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Determination of Maximum Gas Pressure of Collapsing bubbles in Single Bubble Sonoluminescence in Sulfuric Acid

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ABSTRACT

Single Bubble Sonoluminescence (SBSL) in sulfuric acid is investigated with Hydro-Chemical simulation techniques. Gas pressure of SBSL bubbles at the last stage of the collapse is determined for various concentrations of sulfuric acid.The calculated values of the gas pressure are found to be in the range 18755 atm to 33226 atm. Maximum pressure is calculated near 45% concentration of Sulfuric acid. Special attention has been paid to the sensitivity of physical parameters of SBSL bubble to the numerical techniques used. The maximum temperature of the SBSL bubble is found to be less sensitive to the accuracy of the numerical methods. Furthermore, it was found that the bubble pressure is very sensitive to the accuracy of the numerical methods employed.

1. INTRODUCTION

Single bubble sonoluminescence (SBSL) is a remarkable phenomenon in which periodic light emission can be observed from a violently collapsing gas bubble trapped and driven in a liquid by an ultrasound standing sound field. SBSL was discovered by Gaitan and Crum in 1990 [1]. After this discovery, a large number of experimental and theoretical papers have been published describing unusual characteristics of this phenomenon involving emitted light intensity at the bubble collapse, spectrum of the emitted light, temperature and pressure of the bubble at the collapse, and stability of the SBSL bubble.

Investigation of SBSL is twofold, one is experimental and the other is theoretical. Hydro-chemical model is one of the well-established theoretical methods utilized to examine SBSL. Lohse *et al* [2] have improved the Hydro-chemical model which had been developed by other investigators during last two decades[3, 4, and 5] for studying SBSL in water. This model is based on bubble dynamics, thermo dynamics, and chemical kinetics of the gases and vapours in the bubble and the surrounding liquid. Recently, Moshaii *et al* [6] have applied this model for investigating temperature and concentration dependence of light emission of SBSL in sulfuric acid solutions by adding contributions from evaporation and condensation of water and sulfuric acid to the model. They have found that the intensity of the light emission of SBSL bubble first increases as the concentration of sulfuric acid is increased (from 0%). When the concentration reached 65%, the light intensity becomes a maximum. Further increase of concentration of sulfuric acid will decrease the light intensity. This behavior has also been observed experimentally for SBSL in sulfuric acid [7]. However, when the concentration of dissolved Ar in sulfuric acid is low (e.g. 4 Torr), it was observed that

the pressure calculated at the last stage of the bubble collapse with this model is unusually high (e.g. over 100,000 atm) and no explanation for this anomaly is given in [6].

In this paper, the results of our investigation on the dependence of maximum pressure of SBSL bubbles in deferent concentrations of sulfuric acid are presented. We have employed the same model used by Moshaii group, but improved the accuracy of the calculation by using 4th order Runge-Kutta method [8] as the integrator for coupled hydro-chemical equations.

2. HYDRO-CHEMICAL MODEL

This simulation is fundamentally based on a model used by Moshaii *et al* [6]. The radial motion of the bubble is described by the well-known Rayleigh-Plesset (RP) equation. The RP equation, we used in this work is one of the most popular forms of RP equation which was derived by Keller and Miksis [9].

$$\left(1-\frac{\dot{R}}{C}\right)R\ddot{R}+\frac{3}{2}\left(1-\frac{\dot{R}}{3C}\right)\dot{R}^2=\left(1+\frac{\dot{R}}{C}\right)\frac{1}{\rho}\left(P_g-P_a-P_0\right)+\frac{R}{\rho C}\dot{P}_g-4\frac{\mu\dot{R}}{\rho R}-\frac{2\sigma}{\rho R} \quad (1)$$

here, dots denote the differentiation with respect to time, *R, C, ρ, μ,* and σ are the radius of the bubble, speed of sound, density of the liquid, viscosity of liquid and surface tension, respectively. P_g is the bubble internal pressure, P_0 is the ambient pressure, and P_a is the driving pressure which has the form

$$P_a=-p_a\sin(\omega t) \quad (2)$$

where, p_a denotes amplitude of acoustic field and ω denotes frequency. The internal pressure P_g is modeled using van der Waals equation as the equation of state.

$$P_g=\frac{N_{tot}kT_g}{V-N_{tot}B} \quad (3)$$

where N_{tot}, T_g, V and k are total number of particles inside the bubble, gas temperature of the bubble, volume of the bubble and Boltzmann constant respectively. The co-volume $B=5.1\times10^{-29}m^3$ and it is assumed to be equal for all particles.

