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Research Article

Analytical Solutions for Composition-Dependent Coagulation

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Exact solutions of the bicomponent Smoluchowski's equation with a composition-dependent additive kernel $K(v_a, v_b; v_a', v_b') = \alpha(v_a + v_a') + (v_b + v_b')$ are derived by using the Laplace transform for any initial particle size distribution. The exact solution for an exponential initial distribution is then used to analyse the effects of parameter α on mixing degree of such bicomponent mixtures and the conditional distribution of the first component for particles with given mass. The main finding is that the conditional distribution of large particles at larger time is a Gaussian function which is independent of the parameter α.

1. Introduction

Modelling of a number of industrially important processes such as coagulation and growth of aerosols [1], granulation of powders [2], crystallization [3], crystal shape engineering [4], and synthesis of nanoparticles [5] requires particles to be identified with two or more of their attributes, such as mass for two or more different compositions, mass and surface area, mass of primary particles and binder volume, particle volume, and uncapped surface area. In general cases, the coagulation kernel is a function of both size and composition of the particles. In granulation, for instance, the surface properties (surface energy, roughness) of granules determine the efficiency by which granules are coated by the binder [6]. Therefore, components with different wetting properties may exhibit markedly different behavior during coagulation. Compositional effects introduce yet another dimension in the interaction between particles in coagulation.

This multicomponent coagulation problem was brought to focus by Lushnikov [7] and later by Krapivsky and Ben-Naim [8] for systems in which the coagulation kernel is independent of composition. Vigil and Ziff [9] summarized the solutions of Lushnikov and showed that in these cases the compositional distribution is a Gaussian function. More recently, Matsoukas et al. formulated the bicomponent problem in terms of one population balance equation for the size distribution and another for the distribution of components and provided solutions for kernels that are independent of composition [10–12]. These solutions have shown that for such kernels the distribution of components follows Gaussian scaling that is independent of the details of the kernel.

Without loss of generality, only two-component coagulation problem is considered and two components are given by their mass (or volume) (v_a, v_b).

The governing equation for this coagulation problem is the following population balance equation (PBE):

$$\begin{aligned}\frac{\partial N(v_a, v_b, t)}{\partial t} &= \frac{1}{2}\int_0^{v_a} dv_a' \\ &\cdot \int_0^{v_b} K\left(v_a - v_a', v_b - v_b'; v_a', v_b'\right) N\left(v_a - v_a', v_b - v_b', t\right) \\ &\cdot N\left(v_a', v_b', t\right) dv_b' - N(v_a, v_b, t)\int_0^{\infty} dv_a' \\ &\cdot \int_0^{\infty} K\left(v_a, v_b; v_a', v_b'\right) N\left(v_a', v_b', t\right) dv_b'\end{aligned} \tag{1}$$

which is an extension of Smoluchowski's equation for one-component coagulation, where $N(v_a, v_b, t)$ is the number of density functions at time t such that $N(v_a, v_b, t)dv_a dv_b$ represents the number concentration of particles in the size

range of a-component, v_a to $v_a + dv_a$, and the size range of b-component, v_b to $v_b + dv_b$; $K(v_a, v_b; v_a', v_b')$ is the coagulation rate coefficient. Recently, Fernández-Díaz and Gómez-García, by using Laplace transform, obtained an exact analytical solution for (1) with the additive kernel (which is independent of composition) for any initial particle size distribution (PSD) [13]. They further analysed the behavior of the solution for larger sizes and time and found that the scaling solution cannot be used to describe the behavior of the number of the particle size distributions. In this study, we extend Fernández-Díaz and Gómez-García's procedure to solve (1) with a composition-dependent kernel (see (2) in next section) and analyse the effects of parameter α on the properties of bicomponent coagulation.

2. Exact Solution for a Composition-Dependent Kernel

The kernel considered in this study is given as

$$K\left(v_a, v_b; v_a', v_b'\right) = \alpha\left(v_a + v_a'\right) + \left(v_b + v_b'\right), \tag{2}$$

where parameter α determines the relative contribution of composition a to the coagulation. Evidently the additive kernel studied in [13] is recovered if we are letting $\alpha = 1$ of the kernel given in (2).

To seek the exact solution of (1) with kernel given by (2), we can, from (1), obtain the equation for the total number of particles:

$$\frac{\partial M_{00}(t)}{\partial t} = -\left[\alpha M_{10} + M_{01}\right] M_{00}(t), \tag{3}$$

where M_{00} is the total number of particles, M_{10} is the mass of a-component, and M_{01} is the mass of b-component. The solution of (3) is easily obtained: $M_{00}(t) = N_0 \exp(-\phi t) = N_0(1-\tau)$, with $\phi = \alpha M_{10} + M_{01}$, $N_0 = M_{00}(0)$, and the characteristic coagulation time $\tau = 1 - \exp(-\phi t)$.

