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## A R T I C L E S

# Study of the Coalescence of Acoustic Bubbles as a Function of Frequency, Power, and Water-Soluble Additives

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***Abstract:*** The effect that surface-active solutes, such as aliphatic alcohols and sodium dodecyl sulfate (SDS), have on the extent of bubble coalescence in liquids under different sonication conditions has been investigated by measuring the volume change of the solution following a period of sonication. In general, the adsorption of surface-active solutes onto the bubble surface retards bubble coalescence. Within the limitations of the measurement method and the systems studied, bubble coalescence does not appear to be dependent on the applied acoustic power. Also, varying the applied acoustic frequency has a minimal effect on the extent of bubble coalescence in systems where long-range electrostatic repulsion between bubbles, imparted by the adsorbed surface-active solutes, dominates. However, when short-range steric repulsion (or other short-range repulsive forces) is the primary factor in inhibiting bubble coalescence, the dependence on the applied acoustic frequency becomes apparent, with less coalescence inhibition at higher frequencies. It is also concluded that SDS does not reach an equilibrium adsorption level at the bubble/solution interface under the sonication conditions used. On the basis of this conclusion, a method is proposed for estimating nonequilibrium surface excess values for solutes that do not fully equilibrate with the bubble/solution interface during sonication. For the case of SDS in the presence of excess NaCl, the method was further employed to estimate the maximum lifetime of bubbles in a multibubble field. It was concluded that an acoustic bubble in a multibubble field has a finite lifetime, and that this lifetime decreases with increasing applied frequency, ranging from up to $0.35 \pm 0.05$ ms for 213 kHz to $0.10 \pm 0.05$ ms for 1062 kHz. These estimated lifetimes equate to a bubble in a multibubble field undergoing an upper limit of 50–200 oscillations over its lifetime for applied ultrasound frequencies between 200 kHz and 1 MHz.

## Introduction

Ultrasound has a wide range of applications, ranging from the degradation of pollutants,[1–4] polymerization reactions,[5,6] and formation of protein microspheres[7,8] and nanoparticles[9–12] to food science[13,14] and biomedical applications.[15,16] The overall efficiency of these applications depends on the bubble population and sizes, which in turn depend upon various parameters, such as the ultrasonic power and frequency, etc. Our previous study,[17] in which the influence of acoustic power was investigated using sonoluminescence (SL) as a probe, found that the "active" bubble population (i.e., bubbles producing SL) is mainly governed by the degree of bubble clustering leading to "impedance shielding",[18,19] and bubble coalescence.

It is well-known that the extent of bubble coalescence in solution is very much affected by the presence of surface-active solutes.[17,20–22] The adsorption of these solutes on the bubble/solution interface will generally retard the process of bubble coalescence. In addition, the work of Sostaric and Reisz[23,24] concludes that, unlike the case for "static" bubbles in a liquid,

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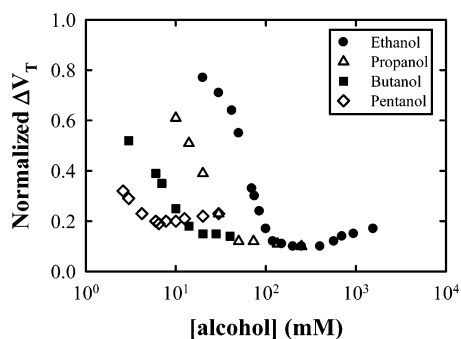
**Figure 1.** Average change in total bubble volume relative to pure water as a function of alcohol concentration in aqueous solutions sonicated at 358 kHz, 0.9 W/cm$^2$.
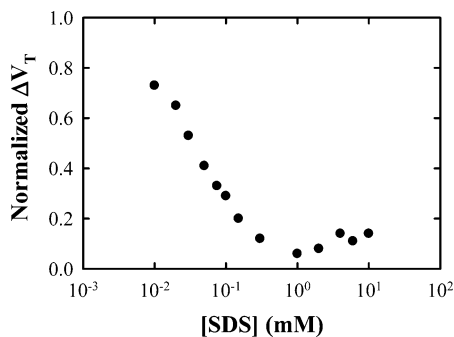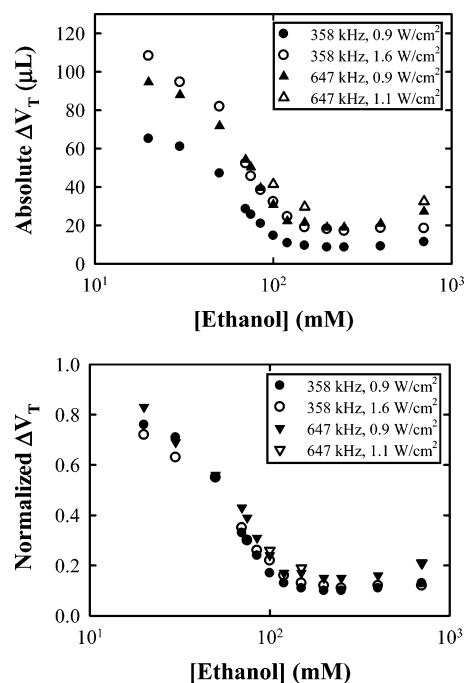


