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Search for neutrons from deuterium-deuterium nuclear reactions in electrochemically charged palladium

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We report on a search for neutrons from *dd* fusion in Pd rods loaded electrochemically with deuterium. In a four-week measurement we observe <0.007 n/(sec g) Pd, compared to 2.6×10^{3} n/(sec g) Pd, claimed recently by Fleischmann, Pons, and Hawkins. Our result implies $<2.2\times10^{-24}$ (*ddn*) fusions/[(*dd* pair) sec], as compared to $\sim10^{-23}$ (*ddn*) fusions/[(*dd* pair) sec] observed by Jones *et al.* in a Ti electrode.

The recent reports by Fleischman, Pons, and Hawkins[1] (FPH) and Jones *et al.*[2] of the observation of neutrons from palladium (titanium in Jones' case) electrochemically loaded with deuterium has stimulated intense interest in the verification of this process, and in the entire possibility of "cold nuclear fusion." In this paper we report our search for neutrons from the electrochemically loaded Pd–D system at room temperature.[3] We set an upper limit on the deuteron-deuteron fusion rate of 2.2×10^{-24} (*ddn*) fusions/[(*dd* pair) sec], with *ddn* indicating the neutron producing branch of *dd* fusion. This is *5 orders of magnitude below* that reported in FPH and approximately a factor of 5 below that reported by Jones. One unique aspect of our experiment, is the use of the high-energy portion of the gamma-ray spectrum from thermal neutrons captured by Na and I in a NaI(Tl)—spectrometer system. Such a method renders the detection system relatively insensitive to ambient natural radioactivity and results in enhanced sensitivity for thermal neutrons. We also report a quantitative nuclear reaction analysis of the deuterium loaded Pd rod to estimate the amount of absorbed deuterium.

We investigated the possible neutron emission from three Pd rods: two wires (0.125 cm diameter by 10 cm long) were mounted separately and parallel to each other in one electrolysis cell. A second cell contained a 0.4 cm diam×10 cm rod. Both cells were made from borosilicate glass (Pyrex, ≈0.2 cm wall thickness) and sealed with Teflon or a rubber stopper to prevent D_2O exchange with atmospheric light water. The electrolytically generated O_2 and D_2 were released through a single bubbler trap, filled with silicone oil. The Pd cathode emerged from the electrolytic solution and then through the seal on the top for electrical connection. In the case of the cell with the two wires, the wires were strapped to a glass rod to prevent electrical shorting caused by shape distortion due to the deuterium loading. The submerged length was 6.9 cm for the 0.4 cm rod and 9 cm for the two smaller wires. The anode was a Pt wire (0.076 cm diameter), coiled onto a 2 cm glass tube cage. This cage was positioned coaxially with the Pd cathode and one of the glass support tubes was fed through (with the Pt wire inside) the top seal for electrical connection.

The temperature in the cells was monitored with conventional mercury thermometers. Since the purpose of this experiment was the detection of neutrons and not a quantitative measurement of any possible "excess" heat effects associated with cold fusion, these thermometers only served to indicate any gross thermal excursions. The electrolytic solution in both cells was nominally the same as that used by FPH: 0.1 molar LiOD in D_2O (99.8 at.% D) with the LiOD obtained by adding Li_2O ($^6Li/^7Li$ ratio unknown) to the heavy water. Both cells were operated in a constant—current mode (separate power supplies) with current densities of 63 and 70 mA/cm^2 for the 0.4 cm rod and the 0.125 cm wires, respectively. Corresponding applied cell potentials varied between 5 and 6 V. The current density values were chosen as an intermediate value of those used by FPH, at which they claimed to have observed a significant neutron flux (4×10^{4} sec^{-1} for a 0.4×10 cm rod).

The Pd wires and the rod in this experiment were prepared in different ways. The two wires were cut from an available wire spool (99.5% Pd), which had been drawn to its final diameter from an ingot. One of the wires was subsequently annealed at 900 °C for ≈1 h in flowing N_2. The rod was cast from Pd powder (99.95% Pd), which was melted under Ar in a radio frequency heated BN crucible and subsequently cold rolled to its final diameter. The rod was then annealed as described for the wires. Finally, both wires and the rod were cleaned with a light mechanical polish (sand paper). After approximately one day's exposure to room atmosphere, they were inserted in the D_2O/LiOD cell electrolyte.

