

**To:** Professor Lefteri H. Tsoukalas, Head of the School of Nuclear Engineering

**From:** The Fact-finding Committee with the Members,

Prof. Chan K. Choi, Franklyn M. Clikeman, and Karl O. Ott

**Date:** Thursday, 23 February 2006

**Subject:** Fact-finding Committee Report



The three-member Fact-finding Committee submits the following report in response to your request dated on February 7, 2006 (see the attached memo from the Head to the Committee) to follow up on the complaints received from within and outside Purdue.

The Committee was charged to inquire about the circumstances surrounding a publication entitled "Bubble Dynamics and Tritium Emission During Bubble Fusion Experiments," by Y. Xu, A. Butt, and S. T. Revankar, which appeared in the conference proceedings of the 11<sup>th</sup> International Topical Meeting on Nuclear Reactor Thermal-Hydraulics (NURETH-11) in Avignon, France during October 2-6, 2005 (Publication 548) (see attached paper). The Committee was specifically requested to address the issues surrounding the data reported, the method of analysis, the actual authorship and the conclusions stated.

The Committee met five times including two sessions with two out of three authors. Meetings were held on February 9<sup>th</sup>, 15<sup>th</sup> (both with the committee members only), February 17<sup>th</sup> (with the committee members and Mr. A. Butt), February 20<sup>th</sup> (with the committee members and Mr. A. Butt and Dr. Y. Xu), and February 22<sup>nd</sup> (with the committee members only). There were no minutes kept for the ongoing nature of the committee activity. The third author, Prof. S. T. Revankar, did not wish to participate.

The Committee focused on two main issues; i.e., 1) issues related to the experiment, and 2) the issues related with the actual authorship of the said paper and its companion paper entitled "Confirmatory experiments for nuclear emissions during acoustic cavitation," by Yiban Xu and Adam Butt, which appeared in the *Nuclear Engineering and Design (NED)*, 235 (2005) 1317-1324 (see attached paper). The NED publication preceded that of NURETH-11.

The Committee finds the following facts and observations:

- 1) The said experiments were claimed to have been done mostly during February 2004 in the Purdue Pharmacy Building by Dr. Yiban Xu. The Committee finds that the claim is consistent with the Laboratory log book time of the experiments – Feb. to April, 2004 (see attachment). There are speculations about the method of the data analysis and the conclusions stated in NURETH-11 paper but the Committee did not dwell on those issues as they are characteristic to the experimentalists. The Committee noted, however, that both the NED and the NURETH-11 papers described the same experiments and the same results.

- 2) About the actual authorship, the Committee finds that i) Mr. Adam Butt did not make contributions to neither NED paper nor the NURETH-11 paper. Adam was asked by Professor Teleyarkhan to be a co-author with Dr. Xu on the NED paper (see the attached statement from Adam Butt dated 2/23/2006). The NED paper was already written by the time Adam saw it and Adam's name was added to the publication the day before submitting it to the NED editor for review. Adam's input was to grammatically review the paper as well as double check the data being used. Adam also learned his authorship on the NURETH-11 paper a week before the conference when he was first given a copy of the NURETH-11 paper by Professor Teleyarkhan. Professor Teleyarkhan asked Adam to go to France give a presentation on the NURETH-11 paper. Adam was not able to travel to France and Professor Revankar instead presented the NURETH-11 paper. ii) Dr. Yiban Xu stated first that he wrote the NED paper and later he re-stated that he only wrote the first draft copy of the paper. Dr. Xu completed his Ph. D. degree in May, 2004 from the School of Nuclear Engineering from Purdue West Lafayette campus and his Ph. D. thesis is readily available. The Committee noted the drastic departure in the command of English language and the writing style in the NED paper from those in the Ph. D. thesis. Dr. Xu stated that he wrote only a draft version of the manuscript. The Committee followed up with an inquiry as to the actual person(s) who provided the writing assistance. Dr. Xu deferred his answer to be given next day (i.e., by Tuesday, February 21, 2006) but as of this report writing the Committee did not receive any response from Dr. Xu even after having reminded him on this question (see the attached email on February 21, 2006). iii) The Committee attempted to reach Professor Revankar (see attached email sent to his attention on February 21, 2006) but he refused to have a meeting as indicated in his email response (see attached email on February 22, 2006). Professor Revankar is a co-author in the NURETH-11 paper, but not a co-author in the NED paper. The Committee had difficulties in understanding why Professor Revankar did not appear as co-authors in both papers since both papers describe the same experiments and the same results. iv) In reference to the actual authorship and the writing styles of both said manuscripts, the Committee finds surprising similarities in other publications, e.g., "Nuclear Emission During Self-Nucleated Acoustic Cavitation," by R. Teleyarkhan, et al., in *Physical Review Letters*, **96**, 034301 (27 Jan 2006) and "Evidence for Nuclear Emissions During Acoustic Cavitation," by R. Teleyarkhan, et al., in *Science*, **295**, 1868-1873 (8 March 2002) (see attached papers). The Committee has only information on the draft of the NED paper but true authorship could not be determined at this point because Dr. Xu refused to provide the information. Dr. Xu was clearly concerned about disclosing the name of the person who wrote the paper as it could jeopardize, in his own words, the "confirmatory nature" of the experiments.

The Committee concludes its findings and observations with a note that the task was a very delicate matter which involved the colleagues in the School and that maintaining the utmost objectivity was the paramount priority.

Attachments as stated

**Subject:** Fact-finding Committee

**From:** "Lefteri H. Tsoukalas" <tsoukala@purdue.edu>

**Date:** Tue, 07 Feb 2006 11:53:13 -0500

**To:** Chan Choi <choi@ecn.purdue.edu>, Frank Clikeman <clikeman@ecn.purdue.edu>, ott@ecn.purdue.edu

Dear Professor Choi,

I would like to request that you chair an faculty fact-finding committee that inquires about the circumstances surrounding a publication entitled "Bubble Dynamics and Tritium Emission During Bubble Fusion Experiments," by Y. Xu, A. Butt, and S. T. Revankar which is presented and found in the Proceedings of the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics (NURETH-11), Avignon, France, October 2-6, 2005 (Publication 548).

Some complaints about the circumstances of this publication from within and outside our institution have come to my attention, including, but not limited to, issues surrounding the data reported, the method of analysis, the actual authorship and the conclusions stated. The allegations range from misconduct to ethical issues. Hence it is important to report factual details surrounding the publication.

This is a very delicate matter and it is important to proceed with the utmost objectivity. I recommend our distinguished faculty emeriti, Profs. F. Clikeman and K.O.Ott as members of the committee. Attached you will find the paper.

Thank you very much.

Lefteri

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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 ( $\sim 1000$  Mbar) pressure and ( $\sim 10^7$  K) 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 Bremsstrahlung radiation composed of excited plasmas in the  $10^6$  K 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  $\mu\text{m}$ ) 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)



The products of D-D fusion reaction are: a neutron ( $n$ ), a proton ( $p$ ), Helium ( $\text{He}$ ) and tritium ( $T$ ). Helium ( ${}^3\text{He}$ ) 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.

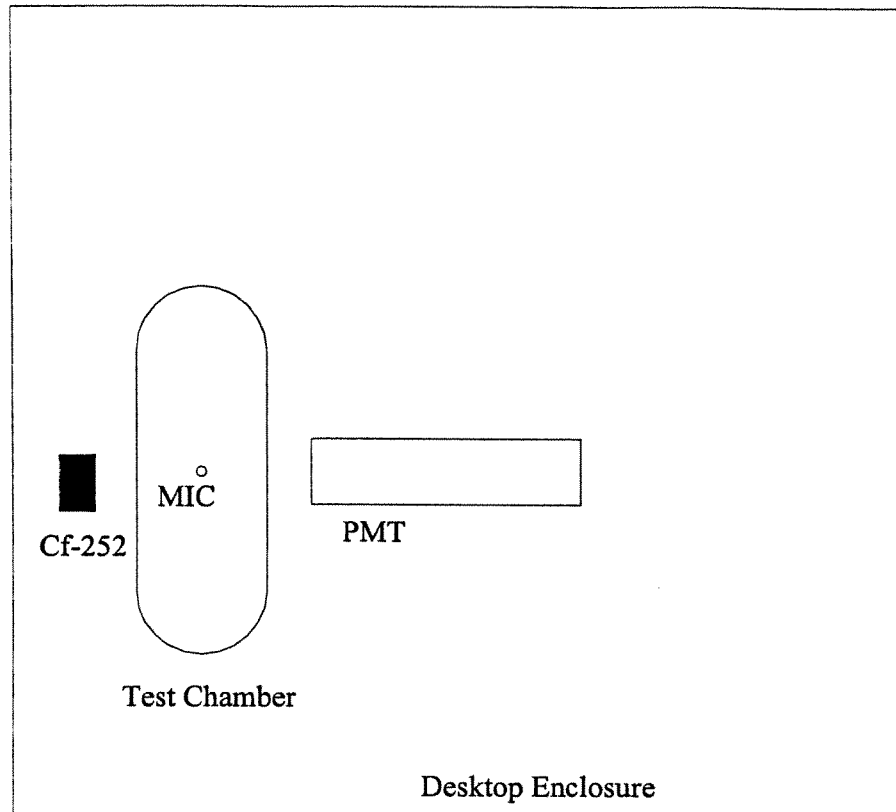
This paper focuses on reporting investigations on two aspects of bubble nuclear fusion: transient bubble dynamics along with SL light emission, and tritium production. These two topics are presented separately. The first part of this manuscript discusses observations of bubble thermal-hydraulics during 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<sup>TM</sup> 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 transmuted 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  $\sim\pm 17$  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 ( $\sim 10^2 \mu\text{m}$ ) 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  $\sim 400 \mu\text{m}$  (Nigmatulin et al., 2002), whereas the size of individual clusters is about 10 times

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.

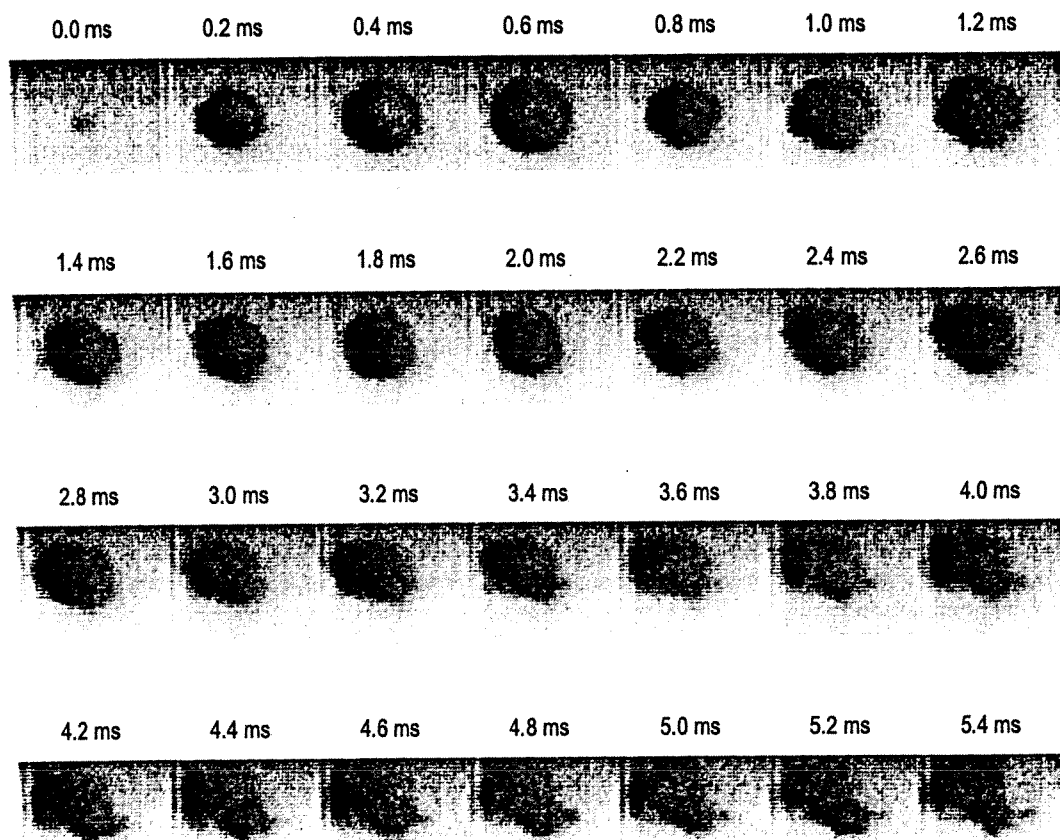


Figure 2: Individual bubble cluster ( $C_3H_6O$ , 4 °C,  $\sim\pm$  17 bars, 16.7 kPa)



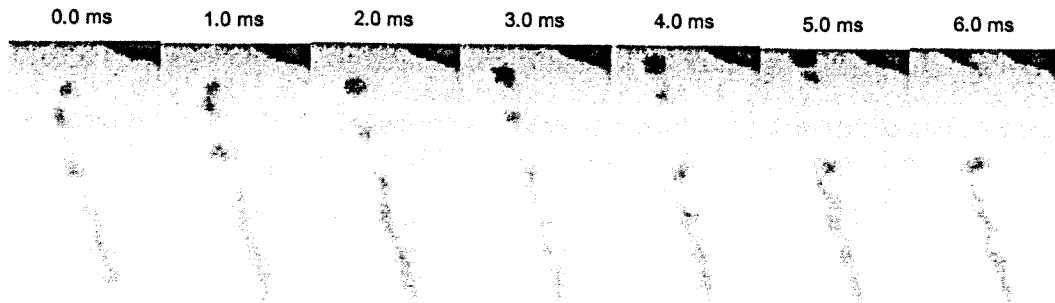


Figure 3: Comet-like streamers ( $C_3H_6O$ , 4 °C,  $\sim\pm$  17 bars, 16.0 kPa)

### 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<sup>TM</sup> 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  $\mu$ s 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.

