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Search for ^{4}He Production from Pd/D2 Systems in Gas Phase

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Abstract

We describe the performance of an apparatus consisting of a cell containing a Pd sheet loaded with D2 in gas phase coupled to a high resolution Q-mass spectrometer. The loading ratio α = D/Pd is increased by applying a constant electric field along the Pd sheet, α is carefully measured by means of the electric resistance variation and of the thermodynamic parameters. In one experiment a signal corresponding to ^{4}He production was observed.

1. Introduction

Since 1989 our Group (TOFUS-INFN Torino) is strongly engaged in the search of unambiguous signals of nuclear ashes from Cold Fusion devices. Our first effort was oriented towards the design, construction and operation of a rather sophisticated neutron spectrometer. The method we adopted to detect neutrons was double scattering among two segmented hodoscopes of plastic NE110 scintillators (60 elements in total). Time-of-flight and localization techniques allowed us to achieve an energy resolution of 1.0 MeV FWHM for 2.5 MeV neutrons, an efficiency of 2.5 10^{-4} and a powerful and reliable rejection of background due to cosmic rays and of environmental noise. Performances and technical details are given in Refs. [l-4]. By means of this spectrometer we studied the neutron emission from cells containing Ti and Pd shavings loaded with D_2 in gas phase and submitted to thermal cycles. Temperatures, pressures and loadings were adjusted in such a way to cross a phase transition line during a cycle. 2.5 MeV neutron emission was observed in different runs, with a maximum statistical significance of 5σ [5-6].

However, the measured neutron fluxes (from ~1 to ~0.1 neutrons/s-gr) were very low, fully inconsistent with the excess power reported by several other experiments. We decided then to concentrate our further efforts on the detection of ^{4}He, which seemed to be the nuclear ash produced at a rate consistent with the excess power [7-9]. To this purpose we designed and constructed an experimental device able to measure on-line the ^{4}He content of gas mixtures from Cold Fusion cells by means of high resolution Q-mass spectrometry. We report in the following the performances of the experimental set-up, the characteristics of the cell and finally the results so far obtained.

2. Experimental set-up

Fig. l shows a sketch of the apparatus. The purpose of the design was to perform all the operations (filling of the cell, use of calibrated volumes, H_2 and D; reduction by means of Ti getter to enhance the ^{4}He percentage, gas spilling from the cell to the spectrometer,..) with one circuit. All the components were of Stainless Steel. In such a way we reduced at minimum the possibility of contamination of ^{4}He from atmosphere, which seemed to be a critical point in other experiments [7,9]. Since we had to operate with different parts of the circuit at once, pressures and temperatures were monitored by several gauges (K-type thermocouples (±1°C), Pt thermoresistances (±0.5% in the range 0 -100 °C) and cold cathode pressure transducers (±5%)).

