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COLD FUSION EXPERIMENTS USING A COMMERCIAL Pd-Ni ELECTROLYSER

M.S. Krishnan*, S.K. Malhotra*, D.G. Gaonkar*,M. Srinivasan‡, S.K. Sikka‡
A. Shyam‡, V. Chitra‡, T.S. Iyengar‡‡ and P.K. Iyengar+
*Heavy Water Division,
‡Neutron Physics Division
‡‡Health Physics Division
+Director

Introduction

The first reports of observation of 'Cold Fusion' during the electrolysis of heavy water[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[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 ≈300 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 up to 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_2O of >99.75% isotopic purity and was prepared by reaction of moisture free Na with D_2O. 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 ≈70 amps. During the initial runs only this was used and operation was restricted to comfortable current values of 60 to 62 amps. 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 BF3 counters embedded in a paraffin moderator block. The second was a 80 mm dia × 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 10 s or less. The neutron yield is deduced by counting the 24 s 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-α-Be neutron source was $\approx 3\times10^5$ neutrons.

Measurement of Tritium Levels in D_2O Electrolyte

Measurements of the absolute levels of tritium in the D_2O 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 withdrawal 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_2O 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 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 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 4×10^7. At the end of this experiment a sample of the electrolyte which was analyzed for tritium content indicated a level of 1.5 μCi/ml of tritium activity in comparison to the initial stock heavy water value of 0.075 nCi/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 nCi/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 amps (current density ~200 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 nCi/ml to 0.12 nCi/ml on 15th June 89. The decrease is attributed to the fact that the electrolyte containing 0.075 nCi/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_2O. 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

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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 different from the 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 BF3 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

Vol. of Pd cathode	= 7 ^{cm3}
Mass of Pd	= 82gm
Area of cathode	= 300 cm^2
Current	= 60 amps
Current density	= ~200 mA/cm^2
Electrolyte	= 20% NaOD in D_2O
Volume of electrolyte	= 250 ml

TABLE II
Neutron Counts Vs Time (21st April 1989)

Interval duration = 5 minutes
Efficiency of BF_3 Bank = 0.06%
Efficiency of NE102A = 0.4%

Int. No.	Neutron counts		Int. No.	Neutron counts	
	BF_3	NE102A		BF_3	NE102A
1	74	683	26	78	636
2	74	659	27	64	606
3	55	605	28	62	640
4	68	672	29	61	659
5	75	686	30	55	603
6	79	693	31	68	631
7	54	650	32	58	755
8	73	612	33	65	647
9	77	641	34	**1680**	**2827**
10	167	727	35	174	788
11	54	643	36	**254**	**1176**
12	124	769	37	269	977
13	74	663	38	**329**	**1290**
14	64	629	39	113	624
15	**429**	**1654**	40	107	622
16	72	627	41	59	626
17	58	630	42	75	606
18	65	641	43	139	619
19	60	655	44	101	714
20	56	663	45	107	958
21	**180**	**1086**	46	74	656
22	**367**	**1185**	47	**17758**	**25872**
23	109	707	48	77	592
24	70	614	49	**1982**	**3874**
25	68	732	–	–	–

TABLE III
Summary of Neutron and Tritium Yields
(Current Density = 200 mA/cm^2)

Run No. & Date	Duration of Electrolysis	Total Neutron Yield	Atoms of Tritium Generated	Neutron to Tritium Yield Ratio
Run l 21st April 89	72hrs	4×10^{7}	8×10^{15}	0.5×10^{-8}
Run 2 12th – 16th June 89	54 hrs	9×10^{6}	$5\times10^{14*}$ $1.9\times10^{15*}$	~1.8×10^{-8} 0.5×10^{-8}

*These were measured 30 hrs and 27 days respectively after the current had been switched off.

