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Experimental Investigation of Cold Fusion Phenomena in Palladium

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Abstract: We conducted at the Paul Scherrer Institute a series of Pd-D_2O electrolysis experiments to investigate the existence of the «cold fusion» phenomenon. In a D_2O-test cell and a H_2O-reference cell, a search for excess heat was made, using a closed-circuit calorimetry set-up, and running currents onto 1 mm and 2 mm palladium cathode wires from 120 to 545 mA/cm^2. Simultaneously, with a NE213 neutron and a BGO gamma detector, we searched for the occurrence of excess neutrons or gammas that might arise from nuclear fusion processes. Our results are negative, i.e. we detected no excess heat within less than 0.1 watt (expectation of *Pons* and *Fleischmann* 1 watt) and we found no nuclear radiation in excess to the background (level of neutron sensitivity < 0.14 n/s or $< 10^{-13}$ W and 5.5 MeV-γ sensitivity < 0.1 γ/s). Mass spectrometric analysis of ^{3}He and ^{4}He in the Pd wire showed no ^{3}He in excess to the background expected from the decay of tritium impurities, contained in the D_2O already prior to our runs. Upper limits of ^{4}He are more than 6 orders of magnitude lower than expected for a neutron free fusion rate as reported by *Fleischmann* and *Pons*.

1. Introduction

Cold fusion of hydrogen isotopes is a fascinating alternative to hot fusion, since the high temperatures (on the order of 100 million degrees) needed for plasma ignition would be avoided. The principle of cold fusion is to overcome the Coulomb potential barrier by shielding the positive charges of protons with negative particles, so that hydrogen nuclei can approach each other close enough for fusion to occur via tunneling. Indeed, cold fusion works very well with negative muons, known as «Muon Catalyzed Fusion» (μCF)[1]. This type of cold fusion was originally suggested in 1947 by *Frank*[2] and discovered in 1956 by *Alvarez* et al.[3]. In the last decade, extensive μCF research has been carried out especially at the Paul Scherrer Institute (PSI) and at Los Alamos[1]. In deuterium/tritium mixtures fusion yields exceeding 100 per muon were observed[4]. This achievement allows already to consider hybrid systems using μCF for energy production[5], however, the present yields are still about one order of magnitude too low for a direct μCF energy cycle[6].

For ordinary electronic deuterium or tritium molecules, it is well known that fusion does *not* occur at an observable rate, since the nuclear distance of ≈ 0.74 Å is too large. Already in 1960, *Zeldovich* and *Gerstein* pointed out in a review paper[7], that reducing that distance by a factor of 5 to 10 would be sufficient to find a significant fusion rate. They also noted that this cannot be achieved in chemical compounds. The shielding provided by a particle with mass five times the mass of an electron increases the charge density at the hydrogen nucleus by a factor of approximately 100. Clearly, such a condition would have to be met, if ever electrochemical cold fusion could become energy productive. Alternative ways, as hypothetical acceleration mechanisms, may also lead to observable fusion processes (neutron bursts?), but these rather belong to «hot accelerator fusion», having an efficiency of $\lesssim 10^{-5}$[8].

Recently, there has been substantial research activity involving heavy water electrolysis at palladium cathodes. Interest in this area was generated by *Fleischmann* and *Pons*[9], who reported that they were able to detect the process of nuclear d + d fusion calorimetrically (several watts of heat), and *Jones* et al.[10], who reported that they detected this fusion reaction by recording neutron spectra. A number of laboratories attempted to duplicate these results; most of their reports could not confirm the initial findings[11]. Some early investigations did report the existence of «excess» heat[12], while other groups indicated that they were able to detect extra neutrons[13-15], though – in terms of energy release – at many orders of magnitude lower levels. A number of theories were proposed to explain the increased probability of cold fusion in palladium or titanium[11]. In a former Chimia edition[16] *Augustynski* discussed some of the difficulties that might arise if these experiments were duplicated, and *Jørgensen* discussed some theoretical aspects. It should also be mentioned, that already in 1926 there was a claim (later retracted) by *Paneth* et al.[17] of detecting helium products from d + d fusion with D_2O electrolysis on palladium electrodes.

At Paul Scherrer Institute (PSI) active research programs in electrochemical en-

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ergy storage and μCF are carried out. An interdisciplinary program was therefore initiated in order to verify the cold fusion results with palladium electrodes in heavy water. A D_2O cell and an identical H_2O cell, for reference, were studied with closed-circuit calorimetry. Simultaneously, well-shielded neutron and gamma counters were positioned near the cells to detect any nuclear radiation from cold fusion. After the runs the electrodes were analyzed for the presence of helium and tritium fusion products.

1.1. Unusual Properties of Palladium

In 1869 *Graham* [18] found that palladium could be reversibly hydrided at room temperature. Palladium is an exceptional hydrogen storage medium; as a hydrogen electrode in a rechargeable battery, it has a theoretical energy density of 250 Ah/kg. Unfortunately, the price of palladium makes palladium hydride batteries uneconomical.

