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Electrolytic Tritium Production

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ABSTRACT

Fifty-three electrolytic cells of various configurations and electrode compositions were examined for tritium production. Significant tritium was found in eleven cells at levels between 1.5 and 80 times the starting concentration after enrichment corrections are made.

INTRODUCTION

The possible production of nuclear products and heat in an electrolytic cell, as first proposed by Pons and Fleischmann[1], and Jones et al. [2], is so hard to accept that extraordinary experimental efforts need to be made to prove that the phenomenon exists at all. This is especially important in view of several complete studies that have failed to report positive results[3; 4; 5]. This study is being reported in order to add to the growing number of observations[6; 7; 8; 9; 10; 11; 12] that tritium, at least, can be made in an electrolytic cell.

Tritium appears to be the least ambiguous and most easily measured product of the “Cold Fusion” effect. In addition, the production of tritium has implications at least as important as the production of heat, although these two products may not originate from the same process. The authors recognize that this work is still incomplete and leaves many questions unanswered. However, the results are supported by such a large and consistent data base that reporting of tritium production is warranted even before a full understanding of the process is available.

The early phase of this study was started shortly after the announcement of possible fusion in palladium. Simple construction and inexpensive diagnostic methods were dictated by funding restraints. Initially we tried to understand how to achieve a high D/Pd ratio. A cell was designed that would allow the cathode to be weighed, thus giving a measure of the deuterium content. This consisted of a wide mouth, 120 ml glass jar with a small electrical socket in the lid that allowed the cathode assembly to be plugged in. Periodically this assembly was unplugged, washed with acetone, dried and weighed. Unfortunately, the electrolyte was found to attack the solder and copper in the plug thereby causing Cu, Zn and Pb to plate onto the cathode surface. Thus, all early cathodes had a surface contamination of these elements. This surface impurity was reduced but not eliminated in some later cells by coating the lead and plug with Torr Seal (an epoxy). Although one electrode having detected Cu, Zn and Pb on the surface produced tritium, subsequent cells were redesigned to eliminate this contamination. An additional problem occurred because the Torr Seal absorbed deuterium causing the apparent D/Pd ratio to be too large. We concluded that D/Pd ratios above ≈0.82 were not only unlikely to occur at room temperature, but appeared to be unnecessary to produce tritium. After the first tritium was observed, emphasis shifted to reproducing this event.

This paper concentrates on those cells that produced tritium. Over 1500 electrolyte samples have been analyzed for tritium which has given an excellent statistical basis for evaluating tritium production.

Three sets of experimental data will be described:

1. sixteen early cells that gave, in two cases, relatively large amounts of tritium and, in two cases, small amounts, but were not well characterized,

2. a set of 13 closed cells with recombination[13] that gave no tritium but provided a good example of counting statistics, and

3. nine cells with external recombination that were followed in detail but gave only small amounts of tritium in seven cells.

EXPERIMENTAL

Data Set 1

The first tritium was produced in the cell design shown in Fig. 1. A 120 ml glass jar contained a 60 ml plastic insert into which the electrolyte [14] (0.2N LiOD[15]) was placed. Gases generated in the cell were allowed to leave though a small hole. The anode was a gauze of Pt metal, approximately 2.5 cm long and 2.5 cm in diameter. The cathode was made by arc-melting powder [16], and rolling the resulting button into a coin-shaped disc ≈2 mm thick and 1.5 cm in diameter. This was spot welded to a Pt wire that was soldered to a small plug. The assembly was plugged into a small socket that was sealed through the lid using Torr Seal. Torr Seal was applied to the Pt lead between the cathode and the plug. In spite of this precaution, Cu and Pb were found on the surface of cathode #30 when the electrode was examined after tritium production had stopped. We do not know whether the continuing increase of these contaminates might have eventually stopped tritium production. However, none of the cells having bare leads, where these contaminates are present in greater quantity, showed signs of tritium production.

Cells of this design typically showed a slow transfer of electrolyte from the plastic insert to the bottom of the glass jar. This transferred liquid is called the spillover. Tritium contained in this liquid would have accumulated gradually as the slow transfer occurred. Usually about 20-30 ml had transferred by the time these cells were terminated.

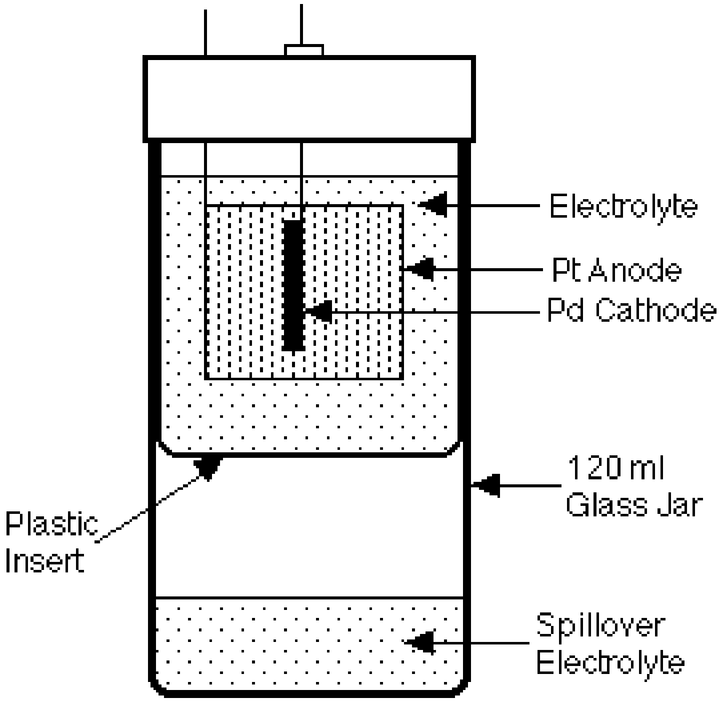

FIGURE 1. Cell design #1.

Several of the Pd coins were pretreated by heating them in H_2S that was made by heating sulfur and paraffin together. The resulting mixture of H_2S and paraffin vapor produced a black layer that, when sufficiently thick, was an electrical insulator. Deuterium uptake would not occur unless the electrode was electrolyzed using reversed current (anodic). This caused most of the black layer to flake off and the subsequent cathodic electrolysis to charge the electrode with deuterium in the normal way. Unfortunately, this work was being done during the early, exploratory stages when minimal funding was available for detailed surface characterization. This treatment was done in an attempt to poison atomic recombination at the surface and increase the D/Pd ratio. Details of this study will be described in subsequent papers.

Tritium measurements[17] were started about 10 days after the first cells of this series were turned on. Table I shows the materials used in the cells and Table II lists the count rate based on 10 min counts. Although many of the cells did not produce tritium, they are included to show that cells running at the same time were not contaminated by the common source of D_2O, Li, electrode materials or materials used for the surface treatment.

TABLE I

Materials used in cells #26 to #44

#	Shape	Wt (g)	Area (cm^2)	Alloy	Surface Treatment	Pre-Treatment	Electrode Poison	Lead Covering	Max D/Pd	Excess T
26	coin	4.9	3.7	Li/Pd=0.047	std	none	thio	torr seal	0.82	n
27	coin	6.4		Ni	std	none	none	torr seal	0.09	n
28	coin	6.4	5.0	Rh/Pd=0.1	std	none	thio	torr seal		n
29	coin	5.0	4.8	Pd	sulfide	H_2S+C	thio	torr seal		y
30	coin	6.8	5.1	Pd	sulfide	H_2S+C	none	torr seal		y
31	coin	6.2	5.0	Pd+Rh+Li	std	none	thio	torr seal		n
33	coin	6.1	5.0	Li/Pd=0.051	std	none	thio,Cu,Pb	no	0.77	n
34	coin	6.2	5.0	Li/Pd=0.023	std	none	thio,Cu,Pb	no	0.83	n
35	coin	6.4	5.0	Li/Pd=0.012	std	none	thio,Cu,Pb	no	0.77	n
36	coin	7.0	6.0	Rh/Pd=0.1	std	none	thio,Cu,Pb	no	0.86	n
37	coin	6.6	5.5	Rh/Pd=0.1	std	none	thio,Cu,Pb	no	0.90	n
40	coin	8.4	7.0	Pd	sulfide	H_2S+C	none,Cu,Pb	no	0.84	n
41	wire	3.2	7.6	Pd	none	none	thio	torr seal	0.86	y
42	coin	6.1	5.0	S/Pd=0.0043	std	none	none	torr seal		n
43	coin			B/Pd=0.026	std	none	As_2O_3	torr seal		y
44	coin	6.5	5.2	Pd	sulfide	H_2S+C	none	torr seal	0.75	n

coin= Arc-melted powder and rolled into coin shape
wire= 0.032” diameter
std=Sanded with 200 grit paper and washed with HNO_3
H_2S+C= Heated in vapor produced by a mixture of paraffin and sulfur
thio= 0.0004 g/ml thiourea added to electrolyte.
Cu,Pb = probable impurity on cathode surface owing to attack of solder and connector
Cells that are thought to produce excess tritium are indicated by “y”
All anodes are Pt and the cells are plastic

TABLE II

Gross count/min-ml in cells running when tritium measurements were started.
(10 min count using 1 ml of electrolyte)

	Cell Number															
DATE	**26**	**27**	**28**	**29**	**30**	**31**	**33**	**34**	**35**	**36**	**37**	**40**	**41**	**42**	**43**	**44**
Start=	6/1	6/1	6/2	6/5	6/5	6/8	6/15	6/15	6/15	6/17	6/17	6/20	6/21	6/24	6/24	6/26
6/13 0	77c	80	78c	c												
6/14 1	77	79	71	**182**		97										
6/15 2	76c	77	74c	**154**	**4204**	68										
6/16 3	103	99	87	**125**	**3711**	81	68	73	71							
6/17 4	80	78c	57	**122**	**3730**	80c	75c	78c	77c							
6/18 5	92	83	88	c	130c	83	91	85	85							

