

OC-48: Freezing Conditions in Inertial Cavitation Bubbles During Post-Collapses Expansions

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The rapid collapse of inertial cavitation bubbles are known to produce huge temperatures owing to the nearly adiabatic compression of the incondensable gas. The main collapse is followed by an expansion and several rebounds. The classical simplified model of bubble dynamics accounting for heat and water transport between the liquid and the bubble predicts that the post-collapses expansions are not isothermal, and freeze the bubble center down to several tenth of °C below zero. The water vapour trapped in the bubble would therefore be driven in conditions where ice crystals could nucleate. This opens the way to a possible new mechanism for ice sonocrystallization, which is discussed.

1. Modelling bubble oscillations

When driven by a large amplitude sound field, gas bubbles undergo a large expansion followed by a rapid collapse. During the expansion, water evaporates into the bubble almost isothermally, whereas along the collapse, water and heat remain trapped in the bubble, since the characteristic timescales for both diffusive transports are large compared to the rapid bubble collapse timescale (Storey & Szeri, 2000). This explains why part of the collapse is almost adiabatic and temperatures of thousands of Kelvin are obtained, and why part of the water vapour trapped near the bubble center can undergo these extreme conditions and decompose into radicals. The rigorous treatment of such process requires solving complex PDEs (Storey & Szeri, 2000), but simplified models based on ODEs have been proposed (Yasui, 1997, Toegel et al., 2000; Storey & Szeri, 2001). They are based on the approximate evaluation of the diffusion lengths in the gas near the bubble walls, allowing to estimate the instantaneous heat and mass flux in conservation equations, which are coupled to a standard bubble dynamics ODE. Such models were found to be in reasonable agreement with complete theory (Storey & Szeri, 2001; Stricker et. al., 2011), SBSL experimental results, and, if coupled to chemical kinetic models, even predict some well-known tendencies in sonochemistry basic experiments (Storey & Szeri, 2001).

2. Bubble temperatures during rebound

Using the ODE-based model, we calculated the response of a 5 µm air bubble driven by a 20 kHz acoustic field of amplitude 130 kPa, in water at 25 °C. Fig.1 shows the classical bubble radius-time curve (left graph) and temperature of the bubble core over one acoustic cycle (right graph). The collapse temperature is slightly above 5000 K, and several secondary temperature peaks appear at the end of secondary collapses. Such results are well-known and have constituted the main matter for the discussion on single-bubble sonoluminescence (SBSL).

However, a generally overlooked information is the low part of the graph, where the minimum temperatures attained in the bubble are visible. The inset displays a magnification around the bubble rebounds and evidence negative temperature peaks of several tenth of °C below zero, originating from the bubble re-expansions after the collapses. Since these expansions occur on a rather fast timescale, heat diffusion from the liquid is limited and the gas near the bubble core is cooled efficiently.

More interesting information can be gathered by drawing the closed trajectory described by the bubble interior conditions in the phase diagram of water (T, p_{H_2O}). Figure 2 displays the water partial pressure in the core of the bubble as a function of temperature superimposed with the vapour-liquid and liquid-solid boundaries.

It is seen that the bubble content enters several times in the solid region of the water phase diagram. This indicates that ice crystals may start to nucleate in the bubble core during these excursions. If the liquid is in ambient conditions, such nuclei would probably redissolve if ever formed. However in undercooled liquid conditions, encountered in freezing experiments (Chow et al., 2003), these nuclei would be able to grow, potentially destroy the bubble, and disseminate in the liquid. This conjecture opens the way to a new interpretation of ice crystallization enhancement by ultrasound.

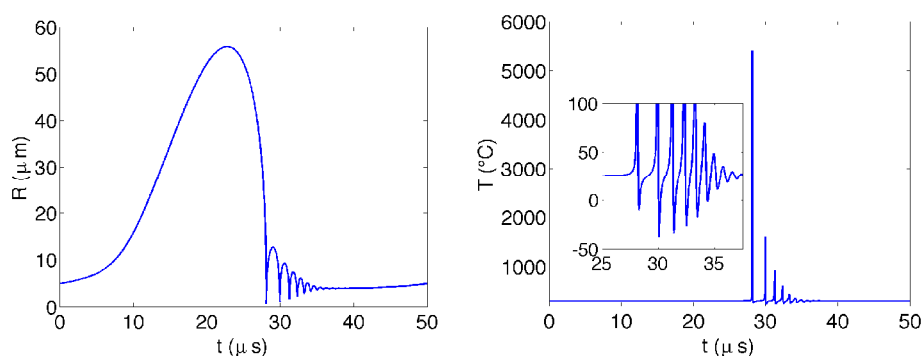


Figure 1: Left : bubble radius vs. time. Right : temperature of the bubble center vs. time. The inset displays a magnification around bubble rebounds.

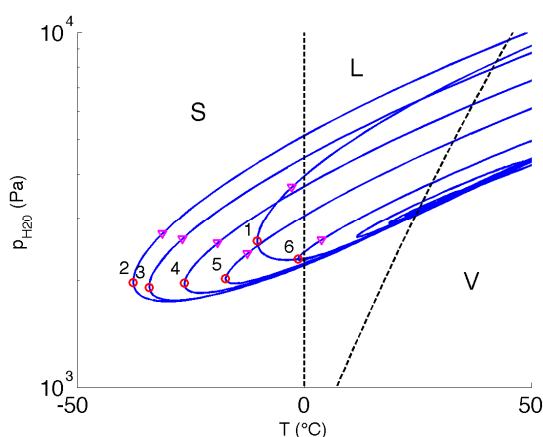


Figure 2: Evolution of water conditions in the phase diagram of water (T , $p_{\text{H}_2\text{O}}$). The circles materialize the six negative peak temperatures visible in the inset of Fig. 1, numbered by increasing time. The triangles materialize the state at a short time before the minimal temperature is attained and are represented to illustrate the direction followed by the trajectories. The dashed lines are the boundaries between the vapour, liquid and solid states of water. The excursion of the trajectories in the solid region are, for paths from 1 to 6 : 114 ns, 205 ns, 213 ns, 207 ns, 199 ns, 73 ns.

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