Iyengar, P.K. and M. Srinivasan. *Overview of BARC Studies in Cold Fusion*. in *The First Annual Conference on Cold Fusion*. 1990. University of Utah Research Park, Salt Lake City, Utah: National Cold Fusion Institute.

OVERVIEW OF BARC STUDIES IN COLD FUSION

P. K. Iyengar
Atomic Energy Commission CSM Marg, Bombay 400 039, India
and
M. Srinivasan
Neutron Physics Division
Bhabha Atomic Research Centre
Trombay, Bombay 400 085, India

ABSTRACT

A wide variety of experiments have been carried out by twelve independent teams employing both electrolytic and gas phase loading of deuterium in Pd and Ti metals to investigate the phenomenon of cold fusion first reported by Fleischmann and Pons in March 1989. The experiments were primarily devoted to the study of the emission of nuclear particles such as neutrons and tritium with a view to verify the "nuclear origin" of cold fusion. In 22 different electrolytic experiments whose cathode surface areas ranged from 0.1 to 300 cm^2, large bursts of neutrons and/or tritium were measured. Some of these gave clear evidence that these two nuclear particles were being generated simultaneously. The neutron-to-tritium yield ratios in the majority of these experiments was in the range of 10^{-6} to 10^{-9}. The specific neutron and tritium yields expressed per cm^2 of cathode surface area also fitted into a systematic pattern. A unique feature of the BARC electrolysis results is that the first bursts of neutrons and tritium occurred (in 8 out of 11 cells) on the very first day of commencement of electrolysis, when hardly a few amp-hrs of charge had been passed.

In gas phase studies copious neutron emission was observed in a Frascati type absorption/desorption mode experiment with Ti shavings. Presence of tritium in D_2 gas loaded Pd and Ti samples was established through the technique of autoradiography as well as Ti K X-ray counting. In the case of Ti, it was noted that RF heating of samples, in lieu of resistance furnace heating somehow promotes tritium formation. Most recently it was found that ≈ 1016 atoms of tritium had been "produced" on the top end surface of the central Ti electrode of a deuterium filled Plasma Focus device after it was subjected to ≈ 80 charge/discharges shots. All in all the BARC studies have unambiguously confirmed the anomalous production of neutrons and tritium in deuterium loaded Pd and Ti lattices.

1. INTRODUCTION

The announcement in March 1989 by Fleischmann and Pons /1/ of the occurrence of (d-d) fusion reactions (or possibly some other unknown nuclear processes) in Pd metal cathodes electrolytically loaded with deuterium, followed by reports of the observation of "2.45 MeV fusion neutrons" independently by Jones et al during the electrolysis of D_2O, resulted in a frenzy of activity the world over to reproduce these measurements. At Trombay several groups having expertise in various areas such as hydriding of metals, electrochemistry, isotope exchange

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 21st April 1989 and since then several different cells have confirmed these results.

Meanwhile reports from Frascati of the detection of neutrons from pressurized D_2 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_2 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_3 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^3 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.

2.2 MILTON ROY COMMERCIAL ELECTROLYZER /5/

A diffusion type water electrolyzer using Pd-Ag alloy tubes as cathodes designed to generate ultra pure (oxygen free) hydrogen gas had been procured from the Milton Roy company of Ireland, sometime in 1988 for the purpose of generating D_2 gas for use in Plasma Focus experiments. Thus it so happened that when news of the cold fusion phenomenon reached Trombay in March 1989, this cold fusion cell was all set to be switched on with D_2O as electrolyte. 5M NaOD in D_2O was selected as the electrolyte based on the recommendation of the suppliers of the Milton Roy cell. A schematic view of this cell is shown in Fig. 1. The outer nickel body along with a central Ni pipe serve as coaxial anodes. The cathode comprises of 16 numbers of specially activated Pd-Ag alloy membrane tubes having a total surface area of 300 cm^2. These tubes are sealed at the top and open at the bottom into a plenum through which the D_2 (or H_2) gas is drawn.

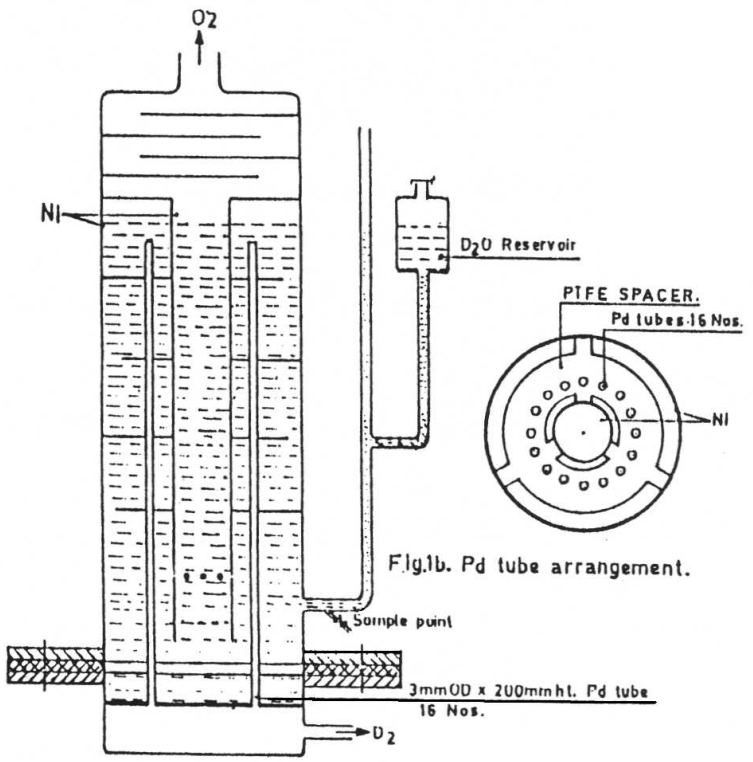

Fig. 1. Schematic View of Milton-Roy Commercial Electrolyzer

D_2O of >99.75% isotopic purity, containing ~0.075 nCi/ml (or ~2.8 Bq/ml) of tritium and moisture free Na were employed for preparing the NaOD electrolyte. The electrolyzer can be operated up to a current level of 100 amps corresponding to a current density of ~330 mA/cm^2, although for continuous operations only 60 amps is recommended in order to avoid overheating.

Run No. 1 (21st April 1989)

The cell was initially operated for about 48 hours with 20% NaOH in ordinary water. It was later flushed with D_2O and filled with 20% NaOD solution in D_2O prior to commencement of electrolysis on 21st April 1989. Following brief operation at 30 amp, the current was slowly raised to 60 amps. After about 3 hours at this current level both the neutron detectors viewing the

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 ~4×10^7.

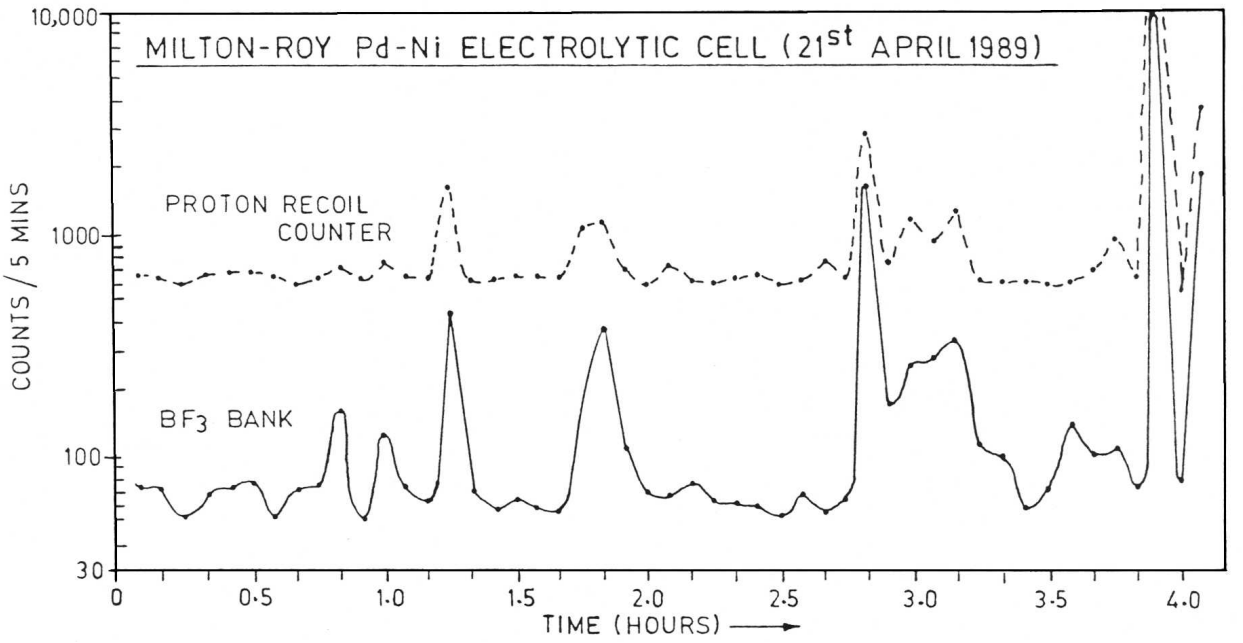

Fig. 2. Neutron Counts Variation During Run No. 1 of Milton Roy Cell (21st April 1989)

A sample of the electrolyte drawn after the run, indicated a tritium content of ~1.5 μCi/ml (55.5 kBq/ml), an increase by a factor of ~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 D_2O added, it is estimated that an excess of ~8×10^{15} atoms of tritium were generated in this run. The corresponding neutron-to-tritium yield ratio works out to be 0.5×10^{-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 (12th to 16th 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 D_2O 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 D_2O 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 ~0.32 nCi/ml (11.8Bq/ml), presumably due to leach out of tritium left over in the cathodes from the large burst of 21st April.

