World Academy of Science, Engineering and Technology
International Journal of Physical and Mathematical Sciences
Vol:2, No:7, 2008

Hydrodynamic Force on Acoustically Driven Bubble in Sulfuric Acid

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Abstract**—**Using a force balanced translational-radial dynamics, phase space of the moving single bubble sonoluminescence (m-SBSL) in 85% wt sulfuric acid has been numerically calculated. This phase space is compared with that of single bubble sonoluminescence (SBSL) in pure water which has been calculated by using the mere radial dynamics. It is shown that in 85% wt sulfuric acid, in a general agreement with experiment, the bubble's positional instability threshold lays under the shape instability threshold. At the onset of spatial instability of moving sonoluminescing (SL) bubble in 85% wt sulfuric acid, temporal effects of the hydrodynamic force on the bubble translational-radial dynamics have been investigated. The appearance of non-zero history force on the moving SL bubble is because of proper condition which was produced by high viscosity of acid. Around the moving bubble collapse due to the rapid contraction of the bubble wall, the inertial based added mass force overcomes the viscous based history force and induces acceleration on the bubble translational motion.

Keywords**—**Bjerknes force, History force, Reynolds number, Sonoluminescence.

I. INTRODUCTION

SINGLE-BUBBLE SONOLUMINESCENCE (SBSL) is the light emission from a trapped violently imploding bubble, in an acoustic standing wave field in liquids [1]. Since its discovery, all the proposed physical and chemical models for SBSL were on the basis of the phenomenology observed in pure water. These models are based on the effective link between the non-linear bubble dynamics and the mechanism of light emission. Recently, a new mysterious state of SBSL from noble gas bubbles in 85 %wt sulfuric acid (from now on we will say sulfuric acid instead of 85% wt sulfuric acid) represented a challenge to the theories of sonoluminescence [2]. SL bubble in sulfuric acid generates an extremely intense light emission with extensive molecular, ionic, and atomic bands and lines and also it is not spatially stable in the standing sound field. Sonoluminescence in sulfuric acid is a typical state of SBSL which Didenko et al. named this phenomenon moving single-bubble sonoluminescence (m-SBSL) [3].

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Recently, numbers of theoretical and experimental studies have been done on the various physical conditions and chemical process occurring in the SL bubble in sulfuric acid [2], [4]-[6]. As well as the wonderful mechanism of light emission, study of m-SBSL dynamics from sulfuric acid is also interested. Experimental reports on m-SBSL from noble gas bubbles in sulfuric acid indicates that in the phase space, driving pressure amplitudes and diffusion equilibrium ambient radii extend relative to what has been observed in pure water. In sulfuric acid the experimental observations show that the SL bubble is not spatially stable and moves in a trajectory [7], [8].

In the present work, we pay attention on the dynamics of m-SBSL by considering the behavior of all forces on the SL bubble in sulfuric acid. For this purpose, Phase space of a sonoluminescing argon bubble in sulfuric acid has been numerically calculated; in the phase space, at the onset of the bubble's spatial instability, translational-radial dynamics of the moving bubble has been calculated. Comparison of the results with those of SBSL in pure water indicates that high viscosity of acid provides an appropriate condition to appearance of non-zero history force on the moving-SL bubble. Relatively high translational and radial Reynolds numbers of pure water in the most part of the acoustic cycle eliminates the history force on the SL bubble. In sulfuric acid the increasing of added mass force around the bubble collapse causes the bubble translational motion to accelerate.

II. NUMERICAL COMPUTATION

A. Computation of translational-radial dynamics

Translational-radial dynamics of the m-SBSL in sulfuric acid has been calculated by using a force balanced equation of motion which recently has been derived by Toegle et al. [9]:

$$R(t)^{3\,\cdot\cdot} = \frac{d}{dt}[(18\mu R + 3R^2\dot{R})(\quad - \quad) + 3R^{3\,\cdot} - 2R^{3}\quad] \tag{1}$$
$$- 3R^2\dot{R}^{\,\cdot} + 3\frac{\Theta_r\Theta_t}{R^2}[(6\mu R + 3R^2\dot{R})(\quad - \quad) + 3R^{3\,\cdot} - 2R^{3}\quad - R^{3\,\cdot}]$$

where, μ is the kinetic viscosity (shear viscosity / density) of liquid, and are the liquid and SL bubble velocity vectors relative to an inertial frame, respectively.

is the gravitational acceleration and $R(t)$ is the temporal radius of the SL bubble which is described by the well-known Rayleigh-Plesset equation [10].

