

Research Article

# Revisiting the Early BARC Tritium Results

Mahadeva Srinivasan<sup>\*,†</sup>

*Bhabha Atomic Research Centre, Trombay, Mumbai, India*

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## Abstract

Within days of the F&P announcement of 1989, several groups at BARC embarked on a program to look for the generation of neutrons and tritium when deuterium (or hydrogen) is loaded into metals such as Pd, Ti and Ni. Electrolytic, gas and plasma loading techniques were deployed. Post run electrolyte samples were analyzed for tritium content using standard liquid scintillation techniques. In the case of gas and plasma loaded “dry” samples, surface tritium content was measured directly using windowless beta counters. Autoradiography was deployed as a very effective tool to monitor the spatial distribution of tritium in the near surface layers of test samples. The first confirmation of copious tritium generation was obtained on 21 April 1989 in a commercial “Milton Roy” Pd–D<sub>2</sub>O electrolytic cell. The results obtained during the first year of the BARC Cold Fusion campaign were presented at ICCF 1 in March 1990 where we reported observing tritium generation in 22 different electrolytic cells, which were set up totally independently by diverse research groups. The present paper revisits the early BARC tritium results obtained in a variety of experimental configurations during the period 1989–1996.

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## 1. Introduction and Scope of BARC Studies

A four-line news item on the Fleischmann and Pons (F&P) announcement carried by the 24 March 1989 issue of the Times of India newspaper, triggered a flurry of activity at the Bhabha Atomic Research Centre (BARC) in Mumbai. Within days, groups with expertise in a variety of disciplines such as hydriding of metals, electrochemistry, isotope exchange processes in upgrading heavy water, fusion plasma experiments and neutron and tritium measurements, set up electrolytic cells with widely different geometrical configurations, in order to verify the nuclear origin of the F&P effect. Because neutrons and tritium were the commonly expected products of fusion reactions, these were the signatures that the BARC experiments sought. BARC, being the premier nuclear R & D centre in India with more than 50 divisions and 3500 scientists and engineers, had the expertise and equipment readily available for study of every aspect of nuclear science.

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<sup>\*</sup>E-mail: chino37@gmail.com

<sup>†</sup>Retired.

Details of the various electrolytic cells setup and the results of the neutron and tritium measurements carried out in the early years are documented in [1–4]. The first publication on the confirmation of generation of neutrons and tritium in the BARC Cold Fusion experiments was presented at the Fifth International Conference on Emerging Nuclear Energy Systems (ICENES V), held in Karlsruhe during July 1989 [1]. A comprehensive paper with 50 authors, published in the August 1990 issue of *Fusion Technology* [4], describes in great detail the Cold Fusion experiments conducted at BARC during the first year following the F&P announcement, inclusive of the gas/plasma-loaded Ti samples studies [5]. An interim progress report, titled “BARC Studies in Cold Fusion,” covering the period April to September 1989 [2] unambiguously confirming the occurrence of nuclear reactions at room temperature in deuterided Pd and Ti samples was published around the same time as the highly negative findings of the report of the Energy Research Advisory Board Cold Fusion Panel set up by the US Department of Energy and played a key historical role in helping keep interest in the subject alive. During 1992–1995, additional experiments were carried out with Ni–H systems, and these also indicated the production of tritium [7–9].

### 1.1. Measurement of tritium levels in solution samples

Tritium levels in the electrolyte samples collected after operation of the cells for some time were measured by specialist groups of the Isotope and Health Physics divisions of BARC using well-known liquid scintillation counting techniques applicable for low energy beta emitters, taking the following precautions: (a)  $^{40}\text{K}$  free counting vials were employed to minimize background counts; (b) For higher count rate cases, 0.1–2.0 ml of sample was added to the scintillator, while for low count rate samples,  $\approx 10$  ml was used. In the higher concentration samples, pH was reduced by diluting with double distilled water, in order to minimize chemiluminescence as well as quenching effects. If warranted, micro-distillation of the samples was resorted to before the addition of scintillation cocktail. Whenever possible, the measured tritium values were cross-checked with results obtained after chemiluminescence cooling; and (c) Instagel scintillation cocktail was used as solvent to minimize chemiluminescence interference effects. Samples of the electrolyte saved prior to the commencement of electrolysis were counted along with the test samples drawn from the operating cells. To compute the excess tritium produced in each run, the following points were considered: (i) Initial tritium concentration in the stock  $\text{D}_2\text{O}$ . (This was typically a few Bq/ml.) (ii) To be on the conservative side, the tritium carried away by the  $\text{D}_2$  gas stream during electrolysis was neglected and (iii) Dilution effects from the periodic make-up of  $\text{D}_2\text{O}$  were accounted for.

