

TABLE I: SUMMARY OF ELECTROLYSIS EXPERIMENTS WITH NEUTRON AND TRITIUM GENERATION

Sr. No.	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11
Division	DD/ HWD NtPD	NtPD/ HWD	NtPD/ HWD	DD/ HWD NtPD	HWD/ NtPD	AnCD	ROMG	ROMG	ApCD	ApCD	IGCAR
Cell (Name)	Ti-SS	MR-1	MR-2	5 Module	Par. Plate	PDC-I	RCS-11	RCS-19	Nafion-1	Nafion-2	RCP-II
Date	1989 21 May	1989 21 April	1989 12-16 June	1989 5 May	1990 15 March	1989 21 April	1989 June - Aug.	1990 Jan. - Apr.	1989 July	1990 Feb.	1989 Dec.
Cathode:											
Material	Ti	Pd-Ag	Pd-Ag	Pd-Ag	Pd	Pd	Pd	Pd	Pd	Pd	Pd
Geometry	Rod	Tubes	Tubes	Discs (5)	Plate	Hollow Cyl.	Cube	Pellet	Ring	Coil	Button
Dimensions (mm)	22 $\Phi$ $\times$ 150 long	3 o.d. $\times$ 200 ht	3 o.d. $\times$ 200 ht	115 $\Phi$ $\times$ 0.1 thk	40 $\times$ 50 $\times$ 1 thk	-	1cm <sup>3</sup>	11 $\Phi$ $\times$ 11.2 ht.	25 $\Phi$ $\times$ 10 ht $\times$ 1 thk	1 $\Phi$ $\times$ 140 long	-
Area (cm <sup>2</sup> )	104	300	300	78	20	5.9	6	5.7	18	4.4	8
Anode	S.S. Pipe	Ni-Pipes	Ni-Pipes	Porous-Ni	Ti Plate	Pt Mesh	Pt Mesh	Pt Mesh	Pt Mesh	Pt Mesh	Pt Mesh
Electrolyte	NaOD (5M)	NaOD (5M)	NaOD (5M)	NaOD (5M)	NaOD (5M)	LiOD (0.1 M)	LiOD (0.1 M)	LiOD (0.1 M)	LiOD (0.1 M)	Li <sub>2</sub> SO <sub>4</sub> (.05 M)	LiOD (0.1 M)
Volume (ml)	135	250	250	1000	300	45	150	120	250	140	-
Current Density (mA/cm <sup>2</sup> )	$\leq$ 400	$\sim$ 300	$\sim$ 300	$\sim$ 800	$\sim$ 200	$\leq$ 340	$\sim$ 100	$\sim$ 700	$\sim$ 60	$\sim$ 50	$<$ 100
Switching On:											
Charge (A-hrs/cm <sup>2</sup> )	1.2	0.6	-	3.2	0.8	3.0	2.5	650	34	0.15	36.7
Time (hrs)	3	5	0.5	4	4	9	24	930	330	3	300
Active Life	Few hrs	$\sim$ 3.5 hrs	$\sim$ 2hrs	$\leq$ 3 mine	$<$ 1 min	$\sim$ 5 hrs	$\sim$ 5d	$\sim$ 100 sec	$\sim$ 40 hrs.	$\sim$ 5 d	8 hrs
Neutron Yield:											
No. of Bursts	Continuum	9	1	1	1	3	17	1	Many	Many	2
Total n/ cm <sup>2</sup>	$3 \times 10^7$ $2.9 \times 10^5$	$4 \times 10^7$ $1.7 \times 10^5$	$.9 \times 10^7$ $1.3 \times 10^4$	$5 \times 10^6$ $1.3 \times 10^4$	$1 \times 10^6$ $5 \times 10^4$	$3 \times 10^6$ $5 \times 10^5$	$1.4 \times 10^6$ $2.3 \times 10^5$	$3 \times 10^6$ $5.2 \times 10^5$	$1.8 \times 10^8$ $10^7$	$5.8 \times 10^6$ $1.3 \times 10^6$	$2.4 \times 10^6$ $3 \times 10^5$
Tritium Yield:											
Total (Bq)	$2.6 \times 10^5$	$1.5 \times 10^7$	$3.8 \times 10^6$	$7 \times 10^6$	-	$1.42 \times 10^5$	$1.3 \times 10^3$	$7.7 \times 10^3$	325	32.5	$6.3 \times 10^3$
Total (Atom)	$1.4 \times 10^{14}$	$8 \times 10^{15}$	$1.9 \times 10^{15}$	$4 \times 10^{15}$	-	$7.2 \times 10^{13}$	$6.7 \times 10^{11}$	$4 \times 10^{12}$	$1.8 \times 10^{11}$	$1.8 \times 10^{10}$	$3.5 \times 10^{12}$
t/cm <sup>2</sup>	$1.3 \times 10^{12}$	$2.7 \times 10^{13}$	$6 \times 10^{12}$	$10^{13}$	-	$1.2 \times 10^{13}$	$1.1 \times 10^{11}$	$5.2 \times 10^{13}$	$1 \times 10^{10}$	$4 \times 10^9$	$4.4 \times 10^{11}$
(n/t) Ratio	$2 \times 10^{-7}$	$0.5 \times 10^{-8}$	$0.5 \times 10^{-8}$	$1.2 \times 10^{-9}$	-	$4 \times 10^{-8}$	$1.7 \times 10^{-6}$	$10^{-6}$	$10^{-3}$	$3.2 \times 10^{-4}$	$7 \times 10^{-7}$

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TABLE II: SUMMARY OF OTHER TRITIUM PRODUCING ELECTROLYTIC EXPERIMENTS

