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On the neutron emission from the Ti/D system(*)(**)

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Summary. — The results of a set of measurements of neutron emission from D_2 gas loaded Ti (metallic and sponge) at different initial values of the D/Ti ratio are reported. The D/Ti system was submitted to thermal cycles in order to pass through phase transitions. We observed no neutron emission with an upper limit of 0.05 neutrons $s^{-1}g^{-1}(1\sigma$ level), in four series of measurements, whereas in two series we observed a neutron emission at a $\sim 2\sigma$ level.

PACS 25.88 – Fusion reaction.

1. – Introduction

In the first announcement of Cold Fusion (CF), Fleischmann, Hawkins and Pons [1] noticed that the production of nuclear ashes (neutrons, ^{3}He) in their cells, even if measured with very simple instruments, was lower by 10^8 to 10^{10} orders of magnitude than what expected by the measurement of the excess heat, in the hypothesis that it was of nuclear origin, following the fusion of two deuterium. In a Pd lattice the (d+d) fusion had to proceed in a way substantially different than in free space, in which case the channels

$$\mathrm{d} + \mathrm{d} \longrightarrow \mathrm{n}(2.45\,\mathrm{MeV}) + {}^3\mathrm{He}(0.82\,\mathrm{MeV}) \tag{1}$$

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and

$$\mathrm{d} + \mathrm{d} \longrightarrow \mathrm{p}(3.02\,\mathrm{MeV}) + {}^{3}\mathrm{H}(1.01\,\mathrm{MeV}) \tag{2}$$

almost equally probable ($\sim 50\%$), are the well-known final products. It must also be noticed that, from the first beginning, the channel with ^{4}He in the final state, known to occur with a frequency lower by a factor 10^{-6} with respect to the (1) and (2) ones, was supposed to be the most important in (d+d) fusions in a metal lattice by some authors [2].

Notwithstanding these facts, our group started its activity in the field by an experiment aiming to measure the energy spectra of neutrons emitted by CF cells with a new, dedicated, detector (TOFUS) featuring the state-of-art techniques. For the CF device we used the gas-loading technique. Pure Ti in different metallurgic conditions was the metal used in almost all the experiments. Detailed descriptions of the spectrometer [3], of the electronics [4] and of the analysis techniques [5] were already published. Results of a first series of experiments were also already reported [6,7]. In sect. **2**, we report a brief summary of the experimental techniques and previous results, in sect. **3**, we describe the upgraded version of the CF device, whereas in sect. **4**, we report the results of the last series of experiments and in sect. **5** the conclusions.

2. – Experimental techniques and previous results

a) Neutron detection technique.

In the design of the neutron spectrometer we focused our attention on the measurement of the neutrons energies. The time-of-flight (TOF) technique is known to be the most reliable to the purpose. However, in CF research its application is far to be straightforward in the absence of an associated particle or a machine signal providing the start of the measurement as for all the other experiments. We overcame this difficulty by adopting a double-scattering technique by means of two arrays of plastic NE 110 scintillators in time coincidence.

The main features of the spectrometer are

- an energy resolution of $\sim 1\,\mathrm{MeV}$ FWHM for neutrons of 2.5 MeV (as expected from (1));
- an overall efficiency of $\sim 2.5 \times 10^{-4}$;
- a background, mainly due to the photomultipliers electronic noise, of ~ 200 triggers/hour ($\pm 8\%$), reduced to ~ 68 events/hour ($\pm 10\%$) after software kinematical cuts;
- a neutron counting rate of the order of 5 MHz (useful for counting "bursts" of neutrons) without energy reconstruction, going down to 250 Hz with full energy reconstruction.

We notice that in the first series of measurements [6,7] we used a tight time coincidence between the two arrays of scintillators optimized for detecting the 2.5 MeV neutrons, and lowering the background due to random coincidences.

b) Metal/D loading technique.

We decided to use the gas-loading technique in our experiments. As a matter of fact the use of electrolytic cells, immersed in water thermostats, would spoil the performances

of our neutron detector since the neutrons emitted at the source with, say, 2.45 MeV would be degraded in energy or even thermalized. Furthermore, gas-loading of D_2 in Ti was claimed to produce strong neutron bursts [8].

c) Summary of previously published results.

