Structure Formation of Magnetic Particles under Magnetic Fields toward Anisotropic Materials

T. Ando¹, D. Katayama¹, N. Hirota², O. Koike³, R. Tatsumi⁴, M. Yamato⁵

¹ College of Industrial Technology, Nihon University, Narashino 275-8575, Japan
² Fine Particle Engineering Group, National Institute for Materials Science, Tsukuba 304-0047, Japan
³ Products Innovation Association, Tokyo 113-8656, Japan
⁴ Environmental Science Center, The University of Tokyo, Tokyo 113-8656, Japan
⁵ Graduate School of Urban Environmental Sciences, Tokyo Metropolitan University, Hachioji 192-0397, Japan

Corresponding author: ando.tsutomu@nihon-u.ac.jp

Abstract
We performed numerical simulation of structure formation of magnetic particles under magnetic field toward anisotropic material development. In this study, we examined the unsteady process of the structure formation and its feature using magnetic particles with diameter of micrometers. This study evaluated the process of structure forming with various particle diameters and particle volume concentrations using the Non-dimensional Boundary Area (NBA). The structure formed by the magnetic particles does not depend on the particle diameter but depends on the particle volume concentration. In the case of particle volume concentration \( \phi = 5 \) vol\%, the bundle of chain-like cluster is not made and each chain-like cluster by magnetic particles is almost single chain. However, in the case of more than \( \phi = 10 \) vol\%, the bundle structure formed by contacts of multiple chain-like clusters is proceeding. On the process of structure formation by magnetic particles, firstly, the single particle rotates by itself with orienting the magnetic moment to the direction of the applied magnetic field. After then, particles connect with each other and the chain-like cluster is formed.

Keywords: Magnetic particles, Chain-like cluster, Anisotropic Material, Discrete element method, Simulation

Introduction
In recent material processing, the use of magnetic fields that can control the orientation of texture and structure of material without contact has received attention²⁻⁴. There are two methods in order to improve the mechanical properties of the material using magnetic fields. One is a method of orienting the magnetic anisotropic substances to the same direction in the material. Another is a method of controlling the direction of the structure formed by the aggregated particles in the medium. For the latter, when a magnetic field is applied to the magnetic particles dispersed in the medium, the magnetic moments of all the magnetic particles are aligned in parallel to the magnetic field, and the particles make chain-like cluster with connection in a parallel direction to the magnetic field by the magnetic dipole interaction. So, the magnetic field is useful tool on development of anisotropic materials.

In this time, we performed numerical simulations of the structure formation of magnetic particles with diameter of micrometer toward anisotropic material development under magnetic fields. We examined the unsteady process of the formation and its feature using DEM (Discrete Element Method)⁶ including the magnetic effect. A lot of simulation researches for magnetic particles has been already published by A. Satoh⁵. But he deals with magnetic particles in magnetic fluids and particle size is on the order of tens of nanometers. Therefore, he calculates using Brownian dynamics method in which particles do not collide directly with each other. He also discusses mainly the rheology of magnetic fluid due to the structure of magnetic particle, and not structure formation in unsteady process. On the other hand, Kawamoto et. al dealt magnetic particles of 35 to 107 \( \mu \)m⁵. They measured the length of the chain-cluster formed by magnetic particles in air and oil under various magnetic fields by experiment and numerical simulation. However, they also didn’t discuss the unsteady process of structure formation and the structure of chain-like cluster on various particle volume concentrations.

Simulation model
Figure 1 shows the schematic model of numerical simulation. The cube of simulation region is a part of the inside of the container and the periodic boundary condition for a three-dimensional system is used. Magnetic particles are randomly dispersed in the container at the initial condition. Here, it was assumed that the magnetic field gradient is negligible in the calculation region. In addition, it is assumed that the particles have a single magnetic domain with saturation magnetization. In this study, we ignored the influence of gravity. In the initial state, the position of particles and the direction of magnetic moments are given randomly. After applying the magnetic field, the direction of the magnetic moment of the particle is aligned parallel to the direction of the magnetic field by the magnetic torque as shown in Fig. 2.
In the figure, $B$ is the direction of the magnetic flux density, and the magnetic moment $m$ of the particle is expressed using the meridian.