Sr. No.	#1	#2	#3	#4	#5	#6	#7	#8	#9	#10	#11
Division Cell (Name)	Heavy Water Division		ROMG	Analytical Chemistry Division					Chemistry Division		
Cell (Name)	MR (Jr)-I	MR (Jr)-II	RCS-18	PDX-0	PDC-II	PDC-III	PDC-IV	PDR-1	CD-4	CD-6	CD-5
Date	1989 21 Sept	1990 5 March	1989 24 Oct.	1989 24 April	1989 10 July	1989 6 Sept.	1989 29 Sept.	1989 9 Nov.	1989 21 July	1989 22 Dec.	1989 24 Oct.
Cathode:											
Material	Pd-Ag Alloy	Pd-Ag Alloy	Cold Rolled Pd	Pd	Pd	Pd	Pd	Pd	Pd ingot	Pd	Pd wire
Geometry	Tubes	Tubes	Hollow Cyl.	Ring	Hollow Cylinder	Hollow Cylinder	Hollow Cylinder	Rod	Cylinder	Pellet	Grid
Dimensions (mm)	3 $\Phi$ $\times$ 150 ht	3 $\Phi$ $\times$ 150 ht	-	2 thk.	-	-	-	4 $\Phi$ $\times$ 19 long	8 $\Phi$ $\times$ 16 long	4 $\Phi$ $\times$ 4 ht.	0.5 $\Phi$ $\times$ 800 long
Area (cm <sup>2</sup> )	113	113	19	14.5	6.37	6.37	6.37	2.75	0.57	0.126	4
Anode	Ni Pipe	Ni Pipe	Pt gauze	Pt Discs	Pt Mesh	Pt Mesh	Pt Mesh	Pt Mesh	Pt Coil	Pt Wire	Pt Wire
Electrolyte	NaOD (5M)	NaOD (5M)	LiOD (0.1 M)	LiOD (0.14M)	LiOD (0.1 M)	LiOD (0.1 M)	LiOD (0.14M)	LiOD (0.1 M)	LiOD (0.1 M)	LiOD (0.1 M)	KOD (Cone) (Paste)
Volume (ml)	150	150	150	65	60	100	80	80	28	3	1.5
Open/ Closed	Open	Open	Open	Closed	Open	Open	Closed	Closed	Open	Semiopen	Closed
Current (A)	40	40	$\sim$ 2	$<$ 2	1 $\sim$ 2 Pulsed	1 $\sim$ 3 Pulsed	1 $\sim$ 2 Pulsed	0.2 $\sim$ 2.2 RF Superposed	100mA	$\leq$ 35mA	30mA
Current Density mA/cm <sup>2</sup>	350	350	$\sim$ 105	$\sim$ 160	$<$ 350	$\leq$ 470	$<$ 350	$<$ 800	$\sim$ 100	$\sim$ 278	7.5
Duration of Electrolysis	12 hrs	30 hrs	13d	7.4 d	366.2 hrs	183.3 hrs	5.8 d	40d	190 d	17 d	80d
Initial Conc. (Bq)	1.44	3.33	3.6	2.7	2.81	2.77	2.70	2.68	4.6	2.0	2.5
Maximum Concn. (Bq/ml)	225.7	18.5	-	$0.93 \times 10^4$	$5.88 \times 10^4$	4.6	-	-	72.1	65.0	22.9
Output to Input Ratio	156.7	5.6	3.36	3425	20,925	1.66	2.5	1.91	15.7	32.5	9.16
(Bq) (Atoms)	$3.3 \times 10^4$ $1.76 \times 10^{13}$	$2.28 \times 10^3$ $1.2 \times 10^{12}$	$2.71 \times 10^3$ $1.44 \times 10^{12}$	$6.02 \times 10^5$ $3.2 \times 10^{14}$	$2.08 \times 10^6$ $1.1 \times 10^{15}$	$2.96 \times 10^3$ $1.56 \times 10^{12}$	$6.29 \times 10^2$ $3.96 \times 10^{11}$	$1.1 \times 10^3$ $5.83 \times 10^{11}$	$10^{12}$	$10^{11}$	$2 \times 10^{10}$
t/cm <sup>2</sup>	$1.6 \times 10^{11}$	$1.1 \times 10^{10}$	$0.8 \times 10^{11}$	$2.2 \times 10^{13}$	$1.7 \times 10^{14}$	$2.4 \times 10^{11}$	$6.2 \times 10^{10}$	$2.12 \times 10^{11}$	$1.8 \times 10^{12}$	$0.8 \times 10^{12}$	$0.5 \times 10^{10}$

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40 hour neutron active phase was  $1.8 \times 10^8$  neutrons. It is interesting to note however that even during an intense phase of neutron production the count rate suddenly dropped to near background levels and remained so for several seconds before abruptly climbing back to levels over 100 times the background value.

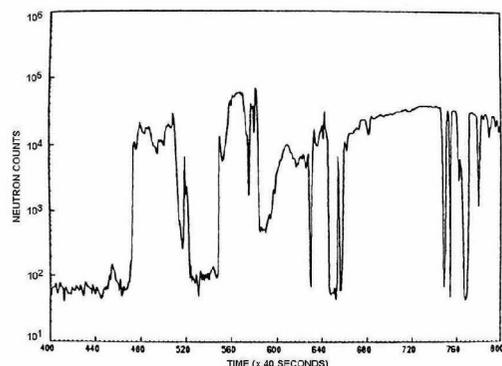


Fig. 9. Expanded View of Portion of Fig. 8

The electrolyte was sampled once in 6 days during the experiment and analyzed for tritium content using standard liquid scintillation techniques. After the neutron active phase, the tritium level of the electrolyte showed an increase from 0.4 to 1.3 Bq/ml. The cell electrolyte volume being 250 ml, this corresponds to an excess tritium generation of  $1.8 \times 10^{11}$  atoms. This does not include the tritium carried away by the gas stream. Degassing of the Pd cathode at 680 K and reformation of water over hot CuO turnings yielded an additional  $3 \times 10^9$  tritium atoms only. Thus an upper limit to the overall neutron-to-tritium yield ratio in this experiment works out to  $\sim 10^{-3}$ .

