Studies on threshold pressures of sonoluminescence for bubbles with different noble gases

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Abstract The noble gases inside bubbles may have a profound effect on the threshold pressure of sonoluminescence (SL). In this work, the SL threshold pressures have been measured experimentally for bubbles with different noble gases. Results show that the threshold pressure increases with the decrease of molecular mass for gases inside the bubbles. The simulating temperature values at the collapse are almost equal to each other for different gas bubbles at the threshold pressures. However, when the pressure is above the threshold one, the SL mechanism satisfies the bremsstrahlung. On the basis of the experiments and simulations, we found that firstly water molecules dissociate in the process of cavitation and light emission follows; then, the noble gases ionize with the increase of temperature and the bremsstrahlung occurs. SL is a process from molecular emission to bremsstrahlung.

Keywords: noble gases, threshold, molecular emission, bremsstrahlung.

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Large oscillations of gas and vapor filled bubbles in liquid during acoustic cavitation. This highly nonlinear bubble motion is accompanied by the emission of light-sonoluminescence (SL)[1, 2]. The noble gases inside the bubble can influence the SL[3−5]. At an acoustic pressure, the intensity of SL increases with the molecular mass of noble gas inside the bubbles[6]. There are several kinds of theories about SL mechanism. At present, the bremsstrahlung mechanism is widely admitted. The bremsstrahlung depends on the degree of ionization of the noble gas. The larger the degree of the ionization, the stronger the SL[7−9].

In the process of cavitation and SL, water vapor inside the bubble also influences the SL strongly. For example, based on bremsstrahlung mechanism and at the same acoustic pressure, one knows that the intensity of light emission from Xe will be 10000 times the one from He, but it was reported that the difference between them is only 10 times[6]. The reason is that water vapor inside the bubble participates in SL process. Here the water vapor will be dissociated at the collapse of bubbles and consume much energy, counteracting the interior temperature increase. On the other hand, the radicals of H and OH generated from dissociated water can emit photons, and the higher the temperature, the more the photons. These photons make the discrepancy of SL intensity from different noble gases faint[10]. Therefore, both the noble gas and the water vapor inside the bubbles take part in the process of SL, and the roles they play are the main reason of the SL. In this paper, we measure the acoustic pressures (threshold pressures) for different noble gases, respectively. The acoustic pressures can just make the bubbles radiate. On the other hand we also simulate the threshold temperatures for the different noble gas bubbles to decide which radiates first, the noble gas or the water vapor and to explore the mechanism of SL.

1 Theoretical analysis

Under an ultrasound pressure a bubble oscillates periodically and emits photons and its motion can be described as the following Rayleigh-Plesset equation[10−12]:

$$r_l\left(R\ddot{R}+\frac{3}{2}\dot{R}^2\right)=p_{\text{gas}}[R(t)]+p_{\text{vap}}-p(t)-p_0+\frac{R}{c_l}\frac{d}{dt}p_{\text{gas}}[R(T)]-4h_l\frac{\dot{R}}{R}-\frac{2s}{R}, \quad (1)$$

where R, $\dot{R}$, and $\ddot{R}$ represent the length, velocity and acceleration of the bubble's radius, respectively, r_l is the density of the liquid, c_l is the velocity of ultrasound in liquid, σ and h_l are the surface tension and viscosity of the liquid, respectively, and p_{gap} and p_{vap} represent the gas pressure and water vapor in the bubble, respectively[10].

The value of interior temperature changes with the bubble's radius. Considering the influence of heat diffusion, one can express the temperature as follows[10]:

$$\dot{T}=-[g\,(R,\dot{R},T)-1]\frac{3R^2\dot{R}}{R^3-h^3}T-c\,\frac{T-T_\infty}{R^2}, \quad (2)$$

where g represents a mutual influence index between the bubble's dynamic change (expand or compress) and the temperature, and can be expressed as follows [13]:

$$g\,(P_e)=1+(\Gamma-1)\exp\left(-\frac{A}{(P_e)^B}\right), \quad (3)$$

where $\Gamma = C_p/C_v$ is the ratio of the specific heats (the Γ for Ar is 5/3), P_e is the Peclet number, A 5.8, B 0.6, and c represents the thermal diffusivity of the interior gas and fits the following formula [14]:

$$c\,(R,T)=\frac{25}{48}\Gamma^{-1}\left(\frac{a^2p\ \Re T}{m}\right)^{1/2}G(x), \quad (4)$$

where m is the molecular weight of noble gas, and a is the effective atomic diameter.

