

# **Analysis of the CR-39 detectors from SRI's SPAWAR/Galileo type electrolysis experiments #7 and #5. Signature of possible neutron emission**

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## **Abstract**

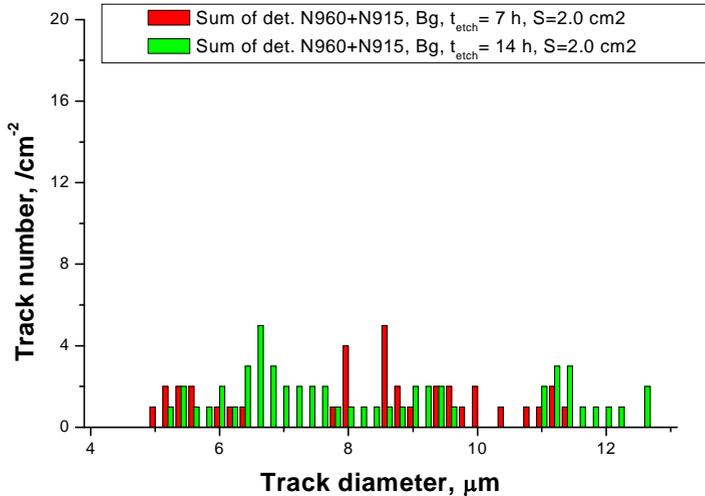
We performed a detailed analysis of two Landauer CR-39 detectors exposed to the cathode wire during palladium deposition experiments at SRI. (Experiments #BE013-7, #7; and BE010-5, #5.) The entire data set obtained from the analysis of the #7 CR-39 detector, including 1) track reading within three removed depths (8.7, 18 and 27  $\mu\text{m}$ ), 2) comparison of foreground #7 track densities and distributions of their diameters with similar parameters of the background, 3) the neutron calibration, as well as 4) the CR-39 efficiency estimate with respect to Cf-252 neutrons, present preliminary evidence for fast neutron emission. The neutron energy is estimated to be in the range of  $E_n \sim 2.2 - 2.5$  MeV with a rate of  $I_n \sim 1-3$  n/s accounting for the  $4\pi$  solid angle. The data obtained from the analysis of detector #5 allow us to conclude that a weak neutron emission from the cathode took place during electrolysis, in addition to some mechanical and electric discharge damage to the front face of the detector.

## **I. CR-39 Reading**

From April to October 2007 we carried out a reading of the #7 and #5 Landauer CR-39 detectors exposed in SPAWAR type Pd electrodeposition experiments at SRI. This was the version of the experiment without an external electromagnetic field. [1,2] We examined the samples at three different removed depths, with three etching times, roughly corresponding to 7, 14 and 21 hr etch in 6 M NaOH at 70°C at  $v_b \approx 1.3$   $\mu\text{m/hr}$ , and we compared the results with that of blank CR-39. The reading was performed manually using the track reading facility "PAVICOM" in the Lebedev Physics Institute, Russian Academy of Sciences, Moscow, Russia. The #7 detector was initially covered with 6  $\mu\text{m}$  Mylar filter, protecting the CR-39 surface from mechanical stress and electrostatic (spark discharge) effects induced by the cathode wire during electrolysis. We carried out the #7 detector reading (and the blank detector reading) from its both sides, including the front face (that was attached to the cathode) and opposite face (that was located from the side opposite to the front side). The reading area of the foreground #7 detector was  $S=1.0$   $\text{cm}^2$  of each side. In case of the blank detector (background) we obtained only a small piece of

CR-39 with readable area of  $S = 0.25 \text{ cm}^2$ . It was found that track density of the blank detector in the track diameter range of interest ( $4.0 < d < 8.0 \text{ }\mu\text{m}$ ,  $t = 7 \text{ hr}$  etch) contain in total 3 track/ $0.5 \text{ cm}^2$  (from both sides). This number is typical for the blank Landauer RadTrack detectors ( $N = 6 \pm 4 \text{ track/cm}^2$  as a result of more than 100 of our measurements), which is why we concluded that the blank detector has not been irradiated by neutrons from an airport security facility or some other unaccounted-for source. This observation allowed us to use our background data obtained with Landauer detectors, in order to increase background statistics used for comparison of foreground #7 detector's results with the blank data.

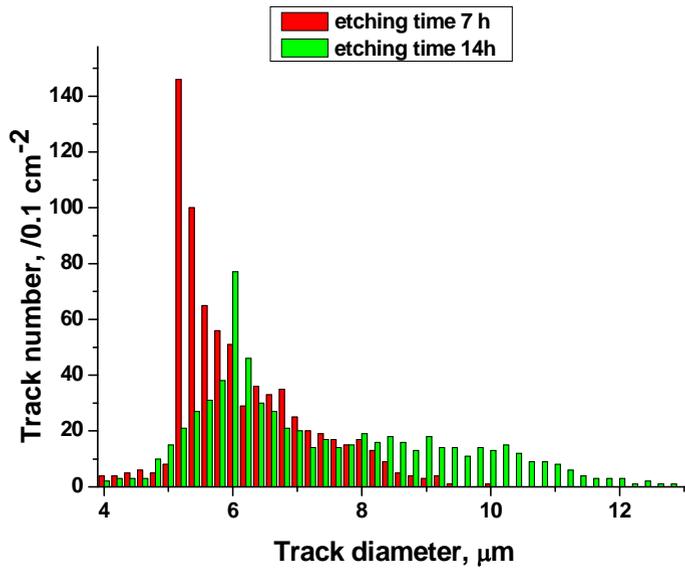
The summary result of the two blank detectors readings ( $S = 2 \text{ cm}^2$  of each side) is presented in Fig. 1. The result of 7 hr etch shows track density  $N = 6 \text{ cm}^{-2}$  in the track diameter range of interest, consistent with the proton recoil track diameters. The track density of 14 hr etch, which is within the  $4.5 - 12 \text{ }\mu\text{m}$  diameter range consistent with Cf-252 14 hr etch proton recoil, is higher ( $N = 26 \text{ cm}^{-2}$ ). This inconsistency between 7 and 14 hr backgrounds is caused by two factors: First, the increase of CR-39 efficiency with respect to fast neutrons (cosmic background) with removed depth in the range down to  $20 \text{ }\mu\text{m}$  from the surface. Second, the presence of other "old" tracks that were stored in the blank CR-39 since the time they were manufactured.



**Figure 1.** Typical background spectra of Landauer CR-39 detector (RadTrack) etched for 7 and 14 hr, respectively ( $S = 2 \text{ cm}^2$ ). The number of proton-like tracks resulting in background neutron irradiation is much smaller than that for SRI's #7 detector: At a 7 hr etch, the mean number of tracks within the range of  $4.5 - 8.0 \text{ }\mu\text{m}$  (consistent with Cf-252 proton recoil distribution) is only  $\langle N \rangle = 6 \text{ cm}^{-2}$ . At a 14 hr etch the number of tracks within the  $4.5 - 12 \text{ }\mu\text{m}$  diameter range (consistent with Cf-252 14 hr etch proton recoil) is  $\langle N \rangle \sim 26 \text{ cm}^{-2}$ . The tracks with diameter  $d \geq 8.0 \text{ }\mu\text{m}$  (7 hr etch) and  $d > 11 \text{ }\mu\text{m}$  (14 hr etch) represent a build

up of stored background alpha activity (mainly radon and triton series). Notice that the background distributions do not contain noticeable increase at 5.2 and/or 6.0  $\mu\text{m}$  track diameter.

