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Separation characteristics of alcohol from aqueous solution by ultrasonic atomization

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ABSTRACT

The generation rate of ultrasonically atomized droplets and the alcohol concentration in droplets were estimated by measuring the flow rate and the alcohol concentration of vapors from a bulk solution with a fountain. The effect of the alcohol concentration in the bulk solution on the generation rate of droplets and the alcohol concentration in droplets were investigated. The ultrasonic frequency was 2.4 MHz, and ethanol and methanol aqueous solutions were used as samples. The generation rate of droplets for ethanol was smaller than that for methanol at the same alcohol molar fraction in the bulk solution. For both solutions, at low alcohol concentration in the bulk solution, the alcohol concentration in droplets was lower than that in vapors and the atomized droplets were visible. On the other side, at high concentration, the concentration in droplets exceeded that in vapors and the atomized droplets became invisible. These results could be explained that the alcohol-rich clusters in the bulk solution were preferentially atomized by ultrasonic irradiation. The concentration in droplets for ethanol was higher than that for methanol at low alcohol concentration because the amount of alcohol-rich clusters was larger. When the alcohol molar fraction was greater than 0.6, the atomized droplets almost consisted of pure alcohol.

1. Introduction

When a liquid is irradiated with ultrasound, a fountain arises from the liquid surface and fine liquid droplets are generated from the fountain [1]. This phenomenon is called ultrasonic atomization. The ultrasonic atomization is utilized in various processes such as humidification, aroma diffusion and nanoparticle synthesis [2]. Recently, it was observed that ethanol was separated from the aqueous solution by ultrasonic atomization [3]. The effects of apparatus [4–7] and sample [8,9] conditions on the ethanol separation performance were investigated. It was reported that surfactants [10,11] and amino acids [12] were also enriched by the ultrasonic atomization. However, the separation mechanism is not fully elucidated because the ultrasonic atomization includes the vaporization from the bulk solution with the fountain in addition to droplets. In order to discuss the mechanism of separation by ultrasonic atomization, the atomized droplet amount and the vaporization amount should be estimated separately.

The droplet diameter in ultrasonic atomization was investigated by Lang [13]. He changed ultrasonic frequency in the range from 13 to 780 kHz and examined the droplet diameter of molten wax by taking photomicrographs. He proposed an equation describing the influence of ultrasonic frequency, surface tension and density of liquid on the droplet diameter. By using Lang's equation, droplet diameters of water and ethanol at 2.4 MHz are calculated to be 2.3 and 1.7 μm, respectively. Effect of viscosity was studied by Ramisetty et al. [14]. Recently, Yano et al. [15] measured droplets of ethanol aqueous solution at 2.4 MHz by X-ray scattering and revealed the existence of nanometer-sized droplets at ethanol molar fractions of 0.2 and 1.0. Kobara et al. [16] used a scanning mobility particle sizer and a hand-held particle counter to measure droplet size distributions of ethanol aqueous solution at 2.4 MHz. They observed only nanometer-sized droplets with diameters of about 30 nm at the molar fraction of 0.50.

Compared with the research on the droplet diameter, the research on the amount of atomized droplets are few. The reason is difficulty in collecting all droplets generated from an ultrasonic atomizer. The estimation of the atomization amount mainly had been conducted by means of mass change [3,4]. In this method, the sample mass is measured before and after the atomization and the amount of mass loss is regarded as the atomization amount. However, this atomization amount includes the vaporization amount from the bulk solution with a fountain since all

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droplets must be ejected from an atomizer vessel by a carrier gas flow at high gas velocity. In our previous study [17], two apparatuses were used to estimate the generation rate of atomized droplets of water. The one was an apparatus with an ultrasonic transducer. Another was an apparatus with a liquid pump and a nozzle to measure the vaporization amount. The generation rate of droplets was able to be estimated by differences of mass change between these two apparatuses. We clarified that the mass generation rate of droplets decreased with increasing ultrasonic frequency at the same ultrasonic intensity. At the same apparent surface area of the fountain, the number of atomized droplets became larger as the ultrasonic frequency increased. The ultrasonic atomization was considered to arise from a combination of the capillary wave at the fountain surface and the cavitation inside the fountain.

Ultrasonic atomization studies concerning alcohol aqueous solution have been mainly conducted against ethanol [3–9]. The information of alcohol concentration in droplets is important to elucidate the mechanism of separation by ultrasonic atomization. However, the amount of atomized droplets and the concentration in droplets have not been reported to our knowledge. In this study, ethanol and methanol aqueous solutions were used as atomizing liquids. The effect of the alcohol concentration in the bulk solution on the generation rate of droplets and the alcohol concentration in droplets were investigated at 2.4 MHz.

