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LA-UR 89-3946

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DEC 04 1989

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LA-UR--89-3946

DE90 003657

TITLE TRITIUM AND NEUTRON MEASUREMENTS OF A SOLID STATE CELL

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SUBMITTED TO NSF/EPRI WORKSHOP ON ANOMALOUS EFFECTS IN DEUTERATED MATERIALS
NATIONAL SCIENCE FOUNDATION/1800 G. St., N. W., Room 543
Washington, DC October 16-18, 1989

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TRITIUM AND NEUTRON MEASUREMENTS OF A SOLID STATE CELL

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Abstract:

A solid state "cold fusion" cell was constructed to test for non-equilibrium fusion in a solid. The stimulus for the design was the hypothesis that the electrochemical surface layer in the Pons-Fleischmann cell could be replaced with a metal-insulator-semiconductor (MIS) barrier. Cells were constructed of alternating layers of palladium and silicon powders pressed into a ceramic form and exposed to deuterium gas at 110 psia resulting in a D/Pd ratio of 0.7. Pulses of current were passed through the cells to populate non-equilibrium states at the MIS barriers. One cell showed neutron activity and was found to have a large amount of tritium, other cells have produced tritium at a low rate consistent with neutron emission below the threshold of observability. The branching ratio for n/p has been about 1×10^{-3} in all the experiments where a substantial amount of tritium has been found.

Introduction:

Recent experiments ^{1,2,3,4} have indicated that electrochemical cells are producing neutrons, tritium, and perhaps heat via an unknown new "cold fusion" process. These cells seem to be remarkably sensitive to palladium surface preparation, electrolyte impurities and exact cell configuration. It is reasonable to assume that the fusion mechanism is a near-surface phenomenon dominated by the high concentration of deuterium electrically driven into the material. It is known ⁵ that electrochemical surface barriers are similar (electrically) to the barrier at metal-semiconductor or metal-insulator-semiconductor contacts. Therefore, it was decided to try and fabricate a MIS device from palladium and slightly oxidized silicon. To achieve a large surface area, palladium and silicon powders were used in a pressed configuration.

Apparatus:

The configuration of the cell is shown schematically in Figure one. The layers were pressed into the ceramic form at a pressure of 11.2 MPa resulting a density of 26% dense material for the palladium and 75% dense material for the silicon. The Belleville washers at each end maintain a constant pressure of 3.3 MPa as the palladium swells during deuteriding. Deuterium gas at a pressure of 0.76 MPa was used to deuteride the samples resulting in a D/Pd. ratio of 0.72. Table one lists the gas analysis of the three bottles that were used to fill the cells as well as the analysis of the major impurities in the silicon and palladium. Both the silicon and palladium powders are coated with an oxide layer which is thought to be

important. The silicon oxide layer is approximately 150 Å thick. Figure 2 shows the unusual morphology of the palladium powder (formed during precipitation) and also the silicon powder size and shape. After pressing, the powder is as shown in figure 3.

A voltage current plot of the cell indicates that the resistance of the cell is primarily controlled by the silicon oxide layer. Shown in Figure 4 are the V-I curves, for cells 10 and 12. Typically the curves are highly non linear suggesting the current is due to tunnelling or a variety of other conduction mechanisms⁶ that have been observed in MIS devices. The V-I curve is taken under pulsed conditions so as not to heat the cell by more than a few degrees.

To enhance the detection of the neutrons and to obtain the highest current densities possible, a pulsed excitation source was used in all the experiments. The voltage and current pulse used was of 1 microsecond to 1 millisecond in duration at up to 3000 V at currents as high as 0.5 amp with a low duty cycle (such as 10 milliseconds) to reduce joule heating. Usually the pulse width and duty cycle were adjusted so that one watt of power was dissipated in the cell, however, the peak power was often 100 or more watts. This limited the joule heating to a few degrees resulting in little change in the average D/Pd ratio. The neutron detector consisted of a bank of 15 helium 3 proportional counters moderated by polyethylene. The efficiency of this system as measured with a ²⁵²Cf source was 1.3%. Instead of energy discrimination, time correlation was used. The pulse to the cell triggered the time of flight electronics of the Low-Q diffractometer at LANSCE. This was intended to give a concurrent background measurement by recording counts well after the current pulses. A schematic of the experimental apparatus is shown in Figure 5.

To test for tritium by-products, a gas line was constructed using an ionization gauge as the measurement device. These devices reject radon and other ionization mechanism that do not produce ionization characteristic of 18.6 KeV electrons. A schematic of the system is shown in Figure 6. The system is used to pump down and fill the cell as well as measure the tritium in the deuterium gas initially and after operation of the cell.

Results:

Eleven cells have been built to date. The most positive results were obtained on cell 2, which was pulsed with 500 to 2000 V at 900 to 25 microsecond pulse widths with pulse rates of 80 Hz or less. The input power from joule heating was usually held at 500 to 2000 milliwatts so that the D/Pd ratio would not change significantly. At one watt the cell heated to about 6 C above ambient in the neutron counter cave.

The neutron counting results did not show any definite time correlation on time scales shorter than the pulse repetition rate. Instead of time correlation, it was found that the count rate when the sample was pulsed was higher than the background with or without the cell or when a resistor was pulsed. Measurements taken early in the life of the cell gave higher neutron counts than those taken five days later. This is shown most clearly in Figure 7 where the neutron count has been roughly correlated

with the total energy dissipation in the sample. After the cell was instrumented for temperature, a long measurement was started and the results are as shown in Figure 8. The temperature of the cell, ambient temperature and the power input were measured as well as the neutron count. While the neutron count rate, delta temperature and input power seem to be somewhat correlated, only the delta temperature and the input power are even approximately consistent. While the neutron data may look suggestive, and many precautions were taken to exclude noise, the possibility that electrical noise from the pulser was causing spurious counts in a stochastic manner cannot be completely discounted.

