Laboratori Nazionali di Frascati

F. Celani, A. Spallone, S. Pace, B. Polichetti, A. Saggese, L. Liberatori,
V. Di Stefano, P. Marini:

FURTHER MEASUREMENTS ON ELECTROLYTIC COLD FUSION WITH D₂O AND PALLADIUM AT GRAN SASSO LABORATORY

FURTHER MEASUREMENTS ON ELECTROLYTIC COLD FUSION WITH D2O AND PALLADIUM AT GRAN SASSO LABORATORY
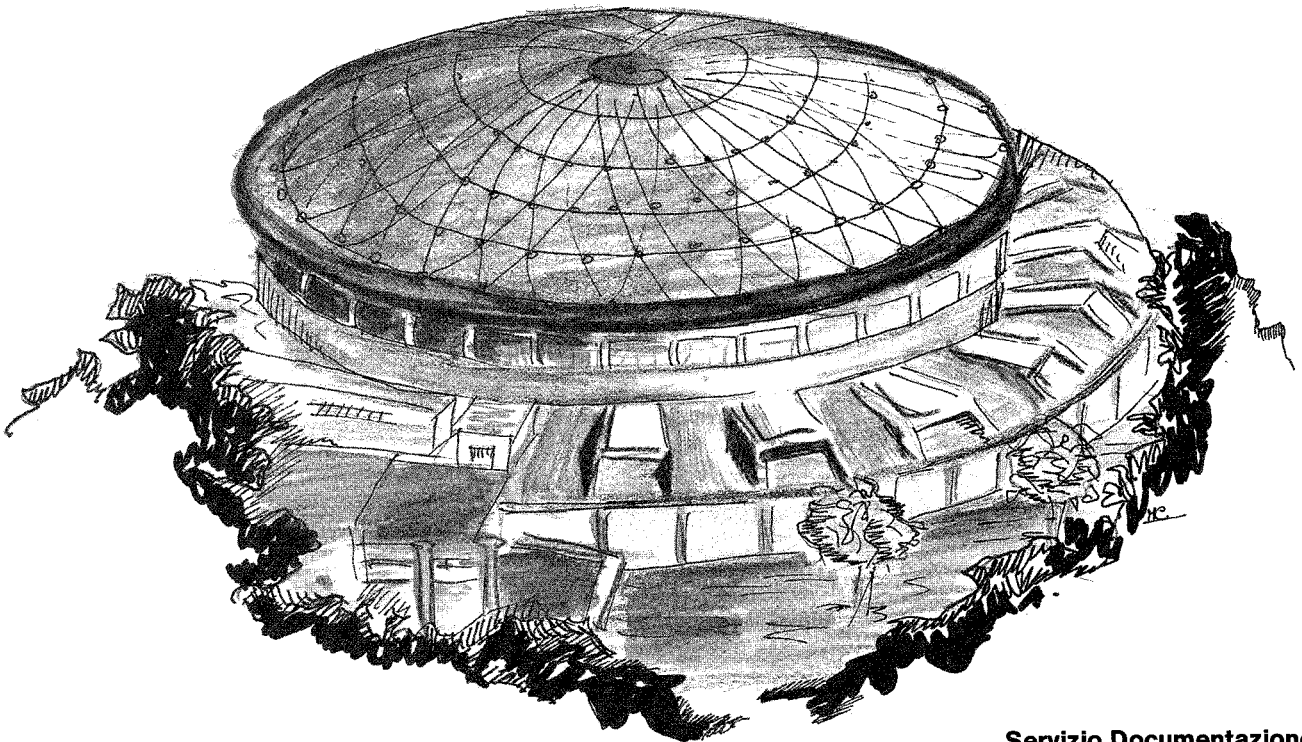
F. Celani, A. Spallone
INFN Laboratori Nazionali di Frascati, P.O. Box 13, 00044 Frascati (Italy)

