TRITIUM MEASUREMENTS IN NEUTRON-INDUCED CAVITATION OF DEUTERATED ACETONE

RADIATION MEASUREMENTS AND INSTRUMENTATION

TECHNICAL NOTE

KEYWORDS: *deuterated acetone, acoustic cavitation, tritium measurements*

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Received February 28, 2006
Accepted for Publication April 11, 2006

An attempt to reproduce the tritium measurements in an acoustic cavitation experiment with deuteratad acetone has shown no evidence of tritium production attributed to D-D fusion. The average number of disintegrations per minute observed is within 1σ of zero.

I. INTRODUCTION

Evidence of statistically significant tritium activity and neutron and gamma-ray emission in cavitation experiments using chilled deuterated acetone have been reported by Taleyarkhan et al.[1] Another research group at Oak Ridge National Laboratory performed the neutron emission measurements with the same experimental setup and reported a negative result.[2] An experiment to reproduce the tritium measurements of Taleyarkhan et al. was carried out at Purdue University between 2002 and 2004.

II. EXPERIMENT

An acoustic chamber was built following the design of West.[3] The chamber was filled with deuterated acetone, degassed, and then cavitated for 7 h at a temperature of 0°C corresponding to the acetone vapor pressure of ~9.4 kPa. The cavitations were initiated using either a 10-Ci Am-Be source or a 1-Ci Pu-Be source. A block diagram of the system is shown in Fig. 1. The cylindrical chamber wall was oscillated by applying an alternating current to the surrounding piezoelectric (PZT). A sine wave of a specific frequency and amplitude was sent from the waveform generator (Agilent Technologies 33120A) to the linear acoustic amplifier (Bogen Gold Seal Series GS3 250) through a series decade inductor (General Radio Company) to drive the PZT. The coils of the decade inductor (ranging from 0.1 to 10 mH) were adjusted to optimize the electrical resonance of the system. A microphone was mounted to the side of the glass chamber. The driving voltage and microphone output were then monitored on an oscilloscope. Continual adjustments were made to the frequency and/or inductor to maximize the microphone output (i.e., the chamber acoustic resonance). Resonant frequencies ranged from 16 to 22 kHz.

Figures 2 and 3 show typical measurements made during the preliminary testing with a hydrophone (Piezotronics 482A21). The voltage output of the hydrophone (measured in millivolts, peak to peak) is proportional to the pressure within the chamber. The measurements were performed to ensure linear dependence between applied power and acoustic pressure in an open chamber. The proper axial and radial pressure profiles were also acquired to ensure acoustic operation at the first resonance mode.

Profile pressure measurements shown in Fig. 2 demonstrate the chamber's ability to operate effectively in the first harmonic resonance. Profiles showing sidebands or distortions were signs that the chamber was off-resonance. Measurements of the chamber's pressure response shown in Fig. 3 indicate that the pressure output is linear with respect to the PZT voltage input. Degassing was performed driving the chamber with the PZT in the presence of seed neutrons and letting the dissolved gases rise to the surface. The atmospheric air was eliminated using the vacuum pump. As the degassing proceeded, the gas stopped bubbling and the pressure in the chamber could be increased gradually to the operating condition (i.e., ±15 bars acoustic pressure in the center). Once the acetone was degassed, neutron induced bubble nucleation onset occurred in the range of ±7 to 8 bars. At that point, the microphone began to capture the shocks, which are caused by bubble collapses. Then, the operating acoustic pressure was achieved by doubling the voltage of the amplifier. A digital oscilloscope was used to monitor pulses from a PZT microphone with a high pass filter placed at the microphone output. Upon completion of degassing, the pulses were <5 ms in width. The cavitation rate at ±15 bars ranged from 25 cavitations/s to 60 cavitations/s. On average, the cavitation rate was 30 cavitations/s. The

