The Influence of Acoustic Power on Multibubble Sonoluminescence in Aqueous Solution Containing Organic Solutes

Devi Sunartio, Muthupandian Ashokkumar, and Franz Grieser*

School of Chemistry, University of Melbourne, Melbourne VIC 3010, Australia Received: May 25, 2005; In Final Form: August 19, 2005

The effect of varying the applied acoustic power on the extent to which the addition of water-soluble solutes affect the intensity of aqueous multibubble sonoluminescence (MBSL) has been investigated. Under most of the experimental conditions used, the addition of aliphatic alcohols to aqueous solutions was found to suppress the MBSL intensity, although an enhancement of the MBSL intensity was also observed under certain conditions. In contrast, the presence of an anionic surfactant sodium dodecyl sulfate (SDS) in aqueous solutions generally enhanced the observed MBSL intensity. For a series of aliphatic alcohols and SDS, a strong dependence of the MBSL intensity on the applied acoustic power (in the range of 0.78–1.61 W/cm²) at 358 kHz was observed. The relative SL quenching was significantly higher at higher acoustic powers for the alcohol solutions, whereas the relative SL enhancement was lower at higher acoustic powers in SDS solutions. These observations have been interpreted in terms of a combination of material evaporation into the bubble, rectified diffusion, bubble clustering and bubble—bubble coalescence.

Introduction

A major goal of sonochemists is to increase the efficiency of ultrasonic processes so that these processes are economically attractive for commercial applications. Sonoluminescence (SL), which is the phenomenon of light emission from cavitation bubbles, can be utilized as a means by which the efficiency of sonochemical reactions under different experimental conditions can be qualitatively monitored. The exact origin of SL emission is still under debate, although in recent years, it has become more widely accepted that SL is probably a combination of blackbody radiation, bremsstrahlung and excited-state emission. Some of the many other processes that have been suggested are comprehensively reviewed by Leighton and Yasui.

Many previous studies have investigated the effect of changing different parameters on the observed SL intensity and sonochemical efficiency. To 22 Vazquez and Putterman reported a decrease in SL intensity as the solution temperature was increased and emphasized the importance of water vapor trapped inside the collapsing bubble. A study conducted soon after by Dezhkunov found that the temperature dependence of SL intensity was different at different acoustic intensities. At low acoustic intensities, SL intensity increased with increasing solution temperature, whereas, at high acoustic intensities, the opposite effect was observed. A balance between an increase in the number of bubbles and a decrease in the intensity of bubble collapse as the solution temperature is increased was suggested as being the cause for these observations.

The driving ultrasonic frequency is another parameter that has been extensively investigated. ^{10–14} Beckett and Hua¹⁰ studied SL and sonochemistry in aqueous solutions over a range of frequencies and concluded that 358 kHz was the optimum frequency for both processes, while the work of Tronson et al. ¹¹ and Price et al. ¹² looked into different types of cavitation

occurring at 20 kHz and 515 kHz. It was observed that the effect of added solutes on the SL intensity was minimal at 20 kHz because of the transient nature of cavitation bubbles at this relatively low frequency. Yasui¹³ recently conducted a series of computer simulations and proposed that the mechanism of SL emission depends on the frequency, with plasma emission largely responsible for light emission at 1 MHz, while both plasma emission and chemiluminescence from excited state •OH radicals give rise to light emission at 20 kHz.

Much of the work undertaken by our research group has focused on exploring the effect of adding various surface-active solutes on the SL intensity generated in aqueous solutions. 12,15-22 It has been found that adding volatile solutes such as alcohols, ketones, carboxylic acids, n-alkylamines and n-alkyl acetates in their neutral forms to the solution undergoing sonication quenches SL emission by lowering the cavitation bubble temperature. 12,15,16,18-22 On the other hand, addition of charged surfactants^{15,16,18,21} and salts¹⁷ enhances SL emission by affecting the structure of bubble clusters and gas solubility in solution, respectively. To this end, however, all of our earlier work has always been carried out over a small range of acoustic power. The present work attempts to extend the previous studies by investigating the significance of varying applied acoustic power on the extent to which the presence of surface-active solutes affects bubble behavior in solution and, consequently, the SL emission intensity.

