Large-time behavior of the generalized Smoluchovski coagulation equations

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(Received 7 September 1994)

We study the large-time asymptotic solutions of the generalized Smoluchovski equations for class I and class II coagulation systems. It is found that, in gelling and nongelling systems of class I, the general solution $c_k(t)$ approaches for $t \to \infty$ the exact solution Cb_k/t (k finite), where the b_k are independent of the initial conditions $c_k(0)$ and can be determined from a recursion relation. In class II systems, if the k and t dependence of $c_k(t)$ factorizes for large time, i.e., $c_k(t) \to c_1(t) b_k$ ($t \to \infty, k$ finite), then the b_k ($\sim k^{-\tau}$) can be obtained from a recursion relation. We show that if $c_k(t)$ is factorizable at large time, then the scaling function method and the recursion relation method give the same result for the τ exponent for the class II systems.

PACS number(s): 05.40.+j, 68.70.+w, 03.20.+i, 05.20.Dd

I. INTRODUCTION

The kinetics of irreversible aggregation and clustering phenomena, in particular the time evolution of cluster size distribution $c_k(t)$ has been studied extensively by using the Smoluchovski coagulation equation [1–7]

$$\dot{c}_k = \frac{1}{2} \sum_{i+j=k} K(i,j) c_i c_j - c_k \sum_{j=1}^{\infty} K(k,j) c_j \ , \qquad (1)$$

where the coagulation kernel $K(i,j)$ represents the rate coefficient for a specific clustering mechanism between clusters of sizes i and j. The large-time asymptotic behavior of the Smoluchovki equation has been studied for the following homogeneous kernels

$$K(ai,aj) = a^{\lambda} K(i,j) \ , \qquad (2)$$

$$K(i,j) \simeq i^{\mu} j^{\nu}, \quad j \gg i, \quad \lambda = \mu + \nu \ , \qquad (3)$$

$$K(x, 1-x) \simeq x^{\mu}[1 + k_1 x^{\mu'} + \cdots] \ , \quad x \to 0, \quad \mu' > 0 \ . \qquad (4)$$

To study long-time properties, two different methods have been used in literature. One is the scaling function method [4,5] the other is the recursion relation method [6,7]. The scaling function method describes the dominant time dependence at large t, whereas the recursion relation method gives the limiting behavior $c_k(t)/c_1(t)$ as $t \to \infty$, but not the approach toward this behavior. It is found that for nongelling class I systems, the large-time behavior predicted from the recursion relation method is in agreement with the results of the scaling function method. However, for general class II systems, the two methods seem to lead to different results [8].

Recently, the generalized Smoluchovski equation (GSE) has been introduced to study the n-tuple coagulation in some idealized dense gas systems, from a mean-field point of view [9–11]. The GSE is given by

$$\dot{c}_k = \frac{1}{n!} \sum_{i_1+i_2+\cdots+i_n=k} K(i_1,i_2,\ldots,i_n) c_{i_1} c_{i_2} \cdots c_{i_n} - \frac{c_k}{(n-1)!} \sum_{i_1,i_2,\ldots,i_{n-1}=1}^{\infty} K(i_1,i_2,\ldots,i_{n-1},k) \times c_{i_1} c_{i_2} \cdots c_{i_{n-1}} \ , \qquad (5)$$

where the coagulation kernel $K(i_1,i_2,\ldots,i_n)$ represents the rate coefficient for a specific aggregation mechanism among n clusters of sizes $i_1, i_2, .., i_{n-1}$, and i_n. Their determination depends on the very particular model of the molecular process involved, and may be obtained by using the relevant fluid theories for some simplified model systems. In the real coagulation system, if the probability of many-body collision is appreciable one should consider a combined n-tuple coagulation processes with all possible $n \geq 2$. The GSE is used to study an isolated many-body coagulation, which is assumed to be the dominant process in the real systems, under favorable conditions. It is found that, when special reaction kernels are considered, GSE can be studied by the standard methods developed for the Smoluchovski equation, and its solutions exhibit properties quantitatively similar to those of the Smoluchovski equation. However, it appears to be difficult to study the GSE for general reaction kernels. So, in this work, we restrict ourselves to the homogeneous kernels.

