



## The effect of bulk viscosity on single bubble dynamics and sonoluminescence

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### ARTICLE INFO

#### Keywords:

Bubble dynamics

Bulk viscosity

Single bubble sonoluminescence

### ABSTRACT

In our previous paper, we derived a new single bubble model including the effect of bulk viscosity. To confront it to experiments, single bubble dynamics was measured here in 30% (v/v) glycerol-water mixture under different acoustic amplitudes and compared to models including or not the effect of bulk viscosity. The results showed that calculated bubble dynamics were not significantly affected by the bulk viscosity within the experimental conditions used in this study. However, there was a noticeable delay for the first rebound when bulk viscosity was considered. The corresponding sonoluminescence intensities were collected and compared with theoretical predictions. The results did not allow to discriminate between the two models (one includes the effect of bulk viscosity, the other does not), confirming the negligible effect of bulk viscosity in this condition (30% (v/v) glycerol-water mixture). Due to the instability of a single bubble in higher viscosity solutions, we could not implement experiments that can discriminate between the two models.

### 1. Introduction

Single bubble sonoluminescence (SBSL), the light emission from a single cavitation bubble, has been studied for decades. A SBSL bubble repeats expansion, compression, collapse and rebounds in response to the pressure oscillation of an ultrasonic wave and emits light within every acoustic cycle [1]. In 1962, Yosioka and Omura reported the light emission from a single bubble driven by ultrasound [2]. In 1964, Wang reported results of experimental investigation of electromagnetic and optical radiations produced by single bubble cavitation [3]. But it was only in the 1990s, after Gaitan's reports on SBSL [4,5] that the phenomenon really started to interest a larger scientific community. In 1997, argon rectification theory proposed by Lohse et al. [6] implied that a single air bubble eventually turned into a pure argon bubble due to chemical reactions that occur within the cavitation bubble. This enrichment in Ar has been proven by several groups [7,8]. Koda [9] measured SL intensity and the formation of nitrites and nitrates during a single bubble experiment. The SL intensity remained constant and the concentrations of nitrite and nitrate ions linearly increased with time over 30 h, which meant that nitrogen and oxygen in the bubble reacted

to form soluble species. According to these experimental results presented in this work, we may assume that most of the gas content inside the bubble is argon with only a small amount of air.

Toegel et al. [10] developed a force balance model for the bubble's translational motion with effective forces, and compared with the results of experimental data. They found that in viscous liquids such as degassed glycol ( $\mu = 20 \times 10^{-3}$  Pa·s), it is not possible to trap the bubble in a stable position and identified the history force (resulting from the unsteady volume change from the bubble oscillation) as the reason of the destabilization of the single bubble in viscous liquids. Liu et al. [11] conducted comprehensive numerical and experimental analyses of the effect of viscosity on cavitation oscillations. The results showed that the increasing of viscosity decreased the maximum bubble radius but increased the minimum bubble radii and the first oscillation time (from  $t = 0$  to the moment  $R = R_{\min}$ ). Englert et al. [12] experimentally studied the luminescence pulses emitted from collapsing laser-induced bubbles in water-glycerol mixtures. They found that the pulse duration increased with the increase of the concentration of glycerol. A likely reason given by the authors was that the speed of the bubble wall in the very last stage of the initial collapse became slower when the viscosity

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<https://doi.org/10.1016/j.ultsonch.2023.106307>

Received 13 December 2022; Received in revised form 18 January 2023; Accepted 18 January 2023