### 1.2. Measurement of tritium on surface of solid samples

Tritium which has a half life of 12.3 years, emits a low energy  $\beta$  having a maximum energy of 18.6 keV corresponding to an average energy of 5.7 keV only. The range of the maximum energy  $\beta$  being as small as  $9 \text{ mg/cm}^2$ , tritium located only within this depth can be detected through direct beta particle counting. Since these  $\beta$ s are stopped even by the ‘thin’ windows of most nuclear particle detectors, for direct counting of tritium windowless detectors have to be employed. In the case of titanium samples, the upper half of the  $\beta$  spectrum is able to excite the characteristic  $\text{K}\alpha$  (4.5 keV) and  $\text{K}\beta$  (4.9 keV) X-rays of titanium. (This does not happen in Pd.) The mean free path of 4.9 keV soft X-rays in titanium metal being  $\sim 10 \text{ mg/cm}^2$  tritium present in deeper layers of the titanium also can get detected through measurement of its characteristic K X-rays.

### 1.3. Autoradiography

Autoradiography is a simple technique to establish the presence of radiation emitting zones and has been used very effectively at BARC to study the spatial distribution of near surface tritium in solid cold fusion samples. To achieve

good spatial resolution the samples are kept in close contact with medical X-ray film and exposed in a dark room for times ranging from a few hours to a few days. In the case of titanium samples, fogging is due to the combined effect of tritium  $\beta$ s and the Ti K X-rays. To rule out the suspicion that image formation could be due to mechanical (scratching of films) or chemical reduction effects caused by the deuterium (or hydrogen), exposures were carried out with a stack of two films in contact with some samples, and it was confirmed that the second (or outer) film also gave a similar but fainter image indicating that image formation is due to nuclear particles only. While autoradiography has the advantage that it gives spatially resolved images of the distribution of tritium (or other radiation emitting substances), quantitative estimates are unreliable due to the saturation characteristics of image formation in photographic films. Highly localized regions of tritium (“hot spots”) containing  $\approx 0.2$  kBq of activity or  $10^{11}$  atoms of tritium each can be barely detected through a 100 h exposure.

## 2. Electrolysis Experiments during 1989–90 [1–4]

As mentioned earlier, several groups at BARC set up electrolytic cells using whatever samples of Pd they could get hold of, within days of the F&P announcement. The Neutron Physics Division had a head-start because, coincidentally, on 24 March 1989, a large cathode surface area ( $300\text{ cm}^2$ ) Pd–D<sub>2</sub>O electrolytic cell with 5 M NaOD electrolyte was all set and ready to be switched on! This off-the-shelf, ultra-pure hydrogen generator with 16 Pd–Ag alloy tubular cathodes arranged along a circle (inner and outer nickel cylinders constituted the anode) had been procured from Milton Roy Co. of Ireland for generating oxygen-free deuterium gas for plasma focus hot fusion experiments. On receipt of the F&P announcement, two different neutron monitors were moved on either side of the cell, while a third neutron detector bank placed over a meter away served as background monitor. The first evidence of neutron emission was obtained on 21 April 1989 when both the foreground neutron detector channels gave a dozen coincident peaks of varying magnitude during a four hour run when the cell current was being slowly ramped up. Four hours after commencement there was a huge neutron burst and the cell operation got automatically terminated by its protection circuit. (A couple of the Pd cathode tubes even got ruptured during this incident due to overheating.) A sample of the electrolytic solution which was later analyzed for tritium content indicated that the tritium concentration had attained a high level of 55.6 kBq/ml ( $1.5\text{ }\mu\text{Ci/ml}$ ), corresponding to a total tritium yield during the whole run of 13.9 MBq ( $\sim 8.10^{15}$  atoms of T). The post run tritium concentration of the electrolyte was  $\sim 20,000$  times the initial value.

Table 1 summarizes details of eight electrolysis experiments in which both neutron and tritium production were observed. The quantum of tritium observed in the electrolytes of 11 additional cells, in which neutron production was not monitored, are summarized in Table 2 of [3]. The magnitude of tritium generated in the various BARC cells (22 cells in all) was in the range of a few kBqs to a few MBqs.

