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TRITIUM AND NEUTRON MEASUREMENTS FROM DEUTERATED Pd-Si

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ABSTRACT

Evidence has been found for tritium and neutron production in palladium and silicon stacks when pulsed with a high electric current. These palladium-silicon stacks consist of alternating layers of pressed palladium and silicon powder. A pulsed high electric current is thought to promote non equilibrium conditions important for tritium and neutron production. More than 2000 hours of neutron counting time has been accumulated in a underground, low background, environment with high efficiency counters (21%). Neutron emission has occurred as infrequent bursts or as low level emission lasting for up to 20 hours. In eight of 30 cells, excess tritium greater than 3 sigma has been observed. In each of these measurements, with the powder system, the ratio of tritium detected to total integrated total neutrons inferred has been anomalously high. Recent cells have shown reproducible tritium generation at a level of about 0.5 nCi/hr. Several hydrogen and air control cells have been run with no anomalous excess tritium or neutron emission above background. A significant amount of the total palladium inventory (18%) has been checked for tritium contamination by three independent means.

INTRODUCTION

Recent experiments have suggested that electrochemical cells are producing neutrons[1,2] tritium[3,4], and perhaps heat[5] via an elusive and unknown new "cold fusion" process in a metal lattice. These cells seem to be remarkably sensitive to palladium surface preparation, electrolyte impurities and exact cell configuration. Since finely powdered palladium exchanges isotopes of hydrogen reversibly and can be nearly completely dehydrided at modest temperatures, we have searched for excess tritium in this system. In contrast to most electrochemical cells, the system used in these experiments is completely sealed during operation and uses no electrolyte. Some of the preliminary results, on this system, were presented earlier at the NSF-EPRI workshop[6].

The results presented here describe in more detail the hydriding and dehydriding and the analysis for tritium contamination in the palladium. In addition, recently, progress has been made in achieving a small but reproducible tritium yield from cells of a particular type. The search for neutron emission from

cold fusion seems to show several common characteristics between different research groups. however, neutron data obtained at the surface of the earth does not seem convincing due to the high spallation rate or the fact that a veto detector has to be employed to reduce background as well as certain other effects enumerated by Anderson et al.[7]. A search for neutron emission in a well shielded underground counting laboratory was therefore initiated with a high efficiency ^{3}He neutron counter.

MATERIALS

Deuterium gas at a pressure of 0.76 MPa or greater was used to deuteride the palladium powder samples resulting in a D/Pd ratio of at least 0.72. In these experiments, no attempt was made to purify the deuterium by cold trapping or isotopic separation of the deuterium from the hydrogen. Several bottles of deuterium were used, but to enhance the effects and to reduce uncertainly in the tritium measurements, most of the cells were deuterided with bottles that had the lowest tritium levels (17-28 μCi/m^3) available at that time. Table I summarizes major impurities in the deuterium gas as well as that in the silicon and palladium powders.

Gas Analysis					
Gas Bottle No.	deuterium	hydrogen	tritium	helium 4	other (all bottles)
Deuterium Bottles:	99.2	0.6	17-110	15 ppm	H2O, O2, CO, CO2, N2 < 0.1%
Hydrogen Bottle:	< 0.1%	99.9	< 1	15+5	same as above
Palladium Analysis	μg/g	(Engelhard)			
Oxygen	928				
Nitrogen	65				
Carbon	47				
Chlorine	80				
Ag = 15, Fe = 20, Pt = 35, Zn = 10, Au = 30, SI = 15, B = 15, Pb all others < 10 ppr wt					
Tritium < 0.02 nCi/gr					
Silicon Analysis	μg/g	(Union Carbide)			
major impurities					
Chromium	600				
Iron	800				
Lanthanum	900-3000				
Tritium < 0.1 nCi/gr (none detected)					
Note Deuterium and hydrogen in terms of mole %. tritium in terms of micro curies per meter cubed.					

Table I. The gas analysis was done by mass spectrometer and a tritium ionization gauge. The palladium and silicon were analyzed by wet chemistry and spectrochemical methods.

The palladium powder was obtained from Engelhard and formed by precipitation from an aqueous solution of Pd(NH$_3$)$_4$Cl$_2$ using reagent quality

chemicals. The raw material was said to be virgin sponge obtained from a South African mine. A total of 468.8 g of palladium has been used in the experiments described in this paper, of that amount, 87.3 g was used in various control experiments for tritium analysis.

