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Experimental Investigation of Ethanol Enrichment Behavior in Batch and Continuous Feed Ultrasonic Atomization Systems*

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The fragmentation of a liquid layer to form a fine droplet mist by high frequency ultrasonic atomization of liquids has been applied to a range of industrial applications such as fine chemical manufacturing, pharmaceutical production, and food processing. A recent development is the separation of alcohol from miscible alcohol-water mixtures using ultrasonic atomization. In this work, the effect of high frequency ultrasonic atomization at 1.6 MHz on the enrichment of ethanol from ethanol-water feed mixtures has been studied. Experiments for evaluating this enrichment process were conducted in batch and continuous feed processing systems. The continuous enrichment process generated product concentrations that were higher than the equivalent vapor-liquid equilibrium curve at feed concentrations greater than 40 mol.-% in a single stage. The role of the ultrasonic jet formed at the surface of the feed solution combined with the ethanol separation characteristics has been discussed.

1 Introduction

When liquids are subjected to high frequency ultrasound, the combination of the cavitation effects through the liquid and the capillary instability at the surface of the liquid jet formed have been identified as contributing towards the formation of micron-sized droplets by Eknadiosyants et al. [1]. Although uniform droplet sizes are formed during ultrasonic atomization, the physical mechanism has been recognized to be quite complex in nature. The key benefit of this atomization process over conventional atomization processes is that the droplets generated have a narrow size distribution and low inertia [2]. Operating parameters, such as frequency and intensity of the ultrasound, in conjunction with physical properties of the liquid, control the droplet size formed. Droplet momentum can be independently adjusted by using a carrier gas at different velocities to transport the droplets. Compared to conventional atomization processes, ultrasound generates micron sized droplets and the droplets formed are uniform in size.

The application of ultrasound for ethanol separation from ethanol-water mixtures has been reported earlier by Sato et al. [3]. The enrichment of the atomized mist from ethanol-water mixtures at different temperatures sonicated at 2.4 MHz was studied and a mechanism for explaining the enrichment by the parametric decay instability of the capillary wave was proposed. However, the importance of the role of cavitation in high frequency ultrasound processes has been illustrated in recent work by Kirpalani and McQuinn [4]. An alternate mechanism to explain the enrichment based on the physical phenomena observed was proposed by Kirpalani et al. [5]. This theoretical mechanism combines the physical formation of cavitating micro-bubbles in the liquid with the oscillating fountain jet at the surface of the liquid during sonication. The proposed theory postulates that cavitating bubbles grow as they travel through the ethanol-water mixture, collapse in the fountain jet and release the alcohol vapor diffused in the bubbles during their expansion. The selective separation of alcohol was explained as a corollary effect of the physical mechanism, whereby a surface excess of alcohol molecules formed at the surface of the cavitating micro-bubbles in a fountain jet.

In previously reported research [3,5] a batch separation system with externally cooled jacketed system was used to maintain the feed temperature. Feed volume, alcohol concentration in the feed solution, and physical properties of the feed were found to change significantly during sonication. As a result, the separation mechanism for these miscible liquids could not be confirmed. Also, for determining the separation mechanism and performing a scale-up of this process, the feed concentration above the transducer needs to be maintained. Other issues to consider during scale-up for ultrasonic process are associated with the complex behavior of acoustic cavitation in liquids [6,7]. To make up for the deficiencies in the batch system, a continuous feed system was designed to validate the earlier proposed alcohol separation mechanism [8].

In this study, detailed experiments were performed to get a better understanding of the ethanol separation characteristics from ethanol-water mixtures, conducting the experiments in two different systems; an internally cooled batch process, where cooling was applied to the feed directly, and a continuous feed system, as shown in Fig. 1. The axial temperature profile in the fountain jet caused by ultrasonic irra-

