CORRELATED MEASUREMENTS OF D$_2$ LOADING AND ^4He PRODUCTION IN *Pd* LATTICE

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Abstract

A series of measurements have been performed on a thin sheet of *Pd* in *D$_2$* gas atmosphere with an electric field applied across its length. The results in terms of correlation between the average loading ratio near the cathode and in the middle of the sheet are reported: they indicate that under particular conditions the application of the electric field succeeds to shorten the loading time and to obtain local *D/Pd* ratios higher than the average value.

1 Introduction

Since 1994 the Torino Group has looked for the ^4He production in *D/Pd* systems by using an apparatus consisting of a vacuum-proof cell, an ultra-high vacuum circuitry and a high resolution spectrometer. Inside the cell a thin strip of *Pd* is shrunk at both ends by two *Cu* electrodes through which an electric field can be applied. The operating principle of the system is the following. After the circuitry and the cell have been emptied up to a high vacuum pressure, a known amount of *D$_2$* gas is admitted into the cell at room temperature. The deuterium, after atomization and ionization at the surface, penetrates inside the *Pd* sheet until the atomic loading ratio reaches the equilibrium value. By applying an electric field along the biggest dimension of the sheet the d^+ ions are forced to migrate toward the cathode where they accumulate. In order to avoid that this locally high concentration causes the deuterium to desorb out, the ends of the sheet are coated by gold which inhibits the flow out. The mean value of the loading ratio of the *D/Pd* system can be easily evaluated from the amount of *D$_2$* initially admitted into the cell and from the temperature and pressure of the gas that are continuously monitored by gauges and thermocouples or thermoresistances. The vacuum circuitry provides to fill in and empty out the cell with D$_2$ as well as with the other calibration gases. The high mass spectrometer is used to detect the gas components and their amounts before and after the admission into the cell. With this apparatus a lot of experiments have been performed in 1995 and 1996 aiming both to look for the most effective conditions (gas pressure, electric field intensity and duration and so on) for obtaining a high loading ratio and to detect the presence of ^4He after the operation in the cell. The results, presented at the previous Cold Fusion conferences (ICCF5 and ICCF6) can be summarized as follows: a) the electric field plays the most crucial role in loading the *Pd* sheet and b) the production of ^4He was observed in 2 measurements over 6. On the basis of these observations, a set of measurements were planned in 1997 with the double goal of increasing the average loading ratio over the equilibrium point with suitable electric fields and of detecting any eventual ^4He excess inside the D$_2$ gas after operated in the cell. This second part of the program could not be fulfilled

due to a partial lack of the financial support which didn't allow the maintenance of the spectrometer; thus only the description and the results concerning the loading technique will be reported in the following.

2 Experimental set-up and measurement procedures

The cell containing the *Pd* sheet (8 cm long, 1 cm wide and 100 μιη thin) was accurately emptied by means of a turbomolecular pump up to a high vacuum of $2 \cdot 10\text{\textasciitilde}^6$ mbar (with running pumps): the vacuum reduces to $2 \cdot 10^{-5}$ mbar after the pumps are switched off. The temperature inside the cell was monitored by 2 thermoresistances T_m and T_c in contact with the *Pd* sheet in the middle and near the cathode respectively and by a thermocouple T_g in the vacuum/gas region. A pressure gauge monitored continuously the pressure P_g of the atmosphere around the sheet. An amount of n_0 moles ($n_0 = 4.89 \cdot 10^{-2}$) of D_2 gas, corresponding to a pressure of 3.2 bar at 20°C in a volume of 373.2 cm^3, was suddenly immitted in the cell and the absorption into the *Pd* matrix started. The heat, produced in this exothermic process, was taken out by a cooling system in contact with the lower surface of the sheet. The average loading ratio $a = D/Pd$ was continuously monitored by measuring T_g and P_g and using the gas equation for calculating the missing D_2 gas in the cell. Obviously, the local concentration $c(x, t)$ along the sheet length χ at the time t could not be measured: in order to have information about it, a model of the absorption and diffusion of the d^+ ions inside the *Pd* matrix under the effect of an applied electric field E has been developed. The absorption/diffusion equation looks like:

$$\frac{\partial c(x,t)}{\partial t} = \frac{\partial^2 c(x,t)}{\partial x^2} - v \frac{\partial c(x,t)}{\partial x} \theta_{el}(x) + K(x,c)[c_s - c(x,t)] \tag{1}$$

where:

$$K(x,c) \equiv \theta_d(x) \cdot [k_{in}\theta_{ass}(c_s - c(x,t)) + k_{out}\theta_{ass}(c(x,t) - c_s)] \tag{2}$$

$c(x, t)$ = local concentration of d^+ in *Pd* at time t;
$D = d^+$ diffusion coefficient in *Pd* matrix;
k_{in}, k_{out} = absorption, desorption coefficients of D_2 in *Pd*;
v = ion velocity under electric field in *Pd* lattice;
cs = thermodynamical equilibrium concentration of d^+ in *Pd* (~ 0.67);
$\theta_{el}, \theta_d(x), \theta_{ass}$ = step functions, taking into account the field and coating geometry.

