MOVING SINGLE BUBBLE SONOLUMINESCENCE IN INORGANIC ACID SOLUTIONS

A. Troia, D. Madonna Ripa, R. Spagnolo
Istituto Elettrotecnico Nazionale "Galileo Ferraris", Acoustic Dept., Torino ITALY
troia@ien.it

Abstract

The phenomenon of sonoluminescence has still some unsolved aspect. Recently [1], it was found that a single cavitating air bubble in polar aprotic liquids (including formamide and adiponitrile) can produce very strong sonoluminescence while moving around the resonator. Here we describe some experiments conducted in aqueous solution of Phosphoric and Sulphuric Acid. In these liquid media, a single trapped bubble can emit light during the well known "jittering" phase or during the Moving-Single Bubble condition; increasing the acoustic pressure a discrete lattice of trapped bubbles generates ("Few Bubble Sonoluminescence"). Each bubble of the cluster emits very intense light flashes and changes its position in the lattice while the overall spatial distribution of the bubbles is maintained stable in time. Some preliminary results obtained by measurements of relative light intensity and Mie-scattering are reported.

Introduction

Since its discovery in 1992, single bubble sonoluminescence (SBSL) has represented a challenge to theoretical and experimental researchers who were trying to grasp the effective link between non-linear bubble dynamics and the light production mechanism. Many physical and chemical models (mainly based on the phenomenology observed in pure water) have been proposed, but it was not possible to definitively test their predictions, owing to the lack of counterexamples, i.d. host liquids -different from pure water- that could sustain stable SBSL.

On October 2000, a paper by Didenko et al. [1] reported stable SBSL from non-aqueous liquids with a series of new phenomena, like moving single bubble sonoluminescence (MSBSL) and molecular emission peaks in the electromagnetic spectra. These features could shed some light on the reliability of the various collapse models at the origin of SBSL.

Finally, T. Lepoint et al. [2] reported their findings about the "ultrabright" multibubble sonoluminescence obtained in concentrated (97%) sulphuric acid, pointing out the possibility of strong single bubble sonoluminescence from the same substance [3]. Following these suggestions by T. Lepoint, we have performed some experiments with concentrate aqueous solutions of H_2SO_4 and H_3PO_4, obtaining strong light signal from levitated bubbles, MSBSL, sonoluminescence from bubbles in the "jittering" condition and the spontaneous formation, at high acoustic pressure, of a tridimensional lattice of emitting bubbles.

Experimental Apparatus

The SBSL set-up used in this work consists of a 50 cm^3 spherical quartz flask driven at a resonance frequency of 37 kHz by a piezoelectric tube glued to the bottom. The bubble activity is monitored by a small piezoceramic disk mounted at bottom-centre of the cell. This "pick-up" microphone was calibrated against a B&K 8103 hydrophone. Light emission is detected by a Hamamatsu R7400 photomultiplier tube (PMT). In order to monitor and record the dynamic of bubbles, a Mie-scattering technique was realised (the same photomultiplier tube is used to detect SL pulses and scattered laser light, see fig. 1).

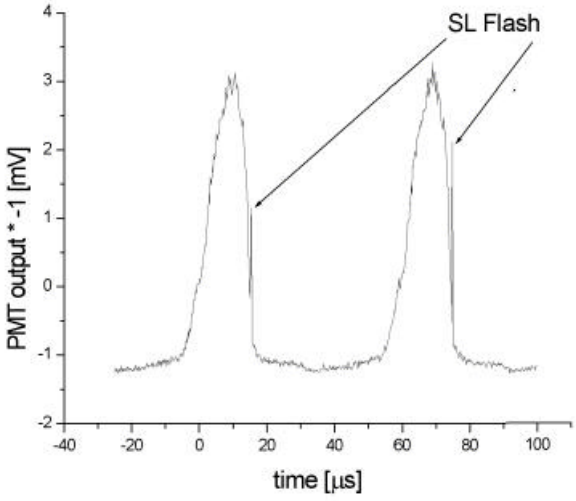

Fig. 1: Mie-scattering signal from stable SBSL in Sulphuric acid solution; frequency 16.8 kHz.