Following [9], the number concentration distribution can be given as below:

$$\begin{aligned} &N\left(v_a, v_b, t\right) \\ &\quad = M_{00}(t) \exp\left(-\frac{\alpha v_a + v_b}{\alpha v_{a0} + v_{b0}}\tau\right) g\left(v_a, v_b, \tau\right). \end{aligned} \tag{4}$$

Substituting (4) into (1), we have

$$\begin{aligned} \frac{\partial g\left(v_a, v_b, \tau\right)}{\partial \tau} &= \frac{\alpha v_a + v_b}{2\left(\alpha v_{a0} + v_{b0}\right)} \\ &\cdot \int_0^{v_a}\int_0^{v_b} g\left(v_a - v_a', v_b - v_b', \tau\right) g\left(v_a', v_b', \tau\right) dv_a' dv_b'. \end{aligned} \tag{5}$$

For (5) we can use two-dimensional Laplace transform

$$\begin{aligned} &L\left[g\left(s_1, s_2\right)\right] \\ &\quad = \iint_0^\infty \exp\left(-s_1 v_a - s_2 v_b\right) g\left(v_a, v_b\right) dv_a dv_b \\ &\quad = G\left(s_1, s_2\right). \end{aligned} \tag{6}$$

Taking the derivative in (6) follows

$$\begin{aligned} &\frac{\partial G\left(s_1, s_2, \tau\right)}{\partial \tau} \\ &\quad = -\frac{\alpha}{\alpha v_{a0} + v_{b0}} G\left(s_1, s_2, \tau\right) \frac{\partial G\left(s_1, s_2, \tau\right)}{\partial s_1} \\ &\qquad - \frac{1}{\alpha v_{a0} + v_{b0}} G\left(s_1, s_2, \tau\right) \frac{\partial G\left(s_1, s_2, \tau\right)}{\partial s_2}. \end{aligned} \tag{7}$$

This is Burgers' equation in multidimension without a diffusive term. It can be solved in the transformed space by the Lagrange-Charpit method [14]:

$$\begin{aligned} w_1 &= s_1 - \frac{\alpha}{m_0} G\left(s_1, s_2\right)\tau, \\ w_2 &= s_2 - \frac{1}{m_0} G\left(s_1, s_2\right)\tau, \\ G\left(s_1, s_2, \tau\right) &= G\left(w_1, w_2, \tau\right). \end{aligned} \tag{8}$$

With multidimensional Lagrange inversion [15], we obtain

$$\begin{aligned} &G\left(w_1, w_2, \tau\right) \\ &\quad = \sum_{k_1=0}^\infty \sum_{k_2=0}^\infty \frac{(-1)^{k_1+k_2}\left(\alpha\tau/m_0\right)^{k_1}\left(\tau/m_0\right)^{k_2}}{k_1! k_2!} \frac{\partial^{k_1+k_2}}{\partial^{k_1} t_1 \partial^{k_2} t_2}\left[F\left(t_1, t_2\right)\right]_{\substack{t_1=s_1 \\ t_2=s_2}} \end{aligned} \tag{9}$$

with

$$\begin{aligned} F\left(t_1, t_2\right) &= G^{k_1+k_2+1}\left(t_1, t_2\right) \\ &\quad - \left(t_1 - s_1\right) G^{k_1+k_2}\left(t_1, t_2\right) \frac{\partial G\left(t_1, t_2\right)}{\partial t_1} \\ &\quad - \left(t_2 - s_2\right) G^{k_1+k_2}\left(t_1, t_2\right) \frac{\partial G\left(t_1, t_2\right)}{\partial t_2}. \end{aligned} \tag{10}$$

That is,

$$G\left(w_1, w_2, \tau\right) = \sum_{k_1=0}^\infty \sum_{k_2=0}^\infty \frac{(-1)^{k_1+k_2}\left(\alpha\tau/m_0\right)^{k_1}\left(\tau/m_0\right)^{k_2}}{k_1! k_2!} \frac{1}{k_1+k_2+1} \frac{\partial^{k_1+k_2}}{\partial^{k_1} s_1 \partial^{k_2} s_2} G^{k_1+k_2+1}\left(s_1, s_2, 0\right) \tag{11}$$

when naming $G(s_1, s_2, 0) = G_0(s_1, s_2)$.

