Lawrence Livermore National Laboratory

September 8, 1989

Dr. William Woodward
Energy Research Advisory Board
U.S. Department of Energy
Washington, D.C.

Dear Dr. Woodard,

Please find enclosed details of the Lawrence Livermore National Laboratory Cold Fusion experiment titled "Cold Fusion Electrolytic Experiments" by Aldridge, Contolini, Ishikawa, and Slaughter of LLNL. In their experiments a sufficiently large number of parameters were measured to meet the requests contained in Dr. Huizenga's letter of August 9, 1989. This experiment was briefly described in the original summary of the LLNL Cold Fusion Program from our Director John Nuckolls to the Department of Energy on June 16, 1989. See page two, paragraph two. Please feel free to call the authors or myself for additional information.

Sincerely,

John F. Holzrichter
Assistant to the Director
for IR&D

cc: J. Nuckolls
J. Bigeleisen

attachments:

Cold Fusion Electrolyte Experiments
Summary - LLNL Cold Fusion Experiments

Cold Fusion Electrolytic Experiments

F. T. Aldridge, R. J. Contolini, M. Y. Ishikawa and D. R. Slaughter

This is a summary of 'O' division attempts to duplicate the Fleischmann-Pons electrolysis cold fusion experiments. Two types of cells were used, one a duplicate of the Fleischmann-Pons cell and another with a much larger flat plate electrode.

The first experiments were run in a duplicate of the Fleischmann-Pons cell, with somewhat smaller dimensions. The cathode was a central Pd cylinder of 6.5mm diameter and 70mm length. Only about 55mm of the Pd cylinder was immersed in the solution. The anode was a Pt coil wound on glass rods which were held by teflon end spacers. The teflon spacers also held the central Pd cathode. The electrode assembly was mounted inside a 24mm inside diameter graduated cylinder. The cell contained 24 cc of 0.1 N. LiOH in 99.8% D_2O.

The first run was made at 4 amperes and 12 volts, with a current density of 0.4 amperes per square centimeter on the Pd electrode. After about 1.3 hours, the Pd rod suddenly glowed red, then white for a brief moment, and the cell exploded with a force similar to a small firecracker. The Pd electrode was discolored, except where it had been covered by the teflon holder. The teflon was not distorted or melted where it had been in contact with the Pd rod. Most of the force of the explosion was probably due to the recombination of electrolytically produced hydrogen and oxygen which was ignited by the hot Pd surface. The discolored layer was 0.5 micron thick and contained Na, K, Ca, and Li. All the metals except Li probably were leached out of the glass vessel by the action of the LiOH solution.

The explosion was possibly a surface reaction not involving the bulk of the Pd rod, or a reaction between the electrolyte or hydrided Pd and a Li-Pd alloy formed on the electrode surface. This may be the same phenomena reported by Fleischmann and Pons.

The apparatus was rebuilt and more experiments run without explosions. In these experiments, current densities were used of 0.21 to 0.39 amperes per square centimeter for periods of up to 32 hours. A NE213 Nuclear Enterprise organic scintillator was used to detect both neutrons and gamma rays. Neither neutron nor gamma counts were seen above the background level.

A water jacketed cell was used in an attempt to measure cell heat output by measuring the temperature of the water entering and leaving the water jacket. The system was too crude to give the accuracy of interest and the attempt was abandoned. Accurate thermal

measurements on this system will require a great deal of careful design work or the purchase of a commercial calorimetry system if valid results are to be obtained. Also, a closed system will be needed, where the hydrogen and oxygen produced are catalytically recombined within the cell and no gas exchange is allowed with the surroundings. (The Fleischmann-Pons type of experiments, which are not closed in this sense, have been widely criticized for their "open" feature.)

Some of our experiments were run in a larger cell. The Pd cathode used was a ten centimeter square plate 0.5 millimeters thick. Two ten centimeter square Pt mesh anodes were spaced one half inch from each side of the cathode. The electrodes were held in place with teflon spacers and the electrode assembly was placed in a glass vessel. This cell was placed in a cooling bath and the electrolyte temperature was approximately 40°C during the run. The cell was run for 65 hours at 10 amperes and 18 volts, with a current density of 0.05 amperes per square centimeter. A NE213 Nuclear Enterprises organic scintillator detector was used to detect gamma rays and neutrons. Several periods of enhanced neutron + gamma emission were seen at levels of 4 to 7 times the background level. A building wide power failure, caused by a bird shorting a power line, ended the experiment an hour after these observations. When the experiment was restarted after power was restored no more enhanced emission episodes were seen.

Since the enhanced count levels could not be duplicated, we do not know if they were significant. They may have been caused by unknown variability in detector sensitivity.

After the experiment was concluded, the electrolyte and the Pd electrode were analyzed for possible fusion products. The Pd electrode showed no ^3He or ^4He above the expected background levels. The electrolyte showed a 13% enrichment in tritium, but this was essentially what would be expected from the electrolytic enrichment of the D_2O, using literature valves for enrichment factors.

The initial volume of electrolyte in the cell was 1350 ml. During the course of the experiment, 560 ml of electrolyte was added to keep the cell volume constant, for a total volume of 1910 ml of electrolyte used. The total current passed through the cell would have electrolyzed 220 ml of electrolyte; 340 ml was lost as vapor or spray entrained in gases liberated by electrolysis. The hydrogen produced by electrolysis would be depleted of tritium. This tritium would remain in the electrolyte, while the vapor or spray losses would not be isotopically selective. Therefore, the remaining electrolyte should be tritium enriched by a factor of ~1.130.

