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EXPERIMENTAL MEASUREMENTS AND MCNP5 MODELING OF TRITIUM PRODUCTION IN DEUTERATED ACETONE EXPOSED TO ISOTOPIC NEUTRON SOURCE

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ABSTRACT

Experimental measurements and Monte Carlo modeling of tritium production in deuterated acetone (D-acetone) exposed to constant neutron fluence were shown. The goal of this study was to evaluate the effect of neutron interactions within the deuterated medium. The experiment is a part of a bigger effort taken at Purdue University to reproduce the experiment described by Taleyarkhan, *et. al*., in *Science*, 8 March 2002. In this paper the efforts were focused on the case where the column of D-acetone was not oscillated, eliminating conditions for any other possible nuclear reactions except neutron scatterings and neutron absorption.

In the experiment a sample of D-acetone was placed next to the neutron source for 1.45×10^{6} sec. A separate sample of D-acetone from the same supply was also kept in a sealed container away from the neutron source. Measurement and comparison of the cocktails yielded difference errors on the order of one count per minute (CPM). MCNP5 was used to model the experimental set-up consisting of a cylindrical glass vessel containing D-acetone exposed to a 9.70 Ci Americium Beryllium (AmBe) neutron source. 7.36×10^{-5} counts per minute (CPM) of additional tritium activity was calculated from the MCNP5 output. It is unlikely that activation will lead to false positive testing during experiments where cavitations are initiated. In addition, MCNP5 was used to estimate gamma flux from a ^{60}Co source emanating from the center of the chamber. The experiments involving the gamma sources are planed to be carried out and will be used to benchmark the MCNP5 model presented in this paper.

Key Words: deuterated acetone, neutron interactions, gamma source, MCNP5

1 INTRODUCTION

The acoustic cavitation experiments involve the oscillation of a column of deuterated acetone (D-acetone) within a glass chamber. The fluid after being well "stretched" (hours of degassing) is exposed to neutron flux (either from an isotopic source or from the pulse neutron generator). The neutron induced cavitations in the acoustically excited D-acetone column are filled with D-acetone vapor and tend to collapse violently during the positive pressure portion of the acoustic cycle[1]. Theoretically, if sufficient temperature is attained during the collapse of cavities (bubbles) the conditions for the fusion of D-atoms may take place[1] leading to the following:

$$D + D \rightarrow \text{He-3} + n \quad (3.27\ \text{MeV}) \qquad (1)$$
$$D + D \rightarrow T + p \quad (4.03\ \text{MeV}) \qquad (2)$$

In this paper we focus at the Monte Carlo modeling using the MCNP5 of neutron interactions with the fluid in the chamber without introducing the acoustic oscillations. A model was develop to estimate the number of $^{2}H(n,\gamma)^{3}H$ reactions that take place due to interactions of the source neutrons used to initiate cavitation within the D-acetone. The model created in MCNP5 was intended to mimic the experimental setup as closely as possible. The MCNP5 result was then compared with the experimental measurements. The results of both the experiment and the MCNP5 model are used to determine how much tritium is produced during the time of a cavitation experiment simply by neutrons being absorbed in deuterium.

2 METHOD FOR ACTIVATION ANALYSIS

An MCNP5 input data deck was created to closely model the acetone cavitation chamber and the surrounding cooling and shielding materials. The set-up consists of a borosilicate glass chamber filled with D-acetone. The acetone is 99.9% ^{2}H. Attached to the glass chamber is a piezoelectric ring (PZT) that is held in place with off the shelf two-part epoxy. The chamber rests on a metal stand made from both aluminum and steel. The source is a 9.70Ci AmBe neutron source and is located 7.62 cm away from the centerline of the chamber. The experiment and source is surrounded by paraffin shielding and air at standard temperature and pressure. A simplified diagram of the cavitation experimental set-up is shown in Figure 1.

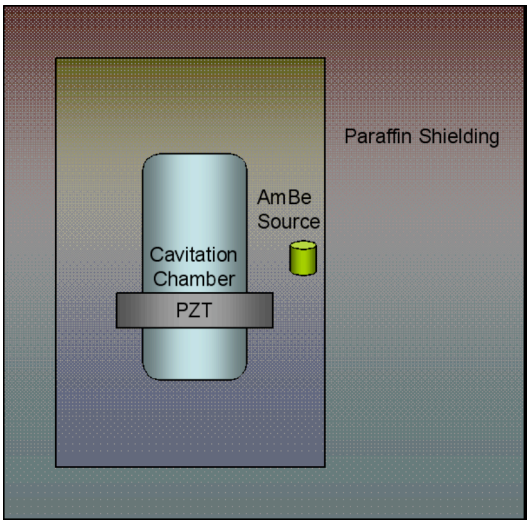

Figure 1. Diagram of a simplified cavitation chamber, source, and paraffin shielding

