Colloids and Surfaces A: Physicochem. Eng. Aspects 483 (2015) 328–340

Contents lists available at ScienceDirect

Colloids and Surfaces A: Physicochemical and Engineering Aspects

journal homepage: www.elsevier.com/locate/colsurfa

Brownian dynamics simulation of a dispersion composed of disk-like hematite particles regarding aggregation phenomena

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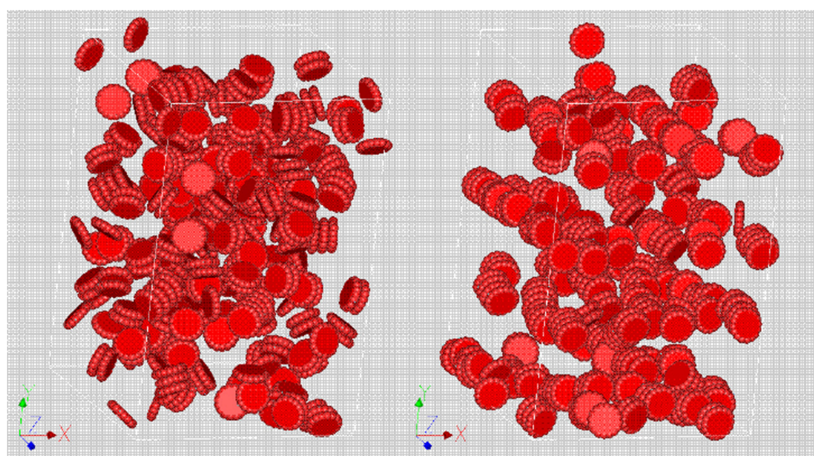
HIGHLIGHTS

- Column-like clusters are significantly formed at strong magnetic interactions.
- The rotational Brownian motion has significant influence on the formation of clusters.
- An applied magnetic field enhances the formation of column-like clusters.
- A shear flow does not have significant influence on the internal structure of clusters.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 4 December 2014
Received in revised form 24 July 2015
Accepted 24 July 2015
Available online 31 July 2015

Keywords:
Magnetic colloidal dispersion
Brownian dynamics method
Disk-like hematite particle
Simple shear flow
Aggregation phenomenon
Internal structure
Orientational correlation function

ABSTRACT

We have investigated aggregation phenomena in a suspension composed of disk-like hematite particles by means of Brownian dynamics simulations. The magnetic moment of the hematite particles lies normal to the particle axis direction, and therefore the present Brownian dynamics method takes into account the spin Brownian motion about the particle axis in addition to the ordinary translational and rotational Brownian motion. These particles are assumed to perform translational and rotational Brownian motion in a simple shear flow with addition of a uniform magnetic field applied in the direction normal to the shearing plane. The main results are summarized as follows. Significant column-like clusters are formed at a magnetic particle–particle interaction much larger than in the case of a magnetic spherical particle suspension. This is because the rotational Brownian motion has significant influence on the formation of clusters in a suspension composed of disk-like particles with large aspect ratio. An applied magnetic field is found to decrease the formation of column-like clusters. A shear flow is found not to have significant influence on the internal structures of clusters, but it does influence the cluster size distribution of the column-like clusters.

1. Introduction

From the viewpoint of developing new magneto-rheological fluids [1], magnetic particles with a variety of shapes and magnetic

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http://dx.doi.org/10.1016/j.colsurfa.2015.07.047
0927-7757/

properties have a significant potential as functional materials over the magnetic spherical particles. Nowadays, the development of the synthesis technology enables one to generate magnetic particles with various shapes such as rod-like, disk-like and cubic-like particles [2–12]. In addition to the application of particle suspensions to magneto-rheological fluids, possible applications are high-density recording materials [13,14], optical units [15–19], and surface modifying technology [20].

Ordinary magnetic rod-like particles are ferromagnetic, and therefore require some techniques to synthesize a stable particle suspension: for example, it may be necessary to employ surfactant agents for preventing dispersed particles from aggregating and sedimenting [21]. In contrast, hematite particles have a weaker magnetization in comparison with magnetite particles, so that if the particle diameter can be controlled in the synthesis process, it is relatively straightforward to obtain a stable dispersion composed of hematite particles.

Spindle-like hematite particles have a characteristic feature in that they are magnetized in the short axis direction [9,10], so that raft-like clusters are formed in the magnetic field direction [22]. Another characteristic feature of a spindle-like particle dispersion is that the viscosity due to magnetic properties becomes negative under a certain condition of an applied magnetic field. This negative magneto-rheological effect was theoretically predicted from the theory based on the orientational distribution function in a simple shear flow [23–25]. Quite recently, by performing an experiment using a cone-plate type rheometer, we have succeeded in experimentally verifying that the negative magneto-rheological effect arises in an actual situation [26]. Our research group has been investigating an effective technique for control of the characteristic aggregate structures and negative magneto-rheological effect.

In addition to spindle-like particles, disk-like hematite particles are also expected to exhibit characteristic orientational features and aggregate structures, resulting into complex magneto-rheological properties that are dependent on these factors. Aggregation phenomena in thermodynamic equilibrium had already been clarified in detail by means of Monte Carlo simulations [27].

