## June 8, 2007

## Analysis of Fran Tanzella's #7 CR-39 detector (Final Report + Appendix)

Andrei Lipson, Alexei Roussetski

### *I. CR-39 Reading*

During April – May , 2007 we carried out reading of the #7 Tanzella's Landauer CR-39 detector at three different removed depths (three various etching time, roughly corresponding to 7, 14 and 21 hr etch in 6M-NaOH at t = 70 °C at $v_b \approx 1.3$ μm/hr) and compare the results with that of blank CR-39. The reading was performed manually using track reading facility "PAVICOM" in Lebedev Physics Institute, Russian Academy of Sciences, Moscow, Russia. The #7 detector was initially covered with 6 μm mylar filter preventing the CR-39 surface from mechanical stress and electrostatic (spark discharge) effect induced by the cathode wire during electrolysis. We carried out the #7 detector (and blank one) reading from its both sides, including the front face (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 $cm^2$ 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 cm2. It was found that track density of the blank detector in the track diameter range of interest (4.0< d< 8.0 μm, t = 7 hr etch) contain in total 3 track/0.5 $cm^2$ (from both sides). This number is typical for the blank Landauer RadTrack detectors (N = 6±4 track/$cm^2$ as a result of more than 100 our measurements), which is why we concluded that the blank detector has not been irradiated by neutrons (in airport security facility). 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 two blank detectors reading (S=2 $cm^2$ of each side) is presented in Fig.1.The result of 7 hr etch shows track density N = 6 $cm^{-2}$ in the track diameter range of interest (consistent with the proton recoil track diameters). The track density of 14 etch (within the 4.5 – 12 μm diameter range consistent with Cf-252 14 hr etch proton recoil) is higher (N = 26 $cm^{-2}$). This inconsistency between 7 and 14 hr backgrounds is caused by two reasons. First is the increase of CR-39 efficiency with respect to fast neutrons (cosmic background) with removed depth in the range down to 20 μm from the surface. The second reflects presence of other "old" tracks that were stored in blank CR-39 since their manufacturing.

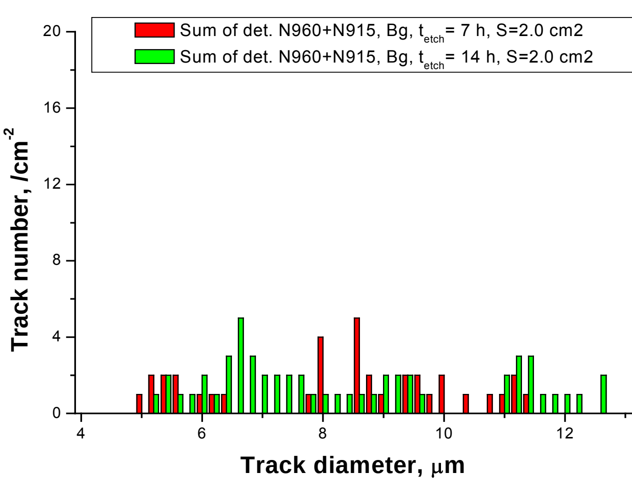


**Figure 1** Typical Background spectra of Landauer CR-39 detector (RadTrack) etched during 7 and 14 hr, respectively (S = 2 $cm^2$). Number of proton-like tracks (resulting in background neutron irradiation) is much smaller than that for Tanzella's #7 detector: At 7hr etch the mean number of tracks within the range of 4.5 – 8.0 μm (consistent with Cf-252 proton recoil distribution) is only <N> = 6 $cm^{-2}$. At 14 hr etch the number of tracks within the 4.5 – 12 μm diameter range (consistent with Cf-252 14 hr etch proton recoil) is <N> ~ 26 $cm^{-2}$. The tracks with diameter d ≥ 8.0μm (7hr etch) and d > 11μm (14 hr etch) represent a build up of stored Background alpha activity (mainly radon and toron series). Notice that the Background distributions do not contain noticeable increase at 5.2 and/or 6.0 μm track diameter.

In contrast to the Background, the Landauer CR-39 spectra representing neutron calibration with 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 (with respect to the neutron source) CR-39 detector side is about 20-30 % higher than that at the opposite one at any etching time (7-21 hr).