Table II, continued

DATE		**26**	**27**	**28**	**29**	**30**	**31**	**33**	**34**	**35**	**36**	**37**	**40**	**41**	**42**	**43**	**44**	
6/19	6	71c	67	70	67	111	72	75	70	67c	76	69						
6/20	7	73	71	72	75	78	73	c		80	80	80						
6/21	8	73	70c	70	70	80	80	69		72	83c	76	65					
6/22	9	64	68	69c	71c		75c	66		67	67	95	111	70				
6/23	10			120	74	64	69	77		75	68	68	77					
6/24	11			80	77	75	76				66	64	70	69				
6/25	12	84	76	83	99	88					129	70	103	80	88	86		
6/26	13	c			110	93								110	102	**112**		
6/27	14	120			**133**	110								128	120	**135**	103	
6/28	15	105			**120**									118	103	**113**	83	
6/29	16	103			**121**									105	103	**126**	83	
6/30	17				**126**									**114**	105	**123**	80	
7/1	18	a			**139**									**118**	102	**128**	97	
7/2	19	77			**131**									**115**	95	**118**	88	
7/3	20	91			**134**									**126**	100	**114**	88	
7/4	21	78			**130**									**117**	95	**122**	100	
7/5	22	80			**131**										108	**128**	116	
7/6	23	69			78									**110**	85	108	91	
7/7	24	72			94									**108**	86	111	90	
7/8	25	85			100									**225**	96	113	134	
7/9	26	83			97											85	102	94
7/10	27	83			96											88	109	88
7/11	28	79			101											94	102	95

Samples showing what we believe to be excess tritium are indicated in bold type. Other, isolated high values are assumed to be caused by sampling errors.
c=electrolyte changed
a=new cell

Four (29, 30, 41 and 43) cells show an indication of tritium production. The values which we believe show excess tritium production are indicated by bold type. Cell #30 gives a disintegration rate[18] of $1.1x10^4$ d/min-ml (x80 increase) which, as indicated, is approximately 80 times the tritium content of unused electrolyte. This level was found when measurements were started 10 days after the cell was turned on. No additional tritium was produced during the next three days. A current reversal after this time produced no additional tritium. Cell #29 showed only a little excess tritium in the electrolyte. This, we believe, was owing to a small amount of original electrolyte in the new solution. A little of the original electrolyte remained in the cell after decanting it from the material that flaked off during anodic operation. This change was made about 30 hours before the first tritium measurements were made. Production of significant tritium in this cell before the change is indicated by activity in the spillover that gave a disintegration rate of $6.5x10^3$ d/min-ml (x47). This spillover had accumulated since the cell was turned on and had not been changed. Some small activity was found in cells #41 and #43, being 540 d/min-ml (x4) and 280 d/min-ml (x2), respectively. Unfortunately, cell #41 was turned off just as an indication of increased production was occurring.

The charging history of the two most active cells is shown in Fig. 2. The sudden increase in the D/Pd shown by cell #29 after 100 hrs and the slow rise shown by cell #30 after 150 hr are, we believe, an artifact caused by the Torr Seal.

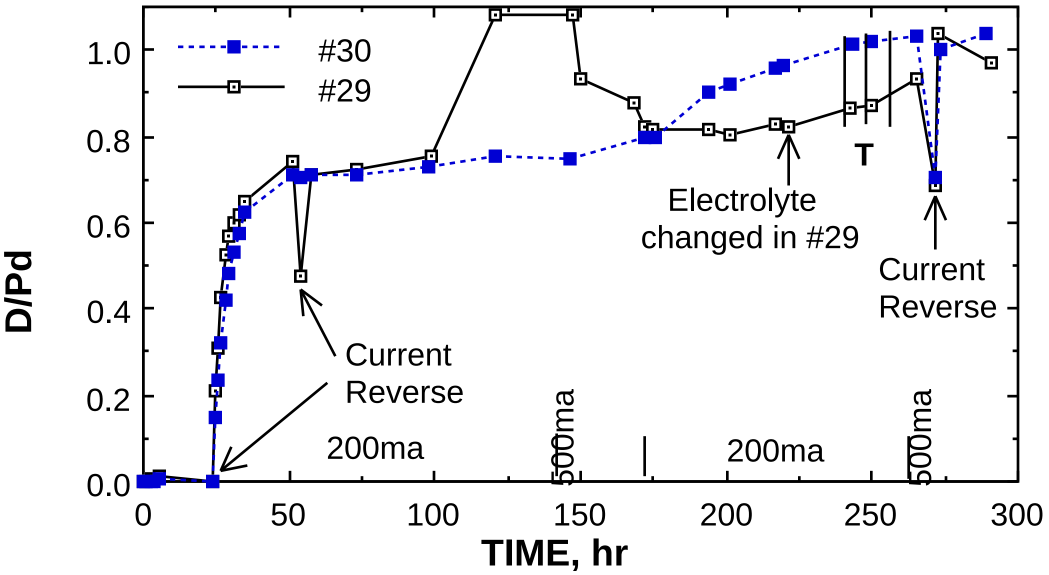

FIGURE 2. Charging history of cells #29 and #30. Tritium measurements were made at times indicated by the vertical lines and the letter T.

The electrolyte in cell #29 was changed two additional times. Another small increase in tritium level was seen after about 30 days from the start. No detectable tritium was found in the electrode at the end of the study.

Cells #41 and #43 also produced minor amounts of tritium. Cell #41 contained about a foot of 0.032” (0.81 mm) diameter wire[19] that was wound into a spiral. Cell #43 was an alloy of Pd and B that was made by arc-melting the elements together. Although the tritium content of these cells is small, the amount is significant because the abrupt increase is not consistent with the gradual increase from enrichment. The amount is well outside the uncertainty in counting error as will be demonstrated later. The enrichment phenomenon also will be discussed in detail later in the paper. The few high, isolated high counting rates were ignored as being possible sampling errors.

Inactive cells running at the same time (#26, #27, #28, #31, #33, #34, #35, #36, and #37), using the same heavy water, gave an average disintegration rate of 154±31 d/min-ml based on 96 data points shown in Table II. This value is somewhat higher than the tritium level later measured in closed cells because of enrichment in these open cells.

In order to verify that the high counts in cells #29 and #30 were not caused by chemiluminescence, three procedures were applied: (1) part of the sample was neutralized, distilled and counted; (2) the light spectrum[20] was measured and compared to a known tritium sample; and (3) the undistilled sample was recounted after sitting a month. All three procedures were consistent with there being tritium in the samples. A similar result was obtained when sample #30 was counted by another group at LANL.

Data Set 2

Gradually the cells were modified. First, plastic was substituted for glass and then recombiners[13] were added. The recombinate ran back into the cell and any excess gas left the cell through a small hole. About 16 cells were studied in this way.

Examples of the counting rate are shown in the Table III.

TABLE III

Gross counting rates (c/min-ml) of closed cells run with recombiners.
(10 min count using 1 ml of electrolyte)

		Cell Number												
DATE	**DAY**	**49**	**51**	**54**	**55**	**56**	**57**	**58**	**59**	**60**	**61**	**62**	**63**	**65**
Start=		8/15	7/4	7/31	7/31	7/31	8/9	8/9	8/9	8/9	8/9	8/9	8/9	8/15
8/11	0		80	70	70	80	60	70	70	70	70	70	70	
8/14	3		75	71	68	71	79c	65	68	66	66	66	*58*	
8/15	4	78	77	87	80	81	78	78	79	91	82	*98*	*89*	69
8/16	5	65	79	65	67	73	67	68	d	d	67	71	75	73
8/17	6	61	71c	69	69	68	66	65	d	d	68	68	68	67
8/18	7	68	70	70	70	71	70	70	d	d	69	69	73	66
8/21	10	70	72	67	71	71	73	68	d	d	70	69	78	69e
8/22	11	71	72	70	70	65	70	66	*106*	71	70	73	80	68
8/23	12	64	68	72	70	69	80	71	82c	68c	63c	65	71	70
8/24	13	66	71	74	75	68	68	69	61	70	65	69	72	70
8/25	14	71	70	72	67	70	66c	66	70	68	70	71	75	66
8/26	15		82	73	75		81	80	65	70	*56*	73	74	71
8/27	16	60	76	70	71	71	*129*	73	69	73	71	72	76	72
8/28	17	68	73	68	73	72	79	72	72	68	59	68	66	66
8/29	18	67	77	72	75	67	69	74	71	74	72	76	78	69
8/30	19	67	72	73	77	71	94	66	70	75	74	68	70	65
8/31	20	73	83	71	71	69	73	70	73	80	75	77	71	63
9/1	21	66	76	72	75	71	84	68	69	77	68	69	71	66
9/5	25	71	76	72	71	74	84	69	68	70	67	70	71	73
9/6	26	71	75	69	74	73	88	71	69	73	69	71	71	69
9/7	27	68	77	74	79	76	90	74	71	72	70	70	73	66
9/8	28	74	81	73	73	82	90	74	75	76	77	73	69	68
9/11	31	73	79	74	74	75	80	77	76	81	78	72	79	70
9/12	32	69		67	74	73	76	72	67	67	72	72	74	69
9/13	33	72		69	68	76	89	70	72	80	71	73	76	67
9/14	34	*50*		70	70	80	90	80	70	80	70	70	70	70
9/15	35	71		71	71	70	82	74	72	75	70	73	73	64
9/18	38	71		71	76	73	83	75	76	74	75	71	73	70
9/19	39	73		75	68	71	82	70	72	77	75	73	75	68

Table III, continued

		Cell Number												
DATE	**DAY**	**49**	**51**	**54**	**55**	**56**	**57**	**58**	**59**	**60**	**61**	**62**	**63**	**65**
9/20	40	81		75	74	77	84	67	80	78	73	77	78	65
9/21	41	67		68	72	76	78	70	73	72	71	70	79	70
9/26	46	82R					86R				85R	79R		70R

e= gas recombination explosion; c=new electrolyte; R= after current reverse

d=Low count because of suspended black nickel oxide
Values in italic were eliminated in the data treatment

