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Separation of a mixture of particles into its individual components with the aid of the magneto-Archimedes separation

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ABSTRACT

The magneto-Archimedes separation allows for separating mixtures of feeble magnetic materials into its components based on the difference of their densities and magnetic susceptibilities. So far, this technique was applied for the separation of relatively large particles of several millimeters in diameter. Here we apply this technique experimentally to the simultaneous quantitative analysis of multiple micrometer-sized particles in a fluid. It was confirmed that the magneto-Archimedes separation can be applied for the separation of mixture of microspheres larger than 20 μm. Further high performance separation efficiency is expected with the optimization of separation conditions including the control of the spatial distribution of the magnetic field.

1. Introduction

Magnetism is a property that all materials have. By applying a high magnetic flux density of more than 10 T, dynamic effects can be imparted even on diamagnetic or paramagnetic materials (feeble magnetic materials). Therefore, the behavior of all materials can be controlled without any direct contact utilizing high magnetic fields. The intensity of the magnetic force acting on feeble magnetic materials becomes large under high gradients of the magnetic field intensity. It is known that diamagnetic materials such as water can be levitated under such conditions called the diamagnetic levitation [1]. In addition, paramagnetic materials can be levitated if the differences of magnetic susceptibilities and densities of objects' materials and the surrounding medium are controlled by selecting surrounding media properly [2]. This method is called as the magneto-Archimedes levitation technique. When a mixture of different materials levitates in the field, the separation of materials can be attained because different substances have different equilibrium levitation positions under a certain magnetic field distribution due to their different materials' properties.

The magnetic ore concentration using magnetic fluids as the surrounding medium is known as a way of separation that based on the levitation of materials in fluid [3–5]. This will require a much lower intensity of magnetic field because of the superparamagnetic nature of magnetic fluids. However, the separation is attained based only on the difference of densities. Furthermore, it seems not suitable for the separation and analysis of biological materials.

So far, the principle of the magneto-Archimedes separation was provided and then, some demonstrations were carried out using relatively large particles of millimeters in size. Smaller micrometer-sized particles are affected heavily by the thermal energy, although it should nevertheless be possible to apply the magneto-Archimedes separation principle. This would allow to replace quantitative batch-run medical analyses (e.g., tests based on the antigen-antibody reaction) concurrently for multiple components. In this study, we experimentally evaluated whether the magneto-Archimedes separation technique can be applied on the separation of micrometer-sized particles in a fluid.

2. Magnetic levitation and magneto-Archimedes separation

To apply high magnetic field of more than 10 T, a superconducting magnet is generally used. The inside of the winding core, the bore, of the superconducting coil is used as the experimental space. The magnetic field in the bore is spatially distributed. Fig. 1(a) shows the spatial distribution of the magnetic flux density in the superconducting magnet bore used in this study, model JMTD13C100 manufactured by JASTEC Inc., as an example. This magnet has a room temperature bore of 100 mm in

