

COMMENTS ON THE SHAPIRA AND SALTMARSH REPORT

by

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Abstract

We have carefully reviewed the data and report of Shapira and Saltmarsh. Our analysis of these data indicates that, contrary to their conclusions, a statistically significant increase of nuclear emissions was actually detected by them during cavitation experiments with chilled deuterated acetone. In particular, the emission rate they measured was ~ 3 n/s (compared to about 8 n/s in our measurements). Shapira and Saltmarsh grossly over-estimated the efficiency of their detector. Actually, their detection efficiency for 2.5 MeV neutrons (based on calibration with a Pu-Be source and corrected for distance and shielding), was found to be $\sim 10^{-5}$. Using this value of efficiency their detected nuclear emission rate was $\sim 2 \times 10^5$ n/s, a value which is comparable to that reported by us, and consistent with the results from our tritium measurements. Statistically significant time-correlated neutron emissions during sonoluminescence (SL) bursts was also observed, however their system was poorly designed for coincidence measurements; particularly, for the most energetic bubble implosions subsequent to cavitation.

Introduction

Two separate experiments were conducted with chilled (0°C) deuterated acetone using most of the same equipment we used¹ (particularly the cavitation chamber and the SL photomultiplier detector), but with a different nuclear emission detection system², which was set up by Shapira and Saltmarsh from ORNL's Physics Division (PD). The PD detector was a large NE-213 proton recoil detector (~ 30 times larger than the detector used in our experiments) which, in principle, was capable of neutron-gamma (n-g) discrimination in the absence of electronic saturation. An independently developed data acquisition system was used with the PD detector. Unfortunately, the significantly larger size of the PD detector made this system much more prone to background radiation and electronic-saturation-related problems.³ Indeed, the recommended³ size detector for 2.5 MeV neutrons is $\sim 100\text{cc}$ (which is the size of the Elscint (ET) detector used by us in our experiments¹).

Setup for the separate PD experiments

The setup of the PD experiments is shown schematically in Fig.1. Also shown for reference is the cylindrical 2"x2" liquid scintillation Elscint (ET) detector used by us. Due to the large size of the PD detector it could not be positioned within the experimental enclosure. Key features and data related to geometry, solid angles, etc. and differences between the ET and PD detectors are summarized in Table 1 and in Fig.1. Significantly, the PD detector was 'shielded' from the experimental chamber by a ~6-mm thick plastic wall covered with a 1.5~2- inch thick refrigeration pack materials for enclosure cooling.

Shapira and Saltmarsh noted in their report² that the PD detector threshold was set based on the 14 MeV proton recoil edge and use of published light output curves³. The use of a ⁶⁰Co source is cited in Reference 2, however the original report written on use of the PD detector made no mention of this calibration⁵, and other versions⁶ claimed the use of ¹³⁷Cs. It is significant that no ¹³⁷Cs or ⁶⁰Co sources existed in our laboratory at the time of the PD experiments. If such calibrations were performed they were not done in-situ and thus are inappropriate for making any case related to knowledge of the lower discrimination threshold. Indeed, calibration quantities are only relevant if made in-situ using the same settings and geometrical configuration as used for actual data gathering. Indeed, this is exactly how the calibrations were performed in our experiment¹.

The 14 MeV proton recoil edge for the PD detector was documented⁴ to be around channel number 2400, and the lower cutoff was set at channel number 200⁵. Using light output curves from Ref. 3 (and presuming the validity for their "direct" applicability for determining the proton recoil edge for 2.5 MeV neutrons for the much larger PD detector) yields a proton recoil edge ratio for 14 MeV and 2.5 MeV protons ranging from 10 to 11. This indicates that the 2.5 MeV proton recoil edge for the PD detector would lie between channel numbers 240 to 220 (i.e., 2400 / (10 or 11)). Using NE-213 light output curves³ we estimate that the threshold was at an equivalent proton energy of 2.2~2.4 MeV, which is below, but very close to, 2.5 MeV. Since this value is so close to 2.5 MeV, this would imply the PD detector had a very small efficiency for detection of 2.5 MeV neutrons since most of the large-angle scattered neutrons would not be counted. Thus, the PD extrapolated-from-14MeV-protons efficiency is considerably smaller than that quoted by the authors². Next we will show that the efficiency of detection of 2.5 MeV neutrons can be significantly lower than that for 14 MeV neutrons and that this efficiency is very sensitive to the threshold setting (bias) on the PD pulse height (see Appendix A).