Figure 1 shows the unusual morphology of the palladium powder and also the silicon powder size and shape. The palladium is composed of small spheres (0.3 to 0.5 Å) in diameter which are welded together at necks to form chains or agglomerates that are up to 30 μ in diameter. The silicon powder size distribution analysis shows that the largest diameter is roughly 20 μ with the peak of the distribution occurring at 7 μ equivalent diameter. A typical cell, made with powders, might contain 12 to 21 grams of palladium in eight layers (one to two grams per layer) and 6 to 8 grams of silicon distributed between seven layers. Silicon layers are typically 0.76 to 2.15 mm thick by 3.17 cm in dia. while the palladium layers vary from 1.16 to 0.48 mm thick by 3.05 cm in dia. for different type cells. Both the silicon and palladium powders were coated with an oxide layer which is thought to be important. The palladium powder was pressed into disk form and then oxidized, in air, at 350 C for 2 hours. The silicon oxide layer is approximately 10 to 20 Å thick as determined by Auger analysis.
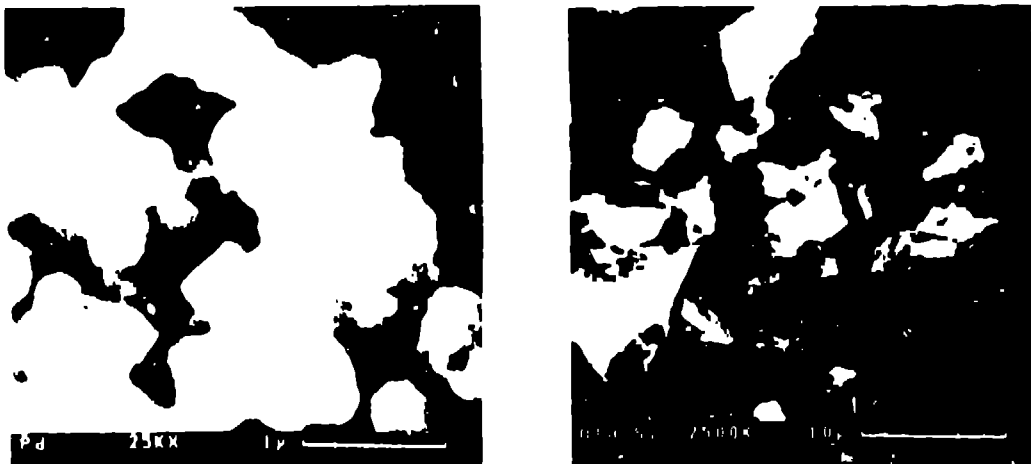

Figure 1. SEM photographs of the palladium (left) and silicon (right) powders used in the cells.

Palladium foil (47.5 and 220 microns thick), as received from Johnson and Matthey, was laser cut into disks and then annealed at 850 C for 2 hours at 10^6 torr. Subsequent to this treatment the foils were oxidized in air, on one surface only, at 800 C for 2 hours. In some cells, Sb doped silicon wafers obtained from Monsanto were used. These wafers were 0.01 ohm-cm in resistivity by 0.5 mm thick disks, 3.07 cm dia..

Only two bottles each of palladium and silicon powder have been used for the experiments, to date, and both have been analyzed for trace amounts of tritium. Three methods for analyzing for tritium were used, first the palladium powder was hydrided (some was also deuterided) in a closed chamber and then

dehydrided into a tritium gas analysis system such that all but 5% of the gas was recovered. This yielded a tritium background level of 0.3 nCi/g (determined by the error in the measurement). The second method involved suspending a small amount of powder (50 mg) in a scintillation gel. No detectible counts over background could be detected by this method. The third, and most sensitive, method involved the dissolution of approximately 200 mg of palladium powder in aqua regia and the subsequent neutralization and counting in a scintillation cocktail. No counts over background could be detected by this method, resulting in a upper limit of contamination of 0.02 nCi/g [8].

Added to the silicon powder was 3% (wt) of either PVA or Dow XUS 40303 binder. These binders and ethanol solvents were tested for tritium contamination by dissolving 132 to 460 mg of binder and solvent in water and placing the resultant mixture in a scintillation cocktail. No tritium could be detected over background by this method. In addition, no counts over background could be detected when the silicon powder (44 mg) was suspended in a scintillation gel and counted.

APPARATUS

To detect tritium, a recirculating gas line was constructed using an ionization gauge as the measurement device. Gas ionization gauges are used extensively in the detection of tritium and have been shown to be stable, reliable and sensitive[9]. The instrument rejects pulse type radioactive events which effectively discriminate against radon and cosmic ray ionization. Due to the small amount of tritium in the deuterium that was used in the experiments, the instrument was operated on the most sensitive range. Despite this, the instrument showed good stability in measuring the tritium background in the deuterium gas (maximum deviation during a year of ± 2.5%).

A schematic of the system is shown in Figure 2. The system is used to evacuate and fill the cell as well as measure the tritium in the deuterium gas initially and after operation of the cell. The total analysis system volume is 2065 cc which includes a 310.9 cc calibration volume. A pump is used to circulate the gas through the loop to ensure good mixing.