S. Pace, B. Polichetti
Univ. of Salerno, Fac. of Science, Dept. of Physics , 84100 Salerno (Italy)

A. Saggese, L. Liberatori
Cons. Inter. Naz. Fis. della Materia, Univ. of Salerno, Dept. of Physics, 84100 Salerno (Italy)

V. Di Stefano, P. Marini
ILVA-Centro Sviluppo Materiali, Via di Castel Romano 100, 00129 Roma (Italy)

Several experiments were performed, at the Gran Sasso Laboratory, on a 0.8-cm-diam x 5-cm-long, hyperpure, high-temperature vacuum-annealed, palladium rod used as cathode for electrolytic infusion of D_2O and 0.1 M LiOH with regular additions of gaseous CO_2 at current density of 60 mA/cm^2. In the very low background radiation environment, several gamma bursts lasting up to 15 min were detected whose intensity, in terms of cold fusion, was > 10^{-20} fusion/(deuteron pair·s). Under normal background conditions, none of these burst signals would have been detected with statistical significance. The shape and intensity of these signals are quite similar to those detected previously.

INTRODUCTION

Following our previous experiments[1] on cold nuclear fusion at the underground Gran Sasso Laboratory,we improved the detection system by decreasing the gamma background by a factor of > 2.5 and by adding another NaI(Tl) gamma detector. We measured, for as long as 40 days, radiation from an electrolytic cell based on a 0.8-cm-diam x 5-cm-long, 99.98% pure palladium rod (cathode) with a platinum anode in an infusion of 50 cm^3 of D_2O with 0.1 M LiOH and regular additions of small quantities of gaseous CO_2 at a constant current density of ~ 60 mA/cm^2.

Due to both an incomplete experimental hall and mis-operation of the digital data acquisition system, we achieved an acquisition time efficiency of ~ 50%. Moreover, several power blackouts left the cell in either an open or a short circuit condition. The short circuit is particularly deleterious because of the possibility of oxigen diffusion inside the palladium electrode having a poisoning effect on hydrogen or deuterium absorption. This condition once lasted as long as 2 days. We recall, however, that this problem is less damaging when the loading value of deuterium in the palladium is increased before the short circuit.

Despite these drawbacks, we observed a total of five burst-type events in addition to the two reported previously, in the experiment started at May 2 1989 and lasting 40 days.

THE NEW EXPERIMENTAL SETUP

Figure 1 shows the geometry of our shielding and detection apparatus. Located in the center, enclosed by more efficient low-activity lead shielding, is the electrolytic cell (Fig. 2), surrounded by 4 cm of light water with 2 cm of heavy water inside the cell to moderate the expected 2.5-MeV neutrons from the electrode. This improved shielding reduced the gamma background measured by the large 5- x 5-in. NaI(Tl) detector from ~ 380 to ~ 140 count/min at an energy > 0.8 MeV. An additional small 3- x 3-in. NaI(Tl) gamma detector was placed ~ 10 cm away from the palladium electrode and set at a 1.2-MeV discrimination energy threshold.

