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Size-selective separation of submicron particles in suspensions with ultrasonic atomization

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ABSTRACT

Aqueous suspensions containing silica or polystyrene latex were ultrasonically atomized for separating particles of a specific size. With the help of a fog involving fine liquid droplets with a narrow size distribution, submicron particles in a limited size-range were successfully separated from suspensions. Performance of the separation was characterized by analyzing the size and the concentration of collected particles with a high resolution method. Irradiation of 2.4 MHz ultrasound to sample suspensions allowed the separation of particles of specific size from 90 to 320 nm without regarding the type of material. Addition of a small amount of nonionic surfactant, PONPE20 to SiO_2 suspensions enhanced the collection of finer particles, and achieved a remarkable increase in the number of collected particles. Degassing of the sample suspension resulted in eliminating the separation performance. Dissolved air in suspensions plays an important role in this separation.

1. Introduction

Irradiation of a high frequency ultrasound in megahertz-range to a liquid brings about atomization into very fine droplets whose sizes are micrometer or below with a narrow size distribution. Unlike conventional pneumatic atomizers, no high pressure and orifice is required in this simple method, and formation of coarse droplet is successfully suppressed. Size of liquid droplets is tunable by varying the ultrasonic frequency and physical properties of liquids [1]. Because of the advantage of producing size-controlled droplets, the method has been applied to inhalation drug delivery [2,3], fuel combustion [4] and analytical nebulizers [5]. The method was even useful for fabricating nanostructured materials [6,7] or nanocomposits, where the liquid droplets are 'mother' of the products.

We had an idea of using the droplets as containers for solid particles of a specific size to be separated from its suspension. Possibility of the idea was proven by the authors with aqueous suspensions of silica and bentonite [8]. Sato's report [9] of ethanol separation from its aqueous solution triggered our interest in applying ultrasonic atomization to separation. They have shown a preferential distribution of ethanol between bulk liquid and fog. Prior to their report, Rasshokin [10] had published the enrichment of a surfactant into the droplets ultrasonically atomized. No such distribution occurs in conventional atomizers, where the whole liquid is atomized. Since after the recognition as a separation technique, ultrasonic atomization has been studied extensively on separating other materials such as alcohols [11], surfactants [12], amino acids [13], and carbon nanotubes [14].

The present study focuses on deeper understanding of the separation of submicron particles of silica or polystyrene latex from suspensions. Our motivation is to develop an easier and simpler method than conventional ones. Summarized in a review article by Fedotov et al. [15] were common methods for separating micrometer- to nanometer-sized particles in analytical chemistry. Generally, ultrafiltration and microfiltration were performed in which membranes were used as a barrier for particles. In field-flow fractionation, a thin channel for liquids is provided and fluid flow is finely designed [16]. A physical force field such as gravitational, centrifugal or electric is applied to enhance separation. Microfluidic systems use confined space for sieving or controlling particle movement in fluids [17–19]. Methods depending on narrow paths for fluids suffer from a serious problem of clogging and pretreatment of crude samples is required for a stable operation. Furthermore, the particle concentration of the sample is limited to low. Even in centrifugal sedimentation [20], which has been applied to larger scale processes, finding a suitable density gradient is difficult. In contrast, ultrasonic atomization is free from difficulties in narrow paths or finding additives for making density gradient, and it could be robust for crude samples. The method is potentially useful for practical particle separation.

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To enhance characterization of this separation method, solid particle size and its concentration were finely evaluated with equipment enables a high resolution analysis. Also, an approach, degassing of the sample suspension was adopted to shed a light on the mechanism.

2. Experimental

Solid samples employed in this study are size- and shape-controlled SiO_2 (Sicastar, micromod Partikeltechnologie GmbH) and polystyrene, PS, latex particles (Micromer, micromod Partikeltechnologie GmbH). The shape was spherical, and the sizes for SiO_2 particle are four different nominal diameters of 50, 70, 100, and 300 nm. The diameters for PS latex particles are 50 and 100 nm. Sample suspensions were prepared by mixing two types of particles of the same material with different sizes in a predetermined volume of deionized water. In the preparation, the number of particle for each diameter was set to be close, and total particle concentration was set constant at 1000 ppm. This particle concentration is much higher than the conditions taken in reported studies on separation of nanometer-sized particles [21–23].

A surfactant, polyoxyethylene(20) nonylphenylether, PONPE20 was used as an additive to modify the separation performance.