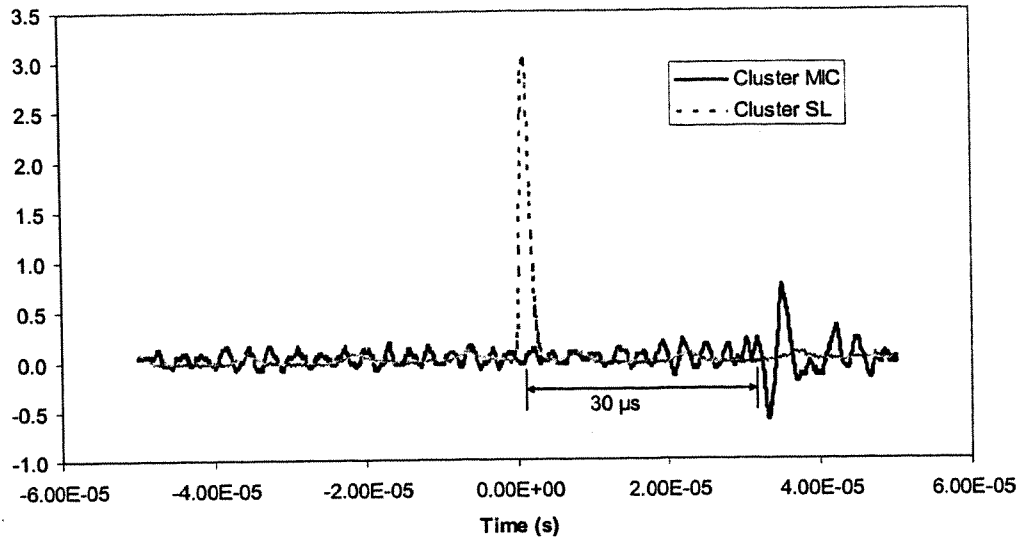


Figure 4: Signals from microphone and PMT of individual cluster

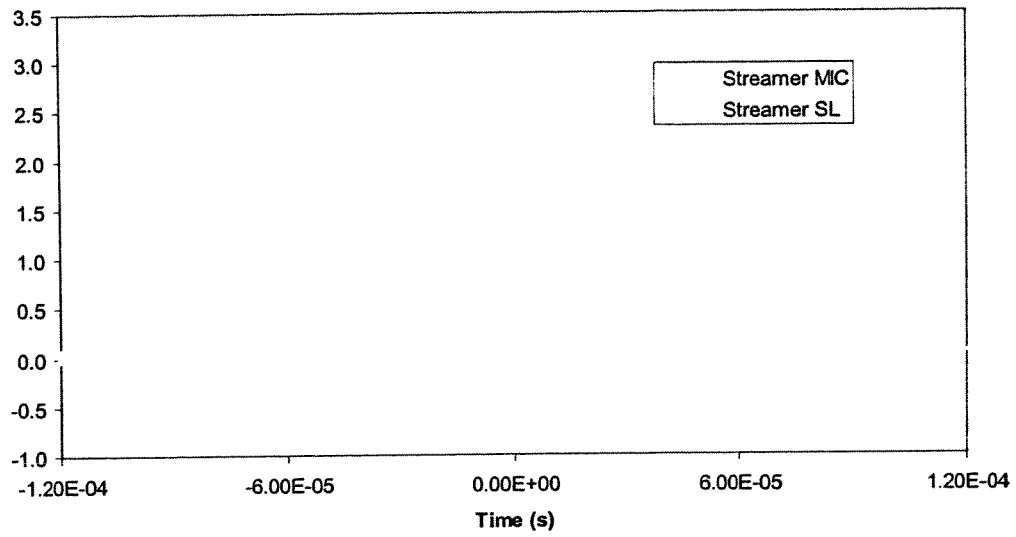


Figure 5: Signals from microphone and PMT of streamers

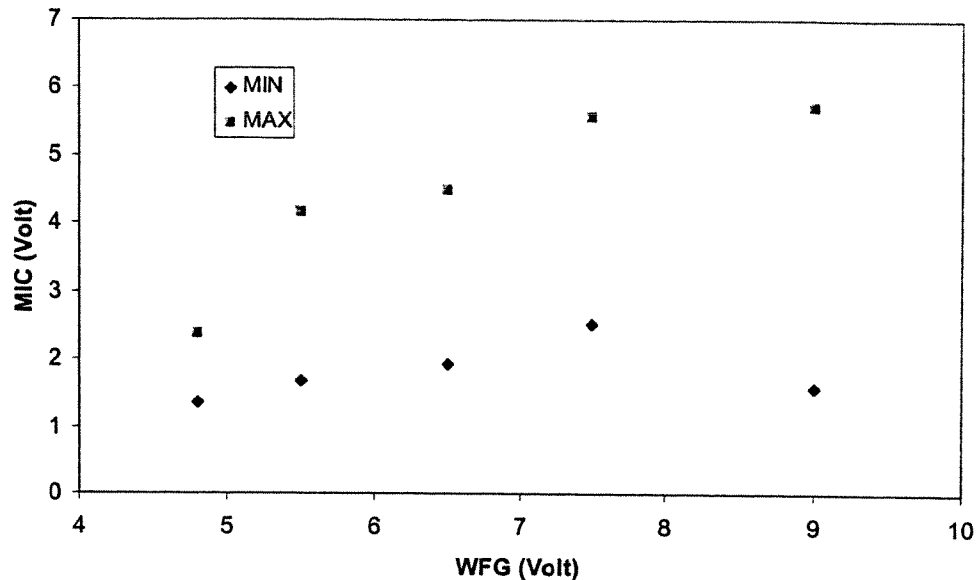


Figure 6: Amplitudes of microphone signals

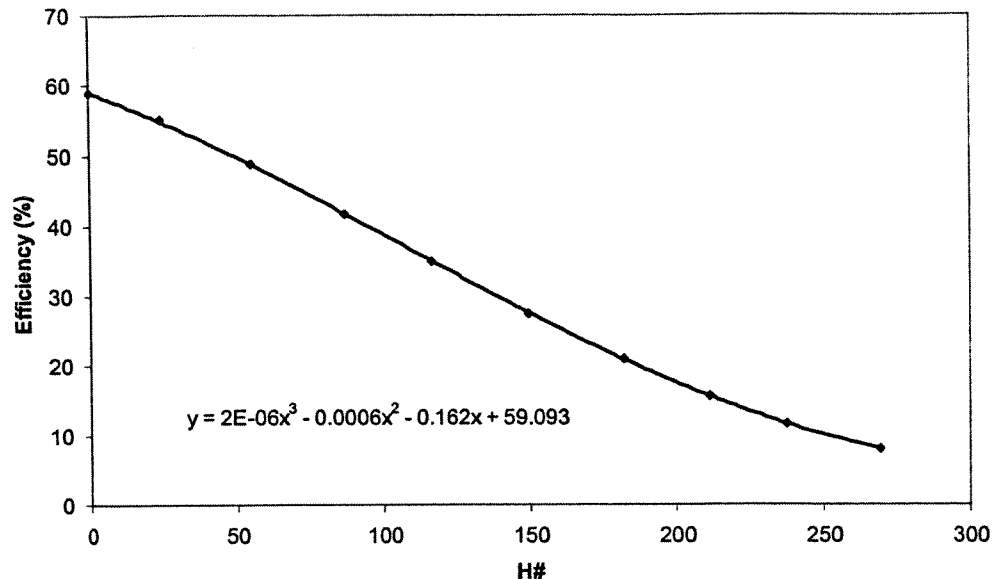
#### 4. RESULTS OF TRITIUM EMISSION

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<sup>TM</sup> 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<sup>TM</sup> 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<sup>TM</sup> 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<sup>TM</sup>, 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  $2 \times 10^6$  n/s) was available and therefore, utilized. The test cell (maintained at  $\sim 0^\circ C$  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  $\sim 6$  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 pipetted and mixed with 15ml of Ultima Gold<sup>TM</sup> 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<sup>TM</sup> liquid scintillation counter. Results of tritium activity changes are displayed in Figure 9. It is seen that a statistically significant increase ( $\sim 4$  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.

## Freezer

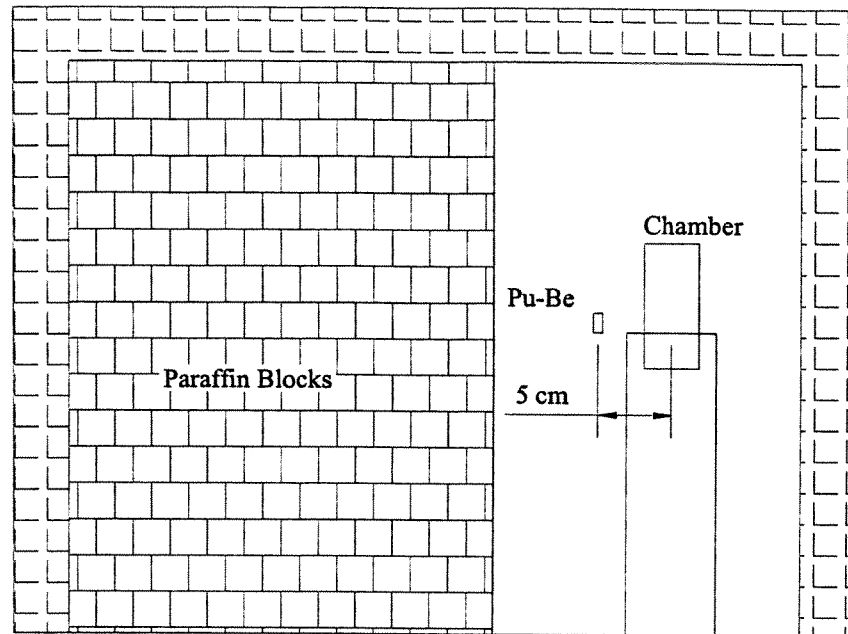


Figure 8: Schematic of experimental apparatus for tritium emission

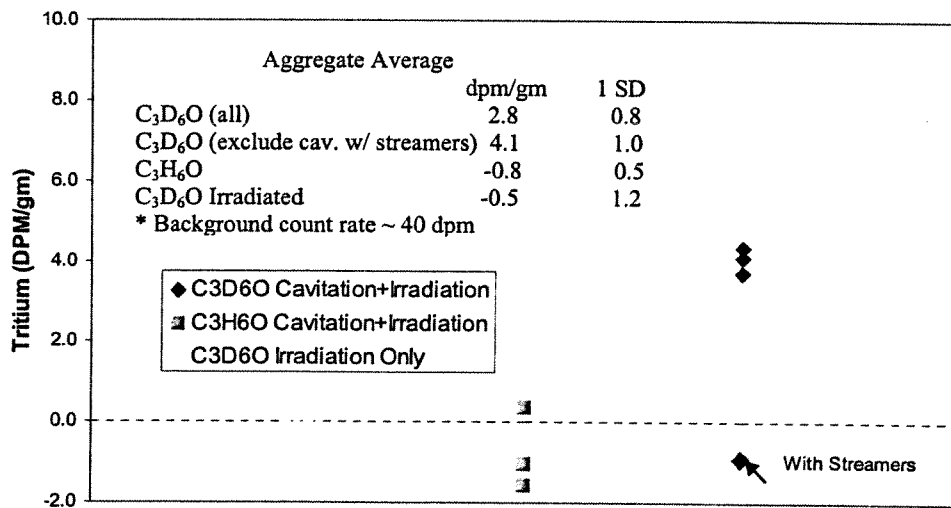


Figure 9: Results of tritium emission counting

## 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 ( $\sim 50 \mu\text{s}$ ) and a contraction was observed at  $\sim 0.8$  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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## Confirmatory experiments for nuclear emissions during acoustic cavitation

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### Abstract

Confirmatory experiments were conducted to assess the potential for nuclear fusion related emissions of neutrons and tritium during neutron-seeded acoustic cavitation of deuterated acetone. Corresponding control experiments were conducted with normal acetone. Statistically significant (5–11S.D. increased) emissions of 2.45 MeV neutrons and tritium were measured during cavitation experiments with chilled deuterated acetone. Control experiments with normal acetone and irradiation alone did not result in tritium activity or neutron emissions. Insights from imaging studies of bubble clusters and shock trace signals relating to bubble nuclear fusion are discussed.

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### 1. Introduction

It is well-known (Gross, 1984) that the thermonuclear fusion of deuterium (D) atoms requires high pressures, high temperatures and sufficient length of confinement time. The intense implosive collapse of bubbles, including acoustic cavitation bubbles, can lead to extremely high compressions and temperatures, and

to the generation of light flashes attributed to sonoluminescence and involves energy focusing of  $\sim 10^{11}$  (Crum and Matula, 1997; Camara et al., 2004). The possibility of using the phenomenon of sonoluminescence for attaining thermonuclear fusion in collapsing gas–vapor cavities has been predicted theoretically as a possibility if appropriate techniques and methodologies were discovered and developed to lead to intense enough compressions and heating (Moss et al., 1996; Nigmatulin et al., 2004; Taleyarkhan et al., 2004b). Taleyarkhan et al. (2002, 2004a) provided experimental evidence of such nuclear emissions using the novel experimental technique and approaches they developed. In this methodology, neutrons are used (much like in a conventional fission reactor where neutrons inter-

*Abbreviations:* DPM, disintegrations per minute; PNG, pulse neutron generator; PRE, proton recoil edge; PSD, pulse shape discrimination; S.D., standard deviation; SL, sonoluminescence

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act with uranium nuclei and produce more neutrons) to seed nanometer size vapor bubbles in a tensioned organic liquid (acetone) which then grow by factors of  $\sim 100,000$  and then intensely implode to produce flashes of sonoluminescence light accompanied with intense localized pressures, and temperatures for the compressed vapor molecules. In a deuterated liquid, the approach resulted in evidence of statistically significant neutron and tritium emissions (Taleyarkhan et al., 2002, 2004a,b).

The aim of the present study and experiments was to confirm if, by following the cited conditions and methodology by Taleyarkhan et al. (2002, 2004a) that nuclear fusion signatures (i.e., statistically significant  $\leq 2.45$  MeV neutrons and tritium emissions) can result during neutron-seeded acoustic cavitation of  $C_3D_6O$  at  $\sim 0^\circ C$ , but not with neutron irradiation alone, nor while conducting corresponding neutron-seeded acoustic cavitation experiments with  $C_3H_6O$  since thermonuclear fusion of H-atoms is not possible (Gross, 1984).

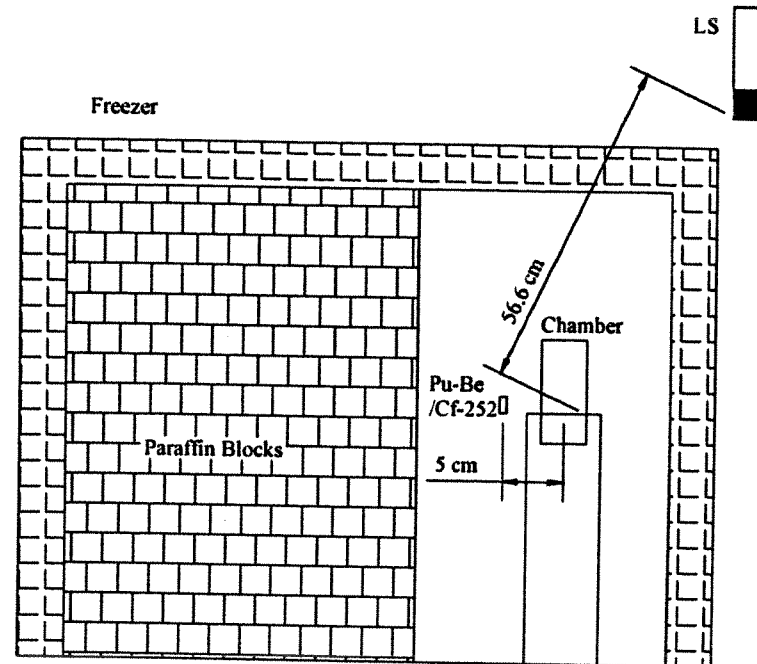
## 2. Experiment set-up

Following the methods and apparatus dimensions reported in the published literature (Taleyarkhan et al., 2002, 2004a), a test cell ( $\sim 62$  mm in diameter and  $\sim 200$  mm in height) made of Pyrex<sup>TM</sup> driven with a cemented PZT piezoelectric driver ring was constructed. The system was driven with a 40 W PiezoSystems<sup>TM</sup> linear amplifier and a Agilent<sup>TM</sup> wave-form generator as shown schematically in Fig. 1. Experiments were conducted to assess if neutrons and/or tritium emissions occur when conducting neutron-seeded acoustic cavitation experiments with deuterated acetone ( $C_3D_6O$ , certified 99.92 at% D). Corresponding control experiments were also devised without cavitation (i.e., neutron irradiation alone) as well with neutron-seeded cavitation in normal acetone ( $C_3H_6O$ , 100% pure). The negative pressure threshold for bubble nucleation by fast neutrons in acetone is  $-7$  to  $-8$  bar (Hahn, 1961). A pressure map of the chamber was obtained by means of a calibrated hydrophone. The drive voltage corresponding to the onset of cavitation (defined herein as the onset of nucleation and collapse of bubbles within a 10 s observation period) in the presence of neutrons was first determined to get

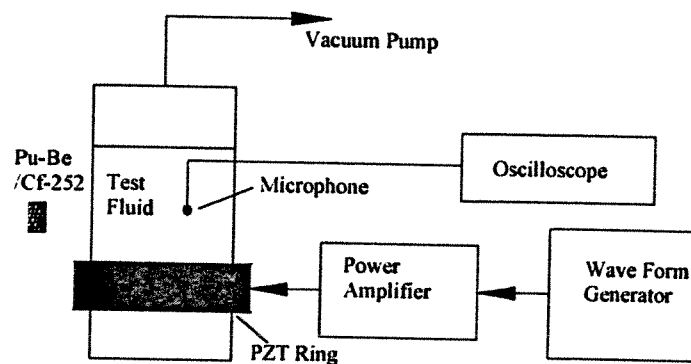
a state point corresponding to  $\sim \pm 7$  bar magnitude, and then doubled (as done by Taleyarkhan et al., 2002) to obtain the approximate drive pressure amplitudes of  $\pm 15$  bar for conduct of the confirmatory experiments.

Unlike the experiments conducted by Taleyarkhan et al. (2002, 2004a) where precise time-based nucleation was performed with a 14 MeV pulse neutron generator (PNG), such apparatus was not available for the present study. Due to this unavailability seeding of bubbles was conducted using an available isotope neutron source. This is considered reasonable since the present study was not focused on timing of sonoluminescence flashes and time-correlation of emitted neutrons with sonoluminescence, etc., but to investigate if the key nuclear fusion signatures (2.45 MeV neutrons and tritium) are possible to detect in statistically significant quantities with neutron-seeded cavitation of  $C_3D_6O$ . The acoustic driving system, filtration (with  $1 \mu m$  filters), degassing and system pressure ( $\sim 10$  kPa) were kept similar to that used in the Taleyarkhan et al. (2002, 2004a) experiments. Upon test cell construction, liquid degassing and performance characterization it was confirmed via counting microphone shock trace histories that  $\sim 10$ – $20$  bubble clusters could be generated per second with a drive amplitude of  $\sim \pm 15$  bar and a resonance frequency of  $\sim 19.6$  kHz for  $C_3D_6O$  and about 20.6 kHz for  $C_3H_6O$ . Although the bubble cluster activity was not as high as reported (Taleyarkhan et al., 2002, 2004a), this performance was considered adequate for overall confirmatory purposes.

As is well-known, the fusion of D atoms (Gross, 1984) results in the emission of a proton, helium-3, a neutron (of 2.45 MeV energy) and tritium. Protons (in the MeV range) are charged particles which cannot traverse more than  $\sim 1$  mm in the liquid before getting absorbed, and therefore, cannot be measured with detectors outside of the apparatus. The same problem holds true for helium-3 atoms which are non-radioactive and difficult to detect in small quantities. Neutrons are uncharged particles which can leak out of the test chamber and can be detected with suitable instrumentation. Also, tritium being a radioactive gas which remains in the test liquid can be counted for beta-decay activity (if a suitable state-of-the-art beta spectrometer is available). Therefore, testing was initiated systematically for monitoring the key signatures consisting of tritium and neutron emissions.



(a) Layout of experimental apparatus



(b) Test cell and acoustic drive chain

Fig. 1. Schematic of experimental apparatus.

