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Nuclear and Thermal Effects During Electrolytic Reduction of Deuterium at Palladium Cathode

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In a galvanostatic experiment of charging deuterium in a palladium cathode, nuclear and thermal effects were found. A sintered palladium electrode shaped as parallelepiped was used. After 6 days of electolysis at 200 mA/cm^2, a simultaneous emission of neutrons, tritium excess in the electrolytic solution, and temperature rapid increase was observed. During the event which lasted 4 minutes, we counted 7.2×10^5 neutrons while the electrode temperature reached 150°C. Electrochemical procedure for charging the palladium electrode by deuterium using galvanostatic pulses as well as the associated electrode temperature trends are shown.

KEY WORDS: Cold fusion; Pd-D system; Pd electrochemistry.

1. INTRODUCTION

On the frame of the F and P[1,2] experiment, we try to perform experiments of so-called "cold fusion" through the electrolytic reduction of deuterium at a palladium cathode. The experimental procedure differs from that used in Ref. 1 in the following points: (1) a sintered palladium electrode (a parallelepiped of 6X5X25mm) was used instead of a cast metal electrode, (2) temperature of the electrode was measured instead of the temperature of the electrolyte solution, and (3) calorimetric measurements were not carried out.

For the detection of nuclear phenomena, neutron and gamma emission were monitored. In addition, data acquisition system was programmed in such a way as to switch off the electrolysis for an electrode temperature up to 80°C.

The experimental electrochemical part is essentially constituted by a three-electrodes assembly to carry out galvanostatic measurements, in which the working electrode is the palladium electrode having a S-type thermocouple embedded in it. The counter electrode is a cylindical platinum mesh while the reference electrode is a mercury–mercury oxide electrode in alkaline solution. The electrolyte is an LiOD 0.1 M solution and the cell is placed in a waterbath which acts as thermostat. In Fig. 1, a scheme of the electrolysis cell is reported. As shown in the figure, the electrode, whose terminal part along the major axis has been machined, is sealed to a glass tube. The thermocouple is also sealed into the electrode. The thermocouple is protected by fused glass in such a way as to eliminate any interaction between thermocouple wires and deuterium. In a previous light-water experiment, in which no thermocouple protection was used, the thermocouple signals were disturbed by high noise.

Referring to the nuclear measurements, we detected neutron emission by a ^{3}He dosimeter with an energy range up to 7 MeV.

The efficiency was measured to be 10^{-4} in isotropic conditions by an Am-Be source. In our experimental geometry, we estimated an efficiency of 5×10^{-5}. This neutron detector is connected to a rate meter counter-recorder system. It was previously checked that each spike on the recording corresponds a count on the counter. The γ-ray measurement was also carried out through a large sodium iodide monocrystal connected to a SILENA spectrum analyzer. The apparatuses for the nuclear measurements were placed around the electrochemical cell

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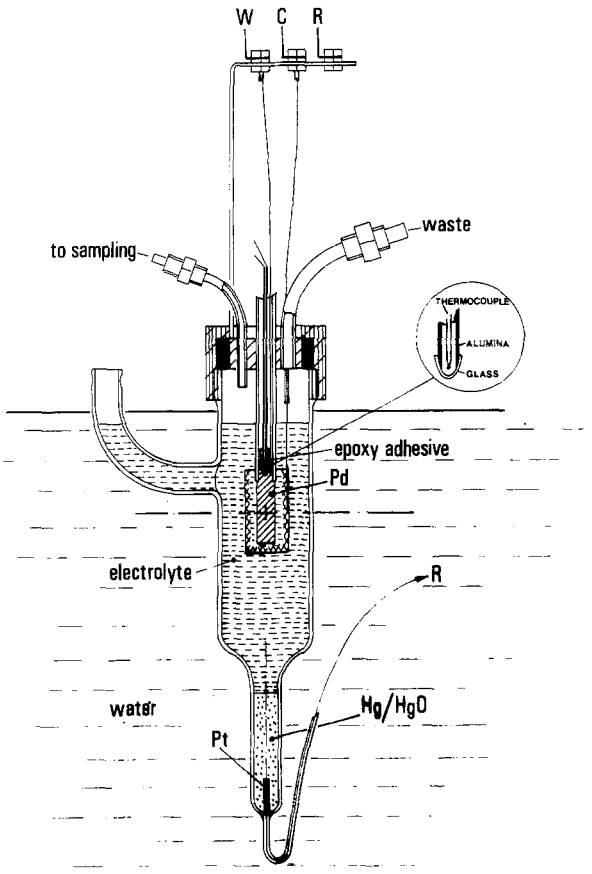

Fig. 1. Electrolytic cell assembly.

