# Multibubble Sonoluminescence in Ethylene Glycol/Water Mixtures

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ABSTRACT: The multibubble sonoluminescence (MBSL) signals generated by 3.5 ms pulses of 515 kHz ultrasound in air-saturated ethylene glycol, water, and ethylene glycol/water mixtures were examined in the absence and presence of a range of solutes, including aliphatic alcohols of various chain lengths (C_3-C_6) and ionic and zwitterionic surfactants. In general, the alcohols quenched the SL in most solvent mixtures and the surfactants enhanced the sonoluminescence signal. However, in some solvent mixtures complex effects were observed in the presence of the solutes. The discussion presented rationalizes the varied behavior of the solutes on the MBSL observed in terms of their influence on inter- and intrabubble effects experienced by bubbles in an ultrasound field.
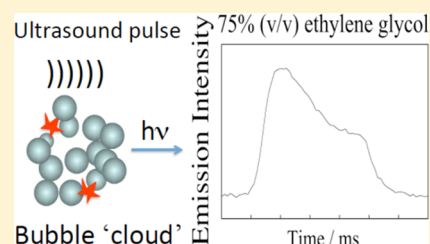
INTRODUCTION

It is now well-established that the emission of light that comes from liquids exposed to ultrasound is associated with the generation and the rapid collapse of microbubbles in the fluid.[1-5] The results from the most recent theoretical studies of this phenomenon have concluded that the genesis of the emission stems from the near adiabatic heating of the gas/vapor inside the bubble during its collapse.[4,5] The high local temperatures realized on bubble collapse lead to a partial ionization (plasma) of the gas/vapor inside the bubble followed by recombinant electron−ion radiative emission and bremsstrahlung. It is also clear that the temperature within a bubble is not an equilibrium temperature, based on studies from single-bubble and multibubble emissions.[6,7] In addition, light emission from vibronically excited states of a variety of molecular entities may also be produced,[7-11] to varying extents, depending on the fluid and the gases present in the system as well as the intensity of the sound field in operation.[5]

Solutes can also exert considerable influence on the intensity of multibubble sonoluminescence (MBSL) generated in a fluid. In water, for example, some solutes can enhance the intensity of the emission whereas others may significantly quench the signal.[12-14] The reasons for the effects are complex, but in recent years some insight into how certain solutes influence the sonoluminescence (SL) generated in aqueous solutions has been gained. The influences of the solutes on the SL have been ascribed to both inter- and intrabubble events within a cluster of multibubbles. Studying the effects of a range of solutes on single-bubble SL has made it possible to distinguish between interbubble and intrabubble events in aqueous multibubble systems.[15-18]

The influence of solutes on SL generated in nonaqueous fluids, or mixed solvents, has received relatively little attention. In this study we present results on the effects imparted by low molecular weight aliphatic alcohols and surfactants on the MBSL generated in ethylene glycol and in ethylene glycol/water mixtures. Ethylene glycol was chosen as it is completely miscible with water in all proportions, and as such allows the

effects of solutes on MBSL to be monitored over the complete range of aqueous to nonaqueous environments. The additives have, in a sense, been used to "probe" the solutions examined to learn more about the physical behavior of multibubbles in a sound field in a nonaqueous environment. The alcohols have been chosen for their character of volatility, a property that is known to influence the intrabubble properties of cavitation bubbles. The charged and neutral surfactants have been selected because they can adsorb to cavitation bubbles and so affect interbubble interactions. Understanding the effects solutes have on cavitation bubbles is of basic use for optimizing and controlling sonochemical reactions in multicomponent systems of interest.