In this experiment the neutron detector was a single NaI(Tl) scintillation crystal (12.5 cm diam×12.5 cm length), completely surrounded with polyethylene moderator, as shown in Fig. 1. The two cells were placed inside this moderator "house" with 5 cm polyethylene between the detector and the cells. Lead blocks were positioned outside this "poly house" to reduce undesired γ background and ≈5 cm thick borax containing boxes were placed outside the lead shield to absorb any external neutrons. Any neutrons resulting from *dd* fusion reactions inside the Pd rods are moderated to thermal energies in the polyethylene blocks. The thermal neutrons then either yield a 2.225 MeV γ ray from the ($np\gamma$) capture reaction in the polyethylene or a nearly continuous spectrum of high-energy γ's in the range 3.5–7 MeV from thermal

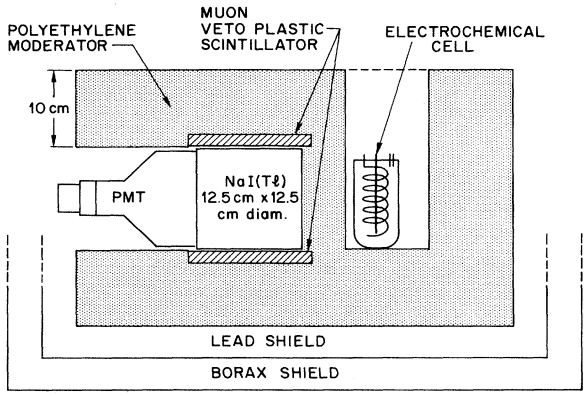

FIG. 1. NaI(Tl) scintillator detector system for measuring electrochemically induced $dd \rightarrow n$ fusion reactions.

neutron capture by ^{23}Na and ^{127}I within the detector.

The advantage of detecting in this high-energy region, rather than directly at the 2.225 MeV line, is the significantly reduced background, which arises mainly from natural radioactivity. This can be seen clearly in Fig. 2(b), which shows a typical pulse height spectrum from the NaI detector. Three lines, at 2.120, 2.204, and 2.447 MeV, appear in this 2.225 MeV region and are associated with the ^{238}U decay chain. In addition, background lines at 2.614 MeV (^{232}Th chain), at 1.764 and 1.847 MeV (^{238}U chain), and the ^{40}K line at 1.460 MeV were observed. The highest energy γ ray from ambient natural radioactivity is 3.35 MeV. Thus at higher energies (> 3.5 MeV) the spectrum is free from these radiation sources except for very weak high-energy fission γ rays and a very low level of α rays from U and Th decay in the crystal itself. Thus, in contrast to the use of 2.2 MeV γ's as an indication of the $(np\gamma)$ reaction, our method practically eliminates the possibility of mistaking a background γ ray for a neutron signal.[4]

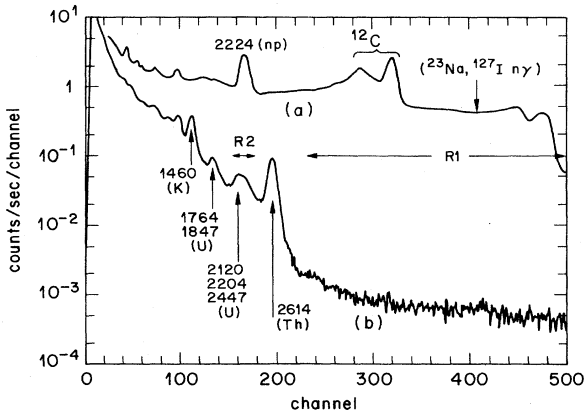

FIG. 2. (a) Pulse height spectrum of a Am–Be calibration source in place of the electrochemical cell measured with a NaI(Tl) detector; (b) typical pulse height spectrum (signal+background) measured during electrochemical deuteration of Pd rods.

The residual background above 3.5 MeV is nearly all due to cosmic ray muons passing through the crystal. In order to reduce this background, two plastic scintillators were installed above and below the NaI detector. Events detected in the NaI crystal were gated in an anticoincidence mode by pulses from the two plastic scintillators. The gated output for a fixed lifetime, typically 1800 sec, was analyzed and stored in a multichannel analyzer. The spectral data (0–7 MeV) was periodically transferred onto a hard disk of a personal computer. From this experimental data base the temporal behavior of the γ-ray intensities in any region can be accessed and examined; regions of particular interest are those around 2.2 MeV (region $R2$) and the n-signal range 3.5–7 MeV (region $R1$).

The neutron detection efficiency was measured with a Am–Be source with a nominal neutron flux of 6×10^4 sec^{-1}. After placing this source in an identical $LiOD/D_2O$ cell in the same position as the electrolysis cells, the calibration spectrum was obtained [Fig. 2(a)]. This shows clearly the 2.224 MeV line due to the $(np\gamma)$ capture, as well as the broadband signal from the reaction of thermal neutrons with ^{23}Na and ^{127}I. The line at 4.4 MeV (and part of the lower energy single escape line peak at 3.9 MeV) are due to $(\alpha,n\gamma)$ reactions in the source and from the reaction $^{12}C(nn'\gamma)$ in the polyethylene by neutrons of energy > 4.4 MeV. This feature is not expected from 2.5 MeV fusions neutrons. A nominal neutron detection efficiency of 0.015 was obtained from the counting rate in $R1$, with the source in place, divided by the neutron flux from the Am–Be source. It must be noted, however, that a substantial fraction of the neutron flux from this source ranges in energy up to 8 MeV, significantly higher than 2.45 MeV, the energy of dd fusion neutrons. The higher energy neutrons are moderated less efficiently and thus contribute less to the signal in $R2$. The value of 0.015 employed in this work should therefore be considered a lower limit to the neutron detection efficiency actually relevant to dd fusion detection.

