Evolution behavior of catalytically activated replication–decline in a coagulation process*

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(Received 30 December 2012; revised manuscript received 1 March 2013)

We propose a catalytically activated replication–decline model of three species, in which two aggregates of the same species can coagulate themselves, an A aggregate of any size can replicate itself with the help of B aggregates, and the decline of A aggregate occurs under the catalysis of C aggregates. By means of mean-field rate equations, we derive the asymptotic solutions of the aggregate size distribution $a_k(t)$ of species A, which is found to depend strongly on the competition among three mechanisms: the self-coagulation of species A, the replication of species A catalyzed by species B, and the decline of species A catalyzed by species C. When the self-coagulation of species A dominates the system, the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form. When the catalyzed replication process dominates the system, $a_k(t)$ takes the generalized scaling form. When the catalyzed decline process dominates the system, $a_k(t)$ approaches the modified scaling form.

Keywords: aggregation, catalytically activated reaction, replication, decline

PACS: 68.43.Jk, 05.40.–a, 82.20.–w, 89.75.Da **DOI:** 10.1088/1674-1056/22/9/096802

1. Introduction

The aggregation phenomena, in which small aggregates merge irreversibly to form large aggregates, have been investigated extensively in the last few decades.[1–9] Real-world examples include crystal growth, chemical bonding of polymeric chains, and the formation of polymeric networks or gels, the formation of clouds in atmosphere, and the growth of droplets. It is thus of theoretical and practical significance to understand the kinetics of aggregate growth. A lot of effort has been devoted to investigating the kinetic behavior of irreversible aggregation through the binary coalescence. An aggregation process typically begins with a huge number of molecular units (monomers) that will join irreversibly to form clusters (polymers), which can be described as $A_i + A_j \xrightarrow{I(i,j)} A_{i+j}$, where A_i denotes the mass of cluster i (a cluster containing i units), and $I(i,j)$ is the reaction kernel which generally depends on the sizes of the two clusters. Assuming spatial homogeneity, the system is characterized by the concentrations $a_k(t)$ of aggregates of mass k at time t. In 1916, Smoluchowski first introduced his eponymous differential equation[10]

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

which describes the evolution of $a_k(t)$ in the mean-field limit. From then on, many works have been done to understand this kind of nonequilibrium aggregate growth. Most intriguingly, many research works have shown that the size distribution of clusters in an aggregation system can approach a scaling form in the long-time limit.[11,12]

Recently, considerable interest has been aroused in a variety of aggregation phenomena involving catalytically activated reactions.[13–16] Lin *et al.*[17] introduced the mutually catalyzed birth of population and assets in exchange-driven aggregation growth to study the interaction between the population and the asset aggregates. In our previous work,[18] we studied the competition between catalyzed birth and catalyzed death in exchange-driven aggregation growth, the results showed that the competition between the two mechanisms gives rise to much more abundant kinetic behaviors. In this paper, we propose a catalytically activated replication–decline model to study the competition between the catalysis-driven replication process and the catalysis-driven decline process, as well as the coagulation process. In this model, there are three distinct species of aggregates in the system, aggregates A, B, and C. Two aggregates of the same species can bond to form a larger cluster when they meet, which can be described by the reaction $A_i + A_j \xrightarrow{I(i,j)} A_{i+j}$ (or $B_i + B_j \xrightarrow{J_1(i,j)} B_{i+j}$, $C_i + C_j \xrightarrow{J_2(i,j)} C_{i+j}$). But the replication of A clusters occurs only with the help of B clusters, which can be described as $A_i + B_j \xrightarrow{K(i,j)} 2A_i + B_j$, where $K(i,j)$ is the catalysis-driven replication rate kernel. Similarly, the decline of A clusters occurs only with the help of C clusters, $A_i + C_j \xrightarrow{L(i,j)} A_{i-1} + C_j$, where $L(i,j)$ is the catalysis-driven decline rate kernel. By discussing the evolution behaviors of the systems, we find

*Project supported by the National Natural Science Foundation of China (Grant Nos. 10875086 and 11145004).
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that the aggregate size distributions $a_k(t)$ of species A depend strongly on the competition between the catalysis-driven replication process and the catalysis-driven decline process, as well as the coagulation process.

