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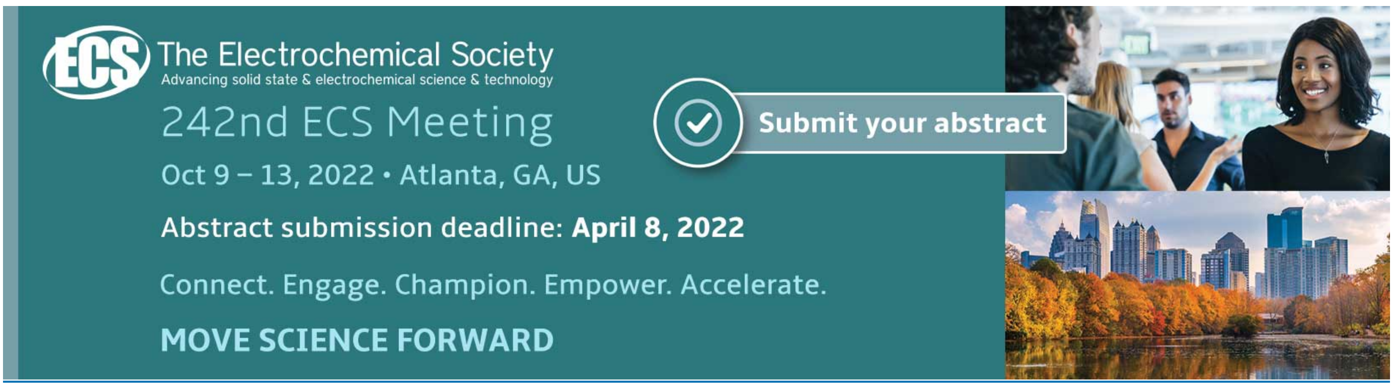

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Experimental Study of the Characteristics of Acoustic Cavitation Bubbles Under the Influence of Ultrasonic Wave

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Abstract. Sonochemistry application in various fields, especially separation industries, are constantly increasing and extensively studied in recent years. In the applications, a detailed understanding of the characteristics and behaviour of acoustic cavitation bubble is crucial. However, acoustic bubbles behave differently in each liquid. Owing to numerous amounts of acoustic bubbles showing dramatic transition during sonication and have complex interaction in a liquid mixture, it is more difficult to observe acoustic bubble consistently and thoroughly. In response to this problem, our study will investigate separately acoustic cavitation bubble form in sonicated water, ethanol and mixture of ethanol-water. Each liquid was sonicated at 20 kHz ultrasound with a transducer attached to the bottom of an acrylic vessel. In order to visualise acoustic bubbles clearly, a high-speed video camera is used to capture the image of acoustic bubbles. Image processing and analysis are by the application of Image Processing Toolbox in MATLAB R2018b. Experimental result reveals that acoustic cavitation bubbles in a mixture of ethanol-water have the largest radius, volume and higher buoyancy force compared to pure ethanol and water. Results obtained coincided well with theory, which indicated that sonicated mixture containing alcohol provide resistance to gas transfer across liquid/bubble interface, leading to an increased bubble radius and hence the buoyancy force.

1. Introduction

Cavitation is a phenomenon of the generation of bubbles due to the decrease in liquid pressure with a consecutive violent collapse of each bubble [1]. These involve bubbles formation, growth, pulsation and collapse of bubbles in liquid during propagation of the sound wave. Chemical effects of ultrasound such as production of free radical and acceleration of a reaction originate in the extreme local condition of collapse bubble has been used in various industries [2]. For many years, the use of ultrasound energy in chemical process industries has developed a growing interest in replacing the conventional method [3] that used a massive amount of energy consumption. Recently, a new approach to the separation process of ethanol-water called "ultrasonic distillation" has been investigated. Many researchers investigated the separation of ethanol from the aqueous solution by ultrasound. It was reported that the application of ultrasonic enhance the total area of vapor-liquid interface and can be served as an input source of thermal energy required for vaporisation [4]. Research done by Yasuda [5] reported that the utilisation of ultrasonic atomization shows an increasing number of ethanol generation rate droplets with increasing ethanol solution. Their works were in alignment with Wakisaka [6] proving that there is a close relationship between evaporation properties in bubbles and cluster-level structure in alcohol-water binary mixtures.

