LNF-90/017

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Estratto da: Conference Proceedings Vol. 24 "Understanding Cold Fusion Phenomena"
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RESULTS OF THE 1st GENERATION EXPERIMENTS, AT GRAN SASSO UNDERGROUND LABORATORY, ON NUCLEAR COLD FUSION.

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ABSTRACT

Since April 15th '89, we have performed several measurements at the underground Gran Sasso Laboratory of nuclear radiation emission from an electrolytic cell of D_2O and LiOH using Pd or Ti(Al) as cathode electrodes, in the framework of the so called Nuclear Cold Fusion. The almost unique characteristics of such underground laboratory, located under about 3500 meter of water equivalent, is an extremely low value of thermal and fast neutron background. In a bunker built with low-activity lead bricks, to further reduce the gamma ray flux coming from the concrete and spurious materials of the experimental set-up, we allocated a set of different nuclear detectors in order to put in evidence gamma rays, neutrons and charged particles arising from the electrolytic cell. We report two emission events:

a) both a "shot" signal and some bursts of signals, simultaneously in almost all of the detectors (may 2, at $\sim$ 23:30), from a just-prepared Pd electrode;
b) burst signals from an over-stressed Ti(Al) electrode (may 1, at $\sim$ 13:30).

The time evolution of these bursts signals is quite similar. A rough evaluation of the fusion rate during the burst, supposing that these emissions are due to cold fusions, is of the order of $10^{-19} \div 10^{-20}$ fusions/deuteron pair/second in the case of Pd electrode and still larger for the Ti(Al) one.

INTRODUCTION

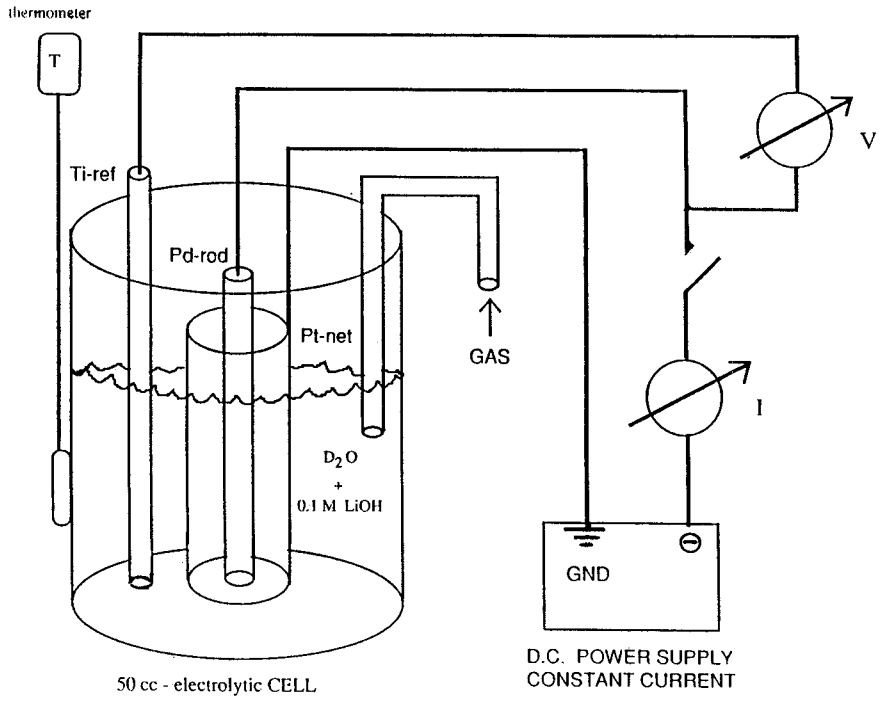
Taking into account the experiments of so called "cold nuclear fusion" [1,2], during electrolytic infusion of Pd or Ti in D_2O, a similar one using thermal stressed titanium in contact with pressurized deuterium gas [3] and some preliminary experiments, performed by us in the Frascati National Laboratory of the Istituto Nazionale di Fisica Nucleare, with an electrolytic cell [4], we realized that low level and unpredictable signals coming from such kind of cell needed a very quiet and "clean" laboratory to be detected clearly and undoubtedly. Following this, we decided to move our experimental apparatus inside the underground Gran Sasso Laboratory having these characteristics:

a) thermal and fast neutron flux about 1000 times lower than normal environments, i.e. of the order of $10^{-6}\,n \cdot cm^{-2} \cdot s^{-1}$, [5].

b) gamma flux about 10 times lower than sea level and further reduced to about 50 times by a bunker of low activity Pb bricks (Bolidn, Sweden).
c) muon flux about 10^6 times lower then sea level (i.e. $\simeq 1.2 \cdot 10^{-8} \mu \cdot cm^{-2} \cdot s^{-1}$) [10].