Applying Laplace inverse transform, we obtain

$$g(v_a, v_b, \tau) = \sum_{k_1=0}^{\infty}\sum_{k_2=0}^{\infty} \frac{(-1)^{k_1+k_2}\alpha^{k_1}}{k_1!k_2!}\left(\frac{\tau}{\alpha v_{a0}+v_{b0}}\right)^{k_1+k_2} \cdot \frac{1}{1+k_1+k_2} L^{-1}\left[\frac{\partial^{k_1+k_2}}{\partial s_1^{k_1}\partial s_2^{k_2}} G_0^{1+k_1+k_2}(s_1, s_2)\right]. \tag{12}$$

By rearranging the multiple series in one, we arrive at

$$g(v_a, v_b, \tau) = \sum_{k=0}^{\infty} \frac{(((\alpha v_a + v_b)/(\alpha v_{a0}+v_{b0}))\tau)^k}{(k+1)!} \cdot L^{-1}\left[G_0^{1+k}(s_1, s_2)\right]. \tag{13}$$

With (4), we finally obtain the general solutions of (1)

$$N(v_a, v_b, \tau) = N_0(1-\tau)\exp\left(-\frac{\alpha v_a + v_b}{\alpha v_{a0}+v_{b0}}\tau\right) \cdot \sum_{k=0}^{\infty} \frac{(((\alpha v_a + v_b)/(\alpha v_{a0}+v_{b0}))\tau)^k}{(k+1)!} \cdot L^{-1}\left[G_0^{1+k}(s_1, s_2)\right]. \tag{14}$$

This solution can be applied for any initial distributions by determining the multidimensional Laplace transform ${G_0}^{1+k}(s_1, s_2)$. If exponential initial PSD is assumed

$$N(v_a, v_b, 0) = \frac{N_0}{M_{10}M_{01}}\exp\left(-\frac{v_a}{M_{10}} - \frac{v_b}{M_{01}}\right). \tag{15}$$

The Laplace transform for this function is

$$G_0(s_1, s_2) = \frac{N_0}{M_{10}M_{01}}\frac{1}{s_1 + 1/M_{10}}\frac{1}{s_2 + 1/M_{01}} \tag{16}$$

and we can obtain the explicit solution as below:

$$N(v_a, v_b, \tau) = \frac{N_0}{M_{10}M_{01}}(1-\tau) \cdot \exp\left(-\frac{\alpha v_a + v_b}{\alpha v_{a0}+v_{b0}}\tau - \frac{v_a}{M_{10}} - \frac{v_b}{M_{01}}\right) \cdot \sum_{k=0}^{\infty} \frac{((v_a v_b/M_{10}M_{01})((\alpha v_a + v_b)/(\alpha v_{a0}+v_{b0}))\tau)^k}{k!(k+1)!}. \tag{17}$$

It is easily verified that the solution in [13] is recovered if we are letting $\alpha = 1$. We will further analyse some interesting properties of solution (17) in the next section and attention is paid particularly to the effects of parameter α on the coagulation properties of such systems.

3. The Effects of α on the Coagulation

3.1. The Total Number and Mass of Particles with the Concentration c. Mixing degree of the mixtures is one of the key issues in bicomponent coagulation problems. To this end, we define two important magnitudes introduced in [7, 13]. One is the total number of particles having the concentration c of the first component:

$$N(c, t) = \iint_0^{\infty} N(v_a, v_b, \tau)\,\delta\left(c - \frac{v_a}{v_a + v_b}\right) dv_a dv_b \tag{18}$$

and the mass of these particles

$$M(c, t) = \iint_0^{\infty} (v_a + v_b) N(v_a, v_b, \tau) \cdot \delta\left(c - \frac{v_a}{v_a + v_b}\right) dv_a dv_b. \tag{19}$$

With the compositional-dependent additive kernel (1) and the initial condition (15), we obtain

$$N(c, \tau) = \frac{1-\tau}{M_{10}M_{01}A^2}\sum_{k=0}^{\infty}\frac{(3k+1)!}{(k+1)!(k!)^2}\left(\frac{D}{A^3}\right)^k, \quad M(c, \tau) = \frac{1-\tau}{M_{10}M_{01}A^3}\sum_{k=0}^{\infty}\frac{(3k+2)!}{(k+1)!(k!)^2}\left(\frac{D}{A^3}\right)^k \tag{20}$$

with

$$A = \frac{c}{M_{10}} + \frac{1-c}{M_{01}} + \frac{\alpha c + (1-c)}{\alpha/M_{01} + 1/M_{10}}\frac{\tau}{M_{10}M_{01}}, \quad D = \frac{\alpha c + (1-c)}{\alpha/M_{01} + 1/M_{10}}\frac{\tau c(1-c)}{(M_{10}M_{01})^2}. \tag{21}$$

As suggested in [13], we observe the process at different average particles size σ with $\tau = 1 - \sigma^{-1/2}$.

Figures 1–3 show the evolution of total number and total mass with concentration c for $\alpha = 0.1$, 1, and 20, respectively. It is shown that the overall behavior of the evolution of total number and mass is similar for different α, while the evolution for total number ((a) in Figures 1–3) and total mass ((b) in Figures 1–3) is obviously different. The curves of total number do not tend to a Dirac-δ function, whereas the curves of total mass do, and this result is consistent with the result in [13]. The effect of α can be clearly seen from comparison between Figures 2(b) and 3(b), and it is shown that the maxima for the total mass approach the overall fraction from the left end for $\alpha = 1$, but from the right end for $\alpha = 20$. This is due to the fact that the larger the parameter α, the bigger the contribution of the a-component to the coagulation, which leads to the quicker growth of particles with higher concentration.