Experimental Details

The ultrasound generator used in all experiments was an ELAC Nautic RF generator type LVG 60 A operating in a continuous mode at 358 kHz. The reaction cell was kept in a light-proof enclosure, which also contained an end-on photomultiplier tube (Hamamatsu, model no. E849—34, detection range 350—600 nm) for detecting the integrated SL emission from the sonicated solution. The emission signal was then displayed on a Tektronix TDS 320 digital oscilloscope.

^{*} Corresponding author. E-mail: franz@unimelb.edu.au.

0.78 W/cm²

△ 0.90 W/cm²

■ 1.13 W/cm²

♦ 1.61 W/cm²

150

200

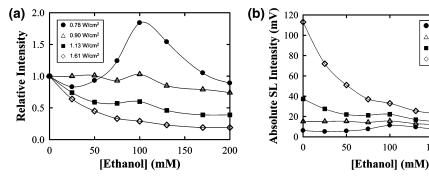


Figure 1. (a) SL intensity relative to pure water as a function of ethanol concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.) (b) SL absolute intensity as a function of ethanol concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.)

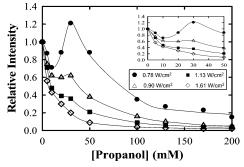


Figure 2. SL intensity relative to pure water as a function of propanol concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers. The inset shows an enlargement of results at low concentrations. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.)

All alcohols used were AR grade, and solutions were made up using Milli-Q water with a conductivity of <10⁻⁶ S cm⁻¹ at 22 °C. Sodium dodecyl sulfate (SDS) was BDH special purity grade and was used as supplied.

Experiments were conducted in air-saturated solutions at room temperature (about 20 °C), and any heating of the solutions during sonication was limited to a few degrees. The volume of solutions sonicated was 200 mL, and the power absorbed by the liquid was determined by calorimetry. In a typical experiment, the SL intensity was obtained by first sonicating the solution for 2 min and allowing it to stand undisturbed for another 2 min before the sonicator was turned on again and the average SL intensity recorded. This procedure was found to give reproducible results.

Results

The emission intensity, relative to pure water, of SL emitted from ethanol solutions of various concentrations sonicated at different acoustic powers is shown in Figure 1a (for comparison, the absolute intensity data is presented in Figure 1b). Consistent with the findings from previous studies^{16,20,21}, the SL emission intensity is affected by the presence of alcohol in the solution. An interesting observation from Figure 1, which has not been previously reported, is the effect of applied acoustic power on the extent of SL quenching. As the applied power increases, the extent of SL quenching also increases. This behavior is also observed with longer chain alcohols, as shown in Figure 2 for propanol and Figure 3 for butanol.

For all three alcohols, it is noteworthy that, at the lower powers used, some enhancement of the SL intensity is observed.

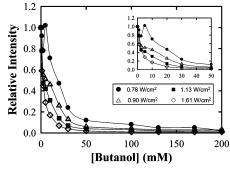


Figure 3. SL intensity relative to pure water as a function of butanol concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers. The inset shows an enlargement of results at low concentrations. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.)

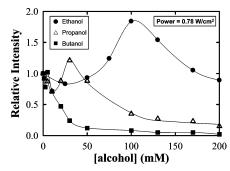


Figure 4. SL intensity relative to pure water as a function of alcohol concentration in aqueous solutions sonicated at 358 kHz at 0.78 W/cm² applied acoustic power. An initial SL enhancement is observed, followed by subsequent SL quenching. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.).

