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**ELECTROCHEMICAL COMPRESSION OF HYDROGEN INSIDE A PD-AG THIN  
WALL TUBE, BY ALCOHOL-WATER ELECTROLYTE**

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### Abstract

Yoshiaki Arata (Osaka University) has since 1955 developed and on 1994 patented in Japan (also USA, N° 5647970, 1997) a procedure for producing ultrahigh pressure H/D gas, based on the electrolytic loading of a hollow cathode made of a Hydrogen and/or Deuterium absorbing material (Pd).

The truthfulness of Arata's results and even the real existence of Arata's device have been recently challenged by an influent Italian scientist and suspected to be merely a "*computer simulation experiment*".

Accordingly, we decided to replicate the Arata experiment in order to confirm his results. A further reason for such a replication is due to the consideration that the electrolytic compression of Deuterium (D) is the basic for most of the Cold Fusion experiments. Moreover, the experiment allows for a direct evaluation of the D<sub>2</sub> (or H<sub>2</sub>) pressure inside the hollow cathode, without making use of indirect measurements, like those based on the well-known "Baranowsky curve" (change of the Pd electrical resistance, versus D or H absorption).

The cathode was a Pd-Ag (75%-25%) tube, closed at one end (diameter 10mm, total length 10cm, useful length exposed to electrolyte 9cm) with a very thin wall (thickness about 50µm), previously used as a Hydrogen purifier (home-made by Dr. Silvano Tosti: ENEA, Frascati, Italy).

The effective volume of the Pd-Ag tube (partially filled with a porous ceramic tube, in order to avoid the vacuum collapse of the Pd-Ag tube), pipes, valves and pressure gauges was about 18ml.

The anode was a Pt (purity >99.99%) wire (diameter 0.25mm) wounded as a pseudo-cylindrical spiral of radius about 4cm. The total length of the Pt wire was about 200cm, number of turns about 5.

The volume of the electrolyte was about 2000ml. The cell was commercial glass (Pyrex type). The solution, according to our long experience in this field, was alcohol-water type (C<sub>2</sub>H<sub>5</sub>OH 85-90%, H<sub>2</sub>O 15-10%) electrolyte. Th(NO<sub>3</sub>)<sub>4</sub> (in 5% wt HNO<sub>3</sub>), SrCl<sub>2</sub> and Hg<sub>2</sub>SO<sub>4</sub> salts were added. Th and Sr salts were added at an amount of some tens of micromoles, and the Hg ion at an amount of only a few micromoles. The electrolytic current density was as low as 2-10mA/cm<sup>2</sup>. The anode-cathode voltage was 20-136Volts. During the experiment the cell temperature ranged between 20 and 60°C.

The Pd-Ag tube surface was previously mechanically cleaned (by fine, silica based, sandy soap), acetone rinsed, washed several times with distilled water, and finally treated at a temperature of about 450°C for a complete degassing.

Before the cathodic H loading operations, the surface was "activated" by proper cycles of anodic oxidation. Vacuum was made inside the tube cavity, connecting pipes, pressure gauges and valves.

We reached a maximum value of pressure inside the hollow cathode of about 8.5atm (absolute 9.5). The maximum value of 8.5atm was imposed by the mechanical strength limit of the 50µm wall of the tube.

We would like to note that the faradic efficiency, i.e. the amount of hydrogen gas produced by the electrolytic current (collected and pressurised inside the tube) reached values as large as 15--20%; the efficiency strongly depends on the Pd surface conditions, type of electrolyte, current density, and temperature.

In comparison, the device invented by Arata (using usual D<sub>2</sub>O - LiOH 0.1M electrolyte, tube 2.5mm thick) had efficiency on the order of 0.5--2.5%.

Arata was able to reach pressures as high as several hundred atmospheres; up to now, we

can't say anything regarding the capability of our electrolyte in obtaining such a high pressures.

In a further set of experiments we are going to use a thicker (0.250mm) ultra-pure (99.99%) Pd tube, specially developed for this purpose by ORIM SpA Company (Italy).

In conclusion:

- 2 The effectiveness of the original device developed by Yoshiaki Arata since 1955 was fully confirmed;
- 3 The surprisingly high Faradic yield for H-D loading in hydro-alcoholic electrolyte might open some ways toward practical applications;
- 4 The measurement of H<sub>2</sub> pressure in the hollow cathode directly confirms our previous results (H/Pd-D/Pd close to one, measured by the change in resistivity of the Pd cathode, and high faradic efficiency), obtained by electrochemical loading of long (typically 60cm) and thin (diameter 50µm) Pd wires in C<sub>2</sub>H<sub>5</sub>OH-H<sub>2</sub>O or C<sub>2</sub>H<sub>5</sub>OD-D<sub>2</sub>O electrolytes.

