Vol 14 No 5, May 2005
1009-1963/2005/14(05)/0999-07
Chinese Physics
© 2005 Chin. Phys. Soc.
and IOP Publishing Ltd

Luminescence of transient single cavitation bubbles in non-aqueous liquids produced by the modified tube-arrest method*

Wu Xian-Mei(吴先梅)†, Ying Chong-Fu(应崇福), and Li Chao(李 超)

Institute of Acoustics, Chinese Academy of Sciences, Beijing 100080, *China*

(Received 6 August 2004; revised manuscript received 13 January 2005)

Large transient single bubbles of effective diameters in centimetres were generated by the modified tube-arrest method in 7 nonaqueous liquids besides water and in glycerin–water mixtures. During collapse, nearly all these bubbles gave off light emissions of various intensities, which in 6 liquids were correlated with the saturation vapour pressure and viscosity of the respective liquid. Bubbles in ethylene glycol and propylene glycol did not follow this rule, but those in the former liquid showed some unusual regularity in luminescence variation, and the bubbles in both the liquids were particularly bright. The luminous properties of the transient single bubble are compared with those of the usual stable single bubble.

Keywords: cavitation, transient single bubble, modified tube-arrest method, luminescence in various liquids
PACC: 4755B, 7860M

1. Introduction

The successful creation in 1992[1] of a single stable cavitation bubble flashing with clock-like accuracy has aroused new and wide interest in the study of the cavitation phenomenon. Much advance has been achieved since.[2−4]

Back in the early 1960s, Wang and Zhang produced a transient single bubble accompanied by optical radiation,[5,6] following a method first designed by Chesterman,[7] which was later named the tube-arrest method.[8] A single bubble is here called transient (TSB) when it lasts in general but one cycle of growth and disruption, in contrast to the well-known "stable" single bubble (SSB) which may survive billions of cycles. Wang and Zhang, however, after Schmid,[9] used a seeded bubble nucleus and obtained single bubbles as large as 2cm in diameter.

Nearly 20 years later, Overton and Trevena applied the tube-arrest method to generate cavitation and succeeded in determining the cavitation thresholds in various liquids.[10,11] They improved Chesterman's work by mounting a piezoelectric transducer at the bottom of the tube, enabling the study of the pressure change in the liquid.

We have in the past few years reapplied this method to the cavitation study with the modifications of adding a seeded nucleus and a pressure-monitoring transducer, in an attempt to evaluate the merits and demerits of so-generated single bubbles.[12] Some of the earlier repetitive experimental results, which had been obtained in co-operation, were reported recently by Chen and Wang[13] and thus will not be described here. We shall also not present here our new findings on the behaviour of this bubble-generating method.[12] We shall focus below mainly on our study of luminescence of the TSB in several non-aqueous liquids. The data reveal that the TSBs in many of the liquids studied are much brighter than the SSBs in the same liquids, as reported in earlier papers. In this respect, the TSBs differ from the SSBs.

2. Experimental set-up

Our experimental arrangement is shown in Fig.1. A glass tube sealed at the bottom and open at the top is partly filled with the liquid to be examined. The tube diameter is usually between 2.6 and 2.8cm

*Project supported by the National Natural Science Foundation of China (Grant No 10434070)
†E-mail: wuxianmei@hotmail.com

while its length is roughly one metre, the liquid level in the following experiments being usually 50cm high. The tube with its liquid content is fastened indirectly to a metallic spring suspended at one end. To seed a bubble nucleus, a rod with a small dent of a diameter of roughly 1mm in its lower end is inserted vertically from air into the liquid so that air is trapped inside the dent. The rod is suspended along the centreline of the tube, with its lower end usually about 5cm from the tube bottom. Being separated from the tube, the rod remains fixed in space whenever the tube slides vertically. For bubble production, the tube is first pulled downward for some distance, 8cm in all the following experiments, with the spring thus stretched. It is then released so as to be quickly pulled upward by the spring. After shooting up for this distance, the tube is suddenly arrested by a stop shown in the figure. In the following experiments, the moving distance of the tube is so fixed that the upward-moving speed v of the glass tube at the instant of being arrested was measured to be consistently 1.7m/s. A tiny single bubble can now be seen to emerge out of the lower end of the axial rod where the dent lies. The dynamic behaviour of the bubble is monitored with, see Fig.1(a), a high-speed CCD camera (Photron FastCam Super

