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**EXPERIMENTAL STUDIES TO ACHIEVE H/PD LOADING RATIO CLOSE
TO 1 IN THIN WIRES, USING DIFFERENT
ELECTROLYTIC SOLUTIONS**

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Abstract

Systematic studies have been performed in order to achieve very high concentration of Hydrogen (or Deuterium) into a Palladium lattice.

In a very diluted acid electrolytic cell a thin Pd cathode wire (100 μm) and thick anode Pt wires (0.5 mm) has been used as electrodes in a coaxial geometry. Normalised resistance (R/R_0) of Pd-H wire system has been measured on-line and used as reference of H/Pd values.

Alcoholic solution (95%) and electrolytic solution (5%) has been used with addition of a very low amount of Sr and Hg ions; high loading results have been achieved with a satisfactory grade of reproducibility.

A typical result of this procedure is to have achieved, stable in the time, $R/R_0=1.15$ (corresponding to an estimation of $H/Pd \approx 1$) with an extremely low electrolytic power supply (7 V, 5 mA).

Transferring this procedure to Deuterated solution, until now a lower loading has been achieved ($R/R_0= 1.55$ as about $D/Pd= 1.97$).

1. INTRODUCTION

Many researchers agree that, in cold fusion experiments, in order to obtain stable and reproducible excess heat, it is necessary to achieve and maintain very high D/Pd ($\gg 0.85$) loading ratios [Ref. 1]. The poor results, especially from the point of view of their reproducibility, generally achieved by the conventional electrolytic techniques, based on the use of LiOH solutions, induced us to develop a completely different approach. In fact, in previous studies, we have reported a reproducible procedure to achieve very high loading using Hydrogen and Palladium wires ($H/Pd \cong 1$) [Ref.s 2,3]. This procedure is based on the increasing of the cathodic over-voltage (which is known to be the main controlling parameter of the H(D) Pd loading) modifying the nature of the cathode, by inducing the formation of a very thin layer of an alkaline-earth carbonate on its surface.

After these encouraging results, we operated the same procedure just substituting light water (H_2O) with heavy water (D_2O), but unsatisfactory results have been obtained ($D/Pd \leq 0.85$); this unsuccessful has been explained [Ref. 2] by the presence of many impurities into the D_2O consisting of many elements, organic substances and bacteria which are interfering with the above reported surface substrate.

In order to overcome this problem, several attempts have been performed purifying the D_2O and reaching interesting values ($D/Pd \leq 0.95$, see Fig. 5), but not still satisfactory. Finally we decided to change approach, changing the nature of the electrolytic solution from the aqueous one to an alcoholic one. In this way, the amount of impurities present into the D_2O are reduced to a few percents and moreover alcohol might reduce the bacteria activity.

This paper shows these new tests about H/Pd over-loading by mean alcoholic solutions.

2. EXPERIMENTAL APPARATUS

A schematic diagram of the experimental set-up is shown in Figure 1. The cylindrical electrolytic cell consists of a central Pd cathode (a thin wire - diameter 100 μm - length 20 cm), a 4 surrounding Pt anodes of the same length (diameter 1 mm) set at a distance of $1.5 \div 2$ cm from the cathode, both plunged in a glass beaker filled with about 400 ml of electrolytic solution. The cell is placed into a two litre water bath in order to reduce the temperature fluctuations around the room temperature (usually kept at 22 $^{\circ}C$). Cell, bath and room temperatures are continuously monitored and recorded.

The power supply can operate as a constant D.C. current generator or, optionally, as a constant D.C. voltage generator. The cathode is grounded. The voltage is applied to the anode electrode through a home-made impedance adapter circuit (impedance booster), in order to avoid a current feed-back from the A.C. measuring circuit. The latter is essentially composed by a pulse generator (having sinusoidal, square and triangular wave selection; we always used sinusoidal wave) and a ground home-made coupling circuit (ground return, both for the D.C. and A.C. generators).

