denote by $a_k(t)$ and $b_k(t)$ the densities of A- and B-clusters, consisting of k monomers. Then the mean-field rate equations for the two-species aggregation-annihilation process read

$$\dot{a}_k = \sum_{i+j=k} a_i a_j - 2a_k(a+b),$$
$$\dot{b}_k = \sum_{i+j=k} b_i b_j - 2b_k(a+b). \tag{3}$$

Here $a(t)$ and $b(t)$ denote the total densities of A- and B-clusters, $a=\sum_{k\geq 1} a_k$ and $b=\sum_{k\geq 1} b_k$, and the overdot denotes the time derivative. In writing Eq. (3) we have assumed that the aggregation rate equals the annihilation rate. For convenience, we set this rate to 2 such that the prefactor of the gain term equals unity.

Summing up Eq. (3) we obtain

$$\dot{a} = -a^2 - 2ab, \quad \dot{b} = -b^2 - 2ab. \tag{4}$$

For the symmetric initial conditions, $a(0)=b(0)$, we get $\dot{a}=-3a^2$ which is immediately solved to find

$$a(t) = b(t) = \frac{a(0)}{1+3a(0)t}. \tag{5}$$

For the asymmetric initial conditions, $a(0) > b(0)$, it is helpful to rewrite Eq. (4) in terms of $u \equiv (a+b)/(a-b)$ and $v \equiv a-b$. This gives

$$\dot{u} = -\frac{1}{2}v(u^2-1), \quad \dot{v} = -uv^2. \tag{6}$$

Expressing v as a function of u yields $dv/du = 2uv/(u^2-1)$, and as a result

$$v = K(u^2-1), \quad K = \frac{[a(0)-b(0)]^3}{4a(0)b(0)}. \tag{7}$$

Inserting Eq. (7) into Eq. (6), we obtain a closed equation for $u \equiv u(t)$ with the solution

$$\frac{2u}{u^2-1} - \frac{2u(0)}{u^2(0)-1} + \ln\left[\frac{u-1}{u+1}\,\frac{u(0)+1}{u(0)-1}\right] = 2Kt. \tag{8}$$

The densities $a(t)$ and $b(t)$ of the total number of A- and B-clusters, $a=v(u+1)/2$ and $b=v(u-1)/2$, cannot be expressed as explicit functions of t. However, asymptotically they exhibit simple power-law behaviors

$$a \simeq t^{-1}, \quad b \simeq a(0)b(0)[a(0)-b(0)]^{-3}t^{-2}, \tag{9}$$

for $t \to \infty$. This indicates that the majority and the minority species evolve very differently. Similarly, in the many-species case the total density of the majority species decays as t^{-1} while the minorities densities decay as t^{-2}.

We turn now to the determination of the cluster densities. Introducing the generating functions,

$$A(z,t) = \sum_{j=1}^{\infty} z^j a_j(t), \quad B(z,t) = \sum_{j=1}^{\infty} z^j b_j(t), \tag{10}$$

transforms the governing equations into

$$\dot{A} = A^2 - 2A(a+b), \quad \dot{B} = B^2 - 2B(a+b). \tag{11}$$

These Bernoulli equations are straightforwardly solved to get

$$A(z,t) = \frac{A_0(z)E(t)}{1-A_0(z)\int_0^t dt' E(t')},$$
$$B(z,t) = \frac{B_0(z)E(t)}{1-B_0(z)\int_0^t dt' E(t')}, \tag{12}$$

with the shorthand notations $A_0(z) \equiv A(z,t=0)$, $B_0(z) \equiv B(z,t=0)$, and $E(t) \equiv \exp\left(-2\int_0^t dt'[a(t')+b(t')]\right)$.