Electrolysis commenced on 12th June at a current level of ~60 amps. The BF_3 neutron detector bank monitored the cell while the 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

continued until 17.45 hrs on Wednesday 14th June when the cell was put off. A couple of hours later there was another small neutron burst lasting ~15 minutes. (These small bursts are shown plotted in Fig. 1 of the companion paper from BARC /6/.) But on the evening of Friday 16th June, there was a large neutron burst (> 10^6 neutrons) lasting for a couple of hours, (see Fig. 3)

The week long experiment was terminated at this point but the electrolyte was left in the cell and the D_2 gas plenum closed leaving the gas at an excess pressure of ~1kg/cm^2 above atmospheric pressure. Samples of electrolyte were drawn every day during the week of the experiment and sent for tritium analysis. The sample drawn on 23rd June indicated a high tritium level of 121 nCi/ml (4.5 kBq/ml). After a lapse of about a month the tritium level in the electrolyte was found to have further increased to a value of ~460 nCi/ml (17 kBq/ml), a four fold increase since the termination of the experiment. Fig. 3 shows the variation of tritium concentration during the entire course of Run No. 2. It may be noted that after the large neutron burst the tritium level has shown a thousand fold jump suggesting that tritium is produced at the same time as the neutrons.

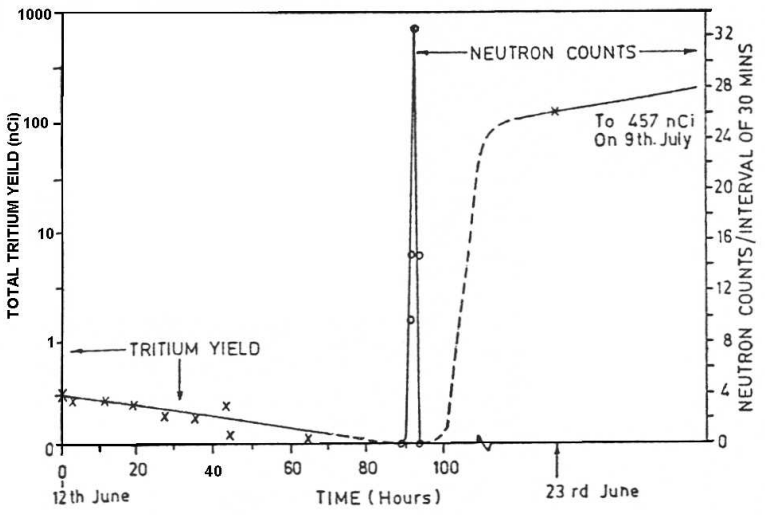

Fig. 3. Neutron and Tritium Output During Run No. 2 of Milton-Roy Cell

The integrated neutron yield during this experiment was ~0.9×10^7 neutrons while the total number of tritium atoms generated was 1.9×10^{15}. The corresponding neutron-to-tritium yield ratio is 0.5×10^{-8}. This is in remarkably good agreement with the results of the earlier Milton Roy run, although the absolute neutron and tritium yields are lower by a factor of ~5. Thus the Pd-Ag cathode appears to have partly lost its capability to support "cold fusion reactions" after the first run.

2.3 FIVE MODULE CELL WITH DISC ELECTRODES /5/

This modular five unit cell represents our early attempts at obtaining experience with an electrolyzer design which can be scaled up to higher capacities if required. Five cathode discs each of 78 cm^2 area and 1 cm^3 volume are fabricated out of Johnson & Matthey palladium (75%)-silver (25%)-alloy sheets (0.12 mm thickness); the anode plates are made of porous nickel. The individual modules of the cell are clamped together and connected in series. Fig. 4 gives a schematic sectional view of the cell. The cell is capable of operating at currents of up to

80 A corresponding to current densities of ~1A/cm^2. Operating electrolyte temperatures close to 100ºC are possible as the unit can withstand internal pressures of a few atmospheres.

Two neutron detectors, namely a 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% NaOD in D_2O on 5th 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 ~5 × 10^6 neutrons were generated during that burst. It was found that the tritium level had jumped by a factor of ~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 ~1 liter, this corresponds to an overall excess tritium production of ~190 μCi or 4 × 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 × 10^{-9}.

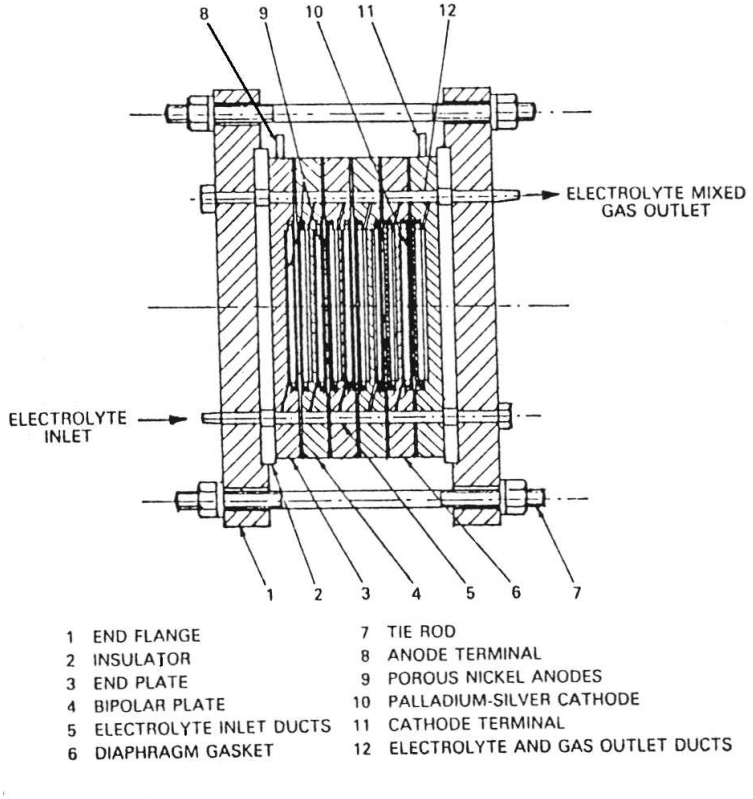

Fig. 4. Schematic Sectional View of Five Module Cell With Disc Electrodes

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 × 150 mm long rod (surface area = 104 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 ~9 mm. PTFE gaskets at the

bottom ensured coaxial alignment, as well as leak tightness. A vent at the top permitted free escape of electrolytic gases. A second cell of identical design was also fabricated for use as a control cell with H_2O.

5M NaOD in D_2O was used as electrolyte. The current density was ~400 mA/cm^2. The main problem with this cell was the continuous deposition of a dull black coating of iron on the cathode which impaired operation. The electrolytic solution also developed a pale greenish yellow colour. The electrode surface had therefore to be cleaned frequently and fresh electrolyte charged, interrupting electrolysis. On the whole it was a messy operation.

The neutron yield during this experiment was monitored by the bank of 3 He^3 counters embedded in paraffin. The count rate was initially about 240 counts/10s, comparable to the count rates observed during an initial H_2O electrolysis run. After about 3 hours of D_2O electrolysis the count rate increased slowly to ~590 counts/10s. Since no big neutron bursts as in the Pd cathode cells were observed it was suspected that this gradual increase in counts could have been due to amplifier drifts, etc. On switching off the cell current it was noted that the count rate came down to ~385 counts/10s, but it did not quite reach the earlier background levels. When the cell was switched on again however the count rate attained levels of about ~590 counts/s once again. Thereafter operation of the cell was terminated and it was confirmed that the count rate decreased to the original background levels when the cell was removed from the vicinity of the neutron detector. Throughout this experiment the counts of the plastic scintillator channel monitoring the background did not show any significant variation. In all ~ 3×10^7 neutrons were generated during this experiment.

A sample of the electrolyte sent for analysis at the end of the experiment indicated tritium activity of ~48 nCi/ml (1.78 kBq/ml), a three order of magnitude increase over the initial stock solution value of ~0.05 nCi/ml (1.9 Bq/ml). The net excess tritium produced after correcting for tritium input through make up D_2O addition etc, works out to ~7 μCi or ~1.4×10^{14} atoms of T. Admittedly this was not a very clean experiment, but even so one can obtain a very rough value for the neutron-to-tritium yield ratio as 2×10^{-7} for this experiment.

2.5 Pd-Ti PARALLEL PLATE CELL

A simple parallel plate cell with Teflon button spacers was fabricated with Pd (0.5 mm thick) and Ti (1 mm thick) plates (40×50 mm^2) as electrodes. The inter-electrode gap was ~2 mm A thin platinum strip was spot welded at the top of the Pd to serve as current feed through, The parallel plate assembly was suspended inside a 300 ml glass bulb having a wide mouth at the top. A vent hole in the stopper permitted escape of electrolytic gases. An advantage of this cell was that either Pd or Ti could be selected as cathode with the other serving as anode.

