Evidence for Fast Neutron Emission During SRI's SPARWAR/GALILEO Type Electrolysis Experiments #7 and #5, Based on CR39 Track Detector Record

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Abstract. We have reported [1] the detailed analysis of the CR-39 detector (Landauer) from SRI's #BE013-7 (#7) Pd deposition experiment where the detector was separated from the cathode wire by a 6 µm Mylar® film. The Mylar® protected the CR-39 surface from chemical, mechanical, and electrostatic (spark discharge) damage during electrolysis. Inthis report we compared those results with that of the CR-39 detector, installed as in #7, in an identically operated cell using light water and with the background detector placed 2 m from the electrolytic cell.

1. Introduction

For the last 3 years claims have been made that during Pd electroplating onto Ag-wire cathodes (or other metal wires) from Pd/D_2O solution, high intensity emissions of charged particles were observed [2]. The emissions have been detected using CR-39 track detectors. The typical pit densities observed in these experiments is 10^7-10^8 pit/cm^2 and the pits are primarily located on surface of the CR-39 nearest where the cathode wire was pressing directly onto the CR-39.
The purpose of this present study is:

- To search for reality of nuclear emissions reported using Pd electrodeposition technique and CR-39 detectors. To separate pits caused by mechanical defects (electric discharge) from that of nuclear origin.
- To test applicability of our track identification technique (see A. Roussetski et al, ICCF-12, Yokohama, 2005 [1], Fig.1) based on in depth destructive etching of the CR-39 detectors, allowing us to obtain track diameter evolution vs. removed depth, to the Pd electrodeposition experiment.
- To apply the CR-39 technique to in-situ neutron detection near an electrolytic cell. To compare the results with that obtained in Blank (H_20 electrolysis) and Background (no cell) experiments as well as with a BF_3 proportional detector count rate.

A series of experiments have been performed where Pd was electrodeposited onto Ag-wire cathodes with CR-39 plastic track detectors separated from the electrolyte and electrodes, with the cell placed in an external magnetic field [3]. The Blank (H2O electrolysis) and Background (CR-39 2 m from the cell) experiments were performed using same series Landauer CR-39 sheet as used in the original D2O experiment. Contrary to earlier studies, the CR-39 detectors in this study were not in contact with the cathode or electrolyte, protecting the CR-39 surface from mechanical stress and electrostatic (discharge) damage during electrolysis. In addition to the CR-39 detector, a low self-efficiency proportional BF3 spherical neutron dosimeter was employed in each experiment.

2. Experimental

The same lot of CR-39 chips (2cm x 1cm, Landauer Inc.) were used in all experiments and calibrations. After exposure the chips were immediately exposed to 6N NaOH at 70°C for 7 hours yielding an etch rate of 1.3µm/hr. Pd was electrodeposited onto Ag-wire cathodes with CR-39 plastic track detectors separated from the electrolyte and electrodes, with the cell placed in an external magnetic field [3]. The detectors from the #7 and #5 experiments were separated from the cathode and electrolyte by sheets of 6 µm Mylar® and 60 µm polyethylene, respectively. The blanks were performed using the same electrochemical parameters, salts, and CR-39 placement as in the #7 and #5 experiments substituting H_2O for D_2O. The background CR-39 detectors were held 2m from the operating cell. A Ludlum proportional BF3 spherical neutron dosimeter with self-efficiency of 2.5x10-3 (with respect to Cf-252 fast neutrons) was employed in each experiment.

Calibration chips were exposed to proton beams of 1.0 - 2.5 MeV from a Van DeGraaf accelerator–and etched at 1.3µm/hr. Fig. 1 shows the raw data and the calibration curve obtained from proton bombardment on CR-39 chips identical to those used in the electrolysis experiments. During analysis the CR-39 detectors were etched three more times, at a bulk etch of rate 1.3 µm/hr. Track detection was performed manually using the "PAVICOM" track processing facility in Lebedev Physics Institute, Russian Academy of Sciences, Moscow.. Proton recoil track distributions from a weak Cf-252 neutron source (120 n/s) are presented in Fig.2.

