Bubbles and Crystals: Time-Resolved Sonoluminescence, Sonocrystallization, and Sonofragmentation

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The application of high-intensity ultrasound on liquids induces acoustic cavitation: the formation, growth, and implosive collapse of bubbles. In a standing wave a single bubble can be trapped. The conditions formed inside such a bubble during collapse are extreme, reaching temperatures on the order of 15,000 K and pressures of over a thousand atmospheres.[1-3] These conditions are extreme enough to excite gas molecules in the bubble, resulting in light emission (single-bubble sonoluminescence, or SBSL).

To extract information about conditions during cavitation, both atomic emission lines (usually from noble gases[1-3]) and the underlying broad continuum[4-5] present in SBSL have been used as spectroscopic thermometers. Unfortunately, these experiments divulge only time-averaged information, whereas the evolution of the conditions created during the collapse of a single bubble has been largely unexplored and the few reports concerning the time evolution of collapsing bubbles are conflicting and problematic.[6-7]

The extremely bright SBSL achievable in sulfuric acid (hundreds to thousands of times more intense than in water[2]) and long emission lifetimes (typically several nanoseconds at low ultrasonic frequencies, compared to as short as 50 picoseconds in water[8]) permitted the acquisition of high-quality time-resolved spectra (Figure 1).

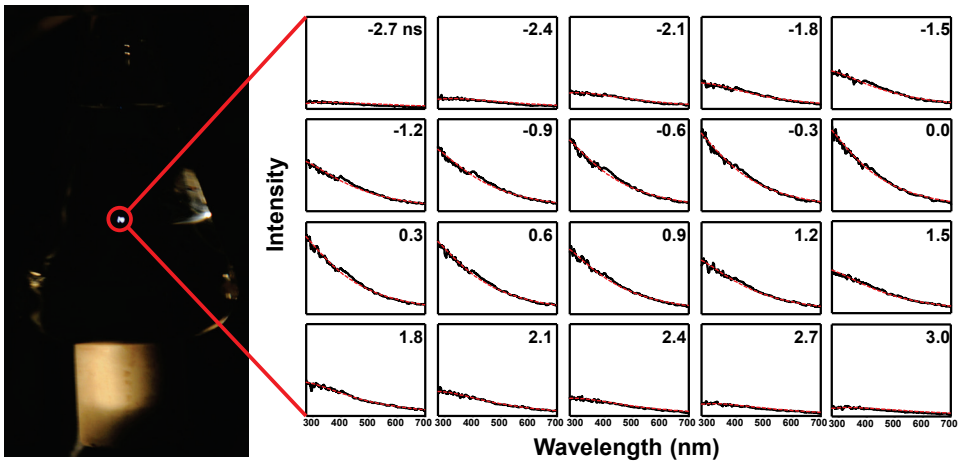

Figure 1. Time-resolved SBSL from a xenon-filled bubble driven at an acoustic pressure of 1.9 bar. Black lines are experimental data, red lines show quality of blackbody fits.

The time-resolved spectra from SBSL in sulfuric acid were consistent with a compressionally-heated bubble and blackbody emission from an opaque plasma. The temperature profile ranged from ~8,000 to 14,000 K (based on the blackbody fits in Figure 1)

with approximate temporal symmetry (the emission during bubble rebound was slightly longer than during collapse). Spectra were qualitatively insensitive to changes in dissolved gas content, resonator geometry, and acoustic driving pressure.

High-intensity ultrasound is also employed in the preparation of molecular crystals, although the field remains in its infancy.[9] The effects of high-intensity ultrasound on the preparation of pharmaceutical compounds are explored, with emphasis on the breakage of particles post-crystallization. A model compound, aspirin, was crystallized under ultrasonic irradiation and particle morphology and crystal size distributions were compared with other crystallization methods.

Particle breakage during sonication of liquid/aspirin slurries was investigated, establishing relationships between particle size and sonication time or sonication intensity. Particle size distributions were modeled using a population-balance method assuming simple binary breakage events. The importance of liquid viscosity and vapor pressure was also established, with the former strongly influencing breakage rates and the latter being negligible.

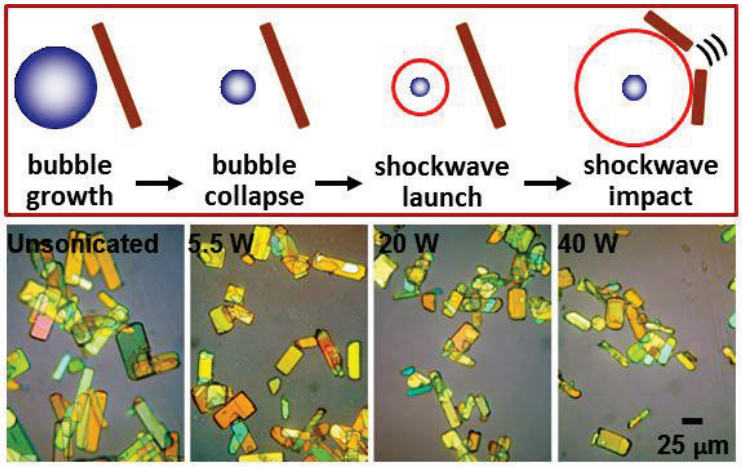

Figure 2. Top: mechanism of ultrasound-induced particle breakage of molecular crystals. Bottom: example of particle breakage by sonicating aspirin crystals for one minute under different ultrasonic power outputs.

The mechanism of particle breakage during sonication was shown to be direct interactions between shockwaves and crystals rather than interparticle collisions (Figure 2), a surprising contrast with superficially similar systems.[10-11] Decoupling and kinetics experiments were performed to rule out particle-particle collisions, particle-horn collisions, or particle-cell collisions as significant contributors.

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