Figure 4, in which the relative SL intensity as a function of alcohol concentration is shown for all three alcohols at 0.78 W/cm² applied power, shows this more clearly. A greater relative enhancement is observed with ethanol compared to propanol, although a higher concentration of ethanol is needed to achieve this effect. Similarly for butanol, its relative enhancement is lower than the other two alcohols, but this enhancement occurs at a significantly lower butanol concentration. Furthermore, comparing the data in Figures 1-3, for a given alcohol concentration, more quenching is observed the longer the alkyl chain of the alcohol. This is in agreement with previous work of Ashokkumar et al.16

Another surface-active solute that has been previously studied is the anionic surfactant SDS. Figure 5a shows the relative SL intensity as a function of SDS concentration at different acoustic powers. (For comparison, the absolute intensity data is presented

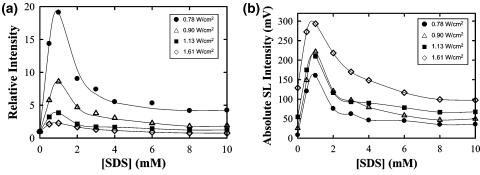


Figure 5. (a) SL intensity relative to pure water as a function of SDS concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers. More SL enhancement is seen at lower powers. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.) (b) SL absolute intensity as a function of SDS concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.).

in Figure 5b.) The signal is seen to increase up to about 1 mM, after which a further increase in the SDS concentration results in a decrease in the intensity of the SL emission. As in the alcohol case, the applied acoustic power plays a major role in determining the extent of SL enhancement in the presence of SDS. The relative enhancement of SL intensity decreases as the acoustic power increases.

Discussion

The data presented in Figures 1–3 illustrate that the extent of relative SL quenching increases as the applied acoustic power is increased. To interpret these results, it is relevant to briefly review our understanding of what factors control the intensity of MBSL.

There are primarily two main factors that govern the MBSL intensity, namely the "active" bubble population and the cavitation bubble temperature. The number of "active" bubbles, which refers to those bubbles that can collapse violently and emit SL, is greatly dependent upon the structure of the bubble clusters that are produced in a multibubble system.^{23–28} The cavitation bubble temperature, which is the maximum temperature that can be attained at the bubble core upon bubble collapse, determines the intensity of SL emitted from the individual bubbles. This temperature depends on the intensity of the bubble collapse, which in turn is a function of a number of factors, such as the symmetry of the collapse, the change in volume of the bubble upon collapse, and the amount and type of gaseous material within the bubble.

The SL quenching by alcohols has been previously suggested 15,16,21,22,29 to be due to the evaporation of these volatile solutes from the bubble—solution interface during bubble expansion, followed by endothermic chemical reactions within the bubble on bubble collapse. At 358 kHz, the bubbles are considered to be "stable", 11 i.e., they live long enough (hundreds to thousands of oscillations 18) to allow the hydrocarbon products produced by these thermally initiated decomposition reactions to accumulate within the bubble over a number of acoustic cycles. The presence of these products and their thermal decomposition results in a lowering of the bubble temperature, leading to a lower SL intensity.

When the applied acoustic power is varied, it can cause a change in the "active" bubble population, the bubble temperature, or both. Two possible reasons for a greater relative quenching of MBSL intensity upon the addition of alcohols at higher acoustic powers are: a greater decrease in the "active" bubble population, or a greater decrease in the bubble temper-

TABLE 1: Amount of Hydrocarbon Products Produced upon the Sonication of 100 mM Ethanol Solutions at 358 kHz for 30 min^a

applied acoustic power (W/cm²)	ethylene concentration (mM)	acetylene concentration (mM)	ethane concentration (mM)	T _{max} (K)
0.90	0.58	1.23	0.06	5500
1.13	1.54	2.49	0.20	4900
1.61	3.91	2.76	0.78	3800

 a The amount produced at 0.78 W/cm² applied acoustic power was too low to allow $T_{\rm max}$ determination and, hence, was not included in the table. $T_{\rm max}$ was calculated according to the method used in previous studies (Hart, E. J.; Fischer, C. H.; Henglein, A. *Radiat. Phys. Chem.* **1990**, 36, 511; Tauber, A.; Mark, G.; Schuchmann, H.-P.; von Sonntag, C. *J. Chem. Soc., Perkin Trans.* 2 **1999**, 1129; Rae, J.; Ashokkumar, M.; Eulaerts, O.; von Sonntag, C.; Reisse, J.; Grieser, F. *Ultrason. Sonochem.* **2005**, 12, 325).

