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MASTER

LOW-BACKGROUND MEASUREMENTS OF NEUTRON EMISSION FROM Ti METAL IN PRESSURIZED DEUTERIUM GAS*

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ABSTRACT

A wide variety of neutron detector systems have been used at various research facilities to search for anomalous neutron emission from deuterated metals. Some of these detector systems are summarized here together with possible sources of spurious signals from electronic noise. During the past two years, we have performed experiments to measure neutron emission from pressurized D_2 gas mixed with various forms of titanium metal chips and sponge. Details concerning the neutron detectors, experimental procedures, and results have been reported previously. Our recent experiments have focused on increasing the low-level neutron emission and finding a way to trigger the emission. To improve our detection sensitivity, we have increased the shielding in our counting laboratory, changed to low-background ^{3}He tubes, and set up additional detector systems in deep underground counting stations. This report is an update on this experimental work.

INTRODUCTION

During the past two years, a considerable amount of work has taken place in an attempt to detect neutron emission from deuterided metal systems. The proposed nuclear reaction is $d + d \rightarrow {}^3He + n$ (2.45 MeV). This reaction competes with the fusion reactions yielding tritium (T) and protons (P) or ^{4}He. In general, neutrons have the desirable property that they readily penetrate the sample, container, and the experimental apparatus.

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High-efficiency neutron detectors have been designed to measure the neutron production rate in bulk samples that include the entire experimental sample. For low-background underground experiments, the detectors are sensitive enough to detect a few d,d fusion events per hour from the samples.[1]

For neutron detection, the detector parameters of interest include the efficiency, neutron energy resolution, pulse time information, sensitivity to gamma-ray and cosmic-ray backgrounds, and noise susceptibility. These characteristics, together with shielding, will determine the sensitivity of the system to measure low-level neutron signals.

This paper describes some typical neutron detectors that have been used for "cold fusion" type experiments, and it gives some of the sources of false signals and some techniques to protect against them. Also included is an update on the recent neutron measurements that we have performed at Los Alamos.

NEUTRON DETECTORS

A wide variety of neutron detectors have been used for the investigation of neutron emission from deuterated metallic-lattice experiments. A summary of the publications corresponding to these experiments can be found elsewhere.[2,3] Table I lists the detector types that have been used in the experiments. The total neutron detection efficiencies range from 10^{-5} to 0.44 with the ^{3}He systems generally giving the higher efficiencies. Note that for neutron coincidence or time-correlation counting, two or more neutrons from a single event must be counted and the coincidence counting efficiency varies as the square of the singles efficiency. Thus, low-efficiency detectors are not well suited to measure neutron coincidence burst events.

There is a basic difference between the thermal-neutron detectors and the fast-neutron detectors listed in Table I. The advantages of the thermal-neutron detectors include

1. higher efficiency,
2. simpler operation,
3. very low gamma-ray sensitivity, and
4. burst detection capability by moderator thermalization time.

The advantages of the fast-neutron detectors include

1. neutron energy spectra,
2. fast time information, and
3. lower neutron backgrounds.

The experiments that use a combination of detector types are especially good for noise rejection because the detectors are usually vulnerable to different types of problems.

Some possible sources of noise that give false neutron signals are listed in Table II. Most of the noise events are electrical but some are nuclear in that cosmic-ray events and radioactive decay might be misinterpreted.

Table III lists some of the techniques that can be used to protect an experiment from noise events and/or to flag noise events in the data analysis. In general, experiments that have been performed during the past year have incorporated more of the noise protection techniques than the experiments during the previous year. As long as the neutron emission results remain intermittent and irreproducible, a great amount of attention must be paid to the noise vulnerability question.

