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Search for Neutron Emission from a Deuterium-Titanium System.

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Abstract. – Search for neutron emission from a titanium-deuterium system has been performed in a wide temperature range (from 77 K to 1100 K), with a D/Ti atomic percentage up to 165%. The sample was continuously monitored for 40 days, both at constant temperature and in nonequilibrium conditions. No neutron emission above background was observed.

The experiment recently reported by De Ninno *et al.* [1] suggested the evidence of neutrons production by a titanium-deuterium system resulting from nuclear-fusion reactions. A first preliminary analysis of the phenomenon [2] suggests that it should not be observed in equilibrium conditions. It might eventually be produced as a consequence of large stresses between zones of different concentrations, or due to transitions between different phases. A mechanism capable of concentrating the required energy to produce the deuteron fusion has been also proposed [3]. It is based on the implosion release of elastic energy, accumulated in a small lattice domain of the host metal. Both these theoretical analyses lead to the conclusion that the possible neutron emission should be time-confined within randomly distributed bursts.

Among the experiments reported so far [1, 4, 5], the largest neutron flux (more than 5000 n/s) has been observed by De Ninno *et al.* (1) using a 100 g titanium sample with D_2 gas. However, the thermodynamic conditions necessary to produce the phenomenon remained unclear, and the amount of deuterium absorbed by the titanium sample was not accurately measured. Moreover the experiment was performed between 77 K and 300 K, while for a strong deuterium absorption in titanium higher temperatures are required [6].

It seems therefore interesting to repeat this experiment, while measuring the D/Ti atomic percentage, and searching for a close correlation between the conjectured neutron emission and the thermodynamics of the system in a wider temperature range, involving phase transitions in the metal lattice.

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(1) The estimated number of deuteron pair wass less than $1.9 \cdot 10^{22}$ (A. De Ninno, personal communication).

1. Experimental apparatus.

1·1. *Titanium and deuterium system.* – The titanium sample is 35.8 grams of Ti powder (2) packed into a stainless-steel cylindric cell (26 mm diameter, 55 mm height). The Ti granulometry spans a wide range of particle diameters (from few μm up to few millimeters). The surface-to-volume ratio is therefore much higher than for the turnings sample used by De Ninno *et al.* The cell is connected to the gas handling circuits by means of a flexible filling line, that allows us to displace the cell, changing the cell-to-detector solid angle by a factor 20. Vacuum lines, valves, sealings and gaskets are all-metal. High vacuum is provided by a turbomolecular pump. The sample can be cooled to 77 K by a liquid-nitrogen bath or heated up to 810 °C by a heater wound around the cell. The temperature is measured by a thermocouple.

The deuterium gas is 99.4% purity (3). The pressure is measured by a capacitance manometer (4) in the range 10 Pa to 10^5 Pa, and by a digital Bourdon gauge (5) above it.

1·2. *Neutron detector.* – The apparatus was selected aiming at the detection of fast neutrons emitted in the fusion reaction D(d, n)^{3}He ($Q = 3.269$ MeV). The block diagram is shown in fig. 1. The neutron detector is a thin-wall aluminium cylinder (5″ diameter, 4″