**Table 1.** Electrolysis experiments in which both neutrons and tritium generation were observed.

Division	Cathode Material	Geometry	Area (cm <sup>2</sup> )	Anode	Yield		Ratio (n/T)
					Neutron yield	Tritium yield	
1. Desalin *	Ti	Rod	104	ss pipe	$3 \times 10^{+7}$	$1.4 \times 10^{+14}$	$2.1 \times 10^{-7}$
2. Neut Phy.*	Pd–Ag	Tubes	300	Ni Pipes	$4 \times 10^{+7}$	$8 \times 10^{+15}$	$5 \times 10^{-7}$
3. HWD*	–	–	300	–	$9 \times 10^{+7}$	$1.9 \times 10^{+15}$	$5 \times 10^{-7}$
4. HWD*	–	5 Disks	78	Porus Ni	$5 \times 10^{+4}$	$4.1 \times 10^{+15}$	$1.2 \times 10^{-9}$
5. Anal. Ch <sup>@</sup>	Pd	Hol Cyl.	5.9	Pt Mesh	$3 \times 10^{+6}$	$7.2 \times 10^{+13}$	$4 \times 10^{-8}$
6. ROMg <sup>@</sup>	–	Cube	6.0	–	$1.4 \times 10^{+6}$	$6.7 \times 10^{+11}$	$1.7 \times 10^{-4}$
7. ROMg <sup>@</sup>	–	Pellet	5.7	–	$3 \times 10^{+6}$	$4 \times 10^{+12}$	$1 \times 10^{-4}$
8. App. Chem <sup>@</sup>	–	Ring	18	–	$1.8 \times 10^{+8}$	$1.8 \times 10^{+11}$	$1 \times 10^{-3}$

Electrolyte: \*5 M NaOD<sup>@</sup> 0.1 M LiOD.

**Table 2.** Tritium production in deuterium gas loaded Pd samples.

Experiment	No. 1	No. 2	No. 3	No. 4	No. 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. 1989	19 Sept. 19 89	7 Mar. 1990	18 Sept. 19 89
Vol. of D <sub>2</sub> absorbed (ml)	1325	34.5	518.4	222	20.2
(D/Pd) ratio	0.63	0.46	0.45	0.20	0.45
Equilibration time (h)	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	$32 \times 10^{-11}$	$1.1 \times 10^{-11}$	$8.7 \times 10^{-11}$	$3.4 \times 10^{-11}$	$8.3 \times 10^{-11}$
Absolute tritium activity (Bq)	410.7	370	429.2	717.8	159.1
Total tritium atoms in Pd	$2.31 \times 10^{11}$	$2.02 \times 10^{10}$	$24 \times 10^{11}$	$4.1 \times 10^{11}$	$8.96 \times 10^{11}$
Tritium atoms per g Pd	$1.2 \times 10^{10}$	$21 \times 10^{10}$	$2.2 \times 10^{10}$	$3.8 \times 10^{10}$	$20.8 \times 10^{10}$

### 2.1. Deuterium gas loaded Pd samples [4]

In these experiments conducted by the Chemistry Division of BARC, D<sub>2</sub> gas was generated by reducing D<sub>2</sub>O 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<sub>2</sub>O used had an initial tritium content of 0.075 nCi/ml (2.8 Bq/ml), corresponding to a (T/D) isotopic ratio of  $3 \times 10^{-14}$ .

Pd samples either in the form of Pd black powder or Johnson & Matthey Pd–Ag foils, were placed 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<sub>2</sub> 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. The concentration of tritium in this water was then measured through standard liquid scintillation counting techniques. The tritium content in the Pd was deduced there from knowing the gram moles of D<sub>2</sub> absorbed in Pd as well as the relevant equilibration constant (*K*).

Table 2 summarizes the results. The tritium activity measured in the distilled water was a fraction of a nCi/ml (5–30 Bq/ml). The total quantity of tritium estimated to have been generated in the Pd foils was in the region of  $10^{10}$ – $10^{11}$  atoms. It is observed that the (D/Pd) ratios attained following D<sub>2</sub> absorption are in the region of 0.20–0.63. The amount of tritium produced per gram of Pd sample varied widely, from  $\sim 1.2$  to  $20.8 \times 10^{10}$  atoms/g. As may be expected the higher values are generally consistent with the longer duration of equilibration time (240 h) between D<sub>2</sub> gas and Pd, but the large Pd foil (column No. 3) which was also equilibrated for 240 h has given only  $2.2 \times 10^{10}$  atoms of tritium per gram of Pd. In all cases the finally attained (T/D) ratios which were in the range of  $0.3 \times 10^{-11}$ – $8.3 \times 10^{-11}$  are two to three orders of magnitude higher than that of the initial gas which was  $3 \times 10^{-14}$ . Thus fresh tritium amounting to about  $10^{10}$  or  $10^{11}$  atoms appears to have been generated in the Pd, presumably following “cold fusion” reactions. It is not possible to conclude whether the tritium was produced during the gas absorption phase or during the subsequent “curing” or equilibration process with water.