A track length estimate of the cell flux of neutrons within the volume of acetone chamber was used to determine the amount of tritium production[3]. We also use the MCNP5 capability to determine how many neutrons travel through and undergo a reaction within the acetone column. A tally multiplier was applied to enable consideration of just the $^{2}H(n,\gamma)^{3}H$ reactions. As shown in Figure 2, carbon and oxygen isotopes have relatively high radiative capture cross-sections. For this reason, the neutron interactions were treated separately for the deuterium and for the D-acetone.

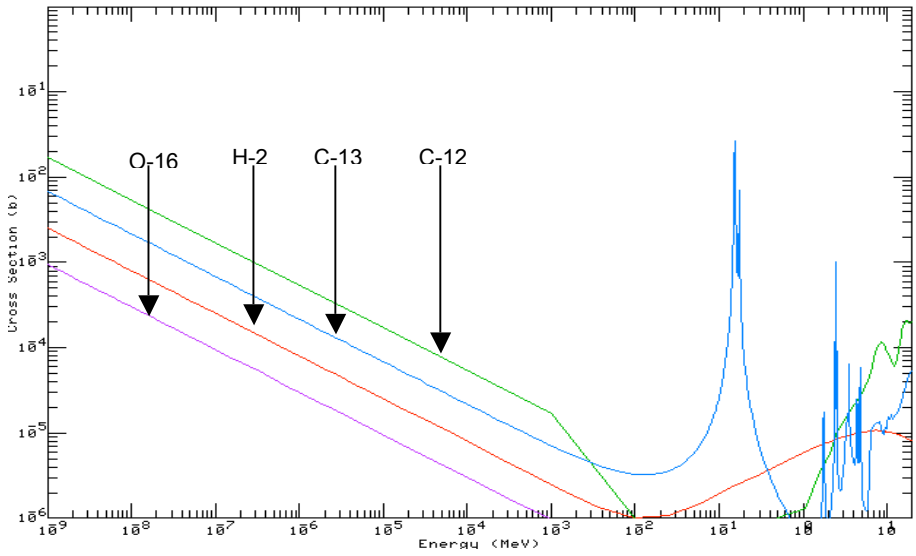

Figure 2. Significant radiative capture cross-sections for D-acetone constituent isotopes[4]

The 9.70Ci AmBe source was modeled using a spectrum obtained from data sheet provided by the source manufacturer. To model this source within MCNP5, an integral approximation was performed using the manufacture's neutron spectrum from Figure 3.

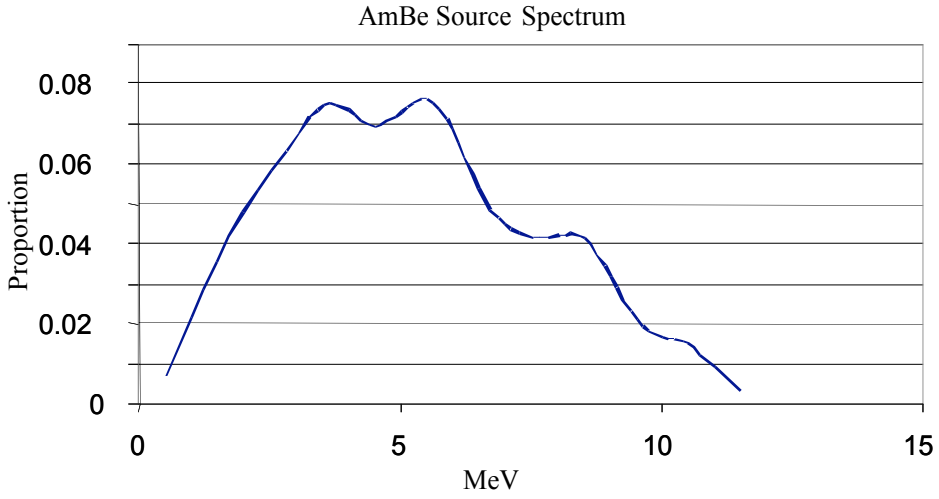

Figure 3. Neutron energy spectrum from AmBe source

The spectrum was broken into 23 different energy bins so that the spectrum of the source in MCNP5 is representative of the actual source. At the time of manufacturing, the source emitted 2.49×10^7 n/s.