Electrolysis was commenced on 15th March 1990 with Pd as cathode and 5M NaOD in D_2O as electrolyte. Current density was adjusted to be ~200 mA/cm^2. Three neutron detectors were available for monitoring neutron output, two for viewing the cell and the third for serving as background monitor. Two consecutive neutron bursts occurred about 4 hours after commencement of electrolysis. The background counts were absolutely flat during this run (see Fig. 5). It was noted that the Pd cathode had buckled outwards and had become extremely hardened. The buckling can be explained on the basis of differential loading of D_2 across the thickness of the metal.

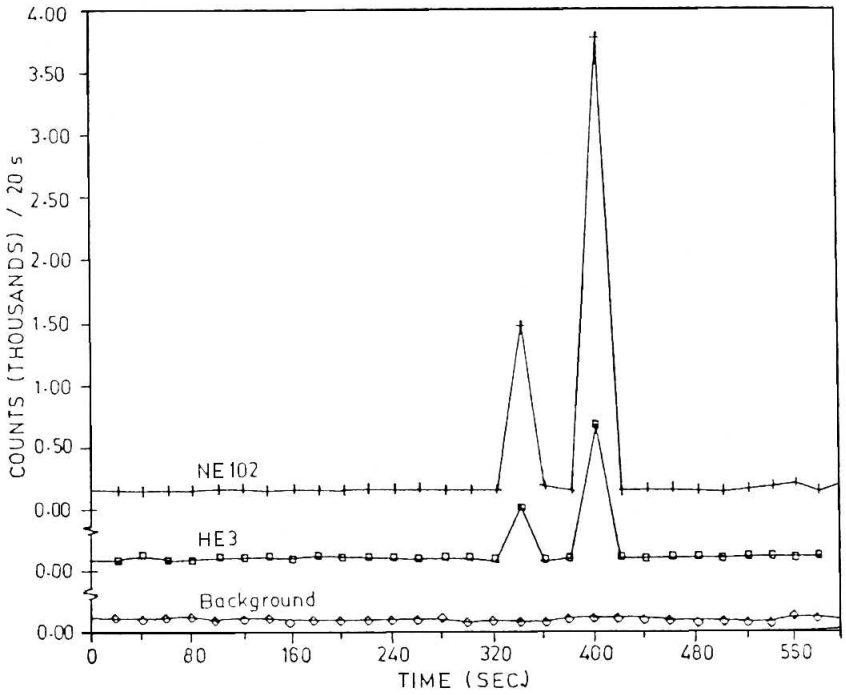

Fig. 5. Neutron Burst of Pd-Ti Parallel Plate Cell (15th March 1990)

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^2; the anode was a Pt gauze; the electrolyte was 0.1 M LiOD in D_2O (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 60ºC, 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 gadolonium capture gamma ray peak. The detector was a 3 × 3 NaI crystal mounted behind a gadolonium compound coated converter plate. After a charge of 17.5 amp-hrs had been passed, the first neutron emission was detected on 21st 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 3×10^6 neutrons. Subsequent analysis of a sample of the electrolyte indicated that a total of 3.85 μCi or 7.3×10^{13} atoms of tritium had been generated in this experiment. This corresponds to a neutron-to-tritium yield ratio of 4×10^{-8}.

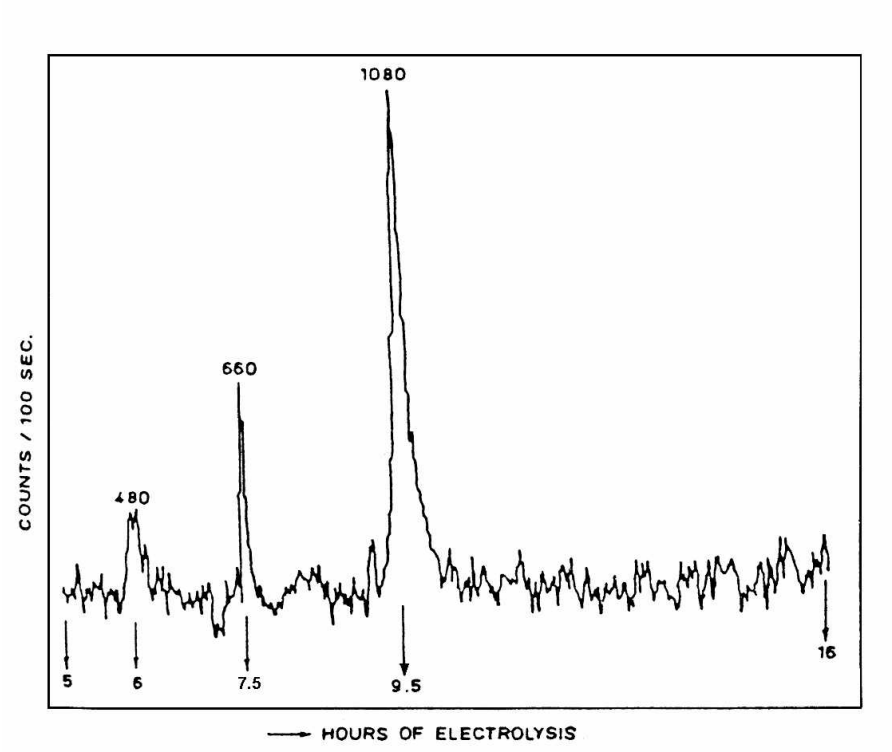

Fig. 6. Neutron Yield PDC-I Cell: 1186 keV Capture gamma Counts

3.2 Pd CUBE EXPERIMENT (ROMG) /5/

In this experiment the cathode was a 1 cm^3 of Pd and anode a Pt wire gauze formed into a cylinder surrounding the cube. The electrolyte was 0.1 M LiOD in D_2O. The electrolytic gases were recombined using a Pd catalyst; excess D_2 was converted into D_2O using hot copper oxide. Although the main objective of these experiments was measurement of excess heat, in this paper only the neutron and tritium results are reported.

The neutron yield was monitored by means of a paraffin encased BF_3 counter mounted immediately underneath the table where the cell was located. A second BF_3 detector placed about a metre away monitored background neutrons. The cell electrolyte as well as the D_2O collected in the various cold traps in the system were periodically sampled for tritium measurements. Electrolysis was carried out at a current of ~0.6 amps. After about 24 hrs when ~14.7 amp-hrs of charge had been passed, bursts of neutrons began to be observed. In all 17 neutron bursts lasting from 2 mins to 55 mins each were recorded. The neutron yield in the bursts varied from 5×10^3 (2 min burst) to 5×10^5 (8 min burst). Altogether a total of 1.4×10^6 neutrons was estimated to have been generated. Thereafter there were no more neutron bursts although the electrolysis continued for a further period of seven weeks (a total of 1365 amp-hrs).

A detailed accounting of tritium distributed in various constituents such as electrolyte, vapour condensate recovered from recombined gases, gases extracted from Pd electrode etc was carried out by this group. It was concluded that in all about 35 nCi or 6.7×10^{11} atoms of excess tritium was produced in this experiment. This corresponds to a gross neutron-to-tritium yield ratio of 1.7×10^{-6}.

3.3 CYLINDRICAL Pd PELLET EXPERIMENT (ROMG) /7/

Here the cathode was a cylindrical Pd pellet 11 mm dia × 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 ~3M and accordingly the applied voltage decreased, with the current being maintained constant at ~4A. The neutron detection set up was the same as in the previous ROMG experiment. On 13th Feb 1990 when ~3400 amp-hrs had been passed, there was a sharp burst of 3×10^6 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_2O 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.

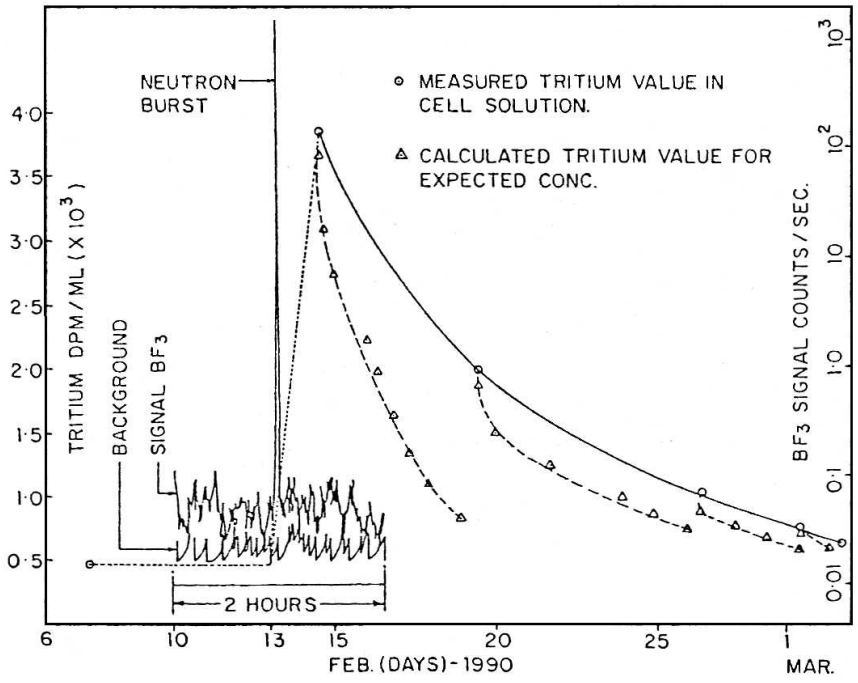

Fig. 7. Neutron and Tritium Output of RCS-11 Experiment

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 900ºC, and from the volume of D_2 liberated the D/Pd ratio at saturation was deduced to be ~0.85.

