

In-situ long - range alpha particles and X-ray detection in Pd thin film-cathodes during electrolysis in $\text{Li}_2\text{SO}_4/\text{H}_2\text{O}$

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Introduction/Objective

- Long-range alphas with energy 8.0 –14.0 MeV were measured earlier at D(H)-desorption from Au/Pd/PdO:D(H).
- Essential X-ray emission should be a signature of massive nuclear reactions that could produce excess heat.
- In-situ measurement of energetic particles, X-ray emissions with CR-39 detectors and calorimetry during electrolysis.

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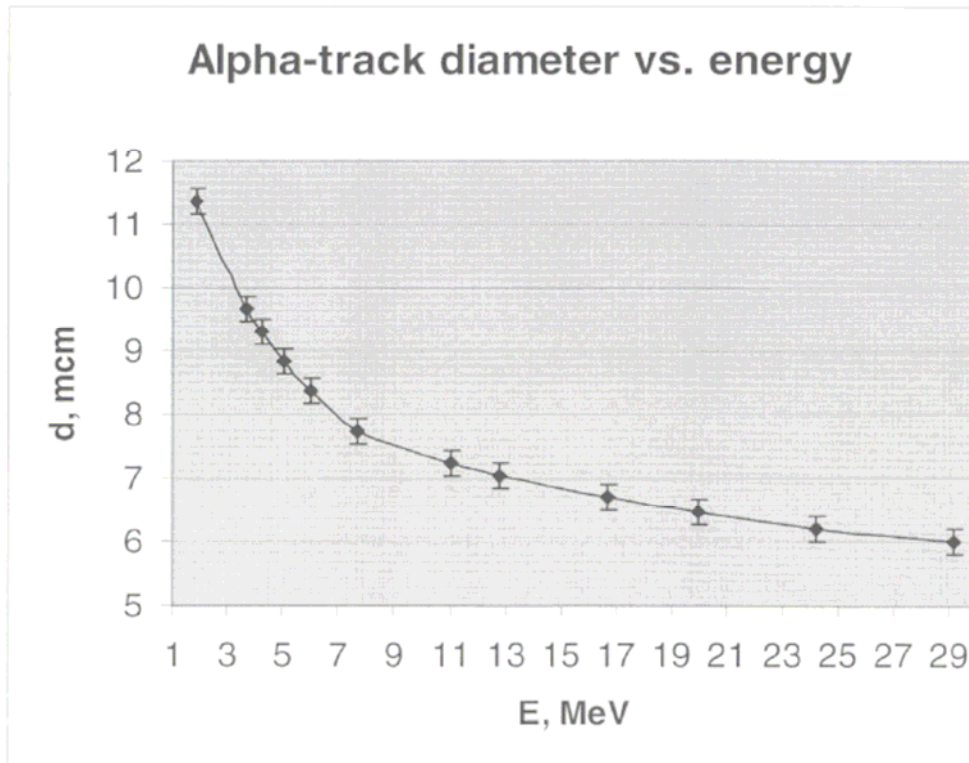
CR-39 track Detectors

- CR-39 detectors “Landauer” rad-track; $S=2.0 \times 1.0 \text{ cm}^2$ attached to Pd/Ni thin film cathode (Foreground); to substrate side or/and immersed in electrolyte in the cell (Background). Low initial Bg before electrolysis: $N(\text{Bg}) < 40 \text{ track/cm}^2$.
- In special experiments used CR-39 covered with $25 \text{ }\mu\text{m}$ Cu-film to identify type of emitted particle

