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BUBBLE DYNAMICS AND TRITIUM EMISSION DURING BUBBLE FUSION EXPERIMENTS

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ABSTRACT

Neutron nucleated, transient bubble cluster dynamics has been studied through direct observations of shock wave and sonoluminescence (SL) signals. Confirmatory bubble fusion-related neutron-seeded acoustic cavitation experiments were conducted with deuterated acetone (C_3D_6O) and non-deuterated acetone (C_3H_6O). Tritium emission monitoring was performed systematically by using a calibrated state-of-the-art Beckman LS6500 beta spectrometer for the samples obtained from bubble fusion experiments of non-deuterated and deuterated acetone with and without cavitation. Statistically significant tritium emission was observed during neutron-seeded acoustic cavitation experiments with deuterated acetone, but not for control experiments involving non-deuterated acetone, nor with irradiation alone, thereby confirming reported observations for the occurrence of thermonuclear fusion reactions in deuterium-bearing imploding cavitation bubbles. Thermal hydraulic conditions of bubble implosions leading to robust SL emission are discussed.

KEYWORDS

Bubble fusion, bubble cluster dynamics, tritium counting.

1. INTRODUCTION

Thermonuclear fusion reactions in imploding bubbles (so called bubble fusion) were observed and reported by Taleyarkhan and his coworkers (Taleyarkhan et al., 2002,2004a; Nigmatulin et al., 2004) but so far have not been confirmed by others. Thermonuclear fusion in highly compressed bubbles is possible only when appropriate conditions are provided: high enough (~1000 Mbar) pressure and (~ 10^7K) temperature and the presence of deuterium (D) atoms which need to be forced close enough, and need to stay together for a sufficient time to permit them to become fused (Gross, 1984). Theoretically, these conditions have been predicted to occur (Moss, 1996; Nigmatulin et al., 2004; Wu, 1993; Taleyarkhan et al., 2004b) and highly depend on bubble dynamics: how these bubbles initiate, grow and implode. Furthermore, recent experiments (Camara et al; 2004) to ascertain temperatures below the surface of SL bubbles have revealed clearly that the emission spectra from the interior resemble those given out by Bremstrahhlung radiation composed of excited plasmas in the 10^6K range. Another study to directly and convincingly demonstrate the existence of plasmas in SL bubbles has recently been published (Flanigan and Suslick, 2005). Based upon these recent

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developments, it is now widely accepted that imploding bubbles can indeed produce extreme states of compression and temperatures.

As is evident, implosions of spherical bubbles produce stronger shock wave compression than aspherical ones; the maximum bubble volume is not only a function of the acoustic pressure amplitude, but can also be affected by the timing of the bubble nucleation (Taleyarkhan, 2004b). Therefore, a comprehensive understanding of bubble dynamics as well as related control variables will be crucial for successful bubble fusion experiments and for future development and optimization of bubble fusion technology.

The process of bubble nucleation, growth and collapse is nonlinear and complicated in general, involving thermal, mechanical, optical, chemical or even nuclear scale phenomena. Depending on the acoustic driving amplitude, a bubble could grow in volume in several acoustic cycles and collapse within one cycle. Huge potential energy accumulated during its growth time can be converted into thermal energy to heat up the bubble's internal contents by shock wave compression. The temperature inside the bubble could be more than 100 million degrees (Nigmatulin et al., 2004) and high enough to accelerate chemical reactions and even cause nuclear fusion reactions. This shock wave continues to propagate in the liquid after the bubble collapses and the evidence can be detected on the chamber walls by an ordinary microphone.

The issue of bubble nuclear fusion thermal-hydraulics becomes even more complicated when a nucleated single bubble grows from ~50 nm by factors of ~100,000 to a large (1000 μm) bubble then implodes and breaks into a cluster of tiny bubbles (Brennan, 1995). These tiny bubbles can stay together as clusters when an acoustic standing wave is applied. From experimental and numerical analyses (Taleyarkhan et al., 2004b) bubble cluster formation can lead to pressure intensification for inner bubbles, causing much higher temperatures and pressures for the bubbles in the center of the cluster than for a single individual bubble. This is attributed to acoustic streaming effects of the shock wave produced by the bubbles along the edge of the cluster (Matsumoto, 2004). Evidently, the assessment of the relative effects of bubble cluster appears crucial for understanding conditions relevant for attaining bubble nuclear fusion, and scale-up of bubble fusion dynamics. This was therefore, attempted for which salient results are presented in this paper.