ature, relative to pure water at the same power. One way to decrease the "active" bubble population is by increasing the bubble-bubble coalescence rate. However, it has been found previously that alcohols retard the rate of bubble coalescence rather than accelerating it.30 Another way to decrease the "active" bubble population is by increasing the density of the bubble cluster. There is no obvious physicochemical reason for the addition of alcohols to increase the extent of bubble clustering, although at this point it is not possible to completely rule this out. One might also argue that, as hydrocarbon products accumulate and the pressure inside the bubble builds up, the average bubble size distribution might be shifted to a larger size and out of the SL "active" range. This will then give rise to a lower number of "active" bubbles and, hence, a decrease in the SL intensity. At higher power levels, this can be expected to occur to a greater extent and thus may explain the greater SL quenching. However, according to the previous works of Guan and Matula,²⁹ and also Ashokkumar et al.,²⁰ for singlebubble systems, complete SL quenching can be achieved without a significant change in the maximum bubble size. Therefore, it can be reasonably concluded that this factor is not significant in explaining the results obtained in the present study.

The possibility, that a decrease in the bubble temperature is responsible for a lowering of the SL intensity, was investigated by measuring the amount of hydrocarbon products produced at the different acoustic powers. Table 1 shows that the amount of hydrocarbon products produced is greater at higher applied acoustic powers. It can be proposed that, at higher powers, as bubbles can be expanded to a relatively larger size during their expansion cycle, more volatile material would evaporate into the bubble before collapse. This, combined with the greater volume change of the bubble upon collapse, would result in

Figure 6. Diagrammatic representation of a bubble being expanded to a relatively larger size during the expansion cycle at higher applied acoustic powers. This leads to a greater level of hydrocarbon products (P) accumulation inside the bubble, lowering the bubble temperature and, hence, quenching the SL intensity to a greater extent.

more hydrocarbon products being produced and, therefore, a greater level of accumulation of these products inside the bubble. The overall increase in complex gaseous chemical products inside the bubble would lead to a lowering of the bubble temperature and, hence, a decrease in the SL emission intensity (Figure 6). This proposition is in agreement with previous results on single bubble SL, 31 where the maximum radius of the bubble ($R_{\rm max}$) was found to vary proportionally with the magnitude of the driving pressure ($R_{\rm max}$ increases when the driving acoustic pressure is increased).

Inhibition of bubble coalescence leads to an increase in the number of bubble nuclei in the solution and, hence, the number of "active" bubbles that would be expected to lead to a relative enhancement of SL intensity. Considering the observation that alcohols inhibit bubble coalescence in solution, both SL quenching (due to material evaporation) and SL enhancement (due to inhibition of bubble coalescence) can take place upon the addition of alcohols to aqueous solutions. The overall outcome with respect to the effect on the SL intensity is determined by the relative importance of these two occurrences. At low applied acoustic powers, it is possible that the SL enhancement dominates over the SL quenching, which accounts for the results presented in Figure 4. Furthermore, by examining the data presented in Figures 1-3, this balance between SL quenching and enhancement in alcohol solutions is also observed at higher powers, albeit less markedly. As suggested earlier, the extent of SL quenching increases as the power is increased, thus, at higher powers, this enhancement is not as prominent as at low powers. It should also be noted that the alcohol concentrations at which the inhibition of bubble coalescence starts to be significant, usually termed the transition concentration, differ slightly from the values obtained by previous studies.^{30,32} A possible reason for the apparent discrepancy is that the previous studies were carried out under steady-state equilibrium conditions, whereas the bubble oscillations and clustering in our case can be expected to give rise to somewhat different interaction conditions, thereby leading to different transition concentrations for the alcohols. The concentrations observed in this work agree very well with those obtained in the recent acoustic study of Lee et al.³³ (Table 2).

TABLE 2: Comparison Between Transition Concentrations of Aliphatic Alcohols Observed in This Work and Those Obtained in Previous Studies

	transition concentration (M)		
solute	this worka	Lee et al.b	Oolman and Blanch ^c
ethanol	0.040	0.05	0.14
<i>n</i> -propanol	0.015	0.02	
<i>n</i> -butanol	3.0×1.0^{-3}		1.8×10^{-3}

^a The transition concentration value denotes the concentration at which the relative SL intensity starts to increase after the initial decrease (see Figure 4). ^b Ref 33. ^c Ref 30.