We believe that our catalysis-driven replication and catalysis-driven decline model for the coagulation process can mimic many real phenomena in nature and society. For example, it has been observed that the end-to-end aggregation of DNA fragments can occur to form long linear DNA chains,[19] which can be described as $A_i + A_j \xrightarrow{I(i,j)} A_{i+j}$. Moreover, it is well known that the replication of DNA is essential during each cell cycle and the replication process cannot take place in the absence of an RNA primer. So, it is reasonable that we consider the replication of DNA as a catalytically activated reaction and the RNA primer as the catalyst. Specially, during the replication of DNA, the DNA helix unwinds at first so that each strand of the double helix could act as a template, then the RNA primer is synthesized (this process can be reasonably considered as a coagulation process, $B_i + B_j \xrightarrow{J_1(i,j)} B_{i+j}$), resulting in the transfer of genetic information from the DNA molecule to the messenger RNA. And after the daughter molecules are created, the genetic information is transferred from the messenger RNA to the daughter molecules. Thus with the help of the primer RNA, a complete replication of DNA is achieved.[20] This whole reaction scheme can be described as $A_i + B_j \xrightarrow{K(i,j)} 2A_i + B_j$. On the other hand, a DNA aggregate may be infected by a virus aggregate.[21,22] We consider a simple case here that the infected individuals cannot recover in the future, which will reasonably result in the decline of the healthy DNA aggregates, the reaction scheme can be described as $A_i + C_j \xrightarrow{L(i,j)} A_{i-1} + C_j$, where C_j denotes the virus aggregates, which may spontaneously coagulate to form larger clusters, $C_i + C_j \xrightarrow{J_2(i,j)} C_{i+j}$. The lower limit for the DNA aggregate is $i = 0$; once the size of a DNA aggregate reaches zero, there is no further decline. Additionally, the catalysis-driven replication and catalysis-driven decline in aggregation growth process also occur in many biological, chemical, and economical systems, and our model may provide a fairly useful description of such natural phenomena.

The paper is organized as follows. In Section 2, we introduce the catalytically activated replication–decline model, and describe the outline of the generalized Smoluchowski's rate equation approach to study the kinetic evolution behaviors of the system. Then a brief summary is given in Section 3.

2. Analytical solution of the catalytically activated replication–decline model

In our model, we assume that the self-coagulation rate kernels of species A, B, and C are all independent of the sizes of reactants, namely, $I(i,j) = I, J_1(i,j) = J_1$, and $J_2(i,j) = J_2$ (here I, J_1, and J_2 are constants). It is sound for some chemical and biological systems, especially for the polymerization of chain-shaped polymers which have only two energetic reactive edges.

Since our system is assumed to be of spatial homogeneity in the mean-field limit, the theoretical approach to the model can be based on the rate equation given by Family *et al.*,[23] which assumes that the reaction proceeds at a rate proportional to the reactant concentrations. The governing rate equations for the model then read

$$\frac{\mathrm{d}a_k}{\mathrm{d}t} = \frac{I}{2}\sum_{i+j=k} a_i a_j - I a_k \sum_{j=1}^{\infty} a_j + a_k \sum_{j=1}^{\infty} K(k,j) b_j + a_{k+1} \sum_{j=1}^{\infty} L(k+1,j) c_j - a_k \sum_{j=1}^{\infty} L(k,j) c_j, \quad (1)$$

$$\frac{\mathrm{d}b_k}{\mathrm{d}t} = \frac{J_1}{2}\sum_{i+j=k} b_i b_j - J_1 b_k \sum_{j=1}^{\infty} b_j, \quad (2)$$

$$\frac{\mathrm{d}c_k}{\mathrm{d}t} = \frac{J_2}{2}\sum_{i+j=k} c_i c_j - J_2 c_k \sum_{j=1}^{\infty} c_j. \quad (3)$$

In Eq. (1) (similarly in Eqs. (2) and (3)), the first term on the right-hand side accounts for the gain in $a_k(t)$ due to the coagulation processes between aggregates $a_i(t)$ and $a_{k-i}(t)$ $(i = 1, 2, \ldots, k-1)$, $A_i + A_{k-i} \xrightarrow{I} A_k$; while the second term accounts for the loss in $a_k(t)$ due to the merging processes that the $a_k(t)$ aggregates bond with any other aggregates, $A_k + A_j \xrightarrow{I} A_{k+j}$. The third term accounts for the gain in $a_k(t)$ due to the catalyzed replication processes driven by catalyst B, $A_k + B_j \xrightarrow{K(k,j)} 2A_k + B_j$. The forth term accounts for the gain in $a_k(t)$ due to the catalyzed decline processes driven by catalyst C, $A_{k+1} + C_j \xrightarrow{L(k+1,j)} A_k + C_j$. And the last term accounts for the loss in $a_k(t)$ driven by catalyst C, $A_k + C_j \xrightarrow{L(k,j)} A_{k-1} + C_j$.

Clusters B and C have the same coagulation processes, so their evolution kinetic behaviors are the same and relatively simple. The kinetics of a simple coagulation process has been studied in the previous work under the monodisperse initial condition $b_k(0) = B_0\delta_{k1}$ and $c_k(0) = C_0\delta_{k1}$ (B_0 and C_0 are the initial aggregate concentrations of species B and C, respectively).[24] The accurate solution to the aggregate size distribution of species B (similarly for species C) is

$$b_k(t) = 4B_0(J_1B_0t + 2)^{-2}\left(1 - \frac{2}{J_1B_0t + 2}\right)^{k-1}. \quad (4)$$

The sketch of the time and size dependence of $b_k(t)$ with given parameters is shown in Fig. 1.

