Srinivasan, M., et al. *Observation of Tritium in Gas/Plasma Loaded Titanium Samples*. in *Anomalous Nuclear Effects in Deuterium/Solid Systems, "AIP Conference Proceedings 228"*. 1990. Brigham Young Univ., Provo, UT: American Institute of Physics, New York.

OBSERVATION OF TRITIUM IN GAS/PLASMA LOADED TITANIUM SAMPLES

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ABSTRACT

The observation of significant neutron yield from gas loaded titanium samples at Frascati in April 1989 opened up an alternate pathway to the investigation of anomalous nuclear phenomena in deuterium/solid systems, complimenting the electrolytic approach. Since then atleast six different groups have successfully measured burst neutron emission from deuterated titanium shavings following the Frascati methodology, the special feature of which was the use of liquid nitrogen to create repeated thermal cycles resulting in the production of non-equilibrium conditions in the deuterated samples. At Trombay several variations of the gas loading procedure have been investigated including induction heating of single machined titanium targets in a glass chamber as well as use of a plasma focus device for deuteriding its central titanium electrode. Stemming from earlier observations both at BARC and elsewhere that tritium yield is 10^8 times higher than neutron output in cold fusion experiments, we have channelised our efforts to the search for tritium rather than neutrons. The presence of tritium in a variety gas/plasma loaded titanium samples has been established successfully through a direct measurement of the radiations emitted as a result of tritium decay, in contradistinction to other groups who have looked for tritium in the extracted gases. In some samples we have thus observed tritium levels of over 10 MBq with a corresponding (t/d) ratio of $> 10^{-5}$.

One of the most interesting findings to emerge out of the autoradiographic imaging of deuterated Ti samples from different experiments is the fact that tritium is invariably concentrated in highly localized spots (fraction of a millimeter or less in size) each containing typically about 10^{12} to 10^{14} atoms (2 to 200 K Bq) of tritium. If this is viewed in the light of the observations of other groups notably, the Los Alamos work, that neutrons are produced in bunches of 30 to 300 within time spans of microseconds and also that the neutron-to-tritium yield ratio is in the range of 10^{-8} to 10^{-9}, it is tempting to speculate that in these titanium samples perhaps some kind of a cascade reaction or micronuclear explosion probably occurs in specific sites in the near surface region

resulting in 10^{10} to 10^{12} fusion reactions during each event. This intriguing possibility warrants further experimental study.

I. INTRODUCTION

Jones and his collaborators at Brigham Young University (BYU), were possibly the first to employ titanium in place of palladium in cold fusion experiments and obtain direct experimental evidence for the occurrence of anomalous nuclear reactions in deuterated metal lattices[1]. They employed an electrolytic loading technique similar to that adopted by Fleischmann, Pons and Hawkins[2] in their Pd electrode experiments. It was however Scaramuzzi and others at Frascati[3] who first demonstrated that a simple high pressure gas loading procedure can be used equally successfully for deuteriding Ti for studying anomalous nuclear phenomena in deuterium/metal systems. A novel feature introduced by Scaramuzzi's group was the adoption of thermal cycling of the deuterated titanium pieces by means of liquid nitrogen cooling followed by warm up phases to create non-equilibrium conditions in the deuterated metal lattice. It was earlier pointed out both by Jones et al.[1] as well as Fleischmann et al.[2] that this helps occurrence of nuclear reactions by causing rapid diffusion and migration of the deuterium ions within the host metal matrix, Both the BYU and Frascati groups have reported the measurement of significant neutron output from deuterated Ti samples. Since then Menlove et al..[4] of Los Alamos in collaboration with Jones have achieved considerable success in carrying out a systematic study of burst neutron emission from deuterated Ti chips, during the warm up phase following cooling to liquid nitrogen temperatures. Their success is attributed not only to the deployment of improved and sophisticated neutron detection equipment but also the use of a larger quantity (up to 300 g) of Ti chips in each experimental bottle[5]. Burst neutron emission from D_2 gas loaded Ti shavings subjected to thermal cycling has also been reported by other groups from Japan[6-7], India[8] and China[9].

At Trombay, within weeks of the commencement of cold fusion research in April 1989, it became apparent that tritium yield was much more prolific than neutrons by several orders of magnitude, a result that was clearly at variance with the commonly accepted branching ratio value of unity in (d-d) reactions. The Trombay groups were amongst the first to report a figure of 10^{-8} for the neutron-to-tritium yield ratio at Karlsruhe as early as July 1989[10]. Bockris et al. of Texas A&M University had earlier reported finding[11] a similar number for this ratio at the Santa Fe Workshop on Cold Fusion Phenomena. Claytor et al.[12] of Los Alamos published a value of 10^{-9} for the neutron-to-tritium yield ratio in their “solid state cell” experiments. Influenced by these findings, right from the inception of gas loading experiments in our group, emphasis was placed on the detection and quantification of tritium levels in gas/plasma loaded Ti targets rather than on measuring neutron output.

In the present paper we start with a brief review of the early results[13-14] which were primarily of an exploratory nature and follow it up with a description of the more recent experiments[15] involving the direct counting of tritium from TiDx chips. The paper also discusses the observation of MBq levels of tritium on the end face of the central Ti electrode of a deuterium filled Plasma Focus device[16].

II. TECHNIQUES OF TRITIUM MEASUREMENT IN SOLID TITANIUM SAMPLES

Tritium which has a half life of ≈12.3 years emits a low energy β having a maximum energy of 18.6 KeV corresponding to an average energy of 5.7 KeV only. The range of the maximum energy β being as small as 9 mg/cm^2 (in medium Z materials), tritium located only within this depth can be detected through direct beta particle counting. Besides, since these low energy electrons are stopped in even the 'thin' windows of most nuclear particle detectors, for direct counting of tritium windowless detectors have to be employed. However one redeeming feature, at least in the case of titanium samples, is that the upper half of the β spectrum, is able to excite the characteristic Kα (4.5 KeV) and K β (4.9 KeV) X-rays of titanium. (Unfortunately this does not happen in Pd to excite whose characteristic K X-rays > 21 KeV of energy is required). The mean free path of 4.9 KeV soft X-rays in titanium metal being ~10 mg/cm^2 it is much higher than the range of beta particles and hence the tritium present in deeper layers of the titanium also can be detected through a measurement of its characteristic K X-rays. In fact the ratio of X-ray counts to β counts is a gross measure of the depth to which tritium is embedded within solid titanium samples. The details of the various experimental techniques that have been employed at Trombay to measure the tritium content in gas/plasma loaded cold fusion titanium targets is described briefly below.