The electrolysis experiment with the Pd coil cathode was carried out for 24 days. The electrolyte was 0.05M  $\text{Li}_2\text{SO}_4$ . Neutron emission started within 4 hours of commencement of electrolysis and lasted for a total period of 15 hrs spread over the first five days of the electrolysis. The integrated neutron yield was  $5.8 \times 10^6$  while the tritium yield at the end of the experiment amounted to  $1.8 \times 10^{10}$  atoms. The neutron-to-tritium yield ratio in this experiment works out to be  $3.2 \times 10^{-4}$ .

It is significant that the neutron to tritium yield ratios of  $10^{-3}$  to  $10^{-4}$  observed in both these experiments are several orders of magnitude larger than the values of  $10^{-6}$  to  $10^{-8}$  obtained in all the previous cells. One possible explanation could be that the employment of Nafion membrane in conjunction with the very low levels of dissolved oxygen in these cells might have been responsible for preventing recombination of T back into DTO and instead have allowed most of the T to escape along with the gas stream. As noted earlier the tritium carried away by the gas stream was not measured in this experiment also.

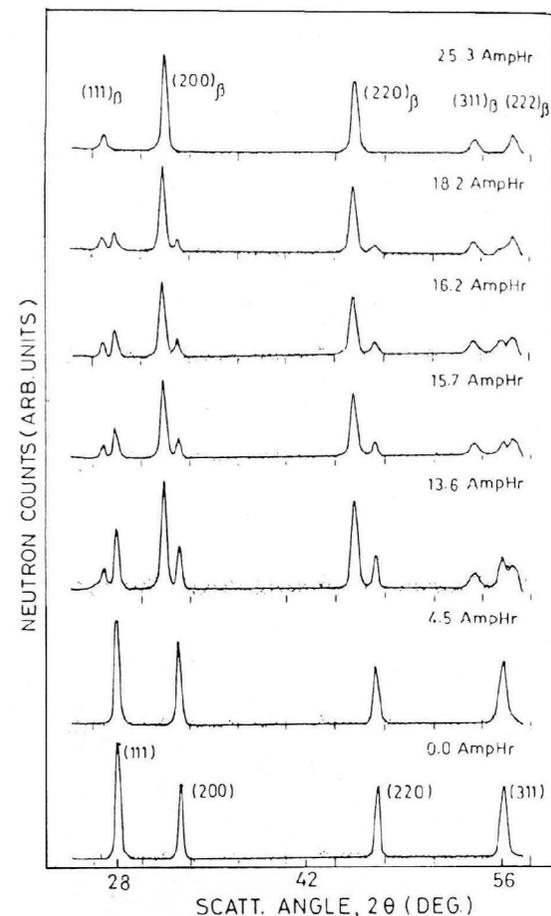


Fig. 10. Neutron Diffraction Patterns at Different Deuterium Loadings of Palladium Rod

Fig. 10 displays the recorded diffraction patterns at various loading stages measured in terms of amp-hrs of electrolysis. It took an hour's reactor time for each spectrum. The Pd electrode was initially in pure fcc (metallic) form with the lattice constant of  $3.89 \text{ \AA}$  characterized by the four peaks corresponding to the  $\alpha$  phase seen in the bottom most pattern of Fig. 10. New peaks indicative of the precipitation of the  $\beta$  phase of the Pd-D system (lattice constant of  $4.02 \text{ \AA}$ ) showed up at around 10 amp-hrs of electrolysis. As electrolysis proceeded the intensity of the  $\beta$  phase peaks built up at the cost of  $\alpha$  phase peaks. It was found that within about 25 amp-hrs the upper part of the Pd electrode projecting underneath the cell was completely converted to  $\beta$  phase wherein the lower part required nearly 60 amp-hrs for this. From the ratio of the structure factors of the odd to even reflections ( $S(111)/S(200)$ ) the stoichiometry in the  $\beta$  phase could be

### 3.3 CYLINDRICAL Pd PELLETT EXPERIMENT (ROMG) /7/

Here the cathode was a cylindrical Pd pellet 11 mm dia  $\times$  11.2 mm height and anode a Pt gauze as before. The concentration of the LiOD electrolyte (120 ml) was increased progressively from 0.1M to  $\sim$ 3M and accordingly the applied voltage decreased, with the current being maintained constant at  $\sim$ 4A. The neutron detection set up was the same as in the previous ROMG experiment. On 13<sup>th</sup> Feb 1990 when  $\sim$ 3400 amp-hrs had been passed, there was a sharp burst of  $3 \times 10^6$  neutrons lasting approximately for about 100 s (see Fig. 7).

A sample of the electrolyte which was taken the day after this burst, showed a clear eight fold increase in tritium level (64.5 Bq/ml vs preburst value of 7.9 Bq/ml). The tritium level thereafter continuously decreased as shown in Fig. 7. But a significant observation was that the rate of decrease was not commensurate with dilution effects caused by make up D<sub>2</sub>O addition. The dotted lines commencing from each experimental point in the figure indicates how one would have expected the tritium level to fall if only dilution was playing a role. This implies that additional tritium is continuously entering the electrolyte for many days after the sharp neutron burst. If this is attributed to diffusion of tritium from the inner regions of the pellet, it would support the theory that tritium (and neutron) generation is not restricted to the surface of the cathode alone.