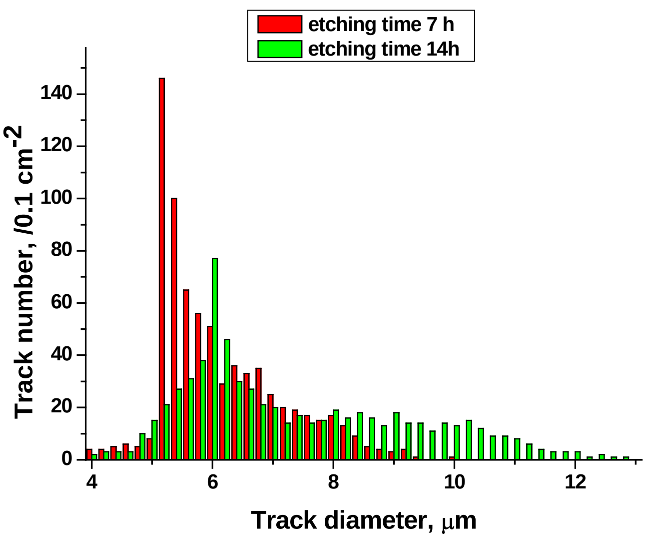



**Figure 2** Cf-252 recoil proton spectra ( Cf -252 neutron source with intensity $I_n$ = (120±20) n/s in 4π-solid angle. Total neutron fluence through the CR-39 chip is $F_n \approx 7\times10^8$ n/$cm^2$. The Landauer CR-39 chip with 200 μm PE radiator has been used. After 7 hr etch in 6N NaOH at t = 70 C the proton recoil spectrum is located between 4.5 and 9.0 μm track diameter with maximum near 5.2 μm. 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 Van DeGraaf accelerator, Fig. 3a). The recoil spectrum of 14 hr etched CR-39 a is broader than that for 7 hr etch and is located within the proton recoil track diameters ranged between 5.0 and 12.0 μm with the maximum near 6.0 μm (Shift in track diameters from 5.2 to 6.0 μm, 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. 3 b). 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 ($\varepsilon_n \sim 1.2\text{x}10^{-4}$) is about factor of 1.3 higher than that at t = 7 hr($\varepsilon_n \sim 0.9\text{x}10^{-4}$) due to increase in proton recoil critical angles with the removed CR-39 depth.

The calibration curve allowing proton (including also recoil protons) energy estimate via track diameters at etch time t = 7 hr is depicted in Fig. 3a. The dependences of track diameter vs.

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

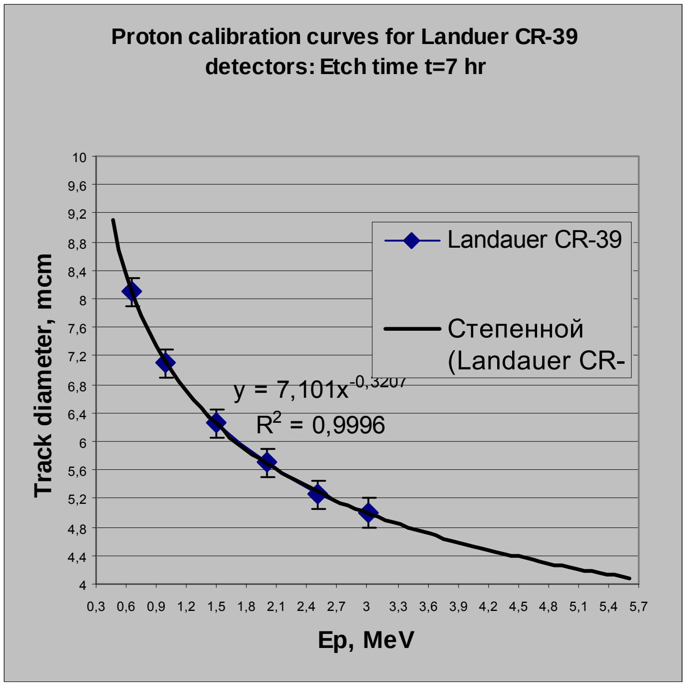



**Figure 3a** Van DeGraaf accelerator calibration curve for the 0.65-3.0 MeV proton beam of normal incidence with respect to the detector surface. The targets are Landauer RadTrack and another Fukuvi –type CR-39 detectors. Etching conditions 6N-NaOH, t = 70 °C, for 7 hr.

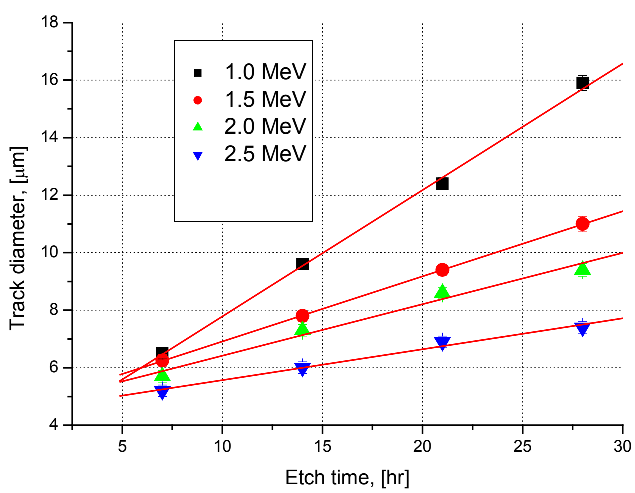



**Figure 3 b** Track diameter vs. etching time (removed CR-39 depth h = 9.2 – 46 µm) for protons of normal incidence in the energy range of 1.0 -2.5 MeV. Note a good linear dependence for Landauer RadTrack detectors. Etching conditions: 6N-NaOH at t= 70 C. The bulk etch rate $v_b$ is derived from empirical 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 the Figure 4 the examples of proton recoil tracks obtained in CR-39 calibration are shown. The proton recoil tracks are represented by bright pits of 5-8 µm in diameter on dark background.