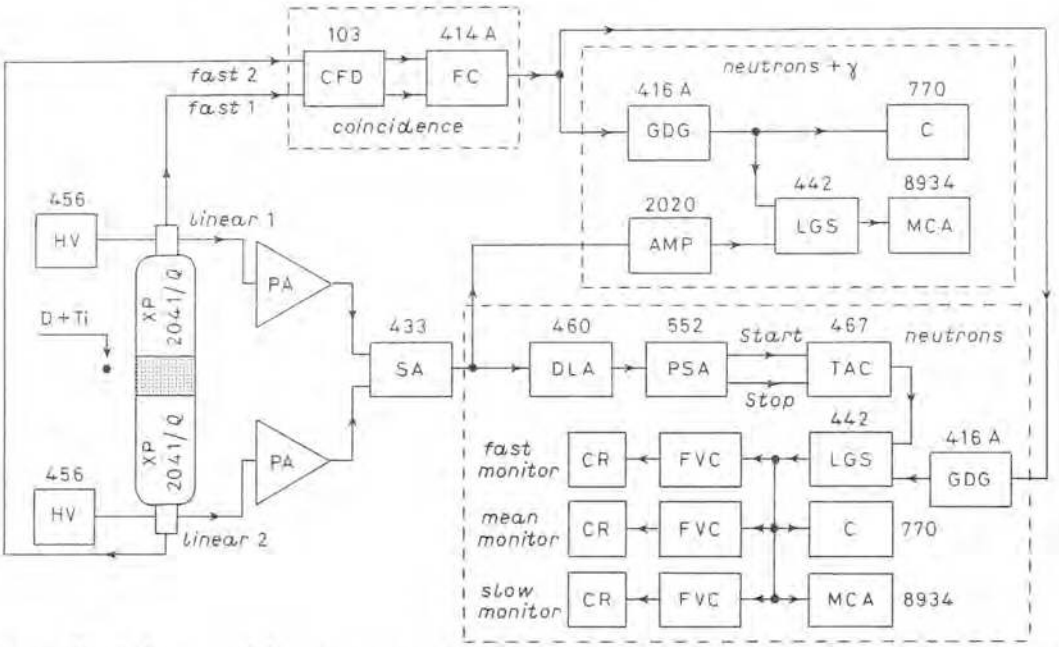

Fig. 1. – Block diagram of the neutron detector electronics. HV: high-voltage power supply, PA: linear preamplifier, SA: summing amplifier, DLA: delay line amplifier, GDG: gate and delay generator, PSA: pulse-shape analyser, TAC: time-to-amplitude converter, LSG: linear gate and stretcher, FC: fast coincidence, CFD: constant fraction discriminator, AMP: spectroscopy amplifier, C: fast counter, MCA: multichannel analyser, FVC: frequency-to-voltage converter, SG: staircase generator, CR: chart recorder.

(2) 98% purity from Carlo Erba, Milano, Italy.
(3) Deuterium N24, Società Italiana Ossigeno, Milano, Italy.
(4) Datametric: head 590 A-1000 Torr.
(5) Dresser: mod. 7100.

length), filled by NE 213 liquid scintillator (6). The lateral inner wall is coated by an efficient diffuse white reflector. The scintillating liquid is optically coupled by two specially-adapted (7) plano-concave quartz windows to the 14-stages, 5″ diameter, photomultiplier tubes. The phototubes are Philips, type XP2041/Q, selected for low dark current. The adopted twin photomultiplier arrangement combines counting efficiency enhancement with phototube noise reduction, while preserving timing resolution. Standard neutron-gamma pulse shape discrimination technique [7, 8] allows us to separate in the time-domain the neutron pulses from the gamma pulses. The bias voltage for each phototube is adjusted to match the two photomultiplier responses.

Linear signals from the 10th dynode of each tube are summed and fed to a multichannel analyser to record the pulse height distribution. Fast signals from the photomultiplier anodes are fed to a twin constant-fraction discriminator (CFD). The CFD thresholds are adjusted to set a lower limit to the signals processed in the pulse-shape n-γ discrimination channel (PSD+TAC). Coincidence between the two signals within 100 ns is imposed by a coincidence circuit (FCD). The lower threshold in the time-to-amplitude converter (TAC) is adjusted to cut most of the gamma pulses, while retaining a reasonable neutron detection efficiency. Integral fast counters (FC) measure the pulse rate at the output of the coincidence channel, and of the neutron detection channel.