Analysis of the electrolyte showed 57.1 picocuries of tritium per ml before the experiment and 63.6 picocuries per ml after the experiment for an enrichment factor of 1.128. Thus, no

tritium production due to cold fusion was seen. The D_2O used was obtained from Sigma Chemical Company and has an isotopic purity of 99.8% deuterium.

June 16, 1989

LLNL REPORT ON COLD FUSION

With the appearance of reports on "Cold Fusion", scientists at the Lawrence Livermore National Laboratory began an increasingly sophisticated series of experiments and calculations to explain these phenomena. These experiments can be categorized as follows: (a) simple experiments to replicate the Utah results, (b) more sophisticated experiments to place lower bounds on the generation of heat, and production of nuclear products, (c) a collaboration with Texas A&M to analyze electrodes and electrolytes for fusion by-products in a cell producing 10% excess heat (we found no by-products), and (d) attempts to replicate the Frascati experiment that first found neutron bursts when high pressure deuterium gas in a cylinder with Ti chips is temperature cycled.

We failed in items (a) and (b) to replicate either the Pons/Fleischmann or Jones phenomena, and appear to have duplicated the Frascati results, (d). Summaries of (b) through (d) are described below, as is a theory effort based on cosmic ray muons.

Electrolytic Cells

A typical experiment involved comparisons of electrolytic cells using D_2O and 0.1 M LiOD with cells using H_2O and 0.1 M LiOH. Palladium wires 1 mm in diameter were used in the two runs described below, the first were simply swaged and drawn, but since there were reports that remelting was necessary we annealed the wire for the second run for 3 hours at 600°C. The first pair of cells ran for 156 hours in a common bath, each with a 35 cm long swaged wire, with current densities starting at 90 mA/cm^2, increasing to 270 mA/cm^2, and then to 450 mA/cm^2. The slight temperature difference between the two cells remained essentially constant throughout the run, that is there was no excess heat difference between them. Using relatively low efficiency neutron detectors, no neutrons above background were observed. An analysis of the Pd for ^{3}He and ^{4}He was negative, setting limits of a few times 10^{10} atoms per gm. After cutting a length of this wire from the above D_2O cell it was run in another thin-walled cell for 4 more days with a Geiger counter near it. No counts above background were observed. A second experiment used a pair of cells with annealed Pd, and it ran for 330 hours at a current density of 230 mA/cm^2 with similar null results. Analysis of that wire showed no helium buildup either. In both cases the thermometry might have missed seeing excess heat at the 10% level.

One cell similarly prepared to the one described above was setup for sensitive neutron counting. The cell ran for 167 hours and featured a proton recoil telescope detector that eliminated all background counts, and it used a sensitive liquid scintillator (7% efficiency, absolutely calibrated, with neutron/gamma pulse shape discrimination). The latter set a limit of 0.2 neutrons/sec from the 27 g Pd wire, half the level reported by Jones at BYU with a 5 g sample.

Several other experiments with electrolytic cells were run in a second independent group. Their experiment was designed to test for helium buildup in Pd, for neutrons, and for excess heat. Calorimetry with a minimum thermal detectivity of about 2% of the input electrical power was used. Similar (null) results were found by this group. If the excess heat is a 10% effect, as observed at Texas A&M, then this calorimeter has sufficient sensitivity and no effect was observed.

Collaboration with Texas A&M

We analyzed samples of Pd from a cell run at Texas A&M by Srinivasan, who reported 340 mW heat flow into their sensitive microcalorimeter with 300 mW heat input, indicating an excess heat generation of 40 mW. However, it should be remembered that an additional power input of 150 mW was needed to separate the D_2O molecules. To the level of 3×10^5 ^3He atoms and 5×10^8 ^4He atoms in the sample (about 20 mg) there was no helium generation in the wire. This is many orders of magnitude below the level associated with 40 mW of fusion power for one hundred hours. Surface analysis with Auger and SIMS techniques revealed a substantial layer of material (Ca, C, Fe, O, Cl, Li, N, Cu and traces of other elements) plated out on the surface. These gave no clue as to possible excess heat generating mechanisms however they indicate suspicious additional electrochemical mechanisms. An analysis of the electrolyte before and after the run showed no change in the tritium level in the original electrolyte.

Neutron Burst Experiments

Experiments using pressurized deuterium cylinders loaded with Ti in various forms, similar to the Frascati experiments, were carried out by several groups at Livermore. In a large number of runs, cycling the temperature from ℓN to room temperature, the results were null. Very recently, with detector efficiency improved to 15%, with higher pressure operation to 60 Atm, and with a pair of coincidence detectors incorporating burst mode triggering in the detection circuits, one group observed what appear to be bursts of a few hundred neutrons. These do not correlate with the temperature of the cylinder at the time of the bursts, which is contrary to data from LANL. Consistent with Sandia, it requires multiple detectors and the observation of coincident signals to place some confidence in the results. Many

spurious signals appear on one detector but not another, and this could be missinterpreted as neutrons. At present, the temporal signals are being studied to assure ourselves that the signals are indeed neutrons. Plans to repeat this in DT gas have been made.

Muon Theory

A theory to explain that the fusion neutrons observed by Jones etc. were generated from cosmic muons was developed here. The theory falls short by about two orders of magnitude to explain the level of neutrons seen by Jones. It was found that charge exchange with deeply trapped electrons around the Pd (or Ti) limits the muon to catalyzing at most a dozen events. In 13% of the muon catalyzed D-D reactions the muon sticks to the resulting He atom, and must subsequently be reionized by deeply trapped (a few kV) electrons. Instead, however, the muon charge exchanges. Further, experiments in Japan with accelerator generated muons, reported recently in Santa Fe, definitively set an even lower limit to muon catalyzed reactions then the above theoretical explanation.