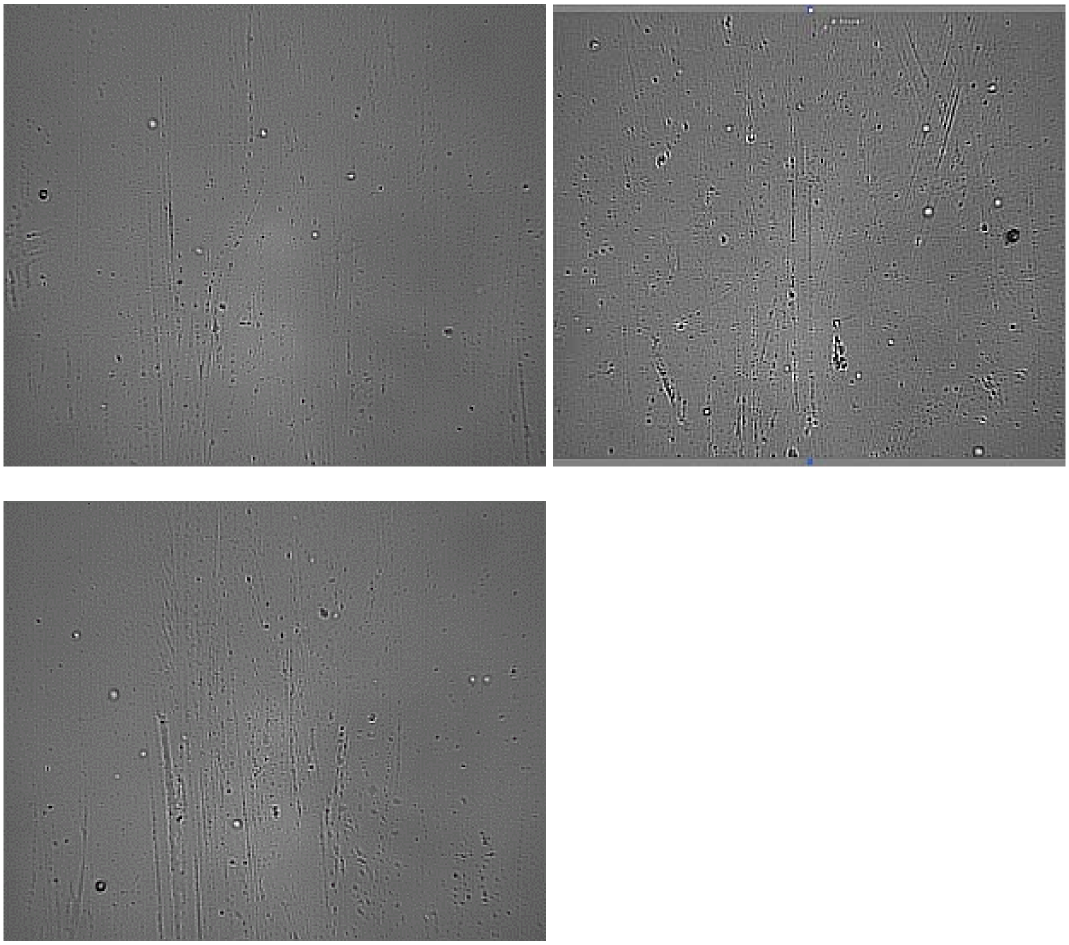
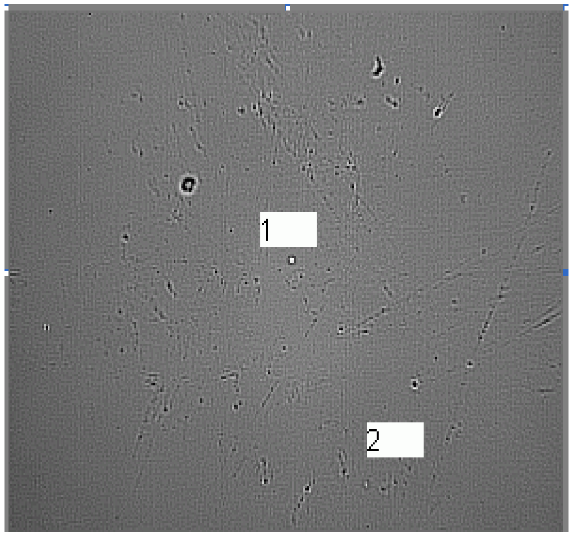
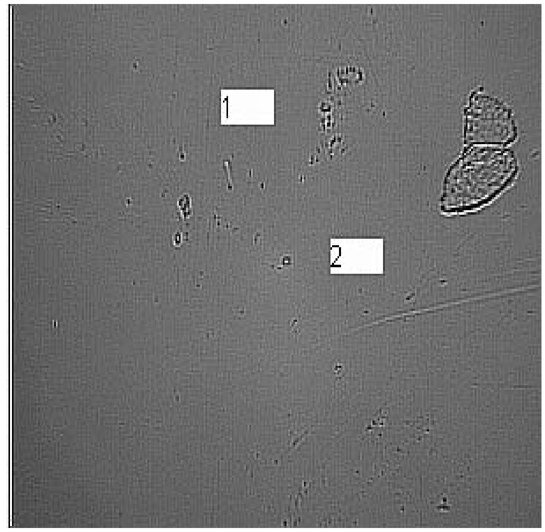


