ON LOW INTENSITY NEUTRON EMISSION FROM TID_X SAMPLES UNDER NON-EQUILIBRIUM CONDITIONS

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ABSTRACT

Several experiments of loading Titanium samples with Deuterium from the gas phase, of changing the temperature of the samples over a wide range and of monitoring the neutron emission were done. Neutron emissions in very low intensity bursts, still significantly above the background were recorded, revealing that low energy nuclear reactions in condensed matter can be produced with a very low rate, which occasionally can be high enough to become detectable.

I. INTRODUCTION

Since the first announcement [1], which stated that nuclear fusion of Deuterium nuclei occurred at low temperature without any external acceleration, experimental work to verify the statement and to investigate the processes has been done all over the world [2] - [5] in sophisticated and in poorly equipped laboratories as well.

In order to verify that nuclear reactions occurred in condensed matter, nuclear radiation should be present; i.e. γ rays, charged particles or neutrons should be detected to a significantly high level during the experiment. The experiments described hereafter were performed to detect fast neutrons emitted from the sample. Our supposition, that the 2.45 MeV fast neutrons are produced in the nuclear reaction:

$$d + d = {}^3He + n + 3.27\ MeV \quad (1)$$

is suggested by the results of the recoil proton neutron spectroscopy performed on other H isotopes absorbing metals loaded with Deuterium, under non-equilibrium conditions, reported in papers like [6]. The experiments were focused on searching for neutrons and not the γ radiation

or charged particles because the neutrons are less attenuated by the 2 cm thick stainless steel wall of the reactor used for loading the samples.

II. EXPERIMENTAL SETUP

A non-spectrometric system, consisting of a BF_3 proportional counter (Nuclear Enterprise), an electronic counter and a printer was used for the fast neutron detection. In order to reduce the background produced by the natural 1.46 MeV γ line of the ^{40}K contained in concrete, a 5-cm thick Lead slab served as a ground shield for the experiments performed in the Nuclear Facility of I.T.I.M. Cluj - Napoca. The BF_3 detector, with a cathode of 2 cm diameter, has a pressure of 400 Torr and an active zone 20 cm in length is placed (by producer) in a 30 cm diameter polyethylene moderator. The sensitivity is 4 (imp/sec)/(n/cm^2 sec) for thermal neutrons. This information is provided by producer and consistent with the results of the check performed on a regular basis. The total detection efficiency of the experimental setup is 509 counts for 10^6 fast neutrons generated where the sample is located. The experimental setup schematic is presented in Figure 1. The sample holder is either an electric heater a Dewar vessel. The thermal insulation consists of a 2 cm thick glass wool placed between two Cooper slabs, 0.1 mm thick each. It maintained a constant temperature at the surface of the polyethylene moderator. This temperature was measured often during both the high and low temperature experiments and was found to be within 1 ^oC of the air temperature

III. CONTROL EXPERIMENTS

The reactor without the Deuterium gas and the sample inside was placed in the sample holder. The neutron background was monitored to determine the occurrence of the counting rates above the background with more than 3 standard deviations (3σ). This is the control experiment for the post-Deuterium loading neutron emission and was repeated three times.

During the second control experiment the reactor was placed in an electric heater and the heater was turned on, the power was maintained for 4 hours and then turned off. The neutron background was continuously monitored during this procedure and long after that. This was the control experiment for the high temperature cycling neutron emissions and was repeated two more times to make sure that neither the power supply nor the heater induced any false neutron counts.

For the third control experiment the reactor without the Deuterium gas and the sample inside was sunk in a Dewar vessel containing liquid Nitrogen, maintained for 4 hours by adding Nitrogen and then the temperature came back to the room temperature. The neutron background was monitored during the procedure and long after. This experiment was repeated three times to verify that the processes did not induce any false neutron counts.

The purpose of both the second and the third experiment was not only to verify whether false neutron counts are recorded but also to verify if the temperature of the detector is changing and if so, if it induces false neutron counts. The temperature on the surface of the polyethylene moderator was measured often during both the high and low temperature experiments. It was found to be within 1 ^{o}C of the air temperature, which suggests that the temperature of the detector placed inside the 30 cm diameter moderator did not change significantly during the control experiments.