Figure 3 shows the linearity of the instrument to two concentrations of tritium in deuterium as a function of chamber pressure. Offsets near zero pressure correspond to ionization currents caused by small amounts of adsorbed tritium on the chamber walls. Absolute calibration was accomplished by inserting the chamber into a circulating loop containing a standard, calibrated ionization gauge while circulating various concentrations of tritium enriched deuterium. Calibration was performed at twelve points from 498 $\mu Ci/m^3$ to 114 $\mu Ci/m^3$. Sensitivity to air contamination was checked by comparing the response of the meter to 600 torr of deuterium with 21 $\mu Ci/m^3$ of tritium to that of 600 torr of deuterium with 10, 20, 50 and 100 torr of air added to the loop. The readings were identical with or without air to within experimental error, ± 1 $\mu Ci/m^3$. Also, the response of the meter to small concentrations of water vapor was measured
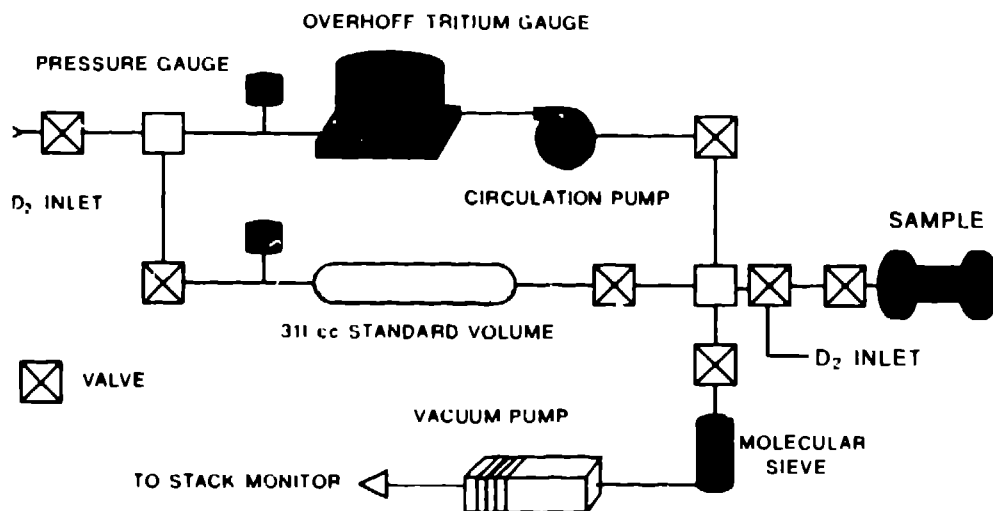
Figure 2. A schematic of the deuterium fill and analysis system. The gas is circulated in the loop during analysis.

to determine if this could give anomalous readings. Up to the equivalent of 38 torr of water vapor, in deuterium, was introduced into the loop resulting in no change to the readings, suggesting that at 38 torr the addition of water had a very small positive effect on the instrument. As this was three times the amount of water contained in a typical cell, the effect of water vapor is thought to be negligible. Very high concentrations of water vapor had the effect of suppressing the output as the instrument suffered from intermittent leakage currents.
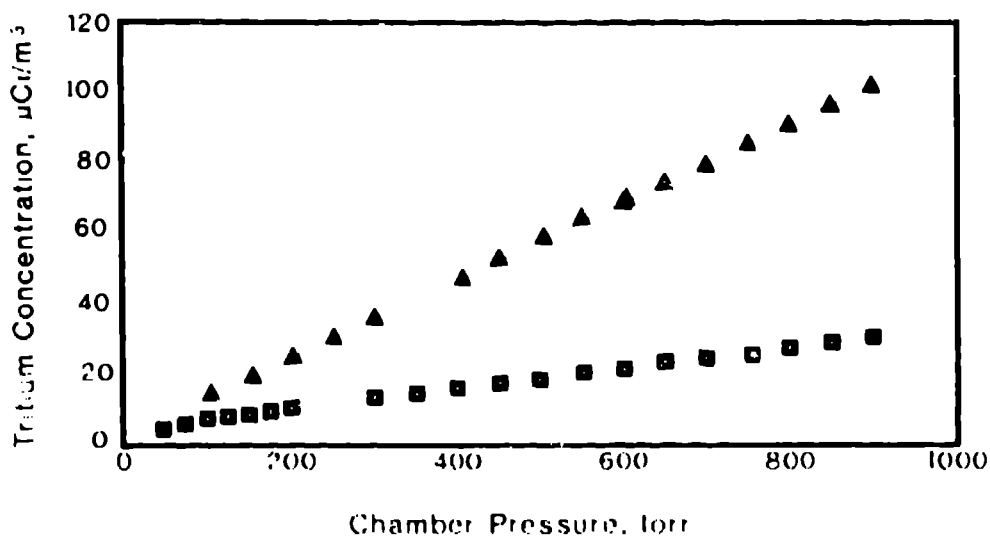

Figure 3. Response of the Overhoff tritium meter showing good linearity over a wide pressure range.