The mobility of hydrogen and deuterium incorporated in palladium is unusually high. The diffusion coefficient for hydrogen und deuterium is about $1-5 \times 10^{-7}$ cm^2/s at room temperature and increases by a factor of 200 at 400°C. These diffusivities are 5 orders of magnitude larger than those observed in other metals such as Ni. Palladium and Pd/Ag alloys are used as hydrogen diffusion membranes that are utilized in the separation of hydrogen from other gases.

Superconductivity of palladium hydrides was first reported by *Skoskiewicz* et al. [19] for $H/Pd > 0.8$ with $T_c = 8.8$ K and 10.7 K (D/Pd), respectively. The high ratios H/Pd and D/Pd (close to 1) were achieved by electrolysis at 193 K [20] or with high pressure (4–5 kbar) at 77 K [16a,21].

Hydrogen acts as an electron donor in palladium, filling first the Pd 4d band. At $H/Pd > 0.6$ the 5s band is filled, which is accompanied by an increase of the Fermi energy level [22]. Not much is known about deuterium interactions in the palladium lattice. Some theories [23] show that the lattice could enhance fusion rates, but the small enhancements are not detectable with standard methods.

Hydrides formed with the elements Ti, Nb, Fe, Ni, etc. [24] are also of some interest in connection with cold fusion. Some important properties of H, D, Li, and Ag alloys of palladium were already discussed by *Jørgensen* recently [16b].

1.2. Cold Fusion Reactions of Interest and their Detection

Table 1 shows the most probable fusion scenarios that «might» be occurring in palladium.

There were some speculations about a strongly enhanced d + d fusion reaction leading to ^{4}He + γ(24 MeV) and some suggested that instead of the energy being radiated as a gamma ray it is released directly to the surrounding lattice as heat. However, there are no theoretical or experimental grounds why the regular d + d fusion channels should be suppressed fully and how such a big electromagnetic energy transfer of 24 MeV to the lattice might occur.

Table 1. List of possible fusion reactions that might arise in Pd electrolysis experiments involving hydrogen isotopes.

p + d	$\rightarrow$	^{3}He + γ(5.50 MeV)	
d + d	$\rightarrow$	^{3}He(0.82 MeV) + n(2.45 MeV)	$\approx 50\%$
	$\searrow$	t(1.01 MeV) + p(3.02 MeV)	$\approx 50\%$
	$\searrow$	^{4}He + γ(24 MeV)	$\approx 10^{-4}\%$
d + t	$\rightarrow$	^{4}He(3.50 MeV) + n(14.1 MeV)	
p + t	$\rightarrow$	^{4}He + γ(19.8 MeV)	
t + t	$\rightarrow$	^{4}He + 2 n(11.3 MeV)	
p + p	$\rightarrow$	(only weak interaction) $d + e + \bar{\nu}_e$	
d + ^{6}Li	$\rightarrow$	^{4}He + ^{4}He + 22.4 MeV	
	$\searrow$	n + Be + 3.4 MeV	
	$\searrow$	(additional channels)	

From the nuclear physics point of view the general statement can be made that there must always be nuclear neutron or γ radiation and that the ^{3}He and ^{4}He end products are always present in nuclear fusion processes as shown in Table 1. Unless these reaction products can be detected there is no sufficient proof for the existence of cold fusion.

Table 1 also provides information about the energies released from the fusion reactions. Neutrons and gammas will not be contained in the cell and should be detectable outside the cell. The charged reaction products (p, t, ^{3}He, ^{4}He, and Be) will cause local heating to the palladium and adjacent electrolyte which can be detected by in situ calorimetry. The helium products can be identified by analysis of the palladium cathode with mass spectrometry since they will mostly remain inside the electrode.

1.3. Electrochemical Considerations

Fleischmann and *Pons* [9] measured the largest «excess heat» with the largest volume sample, suggesting that the fusion reactions occur in the bulk palladium. *Jones* et al. [10] indicated that high surface area palladium or titanium are required for the fusion process, as expected for a surface reaction. An increase in the hydrogen concentration in the lattice would have the greatest effect on bulk reactions. Electrolyte additives or palladium surface treatments can be used to increase the overpotential of the electrode and thereby increase the bulk concentration of hydrogen. High palladium overpotentials can cause lithium alloys to form on the electrode surface [16a], but these films should not interfere with bulk processes as long as diffusion in the lattice is not prevented.

2. Experimental Arrangement

2.1. Cell Materials and Design

The schematic arrangement of the electrolysis experiment and calorimetric measurement is shown in Fig. 1. The cell interior consisted of a cylindrical container of machined Teflon (inner diameter 64 mm, height 70 mm) with an electrolyte volume and head space of approximately 150 and 75 mL, respectively. The anode and cathode were positioned concentrically in this container as described below and they were held in place by vertical Teflon rods supported at the top and bottom of the cell.