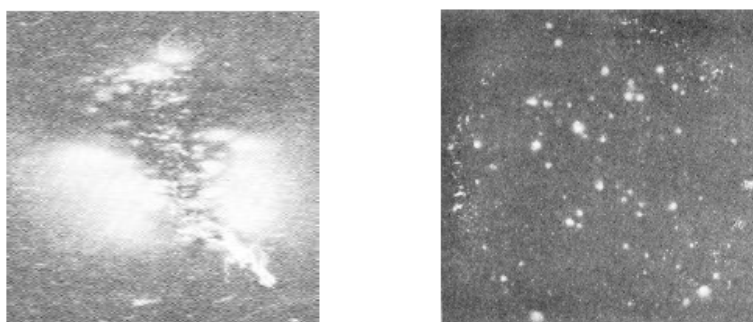
### 2.2. Aged TiD targets [4]

Several groups in BARC had procured deuterated titanium targets on copper backing during 1972–1986 for dosimetry studies with accelerator based neutron sources. Thirteen such targets were available, nine procured from M/s. Amersham International of UK and four fabricated in-house by the Isotope Division of BARC. In view of the ongoing studies

involving deuterated titanium targets it was conjectured that “cold fusion” reactions might have occurred in these “aged” targets over a period of 4–18 years and if so, it was argued, they should contain detectable amounts of tritium. In order to check this hypothesis these aged targets were counted as well as autoradiographed for establishing the magnitude of tritium if any present in them employing the techniques described above. The details of the targets, measurements and results are described in [4]. It was confirmed that all 13 targets indicated the presence of tritium, the absolute content varying between 0.3 and 150 MBq. Inquiries with the suppliers of these targets indicated that while inadvertent contamination during manufacture to the extent of a few hundred Bq was 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  $0.07\text{--}3.5 \times 10^{-4}$ . For comparison the tritium activity of the D<sub>2</sub>O moderator of a CANDU type power reactor is at most 30 Ci/l even at saturation, corresponding to a (T/D) ratio of  $10^{-5}$ . Besides, the (T/D) ratio of fresh D<sub>2</sub>O from a production plant is typically in the region of  $10^{-14}\text{--}10^{-13}$  only. It is therefore postulated that the unexpectedly high tritium levels in aged deuterated Ti targets must most probably be due to the occurrence of “cold fusion” reactions over the intervening years.

### 2.3. Cone and disc-shaped Ti samples subjected to RF heating in D<sub>2</sub> atmosphere

In these experiments, chemically cleaned machined targets of Ti were individually heated to temperatures of up to 900°C in a glass chamber using a surrounding induction heating coil (1–2 MHz frequency, 3–6 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 7 h until a vacuum of  $10^{-5}$  mm was maintained steadily. The Ti samples were then heated to 600°C in H<sub>2</sub> atmosphere of a few mm pressure. The induction heater was subsequently 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 repeated 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 with each cycle, saturating in the third or fourth cycle. The targets typically absorbed  $< 10^{19}$  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 a “skin depth” in the surface region and the (D/Ti) ratio there would be substantially higher. (It is common knowledge that when a metallic object is heated by induction heating, the current distribution and heating rate falls off exponentially with increasing depth.)



**Figure 1.** Autoradiographs of a deuterated Ti cone and a disc target .

After loading, all the targets were subjected to various tests such as autoradiography, K X-ray counting etc discussed earlier, 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  $\times$  2 mm thick) and a couple of conical pieces meant for use as electrodes. Figure 1 depicts the autoradiographs of a deuterated cone and disc respectively. The radiographs show large numbers of “hot spots” randomly distributed within the boundary. The occurrence of spots all along the rim of the machined disc is significant. It is estimated that each spot corresponds to about  $10^9$ – $10^{10}$  atoms of tritium. The total number of tritium atoms in the whole target works out to be  $\sim 10^{11}$ . If this is compared with the  $10^{19}$ – $10^{20}$  atoms of D absorbed in all by the Ti, targets it points to a gross (T/D) ratio of  $\sim 10^{-9}$ . But if only the D present in the “hot spot zones” are considered, the corresponding (T/D) ratio would be significantly higher.

