Effects of Frictional Force on the Formation of Colloidal Particle Monolayer During Drying—Study Using Discrete Element Method—†

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Abstract

This paper presents the effect of frictional force between colloidal particles and a solid substrate on the formation of particle monolayer by a numerical simulation. Discrete Element Method is employed to simulate the dynamics of colloidal particles trapped in liquid film. Forces such as capillary immersion force, van der Waals force and frictional force are included in the simulation model. Isotropic ordering factor and non-dimensional boundary length are introduced to quantify the structures of colloidal particles. In the case where the diameter of colloidal particles ranges from 100nm to 1000nm, the monolayer structures depend strongly on the frictional constant between a particle and a solid substrate. On the other hand, in the case where diameter is about 10nm, large domains of hexagonal close-packed structures are formed because of the Brownian force.

Keywords: Particle Monolayer, Discrete Element Method, Substrate Friction

1. Introduction

In material nanotechnology, techniques for arranging nanoparticles/colloidal particles in a regular pattern are required. For example, a two-dimensional array (monolayer) involving nanoparticles/colloidal particles is used as a mask for lithography. Methods for forming a two-dimensional array include the Langmuir-Blodgett (LB) process, self-assembled monolayer (SAM) process, electrophoretic process and liquid membrane process. The liquid membrane process in which a colloidal dispersion solution is applied to and allowed to dry on a solid substrate is a promising processes. In a drying process within the liquid membrane process, when the liquid membrane is thinner than the particle diameter, a meniscus is formed between particles, thereby a long-distance attractive force known as a lateral capillary force occurs between particles. Mainly by this force, a particle group collects through a self-assembly phenomenon. Other than the lateral capillary force, various factors affect the arrangement structure of a particle group. Examples of these factors include the Brownian force, frictional drag from a substrate and the DLVO force. Clarifying the relation between various factors and resultant structures will make it possible to draw up a guideline for efficient preparation of material.

In order to form a colloidal particle membrane through a liquid membrane process, two-dimensional arrays consisting of polystyrene latex particles or protein particles have been produced as in the case of a series of studies by Nagayama et al. The correlation between process conditions and the structure of particles has been studied; how-

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ever, the various factors that affect the structure have not been studied comprehensively because experiments in which each of a plurality of processes is independently varied are difficult. Nevertheless, with the advance in computer simulations the dynamics of particles can now be directly tracked, and a multiplicity of processes can be studied while varying particular parameters independent of other parameters. We have simulated a colloidal particle arrangement process based on a liquid membrane process, considering the Brownian force, frictional drag from a substrate and the DLVO force by adopting a discrete element method (DEM). As a result, we have found that the actual arrangement structure quantitatively matches those experimentally obtained and have shown that this simulation technique is useful as a means of estimating the structure of particles. In the present study, we focus on the effect of friction from a substrate, which poses a significant effect in an actual system. We perform a series of calculations for particle diameters in a wider range of 10–1000nm and varied the decrease rate of liquid membrane thickness, and studied the correlation between the frictional drags from substrates, particle diameters and drying rates of solvents and the structures.

2. Simulation Technique

2.1 Motion equation for particles

In a system where a group of particles move within a plane, the motion equation used for individual particles in a translational motion is the Langevin’s equation that is used in nonequilibrium Brownian dynamics.\(^1\)

\[
m \frac{d\mathbf{v}_i}{dt} = \mathbf{F}_i - \mathbf{\xi}_i + \mathbf{F}_{\text{B}}(t)
\]

A rotational motion for individual particles can be expressed by the law of angular momentum.

\[
I \frac{d\mathbf{\omega}_i}{dt} = \mathbf{M}_i
\]

\(\mathbf{F}_i\) includes an inter-particle contact force, a lateral capillary force and a frictional drag from a substrate, each considered in the DEM. \(\mathbf{\xi}\) represents the Stoke’s drag coefficient. In our system, the particles are partially immersed in a solvent, and the proportion of immersed particles varies depending on the height of the liquid membrane. Therefore, we took the proportion of the horizontal projected area of the region of the particles immersed in the solvent to the whole area as \(s\), and then defined \(x\) using the following formula:

\[
\tilde{\zeta} = s \times 6\pi\alpha\eta
\]

For time quadrature of a motion equation, the predictor-corrector velocity Verlet method used in the molecular dynamics was employed. This scheme features the 2nd order accuracy for time steps, and can be applied when the force acting on particles, determined by calculating the predicted velocity, is a function of position-velocity. Thus, this method is suitable for our model that incorporates a drag from a solvent and a frictional drag from a substrate.