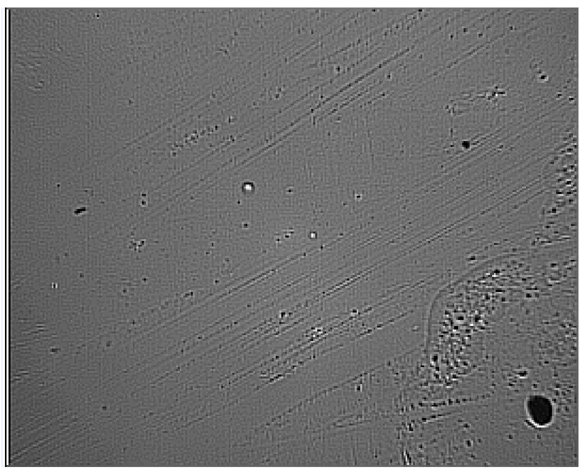
**Figure 4**. Examples of proton recoil tracks of 5-8 µm diameter (bright pits only - all others are the surface defects !!!) for Cf-252 neutron source (7 hr etch, removed depth is h = 9.2 µm): Total neutron fluence is $F_n = 7x10^8$ n/cm$^2$; images size is S= 120 x 90 µm$^2$.

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 calibration CR-39 detector, irradiated with Cf-252 neutron source. Three examples of such tracks are presented in Fig. 5 (a-c).





(a) (b)



(c)

**Figure 5**. Examples of proton recoil tracks (compare with Cf-252 bright pits) of 5-7 μm diameter from CR-39 Foreground detector #7 (removed depth is h ≈ 8.6 μm), images size is S= 120 x 90 $\mu m^2$: (a) - two tracks of normal incidence (1,2); (b) – track of oblique incidence (1), track of normal incidence (2);(c)- two tracks of normal incidence (bright pits in the center of the image).

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

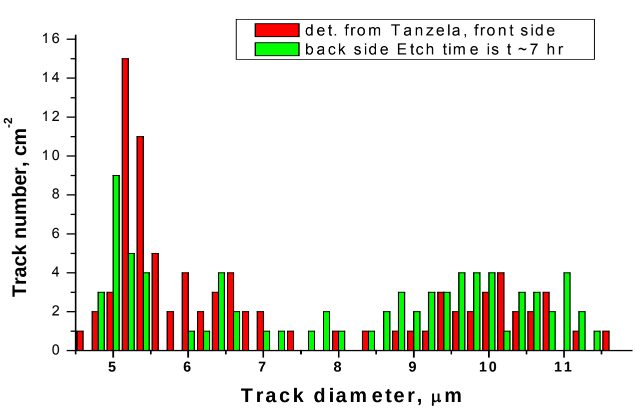


**Figure 6**. Track number distribution vs. track diameter for Fran Tanzella's # 7 CR-39 detector from both front and back sides. Etching time is 7 hr. The spectra contain real nuclear tracks. From the front side: the total number of circular and elliptic tracks (normal and oblique incidence) is N=77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

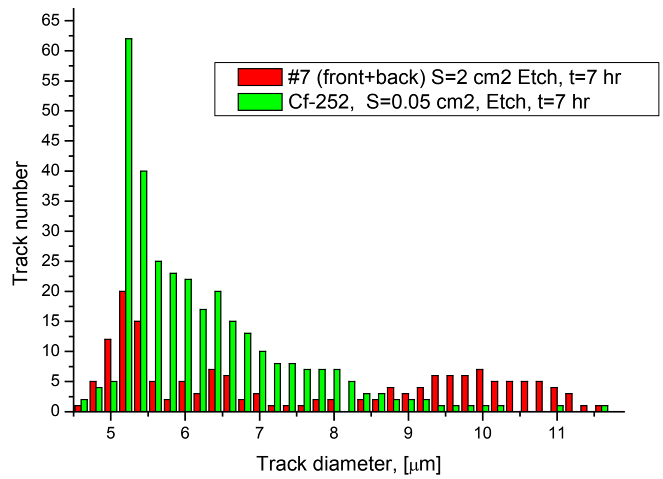



**Figure 7**. Comparison of CR-39 Cf-252 proton recoil spectrum corresponding to 7 hr etching with the sum of spectra obtained from the both sides of #7 CR-39. In the track diameter range of 4.5-8.0 μm the #7 spectrum look similarly to Cf-252 recoil. Note, that the maximum of track diameter distribution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

In order to confirm our finding concerning 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 8,9.

