BARC STUDIES IN COLD FUSION
(April – September 1989)

Edited by
P.K. Iyengar and M. Srinivasan

December 1989

BHABHA ATOMIC RESEARCH CENTRE
TROMBAY, BOMBAY
Typical Autoradiograph of a Deuterated Ti Disc Showing Regions of Tritium Concentration

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Acknowledgement
Energy production in the Universe is mostly based on nuclear reactions especially fusion reactions of light element nuclei. Energy production in the Sun on the basis of fusion of hydrogen, its isotopes and elements up to carbon have been well theorized by now. It is natural to expect that there will be a large variety of nuclear reactions which will lead to the production of nuclear energy. In fact there is a whole gamut of fusion reactions in astrophysics which suggest various combinations of nuclear interactions and modes of decay for energy production. The collapse of binary stars and the transformation of neutron stars into black holes are the ultimate phases of stellar evolution and production of fusion energy therefrom. Even before the discovery of the neutron, scientists had predicted and even tried to prove that nuclear energy could be generated through fusion of hydrogen nuclei (protons). It was however only after detailed accelerator based research in nuclear physics, that the cross sections and Q values for these reactions became available. This enabled many conjectures to be made. Some of the candidate reactions considered for the generation of fusion energy are: (p+p), (p+d), (d+d), (d+t) etc.

The familiarity of scientists with accelerator based nuclear reactions, however led them to believe that fusion reactions can take place only on the basis of overcoming the potential barrier caused by the electrostatic interaction. This demands that the particles have considerable relative velocity and from the analogy of what is happening in the Sun, thermonuclear fusion was considered as the most appropriate technique for releasing fusion energy on a large scale. We are all aware of the many experimental attempts that have been made over the last four decades to obtain conditions appropriate for thermonuclear fusion. The principle of confinement of these hot plasmas by means of complex magnetic fields in special configurations was invented in the early fifties. Of these the Tokamak has been a major success and has almost reached the stage of breakeven in energy production. However, the large size and the expensive equipment needed to attain even this breakeven stage have raised doubts about its commercial viability. The technique of generating temperatures of 100 million degrees using the principle of inertial confinement was first demonstrated in a thermonuclear bomb. The same principle has been borrowed and adapted in making fusion reactions possible in small pellets using lasers, electron beams and heavy ion beams. However even this approach of releasing nuclear energy through a series of fusion micro—explosions has not lived up to its early expectations as the power and energy of the driver beams for obtaining the requisite pellet energy gain became uncomfortably high. Small and more elegant methods are therefore being attempted. Techniques such as Z—pinches, combined magnetic and inertial confinement schemes etc are under experimentation. As an interim measure it has meanwhile been suggested that fusion devices may be employed as a source of neutrons for producing fissile fuel for use in fission reactors. Thus in quest of establishing the best method of producing fusion energy there has been considerable innovations and cross fertilization of new ideas during the last couple of decades. However new ideas are always welcome and must be tried out.

Cold fusion which we are discussing here is one such innovation which on the face of it looks so simple that it seems too good to be true. It has generated considerable speculation on the processes which cause fusion in the solid state at room temperatures. The basic problem is essentially to bring together ions of hydrogen isotopes at distances of a few Fermis so that fusion takes place. It is worth recalling here previous attempts to bring together hydrogen nuclei to distances at which the spontaneous fusing rate would increase considerably. The most effective method has been the replacement of the orbital electron of a molecular hydrogen ion by a μ meson or muon as it is called. Because of its heavier mass, the muon is able to squeeze the nuclei into a more compact molecule and cause a fusion reaction. Besides, the muon is found to have an additional advantage, because of its longer life time (2 μs), freed after a fusion reaction, it is able to catalyze
more fusions. Almost 200 catalyzed fusions per muon have been experimentally observed in (d–t) mixtures to date. It is some of the same scientists who were concerned with the physics of muon catalyzed fusion who have now reported cold fusion in a lattice of palladium.

From the point of view of understanding the physics behind cold fusion, one needs to discuss the lattice structure and its rearrangement when hydrogen is absorbed in palladium, titanium or other alloys used for the storage of hydrogen. It is the fact that enormous quantities of hydrogen can be stored in these materials at densities comparable to or higher than that of liquid hydrogen that first gave a clue that perhaps the internuclear distances can be brought down in such lattices. Many attempts have been made by theorists to evaluate the fusioning rate in such lattices. Some of these are based on an extension of the well known theory for muon catalyzed fusion, wherein both the internuclear distances and the height of the potential barrier are varied. Both will have the effect of increasing the fusion rate from $10^{-64}$ per second/per ion pair in heavy water to something of the order of $10^{-23}$ per second per ion pair which is required in order to explain the experimentally observed neutron production rate in cold fusion experiments. Whether it is possible to have such drastic changes in the fusion probability, which is essentially dictated by quantum mechanics considerations, is a matter of intense debate and discussion.

I would like to invite your attention to a novel application of the principles of quantum mechanics to such a problem. Several years ago Rand McNally had speculated on the feasibility of the occurrence of nuclear reactions at room temperature and was perhaps the first person to coin the phrase "cold fusion" as early as in 1983. To quote his own words "The problem of neutron transfer in solid media is no longer an elementary binary collision process involving Coulomb barriers and brief collision times but rather one in which the nuclei are continuously in each other's proximity. Since $^{135}$Xe has a slow neutron capture radius approaching that of the inter atomic distance the nuclear barrier would perhaps be grossly reduced. Thus it is remotely possible that some combination of natural processes may permit barrier penetration to occur much more readily and a nuclear reaction to ensue". He also proposed the term "de Broglie interaction length" to emphasize the fact that the de Broglie wave length of particles with small kinetic energy is very long. The importance of the de Broglie interaction length can be seen in the extraordinarily high cross section for absorption of slow neutrons by certain nuclei. It is therefore of interest to know what the de Broglie interaction length of a deuteron with very small energy is in a palladium lattice. Further, what happens to the charge distribution of such a deuteron extended in space and the effects of its polarization are too speculative. If the charge distribution has dimensions of the order of a de Broglie interaction length, then the potential barrier due to Coulomb interaction may perhaps become much smaller. If so then fusion should be much more probable at very low temperatures.

From the experimental point of view, the proof of cold fusion must come from a demonstration of the production of neutrons, $^3$He, $^4$He, tritium, gamma rays and other end products of nuclear transmutation reactions. Unfortunately experiments performed so far have used very small electrodes and small cells, and there have not been sufficiently large sized experiments which can give unqualified proof of the number of neutrons or radioactivity produced from this process. Our attempts in different groups at Trombay have however all shown reliable data on neutron and tritium production. These are described in the various papers included in this compilation. It is interesting that fusion reactions also take place when deuterium ions are introduced into a metallic lattice by simply absorbing deuterium gas into titanium or palladium. The group at Frascati in Italy first succeeded in producing neutrons by this method.
It is not our expectation that cold fusion will become an energy source tomorrow or the day after. After all even in 1939 when neutron induced fission in uranium was discovered, it took several years to find out how to set up a fission chain reaction and release fission energy in a large scale. Without detailed measurements of the number of neutrons produced in fission by the Columbia University group and the invention by Fermi of the heterogeneous neutron multiplying system, the nuclear reactor would not have become a reality. Similarly one has to explore and understand the basic mechanisms of fusion in a lattice and determine how this could be used either to produce energy directly or to produce neutrons and tritium in a sustained manner. It is too early to predict the time frame in which this will happen but for those of us familiar with the historical evolution of nuclear technology, one can foresee how it can change our perspective drastically. It is therefore necessary for us to involve ourselves deeply into understanding the mysteries of this new phenomenon. The source of such energy, namely deuterium, is ordinary water which is available in plenty and the technology to separate and concentrate deuterium from water is by now well established in our country.

I would therefore end this Preface by quoting from what Dr. Homi Bhabha said at the first International Conference on Peaceful Uses of Atomic Energy in 1955. He predicted that "in a couple of decades from now fusion energy will become possible and that will ensure energy production for man's needs as long as there is sea water on this earth". It is therefore a historical occasion for us to renew our efforts in research and development in an area so vital for human prosperity. I hope this report will stimulate the interest of scientists and engineers from various disciplines in this centre to channelise their efforts in such a way that we lead in this emerging technology.

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This Preface is an edited version of the Inaugural talk delivered by Dr. P.K. Iyengar, Director, BARC at the one day meeting on 'Cold Fusion' held at Trombay on 18th May 1989.
SUMMARY

This report is a compilation of the work carried out at BARC, Trombay during the first six months of the "cold fusion era" namely April to September 1989. Over fifty scientists and engineers besides a large number of technicians from more than ten Divisions of this Centre have been associated with these studies. This report comprises of three parts. Part A covers cold fusion investigations based on the electrolytic approach. Part B summarises the work based on D₂ loading in the gas phase and Part C covers the theoretical papers. Since most of the theoretical papers have either been published or are in the process of being published in scientific journals they are not commented upon in this Summary.

Electrolysis Experiments

Several groups having expertise in various areas such as hydriding of metals, electrochemistry, isotope exchange processes in the concentration of heavy water and neutron and tritium measurements, devised and set up a variety of electrolytic cells. In the initial experiments the emphasis was on detection of neutrons and tritium rather than excess heat. In a centre such as BARC the equipment and expertise for neutron measurements was readily available. The tritium levels in the D₂O electrolyte after operation of the cells were measured by expert groups (belonging to the Isotope and Health Physics Divisions of BARC) who have been engaged in this type of work for over two decades. Well known liquid scintillation counting techniques applicable for low energy beta emitters were used, taking adequate precautions as described in detail in Paper A9. Prior to the commencement of electrolysis, samples of the initial electrolyte also were analysed for their tritium content.

The first bursts of cold fusion neutrons were detected at Trombay on 21st April 1989 in two separate electrolysis experiments. At the Heavy Water Division a readily available commercial (Milton—Roy) electrolytic cell with Pd—Ag alloy tubes as cathodes and Ni as anode, originally meant for the generation of H₂ gas was adapted for the electrolysis of 5M NaOD in D₂O (see Paper A1). Both a bank of BF₃ detectors embedded in paraffin and a proton recoil fast neutron detector (NE 102A) were employed to look for possible neutron emission. An initial burst of neutrons was detected when the cell current attained 60 amps. Later the current increased to 100 amps and the cell became overheated, resulting in the built—in trip circuit automatically switching off the power. This was followed by a big burst of neutrons approximately two orders of magnitude larger than background levels during a five minute interval. Fig.2 of Paper A1 shows plots of the neutron counts data from the two detectors. From the efficiency of the neutron detection system measured using a standard Pu—Be source, it was surmised that ~ 4x10⁷ neutrons were emitted in all by this cell.

On the same day another cell set up by the Analytical Chemistry Division (ACD) also produced neutron bursts (see Fig.7 of paper A5). Table I presents a summary of the six successful electrolytic experiments conducted at Trombay wherein both neutron and tritium production has been confirmed. These cells have employed a variety of cathode—anode configurations. While some groups selected NaOD as electrolyte, others used LiOD. One of the successful cells has deployed titanium as cathode material. However many cells did not give positive results and these are not included in Table I.

The most important result to emerge out of the Trombay measurements is the surprisingly low overall neutron to tritium yield ratio. As evident from Table I five out of the six successful Trombay cells have given a value in the region of 10⁻⁶ to 10⁻⁹ for this ratio, while one (WCD) has given a comparatively large value of ~10⁻³ for this ratio.
However it must be pointed out that in this cell the absolute amount of tritium generated was very small. On the whole however the Trombay results clearly demonstrate that cold fusion is essentially "aneutronic" in nature.

It is worthwhile carefully scrutinising Table I to see what additional information can be extracted from these results. The current density in all the cells (except the last) was in the region of 170 to 800 mA/cm$^2$ and the total charge passed per cm$^2$ of cathode area prior to the detection of the first neutron burst (the "switching on charge" as it may be called) was in the range of 0.6 to 3.2 amp—hrs/cm$^2$. In the last cell however wherein the charging current density was only 60mA/cm$^2$ the switching on charge was an order of magnitude higher (34 amp—hrs/cm$^2$). Significantly however one ampere—hour or 3600 coulombs approximately corresponds to the charge carried by the number of deuterons required to load a few grams of Pd (associated with each cm$^2$ of cathode area) to a D/Pd ratio of ~0.6. Thus the fact that the switching on charge more or less corresponds to the defined deuterium loading requirement of the cathode is a noteworthy experimental finding.

Since cold fusion in electrolytic cells appears to be essentially a surface phenomenon the integrated neutron yield per cm$^2$ of cathode area may of interest. This quantity lies in the range of 1.7 to 2.9x10$^5$ neutrons/cm$^2$ for four of the cells. For the five module cell (Paper A2) it was an order of magnitude smaller while for the last cell it was two orders of magnitude higher. The specific tritium yield (µCi/cm$^2$) shows a somewhat wider scatter in the data. This quantity lies in the range of 0.5 to 1.3 µCi/cm$^2$ for the three cells wherein significant amounts of excess tritium were generated in comparison to the stock D$_2$O levels. In the case of the titanium cathode cell although the total amount of tritium generated was significant (7 µCi) the specific tritium yield is an order of magnitude smaller (0.07 µCi/cm$^2$). But it must be pointed out that neither the tritium lost via the gas stream nor that entrapped in the solid electrode were accounted for in that experiment. The specific tritium yield is much smaller for the last two cells wherein not much tritium was produced.

All in all it may be concluded that the behaviour of most of the electrolytic cells is reasonably consistent with each other considering the rather wide disparity in their designs, except perhaps for the last cell (WCD) whose behaviour is anomalous in many respects. The reason for this discrepancy is not immediately apparent.

Statistical Characteristics of Neutron Emission

There has been wide speculation in cold fusion literature that the observed neutron emission could be due to the phenomenon of crystal cracking or fracture. In the beginning it was even suspected that cosmic ray produced muons could be catalyzing fusion reactions in the deuterated Pd or Ti cathode. If these mechanisms were the source of cold fusion neutrons then it may be expected that neutron emission would be in multiples or bunches rather than one at a time. It is with a view to throwing some light on this question that the multiplicity spectrum of neutron emission was measured. The number of neutron pulses issuing from a bank of BF$_3$ (or He$^3$) counters (embedded in paraffin) monitoring the cold fusion source was totalled over 20 ms sampling intervals and stored in a personal computer. There were 1000 such sampling intervals during a real time of 5 minutes. Data was simultaneously also recorded from a paraffin encased bank of thermal neutron detectors placed at a distance of ~1.5 m from the source, serving as a background monitor. The data accumulated during periods of significant neutron emission were statistically analysed to yield the probability distribution of neutron counts. The details of these measurements are discussed in Paper A4. It is concluded from these studies that neutron emission essentially obeys Poisson distribution i.e. neutrons are mostly produced one at a time. However it is also found that occasionally 4 or more and even up to 20 neutron counts are registered in a single 20 ms interval. The background monitor has never yielded such high multiplicity
events even once. Careful analysis of these measurements leads to the conclusion that about 15 to 20% of the neutrons produced in cold fusion are due to bunched neutron emitting events, possibly from phenomena such as crystal cracking. But the bulk of the neutrons generated i.e. 80 to 85% obey Poisson statistics implying emission as singles.

Gas Loaded Titanium Target Experiments

Following reports of neutron emission having been detected by the Frascati group with pressurized D₂ gas loaded Ti shavings, two variants of this experiment were carried out at Trombay. One group (Chemistry Division) followed the Frascati procedure with ~20 g of cut Ti pieces and D₂ gas pressures increasing up to 50 bars. These experiments are described in Paper B1. The neutron detection system comprised of 24 He³ counters arranged in a well-like array and having a counting efficiency of ~10%. The neutron count rate reached a peak value of 10⁵/40s as compared to initial background levels of 60 per 40s (see Fig.2 of Paper B1). The neutron emission phase lasted for several hours at times.

In a second variation of the gas loading experiment, described in Papers B2 and B3, small machined targets (discs, cones, cylinders etc) of Ti metal (mass between 0.2 to 1 g) were individually loaded with D₂ gas by inductively heating them to 900°C in D₂ atmosphere at 1 Torr pressure and then switching off the power to the induction coil. D₂ gas was absorbed by the Ti target in the course of its cooling. The quantity of D₂ absorbed could be measured from the observed pressure drop. This corresponded to a gross (D/Ti) ratio of hardly 0.001. However it is believed that most of the absorbed D₂ gas was accumulated in the near surface region. A disc shaped Ti button loaded by this procedure began emitting neutrons on its own, almost 50 hours after loading, producing ~10⁶ neutrons over a 85 minute active phase (see Fig.1 of Paper A4). The background neutron counter did not show any increase in counts during this time.

A new result from the BARC work is the observation of tritium in D₂ gas loaded Pd foils, possibly for the first time. These are described in Paper B4. The tritium produced in these foils was extracted through a novel isotopic exchange technique, by keeping the deuterated foils in contact with a small quantity of ordinary water for several hours. The tritium content in the water was later measured through standard liquid scintillation techniques.

Autoradiography of Deuterated Ti and Pd Targets

An important outcome of the Trombay work is the demonstration of autoradiography as a simple and elegant technique of establishing the presence of tritium in metallic samples wherein cold fusion reactions are suspected to have taken place. Deuterated samples of Ti or Pd were placed over standard medical X-ray films and exposed overnight. The radiographs of Ti discs showed about a dozen intense spots randomly distributed within the disc boundary, besides a large number of smaller spots, especially all along the periphery forming a neat ring of dots (see Fig.1 of Paper B3). Repeated measurements with the same disc target with exposure times varying from 10 to 40 hours gave almost identical patterns of spots, indicating that the tritium containing regions were well entrenched in the face of the titanium lattice. Spectral analysis of the X-ray emissions from such targets using a Si (Li) detector clearly depicts the characteristic Kα (4.5 Kev) and Kβ (4.9 Kev) peaks of titanium excited by the betas from tritium decay.

In the case of Pd samples however the autoradiographs were more uniform. Interestingly it was found that the tritium betas (< 18 Kev energy) were unable to excite the characteristic K X-rays of the Pd. The fact that even the second film of a stack of X-ray films exposed to the deuterated Ti and Pd samples gave a similar image, rules out the possibility that the image formation could be due to pressure or chemical reduction.
effects as may be suspected.

Conclusions

Investigations of cold fusion phenomena carried out at Trombay during April to September 1989, have positively confirmed the occurrence of (d–d) fusion reactions in both electrolytic and gas loaded Pd and Ti metal lattices at ambient temperatures. Neutron emission has been observed at times even when the current to the electrolytic cell was switched off or in case of gas loaded Ti targets when no externally induced perturbation such as heating, cooling, evacuation etc was effected. The main findings of the Trombay investigations to date may be summarised as under:

(a) Tritium is the primary end product of cold fusion reactions with the neutron to tritium yield ratio being approximately $10^{-8}$. The variation of tritium content in electrolyte samples taken at frequent intervals in some cells supports the contention that neutron and tritium production occurs at the same time.

(b) Cold fusion in electrolytic cells appears to be predominantly a surface phenomenon, with a specific charge requirement of a little over an ampere–hour/cm$^2$ to switch on the neutron and tritium producing reactions. The fact that this roughly corresponds to the charge carried by the deuterons in a sample of Pd of a few grams/cm$^2$ loaded to a D/Pd ratio of 0.6 is particularly significant.

(c) Once initiated the cathode produces typically about $10^5$ neutrons/cm$^2$ and almost a µCi of T/cm$^2$, irrespective of the detailed nature of the cell design. It is imperative that we investigate the nature of the quenching mechanism that thwarts the ability of the cathode surface to sustain nuclear reactions.

(d) Neutron emission both from electrolytically loaded Pd and gas loaded Ti is basically Poisson in nature i.e. the neutrons are emitted one at a time. However it is not clear whether the neutrons are generated in the (d–d) fusion reaction itself or whether it is produced in a secondary reaction involving the energetic protons or tritons. In this context it would be of interest to look for the possible presence of $1^+_4$ ev neutrons in cold fusion experiments.

(e) About 10 to 25% of the neutrons produced appear to be generated in bunches of over 100 neutrons each within a time span of less than 20 ms. Viewed in the light of the neutron to tritium yield ratio of $10^{-8}$ noted above, one would be obliged to conclude that a cascade representing approximately $10^{10}$ fusion reactions occurs in under 20 ms. Since this appears very unlikely, lattice cracking or other mechanisms wherein the neutron to tritium branching ratio is close to unity, could be the source of such bunched neutron events.

(f) Autoradiography of gas loaded Ti and Pd targets demonstrates in a simple and elegant manner the presence of tritium. The experimentally deduced tritium to deuterium isotopic ratio in these targets is several orders of magnitude higher than in the initial stock D$_2$O and as such cannot be explained away on the basis of preferential absorption of tritium by the Ti or Pd as may be suspected. The existence of highly localized regions (hot spots) on the target surface as well as all along the periphery of the disc wherein tritium is concentrated, points to the important role of lattice defect–sites in the absorption process, at least in case of titanium.

(g) The very high probability for the tritium branch in cold (d–d) fusion reactions would indicate processes of neutron transfer across the potential barrier as postulated by Oppenheimer over half a century ago and elaborated on more recently by Rand McNally. If
neutron transfer as envisaged by these authors does take place so easily, it may have many implications for the future of nuclear technology, for the deuterium nuclide might very well do the work which free neutrons do in present day fission reactors. In the context of the emerging energy production scenario, aneutronic nuclear reactions such as this may give rise to new nuclear technologies providing a cleaner energy source for the twenty first century.

TABLE I
Summary of Successful Electrolytic Cell Experiments Conducted at Trombay

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*5 Surfaces considered

#Upto first neutron emission
PART A

ELECTROLYTIC CELL EXPERIMENTS
Introduction

The first reports of observation of 'Cold Fusion' during the electrolysis of heavy water\textsuperscript{1,2} using Pd cathodes, resulted in frantic attempts in several laboratories of the world to duplicate these experiments and if possible improve upon them. Electrolytic cold fusion investigations were initiated at Trombay in the first week of April '89 as a collaborative effort between the Heavy Water and Neutron Physics Divisions of BARC. A commercial (Milton Roy) diffusion type Pd—Ag cathode/ Ni anode hydrogen generator which was readily available was employed for this purpose, after loading NaOD as electrolyte in place of the original NaOH. This paper gives details of the electrolyser characteristics, conditions of operation and the neutron and tritium measurements.

The Electrolyser

The electrolyser employed was a diffusion type ultra—pure electrolytic hydrogen generator made by Milton Roy Company of Ireland\textsuperscript{3}. A schematic view of the electrolytic cell is shown in Fig.1a. The anode is of nickel and the cathode consists of specially activated palladium — silver alloy membrane tubes. The outer nickel body of the cell along with a central nickel pipe serve as coaxial anodes. The 16 cathode tubes are mounted with the help of PTFE spacers between these anode pipes as shown in Fig.1b and have a total wet surface area of \(2300\text{cm}^2\). The cathode tubes are sealed at the top and open at the bottom into a plenum through which the deuterium (or hydrogen) gas is drawn. The outlet of the gas plenum can be closed by means of a valve and the deuterium pressure allowed to build up. A pressure gauge reading upto 4 Kg/cm\(^2\) is provided to read this pressure. The important cell parameters are summarised in Table I.

The electrolyte used was 20\% NaOD in D\(_2\)O of \(>99.75\%\) isotopic purity and was prepared by reaction of moisture free Na with D\(_2\)O. The oxygen generated at the anodes during electrolysis escapes through a vent outlet at the top of the cell. A set of baffle plates are provided at the top to prevent alkali carryover into the vent. The hydrogen/deuterium ions which impinge on the cathodes under the influence of the applied electric potential, diffuse through the walls of the Pd—Ag tubes and escape into the gas plenum. The ions recombine inside the tubes to form molecular deuterium which is found to be of very high purity as analysed by gas chromatography. The electrolyte level in the cell is maintained with the help of a connected reservoir. The unit is completely automatic and is equipped with pressure control, a solenoid valve, electrolyte leak detector, low water level signal, temperature control etc.

The inbuilt power supply provided by the manufacturers is capable of giving a current of only \(\approx 70\) amps. During the initial runs only this was used and operation was restricted to comfortable current values of 60 to 62amps. An important feature of the cell design however is its potentially high current carrying capacity. The electrolyser was therefore connected to an external power supply capable of giving over 100 amps. during
subsequent runs. Current levels of ~100 amps were however found to be possible only for short durations as the cell was getting overheated. To overcome this, a heat exchanger and a peristaltic pump were incorporated enabling circulation of the electrolyte.

Neutron Monitoring

Initially there were two types of neutron detectors used to monitor the neutron yield from the electrolytic cell. The first was a bank of 3 BF$_3$ counters embedded in a paraffin moderator block. The second was a 80 mm dia x 80 mm high recoil type plastic scintillator NE102A sensitive, both to fast neutrons as well as high energy gammas. The detectors were mounted at a distance of about 10 cm from the cell. The counting efficiency of these detectors was established using a calibrated Pu—Be neutron source. The counting rates of the detector outputs were totalized for 5 minutes each and printed out continuously on a scroll printer. During the first run there was no separate background neutron monitor. In subsequent runs however a bank of three He$^3$ detectors, also embedded in a paraffin block, was installed about 1.5 m from the cell to serve as background monitor. Also a personal computer became available not only to display graphically the count rate variations but also to accumulate and store the counts data registered in 20 ms intervals, with a view to subsequently perform a neutron multiplicity distribution analysis as described in Ref 4.

In some of the more recent runs a bank of specially fabricated silver cathode GM tubes embedded in a paraffin slab was used as an activation detector for neutrons. This type of neutron detector is ideally suited to measure the yield of a burst of neutrons produced in a time span of about 10s or less. The neutron yield is deduced by counting the 24s half life of Ag$^{110}$ activity induced in the silver cathode of the GM tubes. The threshold sensitivity of the system for the geometry used was determined using a calibrated Pu—a—Be neutron source was ~3x10$^5$ neutrons.

Measurement of Tritium Levels in D$_2$O Electrolyte

Measurements of the absolute levels of tritium in the D$_2$O electrolyte were carried out by the Tritium Group of the Health Physics Division to whom samples were sent. Details of the liquid scintillation counting techniques employed along with precautions taken by them to minimize errors due to chemiluminiscence and other interference effects are discussed in Ref. 5. After the initial electrolysis runs a valve was welded to the cell bottom to enable periodic drawal of samples of the electrolytic solution for tritium assay.

Recently two microprocessor controlled on-line instruments for counting tritium activity deploying low energy sensitive scintillation fibres have been installed one in gas phase analysis and other for electrolytic solution counting. The development, testing and calibration of these instruments was carried out by the Pollution Monitoring Section of BARC. These instruments employ two photomultiplier tubes each in coincidence to suppress noise effects.

Electrolysis Experiments and Observations

Run No.1 (21st April 1989)

To start with the Milton Roy cell was operated with 20% NaOH in natural water as the electrolyte. This operation was carried out for about 48 hours for collecting background data. The cell was then drained, flushed with heavy water and filled with 20% NaOD solution in D$_2$O prior to commencement of experiments on 21st April 1989. The cell was operated initially at 30 amps and later the current was slowly raised to
60 amps corresponding to a current density of \(-200\, \text{mA/cm}^2\). After operation under these conditions for about 3 hours both the neutron counters started showing bursts of neutron counts, well above background values, during some of the 5 minute intervals. After a further couple of hours of operation both counting channels suddenly showed two very large peaks and at the time of the last peak, the current in the electrolyzer had suddenly increased to \(-120\, \text{amps}\) on its own and the electrolyzer immediately got tripped. Later it was found that the PVC insulation of the electric connections between the DC power supply and the electrolyzer had melted and even the soldering at the joints had melted. The diodes of the power supply had also burnt out causing the trip.

The neutron counts data of this run are presented in Table II and also shown plotted in Fig.2. The counting efficiency of the BF$_3$ Bank and Plastic scintillator were 0.06% and 0.4% respectively during this run. The fact that both the counters show identical behaviour in spite of having very different neutron detection characteristics is noteworthy. The total number of neutrons generated during the four hour duration of this run is estimated to have been approximately \(4 \times 10^7\). At the end of this experiment a sample of the electrolyte which was analyzed for tritium content indicated a level of \(1.5\, \mu\text{Ci/ml}\) of tritium activity in comparison to the initial stock heavy water value of \(0.075\, \mu\text{Ci/ml}\). As discussed later, this high build up by a factor of \(-20,000\), is far beyond what can be accounted for by electrolytic enrichment alone.

Run No.2 (12th to 16th June 1989)

The second series of electrolysis runs with the Milton Roy cell commenced during the first week of June 1989. The cell was drained and flushed with heavy water many times for decontamination of tritium. Fresh electrolyte solution prepared using unused heavy water was charged and left in the cell over the weekend. On Monday 12th June a sample of this electrolyte which was analyzed for tritium content gave a surprisingly high tritium level of \(-0.32\, \mu\text{Ci/ml}\). This is attributed to tritium left over in the Pd—Ag cathodes from the 21st April run which must have transferred back into the electrolyte by chemical exchange.

Electrolysis commenced at 11:07 hrs. The cell was initially operated at currents of \(-60\, \text{amps}\) (current density \(-200\, \text{mA/cm}^2\)). During this run the BF$_3$ bank in paraffin was located close to the cell. About 1.5 m away there was a He$_3$ bank in paraffin which served as background monitor. The first neutron burst of this run was recorded within about half an hour i.e at 11:40 hrs. About an hour later two more 5 minute counts indicated high neutron levels. No more neutron bursts were observed for the next couple of days although cell operation was continued until 17:45 hrs on Wednesday 14th June when the cell was put off. But within a couple of hours of this there was a neutron burst lasting 15 minutes. Samples of electrolyte were drawn periodically throughout this operation and sent for tritium analysis. The tritium content of the electrolyte did not show any increase. Rather, it decreased from \(0.32\, \mu\text{Ci/ml}\) to \(0.12\, \mu\text{Ci/ml}\) on 15th June 89. The decrease is attributed to the fact that the electrolyte containing \(0.075\, \mu\text{Ci/ml}\) of tritium was added continuously for maintaining the level of the electrolyte. This obviously diluted the tritium content in the electrolyte.

During the next 48 hours the neutron monitors did not show any increase of counts. On the night of Friday, 16th June more than two days after putting off the cell current however, a large neutron burst was recorded corresponding to a total neutron yield of well over \(10^6\) neutrons. Fig 3 shows a plot of neutron count variation during this burst. Detailed time structure of this burst is presented in Table VII of Ref. 4. A sample of electrolyte was drawn only on 23rd June to ensure that maximum amount of tritium on Pd cathodes would by then have exchanged with the bulk D$_2$O. This indicated a tritium...
level of 121 nCi/ml. The week-long experiment was terminated at this point but the electrolyser was left as such with the electrolyte in the cell and deuterium in gas plenum at a pressure of 1 kg/cm$^2$ above atmospheric pressure. After a lapse of about a month when the electrolyte was removed and analyzed, the tritium level was found to have further increased to 460 nCi/ml, corresponding to a four-fold rise in the tritium level. It is not clear whether this is attributable to additional fusion reactions occurring when the cell was quiescent or whether the earlier built-up tritium continued to leach out into the electrolyte.

