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A revisit to the separation of a binary mixture of ethanol–water using ultrasonic distillation as a separation process

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ABSTRACT

Ethanol separation from binary ethanol–water mixture by utilizing "ultrasonic atomization" or 'ultrasonic distillation' has been investigated and inferred as a case of evaporation. It was assumed that the operation of ultrasonic transducer reveals itself as mechanical agitation where the ultrasonic energy is ultimately regarded as heat input into the separation unit. Thus the local deviations from non-equilibrium owing to the propagation of ultrasonic waves through the bulk liquid were excluded from consideration. The process is accompanied by an enlargement of total vapor–liquid interfacial area due to the generation of atomized mist droplets that are supposed to have the same composition as that of bulk liquid. It contradicts with the previous concept of 'ultrasonic distillation' where the mist droplets were characterized by a higher percentage of volatile (ethanol) fraction. Consequently, this study demonstrates that ethanol enrichment process reported earlier might still be assessed by assuming that initial mist droplets have the same composition as that of the bulk liquid mixture. Thus, either by ultrasonic distillation or by bubbling carrier gas through the bulk liquid or even blowing it over the surface of the liquid, the conversion of liquid into vapour phase occurs and could be interpreted as equivalent to evaporation phenomena.

1. Introduction

Conventional distillation process is associated with significant energy consumption [1] and consequently alternative separation technologies that promise to reduce the energy consumption attract the interests of researchers and practitioners. In recent times, a new method of separation of ethanol from water has been claimed [2–4] using what is referred to as "ultrasonic distillation". In this, the vapour and the mist were produced by employing high frequency ultrasound to water–ethanol mixture which were then carried away by a stream of air. This process entails the production of vapour and mist by employing high frequency ultrasound to water–ethanol mixture, subsequently carried away by a stream of air. Mist generation with the impact of ultrasound on water layer was first described by Loomis and Woods in 1927 [5]. Subsequently, researchers focussed on the enrichment of ethanol with the impact of ultrasound [2–8] and different possible mechanisms for the formation of mist have been discussed [2,6,7,9].

In the quest of identifying a mechanism for the formation of mist, Matsuura et al [2] attempted to explain in terms of parametric decay instability of the capillary wave formed during ultrasonication. Toll and Kirpalani [6] suggested an alternative mechanism for the ultrasonic atomization based on the conjunction theory. This involves the formation of cavitating bubbles in the liquid during sonication and their eventual collapse at the liquid surface generates a cloud of microbubbles. These bubbles then move toward the liquid surface with consequent bursting accompanied by the generation of mist droplets. The selective separation of alcohols has been explained as a corollary effect of the physical mechanism that results into the formation of excess of alcohol molecules at the surface of microbubbles due to surface tension. The alcohol molecules vaporize into microbubbles and with the collapse of these microbubbles, the alcohol-rich mist is released.

Series of experiments were performed to analyze the influence of physical parameters such as temperature, carrier gas flow and position of mist collection on the enrichment. Besides, droplet size measurements of the atomized mists and visualization of the oscillating fountain jet formed during ultrasound application were utilised to understand the separation mechanism [2,3,6,7,9]. Moreover, Matsuura et al [3] and Sato and Matsuura [4] extended

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their work further to the surface characteristics of solution using small angle X-rays and practically applied this technology into a prototype rice wine enrichment facility. Matsuura et al [2] have found that pure ethanol solution can be obtained at a lower temperature (10 °C) over a wide range of ethanol concentration and they have postulated that this could be possible by the simultaneous occurrence of both the atomization and vaporization of pure ethanol. The ethanol concentration from the mist was derived through the mass balance equation based on initial and final composition of mother liquid and illustrated on the ethanol separation characteristics curve of ethanol–water solution. However, their outcomes over water/ethanol distillation revealed much less efficiency of separation. Suzuki and Kirpalani [7] conducted experiments using the feed concentrations of ethanol–water as reported by Matsuura et al [2] and stated that the process did not produce pure ethanol at 10 °C. With very accurate experiments Park et al [8] confirmed the separation using ultrasonic distillation and demonstrated that the separation was less complete relative to that obtained by applying sparging agitation.

Based on the interpretation from the experiments performed in a continuous enrichment system, it was reported [2–4] that the mists generated by atomization indicated a higher concentration of ethanol than the feed and the enrichment ratio was even higher than the vapour–liquid equilibrium curve for ethanol–water feed mixture above 10 mol%. Moreover, pure ethanol was obtained from a solution with different mol% of ethanol–water mixture at 10 °C, although no reasoning for such a remarkable effect had been discussed [2]. The promising results observed from the investigations of Matsuura et al [2,3] along with the patenting of a separator by Sato and Matsuura [4] have motivated other researchers to investigate ethanol–water separation using ultrasonic mist generation [6–8].

