A STUDY OF ELECTROLYTIC TRITIUM PRODUCTION

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ABSTRACT

Tritium production is being investigated using cathodes made from palladium and its alloys (with Li, C, S, B, and Be) to which are applied various surface treatments. Three anode materials (Pt, Ni and stainless steel), and various impurities in the electrolyte have also been used. Tritium has been produced in about 10% of the cells studied, but there is, as yet, no pattern of behavior that would make the effect predictable.

INTRODUCTION

Since the first studies by Pons and Fleischmann[1], evidence for excess tritium production in electrolytic cells continues to be reported [2; 3; 4; 5; 6; 7; 8; 9]. In a few cases, relatively large increases in tritium content ($\times 10^{4-5}$)[2; 4] have been observed in electrolytic cells while smaller but significant increases ($\times 1.5$-80)[3;5;6;7;8] have been seen by many others. Electrode materials from many sources have been used as well as cells of various designs including sealed cells with gas recombiners and a high pressure cell[6]. Tritium also has been made ($\times 1300$) using D_2 gas in a solid electrolytic cell[10]. Although we can now reasonably assume that the effect is real, no patterns have emerged from these efforts to suggest ways to make the phenomenon reproducible. Apparently, a special environment must first be created on or near the electrode surface before the rather novel nuclear reactions can start. Because this special environment is, as yet, seldom achieved, an exhaustive study of the nuclear products using the necessarily expensive equipment is not cost effective and often frustrating. This study was undertaken to learn how the initiating conditions could be produced in order to improve the reproducibility of the nuclear reactions before attempts are made to study them in detail. Tritium production was used as an indication that a nuclear reaction had occurred.

Over 150 cells were examined involving over 5000 tritium measurements. Only about 10% of the cells produced tritium. Although the success rate is still rather low, a number of conditions can be ruled out as not being important.

EXPERIMENTAL

The cell design shown in Fig. 1 was used for most of this work. The cell consists of a 120 ml, wide mouth glass jar with a bakelite lid. Most cells had a paper lid liner. Gases generated by electrolysis flow through a stainless hypodermic needle sealed in the lid with epoxy and are routed to an IV drip system where the recombination catalyst is located. About 1 ml of recombined liquid collects in this region before the excess enters a plastic IV bag. A rubber stopper seals a small hole in the lid and allows the electrolyte to be sampled using a 1.00±0.01

ml disposable, automatic pipette. Thus, the cell was completely sealed except for a small time during sampling. Tritium in the cell was determined daily, and a 1 ml sample of recombinate was taken approximately every other day if the electrolysis rate permitted. These samples as well as tritium-free water were each mixed with 10 ml of Opti-Fluor[11] and counted using a Packard Tri-Carb Liquid Scintillation Spectrometer. Measured quantities of D_2O[12] and electrolyte[l3] were returned to the cell to replace that lost by electrolysis and sampling, respectively.
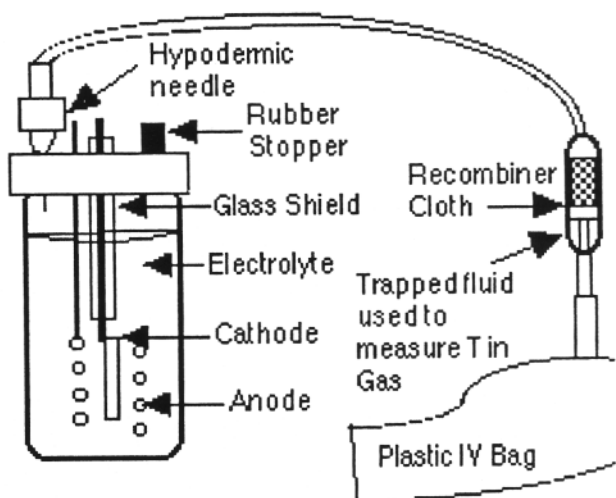

FIGURE 1. Cell with gas collection system

   Palladium alloys were made by arc-melting Pd powder[l4] with the other element in an Ar atmosphere to make the alloys listed in Table I. The X/Pd ratios are indicated for each alloying component. Most of these cathodes were studied using Pt gauze anodes although nickel wire was used in a few cases. The cathodes were either coin shaped, made by rolling an arc-melted button or were strips cut from a coin using a low speed diamond saw. Several of the Pd-Li alloys were swaged into 1- to 2-mm-diameter rods. In most cases, the surface was cleaned with concentrated HNO_3, water, and acetone.

Table I. Pd Alloys Studied as Cathodes

Pd-Li	(0.011, 0.012, 0.013,0.023, 0.051,0.22)
Pd-C	(0.005, 0.012, 0.026, 0.034)
Pd-Li-C	(Li 0.038, C 0.019),
	(Li 0.039, C 0.040)
	(Li 0.012, C 0.012)
Pd-Be	(0.01, 0.04)
Pd-S	(0.0043)
Pd-B	(0.028)

   Unalloyed Pd was studied sometimes as strips cut from a 0.050" thick sheet, coins, and as wires. These cathodes were given various surface treatments as summarized in Table II. Only one treatment was applied to each strip. The paraffin + HS treatment was applied by heating the strip in vapor produced by heating paraffin and sulfur together.

TABLE II. Surface Treatment

Washed with	Heated in
HNO_3,	Parafin + H_2S
HCl,	Parafin followed by H_2S, or
H_2O_2, or	H_2S alone
Acetone	

Various materials were added to the electrolyte in order to plate a component onto the cathode surface. Some were present as unintentional impurities. The known impurities are listed in Table III. Not listed are the impurities in the D_2O and the components electrolyzed from the anode, both of which are largely unknown at the present time. The neutrons were obtained from a [252]Cf source ($\approx$7200n/min) located near the cells.

TABLE III. Materials Present in Electrolyte

Added	Unintentional
Thiourea	Bakelite from lid
Wheat flour	Stainless from hypodermic needle
Fe metal	Glass components
Ag metal	
Hg metal	
Li_2S	
As_2O_3	
Ag_2S	
[238]U metal	
Neutrons	

Several of the electrodes were examined using an electron microscope and an ion probe to determine the surface composition.

RESULTS

The results from the various surface treatments and alloys will be discussed individually starting with the Pd-Li alloys.

