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Catalysis-driven aggregate growth

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Abstract

We propose two-species catalysis-driven aggregation models in which coagulation of one species occurs only in the presence of another species (the catalyst). By means of generalized Smoluchovski rate equations, we study the kinetics of the system with the rate kernel $K_A(i; j; l) \propto l^{\upsilon}$, at which two *A* clusters of size *i* and *j* bond together under the catalytic action of a *B* cluster of size *l*. The results show that the cluster mass distribution of species *A* obeys a conventional scaling law in the $\upsilon \ge 0$ case while it may satisfy the modified scaling form in other cases. Moreover, it is found that the scaling exponents are nonuniversal and dependent on the value of index υ in most cases. On the other hand, we also investigate the scaling behaviour of the mutually catalysis-driven aggregate growth. For the system with the rate kernel $K_A(i; j; l) \propto l^{\upsilon_1}$ and $K_B(i; j; l) \propto l^{\upsilon_2}$, its kinetics depends crucially on the values of the indices υ_1 and υ_2 . Either species scales according to the conventional or modified form in most cases; while the system may undergo a gelation transition in some special cases.

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1. Introduction

The understanding of aggregation phenomena is of considerable importance in many basic and applied problems such as atmospheric aerosols [1], star formation [2] and gelation [3]. In the last few decades, the kinetics of cluster growth through irreversible aggregation has been extensively studied by many sophisticated models [4–10]. It was found that the mass distribution of the aggregates may approach a scaling form in the long-time limit [5, 6]. Most of these research works focused attention on the binary self-coalescence scheme, $A_i + A_j \rightarrow A_{i+j}$, where A_i denotes a cluster consisting of *i* monomers. That is, the clusters A_i and A_j can bond spontaneously and form a larger cluster A_{i+j} . Moreover, Ispolatov *et al* [11]

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and Leyvraz et al [12] proposed a migration-driven growth mechanism which can be described by the scheme $A_i + A_j \rightarrow A_{i-1} + A_{j+1}$ $(i \leq j)$. This different mechanism can provide a natural description for the evolution of city populations, and the results showed that this mechanism gives rise to kinetic behaviour quite different from conventional aggregation processes [11–13]. In addition, there also exist some other classes of growth mechanisms in physics and in social science. For example, in a variety of situations, some species are chemically stable and nonreactive under normal physical conditions, and they may enter into reaction only in the presence of a catalytic substance. Burlatsky et al [14] and Oshanin et al [15] proposed a three-molecule catalytically activated reaction model (CARM), in which the elementary reaction step is $A + B + C \rightarrow$ product (here A and B represent two different types of stable species and C is a catalytic substance), and they incisively analysed its kinetics by extending the Smoluchowski approach. Recently, many developments have made the catalytic reaction mechanism well understood [16, 17]. Motivated by these works, we proposed two distinct catalysis-driven aggregation models with constant rate kernels [18, 19]. The results exhibited that the long-time evolution of the system also obeys a scaling law, but which is somewhat different from the above-mentioned conventional aggregation. All these different mechanisms unfold very rich kinetic behaviour of the aggregation systems.

In this work, we thoroughly investigate the kinetics of the catalysis-driven aggregation models with a general size-dependent rate kernel. There are two types of species in our system, species A and B. Species A is chemically stable and the clusters of this type cannot coagulate by themselves; however, two A clusters can merge with the help of a B cluster. Thus the system evolves according to the catalytic reaction scheme as follows. The first elementary reaction step is a reversible reaction between a pair of A and B clusters, $A_i + B_l \Rightarrow A_i B_l$, and the second step is an irreversible reaction only between the A and AB clusters, $A_i B_l + A_i \rightarrow A_{i+1} + B_l$. It is well known that the reversible reaction may reach its steady state at a very rapid rate. Thus, the catalytic reaction is controlled by the second step. The whole catalytic reaction then reads $A_i + A_j + B_l \xrightarrow{K_A(i;j;l)} A_{i+j} + B_l$, with a size-dependent reaction rate kernel $K_A(i;j;l)$. On the other hand, there also exist some special situations in which both species are non-reactive but one species is the catalyst for the aggregation reaction of another one [19]. Then the catalytic reaction for species A is $A_i + A_j + B_l \xrightarrow{K_A(i;j;l)} A_{i+j} + B_l$, while that for species B is $B_i + B_j + A_l \xrightarrow{K_B(i;j;l)} B_{i+j} + A_l$. In this paper, we shall investigate the dependence of the kinetics of the aggregation system on the catalysis. The catalytically activated processes can contribute to produce required products from the species which is chemically stable under normal conditions and are thus of great practical and theoretical significance.

We shall investigate our models in the mean-field limit. The mean-field assumption neglects the fluctuations in the densities of the reactants and, therefore, applies to the case in which the space dimension d of the system is equal to or greater than a critical dimension d_c . For the irreversible aggregation system, it is found that $d_c = 2$ [6, 7]. In our models, the controlling reaction is also an irreversible binary aggregation-like reaction. Thus, it is reasonable that the critical dimension d_c of our system may also be equal to 2. Based on the mean-field theory, we have derived the asymptotic solutions for the cluster mass distributions in these catalysis-driven aggregation processes. The results show that the cluster mass distribution of the system may satisfy a conventional or modified scaling law, which depends strongly on the details of the rate kernel. As for the $d < d_c$ case, the particle coalescence model (PCM) (see, e.g., [7, 20]) may be used to study the catalytically activated aggregate growth. Since the clusters in the PCM are defined to be single lattice sites and the aggregation reaction occurs only when two or more clusters occupy the same lattice site, the PCM may be considered as a model of aggregate growth through the 'catalytic action' of lattice sites.

The paper is organized as follows. In section 2, we describe a catalysis-driven aggregation model in which the irreversible growth of one species is driven by another species and investigate the corresponding mean-field rate equations to obtain the cluster mass distributions. We then study the kinetic behaviour of a two-species mutually catalysis-driven aggregation model in section 3. A brief summary is given in section 4.