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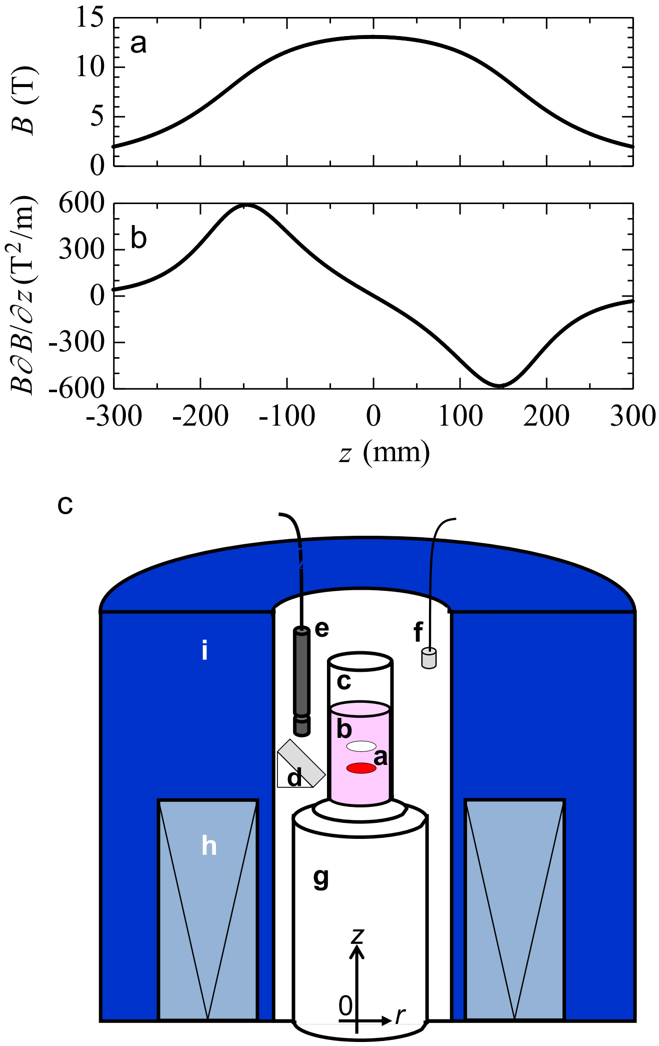

Fig. 1. Distributions of the magnetic flux density (a), the product of the flux density and its gradient (b) along the bore axis of the superconducting magnet used in this study and a schematic figure of the experimental set up (c), where a: samples, b: surrounding media, c: sample container, d: mirror, e: CCD camera, f: LED, g: stage, h: superconducting coil, i: superconducting magnet. The bottom of sample container was fixed at z=140 mm.

diameter and can generate up to 13 T along the bore axis. The horizontal axis in Fig. 1, z, is the position on the bore axis. The magnetic field is maximized at the center of the coil and gradually decreases towards both ends of the bore. Fig. 1(b) shows the product of the magnetic flux density and its gradient that is related to the intensity of the magnetic force acting on materials, along the bore axis. The maximum magnetic force is acting on materials at the off-centered position where gradients are high.

As described above, the levitation of diamagnetic materials can be obtained by exerting large upward magnetic force in vertical direction with applying high gradient magnetic fields. Diamagnetic levitation such as water was reported by Beaugnon and Tournier [1]. In the magneto-Archimedes levitation that takes into account the magnetic contribution of surrounding medium, the condition to attain the levitation becomes easier and even paramagnetic materials can be levitated [2]. By considering the contribution of the effect of surroundings, the condition for magneto-Archimedes levitation is expressed as follows,

$$-\rho_1 g + \frac{\chi_1}{\mu_0} B\frac{\partial B}{\partial z} + \rho_2 g - \frac{\chi_2}{\mu_0} B\frac{\partial B}{\partial z} = 0 \quad (1)$$

where ρ_1 and χ_1 are the density and susceptibility of the levitating substance, respectively and ρ_2 and χ_2 are those of the medium around it, g is the acceleration of gravity, μ_0 is the permeability of vacuum, B is the flux density, and z is the position along bore axis.

Since the magnetic susceptibility of diamagnetic materials is very small as 10^{-5} and negative, high and steep gradient magnetic fields are required to attain the magnetic levitation. Even in the case of water, which can be levitated more easily due to its relatively low density and large diamagnetic susceptibility, the value of$B\cdot\partial B/\partial z$required for levitation is 1400 T^2/m, while typical superconducting magnets with a flux density of 10 T and a room temperature bore of 100 mm can commonly achieve only about 500 T^2/m. This value is approximately proportional to the square of B. As a result, diamagnetic levitation requires a very high field magnet. On the other hand, the magnetic field condition required for the levitation can be decreased by considering the magnetic contribution from surrounding media. For example, by using pressurized oxygen gas of 1 MPa, water can be levitated by ordinal superconducting magnet with a flux density of 10 T [2]. When materials levitate by the magnetic force, there is a stable levitation position. Even if the materials displaced from their stable levitation position, a restoring force will arise and return the material to the original position. Stable levitation position in magneto-Archimedes levitation is determined by the difference in volume magnetic susceptibilities and densities between the material and its surroundings. Therefore, different substances levitated in the field have different equilibrium positions. Even if, some materials levitated accidentally in the same position under certain conditions, it is possible to change the stable point by changing the property of the surrounding medium. So, it seems to be a potential way of separation. Indeed, several trials have been made to apply this separation technique to practical separation processes of feeble magnetic materials [6–8].