Alloys

Pd-Li: The repeated ability of R. Huggins at Stanford University[15] to produce heat using impure Pd containing at least lithium and the observed migration of Li from the electrolyte into the cathode suggests that the presence of Li might be a precondition to a nuclear reaction. Cells were studied using the conditions listed in Table VII located at the end of the paper. No excess tritium was detected in any of the cells. All of the alloys except 22 % Li were ductile and easily rolled into a coin shape and swaged into rods.

Two alloys, Li/Pd = 0.011 (3mm Ø) and Li/Pd=0.034 (2mm Ø) were examined as rods for heat production, by T. Guilinger (Sandia National Laboratory) and by S. Gottesfeld (Los Alamos National Laboratory), respectively. No excess heat was observed in either case.

The presence of lithium was found to increase the ease of deuterium uptake and to increase the limiting D/Pd ratio over that observed using cathodes made with ordinary grade, arc-melted

powder. However, there is no difference in properties when a comparison is made to ultra-pure, arc-melted powder. Apparently the presence of lithium offsets the effect of impurities that are present in ordinary grade palladium.

Pd-C: Carbon, when present in Pd, resides mainly in the grain boundaries as graphite because of its very low solubility. This impurity would be expected to inhibit the diffusion of D through the grain boundaries thereby reducing the uptake rate, increasing the D concentration gradient near the surface, and reducing the limiting, average composition during the early history. When carbon was arc-melted with Pd powder, the resulting coins showed these effects. None of the cells listed in Table VII produced excess tritium.

Pd-C-Li: When Li was added to an alloy containing carbon, the effects produced by carbon were reduced. Thus, the addition of Li to ordinary grade Pd is thought to improve the D uptake rate because it tends to offset the effect of the usual carbon impurity. These alloys, as listed Table VII, did not produce excess tritium.

Pd-Be: Because beryllium has a relatively weak, neutron rich nucleus, a suggestion was made by Edward Teller that its presence might augment the neutron source provided by the deuterium atom. The two listed compositions were sufficiently ductile to be rolled into a coin shape and cut into strips using a diamond saw. These were used as cathodes in cells described in Table VII. No excess tritium was detected.

Pd-S: Although tritium was produced in some cells when a sulfide treatment was applied to the cathode surface, this Pd-S alloy produced no tritium. However, the sulfur does, in both forms, accelerate recombination of the evolving D_2 with oxygen in the air causing intense self-heating.

Pd-B: Two cathodes containing boron were studied as listed in Table VII. One which was run with As_2O_3 in the electrolyte produced a small amount of tritium. The other cathode did not produce tritium.

Surface Treatment

Heated in H_2S: A number of pure palladium samples were heated in H_2S from a gas cylinder. This treatment causes an insulating sulfide layer to form on Pd. Unless the electrode is run as an anode for awhile, the Pd will not take up deuterium. Samples treated only with H_2S, without paraffin, have not produced excess tritium.

Heated in Paraffin Vapor: Several pure palladium samples, both strips as well as wire, were heated in paraffin or paraffin + H_2S from a cylinder. As can be seen in Table VII, several of these cells produced excess tritium. In this series, the electrolyte dissolved some of the bakelite lid because the lid liner had been removed. Surface examination showed the presence of large amounts of Ca which is a major component of bakelite. This Ca, as well as a lesser amount of Zn, formed a complex surface layer on the electrode. Residual paraffin was also observed as clumps. Electrolytic action was uneven where the paraffin was present.

Control Cells

The amount of tritium produced in most of the cells described in this study is so small that the possibility of contamination must be considered. This is especially important because the work is being done in an area where tritium might be present in the air. Two methods were used to determine the contamination level produced by the environment and by the cell construction

materials. In addition, a cell was contaminated on purpose so that the time behavior of such contamination could be studied.

Seven inactive cells listed in Table VII were run during this study using materials from the same lot as were used in the cells that produced tritium (active cells). In addition, many cells of conventional, potentially active design did not produce tritium even though they were being studied at the same time, using the same materials and design as the active cells.

TABLE IV. Control Cells

	Cathode	Anode	Electrolyte	# Cells
A.	Pd	Ni	H_2O	3
	Pt	Pt	D_2O, Aldrich	1
	Ni	Ni	D_2O, MSD	1
	Ti	Pt	H_2O	1
B.	5 open water jars			
C.	1 cell to which tritium water was added			

Open jars containing 60 ml of initially tritium-free water were situated in the room so that any tritium in the environment would be detected by observing an increase in their tritium content.

Finally, a little tritiated water was added to an inactive cell in order to see how this known contamination would behave compared to the time history of active cells.

The results from inactive cells are described first.

(A) Inactive Cells: Three cells (#117, #123 and #132) contained water electrolyte (0.2M LiOH), and construction materials from the lots used in several active cells (#73, #98). Tritium measurements for these cells and for tritium-free water were taken over a 35 day period (23 data points in each set). The average excess tritium in the D_2O cells is 4 count/min-ml greater than tritium-free water (instrument background). Although there may be a slight pickup of tritium from the environment, the amount is well within the uncertainty of ±5 count/min-ml (±14 d/min-ml) found in the tritium measurement for the cells containing D_2O electrolyte. None of the other control cells shows evidence for contamination beyond this level.

(B) Open Jars: One ml samples were taken from each open jar every working day. During times that a small amount of tritiated-water vapor was present in the air, the tritium content of the jar increased. When the air became tritium-free, the tritium content of the jars decreased. This reduction occurred because ≈4 ml/day evaporated in addition to the loss of 1 ml owing to sampling. This loss was replaced by tritium-free water. The rate of change, corrected for dilution, gives a measure of the tritium content of the air. The quantities present in the room are too small to have biological importance and are too small to affect sealed cells as used in this study. This conclusion will be seen more clearly in the following discussion about individual cells.

(C) Addition of tritiated water: A small amount of tritium water was added to an inactive cell (#82) so that the count rate was increased to a value similar to that found in some active cells. Corrections were made for enrichment during electrolysis and dilution by replacement fluid. The result is shown in Fig. 2 as the fraction excess tritium over that at the beginning of electrolysis. No tritium was produced during the 123 days preceding the addition of tritium water. After the

addition, the quantity remained constant, consistent with normal enrichment and dilution. This steady value is in sharp contrast to the behavior of tritium loss from active cells as seen below. After this cell was terminated, the electrolyte was replaced by tritium-free water containing 0.2N LiOH and the current was reversed so that the cathode would lose any absorbed tritium to the water. After electrolyzing in this mode for 24 hrs, no increase in tritium content was seen. Therefore, the tritium which had been added to the electrolyte had not absorbed into the Pd cathode to any significant extent.