To obtain a more definitive estimate of efficiency for detection of 2.5 MeV neutrons, data were taken with the PD detector⁴ located 30cm from our Pu-Be source which emitted $\sim 2 \times 10^6$ n/s. The efficiency of neutron detection was found to be $\sim 2 \times 10^{-4}$. A Pu-Be source emits neutrons which have energies mostly below 5 MeV (with only about 15-20% being in the 6 to 10 MeV range). Thus the value of 2×10^{-4} for efficiency of detection is an overestimate, with the actual efficiency for detection of 2.5 MeV neutrons

being closer to $\sim 10^{-4}$, for the PD detector at a distance of 30cm from the source when there is no significant shielding. It should be noted that this estimate does not account for scattering of 2.5 MeV neutrons by the D-Acetone and glass in the test chamber and also scattering by the 1.5-inch-thick refrigeration pack materials and plastic walls of the experimental enclosure. Hand calculations and complementary Monte Carlo (MCNP) neutron transport calculations shows that only $\sim 50\%$ of 2.5 MeV neutrons would leave the chamber without scattering losses. Again, it is expected that another factor of at least two reduction would result due to scattering of 2.5 MeV neutrons by the plastic wall and refrigeration pack materials. Therefore, the net efficiency of detection of 2.5 MeV neutrons at a distance of 30cm would be $\sim 2.5 \times 10^{-5}$. If the distance from the cavitation source is greater (as was the case for the PD detector - see Figure 1 and Table 1), the detection efficiency of 2.5 MeV neutrons generated in the chamber for the PD detector is estimated to be $\sim 10^{-5}$. What this implies is that, only 10 out of every 10^6 2.5 MeV neutrons would be to detected by the PD detector as a consequence of its location and threshold level.

Coincidence PD measurements were made differently from the method used by us. In our experiments, coincidence was monitored via triggering from the SL signal and then recording the occurrence of a nuclear signal within $\pm 10 \mu\text{s}$ on a 500 MHz digital storage device. However, in the PD experiments Shapira and Saltmarsh initiated a search for coincidences based on the n+g nuclear signal (i.e., not using pulse shape discrimination) and opened up a $\pm 10 \mu\text{s}$ time window to look for the first occurrence of an SL signal, after which the system would stop while data were being transferred to memory. The difference in approach from our method is significant since in these experiments the SL signals occur far less often compared with the n+g signals; the ratio of SL to n+g signals being $\sim 1:600$ for the PD detection system. Searching for coincidences normally requires triggering on the rarest of two signals. Moreover, the PD detector displayed a gamma-to-neutron ratio of $\sim 30:1$. With such a high gamma background level, triggering on the n+g signal (without pulse shape discrimination of gamma rays) would make it quite difficult to obtain coincidence data between SL emissions and the emitted neutrons, and significantly enhances the recording of random coincidences.

Analysis of Experimental Results

We provide next our analysis of the results of the experimental data taken with the PD detector in separate parts as follows:

Analysis of neutron emission rate during cavitation of chilled D-Acetone

Dr. Shapira kindly provided us with his raw data and we analyzed this data for n+g emission in relation to SL emissions for the cases with and without cavitation. The data obtained from these independent experiments had indicated⁵ two key regions of SL light detection. The first region (Region B; Fig. 3 of ref. 2) correlates with the $\sim 25 \mu\text{s}$ time span of bubble implosions after birth of the bubbles due to nucleation by PNG neutrons. The second region (Region D; Fig. 2 of ref. 2)) corresponds to repetitive SL emission at

intervals of 52 μs that occurred later in time and lasted for several hundred microseconds. Figure 2 displays the profile of change in emission for the time spans in and around Regions B and D, respectively. It is seen that a statistically significant (over 10 standard deviations (SD)) increase in nuclear emission occurs correlated with the SL emissions. It was found that the net increase of counts was 2~3 counts/s. Dividing this value by the efficiency of detection of $\sim 10^{-5}$ provides an emission rate of $\sim 2 \times 10^5$ n/s. This value is close to the 8×10^4 n/s we found in our experiment. It should be noted that the value of 8×10^4 n/s emission rate we have reported¹ was not corrected for the 2.5 MeV neutron scattering and subsequent neutron energy losses in our experimental chamber (estimated to be about a factor of two). Estimating counting losses resulting from this scattering of $\sim 50\%$ would imply a neutron emission rate in our experiments of $\sim 1.6 \times 10^5$ n/s. Thus, contrary to what Shapira and Saltmarsh claim², the PD neutron emission rates are in quite good agreement (considering experimental uncertainties) with the emission rate we estimate from our experiments¹ and, with the rates of $\sim 7 \times 10^5$ determined from our tritium data¹.