The D.C. generator can be applied to the edge points A or B of the anodes (electrically connected) whereas the A.C. generator can be applied to the edge points C or E of the cathode. The Pd wire voltage pick-ups C,D,E allow for the measurements of the resistances of the correspondent wire segments (CD= down, DE= up, both of length of 10 cm).

The loading ratio is inferred by means of the well known relationship [Ref.s 4,5,6] between resistance and H(D) content in the Pd matrix (Fig. 2) and continuously monitored.

The Pd cathode, because of its favourable surface/volume ratio, allows for a faster Hydrogen absorption and its high resistance (about 8Ω) improves both the accuracy and the precision of the measurements. Furthermore, the $1/r$ dependence of the electric field around the wire allows for a sharp increase of the pH value just in its proximity, thus promoting the carbonate precipitation in that region only [for more details see: Ref. 3].

3. COMPOSITION OF THE ELECTROLYTE

The electrolyte is mainly composed by alcohol (typically, ethyl: C_2H_5OH or, in a few tests, methyl CH_3OH) as $95 \div 99\%$ of the electrolytic solutions and HCl ($10^{-2} M$) + H_2SO_4 ($10^{-2} M$) as $5 \div 1\%$. Very small amount of $SrCO_3$ (a few mg) and $HgCl_2$ (about $10^{-2} M$), depending by the test performed, were added to the solution (motivations and details of this addition are still reported in Ref. 3).

4. EXPERIMENTAL TESTS

Electrolysis tests have been performed using constant current (in the range of $5 \div 10$ mA) with a voltage drop of about $10 \div 15V$ between the electrodes (corresponding to a electrolytic resistance ranging from 1.5 to $3 K\Omega$).

The procedure used was to load the cathode firstly at low current; in case of the loading ratio did not achieve spontaneously high values ($R/R_o < 1.5$) therefore several electrolytic current cycles (OFF/High/low current) were performed [Ref. 3].

A) - Pd-H loading experiments

In few tests occurred a situation as reported in Figure 3 in which the electrolytic solution was composed by: Ethyl= 400cc + HCl = 2cc + H_2SO_4 = 1cc + $SrCO_3$ = 20mg + $HgCl_2$ = 0.8cc. Quickly (about 1 hour) the normalized resistance reached the peak (H/Pd = 0.75) and slowly (about 10 hours), spontaneously, a high loading (R/R_o = 1.3 corresponding to $H/Pd > 0.95$ for both wire segments “up” and “down”) was achieved. The electrolytic current slowly decreased (from 5 mA to 1 mA) because the electrolysis resistance increased of 5 times (voltage generator was constant at about 10V); that can be consistent with a deposition of Sr^{++} and Hg^{++} ions on to the cathodic surface.

In some other tests occurred a situation as reported in Figure 4 in which the solution was composed by: Ethyl= 395cc + HCl = 21cc + H_2SO_4 = 2.5cc + $SrCO_3$ = 17mg + $HgCl_2$ = 0.25cc.

Quickly (about 20 minutes) the normalized resistance reached the peak (H/Pd = 0.75) and also in very short time (a few minutes) it saturated at R/R_o about 1.6; in this case, to improve the loading a current cycle was performed:

- **OFF** (until a loading discharge to the peak) / **High**= 25mA (until the loading saturation at R/R_o =1.6)
- **OFF** (as above) / **Low**= 2mA (constant for long time);

After that, slowly (about 6 hours) and spontaneously, a high loading (R/R_o = 1.15 corresponding to $H/Pd > 0.95$ for both wire segments “up” and “down”) was achieved (voltage was constant at about 10V).

B) - Preliminary Pd-D loading experiments

Firstly, loading tests using purified D₂O have been performed but we had to notify the difficulty to achieve relevant loadings and the low reproducibility of that; the best result is reported in Figure 5 where the electrolytic solution was composed by: D₂O= 385cc + DCI= 2cc + SrCO₃= 17mg (no Hg added). In a few hours a R/Ro= 1.6 (D/Pd= 0.95) value has been reached and we also operated OFF/ON current cycles but without important improvements.

