SEARCH FOR NEUTRON, GAMMA AND X-RAY EMISSIONS FROM Pd/LiOD ELECTROLYTIC CELLS: A NULL RESULT

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Abstract

We have conducted a series of experiments using state-of-the-art neutron, gamma and x-ray detectors to search for evidence for nuclear reactions occurring in Pd/LiOD electrolytic cells. No evidence for primary or secondary emissions from nuclear reactions was obtained in extended experiments.

I. INTRODUCTION

Since the announcement by M. Fleischmann and B. S. Pons of excess heat production via nuclear reactions in Pd/LiOD electrolytic cells[1], we have stressed the need for state-of-the-art detectors to scrutinize these claims. Here we present new results from our most sensitive detectors. We also caution that compelling results can only be obtained with state-of-the-art systems, which we describe.

II. OVERVIEW OF DETECTOR SYSTEMS

Our primary detector for low-level neutron emissions consists of a combination of a large plastic scintillator core with a surrounding bank of sixteen 3He-filled proportional counter tubes (Figure 1), with all signals digitized at 50 Mhz and stored in computer memory. The central plastic scintillator is 35 cm in length and 8.9 cm in diameter. A central cavity of 4.8 cm diameter admits test cells. Fast neutrons from the sample can generate a recoil proton in the plastic generating scintillations (efficiency about 40%) which are viewed by a photomultiplier tube. Then the neutron slows further in polyethylene moderator 28 cm diam. X 30 cm long, and finally may be captured in one of 16 helium-3-filled proportional counter tubes embedded in the moderator (efficiency for 2.5 MeV neutrons is 34%). These tubes are arranged in four quadrants incorporating 4 proportional-counters in each segment.

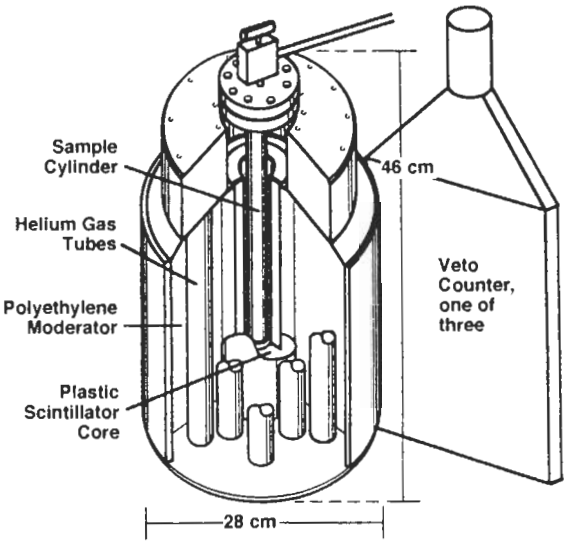

Figure 1. Los-Alamos-type detector with plastic scintillator core detector and cosmic-ray veto paddles (3) added at BYU.

The detector and experiments have the following special features:

1. All signals are digitized using a LeCroy fast-waveform digitizer operating 50 MHz, so that we retain pulse-shape information as well as timing between pulses. Pulse-shape analysis permits excellent noise rejection, along with giving some neutron-energy information (from the prompt plastic scintillator pulse). By rejecting (in software) events having small or no plastic pulses, we strongly discriminate against slow (especially thermal) neutrons. This background-reducing feature is not available to many detectors including those using BF3, 3He and even the Kamiokande detector in Japan[2]. By studying neutron-capture time distributions based on prompt and capture-neutron pulses, we check whether observed distributions agree with those found using a plutonium source.

2. The PC-based data acquisition system records which of the four quadrants of the 3He-type counter showed neutron capture, allowing for checking that the quadrants are hit in equal proportions.

This detector segmentation has, for example, allowed us to throw out apparent large bursts of neutrons

(over 60 "neutrons" in a 320-microsecond window) whose signals unrealistically came from just one quadrant. (Occasionaly two quadrants are involved, due to electronic cross-talk). Even if neutrons are somehow emitted in one or two directions, the polyethylene moderator has the effect of spreading out the neutrons as they slow down, so that detected neutrons of sufficient statistics will necessarily appear in all four quadrants.