It is noteworthy that these autoradiographs are distinctly spotty as against those of the aged targets discussed earlier which were uniform. This difference may be attributed to the fact that the above Ti targets were subject to severe cold working causing defect sites during fabrication (prior to loading) whereas the aged targets were produced through vacuum deposition of titanium thin films onto a copper backing with no cold work involved.

#### 2.4. TiDx shavings subject to thermal shock caused by liquid nitrogen [9]

In the experiments described below, anomalous generation of tritium when deuterated Ti lathe turnings (or chips) were subject to a thermal shock following dunking in liquid nitrogen (LN) was investigated, in a variation of the Frascati type experiment wherein the production of sharp bursts of neutrons had earlier been reported in similar experiments. Prior researchers had made attempts to measure tritium production by degassing the thermally shocked chips and recombining the released gases to form tritiated water. In contrast, in our experiments, the surface tritium content in individual TiDx chips following LN treatment, was measured directly. We speculated [10] that when a single sharp burst of say 100 neutrons is produced within a short duration of less than a 100  $\mu$ s from a cylinder containing thousands of chips, such a burst must most probably be emanating from a single chip. This is because it is very difficult to conceive of any physical process or mechanism by which several individual chips can interact (or communicate) with each other on this short a time-scale so as to generate a neutron-burst simultaneously (“like a symphony orchestra”). Based on the anomalously large ( $10^8$ ) tritium-to-neutron ratio observed in earlier CF experiments, both at BARC and elsewhere, we speculated that at least  $10^{10}$  tritium atoms ( $\sim 20$  Bq of activity) must be present in a few individual TiDx chips.

In the experiments reported here Titanium metal shavings or chips (a few mm long,  $\sim 1$  mm wide and 0.2–0.5 mm thickness) were machined out of a 20 mm diameter rod. A few thousands of these chips weighing about 10 g in all, were chemically cleaned with  $H_2SO_4$ , HCl, etc. before being charged into a stainless steel chamber. The chamber was then heated for 2 h at 800°C under a vacuum of  $10^{-5}$  torr by means of a resistance heater placed around it. During cooling of the chamber, when the temperature cooled down to around 600°C, deuterium gas was admitted. The total quantum of deuterium gas absorbed by the chips during further cooling was estimated from the pressure drop recorded by an oil manometer.

With the objective of causing thermal shock in the deuterated chips, they were dropped directly into liquid nitrogen contained in an aluminum cylinder. After complete evaporation of the LN, when the chips had almost attained room temperature, the container was again refilled; this cycle was repeated 4 or 5 times for each batch of chips. The chips were then analyzed for presence of tritium using a thin window NaI gamma detector. Lots of about 100 chips each from the LN treated and untreated stock of TiDx chips were uniformly spread on the top window of a vertically mounted NaI detector, in such a way as to minimize self shielding effects. While the counts with the untreated chips corresponded to background only, all the lots of the shock treated batch indicated activity above background. Through a process of sub-dividing the active lots into smaller batches and counting again, four highly active chips could be isolated from a

**Table 3.** Tritium activity in highly active Ti chips

Chip ID No <sup>1</sup>	NAI Counter <sup>2</sup>		Gas Counter <sup>2</sup>		Ratio ( $N_{\text{cps}}/B_{\text{cps}}$ )
	Count Rate ( $N_{\text{cps}}$ )	Tritium activity (MBq)	Count rate ( $B_{\text{cps}}$ )	Tritium activity (kBq)	
F6	0.12	0.02	475	4.75	$2.5 \times 10^{-4}$
F55	0.11	0.02	460	4.6	$2.5 \times 10^{-4}$
Z1	63.5	10.7	34665	346	$1.8 \times 10^{-3}$
Z2 <sup>3</sup>	9.7	1.7	–	–	
Z3 <sup>4</sup>	89.2	15.2	–	–	
Z4	140.9	23.9	42415	424	$3.3 \times 10^{-3}$

<sup>1</sup>Indicates a specific number assigned to each of the individually counted chips.

<sup>2</sup>Tabulated count rates are the maximum value averaged over several measurements, corrected for background.

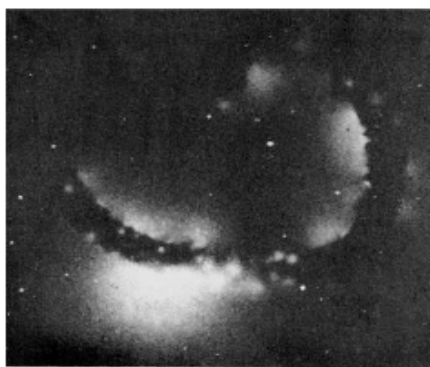
<sup>3</sup>Lost during analysis.

