Investigation of Anomalous Densities of High-energy Alpha-Particles Tracks in CR-39 Detectors during Electrolysis of Heavy Water on Palladium Cathodes.

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Abstract. Recently, several researchers claim the finding of anomalous alpha-particles generation during very simple electrolysis experiments with heavy water and palladium cathodes. The phenomenon seems to improve if deuterium formation on the cathode is associated with deposition of palladium nanostructures coming from chlorides of the same metal present in the electrolytic solution. Due to the relevance of the claims and considered the simplicity of the experimental apparatus, several tests have been performed in order to confirm the claimed results. The results of these tests will be the object of this scientific report.

1. Introduction

Palladium is an element of Group X which has the peculiar characteristic to be extremely permeable to hydrogen; indeed, it is able to adsorb this gas up to 900 times its volume at room temperature. When adsorbed, the gas redistributes itself in the metal lattice, but it is not clear if this behavior is due to the formation of hydride or is only a temporary binding. The very high permeability to hydrogen (and its isotopes) is used in the industry to purify this gas with systems forcing it to flow through palladium membranes, which stop the impurities.
In 1989 M. Fleishmann and S. Pons reported the "strange" result of anomalous temperature rising measurements on an electrolytic cell with a palladium cathode. The electrolyte was LiOH in heavy water D_2O. The sensational impact of their claim was due to the fact that the entity of extra heat generated was not compatible with a chemical origin and consequently its source should be a nuclear reaction.
The concentration of H-D in palladium depends from the thermodynamic parameters; in the cathode, by effect of applied potential, concentration rises up prominently. This mechanism, named "loading", is used to achieve the wanted thermodynamic non equilibrium in order to increase the deuterium concentration in the cathode, the necessary condition to have excess heat production. According to Fleishmann and Pons claim, the temperature raise, which were observed at high "loading" maintaining constant the applied power, would be explained invoking a nuclear mechanism in the metal lattice: D atoms reduce their relative distance, overcoming the electrostatic repulsion of the Coulomb barrier getting to nuclear fusion events. Palladium matrix should act as a sort of catalyst for the reaction. From 1989 many researchers, working both in civil and military field, began an experimental activity on these phenomena, indicated with the acronym LENR (low energy nuclear reactions). In particular, researchers tried to set up excess heat experiments, also varying the experimental embodiment; moreover, theoretical studies were developed to explain the mechanism of low temperature nuclear fusion. The attention was especially dedicated to the production of energetic particles and gamma radiation. In fact, their detection should be a clear clue that nuclear reactions were involved. Concerning particle emission, according the classical theory, during fusion a flux of them (alpha, neutron, gamma ….) has to be generated to carry almost a part of the energy produced. Unfortunately, many trials to detect them were unsuccessful. The prevalent opinion is that LENR does not produce the neutrons flux as for the standard model of this kind of reactions, even if it is hard to explain why. By the way, several scientists continued to look for neutron emission, but the results are not representative. Another research line was the detection of other corpuscular emission, based on the use of well known plastic detectors. Precise protocols are implemented for alpha particles in order to distinguish between the particles generated during the electrolysis and those normally present in the environment as by-product of Radon decay. Radon is a radioactive gas present in particular rocks. Many experiments were performed in the past and are going on to check for alpha emission during palladium loading using a plastic detector, called CR39 (Fig.. 2). When a particle hits the detector, it breaks the polymer chains and the damage becomes visible by a chemical etch in NaOH solution. The etch rate depends on the

temperature of the bath. After etch, a small conical hole appears in correspondence of the incidence path of the particle having the geometrical axis direction depending from the incidence angle.

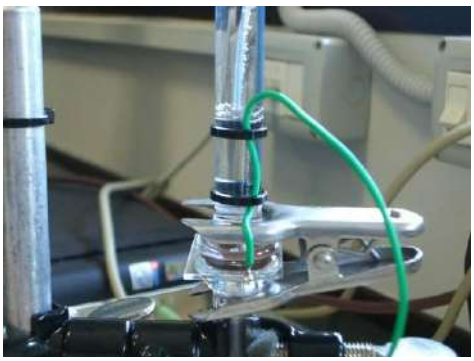

Fig. 1 - Experimental embodiment.

