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Measurement of 2.5 MeV Neutron Emission from Ti/D and Pd/D Systems (*).

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Summary. — A new set of measurements of neutron emission from gas-(D_2 and H_2) loaded Ti and Pd systems has been carried out in the TOFUS experiment. The temperature and pressure controls of the gas loading apparatus were improved. The results concerning the Ti/D system show the presence of a small 2.5 MeV neutron emission, with a signal having a statistical significance of $\sim 5\sigma$. The results on the Pd/D system does not show a statistically significant signal (less than $\sim 2\sigma$).

PACS 25.88 – Fusion reactions.

1. – Introduction.

Since the start of the debate about the occurrence of D-D fusion phenomena in the lattice of some metals like Pd and Ti, the detection of neutrons, in particular 2.5 MeV neutrons, has been considered as the most reliable signature of the effect. In order to clarify this point, a sophisticated neutron detector was designed and built for the TOFUS experiment, carried out in Torino at the Laboratorio Tecnologico of INFN. It started to operate in 1990 and produced a first set of measurements showing a small amount of neutron emission following the loading of Ti shavings with gaseous D_2 [1,2], with a statistical significance of $\sim 2.5\sigma$. A number of improvements has been performed on the apparatus, mainly on the heating system, and during the summer 1991 a second set of measurements has been performed with a better control of the pressure and temperature of both the metal and the gas [3].

We performed measurements not only with titanium but also with palladium,

(*) The authors of this paper have agreed to not receive the proofs for correction.

loaded with gaseous deuterium (and hydrogen for blank measurements). All the blank runs were performed immediately after those with the deuterium filling, in order to avoid the problems due to the possible long time variations of the detector electronics. In sect. 2 the apparatus is briefly described, the improvements are outlined and the thermal operating conditions of the metal/gas system are reported. In sect. 3 the results are shown and discussed and the conclusions are reported in sect. 4.

2. – Experimental set-up and description of the thermal cycles.

The TOFUS apparatus consists of two parts, the neutron detector and the cell with heating and gas loading system.

Concerning the neutron detector, it has been already described in previous papers [3-5] and we just recall here the performances: the neutrons are detected by two blocks of plastic scintillators NE110 in coincidence (double-scattering technique) and their energy is determined using a reconstruction method based on the measurement of the neutron time of flight (t.o.f.) and of the impact position onto the scintillators.

The main feature of this method is that the energy of the neutron «at the emission» is measured and that the resolution is estimated to be 1 MeV FWHM for neutrons of 2.5 MeV by means of a Monte Carlo simulation [3, 6]. The overall efficiency, measured by using an Am-Be neutron source, is $2.5 \cdot 10^{-4}$ for the present geometry: the maximum event acquisition rate is 250 Hz, while the maximum neutron counting rate (without reconstruction) is of the order of 5 MHz. The background, mainly due to the photomultipliers electronic noise, is of the order of ~ 200 triggers/hour ($\pm 8\%$), reduced to ~ 68 events/hour ($\pm 10\%$) after software kinematical cuts.

Other features, like spurious bursts rejection, time distribution of the events, scattering angle *vs.* neutron energy correlation etc., have been described in the quoted references.

A cylindrical cell of 44 mm diameter and 102 mm height is located in front of the first block at a distance of 150 mm and contains the metal. The cell can be loaded with gaseous D_2 or H_2 and can also be degassed up to a vacuum of 10^{-11} bar, during the preliminary stage of the data taking, by means of a rotary and a turbomolecular pump. The gaseous content of the cell can be isolated through valves whose leak rate has been tested to be less than 0.01 bar/day of N_2 for a circuit volume of 2 litres at 3 bar. A heating system, consisting of a set of thermoresistance, is located in contact with the lower basis of the cell. Two *K*-type thermocouples, the first one embedded in the metal and the second one lying in the upper internal part of the cell, allow one to monitor simultaneously the temperature of the metal and of the surrounding gas. The pressure of the gas is monitored too, by means of a piezoresistive pressure gauge located in the upper part of the cell.

In 1991, data were taken first with the Ti/D system while, in a second time, Pd/D was investigated.