We have seen several cases of such large bursts in the past three years of running; but all bursts of over five detected neutrons have proven to be spurious. (see Ref. 3) The large multiplicity "events" are correlated with high-voltage breakdown in the standard electronics of this ^{3}He-type detector. Therefore we retract earlier claims of high-multiplicity time-correlated-neutron events, notably those which appeared to correlate with sample cooling using liquid nitrogen.[4] Compelling data for large neutron bursts require detector segmentation and pulse digitization (allowing signal visualization) as we have done, or other reliable methods of noise elimination.

3. We have added three large cosmic-ray veto counters to show the passage of cosmic rays, which events are rejected off-line. Passive shielding of at least 35 m of rock (12,000 g/cm^2) also greatly reduces cosmic ray-induced events and removes dependence of cosmic-ray rates on fluctuations in atmospheric pressure. After cosmic-ray rejection, the event rate is approximately 0.65 neutron-like singles per hour with an efficiency of 14% for 2.5 MeV neutrons, and 0.07 burst-events per hour with a detection efficiency exceeding 20% (increasing with neutron-burst multiplicity)[5].

4. Two additional highly-sensitive neutron detectors are available in the same deep-underground facility based on a different neutron-capture scheme (capture in lithium-doped glass), to permit checking of any positive results found in the primary detector[5].

III. RESULTS OBTAINED WITH Pd/LiOD ELECTROLYTIC CELLS

A. Experimental Protocols

The neutron data presented below represent 1,054.6 hours (6.3 weeks) of observation of Pd/LiOD cells and backgrounds in our most sensitive neutron detector. Experimental protocols follow those provided by Dr. Thomas Passell[6], for these Pons-Fleischman-type cells, namely:

1. Pd cathodes (6 mm diam. except 4 mm diam rod described in 3 below) were used in a 0.1 M LiOD solution (in D_2O). Electrode spacing of the Pd rods relative to Ni-gauze which formed the cylindrical anode is approximately 2 mm, with a septum used to prevent electrical contacts.

2. Three cells were polarized in series at 40 mA from Sept. 24, 1993 to October 25, 1993, then at 80 mA until October 29, 1993.

3. Following a suggestion of Prof. K. Wolf[7], a fourth Pd/LiOD cell was operated at high altitude (8,500') for three weeks at 20mA/cm2, then added in series connection with the other three cells on October 25, 1993.

4. The palladium cathode rods were scraped/sanded approximately every seven days, and replaced in the cells within a period of about fifteen minutes to minimize deuterium loss from the cathodes during the cleaning procedure. We noticed that the cell potential slowly increased over days of (constant-current) operation, then decreased after the cathodes were cleaned, showing that a resistive surface coating had built up during cell operation. We also observed a gradual rise in electrolytic cell temperature, using a platinum-resistance probe, consistent with increased resistance and joule heating as the resistive surface coating developed on each Pd cathode.

5. A 12-hour cooling treatment was applied to the three primary cells on day 17. The fourth cell (described in 2 above) was subjected to diurnal cooling and heating due to its exposure to a mountain environment; the electrolyte was found to be frozen on two occasions.

6. Boron and aluminum (about 0.001 molar) were added to the LiOD electrolyte on the 18th day.[7]

B. Search for Neutron Burst Events.

A neutron burst event is defined as having a hit in the plastic scintillator core followed by two or more signals in the 3He-filled proportional-counter tubes within 320 microseconds. Since the die-away time for neutrons in the outer detector/polyethylene moderator is 55 microseconds, there is a possibility to see multiple distinct neutron hits there. In effect, the outer detector "de-multiplexes" neutrons should an instantaneous burst occur, as first reported by H. Menlove et al.[4]. A burst is then defined as two or more neutrons captured in 3He within 320 microseconds of a start pulse in the plastic

scintillator. The background rate for bursts is (0.07 +- 0.01) n/hr, all from multiplicity = 2 events, established using Pd loaded with hydrogen in 394 hours of separate runs.

We also scrutinize the time spectra of 3He-captured neutrons relative to the start pulse in the plastic scintillator to determine whether the time distribution corresponds to the 55-microsecond die-away time for neutrons in the 3He-portion of the counter, as seen with a plutonium neutron source.