The objective of the present study is to address a dispersion composed of disk-like hematite particles in order to investigate aggregation phenomena in a simple shear flow by means of Brownian dynamics simulations. The present Brownian dynamics method takes into account the spin rotational Brownian motion about the particle axis, in addition to the ordinary translational and rotational Brownian motion. The validity of the present simulation method is clarified by comparing with results obtained by the fully-established Monte Carlo method. We then investigate the influence of the magnetic particle-field and the particle–particle interaction and the shear rate on particle aggregation phenomena. For a qualitative discussion, snapshots of aggregate structures are used and for a quantitative discussion we focus on the cluster size distribution, the radial distribution function and the orientational correlation functions of both the particle axis directions and the magnetic moment directions.

2. Particle model

As a particle model in the present study, oblate hematite particles are modeled as a disk-like particle with cross section of spherocylinder, as shown in Fig. 1. This disk-like particle is formed of a cylindrical part with diameter d and a torus part with the cross section of a half circle covering the cylinder. The diameter and the thickness of the particle are denoted by d_1 and b_1, respectively. The particle is assumed to be magnetized in a direction normal to the particle axis direction, i.e. in the plane of the disk, with a magnetic

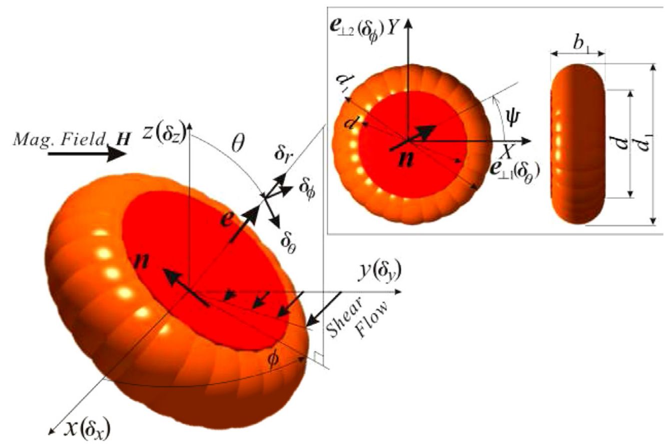

Fig. 1. Magnetic disk-like particles with cross section of spherocylinder in a simple shear flow under the circumstance of an applied magnetic field in the y-direction.

dipole moment at the particle center. We employ the notation $\boldsymbol{e}_i$ for the particle direction of an arbitrary particle i, $\boldsymbol{m}_i$ (=$m\boldsymbol{n}_i$) for the magnetic moment and $\boldsymbol{n}_i$ for its direction. The interaction of the magnetic moment with a uniform applied magnetic field $\boldsymbol{H}$ (=$H\boldsymbol{h}$, where $\boldsymbol{h}$ is the unit vector denoting the field direction) induces the magnetic torque $\boldsymbol{T}_i^{(H)}$ acting on the particle in such a way that the magnetic moment tends to incline in the magnetic field direction. In a many-particle dispersion, the force $\boldsymbol{F}_{ij}^{(m)}$ and torque $\boldsymbol{T}_{ij}^{(m)}$ are due to the magnetic interaction acting on particle i by particle j, and these expressions are shown in Reference [28].

In order to prevent dispersed particles from aggregating and sedimenting, the particles are assumed to be coated with a steric layer of surfactant molecules. In the following, we show a treatment of the repulsive interactions due to the overlap of steric layers in Brownian dynamics simulations. At the present time an expression for the repulsive force acting between two disk-like particles with a steric layer is not known, so that we here apply a known expression for two spherical particles to the model of the interaction of the present disk-like particles. In order to apply this repulsive expression, the model of our disk-like particle is constructed to be composed of spherical particles. However, a simple modeling with constituent spherical particles located as specific positions in the disk-like particle may give rise to a torque that induces unreasonable rotational motion. Hence, we employ the following sophisticated modeling in order to remove this kind of unreasonable behavior arising from the simpler model. Instead of locating spherical particles at predefined, i.e. fixed positions in the disk, the spherical particles are newly located at appropriate positions whenever particles overlap. Firstly, we evaluate the nearest or deepest overlapping positions of the two disk-like particles of interest. For instance, using the figure on the left-had side in Fig. 2, the disk-like particle on the upper side is assumed to be in a configuration where the disk plane (not on the torus section) on the lower disk-like particle is the nearest to the torus part of the upper disk-like particle. In this configuration of the two disk-like particles, a spherical particle with diameter b_1 is placed at each disk-like particle. The spherical particle on the upper disk-like particle is moved along the torus part and another spherical particle of the lower disk-like particle is moved along the disk plane in order to obtain the positions of these spherical particles that give rise to the shortest distance. Then a spherical particle with diameter b_1 is placed at these positions on each disk-like particle. The other spherical particles are placed around the starting spherical particle in a contacting and most-packed situation, completing the formation of each disk-like particle. The concrete way of locating the other spherical particles is explained below in detail. As shown in Fig. 2,