Cells 1,3,4 and 5, which were controls or had mechanical faults, showed no neutron activity, and were not analyzed for tritium. Cell two was kept in storage since it showed definite neutron activity. The subsequent tritium analysis showed that cell 2 had 1300 times the fill gas concentration of tritium. This amounts to 3.5×10^{15} atoms of tritium. While this is a considerable level over background, it only amounts to 65 ppb.

Since both neutrons and tritium were produced and measured, the branching ratio (n/p) may be estimated. An upper limit of 2.7×10^{-9} may be set for this ratio. This is surprisingly close to other data^{7,8} (1×10^{-8} to 1×10^{-9}). In this work, the branching ratio was calculated from the gas left in the cell, but, some of the gas was used for a ^4He analysis prior to the tritium analysis. A correction for the lost gas would bring the branching ratio down to 1×10^{-9} .

A total of 6×10^5 joules were dissipated in the cell. The production of 3.5×10^{15} tritium atoms would have yielded 2200 joules. The efficiency is therefore only 0.3%, however, it should be noted that most of the voltage drop occurs at Si-SiO₂-Si interfaces and only a fraction of the applied voltage is at the Si²-SiO₂-Pd interface. Taking this into account, the efficiency of the surface² layers may have been closer to 20-64%. At this level of efficiency the cell would be highly economic.

Because of these very positive and encouraging results, an attempt was made to reproduce the data in another facility. Specifically, a corroborating measurement of the branching ratio was desired. A noise insensitive high efficiency (21%) channel counter was available at Los Alamos Group N-1 for this purpose. This counter has eighteen ^3He tubes and a background of 850 to 900 counts per hour (4050 to 4280 n/hr) although about half of the background is due to radioactive decay intrinsic to the tube materials. It should be noted that the LANSCE system also had a similar background of about 62 c/h (4770 n/h). At LANSCE, however, the electronics were set to reject most gamma induced background.

Very few, if any, neutrons have been detected at N-1. Some typical plots of the backgrounds are shown in figure 9 a,b,c. These neutron generation rates are far lower than seen with sample 2. Sample 9 showed no tritium, while sample 10 generated enough tritium to be easily measurable and gave a branching ratio that was similar to that of cell 2; however, because the neutron counting statistics are so poor this value is only approximate.

Since sample 10 appeared to be neutron active, the excess tritium was flushed from the sample and it was run at high voltage with very short (5 microsecond) pulses for 140 hours. No excess tritium was found. Then the sample was flushed with fresh deuterium and run for 92 hours at a pulse width of 300 microseconds. The background level was found to be 1.5 times the level of the gas bottle. A subsequent run at 300 microseconds for 160 hours gave a very slight excess over background (20%) and gave two neutron bursts of similar magnitude to the gas cell work of Menlove et. al.¹⁰. These two bursts were 25 and 35 neutron events in a 100 second count. This type of event is statistically quite improbable ($> 5 \sigma$). None of the other counters within a few meters of the sample responded, eliminating extraneous factors. Both bursts occurred within 10 minutes of turning the power off to the sample. Sample 11 was made identically to sample 10, however, it had leaked about 2/3 of the D_2 gas inventory between the time it was filled and analyzed. It was found to have about 4 microcuries/meter cubed of excess tritium, far lower than the 130 microcuries/meter cubed found in cell 10.

Table 2 lists all the cells made to date and summarizes the tritium and neutron measurements. As shown in table 2, a hydrogen control cell was made that showed no neutron production over background when used with the LANSCE counter. While the construction of the cells appears simple, slight variations in pressing pressure, thickness of the layers, oxide layer on the silicon and the exact fabrication details can have large effects on the electrical properties and concomitant effects on the production of tritium.

Conclusion:

While the exact mechanism for fusion in the solid state is not known, this work has helped clarify the situation by virtue of another measurement the branching ratio for cold fusion. An upper limit on the ratio, from this work, is 3×10^{-5} .

The main explanations for D-D fusion in the solid state are microcracking resulting in hot fusion, and various other explanations based on the enhancement of tunnelling due to increased electron shielding resulting from high local lattice pressures (piezofusion), non-equilibrium electron concentration (perhaps at defects) and high D/Pd concentrations. Since all reported measurements indicate a very low branching ratio, the cracking hypothesis is discredited. Since no time correlation was observed with electron injection one can tentatively dismiss the idea of non-equilibrium electron aided fusion due to injected conduction band electrons. This still admits the possibility of some site specific fusion where the local electron concentration is high. The idea of piezofusion or locally high lattice pressures caused by phase transitions or deuterium drift cannot be discounted.

Solid state fusion cells appear to be a viable alternative to the electrochemical cell approach to the production of tritium. Due to the simplicity of the cell and the possibility of creating monolithic structures of Pd and Si there exists considerable room for improvement in efficiency. Based on the power input and the amount of tritium produced in cell 2, it has been

calculated that a factor of 10 improvement in efficiency would result in economic tritium production ¹¹. The reduction in thickness of the Si layers, the elimination of the hydrogen in the deuterium and palladium and increasing the gas pressure of the cell would almost certainly improve the efficiency.

Acknowledgements:

Special thanks to two Los Alamos Fellows, H. Sheinberg and H. Menlove for their participation in this work. Also, this work would not have been possible without the special abilities of our fine technical staff of K. Greichen, W. Ely and J. Ortega.

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Table I. Material analysis of the deuterium, hydrogen and the palladium and silicon used in the experiments. The gas analysis was done with a mass spectrometer and a tritium ionization gauge, the palladium and silicon were analyzed by wet chemistry.