For the Ti measurements, 20 g of high-purity Ti sponge (impurities: Ni: 1 p.p.m.; Cl: 7 p.p.m.; O: 4 p.p.m.; N: 1 p.p.m.; Fe: 3 p.p.m.; C: 2 p.p.m.; Si: 1 p.p.m.) supplied by the Ginatta Torino Titanium S.p.A., were used. The operating thermal conditions of the Ti/gas system were chosen with the aim of exploring the dependence of the

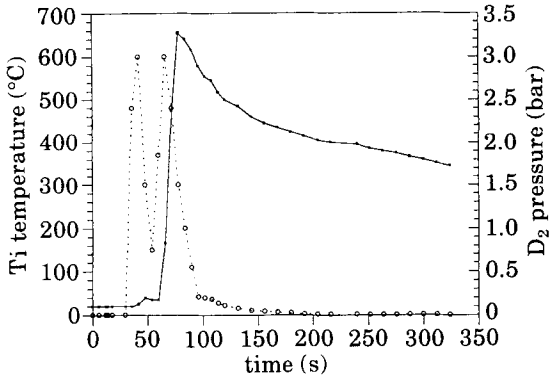

Fig. 1. – Ti temperature (black points, continuous line) and D_2 pressure (white circles, dotted line) as a function of time, during a gas loading phase. The gas has been immitted in two steps.

neutron emission, if any, upon the thermodynamic conditions. The Ti sponge, as received from the manufacturer, is covered by a thick oxide layer. Other impurities like H_2O are also both chemisorbed and physisorbed. This oxide layer acts as a barrier for the diffusion of D (H) from the surface to the lattice: consequently the absorptive capacities of Ti are strongly depressed and the uptake speed is very low or even zero. In order to restore the full absorptive capacity a careful degassing procedure at high temperature is therefore needed to clean the surface. In our experiments this procedure was considered ultimated when a residual static vacuum of 10^{-8} bar is achieved with the Ti bulk temperature at 700 °C. After the degassing step, a known amount of D_2 in the cell was dosed at room temperature. During the immission step, the pressure of the gas inside the cell raised up suddendly and the decreased slowly due to the Ti deuteride formation. As the absorption reaction is exothermic, this leads to an increase of the metal temperature. In fig. 1 the gas pressure and metal temperature are reported as a function of the time during a 2-step immission experiment. After the D_2 immission the valves were closed and the Ti/D

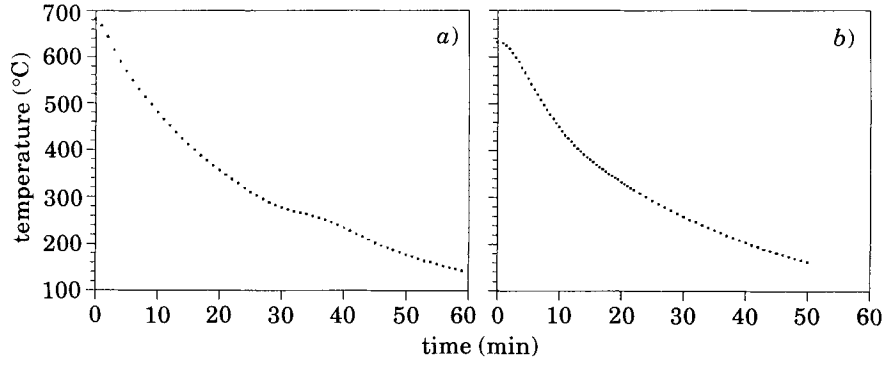

Fig. 2. – *a*) Lattice temperature as a function of the time, during the DOWN part of a cycle of the Ti/D system (initial loading ratio $X \sim 0.7$). The transition from the β to the δ phase is clearly seen as a shoulder of the curve around 270 °C. *b*) Lattice temperature as a function of the time, during the DOWN part of a cycle of the Ti/D system (initial loading ratio $X \sim 1.8$). In this case no shoulder is visible, denoting that the Ti deuteride remains in the δ phase for the whole cycle.

system was submitted to a number of thermal cycles consisting of a heating step from room temperature (~ 25 °C) up to 540 °C at least (called run UP) followed by a cooling step to the room temperature (called run DOWN). During these cycles the gas flowed out and in the metal, as monitored by the increase and decrease of the pressure, respectively. During the temperature cycles, phase transitions occur in both UP and DOWN runs, as illustrated in fig. 2*a*), where the Ti temperature is reported as a function of time during a cycle DOWN. The shoulder around 270 °C indicates the presence of the β-δ phase exothermic transition, which maintains the temperature approximately constant during the phase transformation.

During these repeated cycles, the morphology of the Ti gradually changes from sponge to a powder. This is due to the large strains associated with the hydride formation and phase transformations which cause the formation of internal cracks and fractures and ultimately lead to the crystals fragmentation.