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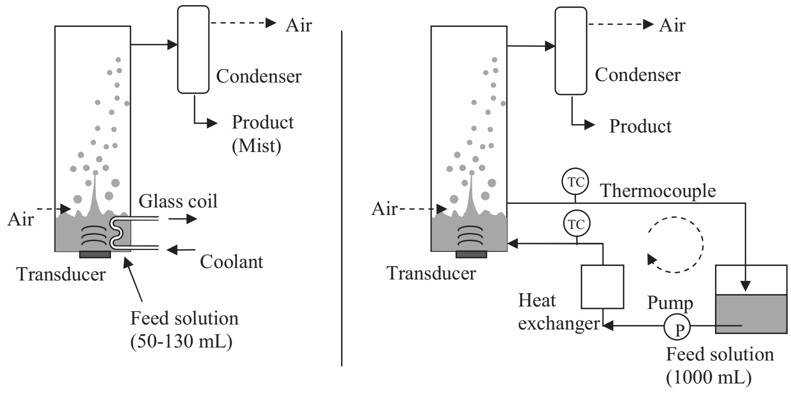

Figure 1. Outline of experimental systems for ultrasonic atomization; (a) Batch system, (b) Continuous feed system.

diation was also studied to develop a better understanding of the separation of ethanol.

2 Experimental

2.1 Batch System for Ethanol-Water Separation

Initial experiments in batch separation of ethanol were performed using an external cooling jacket. However, it was found that the temperature of the feed inside the batch column could not be maintained and a direct cooling glass coil was added inside the system to maintain the temperature of the feed above the transducer. A schematic diagram of the batch system used in this work is shown in Fig. 1a). A high frequency ultrasonic transducer operating at 1.6 MHz with a size of 0.018 m diameter and 0.001 m thickness was installed at the bottom of a glass cylindrical column of 0.05 m diameter and 0.5 m height with a stainless steel bottom. The input power of the ultrasound was 21 W. Ethanol-water solutions with concentrations up to 80 mol.-% ethanol were subjected to ultrasound in this column. The initial volumes of the feed solutions were between 50–130 mL.

During sonication, ultrasonic energy is imparted to the feed solutions and a fountain jet is formed at the surface of the liquid in the column, releasing droplets (mist) from the fountain jet (see Fig. 2). Ultrasonic energy supplied to the liquid, combined with acoustic cavitation in the fountain jet, increases the temperature of the jet. A glass coolant coil was installed to maintain a constant temperature of 297 ± 1 K in the ambient liquid during sonication. The droplet mist generated from the fountain jet was led by carrier gas (air) introduced in the column at a constant flow rate ($2.9 \cdot 10^{-6}$ m^3/s) to a cascaded condenser system and collected. In this batch system, the liquid height from the surface of the transducer decreases during sonication. However, this system differs from earlier work [3], since the liquid was subjected to direct cooling of the feed above the transducer and the velocity of the carrier gas is significantly lower.

Figure 2. Formation of fountain jet and mist during high frequency ultrasonic irradiation.

2.2 Continuous Feed System for Ethanol-Water Separation

The continuous feed system was designed so as to obtain a near-constant feed solution concentration and temperature above the ultrasonic transducer. This system consists of a high frequency transducer located at the bottom of a glass column with a 1 L bulk feed solution beaker that supplies the alcohol-water mixture to the glass column using a peristaltic pump, as shown in Fig. 1b). The condensate collection arrangement was similar to that used in the batch system. The column incorporates a fountain jet collection port for re-circulating the jet into the feed beaker. In this system, the liquid level in the column was kept constant at 0.035 m above the surface of the 1.6 MHz transducer by circulating the ethanol-water solution fed from the feed solution vessel. During sonication, the ambient liquid temperature in the column was kept constant at 297 ± 1 K by using a heat exchanger attached to the feed vessel. The mist generated was collected in both the continuous and batch systems by introducing air into the column at constant velocity. The change of the ethanol concentration in the feed solution in the feed vessel due to recirculation is relatively small due to the large volume ratio between the feed in the glass column and that of the feed solution beaker.

The ethanol concentration in the feed mixtures, condensate products, and residual solution in the glass column at the end of each experiment in each system was analyzed by using a gas chromatograph (Hewlett Packard HP6890 series) and an attached flame ionization detector. The relation between the concentrations of the feed mixtures and condensates was studied and mist collection rates were also recorded. The height of the fountain jet formed above the surface of the feed mixture and local temperature measurements in the jet were made using a K-type thermo couple.