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Based from all the results and the data interpretations from researchers, it is plausible such "ultrasonic distillation" separation technique are feasible. However, separation mechanism in terms of acoustic bubble characteristic is not fully elucidated due to difficulties in characterizing the bubbles in a mixture. In this work, the characteristics of acoustic cavitation bubbles in terms of radius, volume, movement and buoyancy force under the influence of ultrasonic sound wave at 20kHz is examined. To investigate the characteristics of acoustic cavitation bubble, a detailed experimental work of three different liquids with distinctly different properties – water, ethanol and ethanol-water mixture were conducted. Acoustic cavitation bubbles were observed using a high-speed video camera. The results obtained were analysed using MATLAB R2018b, and the results from each liquid were compared thoroughly. Because separation of azeotropic mixtures occurs inside the acoustic bubbles, results of this study may be useful in the separation of azeotropic mixture by ultrasonic distillation, which is the next focus of process intensification in chemical separation industry [7].

2. Experimental Setup

Figure 1 shows the experimental setup for the generation and visualisation of the acoustic cavitation bubble. For bubble generation, ultrasound with a frequency of 20kHz sine wave was supplied by a function generator. The signal was then supplied to a bolt-clamped Langevin-type transducer. The transducer was bonded to a 3 mm thick 316 stainless-steel plate that was located at the bottom of rectangular sono-reactor. The reactor was made from acrylic, and the inner dimensions were 70 mm x 70 mm with 200 mm height. The liquid in the reactor was in direct contact with a stainless-steel plate. Non-degassed water at 20 ^{o}C was filled into the vessel up to 80mm height. A sound pressure field was developed and grew in the reactor by the ultrasound irradiated from the bottom of the reactor. In order to visualise the acoustic cavitation bubbles, we used a high-speed video camera and a flat LED light, mounted facing each other. The setting of the camera was as follows: image size 1024 x 1024 pixels, frame rate 500 fps, exposure time 5s, and spatial resolution 49.3 um/pixel. The recorded images were processed with MATLAB R2018b. To determine the characteristic of an acoustic cavitation bubble, we investigated bubbles formation in three different liquids which are, non-degassed water, pure ethanol and a mixture of ethanol-water 50:50 by v/v. Table 1 shows the properties of each liquid used in the experiment.

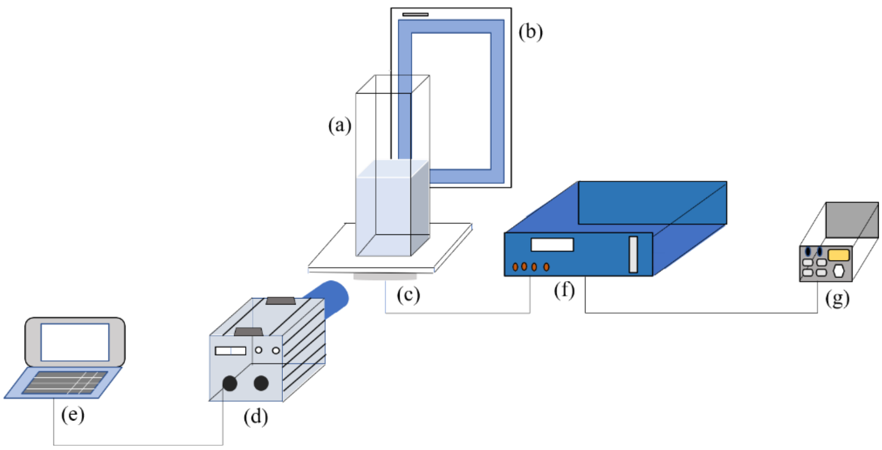

Figure 1. Experimental setup for the visualization of acoustic cavitation bubble. (a) Acrylic reactor (b) LED light (c) Transducer (d) High-speed video camera (e) Computer (f) Amplifier (g) Function generator