EXPERIMENTAL DETAILS

Chemical Reagents. The organic solvent ethylene glycol (EG; AR grade, BDH Chemicals) was used as received. Milli-Q water was used with a conductivity of less than 10^{-6} S cm^{-1} and a surface tension of 72.0 mN m^{-1} at 25 °C. All alcohols used were analytical grade reagents: 1-propanol (BDH Chemicals), 1-butanol (Ajax Chemicals), 1-pentanol (BDH Chemicals), and 1-hexanol (BDH Chemicals). The anionic surfactant sodium dodecyl sulfate (SDS) (special purity grade >99%, BDH Chemicals), cationic surfactant cetylpyridinium bromide (CPBr) (Tokyo Chemical Industry), and zwitterionic surfactant N-dodecyl-N,N-dimethyl-3-ammonio-1-propanesulfonate (DAPS) (Calbiochem) were used as received.

Multibubble Sonoluminescence (MBSL) Intensity Measurements. A hollow bottom cylindrical Pyrex vessel attached with a flat plate transducer, of diameter 35 mm, was used to hold 25 mL amounts of solution. An Undatim 515 kHz generator powered the transducer. The detection unit was an Hamamatsu end-on photomultiplier tube (PMT). The apparatus was contained in a sealable blackened metal box to

shield the PMT from background light. The emission signals received by the PMT were transferred to a LeCroy 7242 oscilloscope. The digital output was recorded and processed on a personal computer.

Sonication of solutions was conducted in pulsed mode (duty cycle of 1:3; 3.5 ms on: 10.5 ms off) to prevent excessive heating of solutions during sonication and to obtain information about the temporal build up of the luminescence intensity. The temperature of the solutions remained in the range of 23−26 °C during a typical measurement period. The power delivered to the solutions was <0.4 W/cm^2. All experiments were conducted in an air atmosphere. To ensure uniformity and reproducibility of the experiments, the solutions were first sonicated for 30 s then allowed to stand for 2 min prior to data collection. Data were collected for 50 pulses and averaged. Standard reference SL experiments were periodically conducted to ensure that conditions remained constant during the sonication of the solutions under examination. Under the air-saturated conditions used in the experiments the SL spectrum in the range of 300−800 nm was featureless and had essentially the same profile in both EG and water. This condition meant we were able to measure the integrated SL generated by the ultrasound pulse as representing the SL spectrum and changes to it in the presence of the additives used.

■ RESULTS

MBSL in Ethylene Glycol/Water Mixtures. The SL intensity as a function of the percentage by volume of ethylene glycol/water is shown in Figure 1. The relative higher signal
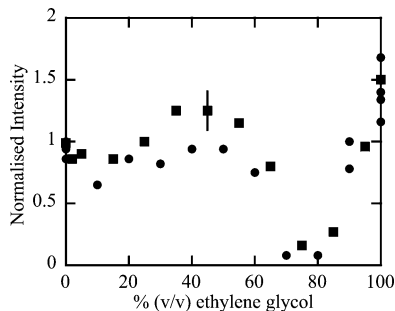

Figure 1. Normalized SL signal (relative to the signal in Milli-Q water) in air-saturated ethylene glycol/water mixtures. The SL signal was an integration of the emission over the entire 3.5 ms pulse. (The data points (●, ■) represent experiments conducted on two separate days. Error bar shown is typical for all data points.).

strength of the SL in pure ethylene glycol compared with that in pure water is consistent with that previously reported.[3] The minimum at about 70 to 80% (v/v) EG/water will be discussed later, although it should be noted that it does not correlate with any singular liquid property, such as gas solubility minimum, viscosity maximum, vapor pressure maximum, or absorption of sound, at this composition. (It is relevant to point out here that the absorption of sound by the liquid mixtures studied increases almost linearly between pure water and pure EG.[19] Approximate calculations on the 515 kHz sound passed through the systems indicate that a maximum of between 3% (water) and 17% (EG) of the sound is absorbed. This would suggest that direct sound adsorption by the liquid is not responsible for the behavior seen in Figure 1.)

In order to help explain the observed behavior, a series of experiments were undertaken using a variety of solvent soluble organic solutes. The concentration ranges of the solutes used were sufficiently low (<1 wt %) so as not to significantly affect the bulk properties of the solvent.