An important consideration in such experiments to evaluate the occurrence of nuclear fusion involves experimental evidence of key signatures. Notably, for bubble fusion experiments (Taleyarkhan et al., 2002, 2004a) the bubble collapse time is so short and the final bubble size during implosion is so small that any attempts of measuring the variables inside a bubble are extremely difficult, if not impossible. Therefore, indirect approaches must be used to identify the possible nuclear fusion reactions in a collapsing bubble. The well-known D-D nuclear fusion reaction proceeds in two branches of roughly equal probability as (Gross, 1984)

$$D + D = \begin{cases} n + {}^{3}He \\ p + T \end{cases} \tag{1}$$

The products of D-D fusion reaction are: a neutron (*n*), a proton (*p*), Helium (*He*) and tritium (*T*). Helium (^{3}He) is a non-radioactive gas and it is difficult to detect and the MeV energy protons (due to them being charged particles) can travel no more than ~1 mm through the test fluid and before getting absorbed. On the other hand neutrons (being uncharged particles) can escape from the test cell, and tritium is a radioactive isotope readily detectable using beta-spectrometry. Therefore, neutrons and tritium become the candidates for fusion reaction detection in bubble fusion experiments as reported by Taleyarkhan et al. (2002, 2004a). However, in bubble fusion experiments, it is to be realized that neutron detection can become difficult due to the presence of large gamma ray fields resulting from the neutrons used to seed bubbles. This requires sensitive on-line detection equipment which can distinguish neutrons from gamma rays, and also distinguish neutrons from nuclear fusion from those neutrons used for seeding bubbles from an external neutron source (PNG or isotopic source). Such issues and complexities are non-existent when monitoring for the radioactive isotope tritium.

This paper focuses on reporting investigations on two aspects of bubble nuclear fusion: transient bubble dynamics along with SL light emission, and tritium production. These two topics are presented separately. The first part of this manuscript discusses observations of bubble thermal-hydraulics during the simulated bubble fusion experiments. These observations were obtained in a desktop test apparatus with isotope neutron-seeding of cavitation nuclei in a test cell. The second part provides confirmatory evidence of tritium emission during neutron seeded acoustic cavitation of deuterated acetone, along with evidence of null results from control experiments.

2. EXPERIMENTAL APPARATUS AND APPROACH

The bubble dynamics experiments were performed in a test apparatus (see Figure 1) similar to what was used by Taleyarkhan et al. (2002, 2004a). The test chamber was placed in a chilled light-tight enclosure. A microphone (MIC) was attached to the outside wall of the chamber for shock wave detection (indicative of bubble implosions) for which the low frequency components were filtered out for counting of cavitation rate. A photomultiplier tube (PMT) was placed ~1 cm away from the test chamber for sonoluminescence (SL) light detection. The PMT was powered by a high voltage supply at -2000 volts and its output was first sent to a preamplifier (ORTEC 113) and then to an amplifier (ORTEC 570). The fluid (normal acetone) was driven and experienced positive and negative pressures at a frequency of ~20 kHz by the acoustic wave generated from a PZT ring epoxied on the chamber. An isotope neutron source (Cf-252 0.5 mCi) was used to seed nuclei in the fluid. A high speed video camera (Fastcam 10K) was used to visualize the bubble behavior.

Following the methods reported elsewhere (Taleyarkhan et al., 2002) before conducting bubble fusion experiments the test cell drive amplitude corresponding to about -7bar for nucleation from multi-MeV neutrons was evaluated after degassing. That is, no bubble nucleation would occur at this acoustic drive power over a waiting time of ~ 30s in the absence of the neutron source. Thereafter, after the baseline drive amplitude was doubled to be ~ +/- 15 bars for each of the cavitation runs (as used by Taleyarkhan et al., 2002, 2004a).

It is well-known that tritium is an extremely rare isotope and can only be produced by via nuclear reactions and hence, becomes a powerful indicator for possible thermonuclear fusion reactions during bubble fusion experiments. Tritium can be examined for its presence in the test fluid after the experiment, but this requires access to expensive and sensitive beta spectrometers. Fortunately, as part of the infrastructure we had access to a state-of-the-art beta spectrometer system, the Beckman LS6500™ system at Purdue University, which was similar to that used in the reported bubble fusion studies at Oak Ridge National Laboratory (Taleyarkhan et al., 2002, 2004a). Therefore, we focused on monitoring for tritium emission during acoustic cavitation experiments to confirm the possible occurrence of bubble nuclear fusion. Along with D-D nuclear fusion producing tritium, it is well-known that D atoms in a deuterated liquid can become transmuted to T atoms in the presence of a very high flux of neutrons (as in a commercial power nuclear reactor). Fortunately, in bubble nuclear fusion experiments transmuting D atoms to T atoms by neutron bombardment is a second order effect, a fact which can be readily validated via conduct of control experiments (i.e., experiments conducted to note changes in tritium content of the test liquid by subjecting the test cell to the same experimental neutron fluence used for seeding bubbles, but without acoustic power turned on such that cavitation is not present). Control experiments were also to be performed under identical experimental conditions, but changing only one parameter at a time (e.g., cavitation on vs. off; alternately, change H bearing liquid to D bearing liquid). The control experiments include non-deuterated fluid tests along with cavitation on or off tests. Evidence for thermonuclear fusion reactions (from tritium emission) in a collapsed bubble needs to manifest only for neutron-seeded cavitation in a deuterated fluid. All tests with a non-deuterated fluid or a test with deuterated fluid without cavitation should not lead to tritium production.