### 3. Tritium detection experiments

Tests were first 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  $2 \times 10^6$  n/s) was available and there-

fore, utilized. The test cell (maintained at  $\sim 0^\circ C$  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 Fig. 1 along with the relative position of the Pu–Be neutron source. Tests were conducted with neutron irradiation alone, followed with tests

with neutron-seeded cavitation—systematically changing only one parameter at a time. Neutron-seeded acoustic cavitation was conducted for  $\sim 6$  h duration. Liquid samples were taken before and after cavitation from the liquid poured into the same test chamber. For each sample  $1 \text{ cm}^3$  of acetone was pipetted and mixed with  $15 \text{ cm}^3$  of Ultima Gold<sup>TM</sup> scintillation cocktail in a borosilicate glass vial. Each vial was counted for 100 min for tritium beta decay activity (i.e., in the 5–19 keV energy emission window) in a calibrated Beckman LS6500<sup>TM</sup> liquid scintillation counter. Results of tritium activity changes are displayed in Fig. 2. As the legend indicates, tritium data for neutron-seeded cavitation tests for 6 h with  $\text{C}_3\text{D}_6\text{O}$  are displayed using solid diamond symbols, whereas solid triangles represent data obtained with 6 h neutron irradiation (without cavitation) for  $\text{C}_3\text{D}_6\text{O}$ . Data obtained for the 6 h control tests with neutron-seeded cavitation of  $\text{C}_3\text{H}_6\text{O}$  are displayed with solid square symbols. It is seen that a statistically significant increase ( $\sim 4$ –5 S.D.) of tritium is only observed for tests with neutron-seeded cavitation of  $\text{C}_3\text{D}_6\text{O}$ . For neutron-seeded cavitation tests with the control liquid  $\text{C}_3\text{H}_6\text{O}$ , as well as for tests with neutron irradiation of  $\text{C}_3\text{D}_6\text{O}$  the tritium activity changes are within 1 S.D. One of the four 6 h tests with neutron-seeded cavitation of  $\text{C}_3\text{D}_6\text{O}$  resulted in no increase of tritium activity. This appears to have been due to the occurrence of significant and persistent ( $>30$  ms) comet-like (see discussion in a later section) bubble streams composed of tiny ( $\ll 1$  mm)

bubble clouds during this particular test alone and not the intensely imploding short-lived ( $<5$  ms) individual and large ( $\sim 6$  mm) spherical bubble clusters required for high-temperature and compression states as was the case for the other remaining three tests. Notably, for the other three remaining 6 h tests with  $\text{C}_3\text{D}_6\text{O}$  large bubble cluster implosions were present not comet-like bubble streams. Therefore, it appears that small-dimension collapsing cavitation bubbles in a comet-like configuration are not conducive for generating intense-enough nuclear fusion conditions and is in line with proposed theory (Nigmatulin et al., 2004).

#### 4. Neutron emission measurements

Upon acquisition of neutron detection equipment tests were also initiated for monitoring changes in neutron activity with and without cavitation using the same closed freezer compartment and acoustic drive train. However, a short while after completing the experiments for tritium monitoring the experiment location had to be shifted to a new building off-campus where the 1 Ci Pu–Be isotope neutron source could not be relocated. Instead, a 0.5 mCi Cf-252 isotope neutron source was available for use. Also, a new test cell (which could produce a relatively lower approximately  $<10$  bubble clusters per second) had to be constructed due to mechanical breakage that developed in the first test cell. A liquid scintillation

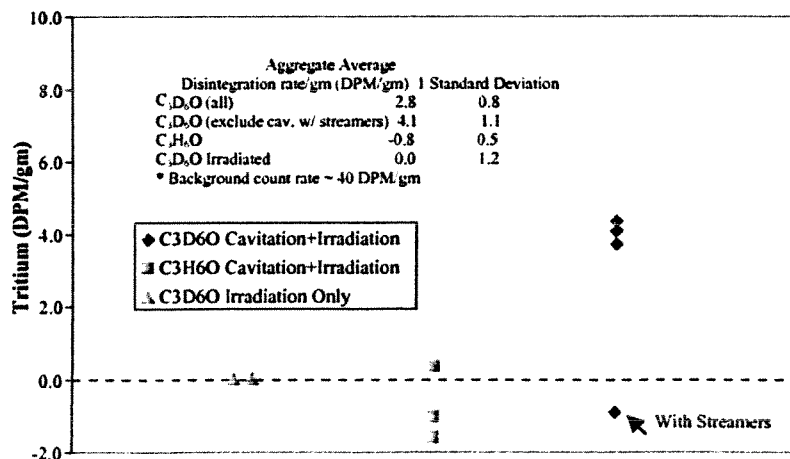


Fig. 2. Results of tritium counting.

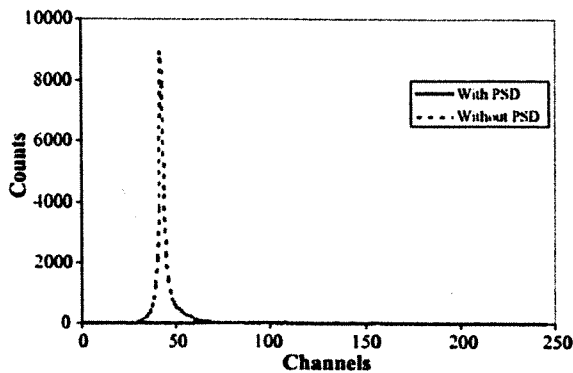


Fig. 3. Pulse shape discrimination (PSD) for gating out gamma ray emissions (using mono-energetic Co-60 source).

(LS) detector from Eljen Technologies Inc. (NE-213 type liquid-based, 5 cm × 5 cm) was procured and used for neutron detection with data acquisition electronics as reported earlier (Taleyarkhan et al., 2004a). Standard pulse shape discrimination (PSD) techniques (Harvey and Hill, 1979) were utilized using an Ortec-552 pulse shape analyzer to gate out gamma rays, as shown in Fig. 3 (where it is seen that the gamma rays component can be convincingly discriminated). For evaluating the proton recoil edge corresponding to 2.45 MeV neutron energy during pulse-height data acquisition calibration was conducted using Co-60 and Cs-137 monoenergetic gamma sources. The

respective pulse height spectra are shown in Fig. 4 from which the 2.45 MeV proton recoil edge was estimated (Harvey and Hill, 1979) to lie around channel 85. The neutron pulses from the LS detector were recorded by a UCS-20™ multichannel analyzer (from Spectrum Techniques Inc.). Typical raw pulse-height data (for total counts collected in individual channels) are depicted in Fig. 5 for C<sub>3</sub>H<sub>6</sub>O and C<sub>3</sub>D<sub>6</sub>O, respectively, with and without cavitation in the presence of the neutron source—keeping all parameters the same between the control experiment and experiments with the deuterated liquid. Notably, it is seen from Fig. 5c that ~50% excess counts (over background) increase takes place only for neutron-seeded cavitation tests with C<sub>3</sub>D<sub>6</sub>O; this mainly occurs at and below channel 85, implying that the neutrons being emitted during cavitation are ≤2.45 MeV. The variation of counts difference between cavitation on and off for the control liquid C<sub>3</sub>H<sub>6</sub>O was found to be random in nature around the zero line. In relation to Fig. 5c for over 25 s of collection time, 2391 counts, and 1629 counts were collected with and without cavitation, respectively, using C<sub>3</sub>D<sub>6</sub>O as the test fluid in the presence of a 0.5 mCi Cf-252 source at the same position. Assuming Poisson statistics, 1S.D. of counts =  $(2391 + 1629)^{1/2} \approx 63$ . Therefore, the change in counts of about 762 (=2391 – 1629) amounts to a statistically significant increase of ~11S.D. (= 762/63).

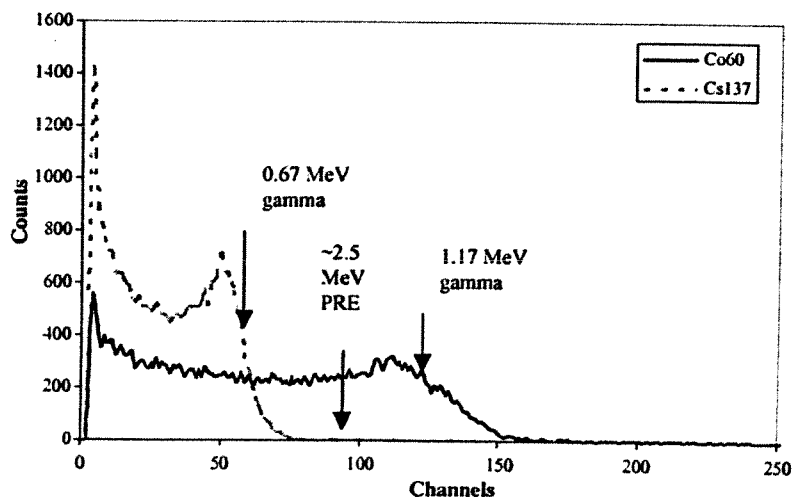


Fig. 4. Calibration of liquid scintillation detector to determine the 2.5 MeV proton recoil edge.

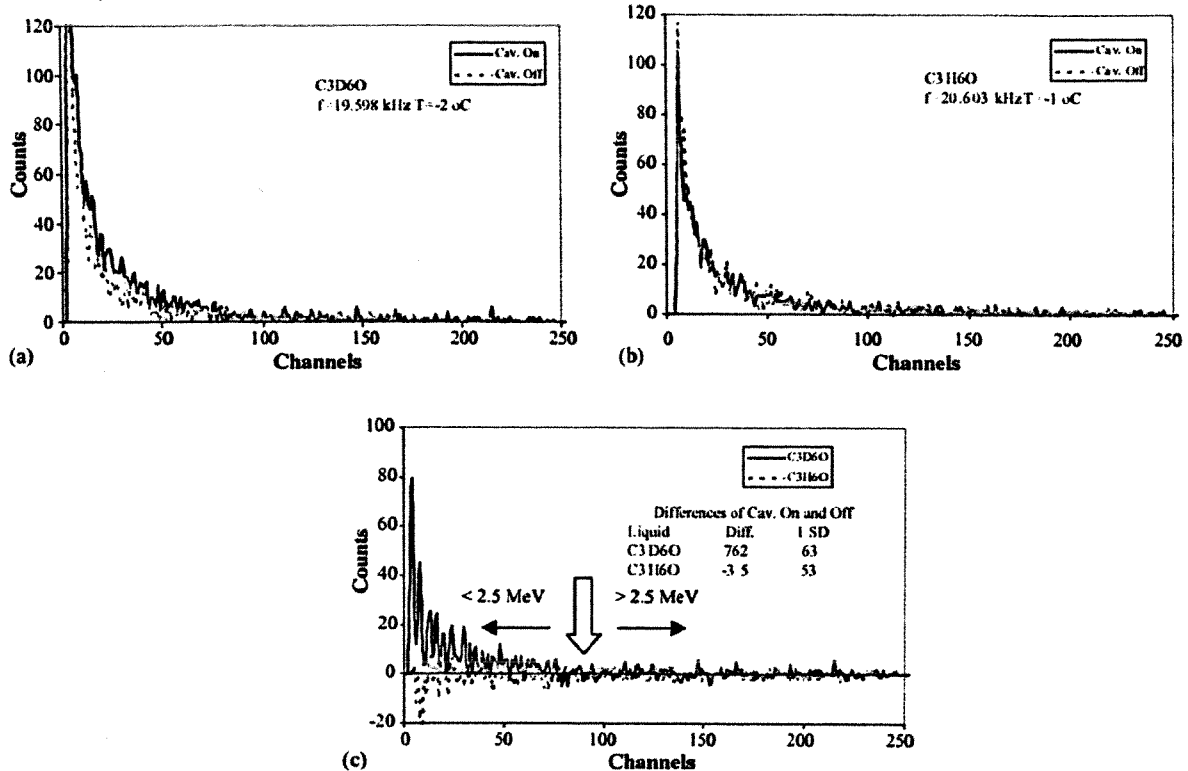


Fig. 5. (a) Representative neutron gated counts below and above 2.5 MeV proton recoil edge for tests with C<sub>3</sub>D<sub>6</sub>O at  $\sim 0^\circ\text{C}$  with and without cavitation; (b) representative neutron gated counts below and above 2.5 MeV proton recoil edge for tests with C<sub>3</sub>H<sub>6</sub>O at  $\sim 0^\circ\text{C}$  with and without cavitation; (c) representative neutron gated counts below and above 2.5 MeV proton recoil edge for tests with C<sub>3</sub>D<sub>6</sub>O and C<sub>3</sub>H<sub>6</sub>O at  $\sim 0^\circ\text{C}$  with and without cavitation.

### 5. Insights into bubble dynamics behavior and possible influence on bubble fusion

During system characterization testing imaging of bubble formation and evolution was also conducted. In addition, a microphone was attached to the test cell walls to record the amplitude of shock waves generated during implosion of the bubbles (which in effect send acoustic perturbations in the liquid and can be picked up when they reach the glass walls of the test chamber). It has been postulated (Brennan, 1995) that large bubbles in the 1 mm range can break apart into clusters of tiny bubbles if the implosive collapse is violent-enough. Clearly, bubble cluster shape and form can change in a complex manner if the drive amplitude or if the pressure amplitude of the test chamber changes. It has been reported (Nigmatulin et al., 2004; Taleyarkhan et al., 2004b) that spherical bubble collapse can lead to sig-

nificantly more intensified implosions than other configurations involving aspherical collapses of bubbles. Therefore, scoping efforts were made to image the evolution of bubble clusters (using a 5000 fps camera) and relate the behavior with shock trace magnitudes to develop possible insights into the complexities involved. It was found that under nominal operating conditions (at  $\pm 15$  bar drive amplitude and test liquid at  $\sim 0^\circ\text{C}$ ) the bubble cluster formation is largely spherical and lasts for about 5 ms. The higher the test liquid temperature, the longer is the life time for the bubbles before they re-dissolve into the bulk liquid (Taleyarkhan et al., 2004b). Fig. 6 shows a typical evolution of bubble clusters under nominal operating conditions for normal acetone (for which the temperature was  $\sim 4^\circ\text{C}$ ). It is seen that the bubble cluster shape is largely spherical for up to about 3.6 ms, after which the bubble cluster starts to lose its spherical shape and tends to dis-

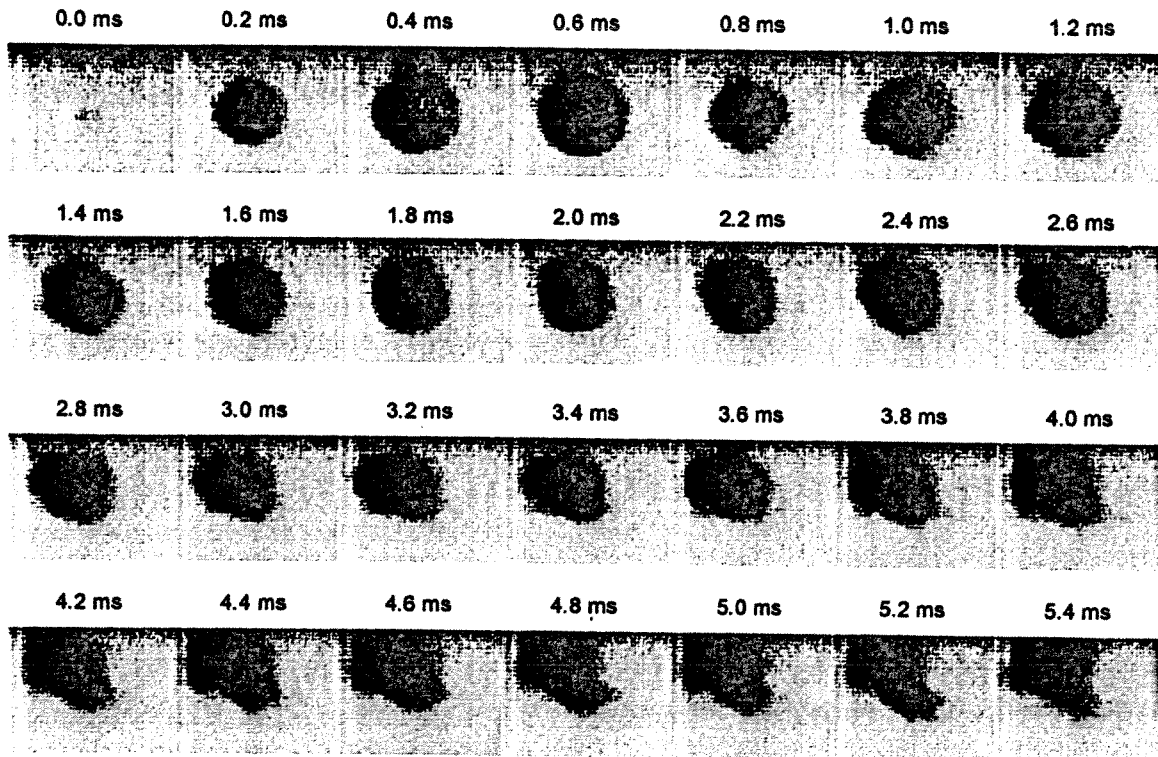


Fig. 6. Individual bubble cluster ( $C_3H_6O$ ,  $4^\circ C$ ,  $\sim \pm 17$  bars, 16.7 kPa).

perse and condenses back into the liquid. On the other hand, experience also indicated that under certain operating conditions the formation of imploding spherical bubble clusters can change radically to form streamers (comet-like structures) shown in Fig. 7. Such comet-like structures can last for several tens of milliseconds

and appear to play a critical role in terms of their ability to induce bubble nuclear fusion.

The transition from spherical bubble cluster shape to the formation of comet-like structures was accompanied with stark changes in the recorded intensity of the shock waves (recorded by the glass-wall mounted

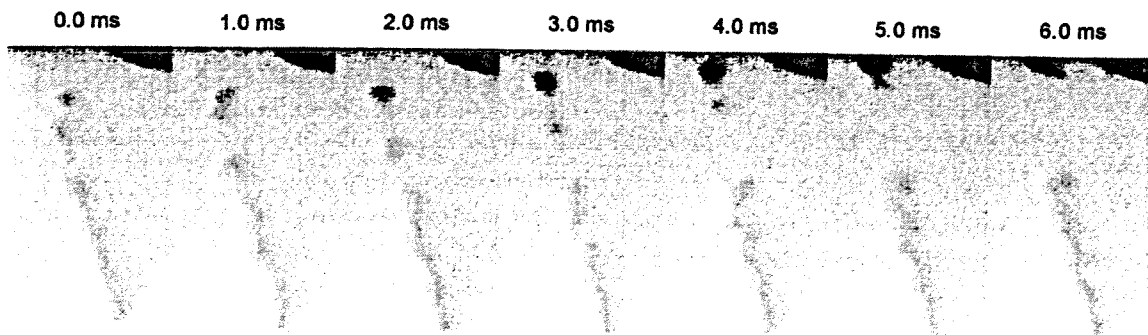


Fig. 7. Comet-like streamers ( $C_3H_6O$ ,  $4^\circ C$ ,  $\sim \pm 17$  bars, 16.0 kPa).