Eq. (12) represents the general solution for arbitrary initial conditions. Consider now the simplest but important case of monodisperse, generally asymmetric, initial conditions

$$a_k(0) = \delta_{k1}, \quad b_k(0) = \lambda\delta_{k1}. \tag{13}$$

These initial conditions imply $A_0(z)=z, B_0(z)=\lambda z$. Expansion of the resulting generating functions $A(z,t) = zE(t)[1-z\int_0^t dt' E(t')]^{-1}$ and $B(z,t) = \lambda z E(t)[1 - z\lambda\int_0^t dt' E(t')]^{-1}$ yields

$$a_k(t) = E(t)\left[\int_0^t dt' E(t')\right]^{k-1},$$
$$b_k(t) = \lambda^k E(t)\left[\int_0^t dt' E(t')\right]^{k-1}. \tag{14}$$

In the symmetric case $\lambda=1$, we find $E(t)=(1+3t)^{-4/3}$ and

$$a_k(t) = b_k(t) = (1+3t)^{-4/3}\left[1-(1+3t)^{-1/3}\right]^{k-1}. \tag{15}$$

Asymptotically, the cluster-mass distribution approaches the scaling form

$$a_k(t) \sim t^{-4/3}\exp(-x), \quad x \sim kt^{-1/3}. \tag{16}$$

The total mass densities, $m_a(t) = \sum_{k\geq 1} k a_k(t)$ and $m_b(t) = \sum_{k\geq 1} k b_k(t)$, decrease with time, $m = m_a = m_b \sim t^{-2/3}$. However, there exists the quantity $I_1 \equiv ma^{-2/3}$ which is conserved by the dynamics of the aggregation-annihilation model subject to any *symmetric* initial conditions (not necessarily the monodisperse one). The conservation of I_1 is verified by a direct computation which makes use of the evolution equations,

$\dot{a} = -3a^2$ and $\dot{m} = -2am$. For pure aggregation processes, the total mass is conserved. Thus the quantity I_1 plays the role of a "mass" in the aggregation-annihilation model. Furthermore, the rate equations (3) actually admit an infinite set of conservation laws which include higher moments, $M_p(t) = \sum_{k\geq 1} k^p a_k(t)$, of the distribution function $a_k(t)$. (The first two moments are the number and mass densities, $M_0 \equiv a$ and $M_1 \equiv m$). These conservation laws are found consequently and read $I_2 = (M_2 - 2m^2/a)a^{-2/3}$, $I_3 = (M_3 - 6M_2 m/a + 6m^2/a^2)a^{-2/3}$, *etc.* We have constructed these integrals I_j recursively. Unfortunately, we understand neither the physical meaning nor the algebraic structure of these integrals.

In the asymmetric case we shall set $\lambda < 1$ thus forcing B-species to be a minority. Making use of Eq. (3) one can express $E(t) = \exp\left(-2\int_0^t dt'[a(t') + b(t')]\right)$ as an explicit function of a and b, $E = (a(0)^{-1} - b(0)^{-1})/(a^{-1} - b^{-1})$. In the long-time limit we use the asymptotic values, $a \simeq t^{-1}$ and $b \simeq \lambda(1-\lambda)^{-3}t^{-2}$, to compute $E(t) \simeq \lambda^{-1}(1-\lambda)b \simeq (1-\lambda)^{-2}t^{-2}$. Therefore,

$$a_k(t) \simeq (1-\lambda)^{-2}t^{-2}\exp(-y),$$
$$b_k(t) = \lambda^k a_k(t), \qquad y = \frac{k}{(1-\lambda)^2 t}. \tag{17}$$

Thus, for arbitrary initial conditions the majority species cluster-mass distribution can be written in the scaling form

$$a_k(t) \sim t^{-w}\Phi[k/S(t)], \quad S(t) \sim t^z, \tag{18}$$

where $S(t)$ is the characteristic mass. The scaling function is exponential, $\Phi(x) = \exp(-x)$, both for symmetric and asymmetric initial conditions. In the symmetric case, the governing exponents are $w = 4/3$ and $z = 1/3$. The asymmetric case is equivalent to single-species aggregation, and the exponents are $w = 2$ and $z = 1$. The minority, on the other hand, does not scale according to the usual definition although it can be expressed in the modified scaling form, $b_k(t) \sim \lambda^k t^{-w}\Phi[k/S(t)]$. The modified scaling form also indicates that two different mass scales are associated with the minority species. A growing scale $S(t) \sim t$, which is forced by the majority species, and a time independent scale $S_\lambda = 1/(1-\lambda)$ which dominates in the long-time limit. The latter scale diverges, $S_\lambda \cong 1/(1-\lambda)$, in the limit $\lambda \to 1$. It is also instructive to compute the total mass densities. By summing Eq. (17) we find that as $t \to \infty$,

$$m_a(t) \to (1-\lambda)^2, \quad m_b(t) \to \lambda(1-\lambda)^{-4}t^{-2}. \tag{19}$$

