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Two-dimensional Monte Carlo simulations of a suspension comprised of magnetic and nonmagnetic particles in gradient magnetic fields

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ABSTRACT

Distributions of particles in a suspension comprised of magnetic particles (MPs) and nonmagnetic particles (NPs) under gradient magnetic fields are vitally important for the preparation of magnetic–nonmagnetic functionally graded materials (FGMs). In the present study, the effects of magnetic field gradient, magnetic interaction between MPs and concentration of NPs on the distributions of particles in the suspension are investigated using a two-dimensional Monte Carlo simulation. The results show that a gradient distribution of MPs is formed under gradient magnetic fields and increases with increasing the field gradient. However, as the interaction between MPs increases, the distribution gradient decreases, accompanied by the formation of chain-like MP clusters. Moreover, NPs are found to hinder the translation of MPs along the field direction. As the NP concentration increases, the translation of MPs becomes difficult.

1. Introduction

Functionally graded materials (FGMs) are a class of special composites with continuous spatial variations of composition and resulting mechanical, physical and chemical properties [1–5]. FGMs have normally been fabricated by means of vapor deposition, powder metallurgy, plasma spraying, self-propagation high temperature synthesis, electro deposition, etc. [6–9]. Another approach has recently been developed in our group to prepare FGMs under gradient magnetic fields, based on the distinct difference in magnetic susceptibility between the components in a suspension [10,11]. In the suspension, magnetic particles (MPs) are driven by the gradient magnetic field to move toward high field regions, resulting in the formation of continuous composition gradient. After subsequent drying and sintering, final FGMs are obtained. Therefore, distributions of both MPs and nonmagnetic particles (NPs) in gradient magnetic fields are vital essentials for the fabrication of FGMs based on this approach.

In the researches of magnetorheological fluids and magnetic fluids, the distributions of MPs in suspensions under various magnetic fields have been studied experimentally and theoretically [12–14]. When a magnetic field is applied to a colloidal suspension comprised of MPs, dipolar interactions are induced within the particles, and various particle distributions are formed. Chain-like structure has been found in low magnetic fields [15,16], while columnar, lamellar and striped structures in high fields [17,18], ellipsoidal structures in pulsed fields [19] and layered structures in rotating magnetic fields [20]. However, understanding of the distributions of MPs under gradient magnetic fields is still weak. Furthermore, even under uniform magnetic fields, very few works have been done on configuration of suspensions containing both MPs and NPs [21].

In our previous work, effects of the magnetic field gradient and concentration of MPs on the final structure of suspension have been studied [22]. However, it is difficult to directly observe the distribution of particles in suspension and, therefore, to study the influencing factors through experiments. Furthermore, not all the factors we are interested in can be reflected by the available experimental conditions, for example, an extremely high field gradient in a large area is unavailable.

For the above-mentioned reasons, molecular simulation is a useful method to investigate the structure of aggregates in colloidal suspension. Dynamics methods, such as the Brownian dynamics method and the Stokesian dynamics, have been normally used to investigate the dynamic behavior of particles in a flow field. The Monte Carlo method is mainly used to investigate the static equilibrium structures of suspension [23,24]. This method is more suitable for our researches than dynamics methods. Satoh et al. [23,24] have investigated particle distribution in a suspension composed of magnetic particles under uniform magnetic fields. In Satoh's work, cluster-moving Monte Carlo algorithm was used. It was developed for use in the

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simulation of a strongly interacting system by allowing a cluster to move as a composite single particle, which was based on the supposition that a particle move rarely overcame strong interactions to escape from its cluster to form larger clusters. It is a further confine based on usual Monte Carlo algorithm, and only can be used in the strong interaction systems. For a strong interaction system, simulation time is shortened by using cluster-moving Monte Carlo algorithm. In this work, the magnetic interaction is not very strong in most cases. Thus, the cluster-moving algorithm seems unsuitable in our work. We have investigated the distributions of particles under uniform magnetic field using usual Monte Carlo algorithm, which gives reasonable results [25]. In this work, the distributions of particles in a suspension comprised of both MPs and NPs under gradient magnetic fields are studied using two-dimensional Monte Carlo simulations.

2. Materials and methods

2.1. Physical model of the suspension

The position of each particle in the suspension is supposed to remain unchanged during the subsequent drying and sintering process for fabrication of FGMs. The model for the suspension is given as follows.

MPs and NPs with identical diameter d are dispersed homogeneously in the suspension. The number of particles is defined as $N = N_m + N_n$, where N_m is the number of MPs, N_n the number of NPs. The particles are coated with a steric layer of thickness δ. When a gradient magnetic field is applied on the suspension, MPs are magnetized with the moments aligned along the field direction; while NPs are not affected. The motion of a magnetic particle is typically caused by three kinds of energy: the interaction energy μ_i^H between particle i and the magnetic field, the interaction energy μ_{ij}^m between magnetic particle i and j, and the interaction energy μ_{ij}^v due to the overlapping of the steric layers. The motion of a nonmagnetic particle is controlled only by the overlapping energy μ_{ij}^v. The above-mentioned energies can be expressed as [23,24]:

