Simultaneous Measurement of the Migration Velocity and Adsorption Force of Micro-Particles Using an Electromagnetophoretic Force under a High Magnetic Field

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A simultaneous measurement technique for determining the migration velocity of a micrometer-sized particle in a capillary and the adsorption force to the inner surface of the capillary has been proposed. This technique is based on an electromagnetophoretic force being exerted on a micro-particle in an electrolyte solution, which is governed mainly by the electromagnetic buoyancy, when a homogeneous magnetic field is applied at a right angle to the electric current through the medium. By the electromagnetic buoyancy, micro-particles such as polystyrene, carbon and yeast were migrated perpendicular to the direction of the electric current and reached a fused-silica wall. A switching of the current direction could desorb the particle from the wall, and allowed to calculate the detaching force from the desorbing current. The migration velocity normalized to the size in the magnetic field of 10 T was increased in the order of yeast, carbon and polystyrene, while reflecting the decreasing order of the apparent conductivity of the particles. The desorption force could be measured up to 1 nN with a sensitivity of pN. The observed interaction forces of polystyrene and carbon were in the range of 250 – 600 pN with large deviations.

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Introduction

For the characterization and separation of colloidal particles, including industrial particles, environmental particles and living cells, a development of a new measurement technique for the physical and chemical properties of particles is highly required in various fields. We have developed new techniques for the specific migration of micro-particles in a liquid medium using a laser beam, an alternative electric field and a magnetic field so far. To characterize the particle surface, a force measurement between a particle and a probe or a substrate surface is a promising approach. Theoretical description of these forces has been widely studied by DLVO theory, and some measurement techniques have been developed.^{1,2} A popular method for directly measuring the interaction force between a micrometer-sized particle and a surface is atomic force microscopy (AFM). In AFM, the interaction force is measured as a function of the particle-surface separation distance by approaching the particle on a cantilever toward the surface. As an alternative method, the interaction force between the cell-cell and the cell-surface has been estimated by cell detachment using a flush shear stress.^{3,4} We focused on an electromagnetic force as a means to control the particle detachment force, which was known as the Lorentz force generated by a homogeneous magnetic field applied at a right angle to the electric current.

The phenomena of electromagnetophoresis (EMP) was studied for the first time by Kolin in 1953.⁵ The general concept of electromagnetophoresis is an application of the Lorentz force to migration analysis. When we apply an electric

current through a conductive fluid including particles in a homogeneous magnetic field perpendicular to the current, a force is exerted on both the fluid and the particles. If the force exerted on the fluid is not equal to that working on the particles, a migration of the particles is caused.

Recently, we developed the electromagnetophoresis of micrometer-sized particles for the migration and characterization of particles.^{6,7} In our previous study using small permanent magnets, we demonstrated that the electromagnetophoretic migration of micro-particles, such as polystyrene and carbon, was predominated by the electromagnetic buoyancy, and the migration velocity was proportional to the electric current, the magnetic flux density and the second power of the radius of the spherical particles.⁵ In the present work, we proposed a simultaneous measurement technique of the migration velocity and the particle-wall interaction force by applying the electromagnetic buoyancy to particles controlled by the current under a high magnetic field of 10 T. The electromagnetic buoyancy was controlled in the range of 10^{-12} – 10^{-9} N in a current of up to 1 mA, as we applied 10 T by a super-conducting magnet.

In this paper, we describe the experimental method used to measure the migration velocity and the desorption force of microscopic particles, such as polystyrene latex, carbon and yeast, which were migrated and adsorbed on the fused-silica capillary wall using the electromagnetic buoyancy. This work has resulted in promising a new principle for measuring the interaction force between a particle and a surface, which is applicable in the field of bio-analytical chemistry.

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Fig. 1 Schematic diagram of the electromagnetophoresis cell: (a) PTFE tube, (b) heat-shrink tube, (c) Ag|AgCl electrodes, (d) fused-silica capillary, and (e) objective. A PTFE tube was connected to a syringe pump. The observation part was the center of the capillary. A homogeneous magnetic field (B) was applied perpendicular to the capillary, and the direction of the current (i) could be alternatively changed.

Fig. 2 Experimental setup for the observation of electromagnetophoresis and the adsorption-desorption of particles: (a) a superconducting magnet, (b) a voltmeter, (c) a current source, (d) an ammeter, (e) a video recorder, (f) a monitor, (g) a syringe pump, (h) a light source, (i) a CCD camera, and (j) an optical microscope.