Coincidence data analysis

As noted, previously, due to the relatively large size of the PD detector (~ 30 times larger volume than ours, which resulted in an n:g ratio of 1:30 compared to the ET detector) and the fact that the detection of coincidences was performed by triggering on the n+g signal rather than the SL signal, the PD detection system should be dominated by random coincidences. This was indeed found to be the case, as documented by Shapira and Saltmarsh for their experiment^{4,5}.

In order to assess whether the PD detector could possibly have detected the ~ 17 net coincidences noted¹ by us in the $\pm 2 \mu\text{s}$ time bins immediately adjacent to the SL signal, an analysis was performed on solid angles, with further corrections made for the time of data gathering and for shielding effects. The results of this analysis is summarized in Table 1. As seen therein, based on geometry and shielding/scattering considerations alone the PD detector would have been able to detect on an equivalent basis only up to 4~5 coincidences in each $\pm 2 \mu\text{s}$ time bin around the SL peak. In fact, this is what was obtained and documented⁵ in the PD experiments.

Due to severe electronic saturation problems⁵ faced by the PD detection system in Region-B (corresponding to the time of the first collapse of the imploding bubbles) pulse shape discrimination could not be performed in this region. However, this was possible to do for Region-D, where close to 20 peaks in SL emission were observed in synchronization with the acoustic drive frequency of our chamber. Obtaining the raw data (from Dr. Shapira) for neutron emission with time for Region-D a detailed analysis was performed by us of Region-D to determine if there was a systematic correlation between the SL peaks observed there and the neutron emission data recorded for this region. As mentioned earlier in the PD measurements, SL flashes in Region-D occur in peaked clusters separated by intervals that are about one period (52 μs for the 19.3 kHz acoustic excitation used). Therefore, the SL spectrum was processed so that data from the 19 SL peaks of Region D near the middle of the spectrum were added together. It

was noted that 2/3 of all the SL counts appeared within +/- 1 μ s of the peaks. In the convoluted spectrum, which is the sum of 19 sections each of 52 channels, we collected the neutron counts (i.e., gammas were rejected by pulse shape discrimination) into bins of these adjacent channels; this was to match the 3 channel width of the individual peaks, while aggregating some of the small number of counts per channel. Overall, it was found that the number of counts in the three channel bins centered on the SL peaks was 476. The average number of counts in all the three channel bins outside the SL peak was 375. The difference is 101 counts, or 27%, in the bin having the most neutron counts, which is higher than the concentrations in the other bins by more than 4 standard deviations (SD). Results of this analysis are shown in Figures 3. It must be noted that every single one of the bins outside the peak was found to have fewer counts by more than 2 SD than the SL peak bin. While not a definitive statement on coincidences, this analysis clearly shows that even though the PD detector had severe gamma saturation problems, the SL emission was time-correlated with statistically significant increased neutron emission for the SL peaks in Region-D.

Conclusions

(1) Statistically significant (over 10 SD) neutron/nuclear emissions were observed in the PD experiments. Using a more accurate efficiency for 2.5 MeV neutron detection by the PD detector we estimate an emission rate of $\sim 2 \times 10^5$ n/s, which is very similar to the magnitude of the neutron emissions detected by our ET detector¹. Moreover, the excess neutron emissions with the PD detector in Region-D were statistically significant (over 4 SD increase) and were time-correlated with the SL emission peaks.

(2) These neutron emission rates are also compatible with the 2.5 MeV neutron emission rates inferred from the tritium emission data reported in our *Science* paper¹.

(3) The large PD detector system suffered from severe electronic saturation issues at least during the time associated with the first bubble implosion Region-B. Analysis of the ET and PD detector systems reveals that the ET detector was much less susceptible to these effects relative to the PD system, and this is why we were able to make valid measurements in Region-B (where the most intense bubble implosions occurred).

(4) Due to reduced "net" detection efficiency for the PD detector, the net coincidences observed in our experiments with chilled deuterated acetone (Region-B) would not have been possible to detect using the PD system.

Closure

An internal audit of our Bubble Fusion experiment¹ was performed by ORNL. It was concluded that both our tritium⁷ and neutron⁸ measurements were valid. However, some additional experiments were performed by the Physics Department (PD) and these measurements initially appeared to yield results which were different from ours (e.g., a lower fusion neutron yield). The resultant internal ORNL report², which was never

subjected to peer review, was unfortunately widely disseminated and this has led to a lot of controversy concerning our experimental findings. Indeed, some people may have reached conclusions concerning the validity of our findings prior to the publication of our paper¹. This is truly unfortunate and is very unprofessional.