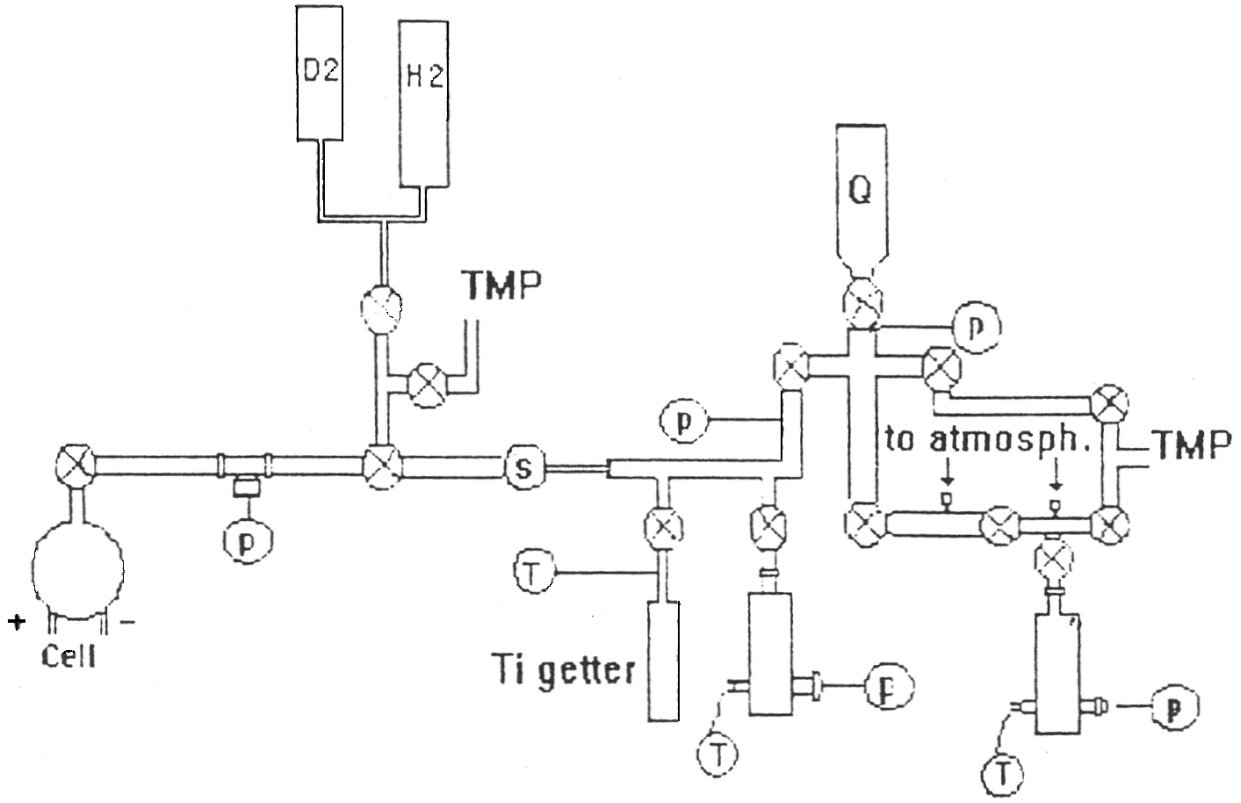

Fig. l Scheme of the experimental set-up (p=pressure gauges, T=temperature gauges, TMP=turbomolecular pump, S=leak valve, Q=spectrometer)

The volumes of the different parts of the circuit were carefully measured by means of gas expansions from a 300 cc calibration volume. Error on each volume is less than ±2%. Vacuum in the circuit was maintained by means of a turbomolecular pump (TMP), and was typically 10^{-6} mbar.

The Q-mass spectrometer is an ULVAC HI-RESOM 2SM, able to analyze masses of gases both in high resolution and low resolution modes. In the first mode the mass resolution at M/e=4 is M/ΔM = 200 (10% P.H.). Fig. 2 shows the peaks corresponding to $^4He^+$ and D_2^+, obtained with a calibration mixture. When used to analyze the gas composition from a Cold Fusion cell, the D_2 (or H_2) percentage, which is overwhelming, must be drastically reduced before immission into the spectrometer. To this purpose we used a getter pump, filled with 24 g

of Ti sponge, well activated under vacuum and at ~ 600 °C before use. We measured in several runs the efficiency of such a device and we found that after about 30 minutes all the content of D_2 (or H_2) was absorbed, within the precision limit due to the residual pressure.

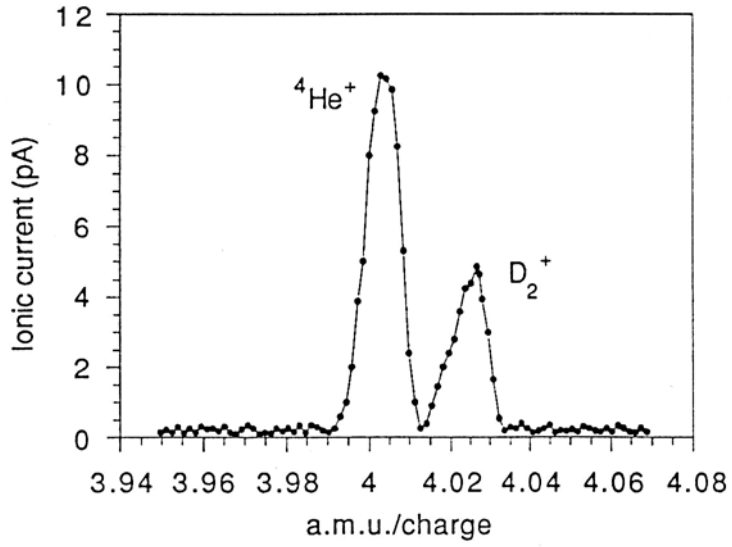

Fig. 2 - Peaks due to $^4He^+$ and D_2^+ obtained with a calibration mixture.

