A STUDY OF "COLD FUSION" IN DEUTERATED TITANIUM SUBJECTED TO HIGH-CURRENT DENSITIES

ROBERT B. CAMPBELL* and L. JOHN PERKINS
Lawrence Livermore National Laboratory, P.O. Box 808
Mail Stop L-644, Livermore, California 94550

Received June 29, 1989
Accepted for Publication July 17, 1989

In response to the startling announcement of fusion reactions occurring at room temperature by Fleischmann and Pons (F-P), the possible role of high-current densities in producing neutrons and excess heat in deuterated titanium maintained near ambient temperatures and pressures is examined. The apparatus used consists of a balanced resistive circuit containing a deuterated "active" element and a hydrogenated "control" element. The use of a simple electrical circuit (no electrolysis) with elements made of chemically stable TiD_x, X = 0.9, removes the complications involved in distinguishing between heat released by chemical versus nuclear processes in an electrolytic cell. This apparatus tests the possibility that the role of high-current density in the F-P experiments is to create such nonequilibrium states as strong pinching due to current microchanneling in the metallic lattice. This strong pinching, in turn, could reduce the deuteron-deuteron separation sufficiently to cause significant fusion. To detect neutrons, an NE-213 liquid organic scintillator spectrometer is used, with gamma counts eliminated by means of pulse-shape discrimination. Samples are subjected to current densities of ~50 A/cm^2 for time periods of 19 h. This current density is a factor of 100 greater than the largest value reported by Fleischmann and Pons. No significant neutron levels are detected above background. The temperature rise of the two samples during the application of the current can be explained by joule heating alone, with no other heat sources present. Based on these experiments, no excess heat is observed within the accuracy of the apparatus, which is estimated to be 10%.

It is concluded that the large quantity of excess heat reported by Fleischmann and Pons is due to the presence of factors other than the current density.

INTRODUCTION

The original announcement of the observation of the "cold fusion" phenomenon in electrochemical cells by Fleischmann and Pons (F-P) sent many researchers around the world to their laboratories to attempt to reproduce the results. The original reports from F-P indicated that about four times as much power was liberated from "fusion" reactions as was consumed by the cell. The excess enthalpy generation was claimed to increase with the applied current density, as well as the sample volume. Large extrapolations in \( Q = \frac{P_{\text{obs}}}{P_{\text{input}}} \) were made for larger samples at higher current density. The large \( Q \) value, as well as the dependence on current density, motivated us to embark on the present study. Based on the F-P results, the excess heat should be easily observed, and therefore measurable by an apparatus that has a relatively low sensitivity. Other workers have demonstrated that the excess heat, if observed at all, is substantially smaller than that reported by F-P, and fairly sensitive calorimetry is necessary to see the effect. Therefore, the results we present here establish that the excess heat released is below a certain level. Based on our experiments, the excess heat was zero within the estimated accuracy of our apparatus.

We adopt the resistive circuit approach with deuterated titanium instead of the electrolytic cell with a palladium cathode for several reasons. First and foremost, the resistive circuit isolates the effect of current and current density from other effects associated with the electrolysis process. Our apparatus tests the possibility that the role of high-current density in the F-P experiments is to create such nonequilibrium states as strong pinching due to current microchanneling in the metallic lattice. This strong pinching, in turn, could reduce the deuteron-deuteron separation sufficiently to cause significant fusion. A second reason for the use of the resistive circuit is that the ability to pass large currents (~50 A) through massive samples (~30 g) is quite easy by using a resistive circuit driven by a constant current power supply. By using a single circuit consisting of a deuterated sample and a hydrogen control sample in series, temperature differences can be measured and related directly to the excess enthalpy generated in the deuterated sample. Estimates of excess enthalpy generation as a fraction of the ohmic dissipation are possible by comparing the temperatures of the active sample and the control sample relative to the ambient temperature. The accuracy of such a scheme is limited by our ability to create identical electrical resistances and heat transfer environments for the active element and the control. We ensure that the electrical resistances are the same by direct measurement, and we equalize the heat transfer environment by measuring temperature differences between two identical samples of

*Employed by TRW, Inc., One Space Park, Redondo Beach, California 90278.
elemental titanium substituted in the circuit. Based on these calibrations, we believe that the error in our apparatus is on the order of 10%, about the same as the upper bound of the fractional excess enthalpy we infer from the loaded sample temperature differences.

