Communication

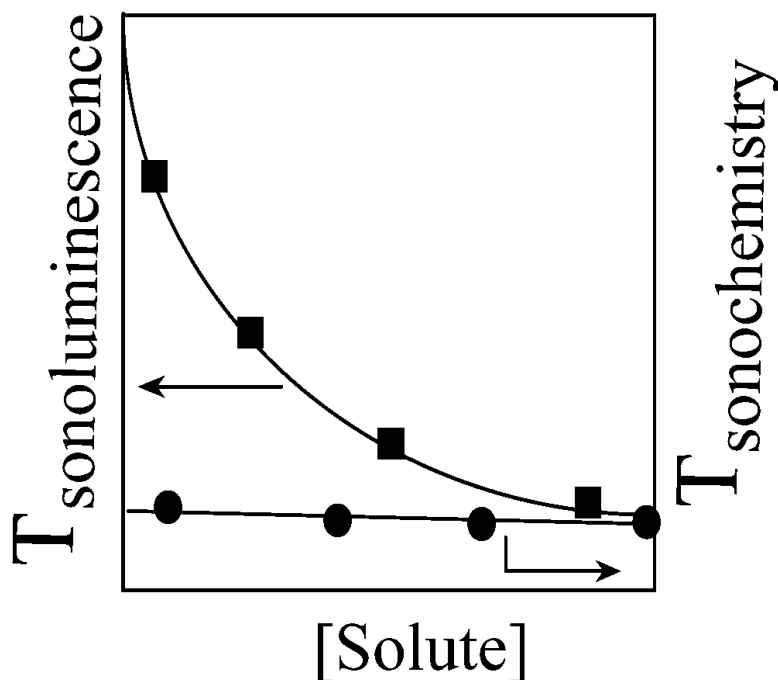# A Comparison between Multibubble Sonoluminescence Intensity and the Temperature within Cavitation Bubbles

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A Comparison between Multibubble Sonoluminescence Intensity and the Temperature within Cavitation Bubbles

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Acoustic cavitation, under certain conditions, is accompanied by the emission of light, commonly referred to as sonoluminescence (SL).[1–3] It is generally accepted that SL arises as a consequence of the high temperatures generated within inertially collapsing microbubbles, known as "hot spots".[1–3] Theoretical modeling results have arrived at maximum bubble temperatures (T_{max}) of around 20,000 K based on a symmetric, and near adiabatic collapse of these bubbles.[1–5] Whereas temperatures of this magnitude may be realized for a single, stable bubble levitated in an ultrasonic field, experimental investigations to date on multibubble clouds report more modest cavitation bubble temperatures, mostly in the range of 2,000–5,000 K.[5–9]

There are a number of theories in the literature on the origins of SL, with perhaps the two most convincing being that the light comes from the relaxation of a highly coupled plasma and/or overlapping emission bands from electronically and vibrationally excited chemical species. In either case, extreme temperatures are required to produce the precursors to the emission. To identify whether there is a direct relationship between the bubble temperature and the SL intensity, we have experimentally determined the cavitation bubble temperatures in aqueous solutions containing various ethanol concentrations and compared these with the SL intensities from the same solutions. To our knowledge, this is the first time such a comparison between the cavitation bubble temperature and the relative SL intensity has been investigated.

The ultrasonic generator/transducer/cell arrangement (frequency = 356 kHz) used for both T_{max} and SL measurements was similar to the one described elsewhere.[10,11] SL data were collected in argon-saturated aqueous solutions following a procedure similar to that reported earlier.[10] For the cavitation bubble temperature measurements, argon-saturated aqueous solutions containing various quantities of ethanol were sonicated in sealed vials.[11] The gaseous products generated by sonication were collected in the "headspace" of the vials and were quantitatively analyzed by GC. The yields of the gaseous hydrocarbon products (ethane, ethylene, and acetylene) were used to calculate the T_{max} using the procedure given by Hart et al.[6] and Tauber et al.[9] We have recently extended this method (referred to as the methyl radical recombination (MRR) method) to estimate the cavitation bubble temperature in the presence of a number of alkyl alcohols.[11]

Figure 1 shows the relative MBSL intensities and cavitation bubble temperatures observed in argon-saturated aqueous solutions, containing different concentrations of ethanol. The temperatures measured by the MRR method are within the range of values measured by other techniques.[5–9] The salient features of the data are that the MBSL intensity decreases substantially with increasing alcohol concentration, whereas within experimental error, the temperature remains essentially constant, 4200 ± 200 K. This significant decrease in the SL intensity from water in the presence of ethanol has been previously reported.[12] Our recent investigations[10–14] on multibubble systems have shown that, in general,
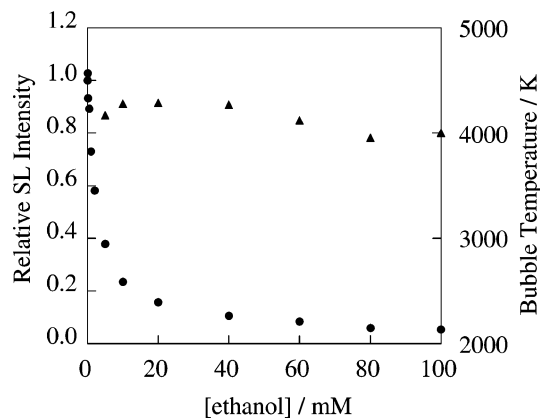

Figure 1. Relative SL intensities and estimated maximum bubble temperatures in argon-saturated aqueous solutions containing lower concentrations of ethanol at 356 kHz.

volatile surface active solutes decrease the SL intensity observed in air-saturated water at high ultrasonic frequencies.

