EVIDENCE FOR NEUTRON PRODUCTION DURING HEAVY WATER ELECTROLYSIS ON PALLADIUM ELECTRODE

NUCLEAR REACTIONS IN SOLIDS

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Neutron burstlike emissions were detected during galvanostatic electrolysis of heavy water. A sintered palladium cathode of cylindrical shape was used. After 166 h of electrolysis at 200 mA/cm^2, two bursts of neutrons with durations of ~200 and 100 s were observed. The cathode temperature did not show any change.

INTRODUCTION

After the work of Fleischmann and Pons[1] and Jones et al.,[2] many groups have been trying to confirm the existence of nuclear fusion at room or even lower temperatures. Some reported positive results[3–8] indicated that the phenomenon was burstlike, lasting a few seconds or less. On the other hand, other experiments found that the neutron emission seemed not to be concentrated in a few bursts but rather distributed over the full duration of the run.[9–12] In this type of research, it is especially important to obtain independent confirmation of the results. However, in most cases, positive results obtained at given laboratories have not been reproduced by others.

In this work, an attempt was made to repeat in as similar conditions as possible the experiment of Gozzi et al.[5] on neutron production and heat generation during heavy water electrolysis on a palladium electrode.

EXPERIMENT

A cylindrical palladium cathode was prepared as in the work of Gozzi et al.[5] Commercially available palladium powder of 1-μm average grain size and 99.95% purity (X-ray fluorescence spectrometry carried out on the powder as received showed traces of iron and copper) was pressed at ~216 MPa in a stainless steel die. The cylindrical pellet was then sintered in vacuum at 1173 K for 12 h and cooled to room temperature. The final mass of the electrode was 8.2 g, and the density was 80% of the bulk palladium. The electrode had a diameter of 11 mm and a height of 9 mm. It was fixed in a shrinking Teflon tube with an epoxy resin with 2.5 cm^2 of exposed area.

A copper-constantan thermocouple was in close contact with the cathode and was isolated from the surface of the electrode by a thin mica layer.

An undivided electrochemical cell (glass) filled with 60 cm^3 of 0.2 M D_2SO_4 solution in D_2O (99.8% purity) was thermostated at 298 K. A cylindrical platinum gauze and a saturated calomel electrode were used as counter and reference electrode, respectively. The electrolysis was carried out galvanostatically at 200 mA/cm^2. Every 2 days, the appropriate amount (15 cm^3) of D_2O was added to the cell to replace D_2O consumed by the electrolysis.

A ^{6}Li-glass scintillation counter,[13,14] similar to those developed by Kedem and Kedem,[15] was used as the neutron detector. It consisted of 2-mm-thick ^{6}Li doped glass scintillator (NE-912), 5.12 cm in diameter, coupled by a light guide to the photomultiplier. The scintillator was 98% opaque to thermal neutrons, which were detected by the $^6\mathrm{Li}(n,t)^4\mathrm{He}$ reaction. A 5-cm-thick polyethylene moderator was selected to optimize the sensitivity to 2.45-MeV neutrons. The sensitivity (number of source neutrons emitted at a 4π solid angle per measured count) of the detector for deuteron-deuteron neutrons and for the geometry used was estimated by using a weak ^{252}Cf spontaneous fission source. Its value was (152 ± 15) neutron/count.

The electronics and the acquisition system were arranged in such a way as to enable simultaneous monitoring of the pulse heights from the neutron detector,

as well as the temperature of the palladium electrode and the potential difference between the palladium and the reference electrodes. The minimum time that could be resolved by the acquisition system was 20 ms; i.e., each event was localized in time with accuracy of 20 ms and in such a way stored.

The experiment was carried out in a large underground laboratory[13,14] under the constant temperature condition (295 K). The detector and the electrolysis cells were shielded against environmental gamma rays (25 cm of lead) and neutrons (9-cm layer of borax powder and 10-cm layer of paraffin).

Electrolysis started on June 30, 1992, at 11 h 15 min and lasted until July 9, 1992, at 8 h 50 min (Greenwich mean time). During this period, 12 measurements were done with an overall duration of 677 660 s. During the measurement, short runs with the ^{252}Cf source were repeated regularly to check the stability of the electronics. Each measurement was carefully analyzed, off-line, by sorting the counts from the neutron region into 10- and 1-s bins. By this procedure, multichannel scaling (MCS) spectra were formed for each measurement.

