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Vapor-liquid equilibrium of ethanol/ethyl acetate mixture in ultrasonic intensified environment

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Abstract−A vapor-liquid equilibrium (VLE) study was conducted on ethanol/ethylacetate mixture as a preliminary step towards developing an ultrasonic-assisted distillation process for separating azeotropic mixtures. The influence of ultrasonic intensity and frequency on the vapor-liquid equilibrium (VLE) of the mixture was examined using a combination of four ultrasonic intensities in range of 100-400 W/cm^2 and three frequencies ranging from 25-68 kHz. The sonication was found to have significant impacts on the VLE of the system as it alters both the relative volatility and azeotrope point, with preference to lower frequency operation. A maximum relative volatility of 2.32 was obtained at an intensity of 300 W/cm^2 and a frequency of 25 kHz coupled with complete elimination of ethanol-ethyl acetate azeotrope. Results from this work were also congruent with some experimental and theoretical works presented in the literature. These findings set a good beginning towards the development of an ultrasonic assisted distillation that is currently in progress.

Keywords: Ultrasonic Wave, Azeotropic Mixtures, Ethyl Acetate/Ethanol, Vapor-liquid Equilibrium, Relative Volatility

INTRODUCTION

Distillation is still a preferred approach in the separation of azeotropic mixtures in chemical and petrochemical industries despite all its associated difficulties. Particularly, separation of azeotropic mixtures by conventional distillation process is either too expensive or impossible to achieve high product purity [1]. Improvements have been proposed by including additional unit operations as in the case of extractive distillation [2-6], azeotropic distillation [7-10] and pressure swing distillation [11-14]. These processes rely on some additional mechanisms, such as the addition of a third component, called a solvent or entrainer, to overcome the azeotropic barrier and modify the relative volatility of the mixture. However, additional distillation columns are required to recover the solvent, leading to an additional cost and higher energy consumption [15]. Alternatively, process intensification and integration techniques, such as thermally coupled distillation columns [16], dividing-wall columns [17], heat-integrated distillation columns [18] and cyclic distillation [19], have been introduced to reduce the energy requirement. In particular, dividing-wall columns have received an increasing attention in the chemical process industry as they can separate more components in a single distillation unit, thereby offering significant energy savings along with substantial capital and space reduction. This technique has also been used in azeotropic separations and extractive distillation [20]. However, this process is marred by high pressure drop and temperature difference caused by the increase in the boiling point [21].

Another new approach is to intensify the process by adding ultrasonic equipment to the distillation system. In this case, acoustic cavitation and the subsequent growth and collapse of bubbles at elevated temperature in microseconds alter the physical properties of the azeotropic mixtures, enhance the mass transfer [22] and heat transfer [23] processes. This has driven interests in applying ultrasonic waves to various separation processes including extraction [24,25], membrane distillation [26] and adsorption [27].

Along the same lines, the potentials of ultrasonic cavitation in enhancing distillation have been recently explored with particular focus on vapor-liquid equilibrium. Although the phenomenon is not well understood, some preliminary results have highlighted its potentials in few occasions. These include the studies involving various vapor-liquid equilibrium systems, including methanol-water [28], methyl tertiary butyl ether/methanol [29] and cyclohexane-benzene mixtures [30]. In all cases, ultrasonic waves were observed to positively change the vapor-liquid equilibrium and alter the relative volatility of azeotropic mixtures.

This paper discusses the effect of sonication phenomena on vapor-liquid equilibrium of another azeotropic system, i.e., ethanol/ethyl acetate (ETOH/ETAC) as a preliminary stage towards the development of a complete distillation process. At this stage, a home designed apparatus equipped with ultrasonic wave generator was used to investigate the effect of microwave frequency and intensity variation on VLE of ETOH/ETAC in which a boiling point difference of only 1.2 °C is used. The results are compared to relevant previous works.