In contrast to the previous experimental setup, where one of the two ^{3}He tubes was fully surrounded by a paraffin moderator, both ^{3}He tubes were bare and very close to the cell in this setup. The bare ^{3}He has a thermal neutron sensitivity, as stated by the manufacturer, of 433 counts n^{-1} cm^2 (Ref. 2).
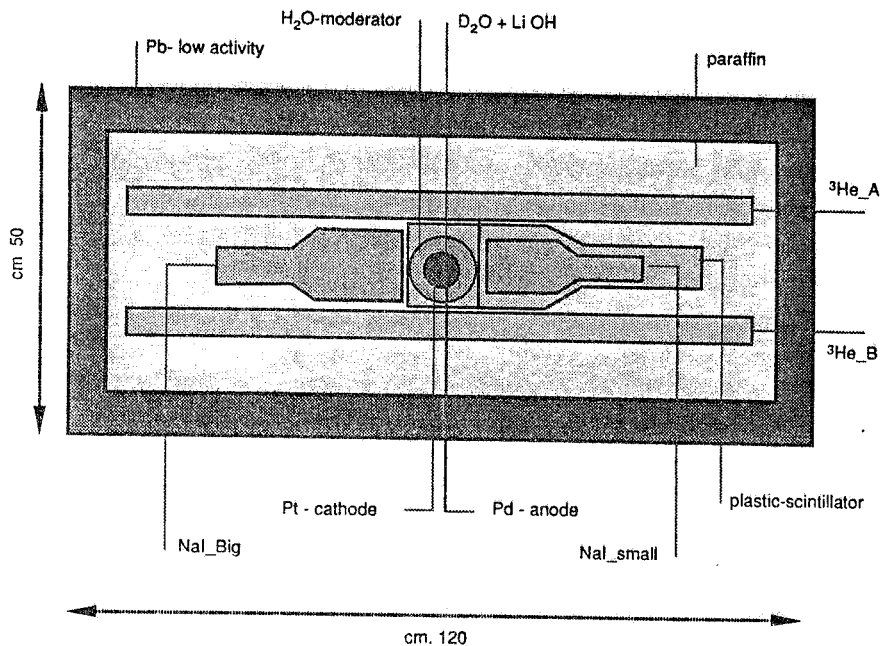

FIG. 1 - Electrolytic cell and detection apparatus setup (top view). The external shielding is made of 5-cm-thick low-activity lead bricks that completely surround the detectors. Paraffin bricks are enclosed between the neutron detectors and the external lead wall. The electrolytic cell, bunged with a rubber plug, is in a 4-cm-deep water bath.
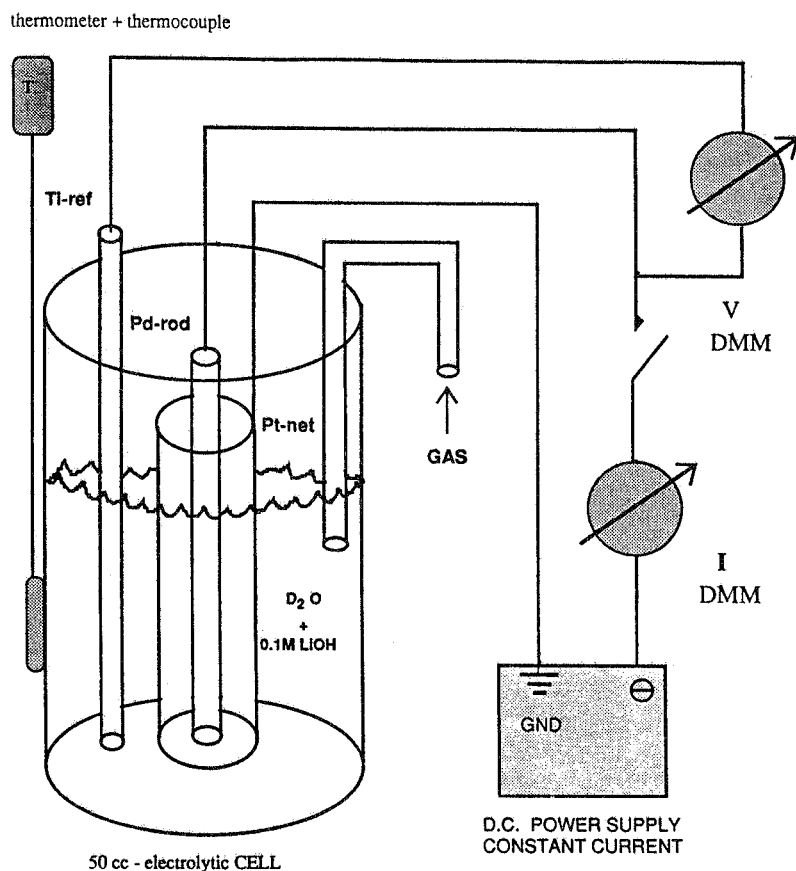
thermometer + thermocouple

FIG. 2 - Experimental setup of electrolytic cell and power supply section (not to scale). All the connections between the electrolytic cell and the power supply or DMM are by coaxial cables to reduce the possibility of picking up external noise or radio-frequency emission from the electrolytic cell itself during the charging procedure. During the nuclear measurements, all Digital Multimeters and the thermometer were switched off to avoid spurious signals due to the sampling.