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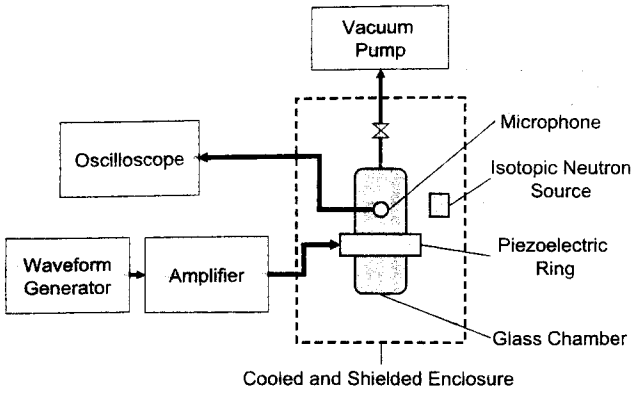

Fig. 1. Block diagram of experiment.

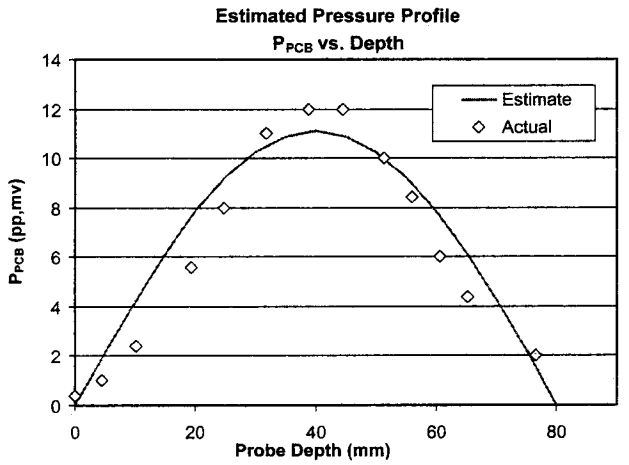

Fig. 2. Axial measured pressure profile.

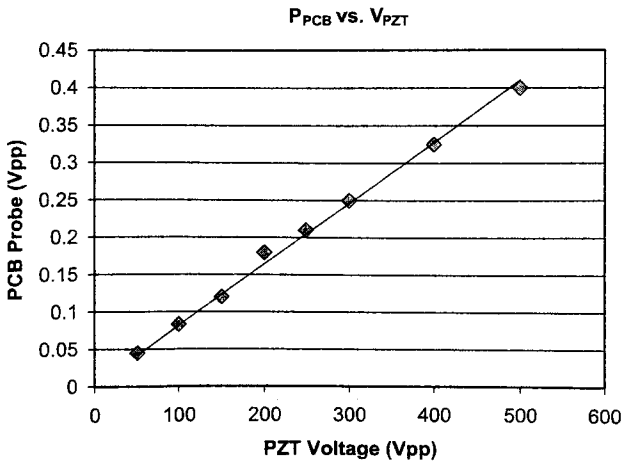

Fig. 3. Measured pressure at the center versus applied voltage.

degassing usually lasted 2 h. Once adequate cavitation was achieved, the operating pressure and cavitation rate were maintained. Each experimental run lasted 7 h.

