

ELECTROCHEMISTRY, TRITIUM, AND TRANSMUTATION

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INTRODUCTION

It is known that nuclear phenomena can be brought about in the cold, e.g., in cells containing D_2O in contact with a palladium electrode, in molten salt cells which contain lithium hydride, in light water cells in contact with nickel electrodes, in cells involving arcs in hydrogen and deuterium, in cells involving high potential differences across a series of palladium electrodes separated by membranes and those involving nickel electrodes under the influence of high frequency radiations.

However, most of these nuclear reactions at low temperatures have been carried out under electrochemical confinement and for this reason a brief outline of these cells is given as a background here.

Two other topics are briefly reviewed: the first published synthesis of tritium from deuterium in the cold, and some aspects of transmutation under conditions which have temperatures in the region of 1000 degrees.

RELEVANT ASPECTS OF ELECTROCHEMISTRY

The Electrochemical Cell

The typical electrochemical cell is shown in Fig. 1. It is filled with, say lithium hydroxide dissolved in heavy water. There are three elements: One is the working electrode (WE) at the interface of which with respect to the solution the electrochemical events occur. Of lesser importance is the counter electrode (CE) which serves to complete the circuit between the working electrode, the power source, and the solution. There is also a reference electrode (RE) as shown in Fig. 1. The potential of the working electrode is measured with respect to that of the reference electrode by means of a high resistance voltmeter.

Most people think that when we refer to the "electrode potential", we are referring to the potential difference between the working electrode and the solution. In fact, "electrode potential" has a complex meaning [1].

The reference electrode is generally the thermodynamically reversible hydrogen electrode. In practice, one sets up a piece of platinum covered with an active form of electrodeposited platinum ("platinum black") at which the reaction $2H^+ + 2e \rightleftharpoons H_2$ occurs and is at equilibrium.

Thus, the so-called electrode potential consists of three potential differences. The first is the one which we would like to know, - and the one which controls the phenomena at the boundary of the working electrode and the solution. But, there is another potential, seldom mentioned, which occurs at the join between the working electrode (consisting, say, of silver) and the reference electrode where the material is platinum. Here, a contact potential difference develops and contributes to the overall potential which comes into what we call the potential on the hydrogen scale. There is, of course, the third potential which occurs at the interface between the reference electrode and the surrounding solution. Of these three potentials, however, the one at the boundary of the working electrode and the solution is the most important.

Equilibrium and the Exchange Current Density

When the working electrode is in equilibrium (at the equilibrium potential, E^0), the rate of electrons leaving the electrode for the solution is equal and opposite to that of electrons leaving the solution for the electrode. Thus there is a dynamic equilibrium. There is no net reaction and the net current is zero. The rate of this equilibrium reaction, in one direction, stated in terms of amps per square centimeter, is the "exchange current density" and it is represented by a symbol, i^0 .

Overpotential

In order that a net electrode reaction takes place, the working electrode potential is to be displaced from the equilibrium value, E^0 , (using an external power supply) either in the negative or in the positive direction. This displacement is called the "overpotential", η . When the potential is displaced in the negative direction, for example, the net reaction is one in which electrons leave the electrode for the solution and a 'net' cathodic current flows. The relationship between this current and the overpotential is given by

$$i = i_0 e^{-\beta\eta F/RT}$$

Here the symbol F means the number of coulombs which reside upon gram ion of univalent ions, R is the gas constant and T is the absolute temperature. The symbol β refers to the "symmetry factor" [2] and the value to use for rough calculations is 0.5.

Suppose we have a negative overpotential of 0.5 V, and an exchange current density of 10^{-6} amps cm^{-2} , the current density is c. 10^{-2} amp cm^{-2} . If one changes the overpotential by about 0.1 volt the current density or rate of reaction, changes by about 10 times.

Fugacity

In discussions in the theory of cold fusion, one sometimes hears the term "fugacity."