Available online 21 January 2023

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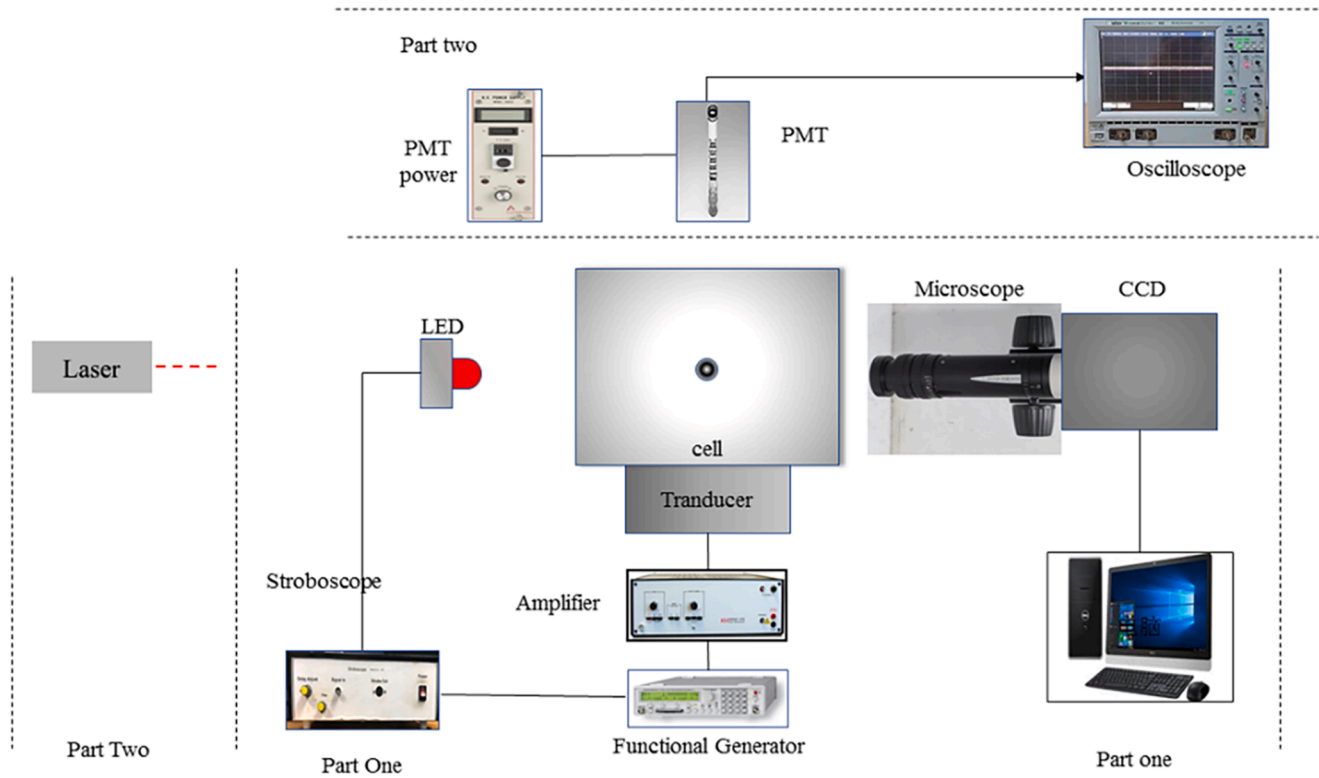


Fig. 1. The schematic diagram of single bubble experiment.

increased, which lead to the plasma remained intact and hot for a longer period of time.

In a previous paper [13], a model of cavitation bubble dynamics had been derived to theoretically study the effect of bulk viscosities on the temperature and pressure inside a collapsing bubble. The results showed that the relative high bulk viscosity had a significant effect on maximum temperature and pressure inside the bubble when the bubble collapsed and the pressure was more sensitive than temperature, and that bulk viscosity of liquid has a nonnegligible effect on single bubble dynamics when it is higher than 6 times viscosity of water (6 mPa·s). Later, Nazari-Mahroo et al. [14] found similar results using Gilmore model with a new modified boundary condition at bubble interface. In the present paper, single bubble experiments were conducted in 30% glycerol-water (v/v) solutions with the viscosity of 3 mPa·s to obtain sonoluminescence intensities. Theoretical calculations of sonoluminescence intensities with or without considering the bulk viscosity were then compared with experimentally observed SL intensities.

## 2. Model

The model for single bubble dynamics used in this study was fully described in Ref. [13] except for three improvements, described here. The first one is to take 89 chemical reactions inside the single bubble into consideration; the second one is to consider the temperature-dependent parameters (viscosity, surface tension and vapour pressure) of the glycerol-water solution [15–21]; the last one is the radical dissolution into the surrounding liquid [1]. In the present study, the uptake coefficient ( $\Theta$ ), the rate of dissolution of chemical products into the surrounding liquid from the interior of the bubble) is assumed as 0.0007 rather than 0.001 (as suggested in Ref. [1,22]). The reason is that the high viscosity may hinder the gases and radicals desorbing from the liquid phase into the bubble or dissolving into the liquid from the bubble. It has been reported that the diffusion coefficients of gases decrease significantly with the increase of glycerol concentration [23,24], which will affect the uptake coefficient. The following is a brief

description of the model.