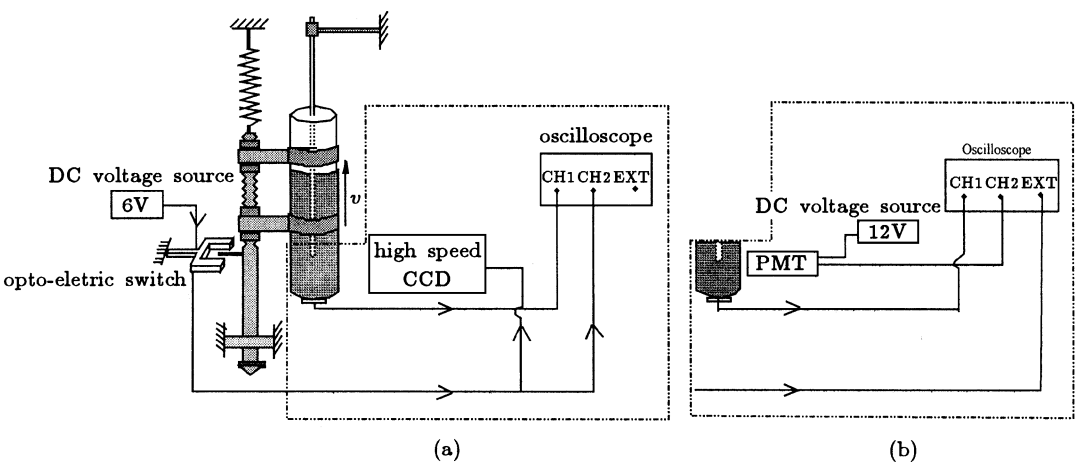


Fig.1. Experimental set-up for generating TSB and for (a) photographing the bubble dynamics, (b) recording the bubble luminescence.

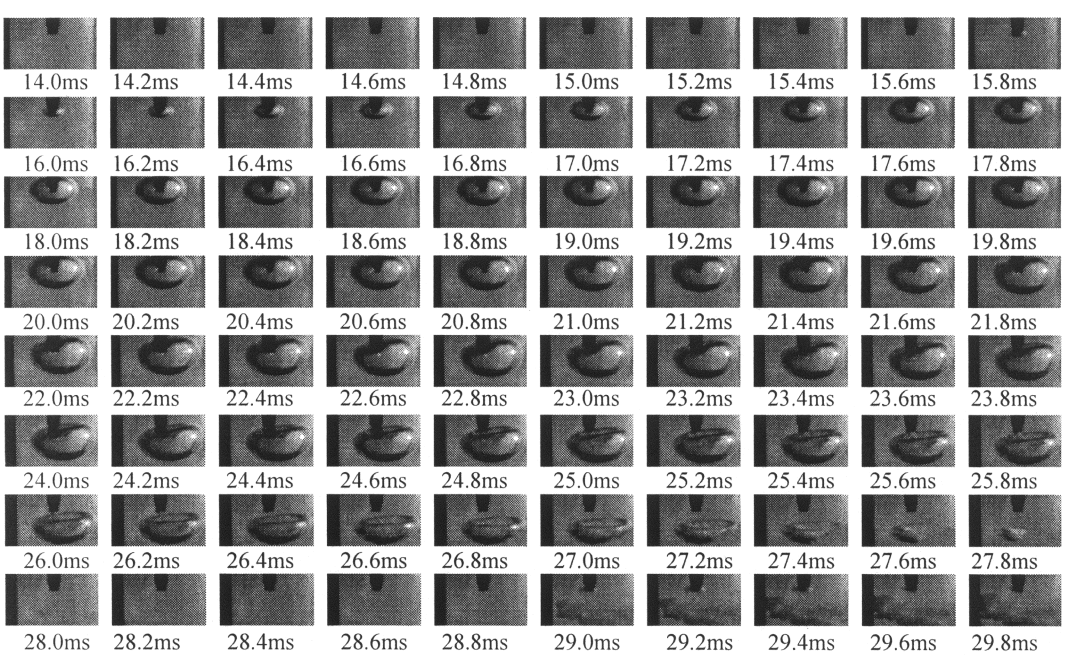


Fig.2. A series of high-speed photos exhibiting the growth, the collapse and the afterbounces of a bubble in acetone. The maximum lateral diameter is measured to be 2.33cm.