Fig. 2 – CR39 detector

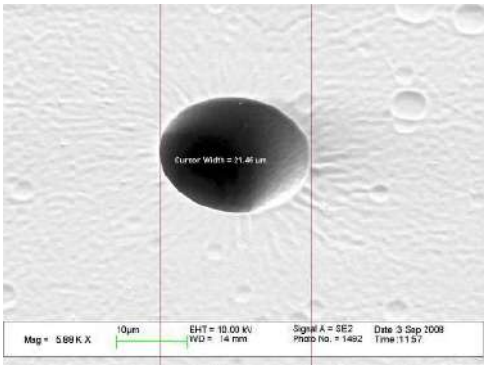

Fig. 3 - SEM photo of an alpha particle track after etch

2. Experimental Details

Two kinds of experiments were performed:

- Electrolysis of LiCl electrolyte in D2O;
- Codeposition of Pd on the cathode adding PdCL2 in the solution. Effects of high and low electrolysis current densities were also tested;
- Comparison of all the test results with CR39 detectors placed in the same embodiment, but without electrolysis.

In the very simple embodiment (Fig. X) the electrolytic cell was a glass tube with an internal diameter of 9 mm having a flange to accommodate an O ring for sealing. Another identical glass tube, empty, was connected to the one with the electrolyte with interposed the CR39 plastic detector having a size 23mm x 35mm x 1.39 mm, produced by INTERCAST EUROPE. CR39 for nuclear measurement is very pure and without defects. The cathode was formed by a palladium wire (diameter 50 µm) fixed on sensible side of detector and between the cathode and CR39 a 11 µm Mylar foil was interposed to prevent the contact of the electrolyte with the detector; all the embodiment parts were fixed together by a metallic clamp. The cell was filled with 8 ml of 0.5 M LiCl/D2O solution. D2O used was undertaken at double distillation to assure its purity. Power supply was a current generator able to generate up to 3 A; voltage and current were continuously monitored by a digital multimeter. The current was incremented in step of 1 mA till 10 mA every 8 hours; at 10 mA the electrolysis went on from 3 to 10 days. To evaluate the counts two reference samples were prepared like experiment 1, 10 mA constant for three days, using only light water for electrolysis.

3. Etch

After any experiment, the detector development process to count the high energy tracks was performed in a NaOH 6.25 M solution, at a temperature of 70°C; the solution was contained in a beaker heated by an hot plate electronically controlled at +- 0.2 °C. The etching time was 6 hours. After the etch the detectors were rinsed in D.I. water, then neutralized in HCl diluted for 10 minutes and finally rinsed again in D. I. water and dried in a flux of filtered nitrogen gas.

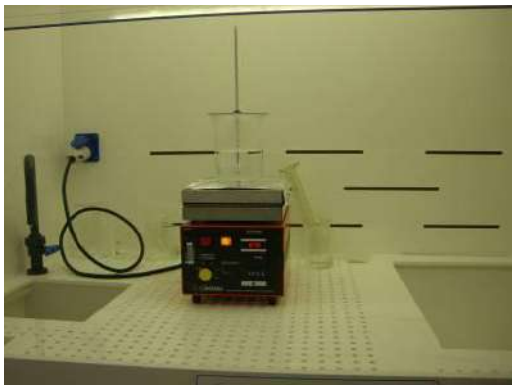

Fig. 4 - Heated etch bath (NaOH)

4. Track Analysis

The count track was performed using a system formed by a Leica binocular microscope in transmission configuration with a webcam and a PC running dedicated software for image acquisition. CR39 detectors were put on a plate with a hole 12 mm wide just to analyze detector area faced to the inner part of the cell during the electrolysis. To analyze the exposed area of 1.186 cm^2 125 photos with 10x objective were taken.
The experiments using CR39 have the aim to demonstrate the existence of nuclear reaction inside the palladium cathode; because the "mark" of these reactions are particles emission, it is necessary that the number/density of them detected on CR39 after deuterium electrolysis are SIGNIFICANTLY higher than the same figures related, for instance, in absence of current flow or using light water. Moreover, because of the direct contact between the cathode and the detector, it is expected to detect a higher density over all in the detector area close to palladium cathode.
The results do not show a significant difference with respect to the detectors used as reference (light water). The Mylar film interposed between cathode and detector acts like a shield for particles with energy less than a specified value which depends on the film thickness. To evaluate the penetration thickness of the particle in Mylar the freeware software SRIM was used. This code calculates stopping power, projected range and other parameters for proton and alpha particles with energy up to 10 MeV. For Mylar 11 μm thick used in the experiments the minimum energy for alpha particle to go across the film is 2.5 MeV. All particles less than 2.5 MeV are stopped. The absence of a significant increase of trace number on detector excludes particles emission with more than 2.5 MeV.