The cycles were performed at two atomic ratios, 0.7 (~ 20% of the total data taking) and 1.8, *i.e.* near the saturation (~ 80% of the total data taking). The phase transition reported in fig. 2*a*) is related to the lower loading runs. For the higher loading runs the cycles in the Ti deuteride phase diagram were such that only the δ phase was concerned and therefore no phase transition was observed, as reported in fig. 2*b*).

The duration of a run UP was ~ 100 minutes and the total number of runs UP was 12; an equal number of runs DOWN was performed, each one of ~ 250 minutes, followed by several hours (~ 13) at steady temperature. Also, 4 runs UP and 4 runs DOWN with hydrogen gas, having the same duration of those ones with deuterium, were performed, for neutron background subtraction purposes, as explained in more detail in the next section.

The total time of measurements for all the Ti runs was 13 933 minutes with D_2 filling and 4631 minutes with H_2 filling.

For the Pd measurements we used 54 g of metallic Pd (99.9: main impurities are Au: 610 p.p.m., Ag: 260 p.p.m.), in form of small cylinders, of diameter 1 mm and length ~ 2 mm. The operating thermal conditions for the Pd/gas system were:

cycles UP from 20 °C to 350 °C,

cycles DOWN from 350 °C to 20 °C.

The total time of measurements in all the Pd runs was 2820 minutes with both D_2 and H_2.

Also in this case the system comes across a transition from the phase β to the phase α (UP) and viceversa (DOWN). The concentration of the D in Pd atoms at room temperature was ~ 0.7. At the end of the experiment the small cylinders of Pd resulted to be transformed into small spheres. This dramatic change of morphology is associated with the lattice strain release during the α-β phase transformation, which induces the formation of internal cracks and dislocations leading ultimately to a change of the morphology of the whole crystallite.

3. – Results of the neutron emission measurements.

The neutron detector can reconstruct the energy spectrum of a source located in front of the first block of scintillators. The background spectrum is mainly due to the electronic noise of the photomultipliers viewing the scintillators and can be measured

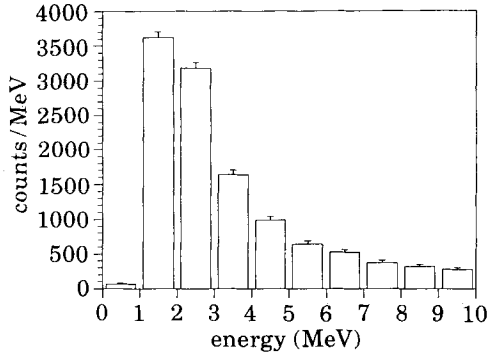

Fig. 3.

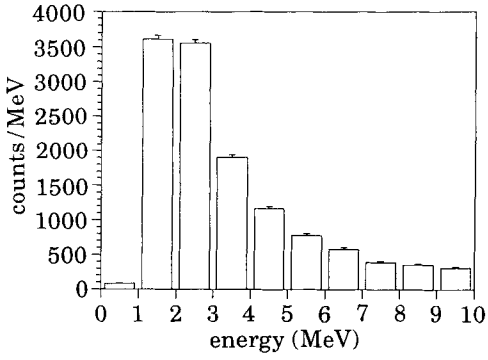

Fig. 4.

Fig. 3. – Background spectrum from the Ti/H system obtained by summing over the yield of all the UP and DOWN runs. The error bars indicate the statistical error and must be intended plus and minus. The ordinate scale is multiplied by a factor 3.0087 (ratio of the measuring time with D_2 and H_2) in order to make easier the comparison with fig. 4.

Fig. 4. – Neutron emission spectrum from the Ti/D system obtained by summing over the yield of all the UP and DOWN runs. The error bars indicate the statistical error and must be intended plus and minus.

in the absence of such a source. In order to operate during these background measurements in the same macroscopical conditions (mainly the temperature) as in the presence of the cell, several cycles were performed by filling the cell with hydrogen instead of deuterium. In this way the influence on the detector of every macroscopical effect was exactly the same for both background and cold-fusion neutron emission.