The Pd/LiOD cells described above were polarized for 708.8 hours. During this time, 24 neutron-like burst events were seen, all having multiplicity = 2. (This represents approximately one burst candidate per 30 hours, a very low rate indeed.) Thus, the neutron-like rate for these events was 48/708.8h = (0.07 ± 0.01) n/hr. These numbers are in complete agreement with those found with hydrogen controls discussed above. There was no significant change in rate for neutron-like burst events between background and runs with electrical currents in the Pd/LiOD cells. There is no indication of a neutron burst signal above a very low background.

To complete the scrutiny for burst-like events, we compare time spectra from these Pd/LiOD electrolytic cell runs with those obtained from H2-control runs and from Pu-source runs. Figure 3 displays the time intervals between each start pulse in the plastic scintillator detector and each stop pulse from the 3He-type outer detector.

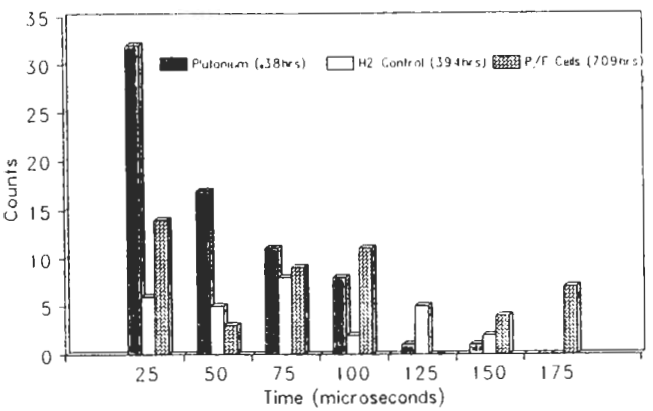

Figure 2. Time intervals between plastic scintillator "start" pulse and ^{3}He-capture "stop" pulse for multiple-neutron ("burst") events. the spectra associated with P/F electrolytic cells and with H_2 controls are consistent and represent background spectra.

The neutrons from the plutonium source follow a pattern consistent with the calculated 55-microsecond die-away time for neutrons in the counter, but neither the controls nor the Pd/LiOD cells show such a distribution (the latter two spectra are consistent with backgrounds.) We conclude that there is no evidence whatsoever for neutron-burst activity in the electrolytic cells. The upper limit on excess power from nuclear, neutron-generating reactions in the electrolytic cells is at the picowatt level.

C. Total neutron-like count rate

Even though there is no neutron-burst signal, there may still be neutron counts above background which we consider "singles." The background rate for such events has been established as (0.65 ± 0.1) counts/hour using Pd loaded with hydrogen. Figure 3 displays results from each run of the electrolytic cells, showing 1-sigma error bars (statistical only). All of the observed rates are entirely consistent with background levels of 0.65 h^{-1}. This exercise has as its conclusion that no neutrons were seen above very low background levels, in a high-efficiency detector. The most important observation may be that state-of-the-art neutron detectors are now available for studies requiring high-sensitivity instruments.

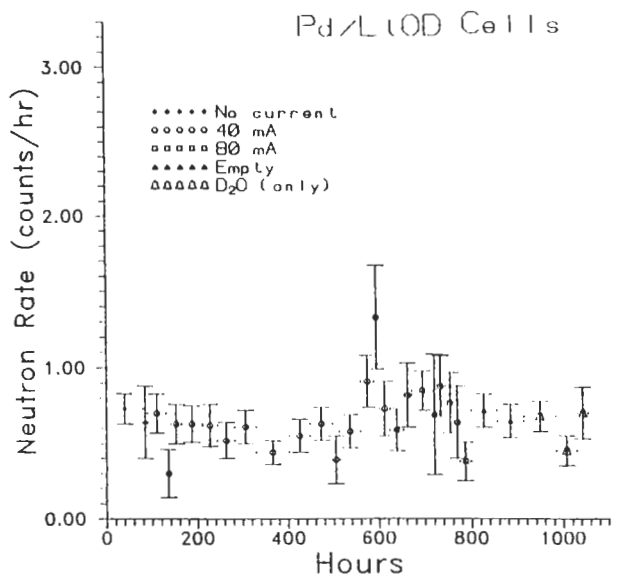

Figure 3. Signal rate in Pd/LiOD cells compared with rates seen for background conditions. Background rates are only 0.7 counts per hour, and there is no evidence of neutron production (above background) in the Pd/LiOD cells.