Between the detectors and the 5-cm lead bricks of the external wall, there are ~ 10-cm-thick paraffin bricks to improve the neutron detection efficiency using the mirror effect of the lead and the moderating effects of the paraffin. The 10 to 20 cm of paraffin previously surrounding the ^{3}He detector was used as a rough estimate of the thickness needed to moderate the expected 2.45- or 14-MeV fast neutrons (from $d + d$ and $d + t$ fusion reactions, respectively) from the cell and increase the total detection efficiency of the counters. Taking into account the geometric factors and the paraffin neutron absorption, we estimate the total detection efficiency of the ^{3}He tubes to be on the order of 10^{-2}. The neutron detectors, however, could not be calibrated with a standard americium-beryllium neutron source because it is forbidden to use neutron sources inside the underground laboratory.

RESULTS

Almost all of the burst events were detected by the large NaI(Tl) gamma detector set at an energy threshold > 800 keV, as described in Ref. 1. Table I lists all the events observed during the test. Between events 2 and 3a, we improved the experimental setup as described above, which decreased the gamma background from ~ 380 to ~ 140 count/min.

The results of the experiment are also reported in Fig. 3. There were many misoperations during the test, and these are indicated in the figure. We assume that each NaI(Tl) count exceeding background is a nuclear fusion event.

We previously reported a significantly long burst event (event 2). In the present experiment, a similar event, event longer and more intense, was detected, as shown in Figs. 4 and 5. All detector counts are reported, but only the NaI(Tl) detectors had excess counts of statistical significance. To avoid storing too much data, we decreased the acquisition rate by setting a software count threshold much (about five times) larger than the average background counts. For the average background counts, we used the mean background count per minute normalized to one bin (0.6 s).

TABLE I - Gamma Detection by NaI(Tl) Detector*

Event and Type	Duration Time[a] (s)	Background Counts per Bin[b]	Total Excess Counts[c,d]	Excess Counts per Bin	Ratio of Signal to Background[e]	Fusion Rate[f] [10^{-20} fusion/ (deuterium pair · s)]
2s (short) (May 2, 1989, 22:40)	0.6	3.86	57	57	15.8	20.4
2 (burst) (May 2, 1989, 23:30)	157	3.82	4092	15.6	5.1	5.6
3a (burst) (May 28, 1989, 0:28)	56	1.33	420	4.5	4.4	3.2
3b (burst) (May 28, 1989, 4:45)	48	1.35	885	11.0	9.2	7.9
4b (burst) (May 28, 1989, 11:57)	337	1.48	7250	12.9	9.7	9.2
5a (burst) (May 28, 1989, 13:23)	28	1.49	540	11.6	8.8	8.3
5b (burst) (May 30, 1989, 11:53)	12	1.47	161	8.0	6.4	5.8

*Set at a threshold > 800 keV.

[a]Duration time indicates the effective duration of the burst (excluding the time between bursts), not the mean time as in Ref. 1. Thus, for event 2, the duration time is only 157 s instead of 205 s as previously quoted and as a consequence, the fusion rate increases from 4.3 x 10^{-20} to 5.6 x 10^{-20} fusion/(deuteron pair · s).

[b]Data acquisition minimum time (bin) corresponds to 0.6 s.

[c]Excess counts exclude the background counts.

[d]The total excess counts are calculated on the effective burst time.

[e]The signal to background ratio indicates the mean intensity of the event.