2.1 Diffusion

The number of particles of species *i* in the bubble change with the time because of the diffusion. The diffusion rate ($\dot{N}_i^d$) of species *i* from the bubble to the surrounding liquid is calculated by the boundary layer formalism of Ref. [4].

$$\dot{N}_i^d=4\pi R^2 D\frac{n_{i,o}-n_i}{l_d} \quad (4)$$

$$D=D_0\frac{n_0}{n_{tot}} \quad (5)$$

where $n_{i,0}$ and n_i are the equilibrium and instantaneous concentration of particles of species *i* respectively. The coefficient D is the diffusion coefficient given by,

where $D_0 = 23.55 \times 10^{-6} m^2/s$ and n_{tot} is the instantaneous total number density of the bubble and $n_0$is the value of n_{tot} at the beginning of the bubble evolution. The quantity l_d is the thickness of the diffusive boundary layer which is considered as common for all species and it is given by,

$$l_d = min\left(\sqrt{\frac{RD}{|R|}}, \frac{R}{\pi}\right) \tag{6}$$

2.2 Chemical Reactions

At the beginning of the bubble evolution, the bubble's contents are argon, water vapour and sulfuric acid molecules. Due to the chemical reactions, five more chemical molecules are created inside the bubble and they are O, O_2, OH, H, and H_2. The rates of the chemical reactions are calculated by the modified Arrhenius laws [4]. Eight chemical reactions occurring inside the bubble due to the dissociation of water vapour molecules were considered in the calculation. Net chemical reaction rate of species *i* is calculated by considering forward and backward reaction rates of species *i*.

$$r_{f,j} = \left[\frac{exp[n_{tot}B/(1-n_{tot}B)]}{1-n_{tot}B}\right]^{t_j} k_{f,j} n_{tot} n_A n_B T^{c_{f,j}} \times exp\left(-\frac{E_{f,j}}{kT}\right) \tag{7}$$

$$r_{b,j} = k_{b,j} n_{tot} n_C n_D T^{c_{b,j}} exp\left(-\frac{E_{f,j}}{kT}\right) \tag{8}$$

$$r_j = r_{f,j} - r_{b,j} \tag{9}$$

here, r_j,$r_{f,j}$, and $r_{b,j}$are the net reaction rate per unit volume, forward reaction rate and backward reaction rate. $n_{A,\ldots,D}$ are the concentration of the participation species. n_{tot} is the number density of the collider M.

2.3 Condensation and Evaporation Changes

Condensation and evaporation changes of water vapour and sulfuric acid molecules at the bubble interface are calculated using following equations.

$$\dot{N}_i = 4\pi R^2 J_i \tag{10}$$

$$J_i = \frac{\alpha_M}{\sqrt{2\pi M_i k_B / N_A}} \left(\frac{P_i^{vap}}{\sqrt{T_0}} - \frac{\Gamma_i P_i}{\sqrt{T_g}}\right) \tag{11}$$

where, $\dot{N}_i$ and J_i are rate of change in vapour species *i* with molar mass M_i and rate of flow of vapour molecule at bubble wall respectively.$P_i^{vap}$is the equilibrium vapour pressure at ambient temperature T_0, P_i is the instantaneous partial pressure inside the bubble.k_B and N_A are the Boltzmann's constant and Avogadro's number respectively. Here $\alpha_M = 0.35$ is the accommodation coefficient for vapour molecules at the bubble surface. The correction factor Γ_i is given by [6].

$$\Gamma_i = \exp(-\Omega_i^2) - \Omega_i\sqrt{\pi}[1 - \mathrm{erf}(\Omega_i)] \tag{12}$$

$$\Omega_i = \frac{\dot{m}_i}{P_i}\sqrt{\frac{N_A k_B T_g}{2M_i}} \tag{13}$$

$$\dot{m}_i = \frac{J_i M_i}{N_A} \tag{14}$$

2.4 Heat Transfer Calculations

Heat transferring between bubble and its surrounding liquid is calculated by the boundary layer formalism and scheme given by Ref. [2].

$$\dot{Q} = 4\pi R^2 \kappa \frac{T_0 - T_g}{l_{th}} \tag{15}$$

$$l_{th} = min\left(\sqrt{\frac{R\chi}{|\dot{R}|}}, \frac{R}{\pi}\right) \tag{16}$$

where, $\dot{Q}$ is the rate of heat transfer at the bubble wall, T_g and T_0 are temperature of the gas and the temperature of the bubble wall respectively. κ is the thermal conductivity coefficient of the gas inside the bubble [2]. l_{th} is the thickness of thermal boundary layer and χ is the thermal diffusivity coefficient of the bubble content given by, $\chi = \kappa / c_p$, with $c_p = \frac{5}{2} n\mathrm{Ar}\,k + \frac{8}{2} n\mathrm{H_2O}\,k$ the constant pressure heat capacity per unit volume of the gas mixture at the wall.