A first exploratory measurement was performed in 1991, with a simplified CF apparatus and Ti in form of metal shavings [6]. The sample was submitted to a number of thermal cycles, each consisting of a heating step from room temperature up to 540° C (called run UP) followed by a cooling step to the room temperature (called run DOWN). A 2.5σ statistical evidence for the presence of 2.5 MeV neutrons, corresponding to an emission of (1.3 ± 0.5) neutrons $s^{-1}g^{-1}$ of Ti was reported. The mean D/Ti ratio (labeled X) in this experiment was 0.32. Mean ratio means that we estimated the mean value of the gas-loading in titanium without any indication of the local behaviour: we got a macroscopical information on the atomic ratio but we did not perform any microscopical investigation.

A second more complete experiment was reported in 1992 [7]. In this case a cell embodying a more careful control of the thermodynamical parameters was utilized. Temperature cycles, as described above were performed starting with Ti sponge at $X = 0.7$ and $X = 1.8$. Adding the statistics cumulated at both values of X, we observed the presence of 2.5 MeV neutrons, with a statistical significance of $\sim 5\sigma$, emitted at an average rate of 0.11 ± 0.03 neutrons $g^{-1}s^{-1}$. No neutron bursts were observed.

Furthermore, the presence of neutrons with energies up to 7 MeV was apparent in the spectra, with a lower statistical signifiance $(2 - 3\sigma)$. We remind that in this experiment the neutron hodoscope was optimized for the detection of 2.5 MeV neutrons, and then the results on the presence of higher energy neutrons could be biased.

We studied also the Pd/D system at the D/Pd atomic ratio of 0.7. A rate of 2.5 MeV neutrons of 0.02 ± 0.01 neutrons $g^{-1}s^{-1}$ with a 2 σ statistical signifiance was measured.

3. – The upgraded version of the cold-fusion device

Aiming at performing a better evaluation of X, we realized some technical improvements in the CF device before starting a new series of measurements. The apparatus is sketched in fig. 1.

The CF cylindrical cell (fig. 1a) of 44 mm diameter and 102 mm height was located in front of the neutron detector. A thermocouple (TC1) (error in the temperature reading: ± 0.1 K), embedded in the Ti sample, measured the D/Ti lattice temperature. A second thermocouple (TC2) monitored the temperature of the deuterium gas surrounding the Ti sample, while a pressure gauge, in the 10^{-1}–10^{2} bar range (class 0.1), measured the gas pressure. A heating system, consisting of a thermoresistance, was located in contact with the lower basis of the cell, while some shields of Al sheets were positioned around the external cylindrical wall of the cell in order to lower the heat flow towards the scintillators of the neutron detector.

The cell was the "core" of a more complete gas-loading and vacuum apparatus, sketched by fig. 1b). The whole apparatus was realized in UHV stainless steel (SS304L), and was composed by the CF cell, the gas-loading system and the pumping circuit.

The gas pumping circuit consisted of a V200 Varian turbomolecular pump (TM) whose prevacuum was obtained by means of a conventional rotary pump. The final residual pressure inside the whole circuit (about 3 l) was 10^{-11} bar near TM and 10^{-10} bar near the CF cell, when the TM pump was active and without Ti sample inside the CF cell; the final residual pressure in the presence of Ti, following a degassing at 700 °C, was