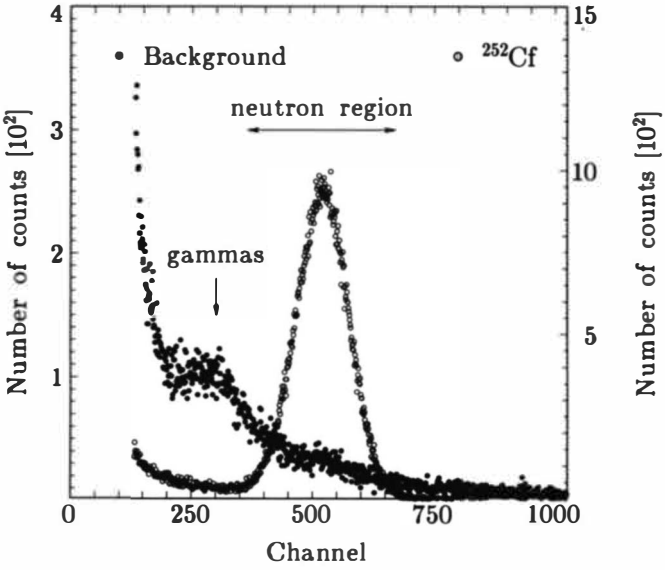

Fig. 1. Scintillation detector pulse-height spectra from background measurement and from the measurement with a ^{252}Cf source.

RESULTS AND DISCUSSION

The experimental setup was tested throughout several months before the beginning of the experiment. Without any shielding and without the electrochemical cell, the background counting rate in the neutron region was 0.203 count/s. Experiments with the described shielding configuration but with the cell out showed that the average background counting rate was (0.127 ± 0.356) count/s (the results in this work are presented in the form $\bar{x} \pm \sigma$, where σ is the standard deviation from the mean value). This value was obtained from the spectra collected during 1 month. When the complete cell with the electrodes and the electrolyte was put in the shield, the value of the average background counting rate did not show a significant difference from this value and varied from 0.125 to 0.128 count/s (calculated for 24-h runs). The test with the cell in but out of operation lasted 2 weeks.

Off-line analysis of all these spectra did not show any burstlike events. All pulse-height and MCS spectra looked like those in Figs. 1 and 2, respectively. In some periods during these tests, high-power pulsing systems (2.5 and 20 MHz) located near the detection system were switched on and off. We did not observe any effect in the pulse height and the MCS spectra.

Figure 1 shows the typical background pulse-height spectrum collected in a 82 920-s measurement during the cell in operation as well as the spectrum obtained when the detector was exposed to the weak ^{252}Cf source for 4300 s (note different scales: background left and ^{252}Cf right). The peak in the ^{252}Cf spectrum corresponds to spontaneous fission neutrons. Two structures observed in the background spectrum are due to environ-

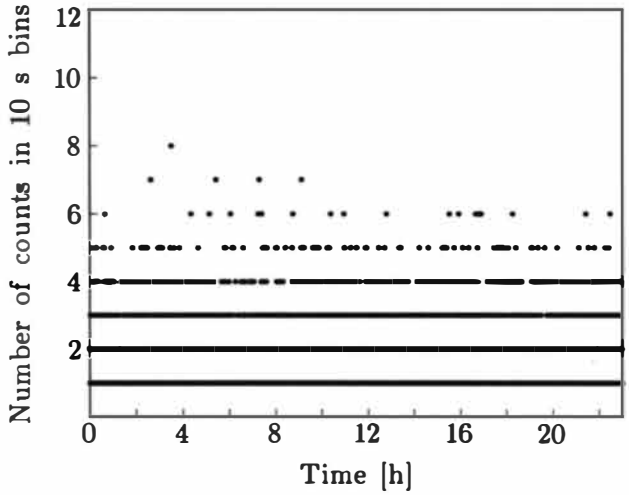

Fig. 2. MCS spectrum of the counts from neutron region of Fig. 1.

mental gamma rays and neutrons. The counts from the neutron region collected during the same time interval and sorted into 10-s bins are shown in Fig. 2. The average background counting rate in the neutron region was (0.126 ± 0.355) count/s with the standard error 0.001. This value was determined from spectra collected during 529 950 s and was taken as the background (BGD) counting rate for this experiment.

