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Measurement of Excess Energy and Isotope Formation in the Palladium-Hydrogen System

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Abstract

A search for the products of fusion reactions that could be triggered by sparking in hydrogen isotopes was carried out. No signatures above background were found. On the contrary, the excess energy production was confirmed in the simple hydrogen/palladium system. The formation of a tightly bound state of the hydrogen (deuterium) atom is hypothesized to explain these results.

1. Introduction

Excess energy production up to several watts have been constantly observed when a metallic hydride forming metal (palladium, stainless steel) is contacted with a discharge struck in an hydrogen isotope ($_1^1H$ or $_1^2H$) at atmospheric pressure [1,2]. Such excess energy production is also observed when electrolyzing water ($_1^1H_2O$ or $_1^2H_2O$) with metallic hydride forming metals (nickel, palladium) [3,4]. Since one possible explanation of this phenomenon is the occurrence of hydrogen isotopes fusion reactions in the metal, we have carried out a search for the products expected from these reactions. No signature above background have been found that can explain the amount of excess energy produced.

We have thus concentrated our efforts on the way this excess energy is produced in the most simple system (pure palladium and pure hydrogen). We have measured the excess energy production per atom of hydrogen. We find that for a given set of experimental conditions, the excess energy production per atom hydrogen involved is constant. Values up to 6000 eV have been measured.

To explain our results, we hypothesize the formation of the Hydrex (Deutex) state of the hydrogen atom, occurring when a high current flows through metallic hydride forming metals containing large amounts of hydrogen isotopes. The properties of these hypothetical atomic species could account for secondary nuclear reactions explaining a number of the weak nuclear signatures observed in similar experiments.

2. Methods

Search for the products of the fusion reactions

This search was carried out using the reactors previously described [1,2]. The metallic hydride forming metal was palladium. Both hydrogen and deuterium were tested. ${}^{3}_{2}He$, ${}^{4}_{2}He$, ${}^{3}_{1}H$ neutrons, X and γ rays were monitored with following methods:

- ${}^{3}_{2}He$ and ${}^{4}_{2}He$ both in the gas phase and in the electrodes were monitored by a high resolution mass spectrometer, equipped with various getters and filters eliminating all gases, except rare gases, thus increasing the resolution. The gas phase was sampled in stainless steel bottles and transferred to the mass spectrometer. The electrodes were melted in a furnace (heated by an ion beam) inside the mass spectrometer thus eliminating air contamination. ${}^{22}_{10}Ne$ was also measured in all samples.
- ${}^{3}_{1}H$ in the gas phase and in the electrodes was first oxidized and the resulting tritiated water was analyzed by liquid scintillography.
- Neutrons were monitored by two ${}^{3}_{2}He$ counters with energy spectroscopy to identify the characteristic peak at 764 keV. Various moderators (water, paraffin) were used.
- X and γ rays were monitored by an ionisation chamber and by solid scintillography with energy spectrometry (Csl and Nal cristals).

Measurement of the excess energy production per atom H

For this, we have measured with precision the excess energy production and the hydrogen consumption in the system hydrogen/palladium (Pure hydrogen – Alphagaz N55, and pure palladium – Johnsonn Matthey Puratronic grade – were used).

- The excess energy production was measured in a system different from the one previously used [1]. The reactors are of the ionizer type, cylindrical (typically 2 cm diameter, 60 cm long) with Pyrex dielectric barriers. We have used either single dielectric barrier type reactors (the electrode in contact with the gas being a 0.25 mm palladium wire) or double dielectric barriers type reactors (the palladium in that case being a 0.1 mm thick foil, at a floating potential between the two-barriers). Fig. 1 is a drawing of the double barrier type of reactor and Fig. 2 of the single barrier one.