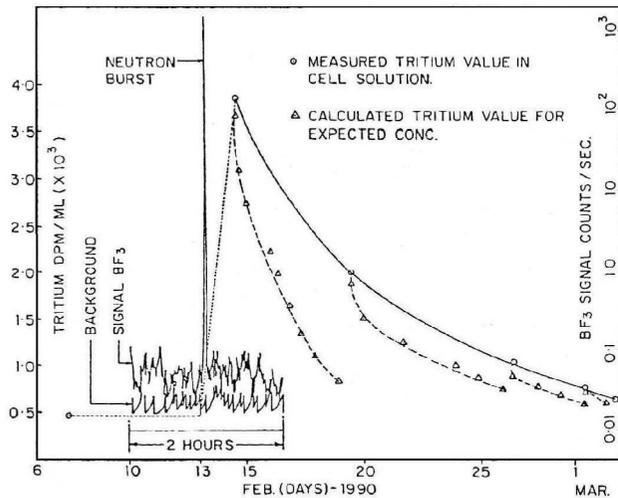


Fig. 7. Neutron and Tritium Output of RCS-11 Experiment

After the experiment was terminated following several weeks of electrolysis, the Pd electrode which was found to have developed many cracks on the surface, was degassed at 900°C, and from the volume of D<sub>2</sub> liberated the D/Pd ratio at saturation was deduced to be  $\sim$ 0.85.

TABLE III

TRITIUM PRODUCTION IN D, GAS LOADED Pd SAMPLES

Experiment No	#1	#2	#3	#4	#5
Nature of sample	Pd black powder	Pd-Ag foil	Pd-Ag foil	Pd-Ag foil	Pd-Ag foil
Mass (g)	20	0.96	10.9	10.8	0.43
Date of loading	20 June 89	24 Aug 89	19 Sept 89	7 Mar 90	19 Sept 89
Volume of D <sub>2</sub> absorbed (ml)	1325	34.5	518.4	222	20.2
(D/Pd) ratio	.63	.46	.45	.20	.45
Equilibration time (hrs)	16	16	240	40	240
Water used for extraction (ml)	50	6	50	50	5
Tritium activity of water (bq/ml)	8.1	5.9	8.5	12.5	32.6
T/D ratio in Pd	$32 \times 10^{-11}$	$1.1 \times 10^{-11}$	$.87 \times 10^{-11}$	$3.4 \times 10^{-11}$	$8.3 \times 10^{-11}$
Absolute tritium activity (bq)	410.7	370	429.2	717.8	159.1
Total tritium atoms in Pd	$2.31 \times 10^{11}$	$2.02 \times 10^{10}$	$24 \times 10^{11}$	$4.1 \times 10^{11}$	$8.96 \times 10^{10}$
Tritium atoms per g of Pd	$1.2 \times 10^{10}$	$21 \times 10^{10}$	$2.2 \times 10^{10}$	$3.8 \times 10^{10}$	$20.8 \times 10^{10}$

The presence of tritium in the Pd-Ag foils has also been independently confirmed through autoradiography. Fig 11 shows the radiograph of a triangular Pd-Ag foil. The image displays some non-uniformity in fogging. It was however observed that the intensity of fogging of these Pd-Ag foils rapidly decreased when attempts were made to reproduce the radiographs on subsequent days, indicating that the tritium retention capability of Pd-Ag is not as good as that of titanium.

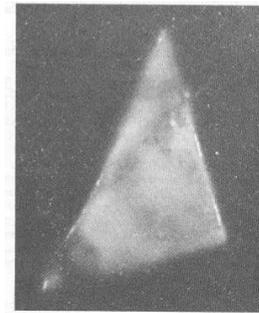


Fig. 11. Autoradiograph of Deuterated Triangular Pd-Ag Foil

### 7.2 FRASCATI TYPE EXPERIMENTS WITH Ti SHAVINGS (CD) /5/

A readily available set up employed earlier for high pressure hydriding studies was used for these experiments/12/. Ti metal pieces cut from a sheet were surface cleaned and subjected to activation treatment prior to loading in the high pressure cell. D<sub>2</sub> gas pressure or temperature was cycled between high and low values using liquid nitrogen. The well type neutron counter employing 24 He<sup>3</sup> detectors embedded in paraffin, along with the associated data acquisition

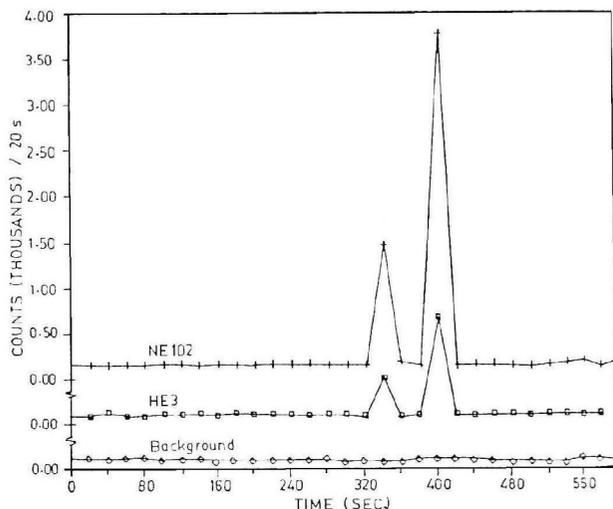


Fig. 5. Neutron Burst of Pd-Ti Parallel Plate Cell (15<sup>th</sup> March 1990)

The Pd cathode was immediately taken for X-ray counting to a low energy NaI detector assembly. Later it was kept overnight in contact with a medical X-ray film for autoradiography. (These techniques are discussed in detail in Ref/5/). However these did not give any evidence of the presence of radioactivity. Samples of the electrolyte taken immediately after the experiment also did not show any significant increase in tritium activity which is indeed very puzzling to the authors. It is possible that the tritium generated had fully escaped along with the electrolytic gases.

