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A solvable two-species catalysis-driven aggregation model

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Abstract

We study the kinetics of a two-species catalysis-driven aggregation system, in which an irreversible aggregation between any two clusters of one species occurs only with the catalytic action of another species. By means of a generalized mean-field rate equation, we obtain the asymptotic solutions of the cluster mass distributions in a simple process with a constant rate kernel. For the case without any consumption of the catalyst, the cluster mass distribution of either species always approaches a conventional scaling law. However, the evolution behaviour of the system in the case with catalyst consumption is complicated and depends crucially on the relative data of the initial concentrations of the two species.

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1. Introduction

The aggregation phenomenon occurs in many natural processes [1, 2]. In the past few decades, considerable works have been devoted to understanding the kinetics of the aggregation processes [3–5]. Many theoretical investigations exhibited that the kinetic behaviour of the aggregation process may obey a scaling law in the long-time limit [6–9]. In general, most of these works focused on the self-coalescence processes in which the aggregation reaction spontaneously occurs between any two clusters of the same species, and a few studies paid attention to the aggregation processes that occur only in the presence of another substance. Kang and Redner [10] introduced a particle coalescence model (PCM), in which the clusters are defined to be single lattice sites and the aggregation or annihilation reaction occurs whenever two or more clusters occupy the same lattice site. In fact, the PCM implies that the aggregation or annihilation reaction occurs only in the presence of the lattice site, namely, the 'immobile catalyst', and it may thus be considered as a catalytically activated aggregation model. Spouge solved the PCM in one dimension and presented a useful method for the

solution of the diffusion–reaction problem [11]. Krapivsky investigated the aggregation–annihilation analogue of the PCM in one dimension and found that the kinetic behaviour of the system is dependent on the whole charge density and the spatial dimensionality [12]. Burlatsky *et al* [13] and Oshanin *et al* [14] introduced the three-molecule catalytically activated reaction model (CARM) and analysed the kinetics of the processes by using an extension of the Smoluchowski approach. Recently, the kinetic and equilibrium behaviours of the CARM have been incisively investigated [15, 16]. The catalytically activated process can contribute to produce the required products from a species which is chemically stable under normal conditions, and it is thus of great practical and theoretical significance. In our previous work, we proposed a single-species model in which the aggregation of the clusters is driven by the catalyst and analysed the scaling behaviour of the cluster mass distribution in several different cases [17].

This work aims at studying the kinetics of a two-species catalysis-driven aggregation model in which the aggregation of one species occurs only in the presence of another species. The first elementary reaction step of our aggregation scheme is a reversible reaction, $A_i + B_{i'} \rightleftharpoons A_i B_{i'}$, and the second step consists of two simultaneous irreversible reactions, $A_i B_{i'} + A_j \rightarrow A_{i+j} + B^*_{i'}$ and $A_i B_{i'} + B_{j'} \rightarrow B_{i'+j'} + A^*_i$. Here, both A and B species are chemically stable species that cannot spontaneously coagulate by themselves but either species is the catalyst for the aggregation of the other, and A^*_i and $B^*_{i'}$ are the products of the above catalytic reactions. In contrast with the irreversible reaction, the reversible one may reach its steady state with a very rapid rate. Thus, the catalytic reaction may be controlled only by the second step of our aggregation scheme. The whole catalytic reactions then read

$$A_i + A_j + B_{i'} \xrightarrow{K_A(i;j;i')} A_{i+j} + B^*_{i'} \qquad B_{i'} + B_{j'} + A_i \xrightarrow{K_B(i';j';i)} B_{i'+j'} + A^*_i$$

where $K_A(i;\, j;\, i')$ and $K_B(i';\, j';\, i)$ are the equivalent catalytic reaction rates. It is well known that after the catalytic reaction, the product of the catalyst can still keep the catalytic ability in some situations while it may lose the ability for other cases. In this work, we thoroughly investigate the dependence of the kinetics of the aggregation system on the catalyst.