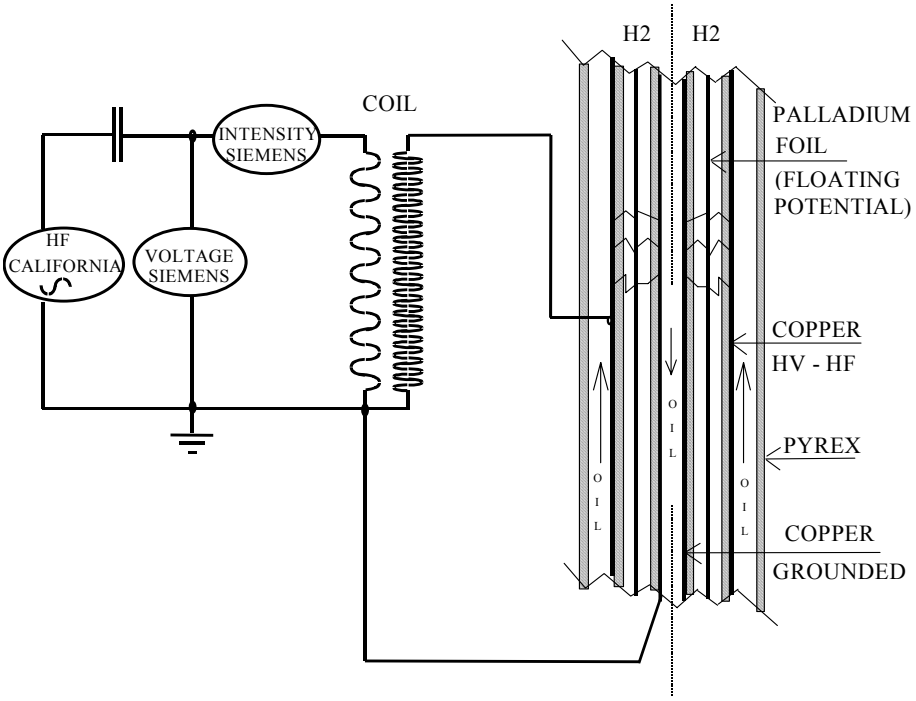

Fig. 1 DOUBLE DIELECTRIC BARRIER

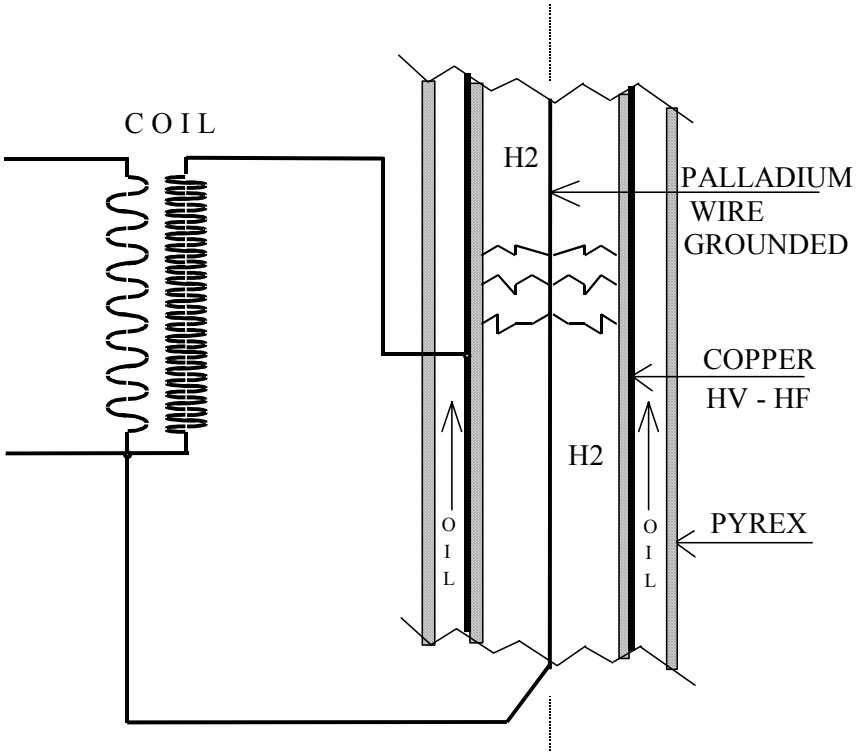

Fig. 2. SINGLE DIELECTRIC BARRIER

The double barrier reactor type allows very representative blanks to be made, by withdrawing the palladium foil. The discharge in that case, still occurs between the two dielectric barriers, with no contact with any metal.

A single calorimeter contains the high voltage coil and the reactor. The high voltage coil is powered by a high frequency (50 to 5000 Hz), low voltage (0 to 220V) generator (Invertron from California instruments) that delivers a quasi sinusoidal signal. The electrical power input is measured at the outlet of the Invertron by a high sampling frequency, numerical wattmeter (Siemens Functionmeter B 1082). The reactor and the coil are housed in a PVC cylinder (10 cm diameter, 110 cm high), that also contains the tubing for circulating silicon oil (Rodhorsil 47V20 Rhone Poulenc) through the reactor (and the coil) and the wires connecting the system to the Invertron. The PVC cylinder is wrapped in a 1 cm thick layer of insulating material (Polyethylen sponge, closed cells, 0.019 kcal/m°C) surrounded by a copper coil (110 cm high) made of joined whorls of copper tube (1 cm diameter) through which the silicon oil circulates. This coil is wrapped in a 5 cm thick layer of insulating material and the resulting cylinder (24 cm diameter is housed in a PVC cylinder (25 cm diameter, 140 cm high), closed at the bottom and the top by two insulating caps, 15 cm thick, with holes for passing the tubing and wires. The outer PVC cylinder is surrounded by a double coil made of joined whorls of PVC tubing (1.8 cm diameter), through which water flows, at a temperature maintained at 20°C (± 0.1 °C) by a cryothermostat (Huber HS 40). The double coil is itself wrapped in a 2 cm thick layer of insulating material. Fig. 3 gives an overall view of the calorimetric system.