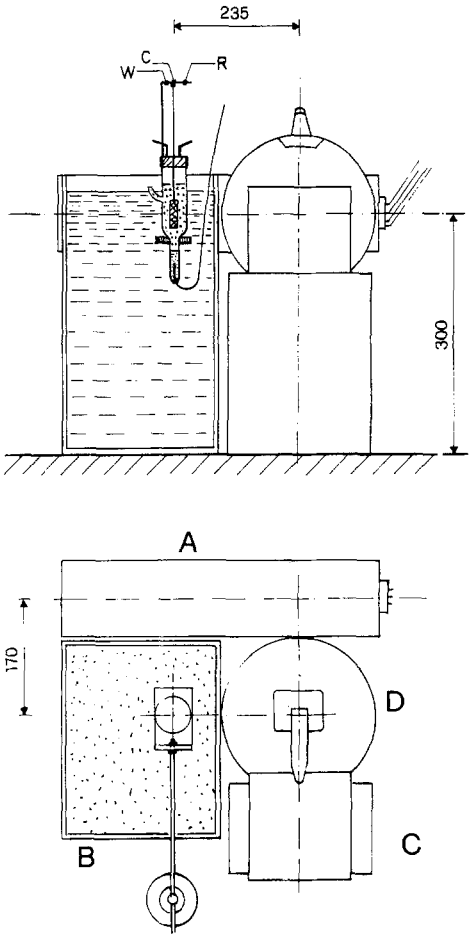

Fig. 2. Neutron and γ-ray detector positioning. (A) NaI (TI) monocrystal, (B) Waterbath, (C) He^3 neutron dosimeter, and (D) polyethylene sphere

as schematized in Fig. 2. Through a data acquistion system, electrochemical parameter and temperature value were transferred in the computer while the neutron detector signals were recorded. Therefore, there is not a synchronized data acquisition for temperature, electrochemical parameters, and neutrons.

During the experiment, some electrochemical measurements were also carried out concerning the possibility of obtaining information about the deuterium content in the palladium electrode, i.e., the palladium "deuterium-charge" state. A well-known electrochemical method was used to charge the Pd electrode electrochemically. This is shown in Fig. 3 where the palladium electrode potential and the temperature is reported vs. time when galvanostatic pulses of different durations are applied to the cell. Two parts are distinguishable in Fig. 3 (upper, part A) in which the current was switched off (EMF measurement) and part B where the current was passing (Fig. 4). In the same Fig. 3 (bottom), the related temperature variations during those steps, are also shown. Figures 4 and 5 which are, respectively, enlarged views of sections B and C of Fig. 3 show that there is a progressive change of the shape of the chrono-potenziometric curve during the D charging in palladium. There is an initial peak which disappears progressively. The associated electrode temperature curves seem to reproduce this behavior. A tentative explanation of this could be found in the change of the D chemical potential gradient in Pd electrode as D charging proceeds. This procedure changes in the diffusion overvoltage which is a part of the total electrode overvoltage. By plotting the EMF values vs. the passed coulomb charge, the charging curve, shown in Fig. 6, was obtained. This curve could be a way to check the time evolution of the deuterium content in the Pd electrode.

After 150 hours of electolysis at 200 mA/cm^2, a nuclear and thermal effect was simultaneously recorded, as shown in Fig. 7.