3 Results and Discussion

3.1 Effect of Processing Time on Product Concentration in the Batch System

The enrichment behavior of ethanol-water mixtures with varying feed ethanol concentrations[1] C_0 in the batch system are shown in Fig. 3. The dotted line in Fig. 3 indicates the vapor-liquid equilibrium of the ethanol-water mixture at 101.3 kPa. The condensate (product) ethanol concentration C after sonication of the feed solution for 1 h, was found to be higher than that of the feed mixture using the internally (direct) cooled batch processing arrangement described earlier. Also, it was found that the condensate ethanol concentration decreased with processing time t as illustrated in Fig. 4. It can be inferred that the ethanol concentration of the product during ultrasonic atomization is process time dependent in the batch system. As a result, batch processing is quite ineffective for processing since constant condensate concentration cannot be maintained during the process. To further determine the batch process characteristics, the ethanol collection rate q has been plotted with respect to processing time, as shown in Fig. 5. The collection rate increases

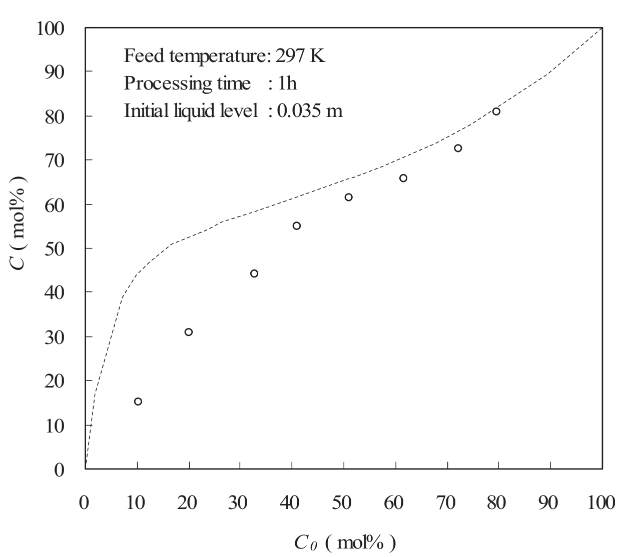

Figure 3. Ultrasonic separation characteristics of an ethanol-water solution by batch ultrasonic processing. The dotted line represents the vapor-liquid equilibrium for ethanol-water solution at 101.3 kPa.

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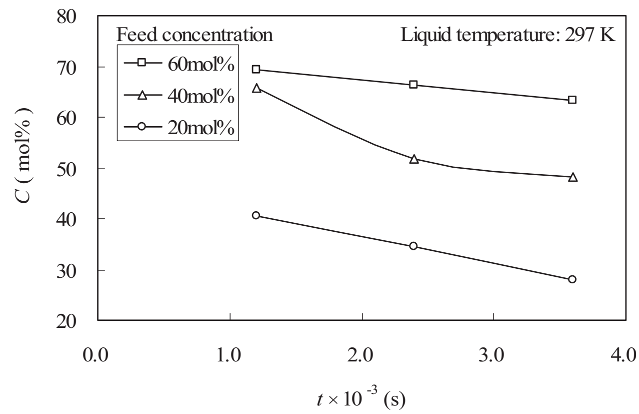

Figure 4. Effect of sonication time on the ethanol concentration of the condensate in the batch system.

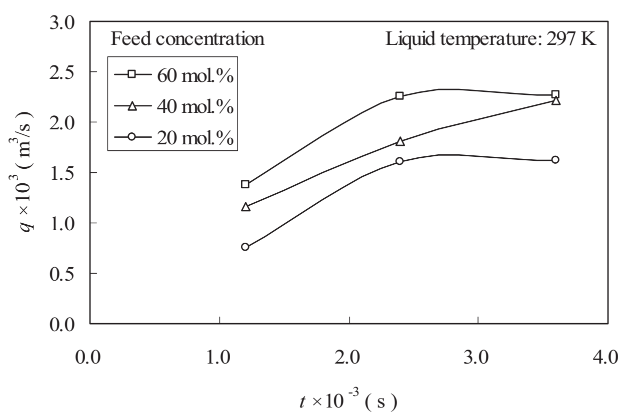

Figure 5. Effect of sonication time on the ethanol collection rate in the batch system.

with processing time and the increase can be attributed to the increase in ultrasound energy density supplied to the decreasing volume of feed mixture by the sonicator during batch processing.