**Discussion**

The results of the neutron and tritium measurements carried out during the above runs are tabulated in Table III. The observed tritium concentrations have been corrected for enrichment effects due to electrolytic separation of deuterium and tritium as well as evaporation losses. It may be noted from this table that tritium production is much higher than the neutron yield, although in ‘hot fusion’ their probability is known to be approximately equal. Our experimental observation is that both neutron and tritium generation seem to be occurring simultaneously because as mentioned earlier from the results of the second run it is seen no neutron and tritium peak was observed for a long duration but the tritium level of the electrolyte was found to have increased multifold after a neutron burst was noticed. The observation in RUN2 of the electrolyzer shows that both neutron peaks and tritium were recorded only about 30 hours after the current had been put off. An important observation of this work is that "spent" Pd electrodes seem to lose their capability to support cold fusion reactions as can be seen from a comparison of the results of RUN1 and RUN2. In the latter case the number of neutrons and tritium atoms produced has decreased. This observation calls for further investigations. A multi-dimensional characterization of the freshly deuterated and spent Pd electrodes, such as measurement of the metallographic and lattice structure will go a long way in understanding this phenomenon and may shed considerable light on the mechanism of ‘Cold Fusion’.

**Acknowledgements**

The authors would like to acknowledge the unstinted cooperation received from several scientists in various aspects of these experiments. They are extremely thankful to each and every one of them. They include C.K. Pushpangathan, V.H. Patil, Arun Kumar and N.P. Sethuram of Heavy Water Division; and R.K. Rout and L.V. Kulkarni of Neutron Physics Division. Shri H.K. Sadhukhan, Head, Heavy Water Division has contributed immensely through fruitful discussions.

**References**

3. ELHYGEN Hydrogen Generator (Mark V), Manufactured by Milton-Roy Company, Shannon Industrial Estate, Clare, Ireland.
Additional Remarks on Neutron Counts Data of Table II and Fig. 2

(1) It is observed from the neutron counts data of Run No. 1 (21st April 1989) plotted in Fig. 2 that while the BF$_3$ bank has recorded at least 9 clearly visible peaks, the plastic scintillator has missed out some of the smaller peaks. The reason for this is obviously higher background level of the plastics scintillator arising from its sensitivity to gammas. The Smaller peaks have apparently got buried in the statistics of the background.

(2) The ratio of the counts under the peaks in the two channels after subtracting the background is found to vary between 1.3 and 2.8 which is considerably value of ~6.7 expected from their efficiencies determined using a Pu—Be neutron source. This discrepancy may be attributed to the following points. (a) The plastic scintillator being sensitive to the gammas of Pu—Be would indicate a higher sensitivity. (b) The energy spectrum of cold fusion neutrons could be different from that of Pu—Be source neutrons. Since the energy response characteristics of the plastic scintillator and BF$_3$ bank are different the efficiency ratios for Pu—Be neutrons and cold fusion neutrons could be quite different.

(3) From a detailed analysis of the neutron multiplicity spectrum (given in paper A4), it has been found that between 10 to 25% of the cold fusion neutrons are emitted in bunches of 100 or more. While the BF$_3$ bank which is surrounded by a moderator is able to resolve and count individually a number of simultaneously incident neutrons, the plastic scintillator will only give a single pulse (although of a much larger height). In fact by comparing the measured ratios of the counts under the peaks in the two channels with the expected efficiency ratio one can approximately assess what fraction of the neutrons emitted in each burst is due to bunched neutronic events. In the 21st April run it is found that in general the larger peaks seem to contain a higher fraction of bunched events.
TABLE I

Details of the Milton Roy Electrolytic Cell

<table>
<thead>
<tr>
<th>Vol. of Pd cathode</th>
<th>Mass of Pd</th>
<th>Area of cathode</th>
<th>Current</th>
<th>Current density</th>
<th>Electrolyte</th>
<th>Volume of electrolyte</th>
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<tr>
<td></td>
<td>7 cm³</td>
<td>82 gm</td>
<td>300 cm²</td>
<td>~200 mA/cm²</td>
<td>20% NaOD in D₂O</td>
<td>250 ml</td>
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TABLE II

Neutron Counts Vs Time
(21st April 1989)

<table>
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<tr>
<th>Interval duration</th>
<th>Efficiency of BF₃ Bank</th>
<th>Efficiency of NE102A</th>
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<tr>
<td></td>
<td>= 5 minutes</td>
<td>= 0.06%</td>
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<tr>
<td></td>
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<td>= 0.4%</td>
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<table>
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TABLE III

Summary of Neutron and Tritium Yields
(Current Density = 200 mA/cm²)

<table>
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<tr>
<th>Run No.&amp; Date</th>
<th>Duration of Electrolysis</th>
<th>Total Neutron yield</th>
<th>Atoms of Tritium generated</th>
<th>Neutron to Tritium yield Ratio</th>
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<td>Run 1 21st April 89</td>
<td>72 hrs</td>
<td>4x10⁷</td>
<td>8x10¹⁵</td>
<td>0.5x10⁻⁸</td>
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<tr>
<td>Run 2 12th–16th June 89</td>
<td>54 hrs</td>
<td>9x10⁵</td>
<td>5x10¹⁴*</td>
<td>-1.8x10⁻⁸</td>
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<td></td>
<td></td>
<td></td>
<td>1.9x10¹⁵*</td>
<td>0.5x10⁻⁸</td>
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</table>

*These were measured 30 hrs and 27 days respectively after the current had been switched off.
Fig. 1a. Electrolytic Cell.

Fig. 1b. Pd tube arrangement.
MILTON-ROY Pd-Ni ELECTROLYTIC CELL (21st APRIL 1989)

PROTON RECOIL COUNTER

BF3 BANK

COUNTS/5 MINS

TIME (HOURS)

FIG. 2
FIG. 3 NEUTRON YIELD VARIATION DURING THE BURST OF 16th JUNE 89
PRELIMINARY RESULTS OF COLD FUSION STUDIES USING
A FIVE MODULE HIGH CURRENT ELECTROLYTIC CELL

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*Desalination Division,
†Heavy Water Division,
‡Neutron Physics Division.

Introduction

In their first cold fusion paper1 Fleischmann et al suggested that an electrolytic cell with large volume and surface area and high current density may cause fusion reactions resulting in the production of significant amounts of heat and nuclear particles. The experiments reported in this paper present the results of our early efforts to design and operate a high current modular Pd—Ni electrolytic cell and look for cold fusion reactions.

Electrolyser and Operation

A five module cell of bipolar filter press configuration having palladium—(25%) silver alloy [procured from M/s Johnson—Mathey (U.K.)] as cathode (0.1 mm thickness) and porous Nickel as anode was fabricated. The electrodes are of circular plate geometry having a sectional area of ~ 78 cm². The five modules of the cell are connected in series and can be operated even upto a high current density of 1 A/cm² and temperatures of 100°C. The mixed gaseous products are carried out of the cell and recombined in a burner—condenser unit and the resultant heavy water recycled back to the electrolyser. The electrolyte can be recirculated via a cooler to keep the operating temperature sufficiently low to reduce evaporation losses. Fig.1 gives a schematic diagram of the modular cell. A flow diagram of the electrolyser and accessories is shown in Fig.2.

The D₂ and O₂ gases produced in the electrolyser and evolving out in a mixed stream enters the recombination unit. This unit is essentially a burner complete with a self igniter and a cooling arrangement so that the product of recombination namely D₂O is condensed and collected. There is a provision to add incremental quantities of oxygen continuously to ensure the complete conversion of D₂ to D₂O.

The system was filled with freshly prepared NaOD in D₂O (20%) upto the preset mark in the level gauge and the electrolyser switched ON and operated continuously at a current of 60 to 65 amps (corresponding to applied voltage of ~ 12.5V) from 5th May 1989 onwards, with a few hours of interruption due to failure of the D.C. power supply. Occasionally the current was raised to 78 A. The summary of characteristics of the cell are shown in Table I.

The present operation was carried out to test the electrolyser, recombination unit and other subsystems. Sustained operation for extended periods of time, particularly in closed loop mode, has not been carried out in this preliminary study.

Neutron and Tritium Measurements

The placement of neutron detectors around the cell was similar to that described in paper A-1. Two neutron detectors namely a bank of three BF₃ detectors embedded in paraffin wax and an 80 mm dia x 80 mm high plastic fast neutron detector were
employed to monitor the neutron output. The counts data was printed out on scroll printer. The BF$_3$ channel was counted for 110 seconds each while the plastic detector counted for 100 secs each. But both counting intervals were commenced at the same time. The cell was operated continuously for 4 hours, when a big burst of neutrons overlapping two counting intervals was recorded in both the detectors. Table II summarises the neutron counts data.

The tritium content of the electrolyte before and after electrolysis as analysed by liquid scintillation counting techniques (see paper A 9) were 0.055 nCi/ml and 190.3 nCi/ml respectively. The significant quantities of tritium carried away by the combined D$_2$ and O$_2$ gas stream has not been accounted for in this. Considering that the total volume of the electrolyte in the system including hold up was ~1 litre, this corresponds to a total tritium build up of 190 $\mu$Ci or 4x10$^{15}$ atoms.

Conclusion

The electrolysis though carried out for limited periods only has shown conclusively that cold fusion occurs in this system also. This is obvious from the neutron bursts obtained (Table II) and the results of tritium analysis in the electrolyte. The latter results have shown an increase of over 3500 times in the electrolyte after 50 hours of operation.

From the efficiency of the neutron detectors and the neutron counts data given in Table II, it is inferred that about 5x10$^6$ neutrons were produced over a 100 second interval. The corresponding tritium yield was -4x10$^{15}$, suggesting a gross neutron to tritium yield ratio of ~10$^{-9}$. But it must be emphasized that a considerable quantity of tritium may have been carried away by the gas stream. Although efforts were made to recombine and recover this D$_2$O for tritium it was not successful. The estimate of 10$^{-9}$ for the neutron to tritium yield ratio may therefore be considered as a lower limit only.

The limited experimental studies conducted with this system has given enough experience for initiating a prolonged closed cycle operation, where simultaneous counting of neutrons and monitoring of progressive build up of tritium during electrolysis is proposed to be carried out.

Acknowledgments

A number of scientists and engineers of the Desalination and Heavy Water Divisions have contributed in the fabrication, installation and operation of this cell. Their help is gratefully acknowledged.


**TABLE I**

Summary of Characteristics of Five Module Cell

<table>
<thead>
<tr>
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<th>Value</th>
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</thead>
<tbody>
<tr>
<td>No. of Cathodes</td>
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</tr>
<tr>
<td>Area of each Cathode</td>
<td>$\approx 78 \text{ cm}^2$ per surface</td>
</tr>
<tr>
<td>Volume of each Cathode</td>
<td>$\approx 1 \text{ cm}^3$</td>
</tr>
<tr>
<td>Total volume of electrolyte in system</td>
<td>1000 cm$^3$</td>
</tr>
<tr>
<td>Current</td>
<td>60 to 65 A with occasional peaking at 78 A</td>
</tr>
<tr>
<td>Current density</td>
<td>750 mA/cm$^2$</td>
</tr>
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</table>

**TABLE II**

Neutron Counts Variation

<table>
<thead>
<tr>
<th>BF$_3$ channel (counts per 110 secs)</th>
<th>Plastic Detector (counts per 100 secs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>131</td>
<td>168</td>
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<tr>
<td>125</td>
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<tr>
<td>148</td>
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<td>17198*</td>
<td>1563*</td>
</tr>
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<td>21113*</td>
</tr>
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<td>138</td>
<td>182</td>
</tr>
<tr>
<td>134</td>
<td>171</td>
</tr>
</tbody>
</table>

* The ratio of counts in the two channels were different in the two consecutive time intervals presumably because the single neutron burst is partitioned unequally in the two consecutive intervals. This was because the counting intervals was longer by 10 seconds in the BF$_3$ channel.
FIG-1 5 MODULE Pd-Ni ELECTROLYSER
Fig. 2 ELECTROLYSER SYSTEM FOR COLD FUSION STUDIES
OBSERVATION OF COLD FUSION IN A Ti—SS ELECTROLYTIC CELL

M.S. Krishnan*, S.K. Malhotra*, D.G. Gaonkar*, M.G. Nayar†
A. Shyam† and S.K. Sikka†

*Heavy Water Division,
†Neutron Physics Division,
†Desalination Division

Introduction

Since the two communications1–2 reporting the occurrence of cold fusion, experiments had been initiated in a number of laboratories to study the electrolysis of D₂O with palladium (Pd) as cathode. In a few cases titanium (Ti) has also been used as cathode. Ti is a material of interest as it can form deuteride up to the composition of TiD₂ (against 0.6 in case of Pd). Further Ti is more easily available and cheaper in our country. Three groups3–5 have reported the use of Ti as the cathode material in their electrolytic experiments. Meanwhile in an interesting paper6 use of Ti in deuterium gas loading experiments has been reported wherein occurrence of neutron bursts under non-equilibrium conditions was observed.

Having obtained very encouraging neutron and tritium yields with electrolytically loaded Pd7, it was decided to investigate Ti as cathode material in the electrolysis of D₂O. The results of both neutron and tritium measurements are reported in this paper. Unlike the Pd—Ni cell described in paper A1 which was a diffusion type commercial hydrogen generator, the Ti electrolytic cell employed for this work was of very simple design and constructed from materials readily available.

Description of the Cell

A schematic sketch of the cell is shown in Fig. 1. A cylindrical rod of Ti of dia 22 mm and length 150 mm constituted the cathode (area 104 cm²) while an SS outer tube of 4 cm ID served as the anode. The two electrodes are fixed coaxially with PTFE spacers and gaskets as shown in the figure. The inter electrode gap is 9 mm (surface to surface). As the cell does not have provision for separating deuterium and oxygen, both these gases are vented out from the top. The cell is also equipped with a thermowell to measure the temperature of the electrolyte. 5N solution of sodium deuteroxide (NaOD) in heavy water (isotopic purity > 99.8%) was used as the electrolyte, to achieve high current density. An external DC power supply was employed. The salient features of this electrolyzer are summarised in Table I.

Operation of the Cell

The electrolytic cell was first operated with 5M NaOD in D₂O for about 8 hours. Only temperature increase was recorded for this run. The current was raised slowly from 20 A (current density 193 mA/cm²) to a maximum of about 40 A (current density of -400 mA/cm²). The cell was then drained, flushed with distilled water several times, and operated for 12 hours with an electrolyte of 5 M NaOH in H₂O. By this time the neutron monitors were installed in position. Subsequently it was cleaned, rinsed and a repeat run with 5 M NaOD in D₂O was conducted. As this cell does not have provision for automatic electrolyte feed, the electrolysis was carried out as a batch process. After a few hours of operation, when the electrolyte level had decreased considerably, fresh electrolyte was added to make up the volume. The cell had operated for 8 hours, when the cathode was found to have attained a dull black coating which when analysed chemically was found to
contain iron as major component. The electrolyte solution also developed a pale greenish yellow colour because of dissolution of Fe from the anode. The cell is therefore being modified to have both anode and cathode of titanium. In addition the inter electrode distance between anode and cathode is being reduced to attain larger current densities.

Neutron Measurements

A bank of three \( \text{He}^3 \) counters embedded in paraffin moderator and a plastic scintillation detector were employed for neutron measurements. Efficiency of the \( \text{He}^3 \) detector viewing the cell was \( \sim 1\% \). The plastic scintillator which was placed away from the cell was employed as a background monitor. Background data was first collected before electrolysis was commenced. The background counts in the \( \text{He}^3 \) channel was \( \sim 240 \) counts per 10 seconds. Neutron measurements carried out with the cell operating with \( \text{NaOH—H}_2\text{O} \) as electrolyte was similar to the background data. But when the cell was operated with \( \text{NaOD—D}_2\text{O} \) as electrolyte, after about 3 hours the level of the neutron counts was higher (\( \sim 590 \) counts per 10 secs.). No big bursts of neutrons were observed as in the case of Pd—Ni electrolyser. When higher counts was continuously indicated, the cell was put off and the neutron counts was observed to come down immediately but it was still above background (\( \sim 385 \) counts per 10 secs.). The experiment was finally terminated when the counts level reverted to background presumably due to fouling of cathode surface by iron deposit. Fig.2 shows a representative plot of the data (for 10 min before and 5 min after switching off) showing the typical drop in the neutron counts when the cell was switched off. The typical background counts when the cell was removed is also indicated in the figure for comparison.

Tritium Measurements

At the end of about 8 hours of electrolysis when the electrolyte was sampled and analysed for tritium content it was found to contain \( \sim 48 \) nCi/ml of tritium activity. This is almost three orders of magnitude higher than the tritium level of the stock heavy water used for preparing the electrolytic solution which was \( \sim 0.05 \) nCi/ml. The tritium level in the electrolyte after about 12 hours of operation with \( \text{NaOH} \) was only \( 0.0676 \) nCi/ml.

Conclusions

The results of the neutron and tritium measurements seem to strongly indicate that cold fusion occurs in the Ti—D system also. When compared with Pd—D systems where big neutron bursts are observed, in the present system the neutrons are produced at a low but more or less steady rate. It is estimated from the neutron counts data, the duration of cell operation (the integrated time for which the cell was active was approximately 150 minutes) and the efficiency of neutron detectors (1%) that in all about \( \sim 3 \times 10^7 \) neutrons had been generated. The terminal tritium level in the electrolyte was \( \sim 48 \) nCi/ml. Since the cell volume is \( \sim 150 \) ml the total tritium inventory in the cell corresponds to \( 48 \times 150 = 7.2 \mu \text{Ci} \). After correcting for the tritium initially present in the \( \text{D}_2\text{O} \) both in the initial charge as well as make up volumes, it is estimated that an excess of approximately \( 7 \mu \text{Ci} \) of tritium was produced due to cold fusion reactions. Considering that the total number of neutrons generated was \( \sim 2 \times 10^7 \), this leads to a neutron to tritium yield ratio of approximately \( \sim 10^{-7} \).

Acknowledgements

The authors are very thankful to Shri S.K. Mitra, and Shri V. Raghunathan of Desalination Division and Kum V. Chitra of Neutron Physics Division for their help in the experimental work. We are also grateful to Dr. M. Srinivasan, Shri H.K. Sadhukhan, Dr. M.P.S. Ramani and Dr.P.K. Iyengar for their keen interest in this work.
References

5 C. Sanchez et al to be published in Solid State Comm.

TABLE - I

Salient Features of the Ti–SS Cell

<table>
<thead>
<tr>
<th>Feature</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume of Ti Cathode</td>
<td>57 cm$^3$</td>
</tr>
<tr>
<td>Area of Cathode</td>
<td>104 cm$^2$</td>
</tr>
<tr>
<td>Current</td>
<td>40 A - 60 A</td>
</tr>
<tr>
<td>Current Density</td>
<td>&lt; 600 mA/cm$^2$</td>
</tr>
<tr>
<td>Electrolyte</td>
<td>20 % (5 Molar) NaOD in D$_2$O</td>
</tr>
<tr>
<td>Volume Of Electrolyte</td>
<td>150 cm$^3$</td>
</tr>
</tbody>
</table>
Fig. 1. Ti-SS Electrolytic cell

- Thermocouple
- Thermowell
- D₂ + O₂
- S S Anode/body
- NaOD in D₂O
- Ti - Cathode
- PTFE Gaskets
- S.S. Flange

Fig.1. Ti-SS Electrolytic cell
Fig. 2. Titanium-Fe Cell: Neutron Counts Variation With Cell On And Off
MULTIPLICITY DISTRIBUTION OF NEUTRON EMISSION IN COLD FUSION EXPERIMENTS

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Introduction

Since the first announcement by Fleischmann & Pons1 and shortly thereafter by Jones et al2 of the observation of cold fusion reactions in palladium electrolytically loaded with deuterium, various theories and speculations have been put forward as possible mechanisms for the same. All the schemes proposed so far may be classified into two broad categories: (i) Those that lead to (d—d) fusion reactions taking place one at a time i.e. wherein occurrence of one fusion reaction does not directly influence the probability of occurrence of another. In this case one can assign a certain probability per second per (d—d) pair for the reaction rate. A figure of $10^{-20}$ has for example been deduced for this by Jones et al. (ii) The second category of mechanisms leads to a cascade or sharp bursts of fusion reactions.

One of the earliest speculations3–4 attributed the cold fusion phenomenon to muon catalysis triggered by cosmic ray produced muons. It was pointed out that each muon could in principle catalyze several hundred fusion reactions within a time span of a couple of microseconds. Yet another mechanism proposed has been lattice crystal fracture or cracking leading to acceleration of deuterons to energies of 20 to 50 Kev in electric fields generated across fracture crevices. Such internally accelerated deuteron beams are then presumed to cause (d—d) reactions. Here again the fusion reactions may be expected to occur erratically leading to bunched occurrence of (d—d) reactions. A brief review of the phenomenology associated with lattice cracking is presented by Kaushik et al in Ref.5.

Since neutrons are one of the end products of (d—d) fusion reactions, it may be expected that statistical analysis such as measurement of the multiplicity spectrum of neutron emission can give valuable insight into the possible origin of cold fusion reactions. The emission of neutrons in bunches of two or more can for example be observed by employing two (or more) fast neutron detectors and looking for coincidences among detected pulses within a gate interval of say 1 or 10 μs. Since the average background count rate is generally small, the chance coincidence rate due to background events would be negligible, making the task of establishing the occurrence of multiple neutron emission events quite easy.

An alternate technique of detecting fast neutron multiplicity is to employ a thermal neutron detector surrounded by a hydrogenous moderator such as paraffin. This type of detector system has the interesting property that a bunch of fast neutrons simultaneously incident on it would be temporally separated due to the statistical nature of the neutron slowing process and get detected as individual neutrons within a time span governed by the neutron die-away time in the moderator-detector assembly. It is this type of thermal neutron detector that was employed in the present studies. This paper summarises the results of the neutron multiplicity spectrum measurements carried out with the Milton Roy commercial electrolytic cell6 as well as D2 gas loaded Ti targets7–8.
Theory of Multiplicity Analysis

If the events producing neutrons are random in time and result in one neutron per event, then the number of counts observed in a time interval $\tau$ would be distributed according to the Poisson law as follows:

$$P_r = \frac{(N_0 \tau)^r}{r!} e^{-N_0\tau}$$

where $P_r$ is the probability of obtaining $r$ counts in time $\tau$ and $N_0$ is the average count rate. Thus when the average count rate is small i.e. when $N_0\tau << 1$, one can set $e^{-N_0\tau} \approx 1$. In this case the probabilities of detecting one, two and three neutrons in the time interval $\tau$ are $(N_0\tau)$, $(N_0\tau)^2/2$ and $(N_0\tau)^3/3$ respectively. In particular note that the ratio of doubles to singles is $(N_0\tau/2)$ while that of triples to doubles is $(N_0\tau/3)$ and so on.

On the other hand if there are events which result in bunches of neutrons, say $\nu$ neutrons per bunch, then in a time interval $\tau$ which encompasses the nuclear event, the probability distribution ($P_r$) of counts would be given by a binomial distribution as follows:

$$P_r = \binom{\nu}{r} (1-\epsilon)^{\nu-r} \epsilon^r$$

It is presumed that the interval $\tau$ is large compared to the neutron die-away time in the detection system. It is also assumed that the source event rate is so low that only one such event occurs in interval $\tau$. Here $\epsilon$ is the overall counting efficiency. If $S$ is the event rate of such multiple neutron producing cascades its contribution to the average count rate would be $Se\nu$.

Thus we can now conceive of a situation wherein there is a steady random background count rate of $N_0$ c.p.s. superimposed on which is a signal event rate of $S$ events producing $\nu$ neutrons each. In such a case the average count rate in time $\tau$ can be expressed as:

$$N_0\tau + Se\nu\tau$$

Likewise the probability of registering two and three counts in time $\tau$ now comprises of two components: One due to random background events and the second due to signal events releasing $\nu$ neutrons each. Table I summarises the expressions for these probabilities. Note that while the probabilities of various multiplicities due to the random background depend on the product $(N_0\tau)$ that due to the bunched neutronic events depend mainly on $\nu$ and $\epsilon$. In the limit of $\nu >> 1$ and $\epsilon << 1$ the expressions simplify as shown in the last column. It is clear from this that the deciding quantity under these circumstances is the product $\nu\epsilon$.

Table II gives the expected frequency distributions of multiple neutron counts for typical values of $N_0$, $\epsilon$ and $\nu$. The counting time interval ($\tau$) is kept fixed at 20 ms while the bunched neutron producing event rate ($S$) is taken as $10^2$/s. The data presented is for a total of $10^8$ sampling time intervals. It is clear from the table that while the average count rate for Poisson events is much higher than for bunched events, the frequencies of higher count multiplicities (>2) is much higher for the bunched events.

Experimental Details

Neutrons were counted by a bank of thermal neutron detectors embedded in a paraffin moderator block. One bank comprised of three BF$_3$ counters while the other was
made up of three $\text{He}_3$ counters. The neutron die-away time in each of these was $25 \mu$s. The $\text{BF}_3$ bank was mounted close to the Milton Roy Pd–Ni electrolytic cell and the $\text{He}_3$ bank near the $\text{D}_2$ gas loading apparatus about 1.5 m away. While one counter bank was being used as signal counter in one experiment the other bank served as background monitor and vice versa. The efficiency ($\epsilon$) of detection for the cold fusion source neutrons was typically in the range of 0.5% to 1.5% depending on the exact distance between the cold fusion source and the detector assembly as well as the pulse height discriminator bias setting.

The outputs of both these counter banks were fed to scalers whose readings could be read off by a Personal Computer (PC) at the end of each counting interval which was controlled by the clock in the PC. By taking the difference in the scaler readings corresponding to the end of two consecutive counting intervals the number of counts recorded in a given counting interval is computed and stored by the PC. It took the PC typically over 250 ms to carry out these operations following each sampling time. Hence a set of 1000 samples consumed a real time of 5 minutes.

The duration of the counting interval selected was 20 ms. This is a compromise between the conflicting requirements of increasing the total number of counts accumulated to get good statistics and minimizing the chance coincidence probability for Poisson events. For the typical count rates met with during the neutron emission phase of the cold fusion experiments 20 ms was a reasonably satisfactory choice. From such data accumulated over several hours, the frequency distribution of counts recorded in 20 ms interval could be computed.

Results and Discussion

Table III summarises the various neutron multiplicity spectrum measurements carried out during May–July '89 in cold fusion experiments. The details of the Milton Roy commercial electrolyser and its operation are described in Ref. 6. Likewise the $\text{D}_2$ gas loading experiments are summarised in References 7 and 8. To begin with the statistics of background counts was studied to ensure that the equipment was functioning satisfactorily. For this purpose all potential cold fusion sources were removed from the room where the neutron detectors were located. Data acquisition of background counts continued in an uninterrupted manner for 63 hours over a weekend (1800 hrs on Friday 2nd June to 0900 hrs Monday 5th June). During this run the average background count rate in the $\text{BF}_3$ bank was 0.023 cps and in the $\text{He}_3$ bank 0.43 cps (The background rate in the $\text{BF}_3$ bank was intentionally adjusted to be lower by setting its pulse height discriminator level high.). Table IV presents the results of the frequency distribution of counts obtained in this long background run. It is heartening to note that not even once out of the 750,000 odd samples were 3 or more counts registered by either of the detector banks as may be expected on the basis of Poisson distribution. The ratio of doubles to singles frequency further conforms to Poisson statistics, indicating that the equipment was functioning properly. Sparking in any of the counters for example would have given rise to significant non–Poisson behaviour.

Table V represents the results of our first attempt to measure multiplicity distributions of neutron signals. The data was accumulated overnight (1805 hrs on 26th May to 0645 hrs on 27th May) with the $\text{BF}_3$ bank viewing the Milton Roy cell which was quiescent i.e. the cell current was not on. Besides, a plastic scintillator (NE 102A) biased to register only neutrons of energy >9MeV monitored cosmic ray and other background events. The first column of the table gives the probability distribution of counts of the $\text{BF}_3$ bank for 20 ms intervals. The average count rate works out to 5.6 c.p.s. It is clear from the frequencies of 2s, 3s and higher multiplicities that there is considerable contribution of
non-Poisson events. The second column of the table gives the frequency distribution of the same counts data whenever there was a pulse recorded by the plastic scintillator also during a 20 ms interval. From this we conclude that only about 1% of the neutron events occurring in the BF$_3$ bank can be attributed to cosmic ray showers.

Table VI presents the frequency distribution results of the Milton Roy cell run of 12th to 14th June. As noted in the companion paper$^6$ six neutron bursts of five minutes duration each were recorded during this period, the first about 50 minutes after cell electrolysis commenced on 12th June, the second and third about an hour thereafter and the remaining three a few hours after the cell current was switched off on the evening of 14th June. During the burst phase, the count rates were in the range of -0.5 to 1.7 c.p.s. which is about 4 to 14 times that of the background value (~0.12 c.p.s.). However it is noteworthy that in 4 out of the 6 bursts observed, count multiplicities of 2, 3, 4, 5 and even 10 have been recorded at least once each. This type of behaviour is indicative of high multiplicity neutron emission events. Throughout this run lasting several days the background counter did not record any noticeable increase of counts.

Table VII summarises the frequency distribution measured during the 2.5 hour long neutron burst recorded on 16th June from 1900 hours onwards with the Milton Roy electrolyser (Fig. 3 of paper A.I gives a plot of the count rate variation during this burst). It may be noted that the cell had not been operated for the previous ~52 hours. The count rate during this wide neutron burst attained a value as high as 20 c.p.s. at the peak. It may be noted that the background neutron monitor which was only 1.5 m away also indicated a small increase in count rate confirming that the neutrons originated from the electrolytic cell. Careful scrutiny of these results indicates that the frequency distribution essentially corresponds to a Poisson distribution. However, the fact that multiplicities of 5 or more are recorded several times again points to the sporadic occurrence of multiple neutron emission events. It is noteworthy that around 1950 hours (close to the peak) there were more than 20 such high multiplicity cascade events within a time span of 5 minutes. (Please note that in Table VII set M corresponds to the early part of the burst and set A close to the peak with absolute time increasing from M to A).

Tables VIII & IX summarize the results of multiplicity distribution measurements carried out with two D$_2$ gas loaded Ti targets. During the weekend run of 9th to 11th June with 15 gms of Ti–Zr deuteride (see Table VIII) the average count rate measured was only 0.42 c.p.s.. Since this corresponds to an N$_0\tau$ value of 0.008, we expect a doubles to singles ratio of 0.004 only. While the high doubles events in both the background and signal counter can possibly be attributed to statistics, the 3s and 4s in the signal counts can be attributed only to high multiplicity neutron emissions from the deuterated Ti–Zr target. Absence of such high multiplicity events in the background channel further strengthens this conjecture.