Microspheres that have antigens on their surfaces with magnetic or fluorescent materials are often used for medical analyses such as immunodiagnosis. To evaluate several different antibodies using such system, batch manner analysis is sometimes applied which requires a long time to analyze many antibodies. If different kind of microspheres with different antigens on their surfaces are introduced into the test sample and then, carried out the magneto-Archimedes separation, different microspheres are expected to levitate into different positions. Each component in the test sample can be analyzed quickly with optical methods in levitated condition or by analysis after collection from the fluid. Such a method might thus be a potential way to analyze multicomponent objects included in a fluid, simultaneously. To test the possibility of magneto-Archimedes separation as a way of simultaneous multicomponent analysis, the separation of a mixture of different microspheres was carried out in this study.

3. Experimental

The schematic of the experimental set up is illustrated in Fig. 1(c). Four kinds of glass microspheres listed in Table 1 were used. The soda-lime glass spheres, Model UB-23L made by Union Co., and the borosilicate glass spheres, UB-23MF made by Union Co., were particles of 50 µm size in diameter. For 20 µm size particles, two kinds of silica spheres, Sicastar produced by micromod, were used. To distinguish the difference of them easily when they were separated, two different colored silica particles, red, Sicastar-red (40-00-204), and white, Sicastar (43-00-204), were chosen. Densities and magnetic susceptibilities of particles were measured with the micromeritics Accupyc 1330 pycnometer and Quantum design MPMS XL magnetometer, respectively, and given in Table 1. Table 1 also contains densities and magnetic susceptibilities of

Table 1
Microspheres and surrounding media used in this study.

Material	Diameter (μm)	Density (kg/m^3)	Magnetic susceptibility (−)
Soda-lime glass	50	2.80×10^3	-4.54×10^{-6}
Borosilicate glass	50	2.63×10^3	-6.20×10^{-6}
Sicastar red 40-00-204	20	2.31×10^3	-12.1×10^{-6}
Sicastar white 43-00-204	20	2.20×10^3	-9.23×10^{-6}
Surrounding media			
2 wt% $MnCl_2$ solution		1.01×10^3	$+27.0 \times 10^{-6}$
3 wt% $MnCl_2$ solution		1.02×10^3	$+35.9 \times 10^{-6}$

manganese dichloride aqueous solutions used in this study as the surrounding media. To prepare these solutions, manganese dichloride tetrahydrate, Wako 133-00725, was used.

In the magneto-Archimedes separation experiment, sample spheres and manganese dichloride aqueous solution were introduced into the sample container and bubbles forming on the surface of the spheres were removed using ultrasonic homogenizer. Then, the sample container was put in a superconducting magnet bore. In the magnet used here, the maximum magnetic force is acting on materials at $z=146$ mm. Therefore, the bottom of the sample container was fixed at $z=140$ mm to apply high magnetic force to spheres. With gradually increasing the applied magnetic field, the behavior of sample spheres was observed using CCD camera inserted into the bore. In the experiments here, mixtures of the soda-lime glass and the borosilicate glass (both 50 μm), the soda-lime glass and the Sicastar red (mixture of 50 μm and 20 μm), and the Sicastar red and Sicastar (both 20 μm) were tested. In the separation of the soda-lime glass and the borosilicate glass, 3 wt% $MnCl_2$ solution was used as surrounding. For the separation experiment of mixtures of the soda-lime glass and the Sicastar red, and the Sicastar red and Sicastar, the pH of the surrounding medium was adjusted to 10.02 with adding 0.1% Tween 20 (Wako, Polyoxyethylene (20)Sorbitan Monooleate, 160-11512) to improve the dispersion of particles in addition to 2 wt% of $MnCl_2$.