3.2. Conditional Distribution for Mass-Given Particles. Conditional distribution is another important issue in bicomponent coagulation problems. Vigil and Ziff [9] postulated the general scaling law for two-component aggregation with

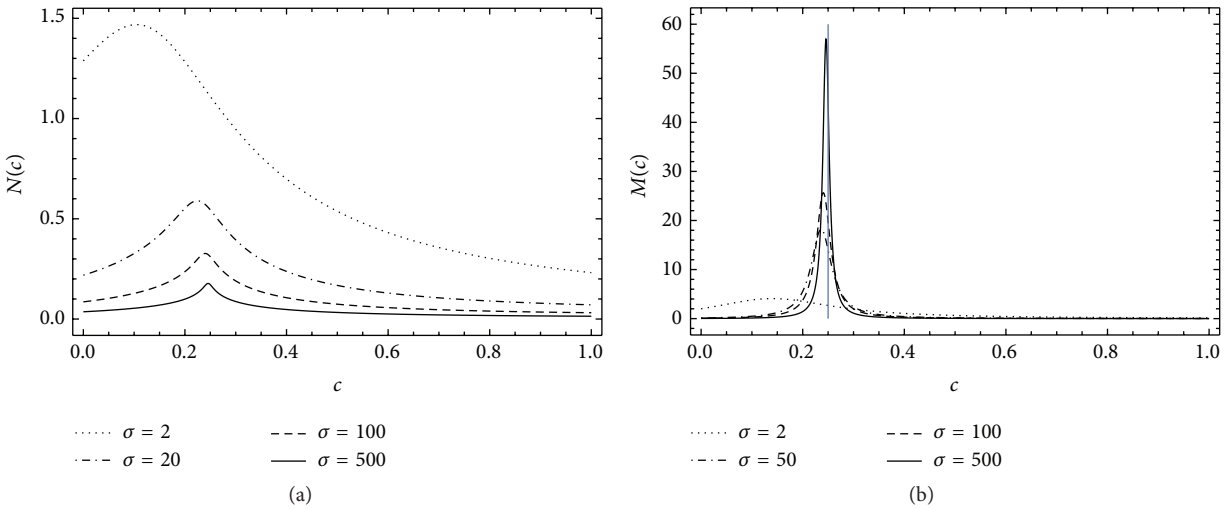

Figure 1: Number (a) and mass (b) for initial exponential distribution with $\alpha = 0.1$ and $N_0 = 1$ and $M_{10} = 1/3$ and $M_{01} = 1$.

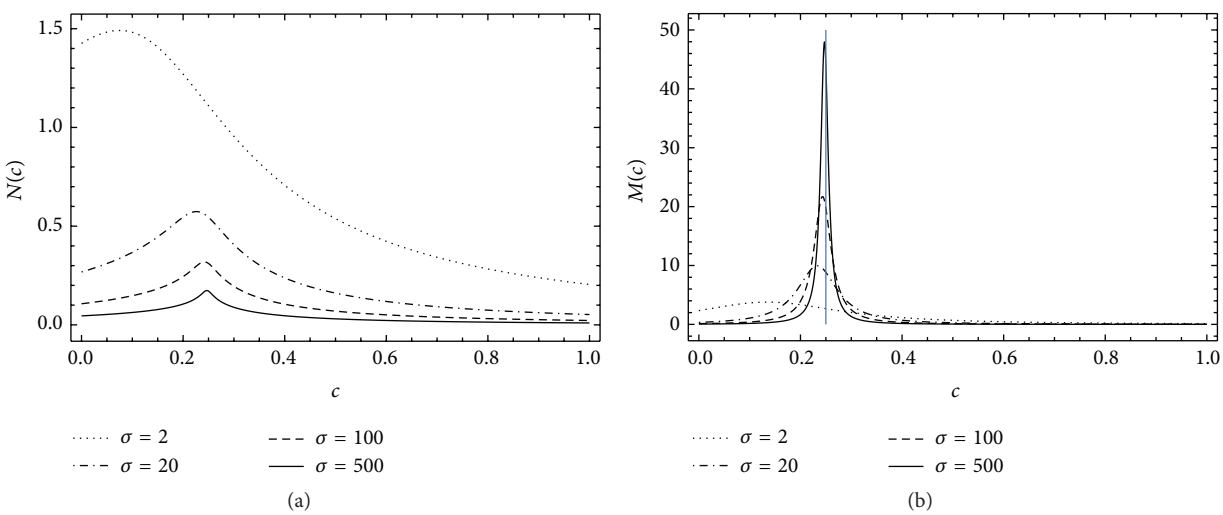

Figure 2: Number (a) and mass (b) for initial exponential distribution with $\alpha = 1$ and $N_0 = 1$ and $M_{10} = 1/3$ and $M_{01} = 1$.