Table 1. Liquid properties for water and ethanol

	Water	Ethanol
Temperature (°C)	20	20
Density, ρ (kg/m^3)	1000	785
Dynamic viscosity, μ (Pa s)	1.0 x 10^{-3}	1.1 x 10^{-3}
Speed of sound, c (m/s)	1482	1100
Surface tension, σ (Nm^{-1})	0.079	0.022
Vapor pressure, p_v (Pa)	2.2 x 10^3	5.3 x 10^3
Thermal conductivity, Ķ (Wm^{-1}K^{-1})	0.58	0.014

3. Results and Discussion

3.1. Visualizations of acoustic cavitation bubbles

Growth of bubble in a system involves two processes, namely rectified diffusion which involves gas diffusion into a bubble and bubble coalescence where bubbles coalesce each other if the number concentration of bubbles is large enough [8]. Bubble coalescence can have a significant influence on the population and size of acoustic bubble [9,10]. Figure 2 shows the differences between acoustic cavitation bubbles formed in each liquid. When non-degassed water is sonicated, a few visible bubbles are readily observed as in Figure 2(a). Acoustic bubble population has shown to increase and move vigorously towards the liquid surface as the liquid change from non-degassed water to ethanol (Figure 2(b)) and mixture of ethanol-water (Figure 2(b)). This can be attributed to mainly the coalescence of bubbles to form more bubbles as the rate of growth is faster in comparison to non-degassed water. During sonication, ultrasound generate forces that are capable to drive bubble into contact and increase the frequency of coalescence. This explains the readily observed bubbles when liquid is sonicated. In liquid containing alcohol, bubble coalescence is much more complicated to evaluate. To the author best knowledge, only a few experimental studies concerning acoustic bubble in alcohol have been investigated [11,12,13]. Results obtained are only limited to the generation of cavitation bubble development and research found that alcohol when sonicated provide resistance towards gas transfer process at acoustic bubble/liquid interface. We will discuss these observations more in detail in the following section.

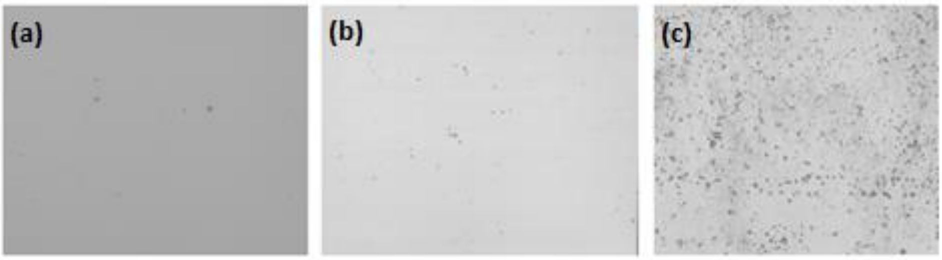

Figure 2. Image of acoustic cavitation bubbles under 20kHz ultrasound, (a) Non-degassed water, (b) Pure ethanol, (c) Ethanol-water mixture

3.1.1. Non-degassed water. Figure 3 shows the movement of the acoustic cavitation bubble in non-degassed water oscillating under acoustic frequency of 20 kHz. The bubbles in figure 3 are more likely to be driven towards the center of sono-reactor. Bubble located at the bottom of frame 1 move slightly upward at frame 2 and remains at almost constant position in frame 3 before moving downward again in frame 4. Movement of bubbles influence each other in a complex way and are related to the growth of the bubble. Under ultrasonic wave, there are two acoustic force that are responsible to drive bubbles towards each other, which are primary Bjerknes forces and secondary Bjerknes forces. Bubbles traveling to the center of sono-reactor are smaller than its resonant radius and directed toward a pressure antinode in a standing wave as shown in Figure 3, where it is a localized region in the liquid volume. Bubbles will continue to expand during compression phase of acoustic cycle and repelled from the antinode. Many experimental works have explained the mechanisms of acoustic cavitation bubble in non-degassed water; thus, we will not explain further on this, but rather focus on bubbles generates in an organic mixture in the following section.

