MRI MEASUREMENT OF DYNAMICS OF THE LIQUID AND A DISSOLVED GAS DURING ACOUSTIC CAVITATION

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1. INTRODUCTION

The defining feature of acoustic cavitation is the presence of the gaseous bubbles produced by the oscillating drop of acoustic pressure in the liquid. It is their evolution and collapse that provide the cavitation with the chemical activity, mechanical erosion, degassing, etc. It is, therefore, natural, that the methods most commonly applied to studies of cavitation are optical and acoustical, i.e. the bubbles are looked at and listened to [1].

The complexity of the cavitation cloud as a collection of many thousands of interacting bubbles makes a thorough theoretical analysis almost impossible; besides, in such practical applications of cavitation as sonochemistry and material processing, there is often a need for a conversion of the visual and acoustical information into parameters relevant to those fields such as gas exchange between the bubbles and the liquid and fluid dynamics of the liquid and the gases during cavitation.

In this work, instead of measuring the bubble dynamics, we attempt to obtain information on dynamics of molecules that constitute the bubbles and the surrounding liquid. Such information is directly acquired via the use of the Magnetic Resonance Imaging (MRI) techniques of measuring the displacement and the relaxation parameters of water and an NMR-sensitive highly soluble gas as a function of the acoustic pressure and the water preparation procedures (filtered, non-filtered, with the addition of surfactants). The obtained data are then examined using a two-site exchange model to extract estimates of the average residence time of gas molecules inside the cavitating bubbles (averaged over the volume).

2. METHOD

An MRI scanner with 2.4 T superconducting magnet (Nalorac, CA), 20 cm i.d. bore, was employed for all our measurements. An ultrasonic transducer with the attached water-filled cuvette, satisfying 2 half-wave length standing wave conditions, was placed in an RF probe of the scanner (at resonant frequencies of 1H for measurements of water and 19F for measurements of the dissolved gas (Freon)). A Langevin-type transducer (SensorTech, ON) was employed for the experiments with the acoustic pressure of up to 1.6 Atm (19.7 kHz), and a magnetostrictive transducer with a cylindrical concentrator (Industrial Sonomechanics, Inc) was used in experiments with the acoustic pressure of up to 6 Atm (19.1 kHz).

We worked with two types of water samples: a distilled water, tap water and the water with 1 mM SDS for the bubble stabilization. The water samples were further filtered to reduce the number of impurities that could server as the nucleation centres. The filtration was done with the syringe 0.2 mkm filter (Sarstedt, Germany).

As an NMR-sensitive gas, Freon-22 was dissolved in the water samples prior to all experiments by bubbling at the ambient temperature and pressure during 20 min, with the resulting dissolution of 0.6 vol/vol (as measured by the NMR signal comparison with the signal from the gas-filled cuvette). Its T2 NMR relaxation times differ by three orders of magnitude for the gas in the free (2 ms) and dissolved (1.5 s) state [2] and hence can be used to differentiate between the two states of the gas molecules.

A PFG SE sequence [3] was employed to obtain propagator (displacement probability) maps both for the water and the dissolved gas. The observation window in the PFG experiments was 88 ms so that only the signal from the dissolved molecules of Freon could be detected. The T2 relaxation was measured with the CPMG [4] sequence for the bulk and with the SE SPI CMPG sequence for the 1-dimensional profile of the cuvette along the acoustic axis.

3. RESULTS

The cavitation onset was detected at 0.6 Atm in the tap water, and at 0.7 Atm in the filtered water by the appearance of the high frequency noise detected by the hydrophone.

MRI Measurements were first performed at the lower acoustic pressures (1.6 Atm) for the samples with the filtered and the unfiltered water. The propagator maps showed a similar dynamics for the motion of the water molecules in the two samples, with the developed acoustic streaming along the acoustic axis.

The distribution of the velocities of the Freon molecules was, however, different in the two samples, with the greater localization of the dissolved gas in the unfiltered water and the greater number of the "stationary" molecules (molecules with the velocities within the resolution limit of the measurement). The displacement of Freon molecules in the filtered water was greater than that of the water molecules, with the almost complete absence of the signal from the "stationary molecules" [5]. The only explanation of the discrepancy in the motions of water and gas molecules dissolved in water was the participation of the gas molecules

in the cavitation process, *i.e.* the diffusion of the gas from water into the cavitating bubbles with which the gas molecules would be able to travel faster than the currents of the surrounding water.

The active participation of the gas molecules in the cavitation meant that a certain fraction of the measurement time they had to spend in the gaseous state inside the bubbles. In that case, the measurement of the T2 of Freon during cavitation should be able to provide us with information on the average duration of the gaseous state, thanks to the profound difference in the T2 between the two states. The changes in T2 during cavitation were reliable yet minor, with a monoexponential decay, indicating that the exchange between the two states was faster than the timescale of the measurement (spin echo time of 0.5 ms). An application of the two-site exchange model resulted in the estimates of very short residence times of molecules of Freon in the gaseous state, on the order of two oscillation periods (~0.1 ms).

At higher acoustic pressures achieved with the other transducer, the dynamics of the water and gas molecules changed again. The motion of the water became more pronounced whereas the motion of the dissolved gas was found to be more and more localized, both at the tip of the concentrator and its sides. At the highest pressures (5.6 Atm), both the gas and water molecules became localized at the tip of the horn, with the active motion of water long the sides.

Measurements of T2 of Freon at the tip of the concentrator showed T2 decrease along the acoustic axis, in the region of the most active cavitation. The application of two-site exchange model to the T2 data yielded the average residence times of Freon molecules of several oscillation periods (100-150 mks).

The addition of SDS to water for the bubble stabilization considerably modified the dynamics of both water and gas molecules. The T2 times of Freon at high acoustic pressures became bi-exponential, indicating two sites with well-defined, quite different molecular environments, which we attribute to the stabilized gaseous bubbles and the surrounding water.

4. DISCUSSION

The observed uncoupling of the water and the dissolved gas dynamics was to be expected, since it is essentially the definition of the cavitation. Still, it was surprising to observe the extent of the involvement of the dissolved gas into the cavitation, with the exchange rate of the total gas content of the vessel on the order of less than a second between the two states.

The observed spatial localization of the gas and water at the elevated pressures can be explained by the increased Bjerknes force action of the acoustic field.

The very short residence times of the gas molecules were unexpected. It is relevant to note that these times are a total time of the gas molecule with all possible translations of the molecule into and out of the bubble over the course of the measurement (~ 1s). With the commonly accepted model of the boundary layer, one would think that the molecules in the proximity of the bubbles will necessarily have a longer exposure to the bubble content. However, the measurements are averaged by volume, and that can mask the boundary layer effect. Also, Freon is a highly soluble gas, so it should be no surprise that as soon as its molecule gets ejected out of the bubble during the collapse phase, it stays in the dissolved state.

The two-site model is quite simplistic, and there is a potential for improving it by adding the boundary layer and the radial diffusive exchange [6]. Another direction of research is the use of other gases with lower solubility: that will be a chance to probe the gas exchange between the bubbles and liquid at higher exchange rate if the experimental problems with the lower signal-to-noise due to the decreased gas concentration will be overcome.

In conclusion, we believe that the presented work demonstrates a potential for the molecule-based approach and MRI measurements in the cavitation research.

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