Following the positive results of our experiment we received the acknowledgement of a large part of the Italian scientific community formerly sceptical about all that concerns Cold Fusion.

Moreover, one of the most diffuse and influential (in Italy) INTERNET blog about environment & clean energy (<http://www.vglobale.it>) has adopted, as front page for 3 months, the photo of the experimental apparatus build at INFN-LNF. The number of down-loads of such photo (and 2 pages of explanations/comments) was quite large (increases of over 500 access/day of the blog, up to now). Such blog is certified by "spazio RP" and was established about 9 years ago.

## 1 Introduction and Motivations

It is well known that any kind of experiment, based on the anomalous effects (the so called “Cold Fusion”) in Condensed Matter Nuclear Science (CMNS) has, as a pre-requisite, the forced absorption of Deuterium (D) and or Hydrogen (H) inside or at the surface of some specific metals at values well over the thermodynamic equilibrium limits. Such metals (M) are usually: (for D loading) Palladium (Pd) and Titanium (Ti); (for H loading) Nickel (Ni).

The most diffuse method for H or D overloading is the electrolytic one.

\* The term “overloading” is used to point out that the value of the H-D/M ratio is larger than that is allowed at the normal equilibrium conditions. Consequently, the extra H or D forced inside the M is in a meta-stable condition, i.e. they were in general rapidly released when the loading driving force is removed.

### 1.1 The Baranowsky curve

In the case of Pd loading, by adopting a proper wire geometry, it is possible to evaluate the amount of H or D absorbed into the bulk of the metal through the relationship between the H-D/Pd ratio and the value of the resistivity of the actual (H-D)–Pd system (“Baranowsky curve”).

We remark that Prof. B. Baranowsky developed such curve since 1969 (further elaborated in 1982 and 1990) by measuring the H<sub>2</sub>-D<sub>2</sub> absorption in Pd rods (up to 50,000 bar), at a temperature close to ambient one. Because of the low diffusion speed due to the relatively large thickness of the used Pd samples, each set of experiment, from vacuum up to 50,000 bar, takes over 100 days of measurements.

It’s noteworthy that the gas pressure method is an experiment performed in *static equilibrium condition*, i.e. the Pressure-Concentration-Temperature relationships are always satisfied.

Recently (ICCF12) Dr. Mike McKubre (Ref.1) has performed a comprehensive review of such an argument.

### 1.2 Electrolytic loading

As previously remarked, the electrochemical Pd loading is the most frequently adopted technique to achieve high (H-D)/Pd ratios; in fact, the cathodic discharge of H-D ions on the Pd surface generally requires a certain over-voltage (with respect to the thermodynamic value for the water decomposition) that can be considered equivalent to a very high H-D pressure; in an open electrolytic cell. It is therefore possible to achieve in a simple way (H-D)/Pd loading ratios similar to that obtained in sealed cell operating at a very high H-D pressure. In this case open-cell means that the pressure inside the electrolytic cell is controlled as to maintain a small over-pressure (usually less than 0.1 bar) needed to avoid that the light water humidity (always present in the air) may contaminate the heavy water (or others deuterated compounds) of the electrolyte.

### 1.3 The Arata experiments

Very recently, during ICCF12 (November 27--December 2, 2005, Yokohama-Japan), Prof. Yoshiaki Arata (Osaka University), Ref. 2, explained in some details about his recent experiments: high-temperature & high-pressure diffusion of D<sub>2</sub> through a thick Pd rod with a

closed “reaction chamber” inside and, for sake of comparison, the previous experimental set-up (based on the forced diffusion of  $D_2$  through a thick Pd rod using an usual electrolytic open-cell).

\* We recall that the experimental set-up developed by Arata and co-workers was named “Double-Structures Cathode” (D-S Cathode) and in some experiments, after very long time (some months) of electrolysis at high current density (about  $500\text{mA}/\text{cm}^2$ ) he measured, inside the thick wall reaction chamber, that the pressure was very large (1000 bar and over); excess heat was detected as well as traces of  $^4\text{He}$ . No photo of the experimental set up, type and position of the pressure gauges are unfortunately available (1990-2004).

Mike McKubre, another well known researcher in the field of CMNS, claimed to have reproduced in USA, in 1999-2000, some of the anomalous effects found by Arata using a Pd cathode lent to him by Arata himself. Unfortunately, no gauge pressure was installed in the D-S Cathode, so no direct measurement was possible.

During ICCF12, Arata made known to everybody that the D-S Cathode experimental set-up was developed by himself since 1955, in the framework of Hot Fusion studies (at Osaka University), and later world-wide-patented in 1994 (Ref. 3). Moreover, at the beginning of the talk, he further specified that he developed such a “high pressure deuterium generator” because in 1954-1955, after the second World War, it was not possible to buy pressurised deuterium gas in Japan even for small amount needed for scientific Hot Fusion experiments. Likely, the Arata devices worked well enough to first observe D-D Hot Plasma Fusion in Japan in 1955 and he was (and still is) very proud of both such scientific and technological achievements.

