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Electrochemical calorimetric evidence for cold fusion in the palladium–deuterium system

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Abstract

Several different types of calorimetric cell designs were used in attempts to measure excess enthalpy during the electrolysis of LiOD + D₂O using palladium cathodes. Control experiments were run by using light water in place of D₂O or by using platinum cathodes in place of palladium. Previous experiments using thin palladium wire ($d = 0.14$ cm, $A = 2.64$ cm²) gave no significant differences between the Pd/D₂O cells and the controls. The use of much thicker but shorter palladium rods (99.96%, $d = 0.635$ cm, $A = 2.64$ cm²) with constant currents of 100 mA/cm² resulted in calorimetric evidence for excess enthalpy in five out of six Pd/D₂O cells. These excess enthalpies are significant at the 99.95% confidence level. The excess rate of heating averaged 0.39 W/cm³ over a 9-day period in one experiment. The total excess enthalpies observed in several studies are difficult to explain by chemical reactions. Various possible calorimetric error sources also failed to provide satisfactory explanations for the excess enthalpies. There were no significant excess enthalpies in any Pd/H₂O cells. These experiments indicate that a tightly-coiled anode surrounding the palladium cathode is an important factor for obtaining excess heat in cold fusion experiments.

INTRODUCTION

The electrochemically induced nuclear fusion of deuterium using a palladium electrode reported by Pons, Fleischmann and Hawkins [1] has sparked a flurry of experimental measurements and considerable controversy [2–6]. The conditions under which this fusion may or may not occur will eventually be determined by many experiments at various laboratories. Enthalpy excesses that can exceed 10 W/cm³ of the palladium electrode have been claimed [1]. Similar experiments by Jones and coworkers [7] also yield evidence for cold nuclear fusion; however, the

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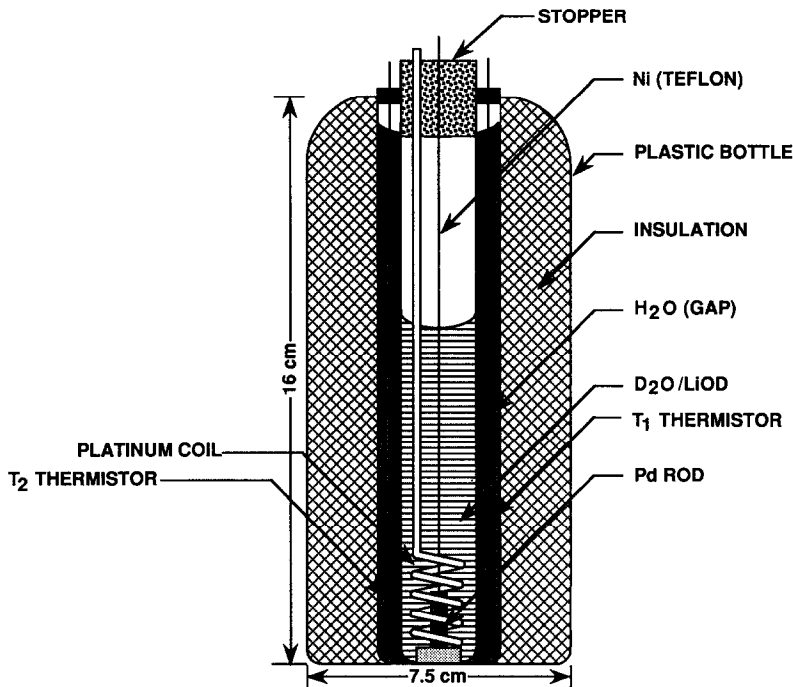


Fig. 1. Electrochemical calorimetric cell design.

fusion rates reported are far too small to be detected by calorimetry. The experiments, described below, constitute an attempt to detect any excess heat output by calorimetric studies during the electrolysis of deuterium oxide containing LiOD at palladium cathodes. Control measurements were run using light water with palladium cathodes or heavy water with platinum cathodes. Radiation levels were also monitored by various methods.

EXPERIMENTAL

The calorimetric cell design used in these experiments is shown in Fig. 1. The electrolysis cell in this configuration can be visualized as a resistive heater with the temperature being measured in the secondary compartment (gap) surrounding the electrolysis cell. The electrolysis cell initially contained 18–20 g of 0.1 M LiOD + D₂O (99.9%, Cambridge Isotope Laboratories) while the gap contained 68–70 g of distilled water. The alkaline solutions were prepared from lithium metal (ROC/RIC, 99.95%). This cell design minimized the decrease in the calorimetric cell constant with the decrease in the electrolyte solution volume which occurs during electrolysis [8]. Both the electrolysis cell and polyethylene bottle were stoppered and wrapped with parafilm to reduce evaporation and contamination. The evaporative losses from both the inner and outer glass vessels were 1% by weight per day. Make up water and heavy water were periodically added to the two compartments. After