[f]This assumes that each excess gamma count corresponds to a nuclear fusion.
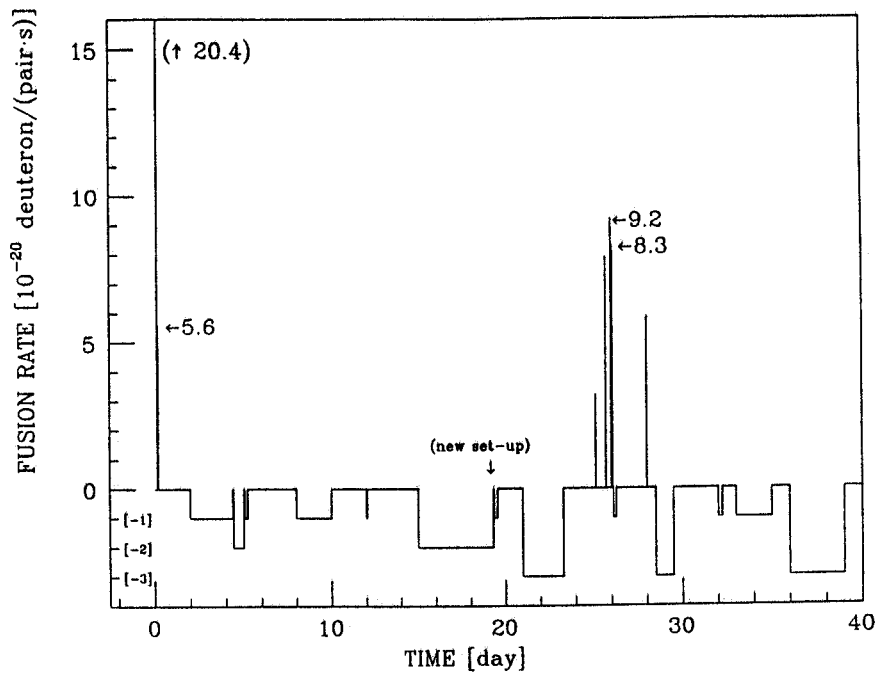
FIG. 3 - Palladium electrode life from May 2 to June 12, 1989. The effective total measurement time was > 20 days. Seven