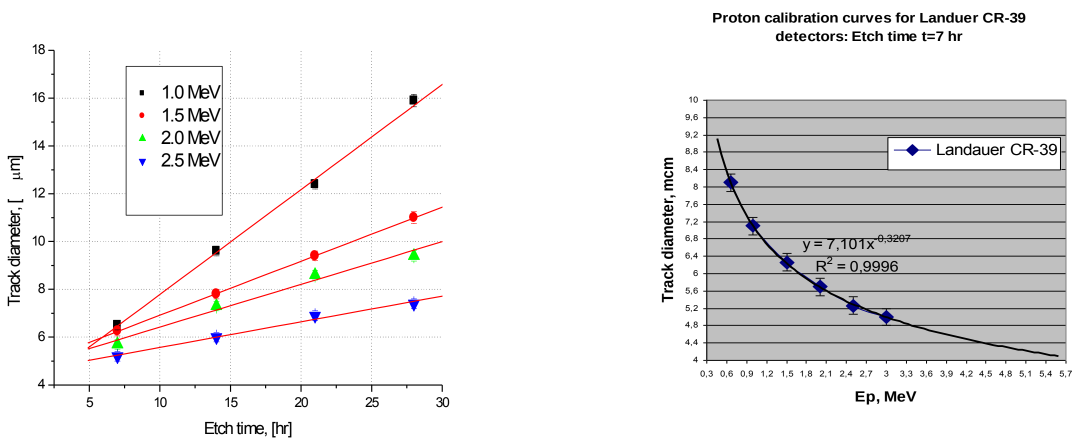

Fig.1 - Track diameter vs. etching time for four different proton energies (left). Proton calibration curve (right).

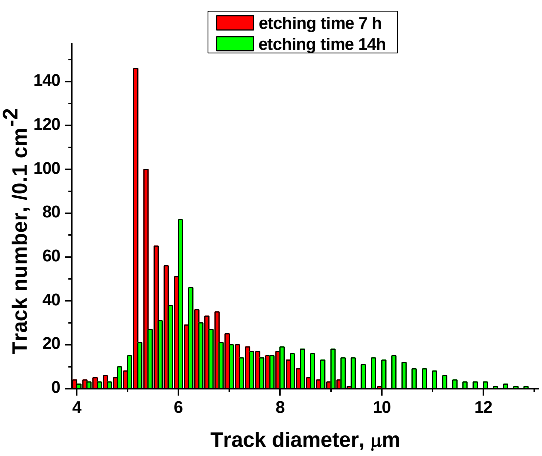

Fig.2 - Proton recoil track distributions (track number vs. track diameter) obtained with Cf-252 neutron source.

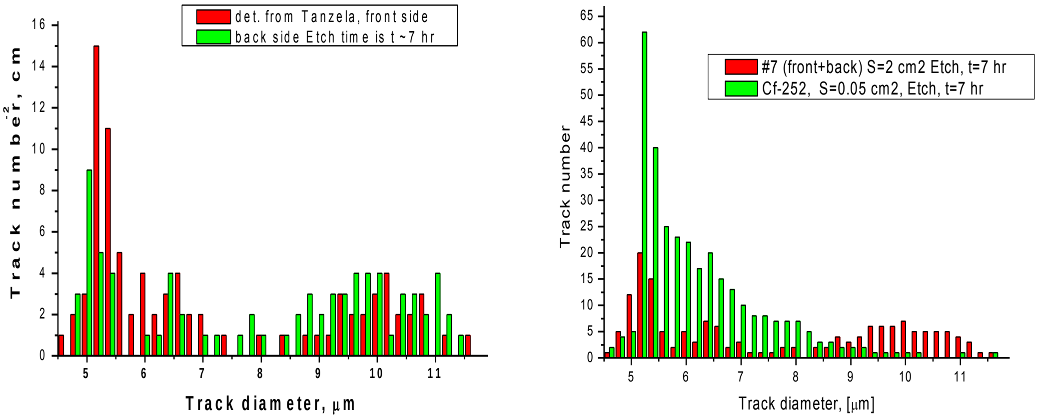

Fig.3 - Tracks from #7 detector, after 1st etch (left picture); Comparison to neutron calibration (right picture).