Gas Analysis

Gas Bottle No.	deuterium	hydrogen	tritium	helium 4
Deuterium Bottle 1:	to be analyzed	tba	110	15 ppm
Deuterium Bottle 2:	99.34	0.66	28	tba
Deuterium Bottle 3:	99.33	0.67	20	tba
Hydrogen Bottle 1:	tba	tba	tba	tba

Palladium Analysis	(ug/g)	(Engelhard)
Oxygen	928	
Nitrogen	65	
Carbon	47	
Chlorine	80	
Trace Metals	127	

Silicon Analysis
To be analyzed

Note : Deuterium and hydrogen in terms of mole %, tritium in terms of micro curies per meter cubed.

Table II. Summary of solid state fusion experiments to date.

Sample Number	Date Built	Neutron Output	Tritium Produced	Hours Run	Cell Type	Fill Gas	Notes on Cell
1	12-Apr	No	Not Analyzed	3	O-Ring, Layered	D2, #1	Sample shorted out
2	14-Apr	Yes, >5 sigma	Yes, 1300 x background	96	O-Ring, Layered	D2, #1	5% Hydrogen added to cell on 4-26
3	19-Apr	No	Not Analyzed	20	O-Ring, Layered	H2, #1	Hydrogen control
4	3-May	No Steady Output	Not Analyzed	<1	O-Ring, Layered	D2, #1	Leaky seal
5	8-May	No	Not Analyzed	<1	O-Ring, Layered	D2, #1	Leaky seal
6	30-May	No, <2 sigma	No	17	O-Ring, Layered-Mixed	D2, #1	Sample shorted out
7	18-Jul	Yes, >2.5 sigma	Yes, 1.5 x background	95	O-Ring, Layered-Mixed	D2, #1	Neutron output varied with voltage
8	9-Aug	No, <1 sigma	No	142	Flange, Mixed	D2, #2	Palladium and silicon powders mixed together
9	14-Aug	No	No	63	Flange, Mixed	D2, #1	Same as 8, but outgassed at 110°C
10	4-Sep	Yes?, <1 sigma	Yes, 4.4 x background	77	Flange, Layered	D2, #2	Neutron bursts, 250 psi fill
11	11-Oct	No	Yes, <20%	162	Flange, Layered	D2, #3	Same as 10 but 70 psi fill
12	8-Nov	in progress			Flange, Layered	D2, #3	Cell can withstand 2000 psi
Sample 2 produced $9.5 \times E6$ neutrons and $3.5 \times E15$ tritium atoms						Branching ratio $n/p = 2.7 \times E-9$, see note one below	
Sample 7 was not monitored for neutrons on a continuous basis, but produced $1 \times E12$ atoms of tritium							
Sample 10 produced 5200 neutrons \pm 7500 and $9.6 \times E12$ atoms of tritium						Branching ratio $n/p = 0.5 \pm 0.8 \times E-9$	
Note 1: A substantial amount of tritium was lost prior to analysis due to leakage from o-ring seal. Correction of the branching ratio for the loss of tritium would decrease the number to $1 \times E-9$.							
Note 2: Gas analysis and material purity listed in Table I.							



CERAMIC ID. 1.25 in.
 TOTAL ACTIVE LENGTH 0.44 in.
 THICKNESS OF Pd LAYERS 0.047 in.
 THICKNESS OF SI LAYERS 0.085 in.

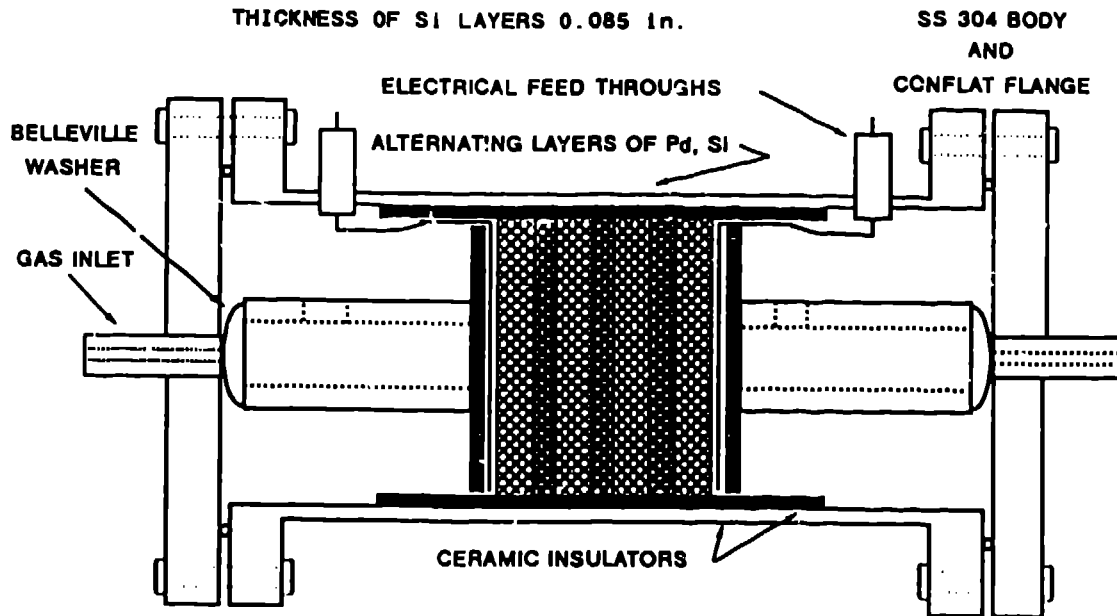


Figure 1. Photo of the experimental cell, and a cross section of the cell showing the layers and the electrical connections.