**Figure 2.** Average change in total bubble volume relative to pure water as a function of SDS concentration in aqueous solutions sonicated at 358 kHz, 0.9 W/cm$^2$.

equilibrium adsorption of some surfactants onto the surface of bubbles exposed to ultrasound is not achieved. Considering that surface-active solutes appear in many sonochemical processes, it is imperative that a sound understanding of the behavior of these solutes in the solution, especially how they influence the bubble population and eventually the efficiency of the ultrasonic processes, is acquired. For this reason, the aim of the present study was to further explore the extent of bubble coalescence in solutions containing surface-active solutes under different sonication conditions by following the method adopted by Lee et al.,[21] while at the same time probing the dynamics of surfactants adsorption onto the acoustic bubble/solution interface.

## Results

Figures 1 and 2 show the typical volume changes observed following sonication of aqueous solutions containing a range of alcohols and SDS, respectively. To see the effect of added surface-active solutes, the $\Delta V_T$ values are normalized with respect to the $\Delta V_T$ for pure water under identical conditions, and the concentrations are plotted on a log scale to show the features of the data points more clearly. The $\Delta V_T$ values described in these experiments are considered to be directly related to the amount of bubble coalescence in the liquid:[21] a smaller normalized $\Delta V_T$ indicates a lower level of bubble coalescence. Consistent with the findings from previous studies,[20−22] it is evident that the extent of bubble coalescence is affected by the presence of the surface-active solutes, as suggested by the significant decrease in the $\Delta V_T$. Furthermore, Figure 1 also shows that the longer is the alkyl chain length of the alcohol, the lower is the concentration needed to achieve the same amount of decrease in $\Delta V_T$.
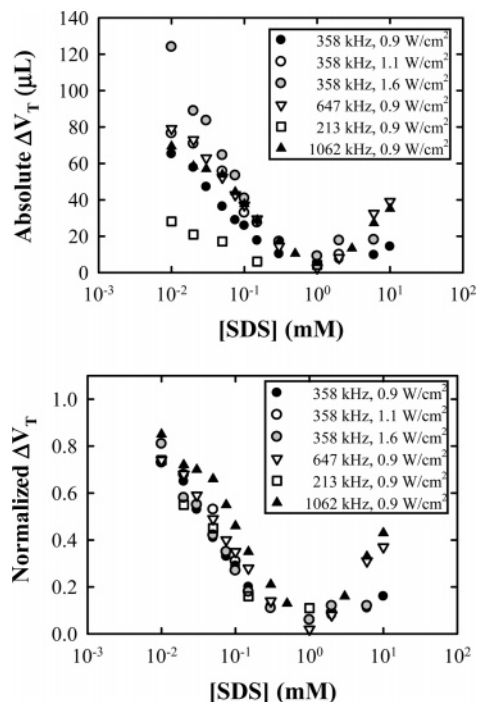
**Figure 3.** Average change in total bubble volume as a function of ethanol concentration in aqueous solutions sonicated at various applied acoustic frequencies and powers expressed as: (a) absolute $\Delta V_T$, (b) $\Delta V_T$ relative to that of pure water under the same conditions.