10K series), the frame rate of which varies between 30fps and 10000fps. During photo taking, the bubble is illuminated externally. The bubble is observed to grow, shrink, fracture, and rebounce more than once not as a single bubble but as a disc-shaped cluster of small bubbles. In the glycerin and silicone oil that we have studied, however, the bubble keeps its entity as a whole bubble for at least 2 or 3 rebounces. In almost all cases, the growing bubble expands to a very large size, of the order of magnitude of centimetres in its effective diameter. Usually, it is apple-shaped rather than spherical, likely due to the existence of the rod from which it originates. Figure 2 gives an illustration of a developing single bubble in acetone inside a glass tube, shown by a series of high-speed photographs. This liquid is included in our study because deuterated acetone has been the active medium in the recent experiments of sonofusion.[14]

We monitor the luminescence of the bubble with a photomultiplier (Hamamatsu R212), as shown in Fig.1(b). The photomultiplier tube (PMT) has a rise time of 2.2ns and a spectral response of 185 to 650 nm. The oscilloscope is a digital type from Lecroy (wave runner LT372).

We measure the pressure field inside the liquid in the glass tube before and after the tube is arrested. A piezoelectric pill is stuck onto the outside surface of the tube bottom as can be seen in Fig.1. We did occasionally insert a small hydrophone into the liquid through the open top of the glass tube. The pressure-time curve measured by the pill, an example of which is given in Fig.3, is similar to that observed in Ref.[10]. We now understand that the part between "A" and "B" in the curve (Fig.3) is the time interval of the bubble's growth. The sudden halt of the glass tube produces a tensile pulse,[8] indicated by "A" of Fig.3, emerging from the tube bottom and travelling upward, which on confronting the bubble nucleus, excites its expansion and then likely exhausts itself. The bubble grows until it collapses when it emits a strong pressure pulse "B" which is usually followed by some rebouncing pressure waves.

For monitoring of the bubble dynamics with the use of a high-speed camera, we need a strong light illumination to the bubble; on the other hand, for the recording of the bubble luminescence, a dark background is requisite. The pressure–time curve felicitously serves as an indirect indicator of the dynamic features of the bubble with or without illumination. So, more often we record the pressure–time curve than taking high-speed photographs.

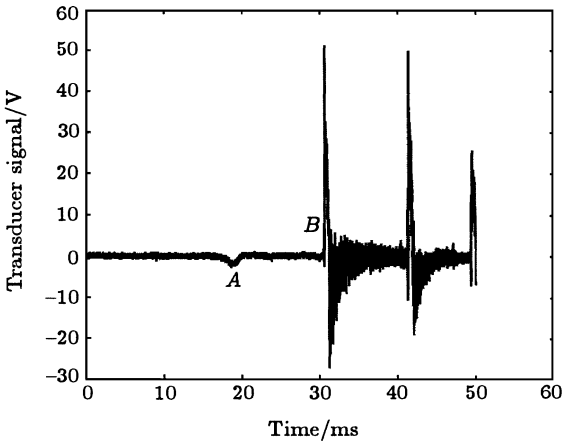


Fig.3. The pressure–time curve in the liquid measured with the piezoelectric pill stuck onto the outside surface of the tube bottom as shown in Fig.1.

3. Luminescence from TSBs in various liquids—measuring procedure and results

We measured luminescence from TSBs generated individually by the tube-arrest method in 8 liquids and one species of liquid mixtures. The 8 liquids were #1 acetone, #2 ethyl alcohol, #3 water, #4 glycerin, #5 dimethyl phthalate, #6 dimethyl silicone oil, #7 ethylene glycol, #8 1, 2-propylene glycol, while the one species of mixtures was water-glycerin mixtures with glycerin concentrations ranging from 0% to 100% in volume. These liquids were of analytic purity except water which was distilled.