Lab benches were swiped where the cells were assembled and no tritium was found over background. However, the disassembly area was also swiped and

found to be about a factor of 2 over background indicating perhaps that some of the early cells (1-10) were not completely dehydrided prior to disassembly. The benches in the disassembly area were newly purchased prior to the beginning of this work and the area was not ever used for any other radioactive work. All materials handling and assembly work was done with latex gloves and disposable paper bench liners. A Packard model 1600 CA scintillation counter was used to analyze for tritium on the swipes and in the binder.

Early neutron detection work was performed at LANCSE (Los Alamos Neutron Scattering Center) using a bank of 15 ^{3}He detection tubes and electronics that were part of the Low Q diffratometer[6]. Since that time, it is thought that the neutron emission is not tightly correlated to the electrical pulsing and may occur randomly and in bursts after some incubation period. To enhance the detection of the bursts and to reduce the cosmic ray spallation background a 21% efficient ^{3}He channel counter containing 18 detection tubes imbedded in polyethylene was installed in two areas of an underground tunnel. The first counting room which was under 9 m of overburden ($\approx$ 1.9 g/cm^3) and later to another room under 75 m of the same tuff. The temperature in the tunnels was normally controlled to within ± 2 °C except for a month long period when the cooling system failed and the temperature rose from 21 °C to 27 °C. No abnormal activity was notice on the counters during this period. The humidity in the tunnel was found to increase to nearly 100% during the rainy season (July Aug.) however, an attempt to correlate the humidity in the tunnel with excess totals or correlated counts was not successful.

The detection equipment is identical to that used by Menlove and has been thoroughly described elsewhere[10],[11]. The main feature of the counter and electronics are that they provide data on neutron totals (total counts accumulated in a specified time) and a number designated as reals which are correlated neutron counts in a 128 μs gate. The reals counts are indicative of a neutron burst. The counters and tube enclosures are environmentally hardened and have shown excellent totals stability in the underground environment in over a year of operation. However, the detector system was found to be sensitive to spurious line transients after the counter was first installed in the tunnel. Thereafter, the counter was connected to a line isolating uninterrupable power source which eliminated the effect of power line noise.

In Menlove's laboratory[11], the average background totals rate and correlated count rate was 870 ± 6 c/h (24 hr) and 3.6 c/h while at the underground laboratory, we found 701 ± 6 c/h and 0.5 c/h while for the still deeper tunnel the rates were 637 ± 6 c/h and 0.15 c/h. For the first two cases above, the results were average totals rates taken over a several month period with a miniumum of 500 hours of data. In the case of the deep tunnel, only 72 hours of background have been accumulated at this time. All of the data were obtained using a counter with aluminum tubes which have a high alpha background. If low background stainless steel tubes were used in the deep tunnel, one could expect the background totals rate to drop to less than 100 c/h. The effect of the low background tubes on the correlated background would be insignificant since

cosmic ray spallation events are the primary cause of the reals background.

Three types of cells have been made, those with palladium powder and silicon powder, those with palladium foil and silicon powder and those with palladium foil and silicon wafers. The physical construction of the cells is shown schematically in Figure 4. Layers of alternating palladium and silicon powder were pressed into a ceramic form at a pressure of 11.2 MPa resulting a density of 26% and 68% of theoretical density for the palladium and silicon respectively. The belleville washers at each end maintain a constant pressure of 3.3 MPa as the palladium swells during deuteriding. The free gas volume of the cell is 69.1 cc excluding the volume of the palladium and silicon.
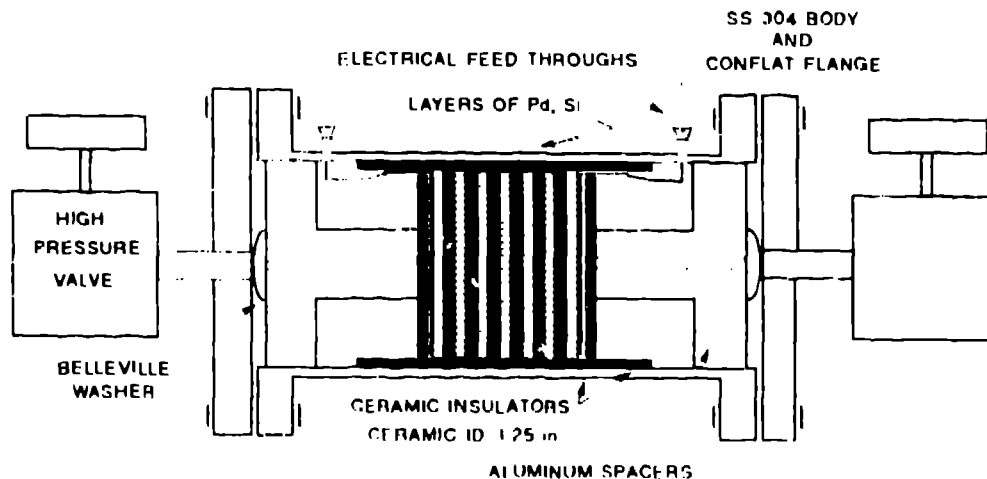

Figure 4. The experimental cell showing the layered structure and electrical connections

A high voltage of 200 to 2500 V generated by a Velonex model 360 pulse generator was used to pulse the cells. In typical operation, a unipolar, square pulse with a width of at least 150 μs at a repetition rate of 100 pulses per second was used at voltages as high as possible before breakdown occurred, typically 1200 to 2500 V.