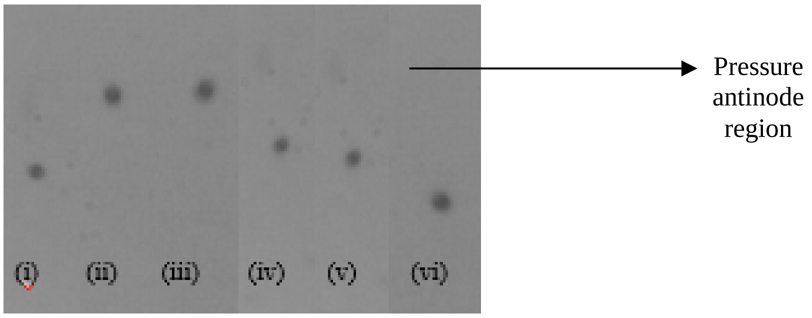

Figure 3. Movement of acoustic cavitation bubble in non-degassed water

3.1.2. Ethanol. Ethanol exhibits a distinct, fast development of cavitation under the influence of ultrasonic wave, making it interesting to discuss. In liquid ethanol, we observed that population of acoustic bubble is more than non-degassed water (figure 2) and generates larger acoustic bubble radius (figure 4). When a bubble is generating from alcohol solution, gas diffusion is strongly retarded. Consequently, lifetime of a bubble becomes much longer and volume of gas increase which is why larger bubble was generated. Sonication towards a high vapor pressure liquid results to a decrease in cavitation collapse thus produce more acoustic cavitation bubbles in sono-reactor. Kenji [14] explained that gas solubilities in organic solvents are much higher compared to water, suggesting that there are different cavitation dynamics occur in the case of organic solvent. A higher vapor content inside the cavitation bubbles leads to more vapor content inside the acoustic bubbles, hence, increases buoyancy force (figure 5).

Figure 6 shows the movement of the bubbles to the liquid surface and the action of buoyancy force when reaching ethanol-air interface. We observed that there are coalescence of small bubbles to form larger bubbles occur during cavitation. Since the gas bubbles can grow large enough, acoustic bubble can leave the system entirely due to buoyancy. Mossaz [15], also in agreement with this, wrote since there are no acoustic bubble streaming, shielding and scattering effect, acoustic bubble movement did not interrupt by ultrasonic sound wave to propagate in liquid. Iwai and Li [16] also reported a lower surface tension in the liquid would decrease the erosion rate of the bubble due to no major collapsing event in it.

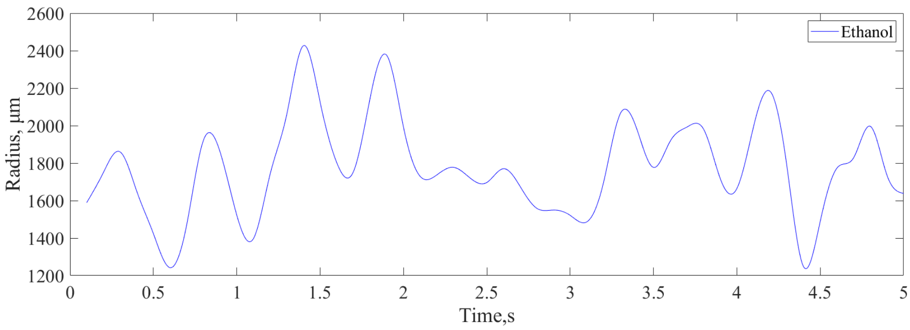

Figure 4. Fluctuation trend of the acoustic bubble radius in ethanol

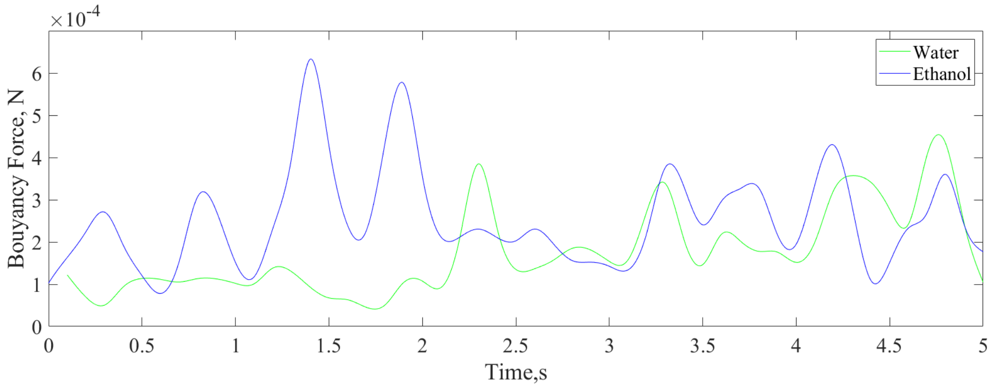

Figure 5. Comparison of acoustic bubble buoyancy force in non-degassed water and ethanol