Considering the pressure-volume relationship of a gas at high pressures, one of the relations which is often given is the equation due to van der Waals [3]. It runs as follows:¹

$$\left(P + \frac{a}{V^2}\right)(V-b) = nRT.$$

Here the pressure P is what one actually measures. But, the reason for the term $\frac{a}{V^2}$ is to allow for the fact that the measured pressure is less than the pressure which occurs inside the gas. When gas molecules strike a sensor, they experience non-isotropic forces, i.e., they are pulled back towards the gas and consequently the pressure registered is less than the pressure within the fugacity.

Fugacity enters electrochemistry and cold fusion in an important way.

Thus, the fugacity of deuterium gas inside a cavity within a palladium electrode is given by the equation [4]

$$f_{H_2} = e^{-x\eta F/RT}$$

The factor x varies according to the mechanism of the hydrogen evolution reaction. It can be as high as two or as low as one half.

Thus, the fugacity inside a cavity for $\eta = -1.0$ V and $x = 1/2$ is 10^8 Atm, a very high value, while the measured pressure will be $\sim 10^4$ Atm.

TRITIUM

The first formation of tritium to be described in a refereed publication was carried out by a graduate student, Nigel Packham, in the Department of Chemistry at Texas A&M University. In those days (April-May 1989), there was enormous excitement in the Electrochemistry Laboratory and the entire group of about eighteen people were working in shifts of about twelve hours each through the night and seven days per week.

We did not know the conditions for bringing about the nuclear reactions reported by Fleischmann and Pons, so we took the attitude that it would be better to try all kinds of different conditions. Thus, we used electrodes which were untreated, annealed, worked and fused. We cleaned the surface in various ways in addition to leaving it untreated.

We had as many as 18 cells working at the same time. Only a few of these cells were properly controlled electrochemical cells with reference electrode, etc. Many of them were simple test tube cells with a palladium wire cathode and a platinum or nickel anode wrapped around it.

We were trying to find the abnormal heat produced by the cells. However, we looked in addition for the formation of tritium because, obviously, if one has deuterium inside a cell

¹The term V in the equation is the total volume available to the gas and the term n allows for the volume of the gas molecules themselves.

under high fugacity, and a nuclear reaction occurs, one would expect to find tritium in the electrode and eventually in the solution.

Electrolyte samples were analyzed in the Department of Nuclear Engineering for tritium. We were at first disappointed, for, the readings showed only an increase which corresponded to that expected by the classical theory of electrolytic enrichment (for the separation factor of 2 between deuterium and tritium, this comes to an increase of about two times). One day, however, still in April, there was a sudden large increase in tritium counts. At the beginning of the experiment these were of the order of 100 but increased to between 10^4 and 10^5 , $10^2 - 10^3$ times above background!

During the next few months, Nigel Packham himself and then, latterly, Ramesh Kainthla and Omo Velev were able to find tritium in a total of 15 cells during 60 runs[5,6]. These experiments were made largely with palladium in D_2O solutions of LiOH 0.1 mole. However, there were some findings of tritium in titanium electrodes.

We often allowed the experiments to run for a very long time, up to eight weeks in some cases. Tritium was seldom found at less than 100 hours and the average time after which we found tritium was around 500 hours. One can easily see that most persons would have judged the experiment a failure long before this time.

The occurrence of tritium was sporadic.² It occurred in bursts. These bursts would last for several hours and on one occasion about one month.

We tried at first to associate the heat with the tritium and indeed in one run (cf. Fig. 2) it seemed there was some parallelism [6]. However, simple calculation showed that the amount of nuclear heat which would be associated with the tritium would only explain about 0.1% of it. This was the first hint that the tritium formation was a side show in the major heat producing reaction. It had importance historically because the finding of other nuclear debris (helium and transmuted metals) is more difficult to discover from the experimental point of view, that the existence of tritium at this early time and its confirmed finding later in many laboratories was the foundation belief in the existence of nuclear reactions under solid state confinement.