As an equation of the bubble radius ( $R$ ), eq.(1), is employed, in which compressibility of a liquid, the effect of evaporation and condensation of water vapour at the bubble wall and the effect of the liquid viscosity on the momentum equation are considered [13].

$$\begin{aligned} & \left(1 - \frac{\dot{R}}{c_\infty} + \frac{\dot{m}}{c_\infty \rho_l}\right) R \left(\ddot{R} - \frac{\dot{m}}{\rho_l}\right) + \frac{3}{2} \dot{R}^2 \left(1 - \frac{\dot{R}}{3c_\infty} + \frac{\dot{m}}{3c_\infty \rho_l}\right) \\ &= \frac{1}{\rho_l} \left(1 + \frac{\dot{R}}{c_\infty}\right) [p_B - p_s(t + \frac{R}{c_\infty}) - p_0] + \frac{\dot{m}}{\rho_l} \left(\dot{R} + \frac{\dot{m}}{2\rho_l} + \frac{\dot{m}\dot{R}}{2c_\infty \rho_l}\right) \\ &+ \left[\frac{R}{c_\infty \rho_l} + \frac{4\mu}{3c_\infty^2 \rho_l^2} \left(1 + \frac{\dot{R}}{c_\infty}\right)\right] \frac{dp_B}{dt} + \frac{4\mu R}{3c_\infty^3 \rho_l^2} \frac{d^2 p_B}{dt^2} \end{aligned} \quad (1)$$

where  $\dot{\cdot}$  denotes the time derivative,  $t$  is the time,  $\rho_l$  is the liquid density,  $p_l$  is the pressure in the liquid,  $\mu$  is the viscosity of the liquid from the bulk of liquid,  $\dot{m}$  is the net rate of evaporation (condensation) per unit and unit time,  $c_\infty$  is the sound speed in the ambient liquid,  $p_0$  is the pressure in the ambient liquid,  $p_B$  is the pressure of the bubble wall.

The liquid pressure on the external side of the bubble wall  $p_B$  is related to the pressure inside the bubble ( $p_g(t)$ ):

$$P_B = P_g(t) - \frac{2\sigma}{R} - \frac{4\mu'}{R} \left(\dot{R} - \frac{\dot{m}}{\rho}\right) - \dot{m}^2 \left(\frac{1}{\rho} - \frac{1}{\rho_g}\right) - \frac{4\mu}{3c^2 \rho_l} \frac{dP_B}{dt} \quad (2)$$

where  $\sigma$  is the surface tension,  $\mu'$  is the liquid viscosity at the bubble interface, and  $\rho_g$  is the density inside the bubble. Note that the third term of eq.(2) includes the liquid viscosity at the bubble interface, the fifth term of eq.(2) includes bulk liquid viscosity.

The gas temperature inside the bubble ( $T$ ) can be expressed as follows:

$$T = \frac{N_A^2 EV + (n_{H_2O} + \sum_i n_i)^2 a}{(n_{H_2O} C_{v,H_2O} + \sum_i n_i C_{v,i})} \quad (3)$$

where  $N_A$  is the Avogadro number,  $V$  is the bubble volume,  $E$  is the in-

ternal energy of the bubble,  $n$  is the amount of molecules,  $a$  is the van der Waals constants,  $C_v$  is the heat capacity at a constant volume, the suffix H<sub>2</sub>O represents water vapour, and the suffix  $i$  represents the gases inside the bubble.

It has been theoretically proven that SBSL is a thermal radiative process rather than blackbody radiation [25,26]. Electron-ion bremsstrahlung, electron-atom bremsstrahlung, radiative recombination of electrons and ions, and radiative attachment of electrons to neutral atoms are considered as the thermal radiative processes in this paper.

Assuming in a first approximation thermal equilibrium and unicity of temperature, the intensity of electron-ion bremsstrahlung is calculated by Eq.(4) [25]

$$I_{e-ion} = \frac{4}{3} \pi R^3 \times 1.57 \times 10^{-40} q^2 N^2 T^{1/2} \quad (4)$$

where  $q$  is the degree of ionization calculated by Saha equation,  $R$  is the bubble radius,  $N$  is the number density of atoms,  $T$  is the temperature inside the bubble.