correcting for evaporation, the measured loss of D_2O due to electrolysis was always within $\pm 1\%$ of the calculated value. The palladium rod cathode (Johnson Matthey, 99.96%, $d = 0.635$ cm, $A = 2.64$ cm²) was spot-welded to a nickel lead. Both the anode and cathode leads were covered with heat shrinkable Teflon tubing to prevent exposure of the bare metal to the gases in the headspace. Two thermistor thermometers (Cole-Parmer, Model 8502-16) calibrated within $\pm 0.01^\circ C$ were inserted into small glass tubes placed in the gap H_2O and positioned 4.6 cm (T_1, T_4) and 1.9 cm (T_2, T_5) from the bottom of the cell. The thermistor tubes were tightly secured to the electrochemical cell with rubber bands, hence the temperatures were always measured close to the cell wall. Two identical calorimetric cells containing two thermistors each as shown in Figure 1 were always run simultaneously in a constant temperature bath (B. Braun Thermomix Model 1460) set at $27.50^\circ C$. Identical coils of Pt -20% Rh (5.35 g, $d = 0.1$ cm) served as the counter electrodes. Other calorimetric cell designs were used in earlier experiments [8].

The constant current source for electrolysis was a Princeton Applied Research (PAR) potentiostat/galvanostat (Model 373) set at 264 mA (100 mA/cm²). Calorimetric cell constants were usually determined during the first day of electrolysis when no excess enthalpy is expected. Experiments using palladium cathodes in H_2O or platinum cathodes in D_2O gave nearly the same cell constants. The calorimetric cell constants were also determined by Joule heating using a 20 ohm resistor. For these calibrations, the resistor (3.51 g) replaced the palladium cathode (4.42 g) and a palladium wire coil (5.34 g) used for stirring replaced the anode coil (5.35 g).

RESULTS

The calibration of calorimetric cell B filled with D_2O by Joule heating is presented in Fig. 2. The results are linear over the power range used (0.5898–1.3797

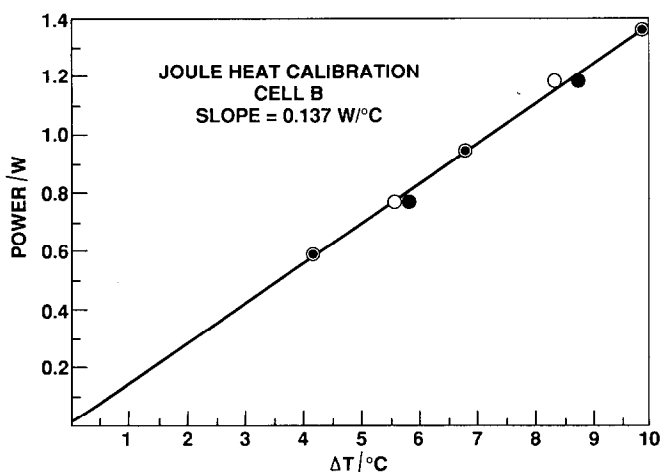


Fig. 2. Calibration of calorimetric cell B filled with D_2O using Joule heating. The thermistor positioned higher in the cell (T_4) tended to give the higher temperature readings (●).

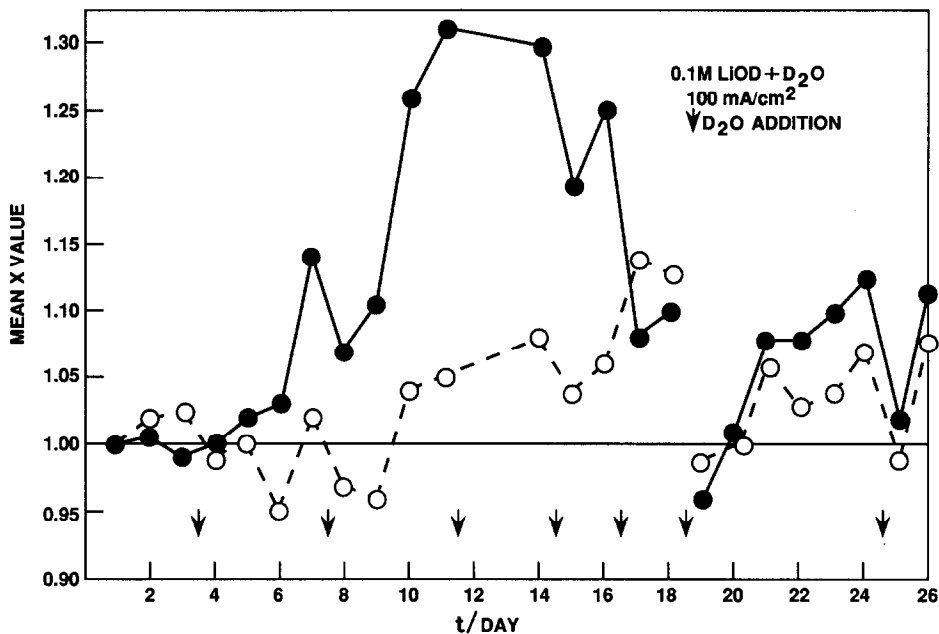


Fig. 3. First cold fusion study using palladium rod (Johnson Matthey, $d = 0.635$ cm). Cell A (— — —, T_2) and cell B (— — —, T_5).