MBSL in Ethylene Glycol/Water Solutions in the Presence of Aliphatic Alcohols. The variation in the relative SL intensity as a function of 1-butanol concentration in solutions of different ethylene glycol/water composition is shown in Figure 2. All the signals have been standardized to the maximum intensity in the particular solvent system being examined.
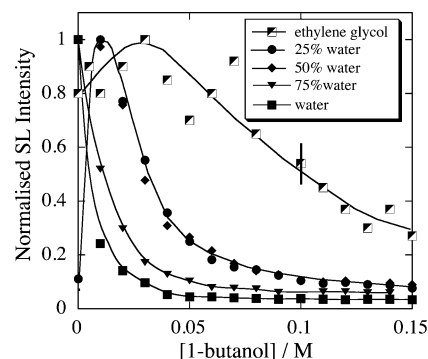

Figure 2. Normalized SL signal (relative to the maximum signal in a particular 1-butanol/solvent system) as a function of 1-butanol concentration in various water/ethylene glycol solvent systems. The lines drawn are only for visual guidance. No line has been drawn through the 50% (v/v) data. The maximum signal for this system is at zero 1-butanol concentration. (Error bar shown is typical for all data points.).

Several trends can be readily observed. Perhaps the most noteworthy being that as the percentage of ethylene glycol in the solvent mixture increases an increasingly higher concentration of 1-butanol is required to reduce the SL signal by an equal amount. Also, at ethylene glycol levels of above about 50% (v/v) there is a lag in the onset of the SL quenching as the 1-butanol concentration increases. The extent of this induction region increases as the ethylene glycol percentage increases.

The effect on the SL intensity on adding different alkyl chain length alcohols at 50% (v/v) EG/water is shown in Figure 3. It
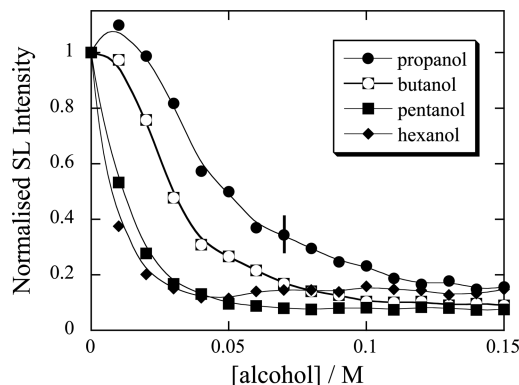

Figure 3. Normalized SL signal (relative to the signal in the solvent in the absence of alcohol) as a function of 1-alcohol concentration. The solvent was 50% (v/v) EG/water. The lines drawn are only for visual guidance. (Error bar shown is typical for all data points.).

can be seen in this figure that as the molecular weight of the alcohol increases the relative extent of the SL quenching becomes more pronounced for any specific concentration of alcohol. This behavior is quite similar to that reported in pure water systems.[12,17]

MBSL in Ethylene Glycol/Water Solutions in the Presence of Surfactants. The SL intensity as a function of surfactant concentration as well as with different head groups was investigated in ethylene glycol/water mixtures, and the results are presented in Figure 4. In all cases it can be seen that the SL signal is enhanced in the presence of surfactant reaching a common level of enhancement at higher concentrations.
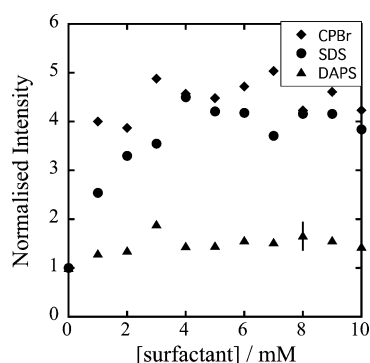

Figure 4. Normalized SL signal (relative to the signal in a 50% (v/v) EG/water solvent mixture) as a function of various surfactant concentrations. (Error bar shown is typical for all data points.).