Thus the final mass difference $\Delta m_\infty \equiv m_a(\infty) - m_b(\infty) = m_a(\infty)$ may be expressed through the initial mass difference $\Delta m_0 \equiv m_a(0) - m_b(0) = 1 - \lambda$ via a surprisingly simple relation, $\Delta m_\infty = (\Delta m_0)^2$. In comparison, for the pure annihilation process, the final mass density is significantly larger, $\Delta m_\infty = \Delta m_0$. More generally, the concentration difference, $c_- = a - b$, is a conserved variable in the pure annihilation process as it is obvious physically and can also be seen from the evolution equations $\dot{a} = \dot{b} = -2ab$. In the aggregation-annihilation process a related "hidden" conservation law exists. From the rate equations, one can verify that the quantity $J_1 = (a-b)(ab)^{-1/3}$ is conserved. This unusual conservation law is trivially satisfied in the case of equal initial densities. There exists another hidden conservation law, $m_a m_b (ab)^{-2/3} =$ const. In the case of symmetric initial conditions this conservation law is transformed into the aforementioned aggregation-like conservation law $I_1 = m_a a^{-2/3} =$ const. Additionally, there is an infinite set of other integrals which generalize the integrals I_2, I_3, *etc.* found for symmetric initial conditions, to arbitrary initial conditions. We conclude that the aggregation-annihilation process exhibits nontrivial conservation laws which are generalization to the usual conservation laws that underly annihilation and aggregation separately. Although the existence of an infinite amount of hidden conservation laws is not customary for irreversible processes, it has been recently found in several such processes [13].

We turn now to the general n-species model. Denote by a^i (m^i) the concentration (mass) of the $i^{\rm th}$ species. One can verify that the quantities $J^{ij} = N(a^i - a^j)/a^i a^j$, with $N = (\prod_{1\leq k\leq n} a^k)^{2/(2n-1)}$, are conserved for every $i \neq j$ independent of the initial conditions. These integrals are reminiscent of the integrals $L^{ij} = (\prod_{1\leq k\leq n} a^k)^{1/(n-1)}(a^i - a^j)/a^i a^j$ which are the conservation laws for the pure annihilation process [6]. In both cases, there are $n-1$ independent conservation laws. The aggregation-type conservation laws can also be generalized to the n-specie case. The quantity M/N is conserved, with $M = (\prod_{1\leq k\leq n} m^k)^{1/(n-1)}$. In the case of the symmetric initial conditions, one recovers the previously established integral $I_1 = m^i(a^i)^{-(2n-2)/(2n-1)}$. Similarly to the first conserved quantity, one can generalize the I_j integrals and it appears that there is again an infinite set of conservation laws.

For asymmetric initial conditions the majority species scales as in single-species aggregation, while the minorities scale only in a modified sense. For the symmetric initial conditions, the cluster-mass distribution approaches the scaling form of Eq. (18) with

$$w_n = \frac{2n}{2n-1}, \qquad \text{and} \qquad z_n = \frac{1}{2n-1}. \tag{20}$$

Thus the exponents depend on the number of species n. When $n = 1$, single-species aggregation is recovered, $w_1 = 2$ and $z_1 = 1$, while the limit $n \to \infty$ corresponds to single-species annihilation, $w = 1$ and $z = 0$.

Generally, we expect that the exponents characterizing the scaling behavior are universal, *i. e.*, they depend only on important aspects of the kinetics. The fact that the exponents do depend on the number of species does not contradict universality. However, the exponents in the aggregation-annihilation system may depend also on the reaction rates. To demonstrate that we consider the general case where the aggregation rate and the annihilation rate are different. We set the aggregation rate to unity as previously and denote the annihilation rate by J. In the n-species case with symmetric monodisperse initial conditions we have

$$\dot{a}_k = \sum_{i+j=k} a_i a_j - 2a_k[1+(n-1)J]a, \qquad (21)$$

with $a_k(t=0)=\delta_{k1}$. By employing the above technique we solve Eq. (21) and find

$$a_k(t) = (1+\nu t)^{-1-1/\nu}\left[1-(1+\nu t)^{-1/\nu}\right]^{k-1}, \qquad (22)$$

where the notation $\nu = 1+2(n-1)J$ has been used. The cluster mass distribution obeys the scaling form of Eq. (18) with the exponents

$$w_n = 2\frac{1+(n-1)J}{1+2(n-1)J}, \quad z_n = \frac{1}{1+2(n-1)J}. \qquad (23)$$