As can be seen in Fig. 3, where two typical cells are plotted as a function of time, the tritium values were essentially constant as would be expected for inactive closed cells. The line is a least squares fit to sample #62. The tritium disintegration rates for these

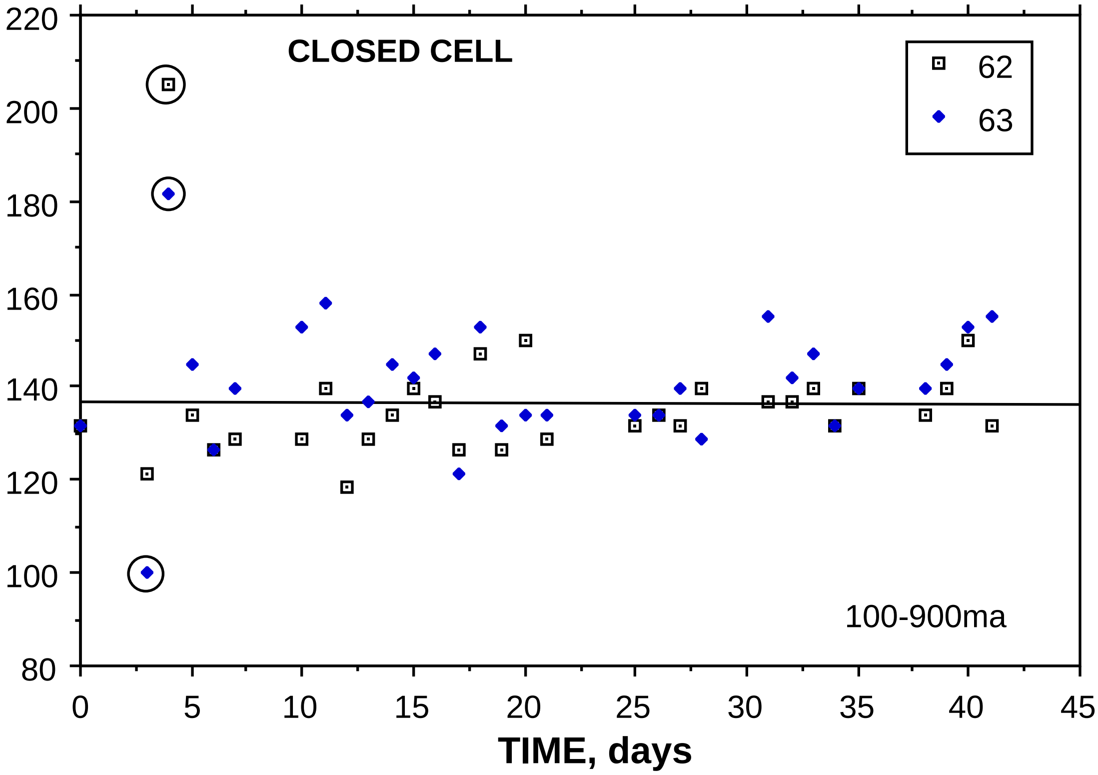

FIGURE 3. Example of closed cell behavior. Encircled points were eliminated in the data treatment.

inactive cells are listed in Table IV along with the standard deviations. The scatter in these data is useful in evaluating the total random errors associated with counting, with transferring the electrolyte to the scintillator fluid and with changes in background. An evaluation of these inactive cells indicates an average standard deviation of ±14 d/min-ml involving a 41 day period, 13 cells and 446 measurements. However, because a few very high or low points are occasionally seen, claim of tritium production is only based on a consistent pattern of values that is well outside this error. In addition, this data set gives an average value of 138 d/min-ml as the disintegration rate of tritium in the electrolyte used in this work. During the duration of the studies described in this paper, approximately 30 one-liter bottles of heavy water[14] were used with no significant difference in tritium activity from this value or between individual bottles.

The background is based on using 1 ml of old well-water[21] in place of the electrolyte and counting for 10 min. Data for 100 days are shown as a function of time in Fig. 4. This time interval overlaps data sets #2 and #3. There is no apparent trend with time and the average value is 19.3± 2.4 c/min. A rounded value of 20 c/min is used in all calculations.

TABLE IV

Tritium disintegration rate and standard deviation for inactive cells in data set #2

Cell Number	**d/min-ml**	**# of Values**	**Cell Number**	**d/min-ml**	**# of Values**
#49	128±15	28	#58	134±11	31
#51	146±11	23	#59	136±12	26
#54	134±10	38	#60	142±14	27
#55	137±12	38	#61	132±14	31
#56	138±12	36	#62	134±7	30
#57	154±23	30	#63	141±9	29
			#65	127±7	28

The absence of any tritium production activity in these cells suggests that the plastic container or the recombiner material added something to the electrolyte that inhibited the reaction. Consequently, future cells were made of glass and were designed so that the recombinate did not flow back into the cell. This design allowed the tritium content of the gas to be determined and, from this information, the distribution factor of tritium between liquid and gas could be calculated.

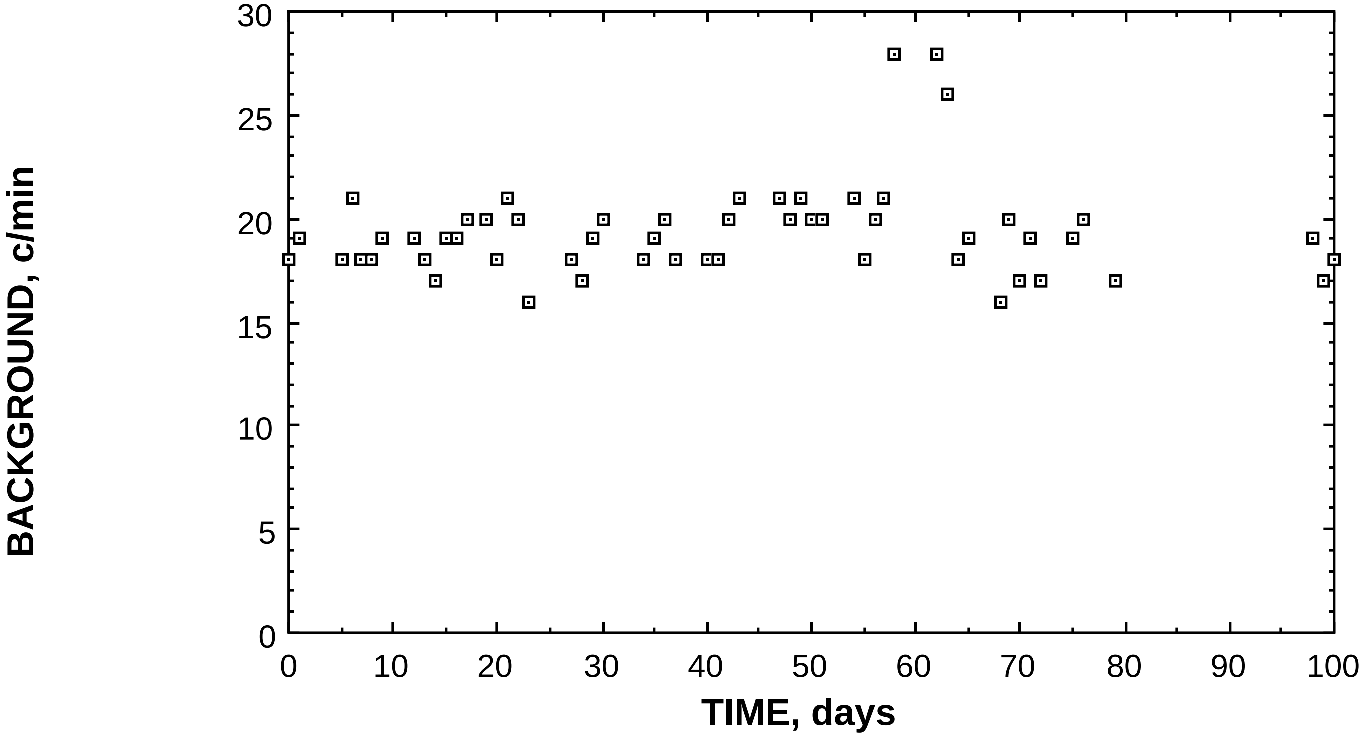

FIGURE 4. Counting background during time interval of data sets #2 and #3 based on 1 ml of tritium-free water.

Data Set 3

The latest group of cells have the design shown in Fig. 5. Here the recombinate is collected in a plastic IV bag. The cell is a glass jar containing 120 ml of fluid and the anodes are either Pt gauze or Ni wire. In each case, there is a minimum distance of about 5 mm between the anode and cathode. Parafilm was wrapped around the jar to seal the lid, a glass tube protected the lead from electrolytic attack and Torr Seal was applied where the leads passed through the lid. A hypodermic needle was used to attach the IV system to the cell. Thus, stainless steel was in the cell in addition to 0.2N LiOD and the anode material. A small disc of silicone rubber was placed, in some cases, on the end of the cathode to prevent shorting to the anode.