II.A AUTORADIOGRAPHY

Autoradiography is a simple technique to establish the presence of radiation emitting zones and has been used very effectively to study the spatial distribution of near surface tritium in deuterated Ti (and Pd) cold fusion samples[17]. The technique is free from electromagnetic interference (pick ups and discharge pulses) as well as chemiluminiscence and quenching effects. 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 based samples the fogging is presumably due to the combined effect of tritium βs and the 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) and not the radiation arising from tritium decay, exposures were carried out with a stack of two films in contact with some of the test 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 somewhat uncertain due to the saturation (or non-linear) behavior of image formation in photographic films. Our experience regarding this is described in Reference (14). We have found that highly localized regions of tritium ("hot spots") containing ≈ 0.2 KBq of activity or 10^{11} atoms of tritium each can be barely detected through a 100 hr exposure.

II.B TITANIUM K X-RAY MEASUREMENTS

The quantum of tritium present in the near surface region of titanium (or TiDx) samples can be conveniently established through a measurement of the characteristic Kα

(4.5 KeV) and Kβ (4.9 KeV) X-rays excited in the Ti[14]. However since the number of X-ray photons produced per beta particle is hardly about 10^{-4}, this technique does not have high sensitivity. Whereas a high purity Germanium (HPGe) or a Lithium drifted Silicon (SiLi) surface barrier detector can clearly resolve Kα and Kβ peaks a NaI (T1) detector gives only a single broad peak encompassing both the X-rays. In the present experiments the following two X-ray detectors were used: (i) A 10 mm thick intrinsic planar germanium crystal (100 mm^2 surface area) with a 75 microm thick beryllium window and (ii) a 1 mm thick 25 mm diameter NaI (T1) crystal having a 125 μm thick Be window. The germanium detector which was coupled through a low noise cryogenic preamplifier to a multichannel analyser (MCA) has a resolution of 170 eV at ≈ 6 KeV X-ray energy. Typical counting time for a sample on this detector was a few tens of minutes. The system has an estimated detection threshold of 25 KBq (for 20 minute counts) and a counting efficiency of 0.7 cps/MBq.

The NaI detector has a comparatively poor energy resolution of about 55% at 6 KeV X-ray energy. The samples to be counted are placed directly on top of the vertically mounted detector. The system which is connected to a PC based MCA along with the associated electronics, was calibrated with a standardized (2800 ± 300 MBq) tritiated Ti (on copper backing) source and found to have a threshold sensitivity of about 7 KBq for 20 minute counting. Owing to its overall higher efficiency of detection (≈ 6 cps/MBq) and ease of use due to vertical mounting, it was the NaI detector that was mostly employed to count the samples while the germanium detector was used only to compare and confirm the values.

II.C BETA COUNTING WITH WINDOWLESS GAS COUNTER

A drawer type 2π geometry windowless gas flow proportional counter using commercial liquefied petroleum gas (cooking gas) as the counting medium[17] has been used to directly count the tritium βs emanating from the surface of the deuterated titanium samples. The counter has a single tungsten anode wire of 25 microm diameter strung horizontally across the chamber and is operated at its plateau region of ≈ 2900 V. The pulse output processed through a low noise preamplifier and spectroscopic linear amplifier is fed both to a personal computer (PC) based Multichannel Analyzer (MCA) and a counting scalar in parallel. The counting efficiency is found to depend not only on the gas flow rate (which determines the dynamic gas pressure within the chamber) but also on the depth of origin and hence energy of the betas emanating from the sample. The counting efficiency for a typical sample in the present experiment was approximately 10%. The background count rate which also depended on the gas flow rate, varied in the range of 50 ~ 100 cps. The estimated threshold for measurement of near surface tritium activity for typical gas loaded cold fusion titanium targets is ≈ 50 KBq, which is significantly lower than that of the NaI detector.

Table I inter compares the threshold sensitivity and counting efficiency of the techniques used for measurement of tritium levels in solid titanium targets.

It must however be pointed out here that the counting efficiency figures noted therein is only a representative value as it is extremely sensitive to the depth at which tritium is present within the samples.

Table I: Inter comparison of different tritium measurement techniques

Detector	Ge(Li)	NaI(T1)	$2\pi\beta$
Source of Data	Ref (14)	Ref (15)	Ref (15)
Background (cps)	0.1	0.5	60-80
Efficiency † cps/MBq	≈ 0.7	≈ 6	≈ 10^5
Threshold *	25KBq	7 KBq	50 Bq
Counting time for estimation (min)	20	20	5

† This is only a typical value.

* For 2σ above background; in case of $2\pi\beta$ detector for 10σ above Bkg.

III. BRIEF RESUME OF EARLIER TROMBAY RESULTS

Our early efforts at establishing anomalous tritium production in D_2 gas loaded titanium samples have been documented in References (13) and (14). These results are briefly summarised here for the sake of completeness and also to bring out the improvements effected in our group both in the techniques of deuteration of titanium as well as in the measurement of tritium levels.

III.A RF HEATED MACHINED TITANIUM TARGETS

Interestingly our initial attempts to load deuterium into machined titanium "electrode" samples were initiated in 1988[13] well before the dawn of the cold fusion era. These attempts were in connection with the investigation of vacuum spark devices which are a class of Z-pinch type plasma fusion configurations and had been under study in BARC for many years. When reports of neutron production from gas loaded titanium shavings carried out at Frascati were first received (in April-May of 1989), we pulled out some of the conical and disc shaped electrodes which had earlier been loaded with D_2 gas and attempts were made to look for the presence of radioactivity through the technique of autoradiography. One of the conical samples gave a sharp spot image repeatedly during five consecutive overnight exposures. Since the spot corresponded to the tip of the cone, we were convinced that some radiation was indeed emanating from the tip. This was subsequently confirmed by K X-ray counting. The technique used for loading deuterium into such electrode samples is briefly described below.

Machined and chemically cleaned targets of Ti were individually vacuum annealed at temperatures of up to 900°C in a glass chamber using a surrounding induction heating coil (1 to 2 MHz frequency, 3 to 6 KW power). The chamber was connected to a vacuum system as well as H_2 and D_2 gas bottles. Degassing was initially carried out at 900°C for several hours until a vacuum of 10^{-5} mm was maintained steadily. The targets were later exposed to H_2 atmosphere at a few mm of pressure at a temperature of ~600°C. The induction heater was then switched off and the target allowed to cool allowing H_2 to be absorbed 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 given with D_2 gas. Fall in pressure 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 3rd or 4th 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 characterizing this phenomenon is given by $\delta = \sqrt{(\rho/\pi\upsilon\mu)}$ where ρ and μ are the resistivity and permeability respectively of the work load and υ 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 of deuterium, the tritium content if any on the target surface was sought to be measured using the technique of autoradiography as well as K X-ray counting discussed earlier. During these early experiments windowless beta detectors were not available in our group. 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, an observation similar to that made by Menlove et al. also[4]. The best results were obtained from a few disc shaped buttons (10 mm dia × 2 mm thick) and a couple of conical targets. Each of these is estimated to have contained more than 10^{12} atoms (>2 KBq) of tritium. The autoradiographs of some of these disc and cone targets have been published in our earlier papers[10,13]. However the most interesting feature of those autoradiographs is its spotty nature indicating that the tritium is entrenched in isolated but individual regions of the titanium, a feature that has since been confirmed in the later experiments also. The quantity of tritium in each of these targets was roughly estimated to have been at least 10^{12} atoms or over a few KBq of activity.