Sunartio et al.[17] showed that the applied acoustic power can have a marked effect on sonoluminescence intensity. It was argued that this arises from the effect on the bubble population, which is largely controlled by the degree of bubble clustering leading to impedance shielding, and bubble coalescence. To further probe this latter aspect, experiments were conducted at various acoustic powers, and the results are presented in Figures 3−5. Absolute $\Delta V_T$ values are greater for higher applied acoustic powers (Figures 3a−5a). However, it is clearly shown in Figures 3b−5b that once these values are normalized to the $\Delta V_T$ for water, they lie on the same curve (within the experimental error of this technique), indicating that there is no significant power dependence on the processes responsible for $\Delta V_T$ values. In addition, the experiments were also conducted at a few different ultrasonic frequencies (213, 358, 647, and 1062 kHz) using an identical cell design. Minimal frequency dependence was observed for ethanol and lower concentrations of SDS (<2 mM) (Figures 3b and 4b). However, dependence on the applied acoustic frequency is apparent for higher SDS concentrations and SDS in the presence of 0.1 M NaCl (Figures 4b and 5b).

The work of Lee et al.[21] also showed that the addition of an electrolyte may partly counteract the effect of surface-active solutes on bubble coalescence (as will be discussed further in the next section). Figure 6 shows the change in $\Delta V_T$ when increasing amounts of NaCl were added to the ethanol and SDS solutions. It can be readily observed that the $\Delta V_T$ increases and then levels off as the NaCl concentration increases, and that the limiting plateau values are different for each SDS solution.

## Discussion

To guide us through the discussion, it is useful to keep in mind Scheme 1, which depicts the fate of individual bubbles in liquids under ultrasonic irradiation. While the full discussion
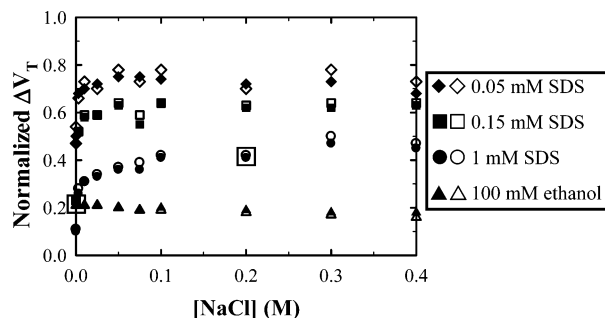
**Figure 4.** Average change in total bubble volume as a function of SDS concentration in aqueous solutions sonicated at various applied acoustic frequencies and powers expressed as: (a) absolute $\Delta V_T$, (b) $\Delta V_T$ relative to that of pure water under the same conditions.
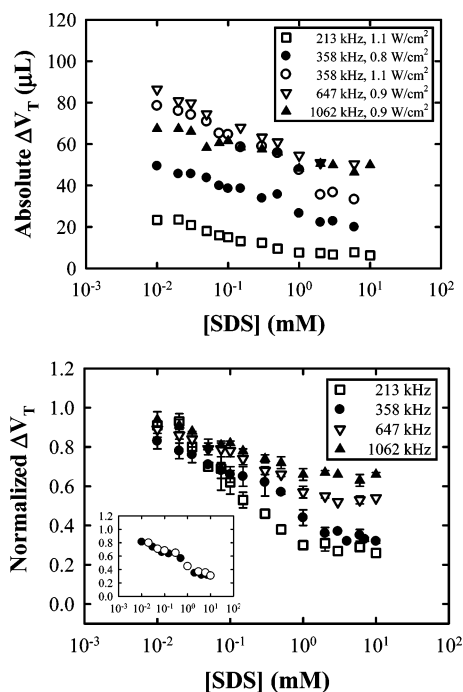


**Figure 5.** Average change in total bubble volume as a function of SDS concentration in aqueous 0.1 M NaCl solutions sonicated at various applied acoustic frequencies and powers expressed as: (a) absolute $\Delta V_T$, (b) $\Delta V_T$ relative to that of pure water under the same conditions. (The inset shows the power comparison for 358 kHz, ● 0.8 W/cm$^2$ and ○ 1.1 W/cm$^2$. All of the data points for the different frequencies in (b) are averages of runs at different powers, with the error bars showing the variation.)

of this scheme has been given in detail elsewhere,[25] the salient features are summarized as follows. As ultrasonic waves pass through a liquid medium, bubble nuclei present in the liquid can grow at the antinodes via rectified diffusion or coalesce with one another to form a larger bubble. Upon coalescence, if



**Figure 6.** Average change in total bubble volume relative to pure water for selected solutions as a function of added NaCl concentration. The aqueous solutions were sonicated at 358 kHz, 0.8 W/cm$^2$ (closed symbols) or 1.6 W/cm$^2$ (open symbols). For the SDS solutions, the relative $\Delta V_T$ increases and then reaches a plateau as the concentration of NaCl added increases. (See text for boxed data points.)