W) with a correlation coefficient of 0.9994 ($n = 5$). Furthermore, the y -intercept is near zero (0.012 W). These results are in agreement with Newton's law of cooling and show that excellent heat recovery can be obtained with this cell design. The power range used in the Joule heat calibrations exceeds that in any electrochemical experiment. Both thermistors (T_4 , T_5) gave nearly the same temperature with cell stirring, hence only the line through the mean temperature values is shown in Fig. 2. From the slope of this line, the mean calorimetric constant is 0.137 W/°C for cell B. Similar Joule heat calibrations for cell A gave a mean calorimetric constant of 0.140 W/°C (correlation coefficient = 0.9995, $n = 5$).

Calorimetric studies using the palladium rod cathodes from Johnson Matthey in 0.1 M LiOD + D₂O are presented in Fig. 3. The equation

$$X = \frac{\text{Heat out}}{\text{Joule heat in}} = \frac{K \Delta T}{(E - E_H^\circ)I} \quad (1)$$

was used where K is the calorimetric cell constant, E is the cell voltage, E_H° is the thermal neutral potential for the cell reaction, and ΔT is the temperature difference measured within the gap by the thermistors (Fig. 1) and in the constant temperature bath ($T_{\text{bath}} = 27.50 \pm 0.02^\circ\text{C}$). Calorimetric evidence for excess enthalpy is found for cell B (Fig. 3). For the two cells run simultaneously, cell B gave a heat ratio of $\bar{X} = 1.31 \pm 0.05$ for the 11th day while the cell A gave a ratio of only $\bar{X} = 1.05 \pm 0.04$ for measurements that day. The major difference between these two cells was the tighter wrapping of the counter electrode coils about the palladium in the cell that

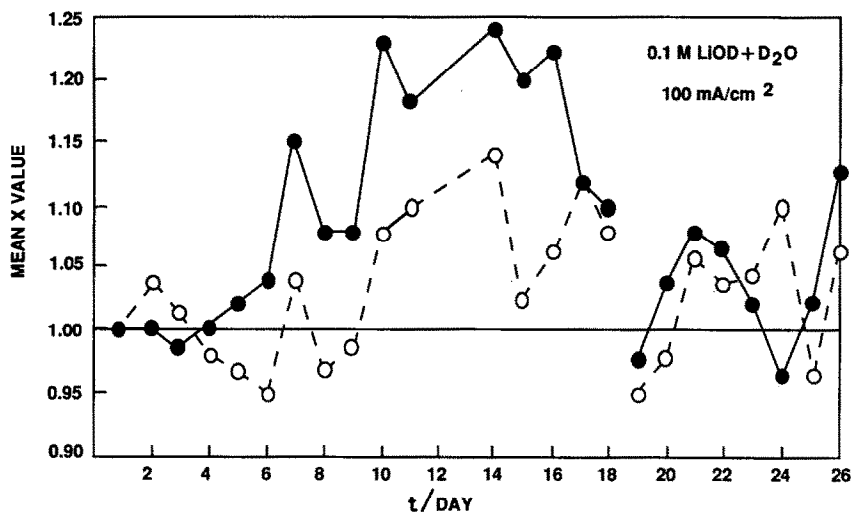


Fig. 4. First cold fusion study using palladium rod (Johnson Matthey, $d = 0.635$ cm). Cell A (---, T_1) and cell B (—, T_4).

gave the greater X values. The discontinuation of the electrolysis overnight after the 18th day showed that the X values returned to near unity. Results from the thermistors positioned higher in the gap were similar (Fig. 4), hence any thermal inversion is unlikely. The daily average for the room temperature ranged from 21.8 to 23.8°C in this study with a mean of $23.0 \pm 0.6^\circ\text{C}$.

A continuation of this cold fusion study following a 10-day shutdown is shown in Fig. 5. The excess enthalpy peaks are not as large and appear to be much more periodic with the D_2O additions than before. The additions of small amounts of ZnO to one cell and sulfur to the other cell had no measurable effects. The room temperature control was not as good in this study since the time frame (October, November) involved the onset of cooler weather. The daily average for the room temperature ranged from 19.7 to 23.4°C with a mean of $22.0 \pm 1.0^\circ\text{C}$.

Earlier experiments using thin palladium ($d = 0.14$ cm, $A = 2.64$ cm²) and platinum ($d = 0.12$ cm, $A = 2.64$ cm²) wire cathodes at the same current density (100 mA/cm²) were performed in heavy water with this same calorimetric cell design (Fig. 1). The overall mean values of $\bar{X} = 0.97 \pm 0.06$ for platinum and $\bar{X} = 1.00 \pm 0.04$ for palladium showed good heat recovery and no measurable excess enthalpy due to the cold fusion effect. Similar results were obtained with other calorimetric cell designs. Details of these earlier studies are reported elsewhere [8].