3.2 Effect of Liquid Height on Product Concentration in the Batch System

The effect of the liquid level on the atomization rate of ethanol in the batch system used was studied to understand the physical separation mechanism. Fig. 6 shows that an increase of the liquid level h caused an increase in the concentration of the condensate. The ethanol collection rate decreased uniformly with increasing liquid level, as shown in Fig. 7.

3.3 Ethanol Separation Characteristics in the Continuous Feed System

Enrichment characteristics of ethanol-water mixtures for varying feed alcohol concentrations C_0 in the continuous

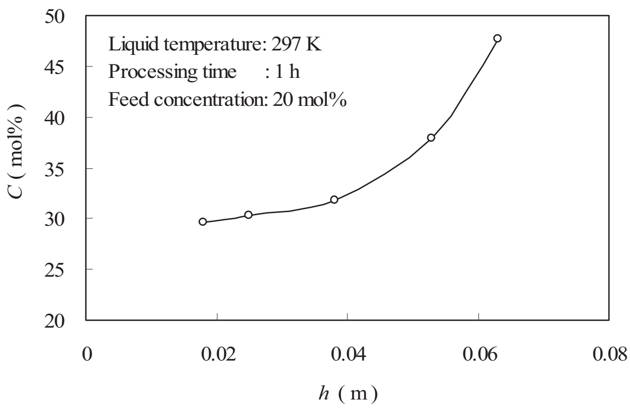

Figure 6. Effect of liquid height on the ethanol concentration of the condensate in the batch system.

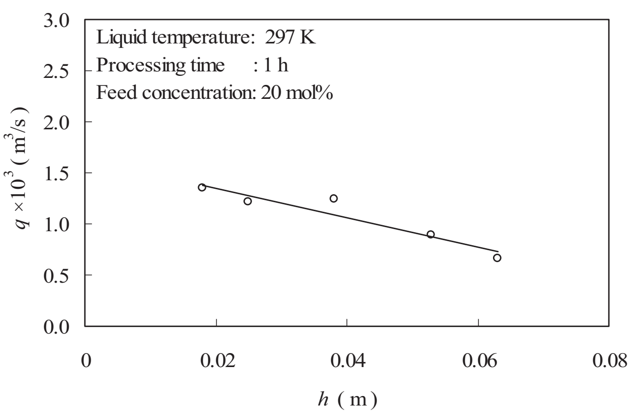

Figure 7. Effect of liquid height on the ethanol collection rate in the batch system.

feed system are shown in Fig. 8. The enrichment curve clearly shows that the ethanol enrichment in the condensate and the ethanol concentration in the condensate C in the continuous system were higher than in the batch system. An interesting aspect of the separation process is the product (condensate) concentration above the vapor-liquid equilibrium curve for feed concentrations above 40 mol.-% ethanol, as shown in Fig. 8. As expected, the condensate concentration over various time intervals (up to 7 h) was found to be constant irrespective of the feed sonication time, and ethanol collection rate q was also constant, as shown in Fig. 9.

As shown in Figs. 4 and 6, the concentration of the condensate in the batch system was influenced by the processing time and liquid level in the column. The enrichment characteristics in the continuous sonication system were found to be repeatable with less than 3 % error typically, as shown in Fig. 8. Also, the product concentration was found to be independent of processing time. This feature of the process is essential for process scale-up and intensification, and has been obtained by maintaining the feed concentration, temperature, and the liquid level in the system.