We use titanium instead of palladium for two reasons. First, the titanium can be loaded to concentrations up to 2:1 D:Ti, substantially larger than that attainable with palladium. Because the titanium becomes very brittle at high loadings, we limit our loading to \(-1:1\), on the same order as the rumored loading of the F-P electrode \(^6\) of 1:1:1 (D:Pd) under the influence of a strong electrochemical overpotential. The second reason for using titanium is that the titanium and hydrogen isotope forms a stable compound in the temperature ranges in which we operate the resistive experiments. Our operating temperature range is from room temperature to \(\sim 100^\circ\text{C}\). Palladium containing hydrogen isotopes, on the other hand, is observed to outgas readily at these temperatures and, in fact, is a catalyst for the exothermic recombination of hydrogen and oxygen into (heavy) water vapor. Therefore, the use of titanium eliminates one possible chemical explanation for the excess heat observed by F-P.

Deuterated and hydrogenated titanium samples are readily available from washer guns used on the mirror device 2X11B (Ref. 3). These rings of titanium are annealed at high temperature \((1100^\circ\text{C})\) for several hours at high vacuum. The \(D_2\) or \(H_2\) gas introduced at these temperatures is easily absorbed in \(\sim 1\) h into the sample. Consequently, annealing of the material occurs naturally as a part of the deuterating process. Annealing is claimed to be an important element in the cold fusion process.\(^2\)

**EXPERIMENTAL APPARATUS**

Figure 1 shows the experimental setup that we constructed to search for cold fusion. The circuit consists of two washers loaded with either hydrogen or deuterium connected in series with No. 6 copper welding cable. Care was taken to ensure that the lengths of wire connecting each of the samples were kept the same, in order to keep the thermal mass and electrical resistance of each sample unit the same. The samples are isolated thermally from each other and the power supply by three heat sinks maintained at room temperature by forced convection. The purpose of these heat sinks is to eliminate conduction heat transfer between the samples and the power supply. Conduction heat transfer would modify the temperatures between the sample and the control. These sinks are denoted by the ground (thermal not electrical) symbols on the diagram. At each of the sinks, the conduction heat transfer into the junction is removed by forced convection, thereby eliminating thermal crosstalk between the two samples. Type K thermocouples are attached to the samples and each of the heat sinks to monitor the temperature differences of all the elements during the experiment. The accuracy of these thermocouples is about \(\pm 0.1^\circ\text{C}\). The temperature data were taken and recorded periodically with a strip-chart-type data logger.

The neutron data are compiled with a NE-213 liquid-scintillator-based spectrometer, with gamma counts eliminated using pulse-shape discrimination. The pulse-height spectrum data were processed and stored automatically by a Toshiba T-1100+ microcomputer. A more detailed description of the neutron spectrometer apparatus can be found in Ref. 4. The deuterated sample to be diagnosed for neutron emissions is housed in a polyethylene "igloo" to reduce the neutron background. The average thickness of the igloo walls is \(\sim 10\) cm, and we estimate that the background counts are reduced by a factor of \(\sim 3\) by this shielding. The distance from the sample to the detector face is such that the subtended solid angle is \(\sim 0.02 \times (4\pi)\).

The igloo tends to change the convective heat transfer environment of the deuterated sample, so each of the samples is housed in a vacuum dewar, and the lids and outer walls of
the dewar are heavily insulated. This step, combined with the careful measurement of lead wire lengths, is important to make the bulk heat transfer coefficients of the two samples the same. This heat transfer coefficient, along with the temperature difference between the active sample and the control, is used to determine the upper limit of the enthalpy production rate due to any fusion process. Based on calibrations with blank titanium rings, we estimate that we can make the resistances and heat transfer coefficients the same to within 10%.