The effect of SDS on the SL signal in solutions of different ethylene glycol/water compositions is shown in Figures 5 and
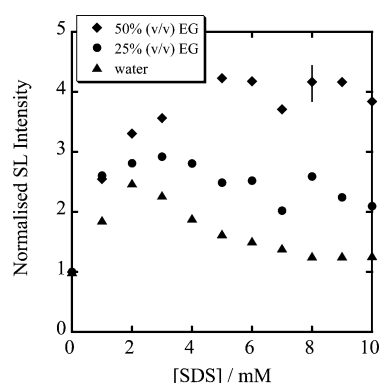

Figure 5. Normalized SL signal (relative to the signal in a particular EG/water solvent system) as a function of SDS concentrations in various EG/water solvent systems. (Error bar is typical for all data points.).

6. It can be seen that in water and below 50% (v/v) mixtures, following the initial rise in the signal intensity, there is a decline in the SL signal at SDS concentrations above about 2 mM. This has been previously noted and is attributed to the change in the electrostatic interactions between charged bubbles as the ionic strength of the solution is raised.[12−14] This will be discussed in greater detail later. In neat ethylene glycol the SL intensity increases with added surfactant, reaching a maximum at around 10−20 mM, and then remains approximately constant at concentrations above that amount. There is also little, if any, effect of surfactant headgroup type on this behavior (see Figure 6).
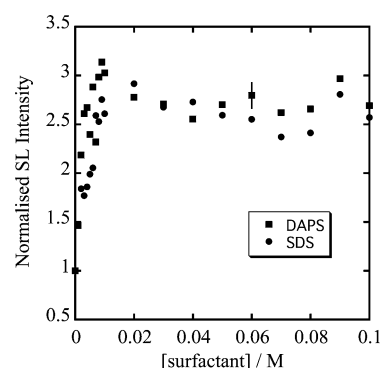

Figure 6. Normalized SL signal (relative to the signal in 100% ethylene glycol) as a function of SDS and DAPS concentrations. (Error bar is typical for all data points.).

■ DISCUSSION

As indicated earlier, the observation that the SL signal in EG is greater than that in water, as seen in Figure 1, has been reported by others.[2,3] A detailed study of the SL signal in mixtures of the two liquids, however, has not been reported before. Considering the level of scatter in the measurements, it is reasonable to make the observation that the SL signal intensity is approximately constant from 0 to about 60% (v/v) ethylene glycol; then at higher EG composition it abruptly decreases and reaches a minimum at around 70 to 80% (v/v) before increasing in intensity again. In 100% EG, the SL signal intensity reaches a level about 1.5 times that obtained in pure water.

In MBSL, for any given level of applied power of ultrasound, the overall signal intensity is due to the product combination of the number of bubbles emitting (i.e., *active* bubbles) and the emission radiated per active bubble. It is also known that the latter is highly dependent on the vapor pressure of the liquid and/or molecular complexity of the gas captured inside an active bubble.[3,4,20,21] The lower the vapor pressure of the solvent, the larger the SL signal. Taking these factors into account, and the two physical properties of EG/water mixtures shown in Figure 7, allows for a plausible interpretation of the intensity variation displayed in the data of Figure 1.
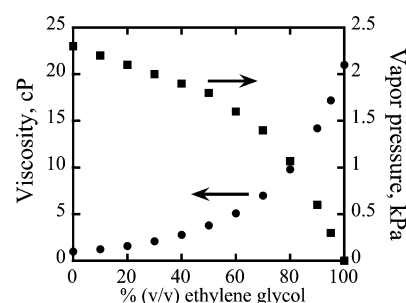
Figure 7. Viscosity (●) and vapor pressure (■) of ethylene glycol/water mixtures at 20 °C. Data taken from www.MEGlobal.biz (product guide on ethylene glycol).