Table IX presents a similar result from a Ti disc target. The gas loading procedure for this target is described in Ref.7, while the measurement of spatial distribution of tritium on the surface of this target by autoradiography is discussed in Ref.8. The neutron active phase of this target lasted almost 85 minutes during which it is estimated to have emitted $5 \times 10^5$ neutrons in all (see Fig. 1). On the whole this target also points to the occurrence of a significant number of high multiplicity neutron emission events.

The last columns of Tables VI, VII and IX give the total frequency distribution for all the 5 minute duration i.e. the sum of all the columns. Also shown are the theoretical frequency distributions expected on the basis of Poisson statistics. It is worth noting that except for the background case (Table IV), in all the cold fusion measurements the frequencies fall according to the Poisson law for low multiplicity events but there is distinct
tendency for them to show a slight peak between the multiplicities of 4 and 6. If we assume that this peak is due to the superposition of bunched neutronic events on a Poisson background, we deduce the value of $\nu$ to be in the range of 400 to 600 since the peak of the binomial distribution occurs at the multiplicity value of $\nu \epsilon$ (see Table I) and $\epsilon$ values in the experiment are typically -0.01. Further, by comparing the observed frequencies with those shown in Table II, it appears that the average source event rate for such events during the neutron emitting phase is very roughly about $10^{-2}$ per second.

Summary and Conclusions

The multiplicity spectrum of neutron counts observed in 20 ms time intervals has been measured so far six times during cold fusion investigations, three with electrolytically loaded Pd electrodes, two with gas loaded Ti targets and once with the background. While the background counts displays strictly Poisson statistics, in the three cases where distinct excess over background neutron bursts were recorded, between 10% and 25% of the emitted neutrons appear to display high neutron multiplicity characteristics. The observed frequency distributions can be explained as being due to bunched neutronic events superposed over a Poissonian background. Analysis of the data leads us to the conclusion that these bunched neutronic events occur on an average once every hundred seconds during the active period and typically produce about 400-600 neutrons per bunch, within a time span of less than 20 ms. Such occasional neutron bunches are also reported to have been observed by Menlov et al at LANL.

If this is viewed in the light of the experimentally deduced neutron-to-tritium branching ratio of $10^{-8}$, we are obliged to come to the intriguing conclusion that during bunched neutronic events upwards of $10^{10}$ fusion reactions occur in under 20 ms. As this appears very unlikely, the authors are inclined to believe that bunched neutronic events are not accompanied by tritium production with a yield ratio of $10^8$ and hence lattice cracking where the branching ratio is close to unity would appear to be the most plausible cause for bunched neutron emission.

References

TABLE I
Expressions for Frequency Distributions of Neutron Counts for Random (Poissonian) and Bunched Neutron Producing Events

<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>Frequency of counts</th>
<th>Random Events</th>
<th>Bunched Events</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Approximate values for $N_0 \tau &lt; 1$</td>
<td>Approximate values for $\nu \ll 1$</td>
</tr>
<tr>
<td>0</td>
<td>$\frac{T}{\tau} e^{-N_0 \tau}$</td>
<td>$\frac{T}{\tau} (1-N_0 \tau)$</td>
<td>$\text{ST}(1-\nu)^{\nu}$</td>
</tr>
<tr>
<td>1</td>
<td>$\frac{T}{\tau} (N_0 \tau) e^{-N_0 \tau}$</td>
<td>$\frac{T}{\tau} (N_0 \tau)$</td>
<td>$\text{ST} \nu (1-\nu)^{\nu-1} \epsilon$</td>
</tr>
<tr>
<td>2</td>
<td>$\frac{T}{\tau} \frac{(N_0 \tau)^2}{2!} e^{-N_0 \tau}$</td>
<td>$-$</td>
<td>$\text{ST} \frac{\nu^2}{2} (1-\nu)^{\nu-2} \epsilon^2$</td>
</tr>
<tr>
<td>3</td>
<td>$\frac{T}{\tau} \frac{(N_0 \tau)^3}{3!} e^{-N_0 \tau}$</td>
<td>$-$</td>
<td>$\text{ST} \frac{\nu^3}{3!} (1-\nu)^{\nu-3} \epsilon^3$</td>
</tr>
<tr>
<td>4</td>
<td>$\frac{T}{\tau} \frac{(N_0 \tau)^4}{4!} e^{-N_0 \tau}$</td>
<td>$-$</td>
<td>$\text{ST} \frac{\nu^4}{4!} (1-\nu)^{\nu-4} \epsilon^4$</td>
</tr>
<tr>
<td>5</td>
<td>$\frac{T}{\tau} \frac{(N_0 \tau)^5}{5!} e^{-N_0 \tau}$</td>
<td>$-$</td>
<td>$\text{ST} \frac{\nu^5}{5!} (1-\nu)^{\nu-5} \epsilon^5$</td>
</tr>
</tbody>
</table>

Mean (Peak) | $(N_0 \tau)$ | $(N_0 \tau)$ | $\nu \epsilon$ | $\nu \epsilon$
<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>Frequency of counts in 20ms intervals for 10^5 samples (8hrs real time)</th>
<th>Poisson Events</th>
<th>Bunched Events (S = 10^-2 per sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>µ = 100</td>
<td>µ = 100</td>
<td>µ = 500</td>
</tr>
<tr>
<td></td>
<td>ε = 0.015</td>
<td>ε = 0.005</td>
<td>ε = 0.015</td>
</tr>
<tr>
<td></td>
<td>Sνε = 0.015</td>
<td>Sνε = 0.005</td>
<td>Sνε = 0.075</td>
</tr>
<tr>
<td>0</td>
<td>99940</td>
<td>99994</td>
<td>99984</td>
</tr>
<tr>
<td>1</td>
<td>60</td>
<td>597</td>
<td>6.6</td>
</tr>
<tr>
<td>2</td>
<td>1.7</td>
<td>6.1</td>
<td>0.3</td>
</tr>
<tr>
<td>3</td>
<td>&lt;10^-5</td>
<td>&lt;10^-2</td>
<td>2.5</td>
</tr>
<tr>
<td>4</td>
<td>1.0</td>
<td>0.03</td>
<td>1.5</td>
</tr>
<tr>
<td>5</td>
<td>-10^-3</td>
<td>-10^-8</td>
<td>0.3</td>
</tr>
<tr>
<td></td>
<td>-10^-4</td>
<td>-10^-8</td>
<td>0.003</td>
</tr>
<tr>
<td>Nature of Cold Fusion Source</td>
<td>Back-ground Study</td>
<td>Milton Roy Cell Quiescent</td>
<td>Milton Roy Cell</td>
</tr>
<tr>
<td>----------------------------</td>
<td>------------------</td>
<td>---------------------------</td>
<td>----------------</td>
</tr>
<tr>
<td>Date of measurement</td>
<td>2nd–5th June</td>
<td>26th–27th May</td>
<td>12th–14th June</td>
</tr>
<tr>
<td>Reference Table no</td>
<td>IV</td>
<td>V</td>
<td>VI</td>
</tr>
<tr>
<td>BF&lt;sub&gt;3&lt;/sub&gt; Bank (BG)</td>
<td>0.023 cps</td>
<td>–</td>
<td>0.12 cps</td>
</tr>
<tr>
<td>He&lt;sup&gt;3&lt;/sup&gt; Bank (BG)</td>
<td>0.43 cps</td>
<td>–</td>
<td>0.1/0.2 cps</td>
</tr>
<tr>
<td>Detector viewing source</td>
<td>BF&lt;sub&gt;3&lt;/sub&gt;</td>
<td>BF&lt;sub&gt;3&lt;/sub&gt;</td>
<td>BF&lt;sub&gt;3&lt;/sub&gt;</td>
</tr>
<tr>
<td>Peak signal count rate</td>
<td>–</td>
<td>5.6 cps (Ave. for 12 hrs.)</td>
<td>0.5 to 1.7 cps</td>
</tr>
<tr>
<td>Times BG</td>
<td>–</td>
<td>–</td>
<td>4 to 14 times</td>
</tr>
<tr>
<td>Main characteristics</td>
<td>–</td>
<td>–</td>
<td>6 nos of bursts</td>
</tr>
<tr>
<td>Fraction of non poissonian neutrons</td>
<td>–</td>
<td>-2%</td>
<td>-25%</td>
</tr>
</tbody>
</table>
TABLE IV

Frequency Distribution of Background Counts in Two Detector Banks

Counting interval 20 ms
Total counting time 63 hrs

<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>Frequency</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>BF$_3$ Bank</td>
</tr>
<tr>
<td>0</td>
<td>750035</td>
</tr>
<tr>
<td>1</td>
<td>339</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
</tr>
<tr>
<td>3</td>
<td>0</td>
</tr>
<tr>
<td>4–20</td>
<td>0</td>
</tr>
</tbody>
</table>

$N_0$                  0.023cps  0.43cps
$N_0\tau$              $5 \times 10^{-4}$  0.0086
TABLE V

Frequency Distribution of Counts in BF₃ Bank and Plastic Scintillator with Quiescent Milton Roy Cell

<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>Gross Frequency</th>
<th>Frequency in those samples in which plastic scintillator records a pulse of large height</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>11941</td>
<td>114</td>
</tr>
<tr>
<td>2</td>
<td>2760</td>
<td>31</td>
</tr>
<tr>
<td>3</td>
<td>111</td>
<td>0</td>
</tr>
<tr>
<td>4</td>
<td>19</td>
<td>0</td>
</tr>
<tr>
<td>5</td>
<td>2</td>
<td>0</td>
</tr>
<tr>
<td>6</td>
<td>13</td>
<td>0</td>
</tr>
<tr>
<td>7</td>
<td>9</td>
<td>0</td>
</tr>
<tr>
<td>8</td>
<td>3</td>
<td>0</td>
</tr>
<tr>
<td>9</td>
<td>5</td>
<td>0</td>
</tr>
<tr>
<td>10</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>11</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>12</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>13</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>14</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>15</td>
<td>1</td>
<td>0</td>
</tr>
<tr>
<td>16</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>
TABLE VI

Frequency Distribution of Counts for 1000 Sampling Intervals Each of 20 ms During Six Periods of High Neutron Activity (Milton Roy Cell Run of 12th to 14th June 89)

<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>12th June</th>
<th>14th June</th>
<th>Total (A to F)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A  B  C</td>
<td>D  E  F</td>
<td>(Observed)</td>
</tr>
<tr>
<td>1</td>
<td>27  0  2</td>
<td>7 29 22</td>
<td>87</td>
</tr>
<tr>
<td>2</td>
<td>0  0  0</td>
<td>0  3  0</td>
<td>3</td>
</tr>
<tr>
<td>3</td>
<td>0  1  0</td>
<td>0  0  0</td>
<td>1</td>
</tr>
<tr>
<td>4</td>
<td>0  0  1</td>
<td>0  0  0</td>
<td>1</td>
</tr>
<tr>
<td>5</td>
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<td>0  0  0</td>
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<td>10</td>
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<td>1</td>
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</table>
### TABLE VII

Frequency Distribution of Counts for 1000 Sampling Intervals of 20 ms Each
During Periods of High Neutron Activity
(Milton Roy Cell Run of 16th June)

<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>Frequency</th>
<th>Total (B to M)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
</tr>
<tr>
<td>1</td>
<td>24</td>
<td>245</td>
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<td>2</td>
<td>7</td>
<td>99</td>
</tr>
<tr>
<td>3</td>
<td>3</td>
<td>16</td>
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<td>3</td>
<td>2</td>
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</tr>
<tr>
<td>6</td>
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<td>0</td>
</tr>
<tr>
<td>7</td>
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<td>0</td>
</tr>
<tr>
<td>8</td>
<td>1</td>
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<tr>
<td>9</td>
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<td>0</td>
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### TABLE VIII

**Frequency Distribution of Counts from Deuterated Zr–Ti Sponge**

<table>
<thead>
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<th>Counting interval</th>
<th>20 ms</th>
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<tbody>
<tr>
<td>Multiplicity of counts</td>
<td>Frequency</td>
</tr>
<tr>
<td></td>
<td>BF$_3$ Background</td>
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</table>

### TABLE IX

**Frequency Distribution of Counts From a Deuterated Disc for 1000 Sampling Intervals of 20 ms Each During Periods of High Neutronic Activity**

<table>
<thead>
<tr>
<th>Multiplicity of counts</th>
<th>Frequency</th>
<th>Total (A to Q)</th>
</tr>
</thead>
<tbody>
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<td>B</td>
</tr>
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<td>9</td>
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<td>0</td>
</tr>
<tr>
<td>6</td>
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</table>
FIG. 1: NEUTRON COUNTS VARIATION FROM DEUTERATED Ti DISC

WEEKEND OF 17th TO 19th JUNE 1989

X — SIGNAL
O — BACKGROUND
SEARCH FOR ELECTROCHEMICALLY CATALYSED FUSION OF DEUTERONS IN METAL LATTICE

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P.K. Iyengar*

Analytical Chemistry Division
*Director

Introduction

Recently, Fleischmann and Pons' have obtained evidence for the fusion of deuterons induced at low temperature, through the electrolysis of deuterium oxide at a palladium cathode. It is believed that this can be achieved if the deuterium loading of the lattice exceeds the value for PdD₀.8. In view of the relevance of this problem to this Centre, a program of work was initiated in this Division. This report is a summary of the behaviour of Pd and other metals during charging in alkaline heavy water.

Experimental

Materials, Reagents and Apparatus

Electrodes of different metals were used. These include a hollow Pd cylinder (area 5.87 cm², thickness 0.4 mm), Pd ring (area 14.5 cm², thickness 2 mm), Pd foil (area 1.5 cm², thickness 0.3 mm), Ti plate (area 8 cm², thickness 0.5 mm) and a triangular piece of Ni—Ti alloy of area 10 cm² and weight 10.5 gm. Platinum gauze, Pt disc or Pt foil of large area was used as the anode. Heavy water (99.87% purity) and pure Li metal were used to obtain 0.1 M LiOD. Iolar—2 N₂ or argon was used for deoxygenation and stirring. All electrodes were lightly abraded with emery, rinsed with acetone and dried before use.

Electrolysis cells were made of quartz for the container and the lids were of PTFE or glass. Provisions and inlets for deoxygenation, addition and removal of solution and temperature measurement were made. Loss of D₂O due to evaporation and electrolysis was compensated.

Equipments

Galvanostatic sources and current pulsing units were made in the laboratory. A Digital Panel Meter (PLA, DM—20), was used for voltage measurements. Neutron counting and tritium activity studies were carried out, as described later. Set—up is shown in Fig.1.

Results

Differential Enthalpimetric Studies

Twin cells composed of Pd as cathode (5.9 cm²) and Pt as anode (gauze) in one cell was coupled in series with Pt as cathode (5.9 cm²) and Pt as anode (gauze) in another cell. The purpose was to assess the magnitude of heat effects due to reactions like deuterium adsorption on the electrode, its recombination, dissolution in the lattice, etc. The cells were insulated by Dewar and 45 ml of 0.1 M LiOD were taken in quartz cells. The temperatures were monitored by matched thermistors using a bridge circuit and also measured with thermometers. The temperature of the two cells during charging is shown in Fig.2. The temperature of Pt—Pt cell remained nearly constant whereas the temperature of Pd—Pt cell
increased and attained a high value. The temperature rise in Cell I (Pd-Pt) is due to absorption, dissolution and interaction of deuterons in Pd lattice.

Calorimetric Measurements of Enthalpy changes

A modified isoperibol solution calorimeter with an accurate thermistor bridge was used. The cell with contents was enclosed in a tight-fitting Dewar and was immersed in a thermostat at 25°C for thermal insulation. Electrodes of hollow cylindrical Pd cathode (5.48 cm$^2$) and Pt sheet anode with 50 ml 0.1 N LiOD were used. Iolar-2 argon was used for deoxygenation and the solution was stirred mechanically. Charging was effected at different current densities and the terminal voltage in each case was noted. The solution temperature was monitored by the thermistor probe and bridge and also by a sensitive thermometer. The temperature increase as a function of time at different c.d. is shown in Fig. 3. The heat capacity of the cell and components was determined by electrical calibration and corrections for heat losses were applied on the basis of Newton's law of cooling (Fig.4). The results are included in Table 1.

Extended Electrolysis with Current Pulsing

Cylinder Electrode:

A hollow cylindrical Pd cathode (5.9 cm$^2$) and a Pt gauze anode were used and 0.1 N LiOD was electrolysed in a quartz cell with nitrogen bubbling. Initially, a constant current of 1A was used and when the temperature reached 60°C pulsing was commenced between 1 to 2A at 1 s. interval. The temperature was controlled to 63°C by forced circulation of air. Neutron flux measurements were made and D$_2$O was added for make up. After 41.8 hours during which 52.2 AH were consumed, the electrolysis was terminated. Measurement of tritium build-up in the final solution showed 3.75 $\mu$Ci.

Ring Electrode:

(1) 65 ml of 0.1 N LiOD in D$_2$O were electrolysed in a quartz cell using a Pd-ring cathode (14.5 cm$^2$) and two Pt discs as anode. At low current densities, the cell voltage was observed as a function of current and this is shown in Fig. 5. Later, electrolysis was carried out for nine days with current pulsing between 1A and 2A (80 h), 3A and 4A (20 h) and between 4A and 4.5 A (7 h) consuming a total of 296 Amp-hours. Neutron activity and capture $\gamma$ measurements were made throughout the duration of electrolysis. At the conclusion of the electrolysis, tritium content in the solution was found to be 16.25 $\mu$Ci.

(2) The above experiment was repeated using 0.1 N NaOD and a total of 231Amp. hour were passed during 77 hours. Neutron counting and tritium activity measurements were carried out.

(3) The same ring electrode, after degassing in vacuum, was subjected to 844amp. hour charging in 382 h. in 65 ml of deaerated 0.1 N LiOD. There was no significant tritium build-up in solution. The electrode was greyish black in colour.

Ni-Ti Electrode:

A Ni-Ti cathode (10 cm$^2$) was used in 0.1 N LiOD and a total charge of 135 amp. hr were passed over 111 hrs. Appreciable neutron activity was observed. The cathode was observed to flake off and disintegrate in powder form.
Ti Electrode:

A Ti sheet cathode (8 cm$^2$ area) and Pt sheet anode were used in the electrolysis of 1 M NaCl in D$_2$O at 34 mA.cm$^{-2}$. The terminal voltage was 4.4 V and temperature rose from 27°C to 36.6°C in 53 minutes. Prolonged electrolysis did not show any significant increase in temperature.

Nuclear Measurements:

Four different types of measurements were made to identify the emission of neutrons from the electrolysis cells.

1. Detection and direct measurement of neutrons were based on the use of He$^3$ detectors arranged in a well-counter as well as a Li$^6$ enriched scintillation detector (Bicron, Model NP-2). The detectors were calibrated by Am-Be source. Near complete rejection of high energy γ-rays was ensured by proper discrimination of the 2.5 MeV Co$^{60}$ sum peak using a Co$^{60}$ source.

2. Detection of high energy capture γ-rays of Gd, Pt and Pd was achieved by using Ge(Li) or HPGe detectors.

3. Measurement of low energy capture γ-rays of energies 199, 944 and 1186 KeV was carried out with a HPGe detector.

4. Gross counting of γ-rays of energy greater than 3 MeV was effected by means of a properly shielded 3" x 3" NaI (TI) detector.

All the above measurements were done in combination to yield cross-validated results. However, in some cells, only a single type of measurement could be made.

Neutron counting was performed both in the MCS mode (using IBM-PC based 8 K MCA system) and in the PHA mode whenever possible. Different dwell times ranging from 0.5 sec. to 60 sec. were selected in the MCS mode to check whether neutron emission is continuous or in "bursts". The complete counting set up is given in Fig. 6. The results can be summarised as under:

a) Palladium hollow cylinder cathode (run 3.1)
   The 1186 KeV gamma ray activity was measured every 100 sec. interval for more than 24 hours. In Fig.7, three definite 'spikes' can be identified. The duration of the 'bursts' was 14 to 20 minutes.

b) Palladium ring cell (run 3.2.1)
   A time correlated analysis of the neutron counts in the Li$^6$ scintillation counter and measurement of γ-rays of energy greater than 3 MeV in the NaI (TI) detector were carried out (Fig.8). The correlation coefficient for the 50 observations was 0.26 which is significant at the 5% level.

c) Ni-Ti cathode cell (run 3.3)
   Neutron counting was carried out using He$^3$ as well as Li$^6$ scintillation detectors with the former in both the MCS and integral modes and the latter in the MCS mode. The cell was run initially at 1A current and when the current was increased to 2 - 2.5 A, three well-defined "spikes" were registered in the MCS mode as shown in Fig.9.
The count rates of He$^3$ and Li$^8$ scintillator are not correlated in any significant manner, possibly because of the different efficiencies and geometries. However, tiny spikes are registered in the He$^3$ detector.

d) Palladium ring cell (run 3.23)

Several spectra for the low energy capture gamma rays were detected using HPGe detector for durations from 10,000 to 50,000 seconds. In some spectra, 199, 944 and 1186 KeV capture $\gamma$-rays could be identified though they were not traceable in other spectra. As the accumulated count rates for these energies are rather small, it appears that neutron emission is low and not continuous.

Discussion

Electrolysis of alkaline heavy water results in the splitting of D$_2$O with evolution of deuterium at the cathode and oxygen at the anode. The electrode process involves charge transfer, adsorption of the intermediates, subsequent reaction and gas evolution(2) On Pd electrode, the discharge reaction in alkaline solution is followed by a rate determining recombination of adsorbed deuterium atoms at low over potential, whereas at high over-potential an electrochemical desorption reaction is favoured. In the case of Ni–Ti, the discharge reaction is always accompanied by rate determining electrochemical desorption reaction.

The uptake of deuterium by metals during charging depends on the reaction

$$\text{MD}_{\text{ads}} \rightarrow \text{MD}_{\text{abs}} \rightarrow \text{MD}_{\text{lattice}} \quad (1)$$

and is therefore, governed by the fraction of the electrode surface covered by adsorbed D, solubility of deuterium in the metal and its diffusivity. The observed rise in temperature of the electrolyte is not due to the exothermic dissolution of deuterium in Pd, Ni–Ti and Ti or due to other chemical factors.

The measured overpotential is composed of ohmic, activation, adsorption, diffusion and concentration overpotential terms. The ohmic resistance and polarisation resistance cause Joule heating and thereby contribute to observed changes in enthalpy. Had the cell systems been reversible, then if the polarisation is reversed, the electrode reactions at Pd and Pt electrodes should also be reversed. In the upper limit the maximum Joule heating is the product of the cell voltage E and cell current i. In the case of Pd–Pt electrodes, the minimum voltage or back E.M.F for electrolysis of D$_2$O was calculated from thermodynamic data as 1.54 V. Therefore, the electrical power available for Joule heating is (E–1.54)i and this is the lower limit. The enthalpy changes calculated for different c.d. and the ratio of thermal output to the power input, expressed as per cent breakeven are also included in Table 1.

The predominant process during d.c. polarisation is the electrolysis of D$_2$O and has been checked by measuring the volume of D$_2$ and O$_2$ mixture liberated in a fixed electrolysis time using a precision integral flow–meter and by gas chromatographic analysis of the composition of the gas mixture. As excess heat is liberated over and above electrolysis, it is clear that some other reactions are responsible for the excess enthalpy observed. Pauling ascribed the excess heat to the formation of palladium deuteride. However, Bockris and coworkers have shown that exothermic effects due to solution of D in Pd, recombination of D atoms, formation of D$_2$O etc.cannot account for the observed heat evolution. Normal
chemical reactions cannot account for the generation of neutrons or the production of tritium during charging of Pd with deuterons. As has been pointed out by Fleischmann and Pons, the results can be rationalised and understood on the basis of cold fusion reactions occurring between deuterons in the Pd lattice as indicated below:

\[
\begin{align*}
{\bf ^1D + ^1D} & \rightarrow {\bf ^3H + ^1H} \\
{\bf ^1D + ^1D} & \rightarrow {\bf ^3He + ^1n}
\end{align*}
\] (2) (3)

In the present work, the emission of neutrons, reasonably above the background level, and the build up of significant tritium activity in excess of the blank value, have been confirmed in four different electrolysis experiments. In certain experiments neither the evidence for significant neutron emission nor any appreciable build up of tritium activity has been observed. It is likely that in such cases charging was not sufficient for ensuring optimum loading of the lattice with deuterons for inducing fusion. However, it appears that in addition to reaction channels (2) and (3), the possible occurrence of a non-emitting nuclear process cannot be precluded. This reaction can be written as

\[
Pd + {\bf ^2D + ^2D}_{\text{lattice}} \rightarrow {\bf ^4He + Pd^*}
\] (4)

which implies that the lattice is excited to a higher energy level to conserve both momentum and energy. It is likely that during the subsequent lattice relaxation, the excess energy stored in the lattice is liberated as heat. This mechanism would lead to the formation and build up of He inside the metal and can possibly account for the observed low yield of neutrons or tritium in certain experiments. Accurate mass spectroscopic analysis of \( {\text{He}}^3/\text{He}^4 \) ratio in the cathode material is needed to substantiate this view. In conclusion, it is necessary to examine in detail the different parameters and optimize important factors like metallurgical history and pretreatment of the cathode, solution chemistry, surface chemistry and electrochemistry to achieve reproducible fusion through electrochemical charging of metals in heavy water.

Acknowledgements

Authors are grateful to Dr. R. M. Iyer, Director, Chemical Group for his keen interest and encouragement. Acknowledgements are due to Shri T. S. Murthy, Director, Isotope Group for kindly providing the data on tritium analysis. Support from Shri S. P. Chaganty of Electronics Division for scintillation detector and associated electronics facility, Shri J. D. Gupta and Ms. Suman Kumari of Computer Division for the 8K PC-based MCA system and Dr. M. R. Iyer of Health Physics Division for \( \text{He}^3 \) well counter and MCS mode acquisition set up are gratefully acknowledged. In particular, we wish to acknowledge the painstaking efforts and support of the following scientists of the Division in providing the reliable data cited in this work: i) Continuous nuclear data acquisition — Smt. G. Leela and Kum. Anna John, Ss. Sanjeevkumar, Sunil Jai Kumar, Rakesh Verma, D. Shreetsan Nair and S. C. Hodwadekar. ii) Nuclear and electrochemical instrumentation — Ss. K. C. Thomas, J. R. Kale, R. G. Dalavi and A. W. Sahani. iii) Calorimetric measurements — Shri S. S. Sawant.
References


### TABLE I
Excess enthalpy generated during electrolysis of LiOD
Area of Pd hollow cylinder: 5.87 cm$^2$, Temp. 25°C

<table>
<thead>
<tr>
<th>Current density mA cm$^{-2}$</th>
<th>Energy input iE Watts</th>
<th>$H_{\text{measured}}$ Watts</th>
<th>Excess enthalpy (in % breakeven) $\Delta H_{\text{measured}} \times 100$ (E-1.54)</th>
<th>$\Delta H_{\text{measured}} \times 100$ Ei</th>
</tr>
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<tbody>
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<td>36</td>
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<td>105</td>
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<td>183</td>
<td>15.46</td>
<td>10.79</td>
<td>78</td>
<td>70</td>
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</tbody>
</table>
FIG. 1 EXPERIMENTAL SET-UP

45 ml D₂O (0.1M LiOO)
CELL I - Pd - Pt
CELL II - Pt - Pt
CURRENT = 1.0 Amp, [C.D. = 160 mA cm⁻²]

FIG. 2 DEPENDENCE OF TEMPERATURE ON TIME
VOLUME OF D₂O = 50 ml (0.1M LiOD)
Pd CATHODE: HOLLOW CYLINDER
Pt PLATE ANODE
C.D. (mA cm⁻²)
1 36
2 73
3 110
4 146
5 183
↓ CURRENT OFF

FIG. 3 TEMPERATURE CHANGES DURING CHARGING
**FIG. 4** TEMPERATURE - RATE OF COOLING RELATIONSHIP

SLOPE $K = 0.022 \text{ min}^{-1}$

**FIG. 5** CELL TERMINAL VOLTAGE AS A FUNCTION OF CELL CURRENT

85 ml D$_2$O/0.1M LiOD, 28 °C

Pd - RING CATHODE

Pt - DISC ANODES

AREA 14.5 cm$^2$
FIG. 6 COUNTING SET-UP FOR NEUTRONS AND GAMMA RAYS

FIG. 7 COUNTS OF 1186 keV CAPTURE Y-RAV OF GADOLINIUM NaI(Tl) DETECTOR
FIG. 8 TIME CORRELATION OF NEUTRON AND GAMMA RAY ACTIVITIES

FIG. 9 DISTRIBUTION OF NEUTRON COUNTS
(MCS MODE 50 SEC. DWELL TIME. Li SCINTILLATION DETECTOR)
TRITIUM GENERATION DURING ELECTROLYSIS EXPERIMENT


*Analytical Chemistry Division
**Isotope Division

Introduction

In continuation of the earlier R&D work carried out in connection with the investigations for electrochemically induced fusion of deuterons using palladium cathode and platinum anode, a series of experiments was carried out. The following is a summary of results and observations for two such experiments:

PDC-II Experiment

Start of the expt. 10.7.89 – Conclusion of the expt 25.7.89.

a) Cathode – Palladium cylinder

i) Thickness – 0.45 mm
ii) Surface area – 6.37 cm²
iii) Volume – 0.143 cc
iv) Weight – 1.7 g

b) Brief history of the palladium cathode

– A cylindrical type of palladium cathode was obtained and it appears that this palladium cylinder had not been used earlier for any tritium work. However, it was used in the analysis of the hydrogen gas samples and hydrogen purification work. – Palladium cylinder of 1 cm length was used.
– The palladium cathode was spot welded to a platinum rod.
– The cathode was cleaned by degreasing, using solvents like acetone and was subjected to heat treatment at 300°C under vacuum for 2 hours.

– The anode was platinum gauze of area 25 cm².

Cell and Accessories

The cell of about 100ml capacity was made of quartz. The pyrex glass lid was provided with cones for introducing thermometer, cathode, entry port for sparging solutions with pure argon gas and vent for liberated gases namely deuterium, oxygen and D₂O vapour for quantitative collection.
– The D₂O vapour collection: The evaporated heavy water from the cell was refluxed back into the cell with a water condenser and the vapour that escaped from the condenser was collected in a moisture trap which was kept cooled with ice.
– The argon, deuterium and oxygen were passed through a column of catalyst, which recombined deuterium and oxygen back to D₂O.