Integrating over the sheet length one obtains the average loading ratio a:

$$\alpha(t) = \frac{1}{L} \int_0^L c(x,t)dx \tag{3}$$

The parameters D, K_{in}, K_{oui} were evaluated by fitting a to the data of 1995 and 1996: the predictions of eq. 3 for the experimental conditions of the 1997 set-up were generally in a good agreement with the measurements.

After 5' of absorption, an electric field was applied along the sheet length by means of a current generator connected to the electrodes at both ends of the sheet. A general view of the current intensity and duration along the time is shown in Fig. 1, where three different intervals, called FIELD I, FIELD II and FIELD III, are indicated.

In FIELD I the current intensity rose up to ~ 90 *A* in ~ 10', in 3 steps, remained constant for ~ 15', then decreased down to 0 in ~ 10' in 2 steps. The corresponding voltage V, measured at the electrodes, rose up to ~ 0.75 *V* and then decreased to 0. In this way an electric field of ~ 1 *V/cm* maximum was applied across the sheet length.

In FIELD II, after the generator was switched-off for ~ 12 hours, the procedure was repeated: in this case the current rose up to ~ 170 *A* in ~ 20' in 9 short steps and then decreased symmetrically to 0. The electric field reached a maximum of ~ 4 *V/cm* but the ratio to the current didn't look constant, indicating some strong variation of the *Pd* sheet resistivity.

Finally, in FIELD III, after ~ 1 hour of switched-off current, the procedure restarted and the current rose up to ~ 200 *A* in ~ 2 hours, in 4 long steps: the electric field rose up to ~ 3 *V/cm* in the first long step, decreased suddenly to ~ 1.3 *V/cm* during this step and then increased as the current increased up to a value of ~ 2 *V/cm*. At this point the generator was switched-off and the *Pd* sheet cooled down to room temperature.
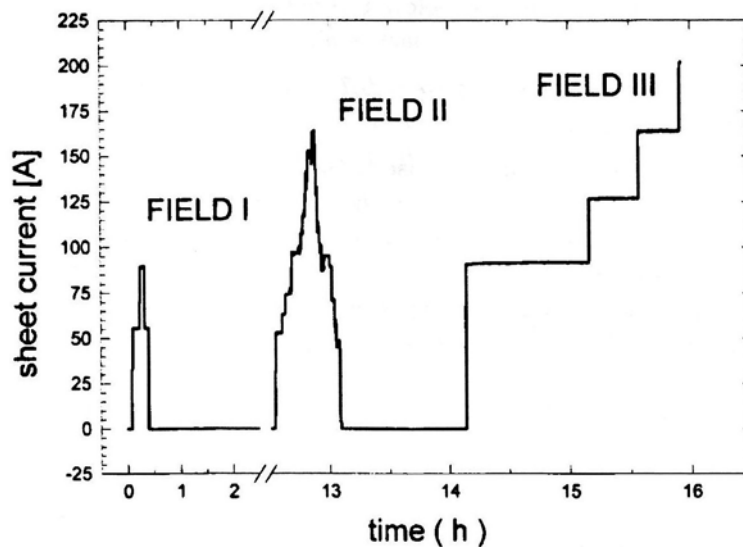

Figure 1: General view of the current applied across the Pd sheet as a function of the time.

The current generator was again switched on some times with the cooling system switched off in order to heat the *Pd* and to produce a complete desorption of the (eventually) produced [4]*He*. At this stage the *D-2* gas (and the produced [4]*He)* were extracted for the mass analysis. As said before, the results of this analysis were affected by a large error due to the resolution of the not maintained spectrometer. In the next section we shall analyse only the results of the temperatures and loading ratios measured during the application of the electric field.

3 Correlation of the loading ratio, temperature and electric field

During the application of the current, the voltage at the electrodes, the temperatures T_m and T_c of the sheet and the mean value α of the concentration were scanned at a frequency

of 0.25 hz. Let's analyse the compared behavior of these variables for each application of the electric field.

- FIELD I: the current is applied for ~ 25' in 5 steps with a maximum at ~ 90 A;

    • V follows linearly the current, indicating a constant resistivity of the Pd, unless for a short non linear behavior near the maximum current;
    • a increases monotonically up to 0.4: the slope is as steeper as the current is higher;
    • $T_c > T_m$ everywhere, suggesting a higher Pd resistivity near the cathode.