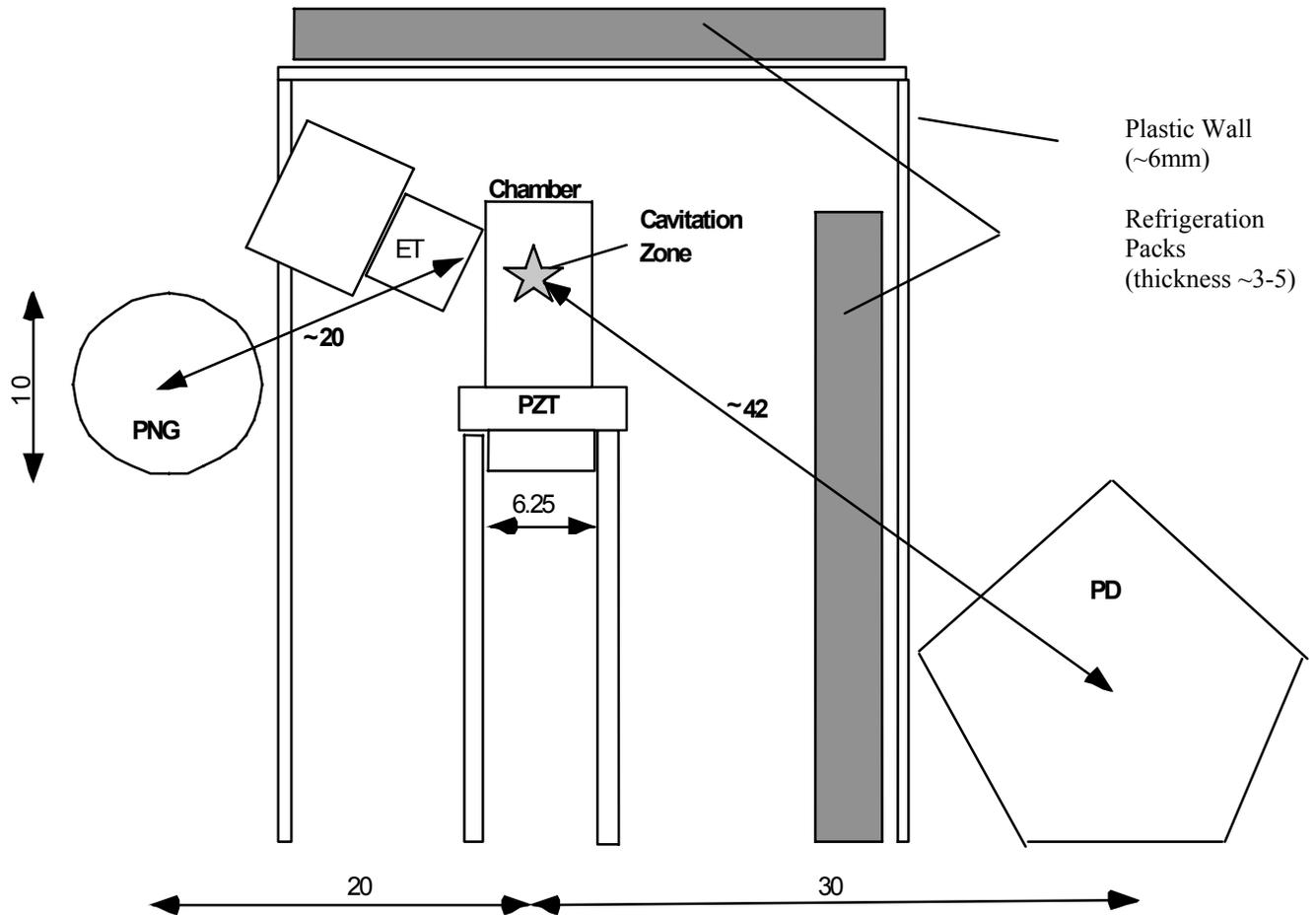
Nevertheless, we have now shown that the PD detection system which was used in the additional TD experiments was poorly designed for the purpose of taking Bubble Fusion data. Moreover, they assumed a detector efficiency which was grossly over-estimated and leads to erroneous conclusions. Nevertheless, when we reanalyzed their data they were found to be completely compatible with ours. It is truly sad that the premature announcements of the results made by the PD investigators² may have confused and/or misled the press, and some scientists and researchers who are genuinely interested in Bubble Fusion¹. In any event, we hope that this on-line report will set the record straight. Finally, we strongly encourage other groups to independently confirm our results, and become engaged in Bubble Fusion research so the exciting possibilities of this new discovery can be fully explored.

References

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2. D. Shapira and M. Saltmarsh, "Comments on the possible observation of D-D fusion in Sonoluminescence" (<http://ornl.gov/slsite>).
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5. D. Shapira and M. Saltmarsh, "Using a Neutron Detector in Sonoluminescence Experiment - Part II," Oak Ridge National Laboratory Internal Correspondence Report, July 31, 2001.
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8. D. Shapira, "Review of Second SL Manuscript by R. P. Taleyarkhan, C.D. West, J.S. Cho, R.T. Lahey, Jr., R. Nigmatulin," ORNL Physics Division Internal Correspondence Review Report, December 19, 2001.