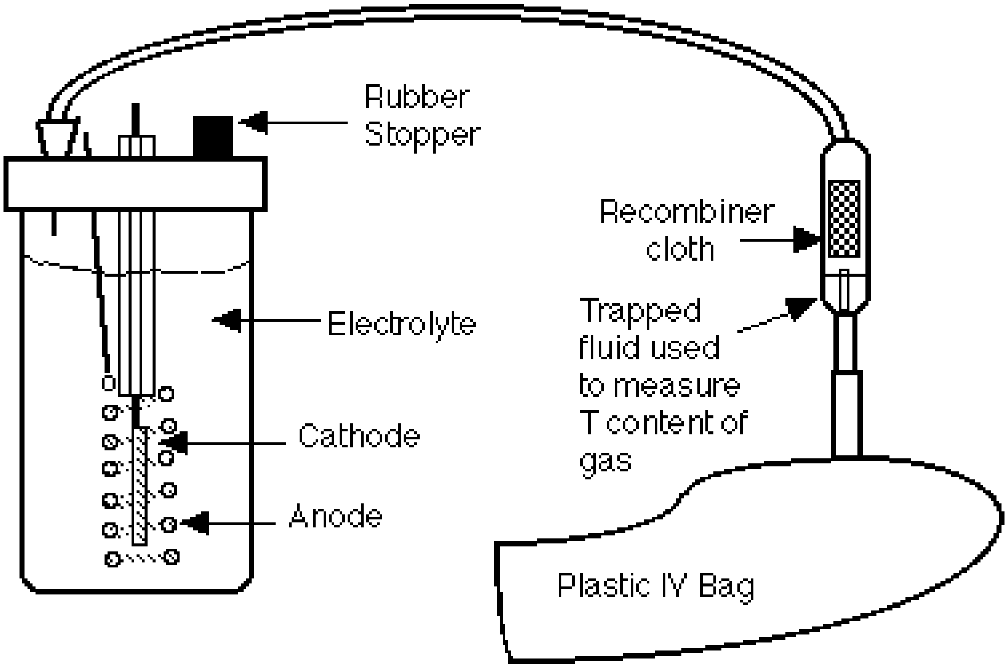

FIGURE 5. Cell design #5.

Seven of the nine cells that were started at the same time and were running in series at constant current[22] show evidence of tritium production. Two were apparently inactive. The design details of this group are shown in Table V and the counting rates are listed in Table VI.

Two cells, #70 and #79, used cathodes made from 1.0 mm wire[23]. Both used a nickel wire anode but #79 had a small piece of silver attached. Cells #71 to #78 had Pd cathodes cut as a 1 mm strip from a 1.3 mm thick sheet[24]. All of the strips except one (#78) were heated in paraffin vapor. A few were subsequently heated in H_2S. Although the temperatures and times were arbitrary, a weight increase caused by interaction with the paraffin and H_2S was noted. This weight change is noted in Table V. At the end of the study, the cathodes were weighed to determine the deuterium content and the listed D/Pd ratios were calculated. The cathode in cell #71 showed a nonuniform surface discoloration and Cell #79 had a heavy, dark surface covering that was probably silver.

Cells #71,#72, #75 and #77 had an anode made from Pt gauze that was formed into a 1 cm diameter tube. The Ni anode in cells #70, #74 and #78 was heated in H_2S before use.

Periodically, 0.5 ml of fluid was removed from the region below the recombiner in the IV drip system where about 1 ml of recombinate was trapped. This was analyzed for tritium and is designated with a “g” in Table VI.

TABLE V

Materials used in cells #70 to #79

#	Shape	Surface Treatment	Method	Anode	Excess Tritium	D/Pd
70	wire	none	HNO_3	Ni+S wire	no	
71	strip	C(0.0001g)-S(0.0013g)	paraffin, H_2S	Pt gauze	yes	>0.55
72	strip	C(0.0001g)-S(0.0000g)	paraffin, H_2S	Pt gauze	yes	0.81
73	strip	C(0.0002g)	paraffin	Ni wire	yes	0.81
74	strip	C(0.0003g)	paraffin	Ni+S wire	no	0.78
75	strip	C(0.0001g)-S(0.0000g)	paraffin, H_2S	Pt gauze	yes	0.71
77	strip	C(0.0002g)-S(0.0004g)	paraffin, H_2S	Pt gauze	yes	0.69
78	strip	none	HNO_3	Ni+S wire	yes	0.79
79	wire	none	HNO_3	Ni wire+Ag	yes	<0.94

All electrode areas = ≈1.3 cm^2

HNO_3= cleaned with dilute HNO_3

Ni+S=Nickel wire heated in H_2S

Ni wire+Ag= A small piece of silver is attached to the nickel wire

All cells are glass.

TABLE VI

Gross count/min-ml obtained from cells #70 to #79

Recombinate counting rates are identified with a “g”

DATE	70	70g	71	71g	72	72g	73	73g	74	74g	75	75g	77	77g	78	78g	79	79g
9/5	Start																	
9/6																		
9/7	64		Start		Start		Start		Start		Start		Start		Start		Start	
9/8	79				74				59		69		84		75		67	
9/11	62		87		56		60		57		66				83		89	
9/12	64		99		80		77				90		80		84		88	
9/13	60		80		86		85		58		94		56		86		86	
9/14	70	80	100	100	90	80	100	100	70	60	100	100	90	100	90	80	100	100
9/15	73		106		97		110		73		101		113		109		103	
9/18	73		129		95		135		63		123		122		98		109	
9/19	65		116		89		128		67		119		122		106		107	
9/20	67		116		97		135		72		128		123		107		112	
9/21	71	62	109	106	93	90	122	93	70	77	119	115	117	96	105	93	101	91
9/22	74		116		105		133		72		122		121		108		112	

Table VI, continued

DATE	**70**	**70g**	**71**	**71g**	**72**	**72g**	**73**	**73g**	**74**	**74g**	**75**	**75g**	**77**	**77g**	**78**	**78g**	**79**	**79g**
9/25	77		118		107		147		67		124		131		120		130	
9/26	77	63	121	98	106	87	145	121	68	73	129	107	117	101	113	96	119	93
9/27	77		121		108		153		74		120		126		120		121	
9/28	83		128		115		164		78		126		133		124		127	
9/29	76	69	118	91	106	90	144	102	72	75	129	110	133	90	124	100	126	99
10/2	86	78	132	99	107	88	147	122	85	75	136	119	123	124	141	132	122	
10/3	79		113		121		167		77		148		123		123		127	
10/4	82	75	121	110	118	95	165	117	76	97	150	124	132	108	129	100	125	
10/5	84		126		115		151		83		151		131		130		136	
10/6	78	80	115	106	106	98	124	107	80	84	122	115	122	144	115	115	118	
10/10	78		111	110	108	92	123	105	81	108	116	128	116	103	111	101	119	
10/11	78		117		107		130		81		112		133		112		116	
10/12	84		118		103		118		80		123		116		108		112	
10/13	81	76	118	103	109	99	116	114	76	77	115	102	118	94	104	102	115	
10/16	82	80	111	89	111	93	118	99	77	80	116	106	116	93	115	96	113	
10/17	76		120		113		110		81		114		115		108		108	
10/18	77		110		107		111		83		118		121		109		106	
10/19	80		116		106		104		81		115		121		109		109	
10/20	82	68	111	90	111	83		85		79	123	88	113	89	111	87	109	90
10/23	83	71	110	89	105	80	116	83	75	82	114	92	115	93	109	81	96	83
10/24	86		106		106		105		79		110		112		106		97	
10/27	79		113		115		110		79		114		116		109		96	
11/3	90	74	116	98	121	98	114	99	84	75	130	99	119	98	105	105	100	91
11/6	84	91	111	91	107	90	105	82	75	95	117	106	117	106	106	101	92	88
11/7	84		109		109		108		75		107		120		107		90	
11/9	80	92	107	108	104	98	105	100	76	91	111	111	110	112	103	103	89	87
11/13	103	80	109	101	109	91	105	106	73	86	102	102	117	90	103	93	87	89
11/15	90		102		107		105		73		106		115		106		90	
11/17	87		109		110		95		74		106		119		109		88	

The gas samples are based on 0.5 ml of recombinate and, therefore, have a larger uncertainty (±0.02 ml) than the electrolyte (±0.01 ml) which is based on 1 ml.

Cells # 70 and #74 showed no detectable excess tritium. Nevertheless, the two inactive cells had a slow, similar increase in tritium content[18] owing to enrichment, as can be seen in Fig. 6. This contrasts with the closed cell behavior seen in Fig. 3. During the first 30 days, the distribution factor of both cells appears to be independent of time and current within the scatter of the data. After the current was reduced at day 31, cell #74 began losing tritium preferentially into the gas. Cell #70 continued to show an expected increase in tritium content. The average distribution factor (gas/liquid) for Cell #74 is 1.12±0.17 (13 data sets) compared to a value of 0.91±0.16 (15 data sets) for Cell #70. These values are rather uncertain because the gas samples are based on only 0.5 ml of fluid taken using a hypodermic syringe rather than a calibrated pipette. Consequently, only the trend to a higher value for Cell #74 compared to Cell #70 can be accepted.

The scatter in counting rate for the electrolytic samples, which is approximately ±5 c/min-ml (±14 d/min-ml), is caused by the sum of errors in sampling, counting and changes in background. This value is consistent with the behavior of data set #2 as described previously.

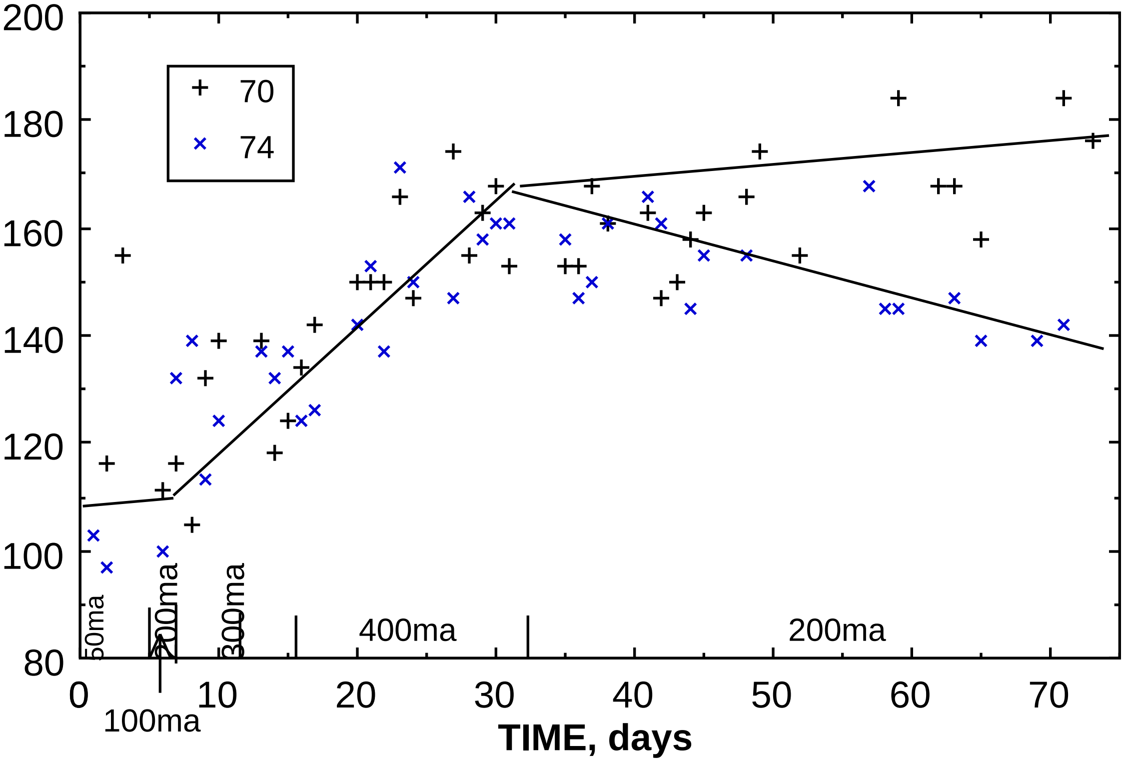

FIGURE 6. Time variation of the tritium concentration in inactive cells.