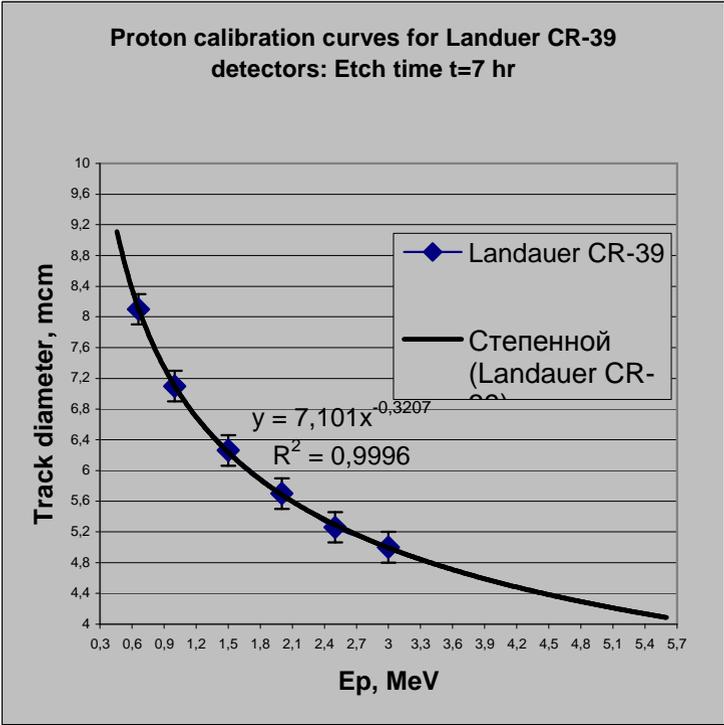
In contrast to the background, the Landauer CR-39 spectra representing neutron calibration with a Cf-252 neutron source (Fig. 2) contain high track density at its both sides. It was found that the number of tracks at the front CR-39 detector side (the front with respect to the neutron source) is about 20-30 % higher than that at the opposite one at any etching time (7-21 hr).



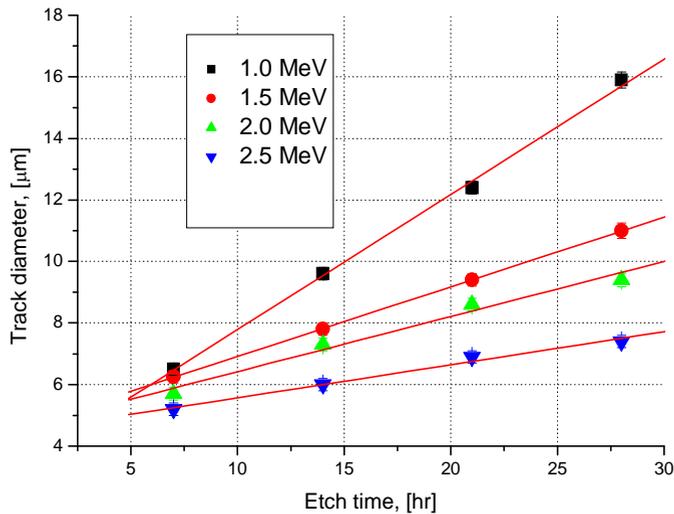
**Figure 2.** Cf-252 recoil proton spectra (Cf-252 neutron source with intensity  $I_n = (120 \pm 20)$  n/s in  $4\pi$ -solid angle. Total neutron fluence through the CR-39 chip is  $F_n \approx 7 \times 10^8$  n/cm<sup>2</sup>. The Landauer CR-39 chip with 200  $\mu\text{m}$  PE radiator has been used. After 7 hr etch in 6 N NaOH at  $T = 70^\circ\text{C}$ , the proton recoil spectrum is located between 4.5 and 9.0  $\mu\text{m}$  track diameter with maximum near 5.2  $\mu\text{m}$ . This maximum is consistent with the mean recoil proton energy in the range of 2.2-2.5 MeV (see Landauer's CR-39 proton calibration curve obtained with a Van de Graaf accelerator, Fig. 3a). The recoil spectrum of 14 hr etched CR-39 is broader than that for 7 hr etch and is located within the proton recoil track diameters ranged between 5.0 and 12.0  $\mu\text{m}$  with the maximum near 6.0  $\mu\text{m}$ . The shift in track diameters from 5.2 to 6.0  $\mu\text{m}$ , compared to 7 hr etch, is consistent with 2.2-2.5 MeV proton track diameter gain at 14 hr etching compared to 7 hr etch (Fig. 3b). Notice that for both etchings at  $t = 7$  and 14 hr about 30 % of all tracks have oblique incidence. The neutron detection self-efficiency of CR-39 at  $t = 14$  hr ( $\epsilon_n \sim 1.2 \times 10^{-4}$ ) is about a factor of 1.3 higher than that at  $t = 7$  hr ( $\epsilon_n \sim 0.9 \times 10^{-4}$ ) due to increase in proton recoil critical angles with the removed CR-39 depth.

The calibration curve allowing an estimate of proton energy (including also recoil protons) based on track diameters at etch time  $t = 7$  hr is depicted in Fig. 3a. The

dependences of track diameter versus etching time 7 - 28 hr of normal incidence protons in the energy range of 1.0 - 2.5 MeV are shown in Fig. 3b.

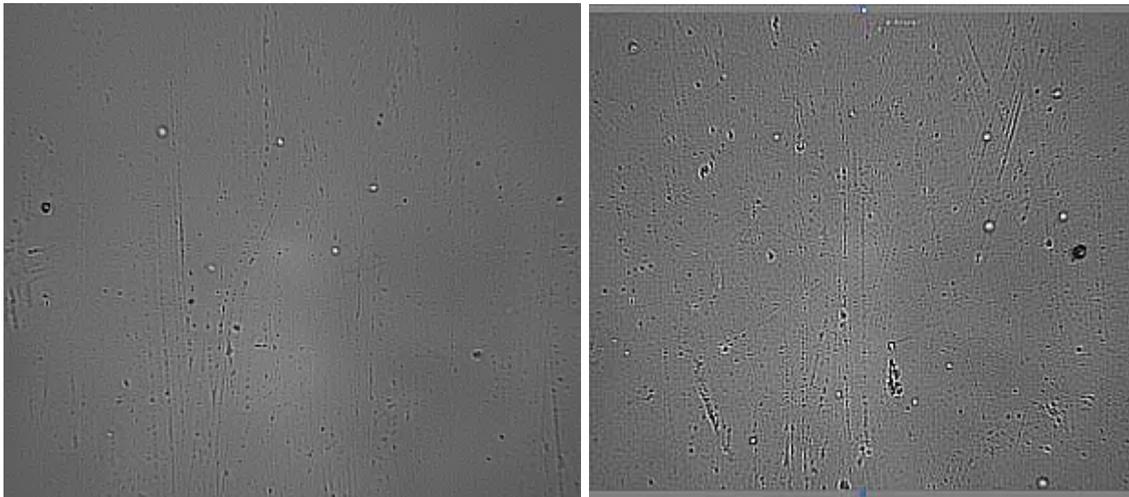


**Figure 3a.** Van de Graaf accelerator calibration curve for the 0.65-3.0 MeV proton beam of normal incidence with respect to the detector surface. The targets are Landauer RadTrack and another Fukuvi-type CR-39 detector. Etching conditions: 6 N NaOH, T = 70°C, t = 7 hr.