EXPERIMENTAL PROCEDURE

Before we begin an experimental run to look for the effects of cold fusion, the detector's energy scale is calibrated with a 207Bi gamma source, and then a background neutron count is taken. The count time is either 18 or 19 h, depending on the case considered. The active run with the sample in place typically begins within 3 h of the completion of this background run, with the same counting time.

The circuit is then balanced between the hydrogenated control and the deuterated active sample with respect to electrical resistance, thermal mass, and heat transfer geometry. The samples are preloaded with hydrogen isotopes to obtain the same electrical resistances. For the loading densities we employ, - 0.9(D,H):1(Ti), the electrical resistance is about twice that of elemental titanium. Although full stochiometric loading [i.e., 2(D,H):1(Ti)] of these samples is possible, they become very prone to fracture due to hydrogen embrittlement above loading densities about 1:1, thus our limit of 0.9:1. The loading of 0.9:1 in our samples is above the room temperature stochiometry of PdD0.7, and approximately equal to the stochiometry of PdD1 that has been rumored to exist in the F-P electrodes under the effects of a strong overpotential. The most difficult step in this balancing process is the equalizing of the contact resistances between the copper lugs attached to the welding cable and the samples. The contact surfaces are polished with emery paper and degreased with Freon. The samples cannot be welded because the hydrogen would be evolved from the metal during heating. We therefore attach the lug and sample with a C clamp. Typical contact resistances we encounter are on the order of 1 mΩ per sample. The heat transfer conditions for the two samples are equalized by the careful insulation of both the dewars and cables exiting them. We establish when the heat transfer environments are comparable in each case by monitoring the temperatures of the two samples relative to the heat sinks. Typically, before we begin a run with current applied, the temperatures of the two samples are within 0.5°C of each other, and a degree or two above the convectively cooled thermal ground.

When the current is turned on, we monitor the rise of the sample temperatures as well as the temperatures of the heat sinks. We also monitor the resistance of the samples to account for the change in resistivity as the sample rises in temperature. The temperatures of the samples exceed the sink temperature by 20 to 40°C; the exact value depends on the applied current. For the cases examined, the temperatures of the samples track each other rather closely, suggesting that the total volumetric heating \( I^2R + P_{\text{ fus}} \) is quite similar. Since no excess enthalpy production is expected in the hydrogen control sample, the small temperature difference between the two samples can be used directly to determine an upper bound on \( P_{\text{ fus}} \).

When the neutron counting has been completed, the current is turned off and the temperature is allowed to decay. This decay time can also be used to estimate the overall heat transfer coefficient.

RESULTS AND ANALYSIS

Neutron Measurements

The pulse-height spectrum from our neutron spectrometer is shown in Fig. 2 for three cases. The raw counts during an 18-h run are shown on the vertical axis, and the recoil proton energy on the horizontal axis. Because of the relatively poor statistics in these neutron counts, we do not unfold this spectrum to obtain an incident neutron spectrum. Shown in this figure are three separate conditions during which we took data.

The first condition is without any deuterated titanium present. These background counts are the average of the results of three 18-h runs. The averaging allows us to improve statistics and account for, in a simple way, the expected natural variations of the neutron background on a day-to-day basis. Data are also shown for two current levels, one at 20 A, the other at 30 A. These two currents correspond to current densities of 30 and 46 A/cm², respectively. There appears to be no statistically significant counts above background for either current level, within the error bars shown. Note that some weak structure in the proton recoil spectrum suggestive of a neutron peak exists in the 2- to 3-MeV range for each set of data. We believe this phenomenon is due to the inability of the pulse-shape discrimination to eliminate completely the background gamma counts. This is a problem

*A step in the proton recoil spectrum can indicate the presence of neutrons at an energy where the step occurs.