3. Results

After the 7-hr etch the proton recoil spectrum from experiment #7 is located between 4.5 and 9.0 μm track diameter with a maximum near 5.2 μm. This maximum is consistent with the mean recoil proton energy in the range of 2.2-2.5 MeV. The broader recoil spectrum after the 14-hr etch is between the between 5.0 and 12.0 μm proton recoil track diameters with the maximum near 6.0 μm. This shift in maximum track diameter from 5.2 to 6.0 μm is consistent with 2.2-2.5 MeV protons. The neutron detection self-efficiency of CR-39 after the second etch ($1.2x10^{-4}$) is about factor of 1.3 higher than that after the first etch ($0.9x10^{-4}$) due to the increase in proton recoil critical angles at the greater depth. Fig. 3 shows the track diameter results measured from both sides of the #7 detector after the first etch compared to that for Cf-252 recoil. For the front side: the total number of circular and elliptic tracks (normal and oblique incidence) is N=77 cm^{-2}, with about 30 % of all tracks having elliptical shape. For the backside: N=40 cm^{-2}. In the range of 4.5-8.0 μm the track diameter plot for #7 looks similar to that for Cf-252.
Fig. 4 shows a comparison of #7 data with that from the H_2O experiment and the Background after the 1st etch. The Blank and the Background data show no sign of proton recoil from fast neutrons proving that there was no accidental neutron irradiation of CR-39 sheet during transit.
Fig.5 compares all above background tracks from the #7 (D_2O) track detector after the 2nd etch with that from the Cf-252 calibration. The above background track distribution in the range of 4.5-11.0 μm is nearly identical to that from the Cf-252 recoil measurements. The D_2O detector showed 101 tracks/cm^2. Fig.6 compares all above background tracks from the #7 (D_2O) track detector after the 2nd etch with that from the H_2O experiment and the background detector. Fig.7 shows a rough reconstruction of the protons recoil spectra for CR-39 detectors obtained during the D_2O electrolysis run #7 and during exposure to a Cf-252 neutron source after the 2nd etch. The reconstruction was

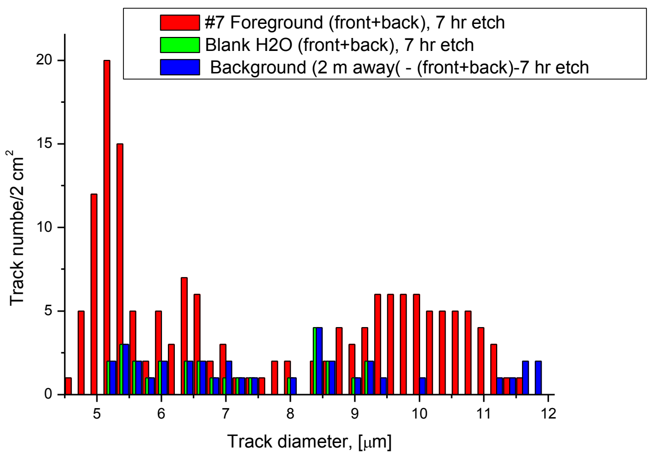

Fig.4 - Comparison of #7 data after 1st etch with that from Blank experiment and Background.

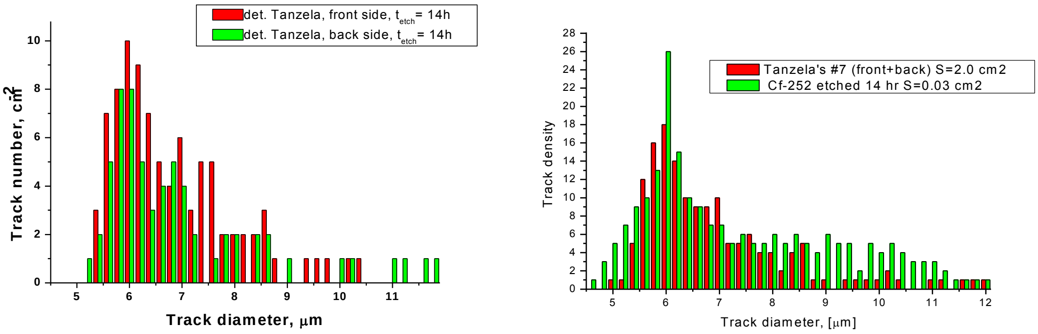

Fig.5 - Comparison of the #7 track detector distribution after the 2nd etch with that of the Cf-252 recoil .

deduced from the track density vs. track diameter histograms, taking into account the track diameter vs. proton energies after the 2nd etch and the critical incidence angle θ_c vs. proton energy.

