

Research Article

An Historical Experiment of Neutron Detection Near an Electrolytic Cell

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Abstract

An electrolysis experiment performed in April 1989, with a hollow palladium cathode in heavy water showed neutrons production. The results were sufficiently reliable to exclude any experimental error. Unfortunately a similar experiment has never been attempted since then. The presence of neutrons is the signature of nuclear reactions.

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1. Introduction

At the time of the March 23 announcement by Fleischmann and Pons [1] of possible nuclear fusion reactions at room temperature inside a palladium cathode, we were working at the Centre d'Etudes Nucleaires de Cadarache in France. Our laboratory was devoted to neutron dosimetry and we had access to suitable equipment for neutron measurements. For us, the easiest method to check the validity of Cold Fusion was to proceed with neutron measurement near an electrolytic experiment. We had several old palladium–silver tubes used to produce ultra pure deuterium gas for an ion accelerator. The palladium–silver tubes were 10 cm long, 2 mm outer diameter, 0.2 mm thick walls and closed at one end. We succeeded in detecting neutrons. In spite of this success, our work was quickly stopped by the head of the French Atomic Energy Commission, and no further experiments were possible.

2. Experimental Details

The electrolytic cell included the palladium-silver cathode (with only 5 cm of the tube immersed in the electrolyte), and a platinum coated anode located around the cathode. The electrolyte was D₂O with NaF. The neutron detector

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located 43 cm from the cathode was a helium-3 type at the center of a polyethylene sphere, 20 cm in diameter. The sensitivity was $0.43 \text{ neutron/cm}^2$, with an uncertainty of $\pm 5\%$. Calibrations were performed with 2.5 MeV neutrons, from a Van de Graaff accelerator. Before each experiment, the calibration of the measurement equipment was verified by means of a reference americium–beryllium source.

Because we lacked a valid hypothesis, we supposed that the neutron emission was in the form of very short bursts. In that case, the best method was to proceed by integration with activation detectors. Several detectors were placed near the electrolysis cell: copper, indium, sulfur and magnesium. The sphere was positioned near a section of the electrolysis cell. Measurements were accumulated for ten minutes before being recorded. To test the counter stability, the polyethylene sphere received a weak and stable high energy neutron flux, originating from two strong sources, americium–beryllium and californium-252 located inside a polythene container. With this type of protection, a very small part of the initial neutron spectrum could escape at high energy. Therefore, the background count was 0.22 pulse/s.

During the first two experiments, we did not measure any increase in the neutron flux with the helium-3 detector. The only observation was a bending of the cathode and cracks on its surface. Since only half of the palladium tube was immersed in the electrolyte, this probably provoked stress inside the section of the palladium loaded with deuterium.

The third experiment was different. Before electrolysis, the third tube was partially loaded with deuterium by placing it in an atmosphere of deuterium at about 2 bar for several days. After some trials of electrolysis current, the amperage was set to 10 mA/cm^2 overnight. During this time (about 16 h), the counts on the helium-3 detector were stable at the background level. Then, the counts showed a surprising and roughly linear increase, for about 5 h, up to a level four times larger than the background level. Unfortunately, the experiment stopped because of an electric power failure in the laboratory due to a thunderstorm. About 30 min later, with the return of the electric power, the counts were about at the same level, but they decreased rapidly to return to the background level in less than 2 h (Fig. 1).

The measurement apparatus seemed to work properly, with background level counts before and after the increase in counts. There was no possibility of parasitic neutrons sources.

Consequently, we conclude that an unexplained neutron emission was certainly observed. The largest net count was: 0.7 pulse/s. The estimate total neutron emission was: $3.8 \times 10^4 \text{ neutron/s}$ with an uncertainty of about 20%. We note that, in the initial paper of Fleischmann and Pons ([1], p. 306), the neutron emission is nearly the same: $4 \times 10^4 \text{ neutron/s}$ with a cathode 10 cm long and 4 mm in diameter.

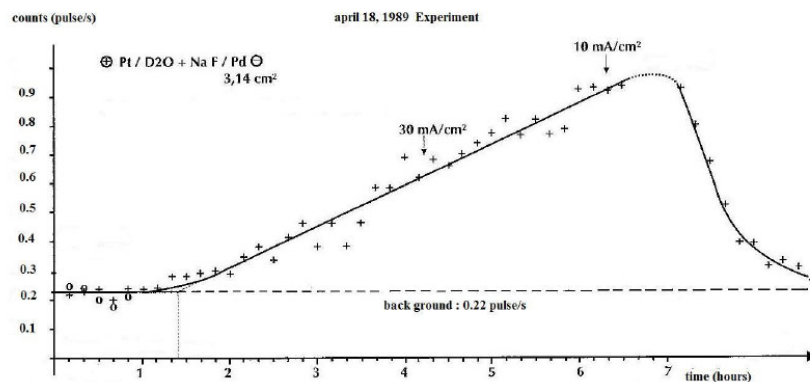


Figure 1. Neutron pulse counts (pulse/s) versus time (h).