In spite of the use of high voltage/high frequency currents, this calorimetric system is electrically tight; that is, no discharge occurs between the high voltage parts and the low voltage ones (except of course in the gaseous gap of the reactor). It is quasi adiabatic, 95 to 99% of the heat being recovered in the flow of silicon oil circulating (volumetric FM1 pump Model RP-D, 1/4″ ceramic head) round (copper coil) and through the high voltage coil and the reactor. This heat flux is calculated from the measure of the silicon oil flow (111 Flo-Meter McMillan Co, checked 4 to 6 times per day with gauge flask and chronometer), and the measures of the inlet and outlet oil temperatures (PT 100 Platinum gauge AOIP). The variations of the specific gravity and specific heat of the oil with temperature are taken into account. The oil stability during the runs is checked by periodic control of its specific gravity and by calibrations as will be described below. The heat flux which is exchanged with the air of the laboratory (5 to 1%) is determined by calibration (correlation between the heat lost and the difference of temperature between the reactor and the laboratory - see calibration).

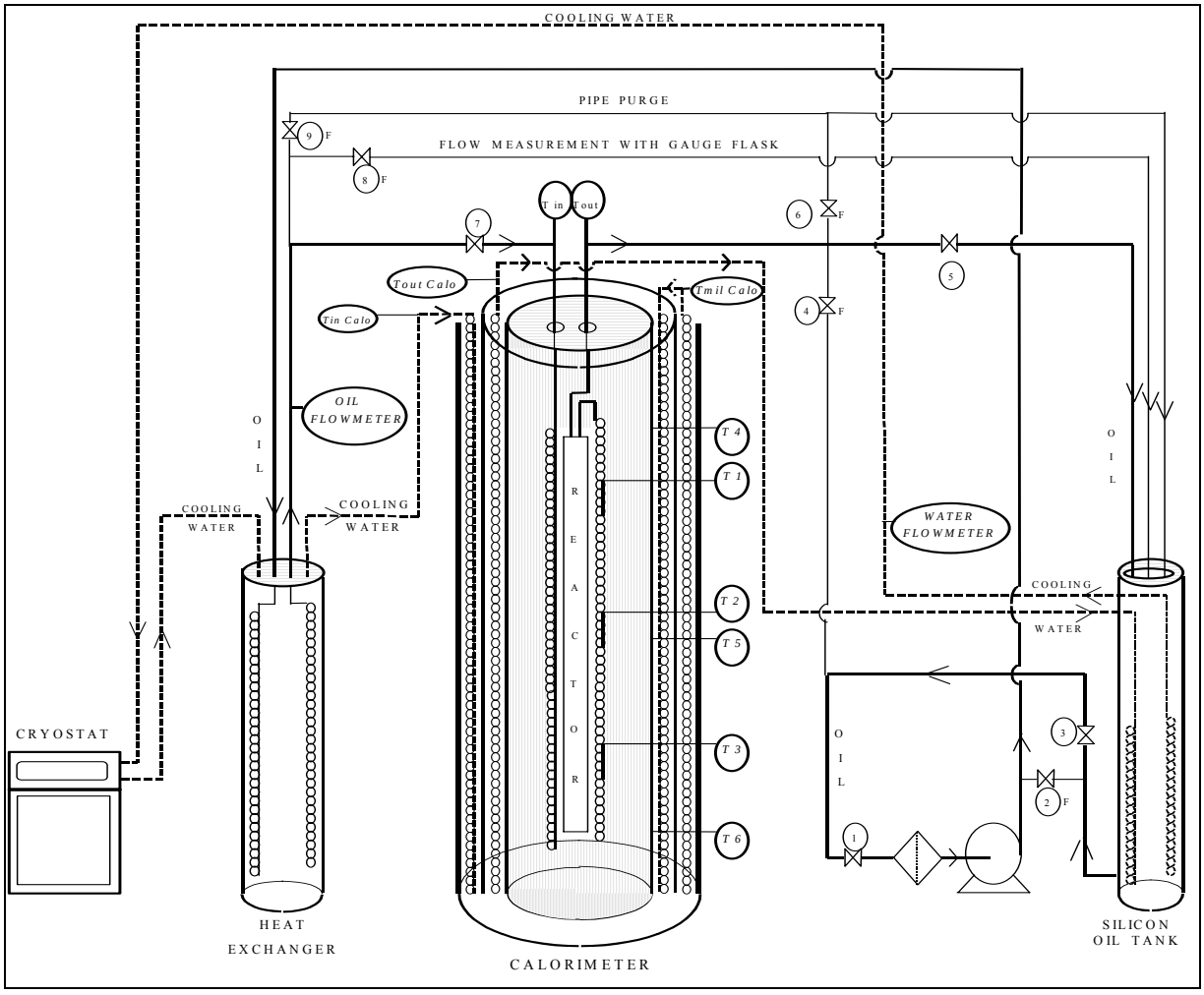

Fig. 3. OVERALL VIEW OF THE CALORIMETER SET-UP

- Hydrogen consumption is measured by a simple pressure balance of the reactor and its feeding reservoir (300 cm^3 initial pressure 5 bars). The reservoir, the pressure regulating valve and the tubing of the hydrogen system are shown on Fig. 4. They are made from stainless steel. A 40 cm long, (1.6 mm ID, 3.2 mm OD) FET Teflon tube connects the stainless steel tubing to the pyrex reactor. The whole system is tight up to 1500 mb of reactor hydrogen pressure and no leakage is detectable on several days tests.
- All data required for the excess power and the hydrogen consumption measurement are collected every 15′ by a data logging system (Centrale d'acquisition AOIP SA 70)