We also noted that statistically significant counts were recorded by the BF_3 dosimeter during the D_2O runs #5 and #7.

3. Discussion

After the 1st etch the average track density on both sides of the detector <N(fg)> is 58.5 tracks/cm^2. The total background track density on both sides of blank detector <N(bg)> is 6.0 tracks/cm^2. After subtraction $<\Delta N>$ is 52.5 ± 8.0 track/cm^2. Assuming a "neutron source" was a cathode wire in a 2π-geometry with respect to the CR-39 surface, the neutron count rate/intensity (I_n) would be $2<\Delta N>/(t \times \varepsilon_s)$, where the CR-39 self-efficiency after the 1st etch (ε_s) is 9.2 x 10^{-5} and t is the electrolysis time. Assuming that the neutrons were emitted when the current was greater than 0.5 mA (t = 15 days), I_n = 0.90 ± 0.14 n/s. If we assume that the neutron emission was observed only when neutron dosimeter read above its background (t = 4 days), I_n = 3.38± 0.53 n/s. So, the neutron emission rate in the D_2O run #7 is in the range of 1.0-3.0 n/s. After the 2nd etch, <N(fg)> = 88 cm^{-2}, while <N(bg)> = 26 cm^{-2}, and ε_s = 1.17 x10^{-4}. I_n is again is in the range of 1.0-3.0 n/s as shown after the 1st etch.

The #5 CR-39 detector used in SRI BE010-5 PdD_x deposition electrolysis experiment had a 60 µm polyethylene film adhered to both faces while immersed in the electrolyte and in contact with the cathode. Although the front face was found to be covered with high density pits (defects) making it almost impossible to distinguish real nuclear tracks from defects, the rear face of #5 detector shows proton recoil tracks similar to those found on both faces of the # 7 CR-39 (with a track density 50 -70% of that of #7). These pit densities are shown in Fig. 8 and actual pits are shown in Fig. 9.

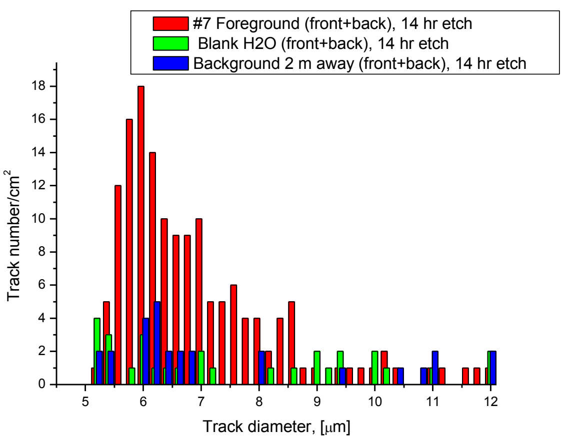

Fig.6 - Comparison of Foreground #7 data (taken from both sides) with that from Blank experiment (H_2O electrolysis) and the Background (detector is placed 2m away of the electrolytic cell) - 14 hr etch

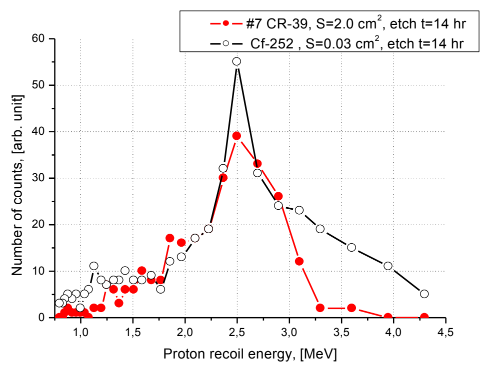

Fig.7 - Rough reconstruction of the protons recoil spectra from both the D_2O run and the Cf-252 exposure.

