NUCLEAR FUSION EXPERIMENT IN PALLADIUM CHARGED BY DEUTERIUM GAS

COLD FUSION

TECHNICAL NOTE

KEYWORDS: *fusion, reaction, palladium, deuterium, protons*

SEBASTIANO AIELLO, ENRICO De FILIPPO,
GAETANO LANZANÓ, SALVATORE LO NIGRO,
and ANGELO PAGANO *Istituto Nazionale di Fisica Nucleare*
Sezione di Catania, Dipartimento di Fisica dell'Università di Catania
Corso Italia, 57 - I 95129 Catania, Italy

Received February 26, 1990
Accepted for Publication April 10, 1990

A palladium-deuterium system subject to different experimental thermodynamic conditions was studied to look for low-temperature d-d fusion reactions. Neutrons, light charged particles, and energetic gamma rays were detected. No significant effects were observed in the neutron and gamma-ray measurements. From the analysis of the light charged particles, a fusion rate $\lambda_f \approx 10^{-23}$ *event/*$(D_2 \times s)$ *was deduced.*

INTRODUCTION

It has been suggested recently[1,2] that deuterium in hydride-forming metals (i.e., palladium and titanium) can undergo nuclear reaction at room temperature. Such a fascinating supposition has stimulated a variety of experimental and theoretical investigations in the scientific community. But, so far, the experimental information is incomplete, and a clear picture of the phenomenon does not yet exist. Furthermore, if we consider the matter from the experimental point of view, the actual controversy seems to be related to arguments concerning such fundamental questions as the reproducibility of the observations and the adopted experimental procedure.[3]

In our case, the metal-deuterium system was obtained by a gas absorption method similar to the one employed by De Ninno et al.[4] However, we investigated different dynamic conditions by changing temperature and pressure of a palladium-deuterium (Pd-D) system. In fact, an enhancement in the *d-d* fusion process rate is expected in hydrogenated metals during gas absorption in nonequilibrium states, as suggested by Cassandro et al.[5]

Assuming that *d-d* nuclear fusion could occur during phase transitions, we focused on studying the nuclear products of the following reactions:

$$d + d \rightarrow {}^3\mathrm{He} + n \ , \qquad (1)$$

$$d + d \rightarrow {}^3\mathrm{H} + p \ , \qquad (2)$$

and

$$d + d \rightarrow {}^4\mathrm{He} + \gamma \ . \qquad (3)$$

Fusion reactions (1) and (2) have remarkably the same rate, as measured[6] in the energy range from 3 to 160 keV and deduced by semiempirical estimations at lower energies.[7] On the contrary, the rate of reaction (3) can be assumed to be four to five orders of magnitude slower, as suggested in Ref. 8.

EXPERIMENTAL APPARATUS AND PROCEDURE

Pd-D System

A schematic of the experimental apparatus is shown in Fig. 1. A thick sample of palladium (99.8%) in a rectangular sheet (1×1 cm^2 of surface area and 1 mm thick) was placed inside a stainless steel cell ~15 cm in diameter.

The cell was connected to a deuterium gas cylinder through a valve and a pressure regulator. The mechanical arrangement was tested for vacuum and high pressure at both room temperature and liquid nitrogen temperature.

The palladium sample was attached to a proper copper support to ensure good thermal contact during a temperature change in the system. The copper support was surrounded by a special dewar during the cooling phase up to the liquid nitrogen temperature and by an electric heater during the heating phase up to ~300°C. An auxiliary line allowed us to evacuate the cell up to 10^{-1} Torr. Both the pressure inside the cell and the temperature of the copper support were continuously monitored.