Note: In this experiment, the gases that are emerging after the recombination step were passed through a water trap without further provision for recombination over copper oxide column.
For temperature measurement, a calibrated thermometer was used.

Electrochemical measurement: The galvanostatic power source with provisions for current pulsing was used. Terminal voltage was measured by a digital panel meter.

Materials used in the experiment:
- Heavy Water: Nuclear grade heavy water was used in the cell for experiments. The isotopic purity was 99.87% with an initial tritium content of 170 dpm/ml. (0.076 x 10^{-3} μCi/ml).

- Electrolyte: Electrolyte was prepared 0.1 normal with respect to LiOD by using E.Merck grade lithium metal, and 60 ml was used for experiment.
- IOLAR:—2 grade argon was used throughout the experiment as carrier gas.
- The cell was cooled by an externally located fan.

Tritium Measurement

- Tritium counting was carried out using LKB system Model 1215, RACKBETA — II system.

Sampling

- Samples (2 ml) were withdrawn from the cell during the experiment from time to time to assess the tritium content.

- Counting procedure: Aliquots from the above samples have been drawn and added to 'Instagel Cocktail' with a total volume of 10 ml for counting the tritium. Known amounts of tritiated water from NBS standard cells were taken into 10 ml of Instagel as reference. The blank sample for tritium was also prepared in Instagel with tritium free water obtained through ground water. Along with these background samples, counting was also done simultaneously for samples drawn from the double distilled water used for diluting and making up the samples for counting. Where interesting results were obtained, the full tritium spectrum was obtained and compared with the NBS standard tritium samples; when the samples gave significant activity, known amounts of the samples were distilled and the water distilled was taken for counting.

- During the electrolysis the cell level was maintained at a constant volume of 60 ml by periodically adding pure D2O as mentioned above. Also, the alkalinity was maintained at 0.1 N by addition of LiOD from time to time.

Electrolysis

The following electrolysis procedure was adopted for the experiment. In the beginning, the electrolysis was continued at a constant current (40 hrs). Later the electrolysis was conducted using pulsing technique of varying current. It may be noted that the pulsing was carried out in this particular experiment only during the day time and in the night time the electrolysis was at a constant current. The details of the electrolysis parameters such as the values of constant current, pulsing current, duration and also the tritium values obtained during the experiment are given below:

From 12.7.89 to 25.7.89 the electrolysis was conducted in the nights with constant current of 1 amp and pulsing of the current was conducted during the day time.
Degassing Procedure

Following procedure was used to heat the electrode to recover the deuterium and tritium.

At the termination of the electrolysis the cathode was transferred to a quartz heating tube and was connected to a vacuum assembly. The moisture was removed from the electrode surface by heating to 80—110°C and condensed in a cold trap. After condensing the moisture, oxygen and nitrogen gases were recovered by standard getters. The electrode was gradually further heated and the gas released was purified and absorbed in a pyrophoric uranium trap. The electrode was maintained at 550°C for 2 hours till no further release of gas took place. Deuterium gas was released from this pyrophoric uranium trap, diluted with about 2.5 l of hydrogen and was converted into water by circulating through heated CuO bed. This water was taken for tritium counting.

Observations : Table IB

— On 11.7.89 between 11.55 and 12.20 hrs. there was power breakdown for 25 minutes.

— The first sample, PDC—II.1, was collected after 75 amp.hr. for tritium assay. It gave 1.59 μCi/ml of D₂O. The total volume of D₂O in the experiment at this time of sampling was 60 ml. The tritium activity accounted would be 95.4 μCi. It may be mentioned that the total D₂O (cell volume of 60 ml and make up volume of 40 ml) works out to be 100 ml. The excess tritium that has been produced at the end of 75 amp.hr. electrolysis works out to 1.25 x 10⁴ times at this stage.

— Second sample was withdrawn at 235.65 amp.hr. and the assay of the sample showed that the activity per ml of the cell came down to 0.76 μCi/ml. With a view to examine the behaviour of the cell, the experiment was continued upto 433 amp.hrs. and intermittent samples were taken:

PDC—II.3 (273.3 amp.hr ), PDC—II.4(381.5 amp.hr ), PDC—II.5(433.32 amp.hr.).

— On 26.7.89 at about 1015 hrs. the lid of the electrolytic cell was thrown out along with the anode and thermometer. The experiment was stopped and a new lid was put with the cathode inside the cell. Oxygen and argon mixture was bubbled through the cell and a final sample PDC—II.5 was taken on 28.7.89 and was assayed for tritium activity (0.31 μCi/ml.)

Observations : Table IC

— The total activity recovered is 55.52 μCi during the electrolysis and after the stoppage of electrolysis 0.71 μCi was recovered from the vapour condensate and deoxo— recombination trap. Electrode degassing yielded 0.03 μCi and in all 56.27 μCi were recovered.

— Total input of tritium activity from (60 ml cell volume + 196 ml make—up volume) 256 ml is 0.02 μCi.

— In all out of 256 ml of D₂O used, 132 ml of D₂O was recovered and accounted for at the end of the experiment.

In conclusion, it may be pointed out that at the end of 75 amp.hrs the electrolysis generated about 95.4 μCi excess tritium (12500 times the input). However, at the end of the electrolysis, that is, after 433.32 amp.hrs. the total excess tritium recovered works out to be 56.25 μCi.
PDC–III Experiment

Start of the Expt. 6.9.89. Conclusion of Expt. 14.9.89

a) Cathode: Same palladium cylinder used in PDC–II experiment was used in this experiment.
b) Cathode preparation:
   To remove deuterium and tritium if any in the cathode the following method was adopted:
   - The cathode was placed in the quartz assembly connected to the high–vacuum line and slowly heated. Subsequently the electrode was heated under continuous vacuum from room temperature to 850°C. The electrode was kept at this temperature for two hours. When the electrode temperature exceeded 750°C, a black deposit was formed on the cooler parts of the quartz assembly. In view of this, heating was continued at 850°C for further two hours. No further deposit was released at this temperature. The black coating formed is being subjected to investigations to identify the nature of the deposit.

   - The electrode was brought to room temperature and the vacuum line was filled with deuterium gas at 1 cm of Hg pressure. The electrode was slowly brought to 800°C and kept for 3 hours at this temperature. After this deuterium reduction process, the deuterium gas was removed and heating was continued for additional 3 hours under continuous vacuum. After this operation the electrode was checked for any further release of gases. No further degassing was observed.

   - It may be pointed out that the black deposit initially released from the electrode turned to a silvery white deposit at this stage and this coating is under investigation.

   - The electrode after this treatment became shining silvery white and was used for this experiment.

c) Anode: The anode was a similar platinum gauze of area 25 cm².

Cell and Accessories

   The cell and accessories remained mostly similar as in PDC–II except for the following:

   - Palladium catalyst of standard design was used for recombination.
   - An additional recombination stage consisting of copper oxide column was used to convert oxygen depleted deuterium gas into D₂O.
   - Electro–chemical measurement. The details are similar as in PDC–II experiment.

Materials used in the Experiment

Heavy water: Similar grade of heavy water as was used in the previous cell was used here.
The heavy water cell volume in this experiment was 80 ml.

   - Electrolyte: Electrolyte was similar as in PDC–II experiment.
   - The other conditions such as cooling the cell and tritium measurements were identical
     with PDC–II experiment.
   - Sampling: The procedure followed and the volume of the samples withdrawn from the
     cell during the experiment remain the same.

In all during this experiment, 12 samples were taken from the start of the experiment to
the end of the experiment. Other samples for tritium counting were collected from various stages of recombination from the catalyst, the water—bubbler solution and the degassed electrode.

**Electrolysis**

Electrolysis procedure followed was similar to PDC-II experiment. However, pulsing was introduced after 67 amp.hrs. of electrolysis at constant current mode. It may also be pointed out that the pulsing was done continuously during the day and night for the entire experiment.

- On 13.9.89 the electrolysis was reverted back to the constant current mode at 2 amp. for 3 hours, followed by pulsing till end of the experiment. The pulsing during the period was between 2 — 4 amps.

- Details of the electrolysis parameters are given in Table II.

**Observations : Table IIB**

- On 8.9.89 between 1300 hrs. to 1350 hrs. there was a power breakdown.
- 9 samples of the cell solution were collected from 7.9.89 to 13.9.89 for monitoring the level of tritium activity. The tritium content in the cell solution did not show an increase between 7.9.89 to 13.9.89, except that there was a marginal decrease in the samples taken on 13.9.89 compared to the samples taken on 12.9.89. In view of this experiment was continued with a higher pulsing parameter as that of Fleischmann experimental conditions.
- During this operation, constant current for 3 hours and pulsing for 2 hours were maintained.
- On 14.9.89 around 1000 hrs. there was a change in the cell behaviour. The bottom of the cell was shattered due to explosion. In all, the total volume of D$_2$O used in this experiment was 185 ml which included 105 ml of make-up D$_2$O and 80 ml of cell D$_2$O.
- Before the explosion it was noticed that the cell temperature shot up from 71°C to 80°C and the bubbling rate was low.

**Observations : Table IIC**

Due to the explosion at the end of the experiment all the electrolyte solution of about 80 ml was completely lost and no sample could be collected for tritium assay. Broken pieces of the cell were collected. The upper part of the cell and the broken pieces were washed and the sample was collected for counting. The D$_2$O condensate and the recombined water from the palladium catalyst, copper oxide and the bubbler solutions were taken for tritium counting.

The total tritium activity counted from all the samples accounted for 2,08,531 dpm while the total input tritium activity in 185 ml of D$_2$O used in this experiment comes out to be 30,170. It may be noted that the activity in the cell solution could not be accounted as it was lost. Even this tritium activity of 2,08,531 dpm, accounted from the recovery and recombination of gases liberated from the cell, has shown excess tritium 5.79 times.

In view of the repeated nature of the explosion at the terminal stages of electrolysis it was decided to subject the palladium cathode for a metallographic examination. Metallographic examination was done at the Physical Metallurgy Division by means of polarised light microscopy after embedding and mounting the electrode with an acrylic
polymer resin. The palladium cathode was then cleaned with acetone and tetrahydrofuran to remove the polymer coating. The electrode on degassing gave about 1.2 ml of gas which accounted for 5,530 dpm of tritium activity. It may be mentioned here that if the electrode had been degassed before the metallographic examination the tritium content would have been significantly higher.

It may be pointed out that had the cell solution not been lost, significant amount of tritium could have been recovered. An important observation from the counting of the cell samples was that large tritium activity was not produced in the cell until 13.9.89 (PDC—III—12) even at 218 amp. hrs. Excess tritium was produced only between 13.9.89 and 14.9.89 (between 218 amp.hrs and 286.2 amp.hrs.) whereas in the PDC—II experiment excess activity was generated at about 75 amp.hrs. in 3 days of electrolysis.

**Results of the polarised light microscopy of the electrode**

Metallographic examination of the palladium cathode used in the experiment and which experienced an explosion showed an extensive twinning within the palladium grains with worm—like microstructure. This is suggestive of an intensive shock—wave impact on the metal. Microphotographs of the two samples are attached for reference(Figs. 1—3)

A similar palladium cylindrical cathode was also metallographically examined. This electrode was given for comparison and did not see any explosion during electrolysis. Results of metallography revealed that it did not show any significant twinning and worm—like microstructure.

**Acknowledgements**

Acknowledgements are due to Dr. P.K. Iyengar, Director, BARC for his keen interest in this work. Acknowledgements are also due to Dr. G.E. Prasad and Dr. M.K. Asundi, Head, Physical Metallurgy Division for carrying out the metallographic investigations.
### TABLE I

#### A. SUMMARY OF ELECTROLYSIS FOR PDC-II

<table>
<thead>
<tr>
<th></th>
<th>Description</th>
<th>Amp.Hrs.</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Constant current mode</td>
<td>10.7.89 - 12.7.89</td>
</tr>
<tr>
<td>2</td>
<td>a) Pulsing current mode (during day)</td>
<td>12.7.89 - 25.7.89</td>
</tr>
<tr>
<td>3</td>
<td>b) Constant current mode (during night)</td>
<td>12.7.89 - 25.7.89</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

#### B. SUMMARY OF TRITIUM LEVELS IN D$_2$O DURING ELECTROLYSIS

- Vol of D$_2$O/0.1 LiOD Electrolyte = 60 ml
- Tritium activity in blank D$_2$O/LiOD = 170 dpm/ml of D$_2$O = 0.076 x 10$^{-3}$ μCi/ml of D$_2$O

<table>
<thead>
<tr>
<th>Date</th>
<th>Sample No.</th>
<th>Amp.Hrs.</th>
<th>Vol. of D$_2$O added ml</th>
<th>Tritium activity μCi/ml D$_2$O</th>
<th>Excess tritium (times)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.7.89</td>
<td>PDC-II-1</td>
<td>75</td>
<td>40</td>
<td>1.59</td>
<td>1.25 x 10$^4$</td>
</tr>
<tr>
<td>19.7.89</td>
<td>PDC-II-2</td>
<td>235.6</td>
<td>111</td>
<td>0.76</td>
<td>3.5 x 10$^3$</td>
</tr>
<tr>
<td>20.7.89</td>
<td>PDC-II-3</td>
<td>273.3</td>
<td>131</td>
<td>0.62</td>
<td>2.56 x 10$^3$</td>
</tr>
<tr>
<td>24.7.89</td>
<td>PDC-II-4</td>
<td>381.5</td>
<td>176</td>
<td>0.39</td>
<td>1.31 x 10$^3$</td>
</tr>
<tr>
<td>25.7.89</td>
<td>No sample</td>
<td>423.17</td>
<td>196</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>26.7.89</td>
<td>No sample</td>
<td>433.32</td>
<td>196</td>
<td>–</td>
<td>–</td>
</tr>
<tr>
<td>28.7.89</td>
<td>PDC-II-5</td>
<td>433.32</td>
<td>196</td>
<td>0.31</td>
<td>0.95 x 10$^3$</td>
</tr>
</tbody>
</table>
C. SUMMARY OF TRITIUM ACTIVITY IN THE OVERALL EXPERIMENT: PDC-II

**INPUT:**
- Initial volume of D₂O electrolyte
- Volume of D₂O added

**Total:**

\[ \text{Total} = 256 \text{ ml} \]

**Tritium activity in D₂O**

\[ = 0.76 \times 10^{-3} \mu \text{Ci/ml D₂O} \]
\[ = 170 \text{ dpm/ml D₂O} \]

**Total input of Tritium activity**

\[ = 0.02 \mu \text{Ci} \]

**OUTPUT**

<table>
<thead>
<tr>
<th>Source</th>
<th>Vol. (ml)</th>
<th>Total activity (µCi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>i) End electrolysis D₂O cell—sample recovered (PDC-II-5)</td>
<td>52</td>
<td>15.96</td>
</tr>
<tr>
<td>ii) Vapour—condensate recovered</td>
<td>16</td>
<td>11.87</td>
</tr>
<tr>
<td>iii) Deoxo—recombined D₂O recovered after termination of electrolysis</td>
<td>16 + 52</td>
<td>20.82</td>
</tr>
<tr>
<td>iv) Vapour—condensate—II recovered</td>
<td>0.5</td>
<td>0.14</td>
</tr>
<tr>
<td>v) Deoxo—recombined D₂O—II recovered</td>
<td>2.8</td>
<td>0.57</td>
</tr>
<tr>
<td>vi) Bubbler (H₂O)</td>
<td>16</td>
<td>1.16</td>
</tr>
<tr>
<td>vii) Electrode gas control extracts after the electrolysis</td>
<td>—</td>
<td>0.03</td>
</tr>
<tr>
<td>viii) Samples drawn during electrolysis</td>
<td>8</td>
<td>5.72</td>
</tr>
</tbody>
</table>

**Excess tritium recovered**

\[ \frac{56.25}{0.02} = 2.812 \times 10^3 \text{ times.} \]
TABLE II

A. SUMMARY OF ELECTROLYSIS FOR PDC III

1. Constant current mode 6.9.87 - 9.9.89
   Amp.Hrs 71.75
2. Pulsing current mode 9.9.89 - 14.9.89
   Amp.Hrs 214.87
             286.62

B. SUMMARY OF TRITIUM LEVELS IN D₂O DURING ELECTROLYSIS

<table>
<thead>
<tr>
<th>Date</th>
<th>Sample No.</th>
<th>Amp.Hrs</th>
<th>Vol. of D₂O added ml</th>
<th>Tritium activity dpm/ml</th>
</tr>
</thead>
<tbody>
<tr>
<td>7.9.89</td>
<td>PDC-III-4</td>
<td>18.0</td>
<td></td>
<td>195 ± 4</td>
</tr>
<tr>
<td>8.9.89</td>
<td>PDC-III-5</td>
<td>42.0</td>
<td>15</td>
<td>276 ± 8</td>
</tr>
<tr>
<td>8.9.89</td>
<td>PDC-III-6</td>
<td>Current stopped</td>
<td></td>
<td>263 ± 8</td>
</tr>
<tr>
<td>9.9.89</td>
<td>PDC-III-7</td>
<td>65.67</td>
<td>30</td>
<td>249 ± 8</td>
</tr>
<tr>
<td>10.9.89</td>
<td>PDC-III-8</td>
<td>106.12</td>
<td>45</td>
<td>216 ± 4</td>
</tr>
<tr>
<td>11.9.89</td>
<td>PDC-III-9</td>
<td>137.87</td>
<td>53</td>
<td>248 ± 10</td>
</tr>
<tr>
<td>12.9.89</td>
<td>PDC-III-10</td>
<td>170.22</td>
<td>65</td>
<td>256 ± 10</td>
</tr>
<tr>
<td>12.9.89</td>
<td>PDC-III-11</td>
<td>185.87</td>
<td>75</td>
<td>260 ± 5</td>
</tr>
<tr>
<td>13.9.89</td>
<td>PDC-III-12</td>
<td>218</td>
<td>80</td>
<td>250 ± 5</td>
</tr>
<tr>
<td>14.9.89*</td>
<td>no sample</td>
<td>286.2</td>
<td>105</td>
<td>End of experiment</td>
</tr>
</tbody>
</table>

* There was an explosion and entire D₂O in the cell was spilled and lost. In view of this no sample could be taken.
C. SUMMARY OF TRITIUM ACTIVITY IN THE OVERALL EXPERIMENT PDC-III

INPUT:

<table>
<thead>
<tr>
<th>Description</th>
<th>Tritium activity (dpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial volume of D₂O</td>
<td>80 ml</td>
</tr>
<tr>
<td>Vol. of D₂O added</td>
<td>105 ml</td>
</tr>
<tr>
<td>Total</td>
<td>185 ml</td>
</tr>
<tr>
<td>Tritium activity in D₂O</td>
<td>166 dpm/ml D₂O</td>
</tr>
<tr>
<td></td>
<td>0.075 x 10⁻³ µCi/ml D₂O</td>
</tr>
</tbody>
</table>

= 30,710 (0.0138 µCi)

OUTPUT:

<table>
<thead>
<tr>
<th>Source</th>
<th>Vol. (ml)</th>
<th>Total activity (dpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>i) Cell wash/ broken quartz pieces</td>
<td>2</td>
<td>8.592</td>
</tr>
<tr>
<td>ii) Vapour—condensate recovered</td>
<td>2.5+5.0</td>
<td>1,14,025</td>
</tr>
<tr>
<td>iii) Palladium catalyst recombined</td>
<td>70+20</td>
<td>68,605</td>
</tr>
<tr>
<td>iv) D₂O recovered after CuO</td>
<td>1+1.1</td>
<td>4,285</td>
</tr>
<tr>
<td>v) Bubbler (H₂O)</td>
<td>—</td>
<td>3,068</td>
</tr>
<tr>
<td>vi) Electrode gas content extracted after explosion</td>
<td>−</td>
<td>5,530</td>
</tr>
<tr>
<td>vii) Samples drawn during electrolysis</td>
<td>18</td>
<td>4,426</td>
</tr>
<tr>
<td>viii) End electrolysis D₂O cell sample*</td>
<td>—</td>
<td>2,08,531</td>
</tr>
</tbody>
</table>

Excess tritium recovered = 1,77,821 = 5.79 times
\[
\frac{30,710}{30,710} = 0.080 \mu Ci
\]

*Calculation of total tritium activity does not take into account 80 ml of D₂O spilled due to the explosion.
Fig. 1. Deuterium loaded Pd sample-1 after explosion
Fig. 3. Pd sample-2 annealed (550°C - 2 hours) but without deuterium loading.
BURST NEUTRON EMISSION AND TRITIUM GENERATION FROM PALLADIUM CATHODE ELECTROLYTICALLY LOADED WITH DEUTERIUM


Introduction

Recently there have been many reports on the observation of neutrons, tritium and excess heat output from palladium and titanium cathodes electrolytically charged with deuterium\(^1\). Steady neutron count rate observed during the operation of the electrolytic cells containing these cathodes and platinum anodes were in some cases only 1.5 to 3 times the background rate\(^3\) whereas in some other cases a much higher steady emission rate \((10^4 s^{-1})\) was observed. In some experiments conducted at BARC earlier, neutron bursts of two orders of magnitude larger than the background have been reported from electrolytic cells\(^5\). The possible nuclear reaction responsible for the emission of neutrons has been inferred to be the \('\text{cold fusion}'\) of D atoms (existing as D\(^+\)) in the metallic lattice of the cathode

\[
\text{D}^2 + \text{D}^2 \rightarrow [\text{He}^4]^* \rightarrow \text{He}^3 + \text{n}^1
\]  

We report here a large burst emission of neutrons (signal/background ratio as high as 2000) from a thin ring shaped Pd cathode during the electrolysis of heavy water at relatively low cell currents and also tritium generation as measured in the electrolyte as well as in the water reformed from the absorbed gas recovered from the cathode.

Experimental

The electrolytic cell design was optimised mainly with respect to observation of fusion products rather than the accurate measurement of excess heat output. This is because the quantity of heat required to raise the temperature of 1 gm of D\(_2\O\) even by 1 K demands the occurrence of about \(10^{13}\) fusions (see Discussion sec.) and hence heat measurement appears to be an in-sensitive method of confirming whether cold fusion occurs in the cathode or not. Fig.1 shows the schematic of the experimental set up. The electrolytic vessel (vol. \(- 250 \text{ cm}^3\)) used was made from quartz with a gas tight nylon cap. The cap had a number of penetrations for inserting electrode lead wires, thermocouple, reference electrode, purge gas inlet and outlet tubes. These tubes were made of 'Corning' glass. The Pd Cathode used was in the form of a hollow ring 2.5 cm in diameter, 1 cm in height and about 0.1 cm in thickness (surface area is 18 cm\(^2\)). It was pretreated by vacuum degassing (<10\(^{-3}\)mm Hg) at 1073 K for a total period of about 10 hr in three heating–cooling cycles (~ 3.3 hr/cycle). The bulk density of the cathode was determined to be 12 g cm\(^{-3}\). The cell configuration was such that the ring cathode could be charged with deuterium from both sides as it was surrounded from inside and outside by Pt gauze anodes. The anodes were loosely sandwiched between pairs of Nafion membrane so as not to allow oxygen evolved at the anode to diffuse to the cathode surface. The electrolyte was D\(_2\O\) of 99.86\%2 of isotopic purity containing 0.1 mol dm\(^{-3}\) LiOD and was kept under constant circulation at a flow rate of 10 cm\(^3\) m\(^{-1}\) using a peristaltic pump. The D\(_2\O\) used for preparing the initial LiOD solution and subsequent makeups during the electrolysis came from a single stock of D\(_2\O\). Incorporation of a heat exchanger in the circulation path along with a thermostated water bath (273—373 K) served to maintain the temperature of the electrolyte (and hence the electrode) at any desired value in this range. Nitrogen gas
was bubbled through the cell to reduce the dissolved oxygen level in the electrolyte. A saturated calomel electrode along with a lugging probe containing 0.1 mol dm$^{-3}$ LiOD/D$_2$O dipped in the electrolyte was used as a reference electrode to monitor the single electrode potential of the cathode. Periodic monitoring of the cathode potential was carried out both when the working electrode was under polarised condition and when the polarisation voltage was momentarily switched off.

A well type neutron detector assembly containing 24 numbers of $^3$He detectors embedded in paraffin filled to sufficient thickness and held between aluminium cylinders for thermalisation of the neutrons was used for n detection. This configuration gave 8.6% efficiency for n detection. The output from the detector–preamplifier–active filter amplifier tie-up was fed in parallel to a scaler with pre–set time to an oscilloscope and to a personal computer operated in a 8K multichannel scaling (MCS) mode with dwell time of 40 seconds per channel (Fig.2b). The MCS mode was specifically aimed at detecting any small burst release of neutrons which can otherwise get averaged out in a longer time duration pre–set time counting. Typically pre–set time counting was done for periods of 6000 s but during the intense burst release of neutrons the pre–set time duration was reduced to around 1000 s to avoid scaler overflow. Any pickup of extraneous signals like those emanating from the operation of pump motors, drilling machines, tesla coils, piezo–electric gas lighter and fluorescent lamps going off and on was thoroughly checked and the counter was found to have good stability and was immune to such electrical disturbances. A constant background of about 1.6 counts/s was registered without the electrolytic cell in operation for a duration of 10 days before setting up the electrolytic cell inside the counter well and also during times when the electrolysis was continuing but no burst emission of neutrons occurred.

Accumulation of tritium in the electrolyte was monitored by withdrawing periodically 1 cm$^3$ samples from the cell at about 6 days intervals and measuring the T content using gel liquid scintillation "cocktail" and a Packard counting system. The counting efficiency was about 25%. The chemiluminescence effect due to the presence of LiOD was seen in the samples and counting was done till such effects died down completely and stable count rates were obtained.

During certain periods of electrolysis the electrolytic gases from the cell were recombined over a Pt/polyester fabric catalyst and collected in a cold trap, and the resulting D$_2$O was checked for the presence of tritium. Loss of D$_2$O due to electrolysis and evaporation was made up by periodic addition of pure D$_2$O while losses due to samples drawn from the cell for tritium analysis were made up by adding 0.1 mol dm$^{-3}$ LiOD solution in D$_2$O. As stated previously the D$_2$O used for the experiment and for replenishments was from a single stock of heavy water. Temperature of the electrolyte was monitored by a chromel–alumel thermocouple encased in a glass tube dipping in the solution.

The cell was run for 32 days mostly at a current density of about 60mA/cm$^2$. After the completion of the electrolysis run the Pd cathode was disassembled from the cell and connecting leads and dried. The absorbed gases from the cathode were recovered by heating it at 680 K in an evacuated chamber. From the pressure volume relationship and gas Chromatographic analysis of the deuterium content the volume of absorbed D$_2$ gas was computed to be 320 cm$^3$ at STP. This gas was then equilibrated over CuO at 680 K till there was no further reduction in volume and the water formed (<~ 0.3 cm$^3$) was collected in a thimble cooled to liquid N$_2$ temperature and this water also was counted for T.
Results

a) Burst neutron emission: Fig. 2 shows the observed burst neutron emission which lasted essentially for about 40 hr during the operation of the cell for a period of 32 days. This figure represents 4000 data points collected in the MCS mode with each of these data points being neutron counts in a 40 s time channel. A portion of the steady background counts recorded prior to the beginning of the burst emission has been included to show the burst intensities in relation to this background. Fig. 2a to 2j represent the observed burst neutron emission in a ten time expanded time axis. Upto 14 days of electrolysis (charge passed -2.24 x 10^6 Coulombs) only background neutron counts were observed and again after the 17th day of electrolysis (charge passed -2.5 x 10^6 Coulombs) till the end of the run i.e., 32nd day (total charge passed -3.65 x 10^6 Coulombs) only background counts were seen. During the neutron emission period (15th-17th day of electrolysis), the bursts were intense up to about 14 hr into the emission with intervening quiescent durations (in which counts were close to background) varying from as short as about 120 s and as long as 3 h. In the later stages the burst intensity decreased but bursts became more frequent so much so near the end they appeared to be continuous. The maximum burst signal (at about 14th hour into the emission) corresponded to 142000 counts in the 40 s time channel as against 50-70 counts registered for the background. This corresponds to a maximum signal to background ratio of about 2000. Using the 8.6% efficiency of the neutron counter the total number of neutrons emitted in these bursts work out to 1.8 x 10^8 in a period of about 40 hr which corresponds to a pseudo average emission rate of 1.3 x 10^3 n/s.

The electrolyte temperature was varied in the initial stages of the run by changing the applied voltage to the cell or by changing the set temperature of the thermostatic water bath. Ten hours before the burst emission and during the burst emission, temperature of the electrolyte was not altered and it remained in between 296 K and 300 K. Electrolyte temperature at any time of electrolysis remained above 293 K.

b) Tritium in electrolyte and electrode: Tritium counting of samples (Table I) drawn from the 5th-17th days after the burst neutron emission (sample nos. 4, 5 and 6 in Table I) showed an increase from 0.4 to 13 Bq cm^-3 Low level counting methods (60 minute count duration) coupled with a stable system background yielded a standard deviation of only 3% on the count rates. The cell electrolyte volume being 250 cm^-3, an increase of 0.4 to 1.3 Bq cm^-3 in tritium activity shows an extra input of T to the extent of 100 to 325 Bq, amounting to 5.6-18.0 x 10^10 extra tritium atoms accumulating in the electrolyte probably as DTO. It is known that at temperatures above 293 K, electrolysis of D_2O does not result in significant enrichment of tritium in the electrolyte. In the present experiment electrolytic gases recombined over a catalyst and counted for tritium have shown tritium escaping from the cell. Hence the observed excess of tritium activity points to an extra source of tritium on the conservative side, i.e. not accounting for the tritium that is carried away in the gas stream during the electrolysis.

Degassing of the Pd cathode at 680 K at the end of the experiment yielded 320 cm^-3 of D_2 gas at STP. By reaction of this gas over CuO turnings at 680 K and quantitatively collecting the reformed water and counting for tritium has shown 5.4 Bq (3 x 10^9 atoms of T) accumulating in the Pd cathode.

c) Electrode potential: Fig. 3 indicates the variation of cathode potential and neutron emission rate (deduced from the scaler counts obtained with pre-set time) with time. A potential of -1.0 V SCE when the polarisation voltage was switched off indicates that the electrode in 0.1 mol dm^-3 alkali concentration behaves as a near hydrogen (in the present case deuterium) electrode. Monitoring potentials some hours into the burst emission period revealed fluctuations in the working electrode potentials. The neutron count rate peaks
seem to coincide with the shift of potentials to more positive values by as much as 0.7 volts in the cell under "off" and by 3 volts in the "on" conditions.

d) Heat Output: No discernible extra heating over the background Joule heating was observed in this experiment.

