Electrochemical Experiments in Cold Nuclear Fusion

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Recently, two scientific papers have reported positive detection of nuclear radiation from similar electrochemical cells operating with deuterated water. We have experimented with similar electrolytic cells and have looked for energetic charged particles which are characteristic of nuclear fusion reactions. We report on six variations of the cell, with an upper limit of 0.005 detected particle/cm^{3}sec. Within background statistics, we observe no nuclear fusion.

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Two recent papers[1,2] have revived interest in cold nuclear fusion (CNF) by reporting the detection of nuclear-reaction products from similar electrolytic cells.[3] Fleischmann and Pons[1] have reported the observation of excess γ rays, neutrons, and ^{3}H from a cell with an anode of Pt, a cathode of Pd, and an electrolyte of heavy water D_2O with LiOD. Their γ-ray spectrum has a pronounced peak at 2.2 MeV, which they suggest may be from a $n+p$ nuclear reaction. They have observed neutrons at about 2.5 MeV which corresponds to a neutron production rate of about 4000/cm^{3}sec (the term "cm^3" refers to the volume of Pd cathode used). Further, they report massive and sustained heat generation—of the order of 4 MJ/cm^3. The heat produced was observed to scale with the Pd cathode volume, so the CNF is presumed to occur throughout the cathode metal. If the heat they observe is due to unobserved nuclear fusions, they have a fusion rate of about 10^{12} fusions/cm^{3}sec.

Jones *et al.*[2] have reported the observation of neutrons during a similar electrolytic experiment, with an anode of Au, a cathode of Pd or Ti, and a complex electrolyte of various metallic salts (chosen to represent typical components of the Earth's crust) in heavy water D_2O. Their neutron spectrometer shows a peak at about 2.5 MeV, exactly the energy of the neutron produced by a $d+d$ nuclear reaction. In an independent work, Jones *et al.* have reported detecting 2.4-MeV neutrons at a rate of 0.7 neutron/cm^{3}sec from an electrolytic cell.

Both of the above experiments are difficult to reproduce in detail since the authors discussed few details of the experimental technique. Both groups have discussed an "incubation period" of charging the cathode, possibly to raise the deuterium concentration within the Pd cathode to the high levels found in β-Pd.

Our approach has been based on the fact that all known exothermic CNF reactions emit energetic charged particles. The above reports have looked at γ rays, neutrons, and excess ^{3}H—all of which have large backgrounds in most laboratories. However, energetic charged particles have a background only from naturally occurring α particles and cosmic rays. Both of these can be reduced to levels of less than five detected particles per day, a level about 10^{-6} less than that usually reported for the detection of γ rays or neutrons. Further, the detection efficiency of particle detectors, such as silicon surface-barrier (SSB) detectors, is about 100% for all energetic incident charged particles in contrast to efficiencies of less than 10% for most γ or neutron spectrometers.

The charged particles from $d+d$ fusion reactions range from 1 to 3 MeV. We have constructed electrolytic cells with Pd cathodes which directly cover SSB detectors, see Fig. 1. Since one $d+d$ nuclear reaction product is an energetic proton (3.02 MeV) which has a range in Pd of about 30 μm, Pd cathodes thinner than 25 μm will