We arbitrarily named the “work of high pressure deuterium generator” as the “*Arata\_1955 experiment*”.

\* The experimental results presented by Arata at ICCF12 were particularly impressive; he showed that it was possible to get continuous excess heat in a pressurised (about 70bar) Pd cell, where few grams of nano-particles of Pd dispersed in a matrix of  $ZrO_2$  are located. The estimated excess heat, by comparison with blanks, was on the order of 10W, at about  $190^\circ\text{C}$ . Despite the starting-up from the ambient temperature in somewhat heat-leaking condition, net input power to the reactor was not necessary to maintain excess heat production.

### **1.3.1 Distribution-in-Italy of Arata’s experimental reports and reactions**

The experimental results, together with the full patent of 1994, were immediately reported and diffused in Italy through a mailing-list (over 130 addresses, F. Celani is the manager) of people interested in Condensed Matter Nuclear Science. Obviously, most of the people inserted in the mailing-list judged the Arata results very interesting, although some criticized the CMNS experiments in-toto.

\* One of the Researchers interested in critics is an important and very influent Nuclear Chemist: Prof. Camillo Franchini. He was Director (over 20 years), of the Laboratory of Nuclear Chemistry and of the Nuclear Reactor installed at the “Centre for Military Applications of Nuclear Energy”, (CAMEN) S. Piero a Grado (near Pisa), Italy. Later, the CAMEN was renamed CISAM.

\* At present, he is the manager of another mailing-list whose main aim is to disprove most of the very innovative results coming out from several advanced fields of research, in Chemistry, Physics and Biology in particular.

BTW, he is one of the strongest opponents to the so-called “Italy-Japan Project” for a physical remediation of nuclear wastes (Ref.4). Aim of this project is to evaluate the effectiveness of the method developed (since 2001 and continuously improved) by Yasuhiro Iwamura (Mitsubishi Heavy Industries, Yokohama, Japan), using natural Sr and Cs (Ref. 5), also for the remediation of the radioactive ashes (i.e.  $^{90}\text{Sr}$ ,  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ ) coming out from nuclear power plant as fission products.

We recall that Iwamura showed transmutation with some amounts (of the order of  $10^{15}$  Atoms/cm<sup>2</sup>) of natural strontium (transmuted to stable molybdenum) and caesium (transmuted to stable praseodymium) using pressurised deuterium gas, permeating through a sophisticated multilayered sample of Pd-CaO-Pd, for about 2 weeks.

We remark that the mailing list of Prof. Franchini includes, among others, several politicians, editors of journals/newspapers, makers of scientific TV programs.

\* Prof. Franchini, together with other important University teachers, claimed (and, de-facto, shared such opinion to several thousand people) that both the Iwamura experiment (the transmutation of natural Sr and Cs) and the Arata one (compression of H, D by electrolysis) are just “*computer simulation experiments*” because they violate the rules of nature. In other words, both Iwamura and Arata, according to Prof. Franchini, deceived the whole scientific community and specially the Italians. In particular, Franchini and his colleagues claimed that it was impossible to get inside a hollowed and closed Pd cathode, using an open cell configuration (i.e. at 1 bar of external pressure), a pressure larger than few bars. If such situation would have happened, it means that Arata has invented the “Maxwell devil” and as a consequence he, and/or whoever else could reproduce the experiment, have to get immediately the Nobel Price.

Because of such critics several doubts/ironies grew in Italy coming from researchers/opinion-makers/industrialists. The meaning was: *all* the experiments, about Cold Fusion, were performed by incompetent or even cheating scientists.

### **1.3.2 Planning replication of the Arata\_1955 experiment**

As a consequence, we decided, at the end of February 2006 to start, as soon as possible, at Frascati National Laboratory of INFN, a scientific rush-program, in order to reproduce the Arata\_1955 claims, at least with a pressure inside the cell not larger than some bars.

\* At that time, the only Pd tube immediately available was a Pd-Ag alloy (Pd at 75%) tube with a very thin wall (only 50µm) and 10mm of diameter: it was manufactured by a cold-rolling and diffusion welding procedure, which is patented (Ref. 6). It was used, for long time, as high temperature (300-400°C) membrane for gas (hydrogen) separation-purification and was developed by Dr. Silano Tosti at ENEA-Frascati (Ref. 7, Ref. 8).

\* Moreover, thanks to long time lasting scientific collaboration, among CMNS, with ORIM SpA, we asked to prepare an extruded tube of ultra-pure Pd (>99.99%). ORIM is one of the

few Companies, in Italy, having long experience about melting and purification of precious metals, specifically Ag, Pd and Pt.