ULTRASONIC SAFETY AND HEALTH

Available data of the potential harmful effects of ultrasound indicates that airborne ultrasonic fields do not appear to be hazardous

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to humans. However, ultrasonic "sickness" appears to be largely psychosomatic, engendered by apprehension or fear of the unknown. Most "awareness" of these processes is due to hearing the "high-audible" components of the noise, not ultrasound. Individuals capable of hearing 20 kHz and above report only a "sensation," rather than discomfort. It should be stressed that the creation of sound waves involves no electromagnetic radiation. The acoustic energy passing through the air is at intensities far lower than those emitted by high-fidelity equipment; there is therefore no reason to fear harm, for example, to a fetus in utero [31].

In mechanical design of ultrasonic equipment processing convertors and cells should be held in elastomeric clamps. Tubing connections to processing cells should always be flexible, both to minimize sound transmission and to avoid interference with the resonant horn. Proper attention to such details will prevent potential annoyance to personnel and complaints about mysterious maladies. Many manufacturers and users of ultrasonic equipment routinely test it in open spaces without acoustic radiation protection; in so doing, they place their workers at risk and are liable for the consequences. Thus, again similarly, anyone working under such conditions must either do something about it or quit, or suffer attendant hearing loss. Unexplained inaudible sound at very high frequencies can cause anxiety and unease, while that at very low frequency can cause depression; understandably, none of these reactions occur when the subject is aware of the situation [32].

MATERIAL AND METHODS

1. Material

Ethanol and ethyl acetate used in this study were supplied by R&M Chemicals (UK). The purity of the chemicals was 99.7 mole% for ethanol and 99.5 mole% for ethyl acetate. These chemicals were used without further purification.

2. Apparatus and Procedure

The apparatus used in the study is shown in Fig. 1. The system consists of distillation flask (500 cm^3), condenser, water bath, ultrasonic wave generator, and thermocouples. This apparatus was designed for a charge binary mixture of 250 cm^3 and can be operated at low or moderate pressure. The distillation flask was immersed in a water bath equipped with an ultrasonic transducer and a heater.

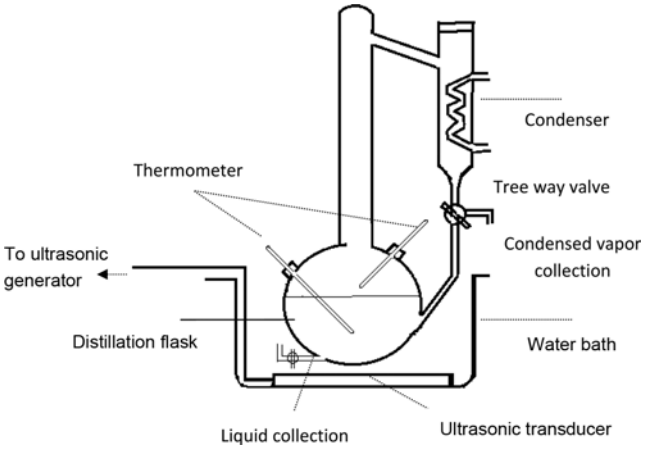

Fig. 1. Schematic diagram of basic ultrasonic distillation unit.

Table 1. Operating conditions of gas chromatograph

Samples analyzed	GC conditions
Ethanol - ethyl acetate	Column: SPB-1, 30 m×0.53 mm ID, 1.0 μm film Oven temperature: 63 °C (6.22 min) to 350 °C at 5 °C/min Carrier gas: Nitrogen 90 psi Detection: Flame ionization detector, 220 °C Injection volume: 1.0 μL, split injection at 220 °C

The ultrasonic transducer, supplied by Crest Ultrasonic Sdn Bhd (Malaysia), was connected to a 500 Watt generator. The system operates at constant temperature maintained by a heater that is equipped with a temperature controller. Vapor and liquid temperatures were measured using thermocouples with uncertainty of ±0.01 °C.

The prepared binary mixture was placed inside the distillation flask heated by water bath. When the liquid mixture is boiling, the vapor produced will ascend and condense forming liquid droplets. The vapor was recirculated until the liquid and vapor temperature remained constant to establish steady-state conditions. Then the condensate was collected through a valve as a distillate sample that represents the vapor-phase composition. Sample taken from the flask was used to represent the liquid phase. The equilibrium temperature corresponding to these two equilibrium compositions was recorded and the product compositions were determined using a gas chromatograph operating at specific conditions as stated in Table 1.