THE ELECTROLYTIC CELL

All our experiments were performed on electrolytic infusion of about 40 cc. D_2O + 0.1 M LiOH with a Pd-Pt or Ti-Pt system and a small regular addition of a proper gas. The geometry of electrolytic cell is coaxial with the Pt used as cylindrical external net and connected to the electrical ground in all the measurements. The Pd or Ti working electrode has been placed at the center of the Pt net electrode axis by proper teflon disks (fig. 1).



[Fig.1] Experimental set-up, not in scale, of electrolytic cell and power supply section. The Digital Multimeters (DMM) are FLUKE mod.77, the power supply is LARCET mod.3/60, the thermometer is J-type thermocouple from Keithley. All the connections between the electrolytic cell and the power supply or DMM are made by coaxial cable in order to reduce the possibility of noise pick-up. During the "nuclear" measurements all the DMMs and the thermometer are switched-off to avoid spurious signals due to the sampling.

The material used, the geometrical dimensions and the composition are:

a) Pt: cylindrical net, $\phi = 15$ mm, l = 50 mm; 95 % Pt, 5 % Ir (Metalli Preziosi - Italy)
b) Pd: cylindrical rods, $\phi = 0.2, 2, 4, 8$ mm, l = 60 mm, 99.98 % (Engelhard)
c) Ti(Al): bar $5 \times 5 \times 50$ mm, 95.5 % Ti, 4.5 % Al (ILVA Terni, Italy)
d) Ti_{ref} : bar $1 \times 1 \times 100$ mm, 99.99 % (ENI, Italy)
e) D_2O : 99.98 % , Tritium < 74Bq/ml, (Carlo Erba, Italy)
f) LiOH: 98 % LiOH, 2 % Li_2CO_3 (Merck, W.Germany)
g) Jar: conic, Erlenmeyer type 50 ml, borosilicate glass (Schott Duran, W.Germany)
h) CO_2: regularly added to the solution in very small quantity.

The Pd electrodes were carefully vacuum degassed at 650 Celsius for 2 hours and slowly cooled down to room temperature under vacuum condition ($1.0 \cdot 10^{-3}$ mm Hg) just before being used in the experiments.

The Ti electrode underwent an heavy treatment at a degassing temperature of 1050 Celsius, in a atmosphere of purified Ar flux with Oxygen content < 1ppM, for 2 hours and slow cooled to room temperature in Ar flux. Both type of electrodes were kept in a dry box until their use.

The hyperpure Ti was used as reference electrode in order to monitor the so called "spontaneous voltage" of working electrode. The cleaning chemical treatment was: HF 30 % concentration, 150 by volume H_2O_2, deionized H_2O, vacuum dried. In the following the "voltage under charge" means the voltage between the reference electrode and the working electrode (Pd or Ti(Al)) when the power is supplied by the constant-current power supply. The "spontaneous voltage" is the same voltage when the power supply is in the open circuit state.

The cell was kept in the dark and the current density was about $60\text{mA} \cdot \text{cm}^{-2}$ related to the working electrode area as suggested by both Fleischmann and Jones [1,2].

We added regularly some gaseous CO_2 to the electrolytic infusion in order to increase both the deuterium intradiffusion in the Pd electrode, which allows a faster diffusion of atomic deuterium into Pd, and the lattice cracking rate [7], in the framework of fracto-emission.

THE DETECTORS AND THE EXPERIMENTAL PROCEDURE

All the detectors surrounded the electrolytic cell as close as possible, within a radius of less than 10 cm. The detection system was based on:

a) one cylindrical 5×5 in. NaI(Tl) gamma detector (*Quartz & Silice*, France);
b) two He^3 neutron detectors, cylindrical shape of 100 cm length and 5 cm diameter, 3 atm. of pressure, nominal thermal neutron sensitivity of $433 \cdot \text{cn}^{-1} \cdot \text{cm}^2$ (model 100 He3/228/50G from *Centronic*,England),the first one was bare and the second one was surrounded of 120 cm long, 30 cm diameter paraffin moderator,[5];
c) one fast plastic scintillators optimized to detect charged particles, $1 \times 10 \times 10$cm.