4. Results and discussion

Fig. 2 shows the result of the separation experiment for the mixture of the soda-lime glass and the borosilicate glass spheres. At the left side of this figure, the spatial distribution of $B{\cdot}\partial B/\partial z$is indicated. The two photographs in the center show the soda-lime glass and the borosilicate glass spheres levitated individually by the magneto-Archimedes levitation, while the photographs on the right side show the result of separating the mixture. The two different samples of glass spheres were levitated to different equilibrium positions in the fluid by the magneto-Archimedes levitation and thus separated clearly from each other. Their levitation end positions were identical to their individual levitation position, as expected. Fig. 3 similarly shows the result of the separation experiment for the mixture of the soda-lime glass spheres and Sicaster red. In addition, microscopic images of the collected particles after separation by magneto-Archimedes levitation are shown. There was a large difference between the stable levitation positions of these particles due to the difference of components, and they could thus be separated from each other relatively easily. Even without adjusting pH and without using the non-ionic detergent Tween 20, the separation of these spheres was observed. However, the separation efficiency was not optimal due to both particles forming agglomerates. These agglomerates were distributed between stable levitation positions of the soda-lime glass and the Sicastar red depending on the composition ratio of agglomerates.

The result of the magneto-Archimedes separation of the mixture of Sicastar red and Sicastar white is shown in Fig. 4. These particles consisted mainly of silica, but their densities and magnetic susceptibilities were different (Table 1). These differences seemed to be caused by the different components used to add color. As expected from these differences in densities and susceptibilities, the individual levitation experiments showed that the stable levitation positions of the different types of silica particles were distinct. In the separation from the mixture, their levitation position broadly distributed and some particles can be found in between their stable levitation positions. Possible reasons of this broadening are a particle size distribution, inhomogeneity of materials in particles, and the agglomeration of particles. The magneto-Archimedes levitation is insensitive with the size distribution of particles because both of the magnetic force and the gravity force are the volume force. As seen in the result of individual levitation, particles levitated at almost same position, therefore, the inhomogeneity of materials in particles seems not large. In general, these particles do not agglomerate, however, it may occur under the coexistence with electrolytes included in the media. Further investigation seems to be required to understand this. However, as seen in the microscope images of collected particles, each colored particles were observed at around their original levitation position. From these observations, we confirmed that the magneto-Archimedes separation can be applied for the separation of microspheres of larger than 20 μm.

Toward the novel application of the magneto-Archimedes separation as a way of the simultaneous quantitative analysis for multiple objects, it was experimentally evaluated in this study whether the magneto-Archimedes separation technique can be applied on the separation of micrometer-sized particles in a fluid.

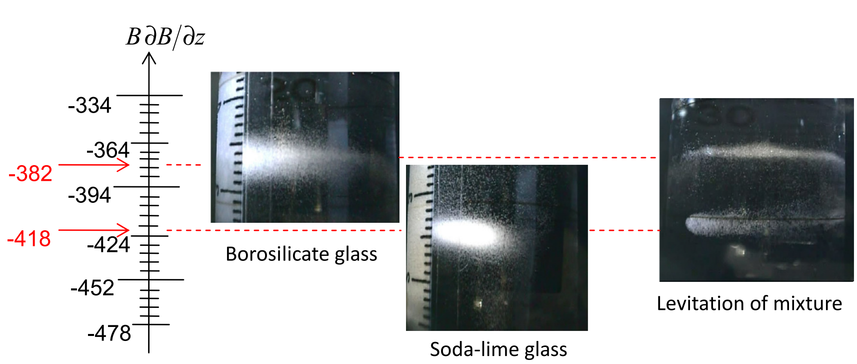

Fig. 2. Result of magneto-Archimedes separation experiments for the mixture of the soda-lime glass and the borosilicate glass spheres. From the left, the spatial distribution of $B{\cdot}\partial B/\partial z$, results when the soda-lime glass and the borosilicate glass were levitated individually, the result of the magneto-Archimedes separation are shown.