In (1), $\Theta_r = 1/(1+(Re_r(t))/(Re_{r,Crit}(t))^4$ and $\Theta_t = 1/(1+(Re_t(t))/(Re_{t,Crit}(t))^4$ are the introduced switches by Toegel et al. [9] and turn on the history force effect on the bubble translational- radial dynamics, at least as soon as $Re_r < Re_{r,Crit}$ and $Re_t < Re_{t,Crit}$. $Re_{r,Crit} = 7$ and $Re_{t,Crit} = 0.5$, are the critical radial and the critical translational Reynolds numbers [11], respectively. Also, $Re_r = R|\dot{R}|/\mu$ is the radial Reynolds number and $Re_t = R| \quad - \quad |/\mu$ is the translational Reynolds number of the liquid. Extraction of (1) has been done on the basis of Magnaudet and Legendre's two asymptotic expressions of the affecting hydrodynamic force on the oscillating bubble [11] and its agreement with the experiment has been shown by toegle et al. [9].

Using a hydrochemical model of sonoluminescence [12]-[16] Rayleigh-Plesset equation of the bubble radial oscillation has been closed by a van der waals equation of state of the bubble interior which includes the gas and liquid vapor pressure. Due to the low vapor pressure of the components of sulfuric acid we did not consider any chemical reaction in the model but for pure water, chemical reactions of the components and mass diffusion of the products have been considered according to. [12], [13]. The physical properties of water and sulfuric acid that we used in the numerical computation have been shown in table I.

B. Computation of Sonoluminescence phase space

The phase spaces of the sonoluminescing bubble in acid and pure water have been numerically calculated from the standard theories of diffusion stability [18], shape instability [19] and positional (Bjerknes) instability [20] for an

TABLE I
THE PHYSICAL PROPERTIES OF WATER AND SULFURIC ACID AT 20^{0}C [17].

Property	Quantity	Water/sulfuric acid 85%
P_{H2O}[Pa]	water partial pressure vapor	2338 /2.45
P_{H2SO4}[Pa]	acid partial pressure vapor	0.0/1.79× 10^{-4}
ρ[kg/m^3]	density	999/1778.6
μ[m^2/s]	viscosity	1.0× 10^{-6}/14.10× 10^{-6}
σ[Kg/m^2]	surface tension	70.7× 10^{-3}/56.03× 10^{-3}
C[m/s]	Velocity of sound	1483/1552

oscillating bubble. A gas bubble with equilibrium radius R_0 will be in diffusive equilibrium with the dissolved gas in the liquid if

$$\frac{C_l}{C_{sat}} = \frac{1}{P_0}\frac{\langle P_g(t)R(t)^4\rangle}{\langle R(t)^4\rangle} \tag{2}$$

Where, C_l and C_{sat} are gas concentration in the liquid and saturated concentration of the gas in the liquid at the ambient pressure P_0. The bubble would be in diffusive stability region in the phase space, if

$$\left.\frac{\partial C_\infty}{\partial R_0}\right|_{\dot{m}} \rangle 0 \tag{3}$$

The pressure threshold of disappearance of an SL bubble can be obtained from the parametric shape instability restriction [19].

To evaluate positional instability of the bubble, we applied the approach of Akhatov et al. [20] to calculate radial primary Bjerknes force, F_{Bj}. If the time average of primary Bjerknes force becomes positive in sign then the bubble would be repelled from pressure anti-node and the bubble's positional instability would take place.

III. NUMERICAL RESULTS

In Fig. 1 to Fig. 5. we discuss on the obtained numerical results of m-SBSL dynamics and effective forces on the bubble translational motion in sulfuric acid and compare these results with those in pure water system.