events were detected with a fusion rate > 3 x 10^{-20} fusion/(deuteron pair·s). The arrow indicates when the new experimental setup was initiated.
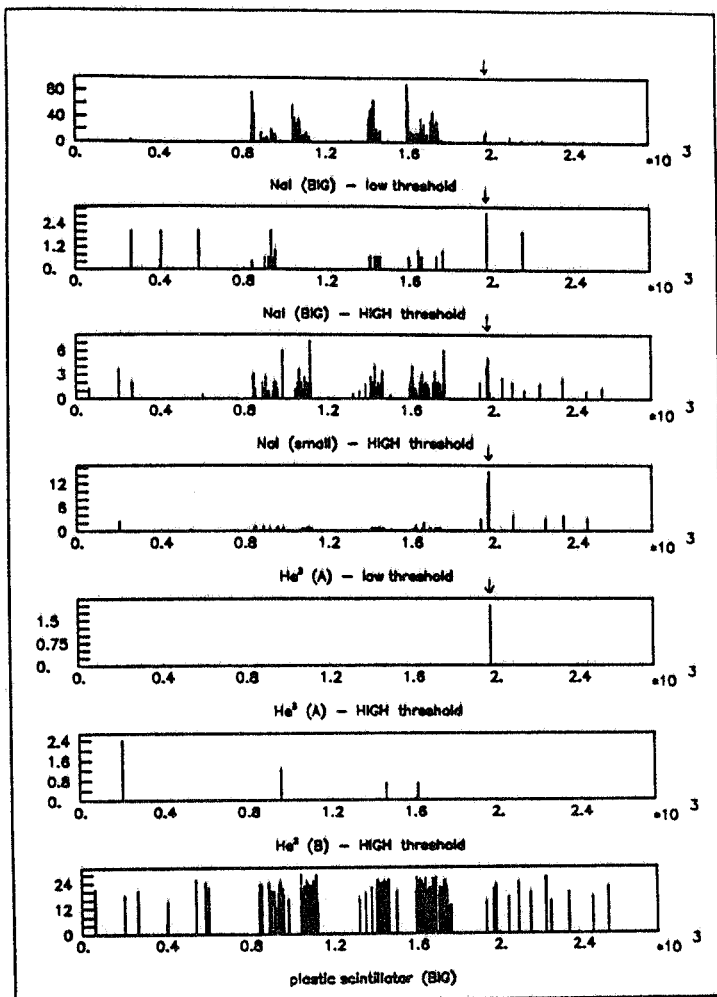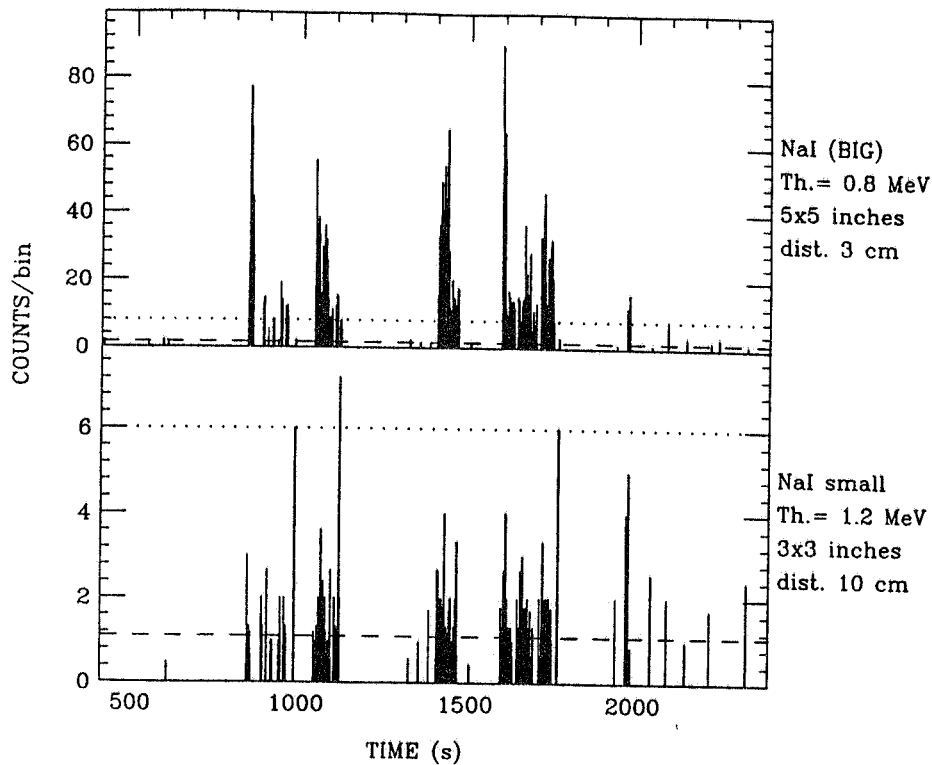