Fig. 5 - System for tracks count

Table 1. Track Density (cm^{-2})

	Blank		Electrolysis
# 1	189	# 1	222
2	154	# 2	296
		# 3	136
		# 4	112
		# 5	74

5. Experiment without MYLAR

In the specific literature, the first experiments based on the electrochemical loading and CR39 detectors were performed without any protection film between the detector and the electrolyte. The solution used was D2O with a small addition of palladium chloride salt (LiCl or LiSO4); the possibility that during electrolysis a surface damage of the detector happened was not evidently taken into account.

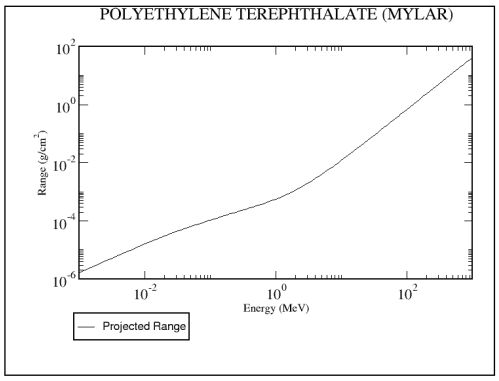

Fig. 6 - Stopping power curve for Mylar

Some researchers reported positive results in terms of high count of traces identified as particle emission from cathode just with experimental arrangements which did not use any interposed film. Comparing images of the reported traces with similar relative at radioactive emission from americium or uranium the difference is very evident. To investigate this point some experiments without Mylar were performed. After electrolysis, just to a naked eye detectors showed some milky and rough appearance; moreover, the area just in contact with palladium wire showed a marked white line. Optical microscope analysis highlighted the presence of many “balls” in coalescence each other; similar structures did not resemble at all with traces from nuclear particle. SEM analysis on some detectors confirmed this strong difference. The surface damage can be so caused from chemical etch of liquid on plastic surface.

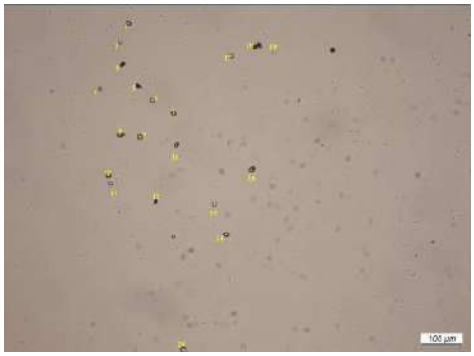

Fig. 7 - Example of CR39 area count

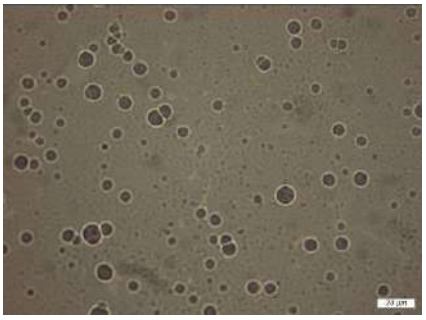

Fig. 8 - Artifact on CR39 surface, without Mylar

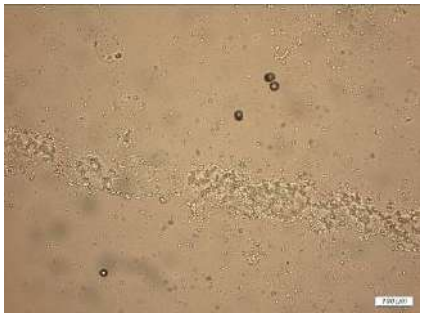

Fig. 9 - Artifacts in CR39 area faced cathode, without Mylar protection

6. Codeposition Pd/D

Szpac and Mosier-Boss performed some experiments in which palladium, coming from palladium salt present in the solution, was codeposited on cathode together with deuterium gas generation with the purpose to have nanostructure of palladium on the cathode were the "loading" could reach an atomic D/Pd ratio close to 1. The same authors reported positive result concerning particle emission during electrolysis. To verify this claim, two trials of codeposition were performed, using as palladium source palladium chloride PdCl2, added in the solution at 0.05 M, concentration. During these experiments two different procedures in increasing the electrolysis current were used. In trial A the starting current was 10 mA up to complete Pd deposition, then raised to 20 mA and maintained at this level for three days; in trial B the electrolysis started at low current – 0.6 mA – then maintained at this level until the palladium deposit on cathode was complete. In both cases it was possible to determine the end of palladium deposition by the solution color change from orange to transparent. So, for the experiment B the current was gradually raised up to 50 mA and the reaction went on for 7 – 10 days. The maximum current density was 0.5 A/cm^2 for experiment 1 and 5 A/cm^2 for experiment 2. At the end cell was disassembled and the detector etched in the chemical bath. In both experiments the Mylar foil was interposed.