No radioactivity was measured with the activation detectors. In order to perform new measurements, but with a reduced background, the experiment was transferred to an underground room. Nevertheless, the sensitivity to parasitic electromagnetic radiation was negligible, for the same equivalent dose, the ratio of neutron counts/gamma counts was greater than 10^4 , tested with ^{60}Co and ^{226}Ra gamma sources. Then again, the arrival of the thunderstorm, coming with a barometric depression, induced a radon release, perceptible on atmospheric controls. The possibility that high energy gammas from ^{214}Bi , daughter of ^{222}Rn , could generate photo-neutrons in the heavy water jar was considered. A similar measurement apparatus was used, in parallel, with an identical Bonner sphere near a 20 liter heavy water tank. No difference of background has been observed in the underground room. Despite numerous attempts, behavior similar to the one reported above was never observed.

If a single experiment was insufficient to understand the observed phenomena, the presence of neutrons appeared real and certified the presence of nuclear reactions.

Below, this experiment is referred to as the “April 18 experiment”

3. Discussion

Later on Biberian utilized a tube similar to the one used for the April 18 experiment in open cell electrolytic experiments. At the end of one experiment, the Dewar type Pyrex cell exploded [7]. The cell enclosed a very small quantity of deuterium and oxygen gas, not enough to cause a chemical explosion of that magnitude. Despite this dramatic event, the amount of energy was not sufficient to induce apparent damage to the palladium tube. Probably this energy was released in a very short burst leading to a production of large power and a shock wave. Incidentally, we note that the tubes have a disturbing tendency to induce nonchemical explosions [9]. More recently, the same type of palladium tube was used during another experiment and 3 W of excess energy was detected for several days [10].

Undoubtedly, the energy released is due to the same phenomenon which can evolve in a very short period of time and even become explosive, or over a time more or less long, allowing excess energy measurements. This phenomenon has characteristics similar to “criticality” in fissile materials, which hints at the possibility it might develop chain reactions under special conditions.

Therefore, we can now understand why the April 18 experiment was not successful in the underground room where the external flux of high energy neutrons was absent.

4. Conclusion

The hypothesis of chain nuclear reactions sustained by protons seems in good agreement with the data resulting from cold nuclear fusion experiments. But this is only a hypothesis, valid as long as an inconsistency is allowed. The direct evidence of cold nuclear fusion requires indubitable and reproducible experiments. Maybe the chain nuclear reactions model is capable of giving a research clue, if however the hypothesis agrees with the reality.

Our interest is to demonstrate unlimited energy resources since the deuterium is extracted from water, no radiation apart some neutrons, and no radioactive product apart from tritium in small quantity.

The development of a line of “cold nuclear fusion reactors” would produce large quantities of helium-4. It would be also possible to recover quantities of helium-3, mainly from tritium disintegration. This element is considered as essential, in the future, for the “hot fusion” reactors.

Appendix A. Neutron Detection

The neutron detector was a Bonner polythene sphere, 8 in. in diameter, with a small size helium-3 detector at its center. Before each experiment, the measurement apparatus was verified by means of a low emission neutron source (7 mCi

Am–Be). Thus, all the equipment was operational for the measurement (Fig. 1). The detector sensitivity was $0,43 \text{ pulse/ n.cm}^{-2} \pm 5\%$ for 2.45 MeV neutron energy (Van de Graaff accelerator calibration). Figure 2 shows a view of the experimental environment.

Measurements were proceeded by a sequence of 10 min periods by a very short time to data transfer. To verify the apparatus stability, a high background was chosen (about 120 pulses in 10 min) by placing the experiment near a container with two strong sources, Am–Be and ^{252}Cf , behind a biological protection (polythene + boron) (Fig. 2).