Studies using the palladium rod electrodes in cells containing H_2O rather than D_2O are shown in Fig. 6. The striking difference from the previous experiments in D_2O are the excursions of the daily mean X values into regions of less than unity. Some rather large differences between the two thermistors in cell B (solid lines) suggest that thermal inversions were occasionally occurring. At times, a temperature

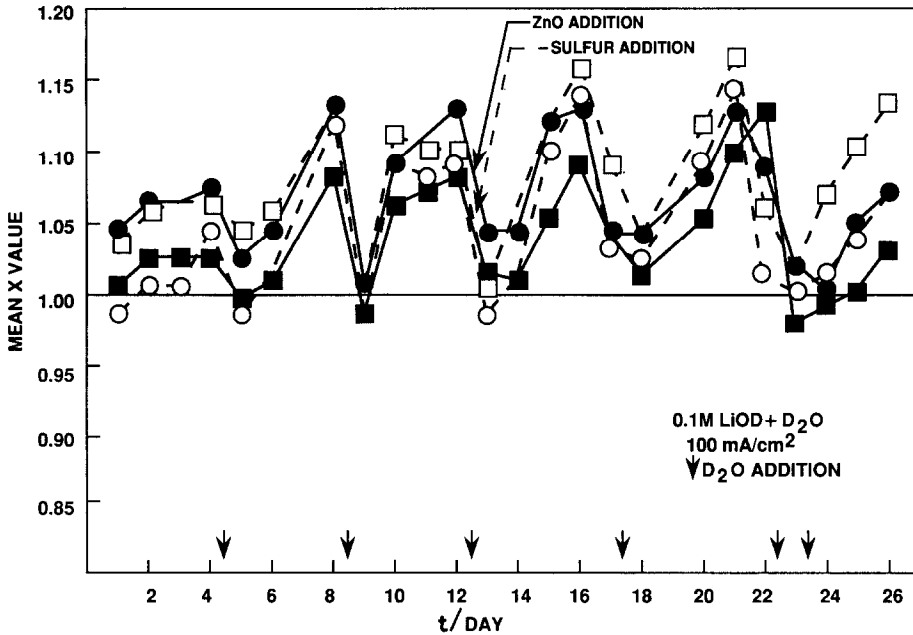


Fig. 5. Second cold fusion study using palladium rod (Johnson Matthey, $d = 0.635$ cm). Cell A (---), cell B (—), T_1 (○), T_2 (□), T_4 (●), and T_5 (■).

instability was noted that was likely due to a mixing of the air in the glass thermistor tube. A portion of the thermistor tube extended above the calorimetric cell and was subjected to cooling by the room air. In later experiments, the thermistor tubes were made flush with the cell top resulting in more uniform temperatures and X values within the same cell. All H_2O measurements were made within an average daily room temperature range of 19.9 to $22.8^\circ C$ with a mean of $21.5 \pm 0.7^\circ C$. Only a weak relationship between the daily X values and the room temperature could be established in this study with a slope less than $0.02^\circ C^{-1}$ and a correlation coefficient less than 0.3.

A third cold fusion study using the same palladium rod cathodes in fresh $LiOD + D_2O$ solutions is shown in Fig. 7. Excellent agreement between the two thermistors in each cell was realized in this study where the thermistor tubes were flush with the cell top. This experiment shows a nearly consistent excess enthalpy production with only a few days yielding near unity for the heat ratio. However the daily mean X values are noticeably less than those for the first study with a freshly prepared palladium cathode (Figs. 3 and 4). The daily average room temperature ranged from 22.4 – $24.5^\circ C$ in this study with a mean of $23.4 \pm 0.5^\circ C$.

Tritium measurements on the $LiOD + D_2O$ solutions following the first two experiments using the palladium rods were negative. The two $LiOD + D_2O$ solutions gave 22.99 ± 2.63 cpm and 22.18 ± 1.06 cpm versus 19.34 ± 0.57 cpm for a