2. Experimental

2.1. Apparatus

Fig. 1(a) shows the schematic diagram of the experimental apparatus with an ultrasonic transducer. The cylindrical vessel was made from transparent polyvinyl chloride resin. The height and inside diameter of vessel were 300 and 100 mm, respectively. A disc-shaped lead zirconate titanate (PZT) ultrasonic transducer (Honda Electronics Co. Ltd.) was installed at the central position of the bottom of vessel. The frequency and diameter of transducer were 2.4 MHz and 14 mm, respectively. Ultrasound was irradiated vertically upwards to the liquid surface. The transducer was driven by a power amplifier (AP400B, ENI) and a signal generator (1941, NF Corp.) to emit a continuous sinusoidal wave. An effective electric power applied to the transducer was calculated from a voltage at the transducer and a current measured using an oscilloscope (TDS3014B, Tektronix Inc.) and a current probe (TCP202, Tektronix Inc.). The effective power applied to the transducer was 20 W. In the case of water, the ultrasonic power determined by a calorimetric method was 13.4 W (8.78 W/cm^2). The atomization threshold intensity was about 3 W/cm^2.

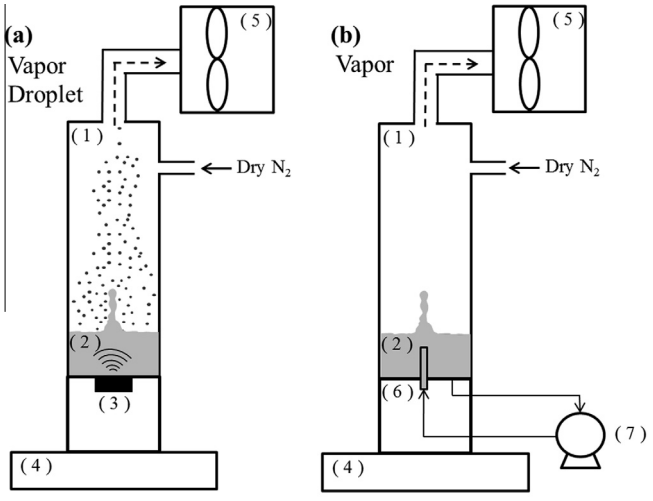

Fig. 1. Schematic diagram of experimental apparatus: (a) ultrasound and (b) pump.

The carrier gas was dry nitrogen and flowed through the vessel to accompany the vapors and atomized droplets by a fan. The gas inlet was fitted at the position of 250 mm from the transducer surface. The initial height of sample was 30 mm. To eject all atomized droplets from the vessel, the flow rate of carrier gas was set at 3.3×10^{-4} m^3/s (the superficial gas velocity based on the vertical cross-section of the vessel was 42 mm/s). The vessel was put on an electronic balance. The ultrasonic irradiation time was five minutes. After ultrasonic irradiation, the mass of the bulk liquid in the vessel was measured by the electric balance and the alcohol concentration was determined by the gas chromatograph equipped with a TCD detector (GC323, GL Science Inc.). The fountain shape and height were observed using a video camera. The sample temperature before ultrasonic irradiation was 293 K and the temperature rise in solution after ultrasonic irradiation was within 3 K.

Fig. 1(b) shows the schematic diagram of the experimental apparatus with a liquid pump. In order to reproduce the fountain formed by ultrasonic irradiation, a nozzle and a liquid pump were used. The carrier gas was dry nitrogen and vapors were ejected from the vessel top by the fan. The flow rate of carrier gas was 3.3×10^{-4} m^3/s. The experimental time was five minutes. After experiment, the mass and alcohol concentration of the bulk liquid in the vessel were measured. Special grade chemical ethanol and methanol were purchased from Wako Pure Chemical Industries, Ltd. These reagents were used without further purification. The distillated water was used.

2.2. Estimation of generation rate of droplets and alcohol concentration in droplets

By using the apparatus with the ultrasonic transducer, the rate of mass change of bulk solution R_u and the alcohol concentration in the discharge solution C_u for the ultrasound are calculated from the material balances as follows:

$$R_u = (M_{u0} - M_{ut})/t \tag{1}$$

$$C_u(M_{u0} - M_{ut}) = C_0 M_{u0} - C_{ut} M_{ut} \tag{2}$$

where M_{u0} and C_0 are the initial mass of bulk solution and initial alcohol concentration in the bulk solution, M_{ut} and C_{ut} are the mass of bulk solution and the alcohol concentration in the bulk solution after ultrasonic irradiation, and t is time.

By using the apparatus with the liquid pump, the rate of mass change of bulk solution R_p and the alcohol concentration C_p in the discharge solution for the pump are calculated from the material balances as follows:

$$R_P = (M_{p0} - M_{pt})/t \tag{3}$$

$$C_P(M_{p0-M_{pt}}) = C_0 M_{p0} - C_{pt} M_{pt} \tag{4}$$

where M_{p0} is the initial mass of bulk solution, M_{pt}, and C_{pt} are the mass of bulk solution and alcohol concentration in the bulk solution after the pump experiment.

In the case of the apparatus with the ultrasonic transducer, the atomized droplets and the vapors, which are generated from the bulk liquid with a fountain, are ejected from the vessel, and the mass change is the amount of droplets and vapors. For the case of the apparatus with the liquid pump, the vapors, which are