Observation of neutrons and tritium in the early BARC cold fusion experiments

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Soon after the Fleischmann-Pons announcement of 1989, a number of groups at the Bhabha Atomic Research Centre (BARC), Mumbai independently set up experiments to confirm if nuclear products such as neutrons and tritium are generated when samples of Pd and Ti metal are loaded with deuterium, using both electrolytic as well as gas/plasma-based loading techniques. Twelve independent research groups involving about 50 scientists were engaged in this massive effort during 1989–95, before institutional support for research on this topic was withdrawn, emulating global trends. This article briefly summarizes the results obtained during those early BARC studies.

Keywords: Cold fusion, electrolytic and gas loading, neutrons, tritium.

Introduction

A four-line news item on the historic Fleischmann-Pons (F&P) announcement carried by the 24 March 1989 issue of the Times of India, triggered a flurry of activity at the Bhabha Atomic Research Centre (BARC), Mumbai. Within days, groups having expertise in diverse disciplines such as hydriding of metals, electrochemistry, isotope exchange processes in upgrading heavy water, fusion plasma experiments and neutron and tritium measurements, set up a variety of electrolytic cells having widely different geometrical configurations with a view to verify the extraordinary claims of F&P. A few deuterium gas-loading experiments were also conducted. The objective of the onslaught was to establish whether the F&P effect had indeed any nuclear dimension to it. Since neutrons and tritium were the commonly expected products of fusion and other reactions such as deuteron stripping, these were the signatures that the BARC experiments sought to detect. The present article is a brief summary of the early BARC cold fusion results^{1–7}.

Electrolytic cells and neutron measurements

The Neutron Physics Division had a head start in the BARC cold fusion campaign because, by a remarkable

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coincidence, on 24 March 1989 a large cathode surface area (300 cm²) Pd–D₂O electrolytic cell with 5 M NaOD electrolyte, which was effectively a 'cold fusion cell', was all set and ready to be switched-on. This off-theshelf, 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 Company of Ireland for generating oxygen-free deuterium gas for plasma focus type 'hot fusion' experiments. On receipt of the F&P announcement, all that we had to do was to mount neutron monitors on either side of the cell and commence electrolysis. A NE 102A plastic scintillator-type fast neutron detector and a bank of three BF₃ counters embedded in a paraffin assembly constituted two independent foreground neutron monitors, while a similar He³ counter bank in paraffin served as background monitor. Figure 1 presents the first evidence of neutron production obtained on 21 April 1989. Both the foreground neutron monitors detected a dozen coincident peaks of varying magnitude while the background monitor counts were almost steady. (The semi-log scale of the Y-axis in Figure 1 actually deceptively suppresses the enormity of the peaks.)

One question that intrigued us even at that early stage was whether these neutrons were being generated one at a time in a random fashion following Poisson statistics or in bursts of 2, 5, 10 or even more, as in a spontaneous fission neutron source. Since the multiplicity distribution of neutron emission could throw much light on the mechanism responsible for neutron generation, a sophisticated



Figure 1. Neutron counts variation during run 1 of Milton Roy cell.

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statistical analysis of the neutron output from several cold fusion devices was carried out. The results and significance of these neutron multiplicity measurements are discussed in detail elsewhere⁸⁻¹¹.

Table 1 summarizes the details of eight electrolysis experiments in which both neutron and tritium production was observed. (The cell listed in the last column of Table was from the Indira Gandhi Centre for Atomic Research (IGCAR), Kalpakkam.) Six groups reported observing the first neutron emission on the first day itself. The neutron emission observed from the cells using Pd or Pd alloy cathodes in the 1989 experiments was generally in the form of spikes superimposed on a steady background. In contrast, the Ti cathode cell set up by the Desalination Division indicated continuous neutron production, in the range 50–100% above the background level^{2,4}.

On several occasions neutron emission was also observed from electrolytic cells, hours or days after the cell current had been switched off. Figure 2 depicts one such neutron output episode from a 'quiescent' Milton Roy cell.

Neutron measurements carried out in 1994 on a newly procured Milton Roy cell (Pd–Ag alloy cathodes) indicated¹⁰ a steady but low rate of neutron production ~9% above background levels throughout a one-month period, as shown in Figure 3. This cell, however, employed 1M LiOD as electrolyte rather than 5M NaOD used by the earlier Milton Roy cell. Neutron production by the new cell was also independently confirmed by the neutron multiplicity measurements carried out throughout the one-month period¹⁰.

Neutron emission in the form of distinct spikes above background level was also observed from an unperturbed titanium deuteride button just sitting on the table (Figure 4). Statistical analysis of the temporal characteristics of this event also was carried out. Emission of neutrons from a shutdown cell or unperturbed TiD button is analogous to the so called 'heat after death' episodes observed



Figure 2. Neutron output of quiescent Milton Roy cell: 16 June 1989.

during calorimetric measurements by the other researchers in the cold fusion field. What this basically means is that once these host metals are loaded with deuterium, nuclear reactions do seem to take place spontaneously even when there is no externally applied triggering mechanism.