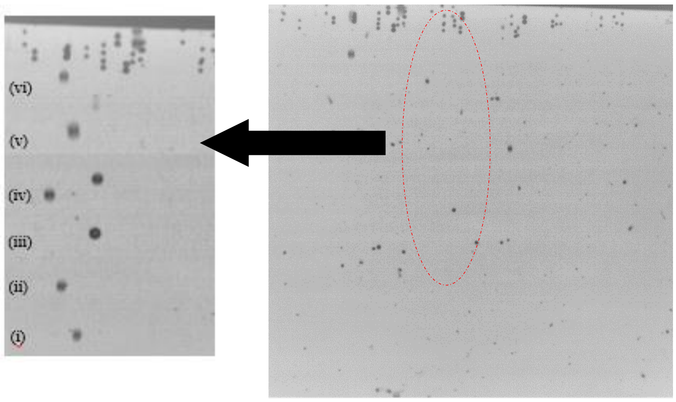

Figure 6. Movement of acoustic bubble in ethanol to liquid-air interface

3.1.3. Ethanol-water solution. Due to the strong demand of its purification from azeotropic mixture, ethanol separation from its aqueous solution is a major subject of discussion in chemical engineering. This section discusses characteristic of acoustic cavitation bubbles in sonicated ethanol-water at 50% concentration. Figure 7 shows time series movement of acoustic bubble in ethanol-water during sonication. We observed that the population of bubbles are much more compare to ethanol and non-degassed water and almost all of the acoustic bubble produced are moving towards the liquid surface as shown in figure 7. The presence of ethanol mixed with water can inhibit the ability of acoustic bubble to coalesce. The inhibition can be explained by the intermolecular interaction with water molecules in ethanol-water mixture. Addition of ethanol in water will break the hydrogen bond network and causes a decrease in surface tension. If alcohol concentration increase, there will be more alcohol rich cluster formed due to gradually ruptured water network in a mixture. Water molecules will losses its hydrogen bond network structure and behave as single-molecule bonded with ethanol molecules [17]. As a result, mixture of ethanol-water has larger volume compare to non-degassed water and ethanol (figure 8).

Alcohol molecules like ethanol, have a hydrophobic nature. Therefore, they adsorb at the liquid/bubble interface. During the expansion of cavitation bubble, adsorption of solute molecule at liquid/bubble interface will evaporate into bubble interior. Molecules will then decompose and accumulate as product thus, reducing bubble temperature when collapse. However, these adsorptions, too, will affect rectified diffusion of gas in the acoustic bubble. Diffusion of gas out of the bubble during contraction will be retarded, causing bubble to be large in volume (figure 8) and lead to higher gas content left in acoustic bubble. Bubbles that become larger may leave the system by buoyancy. Due to the formation of larger bubbles, fluid flow velocity around large microbubbles varies along the reactor (figure 9). Velocity of the fluid inside the reactor is the highest at the center of the sono-reactor. This is due to the presence of localized, high fluid flow created by large cavitation bubble and high -speed acceleration from the propagation of ultrasound through the medium.

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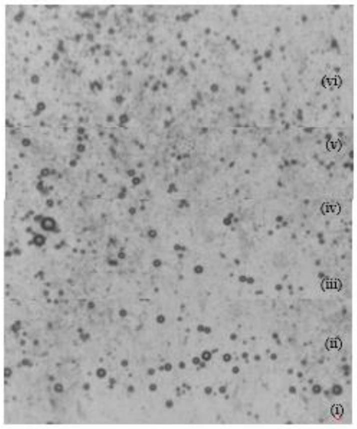

