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Ethanol enrichment from ethanol–water mixtures using high frequency ultrasonic atomization

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ABSTRACT

The influence of high frequency ultrasound on the enrichment of ethanol from ethanol–water mixtures was investigated. Experiments performed in a continuous enrichment system showed that the generated atomized mist was at a higher ethanol concentration than the feed and the enrichment ratio was higher than the vapor liquid equilibrium curve for ethanol–water above 40 mol%. Well-controlled experiments were performed to analyze the effect of physical parameters; temperature, carrier gas flow and collection height on the enrichment. Droplet size measurements of the atomized mist and visualization of the oscillating fountain jet formed during sonication were made to understand the separation mechanism.

1. Introduction

High frequency ultrasonic atomization of liquids is an effective way of generating small droplets by subjecting the liquid to a sufficiently high intensity and high frequency ultrasonic field. During high frequency ultrasonic atomization, a piezoelectric crystal, vibrating at frequencies greater than 500 kHz, supplies the energy required for atomizing the liquid. The piezoelectric crystal is typically installed at the bottom of the sonication vessel and focused acoustic energy is transmitted to the atomizing fluid, either directly or via a coupling liquid. When the ultrasonic vibrations are sufficiently intense, a fountain rises from the surface of the liquid due to acoustic pressure waves and breaks up at the apex as a result of gravitational effects. Large liquid ligaments and drops formed as a result of the breakup are commonly returned into the bulk liquid. In addition to the large liquid ligaments, small low inertia droplets are also released periodically from the jet and quickly surround the jet to form a dense fog. Droplet formation during ultrasonic atomization is often considered to be a consequence of the growth of surface waves generated by spreading a thin layer of liquid on the atomizer surface and are reported to be uniform in distribution. In other words, droplets are produced by the large number of air–liquid interfaces generated due to the high frequency oscillation of the surface. The droplets formed travel at very low velocities and hence a carrier fluid such as air is often used to transport the droplets. Ultrasonic atomization has some distinct advantages over conventional atomization processes for the production of small droplets. The droplet size has a very narrow distribution and is inversely proportional to the frequency of ultrasound. The density of the fog generated and the momentum of the droplets can be independently controlled by adjusting the carrier fluid rate past the liquid surface since the amount of liquid suspended in air as a fog is limited only by the rate at which it falls back into the liquid or by the air flow rate at the liquid surface.

Early studies of droplet sizes formed during high frequency ultrasonic atomization concluded that droplets generated using this atomization process were uniform in size and a mono-dispersed spray could be generated by this method. Numerous reports [1–6] have been published on the mechanism of droplet formation by ultrasonic atomization with this limited understanding in the past. However, it has recognized the complexity introduced by an observed acoustic cavitation zone in the liquid and the periodic release of droplets from the capillary wave (fountain) formed at the surface of the liquid. Also, recent observations have noted that the droplet formation and consequent droplet size distribution are not as uniform as previously considered [1]. The formation of satellite droplets, larger than the primary droplets was reportedly observed in recent experiments and uniformity droplet size distributions over a period of time, based on a physical understanding and models of the atomization process, has been discussed [2].

In general, two theories are considered to explain the mechanism of droplet formation during sonication of liquids based on the resonant ultrasound frequency and intensity. The cavitation

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theory assumes that when a liquid is subjected to ultrasound, small cavitating bubbles formed in the liquid oscillate and collapse implosively close to the surface of the liquid generating high intensity hydraulic shock waves and eject droplets from the liquid surface. This hypothesis was postulated for low frequency ultrasonic atomization. The second theory, based on capillary effects or known as the capillary wave theory [3], was postulated for high frequency ultrasonic atomization (>800 kHz) and hypothesizes that atomization occurs by the breakup of the capillary waves formed at the surface of the liquid due to unstable oscillations that release droplets from the crests of the capillary waves. Hence atomization takes place away from the bulk liquid in the surface capillary waves and the droplets produced at the crests of the capillary wave are proportional to the wavelength of the capillary instability. The capillary wavelength decreases with increasing frequency and produces finer droplets at higher frequencies. For sonically generated capillary waves, whose wavelength can be calculated using Kelvin's equation, the surface wave frequency is equal to one-half of the exciting sound frequency. Hence the droplet size produced by atomization must be approximately one-half the capillary wavelength. A comparison of the number mean diameter with the capillary wavelength indicated that the mean droplet size was a constant fraction of the capillary wavelength. This fraction was determined to be 0.34. Hence Lang [3] proposed the correlation shown below:

$$d_p = 0.34\left(\frac{8\pi\sigma}{\rho f^2}\right)^{\frac{1}{3}}$$

The strong correlation between capillary wavelength and measured droplet size formed favors the capillary wave theory. However, the influence of liquid vapor pressure, amount and type of dissolved gas in the liquid, sonoluminescence and observed droplet breakup on ultrasound induced atomization shows the dependence of the atomization process on acoustic cavitation [4–6].