Work on tritium went on and, later, in 1992, Chien and Hodko [8] found both tritium and helium in the same electrode (cf. Fig. 3). Chien also found He^4 in his electrode.³

Hodko also found later [9], using a technique first originated by Szpak and Boss, that the tritium could be brought up to $\sim 66\%$ reproducible by using gold electrodes on which deuterium

²Thus, in a recent paper by Will and Cedzynska[7], so-called "reproducible" measurements of palladium are reported. However, it is made clear in this paper that the reproducibility obtained was by four measurements on exactly the same kind of palladium and when this palladium was changed to palladium of another origin (perhaps with a different surface structure) no tritium was obtained.

³Determination made by Nate Hoffman and Brian Oliver, at Rockwell International. This finding corresponded to that of Milburn Miles at China Lake who made the more difficult experiment of finding helium in the gas phase. It had importance in confirming that the tritium which had been produced was not from spot contamination of the palladium with tritium, for the helium formed under these conditions would have been 3He and not 4He .

and palladium were simultaneously plated out. However, the amount of tritium then found was a few times above the electrolytic enrichment factor.

TRANSMUTATION

It is possible to regard the reactions which form tritium as $D^+ - D^+$ collisions but the distance apart of the D's and in the Pd lattice is greater than that in D_2 . It may be possible to interpret them better from a reaction of D with Li^6 or Pd.

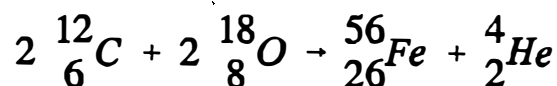
To test the possibility of transmutation reactions with elements of higher atomic weight, several experiments have been tried out in our group. The first was due to the suggestion of J. Champion, who claimed that he had found in 1986 that the imposition of radio frequencies caused the emission of an anomalous heat. Such a phenomenon was witnessed by Kainthla and Velev in 1989 [10].⁴

Champion suggested that, by manipulating the frequencies to match the magnetic moments of individual isotopes, he could provoke a nuclear change in ions dissolved in solution. Experiments of this kind were tried in Texas A&M laboratories in 1992 (under the sponsorship of W. Telander in a project known as the Philadelphia Project) but failed. Champion also suggested heat pulsing and provided a technique of applying a rapid heat pulse to mixtures of mercurous or lead chloride with KNO_3 and carbon. Radioactivity was observed ($\tau_{1/2} \approx 20$ hrs) and in some runs up to 300 ppm of precious metals were found. However, the results were not reproducible.

One of the claims to have provoked a nuclear transmutation reaction concerns conversion of carbon to iron and was recently tried out in my laboratory. The technique was simple. Two spectroscopically pure iron rods with sharpened points were immersed in super-pure water and an arc was struck between them. The carbon detritus which fell to the bottom of the cell was analyzed and was found to contain an amount of iron higher than that had been present initially (as impurity) in the carbon rods.

Table 1 gives the results of this work by Sundaresan and Bockris [11]. Similar results have been reported by others, too [12, 13].

A mechanism suggested by Sundaresan and Bockris is as follows.



Transmutation reactions under solid state confinement have been reported by K. Wolf [14]. It was found that a palladium electrode which had become saturated with deuterium by means of electrolysis contained a large number (more than one dozen) of new nuclei. Kucherov [15] observed that, following a glow discharge between palladium electrodes in D_2 atmosphere,

⁴This work is similar to independent work carried out by D. Letts [16, 17] and also D. Cravens [18]. They describe a procedure in which a palladium cathode, sufficiently charged with deuterium ($D/Pd \sim 0.9$), was subjected to RF fields of moderate (30 - 100 mW) power or magnetic fields of about 200 Gauss; excess heat was observed in a very short time.

many elements were formed in the palladium electrode , some amounting to as much as 0.1 atomic percent.

During 1992-1993, a host of other reports became available which referred to experiments in which the formation of new nuclei under circumstances similar to those described have surfaced. A table comprising some of these reports is given here (Table 2).

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- 18) D.Cravens, *ibid.*, Abstr.# C 3.12.