The 0.4 cm rod was charged for 33 days during which the neutron detection system was operative. At the end of this period the current was discontinued. The sample was allowed to outgas for 24 h in an open, silica container which was flushed with dry nitrogen gas at atmospheric pressure. After this period the 0.4 cm rod was removed and stored in liquid nitrogen. Since the diffusion coefficient of D in Pd at 77 K is $\approx 10^{-17}$ cm^{2}sec^{-1} vs $\sim 3\times10^{-7}$ cm^{2}sec^{-1} at 300 K in the β phase of Pd–D (using the known activation energy[5–9]), storing at 77 K assumes retention of the remaining deuterium. It was noticed upon removal of this rod from the electrolysis cell that the submerged section had clearly expanded and that the rod thus had a "wine bottle shape." The submerged part was ≈ 0.45 cm in diameter, while the unsubmerged part retained the original 0.40 cm diameter. The rod was removed from the liquid N_2 for about 2 h for dicing into disks for subsequent analysis. This process consisted of using a diamond saw to cut three, 0.15 cm disks from both ends of the rod. The different material properties of the deuterated and undeuterated portions were already noticed at this stage, as the deuterated portion cut much more slowly and unevenly. This clearly reflects the brit-

tling effect of the deuterium on the Pd.

The deuterium loading was examined via nuclear reaction analysis using the $d(^3He,p)\alpha$ reaction, a standard ion beam analysis technique.[10] The resulting spectrum is shown in Fig. 3 and indicates a strong proton and alpha particle signal, indicative of a large deuterium concentration. Since the cross section for the analysis reaction is about 10^{-3} of the ^3He elastic (Rutherford) scattering reaction, it is immediately obvious that the deuterium concentration is comparable (within an order of magnitude) to the Pd concentration. It should be noted that the measurement was carried out at the *center* of the disk indicating that the deuterium had diffused through the entire cross section of the rod during the electrochemical loading time. This diffusion (charging) time to reach the rod center is $\Delta t \simeq R^2/D$, with R the radius of the rod and D the diffusion coefficient of deuterium in Pd. For $R = 0.2$ cm and $D = 3\times10^{-7}$ cm^{2}sec^{-1} (at room temperature), this time is 1.5 days. Analysis of the portion of the rod which was not submerged gives no indication of a deuterium signal. The absence of deuterium 4 cm above the loaded area is consistent with a diffusion length of only 0.85 cm for the sample at room temperature for four weeks. The analysis of the nuclear reaction data is complex since the experiment probes through the penetration depth of the ^3He (about 2 μm) with a reaction cross section strongly varying with ^3He energy. Furthermore, the first few microns have lost deuterium due to out diffusion into the atmosphere during the two hours of exposure for cutting and analysis (corresponding diffusion length $\sim$460 μm). Nevertheless the extracted D/Pd ratio is 0.38 which is consistent with typical maximum loading values[5,9,11] and out diffusion during the cutting period. Despite some uncertainties in this analysis, substantial deuterium loading at the center of the 0.4 cm Pd rod is clearly demonstrated.

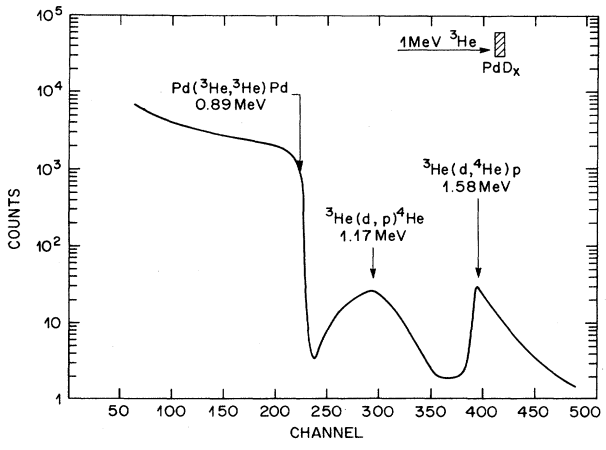

FIG. 3. Spectrum used in the ^3He analysis of the deuterium content of a loaded Pd rod. The spectrum indicates Rutherford backscattering from the Pd and proton and alpha particle signals from ^3He-d interactions. The energy associated with the outgoing proton (1.17 MeV) is determined by the energy loss in the solid-state detector with a depletion layer smaller than the proton range in silicon.