Measurement of tritium levels in electrolyte samples

The tritium levels in the electrolyte samples collected after operating the cells for some time were measured by specialist groups of the Isotope as also the Health Physics Divisions of BARC using well-known liquid scintillation counting techniques applicable for low-energy beta emitters, taking adequate precautions to minimize chemiluminescence interference effects. Whenever warranted, micro-distillation of the test samples was carried out prior to addition of scintillation cocktail. The quantum of tritium observed in the electrolytes of 11 additional electrolytic cells, besides the eight cells of Table 1 in which



Figure 3. Steady neutron production by a new Milton Roy cell: 1994 results.



Figure 4. Neutron emission from an unperturbed TiD button.

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Table 1. Electrolysis experiments with neutron and tritium generation								
Division	HWD	NtPD	NtPD	DD	AnCD	ROMG	ROMG	IGCAR
Cell ID	Ti-SS	MR-1	MR-2	5 Module	PDC-1	RSC-11	RCS-19	RCP-II
Cathode	Ti rod	Pd–Ag tubes	Pd–Ag tubes	Pd-Ag discs	Pd tube	Pd cube	Pd pellet	Pd button
Area (cm ²)	104	300	300	78	5.9	6.0	5.7	8.0
Anode	SS pipe	Ni pipes	Ni pipes	Porous Ni	Pt Mesh	Pt mesh	Pt mesh	Pt mesh
Electrolyte	5M NaOD	5M NaOD	5M NaOD	5M NaOD	0.1M LiOD	0.1M LiOD	0.1M LiOD	0.1M LiOD
Switch on time (h)	~3	~5	~0.5	~4	~9	~24	~930	~300
Neutron yield	3.0×10^{7}	4.0×10^{7}	9.0×10^{7}	5.0×10^{6}	3.0×10^{6}	1.4×10^{6}	3.0×10^{6}	2.4×10^{6}
Tritium (T) yield (atoms)	1.4×10^{14}	8.0×10^{15}	1.9×10^{15}	4.0×10^{15}	7.2×10^{13}	6.7×10^{11}	4.0×10^{12}	3.5×10^{12}
(n/T) ratio	2.0×10^{-7}	0.5×10^{-8}	0.5×10^{-8}	1.2×10^{-9}	4.0×10^{-6}	1.7×10^{-6}	1.0×10^{-6}	7.0×10^{-7}

Fable 1. Electrolysis experiments with neutron and tritium generation



Figure 5. Concomitant generation of neutrons and tritium run 2 of Milton Roy cell.

neutrons too were measured, are summarized in table 2 of Iyengar and Srinivasan³. The quantum of tritium generated in the various BARC cells (22 cells in all) was in the range of a few kilobecquerel (kBq) to a few megabecquerel (MBq). The maximum tritium concentration attained in the electrolyte of any cell was 55.6 kBq/ml, corresponding to a total tritium yield of 13.9 MBq; this was observed in the large cathode area Milton Roy cell soon after the 21 April 1989 neutron emission event described earlier.

Note that while neutrons are measured on-line as and when they are emitted, tritium is measured offline as a cumulative post-run quantity by analysing electrolyte samples drawn periodically. Figure 5 which gives the time variation of neutrons and tritium observed in one of the BARC cells, suggests that neutrons and tritium are both probably generated concomitantly (in the same event). Most research groups collected electrolyte samples every few days and these invariably showed the presence of tritium only in samples collected after a neutron emission event was recorded.

The bottom-most row of Table 1, which summarizes results from those cells in which both neutrons and tritium were observed, displays the neutron to tritium yield ratio. This ratio is seen to be in the region of $\sim 10^{-7}$. Since the ratio for the two main branches of d–d fusion reaction is unity according to conventional nuclear physics data,

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in cold fusion devices this ratio is clearly off by seven orders of magnitude; this experimentally observed 'anomaly' has come to be known as the 'branching ratio anomaly' in cold fusion literature. BARC results were amongst the first to establish this anomaly as a characteristic signature of the physics of CF/LENR devices.

Studies on deuterium gas/plasma-loaded Ti samples⁷

Five different types of gas/plasma-loading approaches were adopted at BARC for loading deuterium into titanium samples. Presence of tritium, if any, on the surface of these samples was established subsequently both through autoradiography as well as direct counting of the beta particles emitted by the tritium as well as through the Ti K_{α} X-rays. A fortuitous characteristic of titanium is that the betas emitted by tritium which have energy up to 18 KeV are able to excite the 4.3 keV K_{α} X-ray line of Ti, which can be easily identified by normal X-ray counting techniques.