In Fig. 1. Phase space of sonoluminescence has been calculated for 4 and 50 torr SL argon bubble in sulfuric acid and pure water. In the numerical computation we used a 38 kHz acoustical frequency near to the acoustical frequency that Flannigan, and Suslik [4] and Troia et al. [8] used in their experiments. Computation of the SBSL phase space of pure water has been done by using the mere radial dynamics [12]-[16] without interrupting any translational effect. For sulfuric acid we used the translational-radial dynamics for bubble oscillation above the Bj. I. Threshold. As we see in Fig. 1a., in the SL phase space of pure water, Sh. I. threshold lays under the positional instability (Bj. I.) threshold. Therefore approximately through all the permissible pressure amplitudes the stationary stable state of SBSL, would take place in pure water. But in Fig. 1. b. we see that in sulfuric acid, this is the Bj. I. threshold that lays under the Sh. I. threshold. It means that in sulfuric acid, m-SBSL with diffusive and shape stability condition would be observable [2], [4], [6], [8].

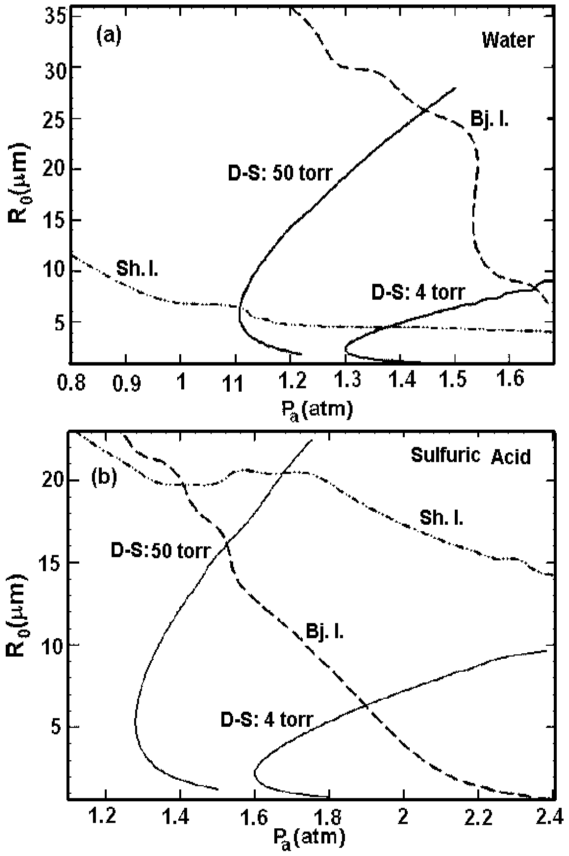

Fig. 1 calculated phase spaces for an argon SL bubble dissolved at the partial pressures of 4 and 50 torr in water and 85%wt sulfuric acid. Above (under) the Bj. I. threshold, the bubble experiences a positive outward (negative inward) Bjerknes force.

Fig. 2. compares the radial oscillation of a 4 torr SL argon bubble in sulfuric acid and pure water, during one acoustical cycle. For sulfuric acid, we made the calculation for a moving SL bubble on the onset of Bj. I. threshold in the phase space. Here the sign of the primary Bjerknes force starts to change from negative to positive. Collective effect of buoyancy, Bjerknes, added mass, drag and history force on the bubble radial dynamics is considered through the force balanced translational-radial dynamics (Eq. 1). As we see in the figure, due to the relatively high viscosity of acid, collapse sequence of the bubble moves deeper in the compression phase of the acoustical field. According to [20], this provides the condition that the time averaged Bjerknes force on the bubble become positive in sign and the SL bubble will repelled from its position in the central antinode of the sound field. In fig. 2. for typical 4 torr SL argon bubble in water, due to the negative time averaged Bjerknes force we did not consider any translational effect. In addition to the effect of the liquid viscosity on the bubble spatial instability, its effect on the translational dynamics of the SL bubble can be investigated through identification of the limits of Reynolds numbers of liquids.

In Fig. 3. and Fig. 4. we see the time variation of $Re_r(t)$ and $Re_t(t)$ for the same bubbles in Fig. 2. To calculate