Voltage current plots indicate that the resistance of the cell is primarily controlled by the silicon oxide layer. At low voltages (400 V) the resistance of a cell might be 100 k Ohms while at 2000 V the resistance would drop to less than 10k Ohms. The amount of pressure applied to the compact by the belleville springs primarily determines the initial resistivity. At high voltages, the assemblies are susceptible to sudden electrical breakdown which often limits the life of the cell.

RESULTS

Tritium Measurements

Table II lists all the cells made to date and summarizes the tritium

measurements. As shown in Table II, two hydrogen control cells have been made with no evidence for neutron or tritium production. In addition to the hydrogen control cells listed in Table II three other hydrogen control cells were made by simply pressing 12 to 30 g of virgin palladium powder into a form and then hydriding with hydrogen and subsequently dehydriding to test for contamination intrinsic to the palladium. All of these tests give a positive excess tritium result from 6 to 12 nCi total. This can be attributed to the effect of the water and

Sample No.	Tritium Total in curies	Hours Run	Cell Type	Fill Gas and Pressure	Notes on Cell
1	Not Analyzed	3	4 Pd layers 12 g	D1. 7.5 atm	Sample shorted out
2	170 nCi	96	4 Pd layers 12 g	D1. 7.5 atm	5% Hydrogen added to cell on 4-26
3	Not Analyzed	20	4 Pd layers 12 g	H1. 7.5 atm	Hydrogen control
4	Not Analyzed	< 1	4 Pd layers 12 g	D1. 7.5 atm	Leaky seal
5	Not Analyzed	< 1	4 Pd layers 12 g	D1. 7.3 atm	Leaky seal
6	No	17	4 Pd layers 12 g	D2. 7.4 atm	Sample shorted out
7	68 nCi	95	4 Pd layers 12 g	D1. 7.2 atm	Repeat of sample 2
8	less than 2 nCi	142	Pd mixed layers 11.6	D2. 13.6 atm	Palladium and silicon powders mixed together
9	less than 2 nCi	63	3 Pd mixed layers 11 g	D1. 8.8 atm	Same as 8, but outgassed at 110 C
10	320 nCi	77	4 Pd layers 12 g	D2. 17 atm	Repeat of sample 2
11	15 nCi	162	4 Pd layers 12 g	D3. 17 atm	Leaky seal
12	44 ± 2 nCi	62	6 Pd layers 18 g	D3. 17 atm	Cell shorted after 62 hours
13	less than 2 nCi	169	6 Pd layers 18 g	D3. 20.4 atm	Repeat of 12 but no oxide on palladium
14	10 nCi	110	6 Pd layers 18 g	D3. 20.4 atm	Repeat of 12
15	6 nCi	306	6 Pd layers 18 g	D3. 40.8 atm	Repeat of 12
16	less than 1 nCi	22.2	7 Pd layers 12 g	D3. 34 atm	Sample shorted out
17	12 ± 3 nCi	112	6 Pd layers 18 g	D3. 40.8 atm	Repeat of 12 but higher fill pressure
18	less than 1 nCi	86	6 Pd layers 18 g	D3. 40.8 atm	Repeat of 12 but with sulfur surface treatment
19	56 nCi	124	8 Pd layers 15 g	D3. 54.4 atm	Si with dow binder and Pd heated to 350 C
20	214 nCi	156	11 Pd layers 19.5 g	D4. 54.4 atm	Si with dow binder and Pd heated to 350 C
21	75 nCi	85	8 Pd layers 14 g	D3. 54.4 atm	Si with dow binder and Pd heated to 350 C
22	11 nCi	49	8 Pd layers 14 g	H2. 54.4 atm	Si with dow binder and Pd heated to 350 C
23	< 1	13	13 Pd layers 18 g	D4. 54.4 atm	Sample shorted out. Material treatment same as 19
24	13 nCi	522	7 Pd foils, Si wafers	D4. 54.4 atm	7-10 mil foils plus two powder end caps. 21.6
25	6 nCi	732	8 Pd foils, Si wafers	D4. 54.4 atm	8-10 mil foils plus two powder end caps. 24.2 g. tot
26	10 nCi	190	10 Pd layers 14.4 g	D4. 54.4 atm	Same as 19 but native oxide and PV binder
27	15 nCi	65	10 Pd foils, Si wafers	D4. 54.4 atm	10-10 mil foils plus two powder end caps. 29 g. PV
28	11 nCi	182	11 Pd layers 15.6 g	D4. 54.4 atm	Same as 20 but native oxide
29	4 nCi	66	10 Pd foils, Si powder	D4. 54.4 atm	26.4 g total Pd. Si powder with dow
30	20 nCi	61	7 Pd layers 13.4 g	D5. 9.5 atm	Repeat of 19 but shorted after 61 hours
31	4 nCi	15	10 Pd foils, Si powder	D5. 43.8 atm	29.6 g total Pd, shorted after 15 hours
32	6 nCi	198	7 Pd foils, Si wafers	D5. 13.6 atm	16.9 g total Pd.