The study was carried out at atmospheric pressure, and the equilibrium data obtained from operations without the presence of ultrasonic waves were used as a reference. To examine the sonication effect, the experiments were repeated using various ultrasonic intensities (100, 200, 300 and 400 W/cm^2) at a fixed frequency of 25 kHz. These were followed by similar experimental studies at two frequencies of 40 kHz and 68 kHz.

RESULTS AND DISCUSSION

1. Baseline Study with Ultrasonic Frequency of 25 kHz

Fig. 2 shows the experimental vapor liquid equilibrium (VLE)

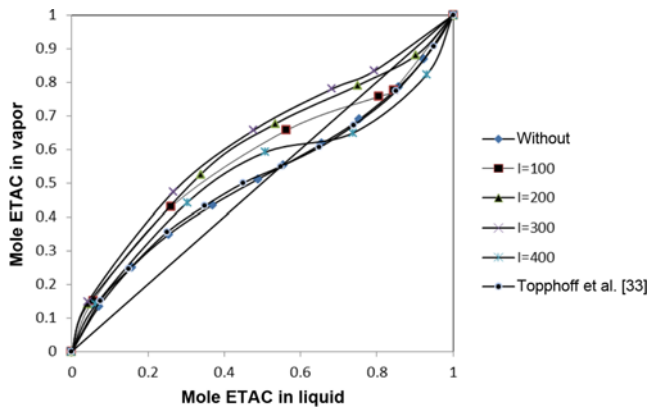

Fig. 2. x–y Diagram of ETOH/ ETAC system without and with different ultrasonic intensities at frequency of 25 kHz.

data of the binary system, ethyl acetate/ethanol at 101.3 kPa. For the base reference (i.e., operation without the use of ultrasonic device), the binary system formed a minimum boiling azeotrope at 72.8 °C and azeotrope point x=0.54 of ethyl acetate. The results obtained in this study agree well with the work in literature [33]. Also note that when the system was operated under the influence of ultrasonic cavitation at a frequency of 25 kHz, the equilibrium curve and azeotropic point of the mixture shifted upwards with the increase in the ultrasonic intensity. This was also true for operation at 200 and 300 W/cm^2, respectively. However, when an intensity of 400 W/cm^2 was used, the situation was reversed and the azeotrope point dropped to x=0.634.

Also, by adding an ultrasonic cavitation effect with the frequency of 25 kHz and intensity of 300 W/cm^2, the azeotropic point of ETOH/ ETAC was completely eliminated. This observation is significant because by eliminating the azeotrope, high purity distillation became possible in a single column. Moreover, by introducing sonication to the system, higher relative volatilities (α) were obtained. The maximum value of α obtained was 2.32, obtained at an ultrasonic intensity of 300 W/cm^2, while the lowest α of 1.33 was obtained at an intensity of 400 W/cm^2.

The phenomena of azeotropic points of the mixture shifted upwards from the standard curve with present of ultrasound waves can also be clearly visualized by Fig. 3, which shows the values of azeotrope point (i.e., showing the composition of ethyl acetate) and the relative volatility against the intensity of ultrasonic used. Since relative volatility is a value derived from equilibrium composition, its variation with changing ultrasonic intensity follows the same trend of azeotrope point with the ethyl acetate composition. This is illustrated by relative volatility values of ethyl acetate which were determined by the following equation:

$$\alpha_{12}=\frac{y_1/x_1}{y_2/y_2} \quad (1)$$

where (α_{12}) is relative volatility of components 1 and 2, (y) and (x) are mole fraction in vapor and liquid phase, respectively.