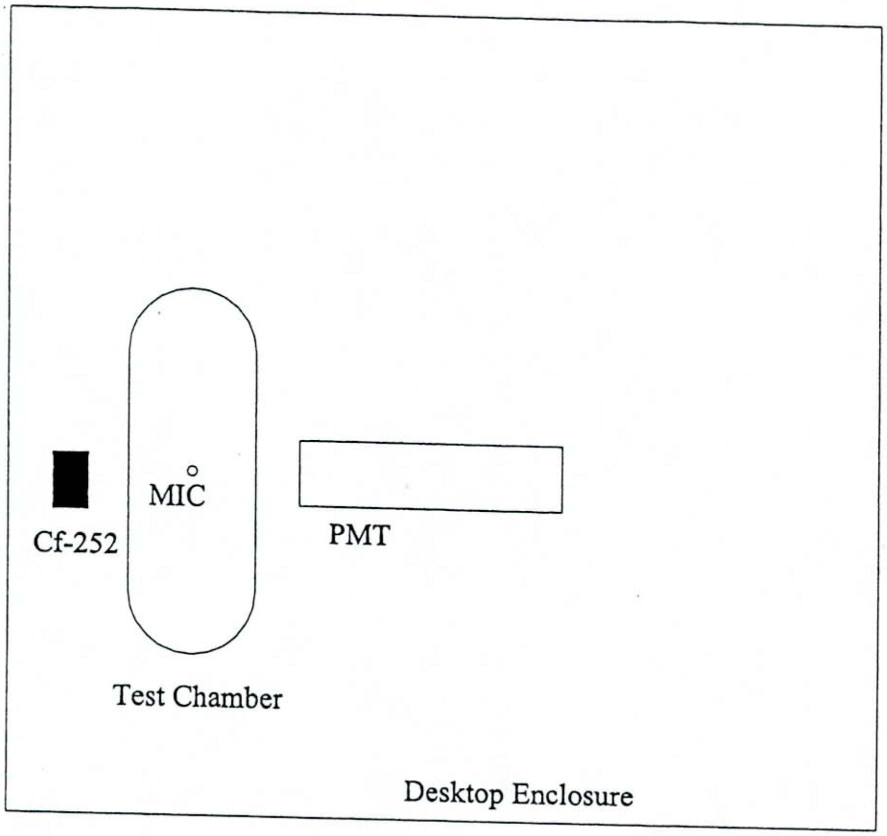

Figure 1: Schematic of experimental apparatus layout (not scaled). Cf-252 – Isotope Neutron Source (0.5 mCi); MIC – Microphone; PMT – Photomultiplier Tube.

3. RESULTS OF BUBBLE DYNAMICS

Following the published approach by Taleyarkhan (Taleyarkhan et al., 2002 and 2004a), the fluid was first properly degassed for about 2 hrs until individual cavitation bubble clusters were achieved. During such evolution, sharp (N-shaped) shock traces were observed on the high-speed digital storage oscilloscope screen coming from the microphone and the PMT. The bubble dynamic behavior has been studied as follows: cavitation visualization by using a high speed video camera (Fastcam 10K), shock wave detection by using a microphone attached on the outside wall of the test chamber and sonoluminescence light emission by using a photomultiplier tube. Typical results are illustrated in the following subsections.

3.1 Cavitation Visualization

Figure 2 displays a typical image sequence of a cavitation bubble cluster of non-deuterated acetone nucleation seeded by neutrons from a Cf-252 isotope source (0.5 mCi of activity) and experienced pressures at ~+/- 17 bars driven by acoustic waves. Note that the images were taken at a speed of 5000 frames per second and 1/20000 s for shutter speed. Since the camera frame speed is smaller than that of the chamber driving frequency, it is believed that the bubble is actually a bubble cluster, which can be verified by quickly turning off the acoustic driving power. The bubble cluster which was otherwise held in place by the acoustic pressure field breaks apart and results in a dispersion of several tiny (~10^2 μm) bubbles. Also, direct numerical simulations for bubble growth using the well established Rayleigh-Plesset formulation indicates that an individual bubble that can reach a maximum of only ~400 μm (Nigmatulin et al., 2002), whereas the size of individual clusters is about 10 times

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larger. The images were compensated for the distortion due to the optical deflection from a cylindrical surface and its scale is about 0.083 mm/pixel. The bubble cluster diameters in the first three images at t=0.0, 0.2 and 0.6 ms are about 0.6 , 2.7 and 3.4 mm, respectively. The first appearance of contraction (perhaps because some of the bubbles in the cluster were imploded in this frame) is seen at t=0.8 ms. The cluster size did not vary much after the first contraction and was diffused out after 3 ms.