The reasons about the use of ultra-pure Pd tube are essentially:

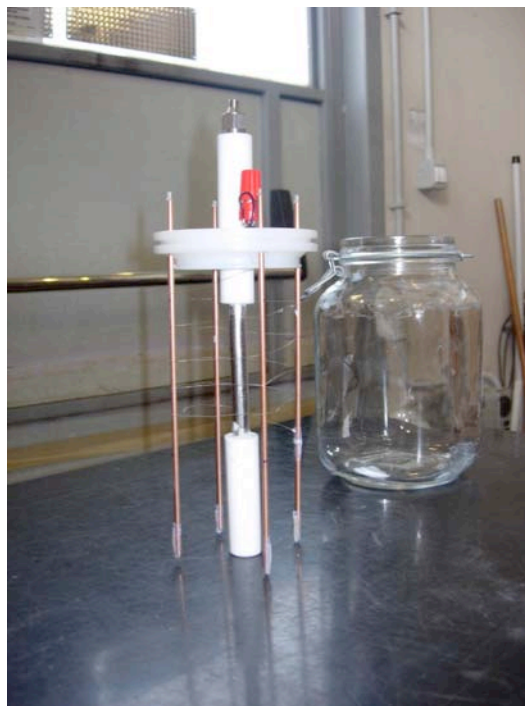
- to compare the results with respect to the Pd-Ag tube;
- to get higher values of pressures due to thicker wall (250 $\mu$ m instead of 50 $\mu$ m) and reduced diameter (8mm instead of 10mm).

The expected disadvantages of pure Pd tube, with respect to the Pd-Ag one are:

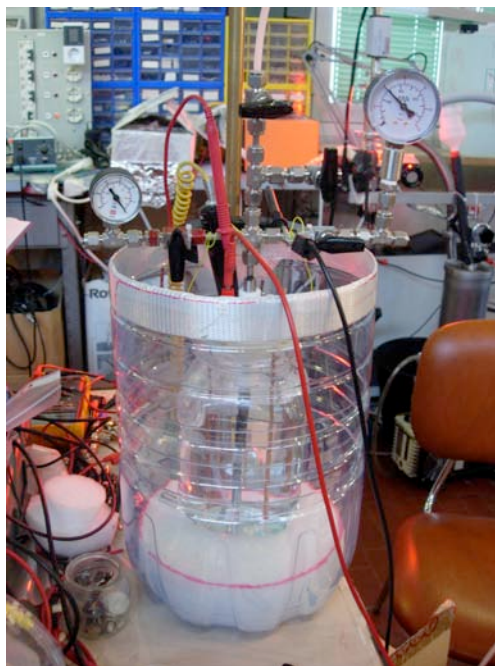
- worse mechanical performances (due to lower tensile strength and higher hydrogen embrittlement behaviour);
- lower diffusion speed.

## 2. Experimental Considerations and Set-up

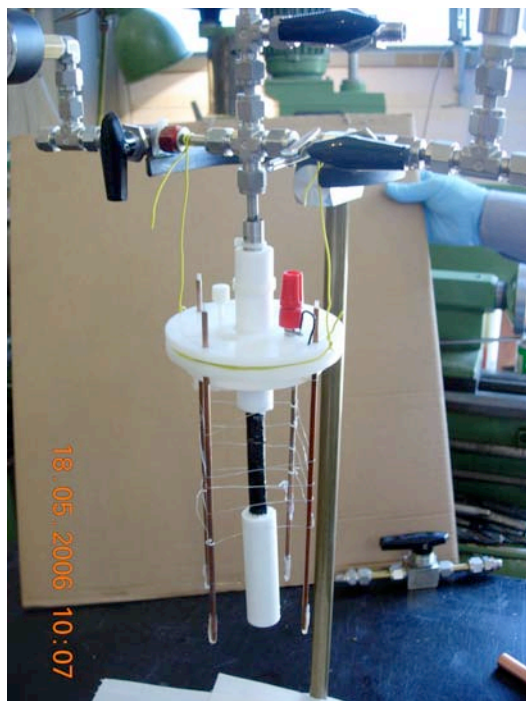
According to our scientific rush-program, we built an experimental apparatus as simple as possible. In Fig.1, 2 and 3 are shown photographs, respectively, of Pd-Ag tube before use, an overview of the whole set-up, the Pd-Ag tube at the end of the 3 months experiments.