In the scaling regime of $k \gg 1$ and $t \gg 1$, Eq. (4) can be asymptotically rewritten as

$$b_k(t) \simeq 4J_1^{-2}B_0^{-1}t^{-2}\exp(-x), \quad x = 2J_1^{-1}B_0^{-1}kt^{-1}, \quad (5)$$

which satisfies the conventional scaling law[11,24]

$$f_k(t) \sim t^{-w}\Phi[k/S(t)], \quad S(t) \propto t^z, \quad (6)$$

with the scaling function $\Phi(x) = \exp(-x)$ and the scaling exponents $w = 2, z = 1$. Here $S(t) = J_1 B_0 t/2$ is the characteristic size of aggregates B, which plays a role analogous to the correlation length in critical phenomena.

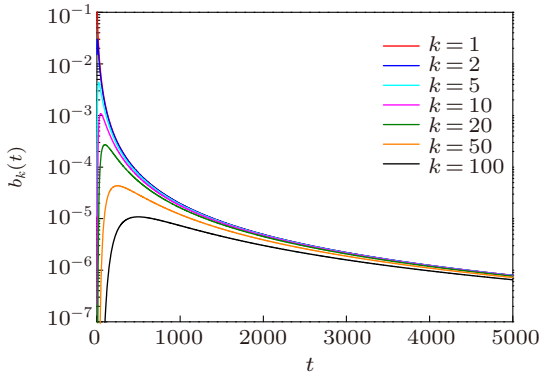


Fig. 1. (color online) Semilog plot of the cluster size distribution $b_k(t)$ versus time t for different size k. The initial condition is $J_1 = 1$ and $B_0 = 0.2$.

Now we focus on the kinetics of clusters A. We consider the natural situation where the catalyzed replication rate kernel is independent of the mother aggregates.[25] While the catalyzed decline rate kernel has a linear dependence on the size of aggregates A (it is reasonable because a larger DNA helix has a bigger probability to be infected by viruses). On the other hand, the two catalysis rate kernels may have a relation with the impacting efficiency of the impacting clusters. In our model, a cluster is characterized only by its size and the impacting efficiency may depend on the size of the impacting cluster.[26] So, in this paper, we restrict our attention to two cases in which the catalysis rates are proportional to the catalyst aggregates' sizes [$K(i,j) = Kj$; $L(i,j) = Lij$] or independent of the catalyst aggregates' sizes [$K(i,j) = K$; $L(i,j) = Lj$]. By comparing the kinetics of the system in the two cases, we find that the aggregate size distribution $a_k(t)$ depends on the competition among three mechanisms: the self-coagulation of species A, the species-B-catalyzed replication of species A, and the species-C-catalyzed decline of species A. In the first case, the aggregate size distribution $a_k(t)$ mainly depends on the competition between the species-B-catalyzed replication of species A and the species-C-catalyzed decline of species A. In the second case, the aggregate size distribution $a_k(t)$ mainly depends on the competition between the self-coagulation of species A and the species-C-catalyzed decline of species A.

2.1. The system with the catalytically activated reaction rates proportional to the catalyst aggregates' sizes

We first study the case in which the catalyzed replication and decline rate kernels are both proportional to the catalyst aggregates' sizes, $K(k,j) = Kj$ and $L(k,j) = Lkj$.

In this case, the governing rate equation (1) can be rewritten in the following form:

$$\frac{\mathrm{d}a_k}{\mathrm{d}t} = \frac{I}{2}\sum_{i+j=k} a_i a_j - IN_A a_k + KM_B a_k + L[(k+1)a_{k+1} - ka_k]M_C, \tag{7}$$

where $N_A(t) = \sum_{j=1}^{\infty} a_j(t)$ denotes the total number of aggregates A at time t, and the total mass of aggregates A is described as $M_A(t) = \sum_{j=1}^{\infty} j a_j(t)$. Similar definitions are used for aggregates B and C.

Under the monodisperse initial conditions, $a_k(0) = A_0\delta_{k1}$, Eq. (7) can be solved with the help of the ansatz (the application of this ansatz has been thoroughly discussed in our previous works[17,18])

$$a_k(t) = A(t)[a(t)]^{k-1}. \tag{8}$$

Substituting Eq. (8) into Eq. (7), we can transform the rate equation (7) to the following equations:

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{IA}{2} + L(a^2 - a)M_C, \tag{9}$$

$$\frac{\mathrm{d}A}{\mathrm{d}t} = [-IN_A + KM_B + L(2a-1)M_C]A, \tag{10}$$

with the initial conditions

$$a(0) = 0, \quad A(0) = A_0. \tag{11}$$

From Eqs. (9) and (10) we can readily obtain

$$A(t) = A_0(1-a)^2 E_B(t) E_C^{-1}(t), \tag{12}$$

with the shorthand notations $E_B(t) = \exp[K\int_0^t M_B(t')\,\mathrm{d}t']$ and $E_C(t) = \exp[L\int_0^t M_C(t')\,\mathrm{d}t']$. Substituting Eq. (12) into Eq. (9), we obtain