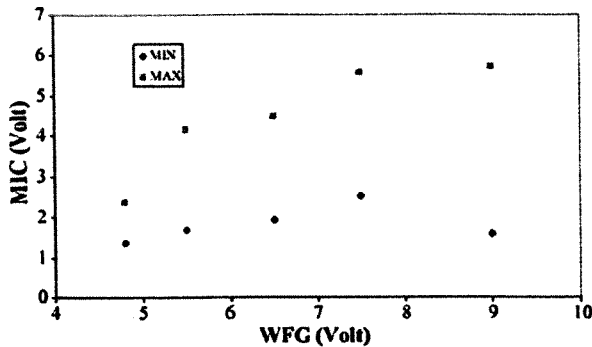


Fig. 8. Amplitudes of microphone signals with increase in drive amplitude.

microphone)—an indirect indication of implosion intensity. Fig. 8 depicts the behavior of mechanical impact-induced shock trace amplitude recorded by the microphone attached to the glass walls of the test chamber. As clearly noted, increasing the drive power from the amplifier initially increases the recorded shock trace magnitude. However, the process is non-linear. Beyond a certain power level the magnitude of shock waves levels off, and indeed, if the bubble clusters turned to comet-like structures no shock traces were recorded and the amplitude of the microphone signals dropped precipitously. These observations provide engineering insights into the tritium emission data from neutron-seeded acoustic cavitation with  $C_3D_6O$  wherein the presence of streamers also resulted in a null-result. However, this conclusion can only be considered as preliminary and more work is clearly needed to understand all the complexities of bubble cluster formation and evolution.

## 6. Conclusions

Statistically significant emissions of tritium and neutrons were measured during neutron-seeded acoustic cavitation experiments with chilled deuterated acetone. Excess neutrons were found to be  $\leq 2.45$  MeV in energy. Control experiments with normal acetone and experiments with neutron irradiation alone did not re-

sult in statistically significant neutron nor tritium emissions.

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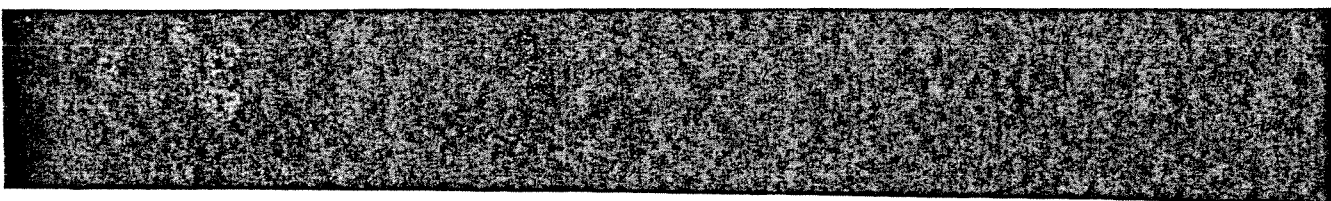
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2/23/04	Josh Walter	11:30	12:00	Y	32	30
1/24/04	ky m... ..	11:35	11:55	Y	20	20
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1/23/04	R.P. Jalyant	4:10	5:00	Y		
1/23/04	Josh Walter	4:17	12:30	Y	0.7	7
1/24/04	Y. Xu	19:00	19:00	Y		
1/25/04	V. Solomya	09:34	09:31	N		
1/25/04	V. Solomya	10:00	10:03	N		
1/26/04	V. Solomya	12:23	13:30	N		
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1/27/04	S. Walter	21:30	22:10	Y	7	7
1/28/04	S. Walter	11:00	12:40	Y	7	7
1/28/04	Y. Xu	11:00	12:40	Y	67	68
1/28/04	Jouffregard	15:0	15:7	N	31	32
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1/21/04	Neelima S.	7:15	9:35	N	40	40
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2/3/04	S. Walter	17:30	19:15	Y	5	5
2/3/04	Nader Sattari	17:30	19:15	N	5	5
2/4/04	Josh Walter	17:00	17:00	Y	10	10
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0.7 6.7	2/12/04	J. Moon	14:15	14:30	Y	08	08
7 7	2/12/04	Y. Xu	21:15	21:30	Y	06	06
7 7	2/15/04	Y. Xu	13:20	13:40	Y	06	06
67 68	2/17/04	S. Neelima	9:05	9:10	N		
31 32	2/17/04	Y. Xu	11:20	12:30	Y	6	6
30 31	2/17/04	J. Walter	17:00	12:18	Y		
31 31	"	Ravi Sathian	17:06	17:18	N		
31 31	2/17/04	Philly Young	17:45	18:50	N		
40 40	2/17/04	Y. Xu	21:00	2:00	Y		
23 25	2/18/04	J. Walter	08:15	17:00	Y	20	20
34 37	2/18/04	Y. Xu	09:30	9:00	Y		
60 60	2/18/04	J.P. VETZEL	10:24	10:36	Y	20	20
34 39	2/18/04	Nador Sathian				20	20
0 0	2/18/04	Jeremy Wiegand	207	213	N	20	20
0 0	2/18/04	Y. Xu	8:15	18:00	N	20	20
5 5	2/18/04	D. Gillies	3:30	4:20	N		
5 5	2/19/04	Y. Xu	9:30	12:00	Y	20	20
4 4	2/19/04	Z. S.	10:05	10:15	Y	159	159
0 0	2/19/04	Y. Xu	13:00	16:20	Y	20	20
07 0.7	2/19/04	R.P. Taleyarkhan	1:45	1:55pm	Y		
06 06	2/22/04	Y. Xu	10:30	11:55	Y	22	22
06 06	2/23/04	Philly Young	10:45	11:55	N	29	30



02/23/04	Y Xu	2:57	3:23	Y	18	18
2/23/04	<del>Y Xu</del>	<del>2:58</del>	<del>3:23</del>	<del>N</del>	<del>29</del>	<del>29</del>
2/23/04	Y Xu	2:40	2:45	Y	58	15
2/24/04	Y Xu	11:34	12:00	Y		
2-24-04	<del>Y Xu</del>	<del>11:40</del>	<del>12:00</del>	<del>N</del>		
2-24-04	Christopher Catto	11:49	12:00	N		
2-24-04	Sash Walker	15:00	15:15	Y	28	27
2-25-04	Neelima S.	9:20	9:25	X	20	20
	Neelima S.	.00	.05	X	29	28
2-25-04	Sung Goo Kang	2:00	2:20	N	18	19
2/25/04	Y Xu	15:00	15:20	Y		
2/26/04	Y Xu	8:15	8:10	Y	28	20
2/26/04	Ryan Bradtke	12:00	12:55	N	19	21
2/26/04	Rust Talayant	1:05	1:20	Y		
2/26/04	S.M. Lee	3:05	3:15	Y	22	22
u	Ravi Sathyan	3:05	3:30	N	22	22
2/26/04	Sash Walker	15:00	15:30	Y	22	20
2/26/04	David Williams	4:07	5:10	N	22	22
2/27/04	Sash Walker	11:00	15:30	Y	27	23
2/27/04	Phil Smagacz	2:48	15:00	N	27	27
2/27/04	Y Xu	2:48	15:00	Y		
2/27/04	Y Xu	10:00		Y	25	25
3/1/04	Phil Smagacz	10:05	4:09	N	27	30
3/1/04	Sharon Rudolph	1:45	2:51	N		
3/1/04	Stephen Paarlhoy	4:30	4:42			
3/02/04	Y Xu	9:45	12:00	Y	26	28
3/02/04	Y Xu	15:50	17:00	Y	28	28
3/2/04	J. Walker	13:50	17:17	Y	28	28
3/2/04	Ryan Bradtke	4:30	17:17	N	29	30
3/2/04	J. Walker	9:30	12:30	Y	27	30
3/13/04	al C...	10:00				

18	18	3/3/04	Nadine S	10:30	10:35	N	20	21
19	19	3/3/04	"	11:45	1:50	N	20	20
18	18	3/3/04	S. Walter	15:50	17:00	Y	20	20
		3/3/04	Yibin Xu	15:50	17:00	Y	20	20
		3/4/04	J. Moon	15:25	15:25	Y	30	20
		3/4/04	S. Walter	16:00	17:15	X	30	30
28	28	3/4/04	Y. Xu	16:00	17:15	Y	30	30
20	20	3/5/04	Y. Xu	11:20	12:20	Y	32	32
28	28	3/5/04	Y. Xu	13:00	14:55	Y	32	32
18	19	3/5/04	D. Gillies	15:10	15:25	N		
		3/6/04	S. Dearborn	15:35	15:55	N		
22	20	3/6/04	A. Roy	17:15	<del>18:30</del>	N	30	30
19	21	3/7/04	Y. Xu	9:30	19:20	Y	30	30
		3/7/04	A. Roy	19:50	20:00	N	30	30
22	22	3/8/04	Y. Xu	9:15	19:00	Y	30	30
22	22	3/8/04	D. Gillies	9:15	11:12	N		
22	20	3/8/04	A. Roy	9:30	9:35	N	30	30
22	22	3/8/04	S. Dearborn	9:35	12:01	N		
27	23	3/8/04	Dray Bregard	11:45	11:48	N		
27	27	3/8/04	Josh Walter	13:30	14:30	Y	22	22
		3/8/04	Nader Salvat	13:30	14:30	Y	22	22
25	25	3/9/04	Y. Xu	14:00	18:20	Y	30	30
29	30	3/9/04	K. P. Lalupichit	14:10	15:10	Y	21	21
		3/9/04	Daniel Gillies	14:50	15:20	N		
		3/10/04	Stephen Dearborn	15:20	18:20			
26	28	3/10/04	Y. Xu	9:00	21:00	Y	30	30
28	28	3/10/04	Th. Smagacz	9:45	5:00	N	21	21
29	30	3/10/04	E. Bhatti	11:12	10:14	Y	12	12
29	30	3/10/04	E. Mouton	12:50	12:50	Y	12	12
28	28	3/10/04	Ryan Bradley	2:35	5:50	N	13	13

3/14/04	MERLIS	08:15		Y		18 20
3/14/04	Y. Xu	11:00	11:00	Y		22 22
3/14/04	E MEMPHIS	12:01	12:06	Y		20 20
3/12/04	E MEMPHIS	12:04	12:06	Y		13 18
3/12/04	E MEMPHIS	12:15	12:18	Y		18 18
3/16/04	Y. Xu	16:30	17:00	Y		22 22
3/18/04	Y. Xu	17:30	18:00	Y		21 24
3/18/04	G. Lee	15:05	15:20	Y		25 25
3/18/04	Y. Xu	17:00	17:00	Y		25 25
3/23/04	Y. Xu	17:15	17:40	Y		25 25
3/24/04	Y. Xu	17:25	18:30	Y		25 25
3/24/04	Paul Ziemer	16:45	16:30	Y		
3/24/04	Ashley Doka	16:45	16:30	Y		20 20
3/24/04	Catherine Wanko	16:47	16:30	Y		69 69
3/24/04	Ayesha Periyadar	16:17	16:30	Y		20 20
	Sachin Reddy			Y		24 24
	Tom Khan			Y		20 20
	Julia Klingler	16:45	17:30			02MSUB200
	Joey Silvers	16:45	17:30		20	20 MR
	Sana Ahmed	16:45	17:30			68 MR
3/24/04	Maureen Pappas	16:45	17:30			0 MR
3/25/04	Y. Xu	2:45	18:30	Y		28 28
3/26/04	Y. Xu	1:45	4:00	Y		28 28
3/26/04	Bill Coon	2:30	3:15	Y		
3/26/04	E MEMPHIS	4:00	4:08	Y		29 29
3/26/04	Y. Xu	17:20	18:00	Y		29 29
3/29/04	Y. Xu	13:35	18:25	Y		29 29
3/29/04	R. Talamon	13:35	14:25	N		32 32
3/29/04	Y. Xu	20:00	4:00	Y		29 30
2/22/06	Y. Xu			Y		



18 20	3/20/04	J. Moon	14:45 <del>14:30</del>	14:30	Y	35	35
22 22	3/31/04	Y Xu	9:20	18:20	Y	30	30
20 20	3/31/04	Phil Smagacz	11:00	4:30	N	32	33
13 18	3/31/04	Phil Smagacz	11:42	19:45	N	35	42
18 18	3/21/04	Rusi Tahyarli	12:05	12:40	.		
22 22	3/31/04	Y Xu	21:40	12:30	N	30	04
22 24	4/1/04	Y Xu	10:20	14:30	Y	04	04
25 25	4/1/04	R. LANDOLT	13:45	15:35	YES	123	123
25 25	4/1/04	R. Gao	15:20	15:45	N	04	04
25 25	4/1/04	Y Xu	15:20	18:45	Y	04	04
25 25	4/1/04	J. Wenter	18:20	18:50	N	4	4
	4/1/04	Ed Morris	15:45		Y	34	
20 20	4/1/04	J. Wenter	16:50	26:30		3	3
67 67	4/2/04	R. Tahyarli	16:15	16:45	N	3	3
20 20	4/2/04	Y Xu	16:15	16:50	Y	3	3
24 24	4/5/04	Y Xu	10:00	18:15	Y	6	2
	4/5/04	Phillip Smagacz	11:10	12:06	N	46	46
2 2	4/5/04	Phillip Smagacz	2:10	5:05	N	44	44
02MSV02MS	4/06/04	Y Xu	8:35	10:30	Y		
20 MR	4/6/04	J.R. Hardy	9:55	10:30		49	50
68 MR	4/6/04	Phillip Smagacz	8:36	10:15		49	50
0 MR	4/6/04	Matt LeBeyec	8:36	10:30		49	50
28 28	4/6/04	A. Pollman	8:37	10:15		99	50
28 28	4/6/04	Sean Finer	8:37	10:20	N	49	50
29 29	4/7/04	Y Xu	9:00	14:45	Y	6	6
29 29	4/7/04	Phillip Smagacz	11:22	4:52	N	48	48
29 29	4/7/04	Stephan Padiborn	13:41	17:10	N	48	48
32 32	4/7/04	Y Xu	15:00	19:10	Y		
29 30	4/8/04	Y Xu	9:20	17:30	Y	6	6
29 30	4/8/04	Phillip Smagacz	9:41	12:14		48	47
38 30	4/8/04	Phillip Smagacz	16:13	17:24		20	21

4/12/04	G. Lee	3:20		yes	25	25
4/14/04	E. MERRIS	3:30	4:15	Y	40	40
4/14/04	A. Roy	5:40pm	5:50pm	Yes	35	
4/14/04	Y. Xu	9:30	18:10	Y	21	22
4/14/04	A. Roy	9:49am	9:50am			
4/14/04	Phillip Smagacz	12:25	2:06		8	
4/15/04	Y. Xu	10:00	15:45	Y	22	22
4/15/04	R.P. Talwar	1:30			25	25
4/15/04	Y. Xu	4:50	8:05	Y	22	24
4/15/04	Y. Xu	23:00	0:30	Y	24	24
4/16/04	Sharon Rudolph	11:26	11:41		30	30
4/16/04	William Coon	2:30	2:40			
4/16/04	Y. Xu	18:30	18:35	Y		
4/18/04	E. MERRIS	3:00	4:15	Y	42	42
4/20/04	Y. Xu	9:30	17:00	Y	24	28
4/21/04	Grey Wiegand	1:30		N	02	09
4/22/04	Y. Xu	9:30	18:30	Y	28	28
4/22/04	Y. Xu	11:00	20:30	Y	28	28
4/22/04	Y. Xu	12:50	12:30	Y	28	28
4/23/04	Y. Xu	13:40	18:40	Y	30	30
4/23/04	J. Wald	15:15	15:30	N	32	32
4/23/04	M. Satwat	15:15	15:30	N	5	6
4/26/04	Y. Xu	10:10	18:30	Y	32	32
4/28/04	S. Nelmina	1:50	12:00	N	30	30
4/28/04	Jatin Kumar Bulwani	1:50	12:00	N	30	50
4/28/04	Nelmina S.	5:50	6:00	N	30	30
4/28/04	" "	8:10	8:15	N	20	30
4/27/04	Y. Xu	9:30	12:30	Y	32	32
4/27/04	Nelmina S.	12:20	12:30	N	30	30
4/27/04	Y. Xu	14:00	18:30	Y	32	32

25	25	4/27/04	Rusi Talayan	3:00pm	3:25	Y		
40	40	4/27/04	ERICO GIOZZO	3:00pm	3:25	Y	32	32
35		4/28/04	J. Moon	2:48 PM	2:22	Y	32	32
21	22	05/04/04	Y. Xu	11:00	12:00	Y		
		05/04/04	Y. Xu	21:00	22:00	Y		
85		05/5/04	Nelma Sarbjan	4:15	4:20	N		
22	22	5/5/04	Jatinder Gulain	11	11	N		
25	25	5/5/04	Nelma S.	7:30	9:35	N		
22	24	5/6/04	Nelma S	10:40	10:45	N		
24	24	5/6/04	Nelma S	1:30	1:35	N		
30	30	5/11/04	William Coon	11:00	12:30	N		
		5/11/04	SeungGoo Kang	1:00	1:20	N		
		5/12/04	Nelma S.	9:35	9:40	N		
42	42	5/12/04	Nelma S.	12:45	12:50	N		
24	28	5/12/04	William Coon	2:15	5:15	N		
02	09	5/14/04	Schwetzer	4:30	4:40	Y		
28	28	5/17/04	vetter	13:13	13:45	Y		
28	28	5/17/04	E. Mewer	13:43	13:45	Y	13	13
28	28	5/18/04	William Coon	10:45	11:20	N		
30	30	5/18/04	G. Lee	14:01	14:20	N		
32	32	5/19/04		15:5	15:5	N		
5	6	5/20/04		7:20		Y		
32	32	5/20/04	W. Coon	12:00	7:30	N		
30	30	5/20/04	Oracy Wegard	150	155	N		?
30	50	5/25/04	W. Coon	4:30	7:00	N		
30	30	5/26/04	S. Rudolf	1:43	1:59	N		
20	30	5/26/04	SeungGoo Kang	3:00	3:15	N		
32	32	5/26/04	Oracy Wegard	135	145	N		
40	40	5/11/04	W. Coon	2:30	7:30	N		
32	32	6/2/04	Schwetzer	10:30	10:55	Y		

Re: Meeting Revisited

**Subject:** Re: Meeting Revisited  
**From:** Adam Butt <butt@purdue.edu>  
**Date:** Thu, 23 Feb 2006 15:02:21 -0500  
**To:** Chan Choi <choi@ecn.purdue.edu>, butt@purdue.edu

Prof. Choi,

Attached is the timeline of events from the standpoint of my involvement in the NED and NURETH-11 papers. Take care,

--Adam--

Chan Choi wrote:

Dear Adam,

I sincerely appreciated your participation to the ongoing committee activities. I have requested for follow up answer to Yiban on the issue of actual revisions of the NED and NURETH-11 papers. I have also sent a request for a meeting to Professor Revankar.