- FIELD II: the current is applied for ~ 33' in 18 steps with a maximum at ~ 160 A;

    • V follows linearly the current before the maximum, then drops down abruptly and becomes linear again in the last ~ 5';
    • α increases monotonically up to ~ 0.7 before the highest current step, then decreases down to 0.4;
    • $T_m > T_c$ everywhere and both rise to 80°C and 190°C respectively when the current reaches the maximum, then they suddenly decrease indicating that the cooling system is not effective for fastly increasing currents; the high temperature could produce some microcracks in the middle of the sheet through which the deuterium flowed out, as suggested by the behavior of a; also the voltage fall could be explained by these combined effects.

- FIELD III: the current is applied for ~ 100' in 4 steps with a maximum at ~ 200 A;
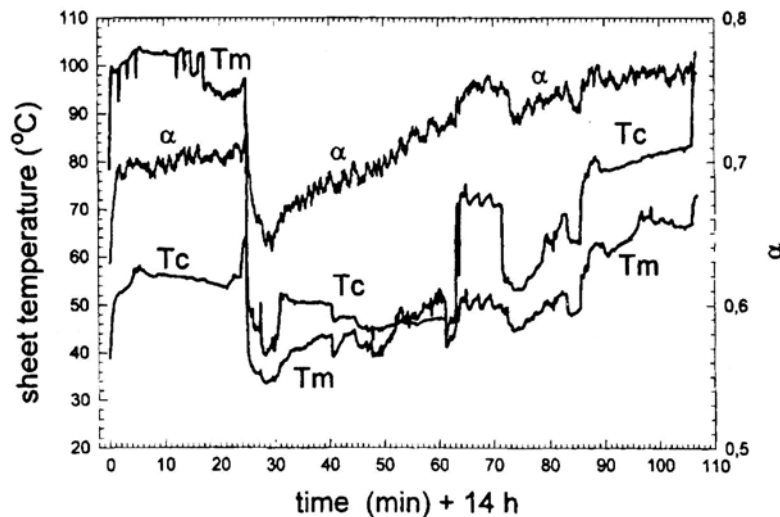

Figure 2: The loading ratio α and the temperatures near the cathode (T_c) and in the center of the sheet (T_m) versus time.

- in the first 30' of the first current step the behavior of V, T_m and T_c is like in FIELD II ; then suddenly V and a drop down and T_c becomes higher than T_m: this phenomenon seems to indicate a recovery of the cracks; in the following:
- V increases with the current;
- $T_c > T_m$ everywhere, indicating higher resistivity near the cathode;
- α reaches the highest value ~ 0.8;
- it must be noticed that a rapid increase and then a decrease of T_c, like a step, appears at the beginning of the second current step, in coincidence with a bump of a, as can be clearly seen in Fig. 2; a possible explanation for this anomalous phenomenon could be a strong accumulation of d^+ near the cathode (under the electric field effect) followed by a partial desorption from the uncoated *Pd* surface; of course more detailed investigations are necessary to clarify this point.

It must be remarked that at the end of the procedures the total time for reaching the loading value a ~ 0.8 is less than 17 hours, while the loading up to the equilibrium (α ~ 0.67) without electric field took more than 5 days in the previous calibration measurements.

4 Conclusions

A set of measurements of the loading under the application of an electric field across the length of a *Pd* strip have been performed for the first time in coincidence with the temperatures in the center and near the cathode of the strip. The analysis of the correlations among all these measured variables has pointed out the following results: i) the application of an electric field has shown the possibility of loading a *Pd* matrix with deuterium well over the thermodynamical equilibrium point, up to ~ 0.8 (averaged over the whole *Pd* volume); this confirms the Coehn effect with D_2 instead of H_1 in *Pd;* ii) furthermore, the difference of the temperature between the cathode and the middle of the *Pd* sheet indicates that the loading ratio is locally higher that the average, as expected by the numerical calculation of a diffusion model; this temperature difference is strongly correlated with the concentration: when the cathode is warmer than the center the concentration increases; iii) finally, it is well established that the application of an electric field greatly shorten the loading time.

The aim of the future work is to take advantage from these information to perform spectrometric measurements on *Pd* samples loaded up to as high as possible local concentration.

References

[1] E. Botta *et al.* in *Proc. ICCF-5* (Monte Carlo, Monaco, April 9-13, 1995), p. 233.

[2] E. Botta *et al.* in *Proc. ICCF-6* (Toya, Japan, Oct. 13-18, 1996) (1996), p. 29.