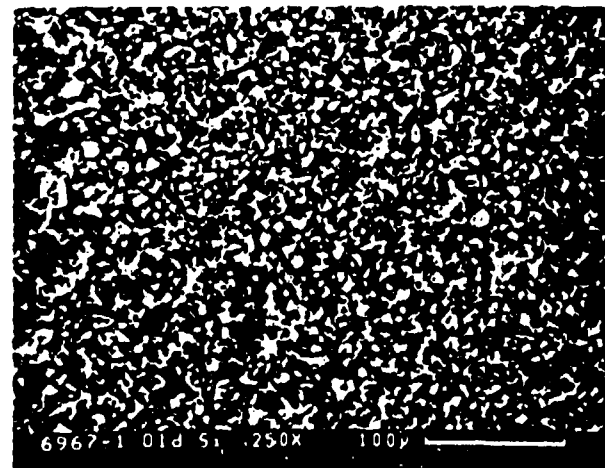
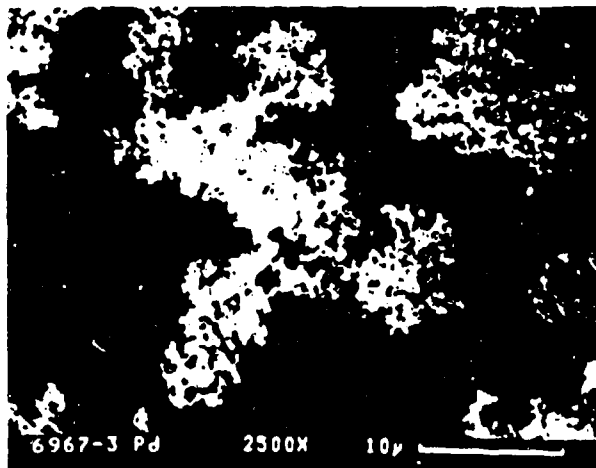
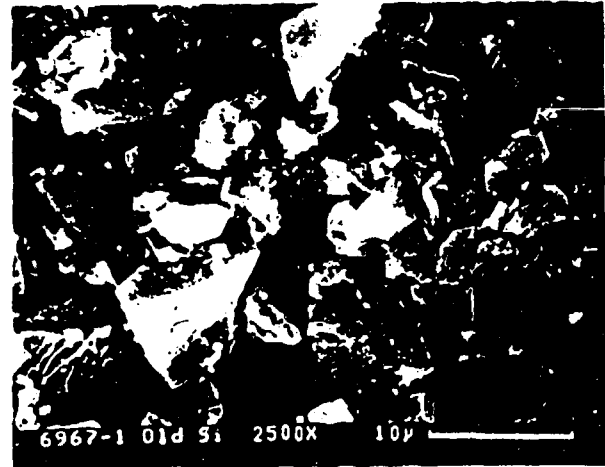
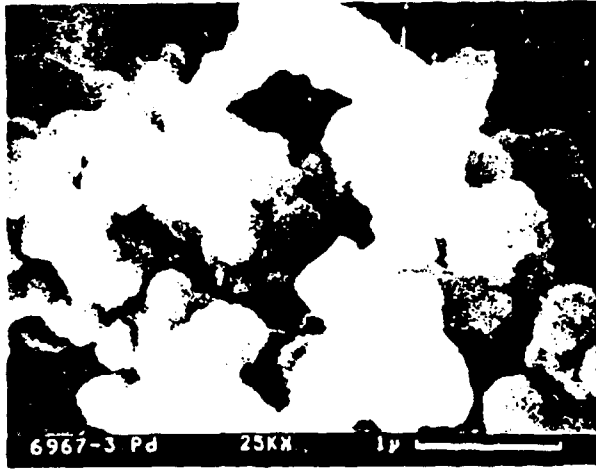


Figure 2. SEM photographs of the palladium and silicon powders used in the cells. The palladium is produced from a precipitation process that leaves the surface coated with an oxide layer. The silicon is ball milled from ingots.

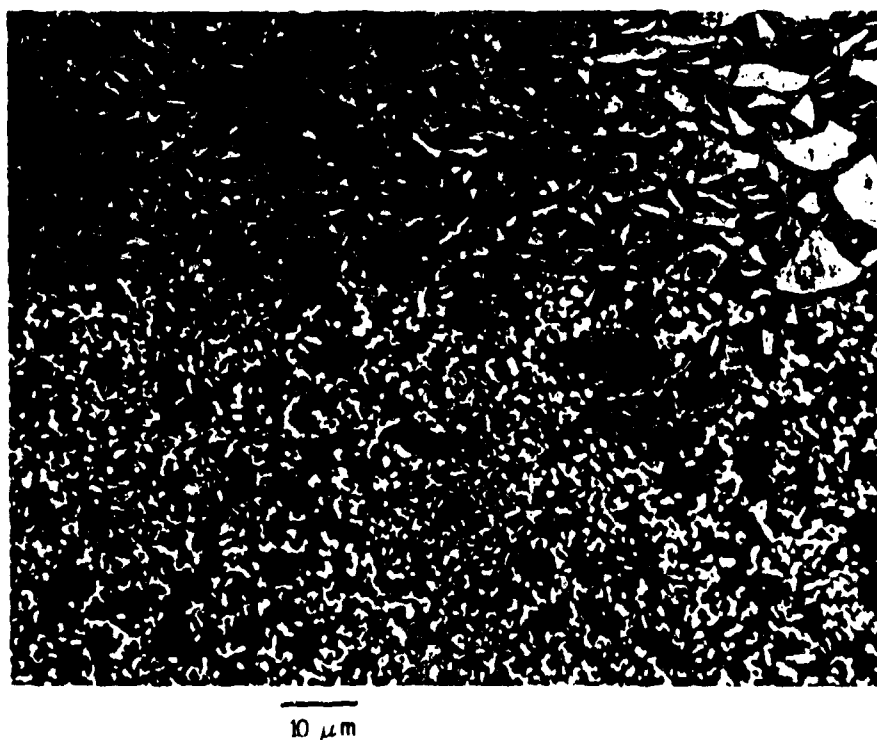
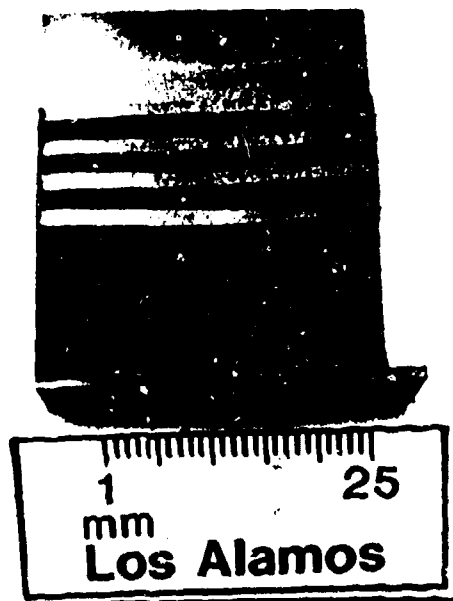