Table 1. Comparison of Key Data for ET and PD Detectors

| Parameter | ET Detector | PD Detector |
|--|-----------------------------|---|
| Projected Area (cm ²) | 20-25 | 270 |
| Volume (cc) | 99 | 2,950 |
| Normalized Area Ratios (normalized to ET) | 1 | 12 |
| Normalized Volume Ratios (normalized to ET) | 1 | 30 |
| Distance from PNG (cm) | 20 | 50 |
| Distance from Cavitation Zone in Chamber (cm) | 5-6 | 42-43 |
| Solid Angle (from Cavitation Zone in Chamber) | 0.072 - 0.05 | 0.012 |
| Normalized Ratio of Solid Angles (normalized to ET) | 1 | 0.16 to 0.24 |
| Neutron Shielding from Refrigeration Packs and Plastic Container | None | Factor of about 2 to 3 reduction in 2.5 MeV neutron flux |
| Time for Coincidence Data Gathering (min), depended on tuning | 18 to ~40 (average 30 min.) | 64 |
| Net Coincidences (in 2 μs bins) | 17 | =17 x (0.16 to 0.24) x (1/3) x (64/(18 to 40)) = ~1.5 to 5 |



- Notes: (1) All dimensions in cm (except where noted)
 (2) Acronyms: ET- Elscint detector; PNG- Pulse Neutron Generator;
 PD- Large pentahedron shaped detector
 (3) PMT and other items such as stands and borated paraffin blocks not shown.

Figure 1. Experimental configuration showing key geometrical parameters and locations for key components.

PD Detector Results for Region B (Emissions Following First Bubble Implosions)

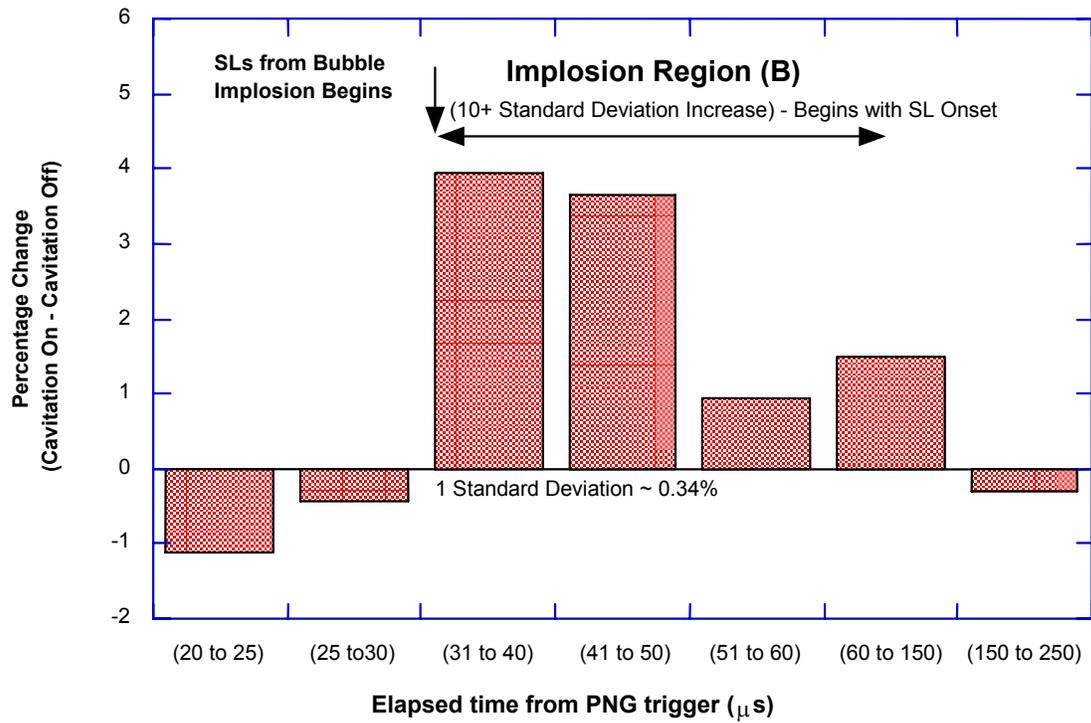


Figure 2a. Variation of increase in nuclear emissions with cavitation in deuterated acetone for Region B (first collapse of nucleated bubbles) - Data taken with PD detection system.