Table 2. Codeposition Parameters

Exp. -A-		**Exp. -B-**	
10 mA	Pd dep.	0.6 mA	Pd dep.
20 mA	3 days	2 mA	1 day
		5 mA	1 day
		20 mA	3 days
		50 mA	2 days

7. Results

In the case of trial A after few hours a non uniform sponge-like palladium deposit was formed on the cathode; in trial B the deposit instead was very uniform and compact. Cathode of trial B, analyzed in a SEM, showed a globular structure (cauliflower type) plus palladium dendrites present over the entire active surface. Same results are reported from Szpak and Mosier Boss with a cell put in a strong electric field. Concerning trace count, no difference was found with respect to reference sample using light water.
For both the experiments an 11 microns thick Mylar foil was interposed between the cathode and the CR39 detector. At the end of the experiments the detectors have been treated as previously described and the cathode submitted to SEM analysis.
In Fig 10 it is shown a SEM image of the cathode surface where it is possible to observe the dendritic and globular structure of the deposited Pd. Similar structures are reported in [2]; moreover it is useful to notice that the dendritic structure, as reported in [2], should be present only in experiment where the cell is placed inside a strong electric field: no electric field was present in our experiments. The track count reported in Tab. 3, does not show significant differences compared with "blank" samples, neither with electrolysis experiments without Pd codeposition.

Table 3. Track Density (Codepos.)

Exp. -A-	113 tr./cm^2
Exp. -B-	146 tr./cm^2

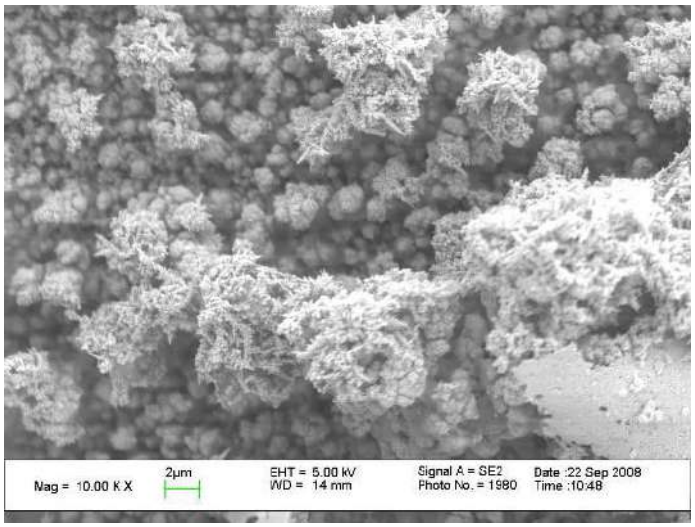

Fig. 10 - SEM photo of dendritic structures on the Pd cathode

8. Conclusions and acknowledgments

The experiments described above did not highlight a meaningful increase of Alfa particles nuclear tracks with energy greater than 2.5 MeV respect to the background of blank experiments, during electrolysis in heavy water with palladium cathodes. Same results came out also in experiments where palladium chloride salts were added to the electrolyte, in order to have nanostructures of Pd deposited on the cathode and increase deuterium loading in the Pd lattice. So, the outcomes of the experiments seem to confirm what is also reported in references [3] and [4].
A new series of test with simple electrolysis and codeposition is planned in the first part of the year 2010 using a different supplier for the CR39 detectors in order to reduce the background tracks level.

We are particularly grateful to Dr. P. Neri of INTERCAST EUROPE for the supply of CR39 detectors and Dr. F. Celani for the supply of heavy water and Pd cell cathode wires.

9. References

[1] R.A. Oriani, J.C. Fisher: *Proceedings of* 11th ICCS, 2004
[2] S. Szpak, P.A. Mosier-Boss, F.E. Gordon: *Proceedings of* 11th ICCS, 2004
[3] http://earthtech.org/experiments/PACA/report.htm
[4] http://earthtech.org/CR39/index.html