*Fig.1 Photo of the Pd-Pt cell. At the centre there is the Pd–Ag tube, with 2 PTFE tube (top and bottom) to avoid contamination (to electrolyte) from SS connections. The anode is 250 $\mu$ m Pt wire, rounded on 4 Cu sticks covered by PTFE sheaths. The cell is just a commercial, Pyrex-type, bottle.*



*Fig. 2. Photo of the very simple experimental set-up in the first series of experiment (without electronic pressure gauge); The second vessel around the electrolytic cell, transparent plastic (0.5mm tick PET with some fibre –glass reinforcing tape) is filled with tap water and is used for safety reasons in the case of the Pd-Ag tube explosion.*



*Fig.3. Photo of the Pd-Ag tube after second series of experiments; The cathode, from silver colour, changed to dark one due to salts deposited at its surface.*



For pressure measurement, we used both mechanical (needle) gauges and electronic sensors (in the second series of experiments). The gas accumulated inside the tube, cf. Fig.2, can be collected into a very light, rubber balloon: it flies! This confines really full of hydrogen.

### 2.1 Mechanical characteristics of Pd and Pd-Ag

The maximum pressure allowable, taking into account the tube dimension and the tensile strength of the Pd-Ag, alloy can be evaluated according to the formula:

$$P=(t*\sigma)/r \quad (1)$$

Where P is the pressure (Pa), r is the tube radius (m), t is the tube thickness (m) and  $\sigma$  is the tangential stress (Pa) in the metal tube, respectively.

In Table-1 are reported, according to Ref. 11 and 15 quoted in Ref. 4, the tensile strength data of annealed and cold worked Pd and Pd(75%)-Ag(25%), at ambient and 400°C temperatures. The positive effect of Ag alloying and worsening of mechanical characteristics (for both Pd and Pd-Ag) due to large temperatures, are evident.

*Table-1 Tensile strength data of Pd and Pd-Ag, annealed or cold-worked, at ambient and 400°C*

Material	Tensile strength (MPa), T=ambient	Tensile strength (MPa), T=ambient	Tensile strength (MPa), T=400°C
Treatments →	Annealed	Cold-worked	Cold-worked
Pd	150	380	150
Pd(75%)-Ag(25%)	380	690	380

\* About the rupture pressure,  $P_r$  (MPa), for a 10 mm diameter tube, the calculated (Ref. 4) values, for a fully annealed and heavily cold worked tube, at ambient and 400°C, are reported in Table-2.

*Table-2: Calculated rupture pressure, in MPa, for 10mm diameter tubes (wall thickness of 50µm and 70µm), Pd (100%) and 25%Ag alloy, at ambient and 400°C*

	$P_r$ , annealed	$P_r$ , annealed	$P_r$ , cold-worked	$P_r$ , cold-worked
Temperature →	Ambient	400°C	Ambient	400°C
Pd, 50µm	1.50	0.67	3.78	1.68
Pd, 70µm	2.1	0.93	5.29	2.35
Pd-Ag, 50µm	3.79	1.68	6.84	3.04
Pd-Ag, 70µm	5.30	2.36	9.58	4.25

\* Taking into consideration that Pd-Ag in our usage worked for long time (over 6 months) at very different operating conditions (pressure, thermal stresses, embrittlement due to hydrogenation and thermal cycles), and adopting the usual safety factor (3) about pressurised devices, we decided that never we would allow a pressure larger than 9 bar (10 absolute).

## 2.2 H<sub>2</sub> Production and absorption

About the evaluation of hydrogen collected into the hollow cathode, we have to consider as follows:

- a) As a proton (H) inside the Pd lattice;
- b) As H<sub>2</sub> inside the cathode cavity.

- a) H absorbed inside Pd lattice:

Pd-Ag tube dimension: radius (R) 5mm; wall thickness 50μm; length (l) 10cm.

The amount of Pd gram atoms is:

$$\text{Pd (gram atoms)} = ((\pi * (R^2 - r^2) * l) * \delta) / M_w \quad (2)$$

Where:  $\delta$  =Pd density (12 g/cc);  $M_w$ = atomic weight (106 Amu)

$$\text{Pd (gram-atoms)} = (3.14 * (0.500^2 - 0.495^2) * 10) * 12 / 106 = 0.01769$$

By considering that the effective amount of Pd is 75% and that the expected H/Pd at 10 bar is about 0.80, the real amount of H gram-atoms in the Pd lattice are:  $0.01769 * .75 * .80 = 0.0106$

As a consequence, the moles of H<sub>2</sub> gas are  $0.0106 / 2 = 0.0053$

- b) Moles of H<sub>2</sub> inside the hollow cathode (filled with porous ceramic, because mechanical strength reasons), pressure gauges, valves, connections:

Total measured Volume = 18cm<sup>3</sup>.

For each bar of H<sub>2</sub> pressure the moles present in the cavity are (at NPT conditions)

$$18 / 22410 = 8.032 * 10^{-4} \text{ Moles of H}_2 \quad (3)$$

The Moles of H<sub>2</sub> gas produced during the electrolysis are proportional to the total amount of charge Q (Coulomb, C) involved:  $Q = It$ , if I is constant.

