Application of the Cluster–Cluster Aggregation model to an open system

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ABSTRACT

Colloid aggregation is often induced by the change of internal or external conditions. In order to account for the dynamic features of the evolutionary open system, a conceptual model for colloid aggregation in open systems was developed based on the classic Cluster–Cluster Aggregation (CCA) model. The extended model allows the important parameters of the classic CCA model, diffusion coefficient D_1 and sticking probability P_1 of primary particles, time-dependent. Consequently, the new model can be used to simulate colloid aggregation in open systems. To demonstrate the applicability of the extended model, the diffusion coefficient D_1 and sticking probability P_1 were defined as a function of solvent evaporation rate and aggregation time in this study. For the simplicity purpose, this study only evaluate $D_1(t)$ while kept $P_1(t)$ as a constant for the simulations. Simulation results indicate that the solvent evaporation altered the aggregation mechanism in various degrees depending on the solvent evaporation rate. This research shows that the extended model based on the classic CCA model is valuable and applicable to open systems.

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1. Introduction

Most of the natural systems are not isolated, but companied with energy or material transfers into or out of the system boundary. Except for the system compositions, colloid aggregation is often induced by the change of internal or external conditions, such as concentration changes due to solvent evaporation, dilution (rainfall as an example), or addition of electrolytes [1,2]. These changes can directly affect the colloidal stabilization and the characteristic structure of the aggregated clusters. It is a great challenge for the researchers to precisely control the dynamic aggregation process in laboratory experiments.

The Cluster–Cluster Aggregation (CCA) model bridges the study of colloid aggregation by computer simulation and laboratory experiment [3–10]. Two distinct and limiting regimes of irreversible colloid aggregation have been identified by computer simulation with the CCA model [11,12]. These regimes generally correspond to the rapid and slow colloid aggregation, which have long been recognized in colloid sciences. One regime is diffusion-limited cluster aggregation (DLCA) corresponding to the rapid colloid aggregation, occurring when there is negligible repulsive force between the particles. In this way, the aggregation rate is limited solely by the time taken for clusters to encounter each other by

diffusion. The other is reaction-limited cluster aggregation (RLCA) corresponding to the slow colloid aggregation, occurring when there is still a substantial, but not insurmountable, repulsive force between the particles. In this way, the aggregation rate is limited by the time taken for two clusters to overcome this repulsive barrier by thermal activation [13–16].

As discussed in the literature, the most important parameters in the CCA model are related to how clusters are selected from the list of clusters and how they are combined [4,17]. Generally, the simulations are performed in a closed system [4,18]. The lattice sites are selected randomly and filled to represent the aggregation particles according to the fraction ϕ . In the simple lattice model simulations, after a cluster has been selected, a random number x uniformly distributed over the range $[0, 1]$ is generated and the cluster is moved if $x < D/D_{max}$, where D is the diffusion coefficient of the selected cluster and D_{max} is the maximum diffusion coefficient for any cluster in the system. If $x > D/D_{max}$, a new cluster is selected at random. It is assumed that, ignoring the shape effects, the cluster diffusion coefficient D_i is proportional to the inverse of the gyration radius R_g of the cluster [13,19], where i is the number of the monomers in the cluster:

$$D_i = D_1 * R_1/R_g \quad (1)$$

where D_1 (constant) is the diffusion coefficient of the primary particle with the gyration radius of R_1 . Thus, the probability that the cluster is selected from the list of clusters is mainly determined

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by its diffusion coefficient. Similarly, the probability that a cluster of size i contacts with a cluster of size j to form a cluster of size $i + j$ may also be size-dependent [3,20]:

$$P_{ij} = P_1 * (i * j)^\sigma \quad (2)$$

where P_1 (constant) is the sticking probability of the primary particle, σ is the sticking probability exponent, and P_{ij} is set to unity if $P_1 * (i * j)^\sigma > 1$. When a collision takes place, a random number y uniformly distributed in $[0, 1]$ is generated and compared with the given P_{ij} . The cluster collision is considered effective only when $y < P$ is verified.