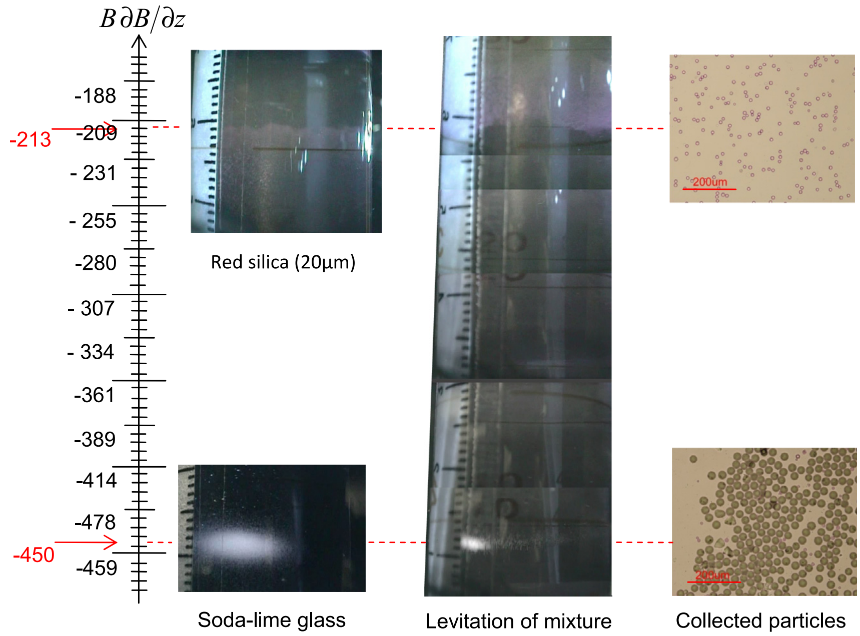

Fig. 3. Result of magneto-Archimedes separation experiments for the mixture of the soda-lime glass spheres and Sicaster red. From the left, the spatial distribution of $B\cdot\partial B/\partial z$, results when the soda-lime glass and Sicastar red were levitated individually, the result of the magneto-Archimedes separation, and microscope images of collected particles are shown.

At present, the required time for separation, which means the time to concentrate in a stable levitation position, took 10–20 min in case of 20 μm size particles. The magnetic force in competition with the fluid resistance or the force caused by thermal energy with decreasing particle size might be responsible for this required equilibration time. Using even smaller particles as is common in currently used medical analyses, equilibration times will become larger. However, in the magneto-Archimedes levitation, the intensity of the magnetic force acting on particles can be controlled by adjusting the properties of the surrounding medium. Therefore, there is still room for improvement in particle size and separation time by selecting proper conditions for the specific analysis. Around the maximum $B\cdot\partial B/\partial z$ position, in general, spatial distribution of $B\cdot\partial B/\partial z$ vary gently compared with the other places, therefore, the resolution of the separation becomes better in this region. By changing the coil design or by putting some magnetic materials around the samples space, even gentler $B\cdot\partial B/\partial z$ will be realized. Further high performance separation efficiency is expected by the control of spatial distribution of magnetic field with the optimization of the separation conditions.

5. Conclusion

In this study, it was confirmed experimentally that the magneto-Archimedes separation can be applied for the separation of a