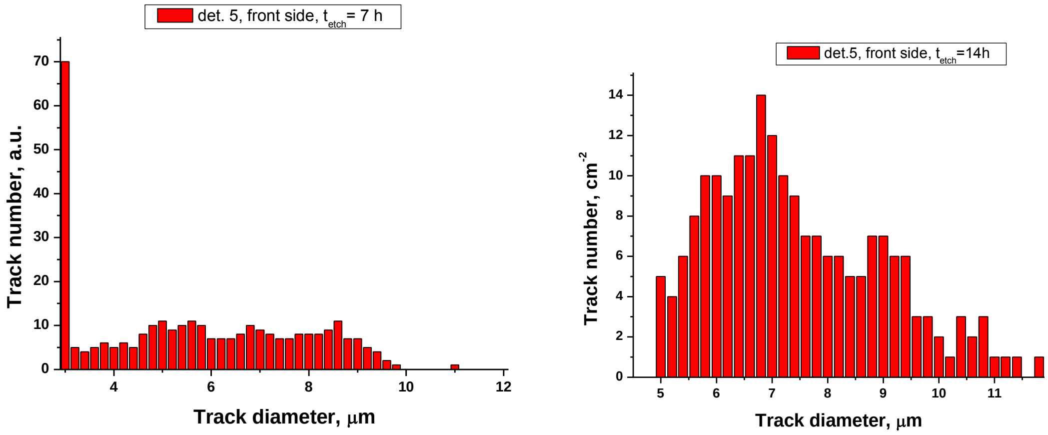

Fig.8 - Individual pit density at the front side of #5 detector: 7 hr (left) and 14 hr (right) etch

The analysis of the back side of detector #5 after the 1st etch showed N(Fg) = (30.0±5.5) recoil protons/cm^2, N(Bg) = 6 ±4 cm^{-2}, ΔN = 24.0 ± 6.8 p/cm^2, and $\langle In \rangle = 2\langle \Delta N \rangle/(t\varepsilon_s)$ = 0.30±0.08 n/s. After the 2nd etch (back side only): N(Fg) = 45 cm^{-2}, front N(Fg) = 63 cm^{-2}, $\langle N(fg) \rangle$ = 54.0±7.3 cm^{-2}. Background $\langle N(bg) \rangle$ = 26±5 cm^{-2}, ΔN = 28±9 cm^{-2} , and $\langle In \rangle = 2\langle \Delta N \rangle/(t\varepsilon_s)$ = 0.29±0.09 n/s in 2π solid angle. If t = 1 day In = 6.0±1.6 n/s in 2π solid angle. These results are shown in Fig. 10.

At a self–efficiency to fast neutrons $\varepsilon_s = 7.6\text{x}10^{-3}$ (R~ 0 cm) and detector-source distance of 10 cm, the total efficiency of BF_3 sphere for fast neutrons (ε_t) would be $7.6\text{x}10^{-5}$. The sensitivity of the detector to fast neutrons (minimal detectable emission rate) can be expressed as: $S = 3[\langle N_b \rangle/(\varepsilon_t 2\tau)\ ]^{1/2}$, where $\langle N_b \rangle$ is the averaged background count rate, and τ is the duration of neutron detection. For detector #7: $\langle N_b \rangle \approx 6.0$ cps, τ = 15 days, resulting in S ≈ 150 n/s, (300 n/s, when t=4 days). This results is 100 times higher than that measured by the CR-39 detector in that experiment. This is also true for the results measured in experiment #5.

4. Conclusions

- Entire results of two CR-39 detectors analysis show that a weak, but statistically significant emission of fast neutrons has been observed in the SRI's #7 and #5 runs replicating SPAWAR Pd-depostion experiment (in the presence of an external electromagnetic field).