Statistical Analysis of Data

A series of closed (recombinate run back into the cell) and, unfortunately, inactive cells allowed the total random error in the tritium measurements to be determined. These cells were unsealed such that there was a small hole to allow excess gas to escape after passing by the recombinate cloth. No enrichment or pickup of tritium was observed. Thus, the recombiner worked properly and no tritium entered from the environment in spite of the hole. The standard deviation based on 446 measurements using 13 cells over a 40 day period is listed in Table V as (A).

TABLE V. Summary of Count Rate and Standard Deviation for Various Inactive Cells

	Data Set	Average d/min-ml	SD	# Values
A	Closed Cells	138	±14	446
B	Open Cell, corrected	120	±14	80
C	Open Cell, uncorrected	140	±31	96

A similar analysis of a sealed cell was made, but where the recombinate was collected separately. The value is based on 80 data points, after correction for enrichment, and is listed as (B). When an enrichment correction was not made, the standard deviation, designated (C) was larger because of the slow increase in count rate over the life of the cell. This study shows that the total random error in the tritium measurement is ±14 decomposition /min-ml (±5.3 count/min-ml) with an occasional outlier near ±25 d/min-ml. Because of such outliers, unusually high or low values are ignored if they are isolated from the trend of the data. In other words, only patterns supported by many points are used as evidence for tritium production. In addition, data are considered significant only if greater than 5 sigma from the behavior produced by enrichment and dilution.
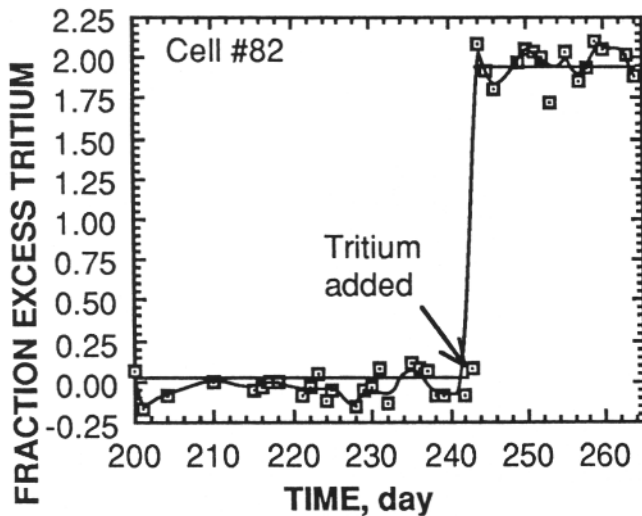
FIGURE 2. Effect of adding tritium water

Distribution Ratio

   During electrolysis, the tritium concentration in the gas is generally less than that in the liquid. This difference causes enrichment of tritium in the liquid. In order to calculate the amount of tritium that is lost to the gas phase, thereby allowing tritium enrichment to be calculated, the distribution ratio (tritium in the gas/tritium in the liquid) must be determined. This quantity was measured directly for a number of active and inactive cells as listed in Table VI. In addition, this quantity was calculated by an indirect approach using the rate at which tritium was enriched in an inactive cell #70 (slope in Table VI). Values as high as 1.1 have been observed in the past (averaged over the life of the cell) although recent cells (>#91) have a mean of 0.55±0.02 based on 13 cells.

Behavior of Active Cells

   A total of 13 cells have shown some evidence for tritium production as of 3/8/90, as listed in Table VII. However, only the more recent observations have been made with sufficient detail to eliminate any doubt about the source of tritium. The time history of various cells is summarized in Fig. 3. The square points indicate the time at which the cell was started, and the corresponding + indicates when the cell was stopped. A few of these point-pairs are connected in the figure to show how the connection should be made. Tritium was produced in some cells during the interval between each set of vertical lines. A box is drawn around the cells that were active during this time. The number of active cells and the total running at the time are also indicated. For example, between 100 and 125 days after 6/1/89, seven cells out of a total of 34 produced tritium. These active cells continued to run after day 125 without producing tritium until they were turned off on day 180. Eleven cells were started during the time when tritium was being produced in these seven cells but none became active. Presence of low level tritium in the room air, as detected by the open water jars, is shown by the boxes at the bottom of the figure. Some tritium could have been in the air before day 125 but no open jars were present to detect its presence. Although some tritium was present in the air when excess tritium appeared in some cells, other cells showed no tritium production. In addition, all of the active cells eventually stopped production even though a small amount of tritium continued to be present in the air.

TABLE VI. Measured Distribution Ratio and Standard Deviation

Cell #	Gas/Liquid	SD	# Values
70 (ratio)	0.91	0.16	15
70 (slope)	0.84		
74	1.12	0.17	13
73*	0.83	0.12	9
82 (before)	0.69	0.09	4
82 (after)	0.44	0.06	3
85	0.53	0.05	10
86	0.52	0.08	8
87	0.76	0.15	21
91	0.53	0.04	7
98*	0.53	0.04	8
110	0.58	0.02	5
118	0.58	0.05	10
119	0.59	0.09	10
120	0.54	0.06	11
121	0.55	0.04	13
124	0.57	0.07	11
127	0.53	0.08	11
128	0.57	0.05	9
129	0.55	0.08	9
130	0.53	0.06	9
131	0.52	0.06	10

Values before and after addition of tritium to cell #82 are shown,
* Active cells

Two active (#73 and #98) and two inactive (#70 and #99) cells are compared in Fig. 4 where the fraction excess is plotted as a function of time from the start of electrolysis. The fraction excess is calculated as described previously[9]. In brief, the amount of tritium lost to the gas phase is calculated from the electrolysis rate and the measured distribution ratio. The small amount of tritium in D_2O that is added to the cell each day to replace fluid loss is also taken into account. The expected inventory of tritium in the cell is calculated and compared to the actual amount. The fraction excess is the difference between these values divided by the initial amount. For example, a calculated excess fraction of 1.0 means that the tritium content has doubled over that in the cell when electrolysis started, taking into account tritium from all known sources. Because the calculated excess does not take into account the small amount of tritium that is in the electrode, the plotted values are lower limits.