As discussed earlier, if and when Professor Revankar agrees to meet with us all, I will inform you hoping to wrap up our duties in good time span.

Thank you very much,

Chan Choi

Statement for Purdue NE.doc

**Content-Type:** application/msword

**Content-Encoding:** base64



Statement from Adam Butt  
2/23/2006

#### Timeline of Events

May 2004 - Adam begins working with Professor Taleyarkhan. Only involvement in Pharmacy lab is to move hardware to INOK lab (was not involved in any experiments there).

January 26, 2005 - Professor Taleyarkhan asks Adam to be a co-author on an article for the Nuclear Engineering and Design (NED) journal. Adam is a co-author to Dr. Xu.

August 27, 2005 – Prof. Taleyarkhan asks Adam to present material related to our paper at the NURETH-11 conference in France.

September 28, 2005 – Yiban Xu sends a copy of his draft for the NURETH-11 conference presentation. No draft of the conference paper was ever sent to me before the conference. I was given a printed version to review around this timeframe, but was not at all involved in the review process for this paper. The first time I saw the paper it was in its final form.

#### Summary,

- All data for the NED publication was recorded before I joined Prof. T's team (pre May 2004).
- My name was added to the publication the day before submitting it to the editor for review (NED paper).
- The NED paper was already written by the time that I saw it (I do not know who wrote it). My input was to grammatically review the paper as well as double check the data being used, which was basically to see if it had been copied accurately from our data acquisition computer to the files used to generate the graphs for the NED paper
- There was a second paper, for the NURETH-11 conference in Avignon, France from 10/2/05 – 10/7/05
- I was asked by Prof. T to go to France give a presentation on our findings
- Up until a week before the conference (9/28/05) I was under the impression that the material being presented would be that contained in the NED paper – I did not know that there was going to be a second separate paper (no one told me, even though I was asked to present).
- A week before the conference I was first given a copy of the NURETH paper by Prof. T, which I was listed as an author (along with Y. Xu and Prof. Revenkar). This time I was not even asked if I wanted to be an author and I had absolutely no input into the paper, nor were asked to review the paper.
- I did not attend the conference

Re: Meeting Tonight

**Subject:** Re: Meeting Tonight  
**From:** Chan Choi <choi@ecn.purdue.edu>  
**Date:** Tue, 21 Feb 2006 17:16:31 -0500  
**To:** Yiban Xu <yiban@ecn.purdue.edu>  
**CC:** clikeman@ecn.purdue.edu, kobott@sbcglobal.net

Dear Yiban,

I would like to express my sincere appreciation for your time and effort in our discussion last night. It was somewhat limited in your delivery as you wanted to have more time to answer some of the pointed questions.

The committee would be grateful for your response to what we had left out, namely, the issue of actual revisions of the NED and NURETH-11 papers.

Sincerely,  
Chan K. Choi

Yiban Xu wrote:

That's good idea. I will meet you at school.

Thanks,

Yiban Xu

Chan Choi wrote:

Dear Yiban,

Could you meet me at the School at 7:15 pm so that we can ride together?  
Or call me (494-6789) if you have better idea.

Chan Choi

Yiban Xu wrote:

Dear Prof. Choi,

I just read your email and I can make it at 7:30 pm. Please let me know the place and the materials need to be prepared.

Thanks,

Yiban Xu

Chan Choi wrote:

Dear Yiban,

It was a good meeting yesterday and we can learn more collectively. For tonight's gathering, please let me know as your time availability gets firm to either 7:00 p.m. or close to 8:00 p.m. I need to inform others about the meeting time tonight.

Re: Meeting Tonight

|||

Thank you,  
Chan Choi

Meeting Requested

**Subject:** Meeting Requested  
**From:** Chan Choi <choi@ecn.purdue.edu>  
**Date:** Tue, 21 Feb 2006 10:34:35 -0500  
**To:** Shripad Revankar <shripad@ecn.purdue.edu>  
**CC:** kobott@sbcglobal.net, clikeman@ecn.purdue.edu

Dear Shripad,

Professors Ott and Clikeman and I were asked by the Head of the School to look into the publication of the NURETH-11 paper on "Bubble Dynamics and Tritium Emission during Bubble Fusion Experiments," that appeared and presented in the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics (NURETH-11) during October 2 - 6, 2005. Records indicate that you are one of the authors and had made the oral presentation.

A formal inquiry was received and it requests for response from the School. The purpose of this three-member committee's charge is a fact-finding mission in that the committee is requested to inquire about the circumstances surrounding the data reported, the method of analysis, the actual authorship and the conclusion stated. This is a very delicate matter and the committee is approaching this matter with utmost objectivity.

I would like to stop by your office to initiate the communication. The meeting itself is not confidential but the contents of the meeting is naturally confidential.

Please advise me of your most earliest convenient time to meet.  
Thank you.

Sincerely,

Chan

Re: Meeting Requested

**Subject:** Re: Meeting Requested  
**From:** Shripad Revankar <shripad@ecn.purdue.edu>  
**Date:** Wed, 22 Feb 2006 09:33:42 -0500  
**To:** Chan Choi <choi@ecn.purdue.edu>

Dear Professor Choi,

I am surprised by the urgency with which you are requesting a meeting for a research paper presented at a conference last October 2005. Due to nature of content in your e-mail I will not be able to meet you on this matter until I have received proper advices and legal counseling. However I will inform you with appropriate reply to your email within 15 days.

Thank you

Shripad

At 10:34 AM 02/21/2006 -0500, you wrote:

Dear Shripad,

Professors Ott and Clikeman and I were asked by the Head of the School to look into the publication of the NURETH-11 paper on "Bubble Dynamics and Tritium Emission during Bubble Fusion Experiments," that appeared and presented in the 11th International Topical Meeting on Nuclear Reactor Thermal-Hydraulics (NURETH-11) during October 2 - 6, 2005. Records indicate that you are one of the authors and had made the oral presentation.

A formal inquiry was received and it requests for response from the School. The purpose of this three-member committee's charge is a fact-finding mission in that the committee is requested to inquire about the circumstances surrounding the data reported, the method of analysis, the actual authorship and the conclusion stated. This is a very delicate matter and the committee is approaching this matter with utmost objectivity.

I would like to stop by your office to initiate the communication. The meeting itself is not confidential but the contents of the meeting is naturally confidential.

Please advise me of your most earliest convenient time to meet.

Thank you.

Sincerely,

Chan

---

Shripad T. Revankar  
Associate Professor  
Purdue University

## Nuclear Emissions During Self-Nucleated Acoustic Cavitation

R. P. Taleyarkhan,<sup>1,\*</sup> C. D. West,<sup>2,†</sup> R. T. Lahey, Jr.,<sup>3</sup> R. I. Nigmatulin,<sup>4</sup> R. C. Block,<sup>3,†</sup> and Y. Xu<sup>1</sup>

<sup>1</sup>Purdue University, West Lafayette, Indiana 47907, USA

<sup>2</sup>Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, USA

<sup>3</sup>Rensselaer Polytechnic Institute, Troy, New York 12180, USA

<sup>4</sup>Russian Academy of Sciences, 6 Karl Marx Street, Ufa 450000, Russia

(Received 19 September 2005; published 27 January 2006)

A unique, new stand-alone acoustic inertial confinement nuclear fusion test device was successfully tested. Experiments using four different liquid types were conducted in which bubbles were self-nucleated without the use of external neutrons. Four independent detection systems were used (i.e., a neutron track plastic detector to provide unambiguous visible records for fast neutrons, a  $\text{BF}_3$  detector, a NE-113-type liquid scintillation detector, and a NaI  $\gamma$  ray detector). Statistically significant nuclear emissions were observed for deuterated benzene and acetone mixtures but not for heavy water. The measured neutron energy was  $\leq 2.45$  MeV, which is indicative of deuterium-deuterium (D-D) fusion. Neutron emission rates were in the range  $\sim 5 \times 10^3$  n/s to  $\sim 10^4$  n/s and followed the inverse law dependence with distance. Control experiments did not result in statistically significant neutron or  $\gamma$  ray emissions.

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PACS numbers: 78.60.Mq, 25.45.-z, 28.20.-v, 28.52.-s

**Introduction.**—Previously, we have provided evidence [1(a),2–4] for 2.45 MeV neutron emission and tritium production during external neutron-seeded cavitation experiments with chilled deuterated acetone, and these observations have now been independently confirmed [5]. Even though the major criticisms associated with our previously published results [1(a)] have been thoroughly discussed and resolved [2], there has been a lingering concern about the use of an external neutron source which strongly suggested the need to demonstrate that thermonuclear fusion could also be attained during acoustic cavitation *without* the use of an external source of neutrons. In addition, it was desirable to demonstrate the ability to attain D-D fusion with alternate deuterated liquids and to measure the emitted neutrons with independent detection systems. In the data presented previously [1(a),2] the neutron measurements were obtained using a NE-213 type liquid scintillation fast neutron detector with pulse-shape discrimination [6–8], and the only test liquid used was deuterated acetone. Our simulations of implosion dynamics indicate that heavy water would not be a good choice [1(b),3] for attaining thermonuclear fusion in imploding bubbles.

In this Letter, we report the results of investigations using independent neutron detection systems and the self-nucleation of bubble clusters in various deuterated liquids including water (without using an external neutron source).

**Experimental system.**—The experimental configuration is shown in Fig. 1. The test liquid was placed in a cylindrical glass test section which was driven harmonically with a lead-zirconate-titanate (PZT) piezoelectric driver ring attached to the outside surface of the test section. This induced an acoustic standing wave in the test section with a pressure antinode of maximum amplitude  $\sim 15$  bar. Test-cell operational aspects are described elsewhere

[1(a),2,9]. Four independent nuclear particle detection systems were utilized in the new study. This included use of a long-established passive-type track-edge fast neutron detector (i.e., CR-39<sup>TM</sup> plastic detector from Landauer, Inc.) that is insensitive to  $\gamma$  rays and that is well-known [9–11] to be able to distinguish between neutrons and  $\gamma$  rays, a boron trifluoride ( $\text{BF}_3$ ) thermal neutron detector (TND), a fast rise-time liquid scintillation (LS) detector to ascertain the energy and quantity of any emitted neutrons, and a NaI  $\gamma$  ray spectrum detector.

A unique feature of the present study was the method used for bubble nucleation. In particular, randomly generated (mainly  $\sim 4$  MeV alpha particles) radioactive decay of dissolved natural uranium was used to nucleate bubble clusters. This completely obviates the need to use an external neutron source and resolves any lingering confusion associated with the possible influence of the previ-

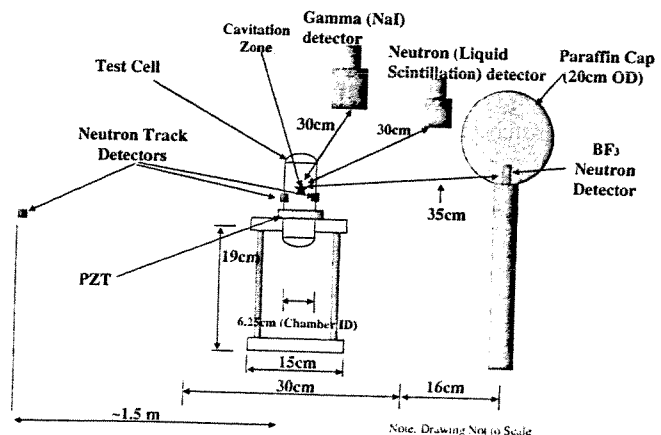


FIG. 1 (color online). Schematic representation of experimental setup.

ously used [1,2] external source neutrons (14.1 MeV) on the emitted neutrons (2.45 MeV).

The test liquids with dissolved alpha emitters were either a mixture of benzene ( $C_6H_6$  or  $C_6D_6$ ), acetone ( $C_3H_6O$  or  $C_3D_6O$ ), and  $C_2Cl_4$  (an organic liquid which has a significantly lower vapor pressure), or water ( $H_2O$  and  $D_2O$ ). The experiments presented herein did not use external neutrons, rather a uranyl nitrate (UN) salt of natural uranium (99.3% U-238) was used to form a solution [9]. The experiments with benzene-based mixtures were conducted at a temperature of  $\sim 7$  C (which is a few degrees above the freezing point of benzene), while the experiments for water were run at  $\sim 5$  C. In contrast with cavitation experiments using fast neutrons and acetone [1(a),2] where we could nucleate  $\sim 30$  bubble clusters per second, in self-nucleation experiments [9] the rate was much lower at  $\sim 1$  per second (for benzene-based mixtures) and around 5 per second for water. It is not clear at the present time why this sort of performance reduction takes place; however, the aim of the present experiments was not to optimize D-D neutron emission but to assess if neutron-gamma emissions were at all possible during bubble implosions with deuterated liquids *without* the use of an external neutron source. See Ref. [9] for additional insights.

**BF<sub>3</sub> detector—experimental observations with various test liquids.**—The BF<sub>3</sub> nuclear particle detector (length = 4.5 cm; diameter = 1.25 cm) was used with a 20 cm diameter paraffin ball moderator over it as shown in Fig. 1.

**Detector calibration.**—The BF<sub>3</sub> detector was calibrated for efficiency of detection using a NIST certified Pu-Be source (emitting  $\sim 2 \times 10^6$  n/s) as well as with 1  $\mu$ Ci Co-60 and Cs-137  $\gamma$  ray sources. Results of pulse height spectra obtained with the Pu-Be source [9] showed a well defined neutron peak above channel 25. At a 10 cm separation the count rate was  $\sim 220$  c/s which, when corrected for solid angle (0.0054), gives an intrinsic efficiency of  $\sim 3\%$  for the detection of fast neutrons in the MeV range. The detector response to  $\gamma$  rays indicated that the majority of pulses would occur [9] below channel 20. The counts collected followed the inverse square law [9], for different separations between the Pu-Be neutron source and the BF<sub>3</sub> detector.

**BF<sub>3</sub> data from self-nucleation experiments with benzene- $C_2Cl_4$ -acetone-UN mixtures.**—Results of control experiments with a  $C_6H_6$ - $C_2Cl_4$ - $C_3H_6O$ -UN mixture indicated [9] that there was no statistically significant net change in counts between cavitation on and off. In contrast, the results of experiments with a deuterated  $C_6D_6$ - $C_2Cl_4$ - $C_3D_6O$ -UN mixture produced a significant increase ( $\sim 400\%$ ) in neutron counts and  $\sim 100\%$  increase of  $\gamma$  ray counts as seen in Fig. 2 and in Table I. Based on the calibrated detector efficiency, the neutron emission rate was estimated to be  $\sim 5$ – $7 \times 10^3$  n/s.

As a cross-check, the distance of the detector from the test cell was roughly doubled from a nominal 35 cm to

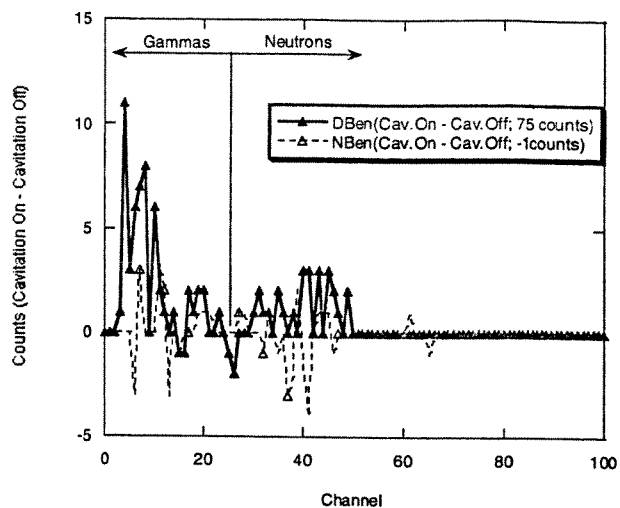


FIG. 2 (color online). Change in counts from Neutron-Gamma Spectra for  $C_6D_6$ - $C_2Cl_4$ - $C_3D_6O$ -UN and  $C_6H_6$ - $C_2Cl_4$ - $C_3H_6O$ -UN with self-nucleation and BF<sub>3</sub> detector (data collected over 3600 s; Cavitation off counts = 49 and 50 counts for deuterated and nondeuterated liquid tests, respectively).