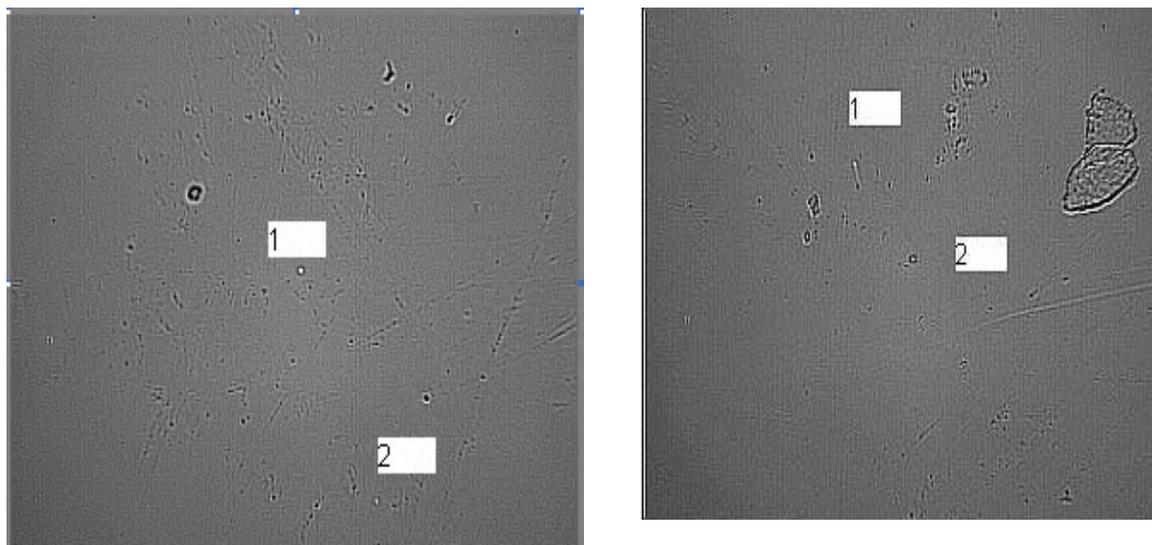
**Figure 3 b.** Track diameter vs. etching time (removed CR-39 depth  $h = 9.2 - 46 \mu\text{m}$ ) for protons of normal incidence in the energy range of 1.0 - 2.5 MeV. Note a good linear dependence for Landauer RadTrack detectors. Etching conditions: 6 N NaOH at  $T = 70^\circ\text{C}$ . The bulk etch rate  $v_b$  is derived from empirical equation:  $v_b = 1.275 \exp[0.828 C + 0.049T - 0.002CT - 17.624]$ , where  $T$  and  $C$  are temperature (in K) and NaOH molarity (mol/l), respectively (D. Nikezic, K.N. Yu, Mat Sci. Eng., R46, 51 (2004)).

In Fig. 4 examples of proton recoil tracks obtained in CR-39 calibration are shown. The proton recoil tracks are represented by bright pits of 5-8  $\mu\text{m}$  in diameter on dark background.



**Figure 4.** Examples of proton recoil tracks of 5-8  $\mu\text{m}$  diameter (bright pits only - all others are surface defects!) for Cf-252 neutron source (7 hr etch, removed depth is  $h = 9.2 \mu\text{m}$ ). Total neutron fluence is  $F_n = 7 \times 10^8 \text{ n/cm}^2$ ; the size of each image is  $S = 120 \times 90 \mu\text{m}^2$ .

The reading of both sides of #7 foreground detector (the front and the opposite one) showed that it contains real nuclear (proton recoil) tracks of both normal and oblique incidence similar to that obtained with the calibration CR-39 detector irradiated with the Cf-252 neutron source. Three examples of such tracks are presented in Figs. 5a and 5b.

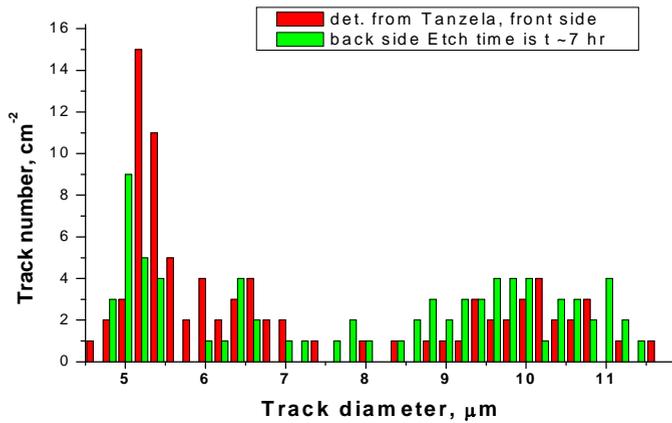


(a)

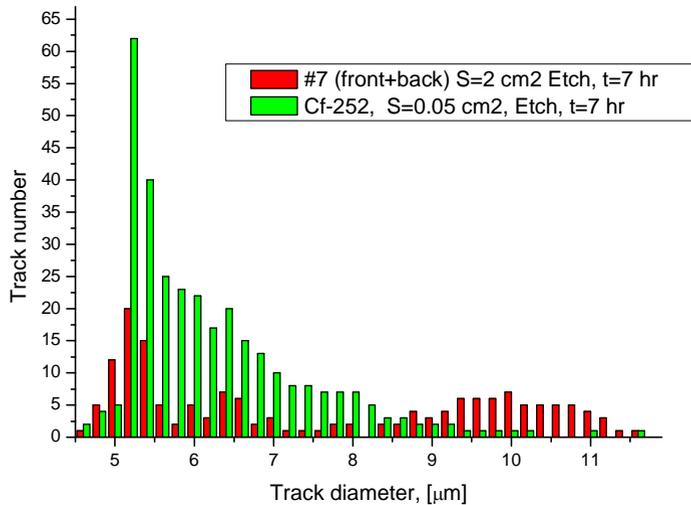
(b)

**Figure 5.** Examples of proton recoil tracks (compare with Cf-252 bright pits) of 5-7  $\mu\text{m}$  diameter from CR-39 foreground detector #7 (removed depth is  $h \approx 8.6 \mu\text{m}$ ), images size is  $S= 120 \times 90 \mu\text{m}^2$ : (a) - two tracks of normal incidence (1,2); (b) – track of oblique incidence (1), track of normal incidence (2).

The total track diameter distribution for the #7 foreground detector is shown in Fig. 6.

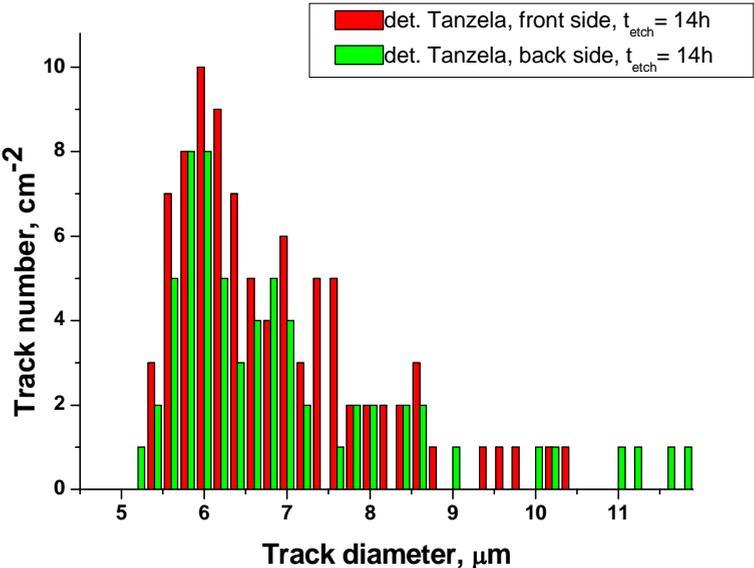


**Figure 6.** Track number distribution vs. track diameter for F. Tanzella's (SRI) # 7 CR-39 detector from both front and back sides. Etching time is 7 hr. The spectra contain real nuclear tracks. From the front side: the total number of circular and elliptic tracks (normal and oblique incidence) is  $N = 77 \text{ cm}^{-2}$ . About 30% of all tracks have elliptic shape. Similarly, from the back side:  $N = 40 \text{ cm}^{-2}$ . Notice that the number of proton recoil tracks at the front CR-39 detector side is higher than the back side.