It is a standard practice to define a neutron emission from a sample, or event, to be a counting rate higher than the background with more than 3σ [7]. The counter worked with counting cycles of 10 minutes, printing each result. The counting time was not changed during the experiments, therefore all the results refer to 10 minutes. The results of three of the control monitoring runs, each lasting more than the experiments that were performed, are presented in Table I. The **f** value means the ratio of the high count rates to the total number of records for the time span of the experiment. These were the control experiments for the post Deuterium-loading neutron emission monitoring experiments.

The highest rate was 0.7%, therefore it might be considered that a rate significantly higher then 0.7 % in an experiment can be associated with neutron emissions.

Another method employed to test if a sample emitted neutrons was to analyze the distribution of the recorded values. A Poisson or a Gaussian distribution indicates that the neutrons are produced by one source. Distortion of a normal distribution indicates a perturbation produced by additional source [7]. For a Gaussian distribution, the theoretical standard deviation can be computed as:

$$s_t = \sqrt{< N >} \quad (2)$$

where <N> is the average of the recorded values. The standard deviation can be computed using the experimental recorded values as:

$$s_e = \sqrt{\frac{\sum_{i=1}^{n} \left(N_i - < N >\right)^2}{n-1}} \quad (3)$$

with n being the total number of recordings. If the two calculated values of the standard deviation are close to each other the distribution is not distorted, but if the relative difference, $(\sigma_e - \sigma_t)/\sigma_t$ is large then the distribution has a distortion. The results of the standard deviation calculations for the neutron background monitoring are presented in Table II. They show that the highest relative difference for the neutron background is 8%.

The next section presents the procedure employed for preparing the samples and for loading them with Deuterium.

IV. SAMPLE LOADING

The samples are placed in a stainless steel reactor that can withstand a 9 MPa pressure from Hydrogen isotopes and temperatures ranging from –250 ^{o}C to 900 ^{o}C. The gas is removed from the reactor using a vacuum pump and the samples are maintained at 10^{-1} Torr and 500 ^{o}C for three hours to ensure a complete sample degazing. All four samples were activated in this manner, but the loading process was conducted differently for each of them as will be discussed. The samples consisted of Titanium small slabs, 1.5 mm x 8 mm x 9 mm.

The absorbed Deuterium mass was calculated using data from pressure and temperature monitoring in a known volume for the first three samples and by weighing the sample precisely for the fourth of them.

After the first sample was activated, the Deuterium gas at a pressure of 7.15 MPa filled the reactor containing the sample at a temperature of 500 ^{o}C and then the temperature decreased slowly as the electric heater was disconnected. Subsequently the reactor valve was closed. The pressure decreased rapidly as Deuterium was absorbed into the sample.

The same procedure was used to load the fourth sample while the second sample after activation was maintained at a temperature of 820 ^{o}C for 10 minutes with the Deuterium at an initial pressure of 8.25 MPa and then cooled at 13 ^{o}C during 3 minutes. These samples presented loading ratios between 0.573 and 1.93. The third sample was activated and then maintained at 680 ^{o}C with the initial Deuterium gas pressure of 6 MPa; the reactor with the sample was maintained at this temperature for 5 minutes and then cooled at 13 ^{o}C during 3 minutes. The

reactor was closed as soon as the Deuterium reached the pressure of 6 MPa. This different treatment was applied to establish a lower value for the loading ratio and a higher pressure of the Deuterium gas in the cold reactor. After this procedure more temperature variation cycles were done with the sample inside, allowing it to absorb Deuterium several more times.

Loading pressure and temperature, sample mass and loading ratio X defined as the number of D over the number of Titanium host lattice ions, for the four samples are presented in Table III.

The Deuterium absorption in the samples labeled as 2 and 3 was very fast, leading to the rapid creation of a high loading ratio region under the surface and producing mechanical strain in the bulk of the sample. The strain induced macroscopic cracks on the surface of the slabs accompanied by sound emission during the first 15 minutes after the beginning of the Deuterium absorption.