It is obvious from Eqs. (1) and (2) that the cluster diffusion coefficient and sticking probability are both size-dependent and the parameters D_1 and P_1 were used to characterize the specific features of the systems. However, there are limitations yet in applying the classic CCA model to mimic the colloid aggregation, although the CCA model is a recognized and widely used ingenious model.

The first limitation of the classic CCA model is that it can not be applied to open systems because the simulations were restrained in a closed square (2 Dimension) or cubic (3 Dimension) without allowing mass transfer between inside and outside of the system [3,4,17]. The second limitation is that the classic CCA model can not reflect the dynamic features of the evolutionary colloid suspension. The parameters D_1 and P_1 in Eqs. (1) and (2) were treated as constants and used to characterize the specific features of the systems during the entire aggregation process. It is reasonable to assume that the values of D_1 and P_1 remain constant in a stable and homogeneous system. However, as the aggregation process progresses, the system becomes heterogeneous with different size clusters. Consequently, the diffusion coefficient and the sticking probability of the primary particles in the heterogeneous system change with time and one can expect that they become different from the initial values. Obviously, the diffusion coefficient and sticking probability of a specified cluster in different environment are not always identical.

Thus, it is of practical significance to develop an aggregation model for the widely existing open systems that can accommodate the dynamic nature of the aggregation process. In the past few years, a number of attempts have been made to investigate some of the effects by extending the CCA model [2,10,21,22]. The philosophy behind most of these works has been to attempt to develop more realistic models while retaining the simplicity and reliability of the basic models for Cluster-Cluster Aggregation [23–25]. But most of the simulations were based on the average-field theory and restrained in a very dilute suspension. These models can not be applied to the high concentration systems and open systems.

The objectives of this research were to (1) develop a time-dependent conceptual model for the open system based on the classic CCA model by adding functions allowing mass transfer out of the system boundaries; (2) apply the extended model to simulate the aggregation process in a system where solvent evaporation occurs; (3) test a new approach to calculate the diffusion coefficient of the clusters during the period of aggregation; and (4) investigate the morphology of the aggregates and the aggregation dynamics in the regimes of DLCA and RLCA, respectively.

2. Model concepts and application

2.1. 1 Conceptual model for the open systems based on CCA

Eqs. (1) and (2) have made the aggregations cluster size-dependent, but omitted the evolution of the system. Since D_1 and P_1 were used to characterize the specific features of the system, to mimic the aggregation process in a more realistic manner, we can extend the classic CCA model as follows:

$$D_i(t) = D_1(t) * R_1/R_g \quad (3)$$

$$P_{ij}(t) = P_1(t) * (i * j)^\sigma \quad (4)$$

where $D_1(t)$ and $P_1(t)$ are the functions of aggregation time, which are different from the classic CCA model by reflecting the dynamic changes of the system.

By considering the time-dependent parameters of $D_1(t)$ and $P_1(t)$, the extended model can better reflect the diffusion coefficient and the sticking probability of clusters. The new model can also be applied to open systems depending on the ways that $D_1(t)$ and $P_1(t)$ are defined. In this study, we use colloid aggregation in a system undergoing solvent evaporation as an example to demonstrate the application of the extended CCA model in an open system.

2.2. Application of the conceptual model for solvent evaporation in an open system

Solvent evaporation can impact the stabilization of the colloidal dispersion mainly in three ways: (1) increasing the electrolyte concentration of the system, which alters the electric double layer interactions between the particles in colloidal dispersion; (2) increasing the particle concentration, which leads to more collisions between particles; and (3) increasing the viscosity of the colloidal dispersion, which decreases the diffusion coefficient of the particles.

The rate of colloidal aggregation is generally determined by the frequency of collisions and the sticking probability during collision [4]. The increase of electrolyte concentration can affect the sticking probability; while the increase of the particle concentration and the increase of fluid viscosity of the system can affect the collision frequency. As mentioned above, the effect of evaporation on the collision frequency and sticking probability can be achieved through the definition of $D_1(t)$ and $P_1(t)$, respectively.