Interestingly, both exponents dependent continuously on the (relative) magnitude of the annihilation rate, J. This nonuniversality contrasts the bulk of previously investigated aggregation models [4] (see, however, [14,10]). Varying the rate ratio represents another way of interpolating between aggregation and annihilation. Indeed, independent of n, when $J=0$, single-species aggregation is recovered, while the limit $J\to\infty$ corresponds to single-species annihilation.

We now consider aggregation-annihilation in the diffusion-controlled limit in low dimensions. We shall study the particle coalescence model (PCM) in which clusters occupy single lattice sites, hop to nearest neighbor cites with a rate independent of the mass, and aggregate or annihilate instantaneously whenever they meet. The PCM allows one to focus on the kinetic aspects of the process and is well suited for numerical implementation. Additionally, in the mean-field approximation the PCM is governed by the same Eq. (3) which has been examined previously.

Let us ignore the mass and the identity of the clusters and denote an arbitrary cluster by C. Then the reduced reaction process can be described by the reaction scheme $C+C\to C$ and $C+C\to 0$ for aggregation and annihilation, respectively. For both processes, the total density $c=\sum_{k\geq 1} c_k$ is inversely proportional to $N(t)$, the number of *distinct* sites visited by a single random walk in d dimensions [5]. $N(t)$ is well known in probability theory [15] and thus the density c, $c\sim N^{-1}$, behaves as

$$c(t)\sim\begin{cases} t^{-1/2} & d=1;\\ t^{-1}\log(t) & d=2;\\ t^{-1} & d>2.\end{cases} \qquad (24)$$

In other words, the time variable t is replaced with a modified time variable $N(t)$. We further assume that the rate equation theory describes the process in low dimensions, with the time variable $N(t)$. Although this approach is a heuristic one, it provides a good approximation for the subcritical behavior. For the symmetric initial conditions, the characteristic mass for the case $n=2$ is given by

$$S(t)\sim\begin{cases} t^{1/6} & d=1;\\ t^{1/3}\left(\log(t)\right)^{-1/3} & d=2;\\ t^{1/3} & d>2.\end{cases} \qquad (25)$$

Using the asymptotic forms of the concentration and the typical mass, a scaling form similar to the one of Eq. (18) can be written. The same analysis can be repeated for the general multi-species case, and we quote the scaling exponents in one dimension only,

$$w_n = \frac{n}{2n-1}, \qquad \text{and} \qquad z_n = \frac{1}{2(2n-1)}. \qquad (26)$$

In the extreme cases of $n=1$ and $n\to\infty$, these exponents agree with the solutions to single-species aggregation and single-species annihilation, respectively [16]. In the case of asymmetric initial conditions, the majority species concentration decays according to single-species aggregation, or equivalently, according to Eq. (24). The minority species, on the other hand, decays much faster. Since the total concentration of the minority species decays as the monomer density of the majority species, $b\sim a_1$, and the latter is known in one and two dimensions (see [16,7]), we find that the minority species concentration decays as $t^{-3/2}$ in 1D, and as $t^{-2}\log^2(t)$ in 2D.

It is interesting to compare the above theoretical predictions with numerical simulations. We have performed simulations for $d=1,2$, and 3. The numerical implementation of the process is simple. Initially all L^d sites on the cubic lattice are occupied with monomers. An elemental simulation step consists of picking a cluster at random and moving it to a randomly chosen neighboring site. If the site is occupied, an aggregation or an annihilation event takes place, depending on the identity of the two clusters. Time is updated by the inverse of the total number of particles in the system after each step. The linear dimension of the lattice used in the simulation was $L=3\times 10^7$, 4×10^3, 2×10^2 in 1D, 2D, and 3D.