The results of neutron monitoring during sample loading are presented in Section V.

V. NEUTRON EMISSION AFTER LOADING TI WITH DEUTERIUM

The sample loading and the experiments described in this section were conducted in the Hydrogen Isotope Laboratory of I.T.I.M.-Cluj and the difference in the experimental setup was that the detector was not placed on the Lead protection, which is part of the floor of the Nuclear Facility.

Neutron emissions from samples 2, 3 and 4 were monitored beginning with the moment the Deuterium reached the Titanium surface for samples 2 and 4, and starting 5 hours after that for the sample 3. The temperatures of the samples decreased as the reactor was losing heat in air. The samples were not subject to any other temperature variation during this type of experiment. Results are presented in Table IV, where No. n emissions is the number of records that are above the background with more than 3 σ, e/t is the ratio of the total emission time to the total experiment time, R is the average counting rate and B is the average background, where both R and B are in counts for 10 minutes.

If we compare Table I describing the control experiment with Table III we notice that e/t is greater then 0.7%, which was the highest value found during the prolonged background monitoring. Therefore we can consider that during all three experiments neutrons were emitted with a very low intensity. The most relevant, in respect to neutron emission, was the experiment

conducted with sample 4 and the variation of the neutron counts during the experiment is presented in Figure 2. An interesting feature of the neutron emission is that it occurred randomly and in bursts throughout the experiment.

The average background was 225.8 neutrons for 10 minutes, with σ=15. The high values appeared both isolated and as groups over time intervals of 30 to 40 minutes. Another interesting feature of the results is that the average counting rate is higher than the background and for the last experiment with as much as 1.64σ.

VI. NEUTRON EMISSION DURING TEMPERATURE VARIATION BETWEEN 20 ^{O}C AND 700 ^{O}C

During these experiments each of the samples was closed inside the reactor where the Deuterium pressure was 3.5 MPa for the first sample, 1.2 MPa for the second sample and 2 MPa for the third sample at the beginning and decreased as Deuterium was absorbed.

One run was conducted for samples 1 and 2 and three runs for the sample 3. Sample 3 was the sample that was loaded at a lower loading ratio to enable it to absorb Deuterium during these experiments. The results of the five runs are presented in Table V, the header being the same that of Table IV. It shows that e/t has values considerably above 0.7%, which was the highest value found during the control experiment.

The first sample was heated and maintained at 700 ^{o}C for 2 hours and cooled slowly to 20 ^{o}C. The pressure inside the reactor was 1 MPa. The second sample was heated and maintained at 700 ^{o}C for 3 hours and cooled rapidly to 20 ^{o}C in 3 minutes; the Deuterium pressure in the reactor was 1 MPa. The third sample was heated and maintained at 600 ^{o}C for 6 hours and cooled slowly to 20 ^{o}C; the initial pressure was 2 MPa and was not monitored during the other runs. The fourth sample was heated and maintained at 670 ^{o}C for 3.5 hours and cooled slowly to 20 ^{o}C.

The variation of the counting rate during the heating experiment performed with sample number 3 is presented in Figure 3. Most of the count rates are above the background.

A spectacular neutron emission was recorded 30 minutes after the heating of the electric heater containing sample 2 began; it lasted 20 minutes and the records were 415687 and 2492 neutrons, while the average neutron background was 400. The emission occurred during the

sample's temperature increase, but the temperature of the sample inside the pressurized reactor was not monitored precisely.

The results presented in Table V show a high e/t ratio, close to 1/3 for sample 2 and an average counting rate higher than the background, especially for sample 3, which are a good indication of neutron emission during the temperature variation experiments.

VII. NEUTRON EMISSION DURING TEMPERATURE VARIATION BETWEEN 293 AND 77 K

This type of experiment consisted of monitoring the neutron emission from Deuterided titanium samples during rapid temperature decrease produced by immersing the samples in liquid Nitrogen.