Table 2 summarizes the electrode configurations and operating conditions for the runs that were conducted. Two different cathode geometries were used. For the

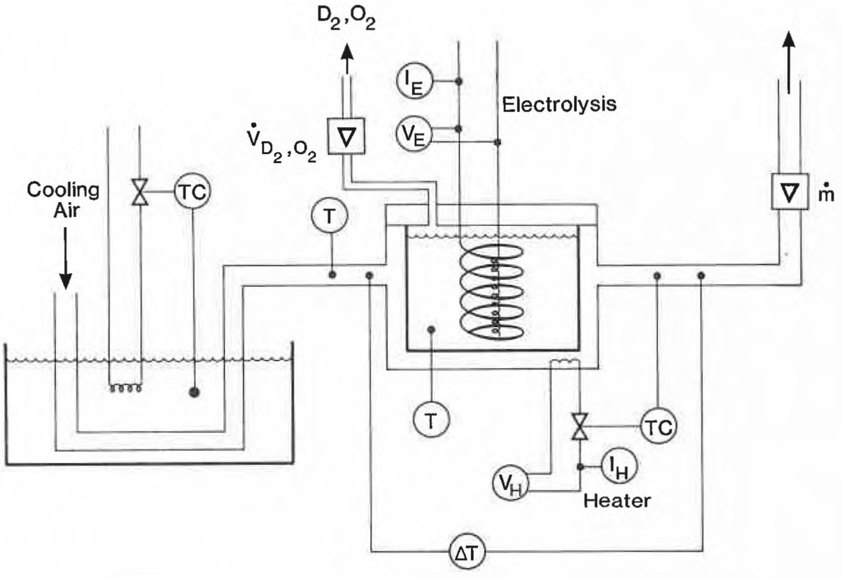

Fig. 1. Equipment arrangement and instrumentation for calorimetry. Two identical cells were used with the same cooling gas stream connected in series. Symbols see text. TC: temperature regulation; I_E, V_E: electrolysis power measurement; I_H, V_H: heating power measurement; $\dot{m}$: cooling gas flow measurement; $\dot{V}_{D_2,O_2}$: electrolysis gas flow measurement; T: temperature sensor; ΔT: NCT differential temperature sensor.

Table 2. Summary of conditions for the cold fusion verification attempts.

Run	Dates (1989)	Cathode Length, Diameter	Anode Length, Diameter	Cathodic Current Density [mA/cm^2]	Electrolyte
A	April 23–May 8	Pd drawn wire 40 cm, 1 mm Pd	Pd drawn wire 80 cm, 1 mm Pd	120–135	Ref.: 0.1 M LiOH Test: 0.16 M LiOD
B	May 8–May 12	Pd drawn wire 40 cm, 1 mm Pd	Pt drawn wire 80 cm, 1 mm Pt	120	Ref.: 0.1 M LiOH Test: 95% (vol.) 0.16 M LiOD 5% (vol.) 0.1 M LiOH
C	May 12–June 5	Pd drawn wire 35 mm, 2 mm Pd	Pt drawn wire 80 cm, 1 mm Pt	545	Ref.: 0.1 M LiOH Test: 0.16 M LiOD
D	June 13–June 29	Pd cast bar 35 × 2 × 2 mm Pd	Pt drawn wire 80 cm, 1 mm Pt	500	Ref.: 0.1 M LiOH Test: 0.16 M LiOD

experiments of runs A and B, 40 cm of 1 mm diameter Pd wire was wrapped around a central Teflon post with a diameter of 5 mm. In experiments C and D, 3.5 cm lengths of a 2 mm diameter Pd rod and a 2 mm square Pd cast bar, respectively, were positioned vertically in the center of the cell. The anode was an 80 cm long piece of 1 mm diameter Pt wire (Pd in experiments A, B) which was spiraled and positioned around the central cathode. The helix diameter was 25 mm. The platinum (99.95%) and 2 mm palladium (99.95%) were obtained from Metaux Precieux, and the 1 mm palladium (99.95%) from Balzers. For experiment D, the cathode was cut by electro-erosion from a Pd ingot which was cast in a arc furnace under argon from 1 mm palladium wire from Brandenberger (99.95%).

Two cells with identical geometry were run simultaneously, a reference (control) cell with H_2O and a test cell with the D_2O based electrolyte. Electrolytes of 0.10 M LiOH (reference) and 0.16 M LiOD (test) were chosen in order to match the electrolyte conductivities at 17 mΩ/cm. The D_2O and H_2O electrolytes were made from 99.7% D_2O and twice distilled water, respectively, and Li was added as a metal powder (99.95%, Strem Chemicals) to the desired concentrations. The potential difference between the cathode and anode was in the range of 8–12 V. From the cell geometry, the conductivity of the electrolyte, and from a measurement with a reference electrode, we conclude that at a current of 1.4 A we had a voltage drop in the bulk electrolyte of about 6 V, whereas the overpotentials at the cathode were 1 V, at the anode 2.5 V. Analysis by thermal ionization mass spectrometry showed that ^{6}Li and ^{7}Li were present in approximately their natural abundance.