Discussion

The present study has shown that for identifying whether cold fusion occurs or not, monitoring neutrons is a much more sensitive method than monitoring tritium, especially when the fusion events generating tritium are not very high. This is because of the small value of the decay constant of tritium which requires an accumulation of $10^9$ atoms to register 1 Bq of tritium activity. In the present case although the neutron emitting events computed from the integrated neutron counts amounted to only $10^8$ as compared to about $10^{10}$ to $10^{11}$ events computed from tritium data, the neutron emission occurred with big spikes and bursts. As against this, $10^{11}$ events through the tritium channel were inferred from an increase of 1.3 Bq/cm³ extra tritium activity observed at the end of the experiment. Since the amount of T held in the Pd cathode at the end of experiment is only 5.4 Bq, it looks as if the generated T (about 325 Bq) mostly diffuses into the electrolyte rather than getting retained in the Pd matrix which is somewhat akin to the neutron emission and suggests the possibility of fusion events occurring close to the surface. From the variation in T activity at different times during the course of electrolysis (Table I) it appears that T input into the electrolyte started after the burst neutron emission occurred. This is because the 16th day sample which was taken during the burst neutron emission registered the same T activity as the sample taken in the initial stages of run. But during the T generation period (17th—32nd day) there was no neutron emission over the background rate.

From the volume of D₂ gas trapped inside the Pd electrode (320 cm³ at STP actually collected), the composition of Palladium—deuteride that would have given rise to the burst n emission was inferred to be between PdD₀.₃–₀.₄. Using the known diffusion coefficients of D < $1 \times 10^{-7}$ cm²/s it was computed that through the 1 mm thick cathode used in this study, D atoms can enter and come out through the other side in about 30 hours time thus pointing to the attainment of at least PdD₀.₂₅ configuration in about a day's time. But, as shown by D₂ gas trapped in the cathode at the end of the experiment (32 days), only at the end of the 32nd days this composition seems to have been reached. The reason for this could be the blackish loose coating forming on the Pd Cathode during the course of the experiment which perhaps acted as a diffusion barrier for the entry of D atoms. EDXF analysis of this coating revealed it to be platinum which might have arisen from anodic dissolution of Pt at the applied cell voltages (which were higher than the reversible Pt/Pt²⁺ potential). The reason for not sustaining the burst neutron emission for more than three days could be summarised as follows: Those sites in Pd matrix in which fusion of D atoms occurred might have been the really active sites and once D atoms got depleted from those sites further replenishment did not take place possibly either because of the barrier coating which only favoured molecular D₂ formation from the cathode surface with no further entry of D atoms into the matrix.

A Pd electrode dipped in an alkaline solution and on whose surface D₂ is evolving is expected to behave as a normal OD⁻/D₂ electrode registering a potential of $-1.01$ V vs SCE. However, in the case of Pd since it absorbs deuterium, the overpotential for D₂ evolution is expected to increase and the polarisation voltage must be adequate enough for evolving D₂ on the electrode surface by electrolysis. However, in the present even after charging the Pd for a number of days, the overpotential obtained was negligible though degassing the cathode at the end of the experiment has shown that it had absorbed 320 cm³
of D₂ gas at STP. The shift to more positive potential values by as much as 0.7 volts in the cell coinciding with the neutron count rate peaks means a possible depletion of deuterium gas at the surface of the electrode. Thus monitoring the cathode potential has thrown some light of what is happening to a Pd electrode which has absorbed deuterium.

In general very few groups have been able to reproduce the large heat output reported by Fleischmann et al¹ and Mathew et al.⁴ Even to match a cooling rate of 0.1 K per minute in a cell of dimensions reported in the present paper it would require the occurrence of fusions at a steady rate of about 10¹³/s. As discussed above, the observations of n bursts revealed that the occurrence of fusions is not at a steady rate of the desired magnitude and hence there is no wonder that the excess heat output went undetected. In our laboratory we have carried out several runs on electrolytic cells using Pd cathodes of various shapes and dimensions (including a solid cylinder 1 cm diameter and 1 cm long) and Pt anodes in 0.1 mol dm⁻³ LiOD/ D₂O along with control as Pd cathodes and Pt anodes in 0.1 dm⁻³ LiOH/ H₂O purely with a view to observe the excess heat output and were not able to observe the same within the limits of uncertainty of our calorimetry, which is estimated to be about ±10%.

Conclusions

The present study has shown evidence for cold fusion phenomenon in an electrolytically charged Pd matrix in terms of neutron and tritium as the signatures. The tritium channel seems to be favoured over the neutron channel. During the period of our experiment a total of 10¹¹ fusion events leading to tritium generation were observed whereas the neutron channel accounted for only 10⁸ fusion events. No significant heat output over the Joule heating could be observed. The present study has also revealed that the effect is small and not a sustained one. Energy production from cold fusion of deuterium in an electrolytically charged Pd matrix in a sustained manner may require more systematic exploration to identify the various parameters governing the occurrence of the process, not the least important among which is the proper pretreatment of the electrode.

Acknowledgements

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References


TABLE I

TRITIUM CONTENT IN THE ELECTROLYTE (0.1 M LiOD) OF Pd–Pt ELECTROLYSIS CELL

<table>
<thead>
<tr>
<th>Sample</th>
<th>Duration of Electrolysis (d)</th>
<th>Cumulums Passed (X 10^-6)</th>
<th>Tritium Activity (Bq/ml)</th>
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<tr>
<td>1</td>
<td>0</td>
<td>0</td>
<td>3.65</td>
</tr>
<tr>
<td>2</td>
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<tr>
<td>3</td>
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<td>4.34</td>
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<td>4.24</td>
</tr>
<tr>
<td>6</td>
<td>32</td>
<td>3.55</td>
<td>5.29</td>
</tr>
</tbody>
</table>

* Standard deviation ± 3%
FIG. 1 EXPERIMENTAL ASSEMBLY - SCHEMATIC.

LEGEND —
1, 2 - INNER CYLINDRICAL ANODE (Pf)
3, 4 - CATHODE RING (Pd)
5, 6 - OUTER CYLINDRICAL ANODE (Pf)
7 - GAS INLET
8 - GAS OUTLET
9 - LUGGIN PROBE FOR CALOMEL ELECTRODE
10 - Cr - Al THERMOCOUPLE
11 - D₂O REPLENISHMENT INLET

WATER FROM WATER BATH
SAMPLING POINT FOR TRITIUM IN ELECTROLYTE

0.1M LiOD ELECTROLYTE (250 ml)
HEAT EXCHANGER

PERISTALTIC PUMP

QUARTZ CELL

TO WATER BATH
Fig. 2 Burst neutron emission in Multi Channel Scaling mode with time
Fig. 2a - 2j Burst neutron emission in multichannel scaling mode with time - expanded plot of Fig.2
Fig. 3 - Burst neutron emission rate and cathode potential variation with time
VERIFICATION STUDIES ON ELECTROCHEMICALLY INDUCED FUSION OF DEUTERONS IN PALLADIUM CATHODES


*Reactor Operations and Maintenance Group.
†Isotope Group.

Introduction

In continuation of the R&D work carried out earlier on electrochemically induced fusion of deuterons, a series of experiments were carried out using palladium cathode and platinum wire gauze anode. Some of the main features of the experiment were as follows:

- The electrolysis experiments were conducted using 0.1 M LiOD solution in D₂O using 1 x 1 x 1 cm cube of palladium as cathode and platinum wire gauze as anode.
- The experiment lasted for seven weeks during which data on heat measurements, tritium production and neutron emission were collected.
- The amount of deuterium-loading on palladium electrode was measured at the end of the experiment.
- Blank experiments with stainless steel cathode in place of palladium and H₂O in place of D₂O were also carried out.

Experimental

Cathode

(a) Palladium Cube Cathode: The spectrographic analysis showed the palladium content to be 98.2%. The impurities were mainly Cu, Ca, Fe, Pb, Ni, etc. The cathode was heated under vacuum and cleaned electrochemically before use.

(b) Stainless Steel Cube Cathode: 1 cm x 1 cm x 1 cm cube of stainless steel (Type 3004 L – Fe 72%, Cr 18%, Ni 8%) was used after polishing the surface and cleaning with distilled water.

The palladium cathodes after use in the electrolysis experiments were generally degassed at high temperatures (850°C) and heated in vacuum for a few hours for reuse.

Electrolyte

The electrolyte solution of 0.1 M LiOD in D₂O was prepared by dissolving lithium metal in D₂O having an isotopic purity of above 99.86% w/w.
- The volume of solution used in the electrolysis cell was 140 ml.
- In the case of some of the blank experiments with H₂O, the electrolyte solution of 0.1M LiOH in H₂O was prepared by dissolving lithium metal in pure double distilled water.

Experimental Assembly

Fig.1 gives a schematic diagram of the assembly. Fig.2 gives the details of the assembly used along with Fig.1 for recovery and conversion of gases (D₂ and O₂) liberated during the electrolysis. It may be mentioned that a palladium catalyst was used for recombination of D₂ and O₂ and hot copper oxide was used for conversion of D₂ into D₂O.
- The recovery assembly enabled assessment of recombination of D₂ and O₂ emerging from the electrolytic cell and also permitted estimation of the absorbed D₂ gas.
liberated from the palladium cathode after the electrolysis was terminated.

- The experimental assembly in Fig. 1 (Cell) was covered with thermocole insulations open to ambient air during the initial stages when data on heat measurements were collected. Subsequently the cell was cooled only by air and during the later part of the electrolysis, the cell was cooled by cold water.
- A resistance wire heater was placed in the cell to collect data on electrical heating without electrolysis.
- Temperature of the electrolyte solution was monitored by a Pt—100 RTD placed in the cell and it was recorded continuously on a chart. In the initial stages of heat measurements, a mechanical stirrer was used for mixing the electrolyte. Subsequently the cell was stirred with the bubbling of nitrogen gas from a cylinder.

Electrolysis

Electrolysis was carried out at constant current and variable voltage. At the end of 1365 ampere—hours, current was changed to pulsing mode (the cycle being 0.5A for 1 second followed by 2.5A for 11.5 second) for about three hours and then to the cycle 0.5A for 1 second followed by 4.5A for 1.5 second.
- The latter lasted for about ten minutes when an explosion occurred inside the cell resulting in the cell lid being thrown out with the electrode assembly. The electrodes were intact but were found electrically shorting. The electrolysis was terminated and the D₂ gas released by the palladium cathode electrode was passed through the assembly of palladium catalyst and copper oxide for conversion of D₂ to D₂O.

Neutron Detection

- For neutron detection, a BF₃ neutron detector with a counting efficiency of 5 X 10⁻³ cps/ fast neutron, was placed below the electrolysis cell. An additional BF₃ detector was kept about two meters away from the cell to monitor background neutrons.

Sampling and Assay

- The cell electrolyte as well as the D₂O collected in the various cold traps in the attached assembly were periodically sampled during the course of the experiment, for tritium measurement. The D₂ gas released by the palladium electrode (after conversion of D₂O) was also analysed for tritium content. All the tritium measurements were done using LKB system Model 1215, RACK BETA—II system.

Blank Experiments

- Blank experiments were carried out following a similar procedure using a combination of palladium and stainless steel cathodes. D₂O and H₂O electrolyte solutions, and following the electrochemical parameters as given in Table I. The approximate duration of each blank experiment was about 16 to 20 hours.

Heat measurements

For measurement of the heating rate in the electrolysis cells, the cell temperature rise versus time data were collected for different current density values till the cells attained thermal equilibrium. Also steady state cell temperature data at different stages of electrolysis with increasing deuterium loading in palladium were collected.
Fig 3 gives the variation of equilibrated cell temperature rise with Joule heating of the cell. The cell thermal sensitivity expressed as the ratio of steady state cell temperature rise (with respect to heat sink temperature) to Joule heat input power, remained almost constant in all experiments with palladium/ stainless steel as cathodes and D₂O/ H₂O solutions of lithium as electrolyte. This seems to indicate that the electrolysis cells behaved similarly so far as their heat production characteristics are concerned. However this observation does not specifically resolve presence or absence of excess heating in the cells. To examine this aspect further, the cell temperature versus time data were analysed using a non-linear least square fit algorithm to obtain the cell calorimetric heating rate employing a "time—constant" method. The calculation method was verified to yield satisfactory results (within about 10%) for simulated electrical resistance heating measurements of a typical cell configuration. Results showed that, compared to the cell Joule heating, the cell calorimetric heatings were consistently higher within experimental and calculational errors for all the cells as shown in Fig. 3.

Table 1 summarizes the results. The ratio between the calculated cell calorimetric heating to the Joule heating rates varied in the range 1.4 to 2.3 for Pd—D₂O system, 1.1 to 1.4 for SS—D₂O systems, 1.6 to 1.9 for Pd—H₂O systems, 1.5 to 1.9 for SS—H₂O systems. (It was also experimentally established that the D₂ and O₂ gases do not recombine significantly within the cell volume which could lead to cell calorimetric heating rate higher than the Joule heating rate).

For the Pd—D₂O cell, the excess over Joule heating rates computed were used to estimate the extent of known nuclear fusion reaction rates. To allow for possible heating contributions from any chemical reactions in the Pd—D₂O cell, the excess over Joule heating rates computed for the blank experiments with Pd—H₂O, SS—D₂O and SS—H₂O cell were least square fitted as a straight line function of cell current. Due to similarity in cell geometry and cell contents, the least square fit results were assumed to be applicable for the Pd—D₂O cell as well.

If one were to assume that the extra excess heating over Joule heating rates thus obtained for the Pd—D₂O cells for various cell currents was due to known nuclear fusion reactions with deuterons, then one would obtain the fusion rates per second per d—d pair to be approximately 7.3 X 10⁻¹¹ assuming only D(d,n)He³ reaction, 6.2 X 10⁻¹¹ for only D(d,p)T reaction and 1.0 X 10⁻¹¹ for only D(d,γ)He⁴ reaction.

When the Pd—D₂O main experiment was conducted in current pulsing mode with low heat sink temperature environment, the cell experienced a temperature transient of about 25°C over a period of 8 minutes. The cell had a mild explosion which dismantled the cell configuration.

Subsequently blank experiments with near simulated heat transfer conditions (as in the main experiments prior to the temperature transient) with low heat sink temperatures were done.

These experiments showed that typically for an electrical power input of about 40 watts,

(a) the cell temperature stabilizes at about 27°C in about 15 minutes with a low heat sink (water bath) temperature of 4°C,

(b) when the heat sink water was drained (as happened during the explosion event) the cell temperature increased at the rate of about 2°C/minute, and
a low temperature heat sink in the form of ordinary ice (as observed prior to explosion) was not efficient to reduce the cell temperature substantially.

Further with an electrical power input of -115 watts into the cell (as existed during the current pulsing conditions) the cell temperature could rise by about 6.5°C/minute even with low (water bath) heat sink temperature of 4—5°C.

These experiments thus showed that the observed cell temperature transient could perhaps be due to inadequate heat transfer from the cell surfaces or due to high electrical energy input into the cell during the pulsing experiments. The reason for the mild explosion however has not been understood.

Tritium formation

For a proper assessment of production of excess tritium in the electrolysis cell, known data for the input tritium in the total D₂O used in the experiment and for the tritium analysed in the various samples including that from the palladium electrode, were used. Table II gives tritium results. For the unaccounted D₂O i.e. D₂O which could not be accounted in the daily D₂O material balance by make up and recovery, a tritium value as shown in Table II was taken, based on the tritium value experimentally obtained for the recovered D₂O. Thus it is seen that a net tritium excess corresponding to 50 percent of the total input tritium was observed. The measured excess tritium production of 0.0348 μCi for the main electrolysis experiment corresponds to an average fusion rate of $2.3 \times 10^{18}$ fusions per d–d pair per second assuming D((d,p)T is the only reaction.

Neutron emission

In the initial stages of electrolysis with Pd–D₂O cell, 17 neutron bursts lasting for 2 to 55 minutes each were observed. The integrated neutron emission in the bursts varied in the range $5.1 \times 10^3$ (burst period = 2 minutes) to $5.4 \times 10^5$ (burst period = 8 minutes). The background neutron level recorded was about 20 neutrons/second during these measurements. However, there was no indication of neutron emission later in spite of the increased loading of palladium with deuterium. In fact, analysis of absorbed deuterium in the palladium electrode after the termination of the experiment gave a value of D/Pd > 2.2 which showed that the electrode was saturated with deuterium. The measured number of neutrons emitted in the bursts observed in the main experiment indicated an average fusion rate of $2.2 \times 10^{21}$ fusions per d–d pair per second assuming D((d,n)³He is the only reaction.

Observations

(a) It may be noted that the fusion rates calculated for the experiment via known nuclear reactions do not appear to tally with each other.
(b) During seven weeks of electrolysis, the Pd–D₂O cell experienced in all three explosions two of which lifted the cell lid.
(c) The amount of D₂ gas released by the palladium electrode after the termination of the electrolysis (duration seven weeks) as well as that obtained by heating the electrode to 800°C, was used to calculate the deuterium-loading value expressed as D/Pd. It was found to be not less than 2.2. The deuterium loading value is an important parameter for calculating the fusion rate per d–d pair. Since some of D₂ absorbed by palladium was lost to atmosphere during the last explosion, the actual deuterium-loading value (D/Pd) is probably close to 3.

Additional experiments are in progress using palladium sheet electrodes to study the cold fusion phenomena further.
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TABLE I

Electrolysis Cells With 1x1x1 cm Cube Cathodes Analysis of Cells Heating Rates

<table>
<thead>
<tr>
<th>S. No.</th>
<th>Date</th>
<th>Cathode</th>
<th>Electrolyte</th>
<th>Cathode Current (mAmp)</th>
<th>T_{cell} (°C)</th>
<th>ΔT_{cell} (°C)</th>
<th>P_{elec}</th>
<th>P_{joule}</th>
<th>P_{calo}</th>
<th>P_{calo/P_{joule}}</th>
<th>Max. # Error</th>
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<td>1</td>
<td>30.6.89</td>
<td>Pd</td>
<td>D_2O</td>
<td>600</td>
<td>59.4</td>
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<td>72.0</td>
<td>48.0</td>
<td>5.8</td>
<td>3.9</td>
<td>7.2</td>
<td>1.85</td>
<td>-0.8</td>
</tr>
</tbody>
</table>

* The cells were operated at the indicated constant currents; T_{cell} denotes stabilized cell temperature and ΔT_{cell} denotes temperature rise w.r.t. ambient air heat sink.

# Denotes maximum deviation in cell temperature computed with least square fitted parameters and corresponding observed cell temperature.

Note: For Pd–H_2O, SS–D_2O and SS–H_2O cells a linear least least square fit gives

\[
(P_{calo}/P_{joule}) = 0.00057 \text{ (mAmp)} + 0.976 \text{ with a standard deviation of 11.7%}.
\]
### Table II

#### Tritium Balance

<table>
<thead>
<tr>
<th>I Tritium Input</th>
<th>Tritium Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>a) Initial volume of D₂O in the cell (ml)</td>
<td>140</td>
</tr>
<tr>
<td>b) Volume of D₂O added for make up (ml)</td>
<td>719</td>
</tr>
<tr>
<td>Total (ml)</td>
<td>859</td>
</tr>
<tr>
<td>Tritium Input from (a) &amp; (b) (μCi)</td>
<td>0.06578</td>
</tr>
</tbody>
</table>

#### II Tritium recovered in the experiment (in μCi)

| a) Tritium in the electrolysis cell at the end | 0.02876 |
| b) Tritium in the vapour condensate | 0.00234 |
| c) Tritium from recombined gases | 0.02297 |
| d) Tritium from D₂O formed in the CuO | 0.00060 |
| e) Tritium from electrode degassing | 0.00922 |
| f) Tritium in samples during electrolysis | 0.00747 |
| g) Tritium in unaccounted D₂O (359 ml) lost during the experiment (i.e. 359 x 181 dpm) | 0.02926 |

Total Tritium in (a) to (g) in II | 0.10062 |

#### III Excess tritium (μCi)

| | 0.03484 |
1. \( \text{D}_2\text{O} \) VOL. IN CELL 140 ml.
2. Pd. 1 cm. x 1 cm. x 1 cm.
3. Pt. WIRE MESH 2 cm. \( \phi \) x 4 cm. HEIGHT.

FIG. 1 CELL FOR ELECTROLYSIS
FIG. 2 ASSEMBLY FOR CONVERTING $D_2 + O_2$ OR $D_2$ to $D_2O$
FIG. 3 Experiments with Pd/SS Cathodes in D₂O/H₂O
TRITIUM ANALYSIS OF SAMPLES OBTAINED FROM VARIOUS ELECTROLYSIS EXPERIMENTS AT BARC

*T S.Murthy, **T.S.Iyengar, B.K.Sen* and T.B.Joseph*

• Isotope Group

** Health Physics Division

Scope

The report summarises the methodology and techniques adopted for the determination of tritium content in various samples obtained during the initial sets of experiments conducted at Trombay in connection with studies on the feasibility of 'Cold Fusion'.

The analyses were carried out at the Isotope Division and Health Physics Division.

Sample Preparation Technique

Sample preparation techniques involved use of the appropriate scintillation 'cocktail' and wherever applicable, the samples were distilled before use. Diluting the sample with double distilled water, even though reduced the number of signal pulses per unit volume of sample, helped in reducing the pH as well as quenching impurities present in the sample. Alternately the sample could be kept for 'chemiluminescence cooling' so that the contribution from the same, if any, is reduced to a negligible level with respect to the sample under counting.

40K free vials are used for low background. Use of Dioxane as solvent was avoided wherever direct counting was adopted, as it tends to show chemiluminescent properties when used in certain samples. The commercially available scintillation 'cocktail' INSTAGEL, (containing a surfactant such as Triton —X—100) was found to be suitable for counting in such situations.

Standardized procedures involved addition of 0.1 to 2 ml of sample in appropriate volume of scintillator in cases where the count rates were high. Larger quantity of sample was taken (8 to 12 ml of sample) in the case of low level samples for better detection limit.

Counting System

There are several LSS systems available for estimation of low-energy beta emitters like $^3$H, $^{14}$C etc. In the present case the LSS system manufactured by M/S Packard Inst. Co. (Model 4530) and LKB system (Model 1215 RACKBETA-II ) were used (Some samples were analysed in an alternate LSS Packard 4530 too, for confirmation of reliability of the method.) The LSS used has facilities for automatic quench correction. The probable errors due to interference due to chemiluminescence is also avoided by adopting appropriate chemical counting methods.

The system stability is checked everyday using sealed tritium standards and sealed background samples in a 'calibration' mode, so that normalization is effected by adjusting the two PMT voltages automatically by the LSS system itself. Standardisation and efficiency of each sample is determined using quench curves developed for each batch of experimental samples by using Quench Standards of the appropriate chemical form.
In the case of heavy water samples used for each set of electrolytic experiments, initial samples were drawn and kept aside and counted along with the samples drawn during the course of the experiments.

The rooms where the experiments are conducted were constantly monitored for tritium contamination from air.

In almost all the experiments samples were drawn at appropriate intervals to follow up the trend in tritium concentration values.

Experiments which have shown definite increase in tritium concentration values are listed separately.

In all the tritium measurements the following factors were considered in arriving at the excess tritium content produced (if any) in the experiment.

a) initial tritium content in the heavy water used for each experiment.
b) concentration of tritium content due to electrolysis.
c) concentration effects due to make-up volumes of the heavy water.

Materials And Method

Tritium Content in Heavy Water Before Electrolysis: Heavy Water used for electrolysis experiments at the Analytical Chemistry Division and Reactor Operations Division are analysed for tritium content. In these two Divisions (ACD and ROD) almost all the experiments were conducted using the heavy water from these stock solutions.

Tritium Content in Palladium Cathodes: For the electrolysis experiments at the Analytical Chemistry Division palladium metal has been used mostly as cathode in different shapes. Before the cathodes were prepared samples of palladium metal was collected for tritium assay. In addition, samples of palladium metal and palladium salts from Radiation Technology Division were also collected and analysed for tritium content.

Palladium metal was dissolved in aqua regia, while palladium salts were dissolved in water and nitric acid. From these solutions suitable stock solutions were prepared. These include

a) acidic solutions
b) neutralised solutions
c) diluted solutions and
d) diluted and neutralised solutions finally made for counting.

In addition, dummy stock solutions were also made with the reagents used except palladium.

The counting was continued for a period of one week. Acidic solutions have shown initially very high chemiluminescence; diluted and neutralised samples after prolonged counting have shown that the palladium and palladium salt do not contain any tritium contamination.

Checking of Lithium Electrolyte: Generally, lithium salts (LiOD) have been used as electrolyte (0.1 Molar) in the electrolysis experiments. In view of this, the lithium solutions were checked for
a) tritium contamination,
b) chemical effects including chemiluminescence etc.,
in the counting of samples from the cells.

For this purpose lithium deuterioxide solution (0.2M) were prepared with known
heavy water (D_2O) stock solution and analysed for both a) and b).

From the counting data the following observations are made:

1. Immediately after preparation, high chemiluminescence was observed. However with
passage of time this chemiluminescence came down to negligible amounts (in about 10 days
time). When these lithium stock solutions were neutralised, the chemiluminescence effect
was greatly reduced to negligible values.

2. When these samples were subjected to distillation and water was collected and
counted, no chemiluminescence was observed. The distilled samples have shown the
original tritium content of the heavy water (D_2O—4) used for this purpose. Therefore no
tritium contamination was noticed in lithium salts used for the electrolysis.

3. However, it has been observed that the electrolyte samples which have been subjected
to few days of electrolysis have shown small amounts of chemiluminescence when compared
to unelectrolysed lithium deuterioxide samples. Initial chemiluminescence which was
observed in some of the electrolysed samples was found to decay rapidly within 24 hours.

4. Therefore, tritium content in the final samples from cells was confirmed by counting
the D_2O distilled from these samples.

Quench Corrections

Due to the presence of chemical impurities even in trace quantities chemical
quenching possibilities exist, which in turn reduce the pulse output. In impurity quench the
components which quench the sample do so either by competing with the fluor for energy
transfer, or by chemically interacting with the fluor molecules to make them less reactive
to energy transfer. Oxidizing agents at high pH can alter oxygen atoms in many of the
fluors so that the fluorescent properties are also changed.

In colour quench, the quenching component absorbs the photons produced by the
scintillation process, before they can be detected by the PMTs. Colour quench usually does
not interfere with the scintillation process but exerts its effect by preventing the photon
being seen by the detector system of the LSS.

In both the above cases the resultant effect will be a compression of the beta
spectrum of tritium and reduction in pulse output, thereby reducing the efficiency of the
LSS. Thus it becomes necessary to correct for such quench errors and arrive at the
efficiency of each one of the samples being counted.

Quench corrections are carried out by any one of the following methods

i) Standard Addition Technique wherein a known amount of standard of appropriate
concentration is added to the sample which determines the efficiency for that particular
sample. In this ‘spike method’ the sample becomes irretrievable.

ii) Sample Channels Ratio Method effectively takes the ratio of two preset channels of
the beta spectrum for the sample and comparing it with a ‘calibration curve’ developed
with a series of known quenchers of appropriate chemical form whose efficiency have been
determined previously. For very highly quenched samples this technique is not quite
suitable as the ratio of the two channels will not reflect true picture of the actual
situations.

iii) Many LSS have built in ‘Automatic External Standard Channels Ratio’ Technique
provided as a facility for quench corrections. The principle depends on pneumatically
shooting a gamma emitting pellet which occupies a place very near the counting vial for a
very short period and produces compton electrons which also undergoes similar quench
effects as that of the sample. Here again a ‘calibration curve’ with a known set of
‘Quenched Standards’ of appropriate chemical composition helps in determining the
efficiency of the sample being counted.

A later version of the same technique developed by Donald Horrocks uses the
compton edge count rates for fixing the channel position for the quenched standards and
calls it ‘H Number’. Each sample being counted will record its H number which provides a
method for determining the efficiency.

Results

Table—I gives the tritium content in the heavy water stock solutions used for
electrolysis experiments conducted at Analytical Chemistry Division and Reactor
Operations Division, and Heavy Water Division, BARC.

Table—II gives the data pertaining to 10 electrolysis experiments conducted at
Analytical Chemistry Division and Reactor Operations Division, BARC.

Table—III From the set of values given in Table II the data where excess tritium has
been observed in the electrolyte solution after electrolysis have been summarised and given.

Table—IV The data from experiments conducted at Heavy Water Division, BARC
are tabulated. It may be pointed out that the correction factors due to make-up volume
contribution in the final tritium content have not been reflected in the table.