TABLE I. Neutron Detectors Used for Cold Fusion Experiments

Type	Neutron Energy	Reaction	Typical efficiency (%)	γ-sensitive
^{3}He tubes	Thermal	^{3}He (n,p)	1-44	No
BF_3 tubes	Thermal	^{10}B (n,α)	0.5-20	No
H_2O + NaI/Ge	Thermal	H (n, γ)	<0.1	No
Activation foils	Thermal	(n, γ) or (n,F)	0.1-5	No
Li glass	Thermal	^{6}Li (n,α)	1-20	No
Liquid scintillator	Fast	n,p recoil	1-25	Yes
Plastic scintillator	Fast	n,p recoil	1-20	Yes
Plastic combination	Fast/Thermal	(n,p) + (n,α)	10-25	Yes/No
Cerenkov	Thermal	(n, γ) + e	15-20	High energy

TABLE II. Potential Sources of False Signals

Electrical Noise
1. Tube high-voltage leakage (moisture seal)
2. EMI* noise pickup (EMI seal)
3. Power line noise (filters, veto counters, etc.)
4. Microphonics
Cosmic-ray Background
1. Total counts (cosmic-ray interactions in the shielding)
2. Coincidence counts (spallation in the detector and shielding)
3. D_2 target reactions
Area Background Neutrons
1. Accelerators and reactors
2. Radioactive sources (manmade)
3. Natural radioactivity (uranium, radon)

*EMI corresponds to electromagnetic interference.

TABLE III. Protection Against False Signals

1. EMI* shielded signal lines and high voltage
2. Hermetically sealed and dried high-voltage components
3. Power-line noise filters
4. Multiple independent counter segments
5. Two (or more) different detectors
6. Neutron spectral energy
7. Pulse time of arrival (slowing down)
8. External veto detectors
9. Variable distance detectors
10. Pulse shape analysis
11. Cosmic-ray shielding (underground)
12. Rigorous control runs

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*EMI corresponds to electromagnetic interference.

To gain better sensitivity in the experiments, it is necessary to reduce the cosmic-ray background signal by electronic means or shielding or both. The true neutron background has a random component from the decay of radioactive elements and a time-correlated component from cosmic-ray spallation reactions in the sample or detector body. The time-correlated background can be greatly reduced by performing the experiment underground. For example, the coincidence background decreases by a factor of 10^3 in the 70-m-deep tunnel at Los Alamos and by a factor of 10^5 at the deep-mine locations at Leadville, Colorado, and Kamioka, Japan[1] (1000 m). The coincidence neutron background rate is only ~0.1 counts/d for a 32% efficient ^{3}He detector in the Leadville tunnel.

HIGH-VOLTAGE-LEAKAGE NOISE TESTS

In response to an observation[4] that electronic noise bursts can be caused by moisture condensation in the high voltage (hv) section of the detector during liquid nitrogen (LN) temperature cycles, we performed a series of experiments to look for this problem. A low-temperature cycle of the sample in the detector can reduce the detector temperature so that moisture condensation might cause hv leakage on the signal line. This problem normally is prevented by the presence of desiccant in the hermetically sealed hv box. However, if there is an air leak into the box, the interior condensation can occur under humid air conditions.

For the tests, we directly applied steel pieces (~2 kg) that had been cooled by LN to the hv junction box and the detector body. The counts from the detector were collected for 8 to 12 hours as the system returned to room temperature. The cooling-warmup cycle was repeated about 10 times for the three ^{3}He-detector systems 1, 3, and 4 listed in Table IV.[5] Detectors 1 and 4 demonstrated no vulnerability to the noise tests. However, detector 3 gave intermittent noise bursts during the warm-up period, but the noise occurred only on humid days (rainy days). To enhance the problem, we placed detector system 3 in a plastic bag containing water to increase the relative humidity to ~100%.