The translational motion of particles governed by Newton’s equation and the rotational motion of particles are expressed by the following equations (1) and (2), respectively.

\[
m_p \frac{dV}{dt} = F^m + F^c + F^w - \xi_{tra} V + R_{tra},
\]

\[
l \frac{d\omega}{dt} = T^H + T^m + T^c - \xi_{rot} \omega + R_{rot},
\]

Where $m_p$ is the mass of the particle, $V$ is the translational velocity of the particle, $t$ is time, $F^m$ is the magnetic dipole interaction force, $F^c$ is the contact force, $F^w$ is the van der Waals force, $R_{tra}$ is the random force by thermal fluctuation of medium, $l$ is the moment of inertia of the particle, $\omega$ is the angular velocity of the particle, $T^H$ is the magnetic torque due to the applied magnetic field, $T^m$ is the magnetic dipole interaction torque, $T^c$ is the contact torque, and $R_{rot}$ is the random torque by thermal fluctuation of medium. The coefficients of fluid resistance for the translational and rotational motions, $\xi_{tra}$ and $\xi_{rot}$, are expressed by the following equations, $\xi_{tra} = 6\eta r l$ and $\xi_{rot} = 8\pi\eta r^3$, respectively. Here $r$ is the particle radius and $\eta$ is the viscous coefficient of the solvent. Note that all the variables in bold represent vectors.

The interactions $F^m$ and $T^m$ acting between magnetic dipoles, which form a coupling structure of particles, are expressed by Eqs. (3) and (4)\(^1\). The magnetic field Torque $T^H$, which align the direction of magnetic moment of particles, is expressed by Eq. (5)\(^1\).

\[
F^m = \sum_{i\neq j}^N \left[ -\frac{3\mu_0 |m_i|^2}{4\pi l_{ij}^3} \left( n_i \cdot t_i - 3(n_i \cdot n_j)(n_i \cdot t_j) \right) t_j - \left( n_i \cdot t_i \right) n_i + \left( n_i \cdot t_j \right) n_j \right],
\]

\[
T^m = \sum_{i\neq j}^N \left[ -\frac{\mu_0 |m_i|^2}{4\pi l_{ij}^3} \left( n_j \times t_i - 3(n_i \cdot t_i) n_i \times t_i \right) \right],
\]

\[
T^H = \mu_0 m \times H,
\]

Where $N$ is the number of particles to be calculated, $\mu_0$ is the space permeability, $m_i$ is the magnetic moment of the particle, $n_i, t_i$ are the unit vector ($\mathbf{n} = \mathbf{m} / |\mathbf{m}|$), $l_{ij}$ are the interparticle distance and the unit vector in that direction, and $H$ is the applied magnetic field. Here, subscripts $i$ and $j$ are used to distinguish particles. The magnetic moment of the particle is expressed as $m = MV_p$, where the magnetization vector of the particle is $M$ and the particle volume is $V_p$. Therefore, the force and torque by the magnetic effect depend on the particle diameter.

**Simulation condition and evaluation method**

Table 1 shows physical properties of particle and solvent. The particle is a spherical body with physical properties identical to those of a nickel particle. The solvent is assumed to be an ultraviolet curable resin. Table 2 shows the simulation condition. The computational domain is $(x, y, z) = (30d_p, 30d_p, 30d_p)$ and the periodic boundary condition is employed. It was assumed that the nickel particles had the saturation magnetization under this applied magnetic field $|B| = 1$ T. In this study, the particle diameter $d_p$ and the particle volume concentration $\phi$ are variable numbers.
Table 1 Physical properties.

| Nomenclature | Value       |
|--------------|-------------|
| Particle     | Density \( \rho_p \) | \( 8.9 \times 10^3 \) [kg/m\(^3\)] |
| (Nickel)     | Saturation  | \( 490.1 \times 10^3 \) [A/m] |
| Magnetization | Diameter \( d_p \) | \( 2.5, 10 \) [\( \mu \)m] |
| Solvent      | Viscosity \( \eta \) | \( 20 \) [mPa\( \cdot \)s] |

Table 2 Simulation conditions.

| Nomenclature                | Value          |
|----------------------------|----------------|
| Computational domain       | \( 30d_p, 30d_p, 30d_p \) |
| Cutoff distance \( R_{\text{off}} \) | \( 10d_p \) |
| Applied magnetic flux density \( |B| \) | 1 [T] |
| Temperature \( T \)        | 293 [K]       |
| Particle volume            | 5, 10, 15 [vol%] |
| concentration \( \phi \)   |                |

As the evaluation method of the structure formed by the magnetic particles, we introduce the Non-dimensional Boundary Area (NBA) defined by Eq. (6).

\[
NBA = \frac{1}{N_{\text{all}}} \left( 1 - \frac{1}{c_{\text{max}}} \sum_{c=0}^{c_{\text{max}}} (c_{\text{max}} - c) n(c) \right),
\]

(6)

Where \( N_{\text{all}} \) is the total number of particles, and \( c \) is the coordination number. The coordination number is maximum value \( c_{\text{max}} = 12 \) when all the particles are perfect spheres and the all particle is same size. \( n(c) \) denotes the number of particle at the coordination number \( c \). The NBA becomes close to 1 when the particles are dispersed in the solvent, and the NBA becomes close to 0 when the particles are aggregated. If the particles form a complete one-dimensional structure, that is, the contact number of all the particles is 2, the value of NBA is 0.8333. Therefore, it means that structure of bundle formed by contacts of multiple chain-like clusters is proceeding when the value of NBA becomes 0.8333 or below.