We used a standard shaping time for NaI(Tl) (0.5μs) and He^3 (2μs) detectors (shaping amplifiers mod 7611L from *Silena*, Italy). The output (rise time < 10ns) from the fast scintillator was sent directly to a level discriminator regulated at 100 ns pulse width. The energy spectrum from the NaI(Tl) detector, is collected by a multichannel analyzer.

The threshold for neutron counters is set well below the capture peak. Two different ("low" and "high") thresholds are used for the NaI(Tl) detector, respectively corresponding to about 0.8 MeV and 2 MeV. The threshold for the plastic scintillator counter is set in order to reject low-energy gammas, while it is fully efficient for " minimum ionizing particles".

The data acquisition system is based on a standard *CAMAC* crate, which houses the modules required for acquisition from digital counters. It is controlled by a *CAVIAR* microcomputer [8]. We acquired counts coming from all detectors, read by the afore mentioned devices.

At the beginning we measured the radiation background, for some hours, and checked it before and after any changing of electrode or apparatus environment. We often repeated this background measurements by putting the cell into the apparatus detection system and very far from it. We also have performed a series of tests on the apparatus to avoid any kind of false "nuclear" signals coming from natural radioactivity of each component of our measurement area or any kind of electrical (dc and ac) thermal or mechanical realistic perturbations. In details, with related results, the tests we performed are the following:

a) we checked each component of electrolytic cell one by one with null results;
b) we sometimes noted a very slight increase of counts when we assembled all the component of the electrolytic cell without external current flow;
c) the effect reported in b) disappeared when we replaced the Pd electrode with a 5 mm diameter 6 cm long iron nail;
d) we supplied the usual dc current of 60mA/cm^2 to the electrolytic cell using the iron nail electrode with null results. This test has been extended up to 3A of total current and 3 hours of running time;
e) we noted a slight increase of counts when the cell is powered at the usual 60mA/cm^2 current density;
f) we ran, in standard "conditions", for more than 4 hours another complete electrolyte cell (with obviously a new Pd electrode, 4mm thick, as prepared) using deionized "light" water obtaining null results.

Single counts are acquired from all detectors. Because of the limited mass storage capability of our system, we use an on-line reduction of data. Single counts integrated over 0.6 s are acquired from all counters. If any of them exceeds a software threshold, the entire set of frequencies is written on the disk, otherwise it is ignored. Moreover, counts from all detectors integrated over 1 min. are always recorded.

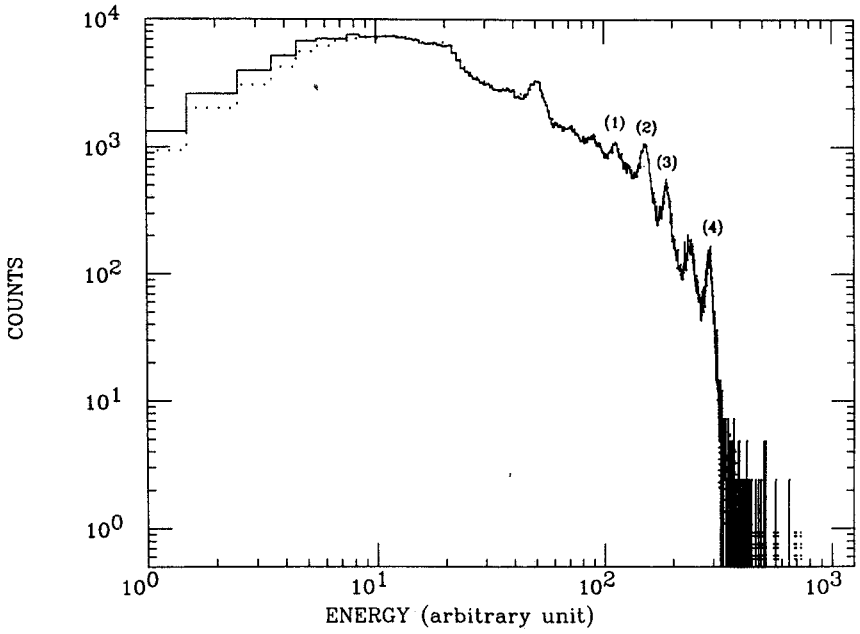
Typical frequencies of count for the detectors (putting the working cell far away from the detection apparatus), and their respective software thresholds for data reduction are:

NaI(Tl) low-threshold	= 6 Hz	(12 counts/0.6 s)
NaI(Tl) high-threshold	= 0.3 Hz	(2 counts/0.6 s)
He^3 bare	= 0.3 Hz	(2 counts/0.6 s)
He^3 with paraffin	= 0.1 Hz	(2 counts/0.6 s)
Plastic scintillator	= 25 Hz	(25 counts/0.6 s)

RESULTS

From the beginning of the first charging-up of the 8 mm diameter Pd electrode, we collect, in the multichannel analyzer (MCA), the signals coming from the NaI(Tl) detector at an energy threshold (about 200 KeV) as low as possible compatibly with the dead-time limit of MCA. Due to the long acquisition time adopted (5116 s) for the MCA, the energy spectra collected during the short emission time of the Pd electrode,i.e. shot and burst events,showed signals with low statistical significance. Anyway, in fig. 2, we report in log-log scales both the background (taken just about 3 hours before the beginning of the experiment), in dotted lines, and the data during the charging-up state, in solid lines. The data are normalized to the acquisition time of background (12268 s) and for a better visual readability we don't report statistical errors. Furthermore, we mark with numbers four of the most intense natural radioactive lines present in our experimental set-up

(Bi^{214}, 1120KeV; K^{40}, 1460KeV; Bi^{214}, 1764KeV; Tl^{208}, 2615KeV).

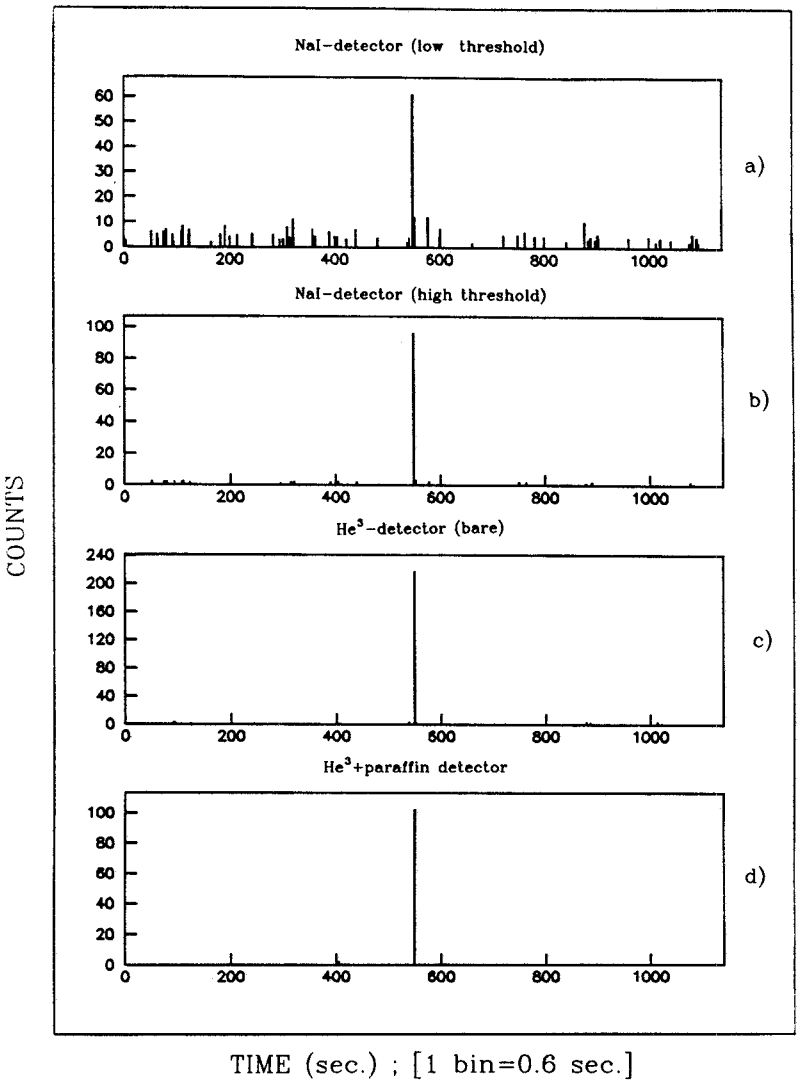


[Fig.2] Log-Log energy spectra of signals coming from NaI(Tl) detector. The background is in dotted line, the data from Pd electrode are in solid line. The data have been normalized to the background acquisition time (12268 s).The numbers (1),(2),(3),(4) correspond to the most intense natural radioactive lines, respectively at (1120,1460,1764,2615) KeV.