3. The Cold Fusion cell

The cell was machined in Al. We used Pd sheets, supplied by Goodfellow (PD000262, 99.95% purity), of dimensions ($8 \cdot 1 \cdot 5 * 10^{-3}$) cm^3. At NTP, the maximum loading ratio $\alpha = D/Pd$ that can be reached is 0.67. Among the methods suggested to increase locally α beyond the thermodynamical limit, electromigration seems to be quite effective. It has been in fact known since long time [10] that protons embedded in a Pd lattice migrate and accumulate near the cathode when a weak electric field is applied through the metal. To this purpose we clamped the Pd sheet among two massive Cu electrodes and an intense electric current (up to 200 A) could circulate in the sheet. To ensure a better electric contact and also in order to have a well defined surface barrier, both ends of the Pd sheet were gold plated (6 - 15 μm thickness) for a length of 1.5 cm. The effective surface of Pd exposed to the D_2 atmosphere was then 10 cm^2. From the value of the specific resistance of Pd at $\alpha = 0$ it is easily seen that with a current of 50 A circulating in the sheet the potential drop between the electrodes is 1.25 V, corresponding to an electric field of 0.25 V/cm. Higher fields could be more effective, but the temperature increase due to Joule heating of the sheet is prohibitive for maintaining a high value of a. Thermal dissipation was ensured by clamping in contact the free lateral surfaces of the Pd sheet with the lateral surface of the cell, cooled by a thermostatized (± 0.5°C) water flow on one side, by a massive Al heat dissipator on the other side. Aluminum surfaces were anodized (~ 20 μm thickness) in order to ensure a good electric insulation. The temperature at the center of the Pd sheet was continuously monitored by a K-type thermocouple (±1°C). The temperature of the gas in the cell was continuously monitored by a Pt thermoresistance (±0.5% in 0+100 °C range) and its pressure was measured by an absolute pressure gauge (±0.1% F.S.).

Fig. 3 shows the increase ΔT of the Pd sheet temperature with respect to the temperature of the water for different values of the circulating current. It can be seen that at 120 A ΔT is ~100°C, and the Pd was rapidly desorbing D_2. Following a long experimental study, we decided to use currents not exceeding 70 A. The measurement of α is of paramount importance in Cold Fusion experiments. It can be measured by means of thermodynamical parameters of the gas (p, T, V), or by the variation of the resistance of the Pd sheet. The ratio R(α)/R(0) at constant T is a bell-shaped function of α, well parametrized by the fifth order polynomial: $(1 + 3\alpha - 15.13\alpha^2 + 44.16\alpha^3 - 49.12\alpha^4 + 17.58\alpha^5)$ [11].

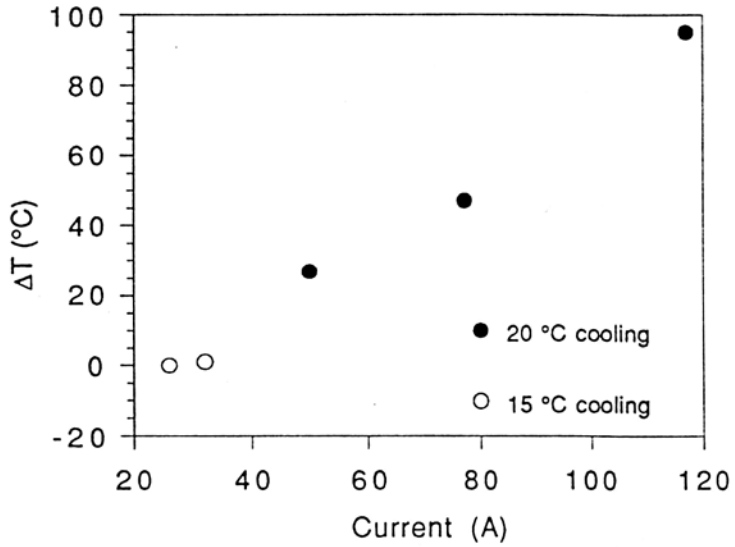

Fig. 3 - Increase of the temperature of the Pd sample as a function of the circulating current.