We prepared different aqueous solutions of sulphuric and phosphoric acid, with concentrations from 25% to 50% and 75% in volume. SBSL was obtained from all these solutions, but the best results, as far as stability and reproducibility are concerned, were obtained from 50% vol. solutions; we also made a few test experiments with pure (98% vol.) sulphuric and pure (84% vol.) phosphoric acid without positive results (it was not possible to produce any SBSL). The liquid samples were degassed under vacuum and then carefully released in the acoustic resonators; measurements were carried out while the gas content

of the samples was slowly increasing, due to diffusion processes, until the saturation level was reached; note the very low solubility of oxygen and nitrogen in sulphuric acid solutions.
We tried other two resonators: a 500 ml spherical flask with a resonance frequency of 16.8 kHz, and a 250 ml beaker driven at 35 kHz.

Observation

As shown in fig. 2, the intensity of light flashes measured during SBSL in sulphuric acid solutions is grater than that obtained in pure water: at high acoustic pressure levels, the luminescence is easily visible without obscuring the laboratory. Moreover, the behaviour of the bubbles is quite different from that seen in water. First of all, SBSL can be sustained in acid solutions at higher acoustic pressure level: bubbles are not destroyed when the 1.5 bar threshold is reached; secondly, new light emitting states were pointed out, like fast moving glowing bubbles (MSBSL) and luminescent jittering bubbles (fig. 3). Lowering the liquid temperature down to 9 °C does not cause an increase in light intensity (see fig. 2) and hysteresis is always present: to reach stable single bubble emission, it is necessary to generate a moving bubble at high ultrasound amplitude and then slowly decrease the acoustic pressure. Bubble stabilisation is not possible starting from a (light emitting) jittering state. If the acoustic pressure is further increased to about 2.0 bar, starting from MSBSL, a tridimensional lattice of intense light emitters appears; this "few bubble" state is quite spatially and temporally stable, but bubbles can move from site to site in the lattice over a time scale of few seconds; fig. 4 is a long-exposure photograph of the phenomenon. To our knowledge, the regular arrangement of SL bubbles can only be obtained in 50% vol. solutions of sulphuric acid.

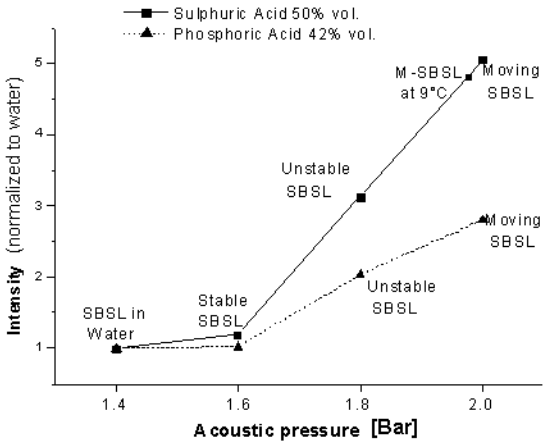

Fig. 2: Intensity of SBSL in acid solutions (related to SBSL in pure water).

We ascribe the lattice generation to a distortion of the spatial structure of the stationary acoustic field when a high amplitude forcing is applied to the resonator.
Finally, we observed sonoluminescence from a single bubble in a non-degassed solution. The bubble was spatially unstable, switching from a fast MSBSL state to an unsteady few bubbles lattice. However, the emission detected by the photomultiplier tube was synchronised with the acoustic field.

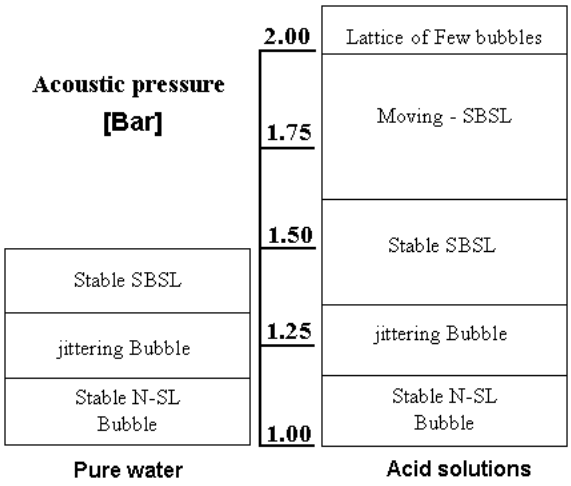

Fig. 3: Behaviour of the bubble increasing the acoustic pressure: pure water and 50% sulphuric acid solution are compared.