The system of eqs. (1)—(4) indicates that the interior temperature of the bubbles is dependent on not only the ultrasound pressure and frequency but also the gas component of the bubbles, for Γ and c are related with gases. Putting the measured threshold pressures of Kr and He, p_{Kr} and p_{He}, and other related parameters into eqs. (1)—(4), one can obtain the values of the bubbles' temperature under this condition. If the values are equivalent, it means that the SL comes from the water vapor. If they are different, the SL comes from the noble gases, because different temperatures are needed to make the noble gases ionized.

2 Experiment

The SL of single noble gas bubbles at different pressures was measured by experiment and the threshold pressures for different noble gas (Kr, Ar and He) bubbles were compared. According to these data the mechanism of SL was analyzed. The experiment setup is shown in fig. 1. It consists of three major components: sample tank, ultrasound generator and ICCD imaging system. The liquid used in the experiment is de-ionized water with saturated single noble gas. In order to keep the content and component of the gas in the bubbles consistent, the water is sealed inside an airtight container seated within a circulating water-jacket with regulated temperature (about 298 K); the ultrasound generation system includes a signal (AFG320, SONY, Japan) power amplifier (ENI CO. Ltd, 2100L, 50-dB) and piezoelectric transducer (5 cm in diameter, Meza Guangzhou, China, resonant frequency: 100 kHz, maximum input power: 70 W); the center of the ICCD imaging system is an ICCD module (ICCD-576-s/1, Princeton Instruments, USA) and its controller (ST-130, Princeton Instruments, USA) is fitted with a photographic lens (Nikon, 50 mm, f/1.8, Nikon, Japan), a micro channel plate (MCP, Princeton Instruments, USA), and a relay lens. In addition, the ambient temperature and sound pressure near the SL bubbles are monitored with a thermometer and a hydrophone (HPM1/1, Precision acoustics, 950 nv/Pa).

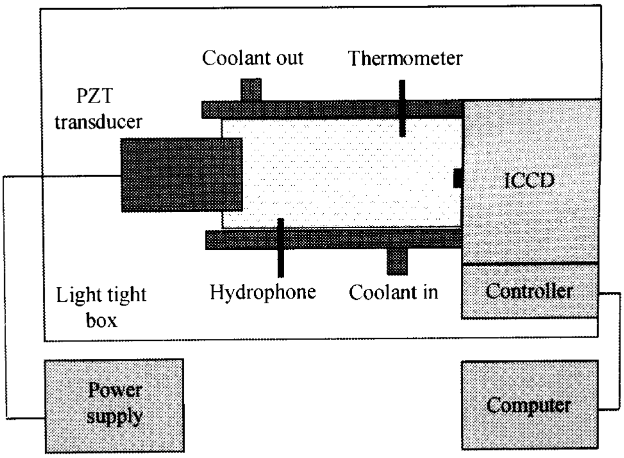

Fig. 1. Experiment setup.

The threshold pressures are determined by measuring the intensity of SL at a series of ultrasound pressures step by step. The ultrasound pressures are adjusted by changing the signal generator's input. To acquire the single noble bubbles, He, Ar or Kr is used to degas the air dissolved in water respectively, the noble gas is continuously puffed into the water for 15 min at 0.2×10^5 Pa and 298 K, then there are only single noble gas and water vapor in the bubbles. The bubbles begin to oscillate periodically under the ultrasound pressure and emit photons, and one can screen the image of SL (at lower ultrasound pressure, there may be no SL, and the ICCD only detects a background) by using the ICCD imaging system, where the ICCD detects for 5 seconds continuously each time. At the same time, the ultrasound pressure near the bubble is detected by means of hydrophone. The input signals begin with 0V and increase each step by 5 mV. The piezoelectric transducer works 5 s continuously during each interval, till the bubble emits intense light. Then one can obtain the relationship of SL intensity versus ultrasound pressure.