After that we tried to repeat the previous tests (operated with Hydrogen-Alcohol solution) but performing a substitution with Deuterium-Alcohol solution. The result is reported in Figure 6 in which the solution was composed by: Methyl= 350cc + D₂O= 30cc + DCI= 20cc + D₂SO₄= 3.5cc + SrCO₃= 17mg (no Hg added). The loading improved a little in respect to the previous one (R/Ro= 1.55 → D/Pd= 0.97) but still below our expectations. Similar tests with the Hg addition (HgCl₂= 0.25cc) and OFF/ON current cycles operation, did not provide better results. Anyway in this case the result was reproduced in other tests.

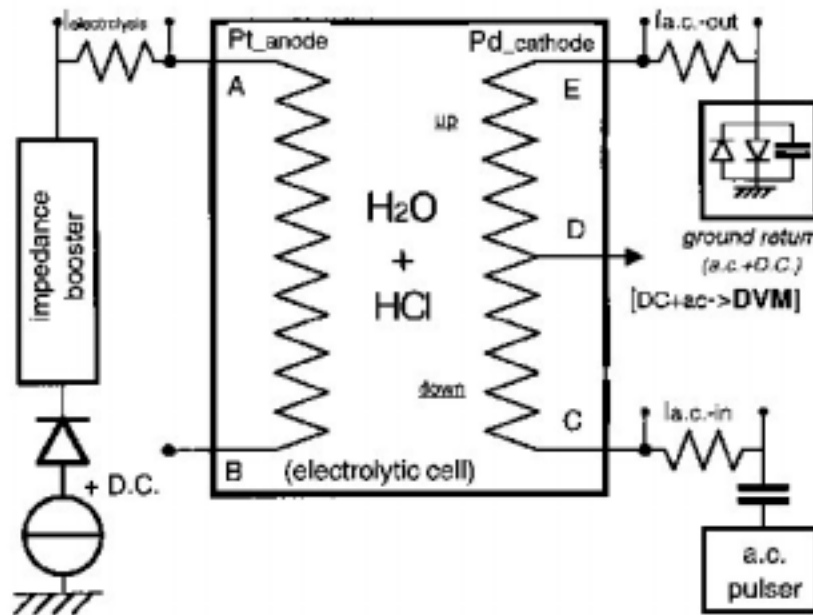


Fig. 1 – Electrolytic cell: a schematic view.

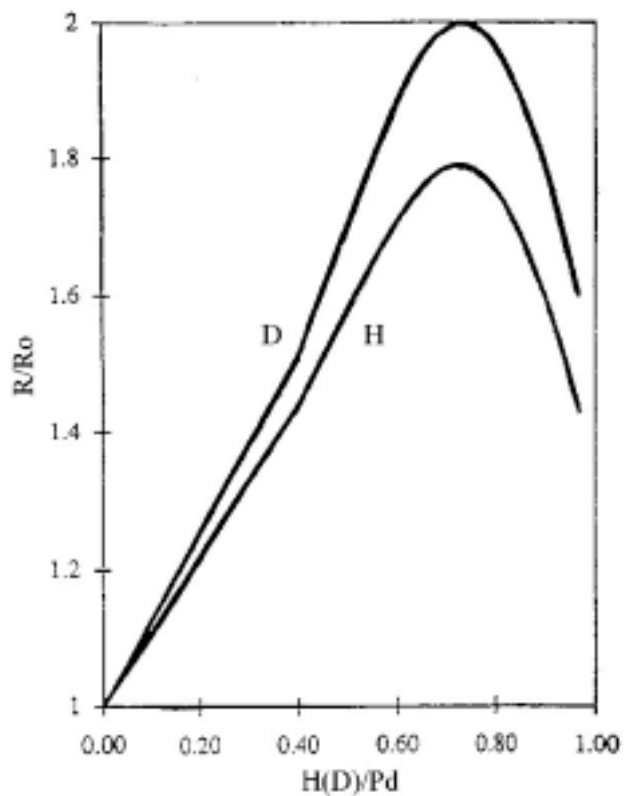


Fig. 2 – Normalized Resistance R/R_o vs $H(D)/Pd$.