During each measurement, the accelerated tube was arrested at 1.7m/s, and the dynamics of the luminescent TSB was inspected with the high-speed CCD camera. The dynamics of the TSBs in the low-viscosity liquids (#1, #2, #3, #5, #7, #8) were rather similar. Thus the maximum lateral radii of bubbles were within the range of 2.0–2.5cm, and the bubble fragmented into a disk-shaped cloud of small bubbles at the very first collapse before any rebouncing. In the high-viscosity liquids, the TSBs, however, behaved differently. In the silicon oil (#6) and the glycerin (#4), the TSBs rebounced before their disintegration. The rebouncing behaviour of TSBs in these two liquids also differed slightly from each other. For example, in one set of observations in the silicone-oil, the maximum lateral radius decreased with successive rebouncing, with the maximum lateral radius during growth being ca 2.3cm. In the glycerin, the

first maximum lateral radius during growth was noticeably small, being only about 3mm, and then a second maximum extended to ca 1.2cm; afterwards the rebouncing bubble shrinked monotonically.

Before the measurement of luminescence, the liquids were degassed by pumping generally for a time from 1 to 2h, while some liquids like acetone were degassed by low-power low-speed pumping to avoid being pumped away. Water was boiled before pumping and so was glycerin which among the liquids studied was the most difficult to degas. This liquid not only was very viscid, but was also known to be very hygroscopic. We noted that in studying the sonoluminescence of multi-bubbles (MB) in glycerin–water mixtures, Young[15] used "a specially distilled supply" of glycerin of 99.9% while Golubnichii *et al*[16] reported to have removed all traces of water from the glycerin by distillation at 150°C. Lacking suitable laboratory set-ups, we first boiled pure glycerin for 20min and then pumped the liquid for ca 6h, combined with occasional violent shaking of the container. Even so, small amount of tiny bubble became visible in the liquid on being shaked, but disappeared on short standing. As to the silicone oil, no remnant bubbles were seen on being shaked after sufficient pumping.

The luminescence intensities of the TSBs (I_{TSB}) in the first 7 liquids were then measured with the photomultiplier. Measurements for each liquid were taken 5 times and the average value is used as representative. Nevertheless, the TSB in the 7th liquid ethylene glycol seemed to vary widely in its luminescence with some unusual regularity. So we studied it more carefully and present the result of measurement separately near the end of this section. For comparison, we add the 8th liquid of 1, 2-propylene glycol, of which the result will also be described later.

The averaged luminescence intensities of TSBs in the first 6 liquids are summarized in Fig.4 (solid circles) with the deviations being indicated by error bars. It can be seen from Fig.4 that the I_{TSB} in these liquids vary at most by up to 3 orders of magnitude. It may be stressed that the stable single bubbles (SSBs) generated acoustically in a few of the above-studied liquids have been reported to emit very weak light. Weninger *et al* in 1995[17] measured the sonoluminescence from SSB in n-dodecane, ethanol, 1-propane, 2-propanol, 1-butanol, 1-pentanol and silicon oil (Dow Corning 200 fluid, 1cSt viscosity) as functions of temperature. The absolute sonoluminescence intensities in these liquids were all found to be lower than that in water with 2×10^4Pa air at room temperature by up to 3 orders of magnitude. In water our comparative measurement showed that the luminescence intensity emitted by SSB is about 10 times weaker than that emitted by TSB.