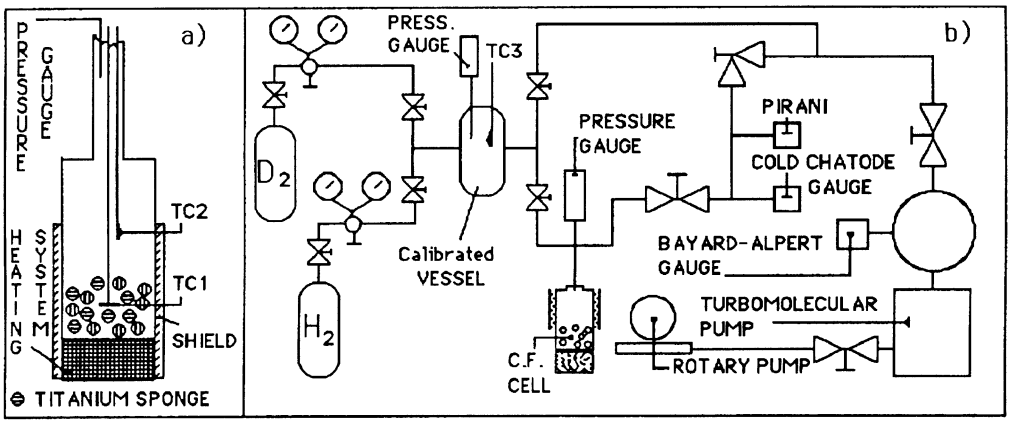

Fig. 1. – a) Scheme of the cold-fusion cell, b) scheme of the full cold-fusion device.

better than 10^{-11} bar, near the cell. The residual pressure was measured near the CF cell by a Cold Cathode Gauge in the range 10^{-11}–10^{-6} bar, near TM by a Ion Gauge type Bayard-Alpert in the range 10^{-12}–10^{-6} bar, and by a Pirani in the range 10^{-6}–10^{-3} bar.

The loading system was composed by two lines, connected with the D_2 and H_2 cylinders. Both lines were connected to the circuit through a vessel of well-known volume ($0.251\,\mathrm{l} \pm 0.0005\,\mathrm{l}$) allowing to determine the number of gas moles (N_{gas}) before immission in the CF cell, by means of pressure, using a pressure gauge operating in the range 10^{-1}–10^{2} bar, and temperature, using a thermocouple (TC3), information. All the thermocouples and pressure devices were connected to a C1732 Multireg Siemens programmable register.

The sequence of operations before starting neutron data-taking run were the following:

1) a cleaning of the Ti sample, received from the manufacturer (Ginatta Torino Titanium s.p.a.) covered by a thick oxide layer, by a heating at 700 °C, following the procedure already described in [7];

2) the immission of the determined number of D_2 moles (H_2 for blank measurements) in the CF cell by opening the valve positioned between the calibrated vessel and the CF cell: the gas expands in the complete volume ($V = 1.056\,\mathrm{l}$), and its absorption from Ti starts.

To obtain a specific initial value of X (called X_0) ratio, neverthless without overcoming the stechiometric atomic ratio, equal to 2, corresponding to D_2Ti, we calculated simply the ratio between the D_2 moles and the Ti moles, multiplied by the factor 2 that accounts for the diatomic nature of D_2 molecules, decomposed near the metal surface before the absoption: then we knew X_0 with an error of $\Delta X_0 = 0.005$.

The variables that characterize our system are X_0 and V.

In order to perform thermodynamical cycles which allow the D/Ti system to pass through different phases (α, β and δ), we choose, as in previous measurements, to submit the samples to a number of temperature cycles at constant volume: each cycle consisted of a heating step (run UP), in which the D atoms were expelled from the lattice, and a cooling step (run DOWN), during which D_2 gas was absorbed from the lattice.

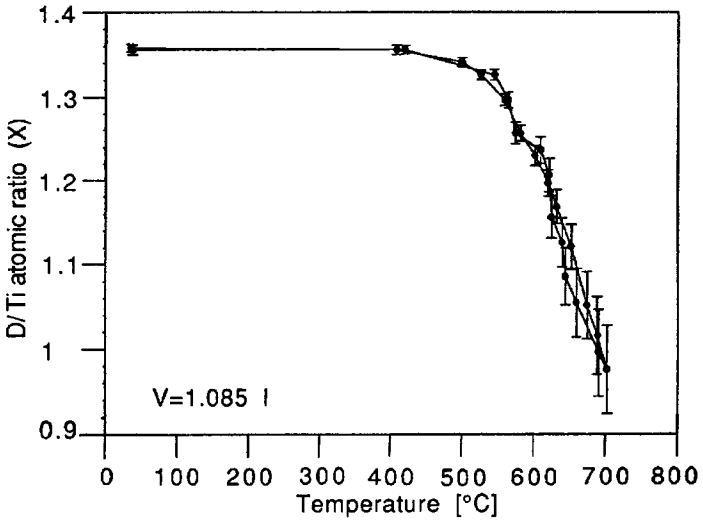

Fig. 2. – Variation of the loading ratio X as a function of the temperature of the Ti lattice in our experimental conditions.