The amount of fluid added to the cells on a daily basis was recorded and compared to the amount lost, calculated using the cell current. Except when a cell occasionally started to leak fluid, the amount added agreed with the amount calculated. Of the two active cells compared in Fig. 4, only cell #98 started to leak after 40 hr. Because this leak occurred after tritium production had stopped, it could not have been the cause of excess tritium.

Cells #70 and #73 were electrolyzed at the same time and at the same current. The cathode in cell #73 had been heated in paraffin vapor before being electrolyzed. Other factors in the cell

design are listed in Table VII. This cell produced tritium over a 25 day period with evidence for two bursts. After the last burst, the tritium content decreased to a constant value over a 10 day period. This decrease is thought to be caused by the removal of tritium that is present as DT gas dissolved in the liquid. The tritium that remained was present as DTO. This initial decrease is in contrast to the stable tritium content that results when tritiated water is placed in a cell as seen in Fig. 2.
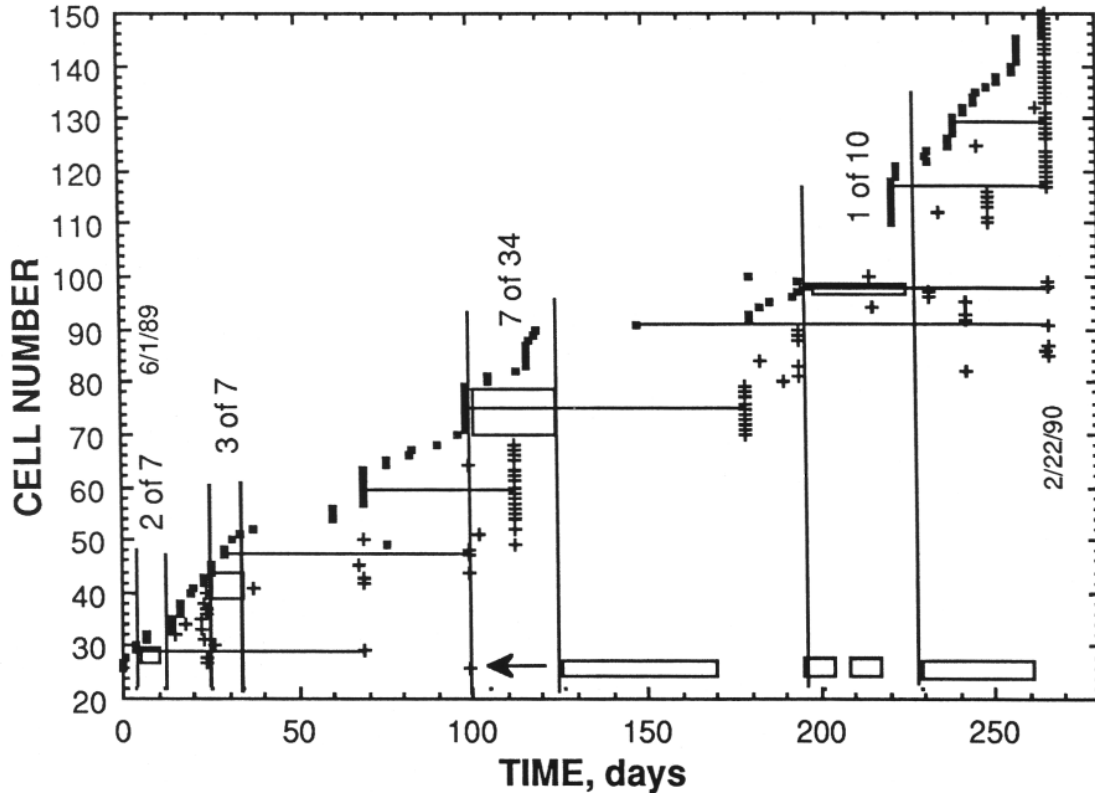
FIGURE 3. Time history of cells
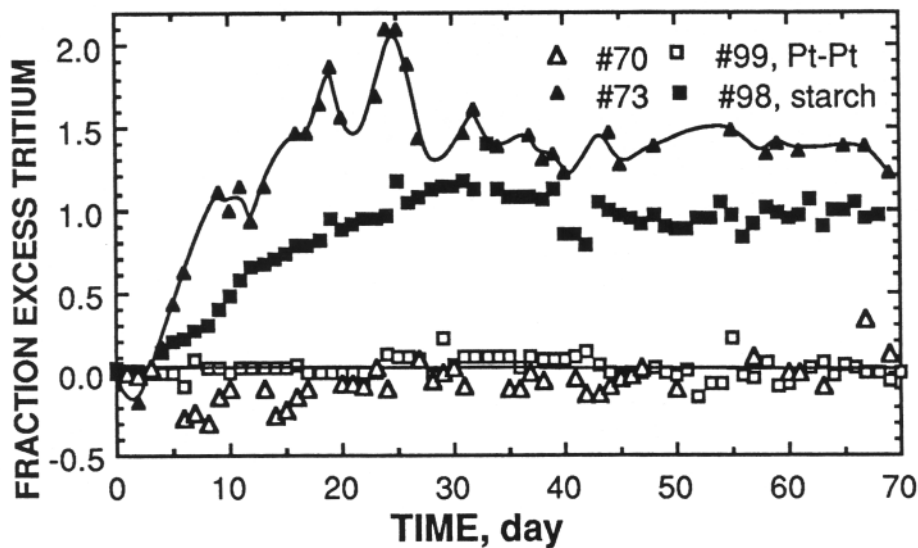
FIGURE 4. Fraction excess tritium calculated for cells #70, #73 #98 and #99.

Cells #98 and #99 were electrolyzed at the same time but not at the same current. The cathode in cell #98 was not pre-treated although 3.4 mg of wheat flour was placed in the electrolyte. Such large molecules are thought to inhibit the growth of surface dendrites. Indeed, the tritium production rate was decreased, production lasted for a longer time, and there were no bursts. However, attempts to duplicate this effect using other cells containing various amounts of wheat flour have failed.

CONCLUSION

We have produced tritium in electrolytic cells containing a Pd cathode, a Ni or Pt anode, and an electrolyte containing 0.1M or 0.2M LiOD. However, we can not yet suggest a design that will have a high probability of success.

A high, bulk deuterium content in Pd does not appear to be essential because tritium has been produced by electrodes having an average D/Pd ratio between 0.55 and 0.80. However, local concentrations might be higher.