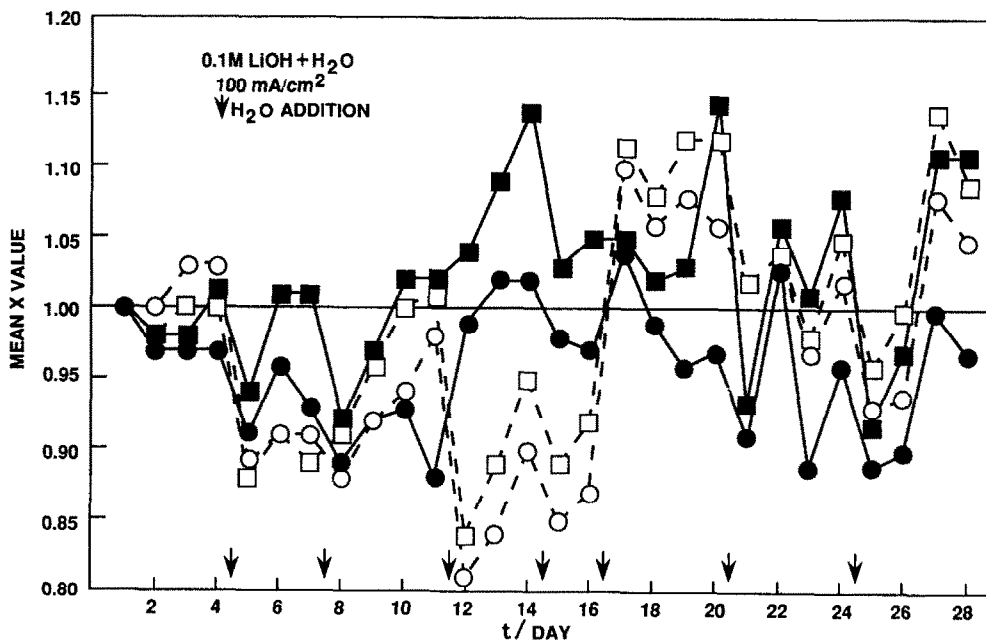


Fig. 6. Studies in water using palladium rod (Johnson Matthey, $d = 0.635$ cm). Symbols for cell A, cell B, T_1 , T_2 , T_4 , and T_5 same as in Fig. 5.

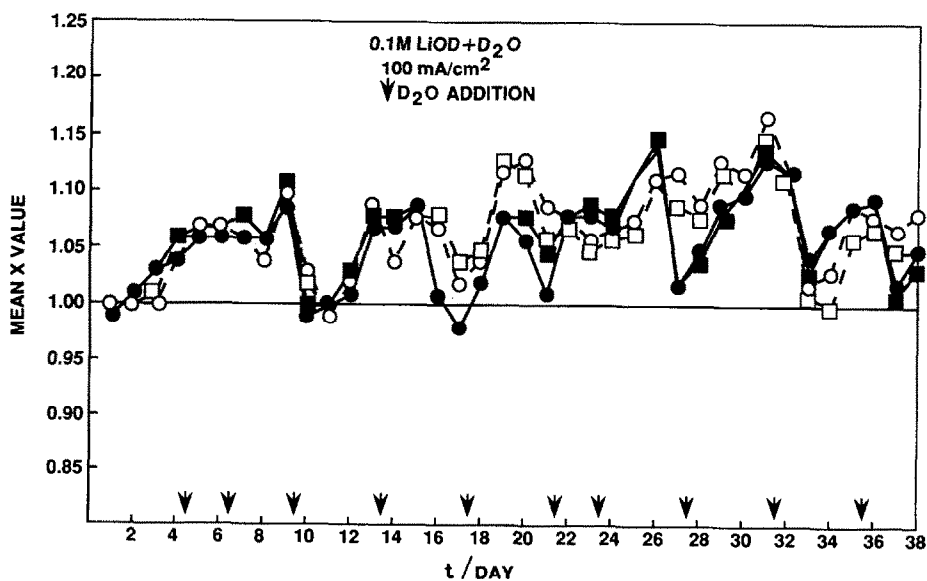


Fig. 7. Third cold fusion study using palladium rod (Johnson Matthey, $d = 0.635$ cm). Symbols for cell A, cell B, T_1 , T_2 , T_4 , and T_5 same as in Fig. 5.

LiOD + D₂O sample that was never subjected to electrolysis. Results from a second laboratory gave similar conclusions. Theoretical calculations based on the total observed excess enthalpy (563 kJ) yield 4.4×10^7 T atoms in ml⁻¹ when the 10⁻⁹ disparity factor between excess heat and tritium production is used [1]. Assuming no loss of tritium to the gas phase and a 100% counting efficiency, this amount of tritium would yield only 4.7 cpm, hence, the tritium produced would have been difficult to detect from the background.

DISCUSSION

Interpolations of the results reported by Pons, Fleischmann, and Hawkins [1] yield an expected heat ratio of $X = 1.95$ for our palladium rod at 100 mA/cm². Such a high X value would unquestionably be well outside the range of any calorimetric errors. Statistical tests must be applied to our smaller X values to determine any significant difference from unity. Nevertheless, the comment [5] that it is most unlikely that the excess thermal energy due to a fusion process would be of the same order of magnitude as the electrical energy input is perplexing. An order of magnitude larger fusion effect involving D₂O electrolysis with a palladium cathode, however, would probably have been discovered years ago. Significantly smaller fusion effects would not be detectable by calorimetric experiments.

The overall mean heat ratios obtained from the various experiments using the palladium rod cathodes as well as the 99% confidence intervals are presented in Table 1. The first five days of data were omitted to allow the palladium to become charged with deuterium (n is the number of days of data considered). The excess enthalpy for cell B (\bar{X}_4, \bar{X}_5) was significant at the 99% confidence level in all three D₂O experiments. For cell A (\bar{X}_1, \bar{X}_2), the excess enthalpy was significant at the 99% level in all except the first D₂O experiment that had the widely spaced counter electrode coils. The experiment in H₂O yields mean heat ratios close to unity for both cells A and B. The overall mean heat ratios and confidence intervals using all data yields similar conclusions. The main difference is that the mean X values are

TABLE 1

Mean heat ratios and confidence intervals after 5 days of charging. $\mu - \bar{X} = \pm 2.58 \sigma/\sqrt{n}$ (99%) (μ is the true population mean)

Experiment	\bar{X}_1	\bar{X}_2	\bar{X}_4	\bar{X}_5	Sample size/days
Pd rod + D ₂ O (I)	1.04 ± 0.05	1.04 ± 0.05	1.145 ^a ± 0.06	1.17 ^a ± 0.08	$n = 11$
Pd rod + D ₂ O (II)	1.06 ^a ± 0.03	1.10 ^a ± 0.03	1.08 ^a ± 0.03	1.05 ^a ± 0.03	$n = 19$
Pd rod + H ₂ O	0.96 ± 0.05	0.995 ± 0.05	0.96 ± 0.03	1.03 ± 0.03	$n = 23$
Pd rod + D ₂ O (III)	1.08 ^a ± 0.02	1.07 ^a ± 0.02	1.06 ^a ± 0.02	1.07 ^a ± 0.02	$n = 33$

^a Excess enthalpy (99% confidence level).