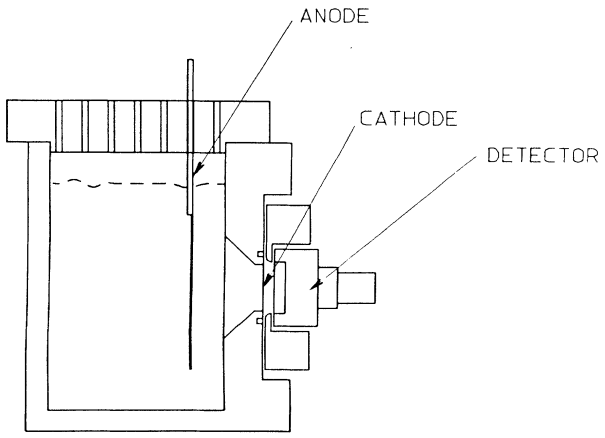

FIG. 1. Cross section of the Teflon electrolytic cell. The anode was always Pt, about 2 cm wide and 4 cm long. A 7-cm^2 conical hole in the side of the cell led to the 2-cm^2 cathode. The anode-cathode spacing was adjustable, but all tests reported here were at a spacing of 2 cm. The cathode was Pd foil of various thicknesses. About 2 mm behind the cathode was a surface-barrier detector to measure energetic charged particles. The detector had a background for charged particles with energies of 1–3 MeV of about 5 counts/d (mostly due to cosmic rays).

allow most of these particles emitted in a backward direction to be detected. Our cells have SSB detectors with a solid angle of about one π and a background of about 5 counts/d (for 1–3-MeV particles).

The electrolytic cells are shown in Fig. 1. Each cell is made from 8-cm-diam cylindrical Teflon with a hollow cylindrical well 5 cm in diameter by 8 cm deep. A 7-cm^2 conical hole in the inner surface of the cell leads to a 2-cm^2 Pd cathode. A surface-barrier detector faces against the back of the cathode.

The anode is made of Pt (99.9%), 2 cm wide by 4 cm long, with a thickness of 0.25 mm. Its distance from the cathode may be varied, but usually it is kept at a distance of 2 cm (as in Ref. 2). A thermocouple was attached to the top of the Pt anode to determine approximate electrolyte temperature without introducing possible contamination to the electrolyte.

The cathode is Pd (99.9%), with thicknesses ranging from 0.025 to 0.5 mm. To prevent leakage of hydrogen gas from the back of the cathode into the air, dense Au films were sputter deposited on the back side of the Pd (the side away from the electrolyte). The Au was made to adhere firmly to the Pd by evaporating an intermediate layer of Cr, 200 Å. These Au films ranged from 1.7 to 6.5 μm in thickness (the effectiveness of these films is discussed later). These films were thin enough to let charged particles pass with modest energy loss (protons at 2 MeV lose 90 keV/μm).

The electrolytic cell holds about 150 cm^3 of electrolyte. The top is closed except for seven 1-mm holes to release gas pressure and to allow adjustment of the anode position, see Fig. 1. The electrolyte consisted of 0.1M LiOD in various mixtures of H_2O (100%) and D_2O (99.5%). The lithium hydroxide was made from Li metal (90% ^{6}Li and 10% ^{7}Li). The pH of the electrolyte was measured as 12.4.

Various mixtures of H_2O and D_2O were tested as electrolytic solutions because conventional nuclear theory suggests that cold-fusion reactions of $d+p$ can have much greater cross sections than $d+d$ reactions. Since both Refs. 1 and 2 did not use pure D_2O, it was possible that they were seeing products of $d+p$ reactions instead of $d+d$ reactions they presumed. It was felt that a mixture of H_2O (10%) with D_2O (90%) would give comparable amounts of ^{1}H and ^{2}H within the Pd cathode metal on the basis of separation factors.[4] We used mostly ^{6}Li for lithium since the cross section for fusion of d with ^{6}Li is much higher than for ^{7}Li. However, the Li metal still contained 10% ^{7}Li.

The surface-barrier detectors were held at a reverse bias of 48–75 V to obtain about 100 μm of depletion. This is adequate for the detection of charged particles up to about 8 MeV. Particles with energies above this are still detected but some energy information is lost. The detectors were energy calibrated and tested using a standard ^{241}Am radioactive source.

The anode-cathode current was held at 150 mA/cm^2 for all of the reported studies. Reference 1 indicates that observed nuclear products scale with current density, thus we selected a current density near the high end of their range of values.

All particle detectors have backgrounds, either due to trace radioactive materials in their fabrication, or due to cosmic rays interacting with the silicon. Reference 3 reviews the background sources in silicon detectors and shows typical cosmic-ray background rates. The rate we observed was statistically within the rate of this study (see Fig. 2). We note that we define "background" as the spectrum observed with our experimental arrangement but with the cell current off. This assumes that there were no CNF particles generated while the cell was not in operation. Only one test was made of background both before and after operation of the cell, and no change was observed.