Most active cells, but not all, have produced tritium using MSD[12] heavy water in contrast to that obtained from Aldrich[16]. In addition, the Aldrich water has a larger variation of tritium content between bottles than does the MSD. This suggests that other, important impurities might have a similar variation.

The presence of Li, Be, C, or S alloyed with Pd does not increase the probability of tritium production at the concentrations studied in this work. A surface layer of only palladium sulfide does not help either. A small concentration of ^{238}U, Hg, Fe, thiourea, Li_2S, or Ag_2S in the electrolyte does not increase the rate of success. However, sulfur in the lattice or on the surface increases the rate at which evolved D_2 reacts with O_2 in the air.

The effect of (1) paraffin alone or paraffin plus sulfide on the surface, (2) wheat flour or As_2O_3 in the electrolyte, or (3) a Pd-B alloy is uncertain, although the chance of tritium production seems to be improved by these conditions.

Distribution ratios for tritium (gas /liquid) have been measured with values between 1.1 and 0.52, the most probable value being 0.55.

ACKNOWLEDGEMENTS

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11. Opti-Fluor was obtained from Packard Instrument Company, 2200 Warrenville Rd, Downers Grove, IL 60515. This fluid shows no chemiluminescence after 20 min when mixed 10 to 1 with 0.2 N LiOD.
12. Obtained from MSD Isotopes, Montreal, Canada (>99.9 % D).
13. Made by mixing Li metal with the D_2O to give a 0.1M or 0.2 M solution.
14. Obtained from Johnson-Matthey Company as 100 % Pd, batch #V7114307 and #V8368501.
15. A. Belzer, U. Bischler, S. Crouch-Baker, T. M. Gur, M. Schrieber, and R. Huggins "Two Fast Mixed Conductor Systems-Deuterium and Hydrogen in Palladium", Cold Fusion Workshop, Santa Fe, NM May 22-25 (1989).
16. Obtained from Aldrich Chemical Company, (99.9 % D).

TABLE VII Description of Cell Treatment

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
	A			B	C	D	E	F	G	H	I	J	K	L
Pd + Li Alloys														
33	Coin	4.87	4.7	Li/Pd(1) = 0.051	std	none	0.5N, thio	MSD	Pt	plastic(2)	none	0.77	6/15	n
34	Coin	4.96	4.2	Li/Pd(1) = 0.023	std	none	0.5N, thio	MSD	Pt	plastic(2)	none	0.83	6/15	n
35	Coin	4.99	4.7	Li/Pd(1) = 0.012	std	none	0.5N, thio	MSD	Pt	plastic(2)	none	0.77	6/15	n
49	Wire	0.74	1.5	Li/Pd(2) = 0.0343	HNO3	none	0.2N	MSD	Pt, small	plastic(3)	Glass		8/15	n
55	Coin	3.67	4.2	Li/Pd(4) = 0.025	std	none	0.2N	MSD, pe	Pt	plastic(4)	Glass	0.73	7/31	n
69	Coin*	10.26	7.9	Li/Pd(1) = 0.013	std	none	0.2N	MSD	Pt	Quartz	Glass		9/5	n
88	Button	4.05	2.0	Li/Pd(1) = 0.22	none	none	0.1N	MSD, Ald.	Pt	glass(5)	Glass		9/25	n
111	Strip	1.26	2.3	Li/Pd = 0.012	HCl	pol., annealed	0.1N	Aldrich	Ni(2)	glass(5)	Glass	0.79	1/9	n
113	Strip	0.82	1.7	Li/Pd = 0.012	HCl	polished	0.1N	Aldrich	Ni(2)	glass(5)	Glass	0.76	1/9	n
Pd + C Alloy														
2	Coin	6.00		C/Pd(1) = 0.0344	std	none	0.2N	MSD	Pt	plastic(3)	torr seal		7/2	n
52	Coin	5.46	5.4	C/Pd(1) = 0.026	Na2S	oxidized	0.2N	MSD	Pt	plastic(3)	1 yes, 2 no	0.88	7/8	n
56	Coin	3.52	3.7	C/Pd(1) = 0.0191	Li2S	oxidized	0.2N	MSD	Pt	plastic(4)	glass	0.66	7/31	n
65	Coin	4.10	4.0	C/Pd = 0.005	std	none	0.2N	MSD	Pt	plastic(4)	glass	0.77	8/15	n
110	Strip	0.92	1.8	C/Pd = 0.012	HCl	polished	0.1N	Aldrich	Ni(2)	Glass(5)	glass	0.73	1/9	n
116	Strip	0.83	1.8	C/Pd = 0.012	HCl	Pol., annealed	0.1N	Aldrich	Ni(2)	Glass(5)	glass	0.75	1/9	n
Pd + Li + C Alloys														
89	Coin	4.90	3.0	Li/Pd(5) = 0.038 C/Pd = 0.019	H2O2	none	0.1N	MSD. Ald.	Pt	Glass(5)	glass	0.70	9/27	n
90	Coin	4.65	3.0	Li/Pd(5) = 0.039 C/Pd = 0.040	none	none	0.002N	MSD, Ald.	Pt	Glass(5)	glass	0.53	9/28	n
114	Strip	0.97	2.0	Li, C/Pd = 0.012	HCl	polished	0.1N	Aldrich	Ni(2)	Glass(5)	glass	0.76	1/9	n
115	Strip	0.80	1.7	C, Li/Pd = 0.012	HCl	pol., annealed	0.1N	Aldrich	Ni(2)	Glass(5)	glass	0.71	1/9	n
Pd-B Alloy														
43	Coin	4.69		Pd(1) + B	Std	none	0.2N, As2O3	MSD	Pt	plastic(3)	torr seal		6/24	y
118	Strip	1.41	2.7	B/Pd = 0.028	HCl	none	0.1N	Aldrich	Ni(3)	Glass(5)	glass		1/9	n
Pd + S Alloy														
42	Coin	4.47		S/Pd(1) = 0.0043	Std	none	0.2N	MSD, pe	Pt	plastic(3)	torr seal		6/2 4	n

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
A				B	C	D	E	F	G	H	I	J	K	L

Pd + Be Alloys

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
92	Strip	1.63	3.0	Be/Pd(2) = 0.01	none	none	0.1N	Aldrich	Ni(1)	Glass(5)	glass	0.75	1 1/28	n
93	Strip	1.66	2.9	Be/Pd(2) = 0.04	none	none	0.1N	Aldrich	Ni(1)	Glass(5)	glass	0.71	1 1/28	n