The changes in the relative volatility and VLE data of the binary mixture are caused by the cavitation activities during transmissions of ultrasonic waves in a liquid medium. When the ultrasonic intensity is increased, greater energy enters the liquid medium, produces micro bubbles, coupled with creation of vacuum effects inside the liquid. Since sonication is a fast transient process that occurs in microseconds, during this short period, heat and mass transfer processes are very rapid. Although it does not give significant net changes in the operating conditions of the distillation process, it does impact the thermodynamics significantly, by altering the vapor-liquid equilibrium of the system. However, since higher ultrasonic intensity produces more bubbles, there will be a limit at which bubble population starts to perturb the cavitation process. The results show that this value was near 300 W/cm^2. At this condition, the tendency of cavitation bubbles to collide is higher. However, since the time available for the bubbles to collapse is insufficient, these bubbles combine to form a bubble 'cushion' at the radiating face of the ultrasonic transducer, which in turn reduces the effect of coupling sound energy to the liquid system [34]. This phenomenon reduces the transmission of ultrasonic energy into the liquid medium and produces less cavitational and vacuum effect. Thus, the increase of ultrasonic intensity beyond 300 W/cm^2 decreased the relative volatility and azeotropic point of ETOH/ETAC and shifted the equilibrium curves towards the reference line.

2. Effect of Sonication Frequency and Intensity on VLE

The previous section illustrated the effect of sonication in shifting the VLE conditions. To better understand this phenomenon, the

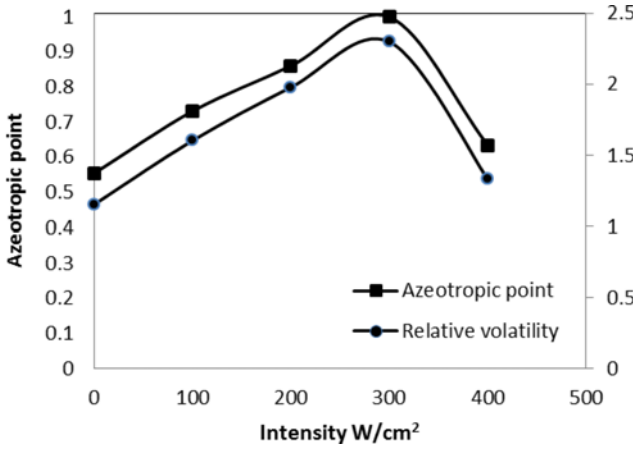

Fig. 3. Azeotropic point and relative volatility of ETOH/ ETAC as a function of ultrasonic intensity at frequency of 25 kHz.

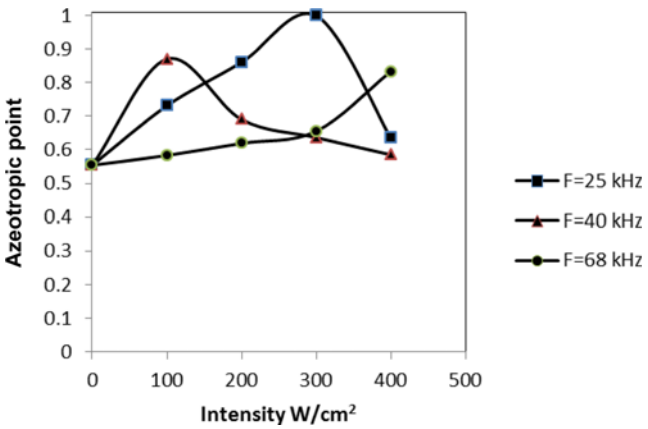

Fig. 4. Azeotropic point of ETOH/ ETAC as a function of ultrasonic intensity and frequency.

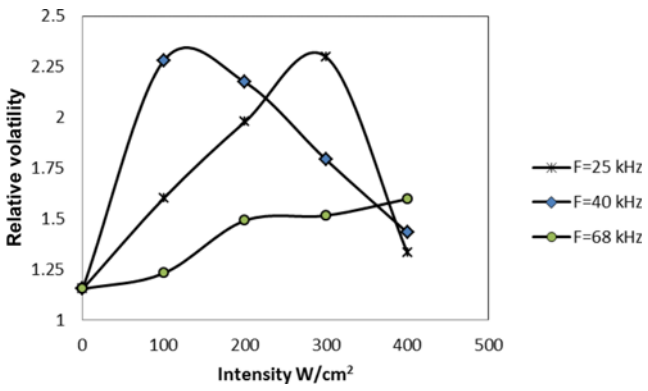

Fig. 5. Relative volatility of ETOH/ ETAC as a function of ultrasonic intensity and frequency.

effect of sonication frequency and intensity on VLE was investigated. Figs. 4 and 5 show the plot of azeotropic point and relative volatility versus the intensity. In all cases, the sonication intensity was found to have a strong effect on the azeotropic point and hence the relative volatility. Considering the results obtained for experiments at 25 and 40 kHz, similar trends are noted. As the intensity was increased, the azeotrope point and relative volatility approached high purity. Nevertheless, after certain values (300 W/cm^2 for 25 kHz and 100 W/cm^2 for 40 kHz) the trend was reversed. This observation is perhaps, best explained by the phenomena described by Mason and Phillip [34]. Further increase in the intensity would result in poorer separation potentials.