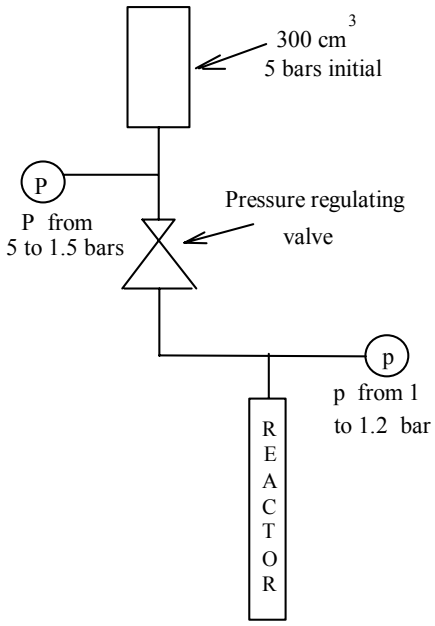

Fig. 4. HYDROGEN CONSUMPTION MEASUREMENT

3. Results

Search for the product effusion reactions

The results of that search can be summarized as follows:

- ${}^{4}_{2}He$ found in the gas phase come from air leaking through the Pyrex or the joints of the reactor. This is clearly evidenced by the ratio ${}^{4}_{2}He/{}^{22}_{10}Ne$ calculated for the "apparent production" of these two isotopes (resulting from a mass balance between the gas input in the reactor and the gas after the experiment, taking into account the hydrogen absorbed by the palladium). Typical values are the following:

air: 3.13 ${}^{1}_{1}H$ experiment: 5.0 ${}^{2}_{1}H$ experiment: 6.0

showing a preferential leakage of ${}^{4}_{2}He$ from air, whatever is the hydrogen isotope used.

- No peak at mass 3 (${}^{3}_{2}He$) was detected in the gas phase.
- ${}^{4}_{2}He$ "apparent production" was also found in the electrodes. In all experiments, this can be explained by ionic implantation from the gas phase. This is evidenced by considering the evolution of the ratio ${}^{4}_{2}He/{}^{22}_{10}Ne$ from the zone of impact of the sparks, to the edge of the electrodes. Given the higher temperature of the impact zone and the evolution with temperature of the diffusion coefficients of ${}^{4}_{2}He$ and ${}^{22}_{10}Ne$, the former should be higher than the latter, when ${}^{4}_{2}He$ and ${}^{22}_{10}Ne$ are implanted by the discharge. This was found to be the case for all experiments

both for ${}_1^1H$ and ${}_1^2H$. However, in all experiments, the impact ratio was higher than the one in the gas phase, with a mean value of 30 (a value of 120 was found in one experiment with ${}_1^1H$). If we attribute to true generation the totality of ${}_2^4He$ found in the electrodes, we arrive at an upper limit of 105 atom/s (for the ${}_1^1H$ experiment mentioned above).

- A peak at mass 3 was detected in the gas from the electrode of one experiment with ${}_1^1H$, and one with ${}_1^2H$. This peak is very likely a HD peak, due to a memory effect of the spectrometer system. It gives an upper value for the true ${}_2^3He$ generation: 5×10^3 atom/s.
- No ${}_1^3H$ was detected above detection limits for ${}_1^1H$ experiments or beyond what is contained in the feeding gas for ${}_1^2H$ ones. This gives the upper limit of production: 2×10^3 atom/s.
- Neutron detection was also negative: the background flux has been shown to fluctuate from 0.5×10^{-3} to 1×10^{-3} n/cm^{2}xs (during a period of 4 months), with no correlation with the excess energy production.
- No X and γ rays were detected with the ionization chamber. Copious signals have been detected with the solid scintillation counters, in the low energy region (<100 keV). These signals are currently under study.