PURE Pd

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
68	Coin	2.09	2.6	ultra pure	Std	none	0.2N	MSD	Pt	plastic(4)	glass	0.79	8/30	n
32	Coin	5.10		Pd(1)	Std	none	0.2N, thio	MSD	Pt	plastic(2)	torr seal		6/8	n
137	Coin	6.21		Pd(3), W sw	sanded, HNO3	vac	0.1N	Aldrich	Pt, Ni(4)	plastic(2a)	glass		2/8	
138	Coin	7.48		Pd(3), W sw	sanded, HNO3	anneal	0.1N	Aldrich	Pt, Ni(4)	plastic(2a)	glass		2/8	
54	Coin	4.08	4.3	Pd(4)	Std	none	0.2N	MSD, pe	Pt	plastic(4)	glass	0.77	7/31	n
50	Coin	5.26		Pd(2)(am)	Std	none	0.2N	MSD	Pt	plastic(3)	torr seal		7/2	n
51	Coin	5.89	5.3	Pd(2)(am)	Std	anneal	0.2N	MSD	Pt	plastic(3)	torr seal	0.80	7/4	n
58	Sheet	1.79	2.5	Pd(2)	HNO3	none	0.2N, thio	MSD	Pt	plastic(4)	glass	0.83	8/9	n
59	Sheet	2.44	3.5	Pd(2)	HNO3	none	0.2N, thio	MSD	Ni gauze	plastic(4)	glass	0.71	8/9	n
66	Strip	1.71	1.5	Pd(2)	HNO3	none	0.2N	MSD	Ni(1)	plastic(4)	glass		8/22	n
104	Strip	0.59		Pd(2)	HNO3	anneal	0.1N	Aldrich	Pt	Glass(5a)	none		12/12	n
106	Strip	1.11		Pd(2)	HNO3	none	0.1N	Aldrich	Pt	Glass(5a)	none		12/12	n
107	Strip	1.16		Pd(2)	HNO3	none	0.1N	Aldrich	Pt	Glass(5a)	none		12/12	n
108	Strip	0.93		Pd(2)	HNO3	none	0.1N	Aldrich	Pt	Glass(5a)	none		12/12	n
109	Strip	0.77		Pd(2)	HNO3	none	0.1N	Aldrich	Pt	Glass(5a)	none		12/12	n
121	Strip	0.68	1.5	Pd(2), sw Ni(4)	HCl	none	0.1N	Aldrich	Ni(2)	Glass(5)	glass		1/1 0	n
127	Strip	0.82	1.6	Pd(2), sw Ni(4)	HNO3	reverse	0.1 N LiOD	Aldrich	Ni(4)	Glass(5b)	glass		1/26	n
139	Strip	1.06	2.0	Pd(2), sw Ni(4)	none	none	0.1N	Aldrich	Pt, Ni(4)	Paraffin	glass		2/12	
140	Strip	1.02	2.0	Pd(2), sw Ni(4)	HNO3	none	0.1N	Aldrich	Pt, Ni(4)	Paraffin	glass		2/1 2	
41	Wire	1.90	7.6	Pd, Marshall	HNO3	none	0.2N, thio	MSD	Pt	plastic(3)	torr seal	0.86	6/21	y
82	Wire	1.66		Pd, Marshall	Std	none	0.05N	MSD.Ald.	Ni(1)	Glass(5)	glass		9/25	n
70	Wire		2.0	Pd, Martin	HNO3	none	0.1N	MSD	Ni(1)	Glass(5)	glass		9/5	n
79	Wire	0.37	2.0	Pd, Martin	HNO3	none	0.2N, Ag	MSD	Ni(1), Ag	Glass(5)	glass	0.94	9/7	y
80	Bar	8.45	9.4	Pd investment	none	none	0.2N	MSD	Pt	Glass(5)	glass		9/14	n
81	Bar	7.44	8.4	Pd investment	none	none	0.2N	MSD	Pt	Glass(5)	glass	0.74	9/14	n

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
				B	C	D	E	F	G	H	I	J	K	L
Paraffin														
73	Strip	0.68	1.0	Pd(2)	C	paraffin	0.2N	MSD	Ni(1)	Glass(5)	glass	0.81	9/7	y
74	Strip	0.69	1.5	Pd(2)	C	paraffin	0.2N	MSD	Ni(1)+S	Glass(5)	glass	0.78	9/7	n
83	Strip			Pd(2)	HNO3	paraffin	0.2N	MSD	Ni(D	Glass(5)	glass		9/25	n
84	Strip	0.54	1.6	Pd(2)	HNO3	paraffin	0.1N	MSD	Ni(1)	Glass(5)	glass	0.76	9/25	n
85	Strip	0.64		Pd(2)	HNO3	paraffin	0.1N	MSD.Ald.	Ni(1)	g!ass(5)	glass		9/25	n
86	Strip	0.42		Pd(2)	HNO3	paraffin	0.1N	MSD.Ald.	Pt	Glass(5)	glass		9/25	n
87	Strip	0.47		Pd(2)	HNO3	paraffin	0.1N	MSD, Ald.	Pt	Glass(5)	glass		9/25	n
91	Sheet			Pd(2)	HNO3	paraffin	0.2N	MSD.Ald.	Ni(1)	Glass(5)	glass		10/27	n
100	Strip	1.13		Pd(2)	HNO3	paraffin	0.1N	Aldrich	Ni(1)	Glass(5a)	none		11/28	n
101	Strip	0.97		Pd(2)	HNO3	paraffin	0.1N	Aldrich	Ni(1)	Glass(5a)	none		11/29	n
102	Strip	0.69		Pd(2)	HNO3	paraffin	0.1N	Aldrich	Ni(1)	Glass(5a)	none		11/28	n
105	Strip	0.87		Pd(2)	HNO3	paraffin	0.1 N	Aldrich	Pt	Glass(5a)	none		12/12	n
Paraffin + H2S														
40	Coin	6.97		Pd(1)	S	H2S+C	0.2N	MSD	Pt	plastic(3)	no	0.84	6/20	n
44	Coin	4.93	4.6	Pd(1)	S	H2S+C	0.2N	MSD	Pt	plastic(3)	torr seal	0.75	6/26	n
45	Coin	5.53		Pd(2)	S	H2S+C	0.2N	MSD	Pt	plastic(3)	torr seal	0.67	6/26	n
46	Coin	5.15		Pd(1)	S	H2S+C	0.2N	MSD	Pt	plastic(3)	torr seal		6/27	n
71	Strip	0.73	2.5	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.55	9/7	y
72	Strip	0.73	2.1	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.81	9/7	y
75	Strip	0.50	1.4	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.71	9/7	y
77	Strip	0.72	1.4	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.69	9/7	y
H2S														
62	Sheet	2.61	3.5	Pd(2)	S	H2S	0.2N	MSD	Pt	plastic(4)	glass	0.56	8/9	n
63	Sheet	2.56	3.5	Pd(2)	S	H2S	0.2N	MSD.pe	Pt	plastic(4)	glass	0.67	8/9	n
64	Sheet	2.27	3.4	Pd(2)	S	H2S	0.2N	MSD	Pt	plastic(4)	glass	0.69	8/15	n
Wheat Flour														
98	Strip	0.79	1.8	Pd(2), sw stainless	HNO3	none	0.1 N, 3.5mg	Aldrich	Ni(2)	Glass(5)	glass	0.93	12/15	y
120	Strip	0.86	1.7	Pd(2), sw Ni(4)	HCl	none	0.1 N, 15mg	Aldrich	Ni(2)	Glass(5)	glass		1/10	n
122	Strip	0.68	1.6	Pd(2), sw Ni(4)	HNO3	none	0.1 N, 5.4 mg	Aldrich	Ni(D	Glass(5)	glass		1/19	n
124	Strip	0.96	1.8	Pd(2), sw Ni(4)	HNO3	none	0.1 N, 4.0 mg	Aldrich	Ni(3)	Glass(5)	glass		1/19	n