The Pd-D system was prepared in the following way:

1. The palladium sample was kept at 77 K under D_2 pressure of 10 bars for ≈15 h.

2. Successively, the system was subject to dynamic transitions for ≈22 h, as shown in Fig. 2. During this period the Pd-D system was expected to change the stoichiometry from Pd-$D_{0.3}$ to Pd-$D_{0.7}$ and then to pure palladium. The compositions were tested on various palladium targets prepared in the same way and measured by Nuclear Reaction Analysis.[9]

3. Finally, the system was left at room temperature under vacuum for ≈9 h.

Detectors

The neutron counting rate was monitored by a liquid NE-213 scintillator (2×2 in.) placed outside the cell. The scintillator was optically coupled with a photomultiplier. To identify neutron counts, the shape of the anodic signal was

Aiello + (90)

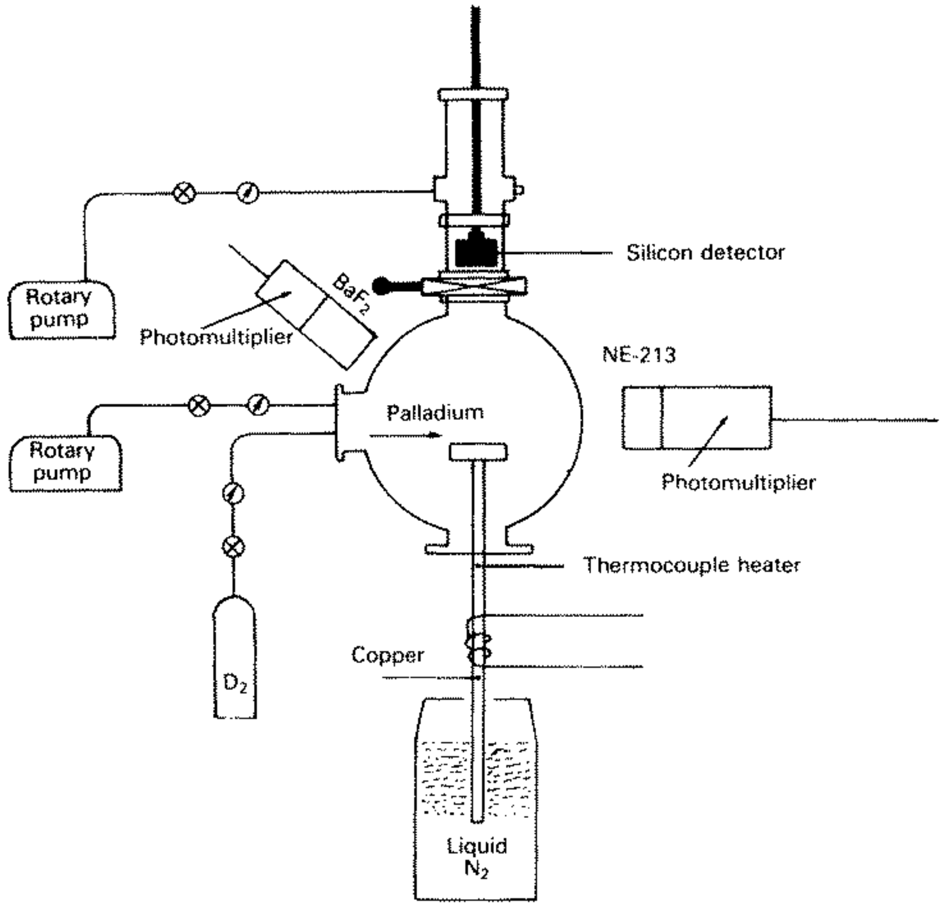

Fig. 1. Schematic of the experimental apparatus.

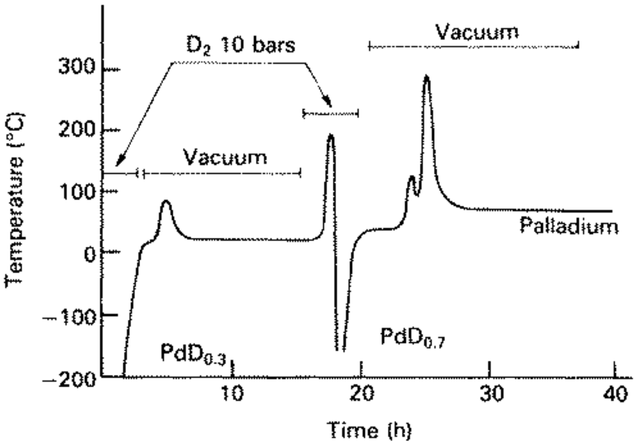

Fig. 2. Temperature of the Pd-D system versus elapsed time. The stoichiometry of the Pd-D sample and the pressure of deuterium gas are also indicated.