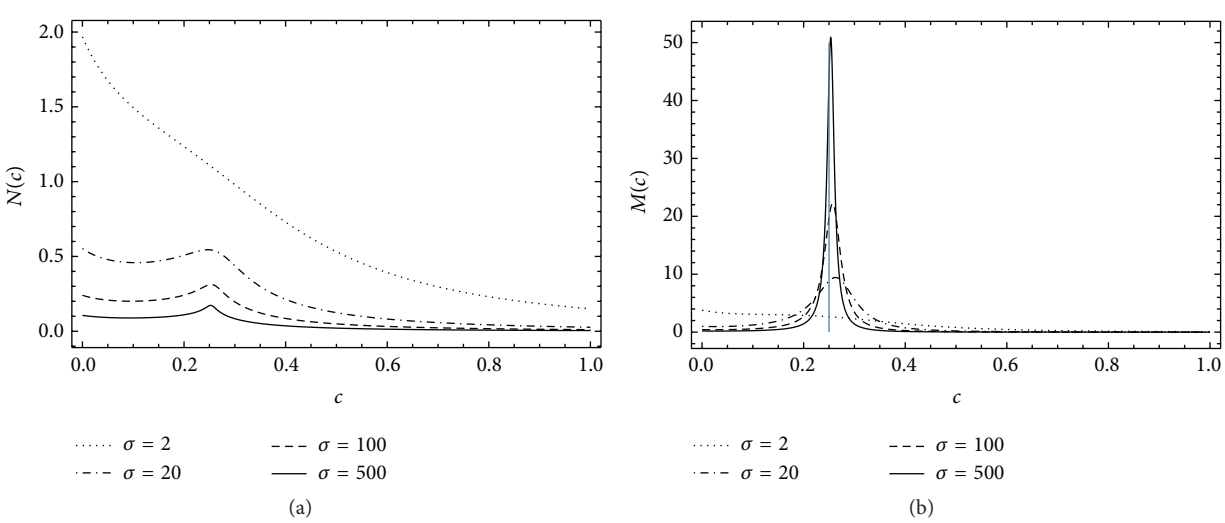

Figure 3: Number (a) and mass (b) for initial exponential distribution with $\alpha = 20$ and $N_0 = 1$ and $M_{10} = 1/3$ and $M_{01} = 1$.