Table 2

Single-tail t tests for excess enthalpy using data after 5 days of charging. $t = (\bar{X} - \mu) \sqrt{n/s}$ where s is the sample standard deviation and $\mu = 1.00$ (no excess enthalpy)

Experiment	t_1	t_2	t_4	t_5	t (99.95%)	Sample size/days
Pd rod/D ₂ O (I)	2.30	2.10	6.64 ^a	5.56 ^a	4.59	$n = 11$
Pd rod/D ₂ O (II)	5.27 ^a	8.27 ^a	7.84 ^a	4.93 ^a	3.92	$n = 19$
Pd rod/H ₂ O	-2.23	-0.28	-4.21	2.25	3.79	$n = 23$
Pd rod/D ₂ O(III)	10.94 ^a	10.25 ^a	8.55 ^a	9.01 ^a	3.65	$n = 33$

^a Excess enthalpy (99.95% confidence level).

generally somewhat smaller for the Pd/D₂O experiments when the first five days are included.

The t -test can also be used to determine if there is a significant difference from unity for the various mean heat ratios in Table 1. Results for the t -test at the 99.95% confidence level are given in Table 2. The same Pd/D₂O cells as before show a significant positive difference from unity at the 99.95% confidence level. Although the third cold fusion experiment (Fig. 7) did not yield the highest heat ratios, the length of this study ($n = 33$ days) along with its small mean standard deviation ($s = \pm 0.02$) yielded exceptionally large t values.

Both cell A and cell B in the Pd/H₂O experiments failed to show any significant positive differences from unity. The value for t_4 (-4.21), however, is a significant negative difference at the 99.95% confidence level. The lower thermistor (T_5) in this same cell (cell B) yields $t_5 = 2.25$, and the average of these two t -values (-0.98) fails the significance test even at a lower 90% confidence level. This difference in t -values within the same cell is further evidence that thermal inversions occurred for cell B in the Pd/H₂O experiments as noted in previous comments concerning Fig. 6.

Tables 1 and 2 provide substantial statistical evidence for excess enthalpy for 5 out of 6 experiments involving the palladium rod in heavy water. It is interesting and instructive to consider the total excess enthalpy produced by the various Pd rod/D₂O cells. For example, using the data from either Fig. 3 or 4, an average of at least 18% excess enthalpy is observed for a 9-day period (days 9-18). This corresponds to an average excess power of 0.14 W (0.39 W/cm³) and a total excess enthalpy of 110 kJ. It can be shown that the complete combustion of the palladium electrode in the form of Pd₂H to yield PdO and H₂O would give an excess enthalpy of only 6.2 kJ. The recombination of D₂ and O₂ to yield 110 kJ of enthalpy would require the formation of 0.37 moles (7.4 g) of D₂O within the cell, yet normal amounts of make-up D₂O were required during this 9-day period. Hence, it is difficult to explain the 110 kJ of excess enthalpy by chemical reactions. Similar calculations based on data in Table 1 yields total excess enthalpies of 213 kJ for cell B in Exp. I, 217 kJ for cell A in Exp. II, and 301 kJ for cell A in Exp. III. No excess enthalpies were measured in any H₂O experiment (Tables 1 and 2).

Our cold fusion studies are unique in that mostly negative results for excess enthalpies were obtained in early experiments that extended over a 5-month period

TABLE 3

Calorimetric cell constants determined in various experiments using $K_{\text{cell}} = (E - E_{\text{H}}^0)I/\Delta T$ where $E_{\text{H}}^0 = 1.53$ V for D_2O and $E_{\text{H}}^0 = 1.48$ V for H_2O . For Joule heating experiments, the calorimetric cell constant was determined from the slope of power vs. ΔT (see Fig. 1)