properly analyzed. A typical bidimensional matrix of the neutron-gamma discrimination established by a standard ^{252}Cf source is shown in Fig. 3. The bidimensional matrix was obtained at room temperature at both the beginning and end of the experiment. In addition, the matrix was verified at both low and high temperatures of the cell.

The overall detection efficiency of the neutron counter (including the geometric efficiency) was estimated to be $\approx 3 \times 10^{-4}$.

The gamma rays with energy higher than 10 MeV were monitored by a 5-cm-diam × 10-cm-long BaF_2 scintillator crystal placed quite close and external to the cell. For this detector, we estimated an overall detection efficiency (including the geometric efficiency) of $\approx 9 \times 10^{-4}$ in the energy gamma-ray range between 10 and 30 MeV. Due to this efficiency and to the very low contribution to the fusion rate from reaction (3), only a possible anomalous enhancement of high-energy photons coming from the reaction (3) could be observed by the BaF_2 detector.

Finally, the charged particles were measured by a large 450-mm^2 × 100-μm-deep silicon surface barrier (SSB) detector. To prevent damages to the SSB detector during the change of pressure and/or temperature in the Pd-D sample, the detector was located in a separate vacuum chamber connected with the cell by a valve. The detector was placed in front of the palladium sheet, at a distance of ≈ 20 mm, only when the pressure in the cell was $< 10^{-1}$ Torr. We estimated a value of 2.3×10^{-2} for the detector efficiency, taking into account the effects of the actual geometry and assuming an intrinsic light charged particle efficiency equal to unity for energies above the electronic threshold.

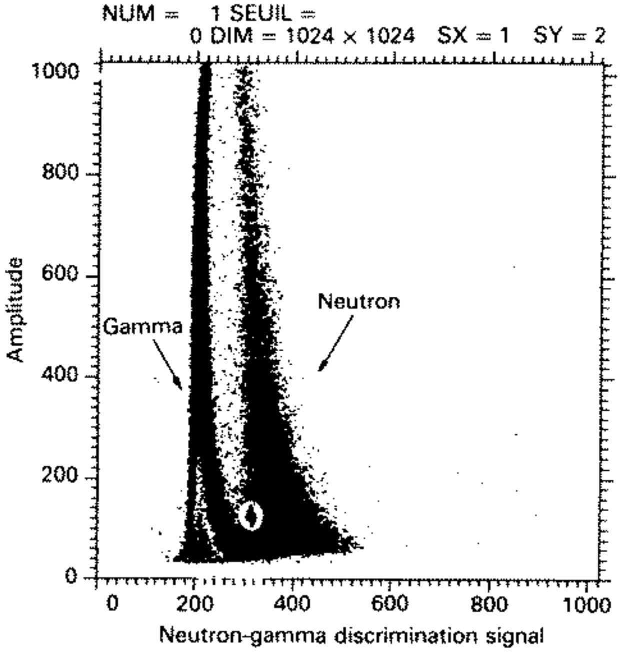

Fig. 3. The obtained neutron-gamma discrimination using a standard ^{252}Cf source. The bidimensional array shows the anodic pulse versus the neutron-gamma discrimination signal.

To have a rough identification of the detected particles, the SSB detector was covered by an aluminum absorber sufficiently thick (≈35 μm) to stop protons of ~1 MeV of kinetic energy.

During the experiment, both the external temperature and the electronic threshold of the detectors were continuously monitored.

A connection of the detection apparatus with a personal computer acquisition system allowed us to store the experimental data in an event-by-event mode.

RESULTS AND DISCUSSION

The response of the BaF_2 scintillator for a total time of 7 days is reported in Fig. 4. The gamma-ray counts were obtained by integrating the spectrum from 10 to 30 MeV at 20-min intervals.