The "incubation" or "charging" period for our cathodes to form β-Pd, Pd hydride, will be shown to be less than 24 h. Longer charging periods might be necessary for Li in the electrolyte to penetrate into the Pd. We accelerated this process by artificially injecting Li into two of the Pd cathodes. In one case Li was introduced by capsule diffusion at 600 °C. A second cathode was ion implanted with Li; half the cathode was implanted with ^{6}Li (200 keV) to a dose of 10^{15} Li/cm^2, and the other half was implanted to 10^{16} Li/cm^2. Since we were looking for any kind of fusion, it was felt that having a mixed concentration of Li in the cathode would not

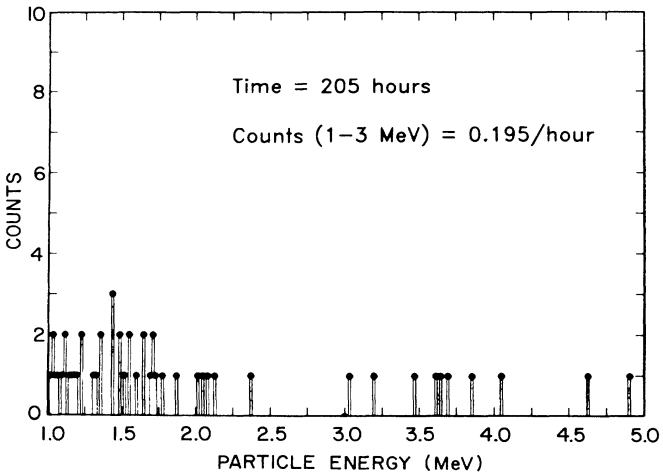

FIG. 2. Charged-particle spectrum for a thin Pd cathode (25 μm thick, backed with 1.76-μm Au) during 205 h of charging. Within statistics, all the counts are due to cosmic-ray neutrons interacting with the silicon detector; see Ref. 5. The background of the detector was determined by making no experimental changes except for turning off the cell current. This background, taken over 82 h, was virtually identical to the spectrum with the cell operating. The upper limit for the fusion rate for this experiment was 0.0038 fusion/cm^2sec.

TABLE I. Experimental results. Electrolytes: 1, 100% D_2O with 0.1M LiOD; 2, electrolyte based on components of Earth's mantle plus volcano lava (see Ref. 2); 3, 90% D_2O+10% H_2O with 0.1M LiOD; 4, 100% H_2O with 0.1M LiOD. Backings: Pd foils were backed with Au films on the side away from electrolyte to prevent out diffusion of hydrogen from the cathode (see text). The table indicates the thickness of the sputter-deposited dense Au films. Calculation of detection limit: In all cases the total signal counts (detected particles from 1 to 3 MeV) were within 3σ of the background counts (taken with the cell current off), assuming Poisson statistics. The quoted particle upper limit of detection is based on 2σ of the signal counts, without background removed, divided by the cathode volume.

Sample description	Electrolyte (D_2O/H_2O)	Duration of run (h)	Fusion rate (fusions/cm^3 sec)
Fleischmann and Pons, neutrons[a]	1	50	4000
Fleischmann and Pons, observed heat[a]	1		10^{12}
Jones *et al.*, neutrons[b]	2	8	0.7
			Particles detected (number/cm^3 sec)
25-μm Pd+1.7-μm Au Prebaked: 900 °C, 1 h, 10^{-6} Torr Pure α-Pd after testing	1 (100/0)	205	< 0.0038
25-μm Pd+6.5-μm Au Pure β-Pd after 24 h	1 (100/0)	250	< 0.0052
0.5-mm Pd+6.3-μm Au Pure β-Pd after 24 h	3 (90/10)	24	< 0.00092
25-μm Pd+6.5-μm Au	3 (90/10)	250	< 0.0051
25-μm Pd+6.5-μm Au Pure H_2O electrolyte	4 (0/100)	250	< 0.0054
25-μm Pd+6.3-μm Au Diffused with ^{6}Li	3 (90/10)	127	< 0.0066
25-μm Pd+1.7-μm Au Ion implanted with ^{6}Li	3 (90/10)	73	< 0.0051

[a]Reference 1. [b]Reference 2.

compromise the experimental results.