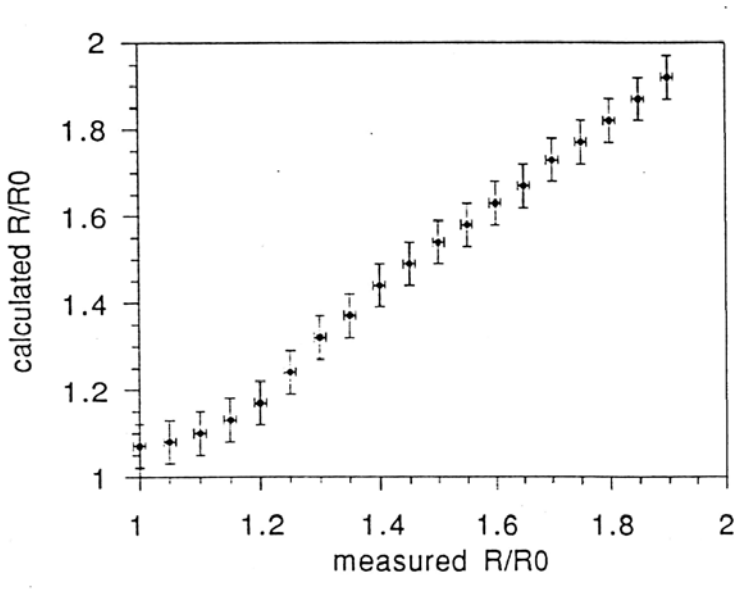

Fig. 4 - Correlation between the measured values of R(α)/R(0) and the calculated ones (see text).

We used both methods to determine α in our experiment. Fig. 4 shows the correlation between the measured values of R(α)/R(0) and those evaluated by means of the polynomial, in which the values of α were taken from the thermodynamical measurements. Within the errors, a satisfactory agreement is observed.

However we notice that both measurements provide α integrated over the full volume of the sheet, including the gold plated ends. At present we do not have direct measurements able to ascertain local enhancements of α. To this purpose a simple model taking into account the absorption, the diffusion and the mobility under application of the electric field of the deuterons into the Pd lattice was developed [12]. The prediction of the model was that α integrated was very slowly increasing with time whereas locally α could reach values exceeding 1, at the cathode, after about one week.

An indirect proof of this behaviour was that two Pd sheets broke just near the cathode, at high currents, and two others showed a local darkening, due to the carbonization on a small Vetronite insulator in contact with the cathode, after long runs at low current. For these four cases we had evidence for a local increase of temperature near the cathode.

4. Experimental results

Table 1 summarizes the conditions corresponding to the different runs, employing five different Pd sheets. The first three sheets were used mostly to study the effect of the different parameters, in particular the electric field, on a. We reached the following conclusions:

1) electromigration seems to be effective both in getting a faster loading of D_2 into Pd below $\alpha = 0.67$ and in exceeding the above value;
2) the effect is more evident with the increase of the pressure;
3) the temperature of the Pd sample must not exceed 70°C;
4) surface treatment helps the loading of D_2.

Pd Sheet N.	Vol. Lxlxδ cm^3	Surface Treatm.	Gold Thick.	Run Time (hour)	P Imm. (bar)	Max α	Analysis of Gas	^4He Excess
1	7.99x0.99 x47*10^{-4}	N	Y 15 μm	74.5	1.117	0.48	N	
2	7.99x0.99 x47*10^{-4}	N	Y 15 μm	91.5	1.566	0.67	N	
3	8.0x1.0 x52*10^{-4}	N	Y 6 μm	1159	1.63	0.74	N	
4	8.0x1.0 x52*10^{-4}	N	Y 6 μm	504	1.696	0.67	Y	N
5	7.98x0.96 x50*10^{-4}	Y $HN0_3$	N	699	1.564	0.69	Y	Y

Table I. Summary of the runs.

Fig. 5 shows the results of two runs. In the first one (a) the application of the current seems very effective in the last part of the run, after a short period during which we performed some rapid variations of the electric field (not shown in Fig. 6a). However, α integrated did not reach the thermodynamical limit of 0.67. In the second one (b) the electric field was applied after a long time, during which the sample of Pd was loaded up to α = 0.67. At this time, the application of an electric field of 0.25 V/cm produced a constant increase of α, up to a final value of 0.75, corresponding to a $\Delta\alpha/\Delta t$ of (6.9+0.2)* 10^{-4} $hour^{-1}$. At a certain time (during a week-end) the acquisition system went down, and we do not have the record. After restart, and switch off of the electric field, α kept constant at 0.75 for more than two days.