No change in the counting rate was observed during the fusion experiment indicated as (a) in Fig. 4. Within the experimental sensitivity of the used detector, any anomalous contribution of reaction (3) was not evidenced.

The SSB detector spectrum (energy calibrated) was obtained by summing the counts of four runs for a total exposure time of 16 h. The spectra of the different runs showed similar shapes within the statistical errors. In particular, we noted the presence of an exponentially decreasing tail in the energy region between 330 keV (the electronic SSB threshold) and 650 keV. This component of the spectrum was also present during various background runs performed by putting the SSB detector in an ≈10^{-1}-Torr atmosphere of hydrogen at room temperature. Figure 5 shows the SSB spectrum where the low-energy noise contribution was subtracted. In the spectrum, it can be observed that a broad component peaked at ~1.8 MeV with 106 total counts. Taking into account both the electronic threshold and the energy loss in the aluminum absorber covering the SSB detector, that bump is consistent with the detection of 3-MeV protons coming from reaction (2), being excluded by the detection of tritons (1.01 MeV).

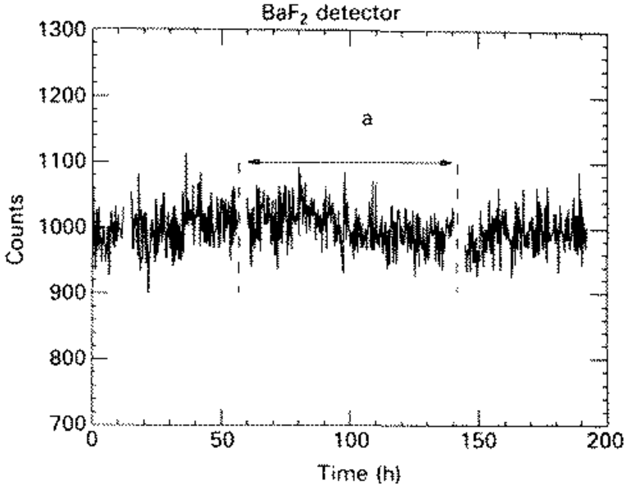

Fig. 4. Time evolution of the counting rates of the BaF_2 scintillator. Region a indicates the results obtained during the fusion experiment.

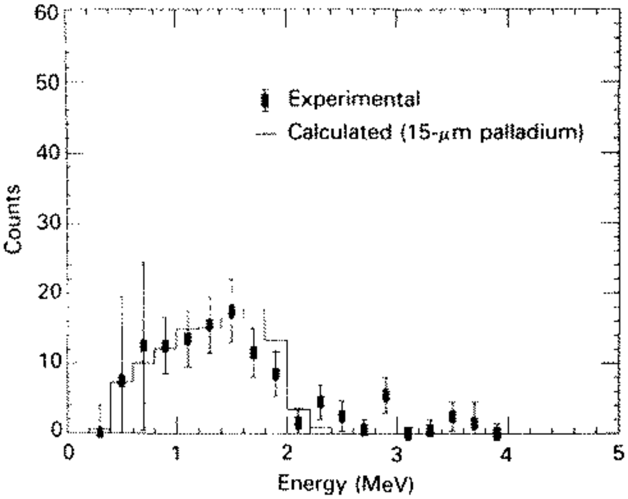

Fig. 5. Energy spectrum of the SSB detector. The background contribution to the spectrum was subtracted. The silicon spectrum time was 16 h.

To test this hypothesis, a simple Monte Carlo calculation was performed. We assumed that 3-MeV protons of energy were emitted isotropically from an internal point of the palladium sample and that, after the particles crossed both the palladium layer and the aluminum absorber (where energy loss was properly calculated by using tabulated energy loss in the materials), they were detected into the SSB. In the calculation, the deuterium was assumed uniformly distributed in a fixed layer of the palladium lattice. In Fig. 5, the calculated spectrum obtained for a 15-μm layer of palladium and 35 μm of aluminum absorber is shown. We note that the experimental and calculated spectra are in good agreement. However, in the frame of these calculations, we observed that palladium layers $>15\ \mu m$ were also compatible with the data. In fact, the increase of the palladium layer strongly modifies only the spectrum shape with an energy less than ≈ 400 keV, near the SSB electronic threshold. So we can estimate only the lower limit of the effective palladium layer.