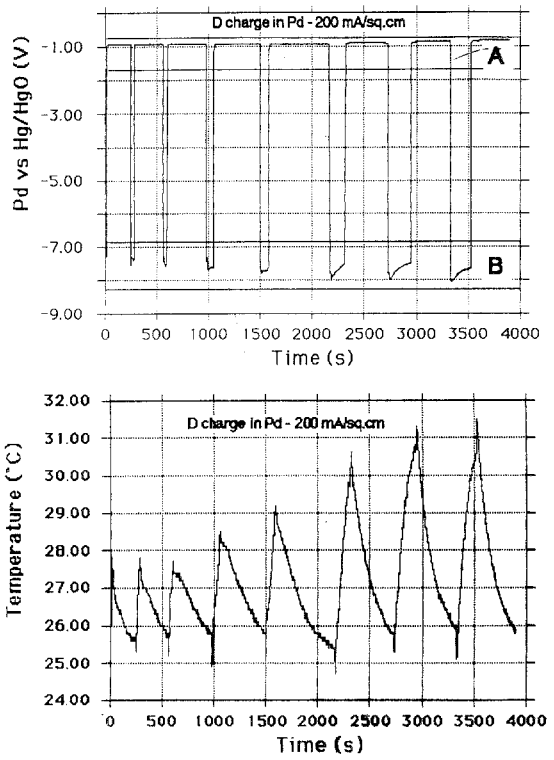

Fig. 3. Palladium potential (upper) and electrode temperature vs. time, recorded when galvanostatic steps were applied to the cell. (upper): Section A: $i=0$, section B: $i=200$ mA/cm^2.

At a certain time, the temperature grew very sharp, and because the recorder was set at the lowest full scale of 1 mV, the pen went full scale. At the same time, a densification of the spikes was measured by the neutron recorder; this is only a part of the total recording. Data refers to a time acquisition of 22^h 5' 54", and 116 was displayed on the counter. If we count each spike in the recording, we have 80 spikes. At a time interval of an estimated of 4 min were concentrated 36 spikes. Before and after the event, the neutron counting was that of the background level of 3.0 $\pm$ 0.2 counts per hour and during the 4 minutes of the event we had 36 counts. Therefore, in that time interval, we had a neutron emission equal to about 180 times the background level. By extrapolating the heating and cooling curves it was possible to obtain the maximum temperature reached by the electrode as shown in Fig. 8. At 135 s, current was switched off by the data acquisition system since temperature was over 80°C while the neutron emission lasted approximately 105 s. If we estimate a temperature increase of about 100°C, by taking into account the mass of the palladium electrode and its specific heat, we have an energy produced of 176 J $=1.1 \times 10^{21}$ eV. In addition, by taking into account the efficiency of the detector we find that 7.2×10^5 neutrons have been emitted. Furthermore, considering the evaluated energy (176J) to be due to the nuclear fusion process $d+d \rightarrow He^3 + n$, we would have a neutron emission of 1.4×10^{15}.

At the time these proceedings were prepared, we can definitely confirm a tritium excess corresponding to $(2.14 \pm 0.04) \times 10^{11}$ atoms related to the solution volume of 41 ml. More detailed information concernig the experimental and other results have been the subject of a paper recently submitted.[3]

2. DISCUSSION

MR. TURKEVICH: Are there any questions?

MR. PETRASSO: Richard Petrasso from MIT. I'm a little confused about your neutron rate. You said first 150 times background and it took an efficiency of 10^{-4}, and I get aroun 10^3 neutrons per second, and that's...

MR. GOZZI: Per hour.

MR. PETRASSO: Right.

MR. GOZZI: 3.5 neutrons per hour.

MR. PETRASSO: Okay, thank you.

MR. TURKEVICH: Any other questions?

MR. GARWIN: Dick Garwin, IBM Research. I can imagine that the catastropic event in which the temperature rose 100 degrees or so was not fusion at all but simply a response to having stuffed deuterons into high-energy sites in the lattice, similar to vigner (phonetic) energy in graphite in reactors. And you see the neutrons, but they may have nothing at all to do with the heat that caused the temperature to rise. To discriminate between these two cases would be to find 10^{14} helium 4.

What has happened to that sample? Have you looked for helium 4 in that sample?

MR. GOZZI: I don't know exactly the process that you mentioned but one can think about several chemical processes that can increase the temperature of the electrode if the effect is concentrated in a short time. But the more realistic process that I can imagine is the direct combination of hydrogen and oxygen due to the high catalytic activity of the deuterated electrode. But this process, also by the experience of some of my colleagues, is possible only if the electrode is exposed to the gaseous atmosphere.

After the event, I switched on the current again, and the current was applied for 24 hours. But any other event was observed.