Continuous monitoring of the neutron pulse rate is provided by these recording systems with different time responses. In the first one («mean monitor»), the pulses from the linear gate stretcher are used to trigger a single-shot generator giving positive pulses of 1 ms width and 12 V amplitude. An RC circuit averages the output signal of the single-shot generator with a time constant of 3 seconds. The mean voltage is measured by a chart recorder which is therefore sensitive to slow changes in the count rate. However, pulses which are grouped within one millisecond are seen as a single pulse. In order to detect also a possible signal made of closely packed pulses we used a second recording system («fast monitor»), working in parallel to the first one. Here the single-shot generator gives pulses 10 μs wide, and the time constant is 1 ms. These two systems are complementary: one is sensitive to slow drift in the count rate, and the other to short bursts of pulses. To measure the count rate in a longer time interval (thus reducing the statistical fluctuation) the integral counter was used. A third monitor («slow monitor») records the number of counts within a fixed time interval. This function is accomplished by feeding the counted pulses into a staircase generator which is reset to zero every 100 s.

2. Calibration and tests.

The efficiency of our detector for monoenergetic neutrons in the energy range 2 MeV to 3 MeV was measured using the ^{7}Li(p, n)^{7}Be reaction with proton energies varying from 3.7 MeV to 4.7 MeV. The target was a LiF layer, 200 μg/cm^2 thick, evaporated onto nuclear grade graphite backing. Efficiency measurements were performed at LNL (8) using the 7 MV Van de Graaff accelerator, and employing the fast-neutron pulsed-beam time-of-flight facility of the laboratory. Absolute efficiency of the detector was determined by measuring the neutron flux emitted at 0° with respect to the proton beam axis using the differential cross-sections for the ^{7}Li(p, n)^{7}Be reaction [9].

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(6) Nuclear Enterprise Ltd., U.K.

(7) The quartz adapters supplied by Philips have been modified to be used as windows in the detector cell.

(8) Laboratori Nazionali di Legnaro, INFN, Padova, Italy.

During the calibration the neutron detector was placed at a distance of 4 m from the neutron source. In the experiment for the search of neutron emission from deuterium-titanium the cell-to-detector distance was much shorter (14 cm centre-to-centre). To account for this difference, we calculated the geometrical factor, and we tested the distance dependence of the detector efficiency by using a ^{252}Cf neutron source (9). The overall efficiency (defined as the ratio between the counted neutrons to the total emitted neutrons over 4π solid angle) of our detector in the experimental conditions results 1.3% for 2.45 MeV neutrons.

The average background counting rate is about 7 counts/s. The sensitivities of the faster monitors are calibrated injecting a test pulse-train superimposed to the background signal at the one-shot trigger input. From these calibrations we estimate that we can detect changes of 1 counts/s above background, within 2 ms to 3 s (mean monitor) and bursts as small as 3 counts within 50 μs to 1 ms (fast monitor). The statistical fluctuation of the counting rate, measured by the integral counter (slow monitor) over a time interval of 100 s, is about 0.3 counts/s, and the long term stability, over few hours, is about 0.5 counts/s. A conservative estimation of the minimum detectable signal due to possible neutron emission from the sample cell is 1 count/s.

3. Experimental procedure and results.

The sample was first baked in vacuum at 800 °C to remove surface oxides, and flushed with pure ^{4}He. Deuterium was then admitted into the cell in five steps corresponding to a n(D)/n(Ti) atomic percentage of 21.2%, 32.0%, 74.8%, 138.0% and 165.5%, respectively. The quantity of absorbed gas was calculated from the repeated pressure drop measured in a calibrated volume.

Gas filling was performed at high temperature (400 °C $< t <$ 800 °C) in order to provide a high diffusion coefficient. Temperature changes due to heat of absorption were clearly observed, but the corresponding enthalpy change was not measured. After the last filling the maximum pressure at room temperature was 647 kPa.