By assuming a deuterium-to-palladium ratio equal to unity, we could estimate the fusion rate from reaction (2) equal to $\approx 10^{-23}$ event/(D_2 × s), which agrees with the value deduced by Ref. 10.

The results on the neutron emission are reported in Figs. 6 and 7. The counting rate of neutrons, as deduced from the neutron-gamma discrimination matrix during the background measurements and the fusion experiment, is shown as a function of time in Figs. 6 and 7, respectively. Each value represents the counts integrated over a 30-min period.

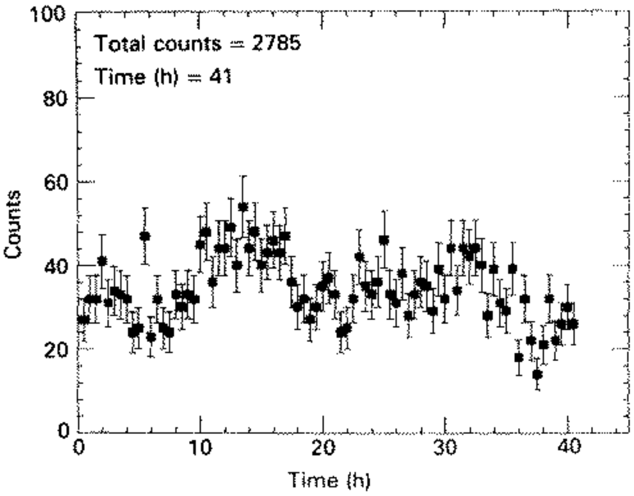

Fig. 6. Neutron counts obtained during the background noise measurements.

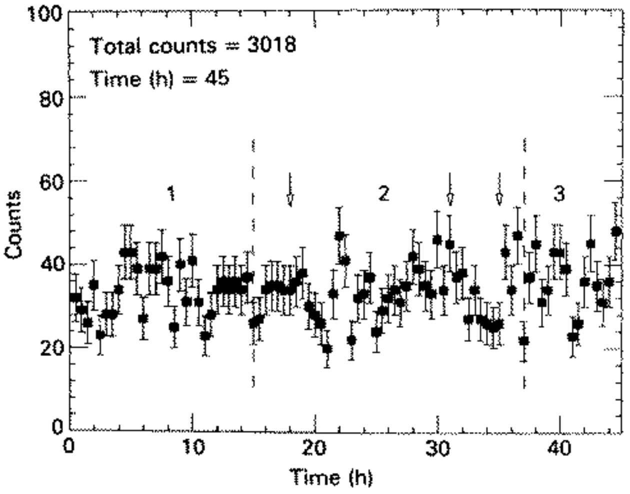

Fig. 7. Neutron counts obtained during the palladium experiment. The three different regions are described in the text. The arrows correspond to the three phase transitions of the Pd-D system.

Background measurements (Fig. 6) were performed for 41 h and 2785 total neutron counts were collected. The mean value and the variance results were $\langle m \rangle = 34.4 \pm 0.2$ and $\sigma = 8.0$ (count/30 min), respectively. This variance could be imputed in part to the method used in order to extract the neutron counts from the neutron-gamma discrimination matrix and in part to the background day-by-day changes.

The fusion experiment was performed for 45 h where 3018 total neutron counts were collected (Fig. 7). The mean value and the variance were $\langle m \rangle = 33.9 \pm 0.1$ and $\sigma = 6.6$ (count/30 min), respectively.

The three phases of the cold fusion experiment, as described in the text and shown in Fig. 7, are indicated by 1, 2, and 3. In region 2, the arrows show the time interval where phase transitions of the Pd-D system are expected.