Let us now describe and discuss first the measurements on the Ti/gas system. The total background spectrum looks like that shown in fig. 3 where all the hydrogen runs, UP and DOWN, are added: recalling that the energy is evaluated from a t.o.f. measurement, the uniform time distribution due to the scintillators noise produces a flat t.o.f. distribution (see fig. 8 in ref. [4]), which was observed, and corresponds to a monotonically decreasing neutron energy distribution. The results obtained after filling the cell with deuterium are shown in the spectrum of fig. 4: one can see that the shape is similar to that of the background but the channel between 2 and 3 MeV is significantly higher. This is expected if emission of neutrons of energy ~ 2.5 MeV from the cell occurs. In order to observe and to measure such an emission, the total background spectrum, normalized in time, has been subtracted from the total spectrum with deuterium and the result is shown by fig. 5*a*): one can see that the energy channel between 2 and 3 MeV contains an excess of ~ 377 counts with a significance of about 3.9 standard deviation. Another way of searching for neutron excesses is that of subtracting from each run with D_2 filling the total spectrum obtained with H_2 filling, properly normalized in time. The counts in each channel were then obtained as the weighted mean of the values obtained for each D_2 run. The error was calculated as the standard deviation. The result is shown by fig. 5*b*) and no substantial difference is apparent between the two methods, apart from the reduction of the errors. The channel between 2 MeV and 3 MeV is again the most populated, at a $5.4\,\sigma$ level. As a further confirmation that the signal in this channel is not due to the

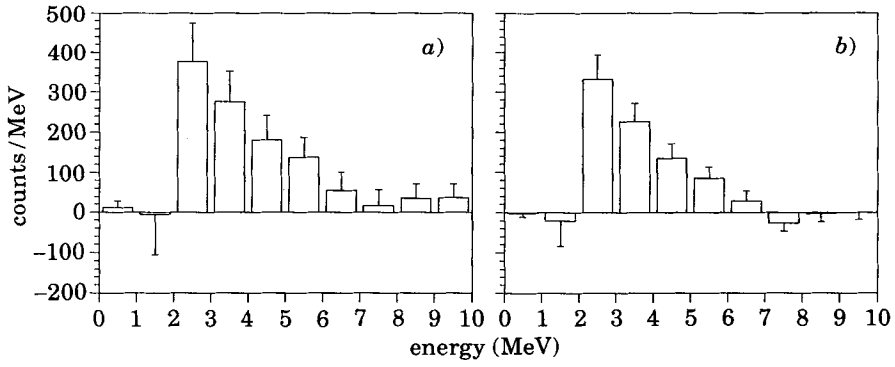

Fig. 5. – *a*) Difference between the spectra of fig. 4 and fig. 3: the error bars refer to the statistical error only and must be intended plus and minus. *b*) Spectrum of neutrons emitted from the Ti/D system calculated by the analysis of the single runs as described in the text.

subtraction method, such a procedure has been applied also to two halves of the total background measurements chosen at random: the result, shown in fig. 6, indicates a statistical fluctuation consistent with zero, as expected. This check has been repeated for several pairs of sets always chosen at random obtaining always an equivalent result.

An estimate of the neutron emission per unit mass and time was made assuming that the neutron production rate was independent of time: on this basis a result of 0.11 ± 0.03 neutrons $g^{-1}s^{-1}$ has been obtained.

As a final remark we point out the total absence of neutron bursts in our measurements, neither detected at the trigger level nor by the fast counters.

Concerning the measurements on the Pd/gas system, the total time for the data acquisition was considerably lower for both D_2 and H_2, with respect to the Ti: this was due to the decision of stopping the cycles when the Pd metal morphology showed to be highly modified with respect to the initial situation.

The same analysis applied to the Ti data was applied to the Pd runs and the result is shown by fig. 7: also in this case a small signal of ~ 70 events appears in the channel between 2 and 3 MeV, with the typical smearing on the nearest channels, but the

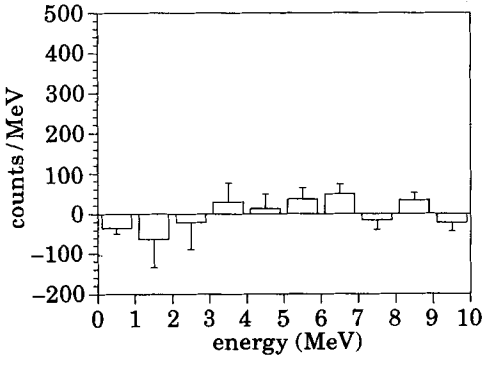

Fig. 6. – The result of the background subtraction procedure applied to two halves of background (Ti/H system) measurements. The error bars indicate the statistical error and must be intended plus and minus.

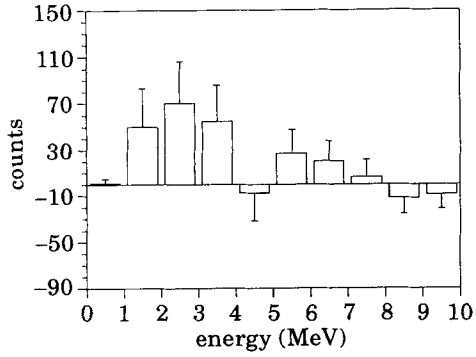

Fig. 7. – Neutron emission spectrum from the Pd/D system after the background subtraction: the error bars refer to the statistical error only and must be intended plus and minus.

statistical significance is small, less than 2 standard deviations. The neutron emission rate would be 0.02 ± 0.01 neutrons $g^{-1}s^{-1}$. Of course no burst has been counted.