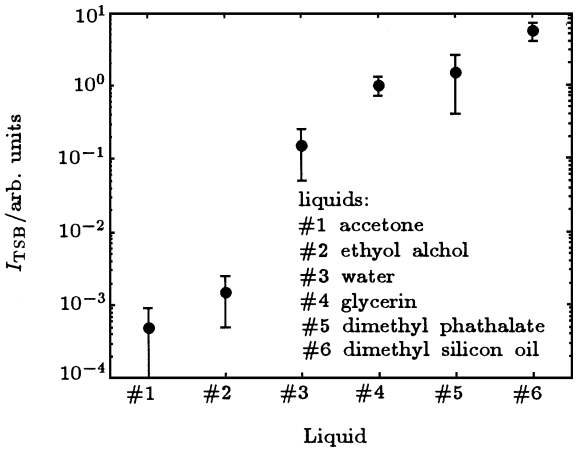


Fig.4. Measured luminescence intensity (I_{TSB}) from TSBs in 6 liquids at room temperature (The vertical ordinate is in logarithmic). Measurements for each liquid are taken 5 times; the average value is denoted by a solid circle while the deviations by the error bar.

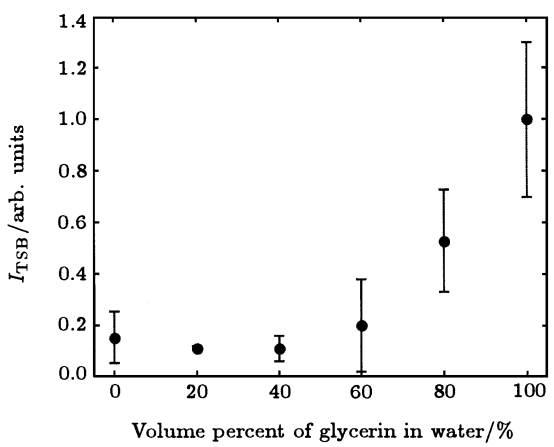


Fig.5. Measured luminescence intensity from TSB (I_{TSB}) in water-glycerin mixture at room temperature versus volume percent of glycerin in water. Also, for each mixture measurements were taken 5 times and averaged.

We measured also the I_{TSB} from TSBs in water-glycerin mixtures with different proportions of glycerin (0, 20, 40, 60, 80, 100 vol%). The measured values are given in Fig.5. It can be seen that the I_{TSB} mainly increases with the increasing content of glycerin. In the case of SSB, Gaitan *et al*[18] have reported the measurement of spectra of single bubble sonoluminescence in water–glycerin mixtures. Their results show that the sonoluminescence intensity of SSB at 40 vol%

of glycerin is roughly two orders of magnitude weaker than that at 0 and 20 vol%. The situation for higher vol% of glycerin was not mentioned.

During the study of the 7th liquid, the ethylene glycol, we noticed, after several trials, that the I_{TSB} increased consistently on repetitively growing new bubbles by using the tube-arrest method. More careful study shows that in other cases, the I_{TSB} might decrease consistently and that there appears certain upper and lower limits. The results of two separate systematic series of study on successively growing new bubbles and their corresponding I_{TSB} are shown in Figs.6 (a) and 6(b). The time interval between consecutive growths of two new bubbles was some 3min. In these two series of study, continuous growing was interrupted at the points "A" marked in the figure, for about 66h in Fig.6(a) and for about 72h in Fig.6(b). The liquid used to obtain Fig.6 (b) was from a new supply different from that in Fig.6(a). The short flashes are so bright that they can be seen by unaided eyes in a dark room and recorded by an ordinary camera and film.

Then we examined the liquid 1, 2-propylene glycol $\mathrm{CH_2\text{-}CH_2\text{-}CH_3}$ (OH OH) but did not observe any behaviour of the I_{TSB} similar to that exhibited by ethylene glycol $\mathrm{CH_2\text{-}CH_2}$ (OH OH). The TSB in the former liquid is, however, nearly as bright as that in the latter.