The purpose of this paper is (1) to study the large-time behavior of GSE for general homogeneous kernels of nongelling class I and class II systems by using the recursion relation method and (2) to show that the recursion relation method and scaling function method give complementary conclusions for class II systems. Our conjecture is that since the scaling theory shows that the k and t dependence of $c_k(t)$ factorizes as $t \to \infty$ for class I and class II systems [4], then the recursion relation method applies and the τ exponent, defined as $c_k(t) \sim c_1(t) k^{-\tau}$, calculated from those two methods should be the same. Thus, combining the results of two different methods we

1063-651X/95/51(3)/1757(5)/$06.00

have

$$\tau=\begin{cases}1+\lambda, & \text{class I}\\ 1+\lambda/2, & \text{class II}\end{cases}, \tag{6}$$

for nongelling systems.

The paper is organized as follows. In Sec. II, we derive the recursion relation from the generalized Smoluchovski equation and show that if the cluster size distribution $c_k(t)$ can factorize at large time, then the recursion relation describes their large-time behavior. In Sec. II, we discuss the large-time behavior from the recursion relation for class I and class II systems. In Sec. IV, we conclude the work by discussing the conjecture, which states that the large-time behavior predicted from two different methods are the same.

II. DERIVATION OF THE RECURSION RELATION

In this section we derive the recursion relation from the generalized Smoluchovski Eq. (5) for the following homogeneous kernels

$$\begin{gathered}K(ai_1,ai_2,\ldots,ai_n)=a^{\lambda}K(i_1,i_2,\ldots,i_n)\;,\\ K(i_1,i_2,\ldots,i_n)\simeq(i_1i_2\cdots i_{n-1})^{\nu}i_n^{\mu}\;,\\ [i_1,i_2,\ldots,i_{n-1}\gg i_n,\ \lambda=(n-1)\nu+\mu]\;,\end{gathered} \tag{7}$$

which represent reaction rates among $n-1$ large and small clusters, and are closely related with the standard classification for binary reaction kernel. In fact, this kernel reduces to the binary reaction kernel for standard Smoluchovski equation if one lets $n=2$. As in the standard scaling theory for the Smoluchovski coagulation equation, we define that $\mu>0$ corresponds to class I, $\mu=0$ to class II, and $\mu<0$ to class III. Note that there are two physical restrictions on the exponents: For n large interpenetrable clusters $K(j,j,\ldots,j)\sim j^n$, which is an upper bound for all $K(j,j,\ldots,j)$ as $j\to\infty$, and thus $\lambda\le n$. Since a j-mer contains at most j monomers, it is required that $\nu\le 1$. There is no restriction imposed on μ except $\mu\le\lambda-(n-1)\nu$. In class I and III, the rate constants for reactions of one large sample with other $n-1$ small and large samples are dominant, respectively. In class II, the reaction rates are the same for large-large and large-small clusters reactions. Nongelling systems correspond to $\lambda\le n-1$, and gelling systems to $n-1\le\lambda\le n$ [9,11].

First we consider the gelling systems, for which there exists a special postgel solution $c_k(t)=c_1(t)b_k$. It can be shown that the size distribution in gelling systems exhibits its universal behavior, independent of the initial distribution $c_k(0)$, namely it approaches the special solution in the following sense

$$\lim_{t\to\infty}c_k(t)/c_1(t)=b_k\;, \tag{8}$$

where b_k $(k=1,2,\ldots,)$ are bounded positive numbers with $b_1=1$.

Since GSE admits an exact solution with a simple time dependence [11]

$$c_k(t)=c_k(t_c)/[1+\beta(t-t_c)]^{1/(n-1)}\equiv b_kc_1(t)\;, \tag{9}$$

where $b_k=c_k(t_c)/c_1(t_c)$ are positive numbers and β and t_c unknown constants, which are related with a set of initial conditions $c_k(0)$. Inserting this solution into Eq. (6) yields

$$\begin{aligned}-\frac{\beta}{n-1}\frac{b_k}{c_1(t_c)^{n-1}}=&\frac{1}{n!}\sum_{i_1+i_2+\cdots+i_n=k}K(i_1,i_2,\ldots,i_n)b_{i_1}b_{i_2}\cdots b_{i_n}\\&-\frac{b_k}{(n-1)!}\sum_{i_1,i_2,\ldots,i_{n-1}-1}K(i_1,i_2,..,i_{n-1},k)b_{i_1}b_{i_2}\cdots b_{i_{n-1}}\;.\end{aligned} \tag{10}$$