**Scheme 1.** Fate of Individual Bubbles in Liquids under Ultrasonic Irradiation$^a$
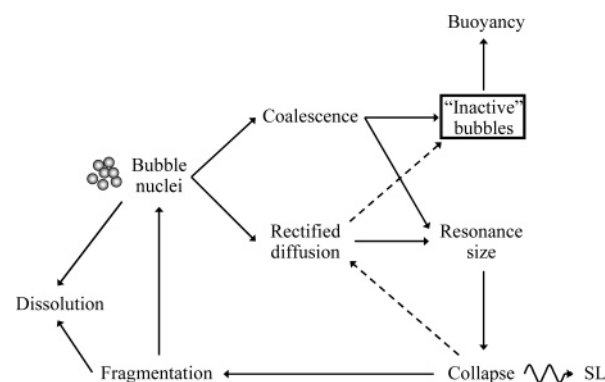


$^a$ Note that "resonance size" here does not necessarily mean the linear resonance radius, but simply the size where bubbles are able to collapse violently and emit SL (Yasui, K. *J. Acoust. Soc. Am.* **2002**, *112*, 1405−141).

the resulting bubble is larger than the resonance size, it will be pushed toward the nodes by primary Bjerknes forces,[26] hence becoming "inactive". These bubbles may eventually float out of the liquid due to buoyancy once the acoustic field is turned off. It is this population of "inactive" bubbles in the nodes that are measured by the $\Delta V_T$ values. On the other hand, if the resulting bubble is still smaller than the resonance size, it can continue to grow via rectified diffusion to reach the resonance size and then collapse violently.

There are two possible pathways that a bubble can undergo after collapse. It can continue to grow further by rectified diffusion following exposure to acoustic cycles, reaching a size larger than the resonance size and then be forced to the node to become "inactive". Alternatively, it can fragment to smaller daughter bubbles that will then either dissolve away or act as further nuclei. However, for multibubble systems, most of the bubbles exist in clusters, which generally lead to asymmetric collapse of the bubbles. This is more likely to make fragmentation the dominant pathway after bubble collapse.

The data presented in Figures 1 and 2 illustrate that, as the concentration of the surface-active solute increases, $\Delta V_T$ decreases until it reaches a minimum and then slightly increases

(25) Ashokkumar, M.; Lee, J.; Kentish, S.; Grieser, F. *Ultrason. Sonochem.* **2007**, *14*, 470−475.
(26) Leighton, T. G.; Walton, A. J.; Pickworth, M. J. W. *Eur. J. Phys.* **1990**, *11*, 47−50.

again at higher concentrations. This behavior has been previously explained[21] as a consequence of the adsorption of surface-active solutes onto the surface of cavitation bubbles.

Bubble coalescence takes place through a sequence of events.[20] As two bubbles in solution are brought into contact with one another, the bubble surfaces flatten against each other, forming a thin liquid film between them. This film then thins further until it is sufficiently thin for rupture to occur via an instability mechanism. The thinning of the liquid film is the rate-determining step. Therefore, if this thinning mechanism takes longer than the bubble contact time, coalescence will not occur.

Adsorption of surface-active solutes on the bubble/solution interface can inhibit coalescence via two different means: steric repulsion (or other short-range repulsive forces), and also electrostatic repulsion if the surface-active solutes are charged. Steric repulsion (or other short-range repulsive forces) only takes place when the adsorbed solutes are in very close vicinity of each other, whereas both long-range and short-range electrostatic repulsions can occur, depending on the nature of the solutes present. All of these repulsion forces act to keep the bubbles sufficiently far apart such that the liquid film between them does not get thin enough for rupture to take place, thus preventing them from coalescing.