We define the degree of orientation of the particle by the following Eq. (7).

\[
\cos \theta = \frac{1}{N_{\text{all}}} \sum_{i=1}^{N_{\text{all}}} (\mathbf{h} \cdot \mathbf{n}_i) .
\]

(7)

Where \( \mathbf{h} \) is a unit vector in the direction of the applied magnetic field \( (\mathbf{h} = \mathbf{H} / |\mathbf{H}|) \). In the case of \( \cos \theta = 1 \), it means that the directions of magnetic moment of all the particles are aligned with that of the applied magnetic field.

**Result and discussion**

As an example of these simulation results, Fig. 3 shows the structure formed by magnetic particles with a particle diameter \( d_p = 2.5 \mu \text{m} \) and a particle volume concentration \( \phi = 10 \text{ vol\%} \) under the applied magnetic field \( |\mathbf{B}| = 1 \text{ T} \). Figure 3 (a) shows that the particles form a coupling structure in the direction parallel to the applied magnetic field, and Fig. 3 (b) shows that the respective coupling structures are hold with some intervals.

Figures 4 and 5 show the time courses of NBA for the particle diameters of \( d_p = 2.5 \mu \text{m} \) and 10 \( \mu \text{m} \), respectively. The representative time \( t_0 \) is derived from the magnetic dipole interaction force \( F_0 \) and the mass of the particle \( m_p \) \( (t_0 = (m_p d_p / |F_0^{\text{rep}}|)^{1/2}) \). Here, \( F_0^{\text{rep}} = |F_0^{\text{rep}}| \) is the repulsion force if the magnetic dipole moments of two particles are same direction and the distance between the surfaces of these two particles is 0. From Figs. 4 and 5, it is seen that both values of NBA for the particle volume concentration \( \phi = 5 \text{ vol\%} \) are around 0.833 and that each chain-like cluster by magnetic particles is almost single chain. However, in the case of more than \( \phi = 10 \text{ vol\%} \), the values of NBA are significantly lower than 0.833, and the bundle structure formed by contacts of multiple chain-like clusters is proceeding. The structures formed by the magnetic particles with diameters \( d_p = 2.5 \mu \text{m} \) and 10 \( \mu \text{m} \) are almost same. From these results, it was confirmed that the structure depends only on the particle volume concentration in the medium.
Fig. 4: Time course of NBA for particle diameter of $d_p = 2.5 \, \mu m$.

Fig. 5: Time course of NBA for particle diameter of $d_p = 10 \, \mu m$.

Figure 6 shows the time course of the particle orientation, $\cos \theta$, for particle diameter $d_p = 2.5 \, \mu m$. This result shows that three lines for different particle volume concentration are overlapped. Therefore, it is seen that the particle orientation does not depend on particle volume concentration. Compared with curves of NBA in Fig. 4, curves of $\cos \theta$ in Fig. 6 are saturated quickly. Therefore, we can see that there are two steps when magnetic particles form a structure under the applied magnetic field. Firstly, the single magnetic particle rotates by itself with orienting the magnetic moment to the direction of the applied magnetic field. After then, particles connect with each other and the chain-like cluster is formed. In addition, in the case of the particle volume concentration more than $\phi = 10 \, \text{vol}\%$, the bundle formations by contacts of multiple chain-like clusters occurs after the single chain-like cluster structure, or occurs at the same time as the structure formation of the chain-like cluster.

**Conclusion**

We conducted a numerical simulation of structure formation by magnetic particles under magnetic fields toward anisotropic material development. The three-dimensional simulation with various particle diameters and particle volume concentrations was carried out using DEM. The following results were confirmed in the present study under the condition that the gravity force was ignored.

1. The structure formed by particles, that is the non-dimensional Boundary Area (NBA), does not depend on the particle diameter but depends on the particle volume concentration.
2. In the case of particle volume concentration $\phi = 5 \, \text{vol}\%$, the bundle formation of chain-like cluster is not occurred and each chain-like cluster by magnetic particles is almost single chain. However, in the case of more than $\phi = 10 \, \text{vol}\%$, the bundle formations are proceeding.
3. On the process of structure formation by magnetic particles, firstly, the single magnetic particle rotates by itself with orienting the magnetic moment to the direction of the applied magnetic field. After then, particles connect with each other and the chain-like cluster is formed.

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