III.B AGED DEUTERIUM TARGETS STUDY[14]

Several groups in BARC had procured a number of deuterated titanium targets on copper backing during 1972 to 1986 for dosimetry studies with accelerator based neutron sources. Thirteen such targets were available, nine procured from M/s. Amersham International of U.K. and the remaining from the Isotope Division of BARC. In view of various studies involving deuterated titanium targets it was conjectured that cold fusion reactions might have occurred in these “aged” targets over a period of 4 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 counted as well as autoradiographed for establishing the magnitude of tritium present in them employing the techniques described in Sec. 2. The details of the targets, measurements and results are described in Reference (14). 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 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 to 3.5×10^{-4}. For comparison the tritium activity of the

D_2O moderator of a CANDU type power reactor is at most 30 Ci/1 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 we have speculated that a plausible explanation for the unexpectedly high tritium levels in aged deuterated Ti targets could be the occurrence of cold fusion reactions.

One noteworthy feature of the autoradiographs of these targets however was that they did not reveal any spotty characteristics. Although some of the targets displayed some degree of non-uniformity in the tritium distribution, the characteristic spotty feature observed in all other solid titanium cold fusion targets was absent. We feel this could be because these targets were produced through vacuum deposition onto a copper backing rather than through machining involving cold working.

IV. GENERATION AND CHARACTERIZATION OF DEUTERIUM GAS

Deuterium gas used in the following experiments was prepared by electrolysis of heavy water in a Milton Roy electrolytic hydrogen generator using a 5M solution of sodium deuteroxide. The isotopic purity of the D_2O used was > 99.8 mole% with a tritium content of ~7 Bq/ml (t/d ratio $\approx 4 \times 10^{-14}$) as determined by liquid scintillation counting. The generated deuterium gas was collected in a metallic cylinder and analysed for its tritium content by first exchanging it over a hydrophobic Pt-C-PTFE catalyst[18], with the heavy water (from the same stock as used for its production) and then counting the liquid phase after equilibration. The tritium content of the gas was deduced from these counts using standard chemical equilibrium equations since the amounts of heavy water and D_2 gas employed for exchange along with the equilibrium constant were known. The tritium concentration in the generated gas was thus deduced to be in the region of 10 to 30 Bq/ml for different batches of gas generated. The gas used for loading Ti lathe turnings described in Sec. VI for example, had a tritium content of 10^{-8} Bq/1, This corresponds to a (t/d) isotopic ratio of 10^{-13} which is 5 times the value of 2×10^{-14}, that is expected in gas phase after accounting for the two-fold isotopic enrichment that occurs in the electrolyte during electrolysis, The slightly higher than expected value of (t/d) ratio is attributed to tritium “contamination” in the Pd cathodes of the Milton Roy electrolytic cell either left over from the cold fusion runs of June 1989[19] or generated in the cathode during the present run itself.

V. ANOMALOUS NUCLEAR EFFECTS IN THE CENTRAL TITANIUM ELECTRODE OF A PLASMA FOCUS DEVICE[16]

In a plasma focus device (described later) a coaxial plasma gun is used to produce a dense (10^{19} to 10^{20} ions/cm^3), hot (≈1 keV temperature) plasma “focus” a few cm long and a few mm in diameter just above the central electrode. The focus itself lasts for less than a microsecond, but before its disintegration (due to instability phenomena) it produces an intense burst of neutrons of hardly a few tens of ns duration, when the device is operated with deuterium gas. The mechanism of neutron production in plasma focus devices has been the subject of investigations in several laboratories of the world since the discovery of the plasma focus effect in 1960. But it is now well established that neutron production in these devices is primarily due to beam target mechanisms caused

by deuterons accelerated within the plasma region itself to energies of a few hundred keV and which get trapped by the self generated magnetic field of the pinch current. In other words the anomalously high fusion reactivity of the plasma in such a device is due to the highly non-maxwellian nature of its ion (or deuteron) energy distribution[20].

In January 1990 an experimental program was underway at BARC, 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, aluminum, stainless steel, tungsten and titanium were investigated for neutron yield systematics under various deuterium filling pressures (1-10 mbar). An unusually high amount of tritium was found on the end face of the central titanium electrode which was exposed to the plasma during the above studies leading to a systematic investigation of the use of a plasma focus device for the study of anomalous nuclear effects in deuterated titanium.

V.A DESCRIPTION AND OPERATION OF PF DEVICE WITH Ti ELECTRODE

A Plasma Focus device consists of a set of coaxial cylindrical electrodes separated by an insulator at the bottom end and left open at the top, the entire assembly being housed inside a vacuum chamber[21]. Fig. 1 shows a schematic diagram of the plasma focus device used in the present experiments. The vacuum chamber is 12.7 cm in diameter and 16.5 cm long. The outer electrode configuration is of squirrel cage type. The central electrode is of 10 cm length and 2.2 cm diameter. Each of the twelve outer electrodes is 11 cm long and 0.8 cm in diameter, placed at a distance of 4 cm from the centre of the inner electrode. The glass insulator used is 4 cm long. The PF chamber is filled with deuterium gas at a pressure typically in the range of 1 to 20 mbar.

The device is driven by a 28 μF low inductance (100 nH at focus formation) 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 length of the insulator. This then develops into a radial current sheath which is accelerated thereafter by JxB forces axially between the electrodes, sweeping and ‘snow plowing’ 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” just above the tip of the central electrode as noted earlier. The time resolved (dI/dt) and voltage which are recorded using a Rogowsky coil and a voltage divider respectively on separate storage oscilloscopes during each shot, indicate the successful formation or otherwise of a healthy “plasma focus” during the discharge.