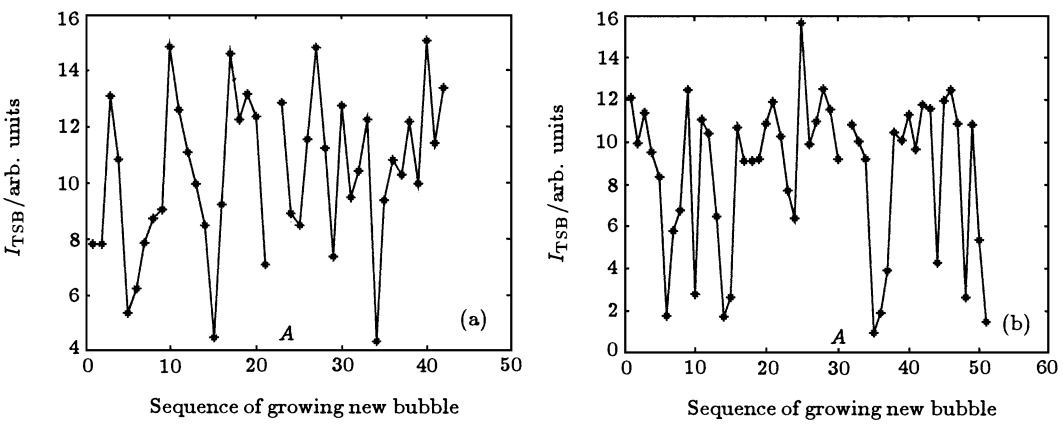


Fig.6. The luminescence intensity from TSB in ethylene glycol, observed to vary with some regularity when new bubble is generated successively. Details are given in the text.

4. Discussion

4.1 Comparison between luminescence intensities from TSB and SSB

We have noted above that the most prominent feature of TSB compared to SSB is its large light-emitting capability in many liquids. As mentioned earlier, Weninger *et al*[17] detected but very weak emissions from SSBs in several organic liquids including low-viscosity silicone oil. Later, Didenko *et al*[19] generated more intense single bubble sonoluminescence (SBSL), of the order of the SBSL in water, in a series of rather special liquids which they chose by deduction to be polar aprotic. On the contrary, we found here that TSB can emit sufficient light in liquids not meeting the criteria set by Didenko *et al.* For example, we found TSB in silicone oil to emit particularly strong light while Didenko *et al* reported that SBSL was not observed from silicone oil.

4.2. Comparison between luminescence intensities from TSBs in various liquids

The intensities I_{TSB} from TSBs in the 6 liquids not including ethylene glycol and propylene glycol can differ by 3 or more orders of magnitude. To explain these wide differences, mechanisms of luminescence from TSB should be taken into account in detail; since this is quite a task at this time of new experimental establishments, we shall take a phenomenological approach to gain some qualitative understanding. Following the steps first introduced by Jarman,[20] we try to see whether I_{TSB} is correlated with some physical properties of the relevant liquid. Some possible significant properties at 20°C of the 6 liquids, including the saturation vapour pressure P_{s}, the surface tension σ and the dynamic viscosity η are listed in Table 1. Two of these physical constants appear in the well-known Rayleigh–Plesset equation while the third (P_{s}) is nowadays extensively discussed.[21,22] The same properties of the peculiar ethyl glycol and the related propylene glycol are also listed in the table for later reference.

Table 1. Three kinds of physical properties at 20°C of the 8 liquids studied in the present paper.

No.	Liquid	Saturated vapour pressure / Pa	Dynamic viscosity / Pa·s	Surface tension / $N \cdot m^{-1}$
1	acetone	2.463×10^{4}	0.33×10^{-3}	2.37×10^{-2}
2	ethyl alcohol	5.85×10^{3}	1.20×10^{-3}	2.24×10^{-2}
3	water	2.3×10^{3}	1.00×10^{-3}	7.28×10^{-2}
4	glycerin	0.027	1.499	6.33×10^{-2}
5	dimethyl phthalate	2.7	1.8×10^{-2}	6.0×10^{-2}
6	dimethyl silicone oil	~ 0	1.075	2.1×10^{-2}
7	ethylene glycol	8	2.1×10^{-2}	4.84×10^{-2}
8	1, 2- propylene glycol	10	6.05×10^{-2}	3.8×10^{-2}

We make a trial of picking out the 2 parameters P_s and η, to define a new parameter $\xi = A\frac{2}{\eta^{1.3}P_s}$ for each of the first 6 fluids in Table 1, the constant A being unity if η is measured in cP and P_s in mmHg, but A=59.6 if η is measured in Pa·s and P_s in Pa. In the lower part of Fig.7, I_{TSB} is plotted as a function of ξ for the 6 liquids. It can be seen that I_{TSB} in each of the 6 liquids is correlated reasonably well with the parameter $\xi = A\frac{2}{\eta^{1.3}P_s}$ for the corresponding liquid.