2. The model of irreversible one-species growth driven by the catalyst

We first investigate the catalysis-driven aggregation processes in which A clusters coagulate with the help of the catalyst B. The theoretical approach to the aggregation processes is based on the mean-field rate equation, which assumes that the reaction proceeds at a rate proportional to the reactant concentrations. The concentrations of A and B clusters of k-mers are denoted as a_k and b_k , respectively. As we aim to investigate the dependence of the kinetics of our system on the catalyst, we assume that the rate kernel depends only on the details of the catalyst. For the cases in which both species A and B are polymers and bonding between any two polymers takes place only at their surfaces, one may consider $K_A(i; j; l) = s_l$, where s_l represents the effective catalytic surface area of a B cluster of size l. For large $l, s_l \sim l^{\upsilon}$ (here υ denotes a geometric index characterizing the catalytic surface of an *l*-mer, e.g., for compact clusters v = 2/3 [21], and it is found that $0 \le v \le 1$. For some other situations, coagulation of the clusters does not merely depend on their geometric surfaces and thus the rate exponent v may not have the above-mentioned constraint. For example, in economics and social science, the overall rate of aggregation between two generalized 'clusters'(e.g., population centres) may also vary as a power law in some scale factor (see [12] and references therein). In order to obtain the explicit solutions of the mean-field rate equations and analyse the scaling properties of the catalysis-driven aggregate growth, we consider here a simple kernel $K_A(i; j; l) = I_1 l^{\upsilon}$ (v is a constant) for all *l*. On the other hand, we assume that the catalyst may coagulate by itself, $B_i + B_j \xrightarrow{J(i;j)} B_{i+j}$, and the self-coagulation rate kernel J(i, j) is set to a constant I_2 . It is obvious that for the v > 1 case, self-coagulation of the catalyst may improve its catalytic ability. Then the mean-field rate equations for the catalysis-driven aggregation processes read

$$\frac{\mathrm{d}a_k}{\mathrm{d}t} = \frac{I_1}{2} \sum_{i+j=k} \sum_{l=1}^{\infty} (l^{\upsilon} a_i a_j b_l) - I_1 a_k \sum_{j=1}^{\infty} \sum_{l=1}^{\infty} (l^{\upsilon} a_j b_l)$$

$$\frac{\mathrm{d}b_k}{\mathrm{d}t} = \frac{I_2}{2} \sum_{i+j=k} b_i b_j - I_2 b_k \sum_{i=1}^{\infty} b_j.$$
(1)

We consider the simplest but important case in which there only exist monomer clusters of species A and B at t = 0 and their concentrations are equal to A_0 and B_0 , respectively. Then the initial condition is

$$a_k(0) = A_0 \delta_{k1}$$
 $b_k(0) = B_0 \delta_{k1}.$ (2)

The rate equations (1) can be solved with the help of ansatz [22]

$$a_k(t) = A(t)[a(t)]^{k-1}$$
 $b_k(t) = B(t)[b(t)]^{k-1}$. (3)

Substituting equations (3) into equations (1), we can transform the rate equations (1) into the following differential equations:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{I_1 A M_{B\nu}}{2} \qquad \frac{\mathrm{d}A}{\mathrm{d}t} = -\frac{I_1 A^2 M_{B\nu}}{1-a} \tag{4}$$

$$\frac{\mathrm{d}b}{\mathrm{d}t} = \frac{I_2 B}{2} \qquad \frac{\mathrm{d}B}{\mathrm{d}t} = -\frac{I_2 B^2}{1-b} \tag{5}$$

with the shorthand notation $M_{B\nu}(t) = \sum_{l=1}^{\infty} l^{\nu} b_l(t)$. Correspondingly, the initial condition for equations (4) and (5) becomes

$$a = 0$$
 $b = 0$ $A = A_0$ $B = B_0$ at $t = 0$. (6)

From equations (4) and (5), one can easily find that $M_{A1}(t) = \sum_{k=1}^{\infty} ka_k(t) = A/(1-a)^2 \equiv A_0$ and $M_{B1}(t) = \sum_{k=1}^{\infty} kb_k(t) = B/(1-b)^2 \equiv B_0$. Hence, both species A and B obey the mass observation law. It is natural for our model without any consumption of the reactants.

Firstly, we discuss the evolution behaviour of the catalyst. From equations (5) one can readily determine the exact solutions of b(t) and B(t) as follows:

$$b(t) = \frac{I_2 B_0 t}{I_2 B_0 t + 2} \qquad B(t) = \frac{4B_0}{(I_2 B_0 t + 2)^2}.$$
(7)

Thus we obtain the exact solution $b_k(t)$ for species B,

$$b_k(t) = \frac{4B_0}{(I_2 B_0 t + 2)^2} \left(1 - \frac{2}{I_2 B_0 t + 2} \right)^{k-1}.$$
(8)

Approximately, equation (8) can be rewritten as

$$b_k(t) \simeq 4I_2^{-2}B_0^{-1}t^{-2}\exp(-y) \qquad y = (2/I_2B_0)kt^{-1}$$
 (9)

which is valid in the region of $k \gg 1$ and $t \gg 1$. Equation (9) indicates that the cluster mass distribution of species *B* approaches the scaling form [22]

$$c_k(t) \simeq t^{-w} \Phi[k/S(t)] \qquad S(t) \propto t^z \tag{10}$$

where $c_k(t)$ denotes the concentration of k-mers and S(t) is the characteristic mass of such an aggregation system. The scaling function for species B is exponential, $\Phi(x) = \exp(-x)$; moreover, the exponents are universal, w = 2 and z = 1. So, species B evolves as in the single-species aggregation case, which indeed follows the way that our model is formulated.