We then performed some test experiments in a 250 ml beaker, filled with a 50% vol. solution of sulphuric acid. If this kind of "resonator" is driven at about 35 kHz, a stationary field with multiple pressure antinodes is excited, producing a regular lattice of emitting bubbles.

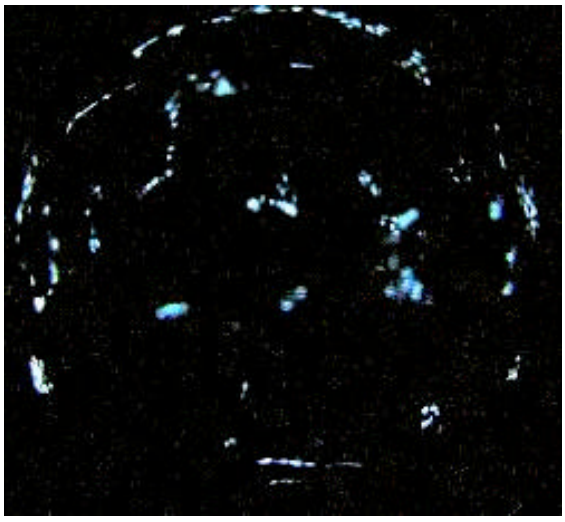

Fig. 4: The "lattice" of few bubbles acquired with a photocamera (after 2 min exposure).

As shown in fig. 5, the signal from the PMT is synchronised with the acoustic field. Since multiple bubbles are present and the various pressure antinodes can be out of phase, a double number of flashes per acoustic cycle is observed.

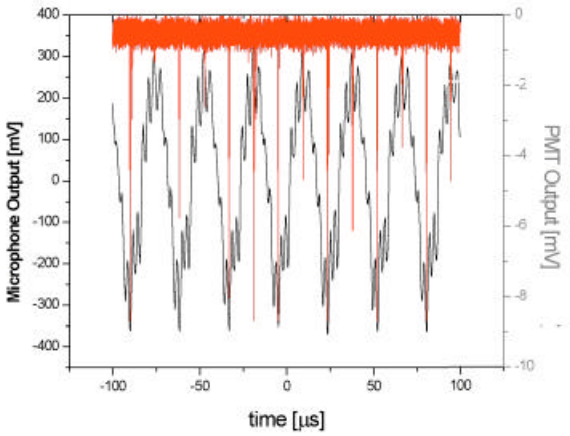

Fig. 5: The double SBSL flashes (red line) and the microphone acoustic signal (dark line)

Discussion

These new preliminary data on inorganic acid solution represent, from our point of view, an interesting link between "classical" SBSL experiments in pure water and recent findings related to complex organic compounds [1].

In a 1997 paper, B. P. Barber et al. [4] claimed that "water is the friendliest fluid for sonoluminescence"; from our experience, it is possible to conclude that concentrate sulphuric acid solutions are far more suited for luminescence production: the range of acoustic pressure sustaining light emission is wider and maximum light intensity greater than the pure water case; interesting dynamical state are observed, like moving-SBSL and jittering-SBSL; sono-luminescence is produced even if the gas content of the liquid sample reaches the saturation level.

The easiness with which SBSL is generated in phosphoric and sulphuric acid solutions seems to confirm some predictions of the model outlined in [5, 6] (the so-called hydrodynamic model of SBSL): since the vapour pressure of a 50% vol. solution of sulphuric acid at ambient temperature is about 8.66 hPa and this value decreases rapidly with the concentration, we can suppose that the drying action of the acid can reduce the presence of water vapour in the bubble [6], enhancing the heating and ionisation processes for the non-condensable fraction of the gas.