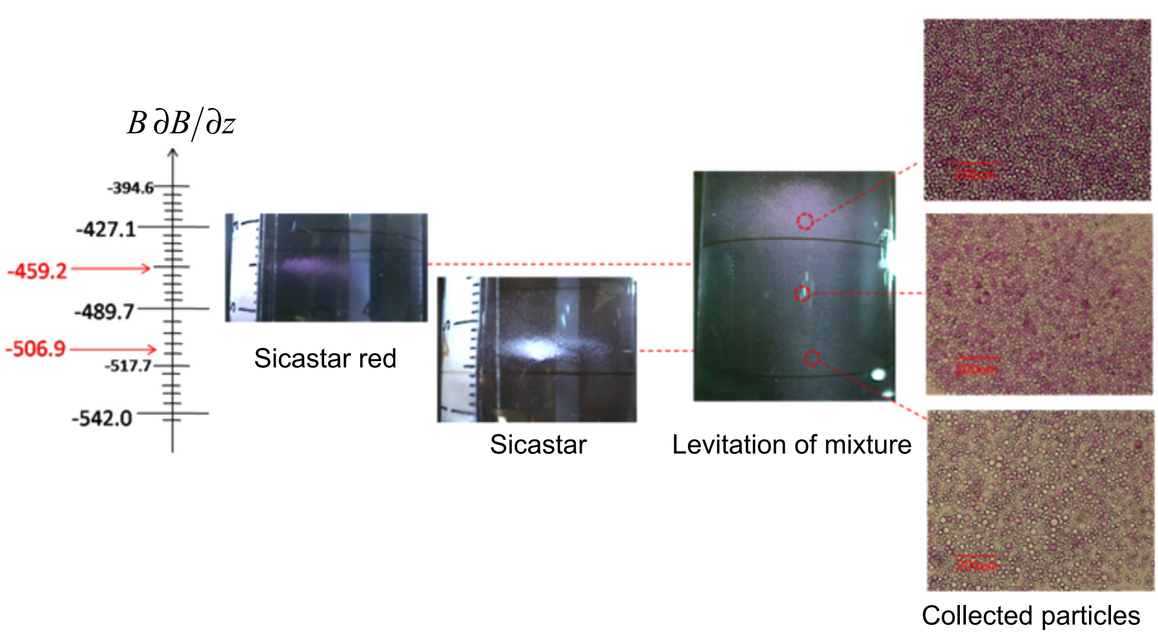

Fig. 4. Result of the magneto-Archimedes separation of the mixture of Sicastar red and Sicastar. From the left, the spatial distribution of $B\cdot\partial B/\partial z$, results when Sicaster red and Sicastar were levitated individually, the result of the magneto-Archimedes separation, and microscope images of collected particles are shown.

mixture of microspheres larger than 20 μm in a fluid. With this observation the possibility of this separation technique was shown to apply as a way of the simultaneous quantitative analysis for multiple objects. To attain faster separation applicable for smaller particles, further optimization of the separation conditions including the control of the spatial distribution of magnetic flux density seems to be required.

The magneto-Archimedes separation is sensitive to both the density and the magnetic susceptibility of the materials and one can choose the principal separation parameter by controlling properties of the surrounding medium. Even materials levitating to the same position at a certain condition can be made to take up another position by changing the property of its surrounding media. This enables the separation of materials into their components independent on particle size and/or size distribution. Therefore, the magneto-Archimedes separation is a potential way for the practical separation of feeble magnetic materials. Using these advantages, we hope that the magneto-Archimedes separation will be applied as a way of the simultaneous quantitative analysis for multiple objects.

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References

[1] E. Beaugnon, R. Tournier, Nature 349 (1991) 470.
[2] Y. Ikezoe, N. Hirota, J. Nakagawa, K. Kitazawa, Nature 393 (1998) 749–750.
[3] R.E. Rosensweig, Ferrohydrodynamics, Dover Publications, New York, 2014.
[4] US Patent US3483968 A, 1969.
[5] US Patent US3483969 A, 1969.
[6] Y. Ikezoe, T. Kaihatsu, T. Suetsuna, H. Uetake, N. Hirota, K. Kitazawa, Proceedings of the International Symposium on New magneto-Science'99 Omiya, Japan, (2000) p. 400.
[7] T. Kimura, S. Mamada, M. Yamato, Chem. Lett. 1294 (2000).
[8] N. Hirota, Y. Ikezoe, H. Uetake, T. Kaihatsu, T. Takayama, K. Kitazawa, Riken Rev. 44 (2002) 159.