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Impact of bubble coalescence in the determination of bubble sizes using a pulsed US technique: Part 2 – Effect of the nature of saturating gas

Rachel Pflieger a,*, Geoffrey Audiger a, Sergey I. Nikitenko a, Muthupandian Ashokkumar b

a ICSM, Univ Montpellier, CEA, CNRS, ENSCM, Marcoule, France
b School of Chemistry, University of Melbourne, Melbourne, VIC 3010, Australia

1. Introduction

Determination of bubble populations and sizes in a cavitation field is of prime importance to optimize sonochemical processes and other ultrasound related applications [1,2]. Among the limited experimental methods available [3–6], only very few allow measurements up to very high US frequencies, i.e., in the range of interest for medical (therapeutic and diagnostics) applications. These indirect methods are based on the correlation between bubble dissolution rate and bubble size. In the first one [3–6], the evolution of the void rate in solution after US irradiation was followed using an electromagnetic method. The second one [7] used an acoustic method to follow the scattering intensity of bubbles vs time. These two methods gave access to a distribution in size of all bubbles in solution, not just to the cavitation bubbles. The third one [8,9] a priori advantageously restricts the focus on sonoluminescing or on sonochemiluminescing bubbles, being based on the measurement of sonoluminescence (SL) or sonochemiluminescence (SCL) intensity under pulsed US conditions. Briefly, by monitoring the evolution of the SL intensity with a decreasing pulse off-time for a constant on-time, the dissolution time of bubbles can be inferred, and from it the corresponding bubble size and size distribution can be determined. Its weaknesses have however been recently underlined [10]. The present study, divided in two parts, aims at a better understanding of the phenomena taking place under pulsed US and at the determination of conditions in which measurements indeed lead to a cavitation bubble size. The first part [10] focused on Ar-saturated water sonicated at 362 kHz. It has brought to light that great care must be taken in the choice of the pulse on-time (t on) and in the experimental geometry. Indeed, if the on-time is not reduced to a minimum (around 1 ms or less for Ar), coalescence of bubbles takes place and leads to the measurement of the size of coalesced bubbles instead of cavitation bubbles. Longer the on-time, larger is the determined size, because more bubbles are formed during t on, which increases the probability of interaction and coalescence. Besides, the formation of a standing wave should be avoided, since it favors coalescence. In optimized conditions, a bubble size of about 2.9–3.0 µm was estimated for Ar-saturated water sonicated at 362 kHz.

This second part (current manuscript) aims at extrapolating the proposed method to other gases (He, Xe, O 2, N 2) showing very different solubilities and physical properties, and to some chosen gas mixtures (Ar-N 2 with different ratios, air). Pre-saturated water was used, without continuous gas flow, since it was previously [10] shown that the presence of a continuous gas flow favors bubble coalescence.

2. Materials & methods

A detailed description of the set-up and of the method can be found...
in Part I of this study [10]. Water was pre-saturated with the gas of interest (He, Xe, O₂, N₂; 99.999%, Air Liquide) by sparging it in the solution for 30 min. Gas mixtures were prepared in situ using a double entry flowmeter (Aalborg). The acoustic frequency used was 362 kHz and the absorbed acoustic power, determined calorimetrically, was 47 W. The water temperature was kept at 10 °C using a thermostat. The on-time was made to vary between 1 and 8 ms, and the off-time, for each \( t_{\text{off}} \) value, between 200 and 1000 ms (it was checked that the SL intensity was very low below 200 ms). Presented evolutions of the SL intensity during \( t_{\text{on}} \) were taken at a \( t_{\text{off}} \) corresponding to the end of the SL intensity increase.

3. Results & discussion

The bubble sizes were measured at an acoustic power of 47 W to avoid standing wave formation. The measurements were also performed at 4.4 W but are not shown here. They confirm that larger sizes are obtained at low acoustic power, similar to the Ar case [10] and that the greater occurrence of coalescence in the standing-wave configuration is not dependent on the nature of the gas.

3.1. Evolution of the SL intensity during \( t_{\text{on}} \)

The evolution of the SL intensity during \( t_{\text{on}} \) (taken at a \( t_{\text{off}} \) corresponding to the end of the SL intensity increase) is presented in Fig. 1 for He, Xe, O₂, N₂ and the gas mixtures air and Ar-N₂ 50–50.