IV. GAMMA AND X-RAY SPECTROSCOPY

A. Search for gamma rays

Immediately following the neutron search, all palladium rods were taken to Los Alamos for gamma-ray spectroscopic analysis. The purpose of this search was to determine whether radioactive isotopes of palladium,rhodium, ruthenium and silver might have been generated during the electrolytic runs, pursuant to claims of Y. Kucherov and others of such transmutations in deuterium-loaded palladium[7,8]. All four Pd rods were placed in a low-background germanium detector operated by Dr. J. Parker and counted for 75,000 seconds. No gamma lines above background were seen, except for a weak 59.5 keV line which represents americium-241. The americium contamination was traced to the nickel gauze used for anodes. The migration of americium from anode to Pd cathode during operation of the electrolytic cells demonstrates that radioisotopes can be picked up by the cathode originating from either the electrolyte or the anode. Therefore, any claims of nuclear transmutation in such cells must first show that the claimed radioisotopes were not originally present in the electrolyte or the anode. Contamination of the cathode must also be carefully scrutinized.

Further gamma-spectrographic analyses of the palladium cathodes used in experiments at BYU and Kamiokande over the past five years have been undertaken: we found absolutely no evidence for radioisotope formation in any palladium cathodes. Careful scrutiny should therefore be applied to any claims that nuclear reactions produce transmutations in electrolytic cells. In particular, claims that radioisotopes are formed far off the line of nuclear stability should immediately arouse suspicion that materials used in the electrodes or electrolyte could have been contaminated or subjected to irradiation by an energetic particle beam. For example, if palladium-100 is found by gamma spectroscopy, then beam irradiation is likely since negative-Q reactions are implicated.

B. Search for X-rays

We have also followed our own challenge[9] of searching for x-rays as would be expected if nuclear reactions are indeed producing measureable heat in electrolytic cells. Nuclear reactions are characterized by release of MeV-scale energies, hence their importance to power-production schemes. Energy release at the nuclear level implies that secondary x-rays will be produced in the environment of a metal lattice, where only tens of keV are required to generate x-rays. That is, if nuclear reactions are indeed producing heat at the levels claimed (>1 mW), then sufficient x-rays should be produced to be detectable, since x-rays arise from ionizing effects of nuclear products on the materials in which the purported heat develops. Thus, x-ray spectral measurements provide a crucial test for the presence, or absence of heat-generating nuclear reactions.

Characteristic x-rays of Pd (K-alpha of 21.1 keV) or Ni (K-alpha of 7.5 keV) which result from K shell vacancies produced by nuclear products are readily detected. We have searched for such lines using two x-ray spectrometers, a 10mmX10mm silicon detector having high sensitivity down to about 4 keV[9] and a lithium-drifted silicon detector with high sensitivity down to approximately 1 keV. We used a Pd/D2O electrolytic cell in which 25 micron Pd foil formed both cathode and external wall. No x-ray production was seen with this electrolytic cell. Similarly, a search for x-rays was conducted using a Ni/H2O cell in which the Ni cathode was placed against a very thin plastic window. Again, no x-ray production was in evidence in the electrolytic cell. Using a montecarlo calculation to determine the overall x-ray detection efficiency from electrolytic cells[9], we set an upper limit of 10 microwatts of excess power from these cells, from <u>any</u> nuclear reactions which produce secondary x-rays.

In a "search for cold fusion using x-ray detection"[10], M.R. Deakin et al. found, as we have, that no x-rays above background were produced by Pd-LiOD electrolytic cells. An important caveat is provided in that paper: "Room background radiation fluoresces the cathode and Pd K x-rays are therefore present as an artifact of background."[10] Thus, the presence of x-rays alone (including fogging of x-ray film) is insufficient to demonstrate the presence of heat-generating nuclear reactions.