2.2 Forces acting on particles

2.2.1 Brownian oscillating force

When in a liquid membrane, particles are in contact with a solvent, and are therefore subjected to the Brownian force that is derived from the thermal agitation of the solvent. The Brownian force is obtained as a probable random force in accordance with normal distribution. When the entire surface of individual particles is fully immersed in a solvent, the Brownian force of particles moving in a two-dimensional plane can be expressed by the formulas below:

\[
\langle F^n(t) \rangle = 0
\]

\[
\langle F^n(t) \cdot F^n(t) \rangle = \frac{24\pi\alpha\eta K_BT}{\Delta t}
\]

In a manner identical to that for the Stoke’s drag, the effective Brownian force was defined, assuming that particles were partially immersed in a solvent, by using the following formula:

\[
F_{\text{B}}^n(t) = s \times F^n(t)
\]

2.2.2 Inter-particle contact force

When particles come into contact with each other, viscoelasticity occurs between particles. With DEM, this force is simulated with the Voigt model, which consists of a spring for representing an elasticity term and a dashpot for representing a viscosity term, wherein the spring and dashpot are situated in parallel. Incidentally, each contact force is determined after division in the normal direction and the tangential direction, wherein a friction slider is inserted in the tangential direction to represent the frictional drag between particles.

2.2.3 Lateral capillary force

The lateral capillary force working between two particles in a liquid membrane can be, if these particles are colloidal particles, approximated with the following formula\(^2\) (Fig. 1).
2.2.4 Frictional drag from substrate

When the vertical component of surface tension is not 0, particles receive static friction and dynamic friction from a substrate. A DLVO force is present between a substrate and particles. For convenience, we supposed that a substrate was inactive to particles. Also, we assumed that the maximum static frictional coefficient and the maximum dynamic frictional coefficient take the same value, and that the frictional drags are governed by the classic Coulomb frictional drag. A static frictional drag is represented by the following formula:

\[ F^s = -F_0 \mu_p Z \]

(\( F_0 \) is the maximum static frictional drag, \( F_0 \) is the magnitude of the resultant of forces other than the frictional drags acting on particles, and \( \mu_p Z \) is the maximum static frictional coefficient.)

At the same time, a dynamic frictional drag is defined as:

\[ F^d = -\mu_p Z \frac{v_i}{|v_i|} \]

where, \( Z = 2\pi\gamma Q \)

When the magnitude of the resultant of forces other than the frictional drags acting on particles is not greater than the magnitude of maximum static frictional drag, the particles remain motionless.

2.2.5 Van der Waals force

Van der Waals force deriving from fluctuation in induced dipole of atoms occurs between particles. The magnitude of the van der Waals force working between particles is defined by the following formula:

\[ F_{vdw} = Aa \frac{12(L-2a)^2}{(L-2a)^2} \]

(\( Aa \) is the van der Waals force constant, \( L \) is the distance between particles, and \( 2a \) is the diameter of the particle.)

When particles are situated in a solvent, an electrostatic repelling force acts between the particles. In the present study, we supposed that the effect of frictional drag from a substrate could be more clearly defined through calculation with a purer system, and did not included the term for an electrostatic repelling force.

2.3 Simulation conditions

In the present study, we assumed monodispersed colloidal silica particles as colloidal particles and water as a solvent. The calculation cell was in the form of a square with one edge measuring 40D, while cyclic boundary conditions were set in two directions. The number of particles within the cell was adjusted...
to 1296. This setting coincided with a coverage ratio of 0.702. Table 1 summarizes the calculation conditions including the physical properties of the particles and solvents. The actual calculation was performed by converting various amounts into nondimensional values including representative values of particle diameter $\alpha$, solvent surface tension $\gamma$ and particle mass $m$.