The cell container served as the insulating liner in a stainless steel cylindrical container. The cell was made air-tight by means of an O-ring with a stainless steel flange fitted with isolated tabs for external electrical connection and openings for electrolyte insertion and electrolysis gas exit.

2.2. Calorimetry

Instrumentation: The calorimetry system was designed so as to achieve a steady-state measurement of the heat produced within the cell. A gas stream flowing through a cooling jacket on each cell was used for heat removal, and the heat produced was determined by the gas temperature difference across the jacket. This arrangement eliminated the need for efficient stirring of the electrolyte.

The two cells were instrumented as shown in Fig. 1. Compressed air, used as the cooling medium, was passed through copper coils in a constant temperature bath (about 17°C) before reaching the cooling jacket on the cell. The outlet temperature of the cooling gas was maintained constant (about 25°C) by adjusting the electrical input to a 5 W heater that was wrapped around the outside of the stainless steel can containing the Teflon cell. This heater allowed operation with a constant cooling load and provided an easy means for calibrations when electrolysis was shut off. Calibrations with a heater inside the cell were also accomplished.

The reference and test cells were run in series with respect to the cooling gas flow. Specifically, the cooling gas was first run through the 17°C thermostat, then through the reference cell, back through another set of copper coils in the thermostat, then through the test cell, and finally to a precision rotameter for the flow measurement. This series arrangement allowed compensation for uncertainty in the gas flow by subtracting the calculated excess heat production of the test cell from that of the reference cell.

Energy balance calculations: The joule heating rate, $\dot{Q}_j$ (W), is the part of the electrolysis power that heats the cell contents. It is given by

$$\dot{Q}_j = I_E \left(V_E - (1 - f_R)\,\frac{\Delta H}{nF} \right) \tag{1}$$

where I_E and V_E are the electrolysis current and voltage, respectively, f_R is the fraction of the products that are recombined back to water within the cell, and $\Delta H/nF$ is the thermoneutral potential of the water splitting reaction (1.527 V, D_2O; 1.481 V, H_2O at 25°C). The latter value has a very weak dependence on temperature only (about 0.2 mV/K) so no correction for variation in electrolysis temperature from 25°C was necessary. This also implies that a large temperature difference between the electrolyte and the electrolysis gas exit could be tolerated without introducing significant error into the heat balance calculation. In practice, the electrolyte temperature was between 30 and 40°C, depending on the current, and the gas outlet temperature was likely within 5°C of this. The value of f_R was determined by measurement of the electrolysis gas flow rate, $\dot{v}$, and was given by

$$f_R = 1 - \frac{p\dot{v}/RT}{3I_E/4F} \tag{2}$$

where p and T are the pressure and the absolute temperature at the flowmeter, and F and R are Faraday constant and the gas constant, respectively. The measured volumetric flow rate was adjusted assuming that the gas was saturated with water. The effect of the associated heat of evaporation on the heat balance was negligible. The rate of heat input from the external heater, $\dot{Q}_H$, is simply given by

$$\dot{Q}_H = I_H V_H \tag{3}$$

where I_H and V_H are the current and the voltage of the heater, respectively.

The rate of heat removal by the compressed air, $\dot{Q}_c$, is given by

$$-\dot{Q}_c = \dot{m}\, C_p\, \Delta T \tag{4}$$

where $\dot{m}$ is the mass flow rate of the air, C_p is the heat capacity of the air (1.0061 J/g K), and ΔT is the temperature increase across the cooling jacket. Because the gas was dried in the compressor, there was negligible error in the C_p value due to water vapor being present.

The excess heat removal rate, $\dot{Q}_{Ex}$, is then given by

$$\dot{Q}_{Ex} = \dot{Q}_c - (\dot{Q}_H + \dot{Q}_j) \tag{5}$$

The excess observed on the test side, $\dot{Q}_{Ex,T}$, was subtracted from that observed in the reference cell, $\dot{Q}_{Ex,R}$, to give the normalized excess heat, $\dot{Q}_{Ex,N}$.

$$\dot{Q}_{Ex,N} = \dot{Q}_{Ex,T} - \dot{Q}_{Ex,R} \tag{6}$$

The data was logged continuously, at least every five minutes, and then read into spreadsheet for the heat balance calculations.

The only departure from strict steady-state operation was the slow diminution of the electrolyte volume. Water was added periodically to the test (D_2O) and reference (H_2O) cells in order to maintain the electrolyte level approximately constant. At the currents used (about 1 A), this amounted to about 10 mL per day.