During the cycle X is evaluated by means of the equation

$$X(p, T_{\text{gas}}) = \frac{2}{N_{\text{Ti}}} \left(N_{\text{gas}} - \frac{pV}{RT_{\text{gas}}} \right) , \tag{3}$$

where R is the gas constant, T_{gas} is the gas temperature in kelvin, p is the gas pressure, N_{Ti} is the number of the Ti moles and V the volume of the gas expansion, while the factor 2 accounts for the diatomic nature of D_2 molecules, as previously explained.

In our system, the critical parameter was the gas temperature, because, during a heating step, large temperature gradients exist in the volume V. In such conditions it was problematic to evaluate the mean value of the T_{gas}: we choose to use the mean value of the information obtained from the two thermocouples located into the volume V, TC2 and TC3 in fig. 1, and the mean deviation was assumed as error. In this way, we obtained the mean atomic ratio with a maximum error $\Delta X = 0.05$ at the higher temperatures.

In fig. 2 we report X as a function of the lattice temperature: the variation of the error of X is apparent.

4. – Experimental results

We performed data taking runs for different samples of Ti (metallic shavings and sponge) and for different values of X_0 as summarized by table I: we submitted the D/Ti system at temperature cycles with a heating step from room temperature to $\sim$ 650 °C, lasting $\sim$ 120 minutes and a following cooling step to room temperature, lasting $\sim$ 250 minutes.

According to different values of X_0, we observed, in both runs UP and runs DOWN, the occurence of the euctectoid formation at a temperature of $\sim$ 280 °C, where α, β and δ phases coexist, and of β-δ phase transitions at higher temperatures.

TABLE I. – *Summary of the series of measurements.* Ti_M*: stands for metallic* Ti*;* Ti_S*: stands for sponge* Ti.

Sample	X_0	Total time of UP runs (minutes)	Total time of DOWN runs (minutes)
Ti_M	1.125	2583.75	3765.46
Ti_M	1.357	3070.15	5880.46
Ti_S	1.069	2103.61	2932.22
Ti_S	1.339	2469.51	2406.74
Ti_S	1.837	2901.82	3179.39
Ti_S	1.987	726.02	1164.36

Due to our heating system, the temperature at which the euctectoid formation occurs and the temperatures of the phase transitions were more visible during the runs DOWN: they apparead as flatings in the otherwise decreasing plots of temperature towards time, while during the runs UP we observed only smooth flexa. We report in fig. 3 the temperature *versus* time plot during a run DOWN for Ti sponge at $X_0 = 1.069$.

Due to the possible presence of neutrons at energy higher than 2.5 MeV, we decided to increase the energy range of detected neutrons up to 8 MeV. To this purpose, we increased the time coincidence width by the proper value. For determining the flux of neutrons emitted by the samples, we adopted the procedure of subtracting the runs DOWN, normalized in time, from the runs UP, as performed in the first experiment [6]. We did not use the runs performed with H_2 for the following reasons:

1) The duration of the runs with D_2 lasted several months and we did not alternate runs with H_2, since we were avoiding possible contamination of the samples: the subsequent values of X_0 were obtained by adding the necessary amount of D_2 moles.