Tritium in Deuterium bottles (in micro curies per meter cubed)

D1	110
D2	58
D3	20
D4	68
D5	17

Table II. Summary of the tritium results described in this paper.

hydrogen gas in displacing small amounts of tritium from the ionization chamber and system walls. In addition, when the same palladium was checked for tritium by the scintillation methods no trace tritium could be found. When palladium is

deuterided and then dehydrided in after a short period of time as seen for those cells that have shorted out (16, 23, 31), one finds either a very small excess tritium or none at all.

The procedure for filling the cell was to evacuate the cell and then fill the tritium analysis loop with deuterium gas at a known pressure less than 1000 torr. The tritium level could therefore be measured in this gas. The cell was then opened and allowed to absorb the gas. Since the pressure in the analysis system and the volume of the system is known, an accurate measure of the gas absorbed by the cell could be made. Subsequent filling with higher pressures allow a determination of the amount of deuterium gas contained in the cell. A comparison of the predicted amount of gas that should be contained in the cell based on the free volume and the amount of palladium and pct curves agrees within 10 percent. The error is thought to be due to the imprecision of the pct curves at pressures greater than 7×10^5 Pa (100 psi)[12]. An additional advantage of this filling technique is that after the gas had been let into the cell the remaining gas could be checked for tritium enrichment or deficit. In all cases that were checked, it was found that the deuterium remaining in the analysis system had the same tritium concentration the original fill gas in the analysis bottle to within experimental error ± 3%. Therefore, when the cell was dehydrided, the same amount of tritium should be released. Indeed this has been the case, in most cells (as in Table II) one finds a equality (within error) between the input tritium and total tritium recovered. However, in several cells (2,7,10,12,19,20,21,30), a significant amount of excess tritium was found.

An energy spectrum has not yet been performed on the gas that causes excess ionization in the system, raising the question of whether or not the gas is actually tritium or not. While the energy spectrum measurement may be definitive, other ways exist to determine that the gas is a radioactive isotope of hydrogen. For instance, the gas from cell 20 was let into the analysis loop, the tritium measured, and then the gas was allowed to readsorb onto the palladium bed. The pressure measured in the loop before and after differed by only 6 torr which was expected since the palladium heated up slightly as the deuterium was readsorbed. The tritium level in the gas left in the analysis loop decreased from 63 μCi/m^3 (at 600 torr) back to 22 μCi/m^3 (at 178 torr) indicating that the radioactivity was following the gas and that the gas could be adsorbed by the palladium. Similarly, gas obtained from cell 21 was cold trapped at 77K as it came from the cell with no increase in excess tritium observed after the cold trap had been warmed up and fresh deuterium passed through. If the tritium was bound as TDO, one would expect the trap to remove the excess tritium.

On occasion, a sample of gas was taken off the cell during operation to determine if excess tritium beyond what was expected from enrichment was present. In most cases this yielded data consistent with enrichment[11]. It has been found, from cell 12 on, that if excess tritium is present it is evolved from the cell when the palladium is dehydrided. This is shown in Figure 5a for cell 20 which is typical of all cells (after 12) that showed significant excess tritium results. The cell contains deuterium gas at 5.51×10^6 Pa. There is (in the case of cell 20) 64.9