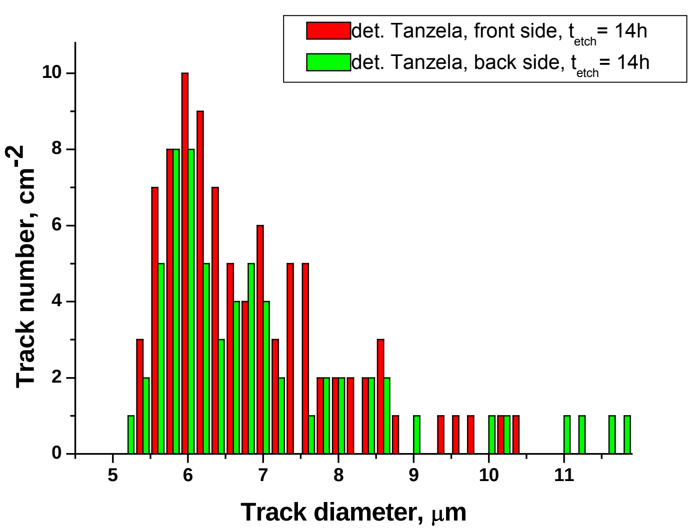



**Figure 8** Track number distribution vs. track diameter for Fran Tanzella's # 7 CR-39 detector from both front and back sides. Etching time is 14 hr. The spectra contain real nuclear tracks.

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 $cm^{-2}$, about 20 % of all tracks have elliptic shape. Similarly, at the back side: N=65 $cm^{-2}$. Notice again that the number of proton recoil tracks at the front CR-39 detector side is higher than at the opposite one

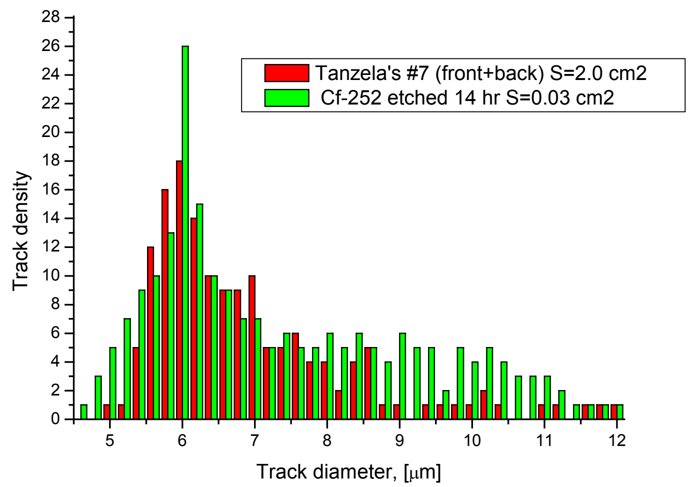



**Figure 9**. Comparison of CR-39 Cf-252 proton recoil spectrum corresponding to 14 hr etching time with the sum of spectra obtained from the both (front and back) sides of #7 CR-39. In the track diameter range of 4.5-11.0 μm the #7 spectrum looks almost identically to the Cf-252 recoil. Note, that the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

For further confirmation of neutron emission effect we carried out one more etching of #7 detector (total etch time $t_{et}$ = 21 hr , corresponding to total removed depth h = 27. 2 μm) The result of track diameter distribution for both the front and the opposite sides of the #7 detector at $t_{et}$ = 21 hr is shown in Fig. 8

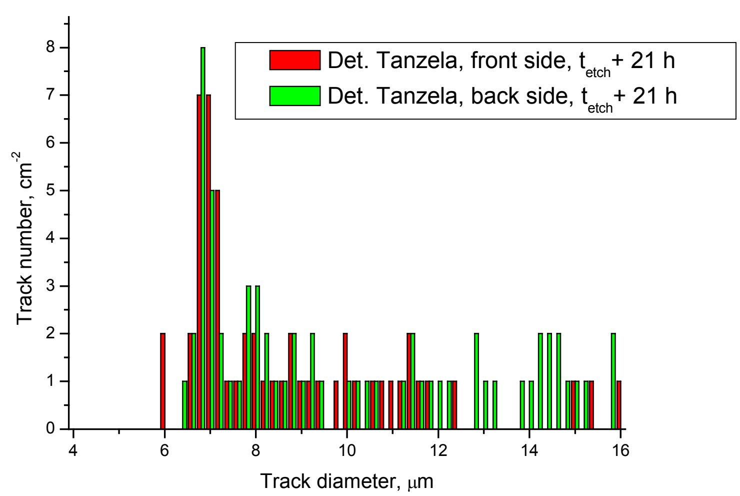



**Figure 10**. Normal incidence track distribution vs. their diameter for Fran Tanzella's # 7 CR-39 detector from both front and back sides. Etching time is 21 hr. The spectra contain real nuclear tracks. From the front side: the total track number (both normal and oblique incidence) in the