We then turn to discuss the mass distribution of A clusters. From equations (4) we obtain

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{I_1 A_0}{2} (1-a)^2 M_{B\nu}.$$
(11)

In the long-time limit, we determine the moments of species B in several cases with different index v,

$$M_{B\nu}(t) = \sum_{l=1}^{\infty} l^{\nu} b_l(t) \simeq \begin{cases} \Gamma(1+\nu) B_0^{\nu} \left(\frac{2B_0}{I_2 B_0 t + 2}\right)^{1-\nu} & \text{for } \nu > -1 \\ \frac{4\ln(I_2 B_0 t/2 + 1)}{I_2 t (I_2 B_0 t + 2)} & \text{for } \nu = -1 \\ \frac{4B_0}{(I_2 B_0 t + 2)^2} \sum_{j=1}^{\infty} j^{\nu} & \text{for } \nu < -1. \end{cases}$$
(12)

We then investigate the evolution behaviour of A clusters in several different cases.

In the first case of v > 0, one can recast equation (11) into the following equation in the long-time limit:

$$\frac{\mathrm{d}a}{\mathrm{d}t} \simeq \frac{I_1 \Gamma(1+\upsilon) A_0 B_0^{\upsilon}}{2} \left(\frac{2}{I_2}\right)^{1-\upsilon} (1-a)^2 t^{\upsilon-1}.$$
(13)

Equation (13) can be directly solved to yield

$$a(t) \simeq 1 - C_1 t^{-\upsilon}$$
 (14)

where $C_1 = [2/I_1 A_0 B_0^{\upsilon} \Gamma(\upsilon)] (I_2/2)^{1-\upsilon}$. Thus we obtain the asymptotic scaling solution of $a_k(t)$ at large times,

$$a_k(t) \simeq A_0 C_1^2 t^{-2\nu} \exp(-x) \qquad x = C_1 k t^{-\nu}.$$
 (15)

Equation (15) indicates that for this case, the cluster mass distribution of species A approaches the conventional scaling form of equation (10) with the typical mass $S(t) \propto t^{v}$. The exponents are nonuniversal and dependent on the value of index v, i.e., w = 2v and z = v. The results also imply that with the help of *B* catalyst all the initial monomers of species A will coagulate each other completely.

In the second case of v = 0, from equation (13) one can derive the following asymptotic solution:

$$a(t) \simeq 1 - C_2 (\ln t)^{-1} \tag{16}$$

where $C_2 = I_2/I_1A_0$. The cluster mass distribution of species A is then obtained as follows:

$$a_k(t) \simeq A_0 C_2^2 (\ln t)^{-2} \exp(-x) \qquad x = C_2 k (\ln t)^{-1}.$$
 (17)

This shows that the evolution behaviour of A clusters obeys a logarithm-correction scaling form, $a_k(t) \simeq (\ln t)^{-w} \Phi\{k/S[\ln(t)]\}$ with $S(t) \propto t^z$ (also see [18]). In this case, the coagulation of all the monomers of species A can also be accomplished finally.

In the third case of 0 > v > -1, we find

$$a(t) \simeq C_3 - C_4 t^{\nu} \tag{18}$$

where $C_3 = 1 - \{1 + 2I_1A_0B_0\int_0^\infty dt (I_2B_0t + 2)^{-2}\sum_{j=1}^\infty j^{\upsilon}[I_2B_0t/(I_2B_0t + 2)]^{j-1}\}^{-1}$ and $C_4 = -(1 - C_3)^2/C_1$. Thus we obtain the asymptotic scaling solution

$$a_k(t) \simeq A_0(1-C_3)^2 C_3^k \exp(-x) \qquad x = (C_4/C_3)kt^{\nu}.$$
 (19)

Equation (19) indicates that species A does not scale according to the conventional definition (10) and the cluster mass distribution satisfies the modified scaling form $a_k(t) \simeq h^k t^{-w} \Phi[k/S(t)]$ (here h is a constant and 1 > h > 0). In this case, the exponents are w = 0 and z = -v. The modified scaling form also indicates that two different mass scales are associated with species A. One is a growing scale $S(t) \propto t^{-v}$, which is forced by the catalyst. Another is a time-independent scale $S = 1/(1-C_3)$, which dominates the evolution behaviour of species A in the long-time limit. Moreover, it can be concluded from equation (19) that all A_k clusters (k = 1, 2, ...) can survive at the end. Hence, no matter how large the initial concentration of the catalyst is, the catalyst cannot make all monomers of species A bond together at the end.

In the fourth case of v = -1, one can recast equation (11) as

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{2I_1 A_0 (1-a)^2}{I_2 t (I_2 B_0 t + 2)} \ln(I_2 B_0 t/2 + 1). \tag{20}$$

Equation (20) can be solved exactly. In the long-time limit, we obtain

$$a(t) \simeq C_5 - C_6 t^{-1} \ln t \tag{21}$$

where $C_5 = 1 - \{1 + \int_0^\infty dt [2I_1A_0 \ln(I_2B_0t/2 + 1)/I_2t(I_2B_0t + 2)]\}^{-1}$ and $C_6 = 2I_1A_0(1 - C_5)^2/I_2^2B_0$. Thus we find that the cluster mass distribution of species A approaches the modified scaling form as follows:

$$a_k(t) \simeq A_0(1-C_5)^2 C_5^k \exp(-x) \qquad x = (C_6/C_5)kt^{-1}\ln t.$$
 (22)

For this case, the growing scale is $S(t) \propto t (\ln t)^{-1}$ while the time-independent scale is $S = 1/(1 - C_5)$. This case is similar to the above third case. All A clusters (the smaller or the larger clusters) can be conserved by the dynamics of the system.

Table 1. Summary of the results of the first model.	
Case	The cluster mass distribution of species A
v > 0 v = 0	Obeys the conventional scaling law with the exponents $w = 2v$ and $z = v$. Obeys the logarithm-correction scaling law with the exponents $w = 2$ and $z = 1$.
$0 > \upsilon > -1$	Satisfies the modified scaling form with the growing scale $S(t) \propto t^{-\nu}$.
$\upsilon = -1$	Satisfies the modified scaling form with the growing scale $S(t) \propto t (\ln t)^{-1}$.
$\upsilon < -1$	Satisfies the modified scaling form with the growing scale $S(t) \propto t$.