### 3. Pd CATHODE CELLS WITH LiOD ELECTROLYTE

#### 3.1 Hollow Pd Cylinder Experiment (AnCD) /5/

In this quartz cell the cathode was a hollow Pd cylinder of 1.7 g mass, having a wet surface area of 5.9 cm<sup>2</sup>; the anode was a Pt gauze; the electrolyte was 0.1 M LiOD in D<sub>2</sub>O (99.87% isotopic purity). To begin with a current of 1 A was used for the electrolysis. After about 30 hours when the temperature attained 60°C, current pulsing between 1 and 2 A at 1 second intervals was adopted. In the absence of a direct neutron detector, this group looked for neutron emission through the 1186 keV window of the gadolinium capture gamma ray peak. The detector was a 3 × 3 NaI crystal mounted behind a gadolinium compound coated converter plate. After a charge of 17.5 amp-hrs had been passed, the first neutron emission was detected on 21<sup>st</sup> April 1989. As seen in Fig. 6, in all three distinct neutron bursts of 14 to 20 minutes duration each were produced amounting to an integrated yield of 3 × 10<sup>6</sup> neutrons. Subsequent analysis of a sample of the electrolyte indicated that a total of 3.85 μCi or 7.3 × 10<sup>13</sup> atoms of tritium had been generated in this experiment. This corresponds to a neutron-to-tritium yield ratio of 4 × 10<sup>-8</sup>.

images are not produced due to mechanical scratching of the film by the sharp edges of the shavings. The aluminium foil converts the tritium betas into soft X-rays which emerge from the foil giving rise to image formation. (See Sec. 8.3).

#### 7.3 EVIDENCE FOR TRITIUM IN Ti TARGETS SUBJECTED TO RF HEATING IN D<sub>2</sub> ATMOSPHERE (TPPED/NtPD) /5/

In these experiments machined and chemically cleaned targets of Ti were individually heated to temperatures of up to 900°C in a glass chamber using a surrounding induction heating coil (1 to 2 MHz frequency, 3 to 6 kW power). The glass chamber was connected to a vacuum system as well as H<sub>2</sub> and D<sub>2</sub> gas bottles. Degassing was initially carried out at 900°C for several hours until a vacuum of 10<sup>-5</sup> mm was maintained steadily. The targets were later heated to 600°C in H<sub>2</sub> atmosphere at a few mm of pressure. The induction heater was then switched off and the target allowed to cool absorbing H<sub>2</sub> in the process. At least three cycles of H<sub>2</sub> absorption/desorption was given "to create active sites for D<sub>2</sub> absorption". Three such heating/cooling cycles were then carried out with D<sub>2</sub> gas. The pressure drop recorded by an oil manometer indicated the quantity of gas absorbed during each cooling cycle. It was observed that the quantity of gas absorbed increased each time saturating in the 3<sup>rd</sup> or 4<sup>th</sup> cycle.

The targets typically absorbed <10<sup>19</sup> molecules of D<sub>2</sub>. Since the mass of Ti was a few hundred milligrams, this corresponds to a gross (D/Ti) ratio of hardly 0.001. However we have reason to believe that most of the absorption would be confined to the surface region. This is because when a metallic object is heated by induction heating the current distribution falls off exponentially with increasing depth. The skin depth  $\delta$  characterizing this phenomenon is given by  $\delta = \sqrt{s/f\pi\mu}$  where  $s$  and  $\mu$  are the resistivity and permeability respectively of the workload and  $f$  is the frequency of the applied electromagnetic field. For a frequency of 1 to 2 MHz,  $\delta$  for Ti is 0.1 mm. Consequently we believe that the (D/Ti) ratio in the near surface region would be much higher than the gross value of 0.001 noted earlier.

After loading, all the targets were subjected to various tests such as autoradiography, K X-ray counting etc in search of tritium. Although several dozen targets were successfully loaded with D<sub>2</sub> gas, only a few of them gave positive evidence for the presence of tritium. Interestingly the samples which soaked up large amounts of D<sub>2</sub> gas did not give any positive results. The best results were obtained from a disc shaped button (10 mm dia × 2 mm thick) and a couple of conical pieces meant for use as electrodes. Table IV summarizes the results. Figs. 13 and 14 are the autoradiographs of a deuterated disc and cone respectively. The radiographs of the Ti disc shows about 50 to 60 spots randomly distributed within the boundary. The occurrence of spots all along the rim of the machined disc is very intriguing. It is estimated that each spot corresponds roughly to 10<sup>9</sup> to 10<sup>10</sup> atoms of tritium. The total number of tritium atoms in the whole target works out to be ~10<sup>11</sup>. This is to be compared with the 10<sup>19</sup> to 10<sup>20</sup> atoms of D absorbed in all by the Ti, pointing to a gross (T/D) ratio of >10<sup>-9</sup>.

80 A corresponding to current densities of  $\sim 1\text{A}/\text{cm}^2$ . Operating electrolyte temperatures close to  $100^\circ\text{C}$  are possible as the unit can withstand internal pressures of a few atmospheres.

Two neutron detectors, namely a  $\text{BF}_3$  bank and a fast neutron recoil detector, were mounted close to the cell to monitor the neutron output. The system was filled with freshly prepared 20%  $\text{NaOD}$  in  $\text{D}_2\text{O}$  on 5<sup>th</sup> May 1989 and electrolysis commenced at a current of 60 to 65 amps (applied voltage was 12.5 V). When the cell had operated for about four hours a big burst of neutrons overlapping two consecutive counting intervals was recorded in both channels. Knowing the neutron detection efficiencies it is estimated that  $\sim 5 \times 10^6$  neutrons were generated during that burst. It was found that the tritium level had jumped by a factor of  $\sim 3500$  from an initial value of 0.055 nCi/ml (2Bq/ml) to a final post burst value of 190.3 nCi/ml (7 kBq/ml). Considering that the total inventory of electrolyte in the system was  $\sim 1$  liter, this corresponds to an overall excess tritium production of  $\sim 190 \mu\text{Ci}$  or  $4 \times 10^{15}$  atoms. It must however be emphasized that this does not include the tritium carried away by the electrolytic gas stream which was allowed to escape. Thus an upper bound to the neutron-to-tritium yield ratio in this experiment is  $1.2 \times 10^{-9}$ .