For SDS, which is an anionic surfactant, both of these repulsion forces contribute to the results observed in Figure 2, whereas for alcohols (Figure 1) only the steric effect (or other short-range repulsive effects) is important. The slight increase in $\Delta V_T$ at higher solute concentrations has been attributed to the increasing level of dissolved air for alcohols,[27] whereas for SDS, it has been proposed that the excess surfactant molecules act as an electrolyte, reducing the strength of the electrostatic repulsion and hence also the inhibition of bubble coalescence.[18]

Figures 3a−5a show that absolute $\Delta V_T$ values are greater for the higher applied acoustic powers. This is most probably due to a greater number of bubbles being created at the higher powers.[17] However, there is no significant difference between the $\Delta V_T$ values for the various powers once they are normalized to the $\Delta V_T$ for water. Based on Scheme 1, it should be possible to qualitatively correlate $\Delta V_T$ to the SL intensity. For SDS solutions in the presence of 0.1 M NaCl, while the $\Delta V_T$ data (Figure 5b in this report) show no power dependence, the corresponding multibubble sonoluminescence (MBSL) data (Figure 8 in ref 17) suggest some dependence on the applied acoustic power. It has been speculated in our previous MBSL studies[17,18,27] that the adsorption of charged surfactants onto the bubble/solution interface leads to the "declustering" of the cavitation bubbles, which is responsible for the enhanced MBSL (under the conditions where surfactant decomposition does not play a significant role in quenching the SL intensity). In a recent acoustic emission spectral study,[28] we have demonstrated that the presence of SDS in the solution affects not only the bubble coalescence, but also the structure of the bubble clusters produced. Our coalescence results also provide further evidence for the hypothesis that the effect of charged surfactants on MBSL is primarily related to bubble clustering. It has been
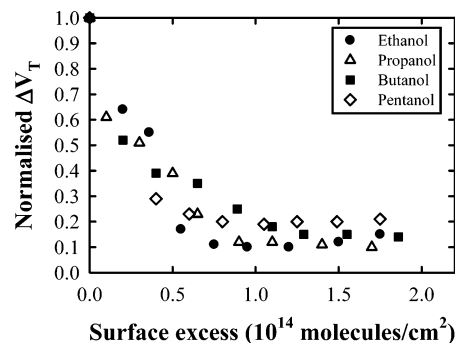


**Figure 7.** Average change in total bubble volume relative to pure water as a function of Gibbs surface excess of aliphatic alcohols in aqueous solutions sonicated at 358 kHz, 0.9 W/cm$^2$.
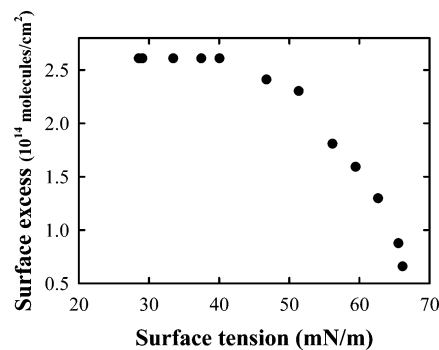


**Figure 8.** Dependence of surface excess on surface tension for SDS solutions in the presence of excess NaCl (ref 33).

observed in our previous MBSL study in the presence of small amounts of SDS ($\sim$1−2 mM) that the normalized MBSL signal returns to that of pure water at higher power levels (1.13−1.61 W/cm$^2$), once the electrostatic repulsion between bubbles is eliminated by the addition of excess electrolyte. However, the normalized bubble coalescence in SDS solutions in the presence of 0.1 M NaCl does not revert back completely to the level observed in pure water, irrespective of the acoustic power used (Figure 5b). This observation supports the hypothesis that the changes to the bubble clustering caused by the adsorption of charged surfactants onto the bubble/solution interface are primarily responsible for the SL enhancement reported in our earlier work (Figure 5 in ref 17).

The data obtained in this work also offer valuable information on the dynamics of surfactant adsorption on cavitation bubbles. As mentioned earlier, the addition of 0.1 M NaCl into SDS solutions is known to screen any electrostatic interactions between bubbles.[18,29] As can be seen from Figure 6, the addition of up to 0.4 M NaCl to 100 mM ethanol solution has no significant effect on the $\Delta V_T$. This is expected because ethanol molecules, although polar, have no formal charge. In contrast, for SDS solutions $\Delta V_T$ increases with increasing NaCl concentration until it reaches a plateau value. This increase in $\Delta V_T$ is consistent with the proposition that the addition of salt reduces the strength of the electrostatic repulsion between the charged surfactant headgroups on the bubbles, leading to a lessening of coalescence inhibition. It should also be noted that at all SDS concentrations used, the limiting $\Delta V_T$ plateau values still fall below that of water (i.e., $\Delta V_T = 1.0$). This confirms that even when the long-range electrostatic repulsion between bubbles is

(27) Lee, J. The Behaviour of Ultrasound Generated Bubbles in the Presence of Surface Active Solutes. Ph.D. Thesis, The University of Melbourne, 2005.