Table—V Information regarding the experiments conducted at various Division/
Sections have been given. The samples counted from these experiments have not shown any
apparent increase in tritium values.
### TABLE I
TRITIUM CONTENT OF INITIAL HEAVY WATER USED

<table>
<thead>
<tr>
<th>STOCK NO.</th>
<th>TRITIUM CONCENTRATION (in uCi/ml)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{D}_2\text{O} - 1$</td>
<td>$1.16 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 2$</td>
<td>$0.489 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 3$</td>
<td>$0.845 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 4$</td>
<td>$0.076 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 5$</td>
<td>$0.117 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 6$</td>
<td>$0.27 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 7$</td>
<td>$0.045 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\text{D}_2\text{O} - 8^*$</td>
<td>$0.055 \times 10^{-3}$</td>
</tr>
</tbody>
</table>

*This value was given by HWD as the average value of initial tritium content in $\text{D}_2\text{O}$ used for the experiments HWD (B) and HWD (C).*
### TABLE - II

**TRITIUM COUNTING DATA IN ANALYTICAL CHEMISTRY DIVISION EXPERIMENTS**

All cathodes: palladium metal  
All anodes: platinum

<table>
<thead>
<tr>
<th>Initial D₂O volume added (ml)</th>
<th>Final D₂O volume (ml)</th>
<th>Electrolyte.</th>
<th>Electrolyte use</th>
<th>Tritium data</th>
<th>Net tritium activity</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>D₂O used uCi/ml</td>
<td>Electrolysis and activity</td>
<td>Tritium sample volume correction uCi/ml.</td>
<td>uCi/ml</td>
<td>C</td>
</tr>
<tr>
<td></td>
<td></td>
<td>A</td>
<td>B</td>
<td>B-C</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>45</td>
<td>50</td>
<td>45</td>
<td>52.2</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>65</td>
<td>52.6</td>
<td>65</td>
<td>296</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>60</td>
<td>85</td>
<td>60</td>
<td>231</td>
<td>0.1 M NaOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>75</td>
<td>55</td>
<td>75</td>
<td>174.08</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>75</td>
<td>326</td>
<td>75</td>
<td>844</td>
<td>0.113 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100</td>
<td>1113.2</td>
<td>100</td>
<td>2510.7</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>100</td>
<td>40</td>
<td>100</td>
<td>61.83</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>78</td>
<td>74</td>
<td>78</td>
<td>76</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td></td>
<td>78</td>
<td>20</td>
<td>78</td>
<td>25.6</td>
<td>0.1 M LiOD</td>
</tr>
</tbody>
</table>
### TABLE III

**TRITIUM COUNTING DATA IN EXPERIMENTS WHERE EXCESS TRITIUM WAS OBSERVED.**

- All cathodes: palladium metal  
- All anodes: platinum

<table>
<thead>
<tr>
<th>Initial D$_2$O volume added (ml)</th>
<th>D$_2$O used by sample (ml)</th>
<th>Electrolyte</th>
<th>D$_2$O activity uCi/ml</th>
<th>Tritium data remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Initial vol. of D$_2$O (ml)</strong></td>
<td><strong>D$_2$O added (nl)</strong></td>
<td><strong>Amp. hrs.</strong></td>
<td><strong>Tr. activity</strong> uCi/ml</td>
<td><strong>Remarks</strong></td>
</tr>
<tr>
<td>Expt(i)</td>
<td>45</td>
<td>50</td>
<td>45</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td>52.2</td>
<td>0.845x10$^{-3}$</td>
<td>0.45</td>
<td>1.76x10$^{-3}$</td>
</tr>
<tr>
<td>Expt(ii)</td>
<td>65</td>
<td>52.6</td>
<td>60</td>
<td>0.1 M LiOD</td>
</tr>
<tr>
<td></td>
<td>296</td>
<td>0.489x10$^{-3}$</td>
<td>0.239</td>
<td>0.385x10$^{-3}$</td>
</tr>
</tbody>
</table>

---

**Remarks**

- RCS-2: 237 74 237 73.3 0.1 M LiOH 0.076x10$^{-3}$ 2.6x10$^{-3}$ 0.1x10$^{-3}$ 2.5x10$^{-3}$ Excess tritium observed.
- RCS-3: 72 20 76 25.6 0.1 M LiOH 0.076x10$^{-3}$ 6.8x10$^{-3}$ 0.098x10$^{-3}$ 6.73x10$^{-3}$ Excess tritium observed.
<table>
<thead>
<tr>
<th>Initial D.O</th>
<th>Final D.O</th>
<th>Tritium data</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>volume added (ml)</td>
<td>D.O used (ml)</td>
<td>Electrolysis activity (uCi/ml)</td>
<td>D.O activity after tritium correction (uCi/ml)</td>
</tr>
<tr>
<td>Expt(A)</td>
<td>250</td>
<td>1650</td>
<td>250</td>
</tr>
<tr>
<td>Expt(B)</td>
<td>115</td>
<td>300</td>
<td>135</td>
</tr>
<tr>
<td>Expt(C)</td>
<td>1000</td>
<td>6000</td>
<td>1000</td>
</tr>
<tr>
<td>Expt(D)</td>
<td>250</td>
<td>1296</td>
<td>250</td>
</tr>
</tbody>
</table>

* These two values were given by IIWD as the average values of initial tritium content in D.O used for the experiments IIWD (B) and IIWD (C).
TABLE V
TRITIUM CONCENTRATION DATA
IN OTHER ELECTROLYSIS EXPERIMENTS

<table>
<thead>
<tr>
<th>Division</th>
<th>No. of Expts.</th>
<th>No. of samples Collected</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>Physic.Chem.Sec.</td>
<td>2</td>
<td>20</td>
<td>Pd-Ni system: LiOD (0.1 M): Ionic membrane used between electrodes: Vol. and cell dimensions changed in different cells.</td>
</tr>
<tr>
<td>Radiation Chem. Sec.</td>
<td>2</td>
<td>14</td>
<td>Pt-Pd system: LiOD 0.01 M Control Exp. with D$_2$SO$_4$</td>
</tr>
<tr>
<td>Water Chem Div.</td>
<td>3</td>
<td>33</td>
<td>Pt-Pd system: LiOD 0.1M Electrode size varied: One cell exploded.</td>
</tr>
</tbody>
</table>

NOTE: NO APPARENT INCREASE IN TRITIUM CONCENTRATION WAS OBSERVED IN THE ABOVE EXPERIMENTS.
ANNEXURE

LIST OF SAMPLES ANALYSED FOR TRITIUM CONTENT AT HEALTH PHYSICS DIVISION (BARC) BY LIQUID SCINTILLATION COUNTING

LSS Used : Packard (Model 4530)
System Bkg for full Channel : <12 cpm (0-19 keV)
Real Time Monitoring for ambient gamma fields? : Yes (in 3rd channel)
System Stability Check? : Yes (everyday)
Corrections made for Quench Corrections :
- a) Chemi- and Photoluminescence
- b) H₂O₂/D₂O presence if any
- c) Chemical and Photon quenching
- d) Volume Quenching (D₂O)

Quench Corrections : By Spectral Index Sample (SIS) and Spectral External Standard (SES) in the Packard LS System. Cross check with Sample Channels Ratio (0-19 keV and 2-19 keV regions) is also done.

<table>
<thead>
<tr>
<th>Division</th>
<th>Experiment Ref.</th>
<th>No. of Samples</th>
</tr>
</thead>
<tbody>
<tr>
<td>Heavy Water Div. *</td>
<td>M Series</td>
<td>55</td>
</tr>
<tr>
<td>Chemistry Div. *</td>
<td>V Series</td>
<td>50</td>
</tr>
<tr>
<td></td>
<td>D Series</td>
<td>35</td>
</tr>
<tr>
<td></td>
<td>P Series</td>
<td>44</td>
</tr>
<tr>
<td></td>
<td>H/HN Series</td>
<td>30</td>
</tr>
<tr>
<td></td>
<td>Gp Series</td>
<td>8</td>
</tr>
<tr>
<td></td>
<td>CG Series</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>K Series</td>
<td>20</td>
</tr>
<tr>
<td></td>
<td>R Series</td>
<td>2</td>
</tr>
<tr>
<td>Analyt. Chem Div.</td>
<td>S Series</td>
<td>4</td>
</tr>
<tr>
<td>Metallurgy Div.</td>
<td>G Series</td>
<td>3</td>
</tr>
</tbody>
</table>

* The identification and interpretation of various sets of experiments are given by the respective groups separately, elsewhere.
NOTE Chemistry Division Experiments are planned such that different parameters are studied independently by different groups.
MATERIAL BALANCE OF TRITIUM IN ELECTROLYSIS OF HEAVY WATER

S.K. Malhotra, M.S. Krishnan and H.K. Sadhukhan

Heavy Water Division

Introduction

Electrolysis of heavy water has acquired great importance during the last few months in view of the discovery of the phenomenon of cold fusion\textsuperscript{1-2}. Though the initial papers reported measurements only of neutron emission and/or heat, lately tritium also has been reported to be forming via this phenomenon\textsuperscript{3}. However any tritium measurements carried out should be corrected for the isotopic enrichment of tritium during electrolysis since it is a well known technique for hydrogen isotope separation. In this short note we have attempted to give a complete material balance of tritium escaping the system in the form of DT gas and also as DTO vapour. Tritium produced in excess of what is predicted from this equation may be attributed to nuclear fusion reactions.

Basic Material Balance Equation

Consider an electrolyser (Fig.1), in which the deuterium gas formed is being let out, D\textsubscript{2}O vapour is also escaping the system along with the gases evolved and heavy water is continuously being added to the electrolyser to maintain constant level of electrolyte in the cell.

For low tritium concentrations, all the tritium in liquid and gas phase will be in the form of DTO and DT respectively. Now the rate of increase in the number of DTO moles in the electrolyser per unit time, can be written as,

\[
\frac{d n_{\text{DTO}}^{\text{(EL)}}}{dt} = \frac{d n_{\text{DTO}}^{(F)}}{dt} - \frac{d n_{\text{DTO}}^{(V)}}{dt}
\]

Where the term with (F) is the rate of addition of DTO in moles per second, being added with the feed, the term with \(n_{\text{DTO}}\) is rate of escape of DT with gas and the term with (V) is the rate of escape of DTO as vapour.

The rate of escape of DT can be calculated from the knowledge of the electrolytic separation factor for tritium. Thus if \(n_{E}\) moles per sec. are electrolysed and \(\alpha_{E}\) is the electrolytic separation factor then for low tritium concentration \(\alpha_{E}\) can be defined as,

\[
\alpha_{E} = \frac{(T/D)_{\text{liq}}}{(T/D)_{\text{gas}}}
\]

The number of DT moles escaping per second will be,

\[
\frac{d n_{\text{DT}}}{dt} = \frac{n_{E}}{\alpha_{E}} x
\]

where \(x\) is the mole fraction of DTO in the electrolyser at time \(t\).

The rate of escape of DTO as vapour can be calculated from the knowledge of the vapour
pressure, \( P_v \) of heavy water at the temperature of electrolysis, pressure \( P \) in the electrolyser and flow rate, \( F \) of the gases evolving out of the electrolyser and the separation factor \( \alpha_v \) for isotopic fractionation of tritium due to evaporation. Thus,

\[
\frac{d}{dt} n_{\text{DTO}} = \frac{P_v}{P - P_v} \frac{F}{g} \frac{x}{\alpha_v}
\]

(4)

where \( F_g \) is the flow of gases from the electrolyser in moles per sec.

The term due to the feed of fresh \( \text{D}_2\text{O} \) to the electrolyser can be written as

\[
\frac{d}{dt} n_{\text{DTO}} = F_L \cdot a
\]

(5)

where \( F_L \) is the feed rate of heavy water in moles per sec. and \( a \) is the mole fraction of DTO in the feed. Equation can now be written as

\[
\frac{d}{dt} n_{\text{DTO}} = F_L \cdot a - \frac{n_E \cdot x}{\alpha_E} - \frac{P_v}{P - P_v} \frac{F}{g} \frac{x}{\alpha_v}
\]

(6)

Also if the electrolyser contains \( N_t \) moles of heavy water at time \( t \) then,

\[
x = \frac{n_{\text{DTO}}}{N_t}
\]

(7)

Therefore,

\[
\frac{dx}{dt} = \frac{1}{N_t} \frac{d}{dt} n_{\text{DTO}} - \frac{d}{dt} \frac{n_{\text{DTO}}}{N_t} \cdot \frac{d}{dt} N_t
\]

(8)

or,

\[
\frac{dx}{dt} = \frac{1}{N_t} \left[ \frac{d}{dt} n_{\text{DTO}} - x \frac{d}{dt} N_t \right]
\]

(9)

Substituting for \( \frac{d}{dt} n_{\text{DTO}} \) from equation 6 in equation 9,

\[
\frac{dx}{dt} = \frac{1}{N_t} \left[ F_L \cdot a - \frac{n_E \cdot x}{\alpha_E} - \frac{P_v}{P - P_v} \frac{F}{g} \frac{x}{\alpha_v} - x \cdot \frac{d}{dt} N_t \right]
\]

(10)

This is the basic differential equation which can be solved for \( x \) knowing all the other parameters. Some typical cases for application to different operational conditions are considered in the following sections.

Case 1: Diffusion Type Electrolyser with automatic Level maintainer: This case particularly refers to an electrolyser used by us (Milton Roy) in our experimental studies on cold fusion\(^4\). In this electrolyser deuterium diffuses through the palladium cathode and is delivered in absolutely dry condition. The level of electrolyte is automatically maintained. For such an electrolyser the total number of moles in the electrolyser, \( N_t \) is constant and therefore \( N_t = N \) and \( \frac{d}{dt} N_t = 0 \). The term \( F_g \) in equation 8 will be the rate of
evolution of oxygen and therefore,
\[ F = \frac{1}{2} n_E \]  
(11)

Also the feed rate of heavy water will be equal to the total of the rate of electrolysis and loss of heavy water due to evaporation. Therefore,
\[ F_L = n_E + \frac{1}{2} n_E \cdot \frac{P_v}{P - P_v} \]  
(12)

Equation 10, now becomes,
\[ \frac{dx}{dt} = \frac{n_E}{N} \left[ \left( 1 + \frac{P_v}{2(P - P_v)} \right) a - \left( \frac{1}{\alpha_E} + \frac{P_v}{2\alpha_v(P - P_v)} \right) x \right] \]  
(13)

Putting,
\[ \left( 1 + \frac{P_v}{2(P - P_v)} \right) = A \]  
(14)
and,
\[ \left( \frac{1}{\alpha_E} + \frac{P_v}{2\alpha_v(P - P_v)} \right) = B \]  
(15)

and rearranging equation 13,
\[ \frac{dx}{aA - Bx} = \frac{n_E}{N} \cdot dt \]  
(16)

Integrating for \( t = 0 \) to \( t \), we get,
\[ \int_{x_0}^{x} \frac{dx}{aA - Bx} = \frac{n_E}{N} \cdot t \]  
(17)

where \( x_0 \) is the mole fraction of DTO in the electrolyte initially. Equation 17 can be rewritten as,
\[ x = a \cdot \frac{A}{B} - \left( \frac{aA}{B} - x_0 \right) e^{-\frac{B}{N} \cdot t} \]  
(18)

This equation gives the mole fraction of DTO in the electrolyte at time \( t \). As \( x, x_0, \) and \( a \) have same units equation 18 can be applied for any common units of these parameters.

It follows from eqn. 18 that \( x \) will increase or decrease with time depending upon the relative magnitudes of \( a \) and \( x_0 \). Thus for \( x_0 > aA/B \), the exponential term will become positive and \( x \) will increase with time; while for \( x_0 < aA/B \), \( x \) will decrease with time. In both the cases the tritium concentration in the cell will try to attain a steady state value equal to the term \( aA/B \) as shown by the asymptotic behaviour of the tritium curves in the two cases (Fig. 2).

If the tritium concentration of the starting electrolyte and that of feed water is same (\( a = x_0 \)) then eqn. 18 becomes,
\[ x = x_0 \left( \frac{A}{B} - \left\{ \frac{A}{B} - 1 \right\} e^{-\frac{B}{N} n_E \cdot t} \right) \]  

(19)

In this case also \( x \) will increase with time because it easily follows from equations 14 and 15 that \( \frac{A}{B} > 1 \) rendering the exponential term negative.

**Case 2: Batch Electrolysers of non-diffusion type:** In most of the electrolysis experiments the cell employed is such that the mixed stream of deuterium and oxygen is coming out of the electrolyser and the cell is operated for a certain period without adding fresh heavy water to it. In this case the first term in eqn. 10 will be zero (as \( \frac{FL}{F_L} = 0 \)) and also \( F_g \) will now be equal to \( \frac{3}{2} n_E \). Also \( N_t \) will no more be constant and will be given as,

\[
N_t = N - (n_E + \frac{P_g}{P - P_v}) \cdot t
\]

(20)
or,

\[
N_t = N - n_E \left( 1 + \frac{3}{2} \frac{P_v}{P - P_v} \right) \cdot t
\]

(21)

Therefore,

\[
\frac{dN_t}{dt} = -n_E \left( 1 + \frac{3}{2} \frac{P_v}{P - P_v} \right)
\]

(22)

Making these substitutions in eqn. 10 and simplifying, we get

\[
\frac{dx}{dt} = \frac{n_E}{N - n_E \left[ 1 + \frac{3}{2} \frac{P_v}{P - P_v} \right]} \left[ \frac{\alpha_E - 1}{\alpha_E} + \frac{3}{2} \frac{\alpha_V - 1}{\alpha_V} \cdot \frac{P_v}{P - P_v} \right] \cdot x
\]

(23)

Integrating and putting the condition \( x = x_0 \) at \( t = 0 \)

\[
L_n \frac{x}{x_0} = -\frac{\frac{\alpha_E - 1}{\alpha_E} + \frac{3}{2} \frac{P_v}{P - P_v}}{1 + \frac{3}{2} \frac{P_v}{P - P_v}} \cdot L_n \left[ 1 - \frac{n_E}{N} \left( 1 + \frac{3}{2} \frac{P_v}{P - P_v} \right) \right]
\]

(24)

Equation 24 can be employed to calculate the tritium enrichment during the electrolysis in terms of time of electrolysis. Alternatively, this equation can also be written in terms of \( N_t \).

Thus
Using eqn. 25, tritium enrichment can be calculated knowing initial and final volume of the electrolyte. Equation 26 is in fact similar to a well known Rayleigh's equation given by:

\[
\ln \frac{N_t}{N_0} = (1 + H) \left[ \frac{\alpha_E}{\alpha_E - 1} \right] \left[ \ln \frac{x_0}{x_t} - \frac{1}{\alpha_E - 1} \ln \frac{1 - x_0}{1 - x_t} \right]
\]

where \(H\) is the humidity of the gases evolving. For low tritium concentrations \(x_0\) and \(x_t \ll 1\) and therefore eqn. 26 becomes

\[
\ln \frac{N_t}{N_0} = (1 + H) \left[ \frac{\alpha_E}{\alpha_E - 1} \right] \ln \frac{x_0}{x_t}
\]

which is similar to eqn. 25 except that it does not include the term due to isotopic fractionation of tritium due to evaporation. A plot of \(x/x_0\) vs \(N_0/N_t\) as obtained from eqn. 25 is shown in Fig. 3.

Let us now consider the different parameters appearing in equations 18, 24 or 25. They are the electrolysis rate \(n_E\), electrolytic separation factor \(\alpha_E\), distillation separation factor \(\alpha_V\) and vapour pressure of D2O, \(P_v\).

The rate of electrolysis \(n_E\) can be calculated from Faraday's laws of electrolysis knowing the current. The electrolytic separation factor \(\alpha_E\) depends upon the temperature of electrolysis and more so on the electrode materials. Not much data is available for electrolytic tritium separation factors. Generally a value of 2.0 is widely accepted.

The distillation separation factor \(\alpha_V\) can be calculated from the vapour pressure of D2O and DTO. Thus,

\[
\alpha_V = \frac{P_{D2O}}{P_{DTO}}
\]

The vapour pressures of D2O and DTO have been computed and are available in literature. Table 1 gives the values of \(P_{D2O}\) (\(P_v\) in eqns. 18, 25 and 26), \(P_{DTO}\) and \(\alpha_V\) at different temperatures.
References


TABLE 1

<table>
<thead>
<tr>
<th>Temp</th>
<th>( P_{\text{D}_2\text{O}} )</th>
<th>( P_{\text{DTO}} )</th>
<th>( \alpha_v )</th>
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<td>20</td>
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<td>80</td>
<td>0.44210</td>
<td>0.44000</td>
<td>1.0048</td>
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</table>
Fig.1. Input and output of tritium in the Electrolyser

\[
X_0 > a \frac{A}{B}
\]

\[
X = a \frac{A}{B}
\]

\[
X_0 < a \frac{A}{B}
\]

Fig.2. Arbitrary plots of $x$ Vs $t$ Eqn. 18
Fig. 3. Plot of $\frac{X}{X_0}$ Vs $\frac{N_o}{N_t}$ (Eqn. 25)
INTRODUCTION

In nuclear fusion reactions involving D₂ helium may be one of the possible products. During electrolytic dissociation of D₂O with Platinum/ Palladium electrodes if any fusion reaction \(^1,2\) is taking place via the helium pathway with helium escaping to the gas phase it should be possible to detect and estimate the yield quantitatively. Helium is reported to have been observed in excess of the background in the electrolysis experiments conducted at the University of Utah\(^1\). The number reported is of the order of \(10^{12}\) atoms per second (- 0.013 µl per hour). In presence of large excess of D₂ and O₂ generated during electrolysis, detection of He³/ He⁴ in the gas phase poses many problems. Among the methods available for the detection of low amounts of helium, gas chromatography (GC) and mass spectrometry (MS) are normally preferred. However in this case mass spectrometry is complicated since helium is in trace concentration and large excess of D₂ will interfere with the signal due to He³ or He⁴ ions unless fractional mass difference method is adopted. In order to concentrate the helium in the gas phase an experimental technique has been devised wherein D₂ and O₂ generated by electrolysis is catalytically recombined in situ facilitating gas collection over long periods of electrolysis. The results obtained by this technique followed by gas chromatographic analysis are given in this paper. Though GC/ MS analysis of the enriched sample may be more conclusive, a suitable GC/ MS gas inlet interface compatible with D₂/ He is not commercially available.

EXPERIMENTAL

The electrolysis cell and the gas manipulation system is shown schematically in figure 1. This consists of a glass cell carrying a ground glass joint with the cup connected to a mercury manometer with a 20 ml expansion bulb at the bottom, a silicone septum carrying vacuum tight electrode leads and a vacuum stopcock connecting the cell to a modified Toeppler pump with facilities to measure the gas pressure, volume and also to pressurize and transfer the collected gas to a syringe sampling manifold through a three way stopcock\(^3\). The third limb of this stop cock is connected to rotary vacuum pump. The catalyst for recombination of D₂ and O₂ at room temperature is a specially prepared platinum catalyst deposited on a thick synthetic fabric. It is freely suspended on the top inner side of the electrolysis cell. The catalyst has been independently assessed for H₂/ O₂ recombination efficiency and found to have T₁/₂ for H₂ reaction of less than 15 seconds in presence of sufficient O₂ with gas volumes upto about 500 ml. The gaseous products are analysed by gas—chromatography using thermal conductivity detector either (1) on a 5 meter x 3 mm i.d. molecular sieve 5A Column at 25°C with argon carrier gas which gave clear separation between helium and hydrogen for the analysis of trace amounts of helium in the sample or (2) on a 2 meter x 4 mm i.d. molecular sieve 5A column at 25°C with helium carrier gas for the analysis of H₂ (or D₂) O₂ and N₂ in the sample. The lowest amount of helium detectable in the presence of large excess of hydrogen under the experimental conditions is about 0.01 µl (or about 1 ml sample with 10 ppm He). The second column provided a reasonably good analysis for the composition of the residual gas with respect to major constituents. Total volume of the electrolysis cell and the gas manipulation system as well as the volume of each segment of the system has been determined to facilitate computation of the gas volumes and composition at any stage of the experiment. The electrolysis experiment has been carried out in three stages. In these
Experiments 20 ml of 0.1 M LiOH or LiOD is used as the electrolyte. Experiments I and II are carried out with stainless steel Cathode and Anode mounted in concentric tubular configuration with 1 mm electrode spacing. The electrodes are approximately 1.5 cm X 3 cm X 0.04 cm in size. These experiments are used to test the system with respect to leak tightness, gas recombination efficiency, trace helium recovery and analysis. Experiments III to VI are carried out with palladium/platinum electrodes. In this case a palladium plate (1 cm X 1.5 cm X 0.15 cm) is sandwiched between two platinum plates (1 cm X 1.5 cm X 0.05 cm).

Prior to starting electrolysis the entire system including the cell with 20 ml electrolyte is evacuated through the three way stop cock to thoroughly degas the electrolyte. After attaining vacuum the cell is isolated from the pump and rest of the system and the electrolysis started and continued for the required duration by connecting the electrodes to a D.C. power supply. At the end of the electrolysis the power is put off, system is allowed to stabilize till no further change in the mercury level of the manometer is noticeable. At this stage by suitable manipulation of the three stop cocks and mercury reservoir gas transferring, compression, measurement of pressure and volume and sampling for analysis are carried out. Whenever necessary desired reactant gas is introduced into the electrolysis cell through the silicone septum carrying the electrodes by means of a syringe. The electrolysis cell isolated under vacuum is found to retain vacuum for more than 48 hours (drop in mercury level -0.5 cm). The results of the experiments with relevant details are given in Table 1.

Conclusions

1) In contrast to the stainless steel electrodes Palladium cathode/Platinum anode configuration resulted in D₂ deficient gas composition and Platinum cathode/Palladium anode configuration showed O₂ deficient gas. In the initial stages of electrolysis much of the hydrogen (or D₂) may be absorbed by palladium reaching equilibrium in about 2.5 hours as indicated by the steady pressure build up.

2) Though all the released gas is contained and concentrated to a residual volume of 1 to 3 ml, no helium could be detected in all the experiments.

3) Hydrogen (or D₂) absorbed on the electrode is slowly released under vacuum (note e).

4) Platinum catalyst can be incorporated in the cell design to recombine the evolved gas thus facilitating D₂O recovery and also possible recovery of any tritium in the gas phase back into the aqueous phase.

5) In case an independent proof, for the cold fusion via the helium pathway is available, this method can be adopted suitably to detect helium and substantiate the finding with the possibility of unambiguous evidence obtainable from GC/MS analysis of the concentrated residual gas.

Explanatory note to the Table

(a) In case of I and II electrolysis is carried out for varying duration to ascertain the viability of the method and recombination efficiency of the catalyst.

(a*) In these cases 150 µl of 1% He in argon is added to the cell prior to electrolysis and the residual gas at the end analysed for He. Recovery is found to be better than 90%.

(b) (i) In case of III to VI initial evacuation of the system is carried out with the D.C.
potential for about 5 minutes, then evacuation continued with the potential off for about 5 minutes, system isolated and the regular electrolysis carried out (system purging).
(ii) In these cases mercury level in the manometer showed a steady drop for the first 2.5 hrs and then slowed down.

(b**) In case III the residual gas is used up in analysing for trace helium. Hence D$_2$:O$_2$ composition is not determined.

(c) At the end of experiment IV the electrolysis cell is maintained in the isolated mode overnight. The mercury level dropped by about 2 cm. overnight and the residual gas on analysis showed 90% D$_2$ indicating release of D$_2$ under vacuum from the electrode.

Acknowledgement

The author is obliged to Dr. N.M. Gupta and A.D. Belapurkar of Chemistry Division for providing the platinum catalyst. The author is also thankful to Dr. R.M. Iyer, Director, Chemical Group and Dr. J.P. Mittal Head, Chemistry Division for their keen interest and helpful suggestions during the course of this work.

References

## TABLE I
Electrolysis Conditions and the Residual Gas Yields

<table>
<thead>
<tr>
<th>Experiment</th>
<th>I&lt;sup&gt;a&lt;/sup&gt;</th>
<th>II&lt;sup&gt;a&lt;/sup&gt;</th>
<th>III&lt;sup&gt;b&lt;/sup&gt;</th>
<th>IV&lt;sup&gt;b&lt;/sup&gt;</th>
<th>V&lt;sup&gt;b&lt;/sup&gt;</th>
<th>VI&lt;sup&gt;b&lt;/sup&gt;</th>
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<tr>
<td>Electrodes</td>
<td>Concentric S.S. Cathode and Anode</td>
<td>Concentric S.S. Cathode and Anode</td>
<td>Palladium Cathode and Anode</td>
<td>Palladium Cathode and Anode</td>
<td>Platinum Cathode and Anode</td>
<td>Palladium Cathode and Anode</td>
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<td>Electrolyte</td>
<td>LiOH, 0.1M in H&lt;sub&gt;2&lt;/sub&gt;O</td>
<td>LiOD, 0.1M in D&lt;sub&gt;2&lt;/sub&gt;O</td>
<td>LiOD, 0.1M in D&lt;sub&gt;2&lt;/sub&gt;O</td>
<td>LiOD, 0.1M in D&lt;sub&gt;2&lt;/sub&gt;O</td>
<td>LiOD, 0.1M in D&lt;sub&gt;2&lt;/sub&gt;O</td>
<td>LiOD, 0.1M in D&lt;sub&gt;2&lt;/sub&gt;O</td>
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<tr>
<td>Electrolysis Voltage, Current and Duration</td>
<td>4.0 V, -250 mA, 1h &amp; 2h*</td>
<td>4.0 V, -250 mA, 2h, 2h, 4h, 1h, 6h, 6h</td>
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<td>Residual gas Volume after Electrolysis and Composition (25°C/76 cm of Hg)</td>
<td>-1 ml, H&lt;sub&gt;2&lt;/sub&gt;O : O&lt;sub&gt;2&lt;/sub&gt; ~ 2:1</td>
<td>-1 to 3 ml, D&lt;sub&gt;2&lt;/sub&gt;O : O&lt;sub&gt;2&lt;/sub&gt; ~ 2:1</td>
<td>26 ml, ~90% O&lt;sub&gt;2&lt;/sub&gt;</td>
<td>16 ml, ~90% O&lt;sub&gt;2&lt;/sub&gt;</td>
<td>48 ml, ~94% D&lt;sub&gt;2&lt;/sub&gt;</td>
<td>28 ml, ~94% O&lt;sub&gt;2&lt;/sub&gt;</td>
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<td>Final gas Volume and Composition</td>
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<td>-1 to 3 ml, D&lt;sub&gt;2&lt;/sub&gt;O : O&lt;sub&gt;2&lt;/sub&gt; ~ 2:1</td>
<td>-3 ml</td>
<td>-1.6 ml</td>
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<td>Volume of Reactant gas Added</td>
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<td>30 ml H&lt;sub&gt;2&lt;/sub&gt;</td>
<td>24 ml O&lt;sub&gt;2&lt;/sub&gt;</td>
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Fig. 1: Electrolysis cell and gas handling system.

1) Electrolytic cell
2) Electrodes
2a) Electrode leads
3) Catalyst fabric
4) Manometer
5) Silicone Septum
6), (7), (8) & (9) Vac Stop cocks
10) Sampling manifold with Septum
11) Expansion bulb
12) Mercury reservoir
13) To vac pump

Hg

Hg
PART B

D₂ GAS LOADING EXPERIMENTS
SEARCH FOR NUCLEAR FUSION IN GAS PHASE DEUTERIDING OF TITANIUM METAL

P. Raj*, P. Suryanarayana*, A. Sathyamoorthy*, and T. Datta†

*Chemistry Division
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Introduction

The possibility of D—D nuclear fusion in some deuterium—metal systems, under ambient conditions, has aroused feverish worldwide interest. Most of the work reported, so far, concerns deuterium charging of Pd metal through electrolysis of D₂O.

In Chemistry Division, we have carried out some experiments on the deuteriding behaviour of Ti metal, through gaseous route, in the absorption as well as desorption modes, with the view to look for the fusion products, neutrons in the present case. This kind of experiments have been reported by Frascatti Group in Italy. These authors detected neutron emission lasting over a period of several hours.

Experiments

Experimental arrangement for deuteriding Ti metal is shown in Fig.1, which is self-explanatory. This set up has been routinely used for high pressure hydriding studies on several systems, reported by us. In some of the experiments reported here, deuterium pressure was cycled between high and low values by simply changing the temperature of the cell housing the sample. Most of the experiments were done in the desorption mode. Ti metal pieces (cut from a sheet) were surface cleaned and subjected to activation treatment before D₂ loading and subsequent desorption treatment etc.

Neutron counting set up consists of an array of 24 He³ counters arranged in a well like geometry. These counters (each 50 cm in length 2.5 cm in diameter and filled with He³ at 4 atm.) housed in paraffin moderators, are all connected in parallel to a single pre-amplifier. The counting efficiency of this system was found to be ≈10%. The counts are recorded in 8192 channel multi-scalers. In the experiments reported here dwell time of 40 sec. was fixed, so that each point in Fig.2(a) to (d) represents the number of counts per 40 sec. The background counts collected for about 10 days, before the start of these deuteriding experiments, was found to be quite steady 60 counts/ 40 secs. This background count rate continues to be the same well after our experiments.

