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ScienceDirect

Ultrasonics 44 (2006) e357–e361

Ultrasonics

www.elsevier.com/locate/ultras

Influence of dissolved-air concentration on spatial distribution of bubbles for sonochemistry

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Available online 2 June 2006

Abstract

The pulsation of ultrasonic cavitation bubbles at various dissolved-air concentration in a sonochemical reaction field of standing-wave type is investigated experimentally by laser-light scattering. When a thin light sheet, finer than half the wavelength of sound, is introduced into the cavitation bubbles at an antinode of sound pressure, the scattered light intensity oscillates. The peak-to-trough light intensity is correlated with the number of bubbles that contribute to the sonochemical reaction. It is shown that as the dissolved air concentration becomes higher, the weighted center of the spatial distribution of the peak-to-trough intensity tends to shift towards the liquid surface. At higher concentration of the dissolved air, a great deal of bubbles with size distribution generated due to coalescence between bubbles disturbs sound propagation to change the sound phase easily. A standing wave to trap tiny oscillating bubbles is established only at the side which is nearer to the liquid surface. Also at higher concentration, liquid flow induced by drag motion of bubbles by the action of radiation force becomes apparent and position-unstable region of bubble is enlarged from the side of sound source towards the liquid surface. Therefore, the position of oscillating bubbles active for sonochemical reaction is limited at the side which is nearer to the liquid surface at higher concentration of the dissolved air.

Keywords: Ultrasound; Sonochemistry; Cavitation bubble; Light scattering; Dissolved air; Sonochemiluminescence

1. Introduction

Intense ultrasound in a liquid causes the generation of a lot of cavitation bubbles. The cavitation bubble repeats expansion and contraction according to the surrounding pressure oscillation of sound [1]. At sound pressure amplitude more than 1 atm the cavitation bubble collapses violently and produces hot spot of local extreme condition with high temperature of several thousand and high pressure of several hundred atmospheric pressure [2–5]. Under these conditions liquid water is easily dissociated to produce oxidants of OH radical, O, O_3, H_2O_2 etc. which are responsible for many kinds of chemical reactions [5,6]. Such chemical reactions with the oxidants from cavitation bubbles by ultrasound are referred to as sonochemical reactions [5]. Also, it is known that with the oxidants a bulk liquid region emits sonochemiluminescence if luminol is added to an alkaline solution, and the luminescence has been studied as a good measure to evaluate yield in a sonochemical reaction [7,8]. Sonochemical reaction is a promising tool for novel method of advanced material technology [9] and environmental processing such as waste water treatment [10].

In order to accomplish high efficiency in sonochemical reaction, it is important to clarify the mechanism of sonochemical reaction based on the bubble dynamics as the ultrasonic cavitation bubble is the origin of sonochemical reaction. Recently, by introducing a laser-light sheet into the ultrasonic cavitation bubbles at various positions in the standing wave field to evaluate the scattered light intensity, we have clarified that a spatially periodic configuration of oscillating cavitation bubbles stably exist in the region near the water–air interface rather than in the region near the sound source [11,12]. We also showed that spatial

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0041-624X/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.ultras.2006.05.002

summation of the intensity of scattered light from oscillating bubbles correlates well with the yield of sonochemical reaction [13]. However, there is no experimental report for correlation in spatial distribution between volumetric bubble oscillation and sonochemiluminescence at different concentration of dissolved air. The dissolved gas concentration is an important parameter related closely with the probability of generation of cavitation bubbles [14].

The present report deals with, for the first time, an investigation on the influence of dissolved-air concentration on spatial distribution of bubbles for sonochemistry through the measurements of light scattering by bubbles and intensity of sonochemiluminescence. In order to evaluate the oscillation characteristics of the bubbles at different position in a glass cell, the measurements of the peak-to-trough intensity for the scattered light waveform by bubbles in the ultrasonic standing-wave field is performed by changing the measuring position in the direction of sound propagation at different concentration of dissolved air. Also, the image of sonochemiluminescence with luminol is captured and the spatial distribution is studied in comparison with the results by light scattering.