cc of free volume resulting in the equivalent of 4450 cc at 600 torr (the analysis pressure). Contained in the palladium would be 1910 cc of deuterium at 600 torr. Thus the total amount of gas contained in the cell is 6360 cc at 600 torr. When an analysis is done, the first gas will be primarily from the free volume as the H/M is very slightly changed when the pressure is reduced from 800 to 500 psia resulting in only 25 cc (of the 2085 cc) coming from the partial dehydriding of the palladium. Most of the excess tritium is either found as the gas is removed from the palladium or the palladium is heated to remove the last gas a.. in Figure 5a.
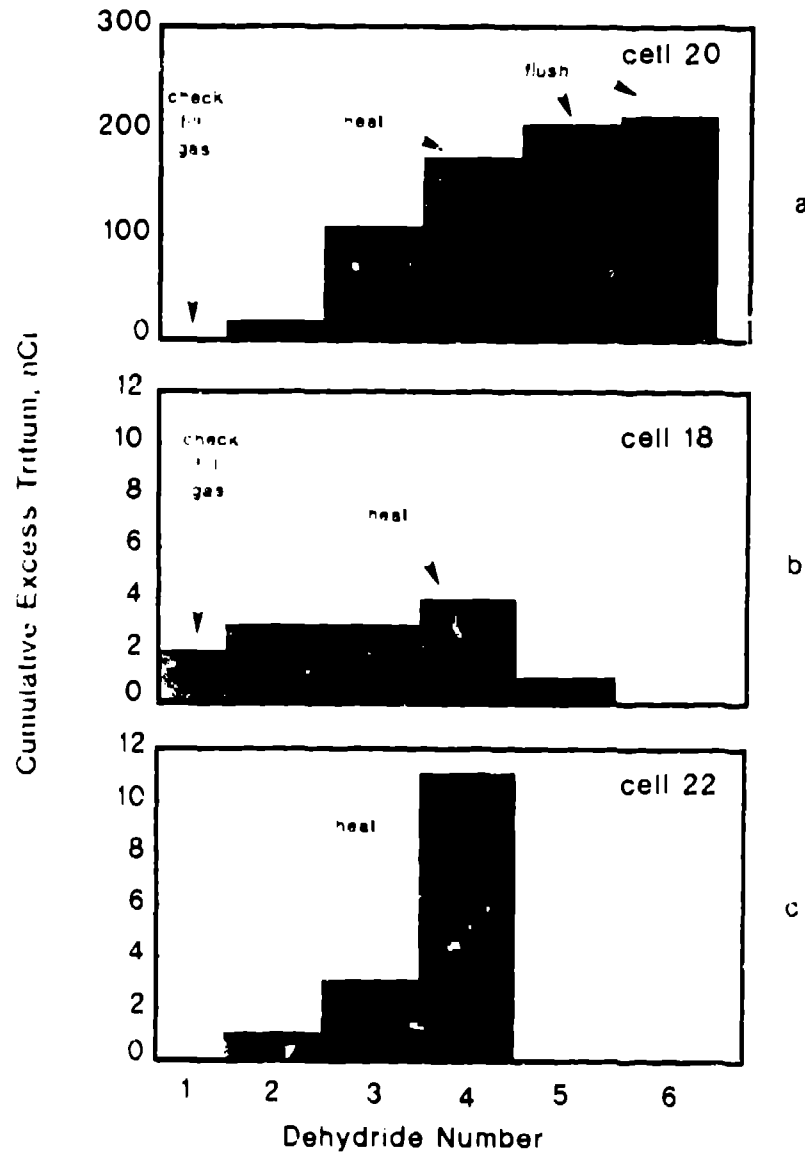

Figure 5. Cumulative excess tritium results for three types of cells during dehydryding.

Figure 5b illustrates the dehydriding behavior for a cell that contains no excess tritium where the data were consistent with enrichment. These results imply that

the tritium is not a contaminant in the original gas or cell walls but must have either been made in the palladium or have been a contaminant in the palladium. The fact that the tritium comes off first during initial dehydriding and not so much after heating implies that the tritium is not strongly bound to the palladium and would have (if it were an impurity) been (mostly) lost from the palladium at room temperature and pressure by oxidation.

The amount of tritium in cell 20 is 540 times the maximum amount found by the dissolution checks. Also, the total amount of excess tritium in cell 20 is 2.2 times the amount contained in all of the deuterium gas used to hydride this cell.

All of the cells that have shown excess tritium have shown the same type of dehydriding behavior in that the excess could be ascribed to evolution from the palladium. Another feature to note is that most of the excess tritium came off before the cell was heated. If water was the cause of this anomalous tritium, one would expect that the major effect would come when the cell was heated which then reduces the oxide layer. The hydrogen control cell (22) showed exactly this behavior when dehydrided (Figure 5c). The first two samples of gas were taken without heating the cell and showed only small increases over background, however the last sample, which had to be heated to remove the hydrogen gave an easily measurable excess tritium reading. This was thought to be due to the effect of H_2O displacing DTO on the walls of the recirculation system. This effect is less pronounced with deuterium control samples since the small amount of tritium in the deuterium is always in deficit when the palladium is heated to dehydride the sample.

Cells 12, 14, 15, 17, 19, 20, 21, 30 were all made similarly except that cells made after cell 19 had a binder in the silicon and the palladium had been oxidized. One finds that the output per hour of cells 12-17 is at most 0.7 nCi/h and averages 0.15 nCi/h. After changes to the cell, to improve the reproducibility, the output has been (19-30) a maximum of 0.88 nCi/h with an average of 0.6 nCi/h. Since, as Table II indicates, there is a wide disparity in tritium production rates one might anticipate future improvements in the rate. The present rate, achieved reproducibly, equates to a generation of 3.4×10^6 tritium atoms per second. Obviously if neutrons were generated at parity with tritium one would expect to easily detect neutrons.