2.2.1. The definition of $D_1(t)$

Assuming that a cube acts as the container of the colloidal dispersion and all the solvent evaporation is from the top surface of the cube, it is predictable that the suspension surface will drop at a specified rate due to solvent evaporation; and the particles at the suspension surface along with the drop will likely collide the particles below the surface. The collision possibility will be greater if there is a big cluster at the surface. Similar to the collisions resulting from the traditional Brownian motion, these collisions can also lead to cohesion. Thus, the height of the suspension surface, $H(t)$, will be a important parameter in this research:

$$H(t) = L - mt \quad (5)$$

where m is the evaporation rate measured by the descent of the suspension surface in unit time Δt ; L is the initial height of the suspension surface, which equals to the side length of the cube; t is the current aggregation time. The unit time Δt is defined as [8]:

$$\Delta t = \frac{l_b^2}{6D_1(0)} \quad (6)$$

where $D_1(0)$ is the diffusion coefficient of primary particles in a homogeneous system with a specific volume fraction; l_b is the step length of the Brownian motion.

The solution viscosity $\eta(t)$ of the system also directly affects the diffusion coefficient of the particles. The diffusion coefficient of the primary particles, $D_1(t)$, used to characterize the specific features of the systems during the entire aggregation process, can be defined as:

$$D_1(t) = \frac{KT}{3\pi\eta(t)d_1} \quad (7)$$

where d_1 is the diameter of the primary particle; T is the absolute temperature of the system, and K is Boltzmann constant.

According to the Einstein's formula [26], the solution viscosity $\eta(t)$ is related to the volume fraction $\phi(t)$ (the number of the particles and clusters per volume), which also represents the system concentration:

$$\eta(t) = \eta_0 * (1 + 2.5\phi(t)) \quad (8)$$

where η_0 is the viscosity of the pure solvent.

Due to the solvent evaporation, the volume fraction $\phi(t)$ can be defined as:

$$\phi(t) = \frac{N(t)}{L^2 H(t)} \quad (9)$$

where $N(t)$ is the number of the remaining clusters in the current system. In a traditional closed system, the volume fraction is doomed to decrease. But in an open system with solvent evaporation, the change of the volume fraction is very complex because $N(t)$ and $H(t)$ are both monotone decreasing functions.

Combining Eqs. (3) and (5)–(9), the time-dependent diffusion coefficient of the cluster with i particles in the system is:

$$D_i(t) = \frac{KT}{3\pi\eta_0 d_1} * \left(1 - \frac{2.5N(t)}{L^2(L - mt) + 2.5N(t)}\right) * \frac{R_1}{R_g} \quad (10)$$

where R_1 is the gyration radius of the primary particles and R_g is the gyration radius of the selected cluster.

2.2.2. The definition of $P_1(t)$

Aggregation kinetics are often quantified in terms of the stability ratio W , which can be considered as the inverse sticking probability for two colliding particles to permanently stick together on contact. The sticking probability is related to the height of the barrier for deposition. Applying the DLVO theory to relate the repulsive energy barrier to the electrolyte concentration c , it follows that for an electric double layer stabilized dispersion $\ln(W)$ varies linearly with $\ln(c)$ until the suppression of electric double layer results in energy barrier reaches zero and rapid aggregation ($W = 1$) is reached [27]. Hence, a plot of $\ln(W)$ against $\ln(c)$ consists of two liner segments, as shown in Fig. 1. The value of c where the liner segments intersect is the critical coagulation concentration c_{ccc} .

Without getting into the details, here we give an approximate expression that provides a simple way to estimate the stability ratio [27]:

Fig. 1. Rate of particle aggregation as a function of electrolyte concentration.

$$\ln(W) = -K_1 \ln(c(t)) + K_2 \quad (11)$$

where K_1 and K_2 are constants when the system temperature is specified.

Due to solvent evaporation, the electrolyte concentration changes with time, which can be expressed:

$$c(t) = \frac{L}{L - mt} c_0 \quad (12)$$

where c_0 is the initial electrolyte concentration of the system.