TABLE IV. Neutron Detector Characteristics

Identification	Shape[a]	Size	No. of ^{3}He Tubes	^{3}He Pressure (atm)	Cavity Size (cm)	Total Efficiency[b] (%)	Singles Bkgd (s^{-1})	Coincidence Bkgd (h^{-1})
System 1	Rectangular	25 x 35 x 35 cm^3 channel	18	4	12 x 23 x 35	21	0.19	0.1
System 2	Cylindrical	23 cm ϕ x 37 cm cavity	6	4	5(diam) x 20	26	0.07	0.5
System 3	Cylindrical	22 cm ϕ x 35 cm cavity	16	6	9(diam) x 28	34	0.16	1.6
System 4	Cylindrical	22 cm ϕ x 35 cm cavity (inside ring)	16	4	9(diam) x 28	31	0.39	1.8
		(outside ring)	8	4	same	5	0.22	0.1

All of the noise events had a time-correlation count greater than 50, and the noise bursts did not satisfy the correlation timing relationship between the totals count and the coincidence gate count. This relationship is $R = N(N - 1)/2$, where R is the coincidence count and N is the totals count that occurs within the 128-μs coincidence gate. The cosmic-ray background rate for coincidence counts was 1-2 counts/h and no excess of small correlation events above background was observed in any of the detector systems during the tests.

We conclude from these tests that the ^{3}He systems can be vulnerable to hv noise from moisture condensation under temperature cycling in humid conditions. Detector systems that have effective air seals and drying agents such as desiccant are not subject to the problem.

Our newer detector configurations have the signal lines segmented to give independent readouts of ^{3}He detector banks and the ratio of the segments easily identifies hv-leakage noise events. However, some of our previous results[5] using detector system 3 were subject to this noise problem.

SAMPLE CHARACTERISTICS

In October 1990, we reported[6] neutron emission results using a consistent sample preparation procedure involving clean samples and high-purity gas preparation. Under these conditions, the titanium (Ti) samples would readily absorb deuterium gas after the oxide layer was breached. For the results included in this paper, we have tried a wider range of sample preparations and experimental procedures in an attempt to trigger the neutron emission by inducing sample disequilibrium with deuterium gas absorption or temperature change. We have attempted to duplicate the reported productive procedures of others as well as our previous[5] positive results. Most of the samples consisted of lathe chips of pure Ti metal; Ti alloyed with 6% aluminum, 6% vanadium, and 2% tin; or Ti alloyed with 6% aluminum and 4% vanadium. Some electrolysis residue samples were used. The cleaning procedure normally included multiple washes with methylene chloride, methanol, and water. For about one-third of the samples, the fill temperature was raised to 400-500°C to activate the Ti for deuterium absorption. For pure gas and clean sample conditions, the samples all absorbed deuterium gas after multiple LN temperature cycles. Nineteen samples containing Ti metal and deuterium gas were prepared during the period between November 1990 and March 1991 for the measurements at Los Alamos.

RESULTS

Of the 19 samples prepared during the current set of experiments, only two gave excess neutron emission above the background levels. Many procedural variations were tried including deuterium gas loading at high temperature (400 to 500°C), gas loading at low temperature (-100° to 2[illegible]°C), and temperature cycling from -197°C to 400°C inside the counting chamber. We gave a typical sample 10 to 20 LN temperature cycles before we stopped the measurements.

Sample DD-17. We measured the highest neutron emission from sample DD-17. This sample contained 304 g of Ti (6,6,2) contained in a 1-ℓ stainless steel (SS) sample bottle. The sample was degassed at a maximum of 230°C using helium to flush out the remaining air and cleaning agents.

During the neutron measurements, LN temperature cycles were performed with a small amount (1 to 4 ℓ) of D_2 gas being absorbed during the warmup from LN temperature. On the seventh LN cycle, 17 ℓ of D_2 were accidentally added to the sample while at LN temperature. About 1 h into the warmup, a portion of the Ti chips went into a hot exothermic reaction excursion when all of the gas was absorbed in about 15 s. A localized spot on one side of the SS bottle was hot; the rest of the bottle was still covered with frost. The bottle was immediately dunked into LN for 10 min and then removed from the LN and allowed to warm up in the detector.