In this paper we report two kinds of observed phenomena. The first one is a so called "shot" event (time duration $\leq$ 0.6s) (fig. 3), followed about 40 minutes later from several bursts (total time $\approx$ 4minutes) on the 8 mm diameter Pd electrode, (fig. 4). The second phenomena are several bursts (total time $\approx$ 6minutes) coming from an over-stressed Ti(Al) electrode, (fig 5).

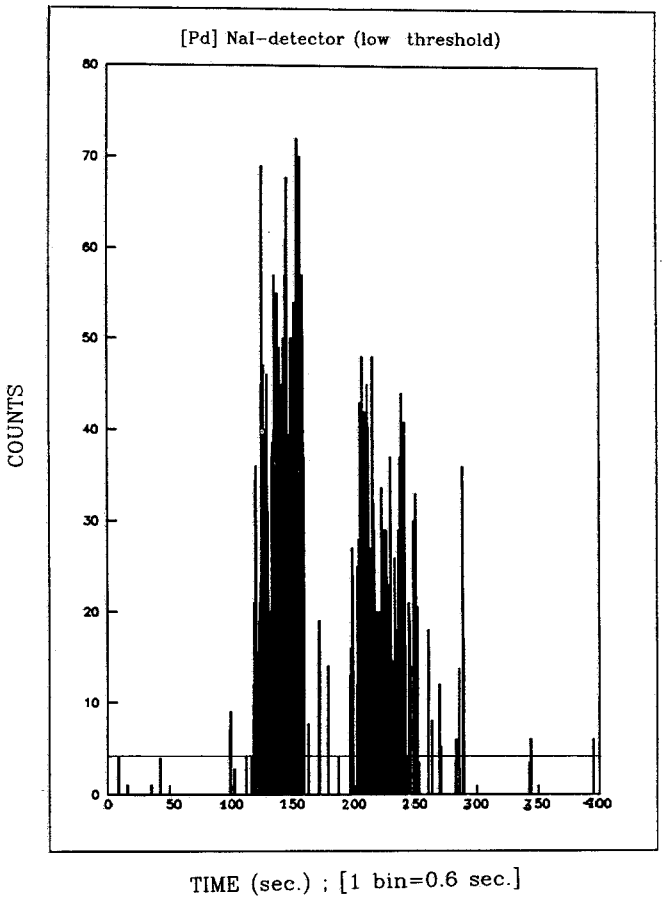
The shot event on Pd occurred about 30 minutes after the *FIRST* charging-up, at a constant current density of 66 mA/cm^2 (maximum allowed voltage Pd $-$ Pt < -3.1volts). The intensity of the shot was really impressive. We got, in an acquisition time of about 0.6 s, the following counts:

NaI(Tl) low thres.	=	61	counts	(typical value in 0.6 s $\leq$ 4)
NaI(Tl) high thres.	=	96	counts	(" " " " $\leq$ 1)
He^3 bare	=	217	counts	(" " " " $\leq$ 1)
He^3 moderated	=	102	counts	(" " " " $\leq$ 1)



[Fig.3] Pd electrode, SHOT event. Time evolution of the signal on NaI(Tl) gamma detector at a) low threshold and b) high threshold. In fig. c) it is shown the result with He^3 bare detector and in fig. d) it is shown the result with He^3 detector surrounded by 12 cm. thick paraffin moderator. The bin of the signal is $\simeq$ 0.6 s. For comparison, it is shown the time evolution of the signal at about 550 s before and after the shot event.