Figs. 4 and 5 illustrate that the characteristics of the VLE vary with respect to both frequency and intensity. For the first two cases, i.e., 25 and 40 kHz, the patterns are similar, but with a shift to the left for higher frequency. Nevertheless, when a higher frequency, i.e., 68 kHz, was applied, the relationships between the azeotrope point and relative volatility, and the intensity differed significantly. The phenomenon of generation of cavitation bubbles can be explained by the earlier findings by Luque de Castro and Priego-Capote [35]. These authors concluded that at high sonication frequency, the time required to create bubbles may exceed that of the rarefaction cycle. At this condition, the overall bubble population is reduced due to higher cavitation rate compared to bubble production. Therefore, at this value and beyond, the sonication intensity must be increased with increasing frequency to create more bubbles, thus balancing the effect of frequency on azeotrope point and relative volatility. For this reason, frequencies in the range 20-50 kHz have traditionally been used for separation purposes [36].

3. Maximum Azeotropic Point

The above findings revealed that there is a need for finding optimum operating conditions to establish optimum separation. Within the limited data available from this experimental work, the trend can be observed in Table 2 and the results show that a high purity separation is possible when a frequency of 25 kHz and intensity is used. This was established when the intensity was fixed at 300 W/cm^2.

As discussed before, the presence of ultrasonic waves changes the VLE data and the relative volatility through the cavitation phenomenon, which involves formations of micro bubbles and generation of a vacuum environment in the liquid medium during rarefaction cycle of ultrasonic waves. At higher frequencies, the rarefaction cycle is shortened, leading to the formation of smaller cavitation micro bubbles and generation of smaller vacuum effects within the liquid medium, and ultimately reduced vaporization of the more volatile component in the liquid mixture [34]. Thus, it is important to note that in Table 2 the highest relative volatility was obtained at lowest frequency and vice versa. Similar trend was observed for the azeotropic point.

Table 2. Maximum Azeotropic point and maximum avarge relative volatility of ETOH/ETAC with different ultrasonic frequencies and intensity

Intensity	Frequency	Azeotropic point	Average relative volatility
300	25	1.0	2.32
100	40	0.87	2.28
400	68	0.84	2.26

Generally, the mass transfer of an individual component from the bulk liquid into cavitation micro-bubbles is more affected by its polarity, and a component with lower polarity will dominate the mass transfer processes [37]. Ethanol and ethyl acetate have very close boiling points, and for cases like this, polarity effect plays a significant role, thus impacting the vaporization process. Since ethanol has a higher polarity index compared to ethyl acetate, the latter will dominate the mass transfer processes from the liquid bulk into cavitation micro-bubbles and during the bubble collapse, ethyl acetate is released and enriched in the vapor phase.

The introduction of ultrasound wave to liquid medium causes the molecules to oscillate about their mean position. During the compression cycle, the distance between molecules decreases due to high pressure, and the reverse occurs during rarefaction. If sufficiently large negative pressure is applied to the liquid (the acoustic pressure on rarefaction), the average distance between the molecules exceeds the critical molecular distance, thus facilitating the separation of liquid molecules away from one another [34]. Note that pressure has significant effect on activity coefficients, which in turn has strong effect on the local compositions of molecules within the vicinity of the cyclical pressure environment caused by sonication, as illustrated by equilibrium equations. Further insight on this issue is also provided in the work of Skolnik [41]. Nevertheless, ultrasound has no direct action on chemical bonds involved in the molecules of the liquid through which it travels [38]. Such attempts would require an additional factor such the use of catalyst to initiate chemical reactions.