III. TRITIUM MEASUREMENTS

Measurements of the tritium were made by comparing the relative activity of the tritium in the acetone samples obtained before and after each run. The measurements were based on 1-ml samples. An illustration of the sampling process is shown in Fig. 4. Before every experiment, acetone was placed into the cavitation chamber, and 5 ml of acetone was extracted from the chamber with a syringe using a stainless steel needle. Upon completion of the experiment, another 5 ml was removed and placed into a glass vial. Four separate 1-ml samples were then added to Ultima-Gold scintillation fluid as shown in Fig 4. Following the counting of the samples in a Perkin Elmer/Packard 1900 Tri-Carb Liquid Scintillation Counter, a known quantity of a secondary National Institute of Standards and Technology (NIST) tritium standard was also counted along with a sample of just Ultima-Gold scintillation fluid for background reference. The information obtained from these sets of counts allows the absolute quantity of tritium in each sample to be determined. For reference, tritium counting of normal acetone without cavitation typically resulted in around 10 count/min (cpm). Counting of deuterated acetone without cavitation resulted in a counting rate of ~50 to 80 cpm, depending on the batch of acetone used. An analysis for systematic variation due to chemiluminescence was performed by monitoring the change in count rate of a freshly prepared cocktail, making continuous 5-min measurements for 1.5 h after sample preparation. A measurable decrease in the counting rate was not observed. Furthermore, each sample in the experiment was counted over a period lasting at least 60 h, ample time for luminescence from chemical reactions to subside. To verify this, the two postprocessed and two preprocessed samples were compared, taking into account that there is a 2-h difference between the times when the samples are counted due to rack position. This analysis did not show a significant change in counts that could be attributed to chemiluminescence. Although a high-accuracy syringe was used, mass differences in the samples were still observed. Therefore, all samples were normalized to the weight of acetone within the cocktail. Empty vials were weighed and found to have negligible variation in mass. It was found that because of quenching effects, lesser amounts of acetone in a sample could lead to higher measured counting rates per unit mass of acetone. Figure 5 shows the dependence between counting rate and acetone mass. Four data points with mass ratios between 0.012 and 0.033 used to determine the effects of quenching are also shown. The ordinate of the plot is the measured cpm value of the sample normalized to the mass of acetone placed in the scintillation fluid, resulting in a value that is a function of activity only. The abscissa is the mass of acetone placed in the sample normalized to the total sample mass (acetone, cocktail, and vial). The resultant plot is thus the activity as a function of acetone mass. Figure 5 shows that the activity per unit mass of acetone decreases due to the effects of quenching. Because of the strong quenching effects of the acetone, a number of liquid scintillators were tested, and Ultima-Gold was found to have the smallest effect. Furthermore, a quenching

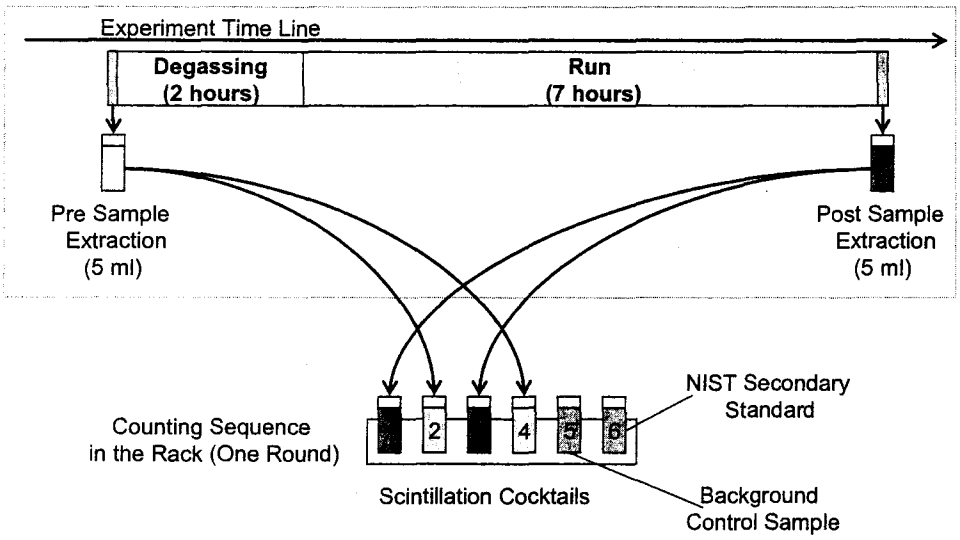

Fig. 4. Sample extraction diagram.

correction technique was employed as follows. An external standard source incorporated into the counter was used to generate a transformed index of the external standard (tSIE) value of each sample before it was counted. Using the measured tSIE values and a calibration curve generated using a set of quenched Ultima-Gold–based secondary NIST standards, the measured cpm values were then converted to corrected disintegrations per minute (dpm). Each set of four samples from each run (two preprocessed and two postprocessed) together with a 16-ml Ultima-Gold sample for background and a secondary standard were counted for 40 min each. The counting cycle was repeated for a minimum of ten times, and the average dpm value together with the standard deviation was determined for each sample. The average dpm value for each sample was first corrected for the background and then for the sample mass. The values of the two preprocessed and postprocessed samples were then averaged, and the preprocessed results were subtracted from the postprocessed results to determine a net change in the activity per gram of acetone due to the processing. The dpm of the secondary standard was used to determine a chi-square value for each series of counts to ensure that the counting system was performing correctly.