The present investigation is performed in the mean-field limit. The mean-field approach to the reaction process assumes that the reaction proceeds at a rate proportional to the reactant concentrations. On the other hand, the mean-field assumption neglects the spatial fluctuation of the reactant densities and, therefore, applies to the case in which the spatial dimension d of the system is equal to or greater than a critical dimension d_c. As for the $d < d_c$ case, fluctuations in the reactant densities may lead to a diffusion-controlled kinetics in the long-time limit. It is found that for an irreversible aggregation system, $d_c = 2$ [10]. In our catalysis-driven aggregation model, the dominant reaction is also an irreversible binary aggregation-like reaction, and it is thus sound that for our model, the critical dimension d_c is also equal to 2. For simplicity, the spatial dimension d of our system is assumed to be greater than 2 and the mean-field approach is thus valid.

It is believed that catalysis-driven aggregation processes are of interest in studying the scaling properties of their evolution behaviour. Based on the mean-field rate equations of the catalysis-driven aggregation processes, we have derived the asymptotic solutions of the cluster mass distributions. The results showed that the kinetics of our model depends strongly on whether or not the catalyst is consumed through the catalytic reaction. In the case without any consumption of the catalyst, the cluster mass distribution of either species satisfies the conventional scaling law; while for the case with catalyst consumption, the evolution of the clusters may fall in a peculiar scaling regime. On the other hand, the results have also exhibited that the kinetic behaviour of this model is quite different from that of the conventional self-aggregation systems [3–5].

The paper is organized as follows. In section 2, we describe an irreversible catalysis-driven aggregation model without any consumption of the catalyst and investigate the corresponding mean-field rate equations to obtain the cluster mass distributions. In section 3, we then investigate the evolution behaviour of an irreversible catalysis-driven aggregation model with catalyst consumption. A brief summary is given in section 4.

2. The model without any consumption of the catalyst

We first investigate the catalysis-driven aggregation model in which the catalyst still keeps its catalytic ability after the reaction. So both species are not consumed in this system. The concentrations of A and B clusters of k-mers are respectively denoted as a_k and b_k. Here, we consider a simple constant-reaction-rate model, in which the catalytic aggregation rates $K_A(i;j;i')$ and $K_B(i';j';i)$ are equal to I_1 and I_2, respectively. The theoretical approach to this aggregation process is then based on the mean-field rate equations

$$\begin{aligned}\frac{\mathrm{d}a_k}{\mathrm{d}t} &= \frac{I_1}{2}\sum_{i+j=k}\sum_{l=1}^{\infty} a_i a_j b_l - I_1 a_k \sum_{j=1}^{\infty}\sum_{l=1}^{\infty} a_j b_l \\ \frac{\mathrm{d}b_k}{\mathrm{d}t} &= \frac{I_2}{2}\sum_{i+j=k}\sum_{l=1}^{\infty} b_i b_j a_l - I_2 b_k \sum_{j=1}^{\infty}\sum_{l=1}^{\infty} b_j a_l.\end{aligned} \tag{1}$$

Let $N_A(t)$ and $N_B(t)$ respectively denote the total numbers of A clusters and B clusters, i.e., $N_A(t)=\sum_{k=1}^{\infty} a_k(t)$ and $N_B(t)=\sum_{k=1}^{\infty} b_k(t)$. By summing up equations (1) we obtain

$$\frac{\mathrm{d}N_A}{\mathrm{d}t} = -\frac{I_1}{2}N_A^2 N_B \qquad \frac{\mathrm{d}N_B}{\mathrm{d}t} = -\frac{I_2}{2}N_A N_B^2. \tag{2}$$

Under the initial condition of $N_A(0)=A_0$ and $N_B(0)=B_0$, from equations (2) we derive the exact solutions

$$N_A(t) = A_0(C_1 t+1)^{-I_1/(I_1+I_2)} \qquad N_B(t) = B_0(C_1 t+1)^{-I_2/(I_1+I_2)} \tag{3}$$

where $C_1=(I_1+I_2)A_0B_0/2$. Equations (3) show that in the long-time limit, the total number of A clusters decays as $t^{-I_1/(I_1+I_2)}$ while that of B clusters decays as $t^{-I_2/(I_1+I_2)}$. We then turn to determine the cluster mass distributions. Introducing two generating functions,

$$G_A(z,t)=\sum_{j=1}^{\infty} z^j a_k(t) \qquad G_B(z,t)=\sum_{j=1}^{\infty} z^j b_j(t) \tag{4}$$

we transform the governing rate equations (1) into the following equations:

$$\frac{\mathrm{d}G_A}{\mathrm{d}t} = \frac{I_1}{2}G_A^2 N_B - I_1 G_A N_A N_B \qquad \frac{\mathrm{d}G_B}{\mathrm{d}t} = \frac{I_2}{2}G_B^2 N_A - I_2 G_B N_A N_B. \tag{5}$$