Considering $P_1(t) = 1/W$, Eq. (4) can be rewritten as:

$$P_{ij}(t) = P_1(0) * \left(\frac{L}{L - mt}\right)^{K_1} * (i * j)^\sigma \quad (13)$$

where $P_1(0)$ is the sticking probability of the particles at the beginning, which can be extracted from Eq. (11):

$$P_1(0) = \frac{c_0^{K_1}}{e^{K_2}} \quad (14)$$

As illustrated in Fig. 1, the critical time for $P_{ij}(t) = 1$ can be achieved from Eq. (12) when $c(t) > c_{ccc}$:

$$t \geq \frac{L}{m} \frac{(c_{ccc} - c_0)}{c_{ccc}} \quad (15)$$

Thus it is very clear that the transformation from slow aggregation to rapid aggregation is totally determined by the colloidal features and the solvent evaporation rate.

In this way, Eqs. (10) and (13) can account for both the effect of solvent evaporation on the diffusion coefficient and sticking probability and the change of the diffusion coefficient and sticking probability in an evolutionary heterogeneous system. Thus, the application of the extended CCA model to open systems is feasible. Its further verification can be demonstrated by the following model simulations.

2.3. Simulation conditions and parameters

There are essential differences between collision frequency and sticking probability on the effect of colloidal aggregation due to solvent evaporation. Furthermore, the equations of the diffusion coefficient and sticking probability contain many free parameters, which are all related to solvent evaporation. It, therefore, would be very complicated if all the parameters are simultaneously considered in one study. Thus, we here focus on $D_1(t)$ changes as a function of time during solvent evaporation since the main aim of this research is to introduce the idea on how to apply the CCA model to open systems. Theoretically, the approach to implement $P_1(t)$ in the algorithm should be similar. We did not do it simply because we want to keep the paper simple and discussion cohesive. But, based on Eq. (2), the initial values of P_1 are still set to vary over the range $[0, 1]$. In this manner, the model evaluation is reasonably simplified without neglecting the important effect of solvent evaporation.

The simulations were performed on-lattice in three-dimension with the following parameters: primary particle diameter $d_1 = 100$ nm, Brownian motion step length $l_B = d_1$, cubic side length $L = 100d_1$ and the initial particle volume fraction $\phi(0) = 0.01$. Therefore, the number of the primary particles $N(0)$ is 10,000 (equal to $\phi(0) * L^3$). To keep the simulation simple (less free parameters), the sticking probability was calculated by Eq. (2), where P_1 was fixed between 0.01 and 1, and the sticking probability exponent σ was set to 0. The evaporation rate m varied in the range of $0.001 - 1d_1 \Delta t^{-1}$, unless otherwise mentioned. For clarity, it was assumed that the initial colloidal dispersion was a mono-disperse system of cubic particles of side length d_1 . Cluster reorganization

was not considered in this research. The simulations terminated when either only a single cluster remained or solvent dried out.

The simulation was made a time-dependent process in the following way: after each cluster has been randomly selected, the time was increased by $1/N(t)D_{max}$.

Customarily, the temporal evolution of the aggregation process was described in terms of weighted average cluster size $S(t)$ [8,28]:

$$S(t) = \frac{\sum_i i^2 N_i(t)}{\sum_i i N_i(t)} \quad (16)$$

where $N_i(t)$ is the cluster-size distribution defined as the number of clusters containing i monomers at time t .

3. Results and discussion

3.1. DLCA with solvent evaporation

The temporal evolution of the weighted average cluster size under different evaporation rates are illustrated in Fig. 2. During their downwards movements, clusters at the suspension surface can sweep up the clusters or particles beneath them in the presence of solvent evaporation. This phenomenon is referred as “sweepage aggregation” here. At the initial stage of solvent evaporation, the surface clusters derived from sweepage aggregation have not been fully formed and the number of the particles below the surface is so big that the perikinetic aggregation made the effect of sweepage aggregation insignificant. As a result, the weighted average cluster size increased very slowly and kept at a relatively small value at the initial stage of evaporation. After a certain long period, the effect of evaporation on the aggregation process became pronounced. The weighted average cluster size increased rapidly and approached the maximum in a short period. This kind of behaviour is very similar to that observed for pure DLCA [3,28].