In the initial state, the particles were arranged at random and their initial velocity was set to 0 so that they did not collide with each other, and the thickness of the liquid membrane was the same as the diameter of particles.

The calculation was repeated until the meniscus was cut off with every particle pair as the height of the liquid membrane decreased.

For the present study, the viscous drag from a fluid acting on particles was expressed using the Stokes drag model. In contrast with an actual system, an effective viscous drag is significantly large because of factors including the lubrication effect, which is important when particles are in closer vicinity. As a result, when the Stokes drag model is employed, the particle move velocity is overestimated. Incidentally, with the our system, the “proportion of particle movement velocity to liquid surface decrease velocity” governs the degree of aggregation of a particle group. Accordingly, we compensated for the overestimation in particle move velocity with an increased liquid surface decrease velocity of $1.0 \times 10^{-2}$ ms$^{-1}$ in order to maintain the above-mentioned proportion at a value obtained from the experiment and to quantitatively express the self-assembly process of particles during the drying process.

### 2.4 Analysis for arrangement structure

In order to evaluate particle arrangement structures, two indices, that is, an ordering factor and a nondimensional boundary length, were introduced.

#### 2.4.1 Ordering factor

Delaunay triangles are obtained by subjecting an arrangement structure to the Voronoi polygonal division, and by connecting particles with line segments that are vertical to the edges of Voronoi polygons. Then, the triangles formed by connecting the centers of all the particles involved are gathered, and polygons composed of these triangles are considered. If the surroundings of particles are most densely packed, the resultant polygons will be regular hexagons. Then, the proportion of the so-formed regular hexagons to all the particles is defined as an ordering factor.

We call this index the Isotropic Ordering Factor (IOF).

#### 2.4.2 Nondimensional boundary length

The characteristics of a particular arrangement structure that cannot be evaluated with an ordering factor can be evaluated using an index that is characterized by the boundary length of an aggregated body. To determine the boundary length of an aggregated body, the coordination numbers of the particles involved are utilized, wherein a coordination number is defined as “a number of other particles with which a given particle is in contact”. As to individual particles, the region not in contact with other particles corresponds with the boundary of the aggregated body. (A shorter boundary length means more progressed aggregation with a particle group.) A Nondimensional Boundary Length (NBL) is defined using the following formula:

$$ NBL = \frac{1}{6n} \sum_{\kappa} (6 - \kappa)N(\kappa) $$  \hspace{1cm} (14)

With formula (14), the solution is 1 with the initial state; 0.83 when all the particles take the form of dimer; and 0 when all the particles form the most densely packed structure.

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Table 1  Physical conditions for the simulations

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
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<tr>
<td>Young's modus of a particle</td>
<td>7.5×10^{10} (Nsm^{-2})</td>
</tr>
<tr>
<td>Poisson ratio of a particle</td>
<td>0.173</td>
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<tr>
<td>Viscosity of liquid, $\eta$</td>
<td>8.91×10^{-4} (Nsm^{-2})</td>
</tr>
<tr>
<td>Surface tension of liquid, $\gamma$</td>
<td>7.20×10^{-2} (Nm^{-1})</td>
</tr>
<tr>
<td>Evaporation speed of liquid</td>
<td>1.0×10^{-2} (ms^{-1})</td>
</tr>
<tr>
<td>Hamaker constant, $A$</td>
<td>8.3×10^{21} (J)</td>
</tr>
<tr>
<td>Contact angle between a particle and liquid</td>
<td>20 (degree)</td>
</tr>
<tr>
<td>Zeta potential</td>
<td>0 (mV)</td>
</tr>
</tbody>
</table>

*) When the solvent is water, the minimum thickness of the liquid membrane that can be stably present on a substrate is approximately 100 nm, and it is impossible to obtain a liquid membrane whose thickness measures several nm. For the present study, we assumed a solid substrate that had a high-energy surface to allow a several nm thick liquid membrane to stably exist.
3. Result and Discussion

3.1 Examples of arrangement process

Fig. 2 illustrates several snapshots for the arrangement process where $D=100\text{ nm}$ and $\mu=0.1$. ($t$ represents a nondimensional time, $lev$ a liquid membrane thickness and $V_{\text{max}}$ the absolute value of the maximum velocity of particles.)