3.4 Pd RING AND Pd COIL CELLS WITH NAFION MEMBRANE (ApCD)

In both these experiments the cathodes were thoroughly degassed and vacuum annealed ($<10^{-3}$ mm Hg, 1070 K, ~10 hours) prior to electrolysis. The anode was a cylindrical Pt mesh covering the cathode on all sides. In the first experiment the Pd ring cathode (2.5 cm dia, 1 cm height and 0.1 cm thickness) was charged from both the sides /5/. In the second experiment a thin Pd rod (1 mm dia × 14 cm length) formed into a coil was employed as cathode /8/. In both these experiments the anodes were loosely sandwiched between pairs of Nafion membranes so as to prevent the oxygen evolved at the anode to diffuse back to the cathode surface. The electrolyte (0.1M LiOD in D_2O of 99.86% isotopic purity) was circulated through the quartz electrolytic cell to reduce the dissolved oxygen level further. A saturated calomel electrode dipping in the electrolyte was used to monitor the cathode potential. The cell was operated at relatively low current density of ~60 mA/cm^2.

Neutron detection was carried out by means of a well type counter containing 24 He_3 detectors embedded in an annular block of paraffin. The test cell was located at the centre of this well, giving a neutron detection efficiency of 8.6%. Data acquisition was carried out with the help of a personal computer having multi-scaling mode facility. The counting time per channel was set as 40s. It was ensured that the overall neutron detection system was immune to extraneous influences which could give false counts. The background reference counts were observed to be steady at ~1.6 cps for about 10 days before start of the experiments.

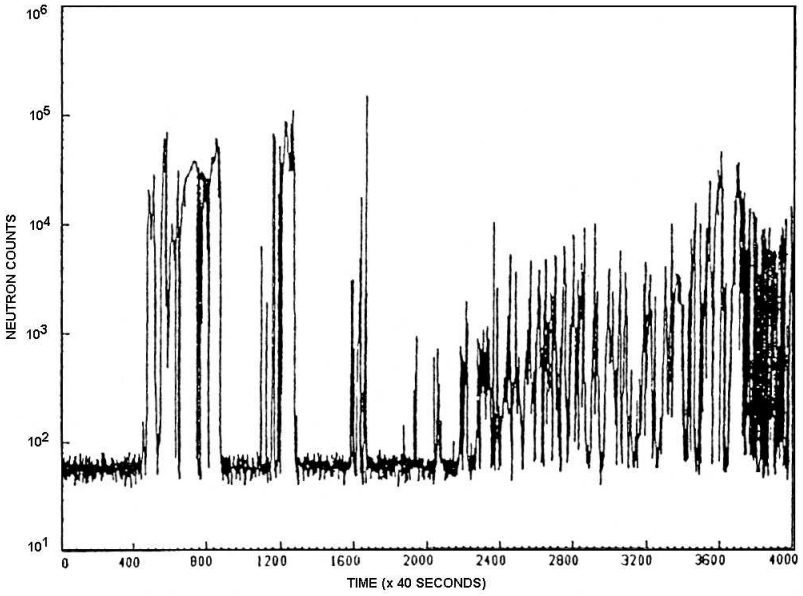

Fig. 8. Neutron Bursts of Pd Ring Cathode Experiment of ApCD

The ring cathode electrolysis experiment was run for 32 days commencing from 6th of July 1989. Between the 14th and 17th days from start of electrolysis copious emission of neutrons in the form of bursts was recorded. Otherwise the count rate remained close to background levels for the rest of the period. Fig. 8 depicts the neutron counts variation over the entire 44 hour (4000 × 40s) duration, while Fig. 9 gives an expanded view of a part of the same data. The log scale of the counts axis should be noted, indicating that the bursts were indeed intense, the peak neutron emission rate being ~1000 times background levels. The total neutron emission over the

40 hour neutron active phase was 1.8×10^8 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.

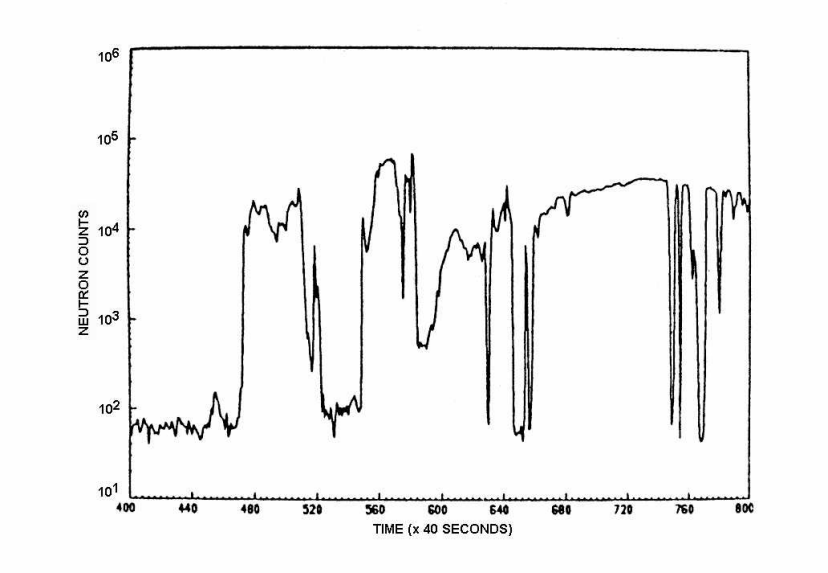

Fig. 9. Expanded View of Portion of Fig. 8

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 1.8×10^{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 3×10^9 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 Li_2SO_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 5.8×10^6 while the tritium yield at the end of the experiment amounted to 1.8×10^{10} atoms. The neutron-to-tritium yield ratio in this experiment works out to be 3.2×10^{-4}.

It is significant that the neutron to tritium yield ratios of 10^{-3} to 10^{-4} observed in both these experiments axe 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.

4. SUMMARY OF ELECTROLYSIS EXPERIMENTS WITH NEUTRON AND TRITIUM GENERATION

Table I presents a summary of the successful electrolysis experiments conducted so far at Trombay wherein significant amounts of both neutron and tritium production has been observed. Also included (last column of Table I) for comparison and completeness are the results of an experiment /9/ conducted at the Indira Gandhi Centre for Atomic Research (IGCAR) in Kalpakkam, Tamil Nadu, a sister Institution of BARC. The main conclusions to emerge out of these results are discussed later.

5. OTHER TRITIUM PRODUCING CELLS / EXPERIMENTS

Besides the above electrolysis experiments wherein both tritium and neutron production has been observed, there have been an additional 11 cells / experiments wherein clear evidence for excess tritium generation has been obtained. The majority of these experiments were carried out in the various divisions belonging to the "Chemical Group" of BARC. Table II summarises these results. In most of these experiments neutron yield, if any, was not monitored due to non-availability of detectors with the groups concerned. In the few cases where neutron detectors were present the increase in count rates if any was not significant enough within the statistics of the background count rate variations. Some of the cells of Table II were of closed type wherein the electrolytic gases were recombined by means of a suitable catalyst.

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 Φ × 150 long	3 o.d. × 200 ht	3 o.d. × 200 ht	115 Φ × 0.1 thk	40 × 50 × 1 thk	–	$1cm^3$	11 Φ × 11.2 ht.	25 Φ × 10 ht × 1 thk	1 Φ × 140 long	–
Area (cm^2)	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_2SO_4 (.05 M)	LiOD (0.1 M)
Volume (ml)	135	250	250	1000	300	45	150	120	250	140	–
Current Density (mA/cm^2)	≤400	~300	~300	~800	~200	≤340	~100	~700	~60	~50	<100
Switching On:											
Charge ($A\text{-}hrs/cm^2$)	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	~3.5 hrs	~2hrs	≤3 mine	<1 min	~5 hrs	~5d	~100 sec	~40 hrs.	~5 d	8 hrs
Neutron Yield:											
No. of Bursts	Continuum	9	1	1	1	3	17	1	Many	Many	2
Total	3×10^7	4×10^7	$.9\times10^7$	5×10^6	1×10^6	3×10^6	1.4×10^6	3×10^6	1.8×10^8	5.8×10^6	2.4×10^6
n/ cm^2	2.9×10^5	1.7×10^5	1.3×10^4	1.3×10^4	5×10^4	5×10^5	2.3×10^5	5.2×10^5	10^7	1.3×10^6	3×10^5
Tritium Yield:											
Total (Bq)	2.6×10^5	1.5×10^7	3.8×10^6	7×10^6	–	1.42×10^5	1.3×10^3	7.7×10^3	325	32.5	6.3×10^3
Total (Atom)	1.4×10^{14}	8×10^{15}	1.9×10^{15}	4×10^{15}	–	7.2×10^{13}	6.7×10^{11}	4×10^{12}	1.8×10^{11}	1.8×10^{10}	3.5×10^{12}
t/cm^2	1.3×10^{12}	2.7×10^{13}	6×10^{12}	10^{13}	–	1.2×10^{13}	1.1×10^{11}	5.2×10^{13}	1×10^{10}	4×10^9	4.4×10^{11}
(n/t) Ratio	2×10^{-7}	0.5×10^{-8}	0.5×10^{-8}	1.2×10^{-9}	–	4×10^{-8}	1.7×10^{-6}	10^{-6}	10^{-3}	3.2×10^{-4}	7×10^{-7}