(28) Ashokkumar, M.; Hodnett, M.; Zequiri, B.; Grieser, F.; Price, G. J. *J. Am. Chem. Soc.* **2007**, *129*, 2250−2258.

(29) Tronson, R.; Ashokkumar, M.; Grieser, F. *J. Phys. Chem. B* **2002**, *106*, 11064−11068.

turned off, there is still some residual coalescence inhibition due to steric repulsion and probably also a very short-range electrostatic repulsion. (For example, the Debye length in solutions with an electrolyte concentration of 0.1 M is calculated to be around 1 nm (Shaw, D. J. *Introduction to Colloid and Surface Chemistry*, 2nd ed.; Butterworths: London, 1970). This, in addition to the steric effects, can possibly inhibit the very last stage of coalescence between two bubbles.)

The strength of the steric repulsion (or other short-range repulsive forces) between bubbles can be expected to depend on the number of surfactant molecules on the surface, which can be directly related to the surface excess of the surfactant. The higher is the surface excess, the greater is the strength of the steric repulsion (or other short-range repulsive forces) between bubbles and hence the greater is the extent of inhibition of bubble coalescence. This statement brings us to an interesting observation. Consider the data points for 2 mM SDS + 0.1 M NaCl and 100 mM ethanol solutions (see boxed points in Figure 6). (Those two points were chosen because the long-range electrostatic repulsion does not play a significant role in these cases, hence allowing the steric effect to be compared.) Steric repulsion (or other short-range repulsive forces) is the principal factor contributing to coalescence inhibition in these two cases. The equilibrium surface excess ($\Gamma_{eq}$) for 2 mM SDS + 0.1 M NaCl[30] is $2.6 \times 10^{14}$ molecules/cm$^2$, while $\Gamma_{eq}$ for 100 mM ethanol[18] is $0.55 \times 10^{14}$ molecules/cm$^2$. It is evident that even though $\Gamma_{eq}$ for 100 mM ethanol is only about one-fifth of that for 2 mM SDS + 0.1 M NaCl, it has a lower $\Delta V_T$, which means it inhibits bubble coalescence to a greater extent! This strongly suggests that, within the lifetime of the bubble, there are more ethanol molecules adsorbed onto the bubble/solution interface than SDS, or in other words, the thermodynamic equilibrium surface excess in the SDS system is not reached. This interpretation supports the work of Sostaric and Reisz,[23,24] who came to the same conclusion based on a series of EPR and spin-trapping studies on some sonicated surfactant solutions.

Figure 5b indicates that not only is the equilibrium surface excess not reached in systems containing SDS, but just how far the system is from equilibrium very much depends on the applied acoustic frequency. It is evident that as the applied acoustic frequency is increased, the bubble coalescence is inhibited to a lesser extent. This indicates that a lower number of SDS molecules adsorb onto the bubble/solution interface at higher frequencies.

The nonequilibrium surface excess is generally difficult to quantify due to the dynamic nature of the adsorption process. The work by Eastoe and Dalton[31] concluded that surface-active solutes need a finite time to equilibrate with the interface. Solutes of low surface-activity were found to equilibrate extremely rapidly. In contrast, surfactants that have a relatively higher $\Gamma_{eq}$ at a particular bulk concentration and reduce the surface tension more significantly than short alkyl chain alcohols require longer times to equilibrate with the interface.[32]

