

# Direct Simulation of Argon Rectification in Single Bubble Sonoluminescence

Brian D. Storey<sup>a</sup> and Andrew J. Szeri<sup>b</sup>

<sup>a</sup>*Franklin W. Olin College of Engineering, Needham, MA 02492–1245*

<sup>b</sup>*Department of Mechanical Engineering, University of California, Berkeley, CA 94720–1740*

While it seems that many of the mysteries around single bubble sonoluminescence (SBSL) are solved, one key question remains, how hot does the bubble really get? Recent computations have reduced the prediction to ~7000 K. This prediction seems to be inconsistent with other well established aspects of SBSL such as the dissociation hypothesis and light emission. This paper will demonstrate that temperatures of 7000 K are not inconsistent with the dissociation hypothesis through direct simulation of argon rectification.

## INTRODUCTION

The quantitative prediction of temperature in the interior of a single, stable, acoustically forced bubble has been evolving ever since SBSL was discovered. While it seems that much of the research community has converged to a consistent understanding of SBSL, there is still an incomplete quantitative prediction of the phenomena.

Recently, the importance of water vapor in the understanding of SBSL has been clarified. Calculations by Moss et al. [1] and Storey and Szeri [2] have shown (independently and through very different methods) that under SBSL conditions the bubble can often be composed of 30–50% water vapor at the time of collapse.

Storey and Szeri [2] included chemical reactions in their simulations showing that the dissociation of the water vapor lowered the prediction of the maximum temperatures from >20,000 K when vapor was neglected to approximately 7,000 K.

There is the commonly held view that the temperature of collapse must be higher than 9,000 K for the dissociation hypothesis to hold true [3]. The hypothesis says that the nitrogen and oxygen in an air bubble will undergo dissociation during the violent collapse when the bubble is hot. The gases will react with water vapor to form soluble species, which will easily diffuse into the liquid water. The result is a net loss of the diatomic components of air and a stable bubble is composed of pure argon. This paper will show through direct simulation that high temperatures are not necessary for the dissociation hypothesis to be valid.

## FORMULATION

In previous work, the authors proposed a simplified cavitation model [4] that took into account inter-species mass diffusion, heat transfer, and

chemistry. This reduced model provided good agreement when compared to a direct numerical simulation [2].

This reduced sonochemical model was originally developed for argon–water but was extended to add air chemistry through a different chemical reaction mechanism (we used a mechanism developed for combustion from [5]). The model [4] was also appended to include the flux of dissolved gases into the liquid. The flux of stable dissolved gas species from the liquid (such as argon, nitrogen, hydrogen, and oxygen) are computed with the long time scale rectified diffusion growth rate from Fyrrillas and Szeri [6]. At steady state, the growth rate from diffusion must match the amount of gas lost to chemical reactions. The argon must be in perfect balance with respect to rectified diffusion.

The very soluble species such as OH, NH<sub>3</sub>, or H<sub>2</sub>O<sub>2</sub> are assumed to be removed proportional to the rate at which they strike the bubble interface. One in ten collisions with the liquid interface is assumed to take in these soluble species [2] (results were insensitive to the rate of uptake by the liquid).

Considering the flux of gases out of the bubble along with the chemical kinetics we were able to search for steady solutions for a given pressure amplitude. The details of this method will not be discussed in detail in the interest of brevity. Once the stable radius and gas concentration were found, the point was shown to be locally attracting by perturbing the solution and watching it move back to the stable point.

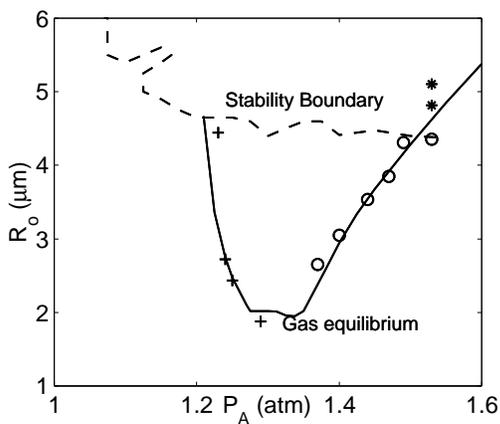
The bubble must also be considered for spherical stability. Finding whether a bubble is stable to spherical perturbations is straightforward and the analysis is covered elsewhere [7]. It was recently shown that the location of the stability boundary is very sensitive to details of the radial dynamics: therefore the complete Navier–Stokes model is used to compute the shape stability boundary [8].