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
				B	**C**	**D**	**E**	**F**	**G**	**H**	**I**	**J**	**K**	**L**
	A													
125	Strip	0.75	1.6	Pd(2), sw Ni(4)	HNO3	reverse	0.1 N, 5.2mg	Aldrich	Ni(3)	Glass(5)	glass		1/25	n
126	Strip	0.83	1.7	Pd(2), sw Ni(4)	HNO3	reverse	0.1 N, 4.7mg	Aldrich	Ni(3)	Glass(5)	glass		1/25	n
128	Strip	0.79	1.6	Pd(2), sw Ni(4)	HNO3	reverse	0.1N, 15mg	Aldrich	Ni(4)	Glass(5b)	glass		1/26	n
129	Strip	0.61	1.4	Pd(2), sw Ni(4)	HNO3	reverse	0.1 N, 7.2mg	Aldrich	Ni(4)	Glass(5b)	glass		1/26	n
130	Strip	0.92	1.8	Pd(2), sw Ni(4)	HNO3	reverse	0.1 N, 4.2mg	Aldrich	Ni(4)	Glass(5b)	glass		1/26	n
Thiourea														
26	Coin	3.63	3.7	Li/Pd(1) = 0.047	Std	none	0.2N	MSD	Pt	plastic(2)	torr seal	0.82	6/1	n
28	Coin	4.88	3.0	Rh/Pd = 0.1	Std	none	0.2N	MSD	Pt	plastic(2)	torr seal		6/2	n
29	Coin	3.63	3.7	Pd(1)	S	H2S+C	0.2N	MSD	Pt	plastic(2)	torr seal	0.72	6/5	y
31	Coin	4.93	4.7	Pd(1) + Rh + Li	Std	none	0.2N	MSD	Pt	plastic(2)	torr seal	0.88	6/8	n
32	Coin	5.10		Pd(1)	Std	none	0.2N	MSD	Pt	plastic(2)	torr seal		6/8	n
33	Coin	4.87	4.7	Li/Pd(1) = 0.051	Std	none	0.5N	MSD	Pt	plastic(2)	no	0.77	6/15	n
34	Coin	4.96	4.2	Li/Pd(1) = 0.023	Std	none	0.5N	MSD	Pt	plastic(2)	no	0.83	6/15	n
35	Coin	4.99	4.7	Li/Pd(1) = 0.012	Std	none	0.5N	MSD	Pt	plastic(2)	no	0.77	6/15	n
36	Coin	5.86		Rh/Pd = 0.1	Std	none	0.5N	MSD	Pt	plastic(2)	no	0.86	6/17	n
37	Coin	5.35	4.8	Rh/Pd = 0.1	Std	none	0.3N	MSD	Pt	plastic(2)	no	0.90	6/17	n
38	Coin	5.13	4.7	Rh/Pd = 0.1	Std	none	0.2N	MSD	Pt	plastic(2)	no	0.85	6/17	n
41	Wire	1.90	7.6	Pd, Marshall	HNO3	none	0.2N	MSD	Pt	plastic(3)	torr seal	0.86	6/21	y
58	Sheet	1.79	2.5	Pd(2)	HNO3	none	0.2N	MSD	Pt	plastic(4)	glass	0.83	8/9	n
59	Sheet	2.44	3.5	Pd(2)	HNO3	none	0.2N	MSD	Ni gauze	plastic(4)	glass	0.71	8/9	n
Uranium														
57	Strip	0.46	1.1	Pd(2)	HNO3	none	0.2N	MSD	Pt, small	plastic(4)	glass		8/9	n
Iron														
61	Sheet	2.08	2.9	Pd(2)	HNO3	none	0.2N	MSD	Pt	plastic(4)	glass	0.80	8/9	n
Silver														
79	Wire	0.37	2.0	Pd, Martin	HNO3	none	0.2N	MSD	Ni(1), Ag	Glass(5)	glass	0.94	9/7	y
Mercury														
95	Strip	1.19	2.3	Pd(3)	HNO3	none	0.1N	Aldrich	Ni(1)	Glass(5)	glass	0.88	12/4	n
Li2S														
60	Sheet	1.98	2.8	Pd(2)	HNO3	none	Li2S+alcohol	MSD	Ni gauze	plastic(4)	glass	0.71	8/9	n
As2O3														
43	Coin	4.69		Pd(1) + B	std	none	0.2N, As2O3	MSD	Pt	plastic(3)	torr seal		6/24	y

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
	A			B	C	D	E	F	G	H	I	J	K	L
131	Strip	1.05	2.0	Pd(2), sw Ni(4)	HNO3	reverse	0.127g As2O3	Aldrich	Ni(4)	Glass(5b)	glass		1/29	n