The cells that have produced tritium are compared in Fig. 7. There is a range of values but each exceeded the count rate of the inactive cells after about 2-3 days and continued to increase in steps. Several of the cells have shown especially

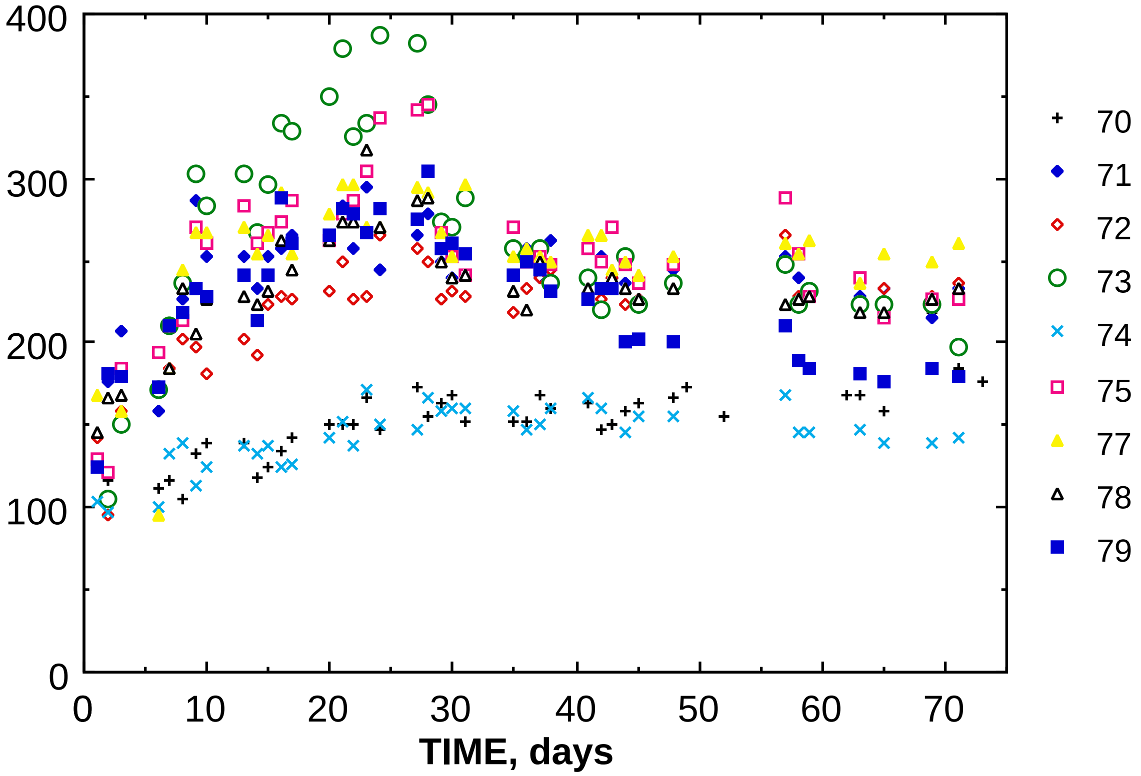

FIGURE 7. Comparison between all cells in this series.

interesting behavior. Two most active cells (#73 and #75) are compared to an inactive cell (#70) in Figure 8. The scatter of values for the active cells is larger than found for inactive cells indicating possible production of tritium bursts. In general, the recombinate followed the count rate of the electrolyte, but had a smaller value. Thus, there is no indication that significant tritium has left the cell by means of the gas phase, in contrast to the experience at Texas A & M University[6] where much higher production rates were observed. Occasionally more tritium is found in the recombinate than in the liquid such as on 10/6 in #77 or on 10/10 in #75. These values might have resulted because of short tritium bursts preceeding the sample or because of sampling errors. The absence of a pattern causes us to ignore such values.

It is interesting that all the cells that produced tritium appeared to start and stop production at approximately the same time. Once tritium production stopped, the amount of tritium appeared to decrease over several days to a constant value that is significantly above that shown by inactive cells.

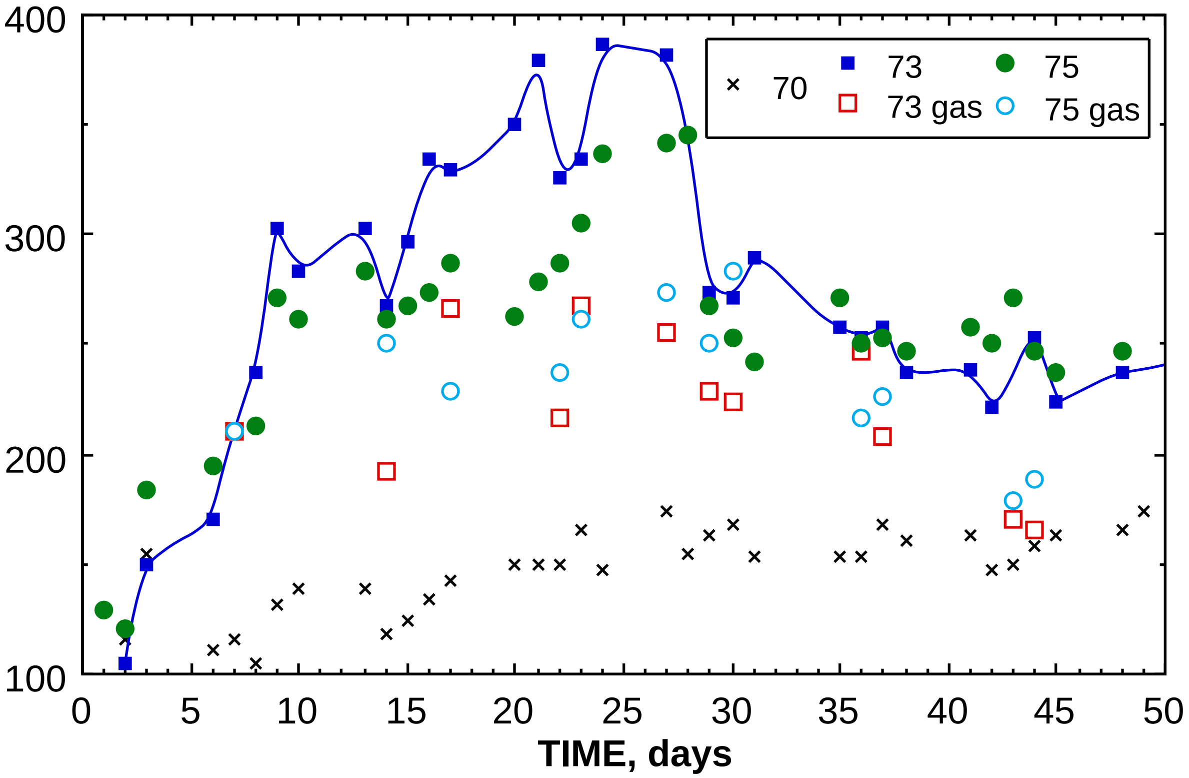

FIGURE 8. Tritium counts for two active cells and one inactive cell. The tritium activity in the recombinate is shown as open symbols.

A cell containing 0.2 N LiOH (normal water electrolyte), a Pd cathode and a Ni anode has been studied recently for 30 days with no increase in tritium concentration. Although this is a relatively short time, it is longer than is necessary to produce tritium in D_2O cells that become active. Such water cells will be included in future work.

DISCUSSION

Production of excess tritium in cells #71, #72, #73, #75, #77, #78, and #79 is evident in the data shown in Fig. 7 where a comparison is made to the two inactive cells. In order to determine the magnitude of this excess, the effect of enrichment must be subtracted. Based on measurements of tritium in the gas and electrolyte of cells #70, #73, #74 and #75, a distribution factor of 0.84 was chosen to calculate the excess tritium in cell #73. This calculation will show that more tritium was found in cell #73 than was present initially or was in the D_2O that was added as electrolyte replacement.

The distribution factor in this study is defined as the tritium concentration of the gas divided by the tritium concentration of the liquid. A value less than unity is calculated when the tritium content of the liquid increases while a value greater than unity results when tritium is preferentially lost by the liquid.