Environmental neutrons originate from various sources.[16] At sea level laboratories, neutrons are produced mostly by cosmic rays, predominantly by a (μ, n)

reaction in environmental materials and in heavy shields (lead in our case). The neutron production rate shows a strong mass dependence of $A^{1.82}$. The other sources of neutrons are (α, n) reactions on light elements induced by alpha particles emitted from uranium and thorium contamination in environmental material. Some neutrons can originate also in spontaneous fission processes, mainly in ^{238}U.

The authors of Ref. 17 measured the flux of fast and thermal neutrons with a germanium gamma-ray detector and found for a 20-cm lead shield the fast neutron flux of 1.5×10^{-2} cm$^{-2}\cdot$s^{-1} and the thermal neutron flux of 2.8×10^{-3} cm$^{-2}\cdot$s^{-1} at the Boris Kidrič Institute, Vinča. Based on these measurements, we made the crude estimate for our detection system (taking into account only fast neutrons) and found 0.135 ns^{-1}. This estimate is in good agreement with the result of our measurement (0.126 ns^{-1}) if we take into account the different shield construction and the different place of measurement.

The apparatus was very stable for the whole interval as it was in similar experiments 3 yr before[13,14] and during the test measurements throughout the several months before this experiment.

The same shape of the spectra was observed in all measurements except one (run 10, collected for 84 710 s). In this case, the MCS spectrum shows that two burstlike events, with durations of ~200 and 100 s, occurred in the 167th hour after the start of the electrolysis (Fig. 3). During these burstlike events, 193 (burst 1) and 63 (burst 2) counts were recorded. So, the total number of recorded counts during 300 s was 256. Observing the 500-s interval (±250 s around the burst 1 maximum), the total number of counts was found to be 290.

To check if the counts from the bursts were neutrons, the counts from the interval ±250 s around the first burst maximum of the MCS spectrum were sorted against their pulse heights. Figure 4 shows that a large part of the counts falls into the neutron region of the spectrum. It should be stressed that with the ^{6}Li detector, one cannot determine the energy that a neutron had before the thermalization process. However, the position of the peak shown in Fig. 4 is the measure of the energy produced in the $^6\mathrm{Li}(n_{th},\alpha)t$ reaction. In this way, the peak in Fig. 4 as well as in Fig. 1 means that the detector detected neutrons and not some spurious events caused by electronics, or electric or thermal noise.

A statistical analysis was performed on the BGD data grouped into 100- and 200-s bins. The Pearson χ^2 test for the goodness of fit performed on 11 separate runs and on all BGD data taken as one measurement showed that the data agree with the Poisson frequency distribution. The same result was obtained for the data collected during the first 80 000 s of run 10. The tail of the experimental and the theoretical frequency distributions of the BGD data grouped into 100-s bins ($\bar{x}$ = 12.6 count/100 s) with designated upper significance limits for significance levels $\alpha = 0.05$ and $\alpha = 0.01$ is presented in Fig. 5.

