A Monte Carlo Simulation Study on: Deposition, Diffusion, and Aggregation

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Abstract. In this paper we present a Monte Carlo simulation program to implement a single model: deposition, diffusion, and aggregation (DDA). By using the simulation program, we show in the following that the DDA model generates a wide variety of fractal structures characteristic of different models such as diffusion-limited aggregation (DLA) or cluster-cluster aggregation (CCA), but such DLA and CCA models do not incorporate the possibility of cluster-mass dependent deposition. Starting from those kernels, aggregation laws to describe the interaction between free and deposited clusters were proposed and a simple cluster-mass dependent deposition law was employed. The simulation results are then discussed in the frame of the relative strength of parameters.

Keywords: Monte Carlo; Deposition; Simulation.

1. Introduction

Fractal aggregation is a universal phenomenon in nature, which has been widely applied to physical, social science and chemical science. Fractal aggregation is a complex process under the influence of many factors, such as system temperature, electrolyte concentration, gravity potential energy, etc. Monte Carlo simulation is an important computer simulation method, which can be used to study on the interaction of colloidal particles[1]. In recent years, a lot of research has been carried out. Witten and Sander[2] proposed diffusion-limited aggregation (DLA). Meakin and Kolb[3] proposed the cluster-cluster aggregation (CCA) model, which is based on the aggregation of the colloidal particles and dusts in the atmosphere. These exist models diffuse and aggregation, but they do not allow the continual injection of new particles via deposition[4-7]. Consequently, Jensen[7,8] developed a model that incorporates the three physical ingredients of clusters: deposition, diffusion and aggregation (DDA). The model taking into account the gravity and Brownian motion, truly reflect the aggregation process of the gravitational field. In general colloid systems, when the clusters reach a certain mass, the effect of gravity cannot be ignored. The effect of gravity on the aggregation process includes two aspects: on the one hand, gravity affects the collision probability of the particles, on the other hand, it will cause the particles to reorganize and form a denser structure. The DDA model assumes that the influence of other factors is not considered, and the dispersed phase particles are only affected by two mechanisms of action. One mechanism is that particles are affected by Brown's force and move randomly around, the other is that the particles are affected by gravity and deposition[8].
There's been a lot of research on DDA model[7-11]. However, the existing DDA model programs are few and most of them are based on two-dimensional space. In our research, the simulation program can dynamically display the movement process of particles by three-dimensional images, which is more convenient for researchers to observe the fractal structure of particles and clusters. Furthermore, in order to make the program more simple and more convenient for users, we used a cross-platform Java program language to implement DDA model.

2. The Method of DDA Model

When implementing the simulation for DDA model, the main work is to implement the simulation of particles’ and clusters’ deposition motion, Brownian motion and aggregation.

The first work is to initialize the system, monomer particles are randomly distributed in a cubic box of height $L$. When give the particle concentration $C$, total number of monomer particles $N$ can be calculated by the following equation:

$$N = C * L^3$$  

There are mainly two kinds of motion of particles in the system: Brownian motion and deposition motion. One particle or cluster will be selected to move is represented by an acceptable probability $P_{move}$. Whether it is diffusion or deposition, the selection is expressed by the following equation[5,6]:

$$P_{move} = \frac{D_i}{D_{max}}$$  

Where $D_i$ is the diffusion coefficient of the cluster consisting of $i$ particles, $D_{max}$ is the maximum diffusion coefficient for any cluster in system. After calculating the moving probability of the selected particles, a random number $x_i$ is generated in the range $[0,1]$, and the cluster is moved only if $x_i < P_{move}$.

The diffusion coefficient $D_i$ is defined as:

$$D_i = D_1 * S^\gamma$$

Where $D_1$ is the diffusion coefficient of the primary particles, $S$ is the mass of cluster, which is expressed by the number of particles contained in the cluster. $\gamma$ is the diffusion exponent, the key factor influencing the particles movement.

In the process of Brownian motion, the clusters tend to disperse, which leads to the selected particle or cluster can move to six different directions (up, down, left, right, front and back) randomly. While in the process of deposition motion, the moving direction of cluster is downwards. Computers can only serialize parallel process. therefore, two motions can be coupled by choosing the direction of moving of the cluster, the process is as follows.