Figure3 shows another type of cavitation consisting of comet-like streamers. Unlike that of individual bubble clusters, the structure of a streamer appears continuous in space and time: bubbles were formed at one end (bottom end in this figure) and ejected outwards from the other end and could last as long as 10 s. Interestingly, and importantly, it was observed that streamers produce neither distinct shock wave peaks in the microphone nor SL light emission. This is described in the next section.

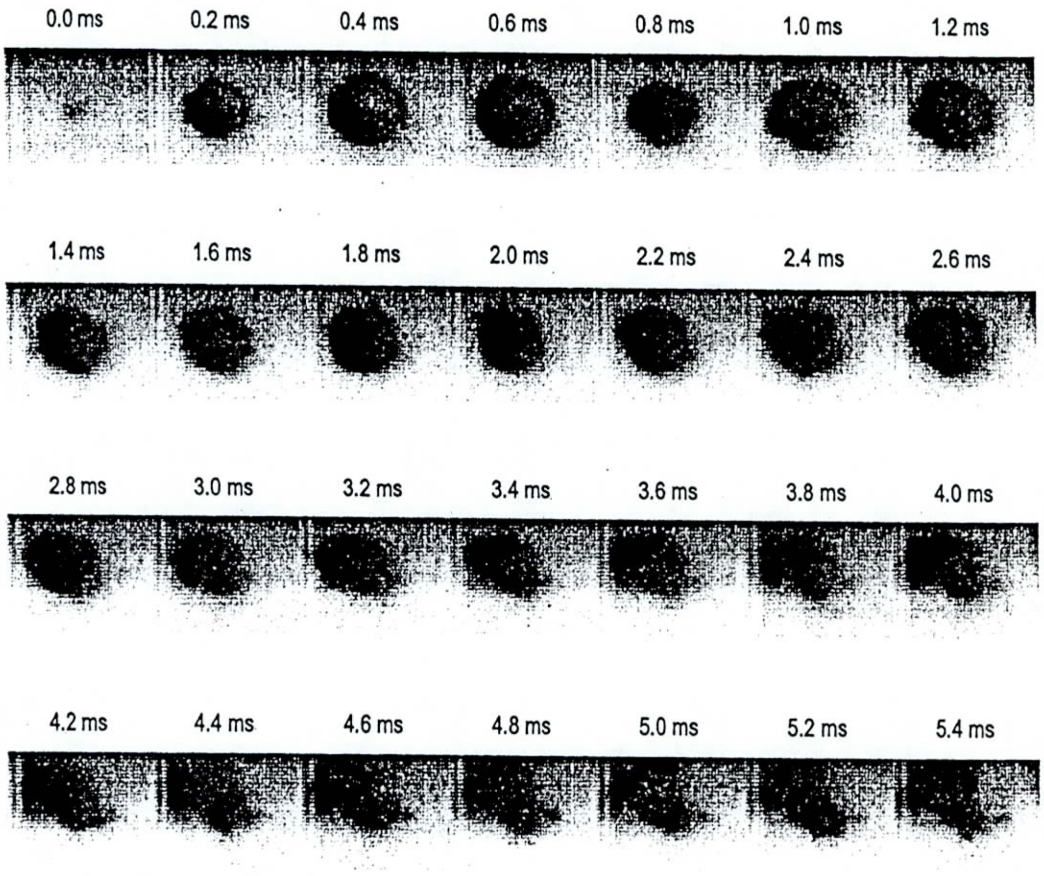


Figure 2: Individual bubble cluster (C_3H_6O, 4 °C, ~+/- 17 bars, 16.7 kPa)

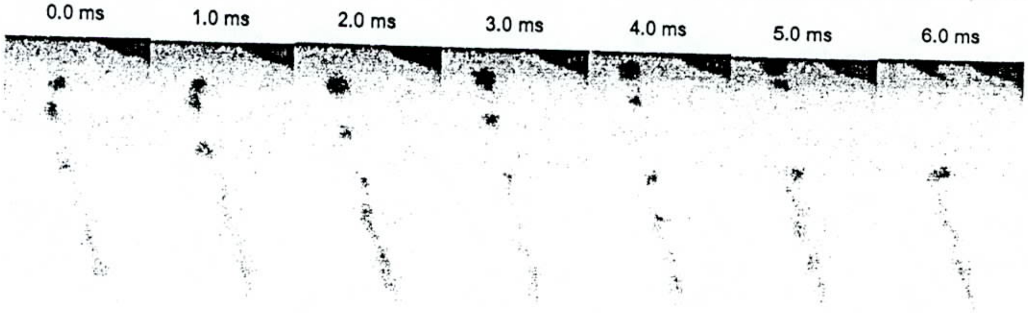


Figure 3: Comet-like streamers (C_3H_6O, 4 °C, ~+/- 17 bars, 16.0 kPa)

