

Frequency Effects on Sonoluminescence of Alkali-Metal Atoms in Sulfuric Acid

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ABSTRACT

Intense orange Na^* emission was observed in different spatial locations from blue emission during multibubble sonoluminescence in sulfuric acid. The color change from blue to orange was observed along the streamer in the filamentous structure of a bubble cloud. By stroboscopic observation, the Na^* emission seemed to occur when a large bubble ejected tiny bubbles at the positions toward a pressure node after bubble coalescence around a pressure antinode. The intensity of Na^* emission in the sulfuric acid increased at lower frequency in contrast with that in water. By comparing a high-resolution Na^* spectrum of sulfuric acid with that of water, the widths of the spectra were almost the same, except for something being superimposed in the water case. The estimations of the temperature and pressure inside the Na^* emission bubbles at 28 and 150 kHz were 1900 K and 100 atm and 2200 K and 150 atm, respectively.

INTRODUCTION

Recently, extremely intense sonoluminescence in sulfuric acid has been discovered in both single bubble sonoluminescence (SBSL) [1-2] and multibubble sonoluminescence (MBSL) [3], which is 1500 times brighter than previous sonoluminescence in water. In sulfuric acid, new emission lines of noble gas atom emission and molecular (SO) and ionic (O^{2+}) emissions were also observed. These studies have given many experimental lines of evidence of theoretical studies and have continued to provide further insight into the physical conditions and chemical processes occurring during SBSL and MBSL [4-14].

One of the most important discoveries of sonoluminescence in sulfuric acid for sonochemistry is the emission from electronically excited metal atoms during SBSL [7] and MBSL [14]. Flannigan and Suslick [7] has provided further insight into how nonvolatile metal cations become heated in a collapsing bubble. In that paper, the observations supported the hypothesis that nonvolatile species can only enter the bubble when the macroscopic bubble motion becomes sufficiently chaotic for energetic surface oscillations to develop and thus entrain liquid droplets in the bubble interior (illustrated as an injected droplet model [7]).

Alkali-metal atom emissions during MBSL are becoming more important in sonochemistry, because Sunartio et al. have suggested that Na^* emission arises from sonochemically active bubbles that are not producing sonoluminescence [15]. The temperature and pressure inside the bubbles that emit alkali-metal emissions were reported by Sehgal et al. [16], but doubted by Flint and Suslick [17]. Although there are many papers concerning the emission from excited alkali-metal atoms observed during MBSL [15-20], the physics and chemistry are still not well understood.

In this study, we have examined alkali-metal emissions during MBSL in sulfuric acid with sodium sulfate, in comparison with those in aqueous solutions. The present study may provide further insight into the origin of alkali-metal emissions, because the extremely intense emission of excited sodium atoms, as well as the intense continuum, can be observed separately. Moreover, the bubble dynamics is relatively slow in the translational motion of bubbles due to the higher viscosity of sulfuric acid than that of water.

EXPERIMENT

The sulfuric acid (H_2SO_4) used was of ultrapure grade (purity: 97.0-98.0%) and sodium sulfate (Na_2SO_4) and sodium chloride were of special pure grade (min. 99.0%). They were purchased from Kanto Chemical Co. Inc., Japan and used as received. H_2SO_4 into which Na_2SO_4 powder was added was degassed under vacuum with stirring and then left under Xe or Ar gas atmosphere for a sufficient time to dissolve the gases. The Na_2SO_4 concentration in sulfuric acid was 1 mol/L, whereas the concentration of NaCl aqueous solution was 2 mol/L; the concentration of the Na^+ cation was the same in both the sulfuric acid and aqueous solutions.

The experimental setup is shown in Fig. 1. Two different experimental apparatus and three different cells were used. Figure 1(a) shows the experimental setup for the simultaneous measurement of MBSL spectra and stroboscopic observation of cavitation bubbles in 1 M sodium sulfate sulfuric acid solution under Xe, which will correspond to the results in Figs. 2-4. Figure 1(b) shows the experimental setup for the comparison of the high-resolution spectra of MBSL under Ar in sulfuric acid solution with those in aqueous solution, which will correspond to the results in Figs. 5-10.