Figure 3. Photo of a cross section of a pressing showing the layered structure of the compact. The photomicrograph shows a boundary between the palladium (lower) and the silicon.

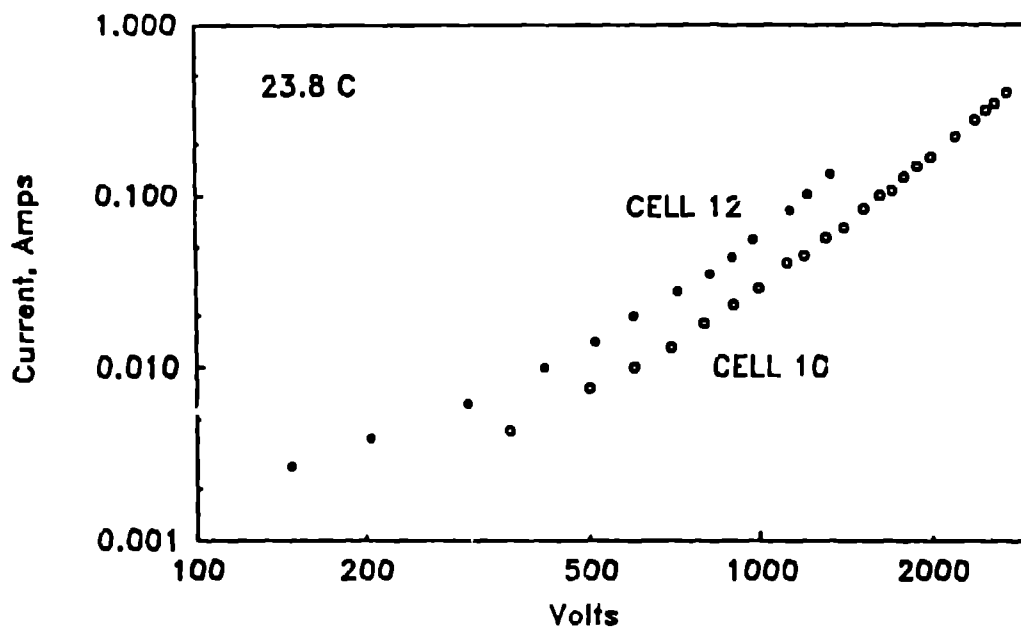


Figure 4. Voltage versus current plots for cell 10 and 12. Slight differences in pressing pressure and silicon thickness have great influence on the electrical properties.