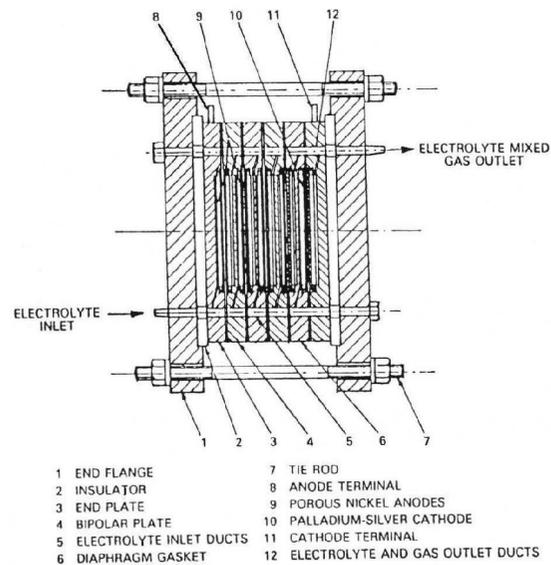


Fig. 4. Schematic Sectional View of Five Module Cell With Disc Electrodes

## 2.4 EXPERIENCE WITH A Ti-SS CELL /5/

A Ti-SS cell was quickly fabricated in April 89 using readily available components in order to obtain some experience with use of Ti as cathode material. A 22 mm dia  $\times$  150 mm long rod (surface area =  $104 \text{ cm}^2$ ) of Ti with a flange at the bottom served as cathode. An SS pipe of 40 mm ID served as anode leaving an annular inter electrode gap of  $\sim 9$  mm. PTFE gaskets at the

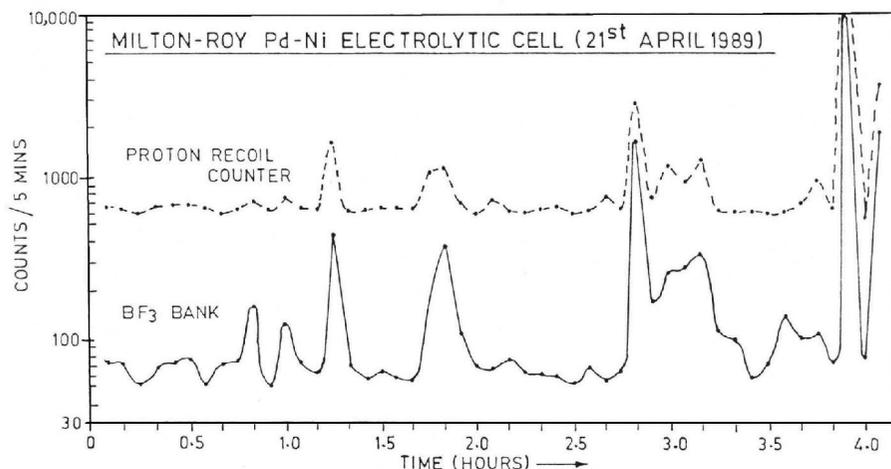


Fig. 14. Autoradiograph of Deuterated Ti Cone

## 7.4 ANOMALOUS TRITIUM PRODUCTION IN TITANIUM ELECTRODE OF A PLASMA FOCUS DEVICE (NtPD) /15/

A Plasma Focus (PF) device forms a high density ( $10^{25}$  ions/ $\text{m}^3$ ), high temperature ( $10^7$  K) plasma which produces an intense burst of neutrons when operated with deuterium gas/16/. This device has a vacuum chamber consisting of coaxial cylindrical electrodes connected through a Corning glass insulator at the bottom end and left open at the top. Fig. 15 shows a schematic diagram of a plasma focus device of the type used in the present experiments. When this coaxial gun is connected to a high voltage (15 to 50 kV) capacitor bank with the help of a spark gap switch, a surface discharge is initiated at the insulator end. This then develops into a radial current sheath which is accelerated by  $\mathbf{J} \times \mathbf{B}$  forces down the length of the electrode system, sweeping the gas ahead of it. On reaching the open end, the current sheath turns around on itself forming a quasi-cylindrical implosion resulting in a dense hot "plasma focus" a few cm long and few mm in diameter just above the tip of the central anode.

cell started showing counts well above background values. At this time the current was raised further and this resulted in a number of distinct neutron peaks appearing in both neutron detector channels (See Fig. 2). The experiment was terminated when the cell current increased on its own to over 120 amps at the time of the last peak, resulting in the power supply getting damaged. The total number of neutrons generated during the four hour duration of this run is estimated to have been  $\sim 4 \times 10^7$ .



**Fig. 2. Neutron Counts Variation During Run No. 1 of Milton Roy Cell (21<sup>st</sup> April 1989)**

A sample of the electrolyte drawn after the run, indicated a tritium content of  $\sim 1.5 \mu\text{Ci/ml}$  ( $55.5 \text{ kBq/ml}$ ), an increase by a factor of  $\sim 20,000$  in comparison to the tritium content of the initial stock heavy water. Taking into account the total volume of the electrolyte (250 ml) as well as the amount of make up  $\text{D}_2\text{O}$  added, it is estimated that an excess of  $\sim 8 \times 10^{15}$  atoms of tritium were generated in this run. The corresponding neutron-to-tritium yield ratio works out to be  $0.5 \times 10^{-8}$ . This was the first indication to us that the neutron-to-tritium yield ratio in cold fusion experiments is anomalously low.