Finally, we investigate the last case of v < -1. From equations (11) and (12) we derive the asymptotic solution of a(t) at large times,

$$a(t) \simeq C_3 - C_7 t^{-1} \tag{23}$$

where $C_7 = 2I_1A_0(1-C_3)^2 \sum_{j=1}^{\infty} j^{\nu} / I_2^2 B_0$. In this case, species A evolves according to the modified scaling form

$$a_k(t) \simeq A_0 (1 - C_3)^2 C_3^k \exp(-x)$$
 $x = (C_7/C_3)kt^{-1}$ (24)

with the time-dependent scale $S(t) \propto t$ and the time-independent scale $S = 1/(1 - C_3)$. The scaling exponents are universal, w = 0 and z = 1. This shows that all the systems with index v in the range of v < -1 evolve in a similar way. To verify this result, we also investigate the special $v = -\infty$ case in which the aggregation of A clusters is driven only by the monomers of the catalyst. By employing the above technique we solve equations (1) exactly and also obtain the same scaling solution (24) of the cluster mass distribution for this special case, where $C_3 = 1 - (1 + I_1A_0/I_2)^{-1}$ and $C_7 = 2I_1A_0(1 - C_3)^2/I_2^2B_0$.

To sum up, we investigate the kinetics of the catalysis-driven aggregation model with a rate kernel $K_A(i, j, l) \propto l^{\upsilon}$ and obtain the asymptotic solution of the cluster mass distribution in several cases with different index υ . A summary of the results is illustrated in table 1.

3. The model of two-species mutually catalysis-driven aggregation processes

In this section, we investigate the kinetics of a two-species mutually catalysis-driven aggregation model. The concentrations of *A* and *B* clusters of *k*-mers are also denoted as a_k and b_k , respectively. We assume that the catalytic aggregation rates are $K_A(i; j; l) = I_1 l^{\nu_1}$ and $K_B(i; j; l) = I_2 l^{\nu_2}$ (ν_1 and ν_2 are two constants). Then the mean-field rate equations for this catalysis-driven aggregation system read

$$\frac{\mathrm{d}a_k}{\mathrm{d}t} = \frac{I_1}{2} \sum_{i+j=k} \sum_{l=1}^{\infty} (l^{\nu_2} a_i a_j b_l) - I_1 a_k \sum_{j=1}^{\infty} \sum_{l=1}^{\infty} (l^{\nu_2} a_j b_l)$$

$$\frac{\mathrm{d}b_k}{\mathrm{d}t} = \frac{I_2}{2} \sum_{i+j=k} \sum_{l=1}^{\infty} (l^{\nu_1} b_i b_j a_l) - I_2 b_k \sum_{j=1}^{\infty} \sum_{l=1}^{\infty} (l^{\nu_1} b_j a_l).$$
(25)

Under the monodisperse initial condition (2), with the help of anastz (3) we can recast equations (25) as the following equations:

$$\frac{da}{dt} = \frac{I_1 A M_{B\nu_2}}{2} \qquad \frac{dA}{dt} = -\frac{I_1 A^2 M_{B\nu_2}}{1-a}
\frac{db}{dt} = \frac{I_2 B M_{A\nu_1}}{2} \qquad \frac{dB}{dt} = -\frac{I_2 B^2 M_{A\nu_1}}{1-b}$$
(26)

with the shorthand notation $M_{A\nu_1}(t) = \sum_{j=1}^{\infty} j^{\nu_1} a_j(t)$ and $M_{B\nu_2}(t) = \sum_{j=1}^{\infty} j^{\nu_2} b_j(t)$. In this model, both species *A* and *B* obey the mass observation law, i.e., $M_{A1} = \sum_{j=1}^{\infty} ja_j \equiv A_0$ and $M_{B1} = \sum_{j=1}^{\infty} jb_j \equiv B_0$. Thus, equations (26) reduce to

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{I_1 A_0 (1-a)^2 M_{B\nu_2}}{2} \qquad \frac{\mathrm{d}b}{\mathrm{d}t} = \frac{I_2 B_0 (1-b)^2 M_{A\nu_1}}{2}.$$
(27)

From equations (27) one can derive the following identical equation:

$$I_2 \sum_{i=1}^{\infty} i^{\nu_1 - 1} a^i \equiv I_1 \sum_{j=1}^{\infty} j^{\nu_2 - 1} b^j.$$
(28)

The system is assumed to reach its steady state at $t \to \infty$ and its steady condition is then given as follows:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{I_1 A_0 (1-a)^2 M_{B\nu_2}}{2} \to 0 \qquad \frac{\mathrm{d}b}{\mathrm{d}t} = \frac{I_2 B_0 (1-b)^2 M_{A\nu_1}}{2} \to 0.$$
(29)

Thus we can conclude that either $a \to 1$ or $b \to 1$ at $t \to \infty$. Without any loss of generality, we assume $v_1 \ge v_2$. We then discuss the solutions of equations (27) in several different cases.

3.1. $v_1 > 0$ and $v_2 > 0$ case

It can be concluded from equations (28) and (29) that for this case, $a \to 1$ and $b \to 1$ at $t \gg 1$. In the long-time limit, equations (27) can be rewritten asymptotically as

$$\frac{\mathrm{d}a}{\mathrm{d}t} \simeq C_8 (1-a)^{2+\nu_1(1-\nu_2)/\nu_2} \qquad \frac{\mathrm{d}b}{\mathrm{d}t} \simeq C_9 (1-b)^{2+\nu_2(1-\nu_1)/\nu_1} \tag{30}$$

where $C_8 = [I_1 A_0 B_0 \Gamma(1 + \upsilon_2)/2] [I_2 \Gamma(\upsilon_1)/I_1 \Gamma(\upsilon_2)]^{(\upsilon_2 - 1)/\upsilon_2}$ and $C_9 = [I_2 A_0 B_0 \Gamma(1 + \upsilon_1)/2] [I_1 \Gamma(\upsilon_2)/I_2 \Gamma(\upsilon_1)]^{(\upsilon_1 - 1)/\upsilon_1}$. Equations (30) show that the solutions of a(t) and b(t) depend crucially on the value of $1/\upsilon_1 + 1/\upsilon_2$.