MR. GARWIN: There is a very good poster upstairs which says if you had helium 4 in there, you still

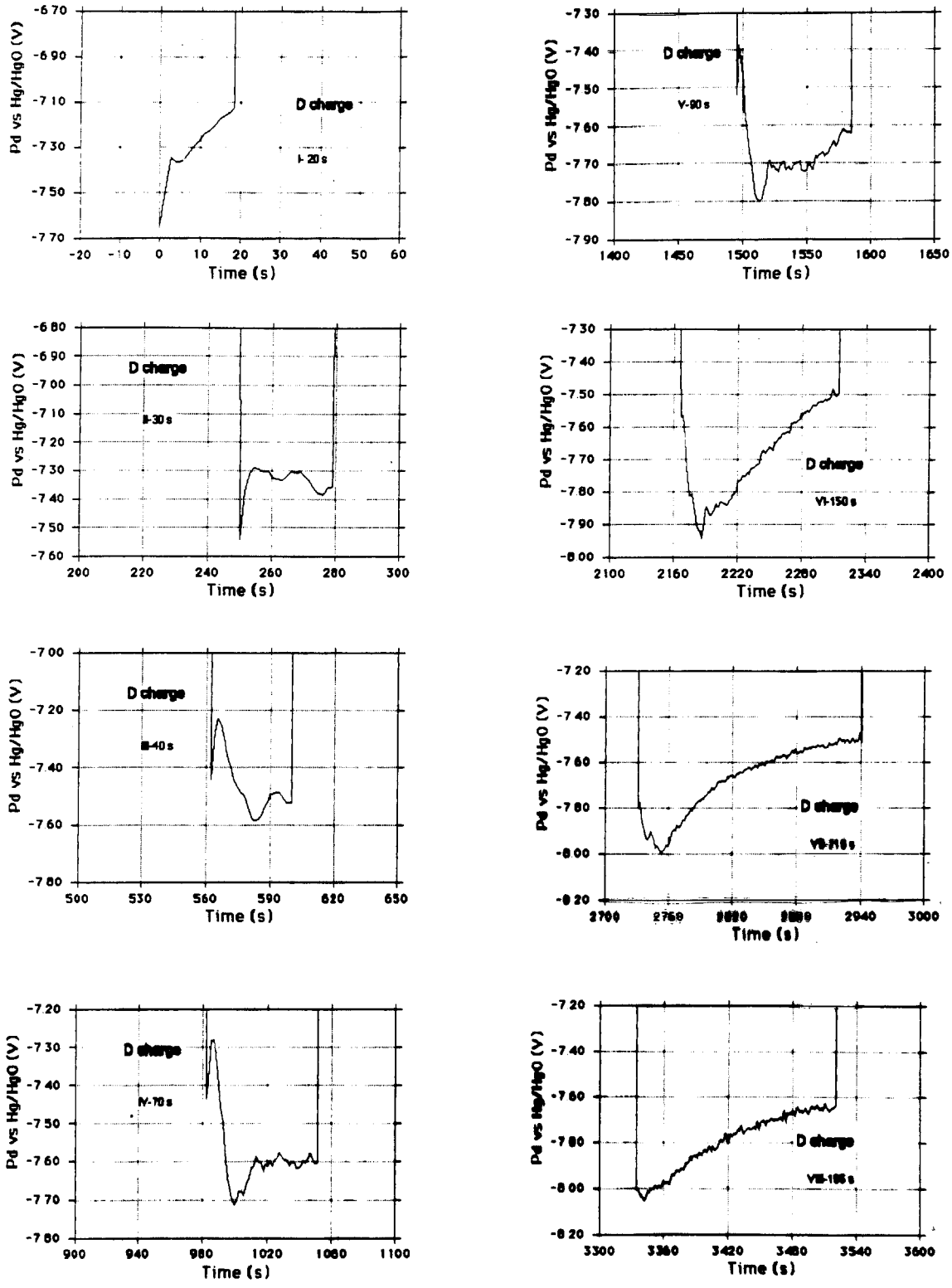

Fig. 4. Magnifications of the $i=0$ region (part B of Fig. 3).