It is worthwhile to compare the data presented in Figure 4 with that of Price et al. 12 They have shown that, at 20 kHz, where SL quenching is negligible because of the transient nature of the cavitation bubbles at this frequency, longer alkyl chain alcohols show marked enhancement in SL intensity compared to that of their shorter chain counterparts. In the present work, however, SL enhancement is more pronounced for shorter chain alcohols. This is because, at the frequency used in this work (358 kHz), SL quenching is an important competing process to the SL enhancement (due to the cavitation being "stable"), and that longer chain alcohols will cause more quenching. As a result, the combined effect of these two competing processes leads to an overall weaker enhancement for longer chain alcohols.

Previous reports^{12,16} have also shown that the amount of material that evaporates into a bubble, hence the extent of SL quenching by alcohols, depends on the amount of alcohol adsorbed at the bubble-solution interface and not the type or bulk concentration of the alcohol in solution. By using the method outlined by Ashokkumar et al., 16 the relative SL intensity is plotted against the air-water Gibbs surface excess for all the alcohols at the different acoustic powers used, as shown in Figure 7. As expected, at higher acoustic powers (Figure 7c and d) there is a good correlation between the relative SL intensity and the Gibbs surface excess because, under these conditions, SL quenching is the dominant process. However, at lower powers (Figure 7a and b), a poor correlation is observed. This can be ascribed to the significant contribution of the inhibition of bubble coalescence by the interfacially adsorbed alcohol molecules on the overall SL emission intensity.

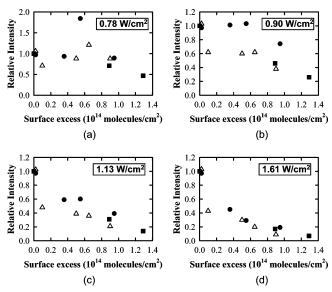


Figure 7. SL intensity relative to pure water as a function of surface excess of alcohols in aqueous solutions sonicated at 358 kHz at: (a) 0.78 W/cm², (b) 0.90 W/cm², (c) 1.13 W/cm², (d) 1.61 W/cm² (● ethanol, △ propanol, ■ butanol).

The effect of SDS on SL intensity has been previously investigated by a number of workers. 16,34-36 While all agree that the origin of the SDS effect is related to the interbubble electrostatic interactions between the charged surfactants adsorbed on the bubble surface, there is a continuing debate on how these interactions actually affect the cavitation bubble field. The various experiments conducted by Segebarth et al.³⁴ lead them to conclude that the presence of SDS primarily modifies the bubble-bubble coalescence processes. They argue that the electrostatic repulsion between the headgroups on the surfactants prevents the bubbles from coalescing, which in turn leads to a shift in the bubble size distribution to a smaller size while increasing the number of available nuclei in the solution. This is also supported by the recent work of Lee et al.,36 in which the average bubble sizes in different systems were estimated by varying the "off" time between acoustic pulses. Thus, clearly, the presence of SDS in the solution does affect the extent of bubble—bubble coalescence and, hence, the overall SL intensity.

On the other hand, it is worthwhile to revisit the earlier work of Ashokkumar et al. ¹⁶ They found that the SL intensity emitted from a 1 mM SDS solution in the presence of 0.1 M NaCl is the same as that in water (all other experimental conditions being kept constant). However, Lee et al. ³³ showed that, under the same experimental conditions, there was still a significant inhibition of coalescence in the SDS/NaCl system. The latter results are important, as they indicate that changes in the degree of coalescence of bubbles in a bubble cluster cannot be the only factor responsible for the change in the SL intensity in the SDS solutions. There must be (at least) another process that comes into play when SDS is present in solution, as previously suggested, a likely explanation rests with the spatial structure of the bubble cluster itself.