Even the phenomenon of light emission from non-degassed solutions can be cast in the "hydrodynamic" framework, since the solubility of oxygen and nitrogen is lower in acid solutions than in pure water: the acid solution undergoes a sort of "spontaneous" degassing.

Nevertheless, the hydrodynamic model, as interpreted in ref. [5], relies on the basic assumption of a perfectly spherical bubble collapse and this aspect of the modern theory of SBSL can hardly be reconciled with the observation of strong light emission during moving or jittering bubble dynamics. As pointed out by A. Prosperetti [8], a collapsing and translating cavity cannot retain a perfectly spherical symmetry and a liquid jet forms in the direction of the bubble translation velocity at the end of the collapse phase. Prosperetti describes sonoluminescence as an effect of high speed impacts of these liquid jets on the opposite bubble wall.

Our experiments confirm that the light emission is synchronised with the acoustic field during MSBSL and jittering SBSL, thus the bubble emits while it is moving in the resonator; these facts cannot be explained by a simple spherical collapse model. Of course, it is possible that moving and stable SBSL, even if apparently identical phenomena in their ultimate effect of flashing light emission, are actually generated by two different physical mechanisms; further evidences of a possible "cold and non-spherical" process [9, 10] underlying sono-luminescence would be found by a forthcoming spectral analysis of the electromagnetic radiation from moving bubbles in sulphuric acids solutions.

Conclusions

Performing bubble cavitation experiments in concentrate sulphuric and phosphoric acid solutions, unusually intense light emission was observed, but SBSL did not appear in pure sulphuric or phosphoric acid. The luminescence was produced by spatially stable or unstable (moving or jittering) bubbles at acoustic pressure higher than the amplitude characteristic in pure water, as recently reported for pure organic compounds like adiponitrile and formamide; we finally noted a less critical role of gas concentration in sonoluminescence excitation. The presence of acid molecules seems to radically change the nature of the fluid as far as cavitation effects are concerned; the light emission stability in the presence of remarkable deformations of bubble shape represents a very interesting challenge to existing theoretical models of SBSL.

References

[1] Y. T. Didenko, K. Suslick, "Molecular Emission during Single Bubble Sonoluminescence" Nature vol. 407, pp.877-879, 2000.

[2] T. Lepoint, F. Lepoint-Mullie, S. Labouret, J Frohly, C. Petrier, M. Comet, H. Fuzellier

"Multibubble sonoluminescence: a symbiosis between chemistry and physics" in Proceedings of ESS8 Confenrence, Villasimuis, Italy 14-19 September 2002, pp. 9a- 9c.

[3] T. Lepoint, private communication.

[4] B. P. Barber, R. A. Hiller, R. Lofstedt, S. J. Putterman, "Defining the unknown of sono-luminescence", Phys. Rep. 281, pp. 65-143, 1997.

[5] S. Hilgenfeldt, S. Grossamann, D. Lohse, "Sono-luminescence light emission", Phys. Fluids vol. 11, pp. 1318-1330, 1999.

[6] R.Toegel,B. Gompf, R. Pecha, D. Lhose "Does Water Vapor Prevent Upscaling Sono-luminescence?", Phy. Rev. Lett. vol. 85, pp. 3165-3168, 2000.

[7] G.E. Vazquez, S.J. Putterman "Temperature and Pressure Dependence Of Sonoluminescence" Phy. Rev Lett. vol. 85, pp. 3037-3040, 2000.

[8] A.Prosperetti, "A New Mechanism For Sono-luminescence" J. Acoust. Soc. Am. vol. 101, pp. 2003-2007, 1997.

[9] M.A. Margulis, I.M. Margulis "Contemporary review on nature of sonoluminescence and sono-chemical ractions", Ultrasonics Sonochemistry vol. 9, pp.1-10, 2002.

[10] A. Troia, D. Madonna Ripa, S. Lago, R. Spagnolo, "Evidence for liquid phase reactions during single bubble acoustic cavitation**",** Ultrasonics Sonochemistry, to be published.