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \frac{IA_0}{2} E_B E_C^{-1}(1-a)^2 - La(1-a)M_C. \tag{13}$$

From this Bernoulli equation and the initial condition (11), we derive the solution of $a(t)$

$$a(t) = 1 - \frac{1}{1 + \frac{1}{2}IA_0 E_C^{-1}\int_0^t E_B(t')\,\mathrm{d}t'}. \tag{14}$$

From Eq. (4), we find that the total masses of aggregates B and C are separately conserved

$$M_B(t) = B_0, \quad M_C(t) = C_0. \tag{15}$$

Then we obtain $E_B(t) = \exp(KB_0 t)$ and $E_C(t) = \exp(LC_0 t)$. So, Eq. (14) becomes

$$a(t) = 1 - \frac{1}{1 - \frac{IA_0}{2KB_0}\mathrm{e}^{-LC_0 t} + \frac{IA_0}{2KB_0}\mathrm{e}^{(KB_0 - LC_0)t}}. \tag{16}$$

From Eq. (12), we can easily derive the exact solution of $A(t)$

$$A(t) = A_0\left[\frac{1}{1 - \frac{IA_0}{2KB_0}\mathrm{e}^{-LC_0 t} + \frac{IA_0}{2KB_0}\mathrm{e}^{(KB_0 - LC_0)t}}\right]^2 \mathrm{e}^{(KB_0 - LC_0)t}. \tag{17}$$

So, the exact solution of the aggregate size distribution of

species A is obtained as

$$a_k(t) = A_0\left[\frac{1}{1-\frac{IA_0}{2KB_0}\mathrm{e}^{-LC_0t}+\frac{IA_0}{2KB_0}\mathrm{e}^{(KB_0-LC_0)t}}\right]^2 \mathrm{e}^{(KB_0-LC_0)t} \times\left[1-\frac{1}{1-\frac{IA_0}{2KB_0}\mathrm{e}^{-LC_0t}+\frac{IA_0}{2KB_0}\mathrm{e}^{(KB_0-LC_0)t}}\right]^{k-1}. \quad (18)$$

Moreover, the total number of species A is obtained as

$$N_A(t) = \frac{A}{(1-a)} = \frac{A_0\mathrm{e}^{(KB_0-LC_0)t}}{1-\frac{IA_0}{2KB_0}\mathrm{e}^{-LC_0t}+\frac{IA_0}{2KB_0}\mathrm{e}^{(KB_0-LC_0)t}}. \quad (19)$$

And the total mass of species A is derived as

$$M_A(t) = \frac{A}{(1-a)^2} = A_0\mathrm{e}^{(KB_0-LC_0)t}. \quad (20)$$

From Eqs. (18)–(20), we find that the kinetics of species A is dominated by the relation of KB_0 and LC_0, we then discuss the kinetics of the system in the following several subcases.

In the subcase of $KB_0 > LC_0$, from Eq. (18), we derive the asymptotical expression of $a_k(t)$ in the long-time limit as

$$a_k(t) \simeq \frac{4K^2B_0^2}{I^2A_0}\mathrm{e}^{-(KB_0-LC_0)t}\left[1-\frac{2KB_0}{IA_0}\mathrm{e}^{-(KB_0-LC_0)t}\right]^{k-1}. \quad (21)$$

The sketch of the time and size dependence of $a_k(t)$ with given parameters in this subcase is shown in Fig. 2.

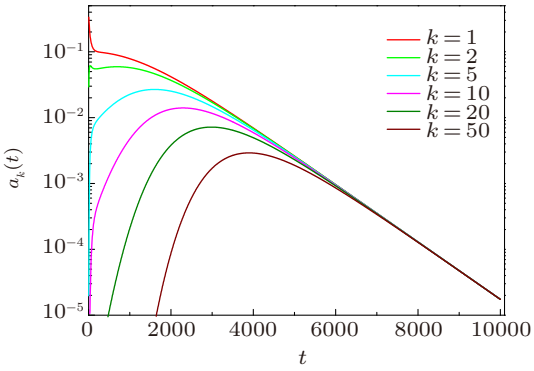


Fig. 2. (color online) Semilog plot of the cluster size distribution $a_k(t)$ versus time t for different size k. The initial condition is $K = L = 0.1$, $A_0 = 0.4$, $B_0 = 0.2$, and $C_0 = 0.19$.