Fig. 3 illustrates the distance-dependency of the magnitude of nondimensional forces associated with the snapshots in Fig. 2, and Fig. 4 shows plotting of the interrelation between the liquid membrane thickness and meniscus cut-off distance. Note that the Brownian forces in Fig. 3 are standard deviations of force.

Fig. 2(a) illustrates the initial state, where the particles appear to have been randomly distributed. When the liquid membrane thickness drops below the particle diameter, a meniscus occurs between particles, thereby a lateral capillary force acts between particles. At the same time, because the component perpendicular to the surface tension is no longer 0, the frictional drag from a substrate also acts on particles.

Fig. 2(b) is a snapshot for $h=1.500$. Several particles are in contact with adjacent particles, forming dimers while most particles remain apart from other particles. From Fig. 3(b), the lateral capillary force appears to be dominant; however, because the screening effect is introduced, the effective lateral capillary force is smaller than $1/10$ the two-body force. Consequently, the resultant force consisting of the lateral capillary force and the Brownian oscillating force is not greater than the frictional drag from a substrate,
and the particles cannot move. With certain particles, the resultant force consisting of the lateral capillary force and the Brownian force is greater than the frictional drag from a substrate, thereby these particles come into contact with adjacent particles. Then, these particles remain motionless as they remain in contact with other particles owing to the frictional drag from a substrate and the van der Waals force.

When the level of the liquid membrane drops to \( h=1.2 \) (Fig. 2 (c), (d)), a plurality of regions that lack particles occur (voids), and these voids grow rapidly.

In particular, certain particles around these voids may have the most densely packed structure. As the effect of the lateral capillary force is greater (Fig. 3(c), (d)), more particles gradually start moving; and at the same time, the size of the voids becomes greater. In particular, the effective lateral capillary force acting on the particles around the voids greatly increases, thereby aggregation of particle groups occurs. When the height of the liquid membrane decreases to about \( h=0.976 \), several domains and larger voids appear, and aggregation seems to be virtually completed. At this time, the Brownian force further decreases (Fig. 3(e)). On the other hand, when the meniscus cut-off distance drops below two times the particle diameter (Fig. 4), the meniscus between domains is cut off, leading to development of particle rearrangement in the domains. Typically by this effect, rearrangement takes place, leading to the final state (Fig. 2(f)). When Fig. 2(e) is compared with Fig. 2(f), clearer boundaries are found in Fig. 2(f). For a two-dimensionally arranged membrane formed by a liquid membrane process too, it seems that when the liquid membrane thickness decreases and the liquid membrane between domains is cut off, the grain boundaries develop.

### 3.2 Effect of frictional drag from substrate on the arrangement process

Fig. 5 illustrates a time-dependent change in NBL for \( D=100 \) nm and \( \mu_{ps}=0.0-1.0 \). If there is no frictional drag from a substrate (\( \mu_{ps}=0.0 \)), and when \( 1.4<h<2.0 \), the NBL simply decreases. This seems to be because the particle groups come into contact with each other owing mainly to the lateral capillary force, and once the particle groups are in contact with each other, the van der Waals force acts between the particles, thereby the particles do not come off from other particles. Then, domains consisting of the most densely packed particle groups are formed, and aggregation does not progress further; as a result, the value of NBL remains unchanged. However, the NBL again decreases when \( 0.8<h<1.0 \). This seems to be because, as mentioned in Sec. 3.1, a meniscus between domains is cut off, leading to development of particle rearrangement in the domains.