PD Detector Results (Region D)
Percentage Change in Counts between
Cavitation On & Cavitation Off

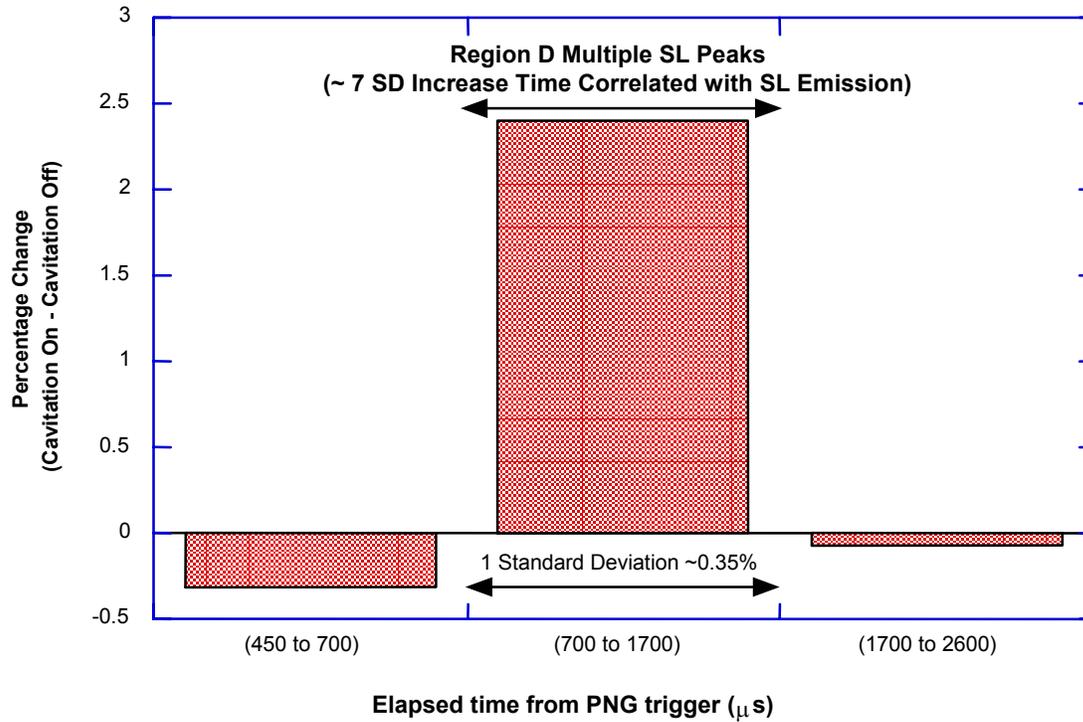


Figure 2b. Variation of increase in nuclear emissions with cavitation in deuterated acetone for Region D (subsequent collapses of nucleated bubbles) - Data taken with PD detection system.