Neutron Detection

In two experiments neutron signals greater than 3 sigma have been clearly detected. In sample 2 which produced a large amount of tritium per hour (1970 nCi/h) neutrons were detected at levels of 8 sigma. Because the detector was a low efficiency (1.3%) detector located above ground, the results were less than satisfactory. Since the high efficiency channel detector has been used, neutron signals for the powder systems have been at background (within 3 sigma) for the totals and slightly above background by up to 3 sigma for the correlated counts. These results would call into question the neutron data from sample 2 except that

the tritium detected in subsequent experiments has also been at a very low level. Since the ratio of neutrons detected to tritium produced in Cell 2 was 3×10^{-9} (an anomalously low value) one can say that the undetectiblity of neutron emission from cells 19-21 is consistent with the results from cell 2. In order to see a 5 sigma effect (in 12 hours in the deep tunnel) in the totals one must have a rate of about 5 nCi/h based on the effect seen with cell 2. Low background stainless steel counter tubes would allow totals at the 5 sigma level to be detected with no further improvements in tritium yield assuming the tritium is correlated with the neutron output.

A clear neutron signal of greater than 5 sigma was seen on sample 25 (a foil sample). In addition to high totals, the reals rate was elevated and a large reals (33) event (for the tunnel) was recorded. After the 5 sigma event was recorded the system was switched off for a day during which time the counter returned to the normal background rate. The cell was turned on again and showed increased activity but only at the 2-3 sigma level. This episode is shown in Figure 6. If counter misbehavior is to be suspected then one would not expect it to be correlated with the pulsing of the cell because Cell 25 was pulsed with air in the cell for about a week before the deuterium was added with no apparent effect on the counter. Also, it is unusual for the reals signal to increase especially if the counter is misbehaving in a slight way. The background rate indicated in Figure 6 is the average of 986 hours of counting before and after cell 25 was run.
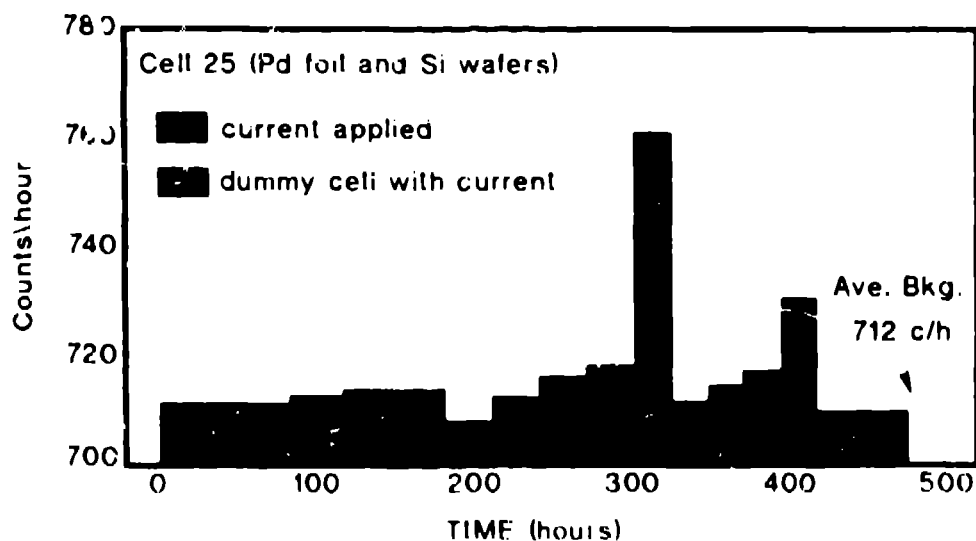

Figure 6. Neutron output from Cell 25 as a function of time. While not apparent because of the time scale, the cell output decayed as a function of time during operation.

Very little excess tritium has been found in the foil samples, especially cell 25 where, since the neutron output was observed, it was expected. This could be construed as evidence that the counter was misbehaving or that the ratio of neutrons to tritium in foil samples is different than in powder samples or is not

casual at all. However, foil samples are being actively investigated in the expectation that the neutron output, as seen from 25, may be reproduced.

CONCLUSIONS

Tritium has been detected at low levels after many hours of cell operation. The cells have been modified to the point where a low level of tritium production can be reproducibly achieved. Checks for tritium contamination three different ways confirm that there is no initial tritium contamination in the powder, foil or other materials used in this study. The tritium and neutron results are self consistent, and consistent with other reports, since only in those powder cells that have produced large amounts of tritium have neutrons been detected. In addition, all the cells that have been dehydrided thoroughly, cell 12 on, have shown that the tritium has been evolved from the palladium and was not a result of gas or cell contamination. While the energy spectrum of the tritium has not yet been measured, chemical methods have indicated that the ionizing gas is tritium.

No evidence for neutron counter misbehavior has been found, although it is highly preferable to operate the neutron counter underground where cosmic ray fluctuations are have been shown to be reduced by a significant factor. Also the correlated counts underground are reduced by a large degree and multiplicities of greater than one occur infrequently revealing more about the nature of the statistics of the bursts.

A reproducible system in which various parameters may be varied will prove invaluable for the further investigation of this phenomena.

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