<sup>4</sup>Due to coil shape of this chip, it was not possible to fit in the drawer of the beta detector.

total of more than 1000 TiDx chips. The activity with different orientations of these four active chips varied by factors of 3–5 with the maximum count rates being in the range of 9.7–140.9 cps as listed in Table 3 (chip Nos Z1–Z4).

All the TiDx chips were again counted in lots of 15–20, by placing them inside the drawer of a windowless  $2\pi$  geometry gas flow type proportional  $\beta$  counter. The results of this gross counting were consistent with the NaI detector measurements. Some of the virgin metal shavings as well as the deuterated but untreated titanium chips were also counted in the detector but none of them gave any counts over the background. Other batches of TiDx chips, cycled later, also failed to show any detectable activity. The four ‘high’ activity chips were also counted individually with the windowless  $\beta$  detector, but in view of their very high activity there was considerable dead time counting losses. The statistical variation in the activity of single chips was studied by counting some of the chips individually from a few of the randomly selected active lots. Of the 66 chips so counted, at least 50% gave count rates more than twice the background.

A careful scrutiny of the X-ray-counts to  $\beta$ -counts ratio for the low and high activity chips, indicated a clear difference between them. Superimposed on the observation of the differences in shapes of the  $\beta$ -spectra between the high and low activity chips, we inferred that tritium in the low activity chips is closer to the surface than in the high activity chips leading to the conclusion that only four (or perhaps a few more chips) had actually “produced” tritium

**Figure 2.** Autoradiograph of chip # Z4.

during the thermal shock treatment whereas most of the other chips must have picked up the tritium as contamination during the LN treatment. The justification for this line of argument is spelt out in detail in our original paper [9].

Autoradiography of the four highly active chips was carried out with a view to gain some insight into the spatial distribution of the tritium on the surface. The test chip was sandwiched between two X-ray films to image the active zones on both sides of it. Fig. 2 which is the autoradiograph of chip No. Z4 for an exposure time of 17 h indicates the presence of about 35–40 hot spots. The radiograph of the other side also gave a similar image but with a lesser number of spots, implying different activity distributions on the two sides, a feature observed during  $\beta$  and X-ray counting also. The hot spots in all these radiographs are similar to those observed earlier at BARC in the autoradiographs of other deuterated titanium targets. However one remarkable feature of the autoradiographs of these chips worth highlighting is the so called “shadowgram effect” resulting in the outline of the chip boundary being clearly visible.

### 3. Plasma focus experiments [11]

In a plasma focus device a coaxial plasma gun (Fig. 3) is used to produce a dense hot ( $\approx 1$  keV temperature) “plasma focus” a few cm long and a few mm in diameter, just above the central electrode. The chamber is filled with deuterium gas at a pressure typically in the range of 1–20 mbar. The device is driven by a fast capacitor bank charged to 12 kV (stored energy of 2 kJ). When the spark gap switch is fired, a surface discharge is first initiated along the surface of the lower glass insulator which then develops into a radial current sheath that is accelerated by  $\mathbf{J} \times \mathbf{B}$  forces axially between the electrodes, sweeping and ‘snow plowing’ the gas ahead of it. On reaching the upper end of the central anode, the current sheath turns around on itself forming a quasi-cylindrical implosion, resulting in a dense hot “plasma focus” just above the electrode tip. The focus itself lasts for less than a microsecond, but before its disintegration it produces an intense burst of neutrons of a few tens of ns duration. The neutron yield in a PF device depends on the quality of focus formation and typically varies in the range of  $5 \times 10^5$ – $10^8$ , depending on the filling pressure and other factors. Since the fusion reactions result from internally accelerated deuterons impinging on a hot deuterium plasma “target”, it is basically a hot fusion (beam-target) process and as such roughly equal numbers of neutrons and tritium atoms are generated.