3 Results and discussion

The curves of SL intensity versus ultrasound pressure for bubbles with different noble gases are shown in fig. 2. There exists a threshold pressure for each kind of noble gas, and the SL intensity increases with the ultrasound pressure quasi-linearly above the threshold one. The threshold pressures of He, Ar and Kr bubbles we got are 1.660, 1.253, and 1.117×10^5 Pa, respectively. After the bubbles begin radiating, the SL intensity of Kr bubbles will be much higher than that of Ar or He bubbles at the same pressure (at 1.70×10^5 Pa, the SL intensity of Kr, Ar and He bubbles are about 64500, 35000, and 16000 counts, respectively). Consequentially, at the same SL intensity, Kr bubbles need lower ultrasound pressure than Ar or He bubbles.

Taking the threshold pressures and the physical parameters of the noble gases into the system eqs. (1)—(4), one can simulate the threshold temperature of the bubbles. In

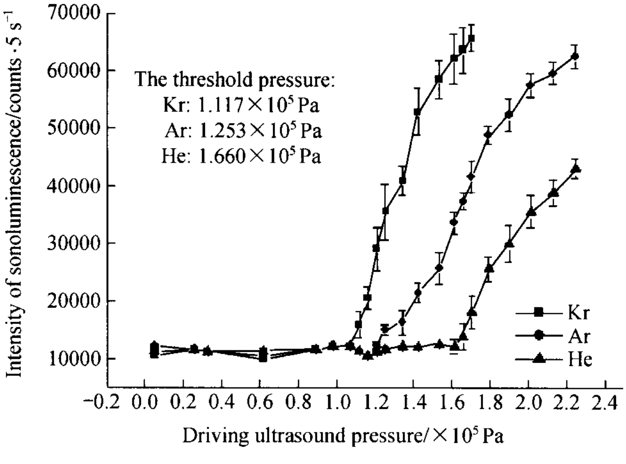

Fig. 2. The curves of SL intensity-ultrasound pressures for Kr, Ar and He bubbles.

the process of numerical simulation, the parameters are f=100 kHz, ambient pressure p_0 = 1.0×10^5 Pa, water density ρ_l = 1000 kg m^{-3}, the velocity of ultrasound in water c_l = 1500 ms^{-1}, σ =0.07 Nm^{-1} and η_l = 0.006 Nsm^{-2}, respectively.

Fig. 3 indicates the radius and temperature changes of Ar bubbles in an oscillation period at its threshold pressure. It can be seen that, at the threshold pressure, the interior temperature of Ar bubbles reaches about 5700 K at the collapse, and the ratio of the maximum radium and minimum radius $R_{\max}/R_{\min} \approx 10$. Using the same method, one can simulate the threshold temperatures of Kr and He bubbles, which are shown in table 1.

Although these threshold pressures are different, the threshold temperatures of the different noble bubbles are almost equivalent (about 6000 K). It indicates that these different noble bubbles begin radiating at the same temperature. Besides, bremsstrahlung hardly takes place at such a low temperature[15]. Therefore, it can be judged that the light emission mostly comes from the water vapor inside the bubble near the threshold pressure. A possible SL process is: adiabatic collapse of a cavitation bubble is accompanied by an increase in the energy of individual species inside the bubble. Water molecules exited into different rotational, vibrational, and electronic states via these inelastic collisions can dissociate into hydrogen atoms and hydroxyl radicals in different ground or excited states. The hydrogen atoms and hydroxyl radicals can emit photons. The hydroxyl radicals release photons and form the 310 nm peak value in the SL spectrum[16].