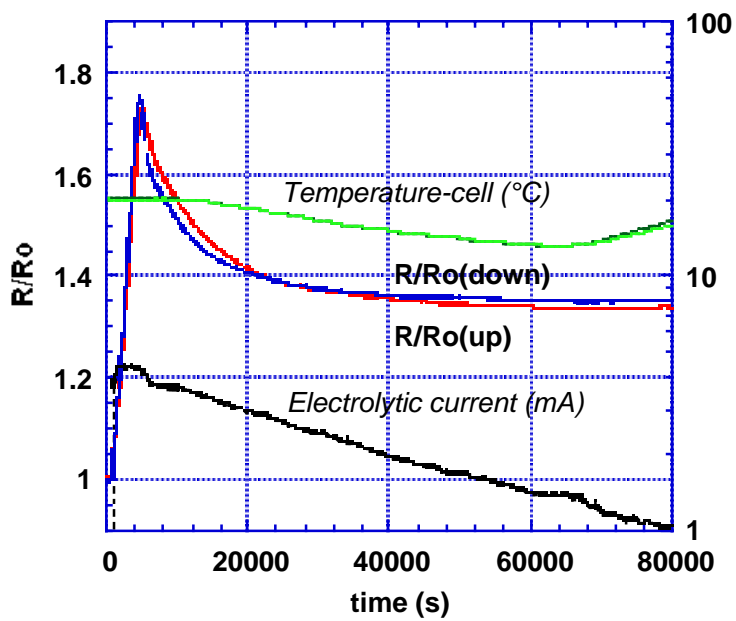


Fig. 3–Ethyl solution H/Pd loading: constant current.

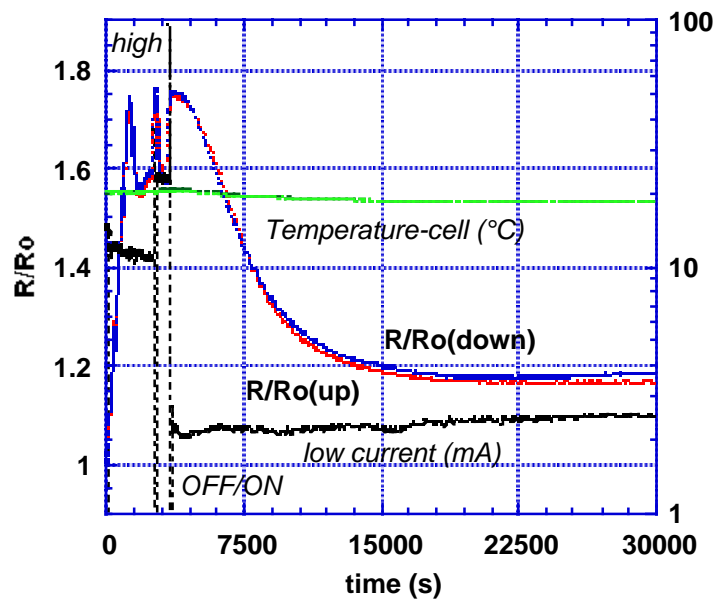


Fig. 4 – Ethyl solution H/Pd-load: OFF/ON cycles.