65 cm and the experiments were repeated whereby the increase of neutron counts came down by a factor of about 4, commensurate with the inverse square law dependence with distance.

**BF<sub>3</sub> data from self-nucleation experiments with water.**—Experiments were also conducted by dissolving UN salt in water [9]. Results of experiments with light water ( $H_2O$ ) and heavy water ( $D_2O$ ) are shown in Fig. 3 and the raw data are provided elsewhere [9]. For these experimental parameters there was no statistically significant evidence of nuclear emissions with cavitation, for either  $H_2O$  or  $D_2O$ , which was in accordance with our expectations [1(b),3,9].

**LS detector data—experimental observations with various test liquids.**—The data obtained with the BF<sub>3</sub> detector system were complemented with data taken with a 5 cm dia  $\times$  5 cm LS detector [2] with pulse-shape discrimination between the neutrons and  $\gamma$  rays. Using well-established light curves [6–9], and with the instrument settings used for these experiments, the 2.45 MeV proton-recoil edge corresponded to channel  $\sim 120$ .

**LS data from self-nucleation experiments with benzene- $C_2Cl_4$ -acetone-UN mixture.**—Raw data from control experiments with  $C_6H_6$ - $C_2Cl_4$ - $C_3H_6O$ -UN mixtures and deuterated  $C_6D_6$ - $C_2Cl_4$ - $C_3D_6O$ -UN mixtures are provided elsewhere [9]. Net results are shown in Fig. 4 and Table I, where we see a statistically significant ( $\sim 17$  SD) emission of neutron counts, corresponding to neutron energies of  $\leq 2.45$  MeV.

**Neutron track detector—direct visible evidence for neutron emission.**—The CR-39<sup>TM</sup> neutron track (NT) detector was also calibrated and used [9] in our studies for neutron detection; the technology has been utilized for over 40 years [10,11] as a passive means for directly confirming

TABLE I. Count Statistics (above background).

Figure No.	Cav. On Counts	Cav. Off Counts	Difference (On-Off)	SD in Difference	Diff/SD	Detector	Fluid
2	50	49	-1	10	-0.10	BF <sub>3</sub>	C <sub>6</sub> H <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> H <sub>6</sub> O + UN
2	124	49	75	13.2	5.7	BF <sub>3</sub>	C <sub>6</sub> D <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> D <sub>6</sub> O + UN
3	39	44	-5	9.1	-0.55	BF <sub>3</sub>	H <sub>2</sub> O + UN
3	37	39	-2	8.8	-0.23	BF <sub>3</sub>	D <sub>2</sub> O + UN
4	913	923	-10	43	-0.23	LS	C <sub>6</sub> H <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> H <sub>6</sub> O + UN
4	2015	1055	966	55	17.4	LS	C <sub>6</sub> D <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> D <sub>6</sub> O + UN
5	Ref. [9]	Ref. [9]	Ref. [9]	Ref. [9]	<1	Track	C <sub>6</sub> H <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> H <sub>6</sub> O + UN
5	Ref. [9]	Ref. [9]	Ref. [9]	Ref. [9]	14 (Ref. [9])	Track	C <sub>6</sub> D <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> D <sub>6</sub> O + UN
Ref. [9]	16 804	16 906	-102	182	-0.5	NaI	C <sub>6</sub> H <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> H <sub>6</sub> O + UN
Ref. [9]	16 196	15 844	352	178	2	NaI	C <sub>6</sub> D <sub>6</sub> + C <sub>2</sub> Cl <sub>4</sub> + C <sub>3</sub> D <sub>6</sub> O + UN

and leaving permanent unambiguous evidence for the presence of neutrons. During each experiment three CR-39 NT detectors were placed as shown in Fig. 1. One CR-39 NT detector was placed approximately 1.5 m away from the test cell to measure the background, whereas two others were affixed to the outside of the glass walls of the test cell.

Experiments were conducted with a deuterated (C<sub>6</sub>D<sub>6</sub>-C<sub>2</sub>Cl<sub>4</sub>-C<sub>3</sub>D<sub>6</sub>O-UN) and control liquid C<sub>6</sub>H<sub>6</sub>-C<sub>2</sub>Cl<sub>4</sub>-C<sub>3</sub>H<sub>6</sub>O-UN. Raw data of the experiments are provided elsewhere [9] and the results are shown in Fig. 5. It was verified that no statistically significant change occurs over background levels without cavitation in deuterated liquid. There were no statistically significant neutron tracks compared with the background sample for the control liquid C<sub>6</sub>H<sub>6</sub>-C<sub>2</sub>Cl<sub>4</sub>-C<sub>3</sub>H<sub>6</sub>O-UN. The average number of preexisting tracks on the background and the test-cell attached detectors (etched for 3 hours each) were both around 13. Significantly, close to a 100% increase [and >5 standard deviation (SD)] in visible neutron tracks were observed for

individual samples, only for samples associated with self-nucleation based cavitation experiments in the deuterated liquid C<sub>6</sub>D<sub>6</sub>-C<sub>2</sub>Cl<sub>4</sub>-C<sub>3</sub>D<sub>6</sub>O-UN experiments. On the aggregate, the production of neutron tracks for the deuterated liquid amounted to a ~14 SD change, whereas, for the control liquid the changes were within 0.5 SD. A confirmatory check was also conducted for cavitation experiments with heavy water (D<sub>2</sub>O), where it was found that no significant production of neutron tracks was observed (i.e., the changes were within 1 SD). Overall statistics are summarized in Table I. Based on the calibrated efficiency of detection using CR-39 NT detectors we estimate a neutron emission rate of ~5 × 10<sup>3</sup> to ~10<sup>4</sup> n/s which agrees with the results from our other detectors.

*Gamma ray spectra data.*—Gamma ray spectra were also obtained [9] using a calibrated Harshaw<sup>TM</sup> NaI (5 cm dia × 13 cm) detector to understand better the neu-

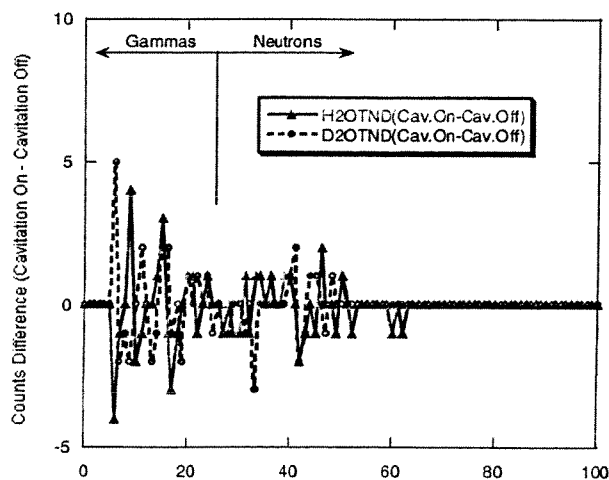


FIG. 3 (color online). Changes in counts from Neutron-Gamma Spectra for D<sub>2</sub>O and H<sub>2</sub>O with self-nucleation and BF<sub>3</sub> detector (counts for cavitation Off/On = 37/39 for D<sub>2</sub>O-UN; = 39/44 for H<sub>2</sub>O-UN).

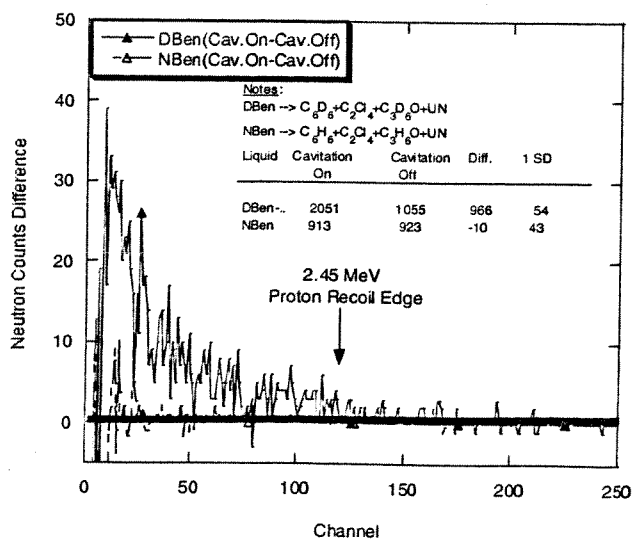


FIG. 4 (color online). Change in counts from pulse height spectra for C<sub>6</sub>D<sub>6</sub>-C<sub>2</sub>Cl<sub>4</sub>-C<sub>3</sub>D<sub>6</sub>O-UN and C<sub>6</sub>H<sub>6</sub>-C<sub>2</sub>Cl<sub>4</sub>-C<sub>3</sub>H<sub>6</sub>O-mixtures with self (alpha recoil nucleation) and LS detector (data taken over 300 seconds).



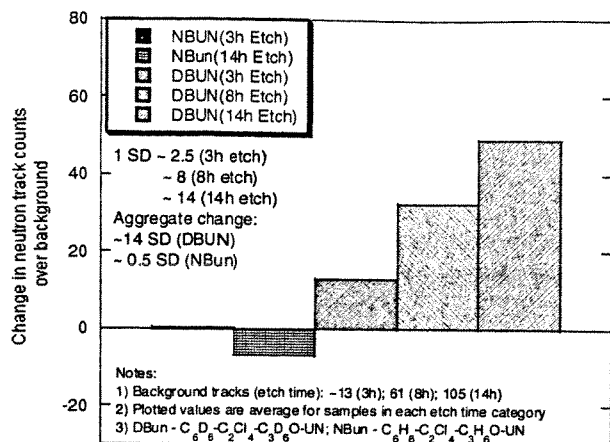


FIG. 5. Variation of neutron tracks for deuterated and non-deuterated test fluids.

tron emission data from the self-nucleation experiments. The results of the control experiments with and without cavitation using  $C_6H_6-C_2Cl_4-C_3H_6O-UN$  indicated [9] no statistically significant change between cavitation on and off. In contrast, the results for the experiments with and without cavitation using  $C_6D_6-C_2Cl_4-C_3D_6O-UN$  showed a noticeable increase in  $\gamma$  ray emissions above background with cavitation on versus off.

*Summary and concluding remarks.*—A novel, new technique was used to develop a stand-alone acoustic inertial confinement nuclear fusion device. Statistically significant emissions of 2.45 MeV neutrons were measured with multiple independent detectors during self-induced cavitation experiments in deuterated benzene-acetone mixtures, but not in corresponding experiments with nondeuterated control liquids or heavy water. The measured neutron emissions ( $\sim 5 \times 10^3$  to  $\sim 10^4$  n/s) obeyed the well-known and expected inverse law dependence with distance. These emission rates are far below break-even levels. As expected, the dissolved emitter did induce cavitation in both  $D_2O$  and  $H_2O$ , but this did not result in statistically significant changes in the measured neutron counts despite far higher bubble cluster nucleation rates.  $\gamma$  ray emissions were also measured during cavitation experiments with chilled  $C_6D_6-C_2Cl_4-C_3D_6O-UN$ . No statistically significant change in neutron or  $\gamma$  ray emissions were observed when there was no cavitation in either deuterated or control liquids.

These data both support and complement the sonofusion data and theoretical predictions that have previously been published [1,2] and clearly demonstrate that thermonuclear conditions can be created during acoustically forced bubble cluster implosions, without the need for an external neutron source.

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## RESEARCH ARTICLES

the lack of a solvent connection. Instead, a proton is taken up from His<sup>Y169</sup> to O<sub>1</sub> and then menaquinol is released to the membrane. (iv) The deprotonated His<sup>Y169</sup> is very unstable, and this is immediately protonated through an extended water channel to His<sup>Y169</sup> after the release of menaquinol.

**Proton motive force generation mechanism in the Fdh-N/Nar system.** The Fdh-N structure, in combination of the Nar topology information, demonstrates that Fdh-N and Nar can form a redox loop where p.m.f. generation is best described as the sum of the following two effects (Fig. 1). (i) Two protons, which are taken up from the cytoplasm at the Fdh-N menaquinone reduction site, are translocated across the membrane and released to the periplasm from the menaquinol oxidation site in Nar. (ii) Two electrons are transferred from the formate oxidation site in periplasm to the NO<sub>3</sub><sup>-</sup> reduction site in cytoplasm. This is not accompanied by an actual proton translocation across the membrane but generates a membrane potential, which is equivalent to 2H<sup>+</sup> translocation across the membrane. The result is consistent with the measured ratio of proton translocation to electron transfer in this system (6).

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## Evidence for Nuclear Emissions During Acoustic Cavitation

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In cavitation experiments with deuterated acetone, tritium decay activity above background levels was detected. In addition, evidence for neutron emission near 2.5 million electron volts was also observed, as would be expected for deuterium-deuterium fusion. Control experiments with normal acetone did not result in tritium activity or neutron emissions. Hydrodynamic shock code simulations supported the observed data and indicated highly compressed, hot (10<sup>6</sup> to 10<sup>7</sup> kelvin) bubble implosion conditions, as required for nuclear fusion reactions.

The intense implosive collapse of gas or vapor bubbles, including acoustically forced cavitation bubbles, can lead to ultrahigh compressions and temperatures and to the generation of light flashes attributed to sonoluminescence (SL) (1–21). Our aim was to study ultrahigh compression and temperatures in bubbles nucleated by means of fast neutrons, whereby bubble nucleation centers with an initial radius  $R_0$  of ~10 to 100 nm are created, and the bubbles grow in an acoustic field to a maximum radius ( $R_m$ ) of ~1 mm (19) before an implosive collapse. This approach builds on the observations that (17) increasing  $R_m$  modestly (for example, by ~50%), or increasing the rate of collapse (16), can result in very large increases in peak gas temperatures and produce light emission during implosions. In contrast to single-bubble SL experiments, in which the initial bubble radius  $R_0$  typically increases to  $R_m$  by a factor of only ~10 (for example, from ~10 μm to ~100 μm), our neutron-induced nucleation technique results in  $R_m/R_0$  of ~10<sup>5</sup>. For a spherical bubble, the increase of  $R_m/R_0$  by a factor of 10<sup>4</sup> implies a related volumetric ratio increase of 10<sup>12</sup> over that produced by conventional techniques. Our expectation was that such an approach, with its vastly increased energy concentration potential during implosions, should give rise to significant

increases in the peak temperatures within the imploding bubbles, possibly leading to fusion and detectable levels of nuclear particle emissions in suitable fluids.

To minimize the effect of gas cushioning by promoting rapid condensation during implosive collapse, we elected to work with highly degassed organic liquids. An organic liquid was chosen [normal acetone (C<sub>3</sub>H<sub>6</sub>O) as the control fluid and deuterated acetone (C<sub>3</sub>D<sub>6</sub>O) as the test fluid] because it permitted the attainment of large tensile states without premature cavitation, and thus a lot of liquid superheat would be present before nucleation. Organic liquids also have relatively large phase change coefficients, which is important, as described later. Unless otherwise noted, the liquid in the chamber was maintained at ~0°C (which was the lowest value obtainable with the equipment we used). The test liquid was degassed and subjected to an acoustic pressure field that oscillated in resonance with the liquid sample and its container. The nucleation of vapor bubbles was initiated with fast neutrons from an isotopic source (Pu-Be) or from a pulsed neutron generator (PNG) that produces 14-MeV neutrons on demand at a predefined phase of the acoustic pressure field.

**Experimental system.** In the experimental apparatus (Fig. 1), the test liquid was placed in an approximately cylindrical glass flask and driven acoustically with a lead-zirconate-titanate (PZT) piezoelectric driver ring attached to the outer surface. Either a plastic or a liquid scintillation detector was used for detection of neutron and gamma signals (22). The light was detected and amplified in a photomultiplier tube (PMT). A

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†Retired.

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liquid scintillator (LS) detector-based system was set up for pulse-shape discrimination (PSD) (23, 24) [see Web Supplement 1 (25)]. The PSD circuit separates neutrons from gamma rays on the basis of differences in the PSD scintillator signal decay time between neutrons and gamma rays. The system could be operated to permit blocking of gamma rays (hereafter, a mode of operation referred to as “with PSD”). The net efficiency for fast neutron detection was estimated to be  $\sim 5 \times 10^{-3}$  (26).

In the experimental sequence of events (Fig. 2), neutrons from the PNG nucleated vapor bubbles in the tensioned liquid when the cavitation threshold was exceeded at the time of the neutron burst. The nuclear radiation detector typically detected a pulse when the PNG was fired. Thereafter, the vapor bubbles grew until increasing pressure in the liquid during the second half of the acoustic cycle caused them to begin to collapse. If the implosion was robust enough, the bubble emitted a SL light flash, which could be detected by the PMT. In theory, if the liquid is composed of deuterium (D) and/or tritium (T) atoms, and the conditions are appropriate for D-D (or D-T) fusion, nuclear particles (neutrons and gamma rays) would be emitted and seen in the response from either the plastic scintillator (PS) or LS detector. Moreover, in cavitation experiments, when a bubble implodes, a pressure wave that travels at about the speed of sound in the test liquid is also generated and can be detected at the chamber walls by microphones. Significantly, our experiments were characterized by bubble dynamics within clouds of many bubbles.

