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Search for cold fusion using x-ray detection

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A search for x rays excited by charged-particle fusion products in a Pt-Pd electrolytic cell with Li-D_2O electrolyte is reported. Electrolytic loading of the palladium cathode was found to be 0.8 deuterium atoms per Pd atom. No x rays above background were detected and the fusion rate was inferred to be less than 1.6×10^{-20} fusions/(sec deuteron).

Reports of "cold" deuteron fusion have been based on the detection of excess heat from electrolysis cells[1] or neutrons from electrolytic cells and solids loaded with deuterium gas.[2] The paper by Fleischmann and Pons[1] and earlier news releases to the press aroused considerable interest in the scientific community and the public. A large power gain was reported from an electrochemical cell consisting of a Pd cathode and a Pt anode operating in a Li+D_2O solution. The paper by Jones *et al.*[2] reported low-level emission of neutrons from an electrochemical cell with similar electrodes but using a more complicated solution of salts dissolved in deuterated water. Since confirmation of the observation of excess heat production will not, in itself, address the nuclear fusion issue suggested by Fleischmann and Pons, we have attempted to find a signature of nuclear events in an electrochemical cell. Reported neutron production levels[1] are far below those expected if deuteron fusion were producing the reported heat excess. Alternative mechanisms for energy production have been considered from among the nuclear reactions normally regarded as responsible for fusion:

$$d+d\rightarrow t(1.008)+p(3.025)\,, \quad (1)$$

$$d+d\rightarrow {}^3\mathrm{He}(0.817)+n(2.452)\,, \quad (2)$$

$$d+t\rightarrow {}^4\mathrm{He}(3.514)+n(14.056)\,, \quad (3)$$

$$d+d\rightarrow {}^4\mathrm{He}(0.076)+\gamma(23.772)\,, \quad (4)$$

where the numbers in parentheses are the laboratory energies of the reaction products in MeV. Reactions (1) and (2) are the usual deuteron fusion reactions observed in which the resulting energy appears as the kinetic energy of the fragments. Reaction (3) has been considered by some authors since tritium occurs in deuterated water. Reaction (4) has been considered as one in which energy results but neutrons are not produced. All the reactions listed except reaction (4) are commonly observed in the laboratory and their cross sections have been measured to energies as low as about 10 keV in the center-of-mass system.[3] A number of searches for heat excess and nuclear reaction products have been unsuccessful[4,5] and have set severe limits on the probability of "cold fusion." Several authors have criticized the experimental techniques of Fleischmann and Pons.[5]

This Rapid Communication reports a search for evidence of charged particle production in an electrochemical cell by detecting characteristic x rays of Pd which result from K shell vacancies produced by fast protons in a Pd cathode. The measurement is motivated by the possibility that at room temperature, reaction (1) could have a much larger cross section than reaction (2), even though at 10 keV the cross sections for the two reactions are about equal.[3]

An electrochemical cell was constructed for our experiment as shown in Fig. 1. The cell was made of Pyrex glass, provided with a thin blown Pyrex window and then

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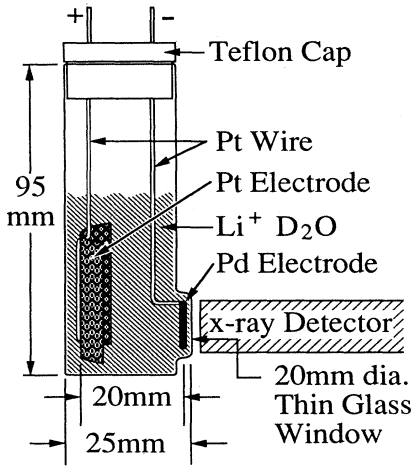

FIG. 1. Construction diagram of the electrochemical cell used in this experiment.

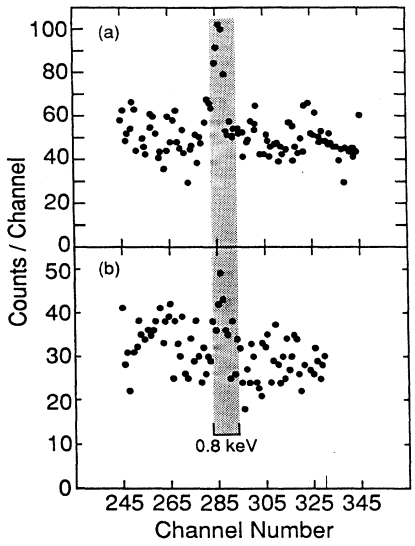

Fig. 2. (a) Portion of the x-ray spectrum produced by fluorescing the Pd electrode with Ba x rays. (b) X-ray spectrum in the vicinity of the Pd *Kα* energy after 333 h of operation of the electrochemical cell.

cleaned with distilled water, rinsed with isopropyl alcohol, and dried in vacuum. The electrolyte used was a D_2O solution, prepared by cutting a piece of lithium metal to the correct volume so that it would form a $0.1M$ solution when added to the D_2O. The lithium had been stored in oil and was cleaned in toluene and dried under nitrogen before dissolving. After the requisite amount was dissolved and the stock solution was made, ≈ 25 ml of the electrolyte was put in the cell and a current of ≈ 60 mA was maintained. The current density was ≈ 300 mA/cm^2. The Pd foil cathode was 5×10^{-3} cm thick and ≈ 1 cm^2 of foil area was viewed by the x-ray detector through the thin window.[6] All operations were carried out at room temperature, and the cell temperature and current were monitored. However, calorimetry on the cell was not attempted.