It was previously shown in Ar saturated water that only cases with a continuous increase in SL intensity during \( t_{\text{on}} \) may correspond to cavitation bubbles. A change in slope indeed indicates interaction between bubbles and in particular formation of coalesced bubbles. In Ar-saturated water, a regular increase was observed for \( t_{\text{on}} = 0.5 – 3 \) ms.

Different behaviors are observed for the different gases. For He, with \( t_{\text{on}} = 1 \) ms, the SL intensity increases continuously during 1 ms. Thus there is no indication of pronounced coalescence.

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3.2. Derived bubble sizes

Bubble sizes were calculated from SL intensity vs. \( t_{\text{off}} \) plots using the equation of bubble dissolution [16], as previously [9,17] described. They are summarized in Table 1.

For all studied gases and as was previously observed on Ar, an increase in the determined bubble sizes is seen when increasing \( t_{\text{on}} \). Its extent depends on the gas nature. For instance, it is very marked for He:

considering the smallest size (1.2 µm) as the ambient radius one gets that a bubble arising from the coalescence of 2 bubbles of radius 1.2 µm will have a radius of \( 1.2 \times \sqrt{2} = 1.5 \) µm. Similarly, the coalescence of 2 bubbles of radius 1.5 µm will give a bubble of 1.9 µm radius (\( 1.5 \times \sqrt{2} = 1.9 \mu m \)), that of 2 bubbles of radius 1.9 µm a bubble of 2.4 µm radius, that of 2 bubbles of radius 2.4 µm a bubble of radius 3.0 µm and so on.

Thus the different values in Table 1 can be explained by bubble coalescence, as was previously discussed for Ar [10]. Owing to He low solubility one may assume that the lowest determined value at 1 ms may be considered as a cavitation bubble size (Rₒ): 1.2 µm.

With Xe as the saturating gas, as was discussed above, strong bubble interactions are present already for \( t_{\text{on}} = 1 \) ms and corresponding bubble sizes are expected to be already sizes of coalesced bubbles. The smallest measured size is 4.9 µm. It may arise from the coalescence of:

2 bubbles, leading to a \( R_\text{o} \) of \( 4.9/\sqrt{32} = 3.9 \) µm

3 bubbles, leading to a \( R_\text{o} \) of \( 4.9/\sqrt{33} = 3.4 \) µm

4 bubbles, leading to a \( R_\text{o} \) of \( 4.9/\sqrt{34} = 3.1 \) µm

The values of bubble sizes obtained for the different \( t_{\text{on}} \) in Table 1 are best explained with 3.1 µm (i.e., one can retrieve them by considering coalescence of bubbles of initial Rₒ 3.1 µm).

In O₂ too, interactions between bubbles are already important for \( t_{\text{on}} = 1 \) ms. The smallest measured size is 3.5 µm, and an initial radius of 2.8 µm (=\( 3.5/\sqrt{2} \)) allows to interpret experimental values for the different \( t_{\text{on}} \).

The nitrogen case is particular and very interesting because bubble size distributions are always very narrow – yet, an intense coalescence is observed: 2.2 µm can be obtained by the coalescence of 4 1.4-µm bubbles, 2.8 µm by that of 8 1.4-µm bubbles (or coalescence of 2 previously formed 2.2-µm bubbles). Also the smallest measured size, 1.4 µm, can be derived from coalescence of two 1.1-µm bubbles, or four 0.88-µm bubbles.

To try and shed light on the nitrogen case, it is useful to look at Ar-N₂ mixtures and at air, for which the SL intensity is high enough to allow measurements with \( t_{\text{on}} = 1 \) ms. Air is mainly composed of nitrogen (78%) and of oxygen (21%). Its behavior is thus expected to show similarities with these two gases. Values presented in Table 1 are much smaller than O₂ values and close to N₂ ones, as expected from air composition. However, it is noteworthy that they are smaller than pure N₂ ones, indicating a smaller extent of coalescence. The minimum measured value of 1.1 µm allows to explain the different radius values obtained: 1.4 µm would correspond to the coalescence of 2 bubbles, 1.7 µm to the equivalent coalescence of 4 bubbles, 2.5 µm to 12. Obviously a \( R_\text{o} \) of 0.88 µm (if one assumes that 1.1 µm corresponds to a coalesced bubble) also allows to explain the different experimental radii.