Generally,  $Q = \int I dt$ , over time t, of I.

In the formula I is the electrolytic current in Ampere (A), t is the time in seconds (s).

The formula about Moles H<sub>2</sub> produced is:

$$\text{Moles H}_2 \text{ produced} = Q / (2 * F) \quad (4)$$

In Formula #4, F is the Faraday constant (96480 C/Moles); the factor 2 is due to the fact the molecule of Hydrogen is H<sub>2</sub>.

\* The efficiency of the total hydrogen collection ( $\eta$ ) is evaluated by the ratio between the sum of H gram-atoms (calculated as H<sub>2</sub>) present in the Pd lattice (formula #2) plus the moles present in the cathode cavity (formula #3) over the gas produced during the electrolysis (formula #4).

Separately, it is given the efficiency about the “useful” gas produced, i.e. neglecting the contribution of H-atoms absorbed by the Pd lattice. Such value of  $\eta$  is, obviously, lower than the total one. In our experiments we compare only such, very conservative, values.

### 3 The cell, electrolytes and experimental results

\* The cell was just a commercial glass, Pyrex type.

All the other materials used inside the cell were protected by PTFE tube or sheath.

The cell was completely open to environment through a hole for gas escaping.

\* Following our long experience (Ref. 9, Ref. 10) on unconventional electrolytes that were able to load thin Pd wires at high value of H, D/Pd, within enough short time, we decided to use our “standard” receipt (ethyl alcohol-water mixtures, concentrations respectively 85-90% and 15-10%) with some amounts of salts of  $\text{Th}(\text{NO}_3)_4$  in 5%wt  $\text{HNO}_3$ ,  $\text{SrCl}_2$ ,  $\text{Hg}_2\text{SO}_4$  at some micro-molar concentrations. The  $\text{Hg}_2\text{SO}_4$  was added at a concentration about ten times lower than Th and Sr. The only important variation, in respect to our usual salt addition using 50 $\mu\text{m}$  tick, 60cm long wire (about 1 $\text{cm}^2$  of surface) was to increase by about a factor 10 of the amounts of salts in order to compensate (partially) for larger surface (exposed area about 27 $\text{cm}^2$ ) of Pd-Ag tube. Moreover, the total amount of liquid solution was about 2000cc in respect to usual 750-1100cc. The pH was adjusted to about 3 adding, *slowly*, some amounts of, diluted in  $\text{H}_2\text{O}$ ,  $\text{HNO}_3$ .

The anode was a pure (99.99%) Pt wire (diameter 250 $\mu\text{m}$ , length about 200cm) rounded on the cathode, see Fig. 1 and Fig. 3. The anode-cathode distance was about 4cm.

\* Before starting cathodic regime on Pd-Ag, there were made several cycles of anodic-cathodic conditions in order to activate the Pd-Ag surface.

We make experiments with current density between 2 and (about) 10 $\text{mA}/\text{cm}^2$ .

The temperature ranged from 20 up to 60°C, depending on current density and conductivity of the solution.

\*We experienced a “conversion factor efficiency” of  $\text{H}_2$  gas stored inside the tube (see discussion of § 2.2, points a) and b)), in respect of that produced (because electrolysis, see Formula #4) typically of 6-8% (Fig.4).

Observing in some details the behaviour of second spontaneous degassing, Fig. 4, at time 420000 (current removed), we can easily understand that the Pd-Ag is very permeable even at room temperature: between 6 and 9.5bar in absolute values, the loading and de-loading times are quite similar.

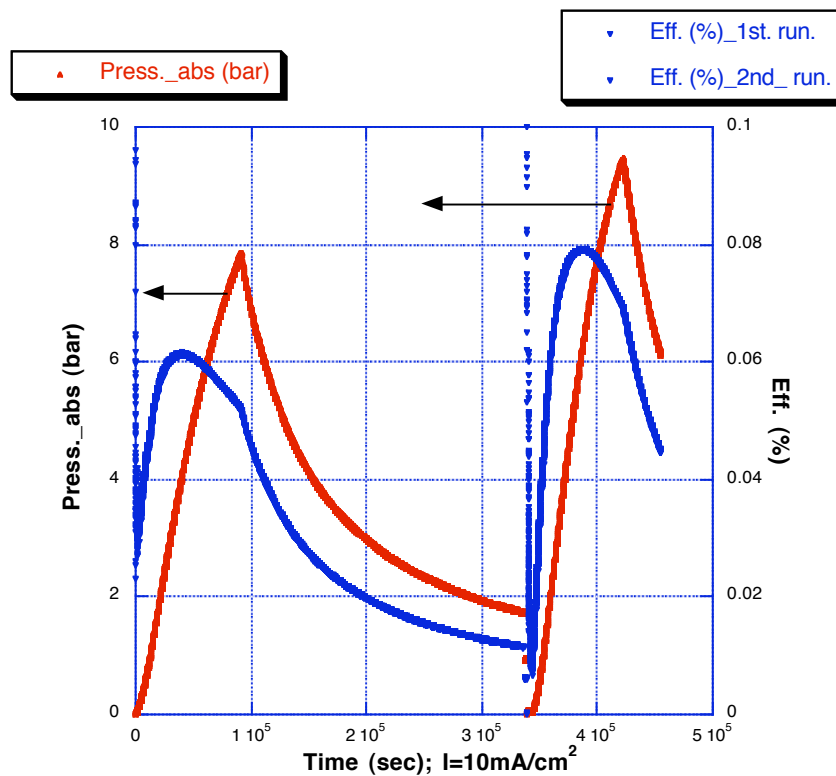
As a consequence, in order to further increase the gas collection efficiency, it can be enough to increase the current density, now at only 10 $\text{mA}/\text{cm}^2$ .