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	1990	1989	1989	1989	1989	1989	1989	1989	1989	1989
	21 Sept	5 March	24 Oct.	24 April	10 July	6 Sept.	29 Sept.	9 Nov.	21 July	22 Dec.	24 Oct.
Cathode:											
Material	Pd-Ag Alloy	Pd-Ag Alloy	Cold Rolled Pd	Pd	Pd	Pd	Pd	Pd	Pd ingot	Pd	Pdwire
Geometry	Tubes	Tubes	Hollow Cyl.	Ring	Hollow Cylinder	Hollow Cylinder	Hollow Cylinder	Rod	Cylinder	Pellet	Grid
Dimensions (mm)	3 Φ × 150 ht	3 Φ × 150 ht	–	2 thk.	–	–	–	4 Φ × 19 long	8 Φ × 16 long	4 Φ × 4 ht.	0.5 Φ × 800 long
Area (cm^2)	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	~2	<2	1~2 Pulsed	1~3 Pulsed	1~2 Pulsed	0.2~2.2 RF Superposed	100mA	≤35mA	30mA
Current Density mA/cm^2	350	350	~105	~160	<350	≤470	<350	<800	~100	~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 Concn. (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	–	0.93×10^4	5.88×10^4	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)	3.3×10^4	2.28×10^3	2.71×10^3	6.02×10^5	2.08×10^6	2.96×10^3	6.29×10^2	1.1×10^3			
(Atoms)	1.76×10^{13}	1.2×10^{12}	1.44×10^{12}	3.2×10^{14}	1.1×10^{15}	1.56×10^{12}	3.96×10^{11}	5.83×10^{11}	10^{12}	10^{11}	2×10^{10}
t/cm^2	1.6×10^{11}	1.1×10^{10}	0.8×10^{11}	2.2×10^{13}	1.7×10^{14}	2.4×10^{11}	6.2×10^{10}	2.12×10^{11}	1.8×10^{12}	0.8×10^{12}	0.5×10^{10}

The MR(Jr) cell was a smaller version of the Milton Roy cell with 6 Pd-Ag alloy tubes. This cell has earlier been used for routine H_2 generation for several years at the Chemistry Division. Cell #11 was a very novel cell wherein the electrolyte was KOD in the form of a paste applied on a multiwire grid electrode system made of alternate wires (0.5 mm dia) of Pd and Pt. Although this gave excess tritium of only 2×10^{10} atoms (lowest in the Table) it must be noted that the total inventory of electrolyte in this microcell was hardly 1.5 ml. In Cell #8 (PDR-I) an RF voltage was superposed on the applied DC voltage with a view to ascertain whether this would help improve the tritium production. As seen from the results there is no evidence of any improvement.

The Cells # 5, 6, & 7 used the same hollow Pd cylinder cathode deployed in Cell # 6 of Table I (Expt. PDC-I). As in the case of PDC-I current pulsing was resorted to in these three PDC series of experiments also. On completion of PDC-I the hollow Pd cathode was degassed at 300ºC in a vacuum furnace for over 2 hrs. Subsequent electrolysis in 0.1 M LiOD (Expt. PDC-II) terminated in an explosion after a charge of 423 amp-hrs had been passed. Careful measurements of the tritium content indicated that over 2 MBq (1.1×10^{15} atoms) of excess tritium had been generated during this experiment, (see Table II). This corresponds to an increase in tritium inventory by a factor of more than 20,000 relative to the total tritium input to this experiment.

This same cathode generated tritium two more times after degassing and reuse. (PDC-III & PDC-IV). But as seen from Table II in each subsequent run the quantum of tritium generated decreased further. For example in PDC-III while excess tritium recovered was 2.96 kBq, in PDC-IV the excess tritium was only 629 Bq even after 123 amp-hrs of charging. Prior to commencement of PDC-IV the electrode had been heated to 850ºC for 4 hrs in vacuum, cooled and again heated to 800ºC in D_2 gas atmosphere at 1 cm pressure for 3 hrs followed by degassing again under vacuum for 3 hrs. This very elaborate pretreatment would have cleansed the Pd of any remnant tritium within its interior, confirming that the fresh amount 629 Bq obtained in PDC-IV must have been generated during this run of electrolysis only. But the more important implication of this result is that even vacuum heating-annealing does not appear to have restored the ability of the Pd cathode to support nuclear reactions.

6. REAL TIME NEUTRON DIFFRACTION STUDY OF DEUTERON LOADING IN A Pd CATHODE /11/

Cell #9 (CD-4 of Chemistry Division) was primarily designed for conducting an online neutron diffraction study of the phase change occurring during the deuteration of a Pd rod. It is comprised of a covered Pyrex glass beaker with an 8 mm dia × 16 mm long Pd cathode of which 11 mm protruded underneath the cell and was set up in front of one of the neutron beams at the Dhruva research reactor at Trombay. The portion of the Pd rod exposed to atmosphere was given a thin protective coating of tin to minimise escape of deuterium. A platinum cell above the cathode served as anode and the electrolyte was 0.1M Li_2O in D_2O. A 0.18 mm thick Nafion membrane between the electrodes helped prevent direct recombination of deuterium and oxygen. Electrolysis was carried out at a steady current of 100 mA extending over a period of more than 8 weeks. Powder diffraction patterns were recorded periodically with the help of a 1 metre wide position sensitive neutron detector mounted so as to provide a 30° angular span. With this arrangement the real time development of the (111), (200) and (311) reflections could be studied.

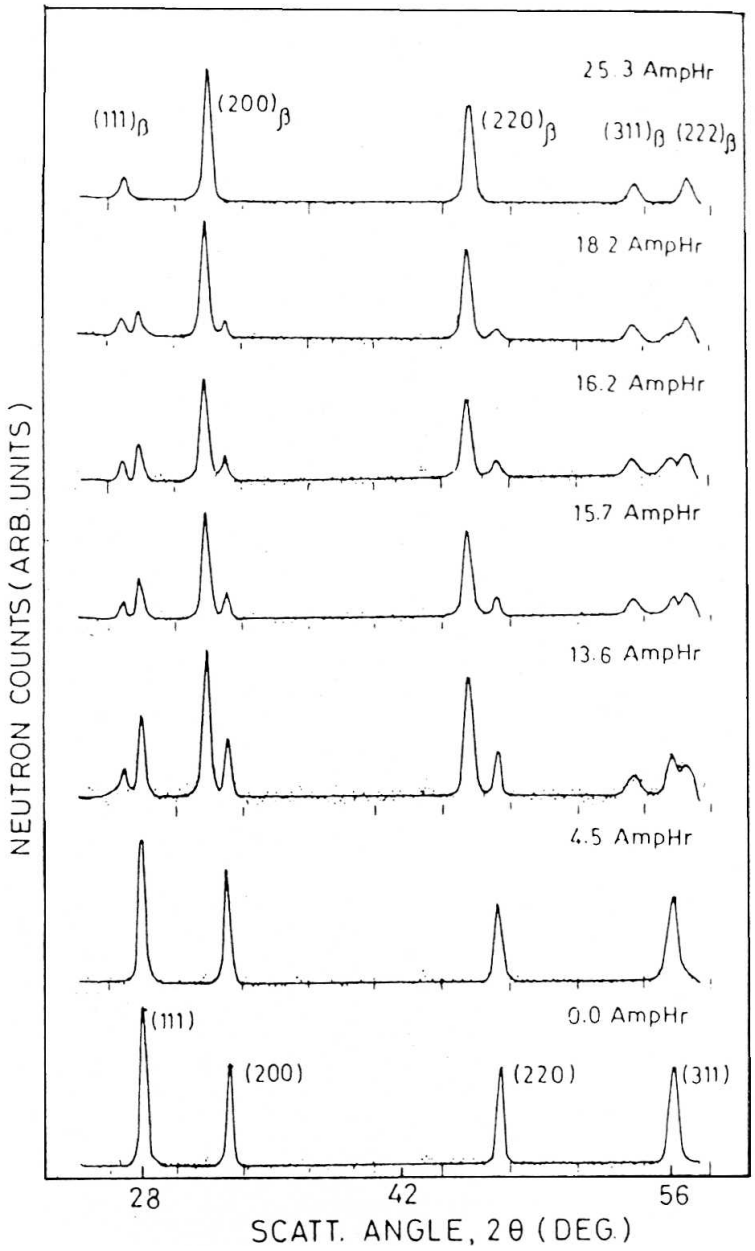

Fig. 10. Neutron Diffraction Patterns at Different Deuterium Loadings of Palladium Rod

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 A° characterized by the four peaks corresponding to the α phase seen in the bottom most pattern of Fig. 10. New peaks indicative of the precipitation of the β phase of the Pd-D system (lattice constant of 4.02 A°) showed up at around 10 amp-hrs of electrolysis. As electrolysis proceeded the intensity of the β phase peaks built up at the cost of α 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 β 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 stochiometry in the β phase could be

deduced. It was summarised from the study that the stochiometry at saturation was $PdD_{0.55}$. The main conclusion of this experiment relevant to cold fusion is that no new phases develop in Pd even after 100 amp-hrs of electrolysis.