3.2 Signals from Microphone and PMT

Shock waves and light emissions from the imploding bubbles were detected by the attached microphone and the PMT respectively. Their signals were displayed and stored by a 100-MHz Agilent™ digital storage oscilloscope. Figure 4 depicts the typical results of these two signals under conditions involving individual clusters. Due to the propagation time required for the sound wave from the location of bubble collapse to the location of the attached microphone on the glass surface, there is a time delay between the microphone signal and the SL signal which is found to be about 30 μs for this chamber. This value corresponds nicely to the time required for a sound wave to travel from the center of the chamber to the walls of the chamber where the microphone is attached. On the other hand, Figure 5 indicates that the corresponding signals are much smaller and random for streamers.

The peak-to-peak amplitudes of the microphone signals were recorded under different driving amplitudes to the PZT ring. The results were depicted in Figure 6. These values indicate the intensities of shock waves generated by the bubble collapse. It can be seen that the shock wave intensity increases with the low acoustic driving amplitudes (implying enhanced levels of implosion) and becomes saturated with increasingly higher drive amplitude. This observation implies that the most intense implosion during cavitation does not necessarily correspond to the highest acoustic driving amplitude.

It was also observed that not every shock wave corresponds to a recorded light pulse. This was found to be especially true for conditions leading to the formation of streamers (which as mentioned earlier look like comets, and consist of thousands of tiny bubbles unlike bubbles in spherical clusters). It was distinctly noted that the presence of streamers did not produce detectable light emission at all, clearly indicating that the intensity of collapse is quite different and much lower (i.e., contents of imploding bubbles were not even hot enough to emit SL light flashes) than that from individual bubble clusters.