Results

In the first set of experiments, starting from 3rd June 1989, after activating Ti metal pieces, D₂ gas was contacted with the sample at a pressure ~10 atm. while keeping the sample at low temperature (~77K). After a soaking time of ~20 min., sample temperature was raised gradually, while simultaneous evacuation was started. Within about 15 min. the neutron counter registered an increase in count rate reaching a max. 3900 (as compared to back-ground counts of ~60), see Fig.2(a). On withdrawing the reactor from the counting well, a considerable reduction in the counts was observed. On re-introducing the reactor after background counts are restored, an additional peak like structure was observed. Although the evacuation was continued, no further increase in count rate over the background could be observed over the next twenty hours.
Next experiment on the same charge was carried out by repeating the conditions of first experiment. The results of this experiment, dated 4th June 1989, are shown in Fig.2(b). Again two peak like structures, each lasting for about 30 min. and separated by 50 min. were seen. However, the intensities of both these structures are greatly reduced, as compared to the first experiment (3rd June 1989), the max. counts being ~ 700.

In the third experiment, with the same charge of Ti pieces, D$_2$ gas pressure was made to cycle between ~50 atm. to ~13 atm. by changing the reactor temperature from room temperature to 77 K. In this case large changes in counts, as a function of time were noticed. An increasing trend of counts initiated at ~2330 hrs on 4th June 1989 lasted for almost 7 hrs. with an estimated integral counts ~6.5x10$^5$. Even after this long bursts like structures, some additional peaks were observed on 5th June 1989. With no further structures observed over the next few hours, desorption was carried out after loading the sample with D$_2$ gas with the sample temperature at ~77K. By raising sample temperature gradually, while simultaneously evacuating, a much bigger structure lasting for ~2 hrs. (from ~1830 to 2030 hrs. on 7th June 1989) was seen Fig.2(c). An approximate estimate of integrated count over this period is 7x10$^5$. Further experiments with this charge, involving D$_2$ loading followed by prolonged periods of evacuation at temperature up to a max. of ~200°C, did not show further structures.

Second series of similar experiments on a fresh charge of Ti from the same source did not exhibit exactly similar behaviour, as found for the first charge. However, one set of experiments on 17th June 1989 see Fig.2(d) involving pressure cycling, followed by evacuation, exhibited increase in count rate lasting over a period of ~100 min. In this case, the scatter in the counts was found to be rather large and maximum counts up to 10$^5$/ 40 s were observed, as compared to background count of ~60/ 40 sec. This charge showed no further increase in count rate even after various treatments.

Further experiments are planned — (i) to study all possible parameters relating to the observed increase in the count rates, (ii) to identify the source of these extra counts, and (iii) to investigate the energy and time structure of the radiation responsible for the observed peak like structures.

Acknowledgement

Authors are indebted to Dr. R.M. Iyer, Director, Chemical group and Dr. J.P. Mittal, Head, Chemistry Division for helpful discussions and encouragement. Sincere thanks are due to Dr. P.R. Natarajan, Head, Radiochemistry Division; Dr. H.K. Sadhukhan, Head, Heavy Water Division; Dr. C.K. Gupta, Head, Metallurgy Division and their colleagues for providing neutron counting facility, Deuterium gas and Titanium metal, respectively.

References

SCHEMATIC DIAGRAM OF THE HYDRIDING/DEHYDRIDING UNIT
FIG. 2 DESORPTION MODE EXPERIMENTS
DEUTERATION OF MACHINED TITANIUM TARGETS
FOR COLD FUSION EXPERIMENTS

V. K. Shrikhande* and K.C. Mittal†

*Technical Physics and Prototype Engineering Division
†Plasma Physics Division

Introduction

Cold fusion experiments were initiated with solid targets made from titanium loaded with deuterium gas on receipt of reports of the successful Frascati experiments. The absorption of deuterium by Ti is a reversible process and when titanium is heated in a deuterium atmosphere, the reaction will continue until the concentration of deuterium in the metal attains an equilibrium value. This equilibrium value depends on the specimen temperature and the pressure of the surrounding deuterium atmosphere. Any imposed temperature or pressure change causes rejection or absorption of deuterium until a new equilibrium state is achieved. If the surface of titanium is clean, the rate of absorption increases rapidly with temperature. At temperatures above 500 °C, the equilibrium is achieved in a matter of a few seconds. However deuterium absorption is considerably reduced if the surface of Ti is contaminated with oxygen. Keeping in view these facts, a procedure was evolved for titanium target preparation and subsequent deuteration. The following sections describe the details of preparation of the targets, their chemical cleaning and degassing followed by deuteration process.

Preparation of the Targets

Titanium targets of different sizes and shapes (planer, conical etc) were prepared. Targets were typically a fraction of a gram in mass and were machined out of a Ti rod using tungsten carbide tools with continuous cooling arrangement. Care was taken to avoid overheating during machining because any overheating could harden titanium and thereby inhibit its capacity to absorb H₂/D₂.

The machined targets were first degreased ultrasonically in trichloroethylene. Then the oxide layer if any was removed by immersing the targets in a 1:1:1 mixture of water, nitric acid and sulphuric acid. They were then rinsed in water and dried in acetone. This was followed by HCl treatment to form an adherent hydride layer on the surface. Targets thus prepared were preserved in a moisture free environment prior to deuterium absorption.

Degassing and Deuteration of Targets

The chemically cleaned targets were first degassed by heating to ≈900 °C in a glass vacuum chamber using a 3 Kw, 2 MHz induction heater. Degassing was continued till a vacuum of less than 10⁻⁵ Torr was achieved. Targets were then heated to ≈600 °C in H₂ atmosphere at a few Torr pressure and allowed to cool. H₂ was absorbed in the targets while cooling. Absorbed H₂ was released again by heating to 900 °C. At least three cycles of H₂ absorption/desorption were given to create active sites for D₂ absorption.

After release of all H₂, the targets were heated to ≈600 °C in D₂ atmosphere at few torr pressure and allowed to cool by switching off the induction heater. D₂ gas was absorbed while cooling. At least three cycles of D₂ absorption/desorption were given, similar to H₂ absorption/desorption. Fall in pressure recorded by an oil manometer is a
measure of the quantity of D$_2$ absorbed. It was found that the quantity of gas absorbed increased in each new cycle and tended to saturate in the 3rd or 4th cycle. Table I illustrates the maximum absorption of hydrogen and deuterium in different Ti targets.

It was noticed that targets could typically absorb $\cong 10^{19}$ molecules of D$_2$. Considering that mass of Ti is a few hundred milligrams, this corresponds to an overall D/Ti ratio of $\cong 10^{-3}$ only. However, if most of the absorption is restricted to the surface, as we suspect, it is likely that the D/Ti ratio is higher than 0.001 in the near surface region.

While preparing the targets, we found that successful deuteration depends on various experimental factors as listed below:

(i) Initial sandblasting of the targets for cleaning and roughening of the surface leads to better absorption of D$_2$. (ii) Impurity content (such as O$_2$, N$_2$ etc) in D$_2$ should be <0.1%. (iii) Since the glass vacuum chamber is isolated from the pumping system during D$_2$ absorption, it is important that the vacuum chamber be leak tight. Small air leaks may contaminate the D$_2$.

The deuterated targets were sent to the Neutron Physics Division for analysis in quest of evidence for cold fusion. Ref. 3 and 4 describe the autoradiography and neutron counting results.

PostScript

As mentioned earlier the deuteration of the titanium targets was carried out using a 3 Kw induction heater operating at 2 MHz frequency. The power supply of this heater became defective in July 89 following failure of the main driver tube. Since then gas loading of targets could not be carried out in this division. Similar experiments were thereafter commenced at the Heavy Water Division using a resistance furnace as described in Ref. 5. However although the loading procedure adopted there was such that very large quantities of D$_2$ gas ($\cong 6$ litres at 1 Kg/cm$^2$) could be successfully absorbed in titanium pieces (mass $\cong 5$ grams), none of the Ti samples have shown any evidence of tritium so far. It is possible that use of high frequency (2 MHz) induction heating may have had some role in causing the detectable levels of cold fusion.

When a metallic object is heated by induction heating, the current distribution within the object is non-uniform with the current density decreasing exponentially from the surface to the centre of the metallic work load. The characteristic penetration or skin depth $\delta$ is defined as that distance over which the current density is reduced to 1/e times the surface value and is given by

$$\delta = (\rho/\pi\mu f)^{1/2}$$

where $\rho$ is the resistivity, $\mu$, the permeability of the workload and $f$ is the frequency of the applied alternating magnetic field. For a 2 MHz induction heater the skin depth in titanium works out to be $\cong 0.1$ mm. It is believed that most of the absorbed D$_2$ gas is accumulated in the near surface region even though the entire sample would have reached high temperatures due to conduction. Hence it is likely that D$_2$ density is very much higher in the near surface region though the gross D/Ti ratio is hardly 0.001.

Further investigations to confirm these conjectures are underway.
Acknowledgements

The authors wish to acknowledge the significant spade work done by Dr. S.K.H. Auluck, in arriving at the optimum conditions for loading of H$_2$/D$_2$ in machined Ti targets. We are also grateful to V.G. Date of the Atomic Fuels Division for fabrication of machined targets.

References


TABLE I

Maximum Absorption of H$_2$/D$_2$ in Different Titanium Targets

<table>
<thead>
<tr>
<th>Sr.No</th>
<th>Target shape</th>
<th>Mass (g)</th>
<th>H$_2$ Absorption</th>
<th>D$_2$ Absorption</th>
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<tr>
<td></td>
<td></td>
<td></td>
<td>mm of oil</td>
<td>mm of Hg</td>
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<tr>
<td>1</td>
<td>Disc</td>
<td>0.980</td>
<td>35</td>
<td>22.5</td>
</tr>
<tr>
<td>2</td>
<td>Cone</td>
<td>0.198</td>
<td>16</td>
<td>5</td>
</tr>
<tr>
<td>3</td>
<td>Cone</td>
<td>0.206</td>
<td>16</td>
<td>5</td>
</tr>
<tr>
<td>4</td>
<td>Kite</td>
<td>0.610</td>
<td>16</td>
<td>5</td>
</tr>
<tr>
<td>5</td>
<td>Cone</td>
<td>0.200</td>
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<td>5</td>
</tr>
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<td>6</td>
<td>Disc</td>
<td>0.875</td>
<td>54</td>
<td>5</td>
</tr>
<tr>
<td>7</td>
<td>Cone</td>
<td>0.460</td>
<td>41</td>
<td>5</td>
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<td>8</td>
<td>Disc</td>
<td>0.875</td>
<td>46</td>
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<td>9</td>
<td>Sponge</td>
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<td>15</td>
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<td>17</td>
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Autoradiography of Deuterated Ti and Pd Targets for Spatially Resolved Detection of Tritium Produced by Cold Fusion

R K Rout, M Srinivasan and A Shyam

Neutron Physics Division

Introduction

For the last few months, hectic activity is underway in various laboratories to study the Cold Fusion phenomenon. De Ninno et al. reported emission of neutrons from titanium metal loaded with deuterium gas under pressure. Similar experiments have been conducted at Trombay. We report here evidence of cold fusion in D$_2$ gas loaded Ti and Pd targets through the use of autoradiography for spatially resolved detection of tritium. Our study employed three different techniques to observe tritium:

(i) Autoradiography using X-ray films.
(ii) Characteristic X-ray measurement of titanium, excited by the tritium $\beta$.
(iii) Liquid scintillation method for tritium $\beta$ counting.

Loading of Deuterium

Titanium and palladium metal samples of various shapes and sizes were loaded with deuterium by two different ways. In the first method an individual titanium target was heated by R F heating up to a maximum temperature of $\simeq$ 900 °C in vacuum and then in deuterium gas atmosphere to absorb deuterium. In the second method the foils of palladium (Pd–Ag alloy) were heated (by ohmic heating) up to a temperature of 600 °C in vacuum (10$^{-5}$ mm of Hg) and then in D$_2$ gas. The deuterium gas used for loading had a tritium content of $\lesssim$5.5 X 10$^{-4}$ Bq/ml of gas, corresponding to a T/D ratio of $\sim$4 X 10$^{-14}$.

Autoradiography

Autoradiography is a simple and elegant technique of detecting the presence of radiation emitting zones. This technique has the advantage of being free from any electromagnetic interference (pick ups, discharge pulses etc), has relatively high sensitivity as it can integrate over long exposure times and can give very useful information in the form of space resolved images. In order to achieve good resolution of the image, the sample was kept very close to the X-ray film. Standard medical X-ray film of medium grain size (10 to 15 $\mu$m in diameter) on cellulose triacetate base was used for this purpose. The exposure time used for the deuterated samples varied from 18 hours to a few days. At times a stack of several films was used. In some cases films were placed on both sides of the sample. For late image formation we used IPC (India Photographic Company Ltd.) made 19B developer and IPC made fixer. The developing time was typically 4 to 5 minutes. Out of many samples which had absorbed D$_2$ gas, only a few showed a latent image. The results are tabulated in Table I.

The radiograph (Fig. 1) of some of the deuterated titanium disc targets showed several spots randomly distributed within the sample boundary. The occurrence of spots all along the rim of the machined target is very intriguing. Repeated measurements over a period of one month, with the same sample with varying exposure times gave almost identical pattern and relative intensity distribution of spots, indicating that the radiation emitting regions were well entrenched in the face of the titanium lattice. The fact that the second film of a stack of films exposed to the target also indicates similar though less intense spots, rules out the possibility of any kind of chemical reduction reaction caused by the deuterium or hydrogen in the target being responsible for causing the spots. The X-ray image (Fig. 2) of a conical target showed a diffused projection of the cone.
The image of Pd—Ag foils (Fig.3) however exhibited a more uniform image. The images however indicated variation in intensity and some spots but on the whole the fogging was more or less uniform. Unlike deuterated titanium targets, the intensity of fogging of deuterated Pd foils reduced very rapidly i.e. within a couple of days the activity reduced below measurement level.

**Measurements of X-ray Emission**

The characteristic X-rays emitted from the deuterated metals (Ti and Pd) were studied with the help of a Si (-Li) (Silicon - Lithium drifted) detector by the Nuclear Physics Division. The detector had a beryllium window of 75 μm thickness. The X-rays of Ti (Kα = 4.5 Kev, Kβ= 4.9 Kev) were observed in case of conical (Fig.4) and disc (Fig.5) samples. The count rate of the conical sample was much more than that of the disc sample. Some of the deuterated Pd—Ag foils indicated the X-ray peaks (Fig.6) corresponding to titanium presumably because of a small amount of titanium impurity picked up by the foils from the D2 loading chamber which had earlier been used for loading of Ti samples. We did not observe the L X-rays of palladium or silver.

**Liquid Scintillation Counting**

This was carried out at the Health Physics Division using the facilities described in Ref.4. The sample was simply dropped into a vial containing liquid scintillator cocktail and the tritium activity was counted by two photomultiplier tubes in coincidence. The typical activities were 50 to 1000 Bq as compared to a background of less than 0.2 Bq. No correction was applied for possible quenching/ shadowing effects.

**Results and Discussion**

The fogging observed in autoradiographs (Figs.1, 2 & 3) is the combined effect of tritium betas and characteristic X-rays of the host material. The radiograph of the disc sample (Fig.1) indicates evidence of tritium localized in the form of microstructures. These spots are unevenly distributed on the face of the titanium, there being about 60 to 70 spots in all. On correlation with the X-ray counts under the peak (K X-ray peak) and liquid scintillation counting results it was found that the each emitting spot corresponds roughly to 10⁹ to 10¹⁰ atoms of tritium. In comparison the total number of deuterium atoms loaded in the disc sample was 10¹⁹ to 10²⁰. The X-ray images (Fig.3) in case of Pd—Ag foils were uniformly fogged and intensity of fogging reduced very rapidly with time unlike with titanium. This type of loss in image may be attributed to the high mobility of tritium in palladium as compared to that in titanium. Observation of K X-ray peaks of titanium (Figs.4, 5 & 6) by Si(-Li) detector was the result of excitation of K-shell by tritium β. L X-ray of palladium (≈ 3.6 Kev) or silver (≈ 3.8 Kev) was not observed because of low fluorescent yield for L X-ray and the detector window being too thick (75 μm) to allow observable quantity of L X-rays. Liquid scintillation counting further confirms the presence of tritium in the samples. It may also be pointed out that the quantity of tritium observed even on the surface of the samples exceeded the total quantity of tritium initially contained in the deuterium gas used to load the samples and hence the gaseous tritium, even if preferentially absorbed by the samples cannot explain this phenomena. Undeuterated metallic targets machined out of the same titanium rods did not indicate any detectable tritium, ruling out any contamination pick up during target fabrication/ handling.

**Summary and Conclusions**

The evidence presented in the paper seems to be indicative of cold fusion reactions occurring in some of the deuterium loaded titanium and palladium targets. It has not been
possible to conclusively establish whether the fusion reactions occur during the deuteration process or subsequently. Also it is not clear whether the reactions occur in sporadic bursts or continuously. However one of the disc targets, which gave impressive spotty radiograph did give rise to a significant neutron burst which produced $10^6$ neutrons over a period of 85 minutes.

Acknowledgements

The authors sincerely wish to express their gratefulness to Dr. P K Iyengar, Director, BARC for his keen interest and constant guidance in the present work. We are also thankful to M S Krishnan, S K Malhotra, S Shrikhande and K C Mittal for supplying the deuterated targets. We also grateful to Drs. V S Ramamurthy and Madan Lal for the X-ray spectral measurements. The authors would like to express their thanks to Dr. T S Iyengar for carrying out the liquid scintillation counting of the targets.

References

1 A De Ninno et al, Europhysics Lett. 9, 221 (1989).

*Estimated from titanium (present as impurity) X-rays; can be inaccurate.

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<th>CS003</th>
<th>PS001</th>
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<td>Ti</td>
<td>Ti</td>
<td>Pd–Ag</td>
</tr>
<tr>
<td>Shape of Sample</td>
<td>Disc</td>
<td>Cone</td>
<td>Foil</td>
</tr>
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<td>Sample Mass (mg)</td>
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<td>110</td>
</tr>
<tr>
<td>D₂ Absorbed (mg)</td>
<td>0.42</td>
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<td>0.73</td>
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<td>D₂ Loading process</td>
<td>Ref.2</td>
<td>Ref.2</td>
<td>Ref.3</td>
</tr>
<tr>
<td>Date of D₂ Loading</td>
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<td>Date of Exposure</td>
<td>23–6–89</td>
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<td>24–8–89</td>
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<tr>
<td>Exposure Duration</td>
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<td>24 Hr</td>
<td>88 Hr</td>
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<td>3</td>
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<tr>
<td>No. of Times Repeated</td>
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<td>3</td>
<td>4</td>
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<tr>
<td>Si(-Li) Result (Bq)</td>
<td>290</td>
<td>1300</td>
<td>2960*</td>
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<td>Date of Measurement</td>
<td>16–6–89</td>
<td>16–6–89</td>
<td>24–8–89</td>
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<tr>
<td>Total Tritium Atoms</td>
<td>$\approx 1.5 \times 10^{11}$</td>
<td>$\approx 6.5 \times 10^{11}$</td>
<td>$\approx 1.5 \times 10^{12}$</td>
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<tr>
<td>T/D Ratio</td>
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<td>$\approx 3.2 \times 10^{-8}$</td>
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<tr>
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<td>850</td>
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EVIDENCE FOR PRODUCTION OF TRITIUM VIA COLD FUSION REACTIONS IN DEUTERIUM GAS LOADED PALLADIUM

M.S. Krishnan, S.K. Malhotra, D.G. Gaonkar, V.B. Nagvenkar and H.K. Sadhukhan

Heavy Water Division

Introduction

After the first announcement reporting the observation of cold Fusion\textsuperscript{1-2} further evidence supporting the same has appeared in scientific literature although many other groups have failed to obtain positive results. Palladium and titanium loaded electrolytically\textsuperscript{1-2} and titanium loaded directly with deuterium gas\textsuperscript{3} have been reported to emit neutrons. Interestingly gas loading experiments involving Pd–D have not been reported so far. Such experiments were therefore conducted recently in our group. Tritium measurements in gas loaded Pd–D targets have been carried out. The present paper summarises the results obtained so far to ascertain whether cold fusion reactions occur in gas loaded Pd targets also.

Preparation of Pd Samples

For loading deuterium gas in Pd, two types of samples were used. One was Pd–Ag alloy supplied by M/s. Johnson–Mathey and was directly used without further surface treatment. The other type of sample used was Pd–black powder which was prepared from PdCl\textsubscript{2}. Absorption of deuterium by Pd–black was very fast and readily gave a stoichiometry of D:Pd = 0.6 as deduced from the drop in gas pressure during loading. But in case of Pd–Ag alloy the absorption was rather slow and also the amount of deuterium absorbed was much less than that corresponding to a D:Pd ratio of 0.6.

D\textsubscript{2} gas Loading Procedure

The D\textsubscript{2} gas used for gas loading was prepared from D\textsubscript{2}O procured from Heavy Water Plant at Baroda (Gujrat state). This D\textsubscript{2}O had a tritium activity of 0.075 nCi/ml. The gas generated from it by reducing with Na in a vacuum system under stringent conditions, was stored in a s.s. cylinder under pressure and liquid N\textsubscript{2} cooling in the presence of activated charcoal. The D\textsubscript{2} gas thus produced was not further analysed for tritium as it was expected to contain not more than 0.038 nCi/l activity. This corresponds to a (T/D) isotopic ratio of 3\times10^{-14}.

The schematic drawing of the experimental set up used for gas loading is given in Fig.1. It essentially consists of a vacuum system equipped with a rotary pump and an oil diffusion pump giving a vacuum of 10\textsuperscript{-6} torr. The s.s. reaction vessel C containing the Pd sample is connected to the vacuum system through an s.s. buffer tank B. Deuterium cylinder D is connected to the vacuum system through needle valve VI. The system is also equipped with a pressure gauge G and a manometer/ pressure gauge VG. The entire system was tested for a vacuum of 10\textsuperscript{-6} torr and pressure of 100 Kg/cm\textsuperscript{2}. A weighed amount of Pd–black or Pd–Ag alloy was taken in the vessel and heated to 600 °C for 2 hours under a vacuum of better than 10\textsuperscript{-6} mm. After cooling to room temperature, deuterium gas was filled at 1 atm. pressure and the system sealed off to attain equilibrium.

After completion of gas loading the vessel containing the Pd sample was isolated from the filling system and transferred into a closed glass container in a dry enclosure, free of moisture or oxygen and kept for equilibration for several hours. Adequate precautions
were taken to avoid inadvertent exposure to moisture since this would lead to catalytic recombination of absorbed D₂ gas with O₂, accompanied by considerable increase in temperature. The entire deuterium absorbed in Pd would be lost if dire precaution is not taken. This was confirmed in one set of experiments where the loaded sample was accidentally exposed to air and the resultant water sample equilibrated did not show any activity.

Tritium Analysis

In order to measure the tritium if any produced due to cold fusion reactions the deuterated Pd samples were kept in contact with distilled water for a few hours to extract the tritium by isotopic exchange into the water. The water samples containing tritium were later sent to the Tritium group of the Health Physics Division for liquid scintillation counting using procedures described in this report elsewhere.

In converting the measured tritium activity in the distilled water to a calculated tritium activity originally present in the Pd samples, there is a conversion factor to be used which is computed as follows:

Taking into consideration the exchange reaction

\[ T_{abs} + H₂O = H_{abs} + HTO \]  

and applying laws of Chemical Equilibrium, one obtains the following relation for low tritium concentration:

\[ Y_i n_g = n_i [X_e (1/K + 1) - X_i] \]  

where \( X \) \& \( Y \) refer to tritium atom fraction in the absorbed and the liquid phases respectively and subscripts \( i \) and \( e \) refer to initial and equilibration conditions respectively. \( n_g \) \& \( n_i \) are the gm moles of gas absorbed in metal and gm moles of water taken for equilibration. \( K \) is the equilibrium constant for reaction (1) and was taken to be same as for exchange of tritium between hydrogen and water, since at equilibrium the system consists mainly of H both in the absorbed and the liquid phases. The value of \( K \) is 6.128 at 30°C. \( Y_i \) calculated from equation (2) gives the number of tritium atoms produced for every deuterium atom absorbed in the metal. The tritium atom fraction \( X \) can be calculated from the tritium activity \( A \) applying equation 3.

\[ x_e = \frac{A}{d \times 3200} \]  

where \( A \) is activity in Ci/ml and \( d \) is density of water in gm/ml (\( d = 0.996542 \) at 27°C).

Results and Discussion

Table I summarises the results of the tritium measurements on the four samples studied by us. The first three rows give the experimental parameters viz. mass of sample, volume (STP) of deuterium absorbed and the volume of water employed for isotopic exchange. The fourth row gives the D/Pd ratio. The fifth row shows the time for which the gas loaded Pd samples were kept undisturbed in the same pressure at which D₂ gas has been loaded. The seventh row gives the tritium activity of the water after equilibration. The last two rows give the idea of absolute amounts of tritium present in the metal before
equilibration. The T/D ratio is about two—three times more in the case of Pd—Ag foils as compared to Pd black (6th row). It should be noted that the T/D ratio in the targets is in the range of \(10^{-12}\) to \(10^{-11}\) which is more than two orders of magnitude higher than that in the initial deuterium gas used for loading.

A fresh lot of Pd black (10 gms) and Pd—Ag foils (0.43 gms) were equilibrated with \(D_2\) gas and after this accidentally they were exposed to air. These became intensely hot and on equilibration with distilled water and analysis of T in the equilibrated water, the T activity observed in both was below detectable limit. The same sample of Pd—Ag foil (0.43) was reactivated (650 °C, \(10^{-5}\) mm) and again loaded with \(D_2\) gas which shows the results as in the 3rd vertical column, proving that the residual activity in Pd is nil. It is also seen that when the equilibration time is more, more tritium production is observed.

The method of isotopic exchange for extraction of tritium from metal to water phase is less cumbersome and the T/D ratio obtained from equation (1) gives at least a conservative estimate of tritium produced.

**Autoradiography of Samples**

The Pd—Ag foils were also subjected to autoradiography to obtain images of tritium distribution as described in detail elsewhere in this report. It essentially consists of keeping the gas loaded sample on x—ray film and developing it after allowing adequate exposure time. The Pd—black samples could not be subjected to Autoradiography as the powder got stuck to the film. But in case of Pd—Ag foils it gave unmistakable fogging of x—ray film corresponding to the geometrical shape of the foils, thus indicating the emission of some radiation from these foils and the only radiation being emitted from these foils which can be thought of is the \(\beta\)—rays from tritium and the characteristics X—rays of the metal excited by tritium \(\beta\)s.

**Summary and Conclusions**

Gas loaded Pd samples have provided evidence for the first time of the presence of tritium, strongly suggesting the occurrence of cold fusion reactions. The Pd—D system does not require high pressure of \(D_2\) gas and also no external perturbation is required to create non equilibrium conditions as suggested by De Ninno et al. Although no quantitative comparisons can be made between electrolytically loaded and gas loaded Pd experiments. The present results confirm that electrolysis is not the only approach to inducing cold fusion in Pd lattice. Unfortunately no neutron measurements were carried out in the present work. A correlation between neutron production if any and tritium production would contribute significantly towards understanding the mechanism of cold fusion.

**Acknowledgements**

Authors sincerely wish to express their gratefulness to Dr. P.K. Iyengar, Director, BARC for his keen interest and constant guidance in the present work. We are thankful to Dr. M. Srinivasan for critically reading the manuscript and offering many suggestions. We also thank Dr. A Shyam and Mr. R.K. Rout for carrying out the autoradiography of the samples. This work would not have been possible but for the unstinted cooperation of Dr. T.S. Iyengar, of the Health Physics Division, who carried out the Tritium counting of all the water samples. The authors also wish to thank Shri. V.H. Patil, Heavy water Division who has been of great assistance in the experimental work.
# References


## TABLE I

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<th>Nature of Sample</th>
<th>Pd black powder</th>
<th>Pd–Ag foil*</th>
<th>Pd Ag foil*</th>
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<td>3 Volume of D₂ gas absorbed (ml)</td>
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<td>0.88</td>
<td>0.23</td>
</tr>
<tr>
<td>7 Tritium activity of water after equilibration (nCi/ml)</td>
<td>3.24 x 10⁻¹²</td>
<td>1.08 x 10⁻¹¹</td>
<td>8.31 x 10⁻¹¹</td>
<td>8.67 x 10⁻¹²</td>
</tr>
<tr>
<td>8 T/D ratio in Pd</td>
<td>11.05</td>
<td>0.97</td>
<td>4.32</td>
<td>11.58</td>
</tr>
<tr>
<td>9 Absolute tritium activity (nCi)</td>
<td>2.31 x 10¹¹</td>
<td>2.02 x 10¹⁰</td>
<td>8.96 x 10¹⁰</td>
<td>2.4 x 10¹¹</td>
</tr>
</tbody>
</table>

*These were the triangular foils also studied by Autoradiography.
†This was a single 11.5 cm dia foil; same as cathode used in cell of paper A2.
Fig. 1. Schematic diagram of equilibration system.
PART C

THEORETICAL PAPERS
MATERIALS ISSUES IN THE SO CALLED "COLD FUSION" EXPERIMENTS

R. Chidambaram and V. C. Sahni

Physics Group

In a recent electrochemical experiment with palladium cathode and platinum anode, immersed in 99.5% D₂O + 0.5% H₂O plus 0.1 M LiOD, Fleischmann and Pons¹ claimed to have seen an 'excess heat', which they ascribed to 'cold fusion' of deuterium nuclei electrochemically infused into the palladium lattice. These authors and Jones et al² have attempted detection of neutrons/³H as signatures of fusion based on the well known reactions

\[ \text{D} + \text{D} \rightarrow ^{3}\text{He} (0.82 \text{ MeV}) + n (2.45 \text{ MeV}) , \]
\[ \text{D} + \text{D} \rightarrow ^{4}\text{He} (1.01 \text{ MeV}) + p (3.02 \text{ MeV}) , \]

and have found some favourable evidence. So far efforts to confirm their findings made in various laboratories, including BARC, have been somewhat inconclusive, although some neutrons appear to have been seen occasionally. The reported neutron production¹ is about six to nine orders of magnitude less than what the 'excess heat' would imply. These observations have generated a great amount of debate³ concerning the nuclear physics of D–D reaction; novel ideas have been proposed including the reaction D + D → ^{4}\text{He}, with the added requirement that the energy (23.8 MeV) be delivered directly to the lattice. Even when they have been observed, often it has been claimed that neutrons appeared only intermittently. In view of these features we deem it prudent to draw attention to some of the relevant solid state aspects of Pd–D system which might be involved in some way.