It is also important to point out that the pioneer work on ultrasonic distillation by Matsuura et al [2] did not show any direct experimental evidence to confirm that the freshly generated droplets of mist are indeed richer in ethanol and no direct analysis of the mist has been performed. Whereas Park et al [8] have shown that the ultrasonic distillation leads to separation but it is somewhat less complete than what is obtained using sparging which lead to the hypothesis that nascent mists have the same concentration as that of bulk liquid but changes due to evaporation of ethanol during the process. Douguchi et al [10] and Matsuura et al [11] employed small angle X-ray scattering measurements during the ultrasonic atomisation of ethanol–water mixtures and detected 1 nm sized ethanol droplets in an environment saturated with ethanol vapour. It is important to note that these nano ethanol droplets should not affect the entire separation process corroborated by the findings from Suzuki and Kirpalani [7] and Bando et al [12], confirming that the major part of the mist consists of droplets having the size of few μm.

Summarising the above we witness that the mechanism of ultrasonic mist formation, which is proposed for the 'ultrasonic distillation' process is not completely understood and still it is the subject of intensive investigation. Looking at the above, our present investigation might help to assess the claims of 'ultrasonic distillation'.

2. Experimental

In this investigation we were looking into whether we can consider the already examined 'ultrasonic distillation' presumably as an evaporation (or humidification) process. We assume that the ultrasonic transducer does mechanical work through vibrations to generate the mist and thereby triggers an enlargement of the interfacial area. This is well supported by the studies [13,14] where it has been observed that the sonication of reaction mixture does not change the chemistry of the process and the beneficial effect is only of physical nature. In other words, in this investigation we have attempted to illustrate whether (1) the results of ethanol enrichment as reported earlier [2,3,6,7] might still be obtained under the assumption that the generated droplets have the same composition as that of the bulk liquid mixture; (2) the already observed effect of 'ultrasonic distillation' might be similarly obtained in an airstream flow without using ultrasound but by other means including mechanical agitation and interface enlargement (bubbling, dispersion) processes; (3) the effect of low temperature on the separation of ethanol and the feasibility of 'ultrasonic distillation' is rather questionable, at least on the basis of the existing data.

To accomplish the above objectives, we have performed detailed experiments on ethanol–water mixtures both at ambient and low temperatures in each of the following three cases: (1) by using ultrasonic mist generator, (2) by using air bubbling and (3) by using cooling tower. The experimental setups used in this study have been shown schematically in Fig. 1(a–f). Ideally an unquestionable estimation from the experiments might be obtained by directly measuring the composition of mist droplets 'in-situ'; however such a technique is quite difficult to implement and thus not existing. In order to eliminate the oxidative effects of sonication of primary alcohols as suggested by Kuppa and Moholkar [13], 99% ethanol (Merck Chemicals) and distilled water were used to avoid the unwanted chemical reactions arising from the presence of oxidative salts. Furthermore, it is well established that sonication of alcohol in the presence of oils or transition metals could lead to chemical reactions in the generation of biodiesel [14–16]. Therefore, all the experiments were performed very carefully to avoid contact with any of the stated materials, and all the glasswares were rinsed thoroughly with alcohols before the start of any experiment. Thus, to determine the mist composition, a quite straightforward and a simple technique was attempted. In this technique, cotton was soaked with coalesced mist droplets when it was placed in the outcoming mist/air stream (Fig. 1a). After every 15 min, cotton with the mist droplets was then squeezed to produce the liquid sample to make the measurements with a refractometer. Ambient air was used as the carrier gas with the flow rate of 4–7.5 L/min. Generally, the mist droplets have the capability to change their composition as they move from the liquid surface to cotton, however we might assume that this effect is negligibly small due to near saturation conditions especially at the low flow rates of carrier gas used in the present investigation. Although soaking the cotton seems to be a primitive technique, but it is a more direct method to assess the composition of mist by comparing to the samples obtained by cooling/condensation of the outcoming carrier air stream in the condenser (Fig. 1b). It is to be noted that in the above cases the samples are a mixture of coalesced droplets and condensed vapour. The data on condensation are useful to estimate the efficiency of the recovery of ethanol from the carrier air stream [6,7,17].

From the observations we noted that the reported enrichment of ethanol [2,3] should not be attributed specifically owing to ultrasound. Thus, similar outcome could be achieved when evaporation takes place even without any application of ultrasound but just by using a continuous injection of air through the liquid which causes an enlargement of interfacial contact between the liquid and gas phases. Such auxiliary experiments were also arranged (shown schematically in Fig. 1(c–f) and carried out. As to the ultrasonic based experiments, a commercial ultrasound mist generator (SC6109, 30 W, 2.4 MHz, Federlite Sdn Bhd, Malaysia) was employed which was connected to a carrier gas input and output ports. Atmospheric air having a relative humidity of 80–95% controlled by RH meter entered the system and passed over the surface of ultrasonicated liquid and exited through an output port