## RESULTS

A frequency of 32,800 Hz and air dissolved at 20% in liquid water were considered in order to model the recent data of Ketterling and Apfel [9]. The computed phase diagram as the pressure amplitude is varied is shown in Figure 1. The solid line shows the ambient radius of the bubble which is stable to mass diffusion. The dashed line shows the shape stability boundary: bubbles below the shape boundary are stable to shape perturbations. The experimental data are shown as points.

The agreement between model and experiment is very striking. The model is able to accurately predict the SBSL phase diagram. There are no new physics introduced in this calculation [3]: this result simply confirms that the understanding of the dissociation hypothesis is correct, the linear stability analysis for shape perturbations is accurate at strong forcings, and temperatures of only  $\sim 5000$  K are adequate for modeling of the dissociation hypothesis.

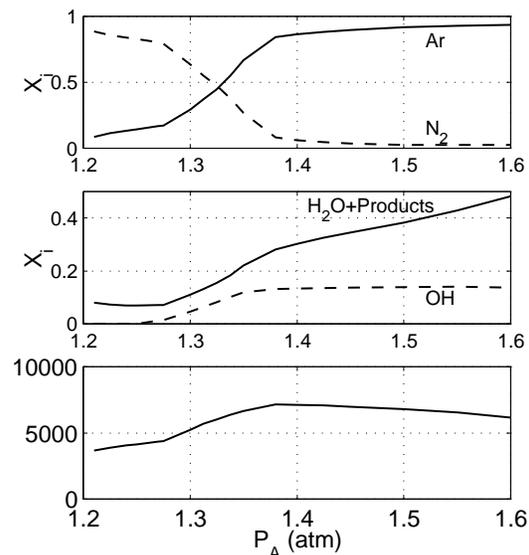
In Figure 2 we show the chemical makeup of the bubble at each pressure amplitude. The mole fraction of the dominant species initially (at the beginning of the acoustic cycle) in the bubble are displayed in the top figure. The transition from a nitrogen to argon bubble is clearly seen. In the middle figure the mole fraction of water vapor and OH are shown at the time of collapse. The temperature of the bubble is shown in the bottom figure. Note from figure 1 that the light emission turns on above 1.3 atm.



**FIGURE 1.** SBSL phase diagram showing chemical-mass transfer equilibrium and parametric shape stability boundary. The "o" points are experimental stable SL measurements. The "+" points show the experimental stable non-SL points while the "\*" points show the unstable SL points. The experimental data from Ketterling and Apfel [9].

## CONCLUSIONS

With respect to the dissociation hypothesis, the sonochemical model predicts all the correct behavior using a complete chemical kinetics reaction mechanism and reasonable heat and mass transfer models. The model also predicts temperatures that are much lower than those needed for currently accepted emission models. Future work could probably resolve the differences in light emission theory with carefully designed experiments.



**FIGURE 2.** Bubble composition and temperature as the pressure amplitude is varied. The top figure is the mole fraction at the ambient state. The middle figure are the fractions of water plus all its dissociated products, and OH at collapse. The bottom figure is the temperature of the gas at collapse.

## REFERENCES

1. W.C. Moss, D.A. Young, J.A. Harte, J.L. Levatin, B.F. Rozsnyai, G.B. Zimmerman, & I.H. Zimmerman, *Phys Rev E*, **59**, 2986–2992, (1999).
2. B.D. Storey & A.J. Szeri. *Proc. Roy. Soc.* **456**, 1685–1709, (2000).
3. S. Hilgenfeldt, D. Lohse, & M.P. Brenner, *Phys Fluids*, **8**, 2808–2826, (1996).
4. B.D. Storey & A.J. Szeri *Proc. Roy. Soc.* To appear, (2001)
5. Gregory P. Smith, David M. Golden, Michael Frenklach, Nigel W. Moriarty, Boris Eiteneer, Mikhail Goldenberg, C. Thomas Bowman, Ronald K. Hanson, Soonho Song, William C. Gardiner, Jr., Vitali V. Lissianski, and Zhiwei Qin [http://www.me.berkeley.edu/gri\\_mech/](http://www.me.berkeley.edu/gri_mech/)
6. M.M. Fyrillas & A.J. Szeri, *J Fluid Mech*, **277**, 381–407, (1994).
7. Y. Hao & A. Prosperetti, *Phys Fluids*, **11**, 1309–1317, (1999)
8. B.D. Storey, *Phys Rev E*. to appear (2001).
9. J.A. Ketterling & R. E. Apfel, *Phys Rev E*, **61**, (2001).