When $1/v_1 + 1/v_2 > 1$, we solve equations (30) and then obtain the scaling solutions of the cluster mass distributions

$$a_{k}(t) \simeq A_{0}C_{10}^{2}t^{-2\gamma_{1}}\exp(-x) \qquad x = C_{10}kt^{-\gamma_{1}}$$

$$b_{k}(t) \simeq B_{0}C_{11}^{2}t^{-2\gamma_{2}}\exp(-y) \qquad y = C_{11}kt^{-\gamma_{2}}$$
(31)

where $\gamma_1 = v_2/(v_1 + v_2 - v_1v_2)$, $\gamma_2 = v_1/(v_1 + v_2 - v_1v_2)$, $C_{10} = [(1 - v_1 + v_1/v_2)C_8]^{-\gamma_1}$ and $C_{11} = [(1 - v_2 + v_2/v_1)C_9]^{-\gamma_2}$. The results show that for this case, the evolution behaviour of either species obeys the conventional scaling definition of equation (10). The scaling exponents for species A are $w = 2v_2/(v_1 + v_2 - v_1v_2)$ and $z = v_2/(v_1 + v_2 - v_1v_2)$ while those for species B are $w = 2v_1/(v_1 + v_2 - v_1v_2)$ and $z = v_1/(v_1 + v_2 - v_1v_2)$. These indicate that the exponents are crucially dependent on the values of indices v_1 and v_2 . Moreover, the typical mass of species A grows indefinitely as $t^{v_2/(v_1+v_2-v_1v_2)}$ while that of species B grows as $t^{v_1/(v_1+v_2-v_1v_2)}$. In the special case of $v_1 = v_2 = 1$, the rate equations (27) can be solved exactly and one can find that the cluster mass distribution of either species evolves according to the conventional scaling form (10) with the constant exponents w = 2and z = 1; that is to say, both species scale as in the two-species non-interacting aggregation system. The results also imply that all the initial monomer clusters of one species can coagulate together under the catalytic action of another species.

When $1/v_1 + 1/v_2 = 1$, the cluster mass distributions of species A and B satisfy the exponential-correction scaling forms

$$a_{k}(t) \simeq A_{0} \exp(-2C_{8}t) \exp(-x) \qquad x = k \exp(-C_{8}t)$$

$$b_{k}(t) \simeq B_{0} \exp(-2C_{9}t) \exp(-y) \qquad y = k \exp(-C_{9}t).$$
(32)

The results show that the governing exponents C_8 and C_9 are nonuniversal and depend on the details of the rate kernel as well as the initial concentrations of both species. The typical mass of species A grows rapidly as $\exp(C_8 t)$ while that of species B grows as $\exp(C_9 t)$. In this case, the clusters of either type can also coagulate completely at the end, which is similar to the result in the above $1/v_1 + 1/v_2 > 1$ case.

When $1/v_1 + 1/v_2 < 1$, we find that the system will come to a gelation transition after a finite time (see, e.g., [21]). The critical time t_c (gel point) can be given by the expression $t_c = (2/I_1A_0B_0) \int_0^1 dx [(1-x)^2(1-y)^2 \sum_j (j^{v_2}y^{j-1})]$ (the relation between x and y is $I_2 \sum_i i^{v_1-1}x^i = I_1 \sum_j j^{v_2-1}y^j$). In this work, we only devote ourselves to the nongelling system and defer the thorough investigation of this gelation case to a future work.

Hence, only when $1/v_1 + 1/v_2 \ge 1$, the system has a permanent evolution in time and the typical mass of either species is always growing.

3.2. $v_1 > 0$ and $v_2 = 0$ case

In this case, we also find $a \to 1$ and $b \to 1$ at $t \to \infty$. By solving equations (27) we obtain the scaling solutions

$$a_{k}(t) \simeq A_{0}C_{12}^{2}(\ln t)^{-2/\nu_{1}}\exp(-x) \qquad x = C_{12}k(\ln t)^{-1/\nu_{1}}$$

$$b_{k}(t) \simeq B_{0}C_{13}^{2}t^{-2}(\ln t)^{2(1-\nu_{1})/\nu_{1}}\exp(-y) \qquad y = C_{13}kt^{-1}(\ln t)^{(1-\nu_{1})/\nu_{1}}$$
(33)

where $C_{12} = [I_1/I_2\Gamma(v_1)]^{-1/v_1}$ and $C_{13} = 2/I_1A_0B_0v_1C_{12}$. Equations (33) show that the typical mass of species *A* grows as $(\ln t)^{1/v_1}$ while that of species *B* grows as $t(\ln t)^{(v_1-1)/v_1}$. It is well known that for an irreversible aggregation system with a constant rate kernel, the typical mass grows as *t*. As contrasted with the typical mass of the general aggregation system, the typical mass of species *B* in this system has a positive logarithm-correction $(v_1 > 1)$ or negative one $(v_1 < 1)$. In this case, the initial monomer clusters of either species can also bond together completely at the end.