Ag2S

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
135	Strip	0.86		Pd(2), sw Ni(4)	HNO3	none	0.1N	Aldrich	Pt, Ni(4)	Glass(5)	glass		2/2	n

CONTROL CELLS

Water electrolyte

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
117	Strip	0.98		Pd(2)	HCl	none	0.2NUOH	H2O	Ni(3)	Glass(5)	Glass		1/9	n
123	Strip	0.85	1.7	Pd(2), sw Ni(4)	HNO3	none	0.2N LiOH	H2O	Ni(1)	Glass(5)	glass		1/18	n
132	Strip	0.90	1.8	Pd(2), sw Ni(4)	HNO3	reverse	0.1 N LiOH	H2O	Ni(4)	Glass(5b)	glass		1/29	n
144	Strip	0.44	4.2	Ti(1), sw Ni(4)	HNO3	none	0.1 N LiOH	H2O	Pt, Ni(4)	Glass(5)	glass		2/14	n

Pt Cathode

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
99	Wire			Pt	HNO3	none	0.1N	Aldrich	Pt	Glass(5)	glass		12/12	n

Ni Cathode

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
27	Coin	6.97	6.4	Ni	Std	none	0.2N	MSD	Pt	plastic(2)	torr seal	0.09	6/1	n

TRITIUM PRODUCING CELLS

#	Form	wt. (g)	area (cm2)	alloy	Cleaning	Pre-Treatment	Electrolyte	Kind	Anode	Container	lead covering	Max. D/Pd	Date Started	T
29	Coin	3.63	3.7	Pd(1)	S	H2S+C	0.2N, thio	MSD	Pt	plastic(2)	torr seal	0.72	6/5	y
30	Coin	5.40		Pd(1)	S	H2S+C	0.2N	MSD	Pt	plastic(2)	torr seal		6/5	y
41	Wire	1.90	7.6	Pd, Marsh. 0.032"	HNO3	none	0.2N, thio	MSD	Pt	plastic(3)	torr seal	0.86	6/21	y
43	Coin	4.69		Pd(1) + B	Std	none	0.2N, As2O3	MSD	Pt	plastic(3)	torr seal		6/24	y
71	Strip	0.73	2.5	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.55	9/7	y
72	Strip	0.73	2.1	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.81	9/7	y
73	Strip	0.68	1.0	Pd(2)	C	paraffin	0.2N	MSD	Ni(1)	Glass(5)	glass	0.81	9/7	y
75	Strip	0.50	1.4	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.71	9/7	y
77	Strip	0.72	1.4	Pd(2)	C-S	paraffin, H2S	0.2N	MSD	Pt small	Glass(5)	glass	0.69	9/7	y
78	Strip	0.66	1.4	Pd(2)	none	none	0.2N	MSD	Ni(1)+S	Glass(5)	glass	0.79	9/7	y
79	Wire	0.37	2.0	Pd, Martin	HNO3	none	0.2N, Ag	MSD	Ni(1), Ag	Glass(5)	glass	0.94	9/7	y
98	Strip	0.79	1.8	Pd(2)	HNO3	none	0.1N, 3.5mg WF	Aldrich	Ni(2)	Glass(5)	glass	0.93	12/15	y

TABLE VII (Continued) Description of Cell Treatment

A   coin = arc melted and rolled into coin shape
sheet = 0.05" thick sheet cut into square shape
Strip = Strip cut from sheet or button to give a parallelepiped
wire = a cylinder having a diameter from 0.03" to 0.04"
button = used as arc-melted
am = used after arc-melting
sw Ni = spot welded to nickel lead
Marshall or Martin = source of wire

B   Alloys made by arc melting Pd with the other component
Pd(1) = Johnson Matthey powder, Batch V7114307
Pd(2) = Englehart sheet
Pd(3) = Johnson Matthey powder, Batch V8368501
Pd(4) = Englehart Powder, Batch A4934
Pd(5) = Englehart Powder, Batch F01567
sw = spot welded using tungsten electrodes, otherwise copper electrodes were used.

C   Various surfaces applied as described under "D"
std. = sanded with 200 grit paper and washed with nitric acid none = not cleaned

D   H2S+C = heated in H2S mixed with paraffin vapor
paraffin, H2S = heated in paraffin vapor first than in H2S
H2S+H2O = heated in H2S that was bubbled through H_2O
H2S = heated in pure H2S to form Pd sulfide on surface
Oxidized = heated in air to produce microfractures on surface
annealed = heated in vacuum

E   thio = thiourea added to electrolyte
pe = current was passed through electrolyte using a Pt dummy electrode before Pd was
used
All cells after #70 contained stainless hypodermic needles
Hg = mercury metal in electrolyte
U = uranium metal in electrolyte
WF = Whole wheat flour added

F   MSD = D_2O from MSD Isotopes, 99.9 at. %
Aldrich - D_2O from Aldrich Chemical Co., 99.9 at %
pe = preelectrolyzed using a Pt cathode and anode

G   Pt = platinum gauze in a large "C" shape around cathode
Ni wire = nickel wire was made into a spiral (0.3" ID) around the anode
Ni+S wire = Ni wire heated in H2S
Pt small = Pt gauze made into 0.3" in diameter tube that surrounded the cathode

Ni gauze = gauze obtained from Texas A and M
Ni(1) = 0.04" wire from Driver-Harris Company
Ni(2) = 0.04" welding wire
Ni(3) = 0.06"
Ni(4) = 0.04" protonic JM batch W12918

H   Glass = flint glass
Plastic = polyethylene
Cell Design #1 = 120ml glass jar
Cell Design #2 = 120ml glass jar with 40 ml plastic insert
Cell Design #3 = 100ml plastic jar
Cell Design #4 = 100ml plastic jar with glass tube covering cathode lead. Recombined
    liquid returned to cell.
Cell Design #5 = 120ml glass jar with recombined gas collected separately.
Cell Design #5a = 10ml glass test-tube with recombined gas collected separately.
Cell Design #5b = 120ml glass jar with recombined gas collected separately in hot water
    bath.
Cell Design #2a = 120ml glass jar with 40 ml plastic insert-recombinate collected
    separately

I   torr seal = Pt wire from Pd electrode was covered with Torr Seal to prevent Cu and Pb
    pickup
glass = lead covered with glass tube no = bare lead

J   Maximum determined from weight gain

K   Month and day cell started

L   y = excess tritium measured
n = no excess tritium measured
? = no tritium measurement made