During the first 2 h after the hot absorption, the sample emitted three bursts of neutrons as shown in Fig. 1. Detector system 4 has both inner and outer rings of ^{3}He detectors as shown in Fig. 2. The 16 inner tubes have a counting efficiency of 31% and the 8 outer tubes have an efficiency of 5%. The ratio for the inner/outer detector efficiency is 6.22 as measured with a ^{252}Cf source (2.3 MeV). The detectors have independent electronics. The collection time bins for the inner detector were 200 s long and the outer time bins were 10 000 s. The ratio of the excess neutron counts in inner/outer rings was 6.2 ± 2.1 that compares well with the calibration ratio of 6.22 for ^{252}Cf neutrons. Based on Monte Carlo calculations,[7] this ratio would be ~2.6 for 14-MeV source neutrons.

After three additional LN temperature cycles with little or no neutron emission above background, sample DD-17 was moved to the ^{3}He detector (system 1) that was located in the underground tunnel (70 m deep) at Los Alamos to obtain a higher counting sensitivity. The first LN cycle in detector 1 consisted of multiple short cycles where the sample was recooled in LN a total of five times during the 6-h warmup period. The cold sample at -197°C was filled with 8 ℓ of D_2 gas and the temperature and pressure were monitored during warmup. The observable gas absorption process began when the temperature reached ~-100°C and the absorption rate increased with temperature. After about 1.5 ℓ of gas were absorbed and the temperature reached -30 to -10°C, the sample bottle was dunked into LN for ~1 min to cool the sample below -100°C and stop the absorption process. This process was repeated five times, after which the sample was left in the detector for ~5 d of counting.

Figure 3 shows the neutron coincidence counts collected in detector system 1 where the first large burst came after the first short cycle. Two bursts were observed during the multiple LN cycles and the excess activity continued for ~50 h with 15 bursts as shown in Fig. 3. In addition to the large bursts, there was an excess of small time-correlated events in which only two neutrons were detected. The average control cell background rate in this detector is ~2 counts/d and during the excess activity period the doublet rate was three times higher than normal. Sample DD-17 was temperature cycled eight more times over a 30-d period with no further neutron emission above background levels.