Table 1. Values of Iron in the Carbon Detritus After Arcing

Experiment No	Wt of carbon detritus (mg)	Fe content (μg)	ppm Fe in carbon
1	269	45	167
2	116	---	---
3	167	---	---
4 ^x	361	45	125
5 ^x	103	15	146
6	231	---	---
7	192	11	57
8	183	5.5	30
9	163	13.5	80
10	143	---	---
11	138	---	---
12	130	5.5	42
13 ^x	471	53	112
14 ¹	477	196	410

^xsame rod

¹ This value is abnormally high; it is possible that an instrument malfunction might have caused the error.

Table 2. Some Attestation of Low Energy Nuclear Change in Condensed Media
(December 1993)

Authors	Date	Reference	Epitome
P.I. Dee Cambridge Un.	1934	Nature, <u>133</u> , 564	Deuterium beam onto (NH ₃ D) ₂ SO ₄ gave DT
Kushi	1976	Michio Kushi, Boston, East West Foundation, Vol 3, (1976)	C → Fe, arc in water
N.J.C. Packham et al. Texas A&M Un.	1989	J. Electroanalyt. Chem. <u>270</u> (1989), 451.	Tritium formed from D dissolved in Pd and at high fugacity
G. M. Miley et al. Un. of Chicago	1991	ICCF 2, paper No.?	Oklo isotopic anomalies interpreted in terms of low energy nuclear reactions such as: ² H + ²³⁸ U → ²³⁹ U + ¹ H.
Enyo & Natoya Hokkaido Un.	1992	ICCF, 4 Paper, N2.1	K Ca
K. Wolf, Texas A&M Un.	1992	Priv. Comm. T. Passal (EPRI), T. Clayton (Los Alamos)	3 Pd electrodes, after D ₂ evolution showed γ and x-rays. Analysis showed many new elements, concentration ~10 ⁹ atoms cc ⁻¹ . But not reproducible.
Kucherov, Lutch Institute, Moscow	1993	ICCF, Paper N3.7	Found new elements after glow discharge Pd in D ₂ Na. In Ag Zr, Nb, Sn + 10 others. Many new materials.
Stringham	1993	ICCF,4, paper C3.8	Sono-illumination of D- saturated Pd in solution He ⁴⁺ + Cd ¹¹⁴
Ohmori & Enyo Hokkaido Un.	1993	ICCF, N 2.3	H ₂ electrolysis on Au electrodes produces heat + Fe. Fe has isotopic abundance significantly different from normal one.
Sundaresan, Texas A&M Un.	1993	Submitted J. Fusion Technology	C Fe, arc in water

Kim & Yoon, Purdue Physics Dept., Zubarov, Hebrew Un., & Rabinowitz, EPRI	1993	ICCF,4 T.2.2	Coulomb barrier is transparent to protons and deuterons in resonance condition. Discusses possible nuclear reaction $D + Pd$ He^4 ; $D + Li$; and involvement ^{39}K and $^{40}Ca_x$ in solid lattice
Chuan-Zan and Yi-Fang, Yunnan Un., Physics Dept., PRC	1993	ICCF,4 T 2.4	Outrightly transmutational. All anomalous heat is due to formation new elements in lattice. > 10 nuclear schemes and reaction sequences suggested. Should expect Ag, Si, Au and many other new nuclei in Pd with H and/or D
Bazhutov, et al., Erzion Center, Moscow	1993	ICCF,4 T 4.9	Outrightly transmutational. There is no cold fusion: only transmutation in the Pd lattice to form numerous new elements
Koretsky, Erzion Center, Moscow	1993	ICCF T 4.10	Treats production of stable isotopes in lattice. States production of gold from mercury-attractive prospect (Sic)
Koretskiy, Erzion Center, Moscow	1993	ICCF.4, Paper 7 4.11	Discusses cold nuclear transmutation reactions in solid lattices. Suggests high speed centrifugation: seems to suggest has found enrichment of Ni-64; Mo-100; Te-130; Xe-136; W-184 and Os-192
F. Will et al. EPRI	1993	J. Electroanalyt. Chem. <u>360</u> (1993), 161.	Tritium formed reproducibly from D dissolved in Pd. Cites 14 papers in peer reviewed journals, report same. Reports extensive testing contamination hypothesis negative.