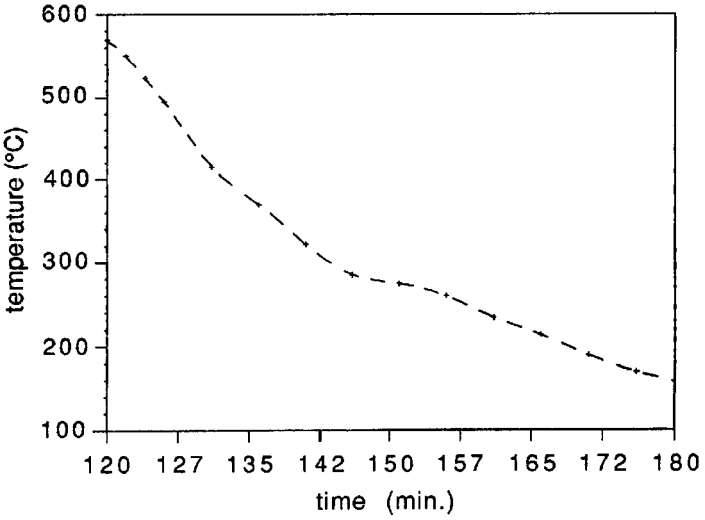

Fig. 3. – Variation of the Ti temperature *vs.* time during a run DOWN (cooling step): the flatings at $\sim 280\,^{\circ}$C and $\sim 380\,^{\circ}$C correspond, respectively, to eutectoid formation, where α, β, and δ phases coexist, and to the $\beta - (\beta + \delta)$ phase transition).

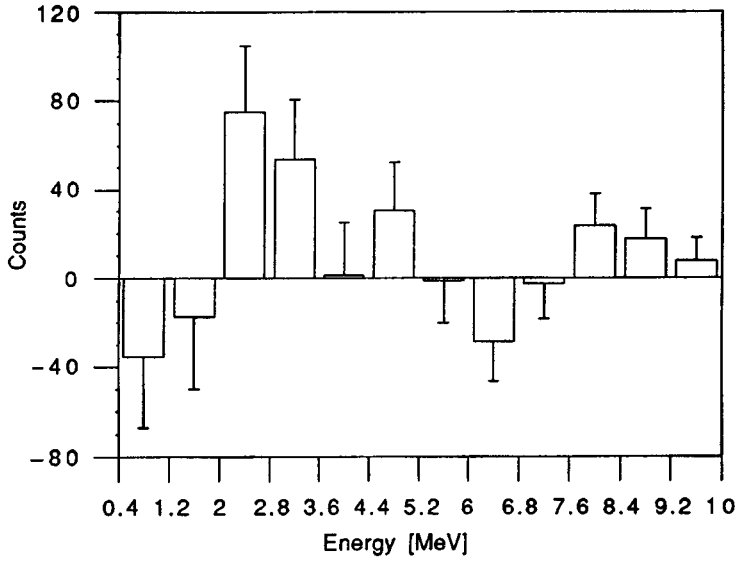

Fig. 4. – Neutron energy spectrum for Ti metallic sample at $X_0 = 1.125$, with D_2 loading.

2) After several months the runs with H_2 showed a background level slightly different, and then the comparison with the corresponding runs with D_2 needed a significant amount of corrections, not easily controlled at the necessary level of precision.

On the contrary, the subtraction of subsequent DOWN and UP runs permits a quite sure elimination of long-term instabilities and drifts of the detecting system. However, such a procedure, instead of measuring the net number of neutrons emitted in a cycle, allows to evaluate differences in the emission occurring in the two parts of the cycle, which are affected by larger statistical errors.

As a matter of fact, in the experiment of [7], the neutron emission evaluated by a difference UP-DOWN was 0.08 ± 0.04 neutrons $g^{-1}s^{-1}$, to be compared with the value of 0.08 ± 0.04 neutrons $g^{-1}s^{-1}$ evaluated by the D_2–H_2 difference method applied to the sum of all cycles (UP+DOWN).

Figures 4, 5, 6, 7, 8 and 9 show the neutron spectra observed with the different samples and loadings. Four of them are consistent with no neutron emission, with an upper limit (1σ level) of 0.05 neutrons g^{-1} s^{-1}, in the range 0.4–7.6 MeV. The spectrum obtained at $X_0 = 1.125$ (fig. 4) shows a 2.5σ signal in the energy range 2–2.8 MeV, corresponding to a neutron emission of 0.10 ± 0.04 neutrons $g^{-1}s^{-1}$.