Sizes derived in Ar-N₂ mixture for \( t_{\text{on}} = 1 \) ms (Table 2) confirm that N₂-Rₒ is most probably 0.9 or 1.1 µm. As soon as some N₂ is present in Ar, bubble sizes strongly decrease. They further decrease with an increase in N₂ content, reaching 1.1 µm for 80% N₂.

3.3. \( R_\text{o} \) and extent of coalescence

Table 3 summarizes the derived \( R_\text{o} \) for different gases and the extent of coalescence at different \( t_{\text{on}} \), i.e., the number of bubbles of equilibrium radius \( R_\text{o} \) leading to the experimentally observed bubble sizes. Obviously, the real mechanism does not involve the coalescence of this (large) number of \( R_\text{o} \)-bubbles in one single step, but successive coalescences of bubbles of growing size.

Let’s first consider equilibrium radii and compare them with data from the literature. This comparison is deliberately restricted to multi-bubble studies since single-bubble measurements are usually performed in (at least partly) degassed solutions and the dissolved gas content is a
Fig. 1. Evolution of the SL intensity during $t_{\text{on}}$ for different $t_{\text{on}}$ for a) He ($V_{\text{PMT}} = 1000$ V), b) Xe ($V_{\text{PMT}} = 550$ V), c) $O_2$ ($V_{\text{PMT}} = 1000$ V (1–4 ms) or 650 V (6 ms)), d) $N_2$ ($V_{\text{PMT}} = 1000$ V), e) air ($V_{\text{PMT}} = 800$ V) and f) Ar-$N_2$ 50–50 ($V_{\text{PMT}} = 1000$ V). The red arrow indicates the beginning of $t_{\text{on}}$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)
Considered R₀ and extent of coalescence (in number of equivalent R₀-bubbles) for the different gases and t₀ms.

Table 3

| Ar       | He  | Xe  | O₂   | N₂   | air | Ar-N₂ 50-50 |
|----------|-----|-----|------|------|-----|------------|
| R₀, µm   |     |     |      |      |     |            |
| 3.0      | 1.2 | 3.1 | 2.8  | 1.1 (or 0.9) | 1.1 (or 0.9) | 1.4 |
| t₀ms = 1 ms | 2-3 | 5.3 | 4.5  | 2.3  | –   | 1.2 (2-4)  |
| t₀ms = 2 ms | 2-4 | 3-10 | 5-8  | 2.6  | 2 (4) | 1-2 (2-4)  |
| t₀ms = 3 ms | 2-3 | 13-40 | 7-16 | 3-6  | 8-10 (16-20) | 4-5 (8-10) |
| t₀ms = 4 ms | 1-4 | 17-21 | 16-22 | 7-17 | 16 (32) | 12-15 (24-30) |
| t₀ms = 5 ms | 4-5 |      |      |      |     | 7-16 |
| t₀ms = 6 ms | 5-9 | 86-159 | –   | 12-26 | –   | 22-30 (44-60) |
| t₀ms = 7 ms | 6-11 |      |      |      |     | – |
| t₀ms = 8 ms | 6-11 | 81-159 | –   | –   | –   | 41-56 (82-112) |
energy concentration, would be necessary to form a plasma out of a gas with high ionization potential or subject to energy losses by dissociation, chemical reactions and vibrational/rotational excitation (i.e. a molecular gas). The gas viscosity may play a role in the bubble dynamics, the formation of surface instabilities and the symmetry of the collapse.

Considering rare gases, Ar and Xe show similar $R_0$ while He bubbles are much smaller. Smaller the bubble size, the more symmetrical is the collapse, thus the strongest it should be – which is necessary for He to form a plasma, due to its high ionization energy (Table 4). Besides, gas segregation [31] inside the collapsing bubbles and enhanced water condensation on the bubble wall at the end of the collapse in the He case may also decrease the necessary $R_0$ to reach a critical energy concentration. It is also to be noted, considering Ar and Xe, that a similar $R_0$ does not imply a similar radius at the maximum of bubble expansion, due to the very different gas solubilities (Xe bubbles will grow more).