PART B: D_2 GAS LOADING EXPERIMENTS

7.1 SEARCH FOR TRITIUM IN GAS LOADED Pd SAMPLES /5/

In these experiments D_2 gas was loaded into Pd samples after thoroughly degassing them and a search was made for the possible production of tritium in the samples. The tritium produced, if any, along with that in the initially loaded deuterium was extracted through isotopic exchange with distilled light water wherein the Pd sample itself served as a catalyst. From the activity measured in the water the amount of tritium "produced" in the Pd was computed,

D_2 gas was generated by reducing D_2O with Na in vacuum and stored under pressure in an SS dewar with liquid nitrogen cooling, in the presence of activated charcoal. The stock D_2O used had a tritium content of 0.075 nCi/ml (2.8 Bq/ml), corresponding to a (T/D) isotopic ratio of 3×10^{-14}. Pd samples either in the form of Pd black powder or Johnson & Matthey Pd-Ag foils were taken in an SS reaction vessel connected to a vacuum system (10^{-5} mm) through a buffer tank of 1 litre volume equipped with a pressure gauge. After degassing and cooling under vacuum, D_2 gas at 1 atm pressure was let into the buffer tank and the system sealed off for equilibration with the Pd contained in the reaction vessel for several hours or days at times. From the pressure drop observed the quantity of gas absorbed in the Pd could be deduced. The deuterated Pd samples were later immersed inside a measured quantity of distilled water for a few hours and the concentration of tritium in the water measured through standard liquid scintillation counting systems. The tritium content in the Pd was deduced therefrom knowing the gram moles of D_2 absorbed in Pd as well as the relevant equilibration constant (K).

Table III summarizes the results. The tritium activity measured in the distilled water was a small fraction of a nCi/ml (5 to 30 Bq/ml). The total quantity of tritium estimated to have been generated in the Pd foils is in the region of 10^{10} to 10^{11} atoms. It is observed that the (D/Pd) ratios attained following D_2 absorption are approximately similar in all the cases (0.20 to 0.63). The amount of tritium produced per gram of Pd sample varies widely, from ~1.2 to 20.8×10^{10} atoms/g. As may be expected the higher value is consistent with the longer duration of equilibration time (240 hours) between D and Pd, but the large Pd foil (column #3) which was also equilibrated for 240 hours has given only 2.2×10^{10} atoms of t/g of Pd. In all cases the finally attained (T/D) ratios which are in the range of $.3 \times 10^{-11}$ to 8.3×10^{-11} are two to three orders of magnitude higher than that of the initial gas value namely ~ 3×10^{-14}. Thus fresh tritium amounting to about 10^{10} or 10^{11} atoms appears to have been created in the Pd, presumably due to "cold fusion" reactions. It is not clear whether the tritium was produced during the absorption process or during the subsequent "curing" or equilibration phase.

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_2 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	32×10^{-11}	1.1×10^{-11}	$.87\times10^{-11}$	3.4×10^{-11}	8.3×10^{-11}
Absolute tritium activity (bq)	410.7	370	429.2	717.8	159.1
Total tritium atoms in Pd	2.31×10^{-11}	2.02×10^{-10}	24×10^{11}	4.1×10^{11}	8.96×10^{10}
Tritium atoms per g of Pd	1.2×10^{10}	21×10^{10}	2.2×10^{10}	3.8×10^{10}	20.8×10^{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.

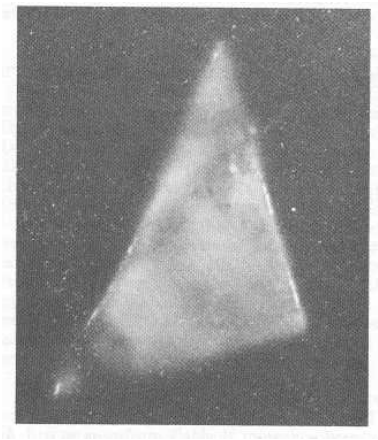

Fig. 11. Autoradiograph of Deuterated Triangular Pd-Ag Foil

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_2 gas pressure or temperature was cycled between high and low values using liquid nitrogen. The well type neutron counter employing 24 He^3 detectors embedded in paraffin, along with the associated data acquisition

system described in Sec 3.4 was used for the neutron yield measurements. The counting efficiency was determined to be 10%.

In these experiments first conducted in June 1989, Ti pieces were to begin with equilibrated with D_2 gas at 10 atm and 77 K for ~20 minutes. The temperature was then allowed to increase slowly to ambient level, with simultaneous evacuation resulting in desorption of D_2 gas from the Ti shavings. This resulted in large neutron bursts lasting between half an hour to ~2 hours each as shown in Figs. 12a to 12d. Prior to the experiments of Figs 12c & 12d, the D_2 gas pressure and temperature were simultaneously cycled. While the first three measurements were carried out with the same charge of Ti, the last one was done with a fresh charge which could be the reason for the slightly different characteristics of Fig. 12d. The integrated neutron yield in these experiments varied in the range of 10^5 to 10^7 neutrons.

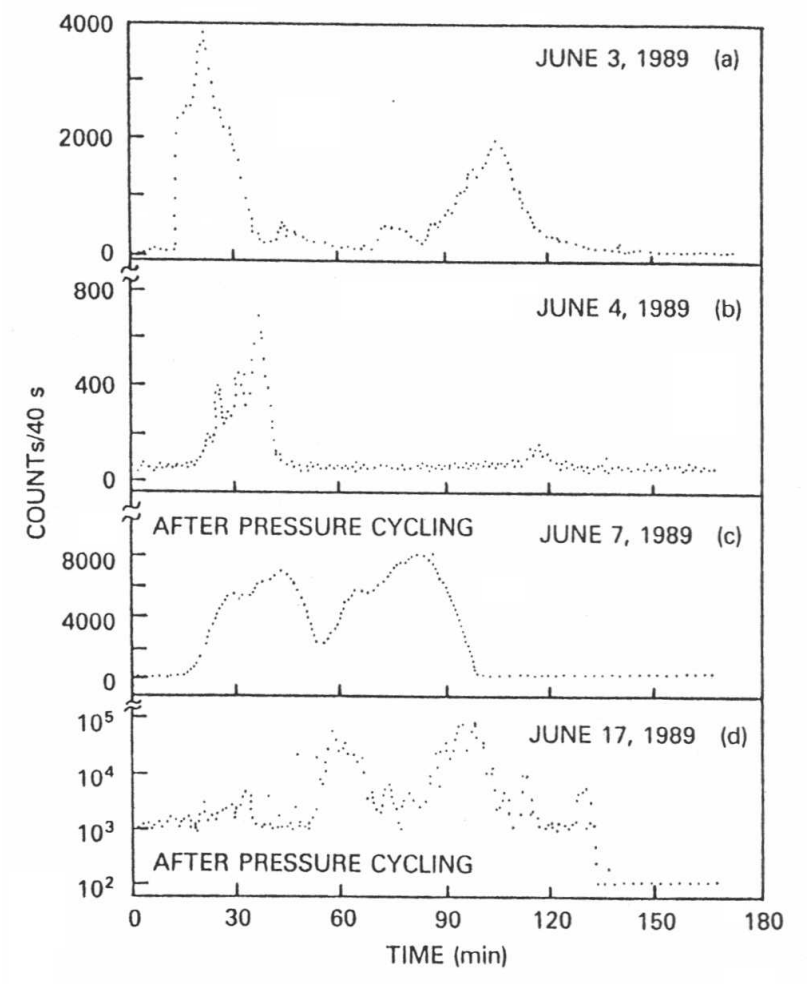

Fig. 12. Neutron Counts Variation During Frascati Type Experiment with Ti Shavings

The authors of these measurements have reportedly been unsuccessful in their attempts to measure neutrons again in repeated attempts to reproduce the earlier results /13/. However they have detected the presence of tritium in some of the newly loaded Ti shavings through the technique of autoradiography. For this they employed a high speed Polaroid camera. A 25 μm thick aluminium foil placed between the shavings and the photo sensitive film ensured that false

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_2 ATMOSPHERE (TPPED/NtPD) /5/

In these experiments machined and chemically cleaned targets of Ti were individually heated to temperatures of up to 900ºC 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_2 and D_2 gas bottles. Degassing was initially carried out at 900ºC for several hours until a vacuum of 10^{-5} mm was maintained steadily. The targets were later heated to 600ºC in H_2 atmosphere at a few mm of pressure. The induction heater was then switched off and the target allowed to cool absorbing H_2 in the process. At least three cycles of H_2 absorption/desorption was given "to create active sites for D_2 absorption". Three such heating/cooling cycles were then carried out with D_2 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^{rd} or 4^{th} cycle.