CR-39 treatment and calibration

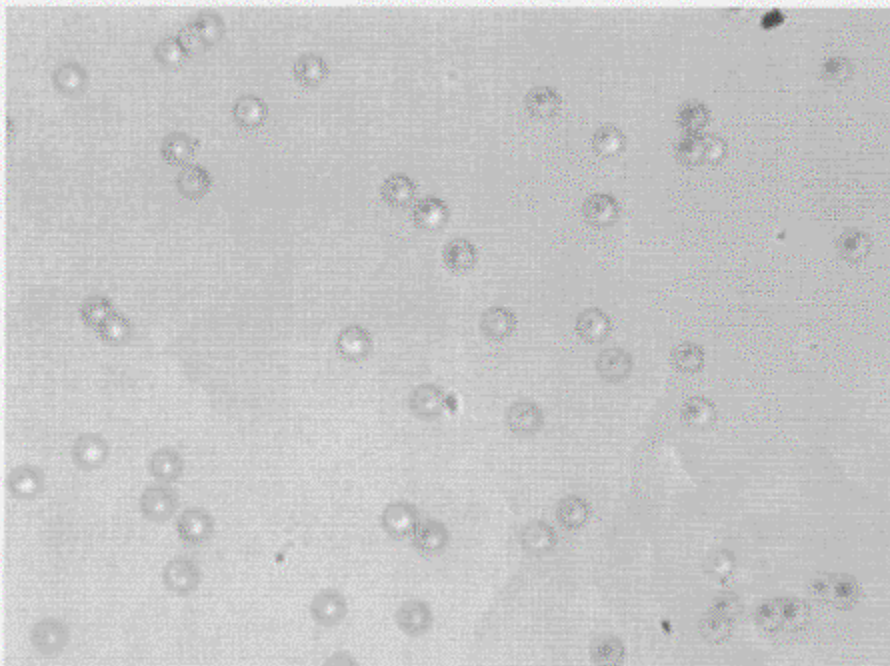
- After exposure detectors were etched in 6N-NaOH at $t=70^\circ\text{C}$ during 7 hrs. and investigated with optic microscope
- Calibration: for alphas $E < 8.0 \text{ MeV}$ – alpha-sources; for $E \sim 8.0\text{-}30.0 \text{ MeV}$ Cyclotron of LNR, JINR, Dubna; for protons $0.75\text{-}3.0 \text{ MeV}$ Van-DeGraaf accelerator, RINF, MSU, Moscow

Alpha calibration



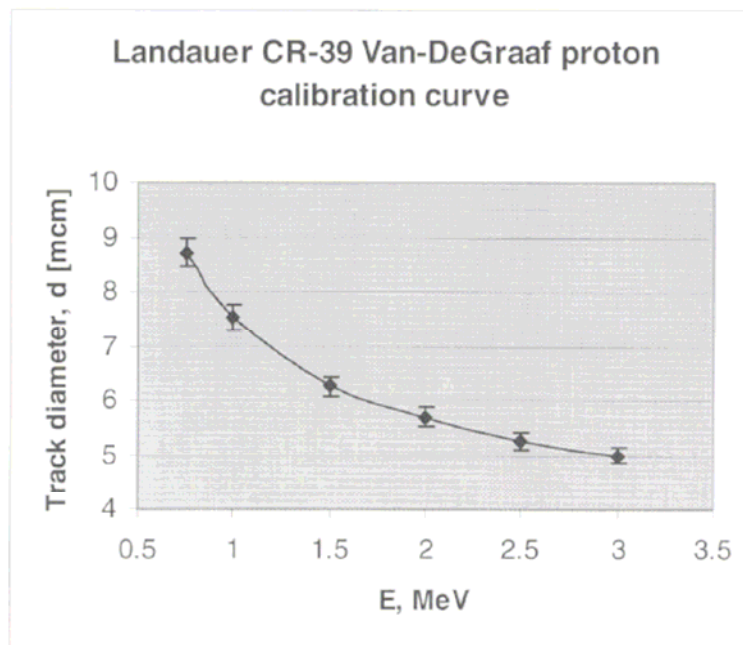
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Tracks from 12.0 MeV α -particles;
image area $S = 0.2 \times 0.2$ mm, (X 700)



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Proton Calibration



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X-ray detection

- TLD: LiF ($2.0 \times 2.0 \times 1.0 \text{ mm}^3$) – sensitivity $\sim 1 \text{ mrem}$; $\text{Al}_2\text{O}_3:\text{C}$ ($2.0 \times 2.0 \times 0.2 \text{ mm}^3$) – sensitivity 0.2 mrem shielded by $50 \mu\text{m}$ PE-film and fixed at Pd-film surface. Background detectors at the bottom of electrolytic cell
- Initial Background (before runs) for LiF – 4.0 mrem , for Al_2O_3 – 10.0 mrem .