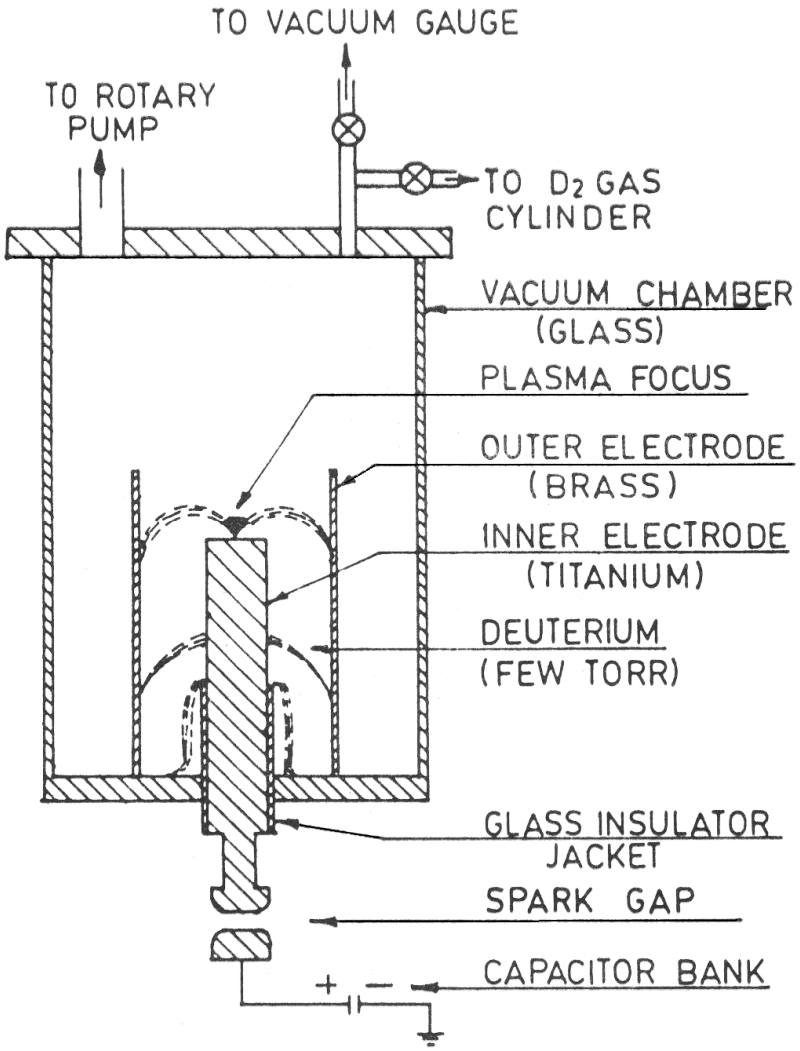

Fig. 1. Schematic Diagram of a Plasma Focus Device With Central Ti Electrode

The neutron yield in a PF device depends on the quality of focus formation. Usually the neutrons are produced in one or at times two sharp bursts of a few tens of ns width but the whole emission occurs well within a micro second. In the present experiments the neutron yield in each shot was measured with the help of a set of calibrated activation type silver cathode Geiger Muller (GM) counters embedded in paraffin and located close to the device. The GM counters count the 23 sec beta activity induced in their silver cathodes by the moderated neutrons. This set-up has a threshold sensitivity of 10^5 neutrons/burst. The neutron yield per shot typically varies in the range of 5×10^5 to 10^8 depending on the filling pressure.

The first experiment with a central Ti rod electrode (TA1) was carried out on the 4th of January 1990. This titanium electrode was initially exposed to 50 charge/discharge shots at different deuterium gas filling pressures with the PF operated in the normal mode, namely with the bank charged to positive voltage (12 kV). Subsequently around 10 shots were taken with the bank charged to negative voltage (-12 kV). The reversed polarity shots were given with the hope that some of the energetic deuterons which are the cause

of neutron production in a PF device may get deflected towards the central electrode and get implanted in it. The second Ti electrode (TA2) was subjected to 25 normal shots at a charging voltage of 12 KV while the third electrode (TA6) was given 25 shots of reversed polarity (-12 KV). A titanium electrode with a central depression (12 mm diameter, 35 mm deep) was also investigated.

V.B TRITIUM LEVELS ON TITANIUM ELECTRODES

Following the first experiment with a Ti anode (TA1), an attempt was made to look for the presence of tritium activity on the tip of the electrode through autoradiography on the same day (4th January 1990) as the plasma focus shots. Unfortunately this gave no results and the experiment was considered a failure and hence plasma focus shots with titanium electrodes were discontinued. However five weeks later (on 9th February 1990) when the anode was scanned using a newly received NaI detector, the titanium anode TA1 was found to contain a surprisingly large activity of more than 10 MBq (10^{16} atoms) of tritium. The overnight autoradiograph taken that night revealed a truly spectacular image displaying many detailed features (See Fig. 2). This result prompted us to resume plasma focus shots with titanium electrodes once again. (We have since found that the reason for the autoradiograph of TA1 not having been recorded on the day of the shots itself was a fault in the developer used during the processing of the X-ray film after exposure).

In order to ascertain whether positive voltage operation or negative voltage operation gives better results, titanium electrodes TA2 and TA6 were given 25 shots each of different polarities. Both these anodes which were counted using the NaI detector immediately after the shots were found to contain significant activity. 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 to us as one would have expected reverse polarity shots to give better results on the basis of improved deuteron implantation. The autoradiographs of TA2 and TA6 taken on the same day as the shots however indicated only a faint image. These autoradiographs are not as impressive as that of TA1, on account of less activity on them. A fourth titanium anode having a depression in the central part which was given over 50 positive and negative shots did not indicate any measurable activity. Table II summarizes the results of the tritium activity measurements on the three solid titanium electrodes studied so far. A tritiated titanium target of known activity (2800 ± 300 MBq) was used as reference for calibration. The error bar on the activity of the tritiated target is due to the uncertainty associated with of quantity of tritium gas absorbed by it, as indicated by its supplier.

Unfortunately these titanium electrodes were too large to fit inside the windowless 2π geometry β counter. Hence neither the β spectrum nor the X-ray to β counts ratio could be measured to get an idea of the depth to which the tritium is embedded. To overcome this limitation a large number of plasma focus experiments were carried out using ~ 1 mm thick Ti planchets (which could fit into the drawer of the β detector) mounted on the top of a standard brass electrode. To our disappointment none of these Ti planchets indicated any tritium activity in spite of many shots being given. However one puzzling feature was that one of these Ti planchets did seem to continue to produce-neutrons for several seconds after the discharge as indicated by the time variation of the activity of the silver GM counters. This phenomenon is under study more systematically.