The targets typically absorbed $<10^{19}$ molecules of D_2. 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 δ characterizing this phenomenon is given by $\delta^2 = (s/f\pi\mu)$ where s and μ 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, δ 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_2 gas, only a few of them gave positive evidence for the presence of tritium. Interestingly the samples which soaked up large amounts of D_2 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 10^9 to 10^{10} atoms of tritium. The total number of tritium atoms in the whole target works out to be $\sim 10^{11}$. This is to be compared with the 10^{19} to 10^{20} atoms of D absorbed in all by the Ti, pointing to a gross (T/D) ratio of $>10^{-9}$.

TABLE IV

TRITIUM CONTENT IN D_2 GAS LOADED Ti TARGETS

Date of loading	14 June 89	9 June 89	21 Mar 90
Shape of sample	Disc	Cone	Cone
Sample mass (g)	.98	.206	.2
D_2 absorbed (mg)	.42	.07	.29
T activity from X-ray counts (bq)	290	1300	5.5×10^{6}
Date of counting	16 June 89	16 June 89	28 Mar 90
Tritium atoms	1.5×10^{11}	6.5×10^{11}	3.0×10^{11}
T/D ratio	1.2×10^{-9}	3.2×10^{-8}	7.1×10^{-5}

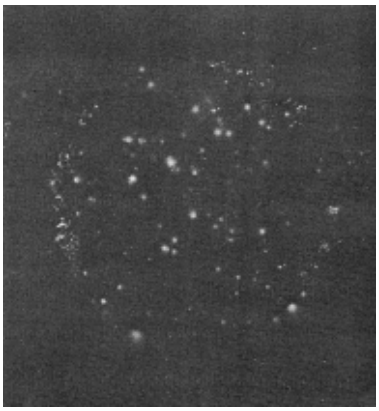

Fig. 13. Autoradiograph of Deuterated Ti Disc

Interestingly one of these disc targets which was kept in front of a paraffin encased BF_3 neutron detector and monitored over a weekend, suddenly emitted a large burst of neutrons on its own lasting over a time span of 85 mins. (See Fig. 2 of companion BARC paper /6/) The most intriguing feature of this experiment is that ever since the RF heating system became defective in September 89, these results could not be reproduced even once using a resistance furnace even though the power was higher and much greater quantities of D_2 gas could be absorbed. It was only in March 90 when a 1MHz furnace became available that a conical target once again indicated the presence of μCi amounts of tritium (See Table IV). The authors are therefore obliged to conclude that oscillatory electric and magnetic fields somehow play a crucial role in stimulating anomalous nuclear reactions on the surface of these machined Ti targets. In this context the recent Coherent Theory of Cold Fusion proposed by Hagelstein /14/ appears to give some theoretical insight as to possible mechanisms which could explain these observations.

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/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 J × 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.

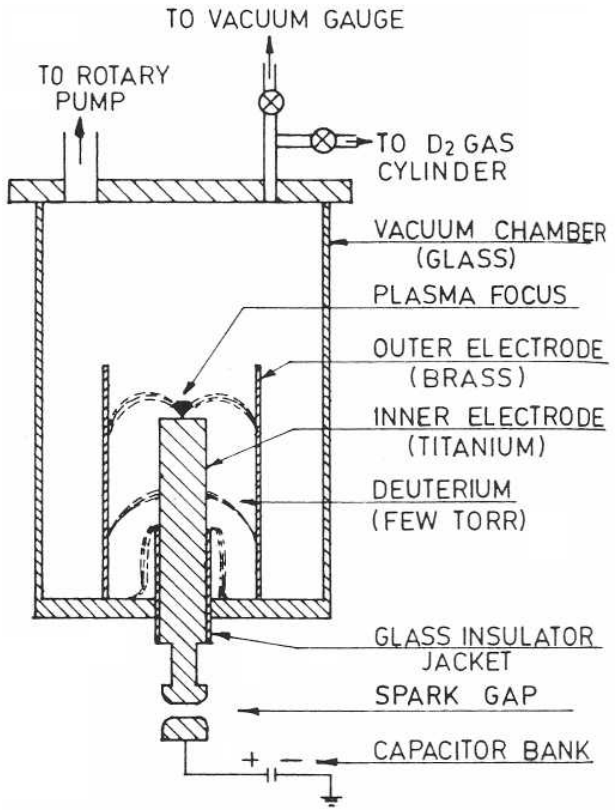

Fig. 15. Schematic Diagram of a Plasma Focus Device With Central Ti Electrode

During January 90 an experimental program was underway to study the influence of anode material on the characteristics of the plasma created and consequently on the neutron production mechanisms of a PF device. Brass, aluminium, stainless steel and titanium were investigated for neutron yield systematics under various deuterium filling pressures (1-10 mbar).

During the experiments with a Ti anode, about 80 charge/discharge shots were carried out. After each shot the chamber was flushed and filled with fresh D_2 gas. Some of these shots were performed with the central electrode operated with negative polarity. This mode of operation would direct the deuteron beams of a few hundred keV energy which are known to be generated in PF devices, towards the central electrode. The neutron yield in every shot was recorded with the help of a bank of calibrated activation type silver cathode Geiger Muller counters located close to the device. Typically with ~3 kJ of stored energy, 10^7 neutrons were produced in each normal shot i.e. when the central electrode is used as anode. In the polarity reversed mode of operation also neutrons were produced but the magnitude of the neutron burst was an order of magnitude smaller.

In view of the special role of Ti in D_2 gas loaded cold fusion experiments, the Ti electrode was tested for induced radioactivity using autoradiography immediately after the experiment on two consecutive nights (4^{th} and 5^{th} Jan. 90). No image was found at that time. But five weeks later (on 9^{th} Feb 90) using a new NaI detector set up it was discovered that a surprisingly high activity of ~392 μCi (~10^{16} atoms of tritium) had built up on the surface of the Ti electrode which had been exposed to the plasma focus shots. To confirm this, the rod was subjected to overnight autoradiography once again. As seen in Fig. 16 a very beautiful and impressive image was obtained. This image has since been reproduced several times through repeated autoradiographic

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 β 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 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 ~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 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.

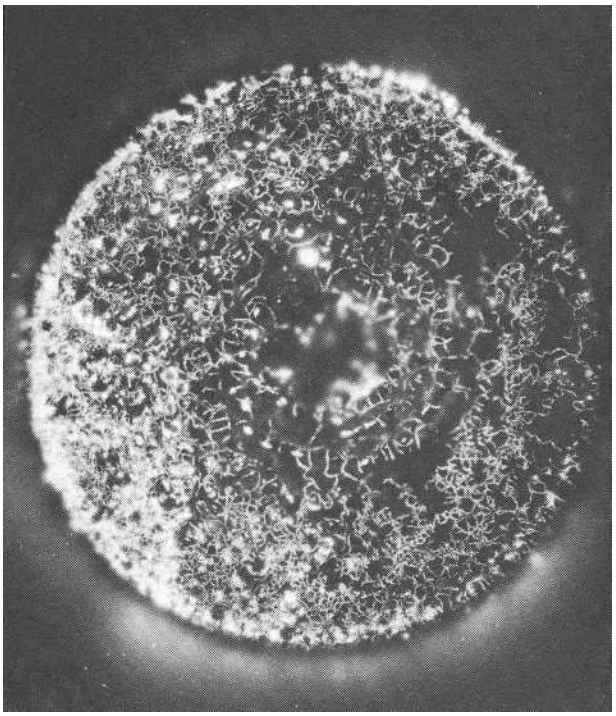

Fig. 16. Autoradiograph of Central Ti Electrode of PF Device (5 Weeks After Charging)

7.5 OBSERVATION OF HIGH TRITIUM LEVELS IN AGED DEUTERATED Ti TARGETS/5/

The Division of Radiological Protection of BARC had procured a number of deuterated titanium targets on copper backing during 1972 to 1981 for dosimetry studies with accelerator based neutron sources. Twelve such targets were available, nine procured from M/S Amersham International of U.K. and the remaining three from the Isotope Division of BARC. In view of the various studies involving deuterated titanium targets described in the earlier sections, it was conjectured that cold fusion reactions might have occurred in these "aged" targets over the past 9 to 18 years and if so, it was argued, they should contain considerable amounts of tritium. In order to check this hypothesis these aged targets were subjected to various studies for establishing the magnitude of tritium in them. Five different techniques namely autoradiography, Ti K X-ray counting with NaI and high purity germanium detectors, β counting with proportional counters and current measurements with an ionization chamber were used. The details of the targets, measurements and results are described in Ref /5/. It was found that the absolute tritium content in the targets varied between 0.3 and 150 MBq. Inquiries with the suppliers of these targets indicate that while inadvertent contamination during manufacture to the extent of a few hundred Bq is in principle a likely possibility, contamination levels in the MBq region is difficult to explain. The tritium levels in these aged TiDx targets expressed in terms of the (T/D) isotopic ratios was seen to vary in the range of 0.07 to 3.5×10^{-4}. For comparison the tritium activity of the D_2O moderator of a CANDU type power reactor is almost 30 Ci/l even at saturation, corresponding to a (T/D) ratio of 10^{-5}. In contrast the (T/D) ratio of fresh D_2O from a factory is typically in the region of 10^{-14} to 10^{-13} only. Hence the authors are inclined to speculate that a plausible explanation for the unexpectedly high tritium levels in aged deuterated Ti targets could be the occurrence of cold fusion reactions.