To avoid heat losses to the atmosphere, the cells were insulated with at least 10 cm of polystyrene on every side. The largest error in the absolute measurement of excess heat rate for each cell was the rotameter calibration which was accurate only to ± 2%. This, however, was compensated by normalizing the excess heat as shown in Equation (6). Other sources of error were error in the thermocouples for the measurement of ΔT, and a ± 2% error in the soap bubble meter measurement of the electrolysis gas flow rate. The room temperature control of the experiment trailer in which runs A, B, and C were conducted was also oscillating by ± 2°C and this introduced noise into the system through heat transfer into the cooling gas loop. We estimate the sensitivity of the calorimetry to be ±100 mW for the normalized excess heat.

3. Neutron and γ Measurements

During the electrolysis runs A, B and C, a NE213 liquid scintillation detector with dimensions diameter 12.5 cm × thickness 10 cm was positioned with its face 15 cm (A,B) and 10 cm (C) from the center of the D_2O cell, giving a sensitivity for detecting 2.5 MeV neutrons from the cell, $\varepsilon_n = 1.4\%$ and 2.6%, respectively (gamma energy threshold 0.2 MeV). The detector signals were electronically analyzed using the pulse shape discrimination method, such that signals from minimum ionizing particles (γ's, cosmic muons, etc.) were eliminated. In this way a sixty fold reduction of the background was achieved.

A BGO γ detector was mounted on the opposite side of the D_2O cell to identify γ-rays from p + d fusion or other reaction channels. The detector, with dimensions diameter 7.62 cm × thickness 7.62 cm was placed 15.6 cm from the cell, with an overall γ efficiency $\varepsilon_\gamma = 0.5\%$ for 5.5 MeVγ's (full energy peak).

The whole set-up was embedded in about 40 cm of boron-polyethylene and boron-paraffin. This shielding strongly reduced neutron background originating from outside the cells to a residual rate below 0.04 s^{-1}.

The neutron and gamma spectra were recorded in event mode on tape during 5 weeks of data taking using a CAMAC-μVAX computer system.

The energy calibrations were made with radioactive sources ^{22}Na, ^{137}Cs and ^{60}Co, while a ^{252}Cf spontaneous fission source allowed the tuning of the pulse shaping electronics. In Fig. 2 a typical proton recoil spectrum is shown, together with the corresponding pulse shape information (insert).

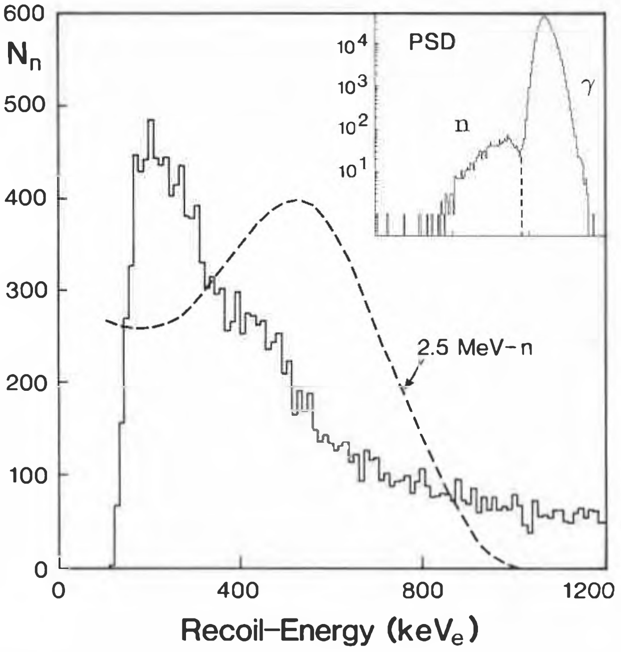

Fig. 2. Neutron recoil spectrum measured with the NE213 liquid scintillation detector ($V = 1.2$ L). The dashed line indicates the shape that would be expected from proton recoils of monoenergetic 2.5 MeV neutrons (shape Monte Carlo generated, as measured in previous dμd fusion runs with the same detector). – Insert: n/γ pulse shape discrimination spectrum. The small peak to the left is due to proton recoils from energetic neutrons.

4. Helium and Tritium Analyses

In addition to the on-line measurements of n/γ nuclear radiation, the fusion process could be identified by the presence of helium (He) or tritium (T) fusion products.

The detection of excess T is limited. The available D_2O contains trace amounts of T ($\approx 10^{-9}$ mol/L) which get enriched during electrolysis due to isotope separation effects. We concentrated our attention on the detection of excess helium accumulated in the palladium, and to measurements of tritium enrichments in the electrolyte during electrolysis. Standard liquid scintillation techniques were used to measure the tritium concentration via its beta-decay.