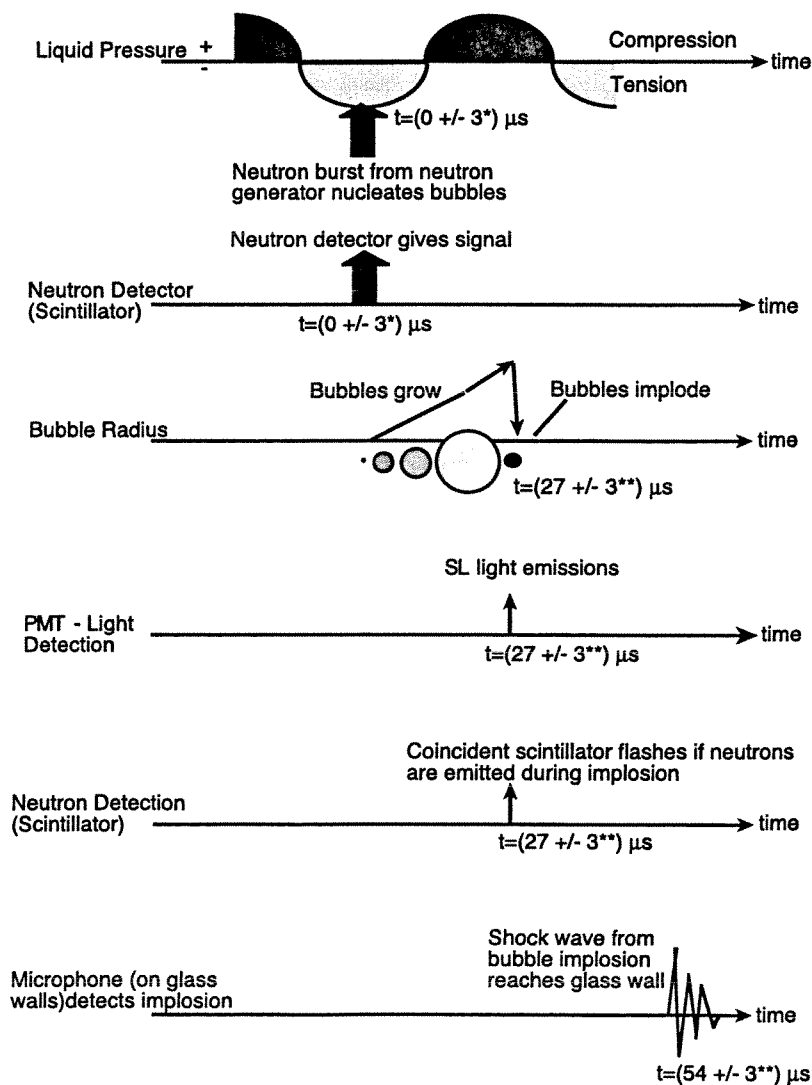
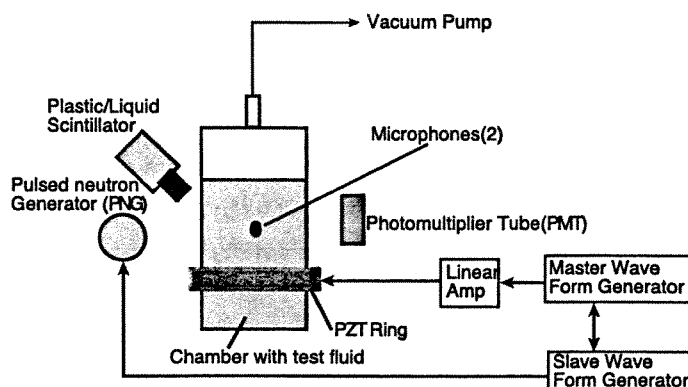
**Timing of key parameters.** With our configuration of the PNG and electronic timing systems, we found, by analyzing the time spectrum of neutrons (25), that neutrons were emitted over a time span of  $\sim 12 \mu\text{s}$  [ $\sim 4$  to  $6 \mu\text{s}$  at full width at half maximum (FWHM)], after which neutron counts were reduced considerably by  $15$  to  $20 \mu\text{s}$  after the PNG fired (25). We initiated the PNG burst when the fluid tension state was greatest. For multiple-bubble implosions, several bubbles can implode and emit closely spaced SL flashes during any given cycle.

The time between a SL flash and the signals received at two microphones set up on diametrically opposite sides of our chamber was found to be  $\sim 27 \mu\text{s}$ , which is in agreement with the time for a shock wave to travel from the center of the chamber to the glass wall (about  $32 \text{ mm}$  away). This result indicates that the bubbles generally nucleated and imploded in or around the central axis of the test chamber (27). The efficiency of SL flash detection was dependent on the PMT bias voltage (which determines the gain) and the chosen discriminator settings (29).

Our data were obtained with a PZT drive amplitude much greater than that required for threshold nucleation. Because of this, and because the PNG pulse width was about  $4$  to  $6 \mu\text{s}$  (FWHM), nucleation could occur a few microseconds before or after the minimum liquid pressure was reached. The timing of

the SL flash relative to the PS pulse was analyzed with a multichannel analyzer (MCA). The PZT drive frequency was about  $19.3 \text{ kHz}$ , which corresponds to a full cycle time of about  $52 \mu\text{s}$ . The time spectrum of events (25) confirmed that the PS flash corresponding to the PNG activation (lasting

**Fig. 1.** Schematic of the experimental setup. The distance from the scintillator head to the PNG is  $\sim 15 \text{ cm}$ ; from the scintillator head to the chamber surface,  $\sim 0$  to  $2 \text{ cm}$ ; from the chamber center to the PNG,  $\sim 20 \text{ cm}$ ; and from the PMT to the chamber surface,  $\sim 5 \text{ cm}$ . The system (the chamber, PNG, and PMT) is  $\sim 1.5 \text{ m}$  above the floor.



(\*) - Full-width at half-maximum

(\*\*) - Can continue for several cycles (to  $5 \text{ ms}$ )

**Fig. 2.** Experimental sequence of events.

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about 12  $\mu\text{s}$ , with 4 to 6  $\mu\text{s}$  FWHM) was followed by a SL flash (lasting about 4 to 6  $\mu\text{s}$  FWHM) about 27 to 30  $\mu\text{s}$  later.

**Experimental observations for  $\text{C}_3\text{H}_6\text{O}$  and  $\text{C}_3\text{D}_6\text{O}$ .** We conducted experiments with  $\text{C}_3\text{H}_6\text{O}$  (100% nominally pure) and  $\text{C}_3\text{D}_6\text{O}$  (certified 99.92 atom % D-acetone), filtered before use through 1- $\mu\text{m}$  filters. Degassing was performed by applying a low pressure of about 10 kPa and acoustically cavitating the liquid for about 2 hours. To ensure continued robust nucleation growth and implosive collapse, the drive voltage to the PZT was set to be about double that needed for occasional cavitation (defined here as the occurrence of nucleation and collapse within a 10-s observation period). The negative pressure threshold for bubble nucleation by neutrons and alpha particles in acetone is  $-7$  to  $-8$  bar (20, 21). A pressure map of the chamber was obtained by means of a calibrated hydrophone. Using the scale factor for induced pressures in our chambers versus drive voltage to the PZTs, and gradually increasing the drive amplitude, we determined that the cavitation began at about  $-7$  bar, which is consistent with the known value (20, 21). The pressure amplitude in our chamber was  $\sim \pm 15$  bar ( $\pm 220$  pounds per square inch).

**T detection, monitoring, and estimation.** The D-D fusion reaction can have one of two outcomes that occur with almost equal probability. The first leads to the production of helium (He) and 2.5-MeV neutrons; the second to the production of T and protons. Therefore, in addition to the evidence collected for neutron or gamma ray activity, the formation of T would provide compelling evidence of D-D fusion.

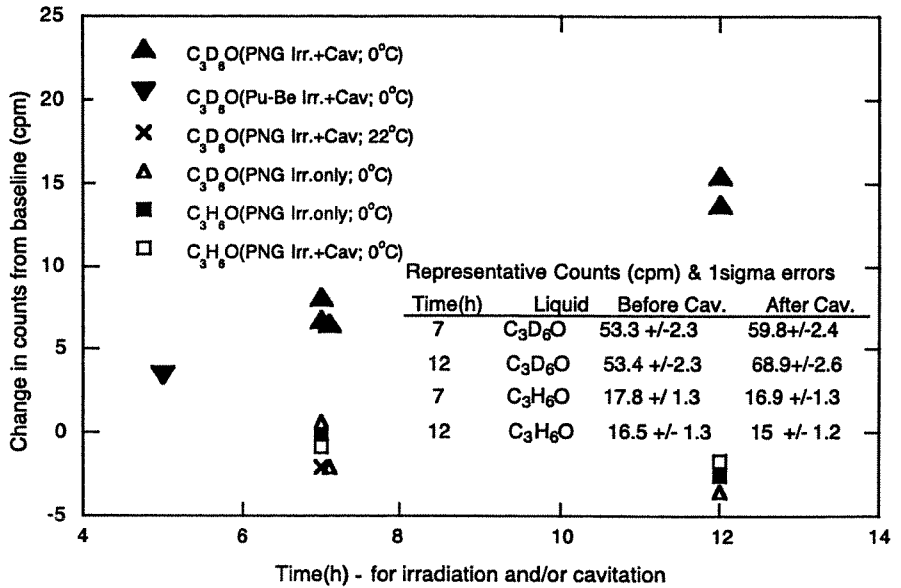
To measure T activity, we sampled the experimental fluid directly with a scintillation counter calibrated for detecting T (28). Unless otherwise noted, experiments were conducted at  $\sim 0^\circ\text{C}$  using 14-MeV neutrons generated at a rate of  $\sim 10^6$  neutrons/s from the PNG spread over the specified time duration. When testing either  $\text{C}_3\text{H}_6\text{O}$  or  $\text{C}_3\text{D}_6\text{O}$  without cavitation (that is, with irradiation alone), we also used the same experimental configuration, including placing the chamber under standard vacuum conditions. In this way, we systematically conducted experiments, changing only one parameter at a time. The chamber was initially filled with  $\text{C}_3\text{H}_6\text{O}$  and irradiated for 7 hours without cavitation. A 1- $\text{cm}^3$  liquid sample was withdrawn from fluid in the top region in the acoustic chamber and mixed with Ecolite and tested for T activity. Thereafter, cavitation experiments were performed for 7 hours. A 1- $\text{cm}^3$  sample of  $\text{C}_3\text{H}_6\text{O}$  was again withdrawn and tested for T activity. This same process was later repeated for 12 hours.

After verifying the absence of T activity from the control tests with  $\text{C}_3\text{H}_6\text{O}$ , we repeated the experiments with  $\text{C}_3\text{D}_6\text{O}$ . The irradiation and cavitation experiments of 7 hours duration with  $\text{C}_3\text{H}_6\text{O}$  and  $\text{C}_3\text{D}_6\text{O}$  were repeated several times at  $\sim 0^\circ\text{C}$ . A separate test was conducted over 5 hours, using a Pu-Be neutron source producing  $\sim 10^6$  neutrons/s, to assess the influence of randomly produced neutrons.

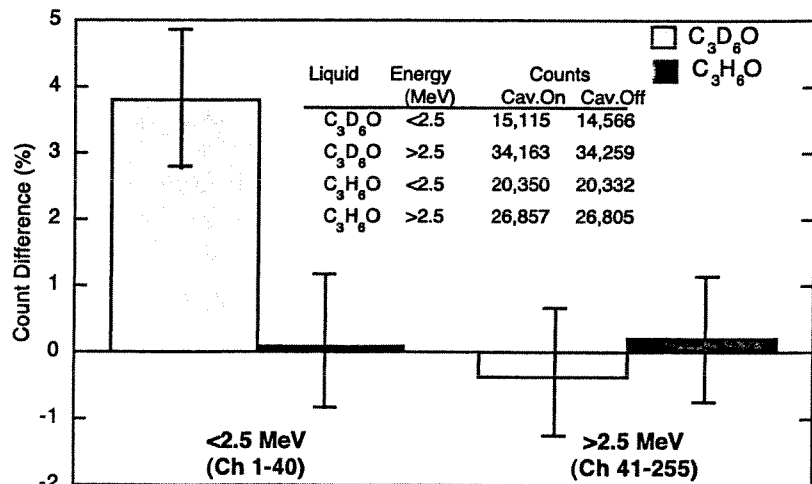
Tests were also conducted to assess the impact of liquid temperature on T activity buildup by testing with  $\text{C}_3\text{D}_6\text{O}$  at  $\sim 22^\circ\text{C}$  (room temperature). Finally, to assess the impact of the time of irradiation and cavitation on T buildup in  $\text{C}_3\text{D}_6\text{O}$ , testing was also conducted for 12 hours with PNG ir-

radiation of  $\sim 10^6$  neutrons/s at  $\sim 0^\circ\text{C}$ .

Results of these experiments are summarized in Fig. 3, which includes values of standard deviation as well as background count rates. The data reveal no significant change in T activity for  $\text{C}_3\text{H}_6\text{O}$  with or without cavitation and irradiation. Under the same experimental conditions, irradiation alone of  $\text{C}_3\text{D}_6\text{O}$  samples with 14-MeV neutrons, or with neutrons from a Pu-Be source, did not result in any statistically significant change in T content. In contrast, in three separate 7-hour cavitation experiments with  $\text{C}_3\text{D}_6\text{O}$  at  $\sim 0^\circ\text{C}$  and  $\sim 10^6$  neutrons/s irradiation with the PNG, an average increase of 7.1 counts per minute (cpm) resulted. Similarly, two separate 12-hour



**Fig. 3.** T activity changes for  $\text{C}_3\text{D}_6\text{O}$  and  $\text{C}_3\text{H}_6\text{O}$  with irradiation (Irr.) and with irradiation plus cavitation (at  $0^\circ\text{C}$ ). For  $\text{C}_3\text{D}_6\text{O}$  and  $\text{C}_3\text{H}_6\text{O}$ , 1 SD =  $\sim 3.5$  cpm and  $\sim 2$  cpm. PNG irradiation was at  $\sim 10^6$  neutrons/s for specified time durations. All testing was conducted under the same configuration, placing the test cell under a vacuum of  $\sim 10$  kPa.



**Fig. 4.** Changes in counts for  $\text{C}_3\text{D}_6\text{O}$  and  $\text{C}_3\text{H}_6\text{O}$  with and without cavitation (Cav.). Error bars are 1 SD; data were taken over 100 s; and the percentage change in the  $<2.5$ -MeV range was found to be similar in magnitude for 300-s data acquisition times.

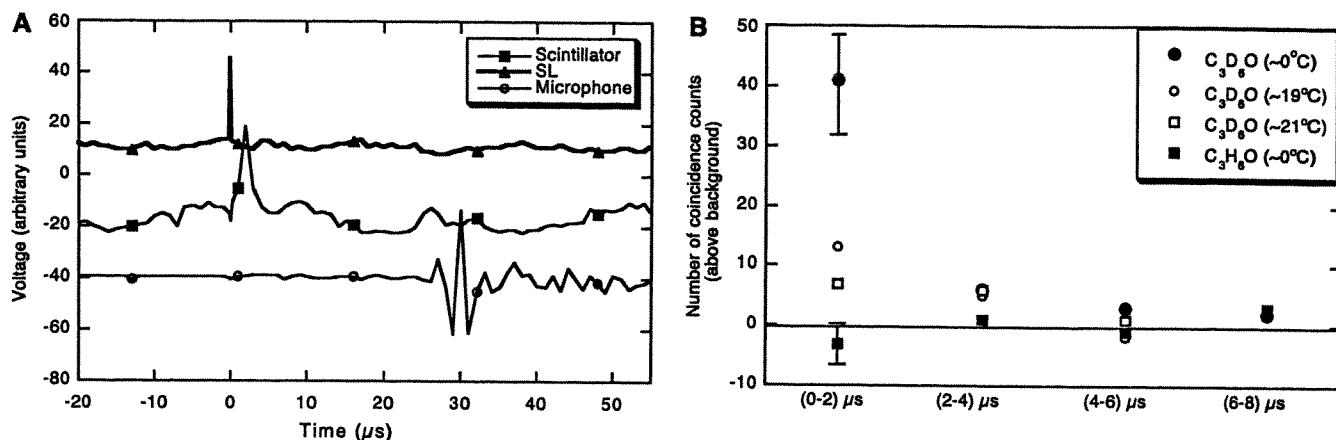
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experiments produced an average increase of 14.6 cpm, the increase being directly proportional, within statistical counting errors, to the increase in duration of the test. Overall, cavitation of  $C_3D_6O$  (at  $\sim 0^\circ C$  with  $\sim 10^6$  neutrons/s PNG irradiation) over 7 hours resulted in increases of up to  $\sim 8.1$  cpm [representing an individual difference of  $\sim 2.5$  standard deviations (SD) and a