3.3. $v_1 = v_2 = 0$ *case*

In this special case, equations (27) can be exactly solved. The solutions of the cluster mass distributions are then determined as follows:

$$a_k(t) \simeq A_0 (C_{14}t)^{-2I_1/(I_1+I_2)} \exp(-x) \qquad x = k(C_{14}t)^{-I_1/(I_1+I_2)}$$

$$b_k(t) \simeq B_0 (C_{14}t)^{-2I_2/(I_1+I_2)} \exp(-y) \qquad y = k(C_{14}t)^{-I_2/(I_1+I_2)}$$
(34)

where $C_{14} = (I_1 + I_2)A_0B_0/2$. Both species scale according to the scaling form (10) with nonuniversal exponents dependent on the values of rate coefficients I_1 and I_2 . Moreover, we again find that all the clusters of either type can bond together at $t \to \infty$. These results are identical with the corresponding results in [19].

3.4. $v_1 \ge 0$ and $v_2 < 0$ case

It is obvious that $I_2 \sum_{i=1}^{\infty} i^{v_1-1} > I_1 \sum_{j=1}^{\infty} j^{v_2-1}$ in this case. So we have $a \to a_{1\infty}$ and $b \to 1$ at $\to \infty$, where $a_{1\infty}$ is a finite constant satisfying the equation $I_2 \sum_{i=1}^{\infty} i^{v_1-1}a_{1\infty}^j = I_1 \sum_{j=1}^{\infty} j^{v_2-1}$. In the long-time limit, from equations (27) one easily obtains the scaling solution of the cluster mass distribution of species *B*,

$$b_k(t) \simeq B_0 C_{15}^{-2} t^{-2} \exp(-y) \qquad y = C_{15}^{-1} k t^{-1}$$
 (35)

where $C_{15} = (I_2 A_0 B_0/2)(1 - a_{1\infty})^2 \sum_{j=1}^{\infty} (j^{\nu_1} a_{1\infty}^{j-1})$. Equation (35) indicates that for this case, species *B* evolves as in the irreversible single-species aggregation process.

Further, by inserting equation (35) into equations (27) we obtain the cluster mass distribution of species *A*, which approaches the modified scaling form

$$a_k(t) \simeq A_0 (1 - a_{1\infty})^2 a_{1\infty}^k \exp(-x)$$
 (36)

with the scaling variable $x = C_{16}kt^{\nu_2}$ for $-1 < \nu_2 < 0$, $x = C_{17}kt^{-1}\ln t$ for $\nu_2 = -1$ and $x = C_{18}kt^{-1}$ for $\nu_2 < -1$. Here the constants are $C_{16} = -\Gamma(\nu_2)(I_1A_0B_0/2a_{1\infty})(1 - a_{1\infty})^2C_{15}^{\nu_2-1}$, $C_{17} = (I_1A_0B_0/2a_{1\infty})(1 - a_{1\infty})^2C_{15}^{-2}$ and $C_{18} = (I_1A_0B_0/2a_{1\infty})(1 - a_{1\infty})^2C_{15}^{-2}\sum_{j=1}^{\infty} j^{\nu_2}$. For all cases with $\nu_2 < 0$, the time-independent scale for species A takes the same form $S_A = 1/(1 - a_{1\infty})$. Meanwhile, the time-dependent scale for species A grows as $t^{-\nu_2}$ in the $-1 < \nu_2 < 0$ case and as $t(\ln t)^{-1}$ in the $\nu_2 = -1$ case and as t in the $\nu_2 < -1$ case. Moreover, equation (36) also exhibits that small A clusters dominate over the large one in the long-time limit.

On the other hand, making a comparison between equations (35) and (36) shows that all the clusters of species *B* will coagulate together finally while *A* clusters of any size (the smaller and the larger) are conserved by the dynamics of the system.

3.5. $v_1 < 0$ and $v_2 < 0$ case

In this case, the solutions of equations (27) depend strongly on the relation between $I_2 \sum_{i=1}^{\infty} i^{\nu_1 - 1}$ and $I_1 \sum_{i=1}^{\infty} i^{\nu_2 - 1}$. We then discuss the solutions of equations (27) in three different subcases.

I. $I_2 \sum_{i=1}^{\infty} i^{\upsilon_1 - 1} > I_1 \sum_{j=1}^{\infty} j^{\upsilon_2 - 1}$ subcase. When $I_2 \sum_{i=1}^{\infty} i^{\upsilon_1 - 1} > I_1 \sum_{j=1}^{\infty} j^{\upsilon_2 - 1}$, we find that $a \to \text{const}$ and $b \to 1$ at $t \to \infty$. Under this condition, by employing the above technique we obtain the same equations (35) and (36) for the cluster mass distributions in this system. Hence, the results of this case are identical with those in section 3.4.

2. $I_2 \sum_{i=1}^{\infty} i^{v_1-1} < I_1 \sum_{j=1}^{\infty} j^{v_2-1}$ subcase. In the case of $I_2 \sum_{i=1}^{\infty} i^{v_1-1} < I_1 \sum_{j=1}^{\infty} j^{v_2-1}$, we have $a \to 1$ and $b \to \text{const}$ at $t \to \infty$. Let $b(t = \infty) = b_{1\infty}$ ($b_{1\infty}$ is a constant satisfying the equality $I_2 \sum_{i=1}^{\infty} i^{v_1-1} = I_1 \sum_{j=1}^{\infty} j^{v_2-1} b_{1\infty}^j$). Using a similar method to that in section 3.4, we find that species A scales according to the conventional form

$$a_k(t) \simeq A_0 C_{19} t^{-2} \exp(-x) \qquad y = C_{19} k t^{-1}$$
(37)

where $C_{19} = \left[(I_1 A_0 B_0/2)(1 - b_{1\infty})^2 \sum_{j=1}^{\infty} (j^{\nu_2} b_{1\infty}^{j-1}) \right]^{-1}$. The cluster mass distribution of species *B* approaches the modified scaling form

$$b_k(t) \simeq B_0 (1 - b_{1\infty})^2 b_{1\infty}^k \exp(-y)$$
 (38)

with the scaling variable $y \propto kt^{\nu_1}$ for $-1 < \nu_1 < 0$, $y \propto kt^{-1} \ln t$ for $\nu_1 = -1$ and $y \propto kt^{-1}$ for $\nu_1 < -1$. In this case, all the clusters of species *A* will coagulate together finally, while the complete coagulation of *B* clusters cannot be fulfilled. The results indicate that this case is in contrast to the above subcase of $I_2 \sum_{i=1}^{\infty} i^{\nu_1 - 1} > I_1 \sum_{i=1}^{\infty} j^{\nu_2 - 1}$.