![Fig. 2. Proton recoil pulse-height spectrum. The dots are an average of three 18-h background runs. The triangles and squares are the spectrum obtained at current densities of 30 and 46 A/cm², respectively.](image-url)
well recognized in the neutron measurement community, and it is particularly problematic at the very low levels we are trying to measure. One possible way to correct this deficiency is to add a coincidence measurement of gamma counts using a NaI detector.

**Excess Heat Measurement**

Figure 3 shows the time dependence of the temperatures in the apparatus as the current is first turned on to 20 A, and then when it is raised to 30 A after ~20 h of operation. The temperatures of each of the samples, as well as the average temperature of the three heat sinks "ground," are shown. Note that the temperatures of the two samples track each other fairly well, and both can rise well above the temperature of the heat sinks. The exponential rise times \( \tau \) of both of the samples from the zero current condition are between 80 and 90 min. The temperatures of the heat sinks remain quite steady at their \( I = 0 \) values for the duration of the experiment.

The determination of excess rate of enthalpy production is facilitated by considering a simple bulk energy balance:

\[
M_{\text{bulk}} C_{p,\text{bulk}} \frac{dT_{\text{bulk}}}{dt} = P(t) - h_{\text{eff}} A_{\text{eff}} [T_{\text{bulk}}(t) - T_{\text{w}}].
\]

(1)

In Eq. (1), \( P \) is the total power release in the sample, and \( T_{\text{bulk}} \) is the bulk temperature of the sample, \( C_{p,\text{bulk}} \), \( h_{\text{eff}} \) and \( A_{\text{eff}} \) are the effective heat transfer coefficients and effective heat transfer area of the sample.

The term on the left-hand side of the above equation is the rate of change of the enthalpy of the sample, and the term on the right-hand side is the rate of change of the enthalpy of the sample due to the power release and the heat transfer to the environment.

By taking the ratio of the \( T_{\text{bulk}} - T_{\text{w}} \) from each sample, we can estimate the upper limit of the excess rate of enthalpy production. For the 20-A case, the excess rate of enthalpy production does not exceed 0.085 W, or 11% of the ohmic dissipation. For the 30-A case, \( P_{\text{Ohm}} \) does not exceed 0.14 W, or 8.5% of \( P_{\text{Ohm}} \). By our calibration of the circuit with titanium blanks, we know that the heat transfer coefficients of the two samples can differ by ~10%. This suggests that the "excess enthalpy" we have calculated above is probably a manifestation of the differences between the values of \( h_{\text{eff}} A_{\text{eff}} \). It is clear from these measurements that any fusion power production is far below a break-even level even at very high current densities.

We now examine what the F-P scaling of the excess enthalpy production with current density and volume would predict. Reference 1 claims that a sample subjected to a current density of 0.064 A/cm² and a volume of 0.078 cm³ has an excess volumetric heating of 1.01 W/cm³. To be conservative in our extrapolations, we ignore the claimed linear dependence on volume of the sample and focus attention on the roughly linear dependence on current density. This extrapolation results in an excess volumetric heating of almost 300 W/cm³ for the 20-A case. The sample volume is ~6.5 cm³, resulting in a total enthalpy production rate of ~3 kW. This amount of power, if present, would make the deuterated sample red hot and make thermal radiation the dominant heat loss mechanism. These high temperatures are not observed in our experiments.

**CONCLUSIONS**

Our experiments show that there is no significant neutron emission or excess enthalpy production in relatively massive samples of deuterated titanium TiD_0.9 subjected to high-current densities of up to 50 A/cm². If we assume that chemically stable TiD_0.9 is fundamentally the same as PdD_0.9, with respect to bringing deuterons close enough together within the lattice structure to cause fusion, our results suggest that current density and volume do not play a direct role in the unknown process producing excess heat reported by Fleischmann and Pons.
ACKNOWLEDGMENTS

We would like to acknowledge the kind assistance of several individuals in obtaining materials and equipment on such short notice for use in our experiment: J. E. Bowman, M. R. Chaplin, M. Malonowski, W. E. Nexsen, Jr., L. T. Summers, and J. H. Thorngate.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory and TRW, Inc.

REFERENCES