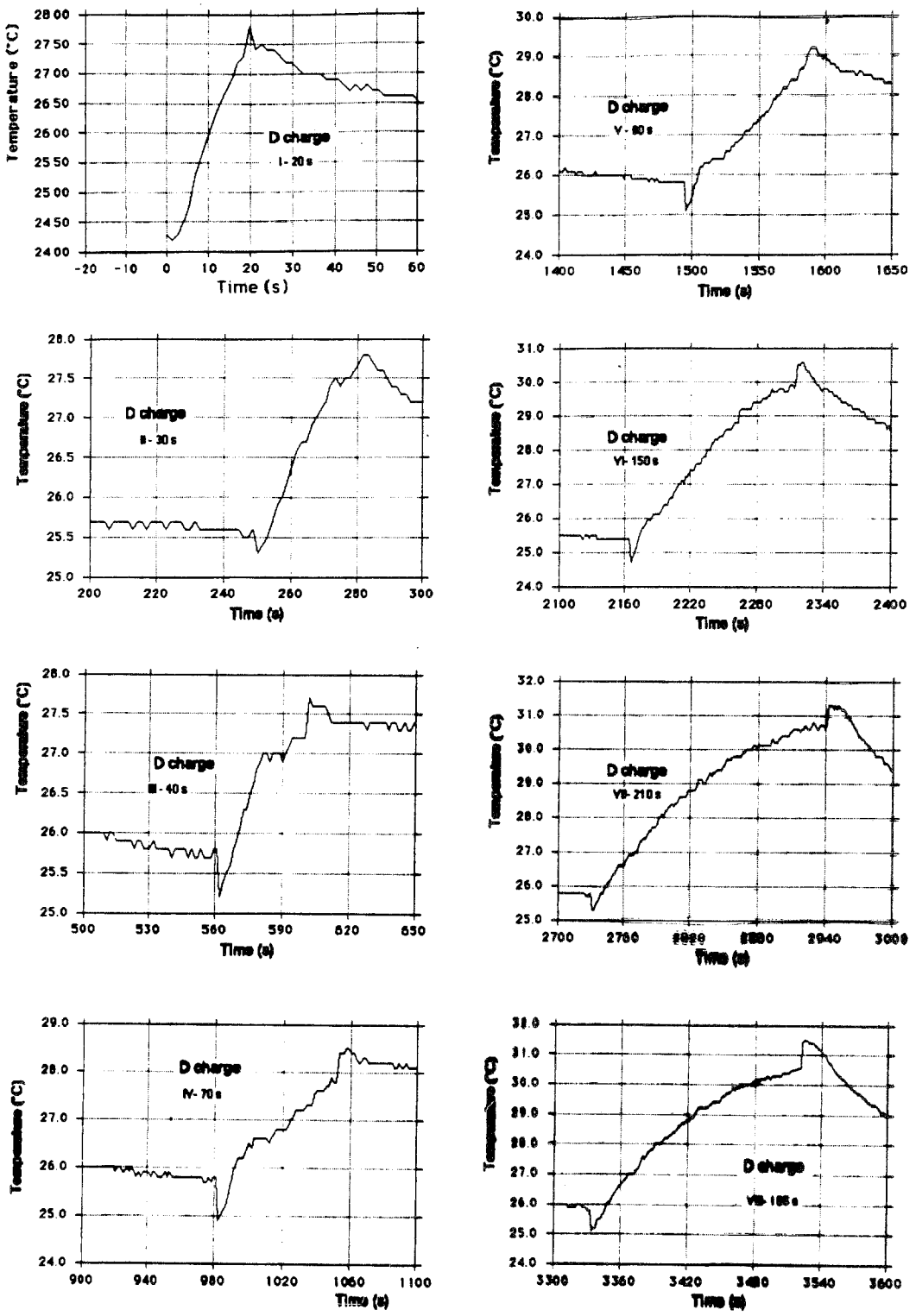

Fig. 5. Magnifications of the electrode temperature during galvanostatic step (bottom of Fig. 3).

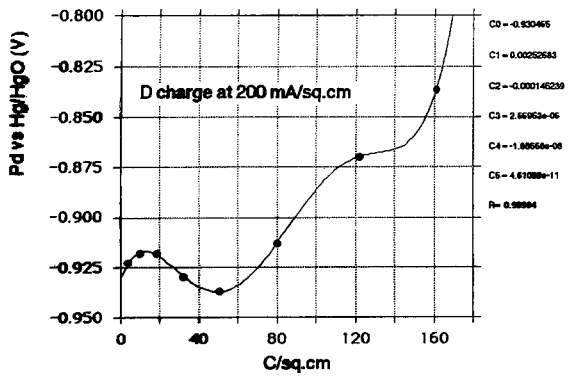

Fig. 6. Pd electrode potential as function of the passed electric charge at 200mA/cm².