This simplified approach allows to shed some first light on possible reasons leading to the experimentally determined $R_0$ for different gases. It however does not explain the particular case of nitrogen and Ar-N$_2$ mixtures where even a very low amount of N$_2$ leads to a strong decrease in $R_0$. This example underlines that phenomena coming into play are complex, not limited to thermodynamics, and deserve detailed studies, such as on the influence of the gas nature on bubble deformation during collapse.

Concerning the extent of coalescence (Fig. 2), it also appears to result from the interplay of different physical phenomena: comparing Fig. 2 with Table 4 shows no direct correlation of the extent of coalescence with any of the listed properties. It also does not seem to be correlated with $R_0$ (Fig. 3). Considering only rare gases, where Ar shows the lowest coalescence, one may speculate that xenon’s much higher solubility leads to a higher number of bubbles and consequently higher probability of coalescence, and that helium’s very high diffusion coefficient may lead to more interactions between bubbles. Oxygen shows a behavior close to Ar, though with some tendency towards more coalescence. This similarity is in agreement with their close solubilities and bubble size. Nitrogen and air show similar ranges of coalescence, at least if the same $R_0$ is considered for both.

It is to be noted that the width of the coalescence range strongly depends on the gas nature, even for a similar mean equivalent number of bubbles of radius $R_0$: for instance the interval is very large for He and ν, though with some tendency towards more coalescence. This fact, combined to the different evolutions of coalescence at larger $t_{off}$ (Table 3), clearly confirms that several parameters play a role in this complex phenomenon: gas solubility, diffusion coefficient, bubble size, but also most probably the bubble dynamics, its deviation from sphericity and the extent of emission of daughter bubbles at collapse.

4. Conclusion

Using the recently developed approach [10] coupling pulsed US and SL intensity measurements for an interval of on-times, ambient radii could be evaluated for different gases: 3.0 μm for Ar, 1.2 μm for He, 3.1 μm for Xe, 2.8 μm for O$_2$, around 1 μm for N$_2$ and air. As previously observed on Ar, increasing the on-time leads to the determination of the size of coalesced bubbles. The extent of coalescence strongly depends on the gas nature. No single physical property of the gas allows to explaining it, but it seems to increase with the gas solubility, which is attributed to the formation of a larger number of bubbles, thus increasing the probability of interactions. Also a high gas diffusion coefficient appears to favor coalescence. A devoted theoretical modelling study would be needed. It would also be interesting to try and link present results with observations of the bubble dynamics and in

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**Table 4**

Considered $R_0$, gas solubility [11], gas density, gas viscosity, speed of sound, diffusion coefficient [12-14] and ionization energy [15].

| Gas  | Ar | He | Xe | O$_2$ | N$_2$ | air | Ar-N$_2$ | $R_0$, μm | Gas solubility, mol/L | $\rho$, kg/m$^3$ | $\mu$, μm/s | Speed of sound in the gas, m/s | Gas diffusion coefficient, m$^2$/s | Ionization energy, eV |
|------|----|----|----|-------|-------|-----|----------|----------|----------------------|----------------|------------|----------------------------|----------------------|------------------|
|      | 3.0 | 1.2 | 3.1 | 2.8   | 1.1   | 1.1 | 1.4      | 3.0      | 1.48     | 10$^{-3}$ | 4.0          | 7.2       | 1.75             | 22.7           | 19.9          | 23.2        | 20.7       | 17.9         | 1.23 | 6.76 | 1.57 | 1.54 | 1.29 | 15.76 | 24.59 | 12.13 | 12.07 | 15.58 | 10$^{-9}$ | 10$^{-9}$ | 10$^{-9}$ | 10$^{-9}$ | 10$^{-9}$ | 10$^{-9}$ |

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Fig. 2. Evolution of the extent of coalescence (in equivalent number of bubbles of radius $R_0$) with $t_{on}$ for Ar, He, Xe, O$_2$, N$_2$ and air.

Fig. 3. Extent of coalescence (in equivalent number of bubbles of radius $R_0$) as a function of the estimated $R_0$, for $t_{on} = 4$ ms.
particular bubble deformation (surface instabilities) and emission of
daughter bubbles at collapse.

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Declaration of Competing Interest

The authors declare that they have no known competing financial
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