Ashokkumar et al. ¹⁶ argued that the repulsive electrostatic interactions between bubbles may result in a *relatively* more open bubble cluster structure. This would then allow the applied ultrasound field to affect a larger number of bubbles in a cluster that would, in turn, lead to a higher number of "active" bubbles and, hence, a higher SL emission. Segebarth et al. ³⁴ disagreed with this view on the basis of their calculations on the magnitude of the Debye length (the distance over which electrostatic forces may act). They indicated that the Debye length is 9.6 nm in a

1 mM SDS solution, and only 1 nm when 0.1 M NaCl is added to the solution. They argued that the usual estimations³⁷ of the bubble volumical fraction, which vary between 10^{-5} and 10^{-2} , means that the bubbles in solution would be too far apart for the presence of SDS molecules on the bubble surface to affect the interbubble distance by repulsive electrostatic interactions. However, the "usual estimation" values for the bubble volumical fraction referred to by Segebarth et al. is an indication of the total volume of the bubbles as a fraction of the total volume of the solution. Because the bubbles responsible for SL emission in a sonicated solution are concentrated within the antinodes, the bubble density within a cluster is very high (as shown by photographic images of bubble clusters obtained by Hatanaka et al.^{24,26}), high enough to have the interbubble distance in the cluster be comparable to the Debye length. Under such conditions, having charged SDS molecules on the surface of the bubbles can cause repulsion between the bubbles. This will give rise to a *relatively* more open bubble cluster structure in the presence of SDS, compared to that in water. It can be argued that an increase in the interbubble distance from 1 nm (in the case of 1 mM SDS + 0.1 M NaCl) to 10 nm (1 mM SDS) is very small and, hence, will not make any difference in the overall spatial structure of the cluster. Nevertheless, having deduced that there must be another factor apart from the bubble—bubble coalescence that affects the cluster, it is possible that this seemingly insignificant increase in the interbubble distance may indeed be a governing factor in explaining the SDS effect on SL intensity.

Figure 5 illustrates that SL enhancement is very much power dependent. Assuming both coalescence and clustering phenomena are important in explaining the experimentally observed results, following a similar approach of reasoning as in the alcohol case, a greater relative enhancement of MBSL intensity upon the addition of SDS at lower powers can be achieved by having a greater increase in the bubble population or a greater increase in the bubble temperature, relative to pure water at the same power. The argument that more open bubble clusters are formed at lower acoustic powers can be rationalized by taking into account that the secondary Bjerknes forces are proportional to the square of the sound pressure. 6,38,39 Therefore, one would expect that, at higher applied acoustic powers, these forces will be much stronger and will bring the bubbles in a cluster closer together. This would result in more compact bubble clusters, approaching that existing in pure water, thereby reducing the number of "active" bubbles and, hence, causing less relative enhancement of the SL emission intensity. The stronger Bjerknes forces will also promote bubble-bubble coalescence, which will further decrease the number of "active" bubbles. The relatively more open bubble clusters structure might also allow a more symmetrical collapse of the "active" bubbles, thereby increasing the bubble temperature and SL intensity emitted from the individual bubbles.

Another possible argument that can explain the lesser SL enhancement at higher power levels is related to the average bubble size distribution. It is known that rectified diffusion increases with acoustic power. As a consequence, the bubble size distribution will increase accordingly. If this new distribution is less "active" than the initial distribution (which can be expected because the separation between the relatively smaller bubbles in the initial distribution would be larger), then the emitted SL intensity would be less. Because all the data presented in this work are normalized to 1 at the zero solute concentration at each power level, any changes in the bubble size distribution upon changing power are already taken into

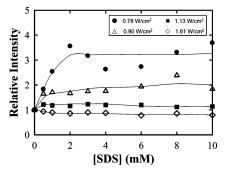


Figure 8. SL intensity relative to pure water as a function of SDS concentration in aqueous solutions sonicated at 358 kHz at various applied acoustic powers in the presence of 0.1 M NaCl. Even though the relative SL enhancement is significantly lower than compared to the solutions containing SDS alone (Figure 5), the power dependence of the SL relative intensity is still apparent. (The lines joining the data points are only intended to guide the eyes and are not theoretical fits.)

account when examining the behavior of each curve. However, having acknowledged that, surfactants such as SDS are known to increase the rate of rectified diffusion. 40,41 Moreover, the increase in the rectified diffusion rate due to the presence of surfactants is greater at higher acoustic powers, 40 which will shift the bubble size distribution and decrease the SL intensity to a greater extent. Nevertheless, it should be noted that, if rectified diffusion is the only parameter that affects the SL intensity, the addition of salt to the SDS solutions should not have any effect. As discussed below, the changes to the SL intensity in the presence of salt suggest that rectified diffusion is not the only factor that affects the SL intensity in the presence of SDS.