In the scaling regime of $k \gg 1$ and $t \gg 1$, Eq. (21) can be rewritten as

$$a_k(t) \simeq \frac{2KB_0}{I}S_A^{-1}(t)\Phi(x), \quad x = k/S_A(t), \quad (22)$$

with $S_A(t) = \frac{IA_0}{2KB_0}\mathrm{e}^{(KB_0-LC_0)t}$. It follows that the aggregate size distribution $a_k(t)$ satisfies the generalized scaling form[18]

$$a_k(t) \simeq S_A^{-1}(t)\Phi[k/S_A(t)]. \quad (23)$$

From Eq. (19), we find that in this subcase, the total number of species A approaches a constant in the long-time limit, $N_A(t) \simeq 2KB_0/I$, which is independent of the initial value $N_A(0) = A_0$. And Eq. (20) shows that the total mass of species A increases with time exponentially, it is just the result of species-B-catalyzed replication of species A. So, we can conclude that in this subcase the dominating mechanism is the catalyzed replication process, and the effective catalyzed replication rate is $K_e = KB_0$, the effective catalyzed decline rate is $L_e = LC_0$. On the other hand, we can easily deduce the conclusion that the aggregate size distribution takes the generalized scaling form (23) when the catalyzed replication (or replication) process dominates the evolution of the system, which was proved in the previous research.[25]

In the subcase of $KB_0 = LC_0$, from Eq. (18), we obtain the aggregate size distribution $a_k(t)$

$$a_k(t) \simeq \frac{4K^2B_0^2I^{k-1}A_0^k}{(IA_0+2KB_0)^{k+1}}\Phi(x), \quad x = k/S_A(t), \quad (24)$$

with $S_A(t) = (1+\frac{IA_0}{2KB_0})\mathrm{e}^{LC_0t}$. It means that in this subcase, $a_k(t)$ satisfies the conventional scaling form (6) in the long-time limit. We can conclude that in this subcase, the effects of species-B-catalyzed replication of species A and species-C-catalyzed decline of species A counteract each other, so the evolution of the system mainly depends on the self-coagulation of species A, and the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form (6). This conclusion (the aggregate size distribution follows the conventional law when the self-coagulation process dominates the evolution of the system) was broadly verified in the previous works.[18,24]

The sketch of the time and size dependence of $a_k(t)$ in this subcase is shown in Fig. 3. It shows that the system will evolve to a steady state at $t \to \infty$ and $a_k(t)$ takes a nonzero steady-state distribution.

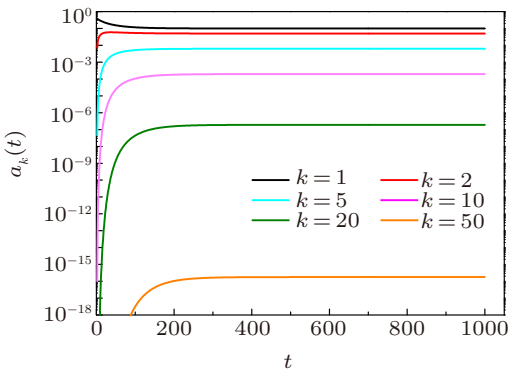


Fig. 3. (color online) Semilog plot of the cluster size distribution $a_k(t)$ versus time t for different size k. The initial condition is $K = L = 0.1$, $A_0 = 0.4$, and $B_0 = C_0 = 0.2$.

Moreover, the total number of species A is obtained as

$$N_A(t) \simeq \frac{2KB_0}{IA_0+2KB_0}A_0. \quad (25)$$

It shows that in the long-time limit, the total number of species A approaches a constant smaller than the initial value A_0. And the total mass of species A is found to be conserved

$$M_A(t) = A_0. \quad (26)$$

Equations (25) and (26) also indicate that in this subcase, the effects of catalyzed replication and catalyzed decline counteract each other, and the dominated process is the self-

coagulation of species A. So, the system will eventually evolve to a steady state.

In the subcase of $KB_0 < LC_0$, the effect of catalyzed decline is stronger than that of catalyzed replication, i.e., the dominating process is species-C-catalyzed decline of species A. From Eq. (18), we deduce the aggregate size distribution of species A

$$a_k(t) \simeq (1-q^{-1})^k \frac{2KB_0}{I} R^{-1}(t)\Psi(x), \quad x = k/R(t), \quad (27)$$

with the scaling function $\Psi(x) = \exp(x)$ and the characteristic size

$$R(t) = q(q-1)\frac{2KB_0}{IA_0}\,\mathrm{e}^{(LC_0-KB_0)t},$$

where

$$q(t) = 1 - \frac{IA_0}{2KB_0}\,\mathrm{e}^{-LC_0 t},$$

and it is obvious that $q(t) \longrightarrow 1$. Equation (27) shows that the usual scaling form (6) breaks down for this case and the aggregate size distribution satisfies the modified scaling form[27]

$$a_k(t) \simeq \lambda^k t^{-w}[R(t)]^{-1}\Psi[x], \quad x = k/R(t). \quad (28)$$

This conclusion of the aggregate size distribution taking the modified scaling form (28) when the catalyzed decline process dominates the evolution of the system agrees with the results of the previous works.[18,28]

The total number and the total mass of species A are both found to decay with time exponentially

$$N_A(t) \simeq q^{-1}A_0\,\mathrm{e}^{-(LC_0-KB_0)t}, \quad M_A(t) \simeq A_0\,\mathrm{e}^{-(LC_0-KB_0)t}. \quad (29)$$

It is obvious that in this subcase, the dominating mechanism is the species-C-catalyzed decline of species A. And species A will die out in the end, which can also be seen in Fig. 4.