Measurement of the excess energy production per atom H

- The excess energy production is measured during "active runs" that last 3 to 6 weeks (in order to eliminate transient situations that can occur at the beginning of a run). "Active runs" are runs were a discharge is struck through an hydrogen isotope, in contact with an hydride forming metal. Before each "active run," the temperature gauges are calibrated. Each "active run" begins with a calibration (2 to 3 days), where the reactor is under Ar or air and the system heated by resistors, powered by the California generator. This calibration ensures that the properties of the silicone oil are stable. The results of these "active run" calibrations are checked to fit the calibration curve, that was established in a way that will now be described.
- *The calibration curve* was established during "calibration runs," where the system was heated in different ways: resistors, discharges in Ar with the two types of reactor (with metal in contact with the discharge) and discharges in hydrogen in the double barriers type reactor, with no metal between the dielectric barriers. In the runs with resistors, the power was delivered either by a DC generator, or by the California generator (with cos φ varying from 0.35 to 1). The power lost to the air is:

Power lost = Power recovered in oil - Power in

and is correlated with the difference of temperature between the reactor and the air of the lab:

$$\text{Power lost} = K (T_{reac} - T_{air})$$

Fig. 5 shows that all calibration points fall on the same correlation, whatever the way the system is heated. Moreover the "calibration runs" with a discharge, show that the system is electrically tight, that is no discharge occurs, that could interact with the various components of the calorimetric system (insulation, PVC ...). Electrical charges transport only occurs in the gas gap of the reactor.

The calibration runs were performed over a period of 4 months, including 3 complete dismantling of the reactor and calorimetric system. Over this period, the standard deviation of the excess power measurement (zero for calibration runs) is 0.7 W, demonstrating the stability of the system (this figure is reduced to 0.3 W, when only a single run is considered).

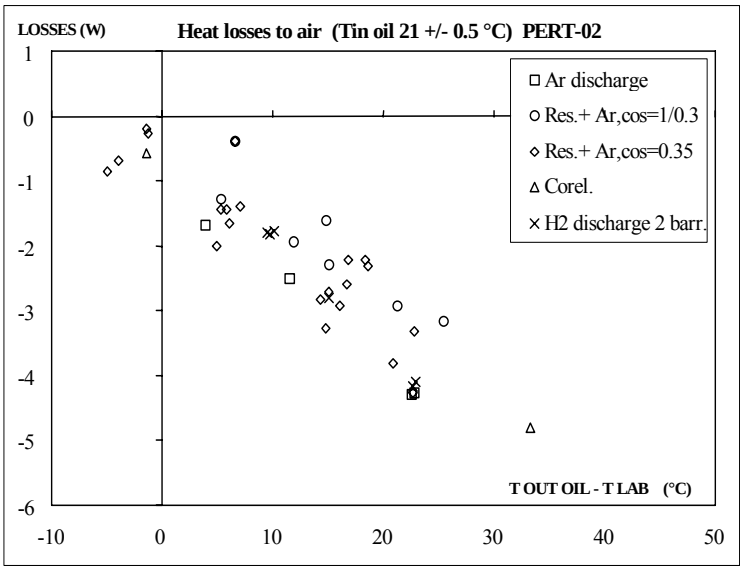

Fig. 5. HEAT LOSSES TO AIR, FUNCTION OF OIL TEMPERATURE

- *The results of the active runs* are very dependant on the type of palladium used:
- runs with foils gave results ranging from 0 to 3 W excess power (for 100 W power input). This erratic performance was shown to be related to the mechanical resistance of the palladium: the palladium that gave negative results was found after the experiment with scales on the surface. The loading ration for these negative experiments was of the order of 0.75. On the contrary, the surface of the palladium that gave positive results was found not altered after the experiment. In that case, strange behaviour of the loading ratio was observed (figures up to 2.5 were measured). This will be discussed later.
- runs with wires gave positive results, the wires being never found altered after the experiment. The strange behaviour of the loading ratio was also observed. Excess power up o 5.5 W were observed (for 150 W power injected). Given the measured standard deviation (0.7 W) these results are very significant.
- *A comparison of two "active runs,"* one with negative results and one with positive ones is shown on Fig. 6 and 7. The negative run was with a palladium foil and a double dielectric barrier. During 1.5 million second, the excess power was 0 ± 1.4 w (99% confidence level). The positive run was with a palladium wire and a single dielectric barrier. The excess energy increased on a period of 3.5 million second, to reach 5.5 ± 1.4 w (99% confidence level). The increase observed at 2.8 million second is due to change in operating conditions (see below).
- The hydrogen consumption together with the weight of palladium used in an experiment, allows the calculation of the loading ratio. For experiments that gave negative results, the behaviour of this loading ratio is normal: values are limited to 0.7/0.75 and once this value is reached, it remains constant for the rest of the experiment. On the contrary, for

experiments giving an excess energy, this ratio always increases. Values up to 5 have been measured in experiments with wires.