Helium analyses were carried out at ETH Zürich, using a mass spectrometer capable to resolve ^{3}He from HD. With this instrument, extraction blanks of ^{3}He and ^{4}He as low as $3 \cdot 10^4$ and $1.5 \cdot 10^9$ atoms, respectively, can be achieved. Hence helium is basically a rather sensitive indicator for fusion reactions. This is particularly due to the low abundance of helium on earth and because helium from fusion can be accumulated in long-term experiments.

We extracted ^{3}He and ^{4}He by fusing the Pd-wire in an UHV furnace at 1850°C for 15 min. The purified noble gases were admitted to a statically operated, single focusing magnetic mass spectrometer designed for high sensitivity and pressure linearity. The magnetic sector field is 90° with a radius of the ion trajectory of 21 cm. To prevent scattered ^{4}He ions to reach the secondary electron multiplier used to count the ^{3}He ions, the analyzer tube is Y-shaped with an appendix for the detection of ^{4}He in a Faraday collector. This geometry allows to record both He isotopes simultaneously. To resolve ^{3}He ions from HD ions the exit-slit on the mass 3 collector is set to a mass resolution of 1000. Sample gas amounts were calibrated by admitting amounts of atmospheric He known to be about 1%, immediately before and after the sample analysis.

Some 50 mg of the Pd-wire used in experimental run A and three additional Pd-wires, as received from the manufacturers, were analyzed. Their ^{3}He and ^{4}He contents were essentially not distinguishable from the signals observed in blank extractions. The same is true for a run A type wire exposed to an electrolysis experiment using H_2O. A sample of the wire exposed in run A (Table 3) was subjected to a stepwise

Table 3. Summary of the helium mass spectrometric analysis. A 38.88 mg sample of Pd wire from experiment A was investigated. (*) = no analysis possible due to very high HD and D_2 amounts. ^{4}He values in parentheses are upper limits, due to interfering D_2.

Date of Extraction	Extraction Temperature [°C]	^{3}He [10^5/g]	^{4}He [10^{10}/g]
30.05.89	20	120	(5)
		16	(9)
30.05.89	50	900	(67)
		350	(15)
30.05.89	100	140	(42)
		120	(19)
31.05.89	200	*	(*)
31.05.89	600	310	(99)
		270	(11)
31.05.89	1000	310	(13)
		100	(12)
31.05.89	1850	1800	16
		260	9
Total		3580	(243)
		540	(27)

extraction at different temperatures to separate residual D and T from helium. Deuterium is degassed well below 1000°C up to which temperature little helium is lost.

5. Results and Discussion

5.1. Calorimetry

Throughout two months of operation, no evidence of excess heat was observed. The recombination rate was consistently less than 3 percent, so the Joule heating rate approached its lower bound. The following are some representative results.

In run A, the 1 mm wire cathode was operated at 1.5 A (120 mA/cm^2) for 14 days. Just prior to the end of the run the current was turned up to 1.7 A (135 mA/cm^2).

In an attempt to see if H-D fusion could be detected, run B, a 4-day experiment was conducted. Fresh electrodes were installed and a mixture of 95% (vol.) 0.16 M LiOD and 5% (vol.) 0.1 M LiOH were inserted in the test cell. A current of 1.5 A (120 mA/cm^2) was used and no excessive heat or evidence of fusion products were observed.

In run C, the electrolysis current was kept constant at 1.2 A (545 mA/cm^2) for 18 days, and Fig. 3a–3d show some of the results from this time period for the reference and test cells. The cooling load varied somewhat due to variations in the cooling gas inlet temperature, caused by a small amount of heat transfer with the room. Fig. 3c shows the normalized excess heat for this period; the average is −65.4 mW and the standard deviation is 103 mW. Based on Ref.[9], a fusion signal of 1 W was expected. Clearly, the calorimetry was sufficiently sensitive to detect such a signal, and it was not observed.

Run D was performed in a laboratory with improved calorimetric stability conditions, but without n/γ detectors. Also no excess heat was observed throughout this run.

5.2. Gravimetric and Surface Analytical Measurements

Gravimetric H and D loading measurements in Pd were made after each of the runs described above. Values of 0.85 to 0.95 for H/Pd and D/Pd were found, in good agreement with other studies[25]. Surface analytical XPS investigations of the electrodes showed that the Pd electrodes were covered by some monolayers of Pb, and minor contaminations of Zn and Hg were also registered. Similar results were obtained at the ABB Laboratory[26]. Small impurities in the D_2O and H_2O could account for this deposition[27]. However, the gravimetric investigation proves that this surface layer did not prevent the incorporation of hydrogen and deuterium.