The intensity of electron-atom bremsstrahlung is calculated by Eq.(5) [25]

$$I_{e-atom} = \frac{4}{3} \pi R^3 \times 4.6 \times 10^{-44} q N^2 T \quad (5)$$

The intensity of radiative recombination of electrons and ions is calculated by Eq.(6) [25]

$$I_{rr} = \frac{4}{3} \pi R^3 q^2 N^2 \sigma_{fb} \bar{v} h \bar{\nu} \quad (6)$$

where  $\sigma_{fb}$  is the cross section of radiative recombination,  $\bar{v}$  is the mean velocity of a free electron, and  $\bar{\nu}$  is the mean frequency of the emitted photon.

The intensity of radiative attachment of electrons to the neutral oxygen atoms is calculated by Eq.(7) [27]

$$I_{raO} = \frac{4}{3} \pi R^3 \times 10^{-26} \times n_e n_o \bar{v}_e h \bar{\nu} \quad (7)$$

where  $n_e$  and  $n_o$  are the number densities of electrons and oxygen atoms inside the bubble,  $\bar{v}_e$  is the mean velocity of electrons. The intensity of radiative attachment of electrons to the hydrogen atoms, OH radicals and oxygen molecules are calculated too.

### 3. Experimental details

60 mL glycerol were added to 140 mL Milli-Q water to prepare 30% (v/v) glycerol-water mixture, which was the highest concentration solution of the mixture where we were able to stabilise a single bubble in. Once fully mixed, the solution was degassed for at least 3 h using a vacuum pump since partial degassing was a key requirement for 'trapping' a single stable SL bubble [4–7,28]. An oxygen meter (HANNA HI 98193) was used to measure gas concentration in the solution. The dissolved oxygen concentration of the degassed solution was about 2 mg/L.

The setup employed for the imaging and SL measurement (Fig. 1) was adapted from previous studies [29,30]. In part one, an acoustic standing wave was generated in a cylindrical cell of dimensions 62 mm diameter and 113 mm height (water level filled up to about 65 mm) connected to a cylindrical Lead Zirconate Titanate (PZT) piezo ceramic transducer at the bottom. The signal generated with a Hameg function generator (HM8131-2) was sent to the transducer through a power amplifier. A simultaneous signal was delivered to a stroboscope (Model 610/1581) connected to a LED such that the LED could flash at the driving frequency. A CCD camera connected to a microscope was used to record the image which was displayed in a computer monitor. Sets of images were taken to calculate the bubble radius [30]. Three images of a 0.82 mm diameter needle were taken under the same focal length to

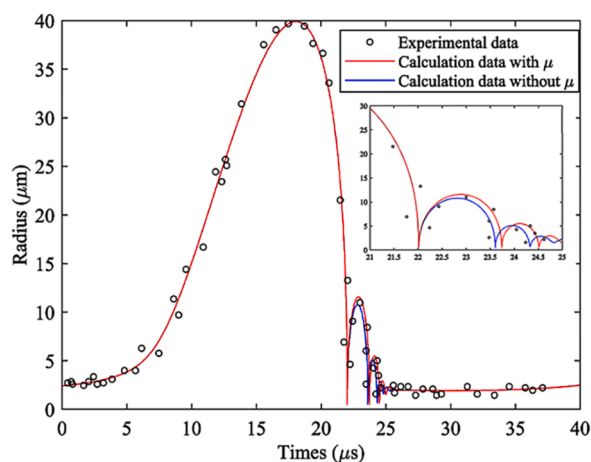


Fig. 2. The bubble radius as a function of time within one acoustic cycle in 30% v/v glycerol at 1.4 bar.