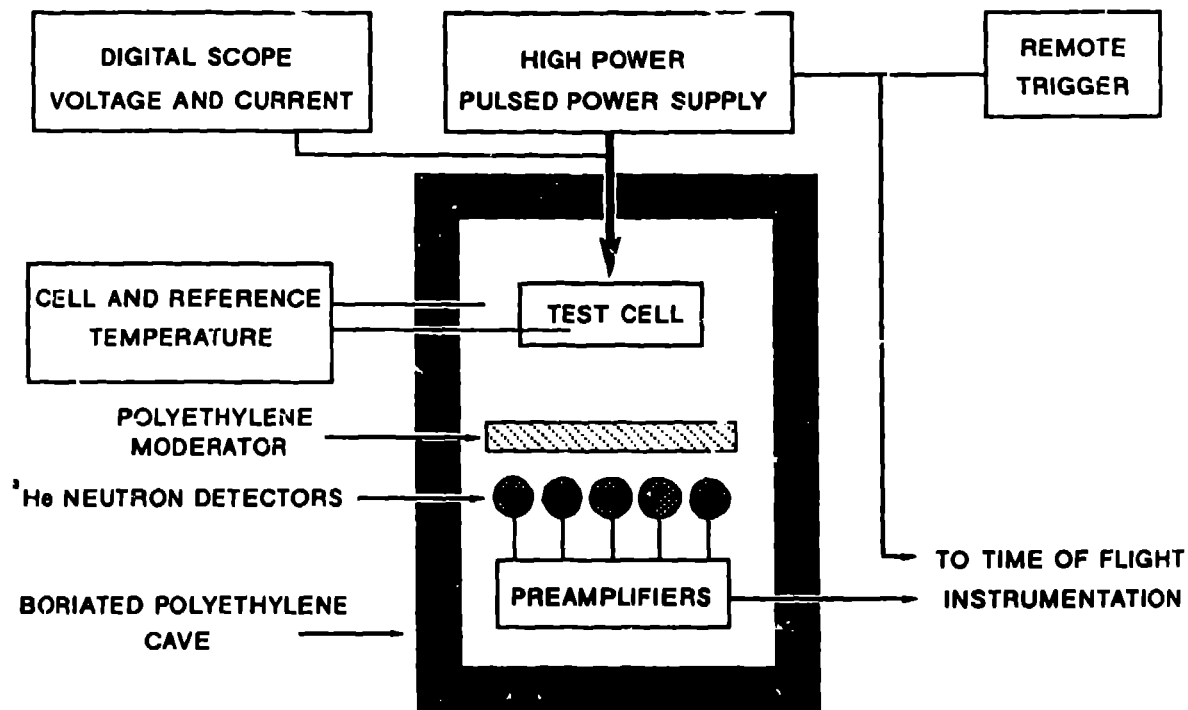


Figure 5. A schematic of the test system for the measurement of neutron emission at LANSCE.

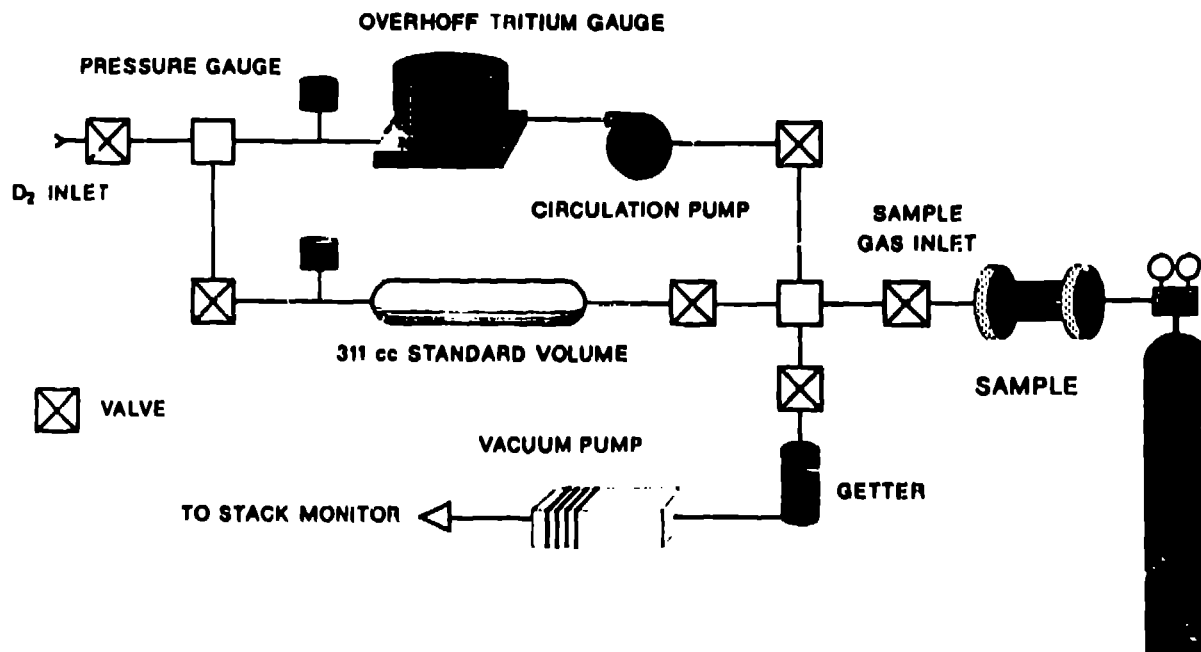


Figure 6. A schematic of the deuterium fill and analysis system. The sample is evacuated and then filled with deuterium from a cylinder. The evacuated analysis system is then filled with the deuterium from the cell. This allows an analysis of the initial trace amounts of tritium present in the gas, cell or palladium.