Absorption of hydrogen/deuterium by Pd has been studied for a long time and is known⁴ to be strongly exothermic. Neutron diffraction⁵ experiment shows that H/D goes into the octahedral sites in the Pd lattice. Accompanying this uptake is a structural change, with the solid changing from the \( \alpha \) phase at low H/D concentrations to \( \alpha + \beta \) phase at higher values to an eventual \( \beta \) phase. Both the phases are f.c.c., with cell constants 3.89 Å and 4.03 Å respectively. Calorimetric studies⁴, using activated Pd and molecular D₂ gas, show that, at 30⁰C, the heat released during the formation of PdDₓ rises from \( \approx 7.50 \text{ kcal per mole of D}_2 \) for \( x \approx 0.023 \) to \( \approx 8.43 \text{ kcal per mole of D}_2 \) for \( x \approx 0.422 \), with the authors asserting that 'these heats show a definite increase for each increment of gas added throughout the mixed phase region'. Thus as more D atoms are loaded into Pd, one may expect that the \( \beta \) phase regions (with a higher lattice constant and presumably better cohesion) would grow in the matrix of \( \alpha \) phase, leading to regions of localized strains. We may conjecture that eventually abrupt atomic readjustments may occur, giving rise to conditions — such as local heating and energetic deuterium motions — that are relatively more favourable for some of the D–D fusion mechanisms proposed in the literature. This description also suggests that the results of the electrochemical experiments could be very much sample—dependent.

Let us next turn to the enthalpy release. Actually there are several adsorption, absorption and desorption processes involved in the experiment using an electrochemical cell. But here we will concentrate only on the heat of formation of PdDₓ, which has not received enough attention. We first note that in electrolysis using Pd as a cathode, because of its special ability to dissolve H/D, one generally does not observe any evolution of H₂/D₂ at the beginning, as these are absorbed by Pd. Also we may bear in mind that the values of the enthalpy release in the formation of PdDₓ quoted above from ref. 4 relate to the situation with \textit{molecular} D₂. If we were to measure these using nascent (i.e. atomic form) deuterium, then the enthalpy released would be larger by the dissociation energy⁶ of D₂, viz
106 kcal per mole of D₂. Assuming that the (cathodic) current in the Fleischmann–Pons experiment is only due to flow of D⁺, we can then infer the amount of deuterium impinging on the Pd cathode and then roughly estimate the rate of enthalpy release due to deuteride formation. We estimate that it is of similar magnitude as the claimed 'excess heat' by Fleischmann and Pons and emphasize the need to include it in the total energy balance calculations.

To sum up, we feel that the neutronic signals reported to have been seen in some of the recent electrochemical experiments deserve to be viewed in the light of the materials science of palladium deuteride. Although, if it is finally confirmed, this so called 'cold fusion' would be physically very interesting, the possibility that it will lead to a significant new energy source appears doubtful at present.

We acknowledge helpful discussions with a large number of colleagues especially Dr. P. K. Iyengar, Dr. S. Gangadharan, Dr.T. P. Radhakrishnan and Dr. S. K. Sikka.

References:


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REMARKS ON COLD FUSION

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In this note we wish to bring to attention certain processes which have been invoked to explain solid state and chemical phenomena and which may have relevance to cold fusion\textsuperscript{1-4}.

Starting with Brownian motion in a liquid we note that the typical momentum gained by a foreign Brownian particle, of 1 size, is hundred times the average momentum of the molecules of the liquid\textsuperscript{5}. This is because the momentum gained by the particle is not due to individual impacts from single molecules but because of collective motion of a number of molecules. We also note that the frequency of the random jumps tells us that such fluctuations in collective motions occur quite frequently over normal observational time-scales. Larger momentum transfers would also be imparted but less frequently.

In a solid, similar fluctuations may be less frequent, but larger energy fluctuations may be expected especially in a system with large anharmonicity; D motion in PdD\textsubscript{x} is known to be very anharmonic. The phase diagram of Pd—D system has been extensively studied and its similarity with gas—liquid transition has been discussed at length by Alefeld\textsuperscript{6}. In the phase, with D/Pd ratio > 0.6, deuterium behaves like a lattice liquid with fairly fast diffusion (D \textasciitilde 10^{-7} \text{cm}^{2}/\text{sec} at \approx 500\textdegree{C} in phase). Lattice Brownian motion has been invoked in the past to explain hydrogen diffusion in metals. The important difference in the present case is that the fluctuations of interest to us now are over atomic length scales (several angstroms) and times are typically lattice vibration times (picoseconds), as against hydrodynamic time scales of Brownian motion referred to earlier. If such large momentum transfers as those observed in Brownian motion (i.e. 100 — 1000 times kT) could be imparted to individual deuterium atoms over very short time scales, one can visualize a situation with a much improved chance of a nuclear reaction.

There are a number of solid state phenomena\textsuperscript{7} which give indication that this could indeed be happening. They have been discussed at length by Khait in a number of papers (see Khait\textsuperscript{7} for a review). He has argued that there are several phenomena—in chemical and solid state physics—to describe which, equilibrium concepts are 'fundamentally qualitatively' inadequate. The phenomena are mostly connected with rate processes involving a large activation barrier. For example, if one chooses to describe the diffusion of Ba in BaO in the temperature range of 1350 to 1500\textdegree{C} by an Arrhenius type of equation, \( D = D_{0} \exp (- EA/kT) \), one obtains EA to be about 12 eV which is about 100 times kT. Similarly the activation energy for diffusion of Si, Ge or Bi in Si is \approx 5 \text{eV} = 6 \times 10^{4} \text{Kelvin}. In spite of such large activation energies measurable diffusion rates are observed. Such rates cannot be explained by assuming small thermodynamic fluctuations at the relevant temperatures. It was therefore proposed that short—lived large energy fluctuations of a small number of particles is necessary to be invoked for explaining these phenomena. The mode of formation and dissipation of these fluctuations is discussed at length in reference 7. The basic point relevant to our discussion is that fluctuations involving large energies over short times do occur in solids. They can occur under 'normal' conditions, but, the probability of getting such large energy fluctuations increases especially near certain transitions, under the presence of gradients (pressure, concentration, temperature, field) etc. It is also enhanced in the presence of high anisotropies and near the surface, in a direction perpendicular to the surface\textsuperscript{7}. In all the 'successful' cold fusion experiments reported to—date, electrolytic\textsuperscript{1,2,4} and absorption/desorption\textsuperscript{3,4} type, these 'favourable' conditions exist. These fluctuations could impart energies which are 100 to 1000 times kT.
to the D atoms in PdD$_x$. Thus, instead of the typical optical phonon energy of 50 meV, the deuteron can get energies ~50 eV due to the presence of a short–lived large energy fluctuation. It can then approach another deuterium to within ~22 Å assuming deuterons are considered to be in the presence of a uniform electron background [Screening length 1 Å, EF = 1 eV]. The frequency of reactions thus induced will depend on the frequency with which the fluctuations are produced in the solid. It is not possible at present to give a realistic number for this, calculated ab initio.

In conclusion, we note that a number of known phenomena give evidence for the presence of large energy fluctuations of small number of atoms over short times and these may be considered as one of the possible modes for ‘starting’ a cold fusion which would then be followed by more nuclear reactions involving the product of the first fusion reaction. Such fluctuations may vary considerably depending on the condition of the solid/experiments, like concentration gradient, field gradient, nearness to a transition etc.

References

THE ROLE OF COMBINED ELECTRON–DEUTERON SCREENING IN D–D FUSION IN METALS

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+Health Physics Division

It is well known that the quantum–mechanical calculation of the fusion rate for D₂ molecule in free space yields a negligibly small value of ~10⁻¹⁰⁷⁰ s⁻¹. The recent electrolytic experiments by Fleischmann and Pons and by Jones et al, however, suggest a possibility of a much higher fusion rate of deuterium in palladium at room temperatures. In this communication we propose that if cold fusion is indeed a reality, it may be explained by the combined screening of the Coulomb interactions by itinerant deuterons and the conduction electrons in metals. The chief assumption of our model namely, that deuterium exists as mobile ionized species in palladium under electrolytic conditions, is based on the following facts. It has been established from the electromigration measurements that in palladium, hydrogen exists as a proton with an effective charge ≈+1. Likewise, deuterium should also exist as deuteron with an effective charge ≈+1. Besides, deuterium is known to be highly mobile in palladium. Under these conditions, the deuterons would participate in screening the electrostatic interactions along with the conduction electrons of the metal. The combined screening reduces the Coulomb barrier between the deuterons separated by a distance r by a factor exp(−kr) (where k is the screening constant), thereby leading to an enhancement in the fusion rate.

For nominal composition PdD with a substantial fraction of deuterium atoms in an ionized state, the condition of charge neutrality in the bulk can be written as

\[ n_p + n_d = n_c \]  

where \( n_p, n_d, n_c \) are the bulk densities of the fixed palladium ions, mobile deuterons and the conduction electrons respectively.

For calculating k, one sets up the Poisson's equation for the potential at a radial distance r from a test charge in terms of the net charge densities induced at r. The induced charge densities are obtained using Fermi–Dirac statistics for electrons and Bose–Einstein statistics for deuterons. Upon linearizing the densities as a function of the potential and using the Fourier transform techniques for the Poisson's equation, it can be shown that

\[ k^2 = k_c^2 + k_d^2 \]  

\[ k_c^2 = 6 e^2 n_c / E_c \]  

\[ k_d^2 = \frac{4 \pi e^2 n_d / k_b T}{g_{3/2}(z)/g_{3/2}(z)} \]  

In the above equations \( E_c = (2m_e)^{-1} (3\pi^2 n_e)^2 / \hbar^2 \) is the Fermi energy of the electron gas, \( k_b \) is the Boltzmann constant and \( T \) is the temperature. The fugacity \( z (0<z<1) \) is related to the chemical potential via the expression \( z = \exp(-\mu / k_b T) \) and the function \( g_{3/2}(z) = [n^{-3/2} z^2] \) is well known in connection with Bose condensation. Equation (4) is valid for temperatures \( T > T_c \) where

\[ T_c = \left[ 2\pi \hbar^2 / m_d k_b \right] [n_d / 2.612]^{2/3} \]  
The Bose condensation temperature of the ideal deuteron gas in PdD calculated from eq. (5) is 6.65K. From eq. (4) it follows that at high temperatures $k_d \propto T^{-1/2}$. At low temperatures $k_d$ can diverge as $(T-T_c)^{-1/2}$ as $T \to T_c^*$. Thus the contribution to screening from charged bosons is strongly dependent on temperature and exhibits a power-law divergence at $T_c^*$. However, this may not be realizable in PdD due to a sharp drop in the mobile fraction of deuterons at cryogenic temperatures.

The Thomas–Fermi screening by conduction electrons is well known in solid state physics. The calculations based on (2)–(4) show that the combined screening by electrons and deuterons is more effective than that due to electrons alone.

To calculate the fusion rate in the presence of the combined screening, we assume the existence of pairs of deuterons in the interstitial sites of palladium lattice. These form metastable $D_2^+$ ions or $D_2$ molecules which undergo spontaneous fusion at a rate

$$R = A[\psi(0)]^2, \quad A = 2 \times 10^{-16} \text{ cm}^3 \text{ s}^{-1}$$ (6)

where $\psi(0)$ is the molecular wave function at the origin. At an internuclear separation $r$

$$\beta(r) = \int_1^r \left[2|2\mu \{E_V - V(x)\} - 1/4x^2|^{1/2} - (1/x)\right] dx$$ (8)

where $\mu$ is the effective mass of the d–d system, $w$ is the vibrational frequency, $E_V$ is the ground state energy, $V(r)$ is the interaction potential between the deuterons and $r$ is the inner turning point on the potential energy curve (figure 1). To take the screening into account, we have used the potential

$$V(r) = (1/r) \exp(-kr) - 1.5/[1+0.127r]^4$$ (9)

which, for $k = 0$ is the same as the potential used by Jackson for an unscreened $D_2^+$ ion. All the quantities in the above equation are expressed in atomic units. In the present calculations we have assumed that almost all the deuterium atoms are ionized and these can be extended to account the actual fraction of deuterons present by using appropriate values for $n_d$ and $n_c$.

The calculations using the above equations show that the fusion rate for $D_2^+$ ions which is $5 \times 10^{-79} \text{ s}^{-1}$ in free space increases to $4 \times 10^{-46} \text{ s}^{-1}$ in palladium when the screening due to the conduction electrons ($k_c = 1.0 \text{ au}$) alone is applied. On the other hand, under the combined screening by deuterons and electrons at 300K ($k = 11.6 \text{ au}$), the fusion rate for $D_2^+$ ion increases to $5 \times 10^{-16} \text{ s}^{-1}$. Similar calculations were performed for $D_2$ molecule with the potential $V(r) = (1/r) \exp(-kr) - 2.18/[1+0.18r]^4$. The fusion rates obtained were $1 \times 10^{-43} \text{ s}^{-1}$ for $k = 0$, $3 \times 10^{-40} \text{ s}^{-1}$ for conduction electron screening ($k_c = 1.0 \text{ au}$) alone and $1 \times 10^{-14} \text{ s}^{-1}$ for the combined screening by deuterons and electrons at 300K ($k = 11.6 \text{ au}$). Thus it is seen that the spontaneous fusion rate increases to $10^{-16} \text{ s}^{-1}$ for $D_2^+$ ion and to $10^{-14} \text{ s}^{-1}$ for $D_2$ molecule at 300K, which lie in the range of values indicated by the cold fusion experiments.

The fusion rate is a sensitive function of $k_d$ which is proportional to $n_d^{1/2}$. Under electrolytic conditions, the density of deuterons is higher on the palladium surface than in the bulk due to the existence of a large concentration gradient at the surface. Moreover, the mobility of deuterons in the surface layers will also be much higher. As a consequence, the fusion rate will be substantially higher in the surface layers than in its bulk.
Suppose an additional electric field is applied along the length of the deuterium-saturated palladium cathode during electrolysis in the Fleischmann and Pons\textsuperscript{1} type cell. Due to electromigration under the applied field, the fraction of the mobile deuterons will be increased. Further, as pointed out by Wipf\textsuperscript{4}, a potential difference of say 500 mV can create a deuteron density ratio of $10^8$ between the two ends of the cathode. Though this factor is probably an overestimate (due to space-charge effects), under the additional electric field, $n_\alpha$ at one end will nevertheless be enormously higher than that in the absence of the field. As a consequence, we expect that under the application of the additional electric field, the screening will be substantially enhanced which would, in turn, lead to a large increase in the fusion rate. Similarly an increase in the fusion rate can be expected in the Ti–D gas experiment\textsuperscript{9} by the application of the electric field across the titanium metal.

We thank Dr. V.K. Kelkar for computer calculations. We are indebted to Dr. J.P. Mittal and Dr. K.S.V. Nambi for their encouragement and support during this work.

References


Figure 1. Potential energy curve and ground state wave function for the relative motion of the two nuclei.
A THEORY OF COLD NUCLEAR FUSION IN DEUTERIUM-LOADED PALLADIUM

Swapan K. Ghosh, H.K. Sadhukhan and Ashish K. Dhara

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An efficient means of achieving fusion of isotopic hydrogen nuclei has only remained a long cherished dream in the field of nuclear science due to the need for high energy acceleration to overcome the associated Coulomb barrier. The recent discovery of cold nuclear fusion by Fleischmann et al and Jones et al during electrolytic deposition and consequent compression of deuterium gas into palladium or titanium electrodes, has thus generated great current interest. The fundamental theoretical problem posed by this spectacular phenomenon is to explain the reported fusion rate (~$10^{12}$ per deuteron pair per sec), which is many orders of magnitude larger than that calculated (~$10^{7}$ per deuteron pair per sec) for subbarrier fusion involving conventional quantum mechanical tunnelling of the Coulomb barrier in a deuterium molecule.

Cold nuclear fusion with even much higher fusion rate is however well known in muon—catalyzed fusion (see, for example, Jones), where the enhancement of the tunnelling probability arises from a drastic reduction of the internuclear distance due to replacement of an electron in a deuterium molecule by a massive particle like muon. An attempt has been made to extend this concept for explanation of cold fusion in a metal lattice by considering the binding particle to be a quasi electron of an effective mass $m^*$ higher than the electron mass $m$. The reported tunnelling probability can be predicted using a value of $(m^*/m)^5$, which is however too high to be realized.

In the present work, we propose a new mechanism for the observed cold fusion in deuterium—loaded palladium. We believe that a consideration of mere tunnelling in an isolated deuterium molecule is not adequate to explain the observed cold fusion. The central theme of our approach is that the fusion in a metal lattice is guided by a collective phenomenon resulting into a screened Coulomb barrier which leads to this drastic enhancement of the barrier penetration probability.

Extensive studies have been reported on various aspects of hydrogen in metals and the palladium—hydrogen system in particular. It is known that the deuterium gas compressed inside a palladium lattice dissociates into atoms, each carrying a net positive charge close to unity ($+e$). Also the quantum mechanical zero point motion of these $D^+$ ions (which are bosons) has large amplitude and thus there emerges the picture of a mobile delocalized charged boson gas of $D^+$ ions, with the effective binding potential provided by the system of electrons and the lattice. For simplicity, in this work we consider a jellium type model in which the positively charged boson gas of uniform $D^+$ ion density is subjected to an attractive potential from the lattice and the electrons, represented by a smeared out uniform negative charge distribution. The resulting system is an example of a quantum plasma of bosons and would be associated with plasma oscillations leading to screening of the interaction potential. The $D^+$ ions now interact with an effective screened Coulomb potential of the form

$$V_{\text{eff}}(r) = \left(\frac{e^2}{r}\right)\exp\left(-\frac{r}{l}\right),$$

characterized by the screening length $l$. The screening parameter $l$ is of the order of $l \approx \left(\frac{v_0}{w_p}\right)$, where $v_0$ is the single particle velocity of $D^+$ ions and $w_p$ is the plasma frequency.
given by

$$w_p = (4\pi n_d e^2/m_d)^{1/2}$$  \hspace{1cm} (2)$$

with $n_d$, $e$ and $m_d$ denoting the number density, charge and mass of the $D^+$ ions. For a classical plasma, $v_0$ is obtained from the average thermal energy, while for fermions, it corresponds to the velocity at the top of the Fermi distribution. For the present boson system, we assume the occupancy of only the ground state, i.e. consider a zero temperature and calculate $v_0$ by equating $(1/2) m_d v_0^2$ to the ground state energy

$$\epsilon_0 = \left(\frac{h^2}{2m_d}\right) \left(\frac{3.142}{r_s}\right)^2$$  \hspace{1cm} (3)$$

of a $D^+$ ion in a sphere of radius $r_s$, defined in terms of the mean volume of the ion in the metal, by

$$(4/3) \pi r_s^3 = n_d^{-1}$$  \hspace{1cm} (4)$$

The screening length $l$ can thus be expressed as

$$l \approx \left(\frac{3.142}{3^{1/2}}\right) \left(\frac{m}{m_d}\right)^{1/2} \left(\frac{r_s}{a_0}\right)^{1/2} a_0,$$  \hspace{1cm} (5)$$

where $a_0$ is the Bohr radius for the electron. The effective screened Coulomb potential in which the $D^+$ ions move is thus defined by eqs (1) and (5). The corresponding Schrödinger equation for a pair of $D^+$ ions can be written as

$$\left[\frac{d^2}{dr^2} + \left(\frac{2(\mu_d/h^2)}{E - V_{\text{eff}}(r)}\right) - \frac{J(J+1)}{r^2}\right] \chi(r) = 0$$  \hspace{1cm} (6)$$

where the centre of mass motion and the angular dependences have been separated out. Here, $\chi(r) = r \psi(r)$ denote the radial wave function, $\mu_d (= m_d / 2)$ is the reduced mass and $E$ is the energy of relative motion of approach. In order to employ the semi-classical WKB approximation to this radial equation, the term $J(J+1)$ in eq. (6) is replaced by $(J+1/2)^2$. For $J = 0$ state, this involves the addition of a centrifugal barrier term $(h^2/m_d)4r_s^2$ to the effective potential $V_{\text{eff}}(r)$. The fusion rate is determined by the ratio of the probability densities $\chi^2(r_n)$ and $\chi^2(r_a)$, where $r_n$ is the distance of closest approach for fusion (equal to the nuclear radius) and $r_a$ is the classical turning point determined by the zero of $Q^2(r)$ defined as

$$Q^2(r) = \left(\frac{2\mu_d/h^2}{E - V_{\text{eff}}(r)}\right) + 1/4r^2$$  \hspace{1cm} (7)$$

The transmission coefficient $T(E)$ for this barrier penetration is given by the well known WKB approximation as

$$T(E) = \exp\left[-2 \int_{r_n}^{r_a} |Q(r)| dr\right], \hspace{1cm} (8)$$

which for convenience can be rewritten as

$$T(E) = \left(\frac{r_a}{r_n}\right) \exp\left[-\int_{r_n}^{r_a} (2 |Q(r)| - 1/r) dr\right], \hspace{1cm} (9)$$

The fusion rate (per deuteron pair per sec) is now obtained from the relation

$$P = S \nu_0 T(E)$$  \hspace{1cm} (10)$$

where $\nu_0$ is the frequency of attack representing how often the two $D^+$ ions approach each other and can be calculated by writing $h\nu_0 = E$, the energy of relative motion ($\sim \epsilon_0$). The
parameter $S$ is a selection factor for a particular channel of $D + D$ nuclear reaction.

We have carried out the integration of eq. (9) numerically and calculated the fusion rates at several values of $D$:$Pd$ ratio. The results obtained by using the density of Pd metal as 12.16 g/ml and a value of $10^{-13}$ cm for the nuclear radius $r_n$ are shown in table 1. For a normal $D$:$Pd$ ratio of 0.6, one has the number density $n_d = 4.13 \times 10^{22}$ /ml; the screening length $l = 0.055$ $a_0$ and the frequency factor $\nu_0 = 7.68 \times 10^{11}$/s. For an $S$ factor of unity (which is used for all the present results), the fusion rate is $3.68 \times 10^{-20}$ per deuterion pair per sec and it increases to $1.07 \times 10^{-18}$ for a $D$:$Pd$ ratio equal to unity that corresponds to high pressure. These preliminary order of magnitude calculations thus lead to results which compare very well with the reported experimental cold fusion rates. Calculations using more exact expressions for the screening length or correct value for the $S$ factor (less than unity) although would modify the numerical results somewhat, qualitative predictions are not expected to change.

The present study reveals that the formation of charged boson plasma state of deuterium in the palladium lattice and the consequent new mode of screening of the interaction of deuterium ion pairs might be responsible for the tremendous enhancement of the fusion probability via quantum mechanical tunnelling. Our work differs from most of the recently reported calculations where existence of deuterium molecule is assumed and either the concept of effective electron mass or electron (fermion) screening has been invoked. It also indicates the role of palladium in cold fusion since in this metal deuterium is present as positive ions, whereas in other metals the charge on each deuterium atom might be zero or even negative in some cases.

In conclusion, we emphasize that the screening mechanism due to a quantum boson plasma can significantly enhance the cold fusion rate. However, due to extreme sensitivity of the predicted rate on the screening length, further studies incorporating the effect of temperature, non adiabatic degrees of freedom, non equilibrium conditions etc are in progress to confirm the role of the suggested mechanism in the observed cold fusion.

References

## TABLE I

Nuclear Fusion Rates in Deuterium-Loaded Palladium

<table>
<thead>
<tr>
<th>D:Pd ratio</th>
<th>( r_a/a_0 )</th>
<th>( r_s/a_0 )</th>
<th>( \varepsilon_0 ) (a.u.) ( \times 10^{-4} )</th>
<th>Fusion rate (per deuterium pair per sec)</th>
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<tbody>
<tr>
<td>0.5</td>
<td>3.61</td>
<td>0.814</td>
<td>1.03</td>
<td>1.07 \times 10^{-20}</td>
</tr>
<tr>
<td>0.6</td>
<td>3.39</td>
<td>0.768</td>
<td>1.17</td>
<td>3.68 \times 10^{-20}</td>
</tr>
<tr>
<td>0.7</td>
<td>3.22</td>
<td>0.730</td>
<td>1.29</td>
<td>1.03 \times 10^{-19}</td>
</tr>
<tr>
<td>0.8</td>
<td>3.08</td>
<td>0.699</td>
<td>1.42</td>
<td>2.49 \times 10^{-19}</td>
</tr>
<tr>
<td>0.9</td>
<td>2.96</td>
<td>0.674</td>
<td>1.53</td>
<td>5.39 \times 10^{-19}</td>
</tr>
<tr>
<td>1.0</td>
<td>2.86</td>
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<td>1.64</td>
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</tr>
<tr>
<td>1.1</td>
<td>2.77</td>
<td>0.633</td>
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<td>1.98 \times 10^{-18}</td>
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<tr>
<td>1.2</td>
<td>2.69</td>
<td>0.616</td>
<td>1.85</td>
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FRACTURE PHENOMENA IN CRYSTALLINE SOLIDS: A BRIEF REVIEW
IN THE CONTEXT OF 'COLD FUSION'

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The reports on detection of neutrons and tritium above background levels in deuterium gas-loaded titanium and palladium targets as well as in electrolytic cells using these metals as cathodes immersed in electrolyte containing heavy water (D₂O), have led to the concept of the so called cold fusion i.e. the occurrence of appreciable fusion reactions among deuterium nuclei at room-temperature. Several theoretical speculations have been put forward, since the beginning of the 'cold fusion era' to explain the existence of such a process. However almost simultaneously it has been suggested, as for example at the AIP meeting held at Santa Fe in May '89, that the fusion may not be 'cold' as such but might be resulting from some kind of beam-target interaction of accelerated particles or hot micro plasma formed at certain sites in the solids due to fracture etc. This proposition, termed as 'fracto-fusion', is based on the past observations of interaction of gases with metals and emissions of all kinds of particles and radiations from cracks or cleaved surfaces of crystalline solids. This note presents a brief survey of this type of studies with a view to assess the role of such mechanisms in 'cold fusion' and plan possible experiments to confirm them.

In solid state science, the process of emissions emanating out of mechanically deformed crystals, specially thin oxide layers, has been known for a long time in the form of 'triboluminescence' where light is emitted on application of stress to the crystals. Several kinds of emissions including electrons, negative and positively charged ions, are found to accompany such process. Subsequent studies by Dickinson and others have correlated these emissions to the appearance of cracks in the materials as shown by the recorded acoustic signals. Besides charged particles even neutral gas is found to evolve in some cases. A burst of as many as $10^4$ particles may occur in a short duration of $10\mu$s which is the time a crack of the size of a few tens of $\mu$m would take to propagate (with the speed of sound) although it may continue for a few minutes at lower rates of emission. Origin of this phenomena has been attributed either to the thermionic emission generated due to very short lived microplasma formed at the crack sites or to the field emission resulting from strong electric fields, developed due to charge layer separation. Some independent estimates of temperature at these sites indicate an appreciably large value of about 1 eV giving credence to a thermionic mechanism. At the same time it has also been fairly well established that the sites of cracks or cleaved surfaces of ionic crystals can result in transient electric fields of more than 15 KV/cm. Sources of such transient fields are attributed to micro- and macro-cracks, besides the clusters of dislocations. All these experiments have been mainly carried out with insulating crystals and as pointed out by Kornfeld, due to neutralization by surrounding gas, it may not be possible to sustain a greater (than 15 KV/cm) field although a much higher value of $10^6-10^7$ V/cm has been quoted by Klyuev et al. Another source of this 'fracto-emission' could be a shock-wave or a stress-wave created by the crack tip motion which may give rise to electrical fields as suggested in ref 10. In brief, as pointed out by Dickenson, fracture of insulators can result in production of highly localized heat, creation of excitations and defects, emission of excited and reactive species in gas phase, separation of charges on crack walls accompanied by intense electric fields and production of acoustic waves.

In the context of 'cold fusion' phenomena, above facts need to be viewed along with the well known phenomena of large cracks found to develop in metal-hydrides and their embrittlement on hydrogenation. An observation by Dickenson on the multiplicity of
the number of electrons emitted in a burst from crack sites in crystals could be very significant to the suggested hot fusion mechanism. One of the evidences in ref.2, analyzed in details elsewhere, about the non-Poisson distribution of neutrons generated from the cold fusion experiments is quite similar to that of the above mentioned electron-emission which is also found to occur in singles as well bunches of many at a time. The mechanism of 'fracto-fusion' has been widely quoted at the Santa Fe meeting of AIP on cold fusion as also by Menlove and Levi.

The experiment of Klyuev et al in 1986 where a striker was accelerated by a gas gun to about 200 m/s and impacted onto a LiD crystal is a clue in this direction. With proportional counters, whose output was analyzed through a pulse height analyzer, neutron signal significantly higher than the background was obtained with an estimate of about 10 neutrons per fracture of the LiD crystal. Since this experiment was basically conducted to check the possibility of the above mechanism i.e. fusion caused by deuterium ions, accelerated through large electric-fields generated at the crystal's cleaved faces, the positive signal of neutrons appears to corroborate it. Sietz has also proposed the possibility of hot fusion in which he indicates that high-temperature (about 20 eV) microplasma might be created at certain sites in the crystal due to the formation energy of D₂ molecule alone else through accelerated ions if conditions favour.

In a recent experiment at Brookhaven Lab, bunches of up to few hundred D₂O ions, accelerated to about 325 Kev energies with an electrostatic accelerator and impinged on deuterated titanium target, are reported to have resulted in yields of 0.1 neutron/second - na of ion current. With various control experiments, the origin of these reactions is suggested as due to cluster-ion impact on TiD target only. The reaction-rate though quite small still shows several orders of magnitude higher value than might be expected on the basis of usual calculations. This discrepancy is sought to be explained by the authors on assumption of certain compression and heating due to impact of cluster -ions in the reaction area. It is possible that this type of conclusions, if studied further, would also help in better analysis and understanding of the probable mechanisms responsible for the observed cold fusion phenomena.

Ikaye and Miyamaru too have tried to investigate if stress in materials could give rise to the observed fusion. In their experiment, an electrolytically charged s.s. electrode, sputtered with palladium was mechanically bent to generate stress in it but the detectors did not show any tritium or neutron signal, although significant amount of excess chemical heat was found to be generated.

Whether the 'fracto-fusion' could really be the process responsible for the fusion in deuterated metals can only be established by further experiments. For this purpose, it may be possible to accelerate a deuterated projectile to a few km/s with the help of an electric gun or railgun and then impact it against another similar target. The main problem in detecting the expected low neutron counts with these accelerators is the accompanying electromagnetic noise. Meanwhile along with fusion signals, the detection of electric fields and related acoustic emission during cracking in the present type of cells or gas-loaded metals, should be able to indicate if the fracturing of metals because of deuterium loading is at least partially, if not completely, involved in the observed cold fusion phenomena.

References

5 Robert Pool in Science 244, 1039 (1989).
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Most of the papers included in this report were transferred into a personal computer directly from the typed manuscript using a digital scanner followed by Optical Character Recognition (OCR) software. This way we could avoid having to retype the papers all over again. The files were then passed through a spelling check program and then transferred into "T3 Scientific Word Processor" before being printed out in Proportional Roman Font (or "Pica font" as it is called).

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