These Bernoulli equations can be directly solved to yield

$$\begin{aligned}G_A(z,t) &= \frac{G_{A0}(z)E_1(t)}{1-\frac{I_1}{2}G_{A0}(z)\int_0^t E_1(t')N_B(t')\,\mathrm{d}t'} \\ G_B(z,t) &= \frac{G_{B0}(z)E_2(t)}{1-\frac{I_2}{2}G_{B0}(z)\int_0^t E_2(t')N_A(t')\,\mathrm{d}t'}\end{aligned} \tag{6}$$

with the shorthand notation $G_{A0}(z)\equiv G_A(z,t=0)$, $G_{B0}(z)\equiv G_B(z,t=0)$, $E_1(t)\equiv \exp\left[-\int_0^t I_1 N_A(t')N_B(t')\,\mathrm{d}t'\right]$ and $E_2(t)\equiv \exp\left[-\int_0^t I_2 N_A(t')N_B(t')\,\mathrm{d}t'\right]$. Substituting equations (2) into the equations of $E_1(t)$ and $E_2(t)$, one can find $E_1(t)=(C_1t+1)^{-2I_1/(I_1+I_2)}$ and $E_2(t)=(C_1t+1)^{-2I_2/(I_1+I_2)}$.

Equations (6) represent the general solutions of equations (5) under arbitrary initial conditions. Now we consider a simple but important case, in which there only exist the monomer clusters of A and B species at $t = 0$ and their concentrations respectively equal to A_0 and B_0. Then the monodisperse initial conditions are

$$a_k(0) = A_0\delta_{k1} \qquad b_k(0) = B_0\delta_{k1}. \tag{7}$$

It is obvious that under the above initial conditions, $G_{A0}(z) = A_0 z$ and $G_{B0}(z) = B_0 z$. Expanding the resulting functions (6), one can then derive the following exact solutions:

$$\begin{aligned} a_k(t) &= A_0 E_1(t)\left[\frac{I_1 A_0}{2}\int_0^t E_1(t')N_B(t')\,\mathrm{d}t'\right]^{k-1} \\ b_k(t) &= B_0 E_2(t)\left[\frac{I_2 B_0}{2}\int_0^t E_2(t')N_A(t')\,\mathrm{d}t'\right]^{k-1}. \end{aligned} \tag{8}$$

Thus we obtain the exact solutions of the cluster mass distributions

$$\begin{aligned} a_k(t) &= A_0(C_1 t+1)^{-2I_1/(I_1+I_2)}[1-(C_1 t+1)^{-I_1/(I_1+I_2)}]^{k-1} \\ b_k(t) &= B_0(C_1 t+1)^{-2I_2/(I_1+I_2)}[1-(C_1 t+1)^{-I_2/(I_1+I_2)}]^{k-1}. \end{aligned} \tag{9}$$

Introducing two scaling variables, $x = k(C_1 t)^{-I_1/(I_1+I_2)}$ and $y = k(C_1 t)^{-I_2/(I_1+I_2)}$, we can further rewrite equations (9) as follows:

$$a_k(t) \simeq A_0(C_1 t)^{-2I_1/(I_1+I_2)}\exp(-x) \qquad b_k(t) \simeq B_0(C_1 t)^{-2I_2/(I_1+I_2)}\exp(-y) \tag{10}$$

which are valid in the region of $k \gg 1$ and $t \gg 1$.