4. Effect of Surface Tension and Viscosity

The minimum acoustic intensity (or acoustic pressure amplitude) required to produce cavitation is called the cavitation threshold or alternatively, the threshold intensity (or pressure). According to Mason and Phillip [34], surface tension and viscosity are important parameters influencing cavitation formations. Eq. (2) shows the effects of surface tension and viscosity on equation of bubble dynamics [39].

$$R\frac{d^2R}{dt^2}+\frac{3}{2}\left(\frac{dR}{dt}\right)^2=\frac{1}{\rho}\left[P_i-P_\infty-\frac{2\sigma}{R}-\frac{4\mu}{R}\left(\frac{dR}{dt}\right)\right] \quad (2)$$

where R is the radius of the bubble, ρ is the density of the liquid, P is the pressure in the liquid at infinity as function of time t, P_i is the pressure in the bubble and P(R) is the pressure in the liquid at the bubble boundary. The surface tension constant and the coefficient of viscosity are σ and μ, respectively.

This equation is widely known as the Rayleigh-Plesset equation, which represents the relationships between important variables that describe cavitation, bubble dynamics and the collapse of the cavity in a large mass of liquid. Employing solvent with low surface tensions and/or low viscosity would lead to a reduction in cavitation threshold, thus leading to lower energy requirement to produce cavitation bubbles [31]. Indeed, the only surface tension and viscous contribution to the Rayleigh-Plesset Eq. (2) comes from the dynamic boundary condition at the bubble surface. Therefore, the corresponding effect of the mixture density can be negligible compared with the large density ratio between the liquid and the gas inside the bubble [40].

The influence of surface tension and viscosity on the azeotrope

Table 3. Effect of surface tension and viscosity: comparison this work with published works [28,29] at frequency 40 kHz

Physical properties	Intensity=50 W/Cm^2 in reference (28)	Intensity=100 W/Cm^2 in this work	Intensity=200 W/Cm^2 in reference (29)
System	MTBE/Methanol	ETOH/ETAC	Cyclohexane/Benzene
Surface tension @ 20 °C (dyn/Cm)	20	23	26
Viscosity @ 25 °C (CP)	0.407	0.756	0.814
Density @ 20 °C (g/cm^3)	0.78	0.84	0.83
Relative volatility	2.06	2.28	2.158
Azeotrope point	0.86	0.87	0.85

point is illustrated by the results in Table 3. In this case, results from this work (i.e., ETOH/ETAC) are compared to those involving MTBE/Methanol [28] and Cyclohexane/Benzene [29], all operating at ultrasonic frequency of 40 kHz, operating temperature of 25 °C, and a hydrostatic pressure of 1 atm. The results shown were obtained from the maximum azeotropic concentration, which corresponds to maximum relative volatility obtained for all systems at the selected frequency (i.e., 40 kHz). It can be seen from the data tabulated that a higher ultrasonic power is required to induce cavitation formation in liquid medium with higher viscosity and/or higher surface tensions [24]. The optimum value for each system would be dependent on the frequency used.

In addition, trends of responses obtained at 40 kHz are as shown in Figs. 3 and 4 are also similar to those obtained in previous works [28,29].

CONCLUSIONS

This paper has illustrated the effect of sonication on the VLE of an azeotropic ETOH/ETAC mixture. It has shown that azeotrope point can be shifted to the advantage of the processing requirement by selecting suitable combinations of frequency and intensity of the ultrasonic transducer, thus providing potential solution to distillation of azeotropic mixtures. For example, a relative volatility of 2.32 was obtained at a frequency of 25 kHz and intensity of 300 W/cm^2 for the EtOH/EtAc mixture considered in this study. At this operating condition, the azeotrope phenomenon was completely eliminated.

The results of this study also revealed the complex nature of the relationships between relative volatility with the sonication frequency and intensity, and the data obtained are consistent with findings of other workers. The findings of this study are in agreement with some published works, and are consistent with fundamental theory. It can be concluded that a lower frequency (25 kHz) is preferred, and by choosing the right operating condition, azeotropic mixtures can be separated in a single column. This study also suggests that cavitation can be considered as one of the methods of introducing discrete energy input, and the cavitation phenomenon offers a novel means for intensification of a variety of physical/chemical transformations including chemical synthesis and chemical processes.

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