By using Eq. (10) for $k=1$, we can eliminate the unknown constant β

$$-\frac{\beta}{n-1}\frac{b_1}{c_1(t_c)^{n-1}}=-\frac{b_1}{(n-1)!}\sum_{i_1,i_2,\ldots,i_{n-1}=1}K(i_1,i_2,\ldots,i_{n-1},k)b_{i_1}b_{i_2}\cdots b_{i_{n-1}} \tag{11}$$

or

$$\beta=\frac{(n-1)c_1(t_c)^{n-1}}{(n-1)!}\sum_{i_1,i_2,\ldots,i_{n-1}=1}^{\infty}K(i_1,i_2,\ldots,i_{n-1},k)b_{i_1}b_{i_2}\cdots b_{i_{n-1}}\;. \tag{12}$$

Thus, the recursion relation is given by

$$\begin{aligned}R(b_k)\equiv&\frac{1}{n!}\sum_{i_1+i_2+\cdots+i_n=k}K(i_1,i_2,\ldots,i_n)b_{i_1}b_{i_2}\cdots b_{i_n}\\&-\frac{b_k}{(n-1)!}\sum_{i_1,i_2,\ldots,i_{n-1}=1}^{\infty}[K(i_1,i_2,\ldots,i_{n-1},k)-K(i_1,i_2,\ldots,1)]b_{i_1}b_{i_2}\cdots b_{i_{n-1}}\;.\end{aligned} \tag{13}$$

As we shall show later on, the solution b_k of the recursion relation at large k, has algebraic k dependence, i.e.,

$$b_k \simeq B k^{-\tau}, \quad k \to \infty \ . \tag{14}$$

This relation defines the τ exponent obtained from the recursion relation, which is the quantity of main interest in this work. Notice that the exact solution (9) in nongelling systems ($\lambda < n-1$) corresponds to an infinite sol mass, and therefore is physically unacceptable. However, asymptotic solutions of the form (14) could be physically acceptable in nongelling systems, because $c_k(t)/c_1(t)$ may approach b_k nonuniformly in k, so that $\sum_k k c_k(t) = 1$, whereas $\sum_k k b_k \to \infty$ since $\tau < 2$.

As a criterion to decide whether asymptotic solutions b_k of form (8) can be determined from recursion relation (13), we use the results from the scaling function method. The most important results of the scaling theory are $\phi(x) \sim \exp(-x^{-|\mu|})$ $(x \to 0)$ for class III systems ($\mu < 0$), and $\phi(x) \sim x^{-\tau}$ $(x \to 0)$ for class I and class II systems. These results imply that class III systems do not admit solutions, satisfying Eq. (8), because $c_k(t)/c_1(t) \to \infty$ ($t \to \infty$; k,j fixed, but large), whereas class I and class II systems admit such solutions since $c_k(t)/c_1(t) \sim (j/k)^{\tau} = \text{const}$ ($t \to \infty$; k,j fixed, but large). It should be noticed that if the k and t dependence of $c_k(t)$ cannot factorize completely, one has, in general.

$$\nu_k(t) = c_k(t)/c_1(t) \ .$$

If this is the case, it has been shown that the class II systems do not admit solutions that satisfy the recursion relation [8].

III. ASYMPTOTIC SOLUTIONS FROM THE RECURSION RELATION

In this section we show that for nongelling models ($\lambda < n-1$) of class I ($\mu > 0$) and class II ($\mu = 0$) systems, the limiting ratio $b_k = \lim_{t \to \infty} c_k/c_1$ satisfies the recursion relation (13) provided that b_k satisfies the strict inequalities

$$E_1 < E_k < \infty, \quad k = 2,3,.., \tag{15}$$

where E_k is defined by

$$\begin{aligned} E_k &= \lim_{t \to \infty} \frac{1}{(n-1)!} \sum_{i_1, i_2, \ldots, i_{n-1}=1}^{\infty} K(i_1, i_2, \ldots, i_{n-1}, k) c_{i_1}(t) c_{i_2}(t) \cdots c_{i_{n-1}}(t)/c_1(t)^{n-1} \\ &= \frac{1}{(n-1)!} \sum_{i_1, i_2, \ldots, i_{n-1}=1}^{\infty} K(i_1, i_2, \ldots, i_{n-1}, k) b_{i_1} b_{i_2} \cdots b_{i_{n-1}} , \end{aligned} \tag{16}$$

where it is assumed that the infinite sums converge.