For the last two samples, we performed an analysis also of the ^4He content in the gas of the cell after a ~ 500 hour run. The second sample showed at the end a signal corresponding probably to the production of ^4He. Fig. 6 shows the spectrometer scan around M/e=4, before and after the immission of the gas from the cell. It can be seen that the peak corresponding to ^4He is increased by 25 - 30%, whereas the residual pressure is lower by ~ 15%. We remind that in mass spectrometers the net number of ions present is proportional to the ratio peak/(residual pressure). We may exclude contamination from the atmosphere. Scans in the low resolution mode, performed before and after the immission, showed that all the peaks corresponding to higher values of M/e were at same level or lower. Apart the obvious case of D_2^+ , $^4He^+$ was the only one showing an increase. However the circumstances of having a significant background even before the immission is somehow disturbing.

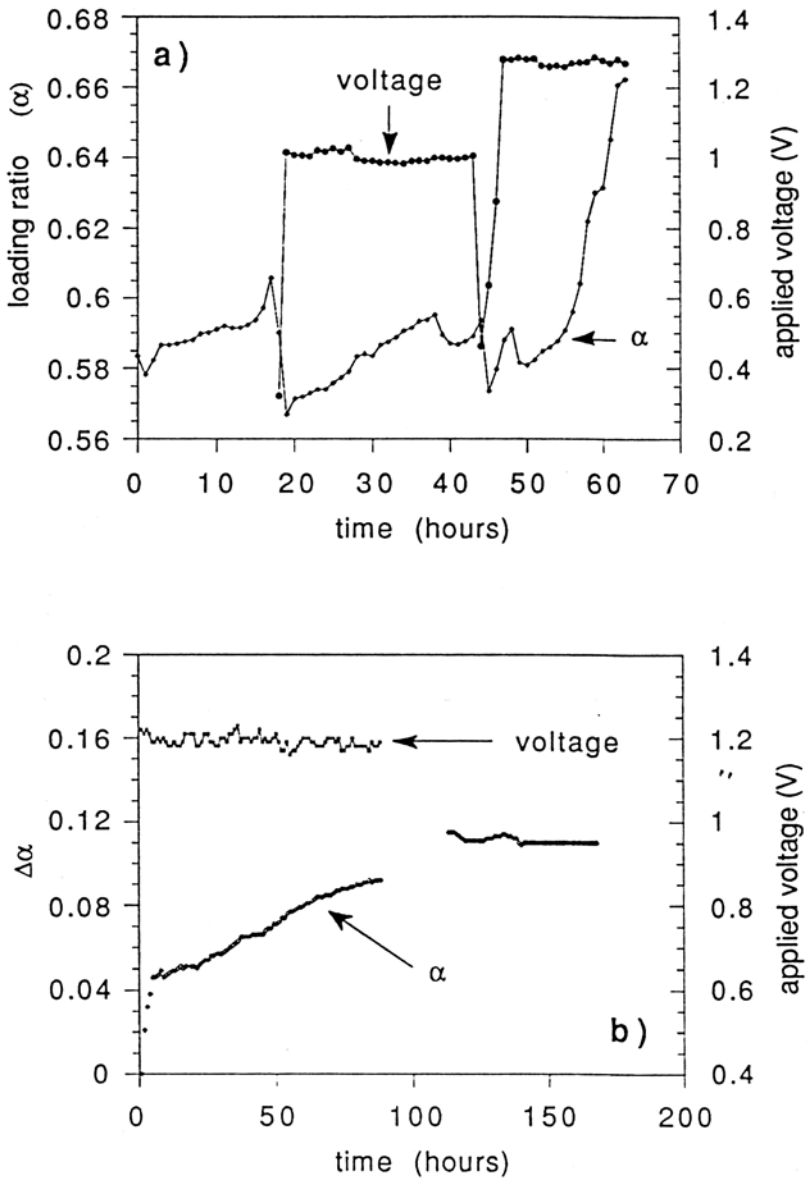

Fig. 5 : a - Measured values of α (lower curve) and of the applied voltage (upper curve) as a functions of time.: b - Measured values of α beyond the thermodynamical limit a= 0.67 (lower curve) and applied voltage (upper curve) as a function of time.