Singh et al. Bhabha Atomic Research	1993	Priv. Comm. from BARC Center. Ms. available	C Fe arc in water
Nakamura Un. of Tokyo	1993	Chemistry Express, <u>8</u> , 341	C Fe arc in water
Bush & Eagleton Pomona Polytechnic	1993	ICCF 4, paper No. 2.2	Ca from K. Sr from Rb (Isotopic distribution radically different from normal)
Dash Un. of Oregon	1993	ICCF, 4 paper M2.1	Light water electrolysis Ag and Au appearing in Pt
Dufour, Shell Oil Co. Research (Paris)	1993	ICCF,4 Paper N1.6	Predicts creation of new metals from Pd-D reactions. Finds 4 new metals
Miles & Bush, China Lake, Navy Station	1993	ICCF,4, paper C,2,9	All steel equipment prevents leaks from air. He ⁴ in gas is 1/3rd that necessary account heat on D+D He ⁴ + γ
Srinivasan et al., Bhabha Atomic Research, India	1993	ICCF,4 c. 3.8	Obtained heat and tritium from light water cells
Romodanov et al., Lutch Institute, Moscow	1993	ICCF,4 Paper N.3	Reproducible tritium from glow discharge
Hagelstein, MIT	1993	ICCF,4 Paper T.1.1	Theory of neutron transfer reactions and lattice induced low energy nuclear changes
Li, Physics Dept., Beijing Un.	1993	ICCF,4 T.2.1	In low energy nuclear change in solids, his theory show no γ radiation (sic). Main expectation is β (sic)

