Search for Excess Heat, Neutron Emission and Tritium Yield from Electrochemically Charged Palladium in D_2O

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ABSTRACT

The electrolysis of heavy water is being investigated with two types of open calorimetric systems. Pd cathodes, Pt anodes and D_2O/0.1M LiOD electrolytes have been used. Until now no clear-cut heat bursts as reported have been observed. One exceptional phenomenon showing abnormal power imbalance without neutron and tritium anomalies was found, but has not been repeated under the similar experimental conditions. Neutron emission, on the other hand, as a very rare case showed an abnormal increase for only short term during one of another series of experiments. The increase of about 3.8σ above the background level lasted for 9 hours on the 20th day after starting the electrolysis. The emission rate amounts to about 27.2 ± 11.2 neutrons s^{-1}, which is equivalent to about 700 times as much as the background level. Neither excess heat nor tritium anomalies were, however, observed. The reason for the lack of repeatability of these experimental results is discussed.

1. Introduction

The generation of excess energy and possible products of nuclear interactions, as proposed by Fleischmann, Pons and Hawkins[2] and Jones et al.[4] has attracted much attention from scientific point of view of metal-hydrogen systems. We were particularly interested in observing by ourselves a "burst" of excess enthalpy lasting a long period of time and in experimentally making clear if it comes from nuclear reaction or not. Aiming to reproduce the claimed excess heat and neutron emission from electrochemical cell, power balance and radiation emission measurements have been continuously carried out[3] for more than 3 and a half years with several sets of open type cells consisting of Pd cathodes, Pt anodes and D_2O/0.1M LiOD electrolytes.

2. Electrochemical Cells

Two types of glass cells have been prepared as shown in Fig. 1; a) water-cooled cells with a cooling water jacket; b) thermally insulated dewar type cells immersed in constant temperature water baths. A variety of Pd rods have been tested as the cathode. Original materials of 99.8 % or 99.95 % were delivered from Johnson

Matthey Company. They were as received, annealed *in vacuo* or remelt in Al_2O_3 crucibles with a UHV induction furnace. Electrolysis has normally been done in constant current mode, although the current was sometimes modulated in various manner. For a) type cells, relatively larger samples were used with a current density up to 1050 mA/cm^2. Power balance was checked by comparing the curve of cell temperature vs. input power with the calibration curve, or by measuring the flow rate and the temperature difference of inlet/outlet water in a cooling channel as shown in Fig. 2. For b) type cells, relatively smaller samples were used with a current density up to 840 mA/cm^2. Excess heat was monitored only by use of the calibration curve.

3. Measurement of Nuclear Products and Chemical Analysis

For neutron monitoring a single or a double rem counter system was used, in which dose meters of BF_3 and/or ^3He type were placed adjacent to the electrolysis cells (See Fig. 3). Tritium concentrations in the cells before, during and after electrolysis were intermitently measured with a liquid scintillation counter by an external standard method[3]. In order to check the hydrogen contamination during long-run electrolysis, a quantitative analysis of H/D ratio was also made by infrared spectrophotometry[1]. The Li concentration was measured by ion chromatography technique. Surface analysis of Pd samples was also done with EDX as well as SEM.

4. Results and Discussions

Although many indications of excess heat have been observed so far, almost all of them except one has been excluded as a ghost by further experimental reexamination. They were caused by; 1) freezing of electrolyte; 2) a change of calibration curve with time due to unexpected escape of Li from cells; 3) an observational error in measurement of temperature difference; 4) a change in calibration of flow rate of the cooling water. A typical example is shown in Fig. 4. Until now we have never observed a clear-cut heat burst as shown in the reference[2]. Only one example of abnormal power imbalance observed in an a) type cell has not been ruled out, however, even after re-calibration of the flow rate. Neutron and tritium showed no anomalies in this case. This phenomenon has not been repeated any more under the similar experimental conditions.

Neutron emission, on the other hand, showed an abnormal increase in one of a series of b) type experiments as summarized in Fig. 5. In these experiments four cells were installed together in each bath and two BF_3 counters (15 and 16 in Fig. 3) were used for neutron monitoring. On the 20th day after starting the electrolysis one (15) of the counters detected the increase of neutron emission, about 3.8σ above the background. It lasted for about 9 hours. Another counter (16) located 3m apart showed a small increase of counting rate, but in the limit of statistical error. No observation of solar neutrons associated with the large flare has been reported on this date. But one (6) of the four cells coincidentally showed a spontaneous increase in current. If this cell is thus assumed to be a neutron emission source, its net emission rate is equivalent to about 27.2±11.2 neutrons s^{-1}. This phenomenon also happened only once without accompanying any heat excess or abnormal increase of tritium.

Experiments were also carried out to check the contamination of D_2O with hydrogen from air during long-run electrolysis. The results proved that the D_2O or the electrolyte, and thus the Pd could be contaminated with unexpected amounts of hydrogen from air. It is reported that 0.9 D per one Pd is the critical threshold value to get the cold fusion phenomena. If this is the case, the hydrogen contamination could account for the lack of reproducibility of above mentioned experimental results.