COUNTS PER HOUR VERSUS CUMULATIVE ENERGY DISSIPATION

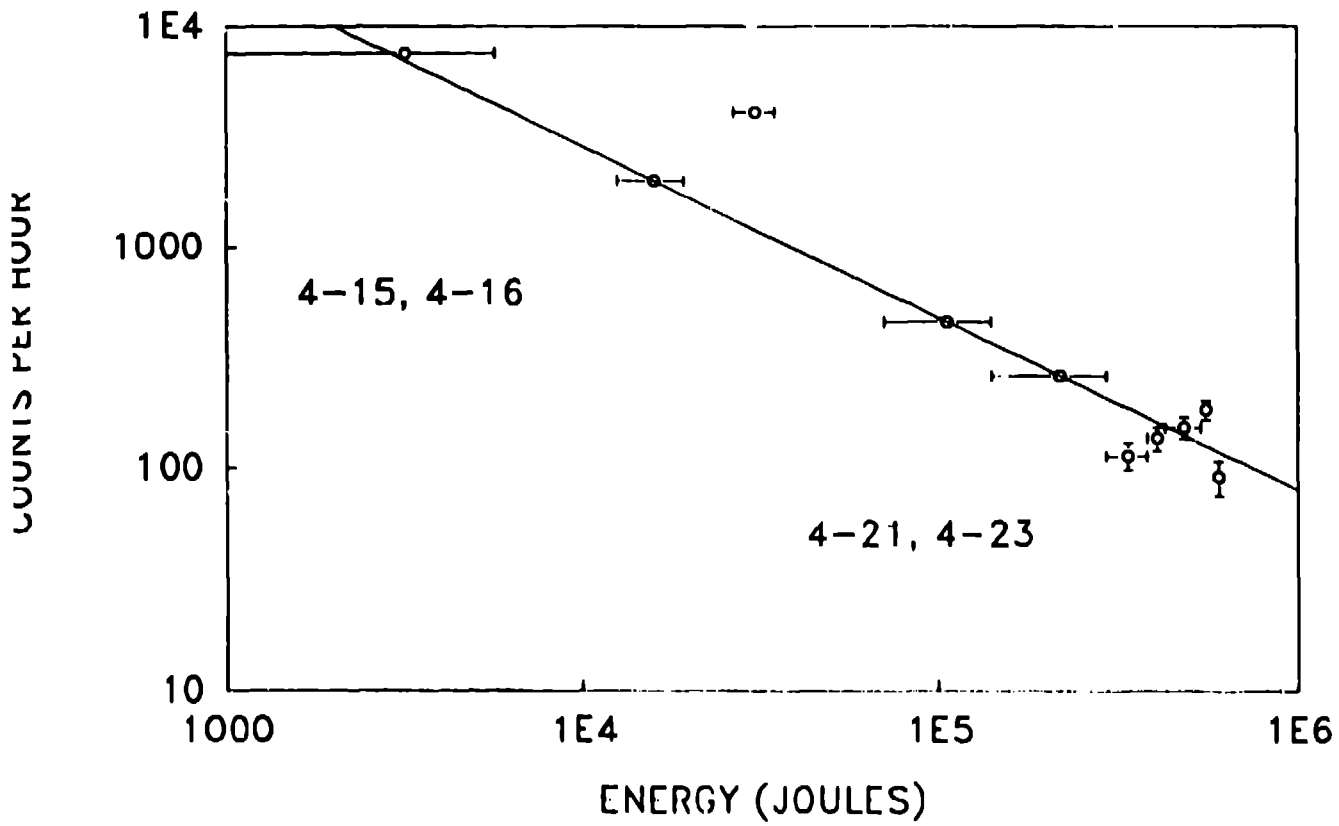


Figure 7. Cell two showed a decreasing neutron output as the cell was operated.

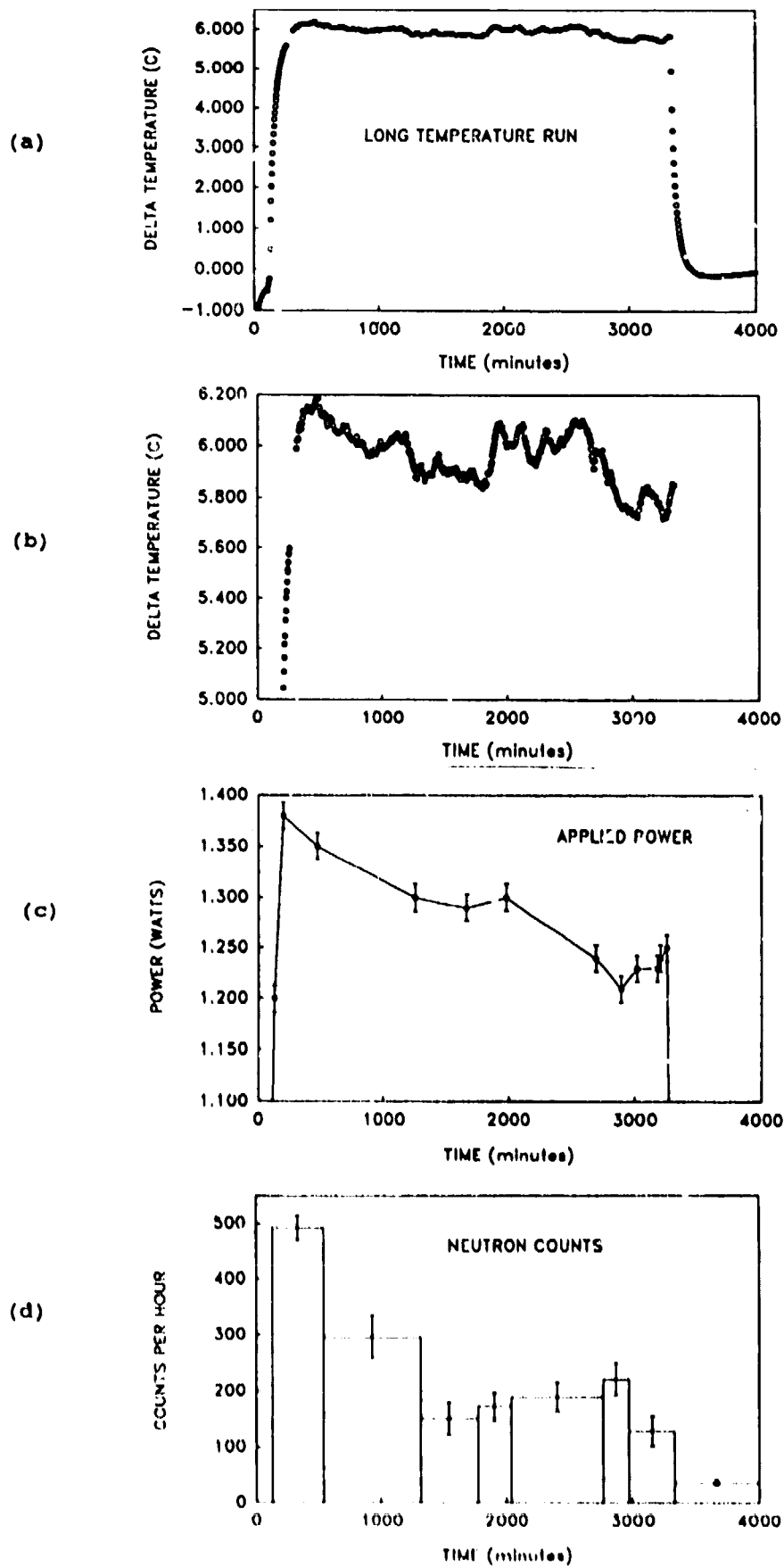


Figure 8. Results from sample 2 during the 55 hour heating and neutron measurement taken over the weekend of 4-21, 23. Curves a and b show the difference between the ambient and cell temperature. The curves roughly correlate with the applied electrical power shown in c. The neutron count rate decreased during the measurement period and was considerable above the background count which is shown as the last bin in d.