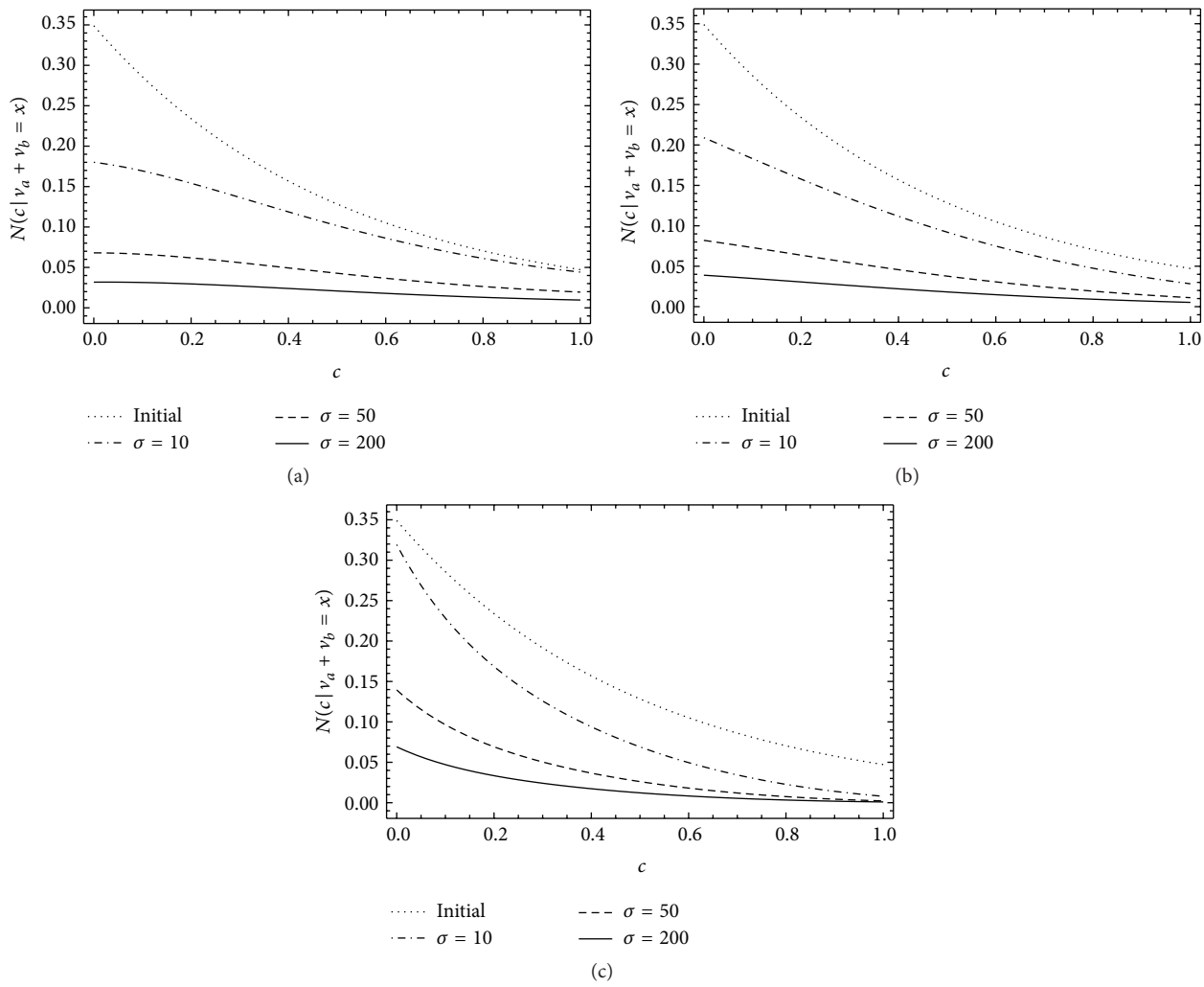

Figure 4: The compositional distributions for $x = 1$ with $N_0 = 1$, $M_{10} = 1/3$, $M_{01} = 1$, and (a) $\alpha = 0.1$, (b) $\alpha = 1$, and (c) $\alpha = 20$.

nongelling kernels that bivariate PSD function can be the product of a normal distribution in particle composition and the one-parameter scaling function for the corresponding homogeneous coagulation problem. And for large time, the compositional distribution in large particles is a Gaussian function. We explore the conditional distribution for a composition-dependent kernel given in (2), and again, the effects of α are analysed in this section. To this aim, we introduce the conditional a-component number distribution $N(c \mid x = v_a + v_b)$, where $c = v_a/(v_a + v_b)$ is a-component concentration for particles given mass x.

Figure 4 shows the conditional distribution for particles given mass $x = 1$ with three different α values. It is shown that the general behavior is similar for all three α values; for example, the total number decreases with the coagulation going on, while the curves become flat. The effects of parameter α in these small particles are nearly negligible. Figure 5 shows the conditional distribution for particles given mass $x = 4$ with three different α values. The difference of α being smaller than 1 and bigger than 1 is clearly seen by comparison of Figures 5(a) and 5(b) with Figure 5(c). The obvious difference is that there exist two extreme points of the curve $N(c \mid x = v_a + v_b)$ for the case $\alpha = 20$, while only one extreme point exists for $\alpha \leq 1$, which means mixing degree is worse for $\alpha > 1$ than $\alpha \leq 1$. Comparison of Figures 4 and 5 shows that a-component is mixed better in larger particles than in minute particles. This tendency can be further illustrated in Figure 6, which shows the distribution for $x = 100$. It is clearly shown in Figure 6 that the mixture is well mixed when $\sigma > 10$ irrespective of the value α. It is also noticed that the distribution curves for 3 different α values collapse each other to a Gaussian function, which can be verified analytically in fact, since from the exact solutions (17) we can obtain for large particles at large time the conditional distribution as below

$$N(c \mid x) \sim \frac{1}{\sqrt{4\pi\left(c_0^2\left(1-c_0\right)^2/x\right)}} \exp\left(-\frac{x\left(c-c_0\right)^2}{4c_0^2\left(1-c_0\right)^2}\right). \quad (22)$$

Equation (22) is a Gaussian function and is independent of the value α.