range of 5-14.0 μm (the diameters consistent with Cf-252 spectrum at $t_{et}$ = 21 hr) is N=91 $cm^{-2}$, about 30 % of all tracks have elliptic shape. At the back side: N=67 $cm^{-2}$. Notice again that the number of proton recoil tracks at the front CR-39 detector side is higher than at the opposite one

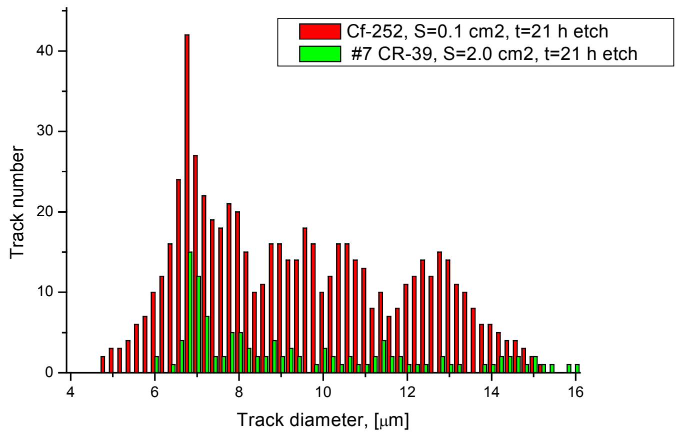



**Figure 11**. Comparison of CR-39 Cf-252 proton recoil spectrum corresponding to 21 hr etch time with the sum of spectra obtained from the both (front and back) sides of #7 CR-39. In the track diameter range of 5-14.0 μm the #7 spectrum looks similarly to the Cf-252 recoil. Note, that the maximum in the #7 detector is localized near 7.0 μm 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 $t_{et}$ = 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 calibration detector at 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. In summary, presented experimental evidence can be considered as a strong, unambiguous proof of #7 detector's fast neutron (2.5 MeV ) exposure.**

***II. Calculation of the neutron emission rate***

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

The total (normal and oblique incidence) Foreground track density in the range of proton recoil diameters (4.5 < d < 7.8 μm): at the front side is $N(fg)_1$ = 77 $cm^{-2}$; at the opposite side is the $N(fg)_2$ = 40 $cm^{-2}$. Average track density is <N(fg)> = 58.5 $cm^{-2}$.
The total Background track density at both sides of blank detector (S = 0.25 $cm^2$ each) is the <N(bg)> = 6.0 $cm^{-2}$ ,which is consistent with usually observed Background in fresh Landauer chips. This consistency allowed us to use our Background data (depicted in Fig.1) representing the result of large area (S=2.0 $cm^2$) reading of two Landauer Background detectors.
Thus, the "effect" with the Background subtracting would be <ΔN> = <N(fg)> - <N(bg)> = 52.5 ± 8.0 track/$cm^2$.
Accordingly to calibration measurements, the CR-39 self-efficiency ($t_{et}$ = 7 hr) with regard to Cf-252 neutrons was found to be $\varepsilon_s$ = 9.0 x$10^{-5}$. 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 (In) would be derived as:

$I_n = 2<\Delta N>/(t \times \varepsilon_s)$, where t- is the electrolysis (Foreground) duration. Assuming that effect of neutron emission was observed during the time when electrolysis current ($j > 0.5$ mA) was turn on ($t = 15$ days), the $I_n = 0.90 \pm 0.14$ n/s in $2\pi$ solid angle (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 count rate above the Background, the $I_n = 3.38 \pm 0.53$ n/s in $2\pi$ solid angle (highest rate estimate). So, the neutron emission rate in the run #7 can be estimated in the range of 1.0-3.0 n/s.