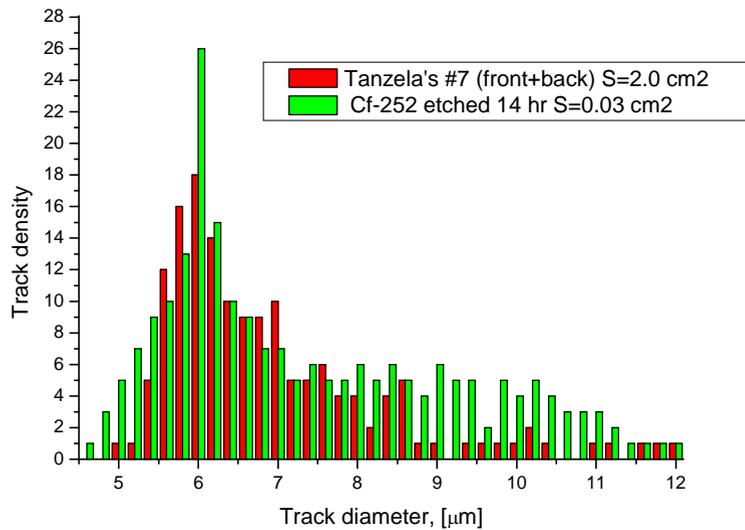


**Figure 7.** Comparison of CR-39 Cf-252 proton recoil spectrum corresponding to 7 hr etching with the sum of spectra obtained from both sides of #7 CR-39. In the track diameter range of 4.5-8.0  $\mu\text{m}$  the #7 spectrum look similarly to Cf-252 recoil. Note, that the maximum of track diameter distribution for the #7 detector is localized near the 5.2  $\mu\text{m}$  value.

In order to confirm our finding concerning the observation of proton-recoil tracks at the 7 hr etched CR-39 we continued in-depth etching adding 7 more hr in the same conditions. The track diameter distributions are shown in Figs 8 and 9.

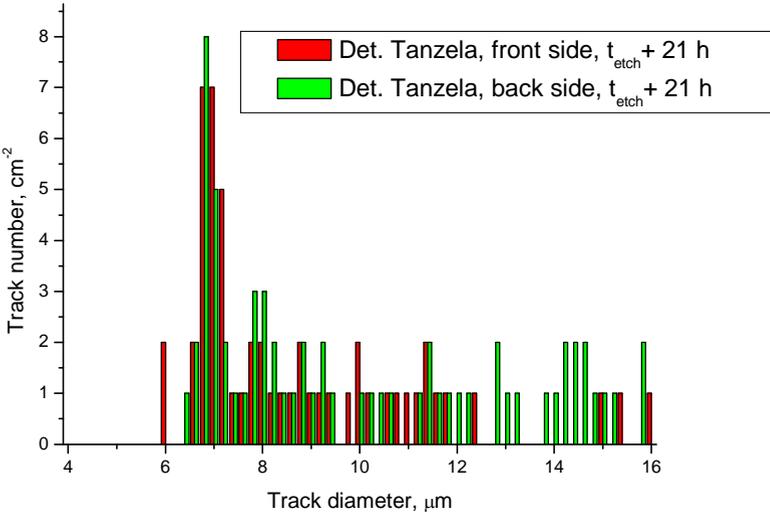


**Figure 8.** Track number distribution vs. track diameter for Tanzella's #7 CR-39 detector from both front and back sides. Etching time is 14 hr. The spectra contain real nuclear tracks. From the front side: the total number of circular and elliptic tracks (the normal and the oblique incidence) in the range of 4.5 - 11.0 μm is  $N = 101 \text{ cm}^{-2}$ . About 20% of all tracks have elliptic shape. Similarly, at the back side:  $N=65 \text{ cm}^{-2}$ . Notice again that the number of proton recoil tracks at the front CR-39 detector side is higher than the back side.

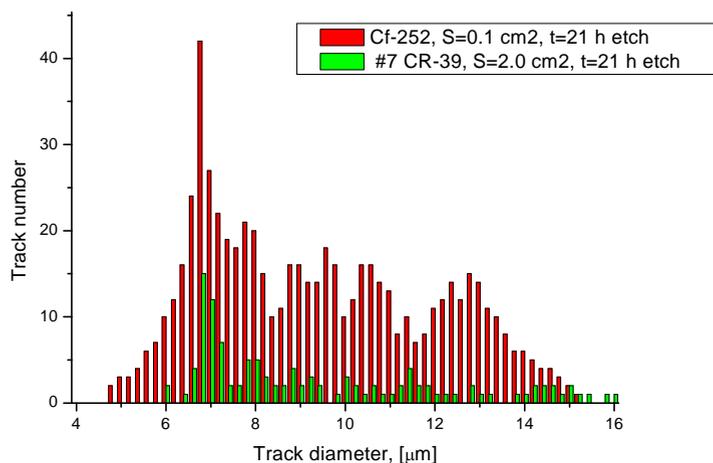


**Figure 9.** Comparison of CR-39 Cf-252 proton recoil spectrum corresponding to 14 hr etching time with the sum of spectra obtained from both the front and back sides of #7 CR-39. In the track diameter range of 4.5-11.0  $\mu\text{m}$  the #7 spectrum looks almost identical to the Cf-252 recoil. Note that the maximum in the #7 detector is localized near 6.0  $\mu\text{m}$  track diameter, as it is for the Cf-252 recoil spectrum. The total number of proton-like tracks at 14 hr etch is about a factor of 1.4 higher than that for 7 hr etch.

To further confirm the neutron emission effect, we carried out one more etching of #7 detector (total etch time  $t_{\text{et}} = 21$  hr, corresponding to total removed depth  $h = 27.2 \mu\text{m}$ ). The result of track diameter distribution for both the front and back sides of the #7 detector at  $t_{\text{et}} = 21$  hr is shown in Fig. 8.

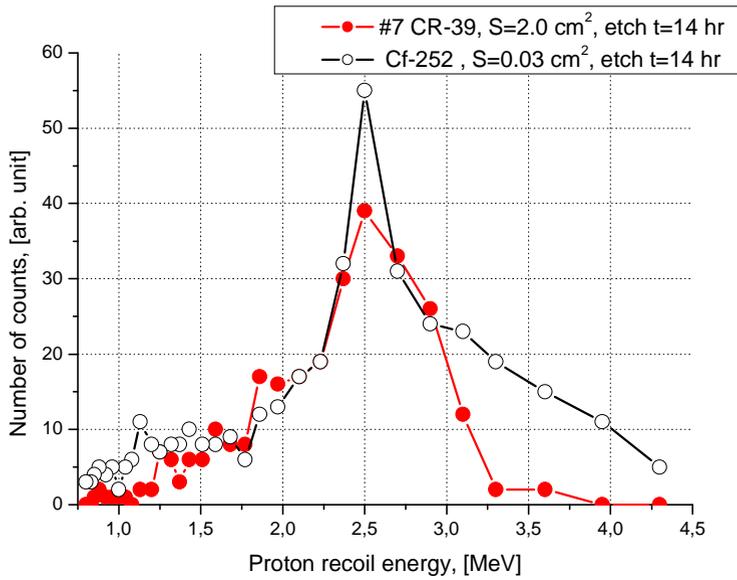


**Figure 10.** Normal incidence track distribution vs. their diameter for Tanzella’s # 7 CR-39 detector from both front and back sides. Etching time is 21 hr. The spectra contain real nuclear tracks. From the front side: the total track number (both normal and oblique incidence) in the range of 5-14.0  $\mu\text{m}$  (the diameters consistent with Cf-252 spectrum at  $t_{\text{et}} = 21$  hr) is  $N=91 \text{ cm}^{-2}$ , about 30 % of all tracks have elliptic shape. At the back side:  $N=67 \text{ cm}^{-2}$ . Notice again that the number of proton recoil tracks at the front CR-39 detector side is higher than the back side.