3. $I_2 \sum_{i=1}^{\infty} i^{\upsilon_1 - 1} = I_1 \sum_{j=1}^{\infty} j^{\upsilon_2 - 1}$ subcase. In the symmetrical subcase of $I_2 \sum_{i=1}^{\infty} i^{\upsilon_1 - 1} = I_1 \sum_{j=1}^{\infty} j^{\upsilon_2 - 1}$, we have $a \to 1$ and $b \to 1$ at $t \to \infty$. The evolution behaviour in this subcase is very complicated and depends strongly on the values of indices υ_1 and υ_2 . In order to investigate the kinetics of this system in a transparent way, we discuss the solutions of equations (27) in several systems as follows.

When $-1 < v_1 < 0$ and $-1 < v_2 < 0$, one can obtain equations (31) of the cluster mass distributions for this system. So, the results of this system are identical with those of the system with $1/v_1 + 1/v_2 > 1$ in section 3.1.

When $-1 < v_1 < 0$ and $v_2 = -1$, we obtain the scaling solutions of the cluster mass distributions

$$a_{k}(t) \simeq A_{0}C_{21}^{2}t^{-2/(1-2\nu_{1})}(\ln t)^{2/(1-2\nu_{1})}\exp(-x)$$

$$x = C_{21}kt^{-1/(1-2\nu_{1})}(\ln t)^{1/(1-2\nu_{1})}$$

$$b_{k}(t) \simeq B_{0}C_{20}^{2}t^{2\nu_{1}/(1-2\nu_{1})}(\ln t)^{-2(1-\nu_{1})/(1-2\nu_{1})}\exp(-y)$$

$$y = C_{20}kt^{\nu_{1}/(1-2\nu_{1})}(\ln t)^{-(1-\nu_{1})/(1-2\nu_{1})}$$
(39)

where $C_{20} = [\upsilon_1/(2\upsilon_1 - 1)]^{(\upsilon_1 - 1)/(1 - 2\upsilon_1)} [-I_1/I_2\Gamma(\upsilon_1)]^{(\upsilon_1 - 1)/(1 - 2\upsilon_1)} [I_2A_0B_0\Gamma(\upsilon_1)(2\upsilon_1 - 1)/2]^{\upsilon_1/(1 - 2\upsilon_1)}$ and $C_{21} = [I_1\upsilon_1C_{20}/I_2\Gamma(\upsilon_1)(1 - 2\upsilon_1)]^{-1/\upsilon_1}$. Equations (39) indicate that both species scale according to the logarithm-correction form. The typical mass of species A grows indefinitely as $t^{1/(1 - 2\upsilon_1)}(\ln t)^{-1/(1 - 2\upsilon_1)}$ while that of species B grows as $t^{-\upsilon_1/(1 - 2\upsilon_1)}(\ln t)^{(1 - \upsilon_1)/(1 - 2\upsilon_1)}$. So, the clusters of one species can bond each other under the catalytic action of another species.

When $-1 < v_1 < 0$ and $v_2 < -1$, the cluster mass distributions approach the conventional scaling forms

$$a_k(t) \simeq A_0 C_{22}^2 t^{-2/(1-2\nu_1)} \exp(-x) \qquad x = C_{22} k t^{-1/(1-2\nu_1)} b_k(t) \simeq B_0 C_{23}^2 t^{2\nu_1/(1-2\nu_1)} \exp(-y) \qquad y = C_{23} k t^{\nu_1/(1-2\nu_1)}$$
(40)

where $C_{22} = \left[(1 - 2\upsilon_1)A_0B_0I_2^2\Gamma^2(\upsilon_1) / 2I_1\sum_{j=1}^{\infty} j^{\upsilon_2} \right]^{-1/(1-2\upsilon_1)}$ and $C_{23} = -I_2\Gamma(\upsilon_1)C_{22}^{-\upsilon_1} / I_1\sum_{j=1}^{\infty} j^{\upsilon_2}$. The governing exponents for species *A* are $w = 2/(1 - 2\upsilon_1)$ and $z = 1/(1 - 2\upsilon_1)$, while those for species *B* are $w = 2\upsilon_1/(2\upsilon_1 - 1)$ and $z = \upsilon_1/(2\upsilon_1 - 1)$. These indicate that the exponents are nonuniversal and dependent on the details of the rate kernel. On the other hand, the typical mass of species *A* in this system grows as $t^{1/(1-2\upsilon_1)}$ and that of species *B* grows as $t^{-\upsilon_1/(1-2\upsilon_1)}$, which indicate that all the clusters of either type can bond together at the end.

When $v_1 = v_2 = -1$, we also obtain the solutions of the cluster mass distributions, which take the logarithm-correction scaling forms as follows:

$$a_k(t) \simeq A_0 C_{24}^2 (t \ln t)^{-2/3} \exp(-x) \qquad x = C_{24} k (t \ln t)^{-1/3} b_k(t) \simeq B_0 C_{25}^2 (t \ln t)^{-2/3} \exp(-y) \qquad y = C_{25} k (t \ln t)^{-1/3}$$
(41)

where $C_{24} = (I_2^2 A_0 B_0 / 2I_1)^{-1/3}$ and $C_{25} = (I_1^2 A_0 B_0 / 2I_2)^{-1/3}$. For this system, the typical mass of either species grows indefinitely as $(t \ln t)^{1/3}$, and therefore the complete coagulation of the clusters of either type will be fulfilled finally.