In order to show that GSE reduces to the recursion relation (13) as $t \to \infty$, we introduce

$$\sigma_k(t) = \frac{1}{(n-1)!} \sum_{i_1, i_2, \ldots, i_{n-1}=1}^{\infty} K(i_1, i_2, \ldots, i_{n-1}, k) c_{i_1} c_{i_2} \cdots c_{i_{n-1}} \tag{17}$$

and

$$S_k(t) = \int_0^t dt' \, \sigma_k(t') \ . \tag{18}$$

Substituting Eqs. (17) and (18) into GSE (6) yields

$$c_k(t) = \exp[-S_k(t)] \left\{ c_k(0) + \int_0^t dt' \frac{1}{n!} \sum_{i_1+i_2+\ldots+i_n=k} K(i_1, i_2, \ldots, i_n) c_{i_1}(t') c_{i_2}(t') \cdots c_{i_n}(t') \exp[S_k(t')] \right\} . \tag{19}$$

The long-time behavior of $S_k(t)$ can be determined from $c_1(t)$ ($t \to \infty$), which is given by Eq. (6) for $k=1$

$$\dot{c}_1(t) = -c_1 \sigma_1 = -c_1^n E_k, \quad t \to \infty \ , \tag{20}$$

provided that $E_1 < \infty$. Thus, from Eqs. (17) and (18) we find, for $t \to \infty$,

$$\sigma_k(t) \simeq c_1^{n-1} E_k \simeq \frac{E_k}{(n-1) E_1 t} \ , \tag{21}$$

$$S_k(t) \simeq \frac{E_k}{(n-1) E_1} \ln(t) \ , \tag{22}$$

provided that $E_k < \infty$. With the help of Eqs. (21) and (22), the dominant large-time behavior of the t integral in Eq. (19) can be estimated as $t^{-\alpha(k)}$ with $\alpha(k) = 1/(n-1)(1 - E_k/E_1)$. The integral diverges as $t \to \infty$ since $E_k > E_1$. Thus, $c_k(0)$ may be neglected in Eq. (19) and the equation reduces to the large-time form

$$b_k c_1(t)\exp[S_k(t)] \simeq \int_0^t dt' \frac{1}{n!} \sum_{i_1+i_2+..6.+i_n=k} K(i_1,i_2,\ldots,i_n)c_{i_1}(t)c_{i_2}(t')\cdots c_{i_n}(t')\exp[S_k(t')] \; . \tag{23}$$

Differentiating Eq. (23) with respect to t leads to

$$b_k(\sigma_k c_1+\dot{c}_1) \simeq \frac{1}{n!} \sum_{i_1+i_2+\cdots+i_n=k} K(i_1,i_2,..,i_n)c_{i_1}(t)c_{i_2}(t)\cdots c_{i_n}(t) \; . \tag{24}$$

By using Eqs. (8), (20), (21), and (22), we obtain

$$B_k(E_k-E_1)=\frac{1}{n!} \sum_{i_1+i_2+\cdots+i_n=k} K(i_1,i_2,\ldots,i_n)b_{i_1}b_{i_2}\cdots b_{i_n} \; , \tag{25}$$

which is in fact the recursion relation (13), $R(b_k)=0$ with $b_1=1$.