$$\mu_i^H = -\mathrm{kT}\lambda_H \mathbf{n}_i \mathbf{H}/H \tag{1}$$

$$\mu_{ij}^m = \mathrm{kT}\lambda_m \frac{d^3}{r_{ji}^3}\{\mathbf{n}_i\mathbf{n}_j - 3(\mathbf{n}_i\mathbf{t}_{ji})(\mathbf{n}_j\mathbf{t}_{ji})\} \tag{2}$$

$$\mu_{ij}^v = 2\lambda_v \mathrm{kT}\left\{1 - \frac{r_{ji}}{dt_\delta}\ln\left[\frac{(t_\delta+1)d}{r_{ji}}\right] - \frac{r_{ji}-d}{dt_\delta}\right\} \tag{3}$$

where k is the Boltzmann's constant, T the absolute temperature of the suspension, H the field strength of the magnetic field $\mathbf{H}$, r_{ji} the magnitude of the vector $\mathbf{r}_{ji}$ drawn from particles i to j, $\mathbf{n}_i$ and $\mathbf{t}_{ji}$ denote the unit vectors given by $\mathbf{n}_i = \mathbf{m}_i/m_i$ $(m_i = |\mathbf{m}_i|)$ and $\mathbf{t}_{ji} = \mathbf{r}_{ji}/r_{ji}$, and t_δ the ratio of the thickness of the steric layer δ to the radius of particle, which is expressed as $2\delta/d$. The dimensionless parameters λ_H and λ_m and λ_v are written as [23,24]:

$$\lambda_v = \frac{\pi d^2 n_s}{2} \tag{4}$$

$$\lambda_H = \mu_0 \mathrm{mH/kT} \tag{5}$$

$$\lambda_m = \mu_0 m^2/4\pi d^3 \mathrm{kT} \tag{6}$$

where n_s is the number of surfactant molecules per unit area on the particle surface, μ_0 the permeability of free space, m the magnitude of the magnetic moment of MPs, which is proportional to the volume $\pi d^3/6$.

2.2. Simulation parameters

In this study, two-dimensional Monte Carlo simulations based on Metropolis's algorithm [26] are conducted to investigate the distributions of particles in the suspension comprised of MPs and NPs. The simulation parameters are as follows.

The side length of the square simulation box L is $61.78d$. Except for Section 3.3, the particle number N and the area fraction of the particles to the system region ϕ are taken as $N = 2916$ and $\phi = 0.6$. The number of MPs is taken as $N_m = 400$ for all the sections. The initial configuration of particles is taken randomly. The thickness of the steric layer δ and the dimensionless parameter λ_v are taken as $\delta = 0.01d$ and $\lambda v = 50$, respectively. The cut-off radius r_{coff} for the particle–particle interactions is taken as $r_{coff} = 10d$. The maximum displacement of a particle in one trial move is $0.005d$.

Fixed boundary condition is applied in the X-direction and periodic boundary conditions in the Y direction. Under the gradient magnetic field, the magnetic field strength changes gradually along the field direction. Thus, the dimensionless parameter λ_H in the simulation is defined as

$$\lambda_H = 10 + g_H x \tag{7}$$

where both g_H and x are dimensionless parameters, g_H the magnetic field gradient, x the distance from the left side in the simulation box, which ranges from 0 to 61.78. According to Eq. (7), the minimum value of parameter λ_H is equal to 10, which means the magnetic field strength is relative high in the simulation. To reduce the computational complexity, the moments of MPs are fixed along the field direction. All simulations are carried out more than 400,000 steps to get a stable distribution of particles. To present simulation results, two-dimensional distribution of particles on the XY plane are plotted with MATLAB 7.3.0 (R2006b) software. The gradient magnetic field is horizontal with its strength increasing from left to right. Red and blue circles represent MPs and NPs, respectively.

3. Results and discussion

3.1. Effects of magnetic field gradient on distributions of particles in suspension

In this section, the interaction between MPs is setting as $\lambda_m = 100$. The effects of the magnetic field gradient on distributions of particles are studied with $g_H = 0$, 10, 100 and 1000, respectively. Simulation results are given in Fig. 1. For $g_H = 0$, which suggests a uniform magnetic field, chain-like MP clusters are formed and distributed uniformly along the field direction (Fig. 1a). For a small field gradient, such as $g_H = 10$, MPs move slightly toward the high field regions, i.e. from left to right (Fig. 1b). With increasing g_H, the distance of particles' translation along the field direction increases (Fig. 1c). In the case of a high gradient, such as $g_H = 1000$, MPs concentrate in the right half of the box and simultaneously compress parts of NPs into a dense mass (Fig. 1d).

In order to give a quantitative description of the effects of the field gradient, the cumulative distributions of MPs along the field direction are analyzed and plotted in Fig. 2. The abscissa of the curve indicates the distance from left side of simulation box, and the ordinate is the dimensionless cumulative distributions of MPs. For $g_H = 0$, the curve starts at $x = 0$ and rises linearly to 1 at $x = 61.78$, suggesting a nearly uniform distribution along the X-axis. As g_H increases, both the starting points and the slopes of curves increase. For a high gradient such as $g_H = 1000$, the curve starts at $x = 41.9$ and rises steeply to 1, indicating a much higher