#### Run No. 2 (12<sup>th</sup> to 16<sup>th</sup> June 1989)

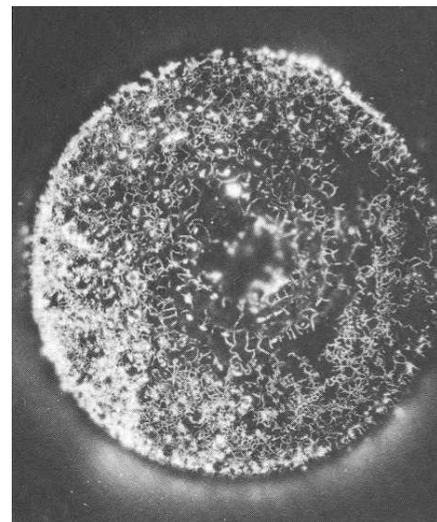
A second series of electrolysis runs was carried out with this cell in June 1989 after it was drained and thoroughly flushed with  $\text{D}_2\text{O}$  several times. Prior to this a drain tap with a valve had been welded to the bottom of the cell to enable periodic withdrawal of electrolyte samples. Fresh electrolyte solution prepared with stock  $\text{D}_2\text{O}$  was charged and left in the cell over a weekend. A sample of this electrolyte taken on the following Monday morning gave a high tritium level of  $\sim 0.32 \text{ nCi/ml}$  ( $11.8 \text{ Bq/ml}$ ), presumably due to leach out of tritium left over in the cathodes from the large burst of 21<sup>st</sup> April.

Electrolysis commenced on 12<sup>th</sup> June at a current level of  $\sim 60$  amps. The  $\text{BF}_3$  neutron detector bank monitored the cell while the  $\text{He}^3$  bank served as background monitor. Except for a few small neutron bursts which were observed within about half an hour of commencement of electrolysis, no neutrons were recorded for the next couple of days, although electrolysis

exposures. There has been no change in the resolution or clarity of the images even after a gap of a few months. The very sharp worm like lines are believed to be due to  $\beta$  from tritium bearing grain boundary regions near the surface, while the intense diffuse spots are due to soft X-rays (Ti K X-rays) emanating from deeper layers of the titanium electrode. As in the case of the RF heated Ti disc autoradiographs (Fig. 13) the presence of spots all along the periphery of the rod is noteworthy. In order to rule out the possibility of image formation due to tritium contamination in the  $\text{D}_2$  gas, the other electrodes (Al, SS and brass) were also tested for radioactivity. But none of them showed any activity confirming that the phenomenon is unique to Ti.

During all the 80 PF shots it is estimated that almost  $10^9$  (d-d) neutrons could have been generated due to hot fusion reactions. Since in conventional (d-d) reactions (both beam target and hot fusion) the neutron-to-tritium branching ratio is  $\sim 1$ , it follows that not more than  $10^9$  tritium atoms could therefore have been produced during the shots. It is totally unrealistic to expect or postulate that all this tritium would have succeeded in getting absorbed on the Ti anode surface. Even if that were true, it still can not explain the  $10^{16}$  atoms of tritium measured on the tip of the Ti anode. But the even more intriguing question is why was it not seen in the radiographs taken on the same night of the experiment as well as on the following night? Although the presence of large amounts of tritium was first detected only five weeks later, it is possible that it might have been produced any time in the intervening period.

The authors suggest that the intense electric and magnetic fields involved in the operation of a PF must have had some role to play in causing "cold fusion" reactions on the tip of the anode. Repeat experiments using planchets of Ti mounted at the top of a brass anode have however not shown any activity so far. A fresh stock of pure  $\text{D}_2$  gas as well as a new Ti electrode are awaited for repeating the experiments under identical conditions prevalent in the earlier successful experiment.



**Fig. 16. Autoradiograph of Central Ti Electrode of PF Device (5 Weeks After Charging)**

processes in the upgrading of heavy water, fusion plasma experiments and neutron and tritium measurements devised and set up a variety of electrolytic cells during the early days of April 1989. In a centre such as BARC which has nurtured the development of the heavy water moderated line of reactors in India for over three decades, equipment and expertise for the measurement of neutrons and tritium was readily available. In the initial experiments the emphasis was naturally on the detection of nuclear particles rather than “excess heat” which required intricate calorimetry. The first positive evidence for the emission of neutrons and tritium was obtained on 21<sup>st</sup> April 1989 and since then several different cells have confirmed these results.

Meanwhile reports from Frascati of the detection of neutrons from pressurized D<sub>2</sub> gas loaded Ti shavings /2/ opened up a second channel of cold fusion investigations. Neutrons or tritium have since been measured in a variety of D<sub>2</sub> gas loaded Ti and Pd targets at Trombay. A brief summary of the early BARC work /3/ was presented at Karlsruhe in July 1989. Report BARC-1500 issued in December 1989 is a compendium of twenty papers, documenting in an informal style the status of ongoing work, and covers “BARC Studies in Cold Fusion” over the period April to September 1989 /4/. The experimental papers of this compilation are also being published in Fusion Technology /5/. The present paper is an overview of all the experimental work done at Trombay during the first year of the ‘cold fusion era’ including new results obtained since the publication of BARC-1500 and summarizes the efforts of about a dozen independent groups comprising over 60 scientists and engineers drawn from different divisions of BARC.

## PART A: ELECTROLYTIC STUDIES

### 2. EXPERIMENTS WITH HIGH CURRENT NaOD ELECTROLYZERS (HWD/NtPD/DD)

#### 2.1 NEUTRON MONITORING

Three different neutron detectors were available for monitoring the neutron yield in these experiments. The first was a bank of three BF<sub>3</sub> counters (each of 25 mm dia × 450 mm length) sensitive mainly to slow neutrons, embedded in a paraffin moderator block. The second was a similar bank of three paraffin encased thermal neutron detectors except that they were of the He<sup>3</sup> type. The third neutron detector was an 80 mm dia × 80 mm high proton recoil type plastic scintillator (NE 102A) sensitive both to fast neutrons as well as high energy gammas. During the electrolysis experiments usually one of the thermal neutron banks and the fast neutron detector were mounted close to the cell while the other thermal neutron bank was located about 1.5 m away to serve as background monitor. The neutron detection efficiency was determined with the help of a calibrated Pu-Be neutron source placed at the cell location and was typically in the region of 0.05% to ~1.5%. In later experiments a personal computer became available to display on line the count rate variations.

chemiluminescence decay curve of one such “difficult” sample which did not cool down even after several hours of dark adaptation and decay time.