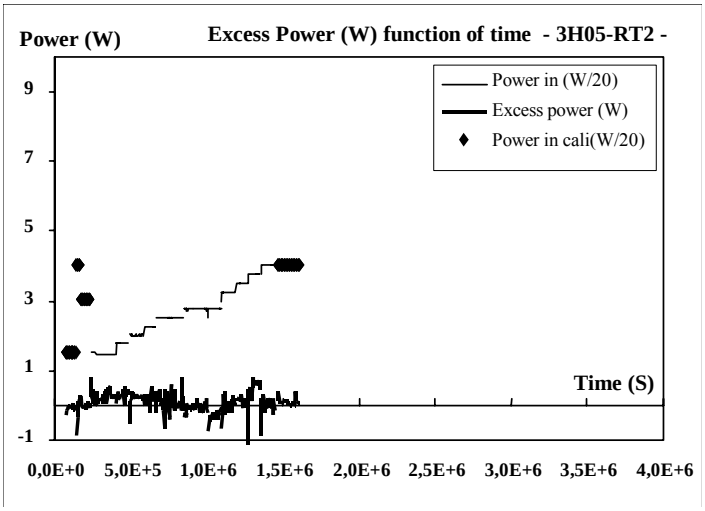

Fig. 6a. EXCESS POWER, FUNCTION OF TIME -3H05-

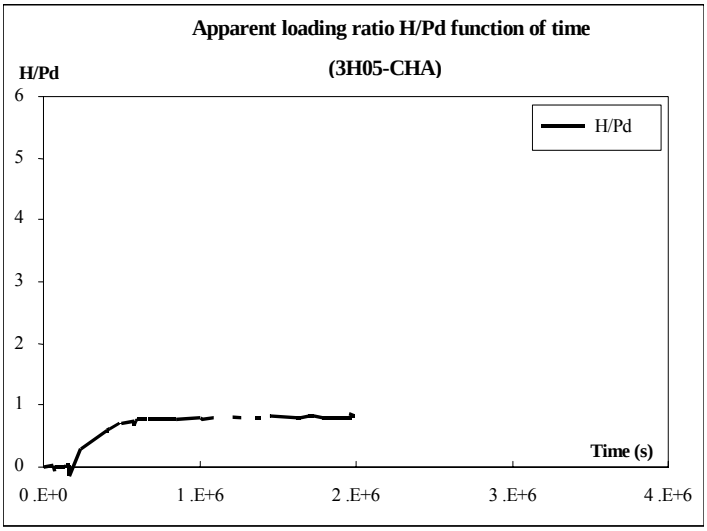

Fig. 6b. APPARENT LOADING H/Pd, FUNCTION OF TIME -3H05-

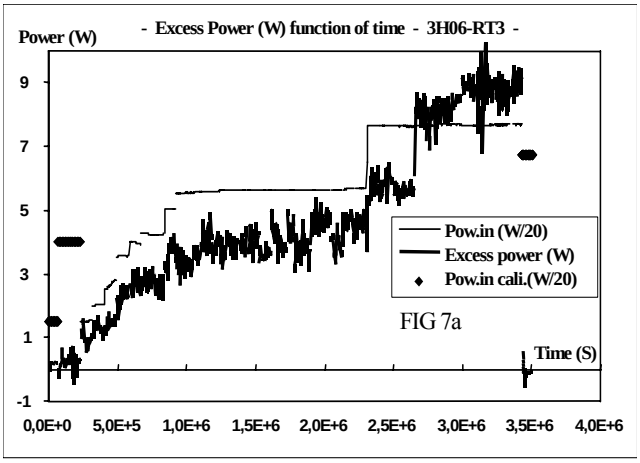

Fig. 7a. EXCESS POWER, FUNCTION OF TIME -3H06-

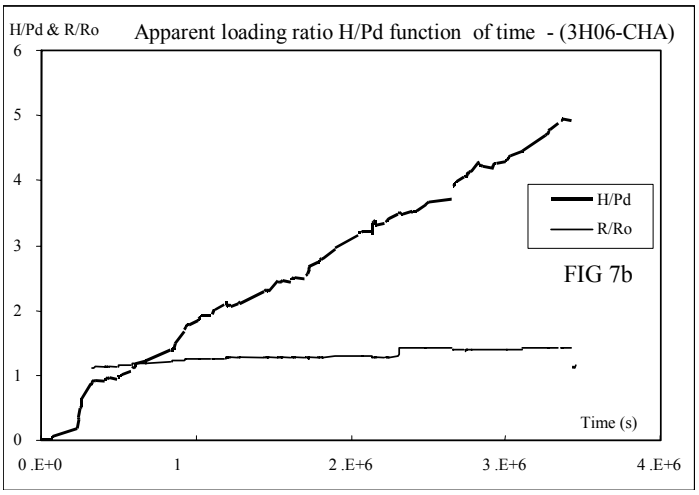

Fig. 7b. APPARENT LOADING H/Pd, FUNCTION OF TIME -3H06-

One obvious cause that could explain this is leakage. We exclude this because when the discharge is cut off, the consumption of hydrogen ceases. Another cause would be combination of hydrogen with material inside the reactor. This seems impossible, because the only material in contact with the discharge is Pyrex (and of course palladium). Small amounts of copper and steel are also present in the reactor, but not in contact with the discharge. The quantities of hydrogen they could absorb, were calculated and found several order of magnitude lower than the missing hydrogen. Their presence cannot explain the phenomenon. Finally, by weighing the palladium before and after the experiment, we saw that, in all cases, the final loading ratio thus measured, did not exceed 0.8. No hydrogen is thus stored in cavities of the palladium after experiment.