IV. RESULTS

Measurement of ten runs with deuterated acetone (C_3D_6O) and three runs with normal acetone (C_3D_6O) were performed. The results are summarized in Fig. 6. All tritium

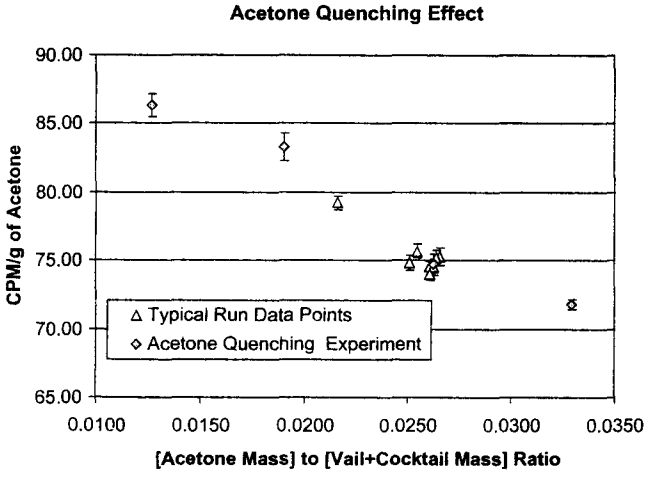

Fig. 5. Acetone quenching effect.

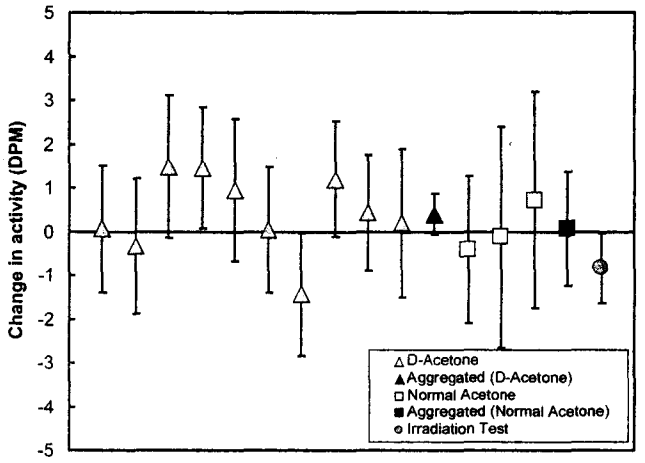

Fig. 6. Tritium measurements.

activity measurements are practically within 1σ of zero. Furthermore, the averaged value of activity change for all the deuterated acetone runs is 0.40 ± 0.47 dpm (i.e., also within 1σ of zero), and the averaged value of activity change for all the normal acetone runs is 0.07 ± 1.30 dpm. Samples from an irradiation test without cavitation were also taken. For this test, a chamber was filled with deuterated acetone and exposed to a neutron source for 3.5 weeks. No measurable increase in tritium activity was observed. This result was confirmed using MCNP5 to analyze neutron capture in deuterated acetone (Walter et al.[4]). In comparison, the closest tritium measurement shown in Fig. 3 of Taleyarkhan et al.[1] is the 5-h irradiation case with the Pu-Be source. That measurement was 3.5 cpm with a standard deviation of 3.5 cpm, which again could indicate a null result.

V. CONCLUSION

Evidence of tritium production attributed to D-D fusion is sought in this study. The average dpm count observed is within 1σ of zero. Therefore, the claim by Taleyarkhan et al.[1] was not confirmed in the experimental study presented.

ACKNOWLEDGMENT

This work was supported in part by BWX Technologies under grant 4000025972.

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