No "cold fusion" experiment anywhere has shown the presence of characteristic secondary x-rays lines (using an x-ray spectrometer) which would characterize fusion or any other nuclear reaction in a metal lattice[11]. There are some experiments that show fogging of x-ray dental film, but such experiments are too crude to provide quantitative information regarding x-ray energies and intensities, and are subject to artifacts.

Thus, we find no compelling evidence to link nuclear reactions to excess-heat production claims. Instead, the lack of significant (primary or secondary) x-rays, gammas and neutrons after five years of searching argues convincingly against claims of excess heat production via nuclear reactions in electrolytic cells (or

equivalent). This conclusion is supported by related experiments at BYU which show up to 750% "excess heat", but which apparent "excess heat" is in fact due to hydrogen-oxygen recombination in the cells coupled with commonly-used (but misleading) analysis techniques for excess-power production in "cold fusion" experiments[12]. Thus, the "excess heat" admits prosaic explanation: it is not nuclear at all.

V. CONCLUSIONS

In order to find compelling evidence for cold-fusion effects, state-of-the-art calorimeters and nuclear detectors are requisite. Table 1 juxtaposes such systems with other systems which are still more generally in use. It is disquieting that some researchers select open electrolytic cells over closed cells, and excessively long sampling intervals (e.g., 5-minute sampling intervals for input voltage used by Pons and Fleischmann in calculating excess heat over a 10-minute boiling period[13]). Some researchers continue to use x-ray films instead of x-ray spectrometers, helium or tritium gas sampling instead of charged-particle spectrometers, Geiger counters rather than silicon or germanium detectors, and neutron survey meters instead of sensitive neutron detectors as described above. It is time to strongly question claims of cold fusion based on crude techniques and to demand tests at a rigorous scientific-proof level. Compelling evidence requires use of the best instruments available, incorporating fast data-sampling and digitization methods, the use of different detectors whose signals agree quantitatively, and presence of signals well above background levels. A real signal should be capable of scaling, and should not shrink as background levels are reduced.

TABLE 1. COMPARISON OF COLD-FUSION RESEARCH METHODS

It is evident that much of the present confusion surround "cold fusion" stems from the continued use of inadequate detectors. This list juxtaposes crude, better and state-of-the-art systems to promote the quest for compelling data, one way or the other. Use of the best available methods is clearly the path-of-logical science.

Crude (simply add to the confusion)	Better (but not good enough)	State-of-the-art (can provide compelling evidence)
Neutron survey meters BF3	Segmented 3He, Plastic scintillators	Segmented 3He 3He or Li-doped glass *plus* scint. and digitizing
Helium gas detection, Tritium gas	Charged-particle det. (Si surface barrier) (requires thin foil)	Thin dE/dx detectro plus Si spectrometer (particle ID & energy)
X-ray film	X-ray film with energy-filters	X-ray spectrometer (Si,HgI2, CdTe,etc.)
Geiger-Mueller counter	see detectors listed above; Ge detector	
Infrequent I*V(t) sampling (e.g., every 300 s)		Integral I*V(t) correct via frequent, redundant sampling
Open cell calorimetry, no H2/D2 +O2 during experiment	Measure H2/D2 +02 simultaneous w/heat	Recombiner inside separate calorimeter
Metal of unknown source quality or purity		Alloyed with known purity and properties
D2O of unknown	D2O from known source,	Highly distilled D2O, known H,O source, isotopes
Visual techniques	Computer-logging several probes	Redundant probes with fast data acquisition
Theories which disregard P, E conservation or light-cone constraints (e.g., "heating lattice")	Fractofusion ignoring e- vs. d+ acceleration	???

With these criteria for state-of-the-art detectors, we find that no compelling evidence for neutron, gamma or x-ray production from deuterided materials currently exists in any cold-fusion experiment, including our own. The only verified form of cold nuclear fusion to date is muon-catalyzed fusion. Nevertheless, having an obligation to resolve remaining issues[14], we will continue our search for several more months. We invite those with evidence for neutron production to accept our invitation to test their systems in the deep-underground neutron detection facility in Provo Canyon in order to confirm results. Gamma and x-ray spectrometers are also available on request.

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