The output of the MCNP5 program gives the number of $^2H(n,\gamma)^3H$ reactions occurring per neutron emitted from the source per cubic centimeter of acetone. Since the process to determine the activity of the D-acetone is done by measuring the activity of 1cc of D-acetone, the output from MCNP5 needs to be multiplied by the source neutron activity to obtain the

number of tritons produced per second of exposure to the source (cell volume specified in appropriate cell).

2.1 Tritium Background and Detector Efficiency

D-acetone has a higher than background level of tritium activity. Depending on the quality of manufacturing, this level varied from ~5 to 10 times the tritium background for non-deuterated acetone. The Perkin Elmer/Packard 1900 Tri-Carb Liquid Scintillation Counter machine was used to determine the counts per minute (CPM). CPM is the number of events within the scintillation cocktail that the machine detects. Based on the detector efficiency, chemical effects due to mixing an organic solvent such as acetone with the scintillation cocktail, and with the geometry of the solid angle, the Packard machine can calculate the number of disintegrations per minute (DPM) in the sample. A 1cc D-acetone sample was measured that gave, for example, 63.59 CPM and 166.0 DPM. Dividing CPM by DPM gives a gross detector efficiency of approximately 38%.

2.2 Experimental Results

A sample of D-acetone was placed next to the neutron source for 1.45×10^{6} sec. A separate sample of D-acetone from the same supply was also kept in a sealed container, well away from the neutron source for the duration of the experiment. At the end of the experiment, 1cc of D-acetone was removed from each container and each sample was used to prepare a scintillation cocktail. Table 1 shows the results of the measurement.

Table I. D-acetone neutron activation analysis

SAMPLE	CPM	ERROR
Irradiated D-acetone	76.74	0.57
Un-irradiated D-acetone	77.58	0.57
Difference	-0.84	0.81

2.3 MCNP5 Results

The result of the tally in MCNP5 gives 4.995×10^{-11} (Realtive Error: +/-0.0005) tritium atoms produced from deuterium per source neutron. A total of 1.98×10^{8} neutrons were tracked and the tally passed the 10 statistical checks for the tally fluctuation chart bin result. At a production rate of 4.99×10^{-11} tritium atoms produced from a source emitting 2.49×10^{7} neutrons per second, over the course of 1.45×10^{6} sec, 1.81×10^{3} tritium atoms will be produced. Considering a half-life of 12.33 years for tritium, the 1.81×10^{3} tritium atoms will have an activity

of 1.94×10^{-4} DPM. Taking into account the approximate efficiency of the tritium counting system of 38%, this number will convert to 7.36×10^{-5} CPM.

3 GAMMA SOURCE MODELING

An additional MCNP5 model was created to determine a gamma flux distribution from a ^{60}Co source located at the center of the chamber. The purpose of this work was to begin development of simplified models using the MCNP5 code which could be verified through the gamma flux measurements in the actual experimental set-up. This will allow for fine tuning of materials and geometry choices within the MCNP5 model and ultimately lead to a fully benchmarked model. The model will then be used to determine what types of in-situ experimental radiation measurements are most feasible.

3.1 MCNP5 Gamma Source Model

The chamber used in the activation model described above was also used for gamma source modeling. For initial calculations, the D-acetone within the chamber and the environment surrounding the chamber were eliminated from the model. A ^{60}Co gamma source similar in shape to a calibration source (cylinder: 3mmH x 25mmD) was placed within the chamber, 5.95cm below the geometrical center and centered within the chamber. Detectors with a 1cm radius were simulated in MCNP5 to measure the gamma flux distribution around the chamber. This distribution was measured in 45 degree increments along each of the three Cartesian planes that pass through the center of the chamber. These planes are the , , and planes and the tally positions are at a fixed distance (15 cm) from the center of the chamber. Figure 4 shows the detector positions for the cases where the XZ and XY planes are analyzed. For the case of the XZ plane, the detector positioned in the positive x-direction represents the detector at 0^{o}. The position then increases to 315^{o} in the counter clockwise direction.