In order to determine the asymptotic solution of the recursion relation, we multiply Eq. (13) with k, and sum over k, and then we obtain

$$E_1\sum_{j=1}^{k} jb_j=\frac{1}{(n-1)!}\sum_{i_1=1}^{k}\sum_{i_2=k-i_1+1}^{\infty}\cdots\sum_{i_n=k-i_1-i_2-\cdots-i_n+1}^{\infty} i_1K(i_1,i_2,\ldots,i_n)b_{i_1}b_{i_2}\cdots b_{i_n} \; . \tag{26}$$

In nongelling class I and class II systems, one finds a consistent solution only if $\tau<2$ is assumed. In this case, by substituting Eq. (14) into Eq. (26), one has

$$BE_1k^{2-\tau}/(2-\tau)\simeq\frac{1}{(n-1)!}\int_0^1 dx_1\int_0^{1-x_1}dx_2\cdots\int_0^{1-x_1-x_2-\cdots-x_{n-2}}dx_{n-1}$$
$$K(x_1,x_2,\ldots,x_{n-1},1-x_1-x_2\cdots-x_{n-1})(x_1x_2\cdots x_{n-1})^{-\tau}(1-x_1-x_2-k\ldots-x_{n-1})^{-\tau} \; . \tag{27}$$

Comparison of the dominant orders in k gives

$$\tau=1+\lambda/(n-1) \; , \tag{28}$$

which is in agreement with the prediction from the scaling function method [11], and consistent with the assumption (15), i.e., $E_k<\infty$.

Now we turn to the gelling systems. Since we are interested in the long-time behavior of the cluster size distribution $c_k(t)$, we are necessarily dealing with the postgel solutions. In gelling systems there is a nonvanishing mass flux $M^{(k)}$ $(k\to\infty)$ transferring clusters with sizes smaller than k to those with sizes larger than k, as $k\to\infty$. In this transfer of sol particles into gel, the mass flux is found to be [10]

$$M^{(\infty)}(t)=-\lim_{k\to\infty}\sum_{i_1=1}^{\infty}\sum_{i_2=k-i_1+1}^{\infty}\cdots\sum_{i_n=k=i_1-i_2-\cdots-i_n+1}^{\infty} i_1K(i_1,i_2,\ldots,i_n)c_{i_1}c_{i_2}\cdots c_{i_n} \; . \tag{29}$$

It must be finite and nonvanishing for all $t>t_c$, where t_c is the gel point. This means that $c_k(t)$ must have an algebraic decay at large k, i.e., $c_k(t)\simeq A(t)k^{-\tau}$ $(k\to\infty)$. Thus,

$$\dot{M}(\infty)(t)=-A^n\lim_{k\to\infty}k^{n+1+\lambda-n\tau}\frac{1}{n!}\int_0^1dx_1\int_0^{1-x_1}dx_2\cdots\int_0^{1-x_1-x_2-\cdots-x_{n-1}}dx_{n_1}K(x_1,x_2,\ldots,x_n)(x_1x_2\cdots x_n)^{-\tau} \tag{30}$$

is bounded and nonzero only if

$$\tau=\frac{\lambda+n+1}{n} \; . \tag{31}$$

It is easy to verify that the result in Eq. (31) can also be obtained from the recursion relation method. Therefore, we conclude that for gelling systems of class I and class II kernels, two methods give rise to the same results.

IV. SUMMARY AND REMARKS

In the previous discussion we proposed that as long as the k and t dependence of the cluster size distribution $c_k(t)$ can factorize at large time, then the recursion relation can be used to the large-time behavior, and the results from the recursion relation method and scaling function method are complementary for class II systems.

Now we demonstrate that the conclusion $\tau=\tau_R$ is self-consistent within the scaling theory of the Smoluchovski equation, where $\tau=2-p_\lambda/w$ is obtained from the scaling function method, and $\tau_R=1+\lambda/2$, is obtained from the recursion relation method. Consider the following moment equation for scaling function $\phi(x)$.

$$(1-\alpha)p_\alpha w=\tfrac{1}{2}\int_0^\infty dx\int_o^\infty dy\, K(x,y)\phi(x)\phi(y)$$
$$\times[x^\alpha+y^\alpha-(x+y)^\alpha] \; , \tag{32}$$

where the αth moment is defined by

$$p_\alpha=\int_0^\infty dx\, x^\alpha\phi(x) \; . \tag{33}$$

Without actually solving the integral equation for the scaling function $\phi(x)$ and calculating the moments, we can determine the upper and lower bounds on τ.