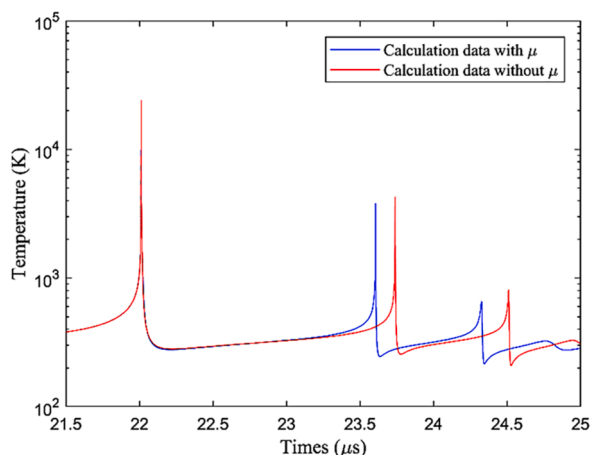


Fig. 3. The temperature inside the bubble at collapsing and rebound phases in 30% v/v glycerol at 1.4 bar.

provide size calibration.

In part two, a photomultiplier tube (PMT) (Hamamatsu R1463 in E849-35 socket powered by Canberra H.V. Supply Model 3002) was used to measure the sonoluminescence intensity. The PMT signal was displayed on an oscilloscope (LeCroy Wavesurfer 452) [30]. A needle hydrophone was used to measure voltage at the bubble position that could then be used to calculate the acoustic pressure amplitude acting on the single bubble (not shown in the figure). A laser beam was used to help find the single bubble in the cell. After finding the SL bubble, it was turned off before measuring SL.

## 4. Results and discussion

### 4.1. Bubble dynamics

The frequency used in experiment was 25.34 kHz, and the acoustic amplitude acting on the single bubble was 1.4 bar as measured by a hydrophone. The initial bubble radius used in theoretical calculation was selected to match the calculated maximum radius with the experimental maximum radius. As mentioned earlier, the temperature-dependent parameters of 30% glycerol-water solution were also considered in the theoretical calculation. The temperature of solution was 19 °C.

A comparison between numerical calculation results of the bubble radius (line) and experimental data (open circles) is shown in Fig. 2.

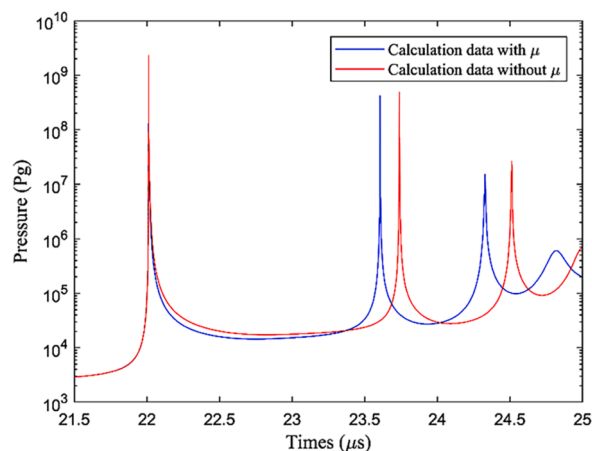


Fig. 4. The pressure inside the bubble at collapsing and rebound phases in 30% v/v glycerol at 1.4 bar.

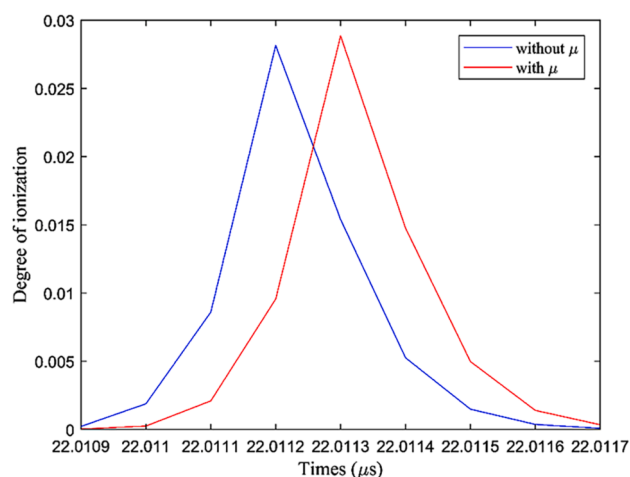


Fig. 5. The degree of ionization at around the minimum bubble radius in 30% v/v glycerol at 1.4 bar.