8. MEASUREMENT OF TRITIUM LEVELS IN AQUEOUS AND METALLIC SAMPLES

As a consequence of the many years of operational experience with heavy water moderated research and power reactors in India, considerable expertise has been built-up in the area of tritium measurements, particularly in moderator and coolant circuits as well as in environmental samples. The status of development of the field of "Tritium Measurement and Applications" was reviewed recently at a Symposium /17/ held in Bombay to mark the golden jubilee of the discovery of tritium in 1939.

8.1 Analysis of Aqueous Samples

The tritium levels in the electrolytes and other aqueous samples was measured by expert groups at the Isotope and Health Physics Divisions of BARC. Commercial liquid scintillation counting systems such as Packard Instruments Model 4530 or LKB Systems Model 1215 (RACKBETA-II) which provide automatic quench correction facilities were employed. ^{40}K free vials were used. Commercially available scintillation cocktail, INSTAGEL, was found rnosff suitable as it, gave minimum chemiluminiscence. Double distilled water was used for diluting samples to reduce PH level as well as quenching impurities. In some electrolyte samples chemiluminiscence effects entirely masked the true tritium signal. Fig. 17 shows the

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

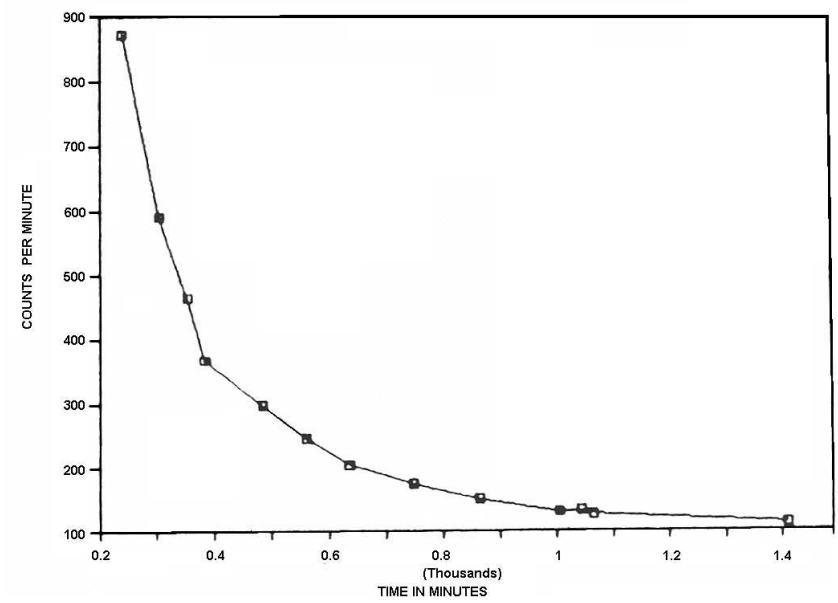

Fig. 17. Chemiluminiscence Decay Curve of a Severe Electrolyte Sample

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 chemiluminiscence 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^2. 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^2. 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.

8.3 Autoradiography

As mentioned already the technique of auto radiography has been employed very effectively at BARC to study the spatial distribution of near surface tritium produced in deuterated Ti and Pd targets. In order to obtain good resolution the samples were kept in contact with medical X-ray film and exposed overnight. The fogging of the film is due to the combined effect of tritium β and the K X-rays excited in the host metal lattice. Sometimes a stack of two films was mounted close to the sample and it was observed that the second film also gives a similar but fainter image, ruling out the suspicion that image formation could be due to mechanical (scratching of films) or chemical reduction effects.

9. CONCLUSIONS

Experiments carried out by a number of totally independent groups employing diverse experimental set ups have unambiguously confirmed the production of neutrons and tritium both in electrolytically loaded and gas loaded Pd/Ti lattices.

Tables I & II present in a nut shell the main results of the BARC electrolysis experiments. It may be noted that in all 22 cells/experiments have yielded excess tritium varying over a wide margin of 10^{10} to 10^{16} atoms. Roughly half of these may be described as having been "doubly successful" since in these both neutrons and tritium were measured. The main conclusions to emerge from the electrolysis experiments may be summarized as under:

(a) The most important observation is the surprisingly low neutron-to-tritium yield ratio, first reported by us at the Karlsruhe meeting in July 1989 /3/. 8 out of the 11 doubly successful cells of Table I have given values in the region of 10^{-6} to 10^{-9} for this ratio, while two experiments have given a comparatively larger value of 10^{-3} to 10^{-4}. These ratios may be considered as overestimates since in most of the experiments the tritium escaping with the electrolytic gases has not been accounted for.

(b) The Trombay electrolytic experiments have also convincingly demonstrated that both neutrons and tritium are generated concomitantly. This is evident from the sharp increase in the tritium concentration of the electrolyte immediately after a large neutron burst in several of the experiments.

(c) Another significant observation pertains to the specific charge passed per unit of cathode surface area, namely amp-hrs/cm^2 at the time of the first neutron burst. This quantity which may be called the "switching on charge" is seen to be in the range of 0.6 to 3.2 amp-hrs/cm^2 in 8 out of the 11 experiments of Table I. In the remaining three cells (all of which have used LiOD as electrolyte) the switching on charge is an order of magnitude or more higher.

(d) It is significant that in all the five experiments which used 5 M NaOD as electrolyte the switching on charge was ≤ 3 amp-hrs/cm^2. In the isolated instance where Li_2SO_4 was used the switching on charge was the lowest namely 0.15 amp-hr/cm^2 among all the experiments reported in Table I.

(e) In 8 out of the 11 cases of Table I the first neutron burst has occurred on the very first day of electrolysis, in fact within 9 hrs of commencement of electrolysis (except for experiment #7 where it occurred after ~24 hrs). This seems to be a unique feature of the Trombay results.

(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 cm^2 of cathode surface) to a (D/Pd) ratio of ~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 ≤ 3 amp-hr/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 #l 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/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 4×10^9 to 1.7×10^{14} t/cm^2. In 10 of 22 cells it is in the range of 10^{12} to 10^{14} t/cm^2 while most of the other cells have given values in the region of 10^{10} to 10^{12} t/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 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 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.

ACKNOWLEDGEMENTS

We are very grateful to all the authors off the individual BARC cold fusion papers for readily supplying experimental details and results for the preparation of this overview paper. We wish to thank Drs R. M. Iyer and T. S. Murthy for many fruitful discussions. The considerable help rendered by A. Shyam, T. C. Kaushik, R. K. Rout, V. Chitra, S. Ranganekar and D. V. Periera in the time consuming task of editing and formatting of this manuscript is warmly acknowledged.

REFERENCES

1. M. Fleischmann et al, J. Electranal. Chem. Vol. 261, pp 301-308 (1989)
2. A. De Ninno et al, Europhys. Lett. Vol. 9, p 221 (1989)
3. P. K. lyengar, "Cold Fusion Results in BARC Experiments, Proc. Fifth Int. Conf. on Emerging Nuclear Energy Systems, Karlsruhe (FRG) World Scientific, Singapore, p 291 (1989)
4. P. K. lyengar & M. Srinivasan (Eds), BARC Studies in Cold Fusion (April-September 1989) Report BARC-1500, (1989)
5. P. K. lyengar et al, Bhabha Atomic Research Centre Studies in Cold Fusion, Technical Note, Fusion Technol. August (1990).
6. M. Srinivasan et al, Statistical Analysis of Neutron Emission in Cold Fusion Experiments, Proc. First Annual Conf. on Cold Fusion, Salt Lake City, Utah March 28^{th}-31^{st} (1990).
7. H. Bose et al, Cold Fusion Cell Experiments with Cylindrical Palladium Cathode at Sub-Ambient Temperatures, Unpublished Note, March (1990).
8. G. Venkateswaran, Personal Communication, 11^{th} March (1990).
9. C. K. Mathews, Personal Communication 2nd March (1990).
10. N. J. C. Packham et al, J. Electroanal. Chem. Vol. 270 pp 451-458 (1989).
11. R. Mukopadhyay et al, Accepted for publication in Solid State Comm. (1990).
12. P. Raj et al, J. Less-Common Metals, Vol. 130 p 139 (1987).
13. P. Raj, Personal Communication 11^{th} March (1990).
14. P. L. Hagelstein, "Status Report on Coherent Fusion Theory", Proc. First Annual Conf. on Cold Fusion, Salt Lake City, Utah. March 28^{th}-31^{st} (1990).
15. R. K. Rout et al, Unpublished Note, (1990).
16. Symposium on "Tritium Measurements and Applications" Feb 22-23, 1990, BARC, Bombay, Bulletin of the Indian Association for Radiation Protection Vol. 13, No. 1, (1990).
17. A. N. Singh et al, An Instrument for On-line Monitoring of Tritium-in-Air in Heavy Water Reactors, Nucl. Instru. & Meth, Vol. 258 p 250, (1987).