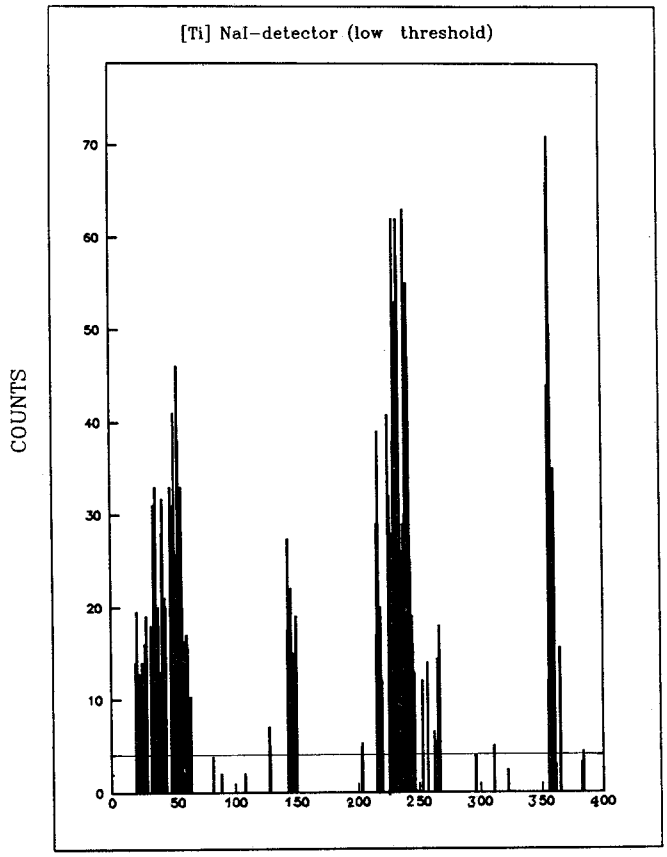
The apparent incongruence on counts between low and high threshold of NaI(Tl) can be understood if we assume that the signal have a very intense burst behaviour:the low threshold retriggerable discriminator-shaper circuitry was "locked" because most of the following pulses arrived between a time shorter than $1\mu s$ (the time we chosed for the digital shaper of gamma signals). About neutron detectors, if we take into account the detector efficiency and unfavourable geometrical factors, we can roughly claim to have produced a number of neutron of the order of 10^4. This large value can be still higher if we take into account the intrinsic dead-time of the conventional shaping amplifier adopted ($\simeq 10\mu s$) in respect to the probable burst structure of the signal.We measured both the voltage-under-charge and the spontaneous voltage just few minutes before and after emission because it was our standard experimental routine during the first charging-up of the electrode: *we noted that both voltages decreased after the emission* (of the order of 1 %).



[Fig.4] Pd electrode, BURST event. For comparison we show the intensity of the signal about 120 s before and after the burst. The total time of the event is about 4 min. The background is shown by an horizontal straight line.

The bursts occurred after 73 minutes from the beginning of the first charging-up, about 1 minute after measuring the spontaneous voltage of the electrolytic cell. Such type of measurement, as pointed before, has two behaviours: a) involves a little "stress" to the system because it is necessary to disconnect the power supply (floating condition) to measure the voltage between the Pd electrode and the pure Ti used as reference one, b) allows a discharging of D from Pd.

As previously described, our "trigger" is any detector (OR condition) that overcomes the stated counts. The "typical" out-of-burst condition is shown with a continuous line in figs. 4 and 5, the amplitude is in counts/bin and one bin correspond to about 0.6 s, i.e. the minimum collecting time allowed from our acquisition system. We show our bursts together with the situation before and after the signal bursts. Fig. 4 shows two main bursts ($\simeq$ 50 s each) with some dead-time in between ($\simeq$ 40 s) and some after-main-burst pulses so that the total duration time of the event is of the order of four minutes.



[Fig.5] Ti electrode, BURST event. For comparison we show the intensity of the signal about 30 s before and after the burst. The total time of the event is about 6 min. The background is shown by an horizontal straight line.

In this particular experiment the Pd electrode was very close to the NaI(Tl) detector ($\simeq$ 2.5 cm) so that we can estimate the overall detection efficiency of the order of some percent. We like to note that after the end of the burst ($\simeq$ 5 minutes) we measured again the spontaneous voltage and we found it slightly lower as before stated. According to us, this fact is related to the coming-out of deuterium from Pd electrode. We didn't note any temperature change (stable at 17 ± 1 Celsius) before and after the event. The thermometer, a J type thermocouple from Keithley, was positioned *external* to the electrolytic cell in open air, well bound to the jar.