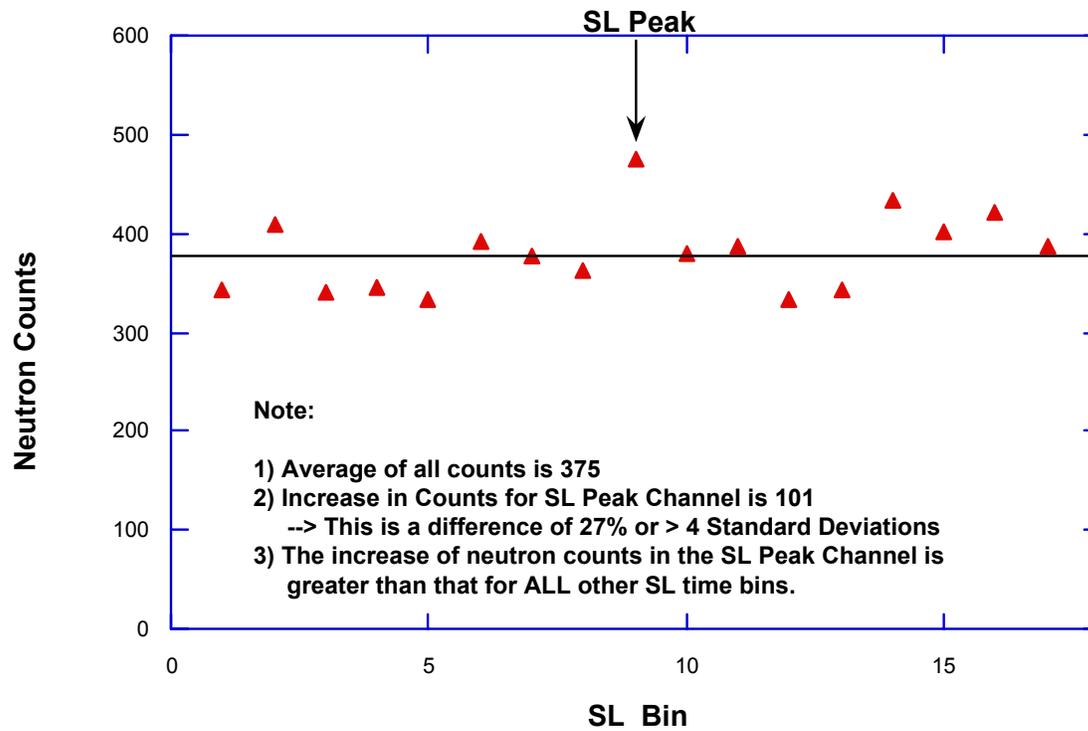


Figure 3a SL time bin correlated neutron emissions for Region D
- PD Detection System

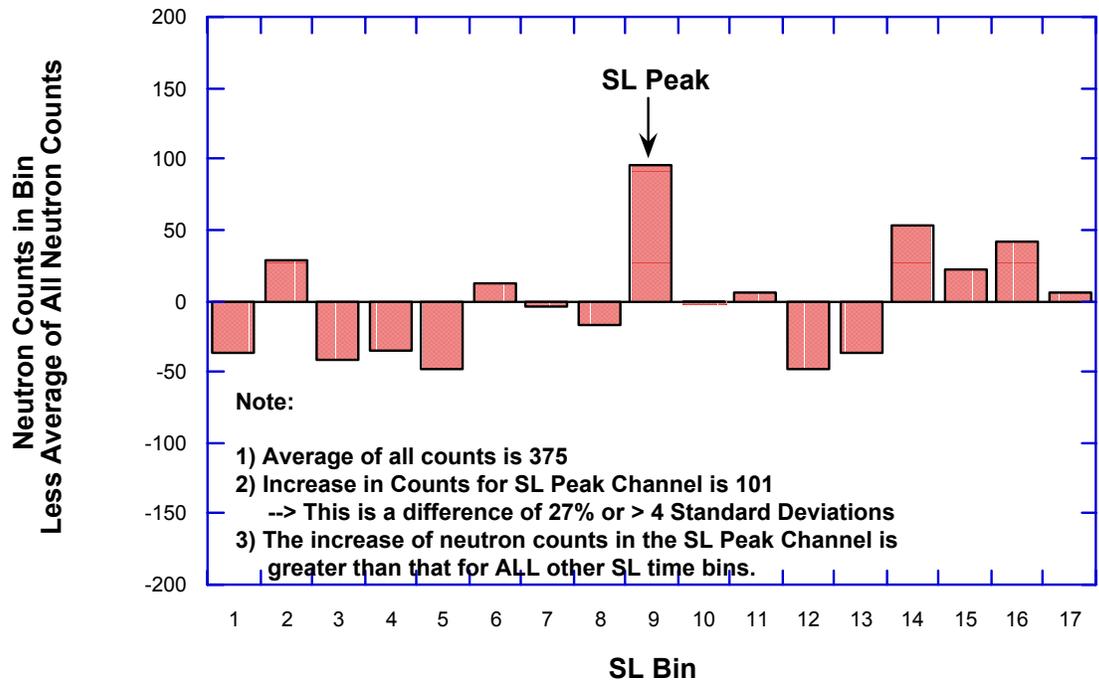


Figure 3b SL time bin correlated difference (actual counts less average of counts) in neutron emissions for Region D - PD Detection System

Appendix A

Efficiency of a Proton-Recoil Detector to 2.5 and 14 MeV Neutrons

Figure A.1 shows the efficiency for detection of a 2.5 or 14 MeV neutron which interacts in the scintillator. For simplicity it is assumed that only H scatters take place and that the pulse height distribution vs proton energy deposited is a uniform rectangular distribution which spans the range from 0 to either 2.5 or 14 MeV. Under these conditions the efficiency per interacting neutron, E_{int} , is given by:

$$E_{int} = [E_n - D_d]/[E_n] = 1 - D_d/E_n$$

Where E_n is the neutron energy (2.5 or 14 MeV) and D_d is the discriminator level (in MeV).

Note that this efficiency is a linear function which is equals unity when the discriminator level is set to zero and equals zero when the discriminator level is equal to (or greater than) the neutron energy. For example, at a discriminator level of 2.0 MeV the efficiency for 2.5 MeV neutrons is only 20% while the efficiency for 14 MeV neutrons is 86%. This illustrates how the discriminator level is much more critical for 2.5 MeV neutrons than for 14 MeV neutrons.