Experiment	$K_1/\text{W } ^\circ\text{C}^{-1}$	$K_2/\text{W } ^\circ\text{C}^{-1}$	$K_4/\text{W } ^\circ\text{C}^{-1}$	$K_5/\text{W } ^\circ\text{C}^{-1}$
Pd rod/ D_2O (I)	0.141	0.145	0.133	0.132
Pd rod/ H_2O	0.135	0.138	0.137	0.134
Pd rod/ D_2O (III)	0.139	0.143	0.133	0.134
Joule heating (D_2O)	0.136	0.144	0.136	0.138
Mean	0.138 ± 0.003	0.143 ± 0.003	0.135 ± 0.002	0.135 ± 0.003

using various Pd-wire cathodes [8], yet mostly positive results were later obtained with the Pd-rod cathodes (Figs. 3–5, 7). The earlier negative experiments involving the longer Pd-wire cathodes had widely-spaced counter electrode coils while all of the experiments that employed compact counter electrode coils about the shorter Pd-rod cathodes gave positive results. The use of a Pd-rod cathode surrounded by a tightly-coiled and symmetrically-arranged counter electrode may charge the palladium in a more uniform manner. In order to prevent the diffusion of deuterium atoms up through the Pd rod and out into the headspace due to the voltage-induced high fugacity [1], the Pd rod should be spot-welded to a different metal lead wire and be kept completely submerged in the heavy water. Differences in impurity levels or in metallurgical methods of preparation are also possible factors for the striking differences in results for the Pd-wire and Pd-rod cathodes.

Table 3 shows that the differences in the calorimetric cell constants measured in various electrolysis experiments were not a significant factor. The cell constants for cell A (K_1, K_2) tended to be slightly larger than for cell B (K_4, K_5). The cell constants determined in H_2O were very close to those determined in D_2O . Joule heating experiments gave good agreement with calorimetric cell constants determined under electrolysis conditions. In agreement with these experimental results, theoretical calculations [9] indicate that enthalpy changes due to the charging of palladium with hydrogen (deuterium) and the evaporation of H_2O (D_2O) aided by the gas evolution reactions (0.2 ml/day) yield a net effect of less than 2% on the calorimetric cell constant.

Possible calorimetric error sources for our cell design (Fig. 1) include room temperature fluctuations, the exposure of the glass thermistor tubes to the room air, the level of H_2O in the gap, the level of the D_2O in the electrolysis cell, thermal inversions in the gap or thermistor tubes, hydrogen/oxygen recombination within the cell, and deviations from Newton's law of cooling. Many of these error sources are small, and they all tend to cancel since cell constant determinations were made under nearly the same conditions. Furthermore, these error sources should affect the Pd/ H_2O experiments in the same manner, yet mean heat ratios close to unity were obtained in the H_2O studies. The H_2O experiments were conducted with every detail as close as possible to the previous D_2O studies (Figs. 3–5). The level of the

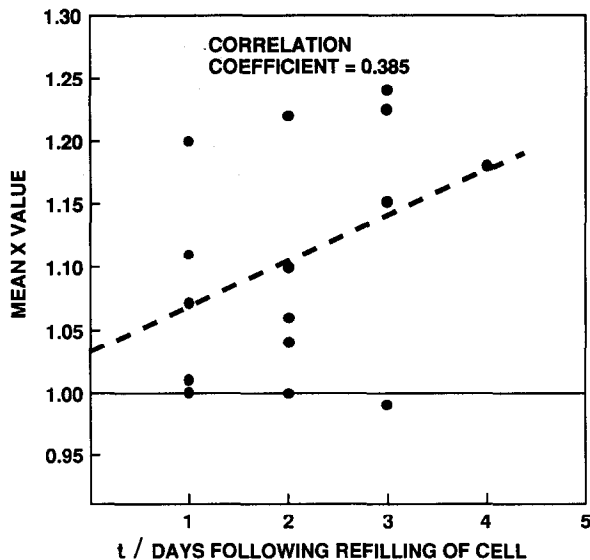


Fig. 8. Test for correlation between the mean X value and the time period between the refilling of cell B in Fig. 4 (solid line, T_4).

gap H_2O must be carefully controlled in our cell design. Experiments where measured amounts of the gap H_2O up to 10 ml were withdrawn showed nearly a 2% increase in X per ml of H_2O removed. Joule heating experiments also showed a 2% increase in ΔT per ml of H_2O removed from the gap. Although the loss due to evaporation was small (and could be entirely eliminated), the effect of the gradual loss of gap H_2O often became experimentally measurable after several days and likely contributed to the periodic trends of Fig. 5. The effect of the D_2O level was much less important for our cell design, hence no correction for this was needed. Joule heating studies gave no measurable change in ΔT when 4 ml of D_2O was removed from the cell. There was never any evidence for deuterium/oxygen recombination within the cell.

Possible correlations between the mean X value and the time period between the refilling of the cell and gap were investigated. A typical result for the Pd-rod cathode in D_2O is shown in Fig. 8. The addition of D_2O to the cell and H_2O to the gap was always on the same day in this experiment (cell B, Fig. 4). Any real correlation between the mean X value and the refilling of the cell and gap is not obvious. For example, the third day following refilling yields both the highest and lowest mean X values. Correlation coefficients for other experiments ranged from -0.023 for H_2O (Fig. 6, X_4) to 0.735 for Fig. 5 (X_4) where a periodic trend was already noted. In general, correlations were rather weak or absent. Similar results were obtained in investigating possible correlations between the mean X value and the cell voltage since there is nearly a linear relationship between the cell voltage and the electrolysis time following refilling of the cell (slope ≈ -0.3 V/day). The

voltage was generally in the range of 5 to 6 V after the cell was refilled. No correlation between the mean X value and the cell voltage could be established.