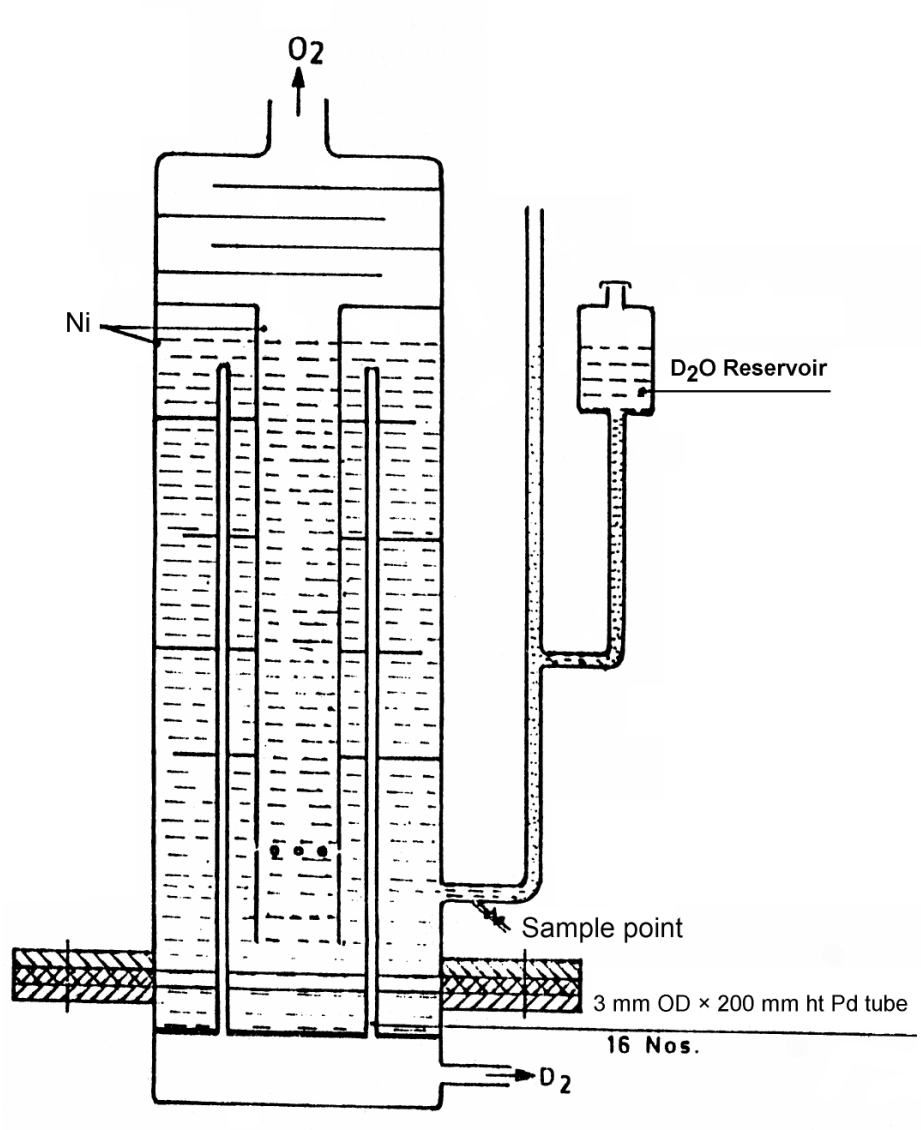

Fig. 1a. Electrolytic cell.

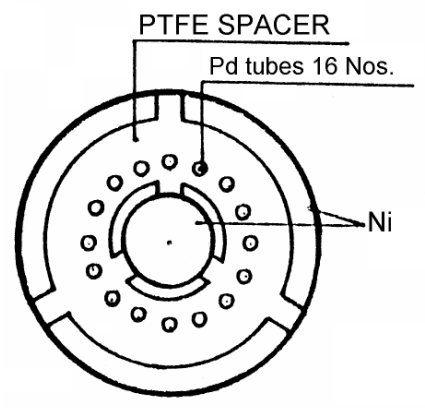

Fig. 1b. Pd tube arrangement.