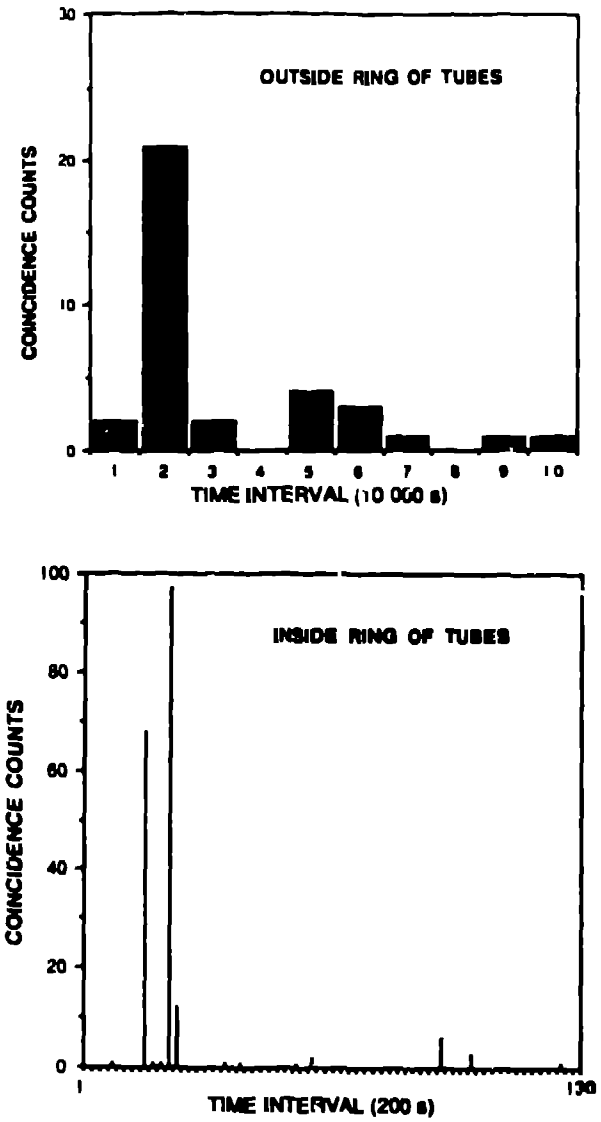

Fig. 1. Neutron coincidence results for sample DD-17 in detector system 4 where the counting intervals are 200 s for the inside ring of tubes (lower graph) and 10 000 s for the outside ring of tubes (upper graph).

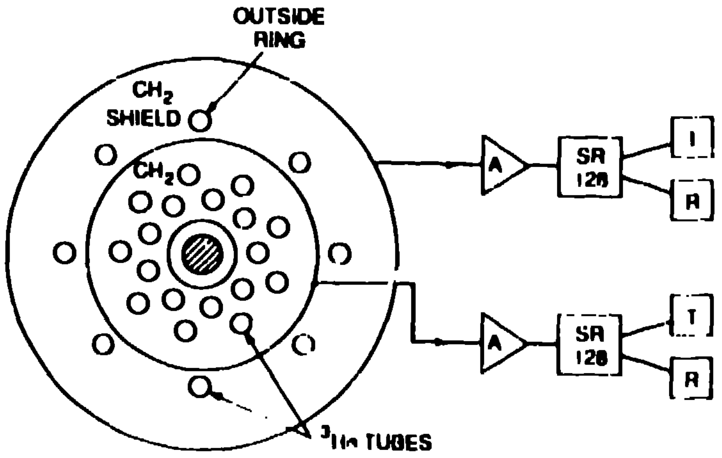

Fig. 2. Schematic diagram of detector system 4 showing the ^{3}He tubes and the signal processing electronics including the amplifiers (A), shift registers (SR), total scaler (T), and coincidence scaler (R). The inside and outside rings of ^{3}He tubes have independent electronics and the inside/outside count ratio is used as a consistency check.