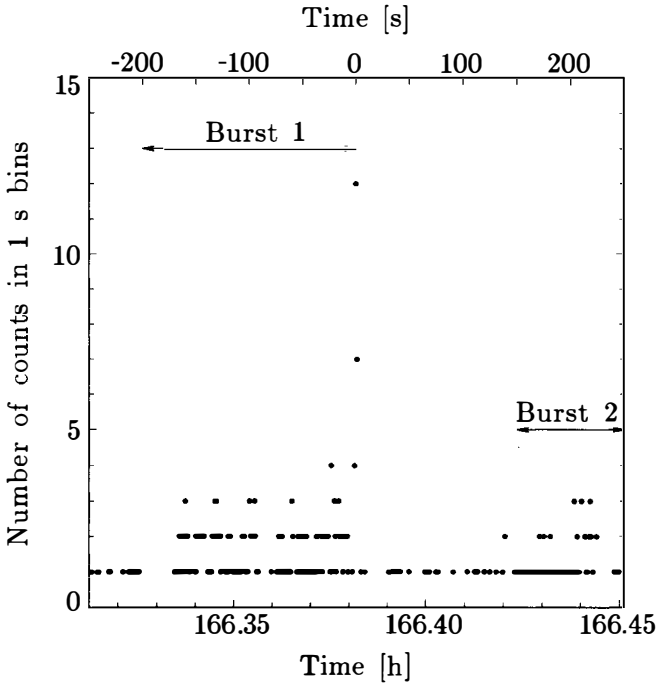

Fig. 3. MCS spectrum of the counts from neutron region from the interval ±250 s around the burst 1 maximum.

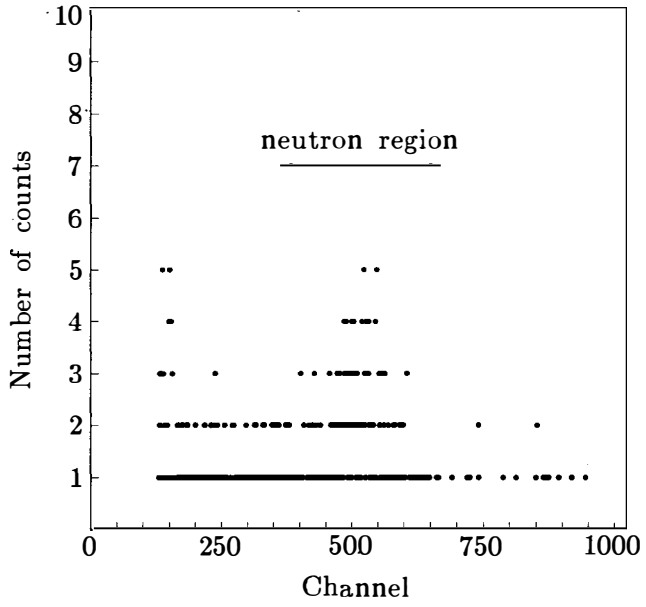

Fig. 4. Pulse-height spectrum of the counts from the interval ±250 s around the burst 1 maximum.

Estimates based on the maximal number of BGD counts observed in 100- and 200-s bins (28 and 43,

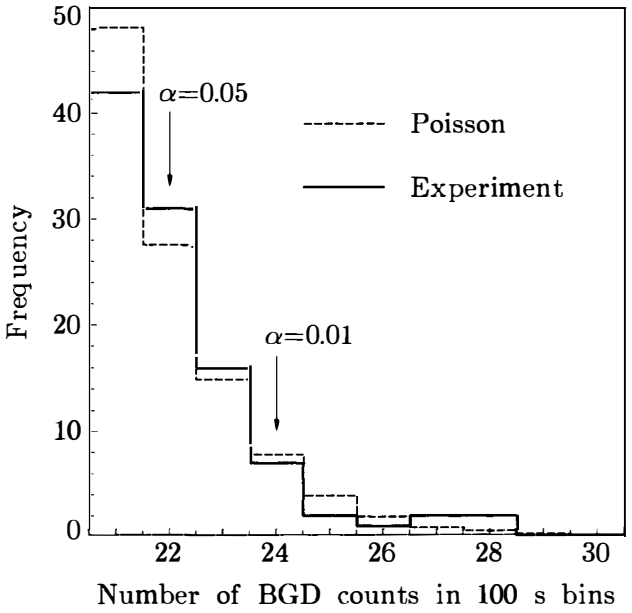

Fig. 5. Tail of the frequency distribution of BGD counts (grouped in 100-s bins) compared with the Poisson distribution ($\bar{x}$ = 12.6 count/100 s).

respectively) showed (a) the number of counts observed in burst 1 (193) is much larger than the upper significance limit of the BGD number of counts for α = 0.05 and even for $\alpha = 0.01$ (74 and 85, respectively) and (b) the number of counts observed in burst 2 (63) is slightly larger than the upper significance limit of the BGD number of counts for $\alpha = 0.05$.