If a substrate provides drag and when \( 1.9<h<2.0 \), the NBL temporarily decreases, and then remains at a particular value. This is because the effective lateral capillary force does not exceed the frictional drag from a substrate, and, as a result, the particles remain motionless. Then, when the thickness of the liquid membrane decreases and the effective lateral capillary force exceeds the frictional drag from a substrate, aggregation proceeds. However, with a greater \( \mu_{ps} \), the start of domain formation delays. In other words, the meniscus cut-off distance is shorter and
the time elapsed before completion of drying is also shorter, the NBL in the final state is much greater.

Fig. 6 illustrates time-dependent change in the NBL for D=10 nm and μps=0.0–1.0. Regardless of whether or not the frictional drag from a substrate is present, the NBL drops to about 0.4 when 1.9<h<2.0. This is because many particles are in contact with each other due to the Brownian force to form a network structure. At this time point, however, there are few domains that are most densely packed. If μps=0.0, 0.1 or 0.25, and when 1.2<h<1.5 and the effective lateral capillary force is greater, the formation of denser domains begins. Consequently, the NBL again decreases, and drops to about 0.10. On the other hand, if μps=0.5 or 1.0, and when h<1.4, progress of aggregation is slower. Nevertheless, when compared with the case where D=100 nm, and when μps is the same, the case where D=10 nm exhibits an apparently smaller NBL in the final state as well as the formation of domains.

3.3 Effect of frictional drag from the substrate and particle diameter on the arrangement structure

Fig. 7 shows snapshots for the terminal state for D=10 or 100 nm, μps=0.0, 0.25 or 0.5. For μps=0.0 (Fig. 7(a)(i), (b)(i)), no significant difference is found between the case with D=100 nm and the case with D=10 nm, and there are a plurality of domains and voids with both cases.

When D=100 nm, the region of domains is smaller with greater μps (Fig. 7(a)(ii), (iii)). With greater μps, the height of the liquid membrane, which is mainly governed by the lateral capillary force, is smaller. More specifically, the meniscus cut-off distance is smaller and the time span where aggregation develops is shorter; as a result, drying ends though the formation of domains does not fully take place.

When D=10 nm and even though μps=0.25, larger domains exist (Fig. 7(b)(ii)). At μps=0.5, the domains are smaller (Fig. 7(b)(iii)) but are still larger compared with the case for D=100 nm. This is because the Brownian force exceeds the frictional drag from a substrate, and, as a result, the particles move at random, promoting aggregation.

Consequently, although a frictional drag from a substrate is present, greater domains are formed.

Fig. 8 illustrates the dependency of IOF on a substrate frictional drag coefficient in the final state for D=10–1000 nm and μps=0.0–1.0. If D=50 nm, the IOF value decreases with a greater μps and becomes closer to 0. Incidentally, if D=10 nm, and when 0.0<μps<0.25, the IOF value is greater than 0.35, and larger domains are formed. When μps>0.5, the IOF value is smaller; however, the IOF value for a same μps is still significantly large. In other words, the motion of particles is inhibited by a frictional drag from the substrate; when D<25 nm, the effect of the Brownian force is significantly exhibited, and the ordering factor increases. In our study, the effect of the Brownian force was apparent only in the case of D=25 nm. It may be possible to determine conditions that allow...
the formation of greater domains in the cases where the particle diameter is 25 nm or greater, by searching for the conditions that allow the Brownian force to work effectively.

3.4 Effect of frictional drag from the substrate and the liquid membrane thickness decrease speed on the arrangement structure

Fig. 9 illustrates the time-dependent change in the NBL for the liquid membrane thickness decrease rates of 1.0, 2.0 and 5.0 $\times 10^{-3}$ ms$^{-1}$ when $D=100$ nm and $\mu_p=0.1$. With a lower liquid membrane thickness decrease rate and when 1.9<$h<$2.0, the degree of decrease in NBL is greater. This is because more particles come into contact with other particles due to a longer time in which the Brownian force can work efficiently. Then, the NBL remains at a constant value, and again decreases when 1.3<$h<$1.4. With the same $h$, a lower liquid membrane thickness decrease rate leads to a lower NBL value.