As the viscosity of the mixture increases there will be an increase in the cavitation threshold, i.e., the ability to form active bubbles.[22−24] Therefore, if the applied acoustic power is relatively weak, as is the case in our experiments, there will be a reduction in the population of active bubbles. In addition, the related effect that the increase in viscosity dampens the bubble

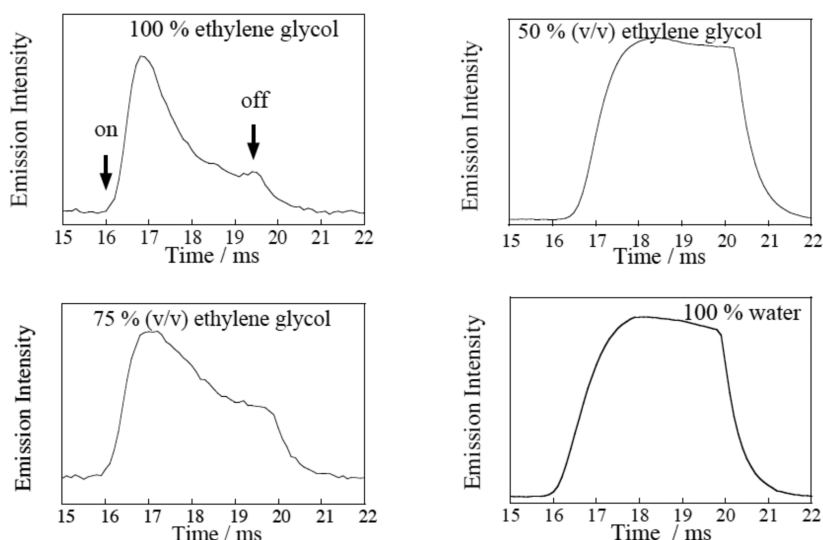**Figure 8.** MBSL pulse shapes generated in different ethylene glycol/water mixtures. The emission intensity is given in nonstandardized arbitrary units. The 3.5 ms pulse "on" and "off" period is shown as an example for the 100% ethylene glycol system. (The slow decay in the SL signal after the power had been switched off is the result of an instrument effect associated with the finite decay in the current supplied to the transducer.).

dynamics (radius−time profile), and in doing so reduces the emission intensity produced in a bubble, can be expected. It has been found that a single bubble in 85% H_2SO_4 (25 cP; cf EG 21 cP) requires a greater driving pressure than a bubble in water (1 cP) to achieve an identical radius−time profile.[20] (However, it must be pointed out that differences in the vapor pressure within the bubble are different in the two systems, and this also affects the bubble dynamics.) Nevertheless, both effects of increasing viscosity with increasing EG content are likely to be responsible for the decline in the MBSL signal seen in Figure 1. Opposing the viscosity-enforced decrease in the SL generated is the significant decline in the vapor pressure of the EG/water mixture as the percentage of EG increases, which will have the effect of enhancing the SL produced within an active bubble. On the basis of the results of Figure 1 it would have to be concluded that the vapor pressure factor begins to dominate at EG percentages above about 75% (v/v), thereby producing the minimum seen in the SL of Figure 1.

This interpretation is *not* what has been previously considered for the increase in the SL signal with increasing viscosity. Young noted the correlation of increasing SL intensity with solvent viscosity, obtained for a number of water/organic liquid mixtures, but simply ascribed the change to a viscosity effect (but unspecified) and did not consider the vapor pressure of the liquids studied.[2] It is relevant to point out that Yasui et al. showed in their theoretical study that increasing the liquid viscosity, under certain conditions, could lead to higher bubble core temperatures on bubble collapse.[24] However, for relatively low acoustic pressures (<3 bar), at both high (300 kHz) and low (20 kHz) frequency ultrasound, and viscosities up to 20 cP, there is little, if any, effect on the intensity of bubble collapse under ambient pressure conditions.[24] Futhermore, at fluid viscosities >20 cP the intensity of bubble collapse decreases. All this supports the contention that the decreasing vapor pressure of the EG/water system at higher compositions of EG in the mixture is responsible for higher core temperatures on bubble collapse and hence more SL per active bubble. This result is the same as is seen in 85% H_2SO_4.[20]

There is one other observation of significance affecting the SL signal that can be ascribed to changes in solvent viscosity.