Experimental

Materials

The particles used in the experiments were polystyrene latex and carbon. Polystyrene latex particles were purchased from Funakoshi (Tokyo, Japan), and carbon particles were purchased from Nippon Carbon (Tokyo, Japan). The diameters of the polystyrene latex particles were $10\ \mu\text{m}$ ($9.14 \pm 0.71\ \mu\text{m}$), $15\ \mu\text{m}$ ($14.6 \pm 1.7\ \mu\text{m}$) and $20\ \mu\text{m}$ ($21.0 \pm 2.7\ \mu\text{m}$), and the diameters of the carbon particles were $20\ \mu\text{m}$ ($22.0 \pm 2.0\ \mu\text{m}$) and $30\ \mu\text{m}$ (29.5 ± 3.1). The electrolyte solutions were $1.0\ \text{M}$ KCl with pH 5.4 (conductivity $0.112\ \text{S cm}^{-1}$) for polystyrene latex particles and $6.0\ \text{M}$ CaCl_2 with pH 6.6 (conductivity $0.110\ \text{S cm}^{-1}$) for carbon particles. These solutions were prepared to have an iso-density with the particles. Polystyrene was an insulator and the carbon, itself, had a higher conductivity than that of the medium. Further, we used common baker's yeasts dispersed in a $1.0\ \text{M}$ KCl solution as an example of biological particles.

Apparatus and measurements

The migration and adsorption-desorption behavior of particles

Fig. 3 Schematic illustration of the migration and adsorption-desorption of a particle by electromagnetic buoyancy. By applying a current, a particle which was adsorbed on the capillary wall was desorbed, and was adsorbed to an opposite side of the capillary wall. Desorption-adsorption was repeated by switching the direction of the current.

were observed in a capillary cell. Figure 1 shows the capillary cell used in the present experiments. The cell was made of fused-silica capillary, which had a $100 \times 100\ \mu\text{m}$ inner section and was $2\ \text{cm}$ long, inserted to PTFE tubes with Ag|AgCl electrodes covered by heat-shrink tubes. The behavior of the particles was observed by an objective lens positioned upward. The focal point was set at the center of the capillary.

Figure 2 shows the apparatus used in the experiments. A magnetic field was generated by a superconducting magnet (JMT, JMTD-10T100HH1, Japan). The cell was set on a holder in the middle of the bore ($10\ \text{cm}$ in a diameter) of the magnet. Sample solutions were provided to the capillary cell by a syringe pump. A galvanostat (Fusoh, HECS317, Japan) device or a potentiostat (Metronix, HSV1K-60, USA) device was used to provide a current. The current was measured by an ammeter (ADVANTEST, Digitalmultimeter R6551, Japan). A microscope attached to a CCD camera (ELMO, ME421R, Japan) was used to observe the behavior of the particles. The CCD image was displayed on a monitor, and was also recorded on a videocassette. The migration velocity was measured from images captured in a computer. The conductivities of the medium were measured by a conductivity meter (TOA DKK, CM-40V, Japan).

In the present experiments, the adsorption force between particles and a fused-silica surface was measured as well as the migration velocity. Figure 3 shows a schematic illustrating the adsorption-desorption behavior of a particle by electromagnetic buoyancy. When a particle appeared in the observation region by the flow, the flow was stopped. At first, the particle was migrated and adsorbed at the inner surface of the fused-silica capillary by applying a current of $200\ \mu\text{A}$. The adsorbed particle was desorbed by a reversed electromagnetic buoyancy by switching the direction of the current, and the particle migrated and adsorbed at an opposite side of the capillary wall. The current was increased from $0\ \mu\text{A}$ to $1000\ \mu\text{A}$ under a fixed magnetic field of $10\ \text{T}$, and the current required for the desorption of particles was measured. Also, the particle was adsorbed again by applying a current of $200\ \mu\text{A}$ for scores of seconds. The desorption and adsorption of the particle were repeated by switching the direction of the current. The desorption current was measured scores of times every for

particle and for several particles by repeating these adsorption-desorption cycles. We then calculated the adsorption force from the desorption current. The applied current was in the range of less than 1 mA to prevent to the generation of Joule heat and the influence of electrophoresis.

All measurements were carried out in a thermostated room at $25 \pm 1^\circ\text{C}$.