Figure 7. Time series of acoustic bubble movement in ethanol-water mixture

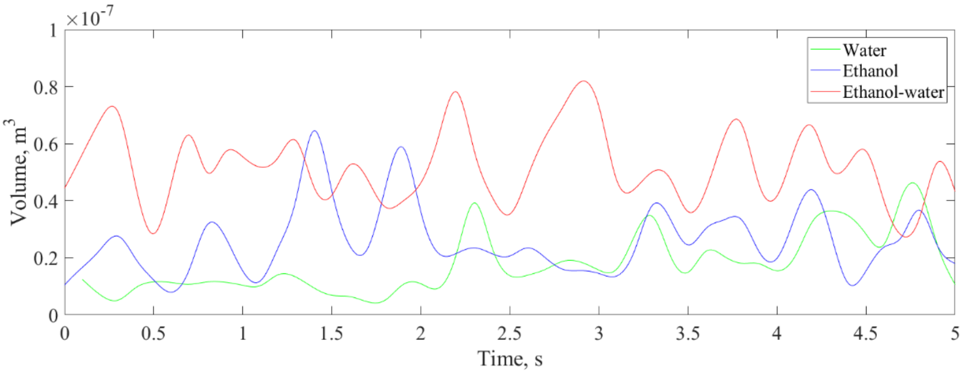

Figure 8. Comparison of acoustic bubble volume in (a) non-degassed water, (b) pure ethanol, (c) mixture of ethanol-water

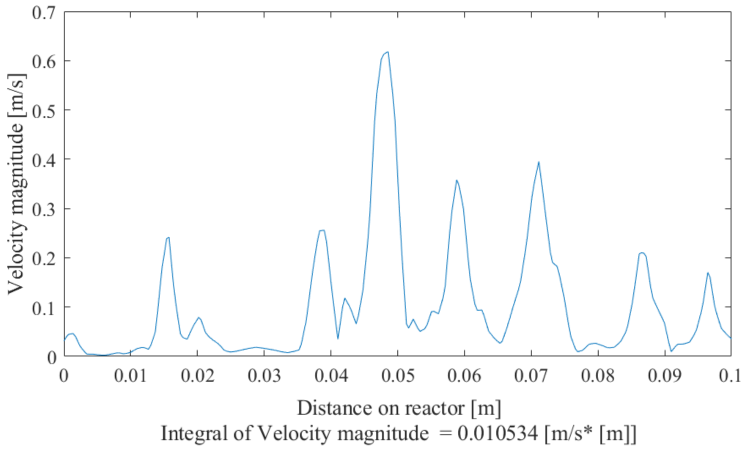

Figure 9. Velocity of fluid along the reactor

4. Conclusion

The formation of acoustic cavitation bubbles and its movement are mainly due to the liquid properties and intensity of ultrasonic sound waves used. In this paper, we studied acoustic cavitation bubbles characteristic in three different liquids which are non-degassed, ethanol and mixture of ethanol-water under a 20kHz ultrasonic sound wave. Results showed that their characteristics are very different due to variations in liquid properties, i.e., surface tension, viscosity, vapour pressure, which play a significant role in the establishment of cavitation bubble. Key findings of the study are:

- The formation of acoustic cavitation in non-degassed water is much less visible compared to the other liquid. There is more acoustic cavitation bubble present in pure ethanol and ethanol-water mixture, and in both of the liquid, acoustic bubbles oscillate vigorously moving towards the liquid-air interface.
- Movement of acoustic bubbles in non-degassed water is primarily due to primary and secondary Bjerknes force. Acoustic bubble experiences less primary Bjerknes Force (attractive force) is repelled from pressure antinode.
- In ethanol, acoustic cavitation bubble can sustain a longer lifespan due to lower surface tension. Surface tension of bubble-gas interface plays a major role in internal bubble pressure.
- Distinctive results in the case of ethanol-water mixture are observed. The addition of ethanol in water provides resistance to gas transfer across liquid/bubble interface, leading to an increased in bubble radius and hence, the buoyancy force.

Results presented by a large area of research, including this paper indicates there might be a possibility of breaking azeotrope using this technique. Due to the complexity in the phenomenon of acoustic cavitation bubble as well as the solution property of ethanol and water mixture, separation mechanism is still under discussion investigation. The results to be uncovered would ascertain whether or not it is feasible to design a system that would consistently breaks azeotropes in organic mixture.

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