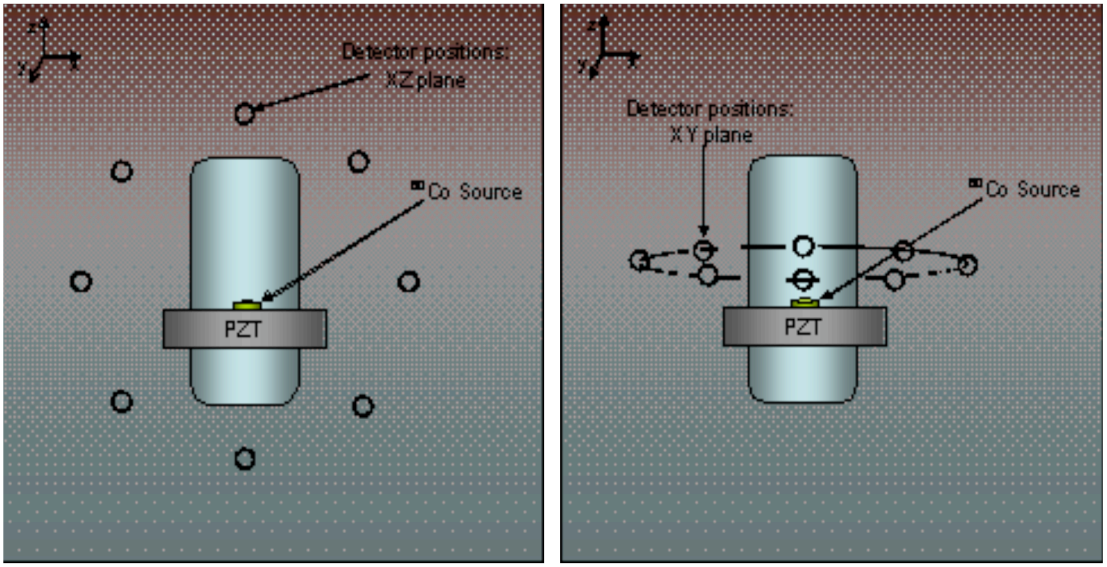

Figure 4. Detector locations for the XZ and XY planes

Figure 5 depicts the gamma flux at different angles along the three Cartesian planes. Only the flux for the 662 keV gamma peak is shown. For the X=0 and Y=0 planes, the flux measurement at 270° is nearly twice the distance from the source as the flux measurement at 180°. It can be seen that the Inverse Square Law verifies the shown flux distribution.

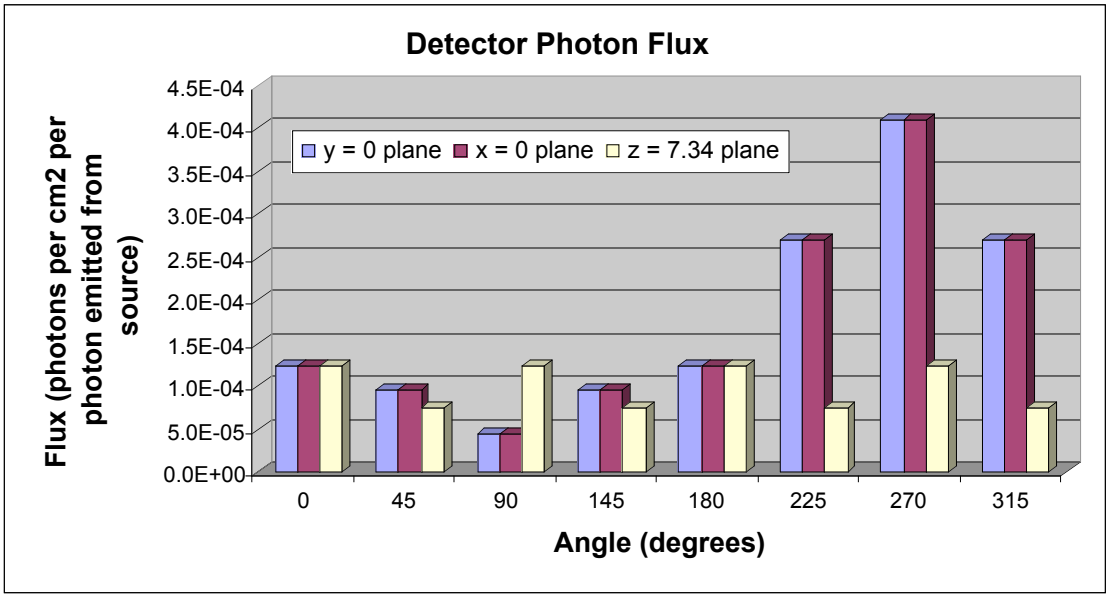

Figure 5. Relative detector flux of 662 keV gammas from ^{60}Co source

4 CONCLUSION

The amount of tritium produced through neutron absorption by deuterium is insignificant to the background level of tritium that exists within the samples of deuterated acetone. This level of activity also falls well below the measurement error of the tritium scintillation counting. It is, therefore, not probable that activation will lead to the presence of excess tritium during cavitation experiments. Future work will focus on relating the gamma flux model to measurements of a ^{60}Co source in experimental set-ups.

5 REFERENCES

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