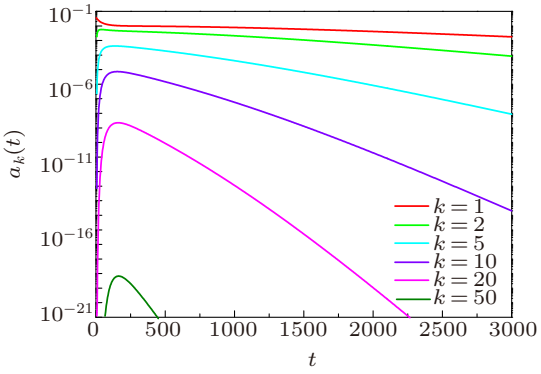


Fig. 4. (color online) Semilog plot of the cluster size distribution $a_k(t)$ versus time t for different size k. The initial condition is $K = L = 0.1$, $A_0 = 0.4$, $B_0 = 0.2$, and $C_0 = 0.21$.

To sum up, in the case of the catalysis rates proportional to the catalyst aggregates' sizes, the aggregate size distribution $a_k(t)$ is found to mainly depend on the competition between the species-B-catalyzed replication of species A and the species-C-catalyzed decline of species A. When the effective catalyzed replication rate $K_e = KB_0$ is larger than the effective catalyzed decline rate $L_e = LC_0$, the catalyzed replication process dominates the system, the aggregate size distribution $a_k(t)$ satisfies the generalized scaling form, and the total number $N_A(t)$ approaches a constant, while the total mass $M_A(t)$ increases with time exponentially. When $K_e = L_e$, the effects of catalyzed replication and catalyzed decline counteract each other, the evolution of the system mainly depends on the self-coagulation of species A, the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form, and both the total number and the total mass approach constants, i.e., the system will eventually evolve to a steady state. When $K_e < L_e$, the effect of catalyzed decline is stronger than that of catalyzed replication, the aggregate size distribution $a_k(t)$ takes the modified scaling form, and both the total number and the total mass decay with time exponentially.

2.2. The system with the catalytically activated reaction rates independent of the catalyst aggregates' sizes

We then discuss the case in which the catalyzed replication and the catalyzed decline rate kernels are both independent of the catalyst aggregates' sizes, $K(k,j) = K$ and $L(k,j) = Lk$. In this case, the governing rate equation (1) can be reduced to

$$\frac{\mathrm{d}a_k}{\mathrm{d}t} = \frac{I}{2}\sum_{i+j=k} a_i a_j - IN_A a_k + KN_B a_k + L[(k+1)a_{k+1} - ka_k]N_C. \quad (30)$$

Similarly to the former case, the governing rate equation (30) can be solved by using the ansatz $a_k(t) = A(t)[a(t)]^{k-1}$. Under the monodisperse initial conditions $a(0) = 0, A(0) = A_0$, we derive the solution of $a(t)$ as

$$a(t) = 1 - \frac{1}{1 + \frac{1}{2}IA_0F_C^{-1}\int_0^t F_B(t')\,\mathrm{d}t'}, \quad (31)$$

with $F_B(t) = \exp[K\int_0^t N_B(t')\,\mathrm{d}t']$ and $F_C(t) = \exp[L\int_0^t N_C(t')\,\mathrm{d}t']$.

From Eq. (4), we can easily obtain the total numbers of aggregates B and C

$$N_B(t) = 2B_0(2+J_1B_0t)^{-1}, \quad N_C(t) = 2C_0(2+J_2C_0t)^{-1}, \quad (32)$$

they both decay linearly with time as t^{-1}. So, we obtain $F_B(t) = (2+J_1B_0t)^{2K/J_1}$ and $F_C(t) = (2+J_2C_0t)^{2L/J_2}$. Then we derive the asymptotic solution of $a(t)$ in the long-time limit

$$a(t) \simeq 1 - \frac{1}{1 - C_1t^{-2L/J_2} + C_2t^{1+2(KJ_1^{-1}-LJ_2^{-1})}}, \quad (33)$$

where $C_1 = [2^{1+2KJ_1^{-1}}(J_2C_0)^{-2LJ_2^{-1}}IA_0]/[2B_0(2K+J_1)]$ and $C_2 = (J_1B_0/2)^{1+2KJ_1^{-1}}C_1$. It follows that the asymptotical expression of $a(t)$ is dominated by the value of $LJ_2^{-1} - KJ_1^{-1}$.