The first experiment involved sample 1 inside the closed reactor. The reactor was placed near the detector and liquid Nitrogen was poured over it. The heat capacity of the reactor caused the temperature decrease to decrease slowly. After the temperature decreased, the reactor was completely covered with liquid Nitrogen and it was maintained at 77 K for 2 hours, then it was extracted and heated slowly to 20 ^{o}C. Then immersing was repeated and the Nitrogen evaporated slowly. Neutron emission was monitored for 24 hours.

During the second run sample 1 alone was cooled very rapidly to 77 K by simply dropping it in liquid Nitrogen near the neutron detector. The same procedure was used for sample 2. The temperature decrease during the last two experiments was very rapid as the temperature variation of 216 K occurred in a few seconds. The results of the data analysis for these three experiments are presented in Table VI, header having the same meaning as in Tables IV and V. Again we notice that e/t ratio had considerable higher values during the low temperature experiments than during the control experiments, meaning that neutrons were emitted.

During the experiment performed with sample 2 the average background was 206. A sequence of neutron emissions occurred when the liquid Nitrogen evaporated and the temperature of the sample was rising slowly. These sequences, located after a burst of 4821 recorded neutrons and detected 18 hours after the immersion, are presented in Figure 4.

VIII. DISCUSSIONS

Simple experiments of monitoring the neutron emission during loading Titanium samples with Deuterium and during temperature variation over a wide range were done. Data from experiments described in this paper was analyzed to check if their distribution is a Gaussian one, as described in Section III. The theoretical and experimental standard deviation were computed for all the experiments performed with samples 2, 3 and 4 and results are presented in Table VII. It shows that the relative difference between the experimental and theoretical standard deviation is considerably larger then the highest value found for the background, which is an indication that the normal distribution was distorted by the random neutron emission from the samples.

Not only the large differences between the theoretical and experimental standard deviations but the higher frequency of the counting rates in the experiments described in this paper clearly demonstrate that the Titanium samples highly loaded with Deuterium, under thermal non-equilibrium conditions, produced anomalous neutron emissions. The neutron emissions occurred in bursts and were randomly distributed during the experiment time.

The anomalous neutron emissions present similar features with the results found in other experiments of loading certain metals with Deuterium, under non-equilibrium conditions, as described in [8] - [15]. A satisfactory explanation for the effect is lacking, to the best of our knowledge. More experimental work is required to determine precisely which are the triggering conditions for the neutron emissions.

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FIGURE CAPTIONS

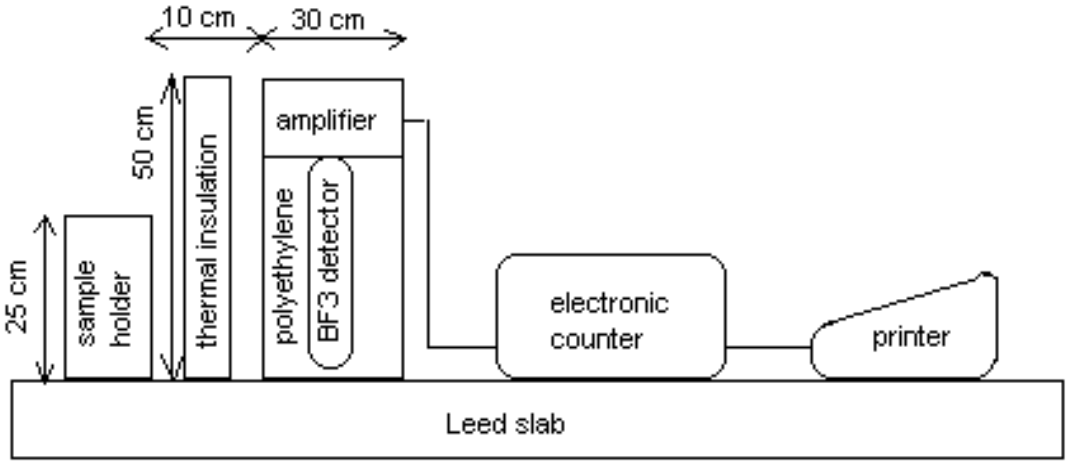

Figure 1. Schematic of the experimental setup.