The change in the SL pulse shape (Figure 8) in going from 50% (v/v) to 100% (v/v) EG reveals that during an actual pulse period the number of active bubbles diminishes. This happens only at solvent percentages above about 65% (v/v) EG. The most plausible explanation for the behavior is associated with changes in the acoustic field once bubble clouds have been formed. Bubble clouds are able to reflect sound, thereby producing a more diffuse acoustic field and therefore weakening the intensity of the sound field propagating through the liquid. The consequence of this, in concert with the increasing viscosity that raises the cavitation threshold, would be to reduce the population of active bubbles. This interpretation is much the same as that given by Henglein and Gutiérrez in their study of MBSL pulse shapes in water at different acoustic powers and external pressure.[25] It is also the same behavior that is seen when the pulse period is lengthened in pure water.[26] In addition, we note that the MBSL signal in glycerine/water mixtures, using continuous wave 21.5 kHz ultrasound at higher acoustic powers than generated by the pulses in the present study, steadily increased in intensity in going from 0 to 100% glycerine, i.e., no minimum in the MBSL was observed.[27] This result is consistent with the suggestion that the higher acoustic power used in the latter study probably maintains an approximately constant active bubble population and the declining vapor pressure of the glycerine/water mixture is the reason for the increase in the SL intensity with increasing glycerine percentage in the mixed fluid.

The trend observed in the quenching of the SL signal by the C_3−C_6 alcohols in Figure 3 is superficially similar to that reported for alcohol quenching of SL in aqueous solutions sonicated at similar frequencies.[12,26] A noticeable difference is that in aqueous solutions considerably lower concentrations of the corresponding alcohols are required to achieve the same degree of SL quenching as seen in Figure 3. In addition, the concentration onset effect seen with propanol and butanol in Figures 2 and 3 is not seen in aqueous solutions sonicated at acoustic powers higher than used in the present study.[12] However, the latter behavior has been observed when using lower applied powers (<0.8 W/cm^2) more comparable to that used in this study (∼0.4 W/cm^2).[28] This onset behavior is

associated with the retardation of bubble−bubble coalescence processes.[29] (More discussion on this will come after dealing with the surfactant data).

The explanation for the SL quenching is that alcohol molecules adsorb to the interface of the bubbles generated by the ultrasound, evaporate into the bubbles during their expansion phase, and are then decomposed during the compression phase to produce hydrocarbon products (methane, ethane, ethylene, etc.). The accumulation of the decomposition products in the core of the bubbles over many oscillation cycles, typical of stable "active" bubbles, results in a decrease of the core temperature of the bubbles, leading to lower levels of SL.[17,21,30] The results of Figures 2 and 3 are largely in keeping with this explanation. The above interpretation is well-supported by SBSL work where intra-bubble SL quenching effects by alcohols are clearly observed.[15−18] Alcohols also hinder bubble coalescence,[31,32] an interbubble process which can be expected to increase the number of active bubbles in the system; however, from our studies it is clear that SL quenching by the alcohols is the dominant process that controls the net SL intensity produced.

The decrease in the extent of SL quenching of 1-butanol with an increase in percent of ethylene glycol, seen in Figure 2, can be attributed to a decrease in the amount of butanol adsorbed at the bubble interface. The higher the ethylene glycol content of the solvent the greater will be the solubility of the butanol in the bulk mixed solvent; therefore, a relatively lower amount is adsorbed at the bubble−solution interface. This is typical behavior of surface-active solutes in EG/water mixtures.[33]

A stylized diagram encompassing the features of the above discussion is given in Figure 9. The diagram depicts bubble
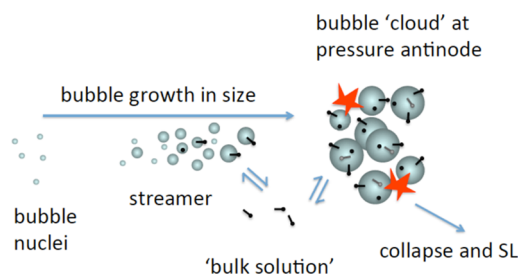

Figure 9. An overview-diagram depicting the growth of acoustic bubbles, from bubble nuclei to the formation of multibubble clusters, in the presence of surface-active solutes (−●). The dark gray units within the bubbles represent volatile solutes drawn into the core during bubble expansion.

nuclei growing in a sound field to become active bubbles, adsorbing surface-active solutes, bubble clustering, and solutes being drawn into expanding bubbles to be decomposed when the bubbles collapse.