After the background subtraction, the number of neutrons, emitted in the 4π solid angle during the burstlike events and the neutron intensities, were estimated. The results of the analysis are shown in Table I. The observed neutron intensities are an order of magnitude lower than those obtained by Gozzi et al.[5]

The analysis of the data concerning the palladium cathode temperature and the palladium electrode potential compared with the reference one showed that there was no change of these parameters during the experiment. After the electrolysis, the palladium pellet was heated, and 602 cm^3 of deuterium was released. This value corresponds to the ratio $N_D/N_{Pd} = 0.7$.

The electrochemical cell operating in steady-state external experimental conditions does not mean that in the volume or at the surface of the palladium pellet, the conditions were steady, at least locally.

X-ray fluorescence spectrometry carried out on the cathode surface showed traces of zinc, mercury, and some quantity of copper besides iron and copper already present in the starting material. A thin platinum layer was detected also on the palladium cathode surface as a result of platinum codeposition during long-term electrolysis.[13,14]

During the period of measurement, the sun flash activity showed some flashes of X rays, which could be accompanied with gamma-ray and particle emissions (particle fluxes are the most intensive at the Earth poles). The most intensive flashes in chronological order (Greenwich mean time) are listed as follows.[18]

June 8	09 h 00 min to 11 h 00 min (intensive)
June 9	16 h 00 min to 18 h 00 min (intensive)
July 3	09 h 00 min to 10 h 00 min (low)
July 5	19 h 58 min to 20 h 10 min, 22 h 10 min to 22 h 20 min, 23 h 39 min to 23 h 50 min (low)
July 6	01 h 47 min to 02 h 00 min (low)
July 7	11 h 20 min to 12 h 00 min (low)
July 8	09 h 45 min to 10 h 00 min (very intensive)
July 9	16 h 35 min to 17 h 00 min (low)

We must say that the most important, so-called "impulsive" phase usually started at the beginning of the flash.

As could be seen from the analysis of the appearance time of the neutron bursts in this experiment, the first recorded burstlike event started on July 7 (166 h 20 min after the beginning of the electrolysis) at 8 h 35 min in Greenwich time scale and reached the maximum 200 s later (8 h 38.5 min). The second one started 134 s later (8 h 40.5 min) and lasted 100 s. Evidently, it is not possible to find any correlation between the results of our measurements and the listed sun flash activity.

TABLE I

Results of the Analysis

	Number of Counts in Neutron Region		Neutrons in 4π Solid Angle ($\times 10^4$)	Neutron Intensity (ns^{-1})
	With BGD	Without BGD		
Burst 1 (200 s)	193 ± 14	168 ± 15	2.55 ± 0.34	128 ± 17
Burst 2 (100 s)	63 ± 8	50 ± 9	0.76 ± 0.16	76 ± 16
Total (300 s)	256 ± 16	218 ± 19	3.31 ± 0.44	110 ± 15
Total (500 s)	290 ± 17	227 ± 23	3.45 ± 0.49	69 ± 10

CONCLUSIONS

From the results of this experiment the following may be concluded:

1. Two neutron bursts were observed.

2. These bursts occurred during the electrolysis of heavy water on a palladium electrode in conditions that were very similar to the conditions in the work of Gozzi et al.[5]

3. The neutron intensities obtained in the current experiment were an order of magnitude lower than those obtained by Gozzi et al., but the durations of the bursts were similar.

4. New positive results in multicell experiments published in the meantime by Gozzi et al.[6] support the results of this experiment.

In light of the worldwide experimental and theoretical efforts in this field of research, it seems possible that this phenomenon is influenced not only with one mechanism but with a few of them. At the moment, it could be said that in some circumstances, the phenomenon is dominated by the mechanism(s) that causes the burstlike events and that in some other circumstances, dominate mechanism(s) that cause random events. Sometimes the phenomenon is influenced by both kinds of mechanisms. Why is this so? At the moment, nobody can answer this question, just as nobody can say anything with certainty about the mechanisms in question.

There is no doubt that some relevant but up-to-now unknown parameters were not under control. In this context, it is very important to repeat the positive-result experiments done at given laboratories by others.

ACKNOWLEDGMENT

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