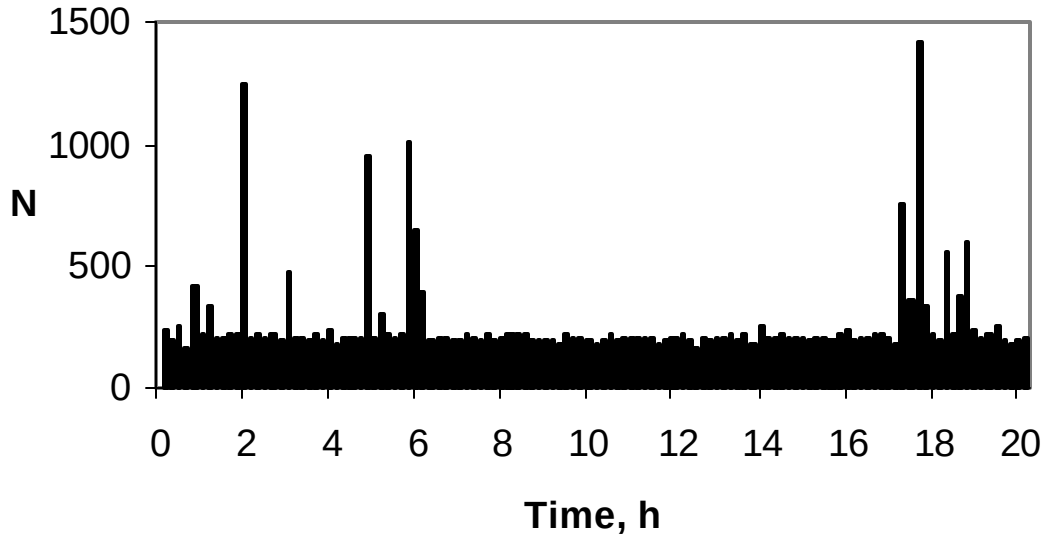

Figure 2. Neutron counts (for 10 minutes) after loading Titanium with Deuterium. The average background was 226 neutrons, with a standard deviation of σ=15.

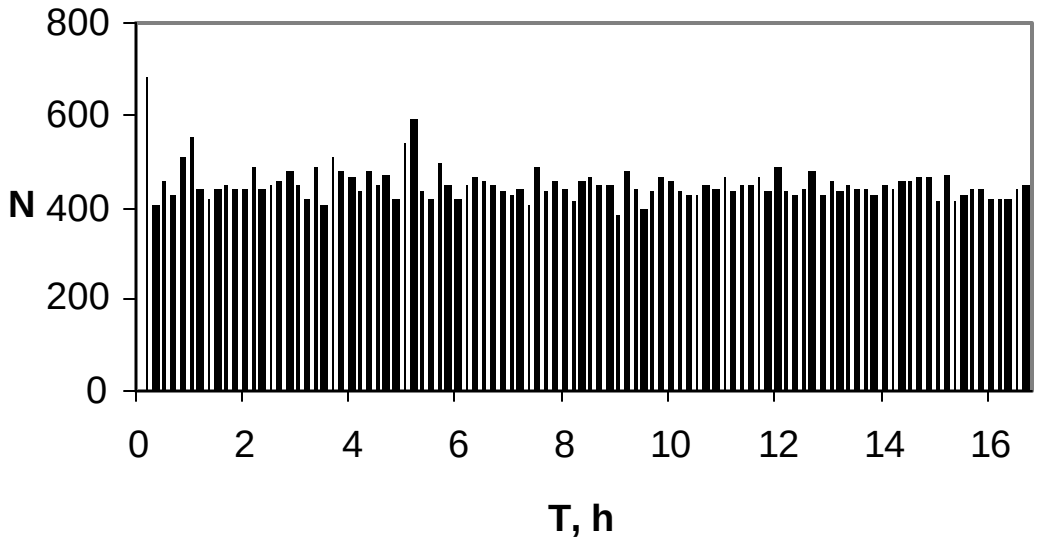

Figure 3. Neutron emission during high temperature cycling with sample number 3. Average background was 400 neutrons, with a standard deviation of σ=20.