In the subcase of $LJ_2^{-1} - KJ_1^{-1} < 1/2$, we derive the asymptotical expression of $a(t)$ from Eq. (33) in the long-time limit as

$$a(t) \simeq 1 - C_2^{-1}t^{-[1+2(KJ_1^{-1}-LJ_2^{-1})]}. \quad (34)$$

The asymptotical solution of $A(t)$ is derived from Eq. (12) as

$$A(t) \simeq A_0 C_2^{-2} C_3 t^{-2(1+KJ_1^{-1}-LJ_2^{-1})}, \tag{35}$$

where $C_3 = (J_1 B_0)^{2KJ_1^{-1}} (J_2 C_0)^{-2LJ_2^{-1}}$. Thus, the aggregate size distribution of species A is derived as

$$\begin{aligned} a_k(t) &= A(t)a^{k-1}(t) \simeq A_0 C_2^{-2} C_3 t^{-2(1+KJ_1^{-1}-LJ_2^{-1})} \Phi(x), \\ x &= k/S(t). \end{aligned} \tag{36}$$

It shows that the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form (6) with the characteristic size $S(t) = C_2 t^{1+2(KJ_1^{-1}-LJ_2^{-1})}$ and the scaling exponents $w = 2(1 + KJ_1^{-1} - LJ_2^{-1})$, $z = 1 + 2(KJ_1^{-1} - LJ_2^{-1})$.

Moreover, the total number of species A is obtained as

$$N_A(t) \simeq A_0 C_2^{-1} C_3 t^{-1}, \tag{37}$$

which decays with time as t^{-1}. And the total mass of species A is

$$M_A(t) \simeq A_0 C_3 t^{2(KJ_1^{-1}-LJ_2^{-1})}. \tag{38}$$

The evolution behavior of the total mass $M_A(t)$ is crucially dominated by the competition between the effective catalyzed replication with rate $K_e = KJ_1^{-1}$ and the effective catalyzed decline with rate $L_e = LJ_2^{-1}$. For the $K_e > L_e$ case, the effect of the catalyzed replication is stronger than that of the catalyzed decline, so the total mass $M_A(t)$ increases with time. While in the $K_e < L_e$ case, the effect of the catalyzed decline is stronger than that of the catalyzed replication, and $M_A(t)$ decreases with time. In the marginal case of $K_e = L_e$, the effects of catalyzed replication and catalyzed decline counteract each other, the evolution of the total mass $M_A(t)$ mainly depends on the self-coagulation of species A, so it is conserved.[24]

In the subcase of $LJ_2^{-1} - KJ_1^{-1} = 1/2$, we derive the asymptotical expression of $a(t)$ in the long-time limit as

$$a(t) \simeq 1 - C_4^{-1} - C_1 C_4^{-2} t^{-2LJ_2^{-1}}, \tag{39}$$

where $C_4 = 1 + C_2$. The aggregate size distribution of species A also satisfies the conventional scaling form

$$a_k(t) \simeq A_0 C_3 C_4^{-2} (1 - C_4^{-1})^{k-1} t^{-1} \Phi(x), \quad x = k/S_A(t), \tag{40}$$

with the characteristic size $S_A(t) = C_1^{-1} C_2 C_4 t^{2LJ_2^{-1}}$.

Moreover, the total number and the total mass both are found to decay linearly with time as t^{-1},

$$N_A(t) \simeq A_0 C_3 C_4^{-1} t^{-1}, \quad M_A(t) \simeq A_0 C_3 t^{-1}. \tag{41}$$

It indicates that in this subcase, the effect of the species-C-catalyzed decline of species A becomes stronger than that in the former subcase of $LJ_2^{-1} - KJ_1^{-1} < 1/2$.

In the subcase of $LJ_2^{-1} - KJ_1^{-1} > 1/2$, we derive the asymptotical expression of $a(t)$ in the long-time limit as

$$a(t) \simeq 1 - q^{-1} + q^{-2} C_2 t^{1+2(KJ_1^{-1}-LJ_2^{-1})}, \tag{42}$$

where $q(t) = 1 - C_1 t^{-2LJ_2^{-1}} \longrightarrow 1$. The aggregate size distribution of species A is derived as

$$a_k(t) \simeq (1 - q^{-1})^k \left(\frac{IJ_1}{4K + 2J_1} \right) t^{-1} R^{-1}(t) \exp[k/R(t)], \tag{43}$$

where $R(t) = q^2(1 - q^{-1}) C_2^{-1} t^{2(LJ_2^{-1}-KJ_1^{-1})-1}$. It means that in this subcase $a_k(t)$ satisfies the modified scaling form in the long-time limit.