Results and Discussion

Electromagnetophoretic velocity

The experimental observations were analyzed by considering the force acting on the particles and the medium. Under the situation of a homogeneous magnetic field and a current applied perpendicular to the magnetic field, a particle in an electrolyte solution in a capillary experiences two types of forces: one is an electromagnetic force working on the particle, itself, and the other is an electromagnetic buoyancy, which has the same magnitude to the electromagnetic force acting on the medium with the same volume to the particle and which is in an opposite direction to the electromagnetic force. Therefore, the total force working on a particle (F_{EMP}) can be expressed as

$$F_{\text{EMP}} = 2 \left(\frac{\sigma_p - \sigma_f}{2\sigma_f + \sigma_p} \right) \frac{iBV}{S}, \quad (1)$$

where B is the magnetic flux density ($\text{N A}^{-1} \text{m}^{-1}$), i is the electric current (A), V is the volume of the spherical particle (m^3) and S is the inner section of the cell (m^2), σ_p is the apparent electric conductivity (S m^{-1}) of the particle and σ_f is the electric conductivity (S m^{-1}) of the medium. Equation (1) indicates that the total force (F_{EMP}) will be proportional to B , i and V . The apparent conductivity included the surface conductivity of the particles. The electric charges on the surface of particles produce an electric double layer.⁸ The effect of a counter-ion cloud around the particles causes the surface conductivity on the particles. Therefore, the apparent conductivity (σ_p) depends on the property of the particle. The acceleration and mass of the particles are so small in our system that the particle migrate with a constant velocity (v) keeping balance between the electromagnetophoretic force (F_{EMP}) and the viscous or frictional force (F_v), which is expressed by Stokes' law,

$$F_f = 6\pi\eta r v C_w^2, \quad (2)$$

where η is the fluid viscosity (Pa s) and r the radius of the spherical particle (m). C_w is the viscous drag coefficient due to the surface of the capillary wall, as shown in

$$C_w = \frac{1}{\left[1 - \frac{9}{16} \left(\frac{r}{h} \right) + \frac{1}{8} \left(\frac{r}{h} \right)^3 - \frac{45}{256} \left(\frac{r}{h} \right)^4 + \frac{1}{16} \left(\frac{r}{h} \right)^5 \right]}, \quad (3)$$

where h is the distance (m) between the center of the particle and the surface of the wall.⁹

From Eqs. (1) and (2), we can obtain the migration velocity v of a spherical particle as

$$v = \frac{4}{9} \left(\frac{\sigma_p - \sigma_f}{2\sigma_f + \sigma_p} \right) \frac{iBr^2}{S\eta C_w^2}. \quad (4)$$

This equation can be used to determine the apparent conductivity of the particle from the experimentally observed

Fig. 4 Electromagnetophoretic velocity of yeast at various applied currents. The magnitude of the homogeneous magnetic field was 10 T. A 1.0 M KCl solution was used as a conductive medium. The conductivity of the medium was 0.117 S cm^{-1} . The diameter of yeasts determined from CCD images was $7.17 \mu\text{m}$.

migration velocity. The apparent conductivities were determined as 0.0287 S cm^{-1} and 0.0947 S cm^{-1} for polystyrene and carbon, respectively.⁷

Figure 4 shows the dependency of migration velocity of yeast on the current under a fixed magnetic field of 10 T. The current dependency of the migration velocity of yeasts was similar to that of polystyrene particles and carbon particles, as suggested by Eq. (4). The form of the yeast was verified to be spherical by microscopic images, and the diameter of the yeast was determined to be $7.17 \pm 1.49 \mu\text{m}$ from images captured in a computer. The apparent conductivity of the yeast was calculated to be 0.0746 S cm^{-1} from the slope of Fig. 4 and Eq. (4).

Desorption force of particles

The electromagnetophoretic migration of particles in a medium is predominated by the electromagnetic buoyancy. The electromagnetic buoyancy could desorb the particles adsorbed on the wall of the capillary when the current was applied at a fixed magnetic field of 10 T. From the current at the time when the particle was just desorbed, the adsorption force could be calculated by Eq. (1).

Polystyrene particles with diameters of $20 \mu\text{m}$ and $15 \mu\text{m}$ and the carbon particles were desorbed by the electromagnetic buoyancy generated under the present conditions. However, the desorption of the smallest polystyrene particles with $10 \mu\text{m}$ diameter was difficult under a current of less than 1 mA. Because the magnitude of the electromagnetic buoyancy was proportional to V , a weaker force was generated in a smaller particle at a fixed current. Therefore, $10 \mu\text{m}$ polystyrene was not enough to be desorbed in a current of less than 1 mA.