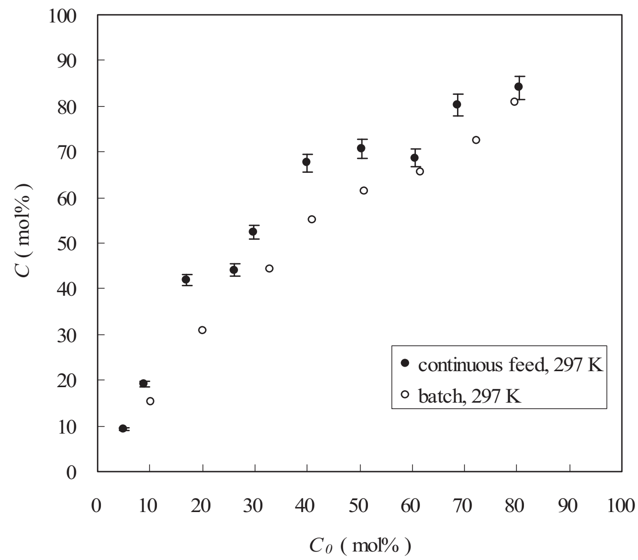

Figure 8. Ultrasonic separation characteristics of ethanol-water in the continuous feed system.

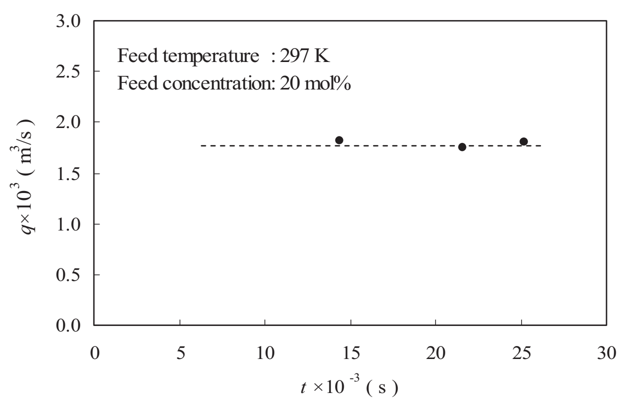

Figure 9. Ethanol collection rate in the continuous feed system.

3.4 Effect of the Temperature in the Ultrasonic Jet on Ethanol Separation

During sonication, the ambient temperature of the liquid in the column was maintained constant, however, localized hot spots were generated due to adiabatic collapse of cavitation bubbles in the jet, and conversion of acoustic energy into heat raises the temperature of the fountain jet formed at the surface of the liquid. The axial profile of temperature in a jet caused by ultrasonic irradiation in a continuous feed system is shown in Figs. 10 and 11. To study the role of temperature on the physical process, ethanol-water mixtures at different concentrations and 297 K were sonicated in an open Petri dish, 0.05 m in diameter, at three different liquid levels of 0.014 m, 0.035 m, and 0.06 m, which were maintained constant.

As shown in Fig. 10, the temperature in the fountain jet increases with distance from the surface of the transducer y. The temperature gradient dT/dy in the jet was found to be

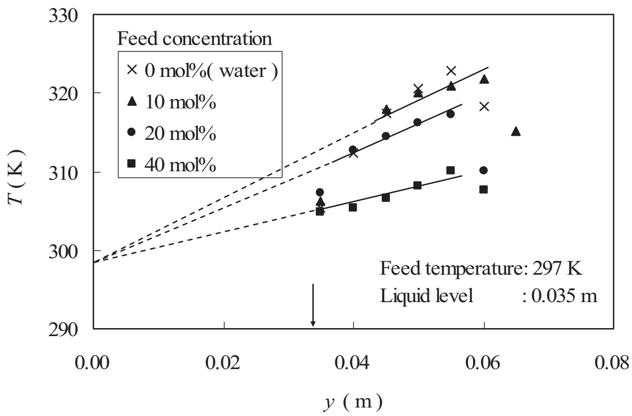

Figure 10. Effect of ethanol concentration on the jet temperature in the continuous feed system.