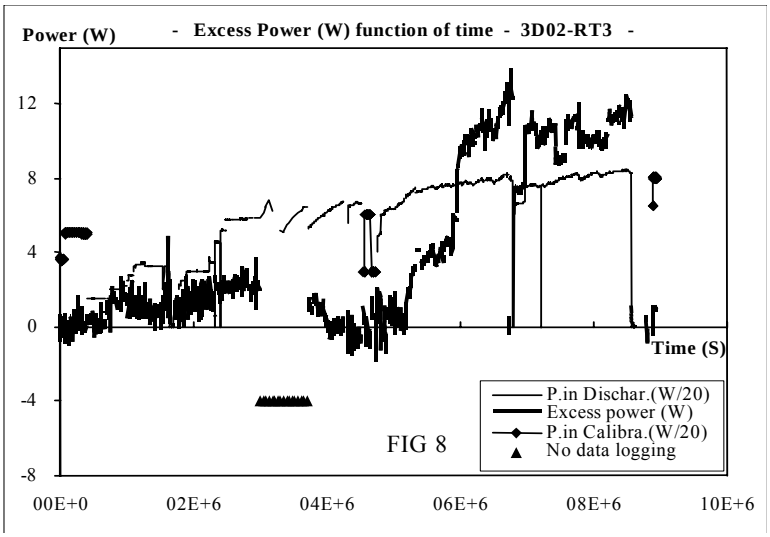

Fig. 8. EXCESS POWER, FUNCTION OF TIME -3D02-

The excess energy production per atom hydrogen can then be calculated from the above measurements. Fig. 8 is a plot of the cumulated excess energy versus the cumulated hydrogen consumption as measured in the experiment with a wire described previously. It can be seen that 3 straight lines can be drawn, corresponding to 3 different sets of operating conditions (see below). This means that under stable conditions, the excess energy per hydrogen atom is constant. These results are summarized in the following tables.

Table I EXCESS ENERGY PRODUCTION PER ATOM HYDROGEN FOR HYDROGEN -3H06-

Period	Duration second	H consumed mMole H	Excess energy on period (J)	Mean excess Power (W)	Exc. Ene. eV per atom H	Power input (W)	Temp. Reactor (°C)
CAL	230 000	0	0			Calib.	
INC	695 000	9.88	1 737 500	2.5	1 800	Increasing	
I	1 390 000	9.63	5 838 000	4.2	6 300	115	56
II	342 000	1.41	1 846 800	5.4	13 600	150	69
III	767 000	5.44	6 596 000	8.6	12 600	150	50
Total	3 424 000	26.36	16 018 300	4.7	9 000*		

* Excluding the “INC” period

Table II EXCESS ENERGY PRODUCTION PER ATOM HYDROGEN FOR DEUTERIUM -3D02-

Period	Duration second	H consumed mMole H	Excess energy on period (J)	Mean excess Power (W)	Exc. Ene. eV per atom H	Power input (W)	Temp. Reactor (°C)
CAL	403 200	0	0			Calibration	
Loading	3 632 400	5.51	3 741 300	1.03	7 000	Increasing	
I	964 800	0.0	0	0	0	125	52
II	864 000	0.62	1 391 000	1.6	23 300	155	56
III	2 744 100	11.03	25 218 300	9.19	23 800	155	45
Total	8 608 500	17.16	30 350 600	4.7	23 700*		

* Excluding the “Loading” period

At the beginning of period III, the decrease of the reactor temperature was achieved by increasing the oil flow. This decrease of temperature was immediately followed by a very fast increase in the hydrogen consumption (few minutes), followed by a slightly slower increase in excess power generation (few hours).

After cutting off the discharge, the hydrogen consumption ceased. Weighing the palladium after experiment showed that it contained 4 mMole of hydrogen (corresponding to a loading ratio round 0.75, the quantity of palladium in the reactor being 5.35 mMole). During the experiment, a total of 22.36 mMole of hydrogen (16.48 during steady state situations) have been lost from the reactor in an unexplainable way.

We conclude from the measurement of the excess energy per atom hydrogen

- that this quantity is a constant for a set of physico-chemical parameters but changes when this set is changed. Values up to 6 000 eV per atom H have been measured.
- and that hydrogen is lost in an unexplained way, during the production of excess energy.