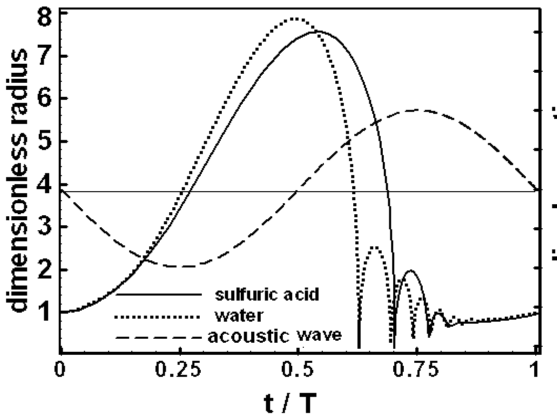

Fig. 2 calculated time-varying radius of a 4 torr argon bubble which is irradiated by a 38 kHz ultrasound in pure water and 85%wt sulfuric acid. Exerting the diffusion stability and shape instability condition, the ambient radius: $R_0 = 5.67\,\mu m$, $6.53\,\mu m$ and acoustical pressure amplitude: $P_a = 1.45$ Pa, 1.92 Pa is used in the calculation for water and 85%wt sulfuric acid, respectively. Also, with the above parameters -2.228×10^{-9} N time-averaged Bjerknes force for SL bubble in water and $+9.784 \times 10^{-9}$ N time-averaged Bjerknes force for SL bubble in 85%wt sulfuric acid has been calculated. The vertical axis of the figure is normalized to the ambient radius of the bubble and the horizontal axis is normalized to the period of sound field.

the Reynolds numbers we examined the hydrodynamic force balanced translational-radial equation (Eq. 1.) for the both SL bubbles in pure water and sulfuric acid. In Fig. 3. lower $Re_r(t)$ of acid system indicates more moderate bubble radial oscillation relative to water system. As we see, over a considerable part of the acoustical cycle, $Re_r(t)$ of the acid system is fewer than $Re_{r,crit}$ which has been introduced by Magnaudet and Legendre equal to 7 [11]. Also, in Fig. 4. for

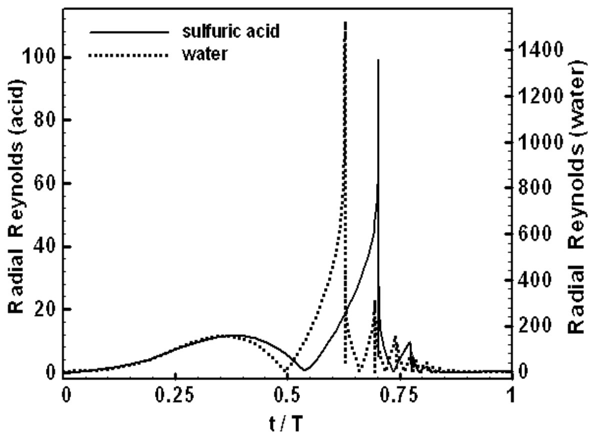

Fig. 3 temporal evolution of $Re_r(t)$ of water and 85%wt sulfuric acid, for the same bubbles in Fig. 2. According to Ref. [11], the under critical amounts of $Re_r(t)$ in some parts of the cycle, provides the necessary (but not enough condition) for appearance of the history force on the bubble.

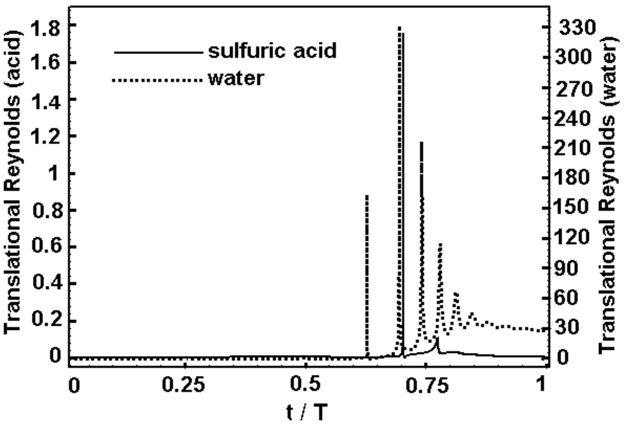

Fig. 4 temporal evolution of the $Re_t(t)$ in water and 85%wt sulfuric acid for the same bubbles in Fig. 2 . Except near the bubble collapse, the oscillating bubble in 85%wt sulfuric acid experiences the amounts of $Re_t(t)$ fewer than 0.5, which is the upscale of translational Reynolds number for appearance of a non-zero history force on the bubble [11].