2.5 Rate of Change of Gas Temperature ($\dot{T}_g$)

Rate of change of temperature is calculated using global energy balance of the bubble interior [2]

$$\dot{T}_g \sum_j \frac{\partial e_{th,j}}{\partial T_g} N_j = \dot{Q} - P_g \dot{V} - \sum_j e_{th,j} \dot{N}_j + \dot{E}_{chem} + \sum_j h_{w,j} \dot{N}_j^d \tag{17}$$

where, $\dot{E}_{chem} = V \sum_j r_j \Delta E_j$ is the rate of change in the chemical energy of the bubble due to eight chemical reactions. $h_{w,j} = \left(1 + \frac{f_i}{2}\right) k T_0$ is the molecular enthalpy of the particles species j at the bubble wall and f_j is the number of translational + rotational degree of freedom $e_{th,j}$ is the thermal energy of the molecule j given by,

$$e_{th,j} = \frac{f_j}{2} k T_g + \sum_l \frac{k\Theta_{j,l}}{e^{(\Theta_{j,l}/T_g)} - 1} \tag{18}$$

here $\Theta_{j,l}$ is characteristic vibrational temperature of the particles species j.

3. NUMERICAL METHODS

We have used 4th order Runge-Kutta method [8] to solve coupled differential equations given above. In the collapse region, bubble parameters change rapidly with time and use of fixed time step size may produce calculation errors. For this reason, adaptive step size control method is used to control the step size of the Runge-Kutta method. This way we make sure that the accuracy of the physical parameters are maintained at the desired level.

In order to start the calculation, initial conditions must be set. For argon, initial number of atoms is calculated by using van de Waals equation and equilibrium bubble pressure. Number of H_2O atoms and number of H_2SO_4atoms are calculated by using partial vapour pressure of H_2O and H_2SO_4 respectively. For all the other species, initial number of atoms is simply set as zero [2]. All other physical parameters used in the current simulation for various concentrations of sulfuric acid and water are taken from Ref. [2 and 6].

4. RESULTS

Based on Moshaii group's previous work [10] and according to source code given on the web, it was found that they have employed a less accurate Euler method for integrating the differential equations. Using 4^{th} order Runge-Kutta method with adaptive step size and properly handling the interdependence of coupled differential equations, we have calculated maximum temperature, minimum radius, total number of species, and the pressure at the bubble collapse very accurately. The results of simulation are given in Table1. Our results are similar to that in [6] both quantitatively and qualitatively, except for the gas pressure at bubble collapse. Experimentally observed temperature is around 25000K [11]. Therefore, the temperature at the bubble collapse determined in this simulation is closer to the available experimental values. The gas pressure at the bubble collapse calculated with our implementation produced more realistic values than in [6]. Our simulation also predicts the same dependency of intensity of the light emission on concentration of sulfuric acid as in [6].

Table I. Values of bubble characteristics at the bubble collapse.

Sulfuric acid concentration (wt %)	85	70	65	45	0(Water)
P_a (atm)	1.89	1.78	1.73	1.60	1.37
R_0 (um)	6.31	6.52	6.65	5.65	4.26
R_{min} (um)	1.13	1.17	1.19	1.02	0.83
R_{max} (um)	47.92	49.77	51.60	46.82	35.20
T_{gmax}(K)	30743	32946	34700	33453	23349
P_{gmax}(atm)	29019	31194	32840	33226	18755
n_{totmax} ($10^{27}m^3$)	6.93	6.95	6.95	7.29	5.90

Here, R_{min} and $R_{max}$are minimum and maximum radius of the bubble. T_{gmax} and P_{gmax} are maximum temperature and pressure at bubble collapse. n_{totmax} is number density at bubble collapse.

5. SUMMARY AND CONCLUDING REMARKS

In this study we have simulated the SBSL bubble collapse in sulfuric acid by solving Rayleigh-Plesset equation with diffusive changes, chemical changes and evaporation and condensation of water and sulfuric acid in the bubble. The simulation was carried out for various concentrations of sulfuric acid and determined the maximum

Temperatures and the maximum Pressures at the last stage of the bubble collapse. Special attention has been paid to the sensitivity of physical parameters of SBSL bubble to the numerical techniques used. The maximum temperature of the SBSL bubble is found to be the highest when the concentration of sulfuric acid is 65% as found experimentally and it is less sensitive to the accuracy of the numerical methods used. The maximum pressure is found to be highest when concentration is 45% and the calculated bubble pressure is very sensitive to the accuracy of the numerical methods employed. We were able to obtain reasonably low values for the bubble pressure by increasing the accuracy of the integrator and introducing variable step size for integration to maintain the accuracy throughout the calculation.

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