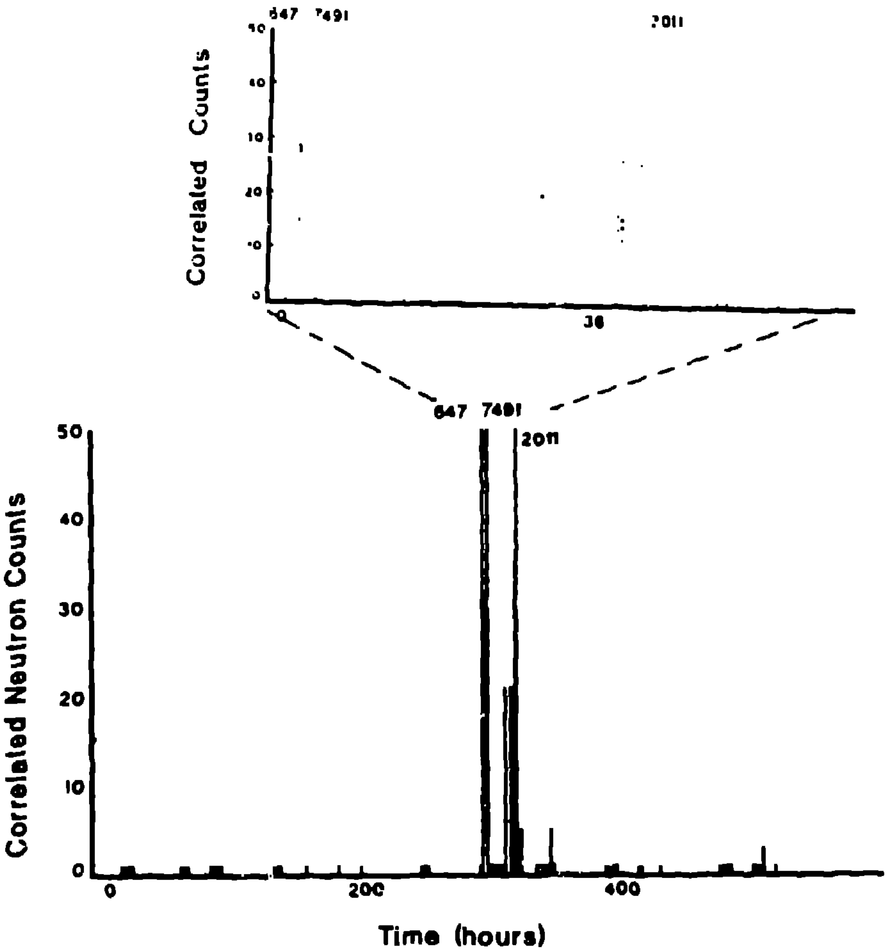

Fig. 3. Neutron coincidence results for sample DD-17 in detector system 1 where the counting intervals are 100 s. The top graph shows a time expansion of the active period. The 280 h of data prior to the active period correspond to the control sample or alternatively an inactive sample (DH-13) in the detector including LN temperature cycles.

Sample Ti-48. Excess neutron emission was observed from sample Ti-48 that contained 56 g of Ti metal and sponge in a 250-mℓ SS bottle. The Ti used in Ti-48 had previously been exposed to deuterium through D_2O electrolysis experiments. The Ti consisted of 35 g of sponge, 11 g of metal pieces, and 10 g of 1.5-mm-thick Ti plate with a thin layer of palladium deposited on one side. The sample was evacuated at 220°C and filled with 53 atm of deuterium gas. The gas pressure slowly decreased to 43 atm during the 90-d measurement period.

The measurements of sample Ti-48 were performed in detector 2 (see Table IV). Figure 4 shows the control runs for system 2 over a six-month period. Each data interval in Fig. 4 corresponds to the average of approximately 24 h of data collection. The control sample was a 300-mℓ SS bottle containing 100 g of Ti chips in air. Previous experiments[5] had demonstrated that control runs with air or H_2 gas gave the same results.

Figure 5 shows the data from sample Ti-48 in detector 2 where each data interval corresponds to the average coincidence rate for ~24 h of data collection. The control runs are shown interspersed between the sample runs. There are several days with excess neutron emission from sample Ti-48 with the highest day having an average yield of 1.12 counts/h

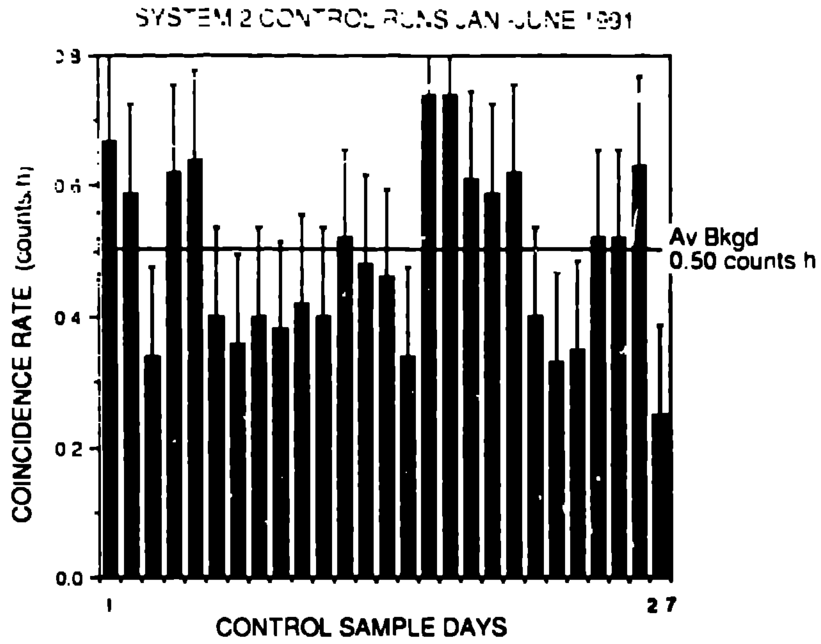

Fig. 4. Neutron coincidence background (control sample) rate in detector system 2. Each data interval corresponds to the average rate for ~24 h of counting. An LN cycle normally began the data interval.