In our SL quenching studies,[10–14] we have proposed that SL quenching by volatile solutes is due to the decrease in the cavitation bubble temperature as a consequence of the endothermic chemical decomposition reactions of the solutes that have evaporated into the bubble core, as well as the presence, and probable continuous decomposition, of accumulated hydrocarbon products within the bubbles, over a number of acoustic cycles.[15] The recent experimental work of Guan and Matula[16] and the theoretical studies of Yasui[5] and Tögel et al.[17] all support our interpretation of SL quenching by alcohols in aqueous solutions.[18]

The data in Figure 1, at first sight, do not appear to be consistent with the interpretation given for the quenching of SL that was briefly described above. A more detailed interpretation of the data of Figure 1 can be made by considering what the two types of measurements actually report, in conjunction with what is known about the heating of the bubble core during the bubble collapse process.

For the case of single bubbles, theoretical calculations indicate that during the early stages of bubble collapse there is little difference between the temperature of the gas/vapor in the bubble and the surrounding liquid. At a collapse radius below the ambient radius of the bubble (R_o), the center of the bubble begins to heat up almost adiabatically.[19] Beyond R_o, a rising temperature gradient is established over many nanoseconds, with the core of the bubble reaching temperatures sufficiently high to lead to ionization of the gas/vapor therein. It is also known that SL only occurs at the very last stages of bubble collapse, and that light emission occurs just prior to the minimum radius being reached.[20] The calculations also show that the peak temperature lasts for less than 1 ns. (Although the theoretical treatments are for single bubbles, the same will be the case for bubbles in a multibubble field with the exception that the peak temperature within these bubbles will not be quite as high as for a single isolated bubble collapse.)[19] What is less clear is

whether a shock wave precedes the collapsing wall of the bubble. Both models have been suggested.[21,22] The significance of a shock wave front is that if it is directed at the core of the bubble, a higher peak temperature would be realized as well as a more pronounced temperature gradient between the very core of the bubble and the wall of the bubble. Yasui's model, on the other hand, predicts that no shock wave exists, and that as the temperature of the core increases on collapse, it is uniform throughout the whole core with only a drop off at temperature at the bubble wall.[21]

The temporal temperature characteristics are important in understanding the results of Figure 1. The methyl radical recombination method that has been used[5,9] to determine the bubble temperature provides an average value of the temperature within the bubble. That is, the formation of hydrocarbon products occurs throughout the volume of the core during the time where and when the temperature is sufficiently high for bond breakage of the solute and hydrocarbon products to occur. SL, on the other hand, occurs only near the final stages of collapse. The two temperature "indicators", therefore, report the bubble core temperature over two substantially different time and volume regimes.

The energy required for C−C bond breakage is of the order of 350 kJ/mol, whereas ionization of gaseous hydrocarbons or water vapor is of the order of 1000 kJ/mol. The data of Figure 1 would suggest that the presence of a small amount of solute/hydrocarbon products has a much larger effect on the short time span ionization processes (during the peak temperature conditions) than on bond breaking that occurs over a longer heating time within the bubble core. The implication is that a small amount of hydrocarbon product in the bubble core lowers the maximum temperature more dramatically than the average bubble temperature, or alternatively, a small change in the peak bubble temperature has a dramatic effect on the ionization and/or excitation processes leading to SL. It is likely that it is a combination of the two that affects the SL intensity.

It is also known from our experiments that there is an increase in the amount of gaseous products with an increase in alcohol concentration, for any given period of sonication of the aqueous solution. On the basis of this observation, it can be anticipated that at alcohol concentrations higher than those used in Figure 1, the mean temperature would be decreased by the presence of a larger quantity of the solutes/gaseous products within the bubble. Further experiments were carried out at higher ethanol concentrations to confirm this, and the results are shown in Figure 2.

The data shown in Figure 2 are consistent with our interpretation of the effect of volatile solutes on the core temperature of an imploding bubble.

In conclusion, it can be said that the two temperature "indicators" examined in this study probe different time and volume regions of a collapsing bubble. The MRR method is clearly not a suitable method for gaining access to the temperature in the volume region within a bubble where SL occurs. The *true "SL temperature"* can be expected to be several thousand degrees higher than the *average* estimated values. The MRR method, by its very nature, does however provide a temperature at which sonochemistry is taking
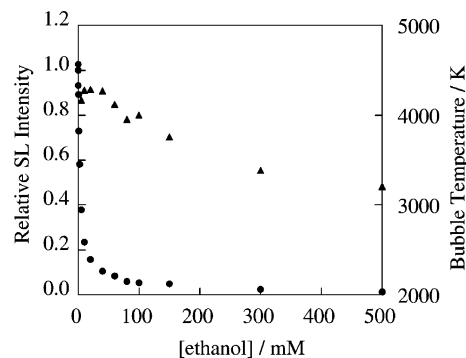

Figure 2. Relative SL intensities and estimated maximum bubble temperatures in argon-saturated aqueous solutions containing higher concentrations of ethanol at 356 kHz.

place, and this will occur even if no SL is detected from the solution. Two other studies[23,24] also support this latter conclusion for both single bubble and multibubble systems.

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