Next to the cells were standard biological neutron monitors, and γ detectors. These were used for personnel safety, and had thresholds of 0.1 mrem/h.

As an independent evaluation of the charging of the Pd cathodes, the crystal structure of the Pd foils was determined by x-ray diffraction, using Zr-filtered Mo $K\alpha$ radiation and a scintillation detector, with a Θ-2Θ diffractometer and symmetrical reflection geometry. Measurements were made for scattering angles 2Θ from 90° to 155°.

All the experimental results are tabulated in Table I. At no time did the neutron or γ detectors near the cells register radiation.[6]

X-ray diffraction measurements were made on several of the Pd cathodes to evaluate the electrolysis time necessary to form β-Pd (above 60% hydrogen) from the normal α-Pd, and to determine the adequacy of the Au hydrogen-sealing layers on the cathode backs. A cathode with a 1.7-μm Au layer gave the diffraction pattern of α-Pd after 200 h of electrolysis. The lattice parameter was 3.92 Å, somewhat larger than the published value for pure Pd (α-Pd), 3.89 Å.

Both thin (25-μm) and thick (0.5-mm) Pd cathodes which were backed with about 6.3-μm Au films, after electrolysis for 24 h, gave a pure β-Pd pattern with a lattice parameter of 4.03 Å, only slightly greater than the published value for Pd monohydride, 4.02 Å, see Fig. 3. There was no evidence for any α-Pd in the diffraction spectrum. This result indicates that 6.3 μm of dense Au provided an adequate hydrogen seal to the back surface of the Pd cathode.

It is not clear whether the formation of β-Pd is essential for the reported CNF, thus we report results from both types of samples.

One sample was preannealed to 900 °C in a vacuum of 10^{-6} Torr for 1 h. This anneal was intended to drive out any preabsorbed hydrogen. The sample showed no unusual results.

Our experiment measured CNF particles with better sensitivity than that of Refs. 1 and 2. Our cell configuration was not identical to those of Ref. 1 or 2 since

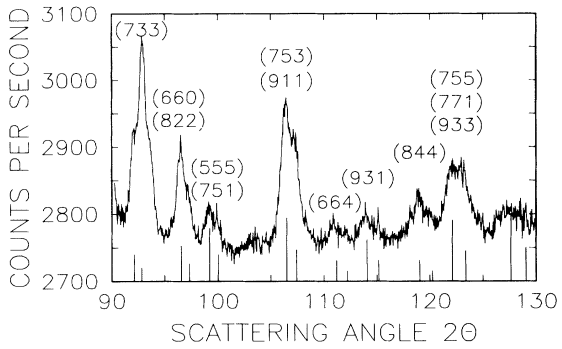

FIG. 3. X-ray diffraction spectrum from the 0.5-mm-thick Pd cathode, backed by 6.3-μm Au. The peaks correspond to a lattice parameter of 4.03 Å, only slightly larger than the published value for Pd monohydride, 4.02 Å. There is no evidence of the original α-Pd metal in the spectrum. This leads us to conclude that complete hydrogenation of the cathode was obtained within the limits of the applied voltage and current density.

our cathode formed a thin window to allow the exiting of energetic charged particles. Our results show that no excess heat and no nuclear fusion products were detected for any of the samples tested. The partial substitution of H_2O for D_2O did not produce nuclear fusion products. Preannealing of the Pd cathode had no effect. The artificial introduction of ^{6}Li into the Pd cathode by either diffusion or iron implantation had no effect.

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[6]Reference 1 states their detected neutrons were 3 times background. Fleischmann has said their neutron signal after 50 h contained 135 counts above background (private communication).