FIG. 4 - Event 4: total counts in all detectors during the longest and most intense event detected after the new setup. Only the counts from the NaI(Tl) detectors are of statistical significance, although all the detectors recorded the event, except for the plastic scintillator used to detect charged particles.

FIG. 5 - Details of event 4: counts from the two NaI(Tl) detectors. The dotted line indicates the software thresholds (8 and 6 count/bin for the large and small detectors, respectively) and the dashed line indicates the average background (~ 1.5 and 1.1 count/bin, respectively).

Note that the intrinsic background counts and photomultiplier noise are quite different for the two NaI(Tl) detectors used. The large (5- x 5-in) detector was specially selected for its low noise. According to its technical characteristics, the mean value of "noise " counts is ~ 410 count/min in the 100 to 3200 keV range, and ~ 65 count/min from 1300 to 3200 keV. To explain the different excess counts detected by the two detectors, we must consider the different active detection areas and the energy thresholds.

In Fig. 4, note that there is a shot event (indicated by an arrow) similar to that reported in Ref. 1, at the end of the long burst event. In Ref. 1, the intense shot event (event 2s) occurred at the beginning of the charging up of the electrode, ~ 50 min before the long burst event (event 2). This shot event was detected by all the detectors except the plastic scintillator and one of the two ^{3}He tubes (set to a high detection energy threshold).

The plastic scintillator counter could not detect (a) 3.02- MeV protons ($d + d \rightarrow + t + p$) because protons at this energy are absorbed by at least 2 cm of heavy water, 4 cm of light water, and ~ 1mm of glass surrounding the electrode and (b) eventual electrons due to gamma conversion or Compton scattering. Except for very large charged-particle fluxes (of the order of 10^6 emission/s at the source), this detector could not detect counts with statistical significance because of its large intrinsic noise.

In Fig. 6 are shown all the burst events occurring after reconfiguring the experimental setup. They seem, at first glance, to be very different in shape and intensity. Observing the last column of Table I, however, we obtain similar values by a factor of < 3. We hypothesize a constant source intensity, but different emissions in time duration and shape.
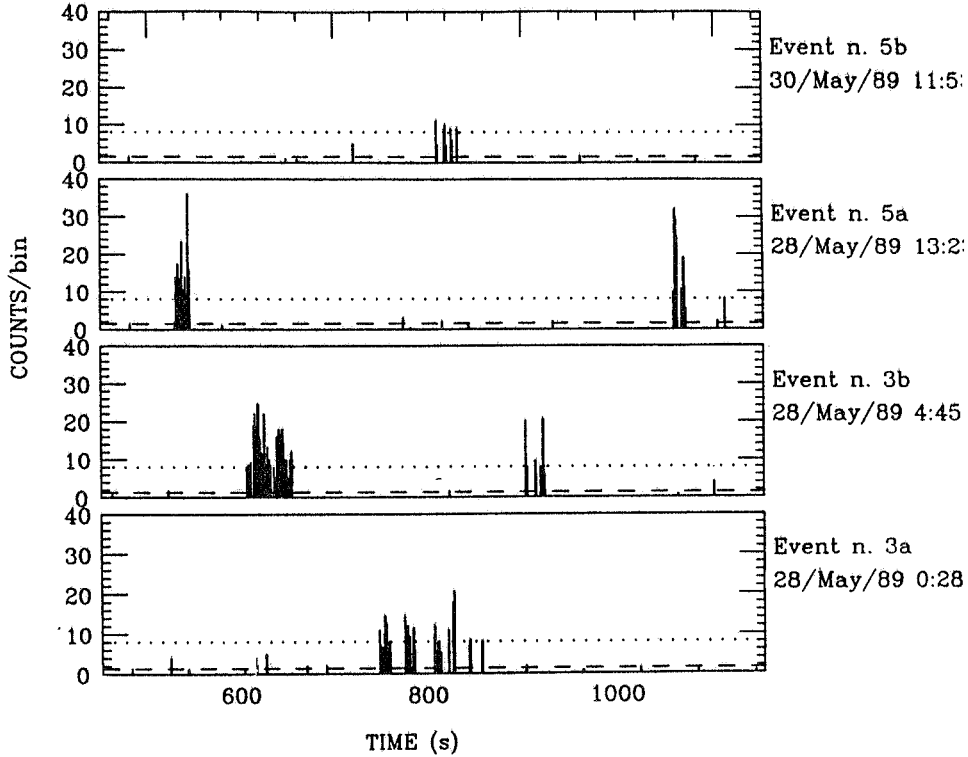

FIG. 6 -The four less intense events detected by the large NaI(Tl) detector. The dotted line indicates the software threshold of 8 count/bin and the dashed line indicates the average background normalized to a bin data acquisition time (0.6 s).