Generally, a function $S(t)$ is used to denote the characteristic average mass of the clusters in such an aggregation process and the concentration $c_k(t)$ of the k-mer clusters at large times can be written in the following scaling form [6]:

$$c_k(t) \simeq t^{-w}\Phi[k/S(t)] \qquad S(t) \propto t^z. \tag{11}$$

Here, the two governing exponents w and z are universally used to describe the scaling nature of the aggregation system in the long-time limit. Equations (10) indicate that for this case, the cluster mass distribution of either species approaches the conventional scaling form of equations (11) at large times and the scaling exponents are

$$\begin{aligned} w &= \frac{2I_1}{I_1+I_2} \qquad z = \frac{I_1}{I_1+I_2} \qquad \text{for } A \text{ clusters} \\ w &= \frac{2I_2}{I_1+I_2} \qquad z = \frac{I_2}{I_1+I_2} \qquad \text{for } B \text{ clusters.} \end{aligned} \tag{12}$$

These exponents are nonuniversal and dependent on the ratio between the aggregation rates I_1 and I_2. When $I_1 = I_2$, the two species have the same scaling exponents, and they thus evolve in similar scaling regimes, which are independent of the relative data of the initial concentrations A_0 and B_0. Moreover, the typical mass $S(t)$ of species A grows as $t^{I_1/(I_1+I_2)}$ while that of species B grows as $t^{I_2/(I_1+I_2)}$.

On the other hand, it is not difficult to find that both species obey the mass conservation law, $M_A(t) = \sum_{k=1}^{\infty} k a_k(t) \equiv A_0$ and $M_B(t) = \sum_{k=1}^{\infty} k b_k(t) \equiv B_0$. This is natural for our system without any consumption of either species.

3. The model with catalyst consumption

We now investigate the catalysis-driven aggregation process with catalyst consumption, in which both the reaction products A_i^* and $B_{i'}^*$ lose their catalytic abilities through the reaction

and then withdraw from the process. For simplicity, we also assume that the details of the initial concentrations and the rate kernels for this model are the same as those for the above model in section 2. The governing mean-field rate equations for this process then read

$$\begin{aligned}\frac{\mathrm{d}a_k}{\mathrm{d}t} &= \frac{I_1}{2}\sum_{i+j=k}\sum_{l=1}^{\infty} a_i a_j b_l - I_1 a_k \sum_{j=1}^{\infty}\sum_{l=1}^{\infty} a_j b_l - \frac{I_2}{2} a_k \sum_{i,j=1}^{\infty} b_i b_j \\ \frac{\mathrm{d}b_k}{\mathrm{d}t} &= \frac{I_2}{2}\sum_{i+j=k}\sum_{l=1}^{\infty} b_i b_j a_l - I_2 b_k \sum_{j=1}^{\infty}\sum_{l=1}^{\infty} b_j a_l - \frac{I_1}{2} b_k \sum_{i,j=1}^{\infty} a_i a_j .\end{aligned} \tag{13}$$

Summing up equations (13), we obtain

$$\frac{\mathrm{d}N_A}{\mathrm{d}t} = -\frac{I_1}{2}N_A^2 N_B - \frac{I_2}{2}N_A N_B^2 \qquad \frac{\mathrm{d}N_B}{\mathrm{d}t} = -\frac{I_2}{2}N_A N_B^2 - \frac{I_1}{2}N_A^2 N_B. \tag{14}$$

From equations (14) we derive the following equations:

$$\begin{aligned} N_B &= N_A - (A_0 - B_0) \\ \frac{\mathrm{d}N_A}{\mathrm{d}t} &= -\frac{I_1 + I_2}{2} N_A (N_A - A_0 + B_0)\left[N_A - \frac{I_2(A_0 - B_0)}{I_1 + I_2}\right].\end{aligned} \tag{15}$$

Equations (15) imply that the quantity $Q = N_A - N_B$ is conserved by the dynamics of this model. For the symmetrical initial case, $A_0 = B_0 = C_0$, from equations (15) we obtain the exact solutions

$$N_A(t) = N_B(t) = C_0(C_2 t + 1)^{-1/2} \tag{16}$$

where $C_2 = (I_1 + I_2)C_0^2$. Equation (16) indicates that for the symmetrical case, the total number of each species decays as $t^{-1/2}$ in the long-time limit. We then consider the asymmetrical initial case. Without any loss of generality, we set $A_0 > B_0$. From equations (15) one can determine the asymptotic solutions of $N_A(t)$ and $N_B(t)$ in the long-time limit,

$$N_A(t) \simeq C_3 + C_4 \exp(-\gamma t) \qquad N_B(t) \simeq C_4 \exp(-\gamma t) \tag{17}$$

where $C_3 = A_0 - B_0$, $C_4 = B_0 A_0^{I_1/I_2} C_3 [I_1/(I_1 A_0 + I_2 B_0)]^{(I_1+I_2)/I_2}$ and $\gamma = I_1 C_3^2/2$. Equations (17) indicate that for the asymmetrical case, the total number of species B decays exponentially with time while that of species A may retain a certain value C_3 in the long-time limit.