To obtain the intrinsic efficiency for a specific detector we have to take into account the fraction of neutrons striking the detector which interact. For the ET cylindrical 2"x2" NE-213 detector¹, the fraction of 2.5 MeV neutrons that interact is 0.615 and for 14 MeV neutrons it is 0.380. Thus the interaction efficiencies shown in Fig. A.1 should be multiplied by 0.615 and 0.380, respectively, to obtain the intrinsic efficiency in this detector for 2.5 and 14 MeV neutrons, respectively. For example, at a discriminator level of 2.0 MeV, the intrinsic efficiency of the 2"x2" ET detector is $0.2 \times 0.615 = 0.12$ for 2.5 MeV neutrons and $0.380 \times 0.86 = 0.33$ for 14 MeV neutrons.

For the much larger PD detector the fraction of 2.5 MeV neutrons that interact is 0.97 and the fraction of 14 MeV neutrons that interact is 0.73. These are the numbers to multiply the efficiencies in Fig. A.1 by. As an example, at a discriminator level of 2.0 MeV, the intrinsic efficiency of the PD detector is $0.97 \times 0.2 = 0.19$ for 2.5 MeV neutrons and $0.73 \times 0.86 = 0.63$.

The intrinsic efficiencies at a discriminator level of 2.0 MeV are summarized in the table below and in Figure A.1.

Intrinsic Efficiency in the ET 2"x2" and PD Detectors for a 2.0 MeV Discriminator Level

| Detector | Intrinsic Efficiency for 2.5 MeV Neutrons | Intrinsic Efficiency for 14 MeV Neutrons |
|-----------------|--|---|
| ET 2"x2" NE-213 | 0.12 | 0.19 |
| PD NE-213 | 0.33 | 0.63 |

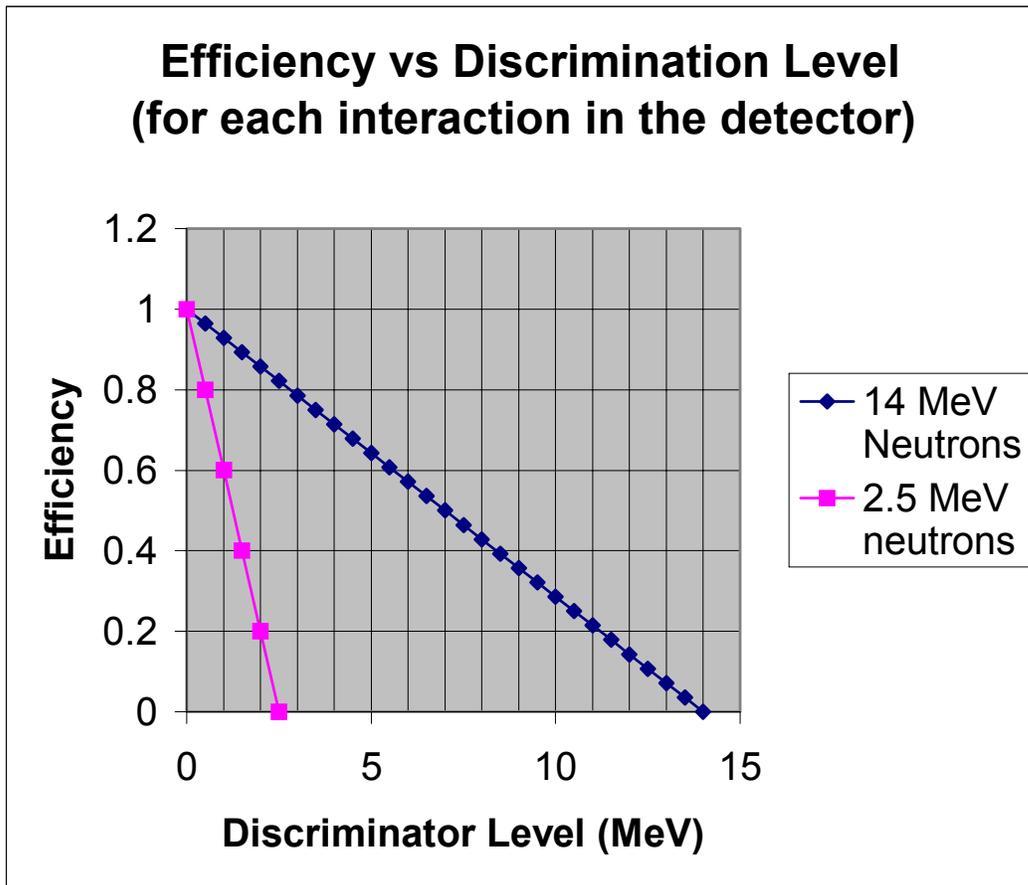


Figure A.1. Variation of efficiency vs discrimination level for neutrons interacting in the detector