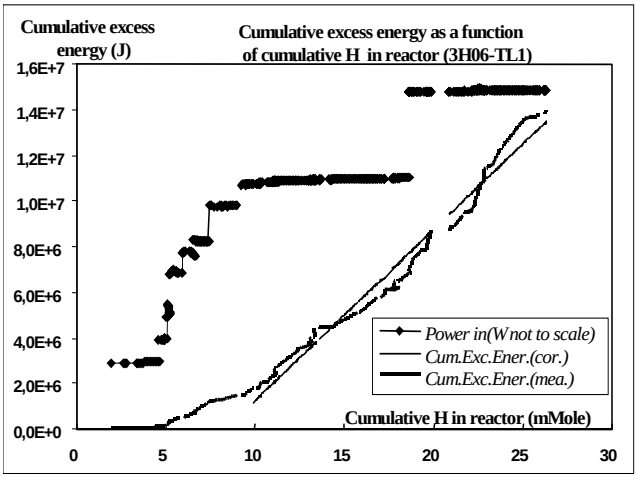

Fig. 9. CUMULATIVE EXCESS ENERGY AS A FUNCTION OF CUMULATIVE H IN REACTOR

4. Discussion

The following experimental evidence has been gathered:

- the upper values found for the ashes expected from the known fusion reactions of ${}_1^2H$ are much too low (many orders of magnitude) to explain 2 W of excess energy (for 1 W these values should be 10^{12} per second for protons, tritons, neutron and ${}_2^3He$, 10^7 for ${}_2^4He$ and gamma rays of 24 MeV or 3×10^{11} for ${}_2^4He$ if this is the only ash). In the case of ${}_1^1H$, no gamma rays of 511 keV were observed, that could have resulted from the annihilation of positrons formed by the known ${}_1^1H$ fusion reaction.
- excess power generation have been measured with high confidence level (99%). Taking into account the total excess energy generated (5 000 000 J), the following explanations have been eliminated (their effect would be several orders of magnitude lower than what is observed): combustion of the hydrogen, combustion of silicon oil, reaction of hydrogen with materials in the reactor, exothermal modification of the Pyrex under the influence of the electrical charges deposited by the discharge.
- this excess power is generated in a very specific way, the excess energy per atom of hydrogen being a constant for a given set of reactor conditions. The value of this energy is in the order of thousands eV. Moreover, this excess power generation occurs simultaneously with an unexplained loss of hydrogen from the reactor.

The Hydrex (Deutex) hypothesis:

The hypothesis of the formation of a tightly bound state of hydrogen (deuterium) in "cold fusion" experiments has been put forward [5,6]. In such bound states, the electron is much closer to the proton than in normal hydrogen. This could explain both an energy of formation and a capacity to diffuse through any material much higher than normal hydrogen. Hence, energies of formation of several thousands eV and loss of hydrogen through the walls of the reactor, could be explained.

We hypothesize that the formation of a one dimension (oscillator) bound state could occur, if the following conditions are met, for one proton and one electron inside the lattice of the palladium:

- their spins are parallel
- they get sufficiently close (100 to 500 fm ?)
- they have the same velocity

These conditions might be met at the occasion of the jump of an hydrogen (proton) from one octahedral site of the palladium to another, when a high electrical current flows through the palladium lattice. The first two conditions could be fulfilled when the current is sufficiently high and create a high magnetic field. Due to collisions, the last condition could be fulfilled for individual protons and electrons of the conduction band of the palladium, although their collective motions are opposite. Due to the action of the strong electromagnetic field created by the electron during its motion to and from the proton, this bound state, once formed, could remain stable, even after the external magnetic field has disappeared. This state being the result of the action of two potentials (electrostatic and magnetic) a continuum could be formed and this could explain that we measure energies of formation varying with operating conditions.

Expected properties of the Hydrex (Deutex) state of hydrogen:

Apart from its high energy of formation and its capacity to diffuse $H\cdot$ (Hydrex), should be a permanent electric dipole of small dimensions (hundreds of fm). When getting sufficiently close to a nucleus present in the metal lattice, it could be confined against it. The probability of electron capture by the nucleus could thus be increased as with ${}^{40}_{19}K$, for example:

$${}^{40}_{19}K + H\cdot = {}^{40}_{18}Ar + {}^{1}_{1}H^{+} + e^{-} + \nu$$

Similar reactions should be expected with ${}^{138}_{57}La$ and ${}^{50}_{23}V$, that give exothermal electron capture reactions.

If $H\cdot$ is confined for a sufficient long time, close to the nucleus, double electron capture could then occur, which could be the case for the following nuclei: ${}^{102}_{46}Pd$, ${}^{56}_{28}Ni$ ${}^{54}_{26}Fe$ and ${}^{40}_{20}Ca$.

These reactions could account for a number of weak nuclear signatures observed in the experiments

5. Conclusion

From these results, we conclude that a very promising reaction occurs in metallic hydride forming metals, when loaded with hydrogen isotopes and submitted to high transient electric currents. We think that explaining the phenomenon by a rearrangement of the binding between the proton and the electron, due to the confinement in the metal lattice, is more plausible (although not known) than invoking highly improbable fusion reactions. We intend to put more efforts in the understanding of this reaction.

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