The excess tritium is determined by taking the initial tritium content, subtracting the tritium lost to the gas phase, owing to electrolysis, and adding the amount used to replace that volume lost by the cell [25]. The excess is the difference between the resulting value and the measured amount of tritium determined daily. Table VII lists the daily inventory of tritium in cells #70 and #73. The gross counting rate, shown in column 2, includes a background of 20 c/min-ml. Column 3 shows the net decomposition rate that is calculated by subtracting 20 from the value in column 2 and dividing by the machine efficiency, 0.38. Estimated values of net decomposition rates are shown in bold type for days when no measurement was made, generally holidays. The uncertainty in these estimates has a negligible effect on the final result. Electrolytic action, owing to the current shown in column 4, caused D_2O to be lost and the daily amount and running sum are shown in columns 5 and 6, respectively. The value is calculated using equation (5) at the end of the table. This lost D_2O contains less tritium than does the electrolyte because it was produced from the gas. Using the distribution factor, the tritium concentration of this electrolyzed liquid is calculated and the running sum is shown in column 8. This volume of D_2O plus that needed to replace the sampling volume (1 ml on days when a sample was taken) was added to the cell. The amount of added tritium is calculated from its known concentration in the fresh D_2O using the factor listed at the end of the table. A running sum of the added tritium is listed in column 7. The excess shown in column 10 is calculated by adding the value in column 7 to the initial tritium content of the cell, subtracting the amount lost (column 8), and substracting the result from the measured daily value in column 9. These data are also given as fraction increase, shown in column 11. This quantity is obtained by dividing the excess tritium value for each day by the excess present on the first day.

TABLE VII

Tritium Inventory and Excess Calculated for Cells #70 and #73
using a distribution factor of 0.84.

Cell #70

1 Day	**2** Gross c/m	**3** Net d/m	**4** I, ma	**5** ml D_2O /day	**6** Sum D_2O	**7** Added T, mol	**8** Lost T, mol	**9** T present, mol	**10** Excess T, mol	**11** Fraction T
0		**150**	50	0.00	0.0	0.0E+00	0.0E+00	2.8E-13		
1		**150**	50	0.41	0.4	9.5E-16	7.9E-16	2.8E-13	-1.5E-16	-0.00
2	76	147	50	0.41	0.8	4.2E-15	3.9E-15	2.7E-13	-5.3E-15	-0.02
3	79	155	50	0.41	1.2	7.5E-15	7.1E-15	2.9E-13	9.4E-15	0.03
4		**150**	50	0.41	1.6	8.4E-15	7.9E-15			
5		**150**	50	0.41	2.0	9.4E-15	8.7E-15			
6	62	111	50	0.41	2.4	1.3E-14	1.1E-14	2.1E-13	-7.5E-14	-0.27
7	64	116	50	0.41	2.8	1.6E-14	1.3E-14	2.2E-13	-6.6E-14	-0.24
8	60	105	50	0.41	3.3	1.9E-14	1.6E-14	2.0E-13	-8.7E-14	-0.31
9	70	132	100	0.81	4.1	2.3E-14	1.9E-14	2.4E-13	-3.9E-14	-0.14
10	73	139	200	1.63	5.7	3.0E-14	2.4E-14	2.6E-13	-2.5E-14	-0.09
11		**140**	200	1.63	7.3	3.3E-14	2.7E-14			
12		**140**	200	1.63	9.0	3.7E-14	3.0E-14			
13	73	139	200	1.63	10.6	4.3E-14	3.5E-14	2.6E-13	-2.8E-14	-0.10

Table VII, continued

Day	Gross c/m	Net d/m	I, ma	ml D_2O /day	Sum D_2O	Added T, mol	Lost T, mol	T present, mol	Excess T, mol	Fraction T
14	65	118	200	1.63	12.2	4.9E-14	4.0E-14	2.2E-13	-6.9E-14	-0.25
15	67	124	200	1.63	13.8	5.5E-14	4.4E-14	2.3E-13	-6.0E-14	-0.22
16	71	134	200	1.63	15.5	6.2E-14	4.9E-14	2.5E-13	-4.2E-14	-0.15
17	74	142	300	2.44	17.9	7.0E-14	5.6E-14	2.6E-13	-2.9E-14	-0.10
18		**140**	300	2.44	20.3	7.5E-14	6.0E-14			
19		**150**	300	2.44	22.8	8.1E-14	6.5E-14			
20	77	150	300	2.44	25.2	8.9E-14	7.2E-14	2.8E-13	-1.7E-14	-0.06
21	77	150	300	2.44	27.7	9.7E-14	7.9E-14	2.8E-13	-1.8E-14	-0.06
22	77	150	400	3.26	30.9	1.1E-13	8.8E-14	2.8E-13	-1.9E-14	-0.07
23	83	166	400	3.26	34.2	1.2E-13	9.7E-14	3.1E-13	1.0E-14	0.04
24	76	147	400	3.26	37.4	1.3E-13	1.1E-13	2.7E-13	-2.6E-14	-0.09
25		**150**	400	3.26	40.7	1.3E-13	1.1E-13			
26		**160**	400	3.26	44.0	1.4E-13	1.2E-13			
27	86	174	400	3.26	47.2	1.5E-13	1.3E-13	3.2E-13	2.2E-14	0.08
28	79	155	400	3.26	50.5	1.6E-13	1.4E-13	2.9E-13	-1.4E-14	-0.05
29	82	163	400	3.26	53.7	1.7E-13	1.5E-13	3.0E-13	6.3E-16	0.00
30	84	168	400	3.26	57.0	1.8E-13	1.6E-13	3.1E-13	1.0E-14	0.04
31	78	153	400	3.26	60.2	1.9E-13	1.7E-13	2.8E-13	-2.0E-14	-0.07
32		**150**	400	3.26	63.5	2.0E-13	1.7E-13			
33		**150**	400	3.26	66.7	2.1E-13	1.8E-13			
34		**150**	400	3.26	70.0	2.1E-13	1.9E-13			
35	78	153	400	3.26	73.3	2.2E-13	1.9E-13	2.8E-13	-2.5E-14	-0.09
36	78	153	400	3.26	76.5	2.3E-13	2.0E-13	2.8E-13	-2.6E-14	-0.09
37	84	168	200	1.63	78.1	2.4E-13	2.1E-13	3.1E-13	3.5E-15	0.01
38	81	161	200	1.63	79.8	2.5E-13	2.1E-13	3.0E-13	-1.1E-14	-0.04
39		**160**	200	1.63	81.4	2.5E-13	2.2E-13			
40		**160**	200	1.63	83.0	2.5E-13	2.2E-13			
41	82	163	200	1.63	84.7	2.6E-13	2.3E-13	3.0E-13	-7.4E-15	-0.03
42	76	147	200	1.63	86.3	2.7E-13	2.3E-13	2.7E-13	-3.7E-14	-0.13
43	77	150	200	1.63	87.9	2.7E-13	2.4E-13	2.8E-13	-3.3E-14	-0.12
44	80	158	200	1.63	89.5	2.8E-13	2.4E-13	2.9E-13	-1.9E-14	-0.07
45	82	163	200	1.63	91.2	2.8E-13	2.5E-13	3.0E-13	-9.1E-15	-0.03
46	83	166	200	1.63	92.8	2.9E-13	2.6E-13	3.1E-13	-4.3E-15	-0.02
47	86	174	200	1.63	94.4	3.0E-13	2.6E-13	3.2E-13	1.1E-14	0.04
48		**160**	200	1.63	96.0	3.0E-13	2.7E-13			
49		**160**	200	1.63	97.7	3.0E-13	2.7E-13			
50	79	155	200	1.63	99.3	3.1E-13	2.8E-13	2.9E-13	-2.5E-14	-0.09
51		**160**	200	1.63	100.9	3.1E-13	2.8E-13			
52		**170**	200	1.63	102.6	3.2E-13	2.8E-13			
53		**170**	200	1.63	104.2	3.2E-13	2.9E-13			
54		**170**	200	1.63	105.8	3.3E-13	2.9E-13			
55		**180**	200	1.63	107.4	3.3E-13	2.9E-13			
56		**180**	200	1.63	109.1	3.3E-13	3.0E-13			
57	90	184	200	1.63	110.7	3.4E-13	3.0E-13	3.4E-13	2.9E-14	0.10
58		**170**	200	1.63	112.3	3.4E-13	3.1E-13			
59		**170**	200	1.63	114.0	3.5E-13	3.1E-13			
60	84	168	200	1.63	115.6	3.5E-13	3.2E-13	3.1E-13	-8.2E-16	0.00
61	84	168	200	1.63	117.2	3.6E-13	3.2E-13	3.1E-13	-7.5E-16	0.00
62		**160**	200	1.63	118.8	3.6E-13	3.3E-13			
63	80	158	200	1.63	120.5	3.7E-13	3.3E-13	2.9E-13	-2.1E-14	-0.08

Table VII, continued

	Gross	**Net**	**I,**	**ml D_2O**	**Sum**	**Added**	**Lost**	**T present,**	**Excess**	**Fraction**
Day	**c/m**	**d/m**	**ma**	**/day**	**D_2O**	**T, mol**	**T, mol**	**mol**	**T, mol**	**T**
64		**170**	200	1.63	122.1	3.7E-13	3.4E-13			
65		**170**	200	1.63	123.7	3.8E-13	3.4E-13			
66		**170**	200	1.63	125.3	3.8E-13	3.4E-13			
67	103	218	200	1.63	127.0	3.9E-13	3.5E-13	4.1E-13	9.3E-14	0.33
68		**200**	200	1.63	128.6	3.9E-13	3.6E-13			
69	90	184	200	1.63	130.2	4.0E-13	3.6E-13	3.4E-13	3.0E-14	0.11
70		**180**	200	1.63	131.9	4.0E-13	3.7E-13			
71	87	176	200	1.63	133.5	4.1E-13	3.7E-13	3.3E-13	1.6E-14	0.06