Previous studies[10,18,33,34] with surface-active solutes in sonicated systems found that a quantitative relationship exists between $\Gamma_{eq}$ of a homologous series of alcohols and a number of sonochemical processes (such as the dissolution of MnO$_2$ particles[33] and the reduction of gold chloride[10,34]) and the quenching of sonoluminescence.[18] This strongly suggests that equilibrium adsorption is achieved for these aliphatic alcohols under the sonication conditions used. On the basis of these studies, we are able to propose a method utilizing the $\Delta V_T$ data obtained to estimate the nonequilibrium surface excess values for some systems. When the $\Delta V_T$ data for aliphatic alcohol solutions are plotted as a function of the Gibbs surface excess, they all lie on a "master curve", as shown in Figure 7. This curve directly relates the amount of surface-active solutes adsorbed on the bubble/solution interface to the extent of coalescence inhibition they impart due to steric repulsion (or other short-range repulsive forces). This direct relationship, however, is not valid in systems where long-range electrostatic repulsions come into play (such as in SDS solutions without the addition of excess electrolyte). Hence, provided steric repulsion (or other short-range repulsive forces) is the dominant factor controlling the extent of coalescence inhibition, one can experimentally obtain the $\Delta V_T$ value for any given system and subsequently use this to estimate the surface excess value from the master curve (Figure 7). Consider the 2 mM SDS + 0.1 M NaCl solution, where the electrolyte is present in excess, thereby eliminating any long-range electrostatic interactions. The normalized $\Delta V_T$ value for this system was found to be around 0.36 for 358 kHz (see Figure 5b or Figure 6). Relating this value to the master curve in Figure 7 gives a nonequilibrium surface excess value of $\sim 0.5 \times 10^{14}$ molecules/cm$^2$ (as compared to $2.6 \times 10^{14}$ molecules/cm$^2$, if true equilibrium had been achieved[30]).

Once this surface excess value is estimated, it can be used further to gain some insight into the lifetime of acoustic bubbles in the antinodes in a multibubble field. It has been proposed[29] that at relatively high frequencies such as the ones used in this work, bubbles exposed to ultrasonic irradiation undergo a large number of oscillations during their active life span. However, the actual number of oscillations is unknown. Tajima's work on SDS solutions in the presence of excess NaCl provides data on both surface tension and surface excess as a function of SDS concentration.[30] These two sets of data were combined, and the result is shown in Figure 8. These data can then be used to convert the dynamic surface tension data for 2 mM SDS + 0.1 M NaCl obtained by Fainerman and Lylyk[35] into the surface excess of SDS as a function of time (Figure 9). It follows then that the time required for a given surface excess value to be achieved in a particular system can be estimated from Figure 9.

It has to be pointed out here that there are limitations associated with adopting this approach. First, the data in the literature are for unstirred systems, whereas constant stirring exists in our systems due to the acoustic microstreaming brought about by cavitation.[19] This will largely affect the transport rate of surfactant molecules toward the bubble/solution interface. Previous studies[36,37] found that SDS adsorption at the air/

(30) Tajima, K. *Bull. Chem. Soc. Jpn.* **1970**, *43*, 3063−3066.
(31) Eastoe, J.; Dalton, J. S. *Adv. Colloid Interface Sci.* **2000**, *85*, 103−144.
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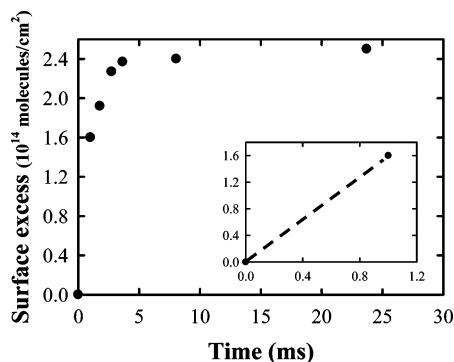(33) Sostaric, J. Z.; Mulvaney, P.; Grieser, F. *J. Chem. Soc., Faraday Trans.* **1995**, *91*, 2843−2846.
(34) Barbour, K.; Ashokkumar, M.; Caruso, R. A.; Grieser, F. *J. Phys. Chem. B* **1999**, *103*, 9231−9236.
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(36) Fainerman, V. B. *Colloids Surf.* **1991**, *57*, 249−266.
(37) Wu, N.; Dai, J.; Micale, F. J. *J. Colloid Interface Sci.* **1999**, *215*, 258−269.