The addition of 0.1 M NaCl into SDS solutions is known to eliminate the SL enhancement because the electrolyte screens any electrostatic interactions between bubbles. 16 To confirm this, the SDS experiments at various powers were repeated in the presence of 0.1 M NaCl, and the relative intensity data is presented in Figure 8. It is evident that, although the overall signal enhancement is much less compared to the case of SDS alone, even after the addition of the salt, some residual enhancement is still observed. Moreover, this enhancement still seems to be power dependent. This shows that, even when electrostatic interactions are completely turned off, some inhibition of bubble coalescence is still present because of the expected steric hindrance from the surfactant molecules that are adsorbed on the bubble-solution interface, which is more pronounced at low powers because of the weaker Bjerknes forces under such conditions.

Conclusions

It has been observed that the extent to which the presence of surface-active solutes affects the SL emission intensity in aqueous solutions is strongly dependent on the applied acoustic power. With aliphatic alcohols, the relative SL suppression is greater at higher powers because of the possibility of expanding the bubble to a relatively larger size during an expansion cycle. This allows more material to evaporate into the bubble core and, subsequently, results in more hydrocarbon product accumulation inside the bubble over many bubble oscillation cycles, leading to a lowering of the bubble temperature and, hence, a greater level of SL quenching. At lower powers, however, some enhancement of the SL emission intensity seen is possibly due to the inhibition of bubble—bubble coalescence.

With SDS as a solute, the greater relative SL enhancement observed at lower powers is argued to be due to the formation of relatively more open bubble clusters, leading to a greater number of "active" bubbles and possibly a more symmetrical collapse of these bubbles, as well as a greater extent of inhibition of bubble coalescence. The results observed in the presence of a salt in SDS solutions suggest that the SDS effect is primarily related to electrostatic interactions between bubbles.

The implication of these findings on sonochemistry is that, depending on the specific application, it is of prime importance to select the correct applied acoustic power in order to achieve optimum reaction efficiency.

Acknowledgment. The authors would like to thank the Particulate Fluids Processing Centre, a Special Research Centre of the Australian Research Council, for financial support. D.S. also acknowledges the receipt of an Australian Postgraduate Award and a postgraduate stipend from The University of Melbourne.