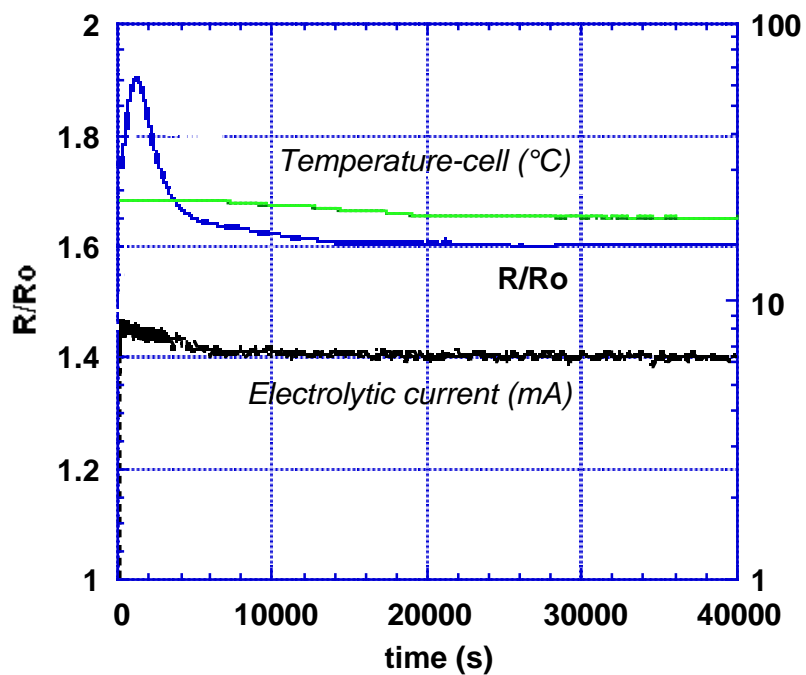


Fig. 5 – $\text{D}_2\text{O} + \text{Sr}$, aqueous electrolysis D/Pd loading.

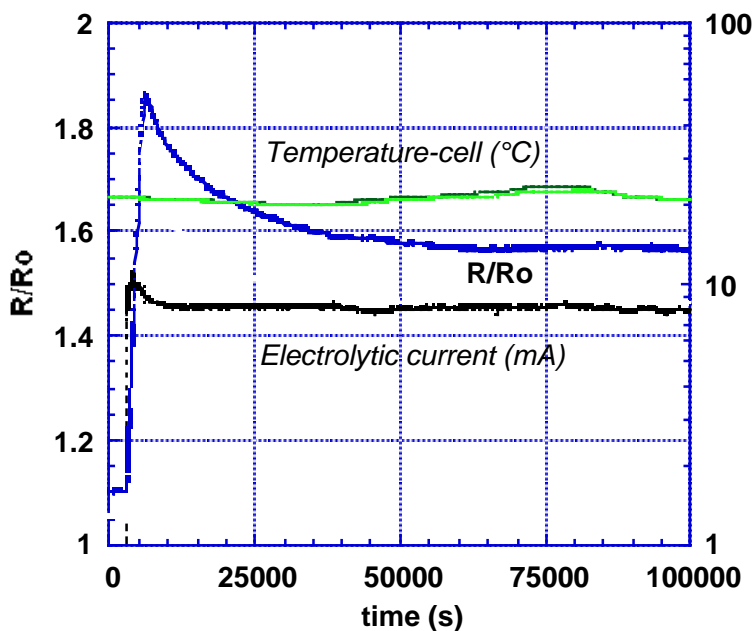


Fig. 6 – D-Methyl alcohol solution D/Pd loading.

5. DISCUSSION

Taking in account also previous studies (performed during last years), we can confirm that our procedure to over-load Pd-H system at 1:1 is effective using both aqueous-solution and alcohol-solution electrolysis. This method is based on an opportune alkaline elements thin deposition onto the Pd cathode surface (independent by the wire section); sometimes OFF/ON current cycles can improve the loading.

This method seems to work “partially” with Pd-D system; we conjecture that the obstacles limiting the over-loading to Pd-D system are due to impurities present into the D_2O and we are looking for solutions to overcome this problem.

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