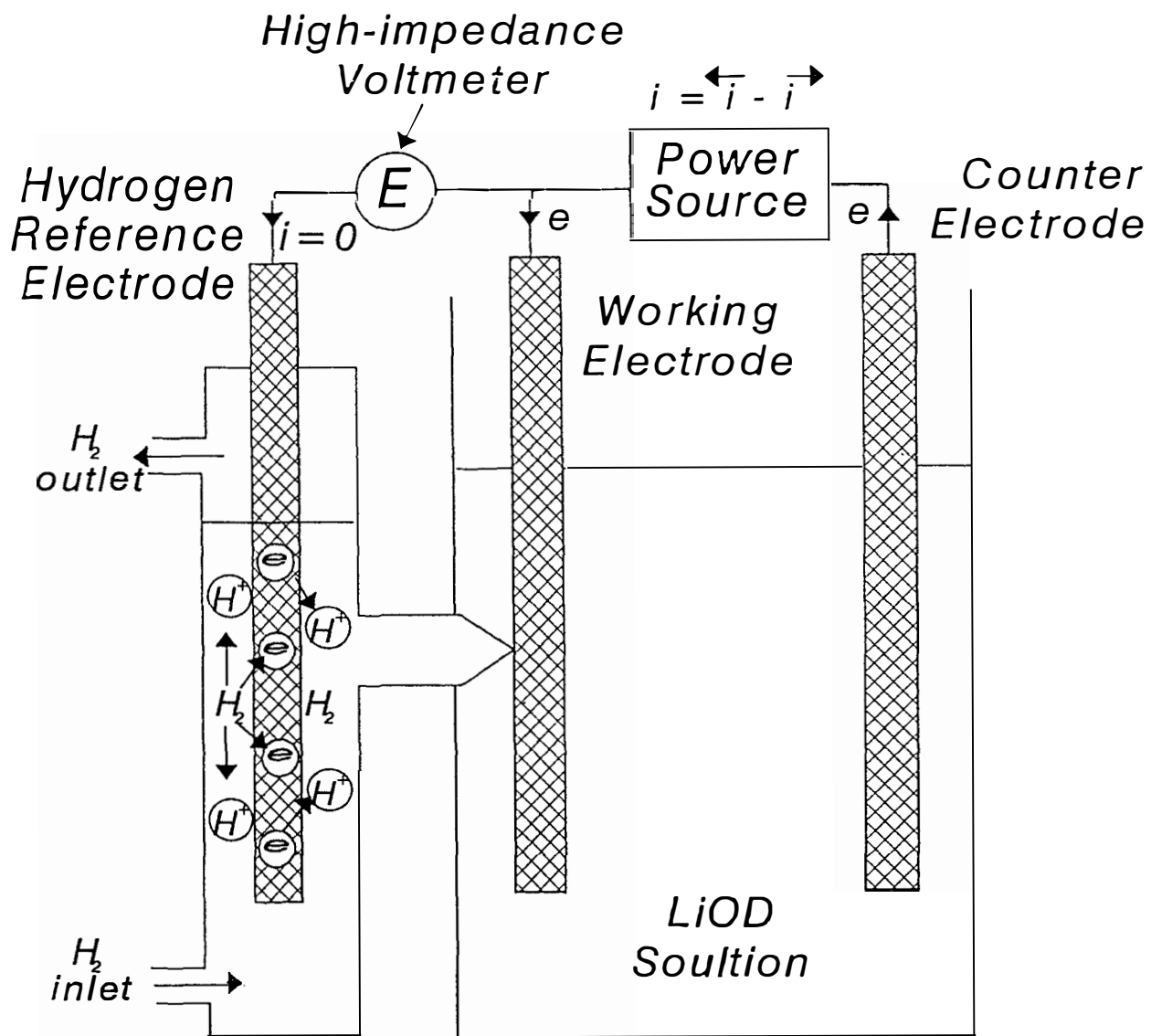


Fig. 1. Electrolysis Cell

CELL 4 % EXCESS HEAT

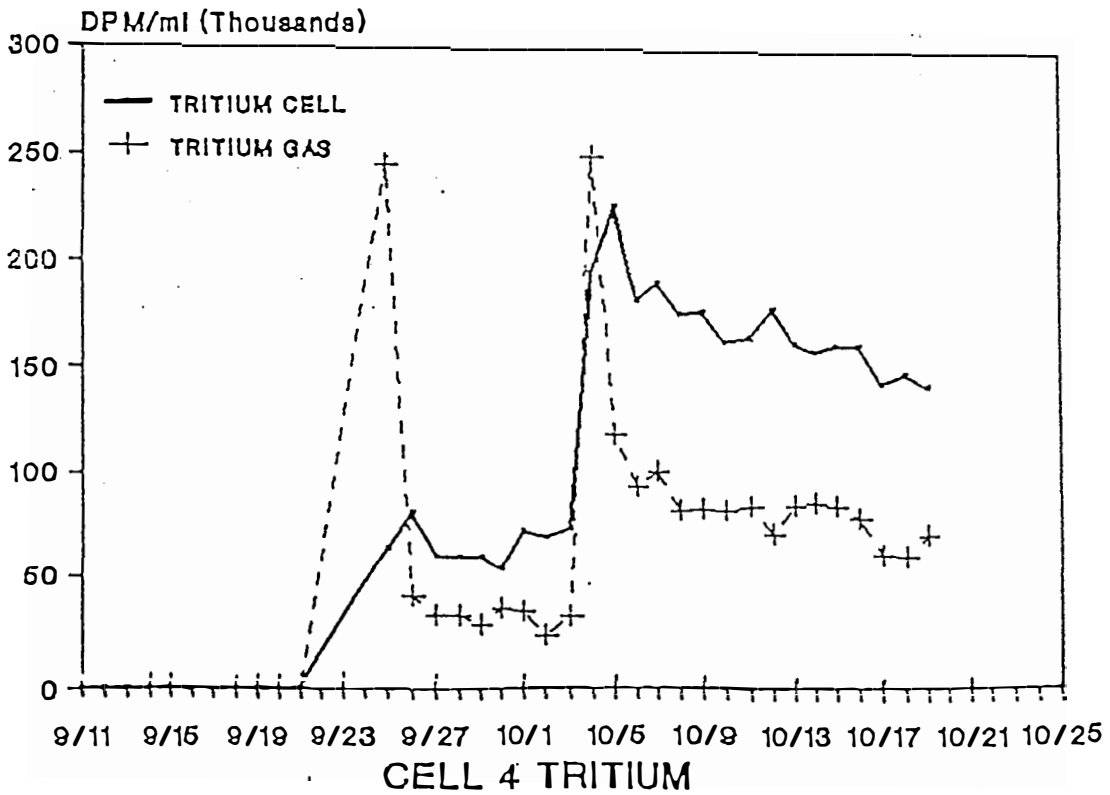