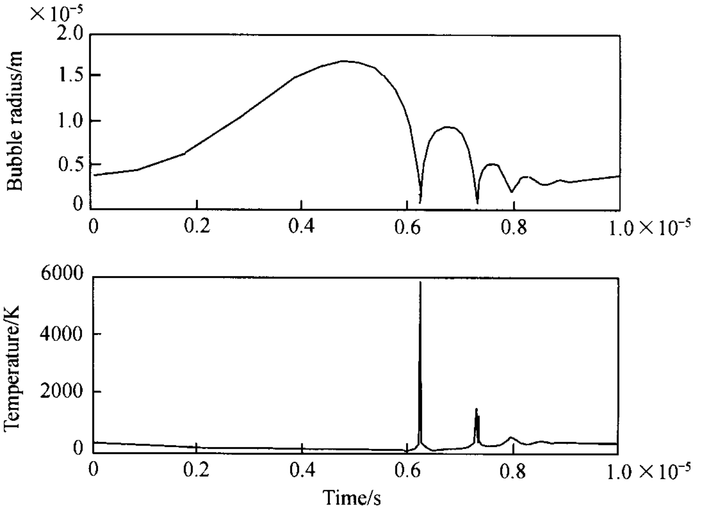

Fig. 3. The *R*-*t* and *T*-*t* curves for a bubble at the threshold pressure at 100 kHz.

Table 1 The threshold temperatures for Kr, Ar and He bubbles

Gas	u/kg • mol^{-1}	a /μm	P/×10^{5}Pa	T/K
Kr	0.058	0.50	1.117	5800
Ar	0.040	0.34	1.253	5700
He	0.004	0.034	1.660	6000

The hydrogen atoms and hydroxyl radicals at different excited states recombine and form the water molecules in different rotational, vibrational, and electronic states, releasing photons and forming the continuum spectrum of the SL. This kind of SL mechanism was considered as molecular emission[16, 17].

In the experiment, if the ultrasound pressure increases, the SL intensity also will be increased quickly. When the different noble gases have the same SL intensity, the needed ultrasound pressures are different. As shown in fig. 2, when the SL is about 35000 counts, the He, Ar and Kr bubbles needed the pressure to be 2.15, 1.70 and 1.22 × 10^5 Pa, respectively. Taking the three pressures into eqs. (1)—(4) and simulating the temperatures, one can obtain the results that the temperatures are different: $T_{\mathrm{He}} > T_{\mathrm{Ar}} > T_{\mathrm{Kr}}$. It indicates that, to produce the same SL intensity, the noble gas that has a larger molecular mass needs a lower temperature. Molecular emission cannot explain this phenomenon. Under this condition, the noble gases participate in the process of light emission. This can be expressed by bremsstrahlung theory[18, 19]. In the bremsstrahlung theory, the noble gas has a weak ionization at high temperature (above 9000 K), light emission predominantly stems from the ionization of a noble gas, and neither molecular gases nor water vapor plays a substantial role. The electrons produced by ionization are absorbed by the near ions and neutral atoms and release photons. The intensity of SL is determined by the degree of ionization: the higher the degree of ionization, the stronger the SL intensity. The degree of ionization not only depends on the temperature, but also on the ionization energy of noble gas[20]:

$$\alpha[T] = \left(\frac{2\pi m_e k_B T}{h^2}\right)^{3/4} \left(\frac{2u_+}{n u_0}\right)^{1/2} \exp\left(-\frac{E_{\mathrm{ion}}}{2k_B T}\right), \tag{5}$$

where m_e is the electron mass and u_+, u_0 are the statistical weights for the ionic and the neutral ground states, respectively. Different noble gases have different ionization energy, for example, $E_{\mathrm{ion}}^{\mathrm{He}} \approx 24.6$ eV, however, $E_{\mathrm{ion}}^{\mathrm{Ar}} \approx 15.7$ eV. It can be seen that from eq. (5), to make the different noble gases have the same degree of ionization, the temperature needed for He is much higher than that for Kr. Therefore, He bubbles need higher ultrasound pressure than Kr. It is consistent with the experiment results.