Fig. 10 shows the IOF values of the arrangement structures obtained from $D=100$ nm, $\mu_p=0.1$ and 1.0 and $V_{\text{evap}}=1.0 \times 10^{-3}$, $1.0 \times 10^{-2}$ m s$^{-1}$.

3.5 The coefficient of friction between the substrate and particles in the experiment

In the present study, we examined the effect of friction from a substrate on arrangement structures of particles. The coefficient of friction between colloidal particles in liquid and a substrate as well as the mechanism of friction between colloidal particles in liquid and a substrate are unknown. One example of results from studies in this field is a report where frictional drags between latex particles (particle diameter 100 or 300 nm) and a mica substrate in water were measured by LFM (Lateral Force Microscopy)$^{10}$. According to this report, if a frictional drag is compatible with Coulomb’s law, the frictional coefficient should be in the order of 1. The reason why the frictional coefficient takes such a value, although a mica substrate is very smooth in the atomic order, is unknown. And, it is likely that with other systems too (in particular, systems with rough-surface substrates), the coefficient of friction between particles and a substrate is not 0. Therefore, we cannot ignore the effect of the substrate involved.

4. Conclusion

We have modeled an arrangement process for colloidal particles with a liquid membrane process, and have studied the effect of frictional drag from a substrate on arrangement structure of particle groups. When a particle diameter is 100 nm or greater, the frictional drag from a substrate inhibits movement of the particles, thereby the coefficient of friction between a substrate and particles significantly affects the arrangement structure. On the other hand, when a particle diameter is about 10 nm, the Brownian force promotes particle movement, thereby densely packed domains are obtained even though the frictional drag from a substrate is present. Even if the ordering factor is otherwise small, the drying rate may be lowered so that the forces including a lateral capillary force are allowed to act on particles for a
sufficiently long time so as to improve the ordering factor.

Acknowledgement

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Nomenclature

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
<th>Unit</th>
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<tbody>
<tr>
<td>( A )</td>
<td>A maker constant of a particle</td>
<td>[J]</td>
</tr>
<tr>
<td>( a )</td>
<td>radius of a particle</td>
<td>[m]</td>
</tr>
<tr>
<td>( D )</td>
<td>diameter of a particle</td>
<td>[m]</td>
</tr>
<tr>
<td>( I )</td>
<td>inertial moment of a particle</td>
<td>[kg \cdot m²]</td>
</tr>
<tr>
<td>( F_b )</td>
<td>Brownian force acting on a particle</td>
<td>[N]</td>
</tr>
<tr>
<td>( F_{\text{eff}} )</td>
<td>effective Brownian force acting on a particle</td>
<td>[N]</td>
</tr>
<tr>
<td>( F_{\text{ext}} )</td>
<td>external force acting on a particle</td>
<td>[N]</td>
</tr>
<tr>
<td>( F_c )</td>
<td>capillary force acting between two particles</td>
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</tr>
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<td>( F_f )</td>
<td>frictional force from a solid substrate</td>
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</tr>
<tr>
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</tr>
<tr>
<td>( m )</td>
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<td>rotational moment of a particle</td>
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<tr>
<td>( n )</td>
<td>number of particles</td>
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<td>( N(k) )</td>
<td>number of particles, the coordinate number of</td>
<td>[\text{--}]</td>
</tr>
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<td>( Q )</td>
<td>capillary charge</td>
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<td>( r_c )</td>
<td>radius of contact circle of three-phase interface</td>
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<tr>
<td>( s )</td>
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<td>frictional Constant between a particle and a</td>
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<td>velocity of a particle</td>
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<td>( \zeta )</td>
<td>coefficient of Stokes drag force</td>
<td>[kg \cdot s⁻¹]</td>
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References


Author's short biography

Hiroyuki Nishikawa

Hiroyuki Nishikawa obtained his Master Degree (2001) in Fundamental Chemistry at Chiba University and his Doctor Degree (2006) in Chemical System Engineering at University of Tokyo. He is now working for Mizuho Information & Research Institute, Inc. as a consultant at Engineering Service Department.

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B.Eng. University of Tokyo in Precision Engineering 1993

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Department of Chemical System Engineering, University of Tokyo

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