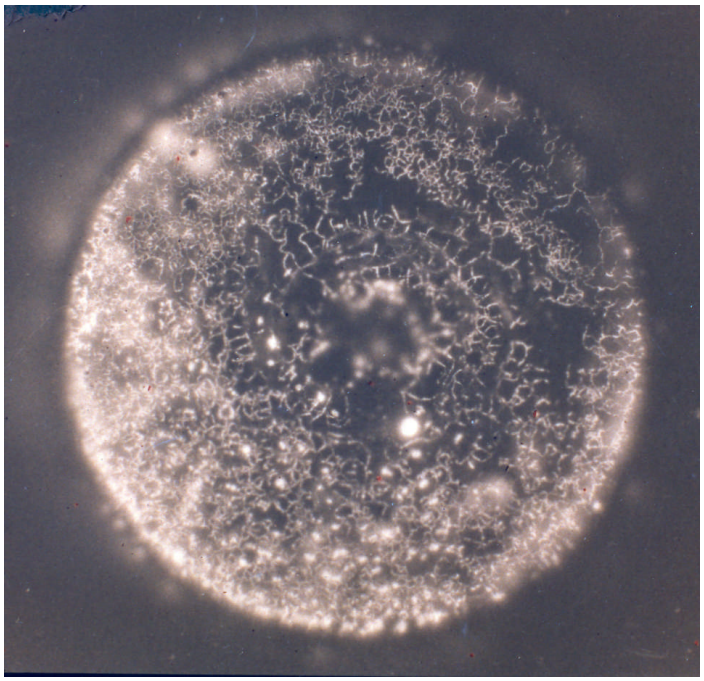

Fig. 2. Autoradiograph of central Ti electrode of PF device.

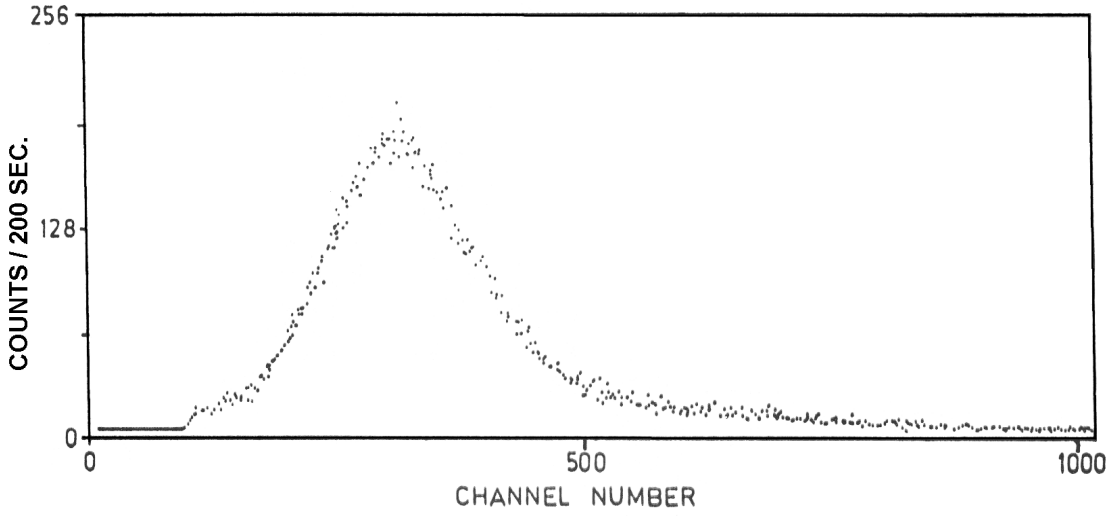

Fig. 3. Typical x-ray spectrum of chip #Z4 on NaI detector.

Table II: Tritium activity on titanium rod electrodes of PF device

Electrodes	TA1	TA2	TA6
Date of PF Shots *	4th Jan.	24th May	4th June
No. of Shots (+ ve)	50	25	-------
No. of Shots (- ve)	10	-------	25
Date of Measurement*	21st Feb.	24th May	4th June
Counting Time (s)	300	300	300
Count Rate with Std. Source ‡ (cps)	6216	6290	11269
Count Rate with Electrodes (cps)	33.3	2.3	1.8
Background (cps)	1.2	1.6	1.5
Tritium Act. (MBq)	14.5±3.3	0.3±0.06	0.07±0.02

* All dates are in the year 1990.
‡ Standard source activity is (2800 ± 300) MBq.

V.C RESULTS, DISCUSSION AND CONCLUSIONS FROM PF STUDIES

While shots with most electrodes produced neutrons, the yield per shot being on an average 10^7, tritium has been observed so far only on three solid titanium electrodes, with TA1 indicating the highest activity (>10 MBq). The deuterium gas used for TA1 was procured from M/S Matheson Company of USA about 8 years back. Unfortunately the deuterium gas of this cylinder was exhausted before tritium activity was detected from TA1 electrode and as such this gas could not be tested for possible tritium contamination. The deuterium used for TA2, TA6 and other electrodes were generated locally by the electrolysis of D_2O as described in Sec. IV, The tritium content of this gas was measured to be < 27 Bq/l.

The autoradiograph of TA1 electrode (see Fig. 2) has been reproduced several times since February 1990 through repeated exposures. Surprisingly there was no noticeable change in the resolution or clarity of the image even after a gap of one year. The image reveals a large number of randomly distributed spots indicating the presence of localized tritium bearing sites. 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.

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 (or rather beam target) fusion reactions. It is totally unrealistic to expect that all of

this tritium would have succeeded in getting absorbed in the central Ti electrode. Even if that were true, it still cannot explain the 10^{14} (TA2) to 10^{16} (TA1) atoms of tritium measured on their end faces, in the present series of experiments. Thus anomalous nuclear reactions would appear to be the most likely source of this high a level of tritium generation in these titanium electrodes.

VI. TRITIUM LEVELS IN TiDx CHIPS CYCLED IN LIQUID NITROGEN

In this section we describe the direct measurement of tritium activity in individual TiDx chips (lathe turnings) in a variation of the Frascati type experiment. Such experiments involving the use of liquid nitrogen (LN) cooling cycles to create non-equilibrium conditions have been successfully repeated by many other groups also, notably Menlove, Jones and their associates. Although these groups mainly concentrated on the study of burst neutron yield characteristics, some of them did attempt to measure tritium production also by degassing the chips and recombining them to form tritiated water. In contrast our emphasis has been on investigating the tritium content in individual TiDx chips following LN treatment, We have speculated that when a single sharp burst of say 100 neutrons is produced within a short duration of less than a hundred microseconds from a cylinder containing thousands of chips then 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 to generate a neutron-burst simultaneously ("like a symphony orchestra"). If now one were to assume that each neutron is accompanied by ~10^8 tritium atoms (as discussed in Sec. I) then we are obliged to come to the conclusion that >10^{10} atoms (or >20 Bq) of tritium activity must be present in a few individual TiDx chips. While this low a level of tritium activity may be somewhat difficult to measure experimentally, tritium levels in the range of 200 Bq have been detected in our laboratory (see Sec. II). In the following we describe the details of the experiments conducted at Trombay using a variation of the Frascati type experiment to verify the above speculations.