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

Similarly to (1), the total (normal and oblique incidence) Foreground track density in the range of proton recoil diameters for 14 hr etch ($4.5 < d < 11.0$ μm): at the front side is $N(fg)_1 = 110$ $cm^{-2}$ and at the opposite side is the $N(fg)_2 = 66$ $cm^{-2}$. Average track density is $<N(fg)> = 88$ $cm^{-2}$.
The Background track density at both sides of blank detector ($S = 0.25$ $cm^2$ each) is the $<N(bg)> = 26$ $cm^{-2}$ (data from Fig. 1)
The "effect" with the Background subtracting is $<\Delta N> = <N(fg)> - <N(bg)> = 62.0 \pm 10.7$ track/$cm^2$.
Accordingly to calibration measurements CR-39 at etch time $t_{et} = 14$ hr the self-efficiency was found to be $\varepsilon_s = 1.17 \times 10^{-4}$.
Then, for t =15 days: $I_n = 2<\Delta N>/(t \times \varepsilon_s) = 0.82 \pm 0.14$ n/s in $2\pi$ solid angle and for $\Delta t = 4$ days $I_n = 3.08 \pm 0.53$ n/s. Thus, the result for 14 hr etch gives approximately the same (within a standard deviation) neutron emission intensity range as that for a 7 hr.

Entire data obtained as a result of analysis of the #7 CR-39 detector, including track reading within three removed depths (8.7, 18 and 27 μm), 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, present strong, unambiguous evidence for fast neutron emission (the neutron energy in the range of $E_n \sim 2.2 - 2.5$ MeV) with the rate of $I_n \sim 1$-3 n/s during electrolysis run #7 (Pd deposition) performed in SRI.

# APPENDIX

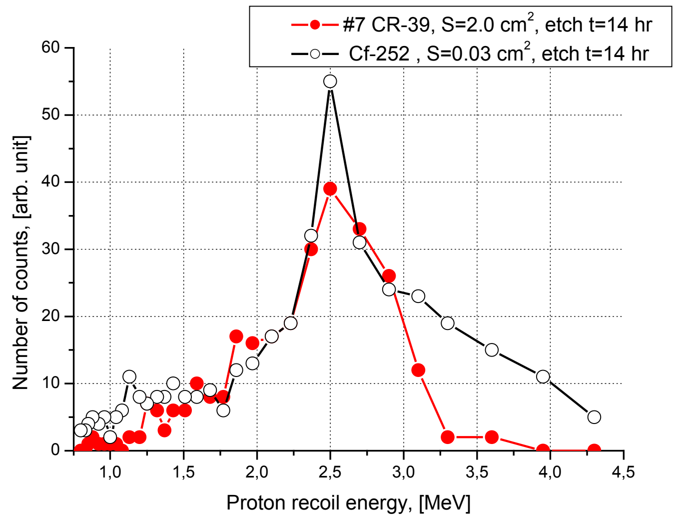



**Figure 1**. Rough reconstruction of the protons recoil spectra for CR-39 Landauer detectors obtained during electrolysis run (detector #7) and during exposure with Cf-252 neutron source. Etch time is t = 14 hr. The reconstruction of the spectra was deduced from the track density vs. track diameter histograms, taking into account two functions: (a) track diameter vs. proton energies at t =14 hr, (Fig. 3) and (b) the critical angle $\theta_c$ vs. proton energy (Fig.4).

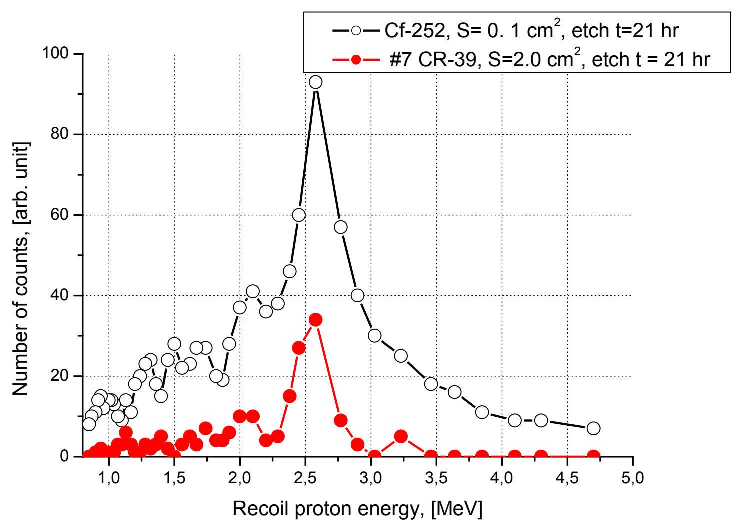



**Figure 2.** Same as Figure 1 but at etch time t = 21 hr.

As seen from Figs 1 and 2, 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 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 to assume that fast monoenergetic neutrons with energy close E = 2.5 MeV have been emitted during electrolysis run. For more accurate numerical estimate of the recoil spectra, a Monte-Carlo calculation must be performed .