References and Notes

- (1) Mason, T. J. Sonochemistry; Oxford University Press: New York,
- (2) Suslick, K. S.; Doktycz, S. J.; Flint, E. B. Ultrasonics 1990, 28, 280.
 - (3) Noltingk, B. E.; Neppiras, E. A. Proc. Phys. Soc. 1950, 63B, 674.
- (4) Yasui, K.; Tuziuti, T.; Sivakumar, M.; Iida, Y. Appl. Spec. Rev. 2004, 39, 399.
- Didenko, Y. T.; McNamara III, W. B.; Suslick, K. S. Nature 2000, 407, 877.
- (6) Leighton, T. G. The Acoustic Bubble; Academic Press: London, 1994.
- (7) Casadonte, D. J., Jr.; Flores, M.; Petrier, C. *Ultrason. Sonochem.* **2005**, *12*, 147.
 - (8) Vazquez, G. E.; Putterman, S. J. Phys. Rev. Lett. 2000, 85, 3037.
 - (9) Dezhkunov, N. V. Ultrason. Sonochem. 2002, 9, 103.
 - (10) Beckett, M. A.; Hua, I. J. Phys. Chem. A 2001, 105, 3796.
- (11) Tronson, R.; Ashokkumar, M.; Grieser, F. J. Phys. Chem. B 2002, 106, 11064.
- (12) Price, G. J.; Ashokkumar, M.; Grieser, F. J. Am. Chem. Soc. 2004, 126, 2755.
- (13) Yasui, K. J. Acoust. Soc. Am. 2002, 112, 1405.
- (14) Price, G. J.; Ashokkumar, M.; Cowan, T. D.; Grieser, F. Chem. Commun. 2002, 16, 1740.
- (15) Grieser, F.; Ashokkumar, M. Adv. Colloid Interface Sci. 2001, 89–90, 423.
- (16) Ashokkumar, M.; Hall, R.; Mulvaney, P.; Grieser, F. J. Phys. Chem. B 1997, 101, 10845.
- (17) Wall, M.; Ashokkumar, M.; Tronson, R.; Grieser, F. Ultrason. Sonochem. 1999, 6, 7.
- (18) Tronson, R.; Ashokkumar, M.; Grieser, F. J. Phys. Chem. B 2003, 107, 7307.
- (19) Ashokkumar, M.; Mulvaney, P.; Grieser, F. J. Am. Chem. Soc. 1999, 121, 7355.
- (20) Ashokkumar, M.; Crum, L. A.; Frensley, A.; Grieser, F.; Matula, T. J.; McNamara, W. B., III; Suslick, K. S. *J. Phys. Chem. A* **2000**, *104*, 8462
- (21) Ashokkumar, M.; Guan, J.; Tronson, R.; Matula, T. J.; Nuske, J. W.; Grieser, F. *Phys. Rev. E* **2002**, *65*, 046310.
 - (22) Ashokkumar, M.; Grieser, F. ChemPhysChem 2004, 5, 439.
 - (23) Leighton, T. G. Ultrason. Sonochem. 1995, 2, S123.
- (24) Hatanaka, S.; Yasui, K.; Kozuka, T.; Tuziuti, T.; Mitome, H. *Ultrasonics* **2002**, *40*, 655.
- (25) Burdin, F.; Tsochatzidis, N. A.; Guiraud, P.; Wilhelm, A. M.; Delmas, H. *Ultrason. Sonochem.* **1999**, *6*, 43.
- (26) Hatanaka, S.; Yasui, K.; Tuziuti, T.; Kozuka, T.; Mitome, H. Jpn. J. Appl. Phys., Part 1, 2001, 40, 3856.
- (27) Matula, T. J.; Roy, R. A. *Ultrason. Sonochem.* **1997**, *4*, 61.
- (28) Moussatov, A.; Granger, C.; Dubus, B. Ultrason. Sonochem. 2003, 10, 191.
- (29) Guan, J.; Matula, T. J. J. Phys. Chem. B 2003, 107, 8917.
- (30) Oolman, T. O.; Blanch, H. W. Chem. Eng. Commun. 1986, 43, 237.
- (31) Akhatov, I.; Gumerov, N.; Ohl, C.-D.; Parlitz, U.; Lauterborn, W. *Phys. Rev. Lett.* **1997**, *78*, 227.
- (32) Zahradnik, J.; Fialova, M.; Linek, V. Chem. Eng. Sci. 1999, 54, 4757.
- (33) Lee, J.; Kentish, S.; Ashokkumar, M. J. Phys. Chem. B 2005, 109, 5095.

- (34) Segebarth, N.; Eulaerts, O.; Reisse, J.; Crum, L. A.; Matula, T. J. *J. Phys. Chem. B* **2002**, *106*, 9181.
- (35) Lee, J.; Kentish, S.; Matula, T. J.; Ashokkumar, M. J. Phys. Chem. B 2005, 109, 16860.
- (36) Lee, J.; Ashokkumar, M.; Kentish, S.; Grieser, F. J. Am Chem. Soc. 2005, (communicated).
- (37) Sutin, A. M.; Yoon, S. W.; Kim, E. J.; Didenkulov, I. N. J. Acoust. Soc. Am. 1998, 103, 2377.
- (38) Mettin, R.; Luther, S.; Ohl, C.-D.; Lauterborn, W. *Ultrason. Sonochem.* **1999**, *6*, 25.
- (39) Lauterborn, W.; Kurz, T.; Mettin, R.; Ohl, C.-D. Adv. Chem. Phys. 1999, 110, 295.
 - (40) Crum, L. A. J. Acoust. Soc. Am. 1980, 68, 203.
- (41) Lee, J.; Kentish, S.; Ashokkumar, M. J. Phys. Chem. B 2005, 109, 14595.