VI.A PREPARATION AND DEUTERATION OF TITANIUM CHIPS

Titanium chips were carefully machined from a 20 mm diameter metallic rod- procured from M/s Mishra Dhatu Nigam Ltd, Hyderabad (India). Typical length of such a chip was a few mm with a width of about 1 mm, while the thickness varied between 0.2 to 0.5 mm. A few thousand of these chips weighing 10 grams, were chemically cleaned with H_2SO_4, HCl etc. before being loaded into a stainless steel chamber. The chamber was heated for two hours at 800°C in a vacuum of 10^{-5} torr by means of a resistance heater placed around it. During cooling of the chamber, when the temperature reached around 600°C deuterium gas was introduced into it. The quantity of deuterium absorbed by the chips was estimated from the drop in gas pressure, the chamber volume being known (1.051). Although the gross (d/Ti) atomic ratio for the entire stock of chips was thus found to be 0.05 only it is likely that some of the chips had a deuterium loading much greater than this average value. Using a similar loading procedure hydrogen loading was also carried out (for a control experiment) having a p/Ti ratio of 0.065. Using an improved loading

procedure recommended by the Las Alamos group a much higher loading ratio of d/Ti ≈ 1.6 has recently been obtained for a new set of experiments.

VL.B LN TREATMENT AND NEUTRON DETECTION

The deuterated titanium chips were dropped directly into liquid nitrogen contained in an aluminium cylinder, 30 mm in diameter and ≈200 mm high placed at the centre of a neutron counting assembly consisting of 10 BF_3 detectors connected in parallel and embedded in polyethylene moderator blocks. There was in addition one plastic fast neutron detector close to the BF_3 counters and a second plastic detector placed further away serving as a background monitor. About 10 minutes after complete evaporation of the LN, when the chips had almost attained room temperature, the container was refilled. This cycle was repeated 4 or 5 times for each batch of chips.

Out of the total stock of deuterated chips available, a bunch of about 100 chips were cycled 5 times in liquid nitrogen on 11 May 1990. The BF_3 assembly registered a large spike which could imply a burst of ~10^4 neutrons in a 5 s counting interval in the first LN cycle itself but due to lack of confirmatory evidence it could not be attributed to a genuine neutron burst. A few days later, the experiment was repeated again with approximately ≈1000 deuterated chips from the original unchilled stock. The previously treated chips were also mixed with it subsequently (in hindsight we realize this was an unwise step) and this “major” batch was further cycled in LN a few more times. No observable neutron bursts were observed. Three more batches containing a smaller number of TiDx chips (less than hundred each) from the same stock, were later similarly processed in liquid nitrogen. No neutron detectors were employed in these cycles as the emphasis in these experiments was to look for tritium.

VI.C MEASUREMENT OF TRITIUM LEVELS IN TiDx CHIPS

Lots of about 100 chips from the LN treated and untreated stock of TiDx chips were uniformly spread on the window of the NaI detector in way to minimize shadowing effects. While the counts with the untreated chips corresponded to background only, all the lots of the major batch were seen to possess measurable activity above background (See Table III).

Table III: Gross X-ray counts of batches of TiDx chips with NaI detector

Group #	With Chips‡ (cps)	Background‡ (cps)
	Major Batch	
A	8.17 (Zl)*	1.80
B	3.74	1.80
C	6.15 (Z2)*	1.80
D	4.54	1.80
E	18.49 (Z3)*	1.80
E	2.30	0.56
F	2.91	1.80
G	3.51	1.80
I†	104.1 (Z4)*	1.80
I†	4.44	0.60
	Other Batches	
S1	0.76	0.98
S1(N)	0.83	0.92
V1	0.55	0.52
V1(N)	0.54	0.52
B1	0.51	0.52

‡ Deduced from 100 to 300 s counts.
# About 100 chips in each group.
* High activity chip present in group.
† Counts after removal of high activity chip.
N: After LN treatment.

Through the process of dividing the active lots into smaller batches and counting again, four highly active chips could be identified from this major batch of more than 1000 TiDx chips. Fig. 3 shows the pulse height spectrum of the K X-rays of one such highly active chip (# Z4) obtained with this detector. The activity with different orientations of these four active chips was observed to differ by factors of 3-5 with the maximum count rates being in the range of 9.7 to 140.9 cps as listed in Table IV (chip Nos Z1 to Z4).

All the TiDx chips were later counted in lots of 15 to 20 which could be accommodated inside the drawer of the windowless 2π geometry gas flow type β counter described in Sec. II.C. The results of the gross counting were consistent with the NaI detector measurements namely only the chips belonging to the major batch showed detectable activity. The four 'high' activity chips were also counted individually with this detector (see Table IV). These counts are however expected to be on the lower side on account of the considerable dead time losses at the high count rates encountered with the high activity chips. Pulse height spectra of a low and high activity chip as obtained with the β

detector, are shown in Figs. 4(a) and 4(b). Note the different scales of both X- and Y-axes in the two figures. The spectrum of the high activity chips (see Fig. 4(b) of #Z4) appears compressed towards the lower energy side, indicating a softer energy of the escaping β-particles. This implies that the tritium is located at greater depths in such chips.