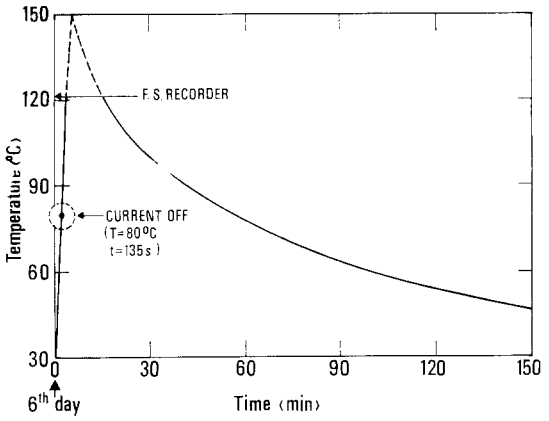

Fig. 8. Extimation of the maximum temperature reached by the palladium electrode during the event.

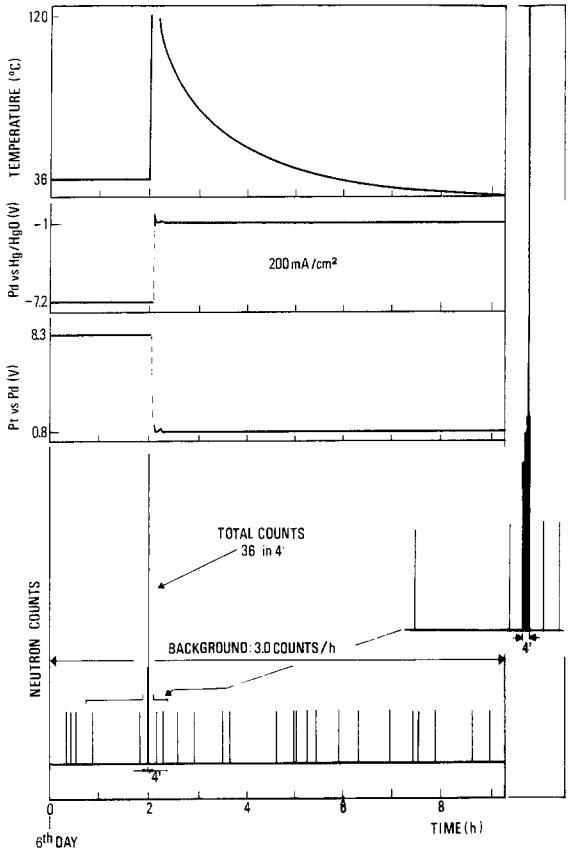

Fig. 7. Experimental variables recorded during the event which produced nuclear and thermal effect.

have helium 4 in there, you should take that cathode and have it analyzed.

MR. GOZZI: Okay. We are trying to do this.

MR. TURKEVICH: Time for one more question, I think, over here.

MR. MELENDRES: Carlos Melendres, Argonne National Laboratory. Have you reproduced this result, and how sure are you that these sudden bursts in neutron counts and heating of the cell is not due to a power surge in your room?

MR. GOZZI: Yes. Before leaving, I started another experiment. During this period, the cell is cycling electrochemically to activate the palladium electrode, I hope to reproduce.

MR. MELENDRES: You have not reproduced the results?

MR. GOZZI: It's the only one. Referring to your question, we did not record any other fluctuation due to some power surge. All the apparatuses are connected to a continuity group which separates from the electrical network as well as stabilizes the power.

MR. MELENDRES: What I'm saying is ...

MR. GOZZI: But you think we observed first neutrons.

MR. MELENDRES: Because the heat and the neutron events are so correlated, I was just wondering whether this was due to a sudden surge in power in your room, and I wonder whether you checked that.

MR. GOZZI: Electrical?

MR. MELENDRES: Did you have your system plugged to the same outlets or something like that? I don't know. I'm just kind of speculating.

MR. GOZZI: I don't understand the question.

MR. TURKEVICH: Thank you. I think we had better go on to the next talk on Calorimetric Measurements on Electrochemical Cells with Pd-D and Pd-H

Cathodes by Redey, Myles, Dees, Krumpelt, and Vissers of the Argonne National Laboratory.

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