It is assumed that the probability of six directions being selected is $P_u, P_d, P_l, P_r, P_f$ and $P_b$, they follow the equation:

$$P_u + P_d + P_l + P_r + P_f + P_b = 1$$  

When only Brownian motion is considered, the probability of each direction being selected is 1/6, the following equation can be obtained:

$$P_u + P_d = P_l = P_r = P_f + P_b = \frac{1}{3}$$  

When deposition motion is taken into account, the probability of the cluster moving downwards increases. According to symmetry, on the horizontal axis of two direction probability should be the same: $P_l$ is equal to $P_r$, and $P_f$ is equal to $P_b$, which can be expressed:

$$P_l = P_r = P_f = P_b = \frac{1}{6}$$

The range of $P_u$ and $P_d$ can be determined:
Consider the combination of Brownian and deposition, the probability of the cluster moving downwards $P_d$ evaluated as:

$$P_d = \frac{1}{6} + P_e \ast i^{-1/D_f}$$

Where $D_f$ is friction coefficient of the cluster, $P_e$ is called the Peclet number of the clusters, which gives the deposition strength of the system of clusters. If the value of $P_d$ we calculate in equation (9) exceeds the range in equation (7), the probability is set equal to $P_d = 1/3$. Following this, the probability of the cluster moving upwards can be achieved from equation (5):

$$P_u = \frac{1}{3} - P_d$$

In this way, equations (6) and (9)-(10) can give the probability of six directions being selected in the system. In the whole process, moving direction is chosen based on Roulette-Wheel Selection strategy[12], six directions filled a section with length of 1, which length is equal to the probabilities respectively. Since then, a random number $y$ over the range [0, 1] is generated, $y$ is used to match the section and chose the corresponding moving direction. If there is no collision between clusters after moving, it will select next one. When collision occurs between two clusters (one is a cluster with mass $i$, and another mass is $j$) they stick together forming a new cluster with the sticking probability $P_{ij}$[5,13]:

$$P_{ij} = P_0 \ast (i \ast j)^\sigma$$

Where $P_0$ is the sticking probability of single particles, and $\sigma$ is the sticking probability exponent. In the process of aggregation, a random number $x_2$ over the range [0, 1] is generated, and the clusters are sticking only if $x_2 < P_{ij}$.

3. The Simulation Results of DDA Model

In our research, we used an on-lattice simulation, one three-dimensional cube represents the system space, which is divided into equal-size little boxes, the side length of the little box is regarded as a unit length. After inputting the initial parameters of the program, $N$ non-overlapping identical particles distributed randomly in the cube with side-lengths of $L$, each particle occupies a little box. When Brownian motion and deposition motion process progresses, the program will update coordinates of particles and display all the particles in the cube.

(a)                       (b)                      (c)
As we can see in equation (9), the value of parameter $P_e$ is the key factor to determine the deposition speed of clusters. It is reasonable to assume that the values of $D_1$ and $P_1$ remain constant when we discuss the value of $P_e$. We set the parameters $L = 100$, $c = 0.01$ and can obtain 10000 particles by the equation (1), at the same time, set the parameters $\gamma = -0.5$ and $\sigma = 0.5$. Subsequently, the results of 10000 particles in various stages are shown in Figures 1, 2 when the parameter $P_e = 0.01$ and $P_e = 0.1$.

It can be seen intuitively from Figure 1 and Figure 2, there are mainly single particles and small clusters in the systems at early stages, and deposition is not obvious. As the diffusion and aggregation process progresses, small clusters become big ones, so that some clusters began to depositing, the clusters anisotropy become very distinct in the vertical direction. At the later stages, the deposition is more
obvious. Afterwards, almost all the clusters have been deposited to the bottom of the cube. In the meantime, it was observed that the deposition motion limits Brownian motion to some extent. By analyzing the difference between Figure 1 and Figure 2, we find that the larger the value of parameter $P_e$ is, the more obvious the deposition motion is, it’s consistent with our previous analysis. When $P_e = 0.1$ (Figure 2), the deposition motion appears earlier (Figure 2c). However, when $P_e = 0.01$ (Figure 1), Brownian motion plays a leading role (Figure 1c).