Several trials confirmed the background stability, except in one case. Before electrolysis, the tube used was partially charged by pressure of about 2 bar during several days. After some trials of electrolysis current (variation $40\text{--}10 \text{ mA/cm}^2$) the amperage was set to 10 mA/cm^2 for a night. During this time, about 16 h, the counts of Bonner sphere were the stable background. Then, the counts showed (Fig. 1) a surprising and roughly linear increasing during about 5 h, up to a level four times greater than background. Unfortunately, the experiment stopped because of a shutdown of the electric power in the laboratory, due to a thunder storm. About 30 min later, with the return of electric power, the counts were about at the same level, but they decreased rapidly to return to the background level in less than 2 h. Taking into account the maximum at 0.92 pulse/s , the net count was $0.7 \pm 0.14 \text{ pulse/s}$. The center of the sphere was 43 cm far from the cathode and the estimation of neutron emission was: $Q = 3.8 \times 10^4 \pm 0.8 \times 10^4 \text{ neutron/s}$ in 4π steradian (about $\pm 20\%$, uncertainty at 3σ). In the initial paper of Fleischmann and Pons (p. 306) the neutron emission is $4 \times 10^4 \text{ neutron/s}$ with a palladium cathode: length 10 cm , diameter 4 mm .

The measurement apparatus seems quite right: stable background before and after the counts evolution, same counts before and after the electric shutdown, no possibility of parasitic neutrons sources in proximity to the laboratory.



Figure 2. Experimental set-up showing a neutron dosimeter calibration. During the 18 April experiment, the neutron sources were located inside the black cubic container about 2 m from the electrolysis cell.

Despite several trials, this experiment is unique and a single experiment is insufficient to understand the observed phenomena.

Appendix B. A Tentative Theoretical Explanation

In conclusion, the presence of neutrons, pointing out nuclear reactions, appears real. The high energy neutrons, going out of the external source container near the experiment, seem to originate according to the hypothesis developed below.

High energy neutrons can move deuterons in the palladium lattice and so create $D + D$ reactions with a proton emission. According to a process described by the Nobel laureate Julian Schwinger a proton can generate a: $p + d \rightarrow {}^3\text{He} + \text{gamma}$ (5.48 MeV) reaction. The ${}^3\text{He}$ may undergo a secondary reaction with another deuteron in the lattice, yielding ${}^5\text{Li}^*$ which is unstable against disintegration into a proton and an alpha particle. Finally, a three body: $p + d + d$ reaction occurs, hypothesis considered by Yamaguchi and Nishioka (*Proc. ICCF3*, Nagoya, 1992). With this process, the proton is not consumed and chain reactions can occur. In the experiments described here, no 5.48 MeV were observed and the photon radiation was negligible. In the $p + D$ reaction all the elements have even parity and with no change of parity electric dipole radiation is forbidden, other effects might intervene but very weakly.

With the lack of 5.48 MeV gamma, the mass energy balance is about 24 MeV per cycle, the same energy for a direct fusion $D + D$. The cross section of the initial $p + D$ is very low but presumably is increased by channeling, where the palladium lattice acts as many particle guides. With the hypothesis of chain reactions, it is necessary to have additional $D(d, p)T$ reactions to implement the growth. The conditions are thus quite similar to the growth in a critical fissile medium with an effective multiplication factor k . This factor k is bound to the charge ratio D/Pd and to the “geometric buckling” of the palladium sample. When k is greater than 1, the growth of chain reactions is exponential. If k is just greater than 1, the growth is approximatively linear as in our experiment. If k is much greater than 1, the growth becomes explosive. This may explain why with a tube similar to the one used for our experiment, in an open cell electrolytic experiment by Biberian, the Dewar type Pyrex cell exploded.

The beginning of chain reactions requires a primary proton, and external radiation is necessary: high energy neutrons in our experiment – high energy gamma ($E > 2.23$ MeV) which divides a deuteron in proton and neutron – cosmic meson, which generate a $D + D$ reaction (Sakharov fusion). Added to the development of criticality conditions (charge ratio, shape and dimensions of the palladium sample), this external intervention can explain why positive experiments are uncertain and not even possible in low background laboratories.

An electrolytic experiment in the presence of a strong source of high energy neutrons can validate the hypothesis of chain reactions, but if the result is positive the interest is only historical, just to say Fleischmann and Pons were right. The use of palladium deuterium system offers many difficulties: palladium is expensive, deuterium is a regulated material, neutron radiation and tritium are produced, and there is a potential risk of explosion. The experiments conducted with hydride of metals such as nickel or tungsten are more promising. Energy excess is observed presumably by nuclear transmutation after proton capture. The palladium is not able, by proton capture, to give transmutations to silver isotopes because of wave function parity.

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