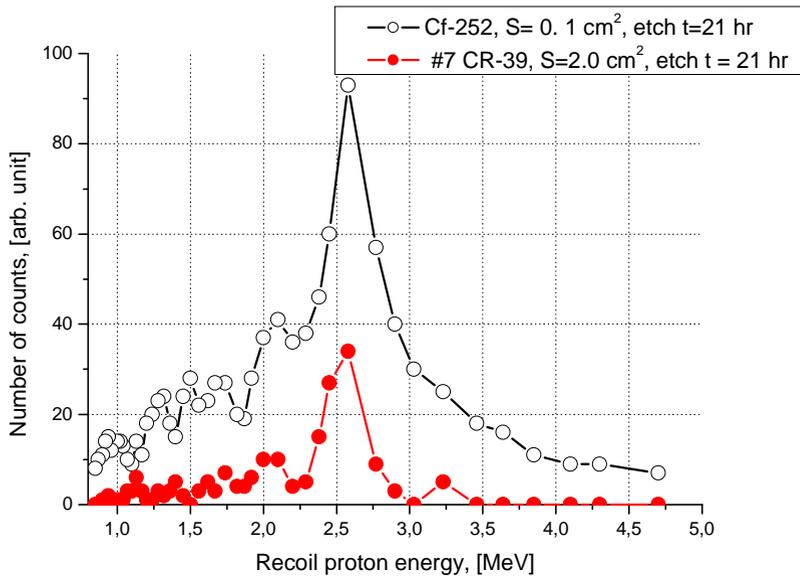


**Figure 11.** Comparison of CR-39 Cf-252 proton recoil spectrum corresponding to 21 hr etch time with the sum of spectra obtained from the both sides (front and back) of #7 CR-39. In the track diameter range of 5 - 14.0  $\mu\text{m}$  the #7 spectrum looks similar to the Cf-252 recoil. Note that the maximum in the #7 detector is localized near 7.0  $\mu\text{m}$  track diameter, as it is for Cf-252 recoil spectrum. This position is consistent with the track diameter for 2.5 MeV proton etched during  $t_{\text{et}} = 21$  hr (Fig. 3b).

Thus, all three etchings of #7 detector at  $t = 7, 14$  and 21 hr show track distributions similar to that obtained by exposure of the calibration detector to a Cf-252 fast neutron source. The position of maximum for track diameter distribution follows to that of Cf-252 source and is consistent with the 2.2-2.5 MeV proton track diameter vs. etching time (Figs. 12a, b). In summary, presented experimental evidence can be considered as strong, unambiguous proof that the #7 detector was exposed to fast neutrons (2.5 MeV).



**Figure 12a.** Rough reconstruction of the protons recoil spectra for CR-39 Landauer detectors obtained during electrolysis run (detector #7) and during exposure with Cf-252 neutron source. Etch time is  $t = 14$  hr. The reconstruction of the spectra was deduced from the track density vs. track diameter histograms, taking into account two functions: (a) track diameter vs. proton energies at  $t = 14$  hr, and (b) the critical angle  $\theta_c$  vs. proton energy.



**Figure 12, b.** Same as Figure a, but at etch time  $t = 21$  hr.

As seen in Fig. 12, the proton recoil spectra of #7 detector are narrower than that of Cf-252 at 14 and 21 hr etch. They contain almost no counts above 3.0 MeV and the peak near 2.5 MeV. In contrast, the Cf-252 spectra look broader due to the presence of high energy neutron/recoil component (up to 12 MeV) and also demonstrate a peak near 2-2.5 MeV.

These comparative results allow us to conclude that fast monoenergetic neutrons with energy close to  $E = 2.5$  MeV were emitted during the electrolysis runs. For a more accurate numerical estimate of the recoil spectra, a Monte-Carlo calculation must be performed.

## II. Calculation of the neutron emission rate

1. For etch time  $t_{et} = 7$  hr (removed depth is the  $h = 8.7\mu\text{m}$ )

The total (normal and oblique incidence) foreground track density in the range of proton recoil diameters ( $4.5 < d < 7.8 \mu\text{m}$ ) is:  $N(\text{fg})_1 = 77 \text{ cm}^{-2}$  on the front side; and  $N(\text{fg})_2 = 40 \text{ cm}^{-2}$  on the back side. Average track density is  $\langle N(\text{fg}) \rangle = 58.5 \text{ cm}^{-2}$ .

The total background track density on both sides of blank detector ( $S = 0.25 \text{ cm}^2$  each) is the  $\langle N(\text{bg}) \rangle = 6.0 \text{ cm}^{-2}$ . This is consistent with the background usually observed in fresh Landauer chips. This consistency allowed us to use our background data (Fig. 1) representing the result of large area ( $S=2.0 \text{ cm}^2$ ) reading of two Landauer background detectors.

Thus, the “effect” with the background subtracted is  $\langle \Delta N \rangle = \langle N(\text{fg}) \rangle - \langle N(\text{bg}) \rangle = 52.5 \pm 8.0 \text{ track/cm}^2$ .

According to calibration measurements, the CR-39 self-efficiency ( $t_{et} = 7$  hr) with regard to Cf-252 neutrons was found to be  $\epsilon_s = 9.0 \times 10^{-5}$ . Assuming a “neutron source” was a cathode wire in a  $2\pi$ -geometry with respect to the CR-39 surface, the neutron count rate/intensity ( $I_n$ ) would be derived as:

$$I_n = 2\langle \Delta N \rangle / (t \times \epsilon_s), \text{ where } t \text{ is the electrolysis (foreground) duration.}$$

Assuming that the neutron emission effect occurred during the time when electrolysis current ( $j > 0.5 \text{ mA}$ ) was turned on ( $t = 15$  days), the  $I_n = 0.90 \pm 0.14 \text{ n/s}$  in  $2\pi$  solid angle. This is the lowest rate estimate. If we assume that the neutron emission was observed only during the time interval  $\Delta t = 4$  days when neutron dosimeter showed a count rate above the background, the  $I_n = 3.38 \pm 0.53 \text{ n/s}$  in  $2\pi$  solid angle — the highest rate estimate. So, the neutron emission rate in run #7 can be estimated in the range of 1.0 - 3.0 n/s.

2. For etch time  $t_{\text{et}} = 14$  hr (removed depth is the  $h = 18 \mu\text{m}$ )

Similarly to (1), the total (normal and oblique incidence) foreground track density in the range of proton recoil diameters for 14 hr etch ( $4.5 < d < 11.0 \mu\text{m}$ ) is:  $N(\text{fg})_1 = 110 \text{ cm}^{-2}$  on the front side; and  $N(\text{fg})_2 = 66 \text{ cm}^{-2}$  on the back side. Average track density is  $\langle N(\text{fg}) \rangle = 88 \text{ cm}^{-2}$ .

The background track density on both sides of blank detector ( $S = 0.25 \text{ cm}^2$  each) is the  $\langle N(\text{bg}) \rangle = 26 \text{ cm}^{-2}$  (data from Fig. 1)

The “effect” with the background subtracted is  $\langle \Delta N \rangle = \langle N(\text{fg}) \rangle - \langle N(\text{bg}) \rangle = 62.0 \pm 10.7 \text{ track/cm}^2$ .