As mentioned previously, during electrolysis the γ-ray flux both at the 2.2 MeV region (region $R2$) and in the region between 3.5 and 7 MeV (region $R1$) was measured. Figure 4(a) shows the count rate with both cells inside the poly "pit" up to 14 days after the start of the experiment. At day 16 the current through the cell with the two wires (cell 2) was discontinued, the cell removed, and the shielding temporarily disturbed in order to facilitate the repositioning of the cell with the 0.4 cm rod in front of the detector. As can be seen in Fig. 4 this resulted in a small but clearly observable spike in the counting rate in the 2.2 MeV region due to enhanced exposure to ambient room background, clearly illustrating the danger of relying on this spectral region to determine any low level γ's from $(np\gamma)$ reactions. On the other hand, no such spike is seen in the n-signal region $R1$.

Counting rates in region $R1$ between days 10 and 33 after the start of the experiment ($t = 0$) are shown in Fig. 4(b). The counting rate is almost an order of magnitude lower than in Fig. 4(a). From day 16, only cell 1 (with the single rod) continued to be monitored up to day 30.

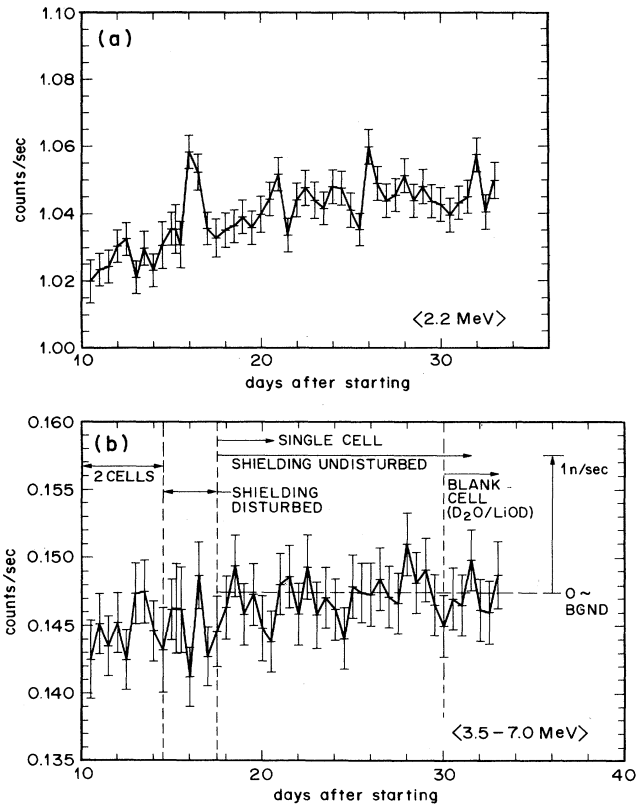

FIG. 4. (a) Counting rate in the 2.2 MeV region showing the vulnerability of this spectral region to natural radioactivity background. The spike occurred by temporarily disturbing the shielding during days 16 and 17 after the start of the electrolysis process; (b) counting rate in the 3.5–7 MeV region during the electrochemical deuterium loading of 3 Pd rods. Data between days 17 and 33 are from a single cell with a 0.4 cm diameter Pd rod. Background data (with identical cell, but without the Pd rod) are shown between days 30 and 33.

During this period the observed signal plus background rate was 0.1472 ± 0.0004 sec^{-1}. The background was determined with a dummy cell, identical (with D_2O/LiOD) except for the presence of a Pd rod, between days 30 and 33 with a measured rate of 0.1472 ± 0.0008 sec^{-1}.

This implies a maximum signal rate of 0.0 ± 0.0012 sec^{-1}, which, together with the measured efficiency of 0.015, results in an upper limit of 0.08 *n*/sec for this particular sample. If a D/Pd ratio of one is assumed, then this neutron flux limit corresponds to a (neutron) fusion rate per deuterium pair of $< 2.2 \times 10^{-24}$ (*ddn*) fusions/[(*dd* pair)sec]. In expressing this ratio in terms of a fusion rate per *dd* pair, it is implied that the effect (real to the extent of the upper limit) is a volume effect.

In this paper we have reported on the search for neutrons from any possible deuterium-deuterium fusion reactions inside electrochemically deuterated palladium rods. Using a NaI(Tl) scintillation detector we have measured < 0.08 neutrons/sec for a 0.4×7 cm cast, cold rolled, and annealed Pd rod. This corresponds to < 0.007 *n*/(sec/g) Pd, compared to FPH's claimed[1] 2.6×10^3 *n*/(sec/g) Pd. Our upper limit corresponds to $< 2.2 \times 10^{-24}$ (*ddn*) fusions/[(*dd* pair)sec], which is approximately five times lower than that claimed by Jones.[2]

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