The *K*-shell ionization cross section of Pd is 1.9×10^{-24} cm^2 for fast protons (averaged from 3 to 1.75 MeV).[7] The cross section for *K*-shell vacancy production becomes very small below 1.75 MeV. The 21 keV palladium *K* x rays can pass through the thin-cell window and are easily detected in a small silicon x-ray detector.[8] The ability of the detection system to register *K* x rays from the Pd cathode was checked by fluorescing the electrode using Ba *K* x rays after the cell had been filled with electrolyte. The x-ray spectrum was continuously accumulated once the current was started. Room background radiation fluoresces the cathode and Pd *K* x rays are therefore present as an artifact of background. An energy spectrum of x rays near 21 keV is shown in Fig. 2(a). This spectrum was produced by fluorescing the Pd cathode with Ba *K* x rays, which appear in channels above 488 and are therefore not seen in Fig. 2(a). The background counting rate is quite time dependent, probably because of its cosmic-ray origin, but the shape of the background spectrum in the region of the Pd x rays is continuous and without structure other than the Pd *K* x rays. The background spectrum could therefore be normalized to, and subtracted from, the spectrum obtained with the cell in operation. Background was determined by counting with the cell in place for 113 h before applying current. The x-ray detector was placed as close as possible to the cathode. Counts within a window of 0.8 keV which included the Pd *Kα* lines were recorded as the cell was operated in the manner described above. The normalized background rate in the energy window was 1.186 ± 0.075 counts per hour. The x-ray spectrum accumulated during the time the cell was in operation (333 h) is shown in Fig. 2(b). The observed counting rate above background was 0.006 ± 0.093 counts per hour. Therefore there is no evidence of fusion induced x rays in this experiment. In order to calculate the maximum fusion rate consistent with these data, we have considered the number of *K*-shell ionizations expected from 3 MeV protons. The fraction of the energy release carried by the triton in the fusion reaction is one-third that of the proton—the *K*-shell ionization probabilities for 1 MeV tritons and the even lower energy ^{3}He ions from reaction (2) are negligible by comparison. We have included an estimate of the fraction of emitted protons stopped in the foil, the fluorescence yield of Pd *K*-shell vacancies, the transmission of 21 keV x rays through the glass window, the solid angle, and efficiency of the detector. The overall efficiency of the detection system is 3.9×10^{-5} x-ray counts per proton.

The loading of deuterium in the Pd cathode was determined by removing deuterium from the loaded cathode. Deuterium which evolved when the applied current was disconnected was captured and its volume measured. The remaining deuterium in the palladium was expelled by heating the sample in a thermogravimetric analysis system (Dupont 951 thermogravimetric analyzer). Based on the total mass of the gas liberated, the loading of the sample was 0.79 ± 0.02 deuterium atoms per Pd atom.

On the basis of our calculations, the measured counting rate corresponds to 0.07 ± 1.1 fast protons released per

second in the cathode. From our measurement of the loading of the Pd cathode with deuterium, the absolute fusion rate detected is consistent with zero with a 2σ upper limit of 1.6×10^{-20} fusions/(sec deuteron), assuming equal probability for reactions (1) and (2).[3]

Another attempt to detect charged particles from electrochemical fusion[9] has also resulted in a null result. Because the charged particles were detected directly, that experiment had inherently greater sensitivity than ours. Their arrangement used the Pd cathode as a thin window on the electrochemical cell with only one side exposed to the electrolyte.

We have run several other electrochemical cells, one for as long as six weeks, in an effort to detect x rays from charged-particle fusion products. No evidence of nuclear fusion has been detected. The data reported here are the result of our efforts to reduce and understand the x-ray background associated with this type of measurement.

While the upper limit we are able to place on the fusion probability, as evidenced by proton production and subsequent excitation of Pd K shell vacancies, is not as stringent as some neutron measurements,[4] the work reported here provides a useful experimental statement regarding fast charged particles from reaction (1). If we accept the proposition that reactions (1) and (2) are dominant in the deuterium fusion process, then the measurements of Gai *et al.*,[4] when combined with our result, rule out nuclear fusion as the energy source observed by Fleischmann and Pons,[1] by a factor of roughly 10^{10}.

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[6]The Pd foil was obtained from Engelhard Corp. and rolled to this thickness. Its purity is 99.9%.

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[8]The x-ray detector used in this experiment was a Tracor model No. 103-22.

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