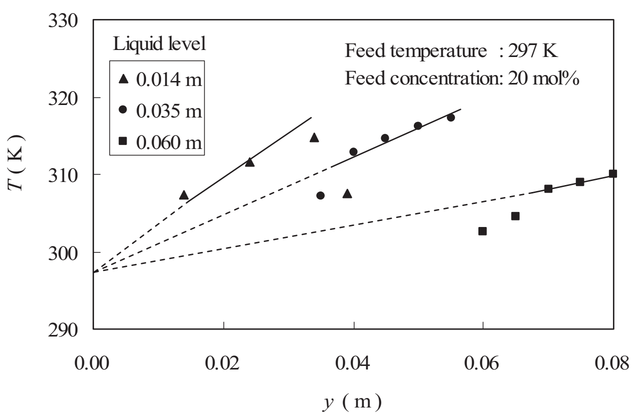

Figure 11. Effect of liquid level on the jet temperature in the continuous feed system.

dependent on the ethanol concentration of the feed solution, and it decreased with increasing ethanol concentration in the feed mixture. In the batch system it was expected that the temperature in the fountain jet would increase leading to a thermal gradient along the jet. Increased droplet vaporization is also anticipated in the system due to the change in thermal profile along the jet. As a result, in the batch system the separation behavior has a higher dependence on temperature than in the continuous process. This explains the difference in the ethanol concentrations of the condensate between the batch and continuous systems.

Fig. 11 shows the effect of the liquid level on the axial profile of the temperature in the fountain jet. When the ethanol concentration was constant, the temperature gradient in the jet decreased with increasing liquid level, and the change in jet height as a result of liquid level above the transducer is illustrated in Fig. 12. The fountain jet height H_j obviously reduces as a result of an increase in liquid level at constant ultrasound intensity. From Figs. 10, 11, and 12 the liquid level above the transducer influences the temperature gradient in the jet proportionately, while causing a decrease in the height of the jet. From these results, it has also been determined that an increase in the initial feed liquid level

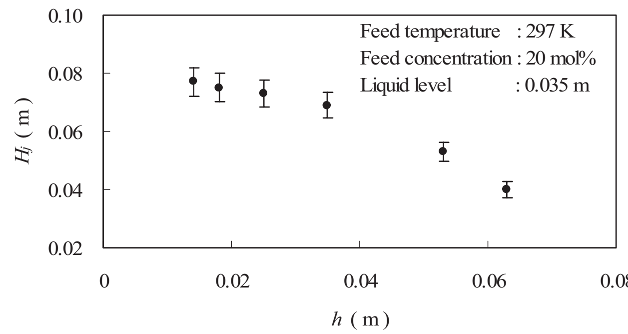

Figure 12. Effect of liquid level on the height of the ultrasonic jet.

increases the condensate concentration in the batch system, as shown in Fig. 6. Hence we can conclude that the liquid level above the transducer has a significant effect on the ethanol separation characteristics described in this work.

4 Conclusions

This study was conducted for further development of a novel enrichment process for ethanol from ethanol-water mixtures using high frequency ultrasonic atomization. The separation characteristics in a newly designed, internally cooled batch system and continuous process feed system by direct collection of the condensate have been determined. The ethanol concentration of the atomized mist formed was found to be dependent on the concentration and the level of feed above the atomizer. Changes in feed properties combined with batch or continuous operation influenced the temperature profile in the ultrasonic jet formed. The temperature in the jet shows that the combination of acoustic cavitation and capillary breakup of the jet can influence droplet mist generation. In the continuous feed system, the ethanol concentration of the mist and the ethanol collection rate were significantly increased and maintained constant due to low variation in the ethanol feed concentration and constant liquid level in the separation vessel.

This process development has demonstrated the application of ultrasound in the separation of miscible liquid mixtures and identified physical parameters that need to be further studied. An understanding of droplet size control and uniform mist concentration from miscible liquids also has potential applications in multiphase reactions such as aero sol-gel synthesis of nano-composites [9, 10].

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Symbols used

C	[mol.-%]	ethanol concentration in condensate mist
C_0	[mol.-%]	ethanol concentration in initial feed solution
H_j	[m]	jet height above liquid level
h	[m]	liquid height from the surface of a transducer
q	[m^3/s]	ethanol condensate collection rate
T	[K]	liquid temperature
t	[s]	time
y	[m]	axial distance from the surface of a transducer

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