4. – Conclusions.

The second set of measurements on the Ti/D system has confirmed the fact that 2.5 MeV neutrons are emitted from a Ti/D system in the gas phase, improving the statistical significance from 2.5σ to 5σ. However, substantial differences occur between the two measurements. The shape of the neutron spectrum is also slightly different. The previous result of 1.3 ± 0.5 neutrons $s^{-1}g^{-1}$ has to be compared with the present one of 0.11 ± 0.03 neutrons $s^{-1}g^{-1}$. The difference can be due or to the different forms of the Ti used (metal Ti shavings in the 1990 experiment, Ti sponge in the present one), or to the different integration times or to both effects.

Concerning the different forms of Ti, we chose this time to use Ti sponge in order to increase the surface/volume ratio. The fact that Ti sponge yields a neutron emission lower by an order of magnitude may indicate that the cold-fusion phenomenon is essentially a bulk and not a surface process, in accordance with a theoretical model [7], following which cold fusion proceeds through the formation of coherence domains in a metallic lattice. One could argue that in the previous experiment, though with an inconsistent value of the D/Ti macroscopic ratio (0.32), we reached in some portions of the lattice the conditions for the occurrence of coherence domains, in which deuterons are delocalized. A very speculative argument in favor of this hypothesis is the fact that the structure of the metal appeared not drastically changed at the end of the thermal cycles in the measurements of [1].

In the present experiment we reached higher values of the D/Ti ratio, fully consistent with those expected from the phase diagram, but at the end of the thermal cycles the sponge was completely transformed into powder. It is possible that this is an indication of the fact that our dominant chemico-physical process in this case was the formation of the hydride TiD_2, with localized deuterons, not favouring the cold-fusion process.

Concerning the background subtraction techniques, different in the two experiments, if we subtract, in the present experiment, from the runs DOWN the runs UP, properly normalized in time, we would obtain an excess of neutrons

emmitted during the runs UP corresponding to 0.08 ± 0.04 neutrons $g^{-1}s^{-1}$, that is an upper limit less than twenty times lower than in the previous case. Furthermore, in the present measurement, the neutron emission is apparently higher during the runs UP than during the runs DOWN.

Concerning the shape of the spectrum, we observe that also the channels from 3 to 6 MeV exhibit a content of events, even if less significant than in the channel from 2 to 3 MeV. A small asymmetry of the neutron peak is expected from the Monte Carlo simulation [5], but not to such an extent. We remind that emission of neutrons in the $(3 \div 6)$ MeV range was observed by Takahashi *et al.* [8] even if with different experimental conditions, namely with electrolytic cells filled with D_2 and with Pd cathodes.

Due to the reduced statistics, we could not attempt a statistically significant correlations between the neutron emission and the thermodynamic conditions. What we can only exclude is that the neutron emission is concentrated in a few bursts. Experimentally, it appears that the neutron emission is distributed along the full duration of the runs, or occurs in many smaller bursts, again distributed along all the runs.

Seeliger *et al.* [9] and Bittner *et al.* [10] reported measurement on neutrons emitted from Ti/D and Pd/D systems. Their results are in a qualitative agreement with ours in the sense that they too observed that the neutron emission rate was higher by about an order of magnitude for the Ti/D system compared with the Pd/D system in gas phase.

Very recently, Prati *et al.* [11] reported a null result for neutron emission from a Ti/D system in gas phase following thermal cycles from 77 K to room temperature, in conditions similar to chose of De Ninno *et al.* [12], and using two forms of Ti metal (shavings and powders). We followed different thermal cycles and then we cannot compare directly the results of the two experiments.

In conclusion, we have confirmed with a greater statistical significance (5σ), the emission of 2.5 MeV neutrons from a Ti/D system submitted to thermodynamic cycles. No such a significant neutron emission was observed for the Pd/D system, again submitted to thermodynamic cycles corresponding to a crossing between the α and β phases. However, the neutron rate observed in this experiment is one order of magnitude lower than that observed in a previous experiment. We attribute this difference to the different nature of Ti metal used in the two experiments and we plan to repeat with the improved cell and with a further improvement in the neutron detector the measurement with the metallic Ti shavings.

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