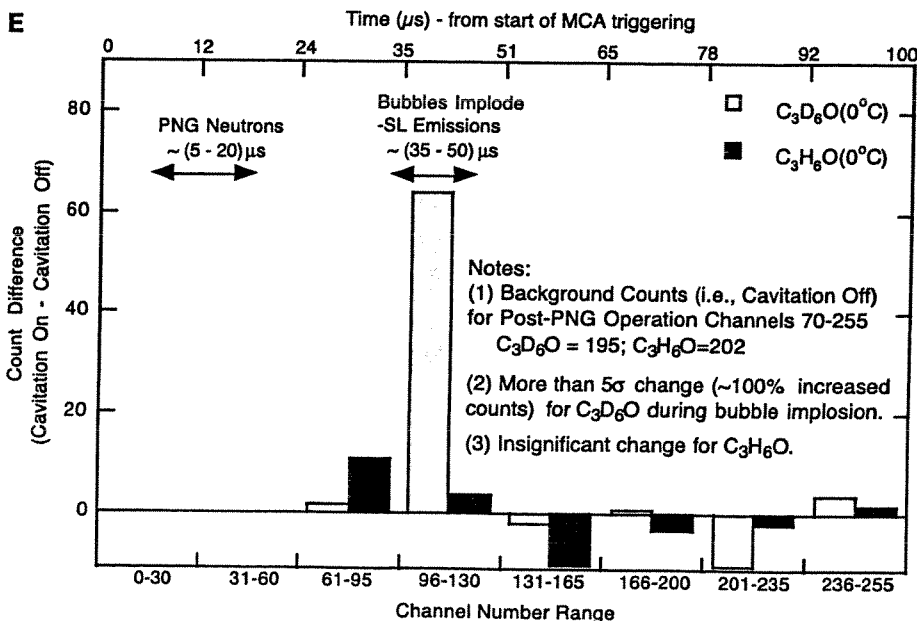
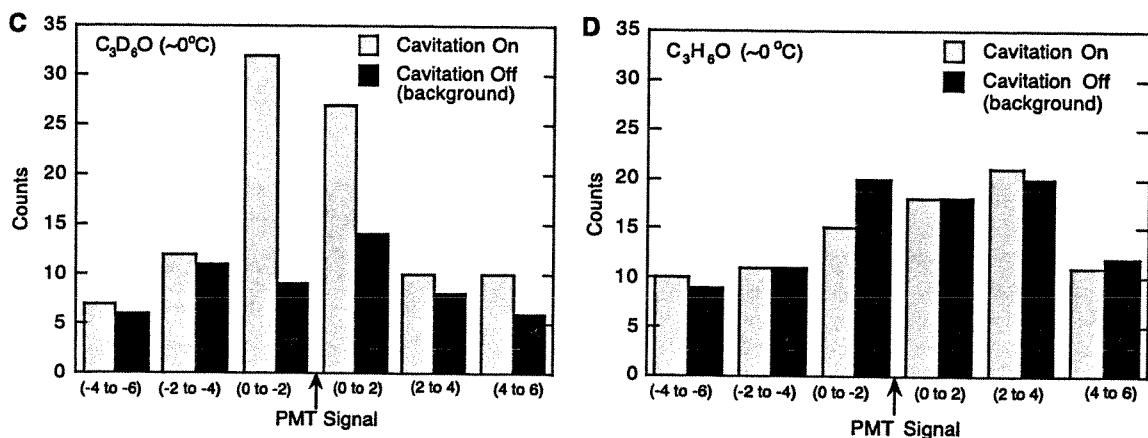
collective change of  $\sim 4$  SD], whereas cavitation and irradiation over 12 hours resulted in an average increase of  $\sim 14$  cpm (representing an individual difference of more than 4.5 SD). Finally, cavitation of  $C_3D_6O$  (at  $\sim 22^\circ C$  with  $\sim 10^6$  neutrons/s PNG irradiation) over 7 hours did not result in any significant change in T activity. As will be described later, this agrees with the

lack of SL activity at this higher temperature and is also consistent with the hydrodynamic shock code simulations to be discussed subsequently.

If none of the T produced reacted with D atoms, an inverse calculation based on the observed T activity indicates that the D-D neutron production rate was  $\sim 7 \times 10^5$  neutrons/s.



**Fig. 5.** (A) Representative time variation of PMT (SL) flashes, the scintillator nuclear signal, and microphone shock trace signals ( $C_3D_6O$  cavitation experiments at  $0^\circ C$ ). (B) Coincidence data for  $C_3D_6O$  and  $C_3H_6O$  (mode 2 operation; PNG operation at  $\sim 10^6$  neutrons/s at 200 Hz). (C) Coincidence spectrum data for  $C_3D_6O$  (mode 2 operation; PNG operation at  $\sim 10^6$  neutrons/s at 200 Hz). (D) Coincidence spectrum data for  $C_3H_6O$  (mode 2 operation; PNG operation at  $\sim 10^6$  neutrons/s at 200 Hz). (E) Time spectrum of nuclear emissions for deuterated and natural acetone [see Web Supplement 1 (25) for further details].



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**Neutron energy spectra data acquisition with PSD.** We used PSD in experiments with and without cavitation to check for neutron production and to decipher the energy range in which significant increases in neutron counts occurred. Tests were conducted with the PSD system so that only neutron counts were accepted by the data acquisition system. For identical settings, it was verified that the PNG neutron output varied by  $\sim \pm 0.2\%$  from measurement to measurement. Tests, with a PNG neutron output of  $\sim 10^6$  neutrons/s, were conducted with  $C_3H_6O$  (as the control fluid) and with  $C_3D_6O$  for data acquisition times from 100 to 300 s, during which neutron counts were accumulated at the rate of  $\sim 500$  counts/s. Changes in counts from the case without cavitation were evaluated in two energy ranges. The first covered the range between the lowest energy detectable and 2.5 MeV. The second was from 2.5 MeV upward (26). Representative results for the sample with  $C_3D_6O$  (that is, the increase in counts and the background counts) are shown in Fig. 4 for observations over 100 s in the two energy ranges. As shown, we observed a 4% increase in 2.5-MeV neutrons in these samples after the onset of cavitation. The variation in counts for the control liquid ( $C_3H_6O$ ) was within  $\sim 0.2\%$  in both energy ranges. These data were repeatable for 300-s acquisition times.

Assuming Poisson statistics, 1 SD from a total population count varying from 50,000 to 150,000 ranges from  $\sim 0.4$  to 0.25%. Therefore, a  $\sim 4\%$  change in counts for a case with cavitation is a significant increase of  $\sim 4$  SD above background. Because the data were repeatable, when taken as an aggregate, the average  $\sim 4\%$  increase in counts represents a statistically significant change of  $> 10$  SD above background. For a  $\sim 4\%$  increase in the case of  $C_3D_6O$  with cavitation (in the 2.5-MeV range), and using a distance-corrected detector efficiency (26), we estimate that the D-D neutron emission rate associated with cavitation was  $\sim 4 \times 10^4$  to  $8 \times 10^4$  neutrons/s. This value is somewhat smaller than the estimated rate of neutron generation from the T measurements ( $\sim 7 \times 10^5$  neutrons/s). At least part of this difference can be attrib-

uted to uncertainties in detector efficiency, such as neutron energy losses by scattering in the test chamber or reduced detection efficiency for large-angle knock-ons from 2.5-MeV neutrons (25). There could also have been a nonuniform T distribution in the chamber after the cavitation experiments.

**Coincidence data acquisition.** We also looked at the coincidence between the SL and PS/LS pulses. Coincidence spectra were obtained by direct visual observations and manual recording of the individual coincidence signals on a digital storage oscilloscope triggered by the SL signal. Two different data acquisition modes (29) were tested. For mode 1 operation, at a low bias voltage for the PMT, it was conclusively determined that no false SL activity occurred during PNG operation, and that coincidence between SL and scintillator signals repeatedly took place for cavitating  $C_3D_6O$  at  $\sim 0^\circ C$  but not for the control liquid. An example of data traces for tests with  $C_3D_6O$  showing coincidence between the SL flash and the scintillator pulse, and the subsequent microphone response, is shown in Fig. 5A. No such coincidence, followed by microphone traces, was seen for tests with the control liquid  $C_3H_6O$ . However, data acquisition with this mode of operation was slow, because many genuine SL signals were rejected as well. In the second mode of operation (mode 2), the bias voltage to the PMT was increased, resulting in some spurious SL signals generated during PNG operation (25, 29). However, this effect was easily accommodated by taking data with and without cavitation, leaving all other parameters the same, and then subtracting the coincidence data taken without cavitation from those taken with cavitation. Figure 5B displays typical coincidence data spectra taken in mode 2 of operation with  $C_3D_6O$  and  $C_3H_6O$  (30). Figure 5, C and D, display the actual counts with cavitation and background counts with PNG operation only. Data gathered in this mode were binned in 2- $\mu s$  bins before and after the PMT signal. The experiments were repeated several times, with sim-

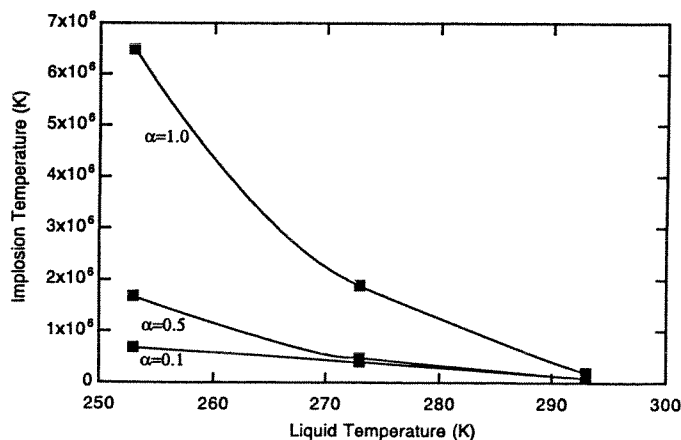
ilar results. Coincidence data taken for each time bin were obtained with and without cavitation (keeping the PNG on in both cases). Counts taken without cavitation (that is, those that occurred during PNG operation, which were all random or false) were subtracted from counts taken with cavitation over the same recording interval. Only for tests with  $C_3D_6O$  at  $\sim 0^\circ C$ , a sharp peak in net coincidences occurred in the 0- to 2- $\mu s$  interval on either side of the PMT (SL) signal, after which the coincidence events tapered off to within 1 SD (29). No such peaking of coincidences was seen for the control fluid ( $C_3H_6O$ ). Similar data for  $C_3D_6O$  were obtained at  $\sim 19^\circ$  and  $\sim 21^\circ C$ . At these temperatures, there was more evaporation and less condensation; as a result, the bubble collapse was less intense, and the number of coincidences was sharply reduced, as seen in Fig. 5B. The influence of random coincidences between SL and scintillator flash signals in the region of bubble collapse was estimated to be insignificant (30).

Results obtained with  $C_3D_6O$ , using MCA time spectrum data (Fig. 5E) and two different modes of data acquisition, show that PS or LS signals detecting penetrating nuclear radiation were coincident with the PMT signals detecting SL light emission during bubble implosion. Such data were not seen in the control tests with  $C_3H_6O$ .

Measurement of 2.5-MeV neutrons in this environment is difficult because of the background of 14-MeV pulsed neutrons and associated gamma rays from the PNG. Independent attempts to reproduce the neutron data using a different detection system and electronics yielded smaller neutron emission and no evidence for real coincidences between SL and neutron events (31). Indeed, in this independent experiment, the analysis of the singles count rates indicated that the coincidences observed may be random in nature. Additional analysis was done (32), but further measurements are required to understand the difference between these two sets of observations.

**The analysis of bubble implosion.** To obtain an estimate of the implosive collapse conditions and to help understand the observed experimental data trends, we developed a one-dimensional hydrodynamic shock (HYDRO) code [see Web Supplement 2 (25)] to numerically evaluate the conservation equations of each phase during bubble growth and collapse. This code includes the Mie-Gruniesen equations of state (33) and Born-Mayer potential functions (33), which are known to be valid for highly compressed fluids (34). In particular, for acetone these equations of state are based on the shock wave adiabat data of Trunin *et al.* (35), and they implicitly specify the effect of the induced radiation field and the dissociation and

**Fig. 6.** Predicted variation of implosion gas temperatures with liquid pool temperature and phase change coefficient. Implosion gas temperatures are quoted for radial locations close to, but not at, the center of the bubble and correspond to the location where neutron production is a maximum [Web supplement 2 (25)]. The gas temperatures at the centerline can be much higher.





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ionization processes that take place during plasma formation within imploding bubbles. Moreover, relevant energy losses and the effect of both molecular and electron/ion conductivity were taken into account, and the resultant HYDRO code [see Web Supplement 2 (25)] allowed for the evaluation of shock wave interaction using the well-established Godunov numerical technique.

Bubble dynamics were studied in  $C_3D_6O$  for conditions typical of those in our experiments. It was found that highly compressed conditions suitable for thermonuclear fusion were predicted, and, as can be seen in Fig. 6, the results were sensitive to the values of the phase change (that is, accommodation) coefficient,  $\alpha$ , and the liquid pool temperature  $T_0$ . The reason why there is a strong sensitivity to  $T_0$  is that at low temperatures there is less evaporation and more condensation of the vapor during bubble expansion and compression, respectively, which in turn reduces the cushioning effect of the compressed vapor during implosions. Similarly, larger values of  $\alpha$  yield more condensation and thus more intense vapor compression. Interestingly, it is difficult to expose water (as opposed to organic fluids such as acetone) to large underpressures without having premature cavitation. Moreover,  $D_2O$  has a relatively low value (36–38) of  $\alpha$  ( $\sim 0.075$ ) as compared to  $C_3D_6O$ , for which  $\alpha \sim 1.0$  (39). Thus,  $C_3D_6O$  appears to be a better test fluid than heavy water.

To obtain an estimate of the D-D fusion neutron production rate, we may evaluate the fusion neutron kinetics equations and fusion cross sections (40, 41) over a range of uncertain parameters to arrive at reasonable estimates for the neutron production rate, varying from  $\sim 10^{-2}$  to 10 neutrons per implosion [see Web supplement 2 (25)]. Direct photographic evidence of the bubble clusters suggests that there were about 1000 bubbles in each bubble cluster in our experiments. Since up to 50 implosions/s were observed during our experiments, the HYDRO code (25, 42) predictions yielded neutron production rates ranging from about  $10^3$  to  $10^6$  neutrons/s, which is qualitatively consistent with the estimates from the T production rate, and the fusion neutrons measured in our experiments.

Many modeling assumptions were necessarily made in the HYDRO code, such as the equations of state, the use of an effective temperature to approximate the behavior of the electrons and ions in the plasma [Web supplement 2 (25)], the relevant energy losses, and various mechanisms for shock wave intensification (43, 44). In particular, a roughly 10-fold increase in the external driving pressure was used in the calculations (25, 42) to appropriately account for the effect of pressure intensification within the imploding bubble clusters. More realistic plasma phys-

ics, nuclear physics, and thermal-hydraulic models can and should be developed for future applications. Nevertheless, the predicted trends and basic physical phenomena that have been modeled agree with our experimental observations.

The observation of statistically significant T activity increases only in chilled ( $\sim 0^\circ C$ ) cavitated  $C_3D_6O$ , coupled with evidence for neutron emissions in chilled cavitated  $C_3D_6O$ , and the absence of neutron emissions and T production in irradiated control tests with  $C_3H_6O$ , complemented by confirmatory modeling and HYDRO code simulations, suggest the possibility of D-D fusion during acoustic cavitation experiments with  $C_3D_6O$  (45).

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22. The Bicon BC404 PS has dimensions of 5 cm by 2.5 cm; the Elscint LS has dimensions of 5 cm by 5 cm. The test cell (a Pyrex flask  $\sim 65$  mm in diameter and  $\sim 200$  cm high, with a PZT driver) was designed, fabricated, and set up at Oak Ridge National Laboratory (ORNL) for the experiments.
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26. The 2.5-MeV proton recoil edge was determined by using cobalt-60 and cesium-137 monoenergetic gamma ray sources (24). For a 255-channel energy scale, the 2.5-MeV threshold was found to lie around channel 40, and the 14-MeV shoulder around channel 110. The efficiency of detection was determined to be  $\sim 5 \times 10^{-3}$  for a  $2 \times 10^6$  neutron/s Pu-Be isotope source positioned near the face of the LS detector. This efficiency, corrected for the actual distance ( $\sim 5$  to 7 cm) of the LS detector from the cavitation region, gives an estimated efficiency of  $\sim 1$  to  $2 \times 10^{-4}$ .
27. Microphones were attached to the outside of the flask on diametrically opposite sides. The frequency of bubble-burst generation varied from  $\sim 15$  to 35 or more bursts/s, depending on the state of tuning. Shock traces from individual bubble bursts lasted for  $\sim 5$  ms.
28. A Beckman LS6500 scintillation counter, calibrated to detect 5- to 18-keV beta ray decays from T, was used.
29. SL detection with the PMT (a Hamamatsu R212 with 2-ns rise time) varied strongly with the bias voltage. At  $-300$  V, about one SL flash every 10 s was detected, whereas about one to five SL flashes/s were detected at a  $-450$ -V bias. The PNG operation caused pseudo SL flashes to occur during the time of neutron production only with  $-450$ -V bias voltage; the time spectrum (25) of SL flashes indicated that about 30% of the recorded SL flashes occurred during PNG operation.
30. The acoustic chamber was a high-Q system and as such required continuous tuning for optimal performance and bubble implosion occurrence. Time to obtain 100 coincidence data points as shown in Fig. 5B averaged about 30 min. Standard deviation was computed by taking the square root of the sum of the counts in each time bin. From the MCA time spectrum, we calculate an instantaneous rate of  $\sim 1$  to 50 neutrons and gamma rays per second during the time of bubble implosion and SL light emission. For a 20- $\mu$ s time window, and a rate of about one SL flash/s for a coincidence gathering time of  $\sim 1600$  s, the number of random coincidences was calculated to be negligible [ $(20 \times 10^{-6}) \times (1 \text{ to } 50) \times 1 \times 1600 \sim (0.03 \text{ to } 1.6)$ ]. In mode 2 operation, data were obtained on a two-channel 500-MHz scope. Simultaneous time spectra data were not possible to obtain for SL and scintillator signals. These were obtained separately with an MCA (25) under identical mode 2 coincidence experimental conditions and revealed insignificant deviation from run to run. These data were then used to estimate random coincidences. It was determined that coincidences occurring during the time of PNG operation would all be random. However, as discussed earlier in this note, the random coincidences during bubble implosion appear to be insignificant.
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