We then turn to derive the solutions of the cluster mass distributions from equations (13). Introducing the generating functions (4), we transform equations (13) into the following equations:

$$\begin{aligned}\frac{\mathrm{d}G_A}{\mathrm{d}t} &= \frac{I_1}{2}G_A^2 N_B - I_1 G_A N_A N_B - \frac{I_2}{2} G_A N_B^2 \\ \frac{\mathrm{d}G_B}{\mathrm{d}t} &= \frac{I_2}{2}G_B^2 N_A - I_2 G_B N_A N_B - \frac{I_1}{2} G_B N_A^2 .\end{aligned} \tag{18}$$

The Bernoulli equations (18) can be directly solved to yield similar resulting generating functions (6) with the different shorthand notation $E_1(t) \equiv \exp\{-\int_0^t \mathrm{d}t'[I_1 N_A(t') N_B(t') + I_2 N_B^2(t')/2]\}$ and $E_2(t) \equiv \exp\{-\int_0^t \mathrm{d}t'[I_2 N_A(t') N_B(t') + I_1 N_A^2(t')/2]\}$. These modified equations can represent the general solutions for the system with the catalyst consumption under arbitrary initial conditions. By making use of equations (14) we can obtain the following equations:

$$\begin{aligned} E_1(t) &= \frac{N_A(t) F_1(t)}{A_0} \qquad \int_0^t E_1(t') N_B(t')\,\mathrm{d}t' = \frac{2[1 - F_1(t)]}{I_1 A_0} \\ E_2(t) &= \frac{N_B(t) F_2(t)}{B_0} \qquad \int_0^t E_2(t') N_A(t')\,\mathrm{d}t' = \frac{2[1 - F_2(t)]}{I_2 B_0}\end{aligned} \tag{19}$$

with the shorthand notation $F_1(t) \equiv \{[(I_1 + I_2)N_A(t) + I_2(B_0 - A_0)]/(I_1A_0 + I_2B_0)\}^{I_1/(I_1+I_2)}$ and $F_2(t) \equiv \{[(I_1 + I_2)N_B(t) + I_1(A_0 - B_0)]/(I_1A_0 + I_2B_0)\}^{I_2/(I_1+I_2)}$. Under the monodisperse initial conditions (7), one can also expand the modified resulting functions (6) to derive the same solutions (8) of the cluster mass distributions for this case.

In the symmetrical initial case, we obtain the scaling solutions of the cluster mass distributions

$$\begin{aligned} a_k(t) &\simeq C_0(C_2t)^{-(2I_1+I_2)/(2I_1+2I_2)} \exp(-x) \qquad x = k(C_2t)^{-I_1/(2I_1+2I_2)} \\ b_k(t) &\simeq C_0(C_2t)^{-(I_1+2I_2)/(2I_1+2I_2)} \exp(-y) \qquad y = k(C_2t)^{-I_2/(2I_1+2I_2)}. \end{aligned} \tag{20}$$

Equations (20) show that both species scale according to the conventional form (11) and the scaling exponents are

$$\begin{aligned} w &= \frac{2I_1 + I_2}{2I_1 + 2I_2} \qquad z = \frac{I_1}{2I_1 + 2I_2} \qquad \text{for } A \text{ clusters} \\ w &= \frac{I_1 + 2I_2}{2I_1 + 2I_2} \qquad z = \frac{I_2}{2I_1 + 2I_2} \qquad \text{for } B \text{ clusters} \end{aligned} \tag{21}$$

which are nonuniversal and depend on the ratio of the aggregation rates. In this case, the typical mass of species A grows as $t^{I_1/2(I_1+I_2)}$ while that of species B grows as $t^{I_2/2(I_1+I_2)}$. On the other hand, we also find that the total mass of species A decays as $t^{-I_2/2(I_1+I_2)}$ while that of species B decays as $t^{-I_1/2(I_1+I_2)}$. For this case, both species cannot survive at the end.