The curves under different evaporation rates scatter gradually as shown in Fig. 2. The weighted average cluster size increase quickly when the evaporation rate increases. For $m \leq 0.01d_1\Delta t^{-1}$, most of the particles below the surface are already aggregated due to the Brownian motion before the surface arrival. Thus, the curves are similar to that of $m = 0d_1\Delta t^{-1}$. For $m \geq 0.05d_1\Delta t^{-1}$, the effects of sweepage aggregation become pronounced. It implies that the aggregation process of the system was significantly accelerated. The aggregations were terminated when solvent dried out,

Fig. 2. A log–log plot of the weighted average cluster size, $S(t)$, as a function of time t , for different solvent evaporation rates. The case of $m = 0d_1\Delta t^{-1}$ corresponds to the classic DLCA.

which resulted in the fact that the curves for high evaporation rates did not reach a plateau value.

In order to compare the aggregation dynamics with the previous literature, the total concentration of the remaining clusters in the system and the concentrations of the clusters of size 1, 2, 3, 4, 5, 6, 7, 8 as a function of time during the simulation were calculated (Fig. 3). The curves for clusters greater than 8 particles were not included since they had a similar pattern. The total concentration of the clusters (number of clusters per volume) and the concentration of the single particles decrease monotonically, and the concentrations of the clusters of size more than 1 are bell-shaped curves. For low evaporation rate, e.g. $m = 0.001d_1\Delta t^{-1}$, the cluster-size distribution is consistent with the result of the classic DLCA [6,28]. But for high evaporation rate, e.g. $m = 0.5d_1\Delta t^{-1}$, the curves were cut off on the right side due to the aggregation process being terminated when the solvent dried out. The basic bell shape of the curves was nearly not influenced by the solvent evaporation, which is still consistent with the scaling theory [6,28].

Morphologies of the final aggregates under different evaporation rates are illustrated in Fig. 4. As the evaporation rate increases, the aggregates appear more flat. For $m = 0.001d_1\Delta t^{-1}$, the aggregate is almost totally isotropic and similar to that of pure DLCA [17]. But for $m = 0.5d_1\Delta t^{-1}$, the cluster anisotropy become very

Fig. 3. A log–log plot of the total concentrations of the remaining clusters, the monomers and the clusters of size 2, 3, 4, 5, 6, 7, 8 (the dashed arrow passed in turn) as a function of time t . $m = 0.001d_1\Delta t^{-1}$ and $m = 0.5d_1\Delta t^{-1}$ characterize the low and high solvent evaporation rate, respectively.

Fig. 4. Morphology of the final aggregates for different evaporation rates, A: $m = 0.001 d_1 \Delta t^{-1}$, B: $m = 0.05 d_1 \Delta t^{-1}$, C: $m = 0.1 d_1 \Delta t^{-1}$ and D: $m = 0.5 d_1 \Delta t^{-1}$.

distinct in the vertical direction. This verified the phenomena of sweepage aggregation mentioned above. The thickness h of the final aggregates (layer) and the evaporation rate m followed well a power law relation as: $h = 16.9 m^{-0.27}$. The results indicated that there was a competition for the aggregation between Brownian motion and the surface falling. For slow evaporation, the particles and clusters mainly undergo Brownian motion, and the aggregation still resembles the pure DLCA. For fast evaporation, the surface clusters can be formed in a short time, and it can sweep up the particles or clusters underneath, insomuch it alters the growth mechanism.

In summary, the solvent evaporation to some extent can accelerate the process of aggregation, reduce the isotropism of the final aggregates, but have inconspicuous influence on the cluster-size distributions. This simulation implies that the extended classic CCA model can be applied to the open system with solvent evaporation in the regime of DLCA.

3.2. RLCA with solvent evaporation

Fig. 5 shows the simulated evolution of the weighted average cluster size versus the aggregation time under different evaporation rates in the regime of RLCA. The shapes of the curves are very similar to that of the classic RLCA ($P_1 = 0.01$, $m = 0 d_1 \Delta t^{-1}$) [6], except for the curve that represents the situation of classic DLCA ($P_1 = 1$, $m = 0 d_1 \Delta t^{-1}$). The effect of solvent evaporation for RLCA was more pronounced than those for DLCA. As expected, decreasing m led to a slower time evolution of aggregation and caused the $S(t)$ to peak later at lower values. In the mean time, it was

Fig. 5. A log-log plot of the weighted average cluster size as a function of time for different solvent evaporation rates. The case of $P_1 = 0.01$, $m = 0 d_1 \Delta t^{-1}$ corresponds to the classic RLCA; the case of $P_1 = 1$, $m = 0 d_1 \Delta t^{-1}$ corresponds to the classic DLCA.

observed that the solvent evaporation also increased the aggregation process, and this effect, to some extent, was more significant than the effect of the sticking probability, since the DLCA curve intersected some of the other curves ($m \geq 0.05 d_1 \Delta t^{-1}$). Similar to those in Fig. 2, the curves for high evaporation rates also did not reach a plateau value due to the drying out of the solvent.