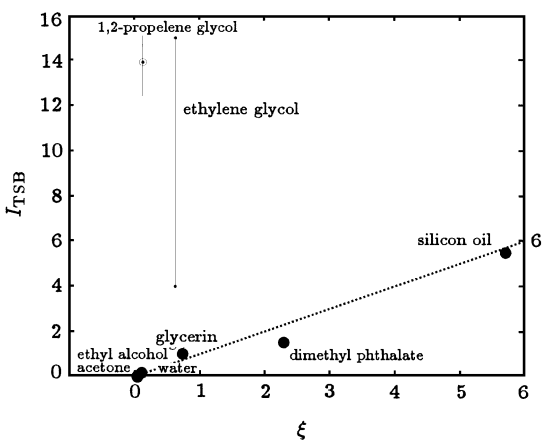


Fig.7. Measured (average) I'_{TSB}s in the 6 liquids given in Fig.4 are plotted as a function of ξ for the 6 relevant liquids. $\xi \equiv A\frac{2}{\eta^{1.3}P_s}$ (see text). Measured I_{TSB} in ethylene glycol and 1, 2-propylene glycol is also plotted; they appear in the upper part of the figure. The values of I_{TSB} in ethylene glycol show up as a vertical line-section because they vary within two limits.

Obviously, the exact form of the parameter ξ should not be taken seriously. What the correlation signifies is but the fact that the lower the saturation vapour pressure of the liquid and the lower the liquid viscosity, the brighter the TSB. It may be noted that, in the case of multi-bubbles (MB), Jarman[20] correlated the intensity with $\frac{\sigma^2}{P_s}$ for 15 liquids, while Golubnichii *et al*[23] correlated for 17 liquids the so-named amplitude of sonoluminescence scintillations with the free energy of molecular interaction $\Delta\varepsilon = RT\ln\frac{RT}{P_sV_m} - RT$, where V_m is the molar volume of the liquid. Flint and Suslick[24] experimentally found, for 3 linear alkanes, a relationship of Be^{-CP_s} for the sonoluminescence intensity dependent on the saturation vapour pressure, B and C being certain appropriate constants.

The correlation between I_{TSB} and ξ might be extended to the case of the various glycerin–water mixtures. Unfortunately, we have not found any experimental or theoretical data on the total saturation vapour pressure of these mixtures (the Raoult diagram) although data on the corresponding η are available, e.g. in Ref.[15]. We thus cannot test the applicability of the correlation formula.

4.3. The unusual behaviour of luminescence from TSB in ethylene glycol

The unusual behaviour of luminescence exhibited by TSB in ethylene glycol reminds one of some two-state phenomenon.[25] It may be that the newly generated bubbles are all confined to the liquid region in the proximity of the tip of the fixed central rod within the glass tube. When the I_{TSB} in this liquid is plotted as a function of ξ in Fig.7, the values show up also as a vertical line-section way off the diagonal line which correlates I_{TSB}'s and ξ's of the earlier 6 liquids. The plots in Fig.7 indicate that TSBs in ethylene glycol and 1,2-propylene glycol can be far brighter than those in other liquids.

5. Conclusion

We have generated TSBs by the modified tube-arrest method in various liquids including glycerin, high-viscosity silicone oil and ethylene glycol, and have obtained very high luminescence intensities from some of the bubbles. This practice is very different from that associated with the usual stable single bub-

ble and will help to greatly extend the area of study of the luminescent properties of various liquids, inorganic or organic. In this respect, the TSB more closely resembles the multiple-bubble (MB), and this promises more simple, incisive circumstance for the study of cavitation luminescence and may provide one with much detailed information. The present paper has obtained some preliminary results which are indicative of new interesting directions for further research.