In the asymmetrical initial case, we determine the asymptotic solutions of the cluster mass distributions as follows:

$$\begin{aligned} a_k(t) &\simeq C_6(1 - C_5)^k \exp(-x) \qquad x = C_7k \exp(-\gamma t) \\ b_k(t) &\simeq C_9(1 - C_8)^k \, \mathrm{e}^{-\gamma t} \exp(-y) \qquad y = C_{10}k \exp(-\gamma t) \end{aligned} \tag{22}$$

where $C_5 = [I_1C_3/(I_1A_0 + I_2B_0)]^{I_1/(I_1+I_2)}$, $C_6 = C_3C_5$, $C_7 = C_4C_5/C_3(1 - C_5)$, $C_8 = [I_1C_3/(I_1A_0 + I_2B_0)]^{I_2/(I_1+I_2)}$, $C_9 = C_4C_8$ and $C_{10} = I_2C_9/I_1C_3(1 - C_8)$. The results indicate that the conventional scaling description of the cluster mass distribution breaks down for either species and both species scale only according to the following modified form [9]:

$$c_k(t) \simeq h^k[f(t)]^{-w}\Phi[k/S(t)] \qquad S(t) \propto [f(t)]^z \tag{23}$$

where h is a constant ($0 < h < 1$) and $f(t)$ is an unusual function of time, such as e^t, $\ln t$, 2^t, and so on. In this case, the scaling function is exponential, $f(t) = \exp(t)$. The governing exponents for species A are $w = 0$ and $z = I_1(A_0 - B_0)^2/2$, while those for species B are $w = z = I_1(A_0 - B_0)^2/2$. These indicate that for the asymmetrical case, the scaling properties of the cluster mass distributions depend crucially on the reaction rate of species A as well as the difference between the initial concentrations A_0 and B_0. On the other hand, the modified scaling form (23) also indicates that two different mass scales, a growing scale and a time-independent one, are associated with either species. From equations (22) we find that both species have the same growing scale $S(t) \sim \mathrm{e}^{\gamma t}$, which is forced by the catalytic reactions. Meanwhile, the time-independent scale for species A is $S_A = 1/C_5$ and that for species B is $S_B = 1/C_8$, which dominate the evolution behaviour of the two species in the long-time limit. Moreover, it is also instructive to make a comparison between the total mass of species A and that of species B,

$$M_A(t) = \sum_{k=1}^{\infty} ka_k \simeq \frac{A_0 - B_0}{C_5} \qquad M_B(t) = \sum_{k=1}^{\infty} kb_k \simeq C_8^{-2}C_9 \, \mathrm{e}^{-\gamma t} \qquad \text{at} \quad t \gg 1 \tag{24}$$

which show that the total mass of species B decays rapidly as $\mathrm{e}^{-\gamma t}$ while that of species A will always retain a certain value after a long time. Hence, only species A, which has the larger initial concentration, can survive at the end.

4. Summary

We have investigated the kinetics of the two-species catalysis-driven aggregation processes with a constant-reaction-rate kernel. Based on the mean-field theory, we have analysed the dependence of the kinetics of the system on the catalysis. The results exhibited that whether or not the catalytic clusters are consumed through the reaction plays an important role in the kinetics of the system.

In the first model without any consumption of the catalytic clusters, we found that the evolution behaviour of the cluster mass distribution of either species obeys a conventional scaling law. The scaling exponents are some nonuniversal constants dependent on the ratio between the catalytic reaction rates I_1 and I_2. Moreover, the total number of either species decays with time while the total mass is always conserved by the dynamics of the system.

In the second model with catalyst consumption, the kinetics of the system depends strongly on the initial concentrations. For the symmetrical initial case, the cluster mass distribution of either species approaches the conventional scaling form with nonuniversal governing exponents; moreover, the total number and the total mass of either species decay with time and no species can survive at the end. For the asymmetrical initial case, both species scale according to the modified form and the scaling exponents are dependent on the initial concentration difference and the reaction rate of the heavy species with the larger initial data; moreover, both the total number and the total mass of the heavy species can respectively retain a certain value in the long-time limit while those of the light species always decay exponentially with time.

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