The total number and the total mass of species A are obtained as

$$\begin{aligned} N_A(t) &\simeq q^{-1} C_3 A_0 t^{-2(LJ_2^{-1}-KJ_1^{-1})}, \\ M_A(t) &\simeq C_3 A_0 t^{-2(LJ_2^{-1}-KJ_1^{-1})}. \end{aligned} \tag{44}$$

They both decay with time as $t^{-2(LJ_2^{-1}-KJ_1^{-1})}$, which implies that in this subcase, the species-C-catalyzed decline of species A dominates the system, and species A will die out in the end.

To sum up, in the case of the catalysis rates independent of the catalyst aggregates' sizes, the aggregate size distribution $a_k(t)$ is found to mainly depend on the competition between the self-coagulation of species A and the species-C-catalyzed decline of species A, When the self-coagulation of species A dominates the whole system ($LJ_2^{-1} - KJ_1^{-1} \leq 1/2$), the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form, and the total number $N_A(t)$ linearly decreases with time as t^{-1}, while the kinetics of the total mass $M_A(t)$ depends on the competition of the effective catalyzed replication and the effective catalyzed decline. For the $K_e > L_e$ case, the effect of catalyzed replication is stronger than that of catalyzed decline, so the total mass $M_A(t)$ increases with time. For the $K_e < L_e$ case, the effect of catalyzed decline is stronger than that of catalyzed replication and $M_A(t)$ declines with time. In the marginal case of $K_e = L_e$, the effects of catalyzed replication and catalyzed decline counteract each other, the evolution of the total mass $M_A(t)$ mainly depends on the self-coagulation of species A, and $M_A(t)$ is conserved. When the species-C-catalyzed decline of species A dominates the whole system ($LJ_2^{-1} - KJ_1^{-1} > 1/2$), the aggregate size distribution $a_k(t)$ satisfies the modified scaling form, and both the total number and the total mass decay with time as $t^{-2(LJ_2^{-1}-KJ_1^{-1})}$, so, species A will die out in the end.

3. Summary

In summary, we have proposed a three-species (A, B, and C) catalytically activated replication–decline model for the aggregation process, in which two aggregates of the same species can coagulate themselves with constant rates (I, J_1, and J_2), an A aggregate of any size can replicate itself with the help of B aggregates, otherwise, a decline of A aggregate occurs under the catalysis of C aggregates. By means of the mean-field rate equations, we derive the asymptotic solutions of the aggregate size distribution $a_k(t)$ of species A in two different cases. The kinetics of the system is illustrated in Tables 1 and 2.

From Table 1, we can derive the following conclusions. In the case of the catalyzed replication and the catalyzed decline rate kernels proportional to the catalyst aggregates' sizes,

$K(k,j) = Kj$ and $L(k,j) = Lkj$, the aggregate size distribution $a_k(t)$ mainly depends on the competition between the species-B-catalyzed replication of species A and the species-C-catalyzed decline of species A. And when the catalyzed replication process dominates the system, $a_k(t)$ satisfies the generalized scaling form; when the catalyzed decline process dominates the system, $a_k(t)$ approaches the modified scaling form; when the two catalyzed processes counteract each other, the evolution of the system mainly depends on the self-coagulation of species A, the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form.

Table 1. The kinetics of aggregate A in the case of the two catalysis rates proportional to the catalyst aggregates' sizes.

	Dominating process	Scaling behavior of $a_k(t)$
$KB_0 > LC_0$	catalyzed replication	generalized scaling
$KB_0 = LC_0$	self-coagulation	conventional scaling
$KB_0 < LC_0$	catalyzed decline	modified scaling

Table 2. The kinetics of aggregate A in the case of the two catalysis rates independent on the catalyst aggregates' sizes.

	Dominating process	Scaling behavior of $a_k(t)$
$LJ_2^{-1} - KJ_1^{-1} < 1/2$	self-coagulation	conventional scaling
$LJ_2^{-1} - KJ_1^{-1} = 1/2$	self-coagulation	conventional scaling
$LJ_2^{-1} - KJ_1^{-1} > 1/2$	catalyzed decline	modified scaling

From Table 2, we find that in the case of the catalyzed replication and the catalyzed decline rate kernels independent of the catalyst aggregates' sizes, $K(k,j) = K$ and $L(k,j) = Lk$, the aggregate size distribution $a_k(t)$ mainly depends on the competition between the self-coagulation of species A and the species-C-catalyzed decline of species A. And when the self-coagulation of species A dominates the system, the aggregate size distribution $a_k(t)$ satisfies the conventional scaling form; when the catalyzed decline process dominates the system, $a_k(t)$ approaches the modified scaling form.

This model is expected to provide some useful theoretical predictions for the evolution of aggregation growth in some practical biological, chemical, and economical systems with catalysis-driven replication and decline.

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