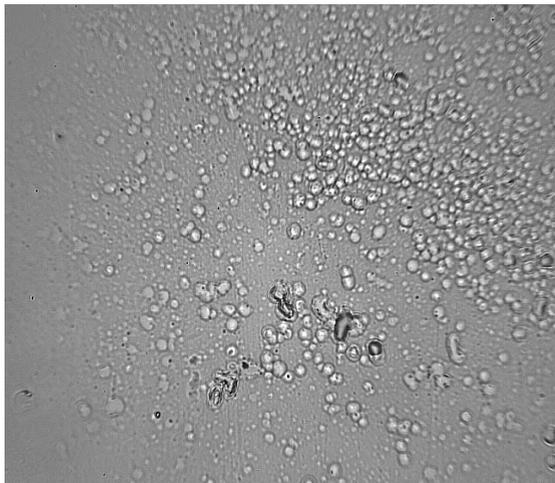
Accordingly to calibration measurements CR-39 at etch time  $t_{\text{et}} = 14$  hr the self-efficiency was found to be  $\epsilon_s = 1.17 \times 10^{-4}$ .

For  $t = 15$  days:  $I_n = 2\langle \Delta N \rangle / (t \times \epsilon_s) = 0.82 \pm 0.14 \text{ n/s}$  in  $2\pi$  solid angle and for  $\Delta t = 4$  days  $I_n = 3.08 \pm 0.53 \text{ n/s}$ . Thus, the result for the 14 hr etch gives approximately the same neutron emission intensity range, within a standard deviation, as that for a 7 hr.

The entire dataset for the #7 CR-39 detector — including track readings with three removed depths (8.7, 18 and 27  $\mu\text{m}$ ); the comparison of foreground #7 track densities and distributions of their diameters with similar parameters of background and neutron calibration detectors; as well as CR-39 efficiency estimate with respect to Cf-252 neutrons — presents strong, unambiguous evidence for fast neutron emission, with neutron energy in the range of  $E_n \sim 2.2 - 2.5 \text{ MeV}$ , at the rate  $I_n$  of  $\sim 1 - 3 \text{ n/s}$  during electrolysis run #7 performed at SRI.

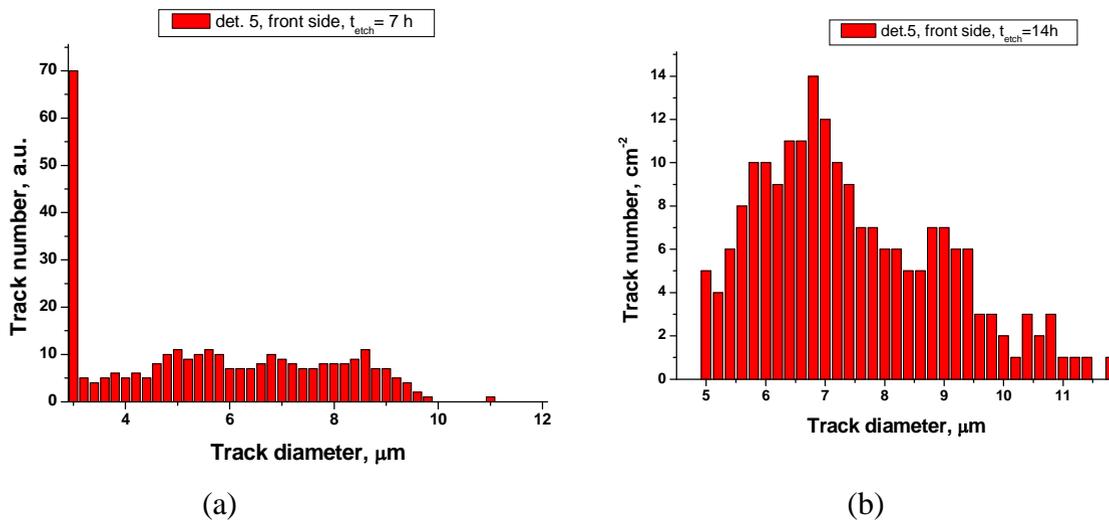
### III. Summary of the #5 detector results (SRI run #BE010-5)

- The #5 CR-39 detector used in SRI #BE010-5 PdDx deposition electrolysis experiment had a 60  $\mu\text{m}$  polyethylene film adhered to both faces while immersed in the electrolyte and in contact with the cathode.
- This detector showed confusing results. 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 the #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).



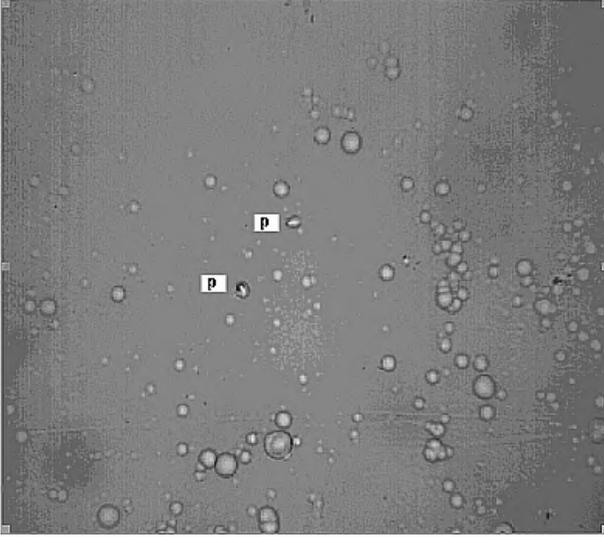
**Figure 13.** Typical image of the front side of the #5 detector at etch time  $t = 7$  hr.

The pit density distributions versus pit diameters at the front side of the #5 detector at 7 and 14 hr etch are very similar to that of a defective CR-39 surface observed during the analysis of another CR-39 sample: the #2 detector from W. Williams. See our paper in these Proceedings. [4]



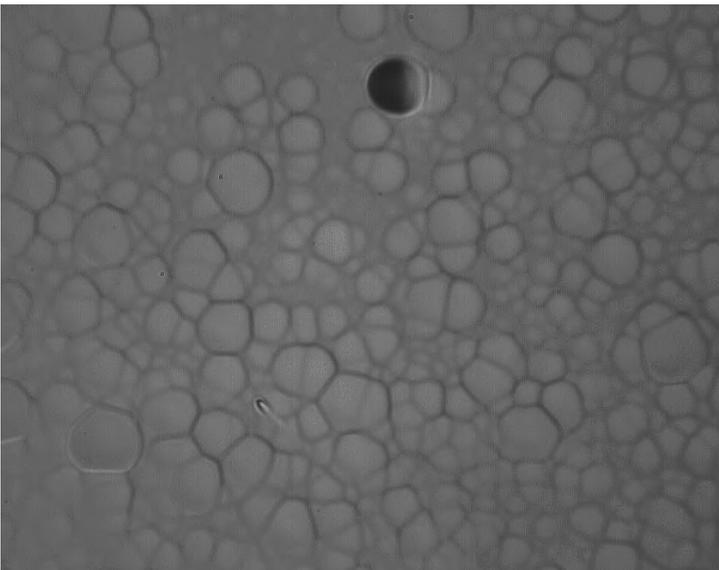
**Figure 14 (a,b)** Individual pit density at the front side of #5 detector: 7 hr (left) and 14 hr (right) etch.

With etching in depth during an extra 7 hr, the real tracks, in particular tracks from the proton recoil caused by fast neutrons, started to appear on the front side of #5 detector, against the defect background (Fig. 15).