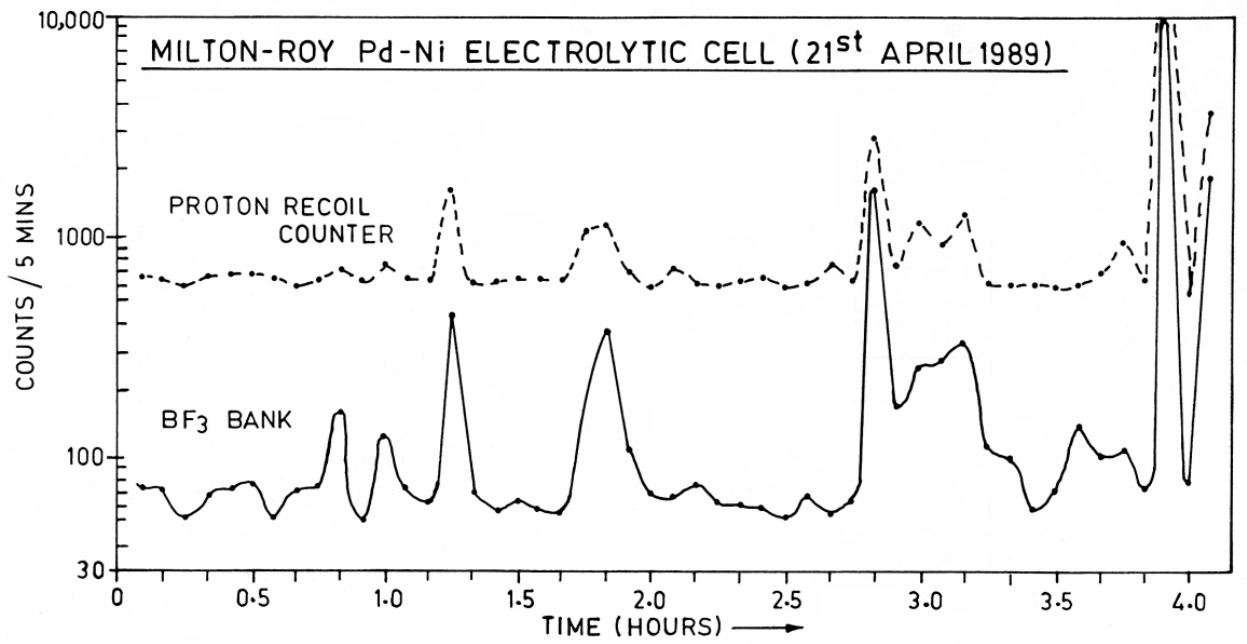

Fig. 2. Milton-Roy Pd-Ni electrolytic cell (21st April 1989).

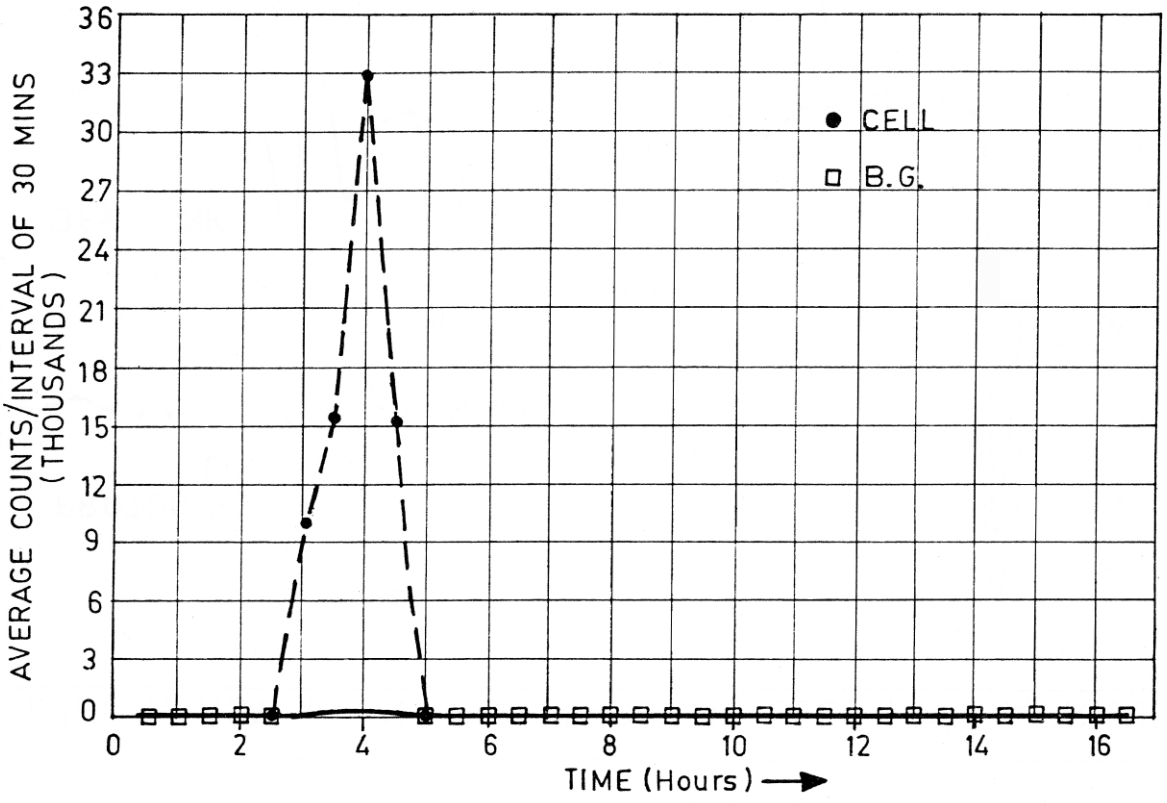

Fig. 3. Neutron yield variation during the burst of 16th June 1989.