CONCLUSIONS

From the signal/noise ratios given in Table I, it can easily be seen that none of the reported signals would have been detected in an usual experimental area due to a large gamma background (~ 50 times more than our experimental setup), except by using a hypothetical Marinelli-like beaker shielding (low-activity lead, oxygen-free copper, and cadmium).

Using the calculation made in Ref.1, except with a reduced gamma detection efficiency (0.5% instead of 1% because the water bath decreases the ~ 2.2 MeV gamma radiation intensity of 30% and increases the distance), we should get for our most intense signal a fusion rate as large as

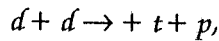$$\lambda_f^{max} = 9.2 \times 10^{-20} \text{ fusion/(deuteron pair·s)}.$$

Our less intense signal (event 3a) gives a noticeable fusion rate as large as

$$\lambda_f^{max} = 3.2 \times 10^{-20} \text{ fusion/(deuteron pair·s)}.$$

Our assumption of one gamma ray detected being equal to one fusion is quite conservative, because if we consider one of the most promising radiation channels[3],

$$d + d \rightarrow + t + p,$$

the gamma yield produced by 3-MeV protons interacting with palladium nuclei is of the order of 10^{-7}.

According to the previous reaction events should detected at 0.3738, 0.4339, 0.5119 and 0.558 MeV, yielding 2.5 x 10^{-8}, 4.5 x 10^{-8}, 2.3 x 10^{-8} and 0.52 x 10^{-8} gamma rays per 3.0 MeV absorbed in palladium, respectively. We recall that the energy resolution (~7%) of NaI(Tl) at few hundred KeV is too poor to allow us to identify those lines. Despite both poor resolution and long integration time (compared to short burst emission time) we detected, with the MCA, like in Ref. 1, a noticeable global increasing of counts at γ energy between ~ 100 KeV and ~ 500 KeV. Unfortunately, we have no gamma spectra to show, of these events at 25th day from starting-up, because the data stored in the tapes were lost.

Except for the anomalous shot event, we did not detect any large neutron counts during the bursts. If we assume nuclear fusion and a branching ratio of 50% between the two fusion production channels ($t + p$ and $^3H + n$), given the above estimated fusion rate, we would expect a large neutron emission to be detected by the neutron counters. Therefore, if we assume $d + d$ fusion, we have to assume a neutronless process.

Fractoemission phenomena can result in detection of very intense emission of charged and neutral particles, as widely reported in the literature[4]. The intense low-energy (1- to 100-keV) gamma can easily simulate signals of enough energy to overlap the 800-keV threshold of our large NaI(Tl) detector.

Thus, it is not yet clear whether all or a fraction of the detected signals arise from nuclear fusion reactions. We plan a new experimental setup with a gamma detector and associated electronics for pulse-height analysis that will be able to follow signals of very intense bursts, but with a very low repetition rate.

ACKNOWLEDGMENTS

We are indebted to S.E. Jones of Brigham Young University for his continuous suggestions and discussions during and after the experiment. We wish to thank Gianfranco Bologna of Turin University for his warm, encouraging critical review of the manuscript and his very useful suggestions.

We also want to express gratitude to all the people at Gran Sasso Laboratory for their warmth and hospitality.

REFERENCES

1) F. Celani et al., "Results of the 1th generation experiments, at Gran Sasso underground laboratory, on nuclear cold fusion". Conference Proceedings Vol. 24 pag. 257 "Understanding Cold Fusion Phenomena" R.A. Ricci, E. Sindoni, F. De Marco (Eds) SIF,   Bologna 1989.

2) A. Rindi et al., "Underground Neutron Flux Measurement", Nucl. Instrum. Methods, **A272**, 871 (1988).

3) D.C. Bayley, "Gamma from Cold Nuclear Fusion", University of Toronto (19xx), submitted to Phys. Rev. C.

4) T. Takeda and T. Takizuka, "Fractofusion Mechanism", J. Phys. Soc. Japan, **58**, 3073 (1989).