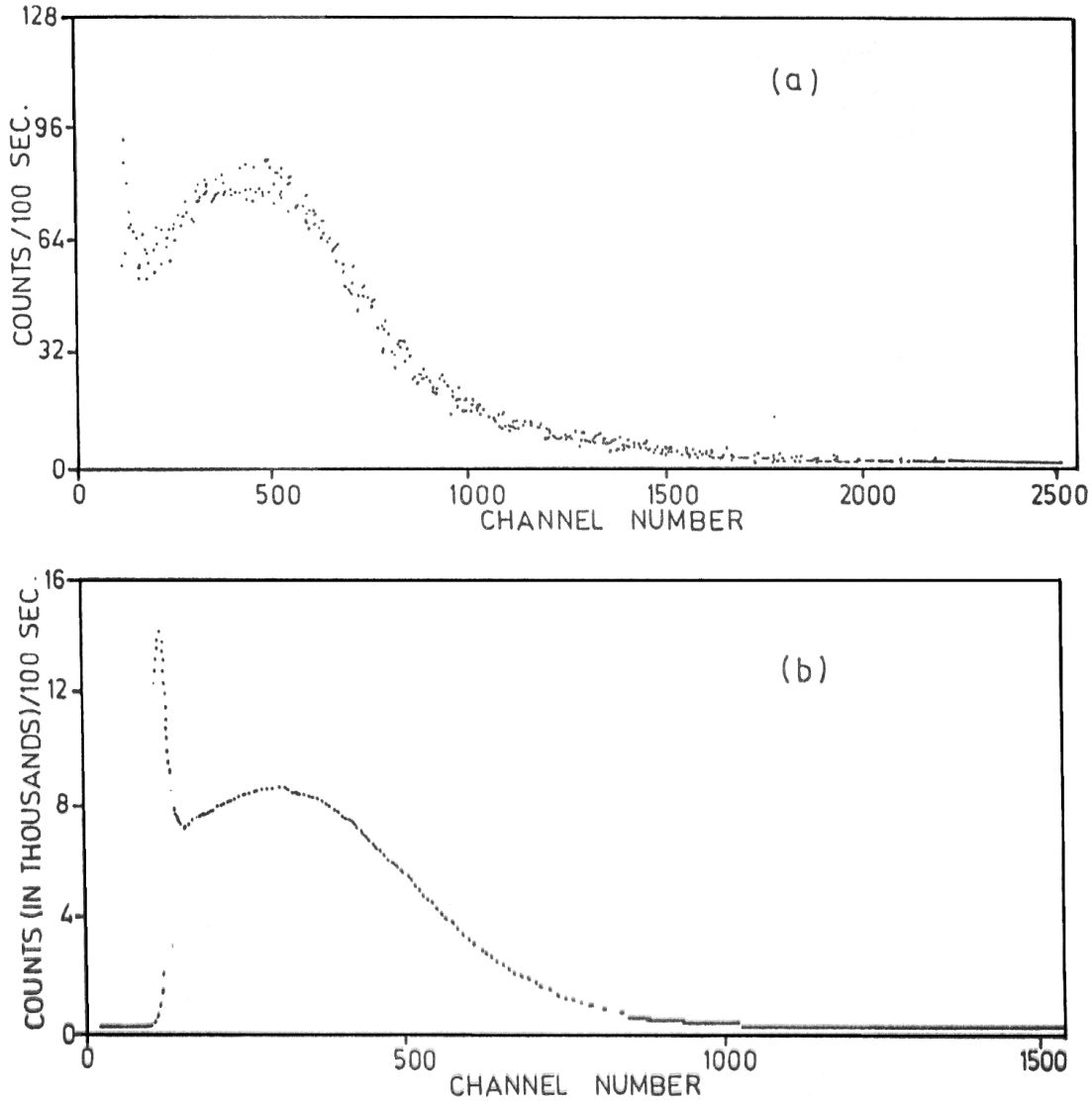

Fig. 4. Typical spectrum of TiDx chip number (a) F55 and (b) Z4, from the β detector.

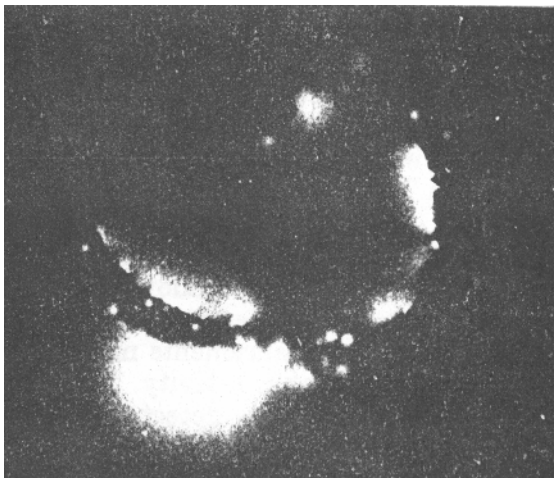

Fig. 5. Autoradiograph of chip number Z4 (9 Jan 1991).

Table IV. Tritium Levels in Individual TiDx chips

Chip I.D. No.[1]	NaI Counter[2] Count Rate (N_{cps})	NaI Counter[2] Tritium Activity (MBq)	Gas Counter[2] Count-Rate (B_{cps})	Gas Counter[2] Tritium Activity (KBq)	Ratio (N_{cps}/B_{cps})
F6	0.12	0.02	475	4.75	2.5×10^{-4}
F55	0.11	0.02	460	4.6	2.4×10^{-4}
Z1	63.5	10.7	34665	346	1.8×10^{-3}
Z2[3]	9.7	1.7	—	—	—
Z3[4]	89.2	15.2	—	—	—
Z4	140.9	23.9	42415	424	3.3×10^{-3}

1) Chip I.D. No indicates a specific no. assigned to each of the individually counted chips.
2) Tabulated counts are the maximum value averaged over several measurements, corrected for background.
3) The chip # Z2 was lost before β counting.
4) Due to coil shape of this chip, it was not possible to fit it in the drawer of the beta detector.

The statistical variation in the activity of single chips was studied[15] by counting all the chips individually from a few of the randomly selected active lots. Of the 66 chips so counted, at least 50% of them resulted in count rates of more than twice the background, Some of the metallic as well as the deuterated but untreated titanium chips were also counted in the detector but none of them (individually or together) gave any counts over the background. The minor batches of TiDx chips, cycled later, also failed to show any detectable activity.

Autoradiography of some of the active chips was then carried out with a view to gain some insight into the spatial distribution of the activity in them. The chip was sandwiched between two X-ray films to image the active zones on both sides of it. Figure 5 is an autoradiograph of one of the high activity chips for an exposure time of 17 h. The autoradiograph indicates the presence of about 35 to 40 hot spots. The radiograph of the other side also gave a similar picture but with a lesser number of spots, implying different activity distributions on the two sides, a feature observed during β and X-ray counting also. Autoradiography of the low activity chips with a long exposure time of ≈100 hours also results in a similar image but with fewer spots, The hot spots in all these radiographs are similar to those observed earlier in the autoradiographs of other deuterated titanium targets. However one remarkable feature of the autoradiographs of these chips worth noticing (see Fig. 5) is the so called "shadowgram effect" resulting in the outline of the chip boundary being clearly visible.

VI.D. DISCUSSION OF RESULTS

The last column of Table IV which gives the ratios of X-ray (Xcps) to β-count rates (Bcps) for a few chips, indicates a clear difference between the values of the low and high activity chips. The ratios for the high activity chips (#Z1 & Z4) are almost 7 times those of low activity chips (#F6 & F55). These are also comparable to the ratios encountered for high (S6-S9 etc) and low activity (S10-S12, also used as references here) Amersham targets14 which have X-ray to β-counts ratios of $\approx 10^{-3}$ and $\approx 10^{-4}$ respectively. The differences in these ratios as well as in the shapes of the β-spectra between the high and low activity chips, indirectly implies that tritium in the low activity chips is closer to the surface than in the high activity chips.

This led us to suspect that the low level activity observed in >50% of the chips of the major batch could have resulted due to the transfer of a small amount of tritium from the high activity chips during LN treatment. To investigate this possibility 'lots' of >50 chips each from the deuterated but inactive, as well as metallic titanium chips were first separately subjected to LN cycling a few times. Following this none of them showed any activity when checked with the NaI detector. The same lots were then treated in LN along with one of the high activity chips (#Z3) and on subsequent counting were surprisingly found to have become active. The gross activity estimated to have been picked up by the metallic and TiDx lots were about 30 KBq and 50 KBq respectively, thus confirming our suspicion that activity transfer does occur during LN treatment. There was no measurable loss in the activity of the high activity chip presumably because the amount of activity lost by it constitutes only a negligible fraction of its total initial activity. We therefore contend that only four or perhaps a few more chips had actually "produced" tritium while all the others that show low level activity must have "received" it from these successful chips when treated together in LN. The observation is very significant from the point of view of the "success rate" in such experiments which is seen to be much less than 1%. However we have no clue as to the mechanism of activity transfer between chips through the medium of liquid nitrogen.