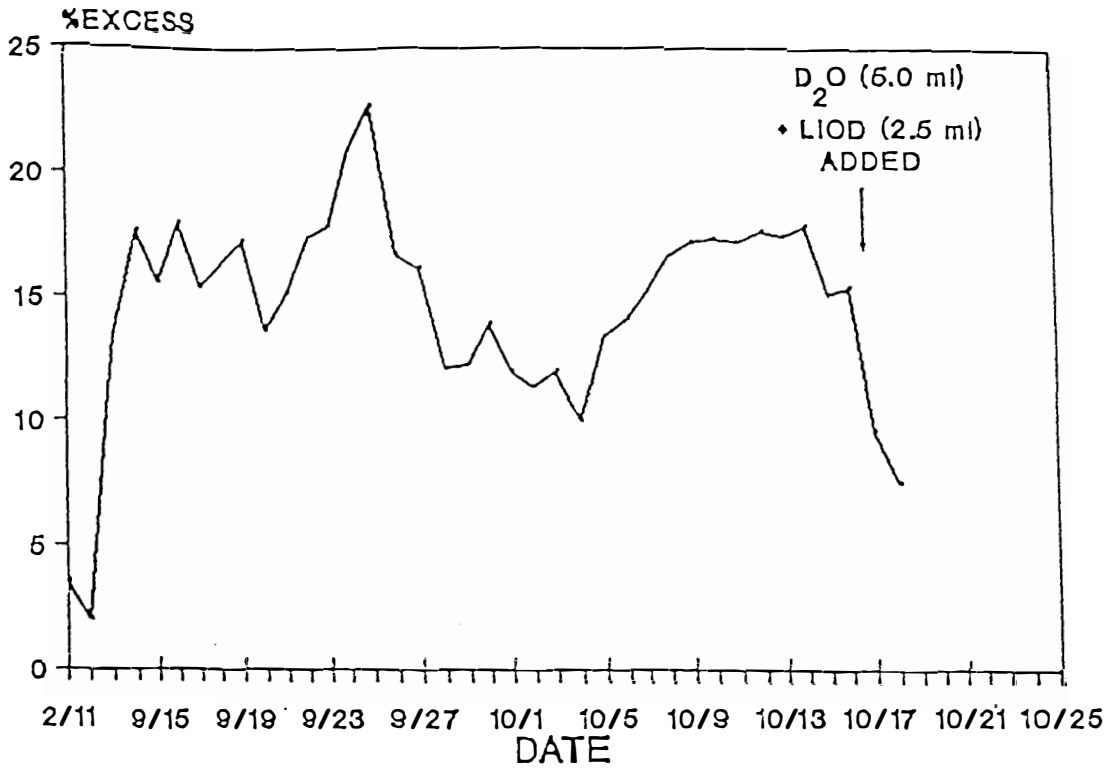


Fig. 2 The Heat Measurements and Tritium Activity Levels Showing a Possible Correlation Between the Two [Ref. 6]