**Figure 9.** Surface excess as a function of time for 2 mM SDS in the presence of 0.1 M NaCl (ref 34). The inset shows the region of very short times, which is assumed to be linear.

aqueous solution interface is mainly diffusion controlled at low bulk concentrations, but the governing mechanism is shifted to mixed diffusion-kinetic controlled at higher bulk concentrations. Therefore, it is expected that using literature data for a higher bulk SDS concentration would be more accurate for our estimations. Because the maximum SDS molecule (monomer) concentration is fixed by the critical micelle concentration (CMC) (the CMC for SDS in the presence of excess NaCl is around 1.62 mM (ref 30)), the estimations of the bubble lifetime are based on the data for 2 mM SDS + 0.1 M NaCl. Using these data, the time estimated from Figure 9 would be an upper limit to the bubble lifetime.[38] As explained earlier in this discussion, however, not all of the bubbles in the antinodes would be active if bubble clustering takes place. In other words, it is likely that the bubbles in the antinodes, whose maximum lifetimes are estimated here, consist of both active and inactive bubbles. Therefore, even though strictly speaking what we are estimating here is the maximum time a bubble can live in the antinodes, it can be inferred that this time is also the upper limit of the active bubble population.

The limited amount of data points available to construct Figure 9 does not cover the range of surface excess values we are interested in. Nevertheless, it is reasonable to assume linearity of the surface excess as a function of time for very short times between 0 and 2 ms (see inset). This gives the time required for $0.5 \times 10^{14}$ molecules/cm$^2$ of SDS to adsorb onto the interface as ~0.30 ms, which equates to around 110 oscillations for the applied frequency of 358 kHz. (It is assumed that the rate of surfactant adsorption and desorption during the active life span of an oscillating bubble is the same as that of a "static" bubble. This is a valid assumption because the rate-determining step of surfactant adsorption onto an interface involves molecular transformations (e.g., the reorientation of the surfactant molecules and the dehydration of the hydrophobic tail) rather than bulk diffusion.)

Referring back to Figure 5b, a similar approach can be followed for all of the other frequencies, and the results are

**Table 1.** Summary of the Estimated Maximum Bubble Lifetimes for the Various Applied Frequencies Investigated

| applied frequency (kHz) | period ($\mu$s) | estimated max bubble lifetime (ms) | estimated max number of oscillations |
|---|---|---|---|
| 213 | 4.7 | $0.35 \pm 0.05$ | $70 \pm 10$ |
| 358 | 2.8 | $0.30 \pm 0.05$ | $110 \pm 20$ |
| 647 | 1.6 | $0.15 \pm 0.05$ | $100 \pm 30$ |
| 1062 | 0.9 | $0.10 \pm 0.05$ | $110 \pm 50$ |

summarized in Table 1. From the table, it is evident that the maximum lifetime of the bubbles decreases with increasing applied frequency. Furthermore, for the range of frequencies investigated, these lifetimes equate to the bubbles undergoing an upper limit of 50−200 oscillations in their active lifetime.

This result is highly significant, in that it indicates that active bubbles exposed to an acoustic field have a finite lifetime in a multibubble system, which is not necessarily the case for a single bubble.[39,40] We had previously estimated that bubbles at a frequency of 515 kHz have an upper limit of 5000 oscillations.[41] However, the present method provides a much better estimate of the persistence time of an acoustic bubble in a multibubble field. It also demonstrates the dependence of the bubble lifetime on the applied frequency.

## Conclusions

Adsorption of surface-active solutes onto bubble/solution interface in liquids under sonication is a dynamic and complex process. It affects the phenomenon of bubble coalescence, which can in turn influence the efficiency of the whole sonochemical process. Within the limitations of the measurement method and the systems studied, bubble coalescence does not seem to depend on the applied acoustic power. Dependence on the applied acoustic frequency, however, is apparent in some systems. The method used also provides important insight into the dynamics of surface-active solutes adsorption onto bubble/solution interface and can be used to estimate the nonequilibrium surface excess values of some surfactants in liquids exposed to ultrasound. An acoustic bubble in a multibubble field has a finite lifetime, as a consequence of bubble−bubble coalescence processes and rectified growth, which is shorter for higher applied acoustic frequency. It is estimated that these bubbles undergo no more than 50−200 oscillations in their lifetime for applied frequencies in the range of 200 kHz to 1 MHz.

**Supporting Information Available:** Complete experimental details. This material is available free of charge via the Internet at http://pubs.acs.org.

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(38) At lower surfactant concentrations, the lifetime of the bubbles will be shorter due to less number of surfactant molecules adsorbed on the interface protecting the bubbles from coalescing, as reflected in the higher $\Delta V_T$ values.

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