2. Experimental

The experimental procedure followed is as shown below. A cw sinusoidal signal of 142 kHz generated by a function generator (NF Electronic Instruments, 1942) was amplified with a 55-dB power amplifier (ENI, 1140LA). The electric output was fed to a plane-type piezo-electric circular transducer (Honda Electronics) of 50 mm in diameter and 5.5 mm in thickness. The transducer was fixed to the center of a stainless steel circular plate of 100 mm in diameter and 2 mm in thickness. A rectangular glass cell of $50 \times 50 \times 150$ mm^3 internal dimensions was set above the transducer through the stainless steel plate, where the small gap of 0.1 mm between the glass cell and the steel plate was filled with water to transport the sound into the cell. The thickness of the cell bottom was 3 mm. The volume of distilled water in the cell was 250 cm^3 and the temperature was 20 °C. Distance from water–air interface to the cell bottom was 100 mm. The air concentration of the distilled water used in the present experiment was adjusted by bubbling air. The concentration of dissolved oxygen (DO) in the distilled water was measured with a DO meter (HORIBA, D-25). Diode laser beam (NEOARK, 50 mW, 684 nm, 1 mm in beam diameter) modified with lens to a sheet light of 0.5 mm in thickness was introduced into cavitation bubbles generated in the cell, where the illuminated area was 25×50 mm^2 of tetragonal and half of plane perpendicular to the sound beam axis in which one side was set closely to the sound beam axis. The intensity of scattered light from bubbles was measured with a photomultiplier tube (Hamamatsu Photonics H7732-10) through a converging lens and a pinhole of 0.5 mm in diameter and changing the position by every 0.87 mm in the vertical direction along the sound propagation using the 3 axes stage (SIGMA KOUKI, LTS-50) was controlled by a computer (NEC, 9801RA). The waveform of scattered light, which was composed of superposition of scattered light intensity according to the scattering cross section of each bubble repeating expansion and contraction due to the acoustic cycle, was recorded with a digital oscilloscope (YOKOGAWA, DL1540C) and controlled by the above computer. According to the relationship between the scattering cross section and the scattered light intensity at different size of cavitation bubbles, small bubble is relatively effective for contributing to the scattered light intensity [7,15]. Also, the size distribution of cavitation bubble effective for sonochemical reactions is restricted due to its shape instability [16,17]. Then, the peak-to-trough intensity corresponds with the number of oscillating bubbles. This will be described with a conceptual sketch in the next section for better understanding. The peak-to-trough intensity for the waveform was measured at different position. Each data obtained was the one averaged over 32 times. In the present experimental condition, ultrasonic power input to the cell was determined calorimetrically and the value was 17 W.

In order to compare the results by light scattering with spatial distribution of sonochemical reaction, images of sonochemiluminescence with luminol were also captured under almost the same conditions as those in the light scattering experiment. The aqueous solution for luminol experiment was prepared by mixing distilled water of 245 cm^3 with 250 mM sodium carbonate-2.5 mM luminol solution of 5 cm^3. The images of sonochemiluminescence with luminol were captured with a digital camera (Nikon, D70). Time of each exposure was 90 s.

3. Results and discussion

3.1. Relationship between peak-to-trough intensity of scattered light and the number of volumetric oscillating bubbles

Fig. 1 shows a conceptual sketch of peak-to-trough intensity of scattered light by bubbles corresponding to the number of bubbles. This is the case of four bubbles

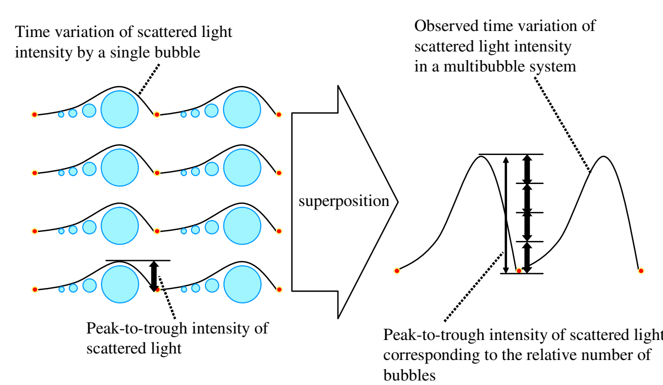

Fig. 1. Concept of peak-to-trough intensity of scattered light by bubbles corresponding to the number of bubbles.