The spectrum corresponding to Ti with $X_0 = 1.837$ (fig. 8) shows no evidence for accumulation of events in particular energy ranges, but perhaps a broad spectrum, extending from 0.4 MeV to 7.6 MeV. The net flux of neutrons in this range is 0.10 ± 0.05 neutrons $g^{-1}s^{-1}$. For this evaluation we adopted an average value of the efficiency of 4×10^{-4}.

We analysed by the same technique of subtracting the runs DOWN from the preceding runs UP, properly normalized in time, also the data taken with H_2 loading. We did not observe statistically significant neutron emission. Figure 10 shows, as an example, the spectrum corresponding to metallic Ti at $X_0 = 1.125$; the total duration of the runs UP was 2017.92 minutes and that one of the runs DOWN was 2422.33 minutes.

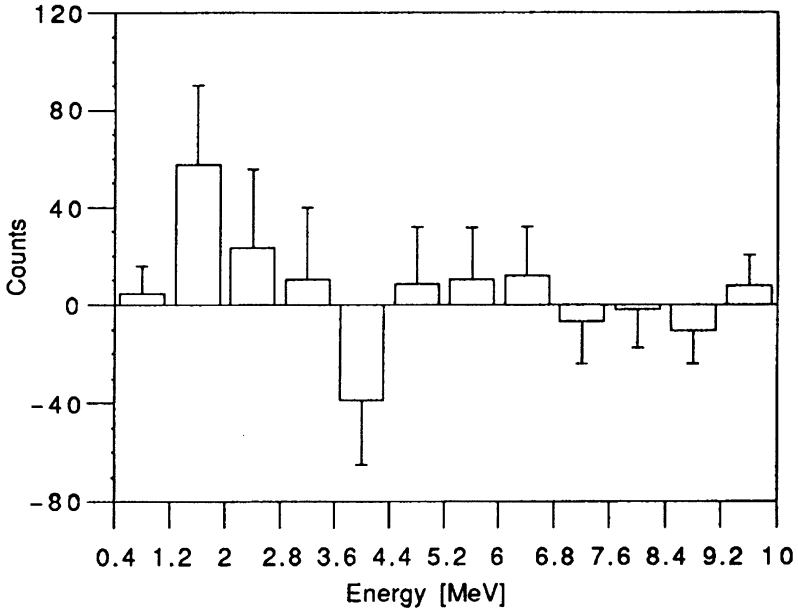

Fig. 5. – Neutron energy spectrum for Ti metallic sample at $X_0 = 1.356$, with D_2 loading.

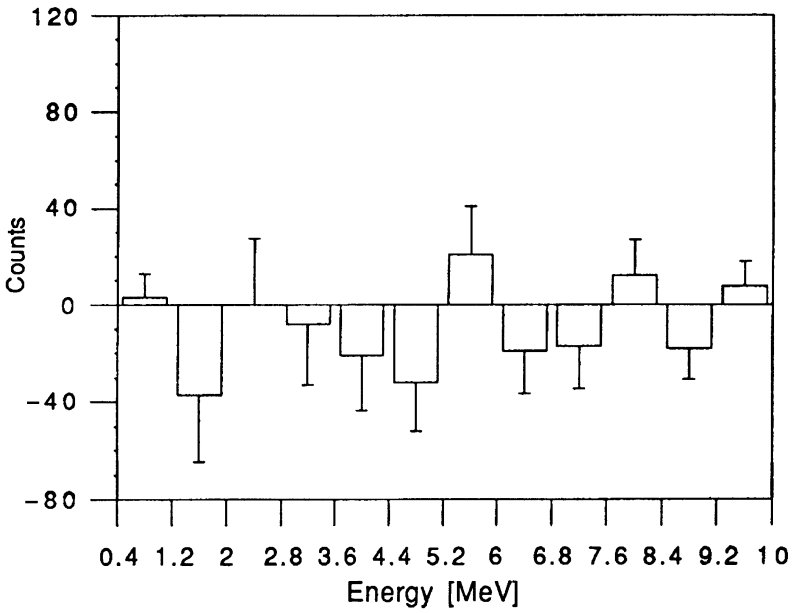

Fig. 6. – Neutron energy spectrum for Ti sponge sample at $X_0 = 1.069$, with D_2 loading.