There are also many problems to be clarified. For example, what is the exact composition of gas and vapour within the bubble and how does the large bubble shine on breaking? (It may be noted that TSB does not need to be large in the present tube-arrest method.) Also, there are many measures which could be taken. One example is that in analogy with the case of SSB, it is natural to think of measuring the light spectrum. This would be easier when in the future even brighter bubble can be expected. Lepoint *et al* in 2003[26] and Chakravarty *et al*[27] more recently reported on super-bright luminescence in sulphuric and phosphoric acids respectively from MB and from bubbles in water hammer.

Acknowledgements

During the early stage of re-establishment of the set-up for TSB generation, close co-operation with Chen Qi-Dai and Deng Jing-Jun and valuable discussions with Wang Long, Shen Jian-Zhong and An Yu are gratefully acknowledged.

References

[1] Gaitan D F, Crum L A, Roy R A and Church C C 1992 *J. Acoust. Soc. Am.* **91** 3166
[2] Barber B P, Hiller R A, Lofstedt R, Putterman S J and Weninger K R 1997 *Phys. Rep.* **281** 65
[3] Hammer D and Frommhold L 2001 *J. Mod. Opt.* **48** 239
[4] Brenner M P, Hilgenfeldt S and Lohse D 2002 *Rev. Mod. Phys.* **74** 425
[5] Wang C H and Zhang D J 1964 *Acta Acoust.* **1** 59 (in Chinese)
[6] Zhang D J and Wang C H 1966 *Acta Acoust.* **3** 14 (in Chinese)
[7] Chesterman W D 1952 *Proc. Phys. Soc.* (London) B **65** 846
[8] Trevena D H 1987 *Cavitation and Tension in Liquids* (Bristol and Philadelphia: Adams Hilger)
[9] Schmid J 1959 *Acoustica* **9** 321
[10] Overton G D N and Trevena D H 1981 *J. Phys.* D: *Appl. Phys.* **14** 241
[11] Overton G D N, Williams P R and Trevena D H 1984 *J. Phys.* D: *Appl. Phys.* **17**, 979
[12] Wu X M 2003 *Post-doctorial Report,* Institute of Acoustics, Chinese Academy of Sciences (in Chinese)
Wu X M and Ying C F 2003 *Tech. Acoust.* **22** supplement 288 (in Chinese)
[13] Chen Q D and Wang L 2004 *Chin. Phys.* **13** 564
[14] Taleyarkhan R P, West C D, Cho F S, Lahey R T Jr., Nigmatulin R F and Block R C 2004 *Phys. Rev.* E **69** 036109
[15] Young F R 1965 *Nature* **206** 706
[16] Golubnichii P I, Goncharov V D and Protopopov K V 1970 *Sov. Phys. Acoust.* **16** 115
[17] Weninger K, Hiller R, Barber B P, Lacoste D and Putterman S J 1995 *J. Phys. Chem.* **99** 14195
[18] Gaitan D, Atchley A H, Lewia S D, Carlson F J and Maruyama X K 1996 *Phys. Rev.* E **54** 525
[19] Didenko Y T, Mcnamara III W B and Suslick K S 2000 *Nature* **407** 877
[20] Jarman P 1959 *Proc. Phys. Soc.* (London) **723** 628
[21] Xie C G, An Y and Ying C F 2003 *Acta Phys. Sin.* **52** 102 (in Chinese)
[22] An Y, Xie C G and Ying C F 2003 *Chin. Phys. Lett.* **20** 575
[23] Golubnichii P I, Goncharov V D and Protopopov K V *Soviet Physics-Acoustics* **16** 323
[24] Flint E B and Suslick K S 1989 *J. Am. Chem. Soc.* **111** 6987
[25] Young F R 1989 *Cavitation* (London: McGraw-Hill) pp105-108
Matheson A J 1971 *Molecular Acoustics* (London: Wiley-Interscience) Chap. 12
[26] Lepoint T, Lepoint-Mullie F, Labowret S, LeveQue J M, Petrier C, Krefting D, Geisler R, Mettin R, Kurz T and Lauterborn W 2003 Abstracts of technical papers, *World Conference on Ultrasonics 2003*, Paris, p115
[27] Chakravarty A, Georghion J, Phillipson T E and Walton A J 2004 *Phys. Rev. E* **69** 066317