the translational Reynolds number of acid we see that except at the bubble collapse, $Re_t(t)$ is fewer than its critical amount which is equal to 0.5 [11]. Therefore, in the most part of the acoustical cycle in sulfuric acid, the limits of $Re_r(t)$ and $Re_t(t)$ provides the necessary condition for appearance of a non-zero history force on the SL bubble (According to [11]). In our calculation for water system, an unexpected increase of $Re_t(t)$ after the collapse sequence and its increase at the end of the acoustical cycle (Fig. 4.), led to an intensive instability on the bubble translational dynamics. Therefore the trapped bubble in water, with negative Bjerknes force, does not experience history force in the most part of the cycle and the SL dynamics in pure water can not be described by (1). Thus, a different force balance with eliminated history force is required for investigation of bubble trapping of SBSL in pure water with a negative Bjerknes force. In our calculations, for a SL bubble in pure water, in a real sonoluminescence condition with negative Bjerknes force, we did not see a regular bubble translation after the first cycle. This identifies that a translational-radial dynamics which has been derived by the equating of all affecting forces is not valid for stationary stable SL bubble in pure water.

Fig. 5 shows the effect of the added mass force as an inertial and history force as a viscous contribution of hydrodynamic force, on the bubble translational motion during three acoustical cycles in sulfuric acid. At the bubble collapse added mass force increases due to the violent increase in bubble radial velocity; In this occasion, history force is omitted. Continuous radial oscillation and region of influence of the history force is shown in Fig. 5a and 5b, respectively. As shown in Fig. 5c. the added momentum of the surrounding fluid increases considerably and as we see in fig. 6d. makes a shift in the bubble translation at the collapse sequence. Because, at this time Bjerknes force on the bubble decreases

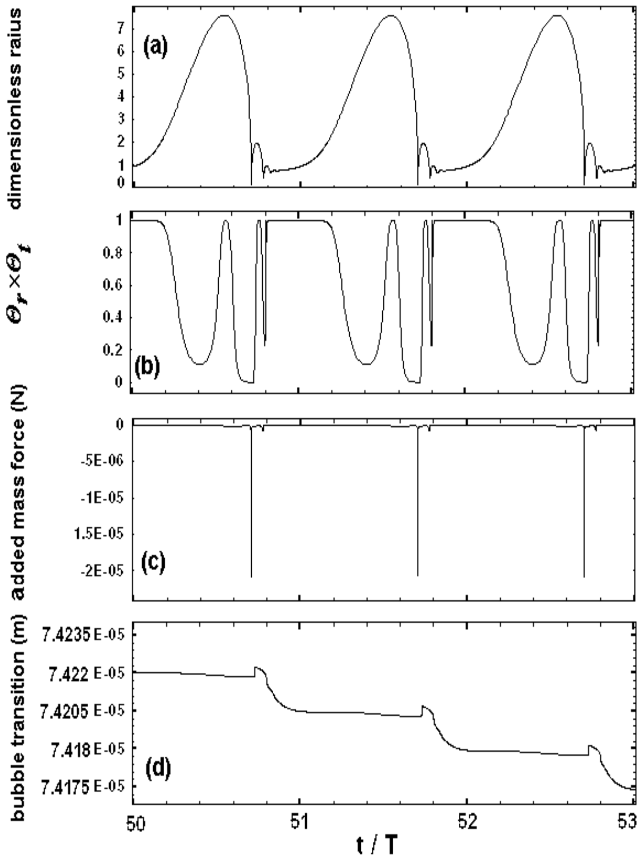

Fig. 5 calculated properties of the moving SL bubble in 85%wt sulfuric acid with the same numerical parameters in Fig. 2. (a) radius of the non trapped bubble with positive Bjerknes force, (b) the region of appearance of history force; Unless at the collapse, the bubble experiences non-zero history force, (c) at the collapse due to the rapid contraction of the bubble, added mass force dominates the viscous contributions of the hydrodynamic force, (d) SL bubble displacement in sulfuric acid.

due to the small surface area upon which the pressure gradient acts, the increased momentum of the surrounding acid, is the main reason of shift in the bubble motion near the collapse sequence.

IV. Conclusion

In conclusion, using a complete force balanced translational-radial dynamics we showed that due to the high viscosity of sulfuric acid, the positional instability threshold lays under the shape instability threshold. This causes that a SL bubble pull out the antinode of the sound wave with a radial-translational dynamics which is influenced from the inertial and viscous contributions of hydrodynamic force all over the oscillation cycle. In pure water, due to the low liquid viscosity, the effect of history force on a trapped SL bubble should be eliminated. Because of the different behavior of the hydrodynamic force, different bubble dynamics is required for m-SBSL in sulfuric acid and stationary stable SBSL in pure water.

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