The neutron counts obtained during the evolution of the palladium system do not show any significant differences from the background measurements. However, if we suppose that in the conditions of our experiment fusion reactions (1) and (2) have the same rate λ_f, one can estimate the number of neutrons expected. By knowing the detector efficiency and by assuming an emission from the whole of the palladium sample, we calculated that the excess of the number of neutrons coming from the fusion *d-d* reaction over the one from the background could be equal to ≈ 12 count/40 h. This counting rate obviously remains hidden in our background signal. In this hypothesis, our neutron measurements do not contradict the result obtained for the proton data.

In conclusion, the obtained fusion rate per deuterium pair λ_f is in good agreement with the upper limit values reported in Refs. 10 and 11, but it is four orders of magnitude smaller than that deduced from Ref. 4. Our experimental apparatus is insensitive to the values of λ_f quoted in Ref. 8.

The results presented stress the importance of performing measurements by improving both the light charged-particle identification system and the sensitivity of the neutron detection[12] with respect to the background in order to obtain more definitive conclusions on the existence of the investigated processes.

ACKNOWLEDGMENTS

The authors wish to express their warmest thanks to S. Leotta, N. Marino, O. Parasole, V. Parasole, S. Reito, A. Trovato, and S. Urso for their skillful technical assistance and F. Arriva for careful drawings. They are also grateful to U. Campisano, P. Baeri, and L. Torrisi for helpful discussions and suggestions during the experiment and for their help in preparing the Pd-D sample. Finally, the authors express their gratitude to G. V. Russo and R. Fonte for their help in operating the acquisition system.

REFERENCES

1. S. E. JONES et al., "Observation of Cold Nuclear Fusion in Condensed Matter," *Nature*, **338**, 737 (1989).

2. M. FLEISCHMANN and S. PONS, "Electrochemically Induced Nuclear Fusion of Deuterium," *J. Electroanal. Chem.*, **261**, 301 (1989).

3. *Proc. Workshop Understanding of Cold Fusion Phenomena*, Varenna, Italy, September 15–16, 1989, R. A. RICCI, E. SIDONI, and F. De MARCO, Eds., Editrice Compositori (1990).

4. A. De NINNO et al., "Evidence of Emission of Neutrons from a Ti-D System," *Europhys. Lett.*, **9**, *3*, 221 (1989).

5. M. CASSANDRO, G. CAVALLOTTI, and G. JONA-LASINIO, Preprint Dipartimento di Fisica, Università di Roma.

6. A. KRAUSE, H. W. BECKER, H. P. TRAUTVETTER, C. ROLFS, and K. BRAND, "Low-Energy Fusion Cross Section of D + D and D + ^{3}He Reactions," *Nucl. Phys. A*, **465**, 150 (1987).

7. G. R. CAUGHLAN, W. A. FOWLER, M. J. HARRIS, and B. A. ZIMMERMAN, "Tables of Thermonuclear Reaction Rates for Low-Mass Nuclei ($1 \leq Z \leq 14$)," *At. Data Nucl. Data Tables*, **32**, 197 (1985).

8. M. GAI et al., "Upper Limits on Neutron and Gamma Ray Emission from Cold Fusion," *Nature*, **340**, 29 (1989).

9. A. TUROS and O. MEYER, "Depth Profiling of Hydrogen by Detection of Recoiled Protons," *Nucl. Instrum. Methods*, **B4**, 92 (1984), and references therein.

10. K. E. REHM, W. KUTSCHERA, and G. J. PERLOW, "Search for Protons from the $^2H(d,p)^3H$ Reaction in an Electrolytic Cell with Pd-Pt Electrodes," *Phys. Rev. C*, **41**, *1*, 45 (1989).

11. L. BRUSCHI, M. SANTINI, G. TORZO, and G. NARDELLI, "Search for Neutron Emission from a Deuterium-Titanium System," *Europhys. Lett.*, **10**, *4*, 303 (1989).

12. A. BERTIN et al., "Experimental Evidence of Cold Fusion in a Measurement Under the Gran Sasso Massif," *Nuovo Cimento*, **A101**, 997 (1989).