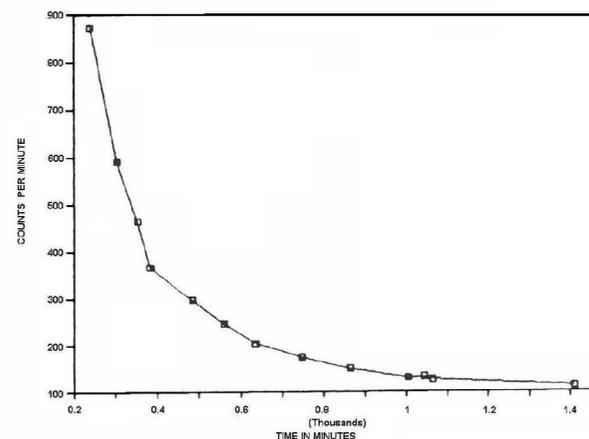


Fig. 17. Chemiluminescence Decay Curve of a Severe Electrolyte Sample

Chemical and photon quenching effects were encountered in other samples resulting in compression of the tritium spectrum. In order to confirm that the spectral output of the electrolytic samples falls well within the tritium spectral region, an experimental sample which did not show any chemiluminescence or quenching effects was spiked with a tritium standard and the pulse height spectrum was checked and compared with that of the test sample. In the case of LiOD based electrolytes a systematic study has been carried out to study the influence of alkalinity on the measurements.

The Pollution Monitoring Section of BARC has recently developed a flow detector for on line measurement of tritium levels in gas and liquid phases /17/. The flow cell has a volume of 15ml and is packed with 5 μm thick plastic scintillating fibres. A pair of photomultiplier tubes viewing from the sides and operating in coincidence measure the scintillation rates, due to tritium.

#### 8.2 β-particle and K X-ray Counting of Deuterated Ti and Pd Targets

The presence of tritium in the near surface region of deuterated Ti and Pd targets has been established through the technique of β particle and Ti K X-ray counting. The β were counted using either a proportional gas counter or a plastic scintillator. In some of the strong sources pico amp levels of ionization current could be measured using an ionization chamber. The range of the tritium β (energy < 18.6 keV) in metallic samples is < 1mg/cm<sup>2</sup>. Some of the β excite the characteristic K X-rays of Ti (~4.5 keV) whose mean free path for absorption in Ti is however 10mg/cm<sup>2</sup>. Thus these X-rays are able to escape from deeper layers of the Ti than the β. Using a high resolution germanium detector or a 1mm thick NaI detector these soft X-rays can be conveniently counted.

(f) It is also worth noting that one amp-hr or 3600 coulombs corresponds roughly to the charge carried by the deuterons required to load a few grams of Pd (associated with each  $\text{cm}^2$  of cathode surface) to a (D/Pd) ratio of  $\sim 0.6$ . In practice since only a fraction (10 ~ 30%) of the deuterons bombarding the cathode actually get absorbed in it, the experimentally observed switching on charge of  $\leq 3$  amp-hr/ $\text{cm}^2$  is consistent with the common sense expectation that a (D/Pd) ratio of at least 0.6 should be achieved before nuclear processes involving deuterons can be expected to take place.

(g) Except for the Ti-SS cell (Cell #1 of Table I) in all the other ten cells neutrons are produced in one or more large bursts of magnitude several times the background values. But in all cases after a limited period of nuclear activity the cells become inactive, no matter for how long the electrolysis is continued.

(h) In spite of the wide disparity in cell designs, it is observed that the specific neutron yield i.e. the integrated neutron yield per unit area of cathode surface, lies in the range of  $10^4$  to  $10^5$  n/ $\text{cm}^2$  except for the two experiments where a Nafion membrane was present between the cathode and anode (Cells # 9 & 10). The specific neutron yield in these two cases is 1 or 2 orders of magnitude higher.

(i) The specific tritium yield (see Tables I & II) has shown a greater overall variation ranging from  $4 \times 10^9$  to  $1.7 \times 10^{14}$  t/ $\text{cm}^2$ . In 10 of 22 cells it is in the range of  $10^{12}$  to  $10^{14}$  t/ $\text{cm}^2$  while most of the other cells have given values in the region of  $10^{10}$  to  $10^{12}$  t/ $\text{cm}^2$ .

(j) The BARC teams generally operated only one cell at a time. The overall "success rate" defined as the percentage of cells which produced tritium or neutrons in relation to the total number of cells operated is estimated to be more than 70%. The groups who used NaOD as electrolyte had perhaps an even higher success rate.

(k) The BARC experiments possibly include the largest sized electrolytic cold fusion cells (measured in terms of either cathode surface area ( $300 \text{ cm}^2$ ) or total current (100 amps)) to have been employed so far.

(l) Unfortunately it has not been possible to conclusively establish whether the neutron and tritium producing reactions occur only on the surface of the electrode or over the whole volume of the cathode. But the delayed appearance of additional tritium in the electrolyte, at times even when the cell was off, indicates that tritium slowly leaches out from the inner regions of the electrode, giving some credence to the volume effect theory.

The gas phase experiments of BARC have spanned a variety of novel approaches and diagnostic techniques. The observation that RF heating of titanium targets in  $\text{D}_2$  atmosphere promotes tritium production is interesting. The large amount of tritium ( $10^{16}$  atoms) found on the top end surface of the central titanium electrode of a plasma focus device, particularly the very impressive high resolution autoradiograph, is puzzling indeed. Finally the unexpectedly large levels of tritium in decades old deuterated titanium targets adds to the pool of puzzling results.

On the whole however, the results obtained by a number of independent experimental groups at BARC during the first year of the 'cold fusion era' have provided ample evidence of the occurrence of anomalous nuclear processes in Pd and Ti lattices loaded with deuterium.