Particle-size distribution of the solid sample was measured with a method of tracking analysis of particles illuminated by a laser light. The apparatus used was Nanosight LM-10R (Quantum Design Japan Co. Ltd.). The method enabled us an observation of individual particles as point-scatterers moving under Brownian motion. Basic principle of size determination is based on Stokes–Einstein's equation. Such a direct observation of particle suppresses background effect, thus attained were both a high resolution and a measurement of concentration of solid particles. Lower detection limit of particle diameter is 30 nm (depending on the sample property) [24].

A schematic diagram of the experimental apparatus is shown in Fig. 1. It consists of an atomization column and a particle collection unit. The column is 0.25 m in height and 0.054 m in diameter. To irradiate an ultrasound with a frequency of 2.4 MHz to sample suspensions, a transducer whose diameter of 0.02 m was mounted at the bottom of the column. A suspension sample with a volume of 50 cm^3 was irradiated with the ultrasound at a constant electric power input of 10 W. Temperature of the suspension was kept constant at 303 K by cooling with a coolant flowing in the coiled tube. As was observed in most liquids, the ultrasonic irradiation to suspensions led to the formation of a fountain. A fog containing solid particles was formed at the surface of the fountain. The fog was taken out of the column by nitrogen flowing at a rate of 0.5 L/min, and was transferred to the particle collection unit for 1 h. In the unit, the fog and nitrogen were passed through an impinger loading with 30 cm^3 of water to capture solid particles. Another type of equipment, a cold trap using liquid nitrogen was also applied for collecting particles.

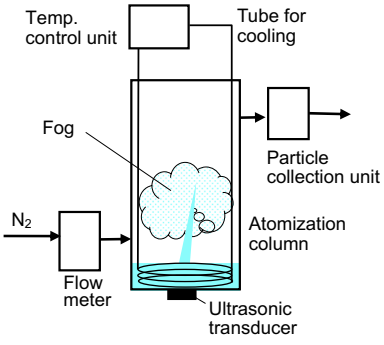

Fig. 1. Schematic diagram of experimental apparatus.

3. Results and discussion

3.1. Separation of mixtures containing SiO_2 particles

A typical experimental result was presented in Fig. 2, where the particle concentration is plotted against the diameter. The sample suspension consists of 100 and 300 nm SiO_2 particles. Two main peaks for 100 and 300 nm particles were detected, which means a high resolution of the present analysis. Concentrations of collected particles were much lower than the sample suspension due to dilution which is inevitable in the collection method. The impinger method is simple but uses water as a medium for particle collection, and fog is brought into contact with water. In spite of a high stability of observed values, concentration of collected particle depends largely on conditions such as water volume, shape of the reservoir and method of dispersing gas. Therefore, another method, a cold trap with liquid nitrogen was applied to confirm the evaluation with impinger method and also to find actual solid particle concentration in fog. The cold-trap method attained a fog collection rate of 80 wt%, and particle concentration was found to about 40% of the original sample, which is much higher than approximately 0.1% from impinger method. Particle size distributions for both collection methods were compared in Fig. 3. Two curves accorded well with a small difference in diameter, thus the result assured validity of the impinger method. Due to the simplicity in operation, impinger method was applied for size determination of particles and relative evaluation of particle concentration.

Collected particles mainly consisted of 100 nm particles. The size distribution was shown in Fig. 4. The median diameter of the collected particles was 140 nm. The result clearly demonstrates that the smaller particles were selectively transferred into fog.

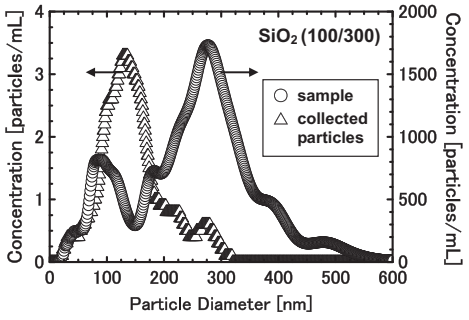

Fig. 2. Concentration of particles contained in original sample mixture of SiO_2 (100 and 300 nm) and of collected particles.

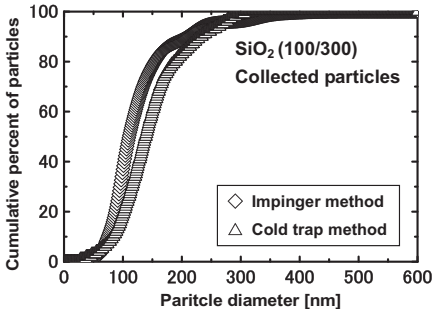

Fig. 3. Comparison between particle size distribution of collected particles with impinger and cold trap.