5. Acknowledgement

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6. References

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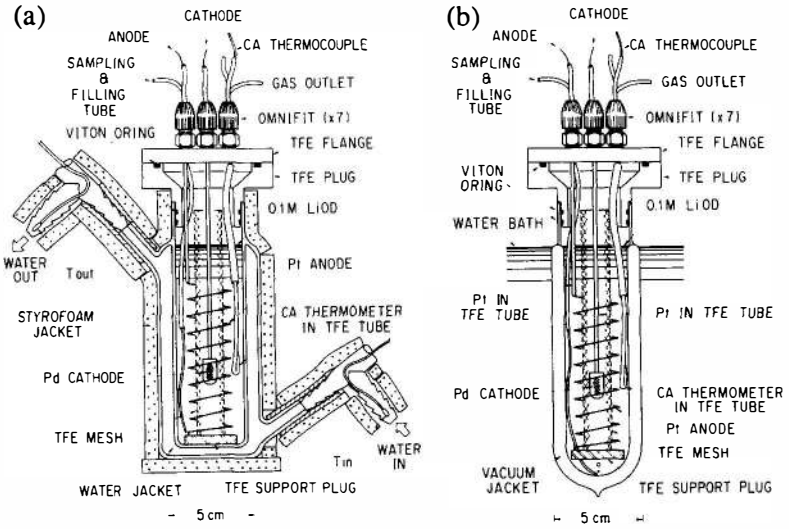

Figure 1.(a) Water-cooled cell, (b) Vacuum insulated dewar type cell.

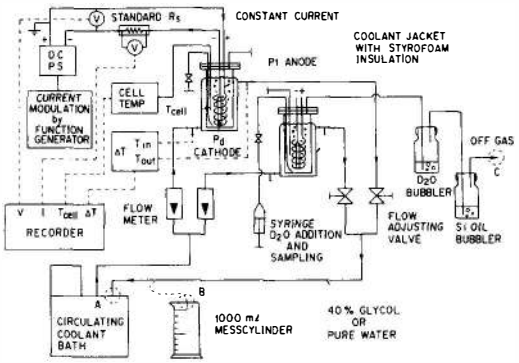

Figure 2. Schematic diagram of calorimetry of a) type cells. Coolant and gas flow rate can be calibrated at B and C positions, respectively.

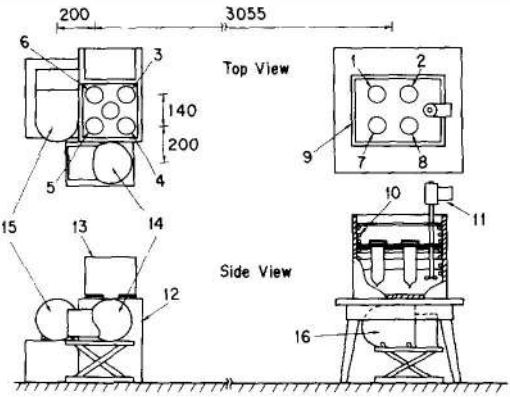

Figure 3. Setup of b) type experiment. 1 ~ 8 Dewar type cells in which 8 is for control experiment, 9 Coleman bath, 10 Cooling pipe, 11 Stirrer, 12 RTE220 bath, 13 Temperature controller, 14 ^{3}He rem-counter, 15, 16 BF_3 rem counters S/N 12254 and 9074, respectively.

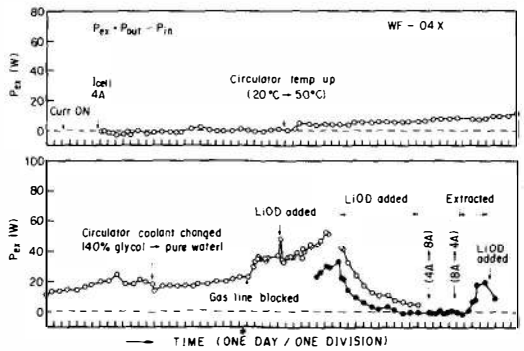

Figure 4. Change of virtual excess power with time. Filled circles denote the same data after flow rate re-calibration. Adjusting the Li content to the original value diminishes the virtual excess power to 0.

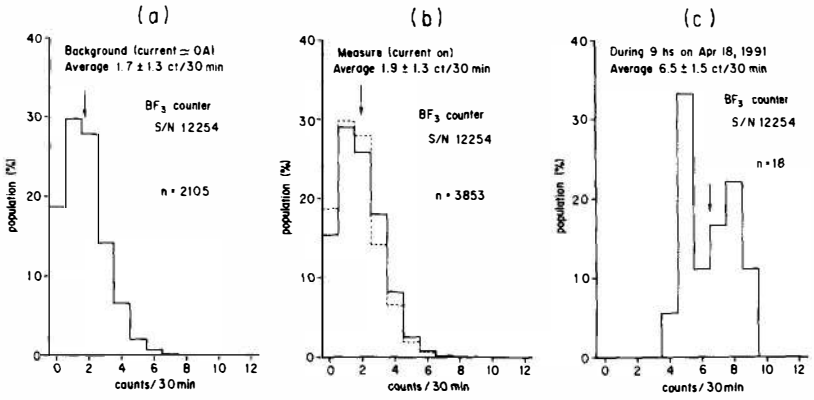

Figure 5. Frequency histograms of the neutron counter in a b) type experiment. Dotted line in (b) indicates the background as shown in (a). Fig. (c) shows the anomalously high counts lasting for 9 hours.