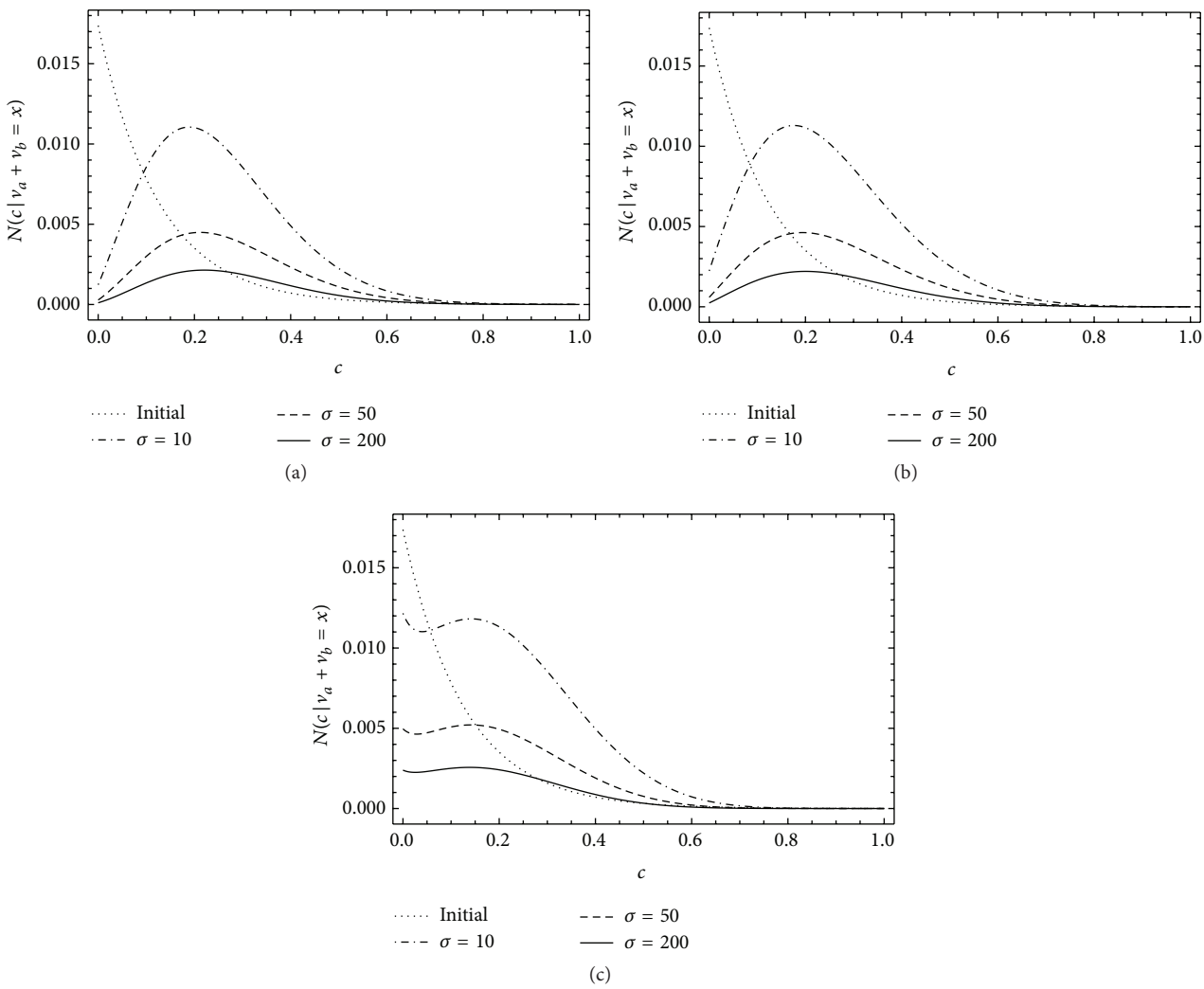

Figure 5: The compositional distributions for $x = 4$ with $N_0 = 1$, $M_{10} = 1/3$, $M_{01} = 1$, and (a) $\alpha = 0.1$, (b) $\alpha = 1$, and (c) $\alpha = 20$.

From Figure 6, we can find an obvious difference between the results for $\alpha = 20$ and the results for $\alpha \leq 1$. The distribution approaches to the asymptotic steady solution (22) from right end when $\alpha = 20$ while the curves approach this steady state from left end. In other words, the equilibrium concentration c of the compositional distribution is bigger than overall a-component mass concentration $c_0 = M_{10}/(M_{10} + M_{01})$ when $\alpha = 20$ and smaller than overall concentration when $\alpha \leq 1$. The mechanism is the same as explained in Section 3.1. In fact it can be proved that when $\alpha > 4.3$ the equilibrium concentration is larger than the overall fraction c_0 at a finite time.

4. Conclusion

In this paper, we have obtained the exact solution of Smoluchowski's continuous two-component equation with a composition-dependent additive kernel which is given as $K(v_a, v_b; v_a', v_b') = \alpha(v_a + v_a') + (v_b + v_b')$ for any initial particle size distributions. The main characteristics of the solution and the effects of parameter α on the bicomponent coagulation have been analysed in detail for an exponential initial distribution.

The effects of α on the total number and mass of particles with concentration c were first investigated, and the results show that the curves for the total number do not reduce to a Dirac-δ function for any given α's while the curves for the total mass tend to a Dirac-δ function at large time. The approaching process is slightly different which depends on the parameter α.

Then, the effects of α on the conditional distribution of the first component in mass-given particles were discussed, and it is found that the conditional distribution is a Gaussian function at large time, which is independent of the value of α. Besides, the conditional distribution in particles with moderate mass is quite different which strongly depends on the parameter α.