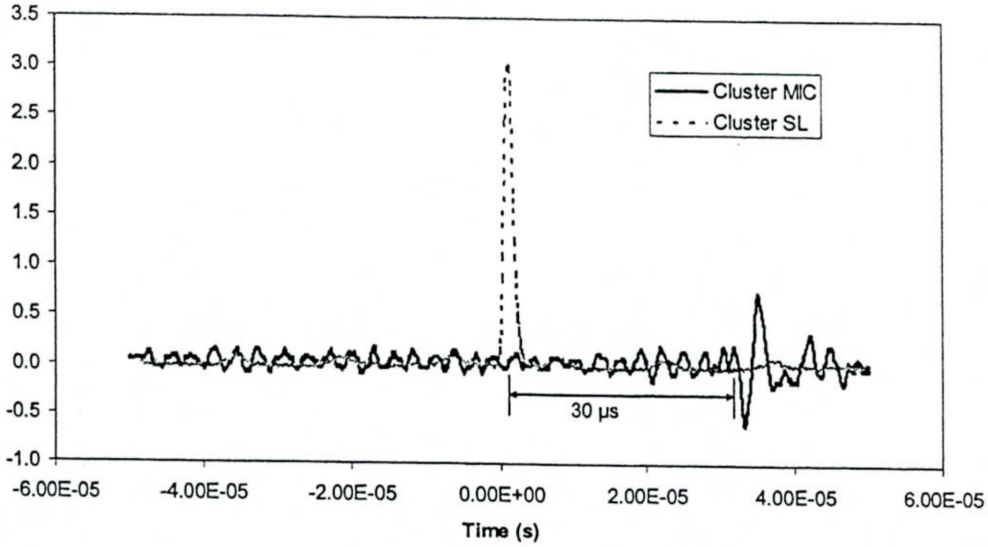


Figure 4: Signals from microphone and PMT of individual cluster

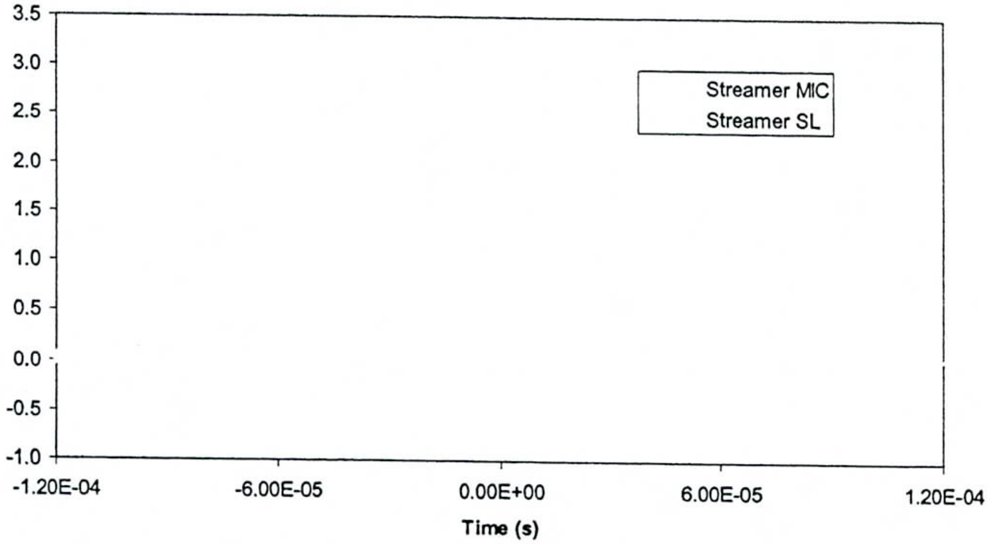


Figure 5: Signals from microphone and PMT of streamers

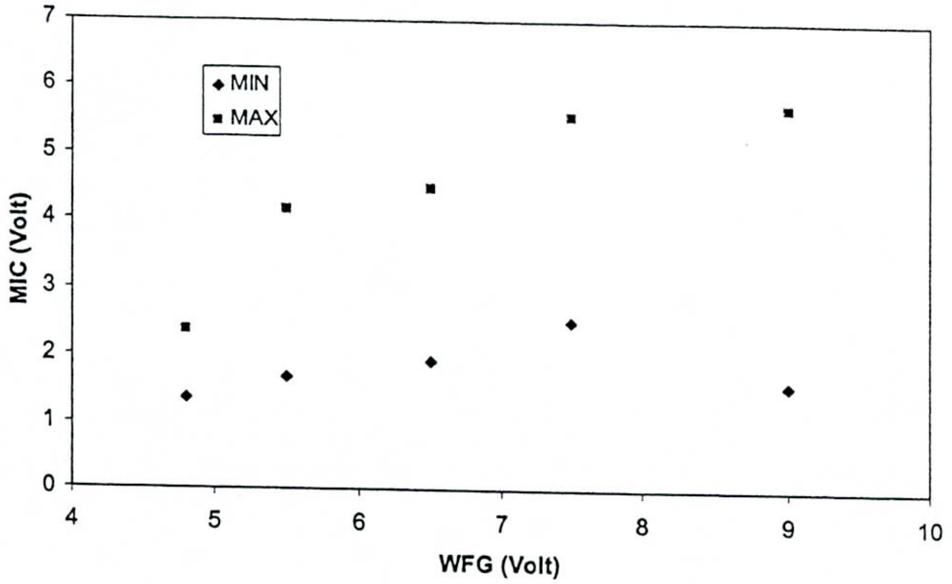


Figure 6: Amplitudes of microphone signals

4. RESULTS OF TRITIUM EMISSION

Similar to the protocol followed for reported bubble fusion experiments (Taleyarkhan et al, 2002, 2004) tests were systematically conducted with deuterated and non-deuterated acetone over six hours duration (to accumulate significant quantities of tritium in the test fluid). The test chamber was positioned in a closed freezer with temperature control, and bubble nucleation was seeded by using a Plutonium-Beryllium (Pu-Be) isotope source (of 1 Ci activity). For each test run lasting for 6h, two samples were systematically prepared by extracting 1 ml of test fluid from the same test chamber before and after each cavitation run and mixing with 15 ml of Ultima Gold™ scintillation cocktail in a 20-ml scintillation vial; therefore, four samples were available for each test run. These samples were analyzed in a scintillation counter for excess tritium emission. The Beckman LS6500™ counter, a sophisticated state-of-the-art system similar to what was used by Taleyarkhan (Taleyarkhan, et al., 2002) was used for these studies. The counter was calibrated with NIST-certified quenched standards and the mass quench effect of acetone was investigated. Each sample was counted over 10 cycles and for 10 minutes during each cycle; therefore, each sample was counted for a total of 100 minutes. There was no interruption for each counting scheme and a sample with 15 ml Ultima Gold™ cocktail alone was also counted simultaneously for validating and ensuring machine stability and for ensuring absence of any unusual background variations.

4.1 Calibration of the Beckman Counter

The Beckman scintillation counter (LS6500) does not directly provide the true measure of radioactive decay in the form of DPM (disintegration per minute). Instead, it conducts a calibration for quenching for each sample (during each cycle) and offers a so-called quench number "H#" along with the raw data for count-rate per minute, i.e., CPM (count per minute) values for each batch. This essentially requires the user to conduct a calibration using known standards (certified by NIST) to obtain the conversion factor from CPM to DPM.

The counter was calibrated with NIST-certified quenched tritium standard vials (procured from PerkinElmer™, 2003). The calibration data were systematically obtained in the same routine as that used for sample counting. The results are shown in Figure 7, where the H# was printed out from the counter accounting for the quenching effect and the efficiencies were calculated from the ratio of the machine CPM and the actual DPM derived from the standards (accounting for radioactive decay).