Fig. 6. Snapshots of a slow aggregation process at the time of the suspension surface height equals to A: $80d_1$, B: $60d_1$, C: $40d_1$ and D: $20d_1$ with $m = 0.1d_1\Delta t^{-1}$ and $P_1 = 0.25$.

In order to visualize the phenomena of the sweepage aggregation, snapshots of a slow aggregation process for $P_1 = 0.25$ and $m = 0.1d_1\Delta t^{-1}$ were taken (Fig. 6). The morphology of the aggregates at different times shows that the aggregation occurred mainly at the top of the suspension. The results suggest that, differing from the rapid aggregation, the low sticking probability provided more chance for solvent evaporation to alter the aggregation mechanism by the sweepage aggregation at the suspension surface.

3.3. Compare DLCA with RLCA

At the same evaporation rate $m = 0.1d_1\Delta t^{-1}$, the cluster-size distributions of rapid aggregation and slow aggregation are compared in Fig. 7. The difference of the cluster size in the rapid aggregation is not evident (Fig. 7A). The particles below the surface can aggregate easily when they have high sticking probability benefited from their freely Brownian motion. This leads to the formation of the aggregates with more branches and smaller fractal dimensions.

Fig. 7. Snapshots of the cluster-size distributions of rapid aggregation (A: $P_1 = 1$) and slow aggregation (B: $P_1 = 0.01$) at the time of the suspension surfaces descended to $80d_1$. Both used the same evaporation rate $m = 0.1d_1\Delta t^{-1}$.

Fig. 8. Effect of different solvent evaporation rates ($m = 0.001, 0.01, 0.05, 0.1, 0.25, 0.5, 1.0$ and $2.0d_1\Delta t^{-1}$) and sticking probabilities on the number of the remaining clusters when the aggregation process terminated.

While in slow aggregation, most aggregations happened at the top of the system due to the fall of the suspension surface, thus the system appears inhomogeneous (Fig. 7B), and its final aggregates tend to have compacted clusters with higher fractal dimension, which is consistent with the earlier results [15,16,28] when the solvent evaporation rate is not very high.

When the aggregation process terminated, the number of the remaining clusters were counted at different solvent evaporation rates and sticking probabilities (Fig. 8). When the evaporation rates are very small ($m \leq 0.01d_1\Delta t^{-1}$), the aggregation processes are slow and are mainly controlled by Brownian motions. The number of remaining clusters is nearly identical for different sticking probabilities. When the evaporation rates are very large ($m \geq 1.0d_1\Delta t^{-1}$), the aggregation processes are mainly controlled by solvent evaporation. The number of the remaining clusters tends to be also identical for different sticking probabilities. However, the number of remaining clusters becomes fluky in the range of the medium evaporation rates. This implies that when the aggregation process is partly controlled by Brownian motions and partly by solvent evaporation, the number of the remaining clusters becomes uncertain, which becomes more evident at the low sticking probabilities (e.g. $P_1 = 0.01$).

Comparatively, the coupled perikinetic aggregation and sweep aggregation processes in the regime of RLCA are more complicated than that in the regime of DLCA.

4. Conclusions

The focus of this research is to extend the classic CCA model to the open systems. To verify the applicability of the extended model

for open systems, a case of solvent evaporation was simulated. From the definitions, it can be found that the definitions of $D_1(t)$ and $P_1(t)$ are flexible to account for the features of the open systems. Although the simulations were mainly based on the definition of $D_1(t)$ to demonstrate the extended CCA model to open systems, a similar approach can be used to show that the new model can be used to simulate aggregation in open systems with the definition of $P_1(t)$.

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