We discuss first results for the symmetric initial conditions involving two species. To verify that the process belongs to the PCM universality class we measured the density of particles as a function of time. Indeed, Eq. (24) appears to hold asymptotically (see Figure 1). We also measured the average cluster size $\langle k(t)\rangle = \sum kc_k/\sum c_k$. We expect that the average cluster size is proportional to the characteristic size $S(t)$, asymptotically. For $d=3$, the mean-field prediction, $\langle k(t)\rangle\sim t^{1/3}$ is verified (see Figure 2). Note that the simulation results are reliable up to time $\propto L^2$ due to finite size effects. At the critical dimension, $d_c=2$, the average mass grows slightly slower, consistent with the logarithmic correction of Eq. (25). In 1D, the growth exponent z as determined by a least square fit is $z = 0.190 \pm 0.002$. However, the corresponding value from the approximate theory is lower $z = 1/6$. The simulation was carried over a relatively large temporal range, suggesting that the growth exponent is indeed different than suggested by the heuristic theory. A similar trend is observed when more than two species are involved. We find that the numerically determined exponents $z_3 \cong 0.12$ and $z_4 \cong 0.09$ are slightly higher than their theoretical counterparts $z_3 = 1/10$ and $z_4 = 1/14$. We have made some additional consistency checks. For example, we verified that the cluster mass distribution follows the scaling form of Eq. (18). Above the critical dimension the scaling function is indeed a simple exponential, in agreement with the rate equation predictions. Also, we verified numerically that both the aggregation-type and the annihilation-type conservation laws are satisfied by the PCM above the critical dimensions $d_c = 2$. This suggests that existense of hidden conserved quantities is an intrinsic property of the model.

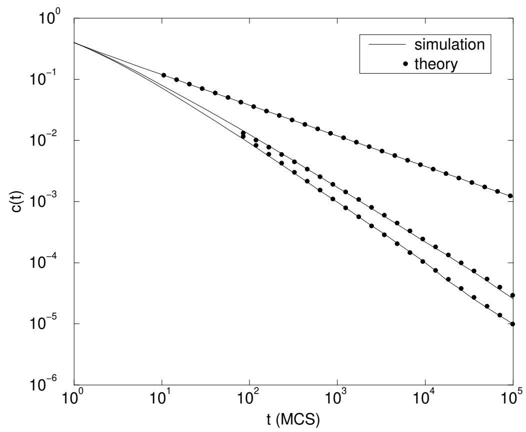

FIG. 1. The total density versus time in 1D (top), 2D (middle) and 3D (bottom). Shown are the simulation data (bullets) and the theoretical prediction of Eq. (24) (solid line). Time is given in terms of Monte-Carlo Steps (MCS).

In summary, we introduced an aggregation-annihilation model and presented the solution to the governing rate equations. The mass distribution follows a general scaling form with nontrivial growth exponents, and the process is characterized by unusual conservation laws. The kinetic behavior depends both on the number of species and on the specific rates of the annihilation and the aggregation processes. Numerical simulations confirm the rate equation predictions above the critical dimension. Below the critical dimension, a heuristic argument provides a good estimate for the mass distribution. Nontrivial exponents describe the process in one dimension and it would be interesting to study this system rigorously. It is plausible that further investigation of the underlying conservation laws below the critical dimension will pave the way for understanding the long time kinetics.

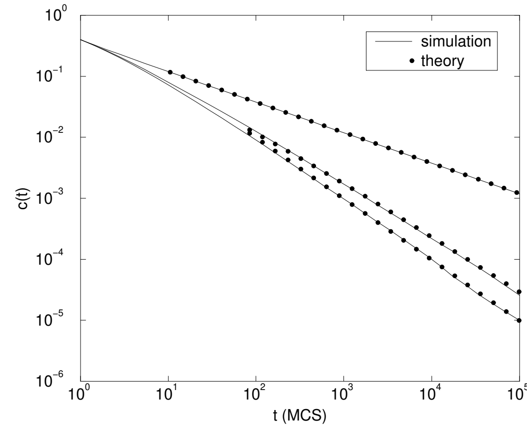

FIG. 2. The average cluster size $\langle k(t) \rangle$ versus t in 1D (bottom), 2D (middle), and 3D (top). Shown are simulation data (bullets) and lines of slope 0.19 (solid line) and 1/3 (dashed line) for reference.

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