Deviations from Newton's law of cooling have been recently discussed as a possible calorimetric error source [10]. Although this concern applies mainly to Dewar calorimeters where heat transfer occurs mostly by radiation, it is interesting to see how such errors would affect experimental X values in any type of calorimeter. If heat transfer from the calorimetric cell occurs only by radiation (Q_R) and conduction (Q_C), then

$$Q_R + Q_C = K_R(T_i^4 - T_o^4) + K_C(T_i - T_o) \quad (2)$$

The substitution $T_i = T_o + \Delta T$ yields

$$Q_R + Q_C = [K_R^0(1 + \Delta) + K_C] \Delta T \quad (3)$$

where $K_R^0 = 4K_RT_o^3$ and $\Delta = 3\Delta T/2T_o + (\Delta T)^2/T_o^2 + (\Delta T)^3/4T_o^3$. Thus the ratio of heat in to heat out becomes

$$X = \frac{[K_R^0(1 + \Delta) + K_C] \Delta T}{(E - E_H^0)I} \quad (4)$$

rather than eqn. (1). If there is Joule heating only, then $X = 1.00$, $\Delta T = \Delta T_J$, and $\Delta = \Delta_J$. If there is a fusion contribution to the heat, then $X > 1.00$, $\Delta T = \Delta T_J + \Delta T_F$, and $\Delta = \Delta_F$. Thus

$$X = \left(1 + \frac{\Delta T_F}{\Delta T_J}\right) \left[\frac{(1 + \Delta_F) + K_C/K_R^0}{(1 + \Delta_J) + K_C/K_R^0}\right] \quad (5)$$

If the heat transfer occurs mainly by conduction as in our calorimeter, then $K_C \gg K_R^0$, hence

$$X = 1 + \frac{\Delta T_F}{\Delta T_J} = X_N \quad (6)$$

which corresponds to Newton's law of cooling. If the heat transfer occurs mainly by radiation, then $K_C \ll K_R^0$, hence

$$X = \left(1 + \frac{\Delta T_F}{\Delta T_J}\right) \left(\frac{1 + \Delta_F}{1 + \Delta_J}\right) \quad (7)$$

Since $\Delta_F > \Delta_J$, larger X values would be obtained using eqn. (7) rather than Newton's cooling law (eqn. 6). For example, assuming $\Delta T_J = 10$ K, $\Delta T_F = 2$ K, and $T_o = 300$ K yields $X_N = 1.20$ from Newton's law and $X = 1.21$ from eqn. (7). Hence calibrating a Dewar type cell under conditions of Joule heating only and measuring a fusion heat contribution would yield somewhat higher X values if the more exact expression (eqn. 7) is used rather than Newton's cooling law.

The exposure to neutrons and other harmful radiation can be a matter of concern in cold fusion experiments. Theoretical calculations of neutron flux levels and dose rates were performed for our cell design (Fig. 1). Details of these calculations are

reported elsewhere [11]. Assuming a source of 10^4 neutrons s^{-1} from the palladium, the calculation dose rate was 0.28 mrad h^{-1} at the outer surface of the bottle ($1 \text{ rad} = 0.01 \text{ J/kg}$). These calculations suggest safe dose rates for people near the cold fusion experiment when the source level is 10^4 neutrons s^{-1} . These theoretical calculations also show that the flux of thermal neutrons would not be sufficient to activate gold or copper metal placed at the surface of the bottle [11]. Gold and copper metal samples were always kept near the cold fusion experiments, but radiation measurements gave essentially only background levels. Neutron monitoring for safety concerns carried out with a Ludlum Model 15 counter never gave any response significantly above the background levels.

The upper limit for transmutations or isotopic changes in metals due to an electrochemical fusion process that produces neutrons can be illustrated by a simple calculation. Assuming an excess power of 1 W/cm^3 resulting from the fusion reaction



yields 1.91×10^{12} neutrons $\text{cm}^{-3} \text{ s}^{-1}$. If all of these neutrons were absorbed by the palladium electrode, then the time required to convert 1% of the palladium to other isotopes is given by

$$t = \frac{(6.79 \times 10^{22} \text{ Pd atoms cm}^{-3})(0.01)}{1.91 \times 10^{12} \text{ neutrons cm}^{-3} \text{ s}^{-1}} = 3.55 \times 10^8 \text{ s} \quad (9)$$

which translates into 11 years. Considering the relatively short duration of most electrochemical fusion experiments and the low thermal neutron capture cross-section for the various palladium isotopes, any transmutation or change in the isotopic composition of the palladium electrode would be difficult to detect. The rate of ${}^3\text{He}$ production would be equally small and difficult to detect, especially considering its possible escape through cracks and fissures.

CONCLUSIONS

Heat ratios greater than unity can be measured in calorimetric experiments for Pd rod/ D_2O cells that are significant at the 99.95% confidence level. However, these heat ratios are small in many experiments. Nevertheless, the total excess enthalpies are difficult to explain by chemical reactions. There were no significant excess enthalpies in Pd rod/ H_2O cells. A tightly-coiled anode surrounding the Pd cathode is possibly an important factor for obtaining excess heat in cold fusion experiments.

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