However, not all of the features of the results in Figures 2 and 3 can be qualitatively accounted for by the explanation given above. In particular, as ethylene glycol is itself a low molecular weight alcohol with a room-temperature vapor pressure similar to that of 1-octanol, it is noteworthy that there is no SL quenching observed when ethylene glycol is added to water until about 70% (v/v) (see Figure 1). As already discussed, this decrease can be attributed to factors associated with changes in the solvent composition. Moreover, the apparent lack of SL quenching by ethylene glycol is not due to its poor adsorption disposition at the air−water interface. We

have previously investigated MBSL quenching by aliphatic diols in aqueous solutions and concluded that because of their greater degree of hydrogen bonding with water compared with aliphatic alcohols, the rate of evaporation of diols into an expanding bubble (Figure 9) is not significant over the lifetime of a stable active bubble.[34] In essence, it can be concluded that the thermal decomposition of diols does not occur to any significant extent, nor does the accumulation of hydrocarbon products over multiple oscillation cycles of stable active bubbles. Hence, EG does not quench the SL of stable active bubbles as is found to be the case for aliphatic alcohols.[12,17,34]

One other obvious difference with the results of Figure 3, compared with SL quenching by aliphatic alcohols in water, is the apparent residual SL signal at higher alcohol concentrations (>0.1 M). This is particularly noticeable for the C_4 to C_6 alcohol-containing systems. This result suggests that there is a significant population of transient active bubbles in the EG/water mixture that are active only for a few cycles and do not accumulate decomposition products and therefore do not affect the SL produced. This type of bubble population has been observed previously when water is exposed to 20 kHz ultrasound, but not at higher frequencies under such operating conditions as used in the present study.[26,35]

The resultant effects of the surfactants on MBSL shown in Figures 4 to 6 are similar to those seen previously when these types of surfactants were added to water.[12,14] The rise in MBSL with the addition of the ionic surfactants SDS and CPBr to the mixed solvent seen in Figure 4 is consistent with these charged surfactants adsorbing to active bubbles, thereby creating an electrical double layer, and then through interbubble electrostatic interactions retarding bubble−bubble coalescence and reducing the density of bubble clouds. Both situations will have the effect of increasing the population of active bubbles and therefore also the MBSL signal. Further evidence that the increase in the MBSL signal is due to interbubble electrostatic interactions comes from the DAPS data shown in Figure 4. DAPS is a net neutral surfactant and when adsorbed at the bubble/solvent interface cannot exert any long-range electrostatic interactions between bubbles.[12] Hence this surfactant does not have the same pronounced effect on the MBSL signal as seen for SDS and CPBr.

The change in the shape and signal intensity of the MBSL in Figure 5, as a function of SDS concentration, as the EG percentage in the solvent mixture is changed from 0 to 50% is also consistent with interbubble electrostatic interactions affecting the active bubble population. The 100% water results seen in Figure 5 show a maximum in the MBSL signal. This maximum can be ascribed to two opposing effects. Increasing amounts of added SDS will increase the amount of adsorbed anionic surfactant on the active bubbles; these bubbles will gain electrostatic charges and repel each other. The consequence of this has already been explained above. However, the ionic strength of the solution will also rise (SDS is a strong electrolyte), and this will have the effect of reducing the electrostatic repulsion between bubbles. At SDS concentrations >2 mM the latter effect dominates, hence the behavior seen. In support of this interpretation, as has been previously shown, adding an electrolyte, such as NaCl, to 2 mM SDS solutions reduces the MBSL signal to a constant value at all concentrations of SDS in the range of Figure 5.[12]