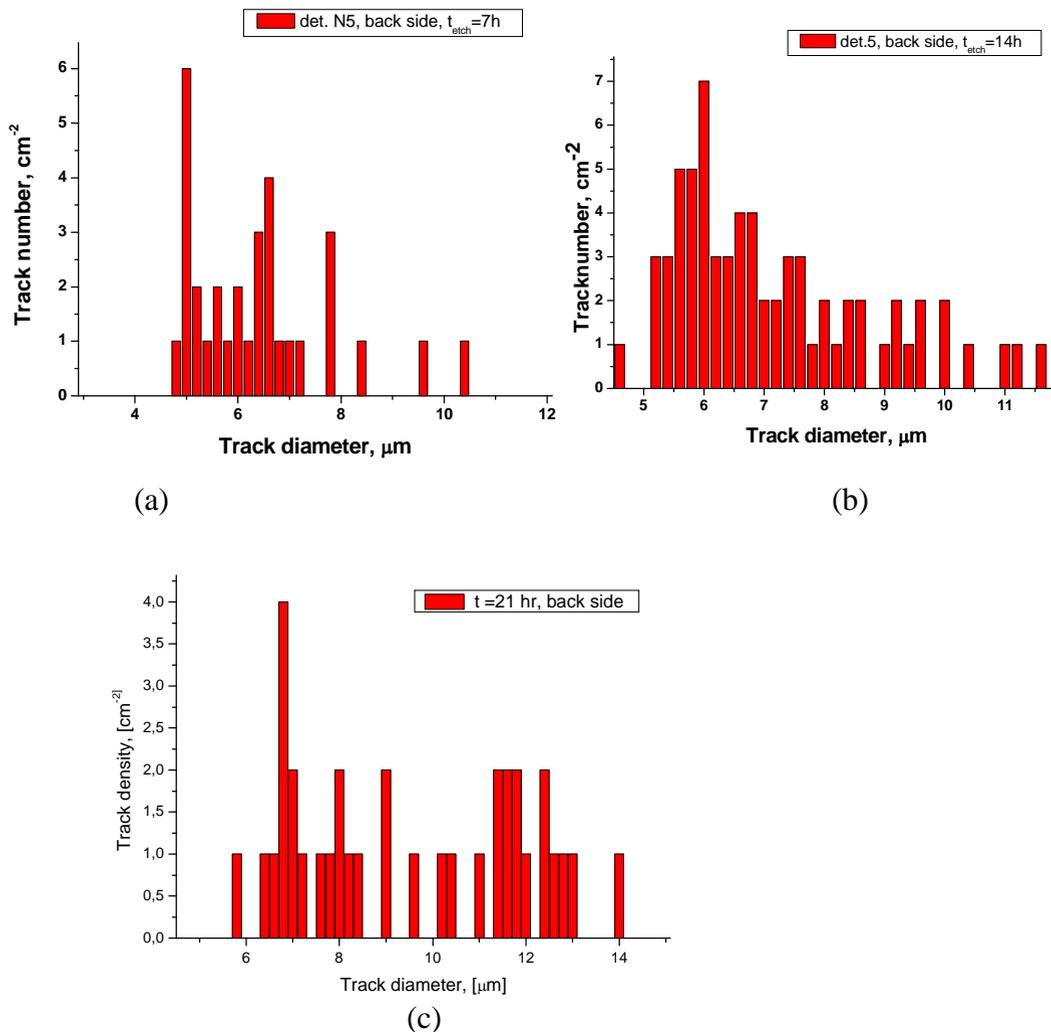
**Figure 15.** Proton recoil tracks at the front side of #5 detector well distinguished among the defects (so-called “ground beef”) background at  $t = 14$  hr etch.

After total etching of  $t = 21$  hr, the defects at the front surface of #5 detector lose contrast and assume a non-circular shape, while nuclear tracks (alphas and protons) become sharply distinguished compared to this low contrast background.



**Figure 16.** Typical image of the front side of #5 detector after total 21 hr etch (real nuclear tracks in the background of the “ground beef”). The bright oval track at lower part of the figure was made by a proton with oblique incidence. The dark circle at the upper part of the figure was made by an  $\alpha$ -particle.

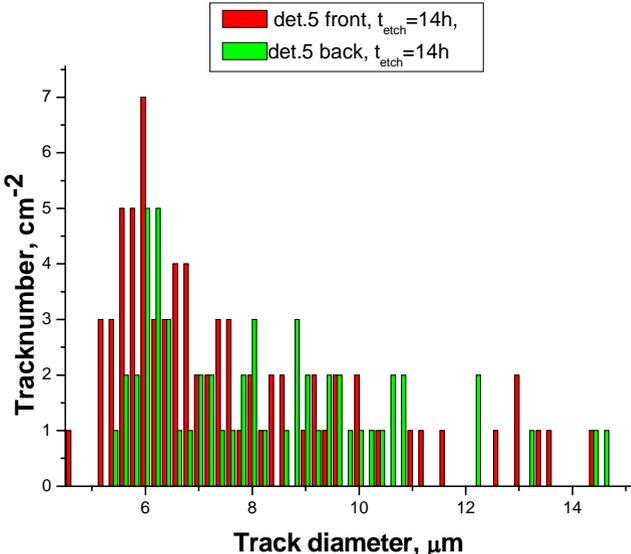
The back side of the #5 detector does not contain defect areas (which are called “ground beef” in CR-39 jargon). The side shows proton recoil tracks similar to those found at the both faces of the # 7 CR-39 detector analyzed above. The proton recoil distribution on the back side of #5 detector are presented in Figs. 17a, b and c.



**Figure 17(a-c).** Proton recoil tracks from the “clean” back side of #5 detector: (a) 7 hr etch; (b) 14 hr etch; (c) 21 hr etch.

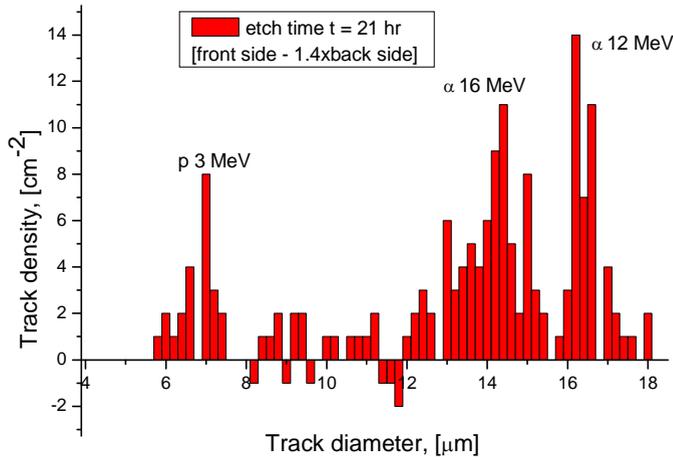
A comparison of the nuclear tracks obtained both at the front and at the back side of the #5 detector (at  $t = 14$  hr etch) show the remarkable similarity of their distributions versus

track diameter (Fig. 18). This suggests that the #5 detector has actually been exposed to a low intensity fast neutron flux.



**Figure 18.** Comparison of the front and the back sides of the proton recoil spectra of the #5 detector at t = 14 hr etch.

During in-depth etching at t = 21 hr (when most of defect pits already loose their contrast) it was found that the number of nuclear tracks at the front side of #5 detector far exceeded the number of proton recoil tracks on the clean back side of the detector. To obtain a clue about the nature of those excess tracks (besides proton recoil), we have subtracted the back side track distribution vs. their diameters from that of the front side of the same detector (Fig. 19).



**Figure 19.** The front side spectrum of nuclear tracks of #5 detector subtracting the neutron induced proton recoil spectrum from its back side, at  $t = 21$  hr etch.

As seen from Fig. 19, after subtracting the neutron proton recoil, the front side nuclear track spectrum still shows a two-band structure consisting of the 3 MeV DD-protons and high energy alphas [3]. One possible way these nuclear signatures may have originated could be the charged particle emissions from PdDx powder deposited on the PE protective sheet of the CR-39 in the area where cathode is in contact with the cathode. Indeed, a metallic coating on top of PE film covering the front side of the #5 detector has been found.