**Figure 3.** The results of 10000 particles in three stages: the numbers of clusters = (a)5000; (b)1000; (c)1.(parameter $\gamma = -1$).

**Figure 4.** The results of 10000 particles in three stages: the numbers of clusters = (a)5000; (b)1000; (c)1.(parameter $\gamma = 0$).

**Figure 5.** The results of 10000 particles in three stages: the numbers of clusters = (a)5000; (b)1000; (c)1.(parameter $\gamma = 1$).
From the equation (3), we can obtain that the cluster’s diffusion coefficient $D_i$ is not only related to the mass of cluster, but also is influenced by the parameter $\gamma$. We have done the simulation test with different values of parameter $\gamma$ for 10000 particles and the results are shown in Figures 3,4,5 when particles in different stages. Three cases can be distinguished:

- $\gamma < 0$: by analyzing equation (3), small clusters diffuse faster than large ones. Figure 3a and Figure 3b shows that there is little difference in cluster size at different stages in the system. Consequently, the deposition caused by gravity has little difference, clusters are deposited layer by layer to the bottom. As we can be seen from Figure 3c, all particles aggregate into a large cluster and deposit to the bottom. The whole simulation process can be completed quickly.

- $\gamma = 0$: there has nothing to do with the mass of cluster. As shown in Figure 4b, the cluster sizes in the system are quite different, large clusters can deposition to the bottom earlier due to the gravity effect, single particles and small clusters are still distributed above the system. Comparing the Figure 3c and Figure 4c, when $\gamma = 0$, the final cluster formed at the bottom is relatively dense (Figure 4c).

- $\gamma > 0$: because of the large clusters are easier to diffuse, there is always a large cluster in the whole process, and the others are almost the single particles and small clusters (Figure 5b). At the middle and later stages, the small clusters near the bottom are gradually combined with the large clusters at the bottom, and gradually aggregate layer by layer, so that the final large cluster is branched as shown in Figure 5c.

From the equation (11), we can see that the parameter $\sigma$ is the key factor to determine sticking probability. Similar to parameter $\gamma$, we have done the simulation test with different values of parameter $\sigma$ and the results are shown in Figures 6,7,8 when they are in different stages, three cases can be distinguished:

![Figure 6](image1.png)

**Figure 6.** The results of 10000 particles in three stages: the numbers of clusters = (a)5000; (b)1000;(c)1.(parameter $\sigma = -1$.)

![Figure 7](image2.png)

**Figure 7.** The results of 10000 particles in three stages: the numbers of clusters = (a)5000; (b)1000;(c)1.(parameter $\sigma = 0$.)
• $\sigma < 0$: there is a negative correlation between sticking probability and cluster size, that is to say, small one has great sticking probability. on the contrary, large clusters are not easy to aggregate. As the simulation goes on, the sticking probability of clusters becomes very small, and clusters are mainly conducted by deposition motion. At last, a very dense cluster is formed and very close to the bottom as shown in Figure 6c.

• $\sigma = 0$: the mass of clusters has no effect on the sticking probability, the rate of aggregation mainly depends on the parameter $P_0$.

• $\sigma > 0$: with the increase of the cluster size, the sticking probability will increase, when the cluster becomes large enough, two clusters will aggregate after they collide. Compared with the case of $\sigma < 0$, aggregation has a greater influence on the process of cluster diffusion (Figure 6 and Figure 8).

4. Conclusion

The focus of this research is to propose a Monte Carlo simulation program for describing a single model: deposition, diffusion, and aggregation (DDA). We find that the model permits one to distinguish the effects of deposition, diffusion and aggregation, and that tuning the relative strength of parameters, generates a rich range of fractal structure and close to real physical phenomena. In general, the simulation program is useful when the fractal nature of the deposited clusters plays a key role in the subsequent uptake of free clusters.

Acknowledgements

This work was financially supported by the National Natural Science Foundation of China (41271292), the Key Project of Chongqing Science and Technology Bureau (cstc2019jscxgksbX0103), and the Fundamental Research Funds for the Central Universities of China (SWU2009107).

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