Prior to LN treatment, the chips were not individually analysed for tritium activity in a systematic way to rule out presence of activity in any of them, However looking at all the possibilities as discussed in reference (15) it seems reasonable to conclude that tritium in

at least the four high activity chips was somehow "produced" within them and further in view of results, of other similar experiments, the LN treatment appears to be the most likely stage where the "generation" should have occurred. The conclusion is based on the fact that the total amount of tritium activity in the four high activity chips (a minimum of $\approx 10^{15}$ tritium atoms) is several orders of magnitude higher than the number of tritium atoms in the whole of the 1.05 l of D_2 gas (which contained $<10^{10}$ tritium atoms, as per the measured gas phase (t/d) ratio of $\approx 10^{-13}$) used for deuteration of the full stock of 10 g of Ti turnings. Hence even an assumption that the whole of the tritium from the gas might have somehow been preferentially absorbed in a few chips, fails to account for the measured high levels of tritium activity. Initial metallic phase contamination of a few chips, although not completely ruled out, also appears to be highly unlikely especially since the chips were degassed for 2 hours at 800°C.

VII OVERALL SUMMARY AND CONCLUSIONS

This paper has given an overview of the gas/plasma loaded titanium target experiments carried out at Trombay during the period May '89 to October '90 wherein the thrust was primarily towards the detection and measurement of anomalous tritium generation in deuterated titanium samples, Besides the commonly used procedure for deuteration of Ti metal shavings, two different methods have been adopted for "surface loading" of deuterium into Ti, namely induction heating of single machined Ti electrode targets such as discs and cones and plasma focus loading of the end face of its central Ti electrode. While the lathe shavings experiment followed a variation of the Frascati methodology of liquid nitrogen treatment for thermal cycling (the deuterated shavings were directly dipped into LN in our case) the other two experiments did not resort to any special treatment of the deuterated samples to induce non-equilibrium conditions in the metal lattice. However it must be remembered that in the latter two experiments the titanium metal surface was subjected to transient electromagnetic fields which may have had some "sloshing" effect on the deuterons within the Ti metal, possibly resulting in enhanced deuteron collisionality.

In all the different types of experiments conducted by us so far, several samples were found to have developed MBq levels of tritium. The presence of tritium was established through direct counting of the radiation (β and Ti K X-rays) emanating from the sample surface. Furthermore in all the "successful" titanium samples (except the vacuum coated thin film type "aged" deuterium targets) the tritium is found to be lodged in highly localized sites, a fraction of an mm or less in size, as seen through autoradiographic imaging. The real size of the titanium bearing site is obviously much less than its apparent size as seen on the radiographic image due to the finite grain size and resolving power limitation of the X-ray films. The quantum of tritium content in each of these 'hot spots' is deduced to be in the region of 10^{12} to 10^{14} atoms each. One of the remarkable features of all these titanium samples is the apparent stability of the tritium within the titanium metal lattice for periods of several months to a year (as evident from the reproducibility of the autoradiographs) in spite of being exposed to the humid atmosphere of Bombay. For example the radiograph of Fig. 5 was taken on 9th Jan '91, almost 8 months after LN treatment of the chip. This observation has an important bearing on the mobility (or rather lack of it) of tritium (and by implication deuterium also) within the titanium metal lattice. This perhaps explains the important role of artificially induced

non-equilibrium conditions in causing anomalous nuclear reactions in deuterated titanium samples.

The characteristic spotty nature which is a manifestation of the highly localized occurrence of tritium in clusters of 10^{12} to 10^{14} atoms each is very intriguing. Equally puzzling is the very small success rate of < 1% in the chips experiment. In other words what we learn from the Ti chips experiments is that not only do these anomalous nuclear reactions seem to occur in a very few chips, even in those successful chips it occurs in specific sites only.

While seeking a theoretical explanation for these observations we have to keep in view two other findings which have emerged from similar and related experiments, namely (a) TiDx chips subjected to LN cooling/warm up cycles have been found to produce bursts of 30 to 300 neutrons each in "microsecond time spans" and (b) that the neutron-to-tritium yield ratio in cold fusion reactions is in the range of 10^{-8} to 10^{-9}. This latter observation has been seen not only in electrolysis experiments but also in solid state cell measurements[12]. Similarly neutron bursts have also been observed in D_2O electrolysis experiments with Pd cathodes. If now we assume that the neutron bursts seen by Menlove et al. as well as at least six other groups in the world and the tritium in the spots observed by us are generated "simultaneously" (or concomitantly) all the above experimental observations seem to fit into a nice picture pointing towards the interesting speculation that each spot on the autoradiograph is possibly the result of one or more micronuclear explosions or cascade reactions wherein 10^{10} to 10^{12} fusion reactions occur within a very short time, most probably in less than a ns! The experimentally observed microsecond time scales is obviously imposed by the limitation of the neutron detection equipment used so far in such experiments namely thermal neutron detectors (He^3 or BF_3 type) embedded in a polyethylene moderator assembly. If a sharp burst of neutrons of say < 1 ns width were to impinge on such a detection system, on account of the neutron slowing down time in the moderator assembly, the correlated neutron detection times will be stretched out to microsecond time spans.

The above microexplosion postulate is clearly at variance with one of the commonly floated mechanisms to explain the production of neutron bursts, namely fracto-fusion caused by fracture of the TiDx lattice. It is very difficult to reconcile the fracto-fusion hypothesis with a neutron-to-tritium yield ratio as small as 10^{-8}. We first speculated on the possible occurrence of microexplosions as early as in July '89 [10] but ourselves felt that perhaps it was too radical a concept and hence "conceded" subsequently that fracto-fusion may after all be the source of burst neutron emission[22]. But the fresh results from the LN cooled TiDx chips has prompted us to once again revive the microexplosion theory. The authors would therefore recommend that future measurements with TiDx chips should be designed to settle which of the two mechanisms is a truer description of the phenomena occurring in deuterated titanium chips. It may of course be argued that both phenomena may be taking place simultaneously but independently. This scenario is indeed possible provided the neutron-to-tritium ratio is not constant at 10^{-8} but varies from experiment to experiment. It is also possible that the 5 MeV tritons observed by Cecil[23] and the 4-6 MeV energy neutron component observed by Takahashi et al.[24], both of which were reported for the first time at the present conference, may provide a clue to this puzzle.

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