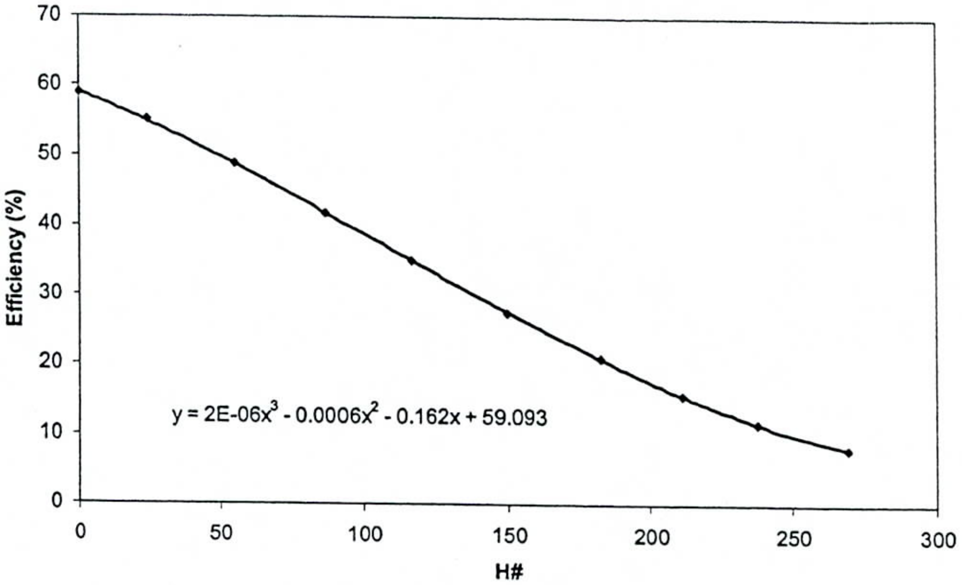


Figure 7: Beckman LS6500 scintillation counter calibration curve. The dots are calibration data points and the solid line is a curve fitting with a third-order polynomial as shown in the figure, which was used to convert the CPM into DPM in tritium counting

4.2 Tritium Counting

Several six-hour duration tests were conducted to confirm if statistically significant quantities of tritium are generated only when conducting neutron-seeded cavitation in C_3D_6O. For these experiments a 1 Ci Pu-Be neutron source (emitting about $2x10^6$ n/s) was available and therefore, utilized. The test cell (maintained at ~ 0°C temperature) was placed in a closed freezer, which was furthermore, surrounded with paraffin blocks for radiological safety. A schematic of the experimental arrangement is shown in Figure 8 along with the relative position of the Pu-Be neutron source. Tests were conducted with neutron irradiation alone, followed by tests with neutron seeded cavitation – systematically changing only one parameter at a time. Neutron-seeded acoustic cavitation was conducted for ~6 h duration. Liquid samples were taken before and after cavitation from the liquid poured into the test chamber. For each case 1 ml of acetone was pippetted and mixed with 15ml of Ultima Gold^{TM} scintillation cocktail in a borosilicate glass vial. These vials were counted for 100 minute for each sample for tritium beta decay activity (5 to 19 keV energy emission window) in a Beckman LS6500^{TM} liquid scintillation counter. Results of tritium activity changes are displayed in Figure 9. It is seen that a statistically significant increase (~ 4 to 6 SD) of tritium is only observed for tests with neutron-seeded cavitation of C_3D_6O. Null results are obtained for all other control experiments. For neutron-seeded cavitation tests with the control liquid C_3H_6O, as well as for tests with neutron irradiation only (without cavitation) of C_3D_6O the tritium activity changes are within 1 SD. Interestingly, one of the four 6h tests (where bubble activity was in the form of streamers, not individual large bubble clusters) with neutron-seeded cavitation of C_3D_6O also gave a null result. This appears to have been due to the occurrence of significant comet-like bubble formations during this particular test. As was mentioned earlier, the presence of streamers also does not give rise to any SL light emission. It is not clear why this particular test gave rise to streamers but the net effect of the change in thermal-hydraulic conditions is unmistakable and goes a long way towards underscoring the importance of attaining appropriate bubble cluster formations to attain bubble fusion.