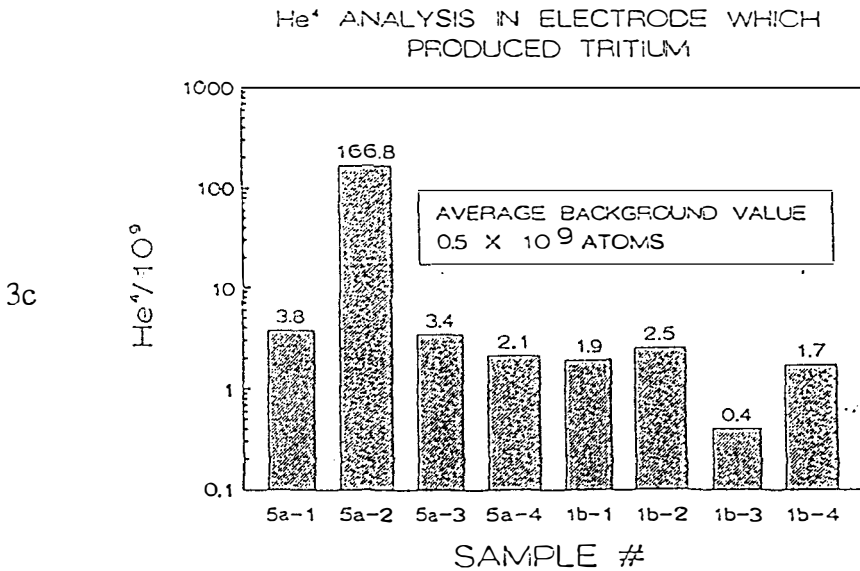
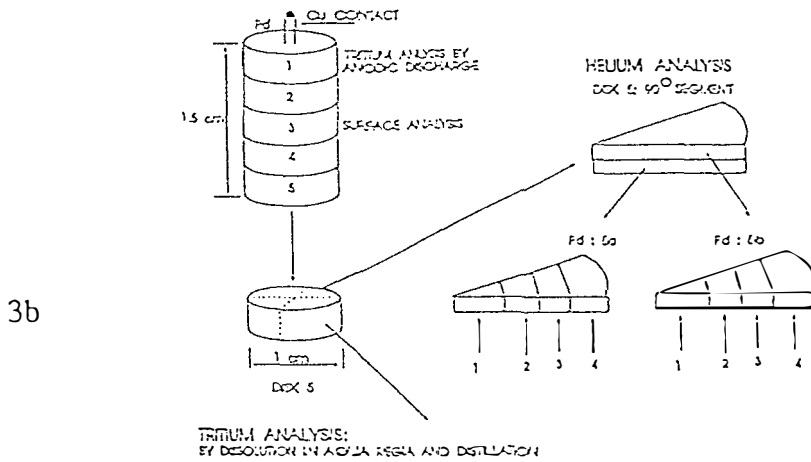
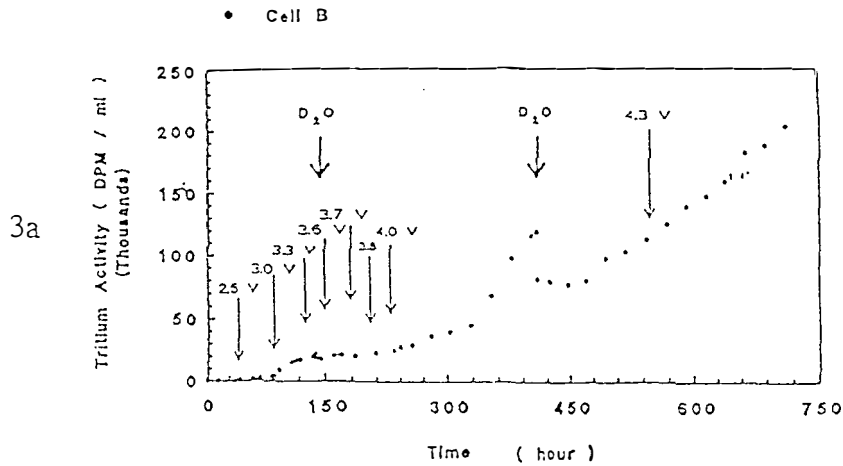


Fig.3 Helium Analysis of the Electrode Which Produced Tritium : a) Tritium Activity in the Electrolyte as a Function of Time; b) Electrode Segments; c) Helium Analysis of Segments. [Ref. 8]