The gas component inside the bubbles influences SL mechanism. Bremsstrahlung takes place more easily in Kr bubbles than in He. The reason is that the molecular mass of Kr is much larger than that of He, and then Kr bubbles have a higher adiabatic exponent and lower thermal diffusivity, leading to the high temperature at bubble collapse. Although the water vapor consumes a certain amount of energy in the process of dissociation, and baffles the interior temperature increase, the temperature will finally be high enough to make the gas ionized and lead to bremsstrahlung. If there is plenty of gas that can be dissociated easily in the bubble, the thermal chemical reaction may lead to no radiation or only molecular emission of the bubble. Fig. 4 is the curves of SL intensity change with the ultrasound pressure for two species of experiment gas. The gas for curve a is air, and for curve b is mixed gas of nitrogen and oxygen, with the volume ratio for the two gases at 1:1. As shown in fig. 4, the air bubble also can emit light. However, its threshold pressure is much higher than that of Ar or Kr bubbles (fig. 2). And the threshold temperature obtained by numerical simulating is about 5500 K. When the SL becomes stronger at a higher ultrasound pressure (intensity: 21000 counts, ultrasound pressure: 2.975×10^5 Pa), the simulated temperature is only about 6900 K. Therefore, in this luminescent bubble, the photons mostly came from molecular emission.

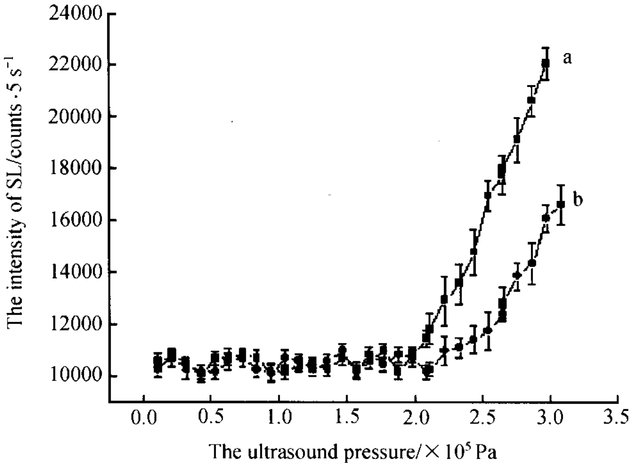

Fig. 4. The SL intensities of air and N_2+O_2 bubbles change with the ultrasound pressures.

From curve b in fig. 4, the bubbles filled with the mixed gas of nitrogen and oxygen can hardly radiate even at a higher ultrasound pressure. The reason is that as for the nitrogen and oxygen inside bubbles, thermal chemistry reaction at high temperature: $N_2+O \rightarrow NO+N$, $N_2+O_2 \rightarrow 2NO$ can easily take place, the needed energy is 3.27 and 1.90 eV, respectively[4]. The energy produced in the bubbles at the collapse is mostly for the two reactions. Consequently, the higher temperature is prevented.

Therefore, the SL mechanism can be analyzed by measuring the threshold pressure and simulating the interior temperature of the bubbles. The possible process may be that the temperature inside the bubbles increases quickly at the collapse of the bubbles; when the temperature reaches about 6000 K, the water molecules are dissociated to hydrogen atoms and hydroxyl radicals. The hydrogen atoms and hydroxyl radicals at different excited states recombine and form the water molecules in different rotational, vibrational, and electronic states, releasing photons and forming the spectrum of the SL. This is a typical molecular emission. Although the process of water dissociation consumes certain energy and prevents the temperature increase, the final temperature could reach high enough to make the noble gas ionized; then, the bremsstrahlung occurs. In fact, SL is a process from molecular emission to bremsstrahlung.

4 Conclusion

In this paper, we measure the threshold pressures and intensities of SL at different ultrasound pressures for different noble gases, simulate the interior temperatures based on the experiment data, and analyze the SL mechanism by comparing the interior temperatures of different noble gas bubbles. The result shows that the SL mainly comes from molecular emission near the threshold pressure, the interior temperature increases quickly with the increment of ultrasound pressure, and the SL is mainly contributed by bremsstrahlung. The whole process of the bubbles' emission is the one of molecular emission to that of bremsstrahlung.

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