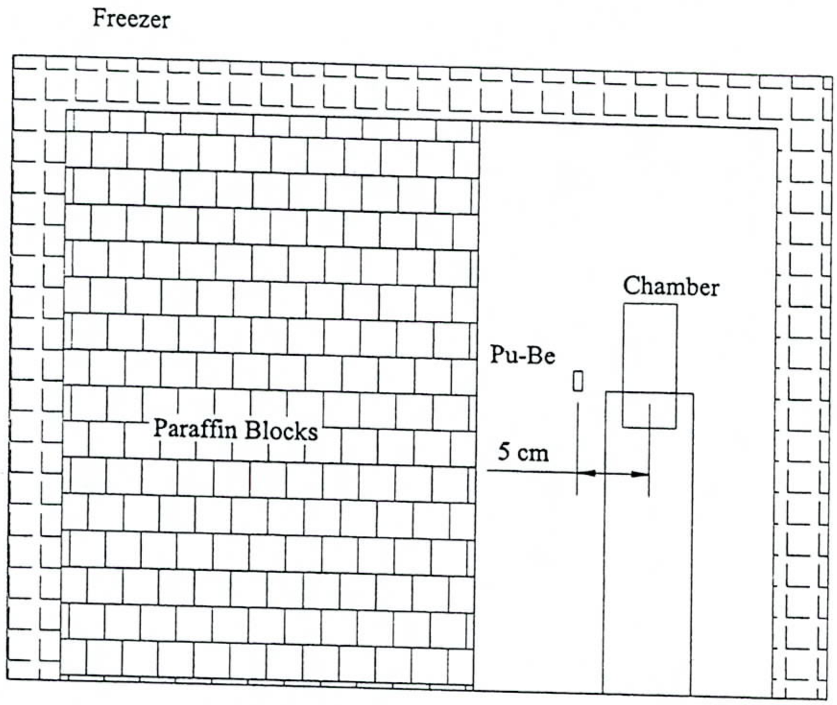
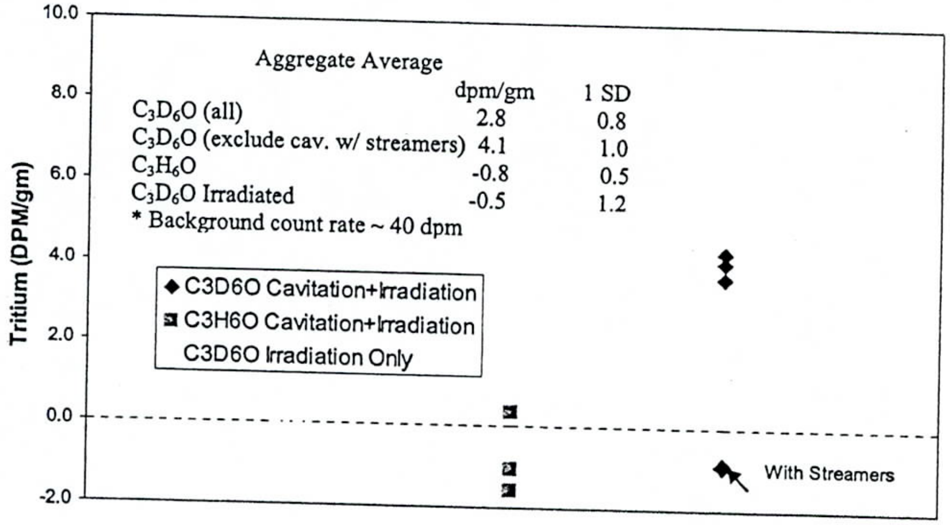


Figure 8: Schematic of experimental apparatus for tritium emission


Aggregate Average

	dpm/gm	1 SD
C_3D_6O (all)	2.8	0.8
C_3D_6O (exclude cav. w/ streamers)	4.1	1.0
C_3H_6O	-0.8	0.5
C_3D_6O Irradiated	-0.5	1.2

* Background count rate ~ 40 dpm

Figure 9: Results of tritium emission counting

5. DISCUSSION AND CONCLUSIONS

Bubble thermal-hydraulics was studied in relation to sonoluminescence light emission and shock wave signals. It was found that strong shock waves from spherical bubble cluster implosions correspond to the generation of significant sonoluminescence light emission, whereas streamer-like bubble formations produce neither distinct shock waves nor sonoluminescence light signals. The bubble cluster lifetime (typically 2 to 5 ms) was much longer than the acoustic driving cycle period (~50 μs) and a contraction was observed at ~0.8 ms, indicating the presence of complex thermal-hydraulic phenomena.

Tritium counting was conducted systematically by using a Beckman LS6500 scintillation counter for the samples obtained from the multiple 6-h bubble fusion experiments with deuterated acetone as well as for the control experiments with non-deuterated acetone. Irradiation only experiments were also performed for deuterated acetone in the presence of the neutron source, but without cavitation. Results of tritium measurements confirmed reported results (Taleyarkhan et al., 2002, 2004a) that the production of statistically significant emissions of tritium occurs only during neutron-seeded acoustic cavitation of deuterated acetone. Control experiments with irradiation alone, and neutron seeded cavitation of non-deuterated (H-bearing) acetone produced null results. The results indicate the possible occurrence of thermonuclear fusion reactions in neutron-seeded acoustic cavitation with deuterated acetone.

NOMENCLATURE

C_3D_6O	Deuterated Acetone
C_3H_6O	Non-deuterated Acetone
D	Deuterium
DPM	Disintegrations per minute
^3He	Helium-3
MIC	Microphone
n	Neutron
p	Proton
PNG	Pulse Neutron Generator
PZT	Lead-Zirconate-Titanate
SD	Standard Deviation
SL	Sonoluminescence
T	Tritium

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