The data in Figure 5 reveal a decreasing influence of the ionic strength effect as the percent of EG in the solvent mixture increases. Also, in 100% EG the SDS data are virtually identical

to the neutral headgroup DAPS results. This trend can be explained by noting that in progressing from 100% water to 100% EG the dielectric constant of the solvent changes from 78 to 38. This condition favors the ion pairing of Na^+ and $CH_3(CH_2)_{11}SO_4^-$, effectively electrostatically neutralizing SDS in 100% EG, and thereby eliminating interbubble electrostatic interactions.[33] That there is still significant MBSL enhancement observed (Figure 6) even when interbubble electrostatic interactions are not operating is not surprising, and the reasons for this have been considered in some detail elsewhere.[29] In brief, the adsorption of solute molecules (neutral or charged) at the acoustic bubble interface retards the fluid flow in the thin film between two bubbles as they approach each other. This change in flow conditions hinders bubble—bubble coalescence processes, and as mentioned above, this may increase the number of active bubbles in the system and therefore the MBSL signal.

The possibility that the free monomer surfactant concentration is limited by micelle formation, and so affect the maximum in the sonoluminescence observed in the systems examined, can be dismissed. For the SDS case, for example, the critical micelle concentration (cmc) is 8 mM in water, 12 mM in 25% EG, 25 mM in 50% EG, and 67 mM in pure EG.[32] All the surfactant concentration effects that are observed are below the cmc of SDS in the systems studied. Also, surfactant adsorption can be expected to change the interfacial tension of acoustic bubbles and in doing so affect bubble coalescence behavior. However, as we have previously noted the effect must be relatively minor compared with the other effects in play when surfactants adsorb to the bubble interface.[12]

Finally, on the basis of the above, the MBSL maxima seen in Figures 2 and 3 can now be ascribed to a combination of adsorbed alcohol molecules hindering bubble coalescence, hence increasing the number of active bubbles, and alcohol molecules evaporating into stable active cavitation bubbles, resulting in the quenching of SL, that is, competing effects of interbubble and intrabubble processes. The actual amount of alcohol molecules adsorbed to a cavitation bubble interface will depend on the surface activity of the alcohol, which in turn will depend on the concentration of the alcohol, its alkyl chain length (hydrophobicity), and the polarity of the mixed solvent. This mix of factors will govern the extent and position of the SL maxima seen in Figures 2 and 3.

CONCLUSIONS

It can be concluded that both volatile and nonvolatile surface-active solutes have a similar effect on acoustic bubbles in EG as they have on bubbles in aqueous solutions. Volatile solutes (e.g., low molecular weight aliphatic alcohols) quench MBSL in EG by the same intrabubble processes that take place within bubbles in water. The adsorption of solutes at the cavitation bubble—solution interface results in interbubble effects in EG that are similar to those in water, and gives rise to the enhancement of MBSL. The most significant difference between ionic surface-active solutes in water and EG is that in pure EG interbubble electrostatic interactions between bubbles are essentially eliminated. This is attributed to the enhanced level of ion pairing between surfactant ions and their counterions in EG because of its lower dielectric constant (lower polarity) compared with water. The small differences in the effects on the MBSL signal of the solutes as a function of concentration in EG compared with water arises because surface-active solutes, at any particular concentration, will

adsorb to a lower extent to the bubble—solution interface in EG than to bubbles in water. This simply means that higher levels of solute are required in EG to affect the same degree of inter- and intrabubble events as in water. The effect of the higher viscosity of EG seems to primarily affect the number of active bubbles produced, but this can, in principle, be overcome by using higher acoustic powers. The lower vapor pressure and the slower evaporation rate of EG are responsible for the greater SL intensity generated in pure EG, compared with that in pure water, despite the relatively higher viscosity of EG.

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Notes

The authors declare no competing financial interest.

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