Boguslavskii and Eknadiosyants [4] extended the capillary theory for high frequency ultrasonic atomization and proposed a "conjunction" theory that attributes droplet formation to be an effect of periodic shocks generated by the implosive collapse of cavities in the capillary waves leading to the breakup of capillary wavelength scale droplets. The cavitational disturbances are assumed to be responsible for the broadening of the droplet size distribution. Due to the random nature of cavitation, limited experiments that show the effect of cavitational disturbances have been reported and majority of research has relied on the capillary wave theory and Lang's equation [3] for the estimation of mean droplet size.

Selective separation of ethanol from ethanol–water mixtures using ultrasound was first introduced by Sato et al. [7]. They subjected ethanol–water mixtures of 10–90 mol% ethanol at 10 °C to high frequency ultrasound at 2.4 MHz. The atomized mist was reported to be pure ethanol and a mechanism based on parametric decay instability of the capillary wave, in which local accumulation of the acoustic energy occurs and this leads to the formation of an atomized mist of pure ethanol was proposed. Remarkably, unlike distillation processes, much less heat is required since phase change is less important in the process. In our previous reported work [8], an alternate mechanism based on the conjunction theory has been postulated for the process of ultrasonic atomization. This mechanism involves the formation of micro-bubbles in the liquid during sonication and the growth of the bubbles as they travel through the liquid mixture and collapse at the liquid surface into a cloud of micro-bubbles moving upwards in a capillary fountain jet and releasing the alcohol vapor diffused in the bubbles. The selective separation of alcohols was explained as a corollary effect of the physical mechanism whereby a surface excess of alcohol molecules formed at the surface of the micro-bubbles. The alcohol molecules at the walls of the bubbles were expected to diffuse into the bubbles and fill the bubbles with alcohol vapor until their collapse in regions of high accumulation of acoustic energy. However, this theory limits the ethanol concentration of the mist to the surface excess over the bulk concentration.

In this study, our previous work on evaluation of ethanol enrichment [9] was extended with experiments conducted in a continuous enrichment system to evaluate ethanol enrichment and also develop a reliable process with a constant enrichment quality for a given feed ethanol concentration. In addition, droplet size measurements of the enriched ethanol mist and local temperature measurements of the ultrasonic jet formed were also performed.

2. Experimental setup

A schematic of the experimental arrangement used in this study is shown in Fig. 1. A high frequency ultrasonic transducer, operating at 2.4 MHz and 18 mm diameter, was installed in the bottom of a glass cylindrical column, of 0.5 m diameter and 1 m (height), with a stainless steel bottom. Ethanol–water solutions with concentrations up to 90 mol% ethanol were subjected to ultrasound in this column fed from a 1-L beaker during the study. During sonication, sonic energy is imparted to the feed liquid and a fountain is formed at the surface of liquid in the column and a droplet mist is released from the fountain. Ultrasound irradiation of the liquid combined with acoustic cavitation in the fountain increases the temperature of the fountain. The column incorporates an opening to allow the fountain to pass through and fall into the bulk solution thereby avoiding avoid temperature and concentration changes during the process as shown in Fig. 1. Feed liquid level in the column was kept constant at 3 cm above the transducer level by adding fresh feed liquid from the beaker to compensate for the amount of liquid atomized and the ultrasonic fountain re-circulated to the beaker. The input power for the transducer was maintained at 20 W. Air is introduced in the system at a very low velocity (0.086–0.271 L/min) as a carrier gas to collect the mist produced by atomization to a series of condensers. Mist collection outlets were provided at different elevations (at 0.3, 0.5, 0.9 m) above the surface of the transducer in the column. Time to the collect a condensate sample was typically 1 h to ensure that a large enough sample was made in analysis and the concentrations, quantity of mist and temperature distribution in the solution were measured. The ethanol concentration in the feed mixtures and condensate

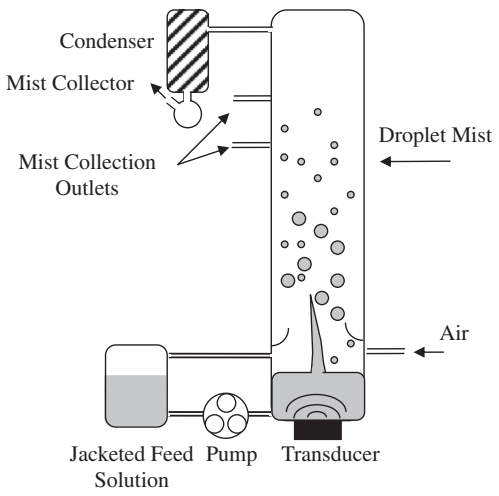

Fig. 1. Schematic representation of experimental apparatus used for ultrasound studies on ethanol–water mixtures.