5.3. n/γ Results

The measurement of n/γ spectra during the calorimetry runs A, B, and C, with

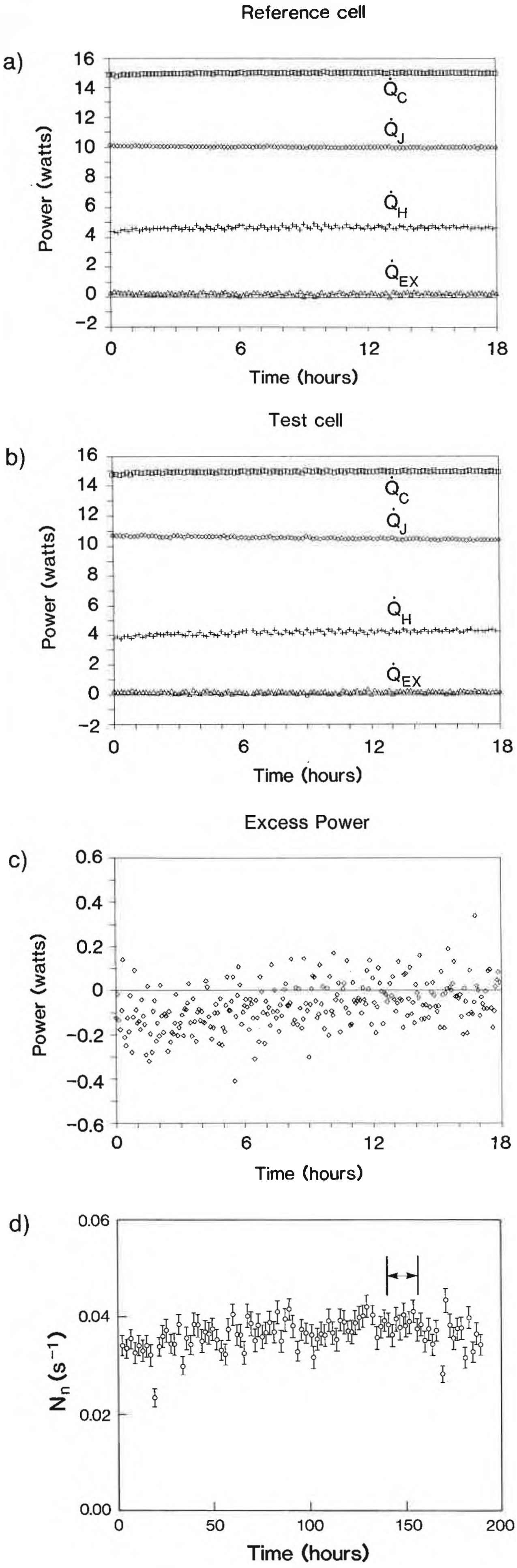

Fig. 3. Representative results for run C (18 hour period ending 14:00 on June 3). Calorimetry results from a) reference and b) test cells, c) the normalized excess power, and d) neutron counting results (8 days; the arrow shows the period of displayed calorimetric data).

intermittent electrolysis running at various currents, showed no correlations and in fact no excess of neutrons nor gammas. The n/γ spectra (for neutrons see Fig. 2) clearly show no evidence of monochromatic neutrons or gammas as would be expected from fusion reactions. The neutron rates are shown per 2 h time bin in Fig. 3d. They were quite stable, never showing significant excursions. Taking into account all possible sources of systematical errors (shifts, gain drifts, etc.) we can exclude with these data any excess neutrons above 10% of the residual rate in the sensitive region. Thus we get as upper limits the following n/γ rates (assuming its origin in the center of the cell):

2.5 MeV-n	I_n^{excess}		< 0.26 n/s	run A, B
		or	< 0.06 n/g·s	run A, B
			< 0.14 n/s	run C
		or	< 0.10 n/g·s	run C
5.5 MeV-γ	I_γ^{excess}		< 0.10 γ/s	run B
		or	< 0.02 γ/g·s	run B

The latter numbers are normalized to 1 g of the Pd cathode material used for electrolysis. These upper limits reflect essentially systematical uncertainties, while the statistical accuracy over a whole run period was about one order of magnitude smaller. Since our n/γ measurements were subdivided in runs of 20 min, we also can exclude any neutron bursts, exceeding 1000 neutrons per 20 minutes.

Any neutron or γ excess rate of the magnitude given here would have nothing to do with the heat reported by *Fleischmann* and *Pons*, as discussed below. Our conservatively chosen upper limits are clearly below the positive observations reported by *Jones* et al. [10] (e.g. 0.4 n/s from 3 g of titanium), although a direct comparison is difficult due to different experimental arrangements and choice of materials.