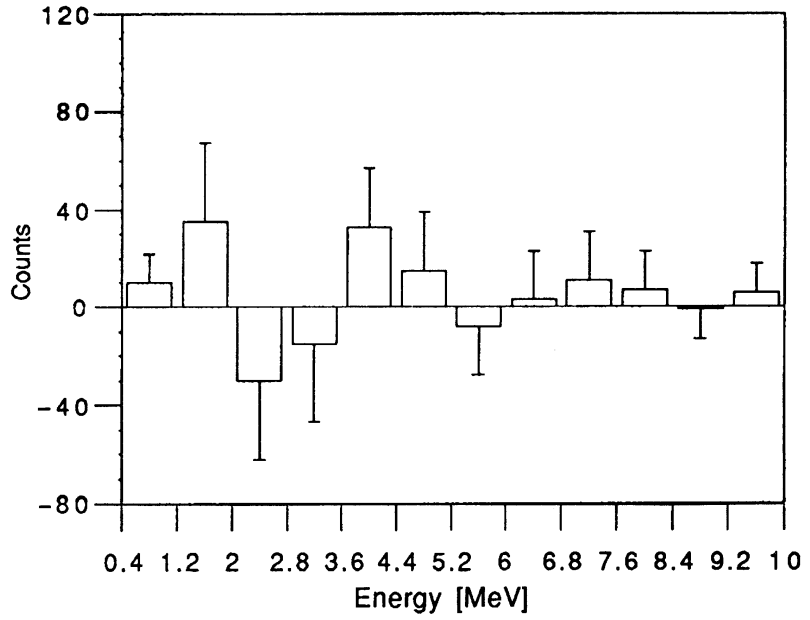

Fig. 7. – Neutron energy spectrum for Ti sponge sample at $X_0 = 1.339$, with D_2 loading.

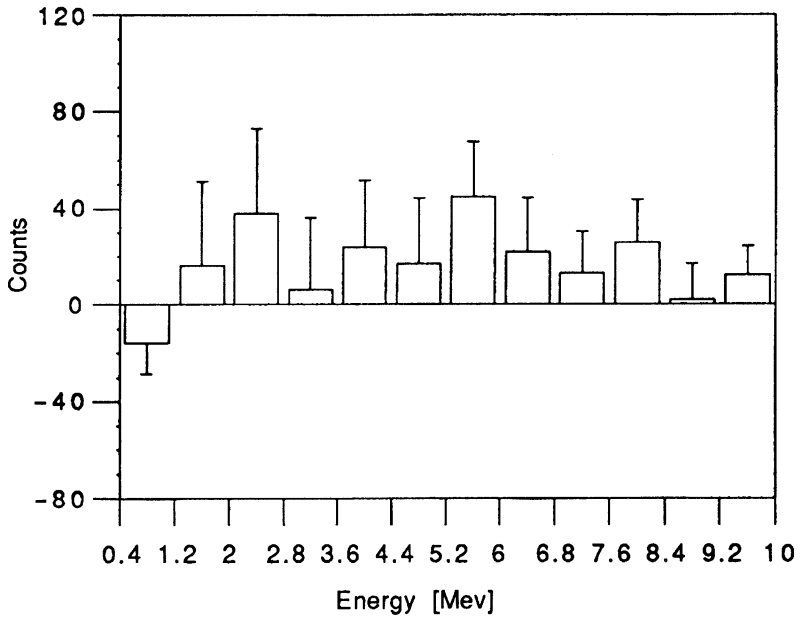

Fig. 8. – Neutron energy spectrum for Ti sponge sample at $X_0 = 1.837$, with D_2 loading.