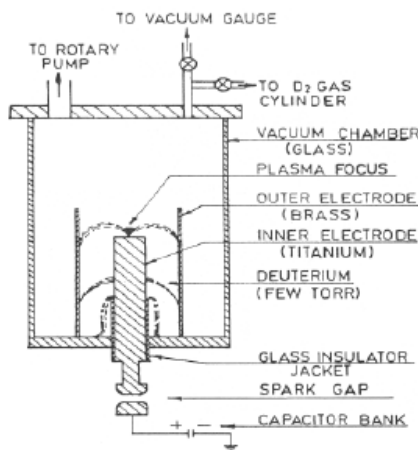
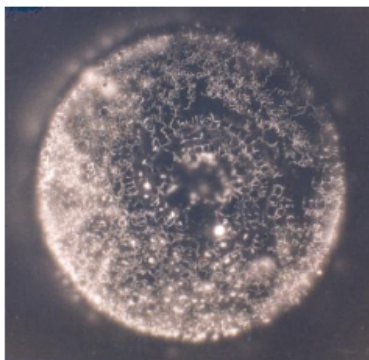


Figure 3. Schematic of a PF device.





**Figure 4.** Autoradiograph of Ti anode TA1.

In January 1990 as part of a program to study the influence of anode material on the characteristics of the formed plasma focus, experiments with a central Ti rod electrode were carried out. This first titanium electrode (TA1) was exposed to 50 charge/discharge shots at different deuterium gas filling pressures. The electrode was then removed and scanned using a NaI detector. It was found to contain a surprisingly large activity of more than 10 MBq ( $10^{16}$  atoms) of tritium. The overnight autoradiograph taken on 9 Feb. 1990 confirmed this and gave a truly spectacular image (Fig. 4). The image revealed a large number of randomly distributed tritium bearing hot spots. Two kinds of structures are discernible in the image : the first type is sharp worm-like lines which appear to be created by betas emanating from the surface. The second variety are diffused spots, which could be attributed to soft X-rays emanating from deeper layers.

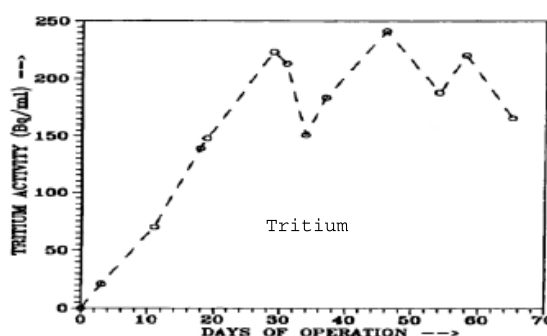
In order to ascertain whether positive voltage operation or negative voltage operation gives better results, two fresh titanium electrodes (TA2 & TA6) were exposed to 25 discharge shots each of opposite polarities. The reversed polarity shots were conducted following speculation that some of the energetic deuterons which are the cause of neutron production in a PF device may get directed towards the central electrode and get implanted in it. Both these anodes which were then scanned using the NaI detector. TA2 subjected to positive voltage shots gave 0.3 MBq while TA6 which was given negative voltage shots had 0.07 MBq. This was somewhat puzzling as one would have expected reverse polarity shots to give better results on the basis of improved deuteron implantation. The autoradiographs of these electrodes however indicated only faint images, confirming the presence of less tritium activity as compared to TA1 anode.

As mentioned already the 2 kJ Plasma Focus device typically produces  $10^7$  neutrons/shot. In the present experiments assuming 100 shots as upper limit, not more than  $10^9$  neutrons could therefore have been produced in all, with each electrode. Since the tritium-to-neutron branching ratio in conventional (d–d) reactions is unity, the same number, namely  $10^9$  tritium atoms would have been produced at the most as a result of the hot fusion reactions. Besides it is totally unrealistic to expect that all of this tritium would have succeeded in getting absorbed on the top surface of the central Ti electrode. Thus the  $10^{14}$  (TA2) to  $10^{16}$  (TA1) atoms of tritium measured on the end faces of the Ti anodes cannot be attributed to known PF type hot fusion processes. Besides the initial tritium content of the deuterium gas used in these experiments which was measured to be  $< 27$  Bq/l too cannot account for the large amounts of tritium measured on the top surface of these Titanium anodes. It is concluded that anomalous “cold fusion” type nuclear reactions would appear to be the most likely source of this high a level of tritium generation in these titanium electrodes.