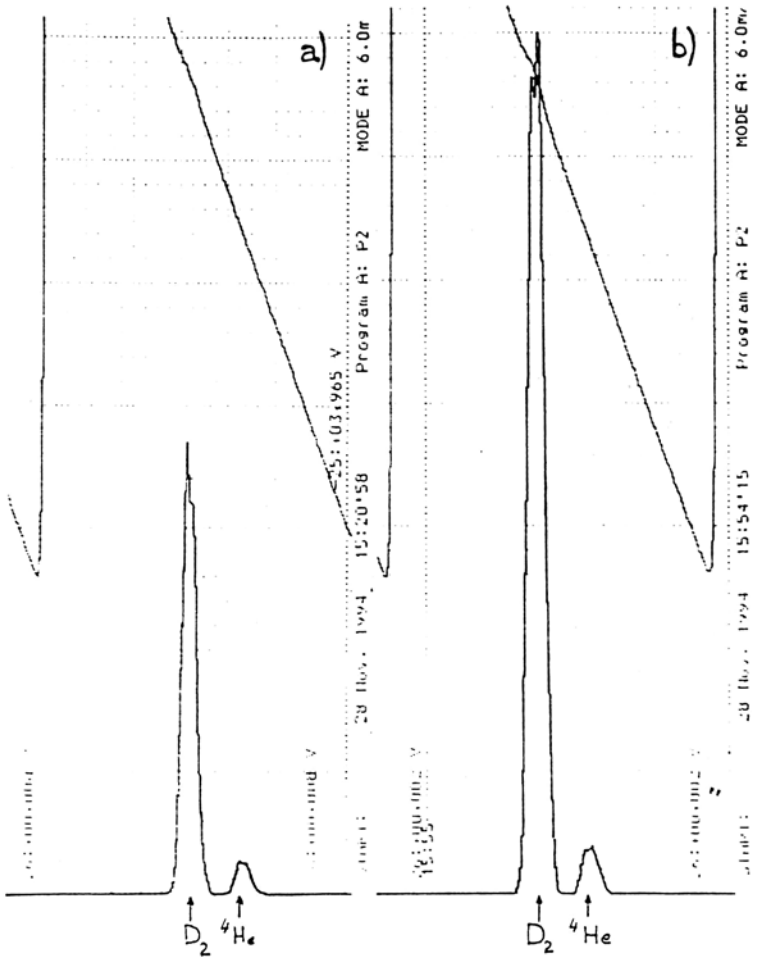

Fig. 6 - Spectrometer scan around M/e=4 before (a) and after (b) the analysis of the gas sample from the cell.

In the hypothesis that the 25% increase of the peak is real, it would correspond to ~$3*10^{10}$ ^{4}He atoms in the analysis volume of the Q-mass spectrometer and to $7.5*10^{16}$ ^{4}He atoms in the total volume of the cell, in the very simplified hypothesis that the ratios of the partial pressure of ^{4}He to the total pressure in the cell volume and in analysis volume are the same.

The above number of the ^{4}He atoms must be considered as an upper value, since the effect of the different pumping rate of the TMP for different gases (even a few units) has not been taken into account. For this reason we have not quoted errors, and we will not do it in the following.

Converting the numbers of ^{4}He ions into power due to the reaction d+d $\rightarrow$ ^{4}He+23.8 MeV, we obtain an average value of about 1 watt for three days, too weak to be signaled by an increase of temperature of the cooling water of the cell. The average specific power is 25 watt/cm^3. However, since the profile of α along the Pd sheet is expected to be peaked near the cathode, and we may quite reasonably expect that Cold Fusion occurs in regions with high α, specific powers larger by at least one order of magnitude may be deduced.

The neutron spectrometer was active during these runs. No significant peak corresponding to 2.5 MeV neutrons was observed. The upper limit is 0.4 neutrons s^{-1} gr^{-1} at 1 σ level. The ratio ^{4}He/n is then larger than 10^{12}.

We stress again that all the above considerations follow from the hypothesis that the observed increase of the ^{4}He peak is not an instrumental artifact, a possibility that looks remote but cannot be excluded.

4. Conclusions

First round of runs of a novel Cold Fusion cell, with gas loading of D_2 assisted by electromigration, looks very promising. The cell is directly coupled in vacuo to a high resolution Q-mass spectrometer. In one experiment production of ^{4}He was observed. Improvements on the cell design (higher pressure of the loading gas, control of the temperature at both ends of the Pd sheet) were performed and we intend to start a new series of runs, both with H_2 and D_2, to confirm without doubts our observation.

Acknowledgments

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