5.4. Helium Results

From the concentration of ^{3}He in the Pd-wire used in run A (Table 3) and from the duration of this experiment of 15 days (Table 2) we deduce a production rate of ^{3}He of

$$P3 = (280 \pm 40)\ ^3\text{He/g·s} \qquad (7)$$

Unfortunately, the tritium concentration of the heavy water used in run A ($180 \cdot 10^{-6}$ curie/L or $56 \cdot 10^6$ TU or T/D $= 5.6 \cdot 10^{-11}$ in 1980) is relatively high. Furthermore, neither the mean T/D ratio in the electrode nor its temporal change during the experiment are known. By simply assuming the T/D ratio in the Pd electrode to be the same as in the heavy water and a D/Pd ratio of about 0.9, we deduce a production rate of ^{3}He from the beta-decay of tritium to be about:

$$P3' = 310\ ^3\text{He/g·s} \qquad (8)$$

This is remarkably close to the measured ^{3}He production rate, suggesting that, within error, all ^{3}He detected is due to the tritium initially present in the heavy water. If fusion reactions on the level reported by *Jones* et al. [10] are to be confirmed by ^{3}He as a fusion product, the T/D ratio in the heavy water should be considerably less than 10^{-14} and the heavy water must be free of atmospheric helium.

The determinations of ^{4}He are hampered by the substantially larger extraction blanks and by interfering D_2. Nevertheless, the 1850 °C extraction step of the run A sample yields an upper limit for the production of ^{4}He by fusion of about

$$P4 = 4 \cdot 10^5\ ^4\text{He/g·s} \qquad (9)$$

Even though this limit may be much too high because of the D_2 contribution, it clearly precludes a fusion rate of 10^{12} reactions per second as reported by *Fleischmann* and *Pons* [9] by more than 6 orders of magnitude.

5.5. Tritium Enrichment in the Electrolyte

The D_2O electrolyte solution was not completely free of tritium. A significant increase of the tritium concentration during the electrolysis was noticed. This is expected due to the isotope separation effects between liquid and gaseous phase during electrolysis. In run D we measured the increase of tritium in the electrolyte of the test cell. Taking into account all D_2O refills and the initial and final electrolyte volumes, the observed enrichment of tritium during electrolysis can be explained with an isotopic separation factor of about 2 (N.B. approximately 10 mL D_2O were electrolyzed per day and had to be replaced periodically). Isotope separation is due primarily to different desorption activation energies of tritium and deuterium at the palladium electrode. An isotope separation factor between deuterium and tritium of 2 is considered reasonable and is in good agreement with the results obtained by *Comninellis* and *Friedli* [28].

6. Conclusions

The reports of evidence for cold fusion observations initiated by *Fleischmann* and *Pons* [9] and *Jones* et al. [10], have to be grouped into two categories [8]:

(a) Excess heat generated in D_2O electrolysis on Pd electrodes.
(b) Neutrons or neutron bursts produced in or on deuterium-loaded metals (Pd, Ti, etc.) exposed to stresses like pressure, thermal cycling, phase change, high-current electrolysis.

The two categories are to be considered as separate since they differ with each other by 9 to 13 orders of magnitude in the energy scale!

Category (a) – reported by Ref. [9] and in part also by Ref. [12] is at odds with present knowledge of physics:

(a1) No explanation has been suggested for how the necessary shielding of hydrogen nuclei can be accomplished with electrons [23].
(a2) No way exists, according to all knowledge of nuclear physics to exclude the $d + d \rightarrow n + {}^3$He channel, nor the $d + d \rightarrow p + t$ channel (see Table 1).
(a3) There is no mechanism known which could enhance the $d + d \rightarrow {}^4\text{He} + \gamma$ channel over the regular channels (a2) by many orders of magnitude.
(a4) There is also no mechanism conceivable that could transfer a 24 MeV electromagnetic quantum to the lattice (assuming the $d + d \rightarrow {}^4\text{He} + \gamma$ reaction to occur).

It is thus not surprising that the worldwide attempts to reproduce *Pons & Fleischmann's* announcement failed, including this and many other groups reporting [26,27,29].

Our calorimetric method allowed continuous power measurements by measuring the temperature difference of the cooling media before and after the cell at fixed flow rate. Inhomogeneous electrolyte temperatures inside the cell did not affect the measurements. We determined the recombination rate by measuring the rate of gas evolution periodically. We thus excluded the two main criticisms of the experiments carried out by *Fleischmann* and *Pons*. They had no control over the recombination rate and they measured heat production by following the temperature rise inside the cell, where the only mixing was due to gas evolution. In such an experiment inhomogeneous temperature in the electrolyte could easily lead to wrong calorimetric results. The tritium enrichment in the electrolyte can be explained with a reasonable isotope separation factor.

The second category (b) of possible neutron observations has not yet come to a clear conclusion. At the rates reported we cannot exclude some acceleration mechanism which in certain cases may yield «hot fusion» neutrons [8]. Again we would like to point out that such processes cannot lead to energy production.

The existence of category (b) fusion is, however, not established, since most reports are based either on insufficient statistics or irreproducible conditions.

With the «Pons-Fleischmann» set-up, as reported in this paper, we can exclude from our n/γ observations any continuous fusion rate above the 10^{-13} W level, which is below the effect reported by *Jones* et al. [10].

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