\* The best value up to now obtained, about  $\eta$ , was larger than 18% (Fig. 5) at the beginning of the experiment when the cathode was just cleaned and properly activated. At that time we were very prudent about pressure increase: we stopped the electrolysis when the pressure reached a value of over 3.8bar relative to ambient.

\* The pressure value of 3.8bar was judged sufficient to demonstrate that *Arata* was a **fully honest Scientist** and that *Franchini* (together with some of his University Colleagues) **made completely wrong assertions**.

\* We realised that the efficiency depended strongly on: electrolyte compositions, surface conditions of Pd-Ag, current density, and temperature. Such conditions are very similar to what we found using thin and long Pd wires.

As a comparison, the efficiency of original Arata device, using D<sub>2</sub>O and LiOH 0.1Molar, was of the order of 0.5-2.5%. Anyway, Arata was able to get pressure as high as 1000bar (using very thick Pd, several mm) and, up to now, we get a maximum pressure of only 8.5bar (value due to mechanical limits of 50µm Pd-Ag tube).



*Fig.4. Second series of experiment: electronic gauge pressure (apart the usual mechanical “needle” gauge in parallel). Behaviours of pressure and H<sub>2</sub> gas efficiency, versus time. At time 95000sec the electrolysis was interrupted and the tube was allowed to degas for over 2 days. The second loading cycle, at the same current density, gave better results about pressure and gas loading efficiency (improved from 5.5% at 7.8bar to about 7.5% at 9.5bar). At time 420000sec the electrolysis was interrupted and was studied, again, the degassing behaviour.*

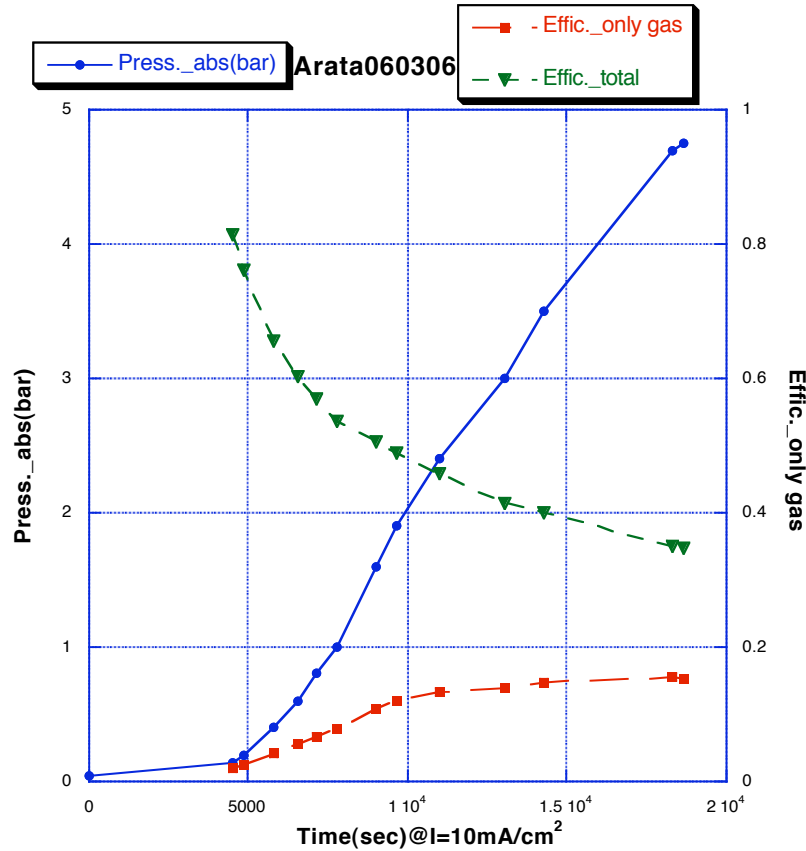


Fig. 5. Behaviour of pressure increasing, from vacuum, at the beginning of the experiment; The current density was  $10\text{mA}/\text{cm}^2$ . They were shown also the efficiencies of absorption, considering both the total amounts of hydrogen dissolved in the Pd lattice and the only gas in the chamber (the so called  $\eta_{\text{useful}}$ ).