When $v_1 = -1$ and $v_2 < -1$, both species also scale according to the logarithm-correction forms

$$a_k(t) \simeq A_0 C_{26}^2 t^{-2/3} (\ln t)^{-4/3} \exp(-x) \qquad x = C_{26} k t^{-1/3} (\ln t)^{-2/3} b_k(t) \simeq B_0 C_{27}^2 t^{-2/3} (\ln t)^{2/3} \exp(-y) \qquad y = C_{27} k t^{-1/3} (\ln t)^{1/3}$$
(42)

where $C_{26} = (I_2^2 A_0 B_0 / 6I_1 \sum_{j=1}^{\infty} j^{\nu_2})^{-1/3}$ and $C_{27} = I_2 C_{26} / 3I_1 \sum_{j=1}^{\infty} j^{\nu_2}$. Equations (42) show that large *B* clusters dominate over the corresponding *A* clusters, which is independent of the initial concentrations A_0 and B_0 . From equations (42) we also find that the typical mass of species *A* in this system grows as $t^{1/3} (\ln t)^{2/3}$ while that of species *B* grows as $t^{1/3} (\ln t)^{-1/3}$. So, the clusters of either type in this subcase will also coagulate completely at the end.

When $v_1 < -1$ and $v_2 < -1$, the scaling solutions of the cluster mass distributions are obtained

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Table 2. Summary of the results of the second model.	
Case	Summary of the results
$\overline{\upsilon_1 \geqslant 0, \upsilon_2 \geqslant 0}$	(i) If $v_1 = 0$ or $v_2 = 0$, both species scale according to the logarithm-correction form (ii) If $v_1 = v_2 = 0$, both species scale according to the conventional form (iii) If $\gamma > 1$, both species scale according to the conventional form (iv) If $\gamma = 1$, both species scale according to the exponential-correction form (v) If $\gamma < 1$, both species fall in the gelation transition after a certain time
$v_1 \ge 0, v_2 < 0$	(i) Species <i>B</i> scales according to the conventional form with constant exponents (ii) If $0 > v_2 > -1$, species <i>A</i> scales according to the modified form with $S(t) \propto t^{-v_2}$ (iii) If $v_2 = -1$, species <i>A</i> scales according to the modified form with $S(t) \propto t (\ln t)^{-1}$ (iv) If $v_2 < -1$, species <i>A</i> scales according to the modified form with $S(t) \propto t$
$\upsilon_1 < 0, \upsilon_2 < 0$	(i) If $\eta > 1$, the cluster mass distribution of species <i>A</i> obeys the modified scaling form while that of species <i>B</i> obeys the conventional one (ii) If $\eta < 1$, the cluster mass distribution of species <i>A</i> obeys the conventional scaling form while that of species <i>B</i> obeys the modified one (iii) If $\eta = 1$, the cluster mass distribution of either species obeys the conventional scaling form

$$a_k(t) \simeq A_0 C_{28}^2 t^{-2/3} \exp(-x) \qquad x = C_{28} k t^{-1/3}$$

$$b_k(t) \simeq B_0 C_{29}^2 t^{-2/3} \exp(-y) \qquad y = C_{29} k t^{-1/3}$$
(43)

where $C_{28} = \left[3A_0B_0\left(I_2\sum_{j=1}^{\infty} j^{\nu_1}\right)^2/2I_1\sum_{j=1}^{\infty} j^{\nu_2}\right]^{-1/3}$ and $C_{29} = \left[3A_0B_0\left(I_1\sum_{j=1}^{\infty} j^{\nu_2}\right)^2/2I_2\sum_{j=1}^{\infty} j^{\nu_1}\right]^{-1/3}$. Equations (43) show that for this system with indices $\nu_1 < -1$ and $\nu_2 < -1$, both species always scale according to the conventional form of equation (10) with the universal exponents w = 2/3 and z = 1/3. The results also indicate that for this subcase, the clusters of one type can bond together completely with the help of clusters of another type.

To sum up, we investigate the kinetic behaviour of the two-species catalysis-driven aggregation processes in several different cases. A detailed summary of the results is given in table 2, where $\gamma = 1/v_1 + 1/v_2$ and $\eta = I_2 \sum_{i=1}^{\infty} i^{v_1-1} / I_1 \sum_{j=1}^{\infty} j^{v_2-1}$.

4. Summary

We have studied the kinetic behaviour of the two-species catalysis-driven aggregation processes. Based on the mean-field rate equations, we analysed the dependence of the kinetics of the system on the catalyst. It was found that the evolution behaviour of the cluster mass distributions depends crucially on the details of the rate kernel.

For the first model in which aggregation of A clusters proceeds at a rate $K_A(i; j; l) \propto l^{\upsilon}$ (*l* is the size of *B* catalyst), the evolution behaviour of species A is dependent on the value of index υ . We found that the cluster mass distribution obeys the conventional scaling law in the case of $\upsilon \ge 0$ while it approaches the modified scaling form in the $\upsilon < 0$ case.

We also investigated a two-species mutually catalysis-driven aggregation model with the reaction rate kernel $K_A(i; j; l) = I_1 l^{\nu_1}$ and $K_B(i; j; l) = I_2 l^{\nu_2}$. The results indicated that the kinetics of the system depends strongly on the values of υ_1 and υ_2 . In some special cases, the rate coefficients I_1 and I_2 also play an important role in the kinetic behaviour of the cluster mass distributions. For the cases of υ_1 , $\upsilon_2 \ge 0$ or $I_2 \sum_{i=1}^{\infty} i^{\upsilon_1-1} = I_1 \sum_{j=1}^{\infty} j^{\upsilon_2-1}$, the cluster mass distribution of either species approaches the conventional scaling law; while for other cases the species with the smaller value of $\sum_{i=1}^{\infty} i^{\upsilon_i-1}/I_i$ (l = 1, 2) obeys the conventional scaling law while another species scales according to the modified form.

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