Figure 5 shows a histogram of the desorbed numbers of polystyrene particles of $15 \mu\text{m}$ and $20 \mu\text{m}$ diameters from the silica-capillary wall, when the current was increased with every $10 \mu\text{A}$. The total event numbers were 128 and 160 for $15 \mu\text{m}$ and $20 \mu\text{m}$ polystyrene particles, respectively. Because the total numbers were different, the desorption number (N) was normalized divided by the maximum desorption number (N_{max}). The desorption current of polystyrene particles of $15 \mu\text{m}$ diameter was larger than that of $20 \mu\text{m}$ particles. The average values of desorption currents were $397 \mu\text{A}$ and $124 \mu\text{A}$ for 15

Fig. 5 Histogram of desorption event of polystyrene particles with diameters of (a) 15 μm and (b) 20 μm for the every increment of 10 μA . Total desorption events were 127 and 160 for 15 μm and 20 μm polystyrene particles, respectively.

Fig. 6 Histogram of desorption events of carbon particles with diameters of (a) 20 μm and (b) 30 μm for the every increment of 10 μA . The total desorption events were 187 and 107 for 20 μm and 30 μm carbons, respectively.

μm and 20 μm polystyrene particles, respectively.

Figure 6 shows a histogram for the desorption of carbon particles, whose diameter were 20 μm and 30 μm for the current increment of every 10 μA . The total desorption numbers were 187 and 107 for 20 μm and 30 μm of polystyrene, respectively. The desorption current of carbon particles with 20 μm diameter was larger than that of particles with 30 μm diameter, as in the case of polystyrene. The average values of the desorption current were 309 μA and 192 μA for carbon particles with diameters of 20 μm and 30 μm , respectively. It was found that the desorption current of 20 μm carbon particles was larger than that of polystyrene particles, whose diameter was equal to that of carbon.

Figure 7 shows the desorption forces of polystyrene particles and carbon particles from the capillary wall, calculated from Eq. (1) using i_D determined from Figs. 5 and 6. The adsorption force of polystyrene particles was larger than that of carbon particles. Furthermore, the adsorption force showed a tendency that the larger particle adsorbed slightly stronger for both polystyrene and carbon. The average values of the adsorption

Fig. 7 Adsorption forces of polystyrene particles and carbon particles which were calculated from Eq. (1) using the results shown in Figs. (3) and (4). The filled circles indicate polystyrene particles and the open circles indicate carbon particles.

forces of polystyrene particles with 15 μm and 20 μm diameter were 420 pN and 468 pN, respectively. If the adsorption force of polystyrene of 10 μm is about 400 pN from the results for 15 μm and 20 μm polystyrenes, the current required for the desorption would be 1200 μA , which is out of the current region in the present experiment.

Next, we tried to desorb yeasts adsorbed on the capillary wall. However, we could not desorb yeasts under the present experimental conditions. Because the size of the yeast was as small as 7.17 μm and the apparent conductivity of the yeast was as large as 0.0746 S cm^{-1} , the electromagnetophoretic force was not enough to desorb yeasts in as current of less than 1 mA. The electromagnetophoretic force, F_{EMP} , calculated from Eq. (1) at a current of 1 mA was only 53.6 pN. When the particle's size and/or the difference between the conductivities of the particle and the medium is so small, the electromagnetophoretic force required for the desorption of particles is not generated.

The adsorption or desorption forces measured in this study had a large deviation. One of the reasons could be the roughness or inhomogeneity of both surfaces. This means the interaction between the surfaces of the particle and the wall is not the same in every adsorption event.

The dependence of the adsorption force on the nature and size of particles could be mainly governed by their van der Waals interaction.¹⁰ Therefore, this technique will provide a new insight into the interaction force and some specific properties of the surface of particles, especially for biological cells.

Conclusion

We proposed a new technique for measuring the migration velocity and desorption or adsorption forces of micro-particles in liquids in a closed cell by using an electromagnetophoretic force. The uniqueness of this method is a non-contact method with a high sensitivity of pN, with more simplicity, versatility and sensitivity than AFM. We demonstrated for the first time by this technique that the adsorption force between particles, such as polystyrene, carbon and yeast, and the inner surface of a capillary could be measured in a magnetic field of 10 T. The adsorption-desorption cycle of particles could be controlled mainly by the electromagnetic buoyancy changing the magnitude of the current. This new technique is highly

promising for analyzing the surface properties of biological cells. Various chemical modification of the capillary wall will provide useful information on the chemical composition and its functions. Furthermore, this method suggested the possibility to separate particles in a capillary by the difference in their adsorption force. The repeated adsorption-desorption of particles traveling in a capillary will result in a new concept of micro-particle chromatography.

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