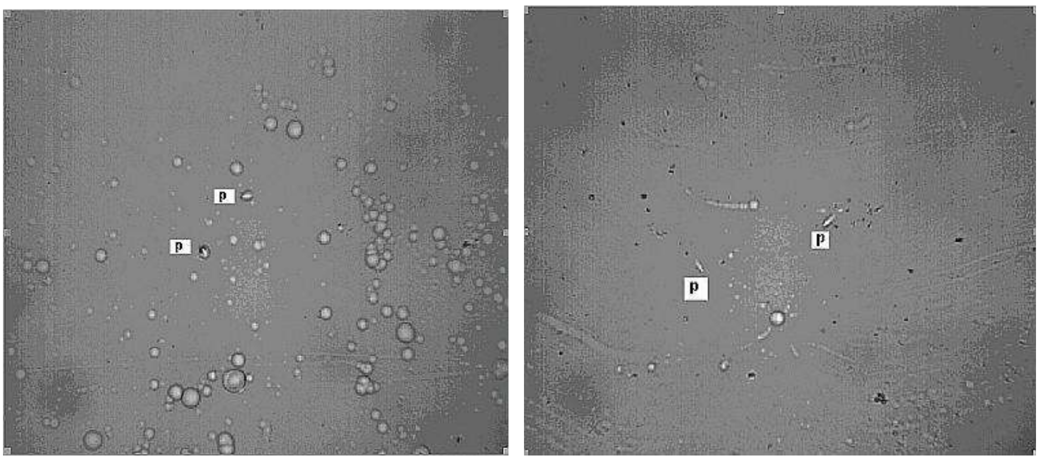

Fig.9 - Proton recoil tracks at the front side of #5 detector and the defects ("ground beef") after 2nd etch.

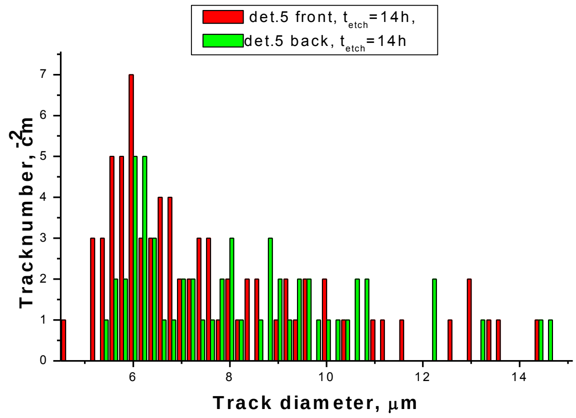

Fig.10 - Comparison of the back and the front side proton recoil spectra at t = 14 hr etch

- The #7 detector separated from the elctrolyte by a 6 µm Mylar® film shows “clean” front and back faces, containing only nuclear tracks (proton recoil). While the #5 detector, submerged in the electrolyte but covered with a 60 µm PE film, had vast areas covered by defects (“ground beef”) on the front face lower (than #7) proton recoil density at the back side
- The small diameter defect pits were eliminated by in-depth etching (>18 µm) allowing us to distinguish real proton recoil tracks caused by neutrons as well as by energetic charged particles (protons and alphas) emitted from the PdD_x powder being deposited on top of the detector during electrolysis.
- Comparison of proton recoil spectra (track number vs. their diameter) of the analyzed detectors with that of their respective Blank and Background runs and with the tracks from Cf-252 source gives solid evidence for a fast neutron emission taking place during the runs #7 and #5.
- Comparison of the neutron emission rates obtained from CR-39 analysis with that deduced from SRI (proportional BF3 detector) measurements shows a of the results, suggesting orders of magnitude higher neutron emission recorded by SRI detector than that calculated from the noiseless CR-39 measurement data .
- Because of the huge discrepancy between the CR-39 and BF_3 dosimeter results and since the dosimeter has very low neutron sensitivity we assume that the excess dosimeter counts contain significant electromagnetic noise, induced by the electrolysis power supply.
- In order to provide additional confirmation for our CR-39 results on neutron emission in SRI experiments, a high-efficiency neutron spectrometer would be desirable.

Acknowledgements

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5. References

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[3] F.L. Tanzella et al, *Presented at the 8th International Workshop on Anomalies in Hydrogen/Deuterium Loaded Metals*, *Catania*, *2007*.