Cell #73

	Gross	**Net**	**I,**	**ml D_2O**	**Sum**	**Added**	**Lost**	**T present,**	**Excess**	**Fraction**
Day	**c/m**	**d/m**	**ma**	**/day**	**D_2O**	**T, mol**	**T, mol**	**mol**	**T, mol**	**T**
0		**147**	50	0.00	0.0	0.0E+00	0.0E+00	2.7E-13		0.00
1		**150**	50	0.41	0.4	9.5E-16	7.9E-16			
2	66	121	50	0.41	0.8	4.2E-15	3.3E-15	2.3E-13	-4.9E-14	-0.18
3	77	150	100	0.81	1.6	8.4E-15	7.2E-15	2.8E-13	4.4E-15	0.02
4	85	171	200	1.63	3.3	1.5E-14	1.4E-14	3.2E-13	4.4E-14	0.16
5	100	211	200	1.63	4.9	2.1E-14	2.1E-14	3.9E-13	1.2E-13	0.43
6	110	237	200	1.63	6.5	2.7E-14	3.0E-14	4.4E-13	1.7E-13	0.62
7		**300**	200	1.63	8.1	3.1E-14	3.6E-14			
8		**300**	200	1.63	9.8	3.4E-14	4.3E-14			
9	135	303	200	1.63	11.4	4.0E-14	5.4E-14	5.6E-13	3.0E-13	1.11
10	128	284	200	1.63	13.0	4.7E-14	6.4E-14	5.3E-13	2.7E-13	1.00
11	135	303	300	2.44	15.5	5.5E-14	7.8E-14	5.6E-13	3.1E-13	1.15
12	122	268	300	2.44	17.9	6.3E-14	9.1E-14	5.0E-13	2.5E-13	0.93
13	133	297	300	2.44	20.3	7.1E-14	1.1E-13	5.5E-13	3.1E-13	1.15
14		**300**	300	2.44	22.8	7.6E-14	1.1E-13			
15		**300**	300	2.44	25.2	8.2E-14	1.2E-13			
16	147	334	400	3.26	28.5	9.2E-14	1.4E-13	6.2E-13	4.0E-13	1.46
17	145	329	400	3.26	31.7	1.0E-13	1.6E-13	6.1E-13	4.0E-13	1.46
18	153	350	400	3.26	35.0	1.1E-13	1.8E-13	6.5E-13	4.5E-13	1.64
19	164	379	400	3.26	38.3	1.2E-13	2.0E-13	7.0E-13	5.1E-13	1.88
20	144	326	400	3.26	41.5	1.3E-13	2.2E-13	6.1E-13	4.3E-13	1.56
21		**330**	400	3.26	44.8	1.4E-13	2.4E-13			
22		**330**	400	3.26	48.0	1.5E-13	2.5E-13			
23	147	334	400	3.26	51.3	1.6E-13	2.7E-13	6.2E-13	4.6E-13	1.69
24	167	387	400	3.26	54.5	1.7E-13	2.9E-13	7.2E-13	5.7E-13	2.10
25	165	382	400	3.26	57.8	1.8E-13	3.2E-13	7.1E-13	5.8E-13	2.11
26	151	345	400	3.26	61.0	1.9E-13	3.4E-13	6.4E-13	5.2E-13	1.89
27	124	274	400	3.26	64.3	2.0E-13	3.5E-13	5.1E-13	3.9E-13	1.43
28		**275**	400	3.26	67.6	2.0E-13	3.6E-13			
29		**275**	400	3.26	70.8	2.1E-13	3.7E-13			
30		**275**	400	3.26	74.1	2.2E-13	3.9E-13			
31	123	271	200	1.63	75.7	2.2E-13	4.0E-13	5.0E-13	4.0E-13	1.47
32	130	289	200	1.63	77.3	2.3E-13	4.1E-13	5.4E-13	4.4E-13	1.61
33	118	258	200	1.63	79.0	2.4E-13	4.2E-13	4.8E-13	3.9E-13	1.41
34	116	253	200	1.63	80.6	2.4E-13	4.3E-13	4.7E-13	3.8E-13	1.39
35		**250**	200	1.63	82.2	2.5E-13	4.3E-13			

Table VII, continued

Day	Gross c/m	Net d/m	I, ma	ml D_2O /day	Sum D_2O	Added T, mol	Lost T, mol	T present, mol	Excess T, mol	Fraction T
36		**250**	200	1.63	83.8	2.5E-13	4.4E-13			
37	118	258	200	1.63	85.5	2.6E-13	4.5E-13	4.8E-13	4.0E-13	1.45
38	110	237	200	1.63	87.1	2.6E-13	4.5E-13	4.4E-13	3.6E-13	1.31
39	111	239	200	1.63	88.7	2.7E-13	4.6E-13	4.5E-13	3.7E-13	1.34
40	104	221	200	1.63	90.3	2.8E-13	4.7E-13	4.1E-13	3.3E-13	1.22
41		**230**	200	1.63	92.0	2.8E-13	4.8E-13			
42		**230**	200	1.63	93.6	2.8E-13	4.8E-13			
43		**240**	200	1.63	95.2	2.9E-13	4.9E-13			
44	116	253	200	1.63	96.9	2.9E-13	5.0E-13	4.7E-13	4.0E-13	1.46
45	105	224	200	1.63	98.5	3.0E-13	5.0E-13	4.2E-13	3.5E-13	1.27
46		**230**	200	1.63	100.1	3.0E-13	5.1E-13			
47		**230**	200	1.63	101.7	3.1E-13	5.1E-13			
48	110	237	200	1.63	103.4	3.1E-13	5.2E-13	4.4E-13	3.8E-13	1.38
49		**240**	200	1.63	105.0	3.2E-13	5.3E-13			
50		**240**	200	1.63	106.6	3.2E-13	5.3E-13			
51		**240**	200	1.63	108.3	3.2E-13	5.4E-13			
52		**240**	200	1.63	109.9	3.3E-13	5.4E-13			
53		**240**	200	1.63	111.5	3.3E-13	5.5E-13			
54		**240**	200	1.63	113.1	3.4E-13	5.5E-13			
55	114	247	200	1.63	114.8	3.4E-13	5.6E-13	4.6E-13	4.1E-13	1.49
56		**230**	200	1.63	116.4	3.5E-13	5.7E-13			
57		**230**	200	1.63	118.0	3.5E-13	5.7E-13			
58	105	224	200	1.63	119.6	3.5E-13	5.8E-13	4.2E-13	3.7E-13	1.34
59	108	232	200	1.63	121.3	3.6E-13	5.9E-13	4.3E-13	3.8E-13	1.41
60		**230**	200	1.63	122.9	3.6E-13	5.9E-13			
61	105	224	200	1.63	124.5	3.7E-13	6.0E-13	4.2E-13	3.7E-13	1.36
62		**220**	200	1.63	126.2	3.7E-13	6.1E-13			
63		**220**	200	1.63	127.8	3.8E-13	6.1E-13			
64		**220**	200	1.63	129.4	3.8E-13	6.2E-13			
65	105	224	200	1.63	131.0	3.9E-13	6.2E-13	4.2E-13	3.8E-13	1.38
66		**225**	200	1.63	132.7	3.9E-13	6.3E-13			
67	105	224	200	1.63	134.3	4.0E-13	6.4E-13	4.2E-13	3.8E-13	1.39
68		**225**	200	1.63	135.9	4.0E-13	6.4E-13			
69	95	197	200	1.63	137.6	4.1E-13	6.5E-13	3.7E-13	3.3E-13	1.22

Estimated values are shown in bold type.

Values used in calculation:

(1) 1.55×10^{-17} mol T = 1 d / min,

(2) Fresh electrolyte = 2.3×10^{-15} mol T / ml,

(3) Density D_2O = 1.1 g / ml

(4) T half-life = 12.3 y

(5) Mol D / day lost owing to electrolysis = 86.4 I / 96500, where I is the cell current in ma

Cell #70 shows no sign of excess tritium production and is used to determine the distribution factor. A value of 0.84 produces a minimum in the square of the excess tritium value for data taken over 71 days. Thus, by using this value, the calculated excess tritium value fluctuates around zero with a minimum standard deviation. The distribution factor can also be obtained by comparing the tritium content in the cell to that found in the recombinate. The average gas/liquid ratio based on 15 data sets is 0.91±0.16. Thus, the distribution factor can be calculated using two independent methods that give similar results. A distribution value of 0.84 was used to calculate the behavior of cell #73.

Cell #73 was the most active of the seven active cells. Tritium production started after two days and continued off and on for about 22 days with evidence of bursts as can be seen in Fig. 9. The tritium content of the recombinate was less than the electrolyte and gave a somewhat lower distribution factor (0.82±0.12) than that obtained from cell #70. This difference is within the uncertainty in the measurement. Thus, the excess tritium calculated for this cell is not caused by an unusually small distribution factor compared to the inactive cell.

The decrease in tritium content after the second burst is significant because workers at Texas A & M University[26] have seen the same phenomenon. We believe that part of the tritium was produced on the electrode as DT gas and part exchanged with D in D_2O. The DT gas was produced at such a low rate that most of it was able to dissolve in the electrolyte from the gas bubbles, thereby replacing D_2 in the saturated solution. When tritium production stopped, the dissolved DT gas was gradually flushed out of solution (replaced) by the steady electrolytic production of D_2 gas. This reduction is seen clearly after the 22^{nd} day where the tritium excess shows an exponential drop which approaches a constant value of 1.4. Thus, approximately one third of the tritium contained in the cell on the 22^{nd} day apparently had been present as dissolved DT and about two thirds had exchanged with D in D_2O and remained fixed in the liquid. The other active cells had a different proportion between these two chemical forms.

It is interesting and perhaps significant that these cells all started and stopped producing tritium at approximately the same time. The deposition of surface impurity might have been similar and may account for the similar behavior. This possibility will be explored in the future. No clear pattern could be found in the cell designs that gives an explanation or a significant probability for reproducing the behavior. However, cathodes treated with paraffin appear to have a higher probably of success than those not treated this way.