The cell temperature and the deuterium concentration during the first part of our investigation is schematically reported in fig. 2. The system temperature has been suddenly changed many times after each filling. The sample was prevalently kept at three temperatures: 77 K, 300 K and 1000 K. Assuming the Ti-D phase diagram to be similar to that of Ti-H, each temperature change should involve at least one phase transition [6]. For example in regions 1 and 2 of fig. 2 the system should cross both the $\beta \rightarrow (\alpha + \beta)$, and the $(\alpha + \beta) \rightarrow (\alpha + \gamma)$ phase boundaries when temperature is lowered from the highest to the lowest value.

After nearly 330 hours of investigation, deuterium was pumped out, while keeping the cell at various temperatures, for 4 days. At this point the residual atomic percentage was evaluated to be approximately 20%, by measuring the pressure *vs.* temperature and comparing the data with the Ti-H isochores. In this condition the sample was continuously monitored for more than ten days, switching temperature several times between 77 K and 1100 K. Finally the cell was filled again up to 50% atomic percentage, and thermally cycled three more times. At the end of the experiment the cell was opened and the Ti powder was found to be sinterized.

The correct performance of our apparatus was periodically checked by recording the signal produced by the ^{252}Cf source, placed near the detector symmetrically with respect to

(9) Calibrated 252-Californium source (actual emission: 1400 neutron/s).

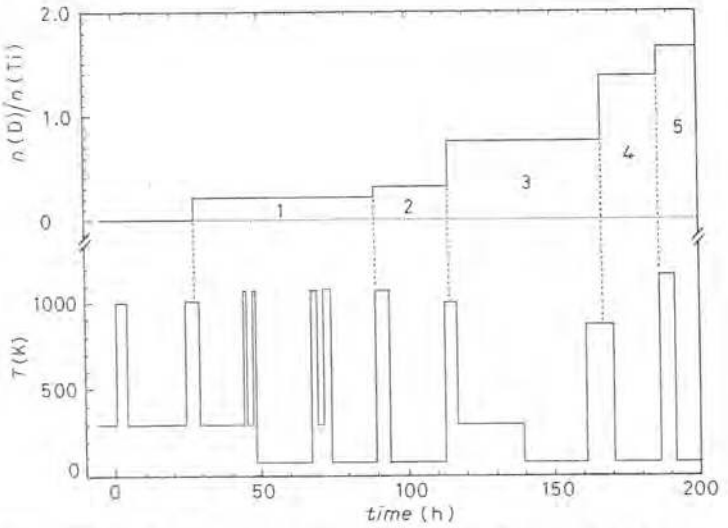

Fig. 2. – Upper side: the deuterium/titanium atomic ratio as a function of elapsed time. Lower side: schematic temperature history of the sample cell. Similar temperature changes, for a total of 80, have been performed during the successive 750 hours. Heating and cooling were performed at various speeds: the full temperature span was accomplished within time intervals ranging from 10 minutes to 1 hour.

the sample cell. The maximum change in the background count rate, during the whole experiment, was less than 2.5 counts/s, with day-by-day changes smaller than 1 count/s. We proved, however, that these changes were not due to neutron emission from the deuterated titanium, simply by moving the sample cell far from the detector.

During our uninterrupted investigation lasting 40 days (from June 6 to July 16) we did not observe mean rate changes related to the D/Ti system greater than 1 count/s, nor fast spikes greater than 3 counts. Taking into account the overall detector efficiency for 2.45 MeV neutrons, we may conclude that the mean neutron emission rate from our sample, if any, was always smaller than 75 neutrons/s.

In the work of De Ninno *et al.* the maximum neutron emission was $5000\,s^{-1}$ corresponding, with a deuterium percentage not greater than 3%, to a fusion rate for the neutron production branch $\lambda_F > 2.6 \cdot 10^{-19}$ neutrons per second per pair.

In our experiment, at our maximum deuterium concentration (165% D/Ti atomic percentage), the neutron emission that could remain hidden in our background signal should correspond to $\lambda_F < 2.0 \cdot 10^{-22}$ neutrons per deuteron pair per second, three orders of magnitude smaller than in Frascati[1] experiment. We have no explanation for this disagreement.

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