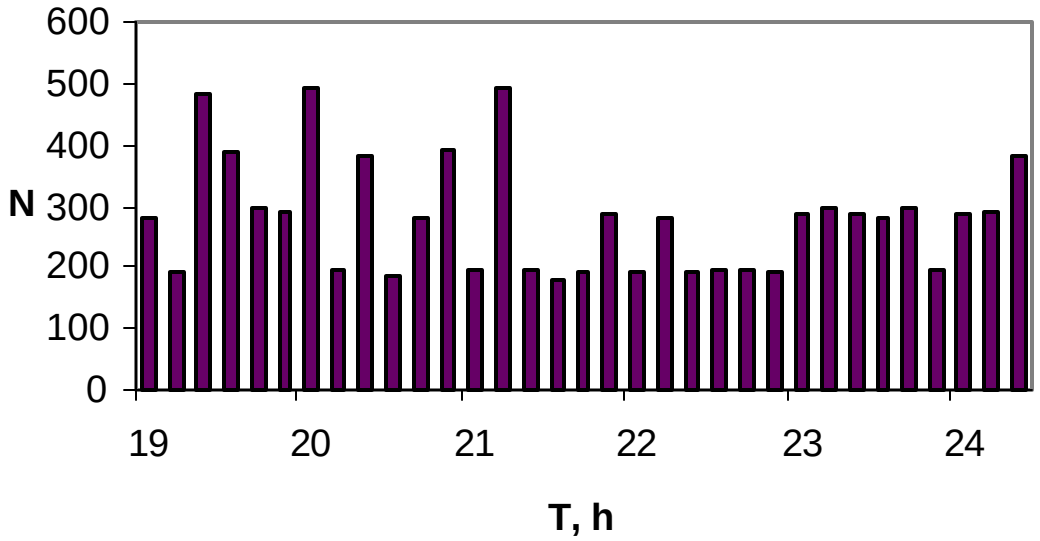

Figure 4. A sequence of neutron bursts during slow temperature increase from 77 K, for sample number 2, at the end of the experiment. The average background was 206 neutrons with a standard deviation of σ=14.4.

Table captions

No.	Time, h	No. records	No. of events	f, %
1	23.5	140	0	0
2	22.17	133	0	0
3	73.17	433	3	0.7

Table I. The neutron background.

No. Exp.	No. of records	<N>	σ_t	σ_e	$(\sigma_e-\sigma_t)/\sigma_t$, %
1	140	236,76	15,39	16,29	6
2	132	379.71	19.49	20.22	4
3	433	290.24	17.04	18.34	8

Table II. Results of statistical calculations for the neutron background monitoring.

Sample	Initial Pressure, Mpa	Initial Temperature, ^oC	M_{Ti}, g	M_D, g	$X=N_{Ti}/N_D$
1	7.15	500	9.70	0.2322	0.573
2	8.25	820	6.90	0.556	1.93
3	6	680	3.90	0.308	0.31
4	7.15	500	4.9350	0.2771	1.345

Table III. Loading conditions, mass and loading ratios for the four samples.

Sample	No. of records	No. n. emissions	e/t, %	(R – B)/ σ
2	102	3	2,9	0,2
3	110	3	2,7	1,1
4	121	16	13	1,64

Table IV. Results of the post-Deuterium loading neutron emission.

Sample	No. of records	No. n. emissions	e/t, %	(R – B)/ σ
1	110	2	1.8	0.83
2	25	8	32	2
3	100	26	26	2.5
3	127	13	10.2	162
3	162	16	3.7	0.12

Table V. Neutron emission as a consequence of temperature variation between 20 ^{o}C – 700 ^{o}C.

Sample	No. of records	No. of emissions	e/t, %	(R – B)/ σ
1	140	2	1.4	0.014
1	22	7	31.8	3.6
2	146	30	21	3

Table VI. Neutron emission during rapid temperature decrease.

Sample	No. of records	<N>	σ_t	σ_e	$(\sigma_e-\sigma_t)/\sigma_t$,%
2	272	214.01	14.63	56.95	289
3	282	294.71	17.17	22.92	33
4	116	231.37	15.21	177.11	407

Table VII. Statistical calculations of neutron emission data for samples 2, 3 and 4.