Since the expression of the exponent τ is the same from two different methods, for class I systems we, therefore, restrict ourselves to the class II systems. As an example of a class II kernel, let us consider $K(i,j)=(i+j)^{\lambda}$ with $\lambda<1$. An upper and lower bound on p_{λ} can be obtained as follows. For $\lambda<1$ and x,y positive, one has

$$x^{\lambda}+y^{\lambda}>(x+y)^{\lambda} ,$$

so that

$$x^{\lambda}+y^{\lambda}-(x+y)^{\lambda}<\frac{(x^{\lambda}+y^{\lambda})^2-(x+y)^{2\lambda}}{(x+y)^{\lambda}} =\frac{2x^{\lambda}y^{\lambda}}{(x+y)^{\lambda}}+\frac{x^{2\lambda}+y^{2\lambda}-(x+y)^{2\lambda}}{(x+y)^{\lambda}} .$$

When $\lambda>\frac{1}{2}$ one has

$$(x+y)^{2\lambda}-(x^{2\lambda}+y^{2\lambda})>(2^{2\lambda}-2)(xy)^{\lambda} .$$

Thus, if $\lambda\leq\frac{1}{2}$, we have following inequalities

$$2^{\lambda}(2-2^{\lambda})(xy)^{\lambda}(x+y)^{-\lambda}\leq x^{\lambda}+y^{\lambda}-(x+y)^{\lambda} <2(xy)^{\lambda}(x+y)^{-\lambda} , \tag{34}$$

and if $\frac{1}{2}<\lambda<1$, then

$$2^{\lambda}(2-2^{\lambda})(xy)^{\lambda}(x+y)^{-\lambda}\leq x^{\lambda}+y^{\lambda}-(x+y)^{\lambda} <2(2-2^{2\lambda-1})(xy)^{\lambda}(x+y)^{-\lambda} . \tag{35}$$

Substituting these inequalities into the moment Eq. (32) yields

$$2^{\lambda-1}(2-2^{\lambda})p_{\lambda}^2<(1-\lambda)p_{\lambda}w<p_{\lambda}^2, \quad 0<\lambda\leq\tfrac{1}{2} \tag{36}$$

and

$$2^{\lambda-1}(2-2^{\lambda})p_{\lambda}^2<(1-\lambda)p_{\lambda}w<(2-2^{2\lambda-1})p_{\lambda}^2, \quad \tfrac{1}{2}<\lambda<1 . \tag{37}$$

Thus, one finds that p_{λ} must satisfy the following inequalities:

$$1-\lambda<p_{\lambda}/w<\frac{1-\lambda}{2^{\lambda-1}(2-2^{\lambda})} \tag{38}$$

for $0<\lambda\leq\frac{1}{2}$, and

$$\frac{1-\lambda}{2-2^{2\lambda-1}}<p_{\lambda}/w<\frac{1-\lambda}{2^{\lambda-1}(2-2^{\lambda})} \tag{39}$$

for $\frac{1}{2}<\lambda<1$. From our conjecture it follows that

$$p_{\lambda}/w=1-\lambda/2 . \tag{40}$$

It is an easy matter to verify that Eq. (40) satisfies the inequalities (38) and (39) for all $\lambda<1$. So we argue that at least for factorizable $c_k(t)$, the class II systems can be studied from the recursion relation method and $\tau=\tau_R$.

In summary, we have studied the large-time properties of n-tuple coagulation process for specific homogeneous reaction kernel defined by Eq. (7). We found that if the k and t dependence of the cluster size distribution $c_k(t)$ can factorize, the long-time behavior of $c_k(t)$ can be determined from the recursion relation, derived from GSE. We have also studied the complementary property of the scaling function method with the recursion relation method for class II systems. We show that the conjecture $\tau=\tau_R$ is consistent with the prediction of scaling function theory, for factorizable class II systems. It, however, is worthwhile to stress that if $c_k(t)$ cannot factorize into k and t dependence, τ is, in general, not equal to τ_R, as has been shown in Ref. [8].

As far as the properties of n-tuple coagulation processes are concerned, it should be noticed that, only the simplest type of coagulation kernels have been discussed in this work. A general classification of the multipolymer coagulation is expected to be much involved, due to the diverse coagulation kernels. The large-time behavior of n-tuple coagulation process for more complicated reaction kernels will be discussed in a future work [12].

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