#### 4 Conclusions

- 1) The effectiveness of Arata procedure was fully proven by an experimental set-up, which was done by Researchers/Country completely different from original one.
- 2) Within the limits of pressure up to now explored (up to 9.5bar in absolute value), we have found a conversion efficiency, of  $\text{H}_2$  collected inside the hollow cathode, about 5-10 times larger than that reported by Arata in his experiments.
- 3) The method of gas collection inside a hole is a powerful method to demonstrate directly (and without further questions) that also the electrolysis is able to load, at large amounts (over the thermodynamic limit) of Hydrogen or Deuterium inside proper materials. In other words, also the original claim of M Fleischman and S. Pons about possible nuclear fusion using Pd rods that underwent electrolysis, in  $\text{D}_2\text{O}$ -LiOD solution for long time, can be reinforced by this experiment.

4) The next experiment, now in the calibration state, with ultra-pure (>99.99%) Pd, will add very precious information on the peculiarity of H, D forced absorption (and spontaneous desorption) by electrolysis.

5) We have to thank Prof. Camillo Franchini, and his University Colleagues (at Rome and Pisa), that, de-facto, forced us to reproduce the Arata\_1955 experiment: we learn, by experiments, very precious information (some of them not written in the text books) that we are happy to share between the world wide CMNS Researchers.

## 5 Acknowledgements

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## References

- 1) M.C.H. McKubre, F.L. Tanzella: *Using resistivity to measure H/Pd and D/Pd loading: method and significance*, Invited Paper at ICCF12, Nov.27-Dec.2, 2005, Yokohama, Japan, to be published by World Scientific Publ. Co., 2006.
- 2) Yoshiaki Arata, Yue-Chang Zhang: *Development of “DS-Reactor” as the practical reactor of “Cold Fusion” based on the “DS-cell” with “DS-Cathode”*, Invited Paper at ICCF12, Nov.27-Dec.2, 2005, Yokohama, Japan, to be published by World Scientific Publ., 2006.
- 3) Yoshiaki Arata: *Method of producing ultrahigh pressure gas*, Japan Patent 6-324459, December 27, 1994, United States Patent: 5,647,970. July 15, 1997.
- 4) A. Takahashi, F. Celani, Y. Iwamura: *The Italy-Japan Project: fundamental research on cold transmutation process for treatment of nuclear wastes*, Invited Paper at ICCF12, Nov.27-Dec.2, 2005, Yokohama, Japan, to be published by World Scientific Publ., 2006.
- 5) Y. Iwamura, T. Itoh, M. Sakano, S. Sakai, S. Kuribayashi: *Low energy nuclear transmutation in condensed matter induced by D<sub>2</sub> gas permeation through Pd complexes: correlation between deuterium flux and nuclear product*, ICCF10 Conference Proceedings, 2003. pgg. 435-446, World Scientific Publ., ISBN 981-256-564-7 .
- 6) Tosti S, Bettinali L, Lecci D, Marini F, Violante V: *Method of bonding thin foils made of metal alloys selectively permeable to Hydrogen, particularly providing membrane devices, and apparatus for carrying out the same*, Italian Patent IT20RM00412, 25 July 2000, European Patent 1184125 A1, 12 July 2001.

- 7) S.Tosti, L. Bettinali, V. Violante: *Rolled thin Pd and Pd-Ag membranes for hydrogen separation and production*, International Journal of Hydrogen Energy 25 (2000) 319-325.
- 8) S. Tosti: *Supported and laminated Pd-based metallic membranes*, International Journal of Hydrogen Energy 28 (2003) 1445-1454.
- 9) F. Celani, A. Spallone et al: *“Innovative procedure for the, in situ, measurements of the resistive thermal coefficient of H(D)/Pd during electrolysis; cross-comparison of new elements detected in the Th-Hg-Pd-D(H) electrolytic cells”*  
ICCF11 Conference Proceedings 2004, pp. 108-12.  
World Scientific Publ., ISBN 981-256-640-6.
- 10) F. Celani, A. Spallone et al.: *“New procedures to make active, fractal like, surfaces on thin Pd wires.”*  
Invited Paper at ICCF12, Nov.27-Dec.2, 2005, Yokohama, Japan, to be published by World Scientific Publ., 2006.