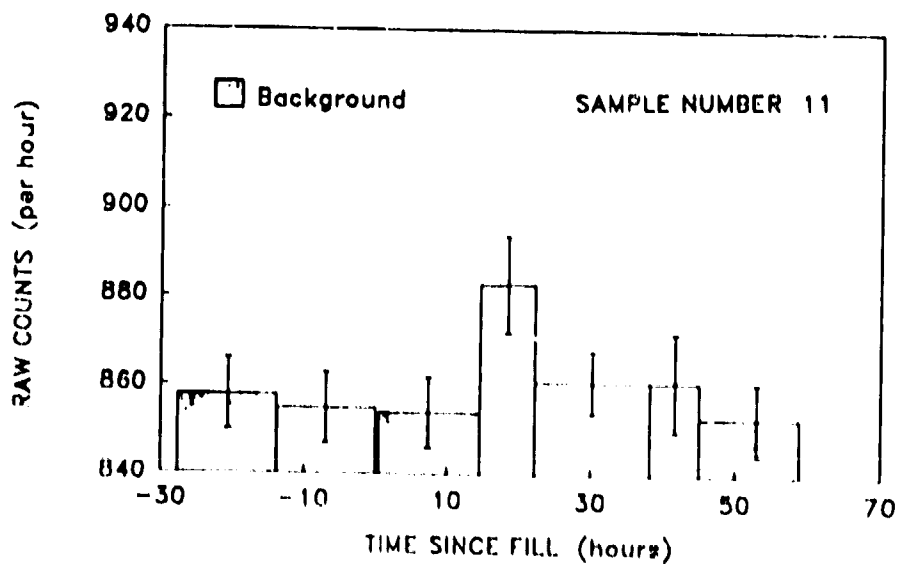
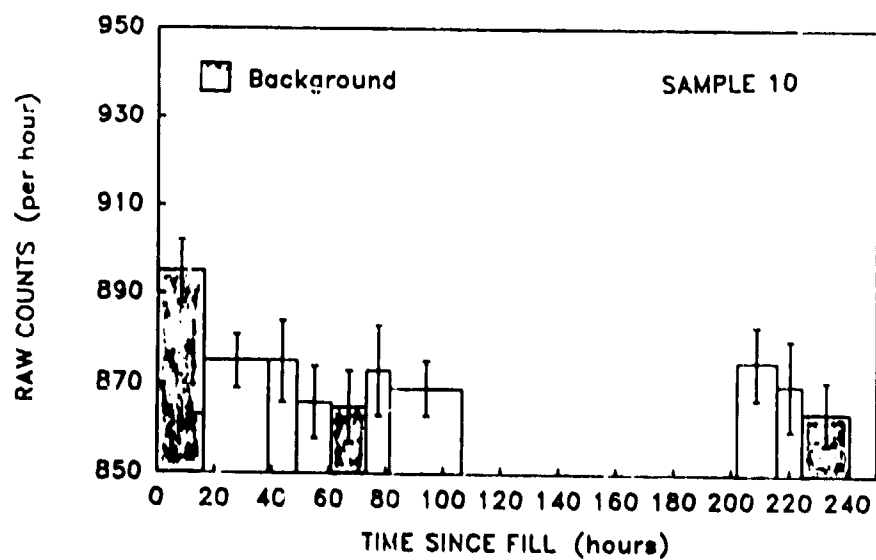
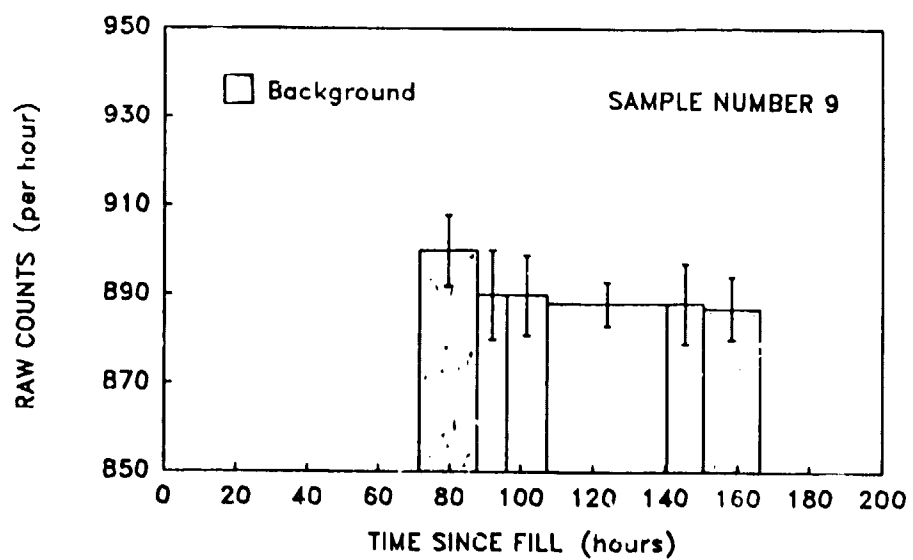


Figure 9. Recent neutron results from Menlove's counter. With the exception of the bursts recorded from cell 10, the neutron output has been at the statistical level which is consistent with the tritium measurements, given the branching ratio observed for cell 2.