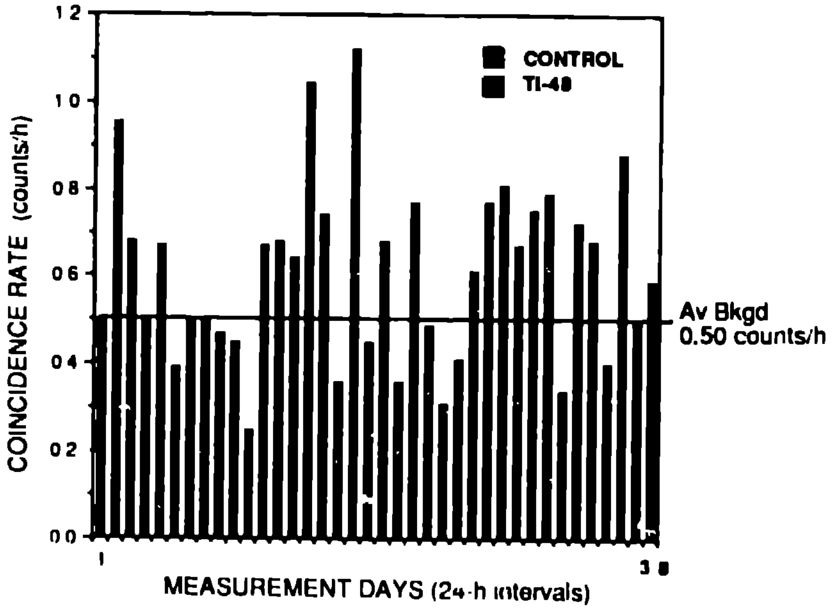

Fig. 5. Neutron coincidence results for sample Ti-48 in detector system 2. Each data interval corresponds to the average rate for ~24 h of counting. Most of the time intervals were initiated with an LN temperature cycle.

and a statistical significance of 3 σ. If we take the average of all the Ti-48 sample days and compare it to the control sample backgrounds, we obtain a 4-σ significance level.

SUMMARY

During the past two years, we have performed experiments to measure neutron emission from pressurized D_2 gas mixed with various forms of Ti metal chips and sponge. Our recent experiments have focused on increasing the anomalous low-level neutron emission. Thus far we have been unsuccessful in finding a way to trigger the emissions, although we have measured several samples that yielded excess neutrons above background. To improve our detection sensitivity, we have increased the shielding in our counting laboratory and we have located additional detector systems in deep underground counting stations.

Our overall detector efficiencies range from 20% to 44% for the four separate detector systems that are operating in parallel experiments. Two of the detector systems are segmented to provide separate signal outputs for a consistency check on the origin of the signals. Our coincidence background depends on the detector and shielding location and ranges from 2 counts/h to less than 0.5 counts/wk in the deep mine locations.

Only two of the 19 samples emitted excess neutrons during the current series of experiments; however, the excess yields were observed in three independent detector systems (detectors 1, 2, and 4). The neutron yield from sample DD-17 in detector 1 was several orders of magnitude above the control-run background levels, and the yield was the largest that we have observed during two years of experiments. This result was obtained in the low-background underground laboratory at Los Alamos.

Our search for a trigger mechanism for the neutron emission has been unsuccessful and our sample success rate is less now than it was one year ago. We think that part of the reason for the low success rate is that we have tried a large variation in sample types and experimental procedures. The number of experimental variables far exceeds our capacity to investigate the parameters.

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