The rate of possible neutron emission that was detected by #5 CR-39 (after 7 and 14 hr etch) in electrolysis run # BE010-5 (for  $t = 20$  days) is presented below ( $\epsilon_s$  – is the self-efficiency of the CR-39 with regards to Cf-252 neutrons):

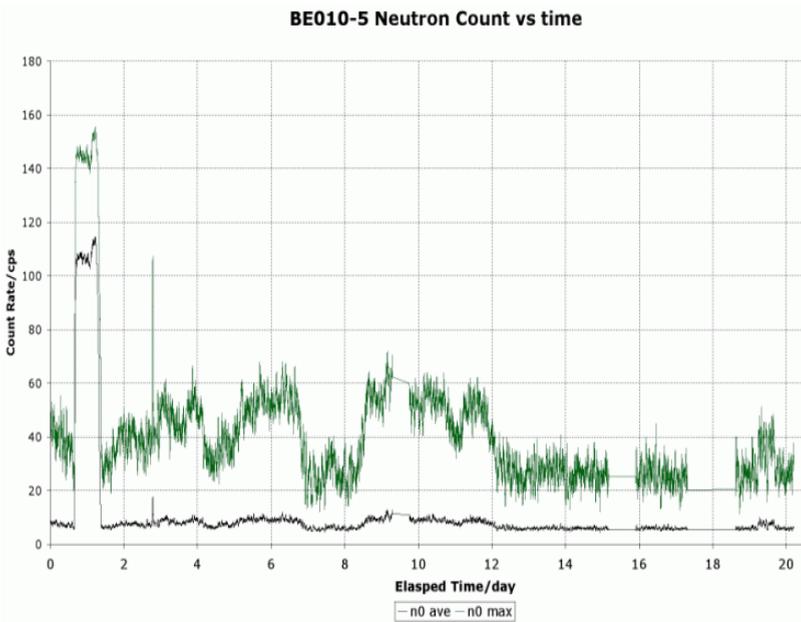
#### **t ~ 7 hr etch**

- only on back side:  $N(\text{fg}) = 30.0 \pm 5.48$  recoil protons/cm<sup>2</sup>
- $N(\text{Bg}) = 6 \pm 4$  cm<sup>-2</sup>
- $\Delta N = 24.0 \pm 6.8$  p/cm<sup>2</sup>
- $\langle I_n \rangle = 2\langle \Delta N \rangle / (t\epsilon_s) = 48 / (1.73 \times 10^6 \times 9.2 \times 10^{-5}) = 0.30 \pm 0.08$  n/s in  $2\pi$  solid angle

#### **t = 14 hr etch**

- back:  $N(\text{Fg}) = 45$  cm<sup>-2</sup>, front  $N(\text{Fg}) = 63$  cm<sup>-2</sup>  $\langle N(\text{fg}) \rangle = 54.0 \pm 7.3$  cm<sup>-2</sup>
- Background  $\langle N(\text{bg}) \rangle = 26 \pm 5.1$  cm<sup>-2</sup>
- $\Delta N = 28.0 \pm 8.9$  cm<sup>-2</sup>
- $\langle I_n \rangle = 2\langle \Delta N \rangle / (t\epsilon_s) = 56 / (1.73 \times 10^6 \times 1.2 \times 10^{-4}) = 0.29 \pm 0.09$  n/s in  $2\pi$  solid angle
- If  $t = 1$  day  $I_n = 6.0 \pm 1.6$  n/s in  $2\pi$  solid angle.

In Fig. 20 the results of neutron measurements carried out in SRI with the BF<sub>3</sub> sphere (a simple neutron dosimeter) during run #BE010-5 are presented.



**Figure 20.** Neutron protocols of the run #5 obtained in SRI with the BF<sub>3</sub> detector.

As seen in Fig. 20, the neutron count rate in this run according to the BF<sub>3</sub> measurements was above background for 20 days, and showed a maximum in the first day of electrolysis. The neutron count rate in the run #BE-013-7, when the CR-39 detector #7 was exposed, showed similar characteristics. The analysis of the SRI data and their comparison with the CR-39 results is below.

### Sensitivity to neutrons of SRI's BF<sub>3</sub> sphere and the CR-39 neutron results

- At self-efficiency to fast neutrons  $\epsilon_s = 7.6 \times 10^{-3}$  ( $R \sim 0$  cm) and distance between the detector and the hypothetical neutron source (the cathode wire)  $R = 10$  cm, the total efficiency of the BF<sub>3</sub> sphere with respect to fast neutrons would be  $\epsilon_t = 7.6 \times 10^{-5}$ .
- The sensitivity of the detector to fast neutrons — reflecting the minimal neutron emission rate that can be distinguished from the background and equal to, at least, 3 standard deviations of the background count rate — can be expressed as:  $S =$

$3[\langle N_b \rangle / (\epsilon_t^2 \tau)]^{1/2}$ , where  $\langle N_b \rangle$  is the averaged count rate in the background measurement and  $\tau$  is the duration of neutron detection.

- In the case of experiment #7:  $\langle N_b \rangle \approx 6.0$  cps,  $t = 15$  days, resulting in  $S \approx 150$  n/s, (300 n/s, assuming neutron emission continued for 4 days) showing the figure that is a factor 100 higher than the fast neutron emission rate resulting in recoil protons detected by CR-39 detector during the #7 run.
- In the case of experiment #5: for 20 days of exposure,  $S \approx 130$  n/s (a factor of 400 higher than CR-39 showed). If we assume that neutrons were emitted only in the one day (according to n-count peak), then the sensitivity would be  $S \sim 600$  n/s, which is also  $\sim 100$  times higher than #5 CR-39 shows.

## IV Conclusions

- The entire set of results from the analysis of two CR-39 detectors show that a weak but statistically significant emission of fast neutrons has been observed in SRI's #7 and #5 runs replicating SPWAR Pd-deposition experiment.
- The #7 detector, which was protected by 6  $\mu\text{m}$  Mylar film, shows "clean" front and back faces, containing only nuclear tracks (proton recoil + background radionuclides).
- The #5 detector, which was protected by 60  $\mu\text{m}$  PE film, shows mixed zones of defects ("ground beef") and nuclear tracks at its front side, and proton recoil density lower than #7 on the back side. The small diameter defect pits can be eliminated by in-depth etching (removed depth  $h > 18$   $\mu\text{m}$ ) allowing us to distinguish real nuclear tracks of proton recoil, caused by neutrons as well as by energetic charged particles (protons and alphas) emitted from the PdDx powder being deposited on top of the detector during electrolysis.
- A comparison of proton recoil spectra (track number versus their diameter) of the analyzed detectors with that of the background runs and tracks of Cf-252 calibration gives preliminary evidence for fast neutron emission in the runs #7 and #5.
- A comparison of the neutron emission rates obtained from CR-39 analysis with that deduced from SRI's proportional BF3 detector shows a huge discrepancy of results, suggesting orders of magnitude higher neutron emission was recorded by the SRI detector than we calculated from the noiseless CR-39 measurement data.
- Due to the low sensitivity to neutrons of the SRI detector, and the absence of pulse-height/pulse shape analysis, we assume that the signal of the BF3 sphere contains a significant electromagnetic noise fraction, induced by the electrolysis power supply.
- In order to provide confirmation of our CR-39 results on neutron emission in SRI experiments, additional high efficiency measurements with a more sophisticated type of neutron detector would be desirable.

## Acknowledgements

This work was supported by New Energy Institute. We also personally thank S. Krivit, E. Greenspan, and L. Forsley for arranging this work as well as for valuable discussions.

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