The exact solutions obtained in this paper are of potential help to understanding the coagulation in a bicomponent system. The solutions can also be used to test the validity of

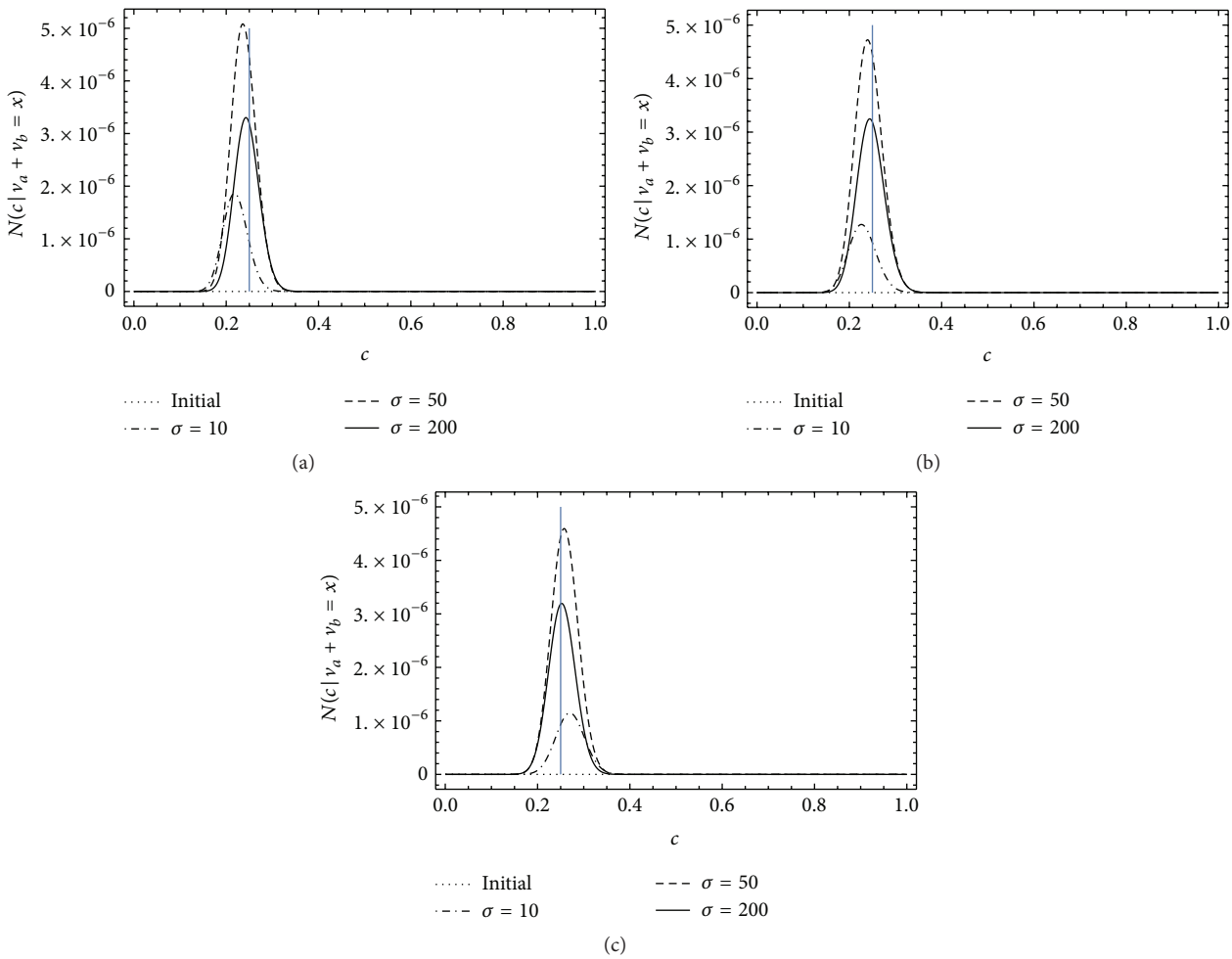

Figure 6: The compositional distributions for $x = 100$ with $N_0 = 1$, $M_{10} = 1/3$, $M_{01} = 1$, and (a) $\alpha = 0.1$, (b) $\alpha = 1$, and (c) $\alpha = 20$. Compared with overall a-component mass concentration $c_0 = M_{10}/(M_{10} + M_{01})$ (here $c_0 = 0.25$).

the numerical methods developed to solve multicomponent Smoluchowski's equation.

Symbols

v_a, v_b:	The size of particles a, b
$M_{00}(t)$:	Particle number at time t
M_{10}:	Mass of a-component
M_{01}:	Mass of b-component
N_0:	Initial number of particles
ϕ:	Equivalent total mass of particles
τ:	Characteristic coagulation time
α:	Parameters of component effects on coagulation
c:	a-component mean concentration
c_0:	Overall a-component mass concentration
σ:	Average particles size
x:	Mass of given particles.

Competing Interests

The authors declare that they have no competing interests.

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