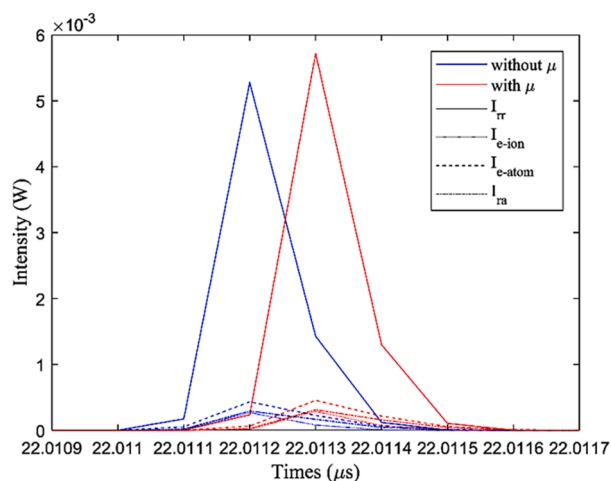


Fig. 6. The SL intensities at around the minimum bubble radius in 30% v/v glycerol at 1.4 bar.

Partial highlighted view at bubble collapse and rebound phases is also nested in this figure. It is clearly seen that the numerical calculation results with and without considering the bulk viscosity ( $\mu = 0$  in Eq. (1)

and (2)) (hereinafter case 1 and case 2) both fit well with the experimental data. The difference between both theoretical curves is not big enough to show theoretical curve fits better the experimental one.

#### 4.2. Effect of bulk viscosity on the calculated $T$ , $p$ and ionisation degree

The theoretical temperatures inside the bubble at collapse and rebound phases are shown in Fig. 3. It can be seen that the temperatures inside the bubble increase up to 24,096 K (case 1) and 23,928 K (case 2), so up to very close values.

The theoretical pressures inside the bubble at collapse and rebound phases are shown in Fig. 4. It can be seen that the pressures decrease down to 2,000 Pa for both cases and increase up to 2.43 GPa (case 1) and 2.40 GPa (case 2), confirming that the pressure is more sensitive than the temperature to taking bulk viscosity into account in the model.

In Fig. 5, the degree of ionization ( $q$ ) at around the minimum bubble radius is shown. The degrees of ionization increase up to 0.0289 (case 1) and 0.0281 (case 2) when the temperature inside the bubble reaches its maximum.

In Fig. 6, the intensities of the light emitted by electron-ion bremsstrahlung (dotted line), electron-atom bremsstrahlung (dashed line), radiative recombination (solid line) and radiative attachment (dash-dot line) are shown. From this figure, it can be seen that the main contribution according to the present model of single bubble sonoluminescence is radiative recombination which is much larger than the other three radiative processes, in agreement with previously reported data on water [25]. The duration of SL flash is about 300 ps in both cases, which is consistent with the experimentally measured results in Reference [31].

It should be noted that there is a delay of about 100 ps between two cases at the very last stage of strongest collapse as shown in Figs. 5 and 6. The reason of the exist of a delay is that when considering the bulk viscosity, the pressure inside the bubble is higher, slowing the speed of the bubble collapse. A similar delay is observed after each rebound, as seen in Figs. 2-4. It is the most pregnant difference introduced by taking the viscosity into account in the model.

Theoretical calculations show no significant changes to the bubble dynamics in 30% glycerol between the results with or without considering the bulk viscosity – both fairly reproduce experimental data. The key observation is a relatively longer induction time is required for the collapsing phase in viscous solutions when the viscosity is considered in the model. This is because with the increase of the viscosity of liquid, the pressure inside the bubble increases, decreasing the speed of the bubble wall. Fig. 2-Fig. 6 show that there are delays in reaching the minimum radii in the rebound phases when bulk viscosity is included.

#### 4.3. Comparison of experimental SBSL intensities with calculated ones

As mentioned earlier, the bubble radius is not a sensitive parameter to observe the effect of bulk viscosity in the present analysis. In order to see if SBSL intensities could be used as a parameter to observe the effect of bulk viscosity in the model, experiments were conducted to measure the intensities of SBSL under different acoustic amplitudes. In the calculation, because the bubbles at higher amplitudes were not as stable as the bubble when the amplitude was 1.4 bar, both the amplitudes and initial radius at higher amplitudes were calibrated by the Eller-Flynn formula [26]:  $\int_0^{T_b} R^4 (p_g - p_v - p_0 c_i / c_0) dt = 0$ , where  $T_b$  is the period of ultrasound,  $R$  is the bubble radius,  $p_g$  is the total pressure inside

Table 1

The amplitudes and initial radius used in theoretical calculations of SBSL intensities.