The bursts coming from Ti (fig. 5) electrode arose after a long restoring cycle of this electrode subsequent to several severe stresses: -196 $\div$ +200 Celsius thermal cycling, cathodic and anodic current up to 3A, mechanical stress. We get the signal after 3 days of low current infusion at 50mA/cm^2 and we can't say anything about spontaneous voltage or temperature conditions because we were absent during the emission and we hadn't automatic record of them. The distance between electrode and NaI(Tl) was about 10 cm, so that the overall efficiency for gamma signal is less than 1 % in the energy range recorded.

The time evolution of the signal is similar to that appeared for Pd electrode and, in this case, the total time of the event is about 6 minutes.

We never got statistic evidence of excess counts from the charged-particle detectors, i.e. the plastic scintillator, using both Pd or Ti(Al) electrodes.

CONCLUSION AND DISCUSSION

We observed gamma and neutron emissions from electrodes made of Pd and Ti(Al) during non-equilibrium conditions due to electrolisys of D_2O and LiOH: all the emissions detected had a bunched structure.

About the calculation of "fusion rate" λ_f, if we use the *emission data of Pd during the burst (fig.4) and we assume that all the excess gamma counts arise from any kind of fusion*, adopting a mass of Pd 30 g, excess gamma counts 4100 in 205 s, gamma detection efficiency 1 %, D/Pd concentration in beta phase about 0.6, we should obtain

$$\lambda_f = 4.3 \cdot 10^{-20} \text{ fusion/deuteron pair/s}$$

This value is more than 1 order of magnitude larger of results of P.Perfetti et al. [9] and about 3 order of magnitude lower than the results of M.Fleischmann et al. [1]. We remark that it is not clear, at this time of the research, if our assumption of excess gamma counts is equivalent, from the point of view of λ_f calculation, to the excess neutron counts reported from P.Perfetti or M.Fleischmann.

We note that the results by M.Fleischmann arise from *continuous* emission, not *burst* as in our or P.Perfetti calculation.

Moreover, if we consider the evaluation of λ_f during the shot, using the gamma count value at low threshold (still neglecting the saturation effect), we reach a fusion rate even larger then the value reported by M.Fleischmann [1]. Furthermore, the neutron counts during the shot, corrected by the proper detection efficiency, is roughly the same value reported by M.Fleischmann. As before stated, the count of excess neutrons during the burst has not sufficient statistical significance because of our low sensitivity and non optimised energy threshold.

The calculation of λ_f on Ti(Al) electrode is quite complicated when compared with Pd one, even ignoring completely the effects due the heavy Al alloying (4.5 %) and is less accurate because we adopted for the deuterium the hydrogen diffusion coefficient. At first, we remind that the diffusion velocity of the deuterium in Ti is very low, so in our experimental conditions, supposing a charging time as long as 48 hours, the penetration depth of deuterium in Ti is only 0.32 mm [10]. The maximum measured concentration of deuterium in Ti at 23 Celsius is 1.73, determining an oversimplified mean value of 0.85 in our calculation (this condition is called the $\alpha + \gamma$ phase) [10]. As a consequence, the effective active volume of the

electrode is only 5 mm^3; taking the electrode density about 4.5 g/cm^3, the atomic weight 48, gamma detection efficiency about 0.5 % , excess count of 2800 counts in 360 s, we should obtain:

$$\lambda_f = 1.3 \cdot 10^{-17} \text{ fusion/deuteron pair/s}$$

This value is about three order of magnitude larger than the largest value estimated by S.Jones [2] taking into account burst effect, but, again, we remark that, in this calculation, we consider a system composed by hydrogen and pure Ti instead of deuterium and Ti(Al).

The nuclear origin of the reported signals is however still an open question, since the gamma spectra collected by the MCA do not show peaks at the foreseen energies. Pile-up of signals and dead-time of the MCA during the very intense emissions could be an argument.

During the experiment we had some others, less intense, events with a behaviour very similar to these quoted.

In any case, we are strongly certain that almost all the possibilities of fake signals originated by electrical or mechanical perturbation and instability of the detectors and associated electronics, as well as any abrupt changing of environmental nuclear activity (thanks to the underground laboratory) were deeply investigated and eliminated if any. Obviously, there is a remote possibility that some source in the electrodes (ultrasounds, fractures, phonons, etc.) could originate some kinds of the signals reported.

We like to note about the hypothesis of muon-induced fusions that , due to the extremely low value of muon flux inside the Gran Sasso Laboratory, it is very difficult to think it was the main cause of signals in our experiment.

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