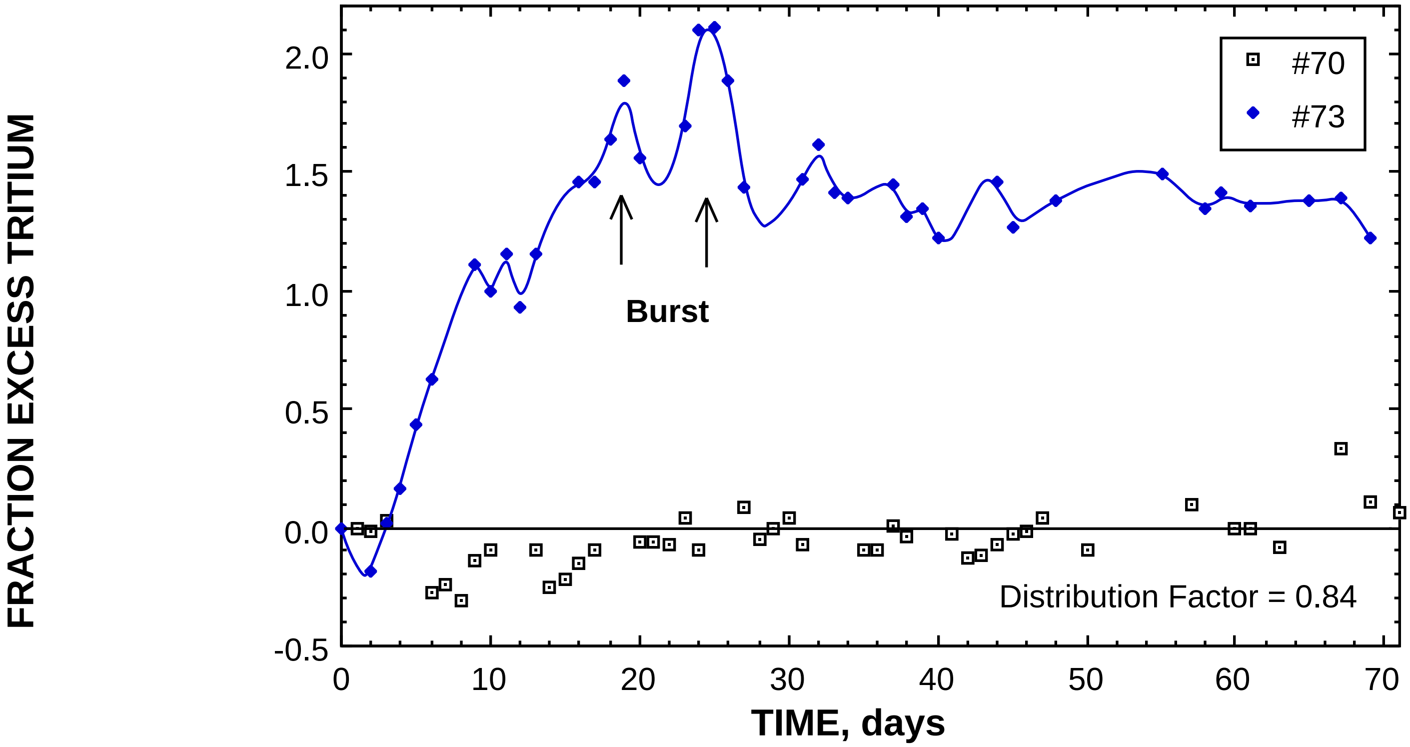

FIGURE 9. Fraction excess tritium in cells #70 and #73 using a distribution factor of 0.84.

The possibility that tritium could result from contamination of the materials used in the cells is not supported because materials from the same chemical lots were used in cells that did not show the presence of tritium. Twenty other cells, running at the time tritium was being produced in the seven active cells, showed no excess tritium. In addition, these cells were completely sealed so that no external tritium could enter.

The possibility that the recorded counts were caused by chemiluminescence owing to active chemical species is not realistic. First, the light spectrum emitted by the scintillator fluid is characteristic of tritium which, because it has an unusually low energy, cannot be confused with other radioactive materials or with chemiluminescence. Second, the count rate becomes independent of time about 20 minutes after the scintillator fluid and the electrolyte are mixed, indicating that chemical effects are short lived in the scintillator fluid used in this work[27]. Counts were recorded only after this effect had died away. Third, the eventual reduction in activity within the cell over a period of time after the second burst (Fig. 9) is only consistent with the source of activity being removed from the cell, not added as would be expected if active chemical species were being produced.

Although the amount of tritium made in this study is small, it is well outside of the uncertainty in the measurement based on a large and consistent data base. The standard deviations for the various data sets are compared in Table VIII. Evidence of tritium production is based on samples having excess tritium between 5 and 785 times the standard deviation based on a total of all random errors. In addition, the pattern of tritium behavior with respect to distribution and exchange is consistent with its known characteristics. Occasional, isolated high values occurred. They were ignored as being possible sampling errors.

It is evident that tritium production in these cells is still very inefficient and probably isolated to a few special locations on the cathode. We can not yet say what special conditions are required for its production.

TABLE VIII

Standard deviation and number of data points of inactive cells compared to excess tritium values

Description of Data Set	Average value (d/min-ml)	Standard Deviation (d/min-ml)	Number of Data Points
Background	19.3*	±2.4*	55
Closed cell	138	±14	446
Open cell, corrected for enrichment	120	±14	80
Open cell, uncorrected for enrichment	140	±31	96

* Values in c/min-ml

Excess Tritium Production
70 - 1.1x10^4 d/min-ml

CONCLUSION

Tritium was produced in 11 cells at levels between 1.5 and 80 times the starting concentration. Over 1500 tritium measurements were made on 53 cells of various designs. As can be seen in Table VIII, the total uncertainty in tritium content for these measurements is ±14 d/min-ml which is 0.1 times the starting concentration. Thus, the proposed tritium excess is well outside the uncertainty in the total measurement.

Fourteen inactive cells are described in this work and are used as reference standards. In addition, a cell containing normal water (0.2N LiOH) has been studied recently and shows no tritium increase after 30 days. The effect of chemiluminescence, counting efficiency, and sampling error have been studied, but are not described in this paper. Based on this background, we believe that the tritium is real, it is not caused by contamination and it is not a product of normal electrolysis.

The distribution factor (gas/liquid) for tritium in the electrolyte is not a constant but appears to be influenced by the cell current density and, perhaps, by the cathode surface characteristics. This work produced values between 0.82 and 1.1.

Tritium production is found to occur after times as short as two days after electrolysis is started and with cathodes having an average D/Pd ratio as low as 0.70. Only about 1 cell in 10 is found to be active.

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[13] The catalytic recombiners are graphite cloth containing teflon on which Pt is deposited. These were purchased from Prototech Company, 70 Jaconnet St., Newton Highlands, MA, 02161 (Phone [617] 965-2720) as 20% Pt on carbon (0.35 mg/cm^2 on woven cloth).

[14] Heavy water (>99.9 % D) was obtained from MSD Isotopes, Montreal, Canada. An analysis showed the following values in ppm: (Ag, Al, As, Au, Be, Bi, Ca, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Fe, Ga, Gd, Ge, Hf, Hg, Ho, Ir, La, Li, Lu, Mg, Mn, Mo, Nd, Ni, Os, Pb, Pd, Pr, Pt, Rb, Re, Rh, Ru, Sb, Sc, Se, Sm, Sn, Sr, Ta, Tb, Te, Th, Ti, Tl, Tm, U, V, W, Yb, Zn, Zr)<1, (Si, B, Na) =3.

[15] The lithium metal contained the following in wt. %: 99.8689% Li (99.982 at. %^{7}Li) , 0.0100% heavy metals, <0.010% Cl, 0.0265% C and 0.0732% N. The following metals were detected in ppm: Ca=84, Fe=20, K=8, Na=77, Si=25

[16] The powder was obtained from Johnson Matthey (Stated as 100% Pd), batch #V7114307. Detected elements in ppm (wt) Ag=60, Al=20, Fe=25, Na=65, Pt=35, Si=35. Undetected elements: As, Au, B, Ba, Be, Bi, Ca, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Ga, Gd, Ge, Hf, Hg, Ho, Ir, La, Li, Lu, Mg, Mn, Mo, Nb, Nd, Ni, Os, Pb, Pr, Rb, Re, Rh, Ru, Sb, Sc, Se, Sm, Sn, Tl, Tm, U, V, W, Yb, Zn, Zr (all <10 ppm).

[17] Tritium was determined by mixing 1 ml of the sample with 10 ml of scintillator fluid in a plastic vial. The light pulses produced by the tritium β-scintillator fluid interaction were counted using a Packard Tri-Carb Liquid Scintillation Spectrometer, Model #3255. Standards obtained from NBS were run with the samples and used to calibrate the machine.

[18] The decomposition rate is calculated by subtracting 20, the average background, from the sample counting rate and dividing by 0.38, the counting efficiency. Conversion to other units can be done using 100 d/min = $4.50x10^{-5}$ µCi = 1.67 Bq.

[19] Supplied by J. Marshal, Marshal Laboratories, 5854 Rawhide Court, Suite B Boulder, Colorado 80302. Detected elements in ppm(wt) Ag=20, Al=35, Au=115, Ca=10, Fe=95, Pt=140, Si=40, Zn=25. Undetected element: As, B, Ba, Be, Bi, Cd, Ce, Co, Cr, Cs, Cu, Dy, Er, Eu, Ga, Gd, Ge, Hf, Hg, Ho, Ir, La, Li, Lu, Mg, Mn, Mo, Nb, Nd, Ni, Os, Pb, Pr, Rb, Re, Rh, Ru, Sb, Sc, Se, Sm, Sn, Tl, Tm, U, V, W, Yb, Zr (all <10 ppm).

[20] The light spectrum is different when luminescence is caused by beta emission compared to chemical effects.

[21] Very old (>2000y) well water is available at Los Alamos and was used because of its very low tritium content.

[22] The power supplies were Hewlett-Packard 6038A.

[23] Supplied by Professor Martin, Texas A&M University.

[24] Purchased from Englehard Company.

[25] One ml of electrolyte, to replace that taken during sampling, and D_2O, to replace that lost owing to electrolysis, were added on a daily basis, generally before the tritium sample was taken. Because the cell contained 120 ml of electrolyte and the electrolysis rate was generally low, these periodic additions of fluid had only a small perturbation on the tritium concentration.

[26] John Bockris, private communication (1989).

[27] 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 1with 0.2 N LiOD.