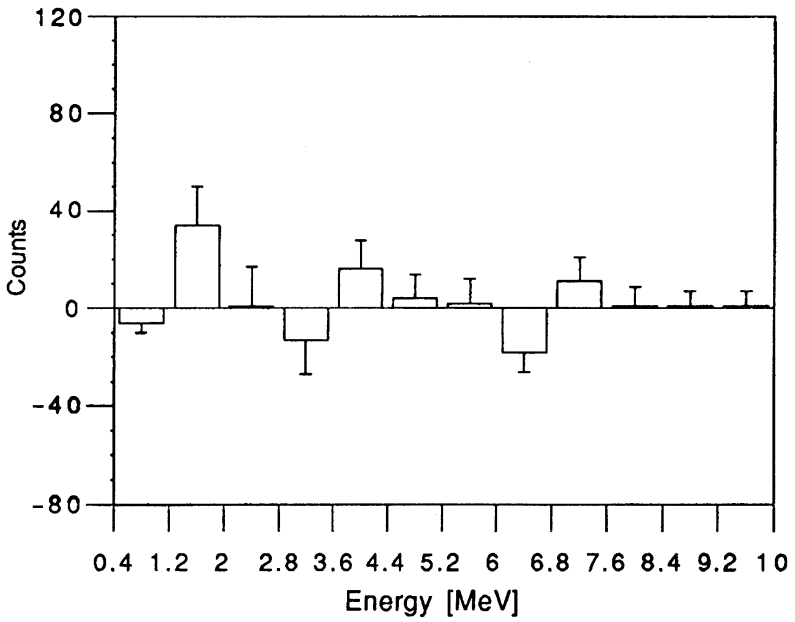

Fig. 9. – Neutron energy spectrum for Ti sponge sample at $X_0 = 1.986$, with D_2 loading.

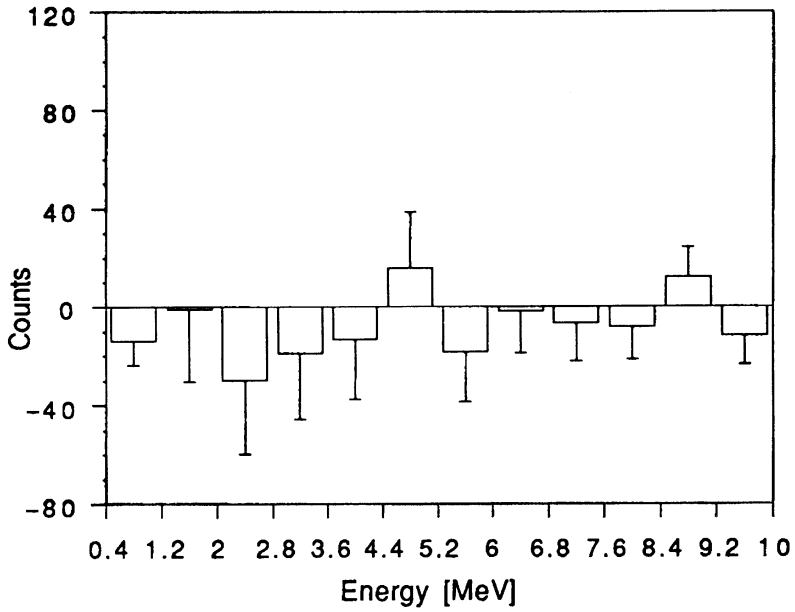

Fig. 10. – Neutron energy spectrum for Ti metallic sample at $X_0 = 1.125$, with H_2 loading.

5. – Conclusions

These series of measurements, performed with an advanced neutron detector and with a systematic study of possible effects due to the Ti/D atomic loading ratio, confirm the results of many experiments on neutrons emission by CF devices published in the last years. As examples, we may quote the experiments of Mengoli *et al.* [9] and Oya *et al.* [10].

Our conclusions are:

1) A weak emission of neutrons (of the order of 0.10 neutrons $g^{-1}s^{-1}$) is observed at a 2–3 σ level in some experiments, however without a clear correlation with the loading ratio achieved.

2) Emission of neutron bursts is excluded.

A final remark is that the neutron measurements seem not so useful for ascertaining the nuclear origin of CF phenomena. Emission of neutrons, apart from the problem of reproducibility, is so weak that their detection cannot fulfill at the same time the requirements of energy measurement and high efficiency and then statistical signifiance. We chose the option of a precise energy measurement in spite of efficiency, but we ended with several problems of statistics.

The search for other nuclear ashes, like ^{4}He, observed in some cases in considerable amounts, seems more fruitful for ascertaiming the nuclear origin of Cold Fusion [11, 12].

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