### 3.1. Ni hydrogen electrolysis experiments [7–9]

Experiments were initiated with light-water electrolytic cells using Ni cathodes and  $K_2CO_3$  solutions as electrolyte, following reports of observation of excess heat in such systems by Mills and Kneizys [12] and other groups in the early 1990s. Although observation of excess heat in open electrolysis cells with Ni cathodes and  $K_2CO_3$  electrolyte was reported by us at ICCF 3 in Nagoya in 1992 [7], we subsequently came to the conclusion that the apparent excess heat reported at that time can be attributed to hydrogen-oxygen recombination effects within the open cell. We were, however, probably the first to report observation of tritium in light water electrolytic cells. At Nagoya, we reported that 18 out of 29 cells indicated tritium levels in the range of 46–3390 Bq/ml after a few weeks of cell operation. The high value of 3390 Bq/ml was produced in a cell in which  $D_2O$  was added to the extent of 25%, but three other identical cells with 25%  $D_2O$  did not give any tritium. One cell with enriched  $Li_2CO_3$  in  $H_2O$  (54% enriched  $^6Li$ ) gave 1454 Bq/ml, whereas two others with the same type of cathode and electrolyte gave no tritium whatsoever. Cell no. OM-3 with online sampling facility showed a steady increase of tritium level, reaching a value of 224 Bq/ml over four weeks; further operation of this cell however showed a saw tooth type variation of the tritium activity (see Fig. 5).

Some skepticism was expressed even within BARC circles regarding the reliability of low level tritium measurements in light water cells. Suspicion was voiced that the result could have been either from cross-contamination at the tritium measurements lab, where samples from different research groups were handled, or from “tritium in the BARC atmospheric air”. Hence, starting in June 1993, a completely new set of Ni light water cells were set up and operated at two separate laboratory sites, over a three month period. Six cells were set up at the Chemical Engineering Division, and 17 cells (five at a given time) at the Process Instrumentation Division, specifically for the purpose of verifying tritium production. This time, a new captive liquid scintillation counting facility was installed in the Chemical Engineering Division and extra caution observed to avoid any possible external contamination. All electrolyte samples were micro-distilled, with researchers taking adequate precautions to avoid alkali carry over before mixing with the scintillation cocktail for counting. With these procedures, tritium levels down to 0.5 Bq/ml could be measured. Details regarding these repeat measurements are presented in [7]. In all, 10 of 23 of the new cells showed low levels of tritium in the 0.5–4.8 Bq/ml range. In three of the Chemical Engineering Division cells, from which electrolyte samples could be drawn online without disturbing cell operation, an intriguing saw-tooth oscillatory variation of tritium levels of the electrolyte was observed. This observation, pointing out, with cross references, that such oscillatory behavior of tritium levels in electrolyte has been observed by other laboratories also in Pd- $D_2O$  cells are discussed in [7].



**Figure 5.** Variation of tritium activity in electrolyte of Ni cathode light water cell No. OM-3.

### 3.2. Tritium in H<sub>2</sub> gas loaded Ni wire experiments [8]

During 1994–1995, experiments were also conducted with self-heated thin Ni wires in a hydrogen gas atmosphere by subjecting the wires to several cycles of hydrogen absorption–desorption. The wires were then cut into 1 cm bits and analyzed for tritium content after dissolution. The results again showed, as in the case of titanium, a non-uniform production of tritium along the length of the wire, with some sections showing no tritium and others containing significant amounts; one bit had as much as 2,333 Bq. Details are given in [8].

## 4. Conclusions

This paper gives a brief overview of the variety of experiments conducted independently by many different groups at BARC during the period 1989–1996 wherein the anomalous generation of tritium in selected metallic deuterides/hydrides was confirmed. The experiments spanned both electrolysis studies as well as gas/plasma loading techniques in which Pd, Ti and Ni were used as the host metal. Production of tritium was confirmed both in Pd–D systems as well as Ni–H configurations, and in each case with both electrolysis as well as gas/plasma loading. The fact that all the dozen groups who set up electrolytic cells totally independently and confirmed finding tritium in the electrolyte, as well as in a variety of gas/plasma loaded target studies, is very significant.

Looking back, BARC can perhaps claim credit for being the *first* to report the following experimental findings:

- (1) Production of tritium in Pd–D<sub>2</sub>O electrolytic cells (Prof. Bockris has conceded this in private correspondence).
- (2) Tritium production is over a million times more probable than neutrons (so called “branching ratio anomaly”)
- (3) Generation of tritium in Ni–H<sub>2</sub>O electrolytic cells.
- (4) With machined Ti targets tritium production invariably occurs in isolated hot spots (first hints of NAE concept ?).
- (5) Use of a Plasma Focus device for loading and triggering anomalous tritium production.
- (6) Application of autoradiography as a very effective tool to record the spatial distribution of tritium in Titanium targets.
- (7) Lastly observation of neutron emission in “bunches” in a non Poissonian manner, leading to the speculation that micro-nuclear explosions may be occurring. This aspect is dealt with in detail in [10].

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