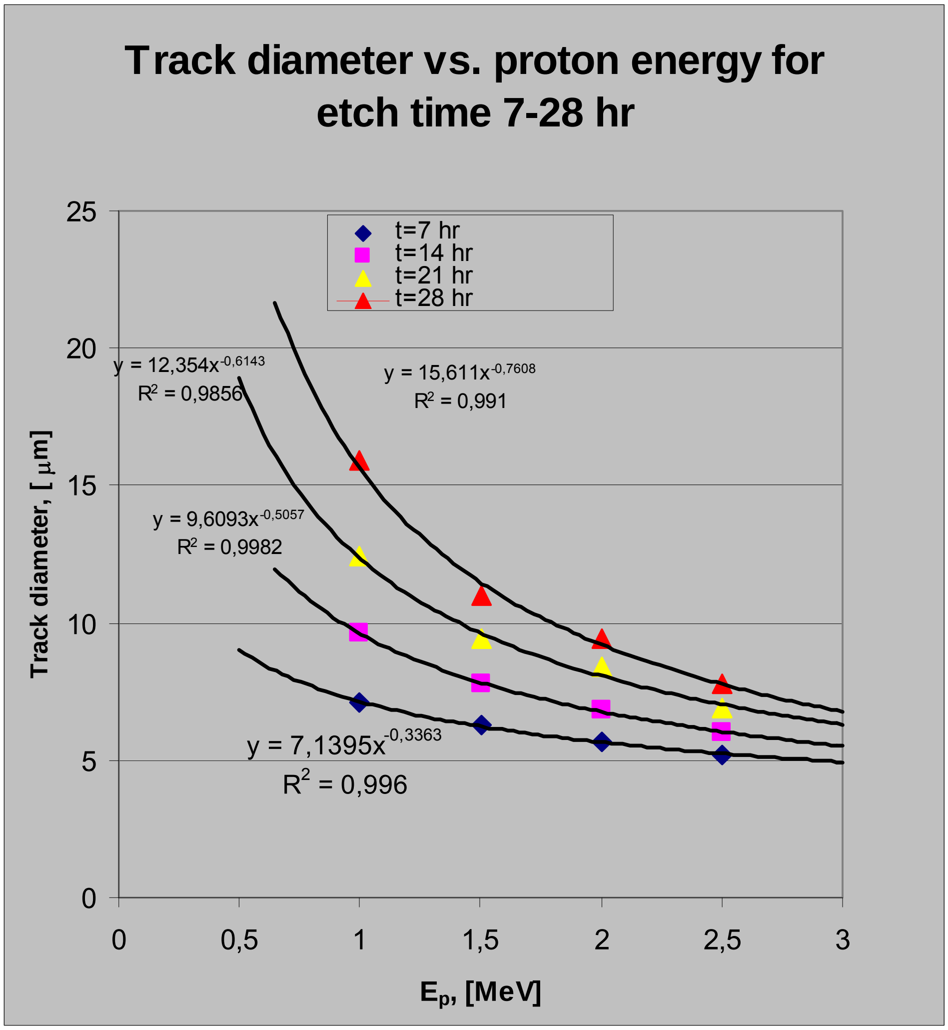



Figure 3. Track diameter vs. proton energy (E > 0.5 MeV) dependences for etch time 7-28 hr.

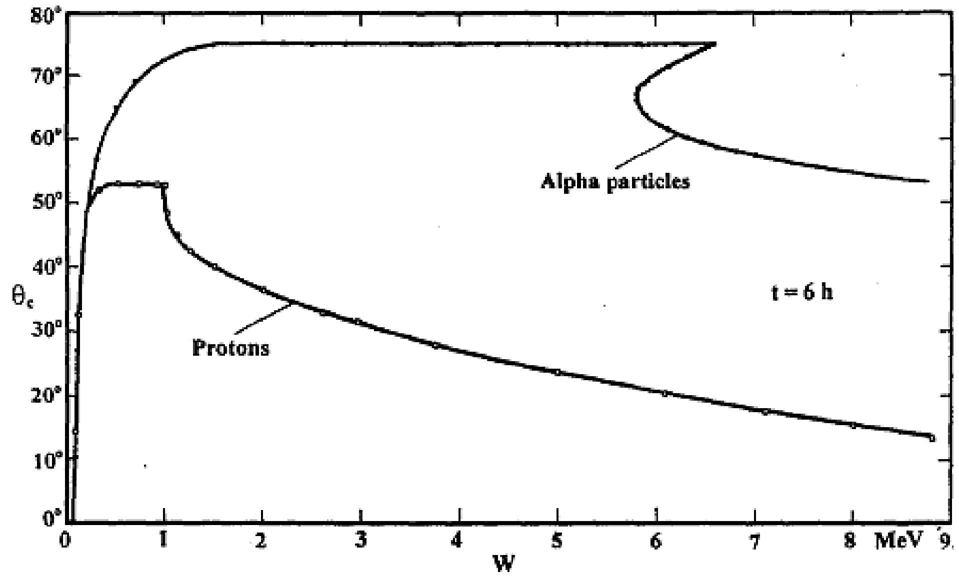



Fig. 4. Critical angle, $\theta_c$, as a function of the initial particle energy, W, at an etching time of t = 6 h for protons and alpha particle.