In the first approach¹² which used standard procedures for gas loading, titanium samples were thoroughly cleaned using acids, then vacuum annealed, degassed and then heated to 800°C using a resistance furnace and then allowed to cool in D₂ gas atmosphere. In the lathe shavings (or chips) experiment^{5,12}, deuterium loading was carried out by this procedure. In this experiment after loading, the deuterated chips were dropped directly into liquid nitrogen; subsequent measurements indicated presence of large amounts of tritium (tens of megabequerels) in four out of about 1000 chips. The autoradiograph of these four chips showed several remarkable hot spots which could be repeatedly radiographed over several months¹². Figure 6 shows the image provided by chip no. Z4, which was found to contain 424 kBq of tritium using a windowless flow-type proportional beta counter¹². The chips belonging to the control lot which were not loaded with deuterium nor treated in liquid nitrogen did not indicate presence of any tritium.

In the second approach, machined Ti electrode samples (discs and cones) were heated in deuterium gas using an

induction heater. Some of these subsequently gave characteristic spotty autoradiographs. Measurements using X-ray detectors confirmed the presence of tritium in the spotty regions, especially along the edges which displayed a ring of dots along the periphery in case of the disc and in case of a cone, an intense hot spot corresponding to the tip the cone.

The third method of deuterium loading was through a direct current plasma glow discharge between a pair of titanium electrodes, which leads to absorption of deuterium by the cathode.

The fourth method employed a high voltage (40 kV) discharge shot in a plasma focus device¹³, wherein a large Ti rod served as the central anode. In this method during the electrical discharge a hot and high-density deuterium 'plasma focus' is formed just above the top surface of the rod, following a Z-pinch initiated magnetic compression phenomenon in the gas. During this process the anode is subject to transient but intense magnetic and electric



Figure 6. Autoradiograph of a deuterated Ti chip showing tritium-containing hot spots.



Figure 7. Autoradiograph of Ti electrode of a plasma focus device after 50 discharge shots.

fields, and the anode top surface gets effectively cleaned resulting in efficient absorption of deuterium on the surface region, in spite of the fact that the rod is positively biased during the discharge transient. Since the electromagnetic fields last hardly for a microsecond, the anode rod which remains exposed to the gas in the chamber after each discharge shot, gets deuterated on the surface.

One Ti rod which was subjected to 50 such plasma focus shots was later found¹³ to contain 14.5 MBq of tritium which remained entrenched on the surface for several years subsequently. Figure 7 shows on autoradiograph of the top surface of this rod. Two other Ti anodes were also found to contain tritium after several discharge shots, but the magnitude of tritium yield was lesser. A detailed paper¹³ discusses the arguments as to why the tritium generated is attributable primarily to anomalous cold fusion-type fusion reactions and not hot fusion-type reactions which are known to occur during the focus formation phase but of a much lesser magnitude.

In the so called 'aged TiD targets' study³, anomalously large amounts of tritium (varying from a fraction to a couple of hundred megabecquerel) were detected on the surface of all 12 TiD targets which had been manufactured several (9-19) years earlier by loading deuterium onto thin Ti films coated on copper backings, for use as targets in accelerator experiments. The presence of tritium was confirmed through five different measurement techniques inclusive of autoradiography. Each one of the 12 'aged' targets gave an impressive coin-like circular image. Interestingly, the images were quite uniform, not spotty as in the deuterated, cold-worked Ti samples discussed earlier. We postulate that the tritium could have been produced by CF/LENR processes in the TiD thin films even while merely sitting in the drawer over the years. The possibility that tritium could have entered as contamination during the manufacturing stage of the TiD targets at the suppliers end (nine were from M/s Amersham Company, UK and three had been fabricated at BARC by the Isotope Division), has been ruled out after discussions with the manufacturers.

Conclusion

The fact that a dozen experimental groups set up cells using Pd samples lying about in the laboratory and yet detected neutrons and tritium within the first day itself is noteworthy. Table 1 which tabulates the 'switch on time' for each of the electrolysis experiments shows that six out of the 11 cells listed detected the first neutron signal within 9 h, and one within 24 h. However, all these cells stopped yielding any more neutron signals on continued electrolysis, indicating the crucial role of 'poisoning' effects during electrolysis, which has been noted by others too. Of course, there were several other cells which did not produce any neutrons or tritium at all. Thus like most other workers in the cold fusion field, the BARC groups also could not replicate the results.

BARC can claim credit for being the first to report the following experimental findings: (a) Production of tritium in Pd-D₂O electrolytic cells. (This has been finally conceded by Bockris of Texas A&M University, who was the other contender for this claim.) (b) Tritium production is over a million times more probable than neutrons (the so called 'branching ratio anomaly', where one of several possible explanations could be the Philips Oppenheimertype deuteron-stripping reactions). (c) Generation of tritium in Ni-H2O electrolytic cells (BARC work in Ni-H systems has, however, not been covered in this short article.) (d) With machined Ti targets tritium production invariably occurs in isolated hot spots (first hints of NAE concept?). (e) Use of a plasma focus device for loading and triggering anomalous tritium production. (f) Application of autoradiography as an effective tool to record the spatial distribution of tritium in titanium targets. (g) Lastly, observation of neutron emission in 'bunches' in a non-Poissonian manner, leading to the speculation that micro-nuclear explosions⁸⁻¹¹ may be occurring.

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