Pressure (bar)	1.416	1.459	1.476	1.512	1.565
$R_0$ ( $\mu\text{m}$ )	2.43	2.82	3.03	3.60	4.20



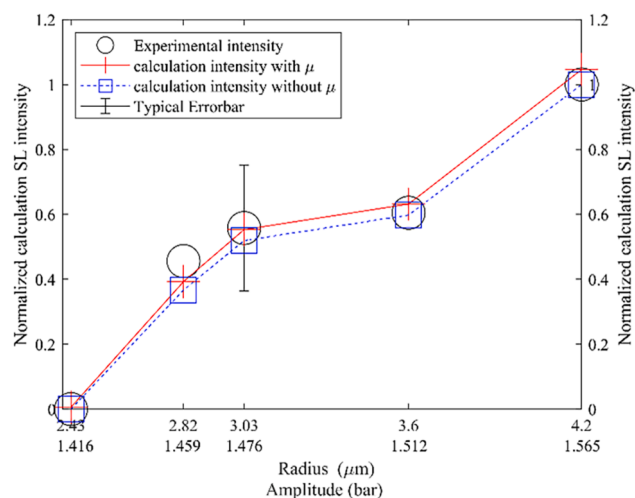


Fig. 7. Comparison of normalized experimental and calculated SBSL intensities.

the bubble,  $p_v$  is the vapor pressure inside the bubble,  $p_0$  is the ambient pressure (1 atm),  $c_i$  is the actual gas concentration in water apart from the bubble,  $c_0$  is the solubility of gas in water. The amplitudes and initial radii used in theoretical calculation are shown in Table 1.

A comparison of experimentally measured and calculated normalized SBSL intensities is shown in Fig. 7. The open circles are normalized averaged intensities over 200 cycles; red plus and blue squares are calculated normalized SBSL intensity with and without considering the bulk viscosity, respectively. Correlation coefficients of the experimental intensities and the calculated ones with and without considering bulk viscosity are 0.9901 and 0.9879, respectively.

A comparison between experimental SL intensities and calculated ones under different amplitudes shows that the model with considering bulk viscosity has no obvious advantage over the one without considering bulk viscosity because the difference between correlation coefficients is really small, which means that SBSL intensity may not be a good parameter due to the very small difference between the calculated value in the absence and presence of viscosity term.

The experimental SBSL intensities do not allow to discriminate between the two models, confirming that the negligible effect of bulk viscosity in low viscous solutions [13,14]. Failure to stabilise a single bubble under relatively higher viscous conditions (about 60% glycerol solutions) lead to the impossible of discriminating between the two models in higher viscous. Such experiments are required in future.

## 5. Conclusion

In this study, single bubble experiments were conducted in 30% (v/v) glycerol-water mixture solutions under different acoustic amplitudes. The corresponding sonoluminescence intensities were collected to compare with theoretical SL values calculated under the same conditions. The results show that taking the bulk viscosity into account in the model does not affect the bubble dynamics in expansion and compression phases but show minor changes in the collapse and rebound phases that are smaller than the experimental scatter. In the previous work, we theoretically confirmed that the bulk viscosity plays a nonnegligible role of the bubble dynamics when it is high enough. In this work, however, the comparison of SBSL between experimental results and theoretical ones did not allow to discriminate the previous theory. The main reason is that the viscosities used in this work were not high enough, and unfortunately we were unable to stabilise a single bubble under a more high viscous conditions. In order to obtain more clear effect of bulk viscosity, single bubble experiments in higher concentration glycerol solutions or at higher amplitude or other solutions (like  $H_2SO_4$  aqueous

solution) should be conducted in the future.

## CRediT authorship contribution statement

**Yang Shen:** Conceptualization, Methodology, Software, Data curation, Writing – original draft. **Rachel Pflieger:** Supervision, Formal analysis, Writing – review & editing. **Weizhong Chen:** Funding acquisition, Writing – review & editing. **Muthupandian Ashokkumar:** Methodology, Supervision, Formal analysis, Writing – review & editing.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgement

This work was supported by National Natural Science Foundation of China (No. 12074185) and the authors want to thanks Sergey Nikitenko for his useful discussion.

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