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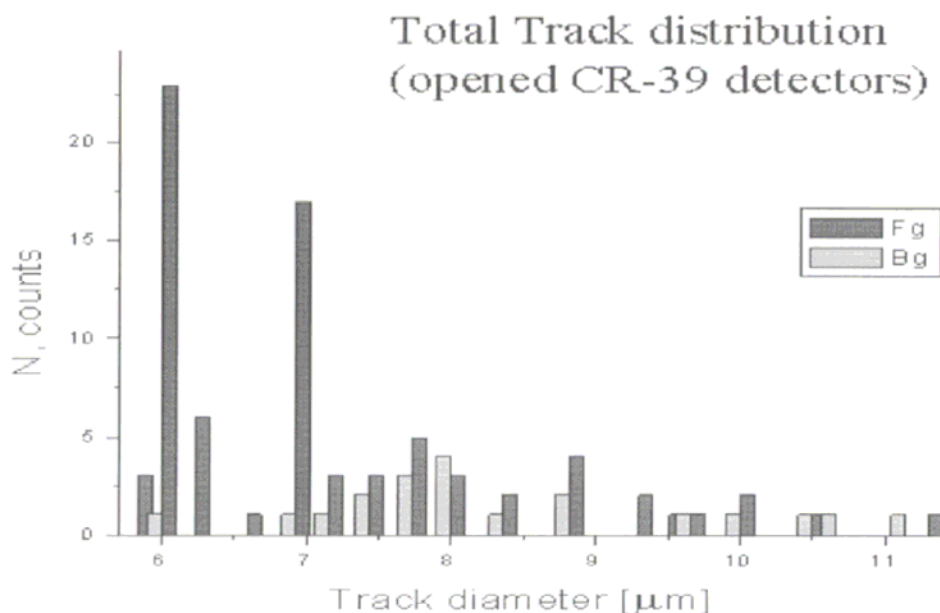
Experimental Results

- Two significant peaks at 7.0 μm and 6.0 μm observed only in Foreground runs with opened CR-39 detectors.
- Almost no counting for tracks with $d < 7.5$ μm in Background runs.
- With Cu-film shielded CR-39 7.0 and 6.0 μm peaks disappeared. But 3 other peaks ranging from 7.5 to 11.4 μm remain after Background subtracting.

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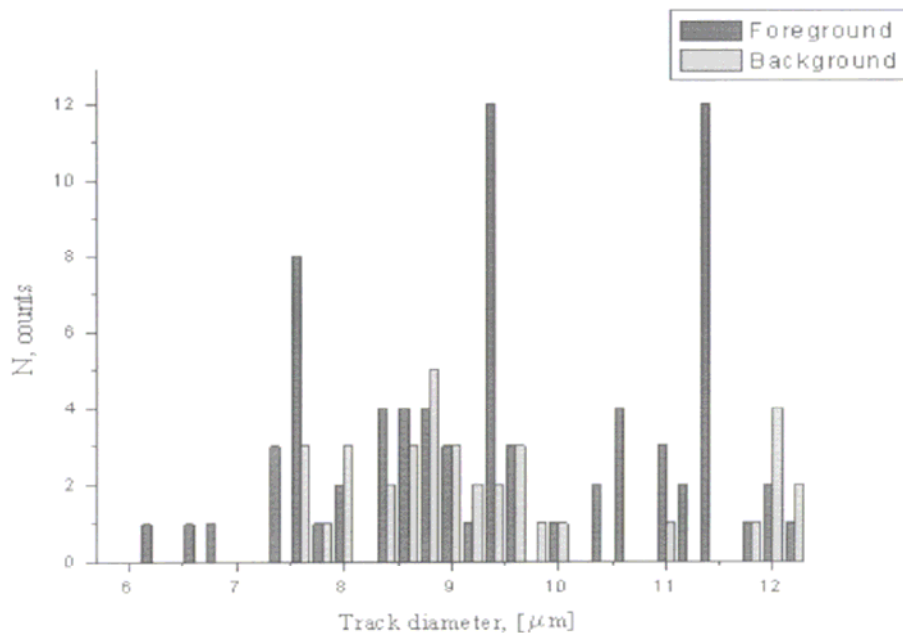
Histograms of track distributions with opened CR-39 detectors



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Histograms of track distributions with Cu-shielded CR-39 detectors



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X-ray results

- Reproducibility of results is not satisfied. Best result with LiF TLD for Pd/glass-sample (fractured) with Background subtraction:

$$\Delta n_x = 2.0 \pm 1.0 \text{ mrem,}$$

This dose consistent to 5.0-10.0 keV X-ray quanta yield upper limit $Y_x \leq 5.0 \text{ s}^{-1}\text{xcm}^{-2}$.

Expected X-ray production from alpha and proton emission is: $N_x \sim 0.4 \text{ s}^{-1}\text{xcm}^{-2}$.

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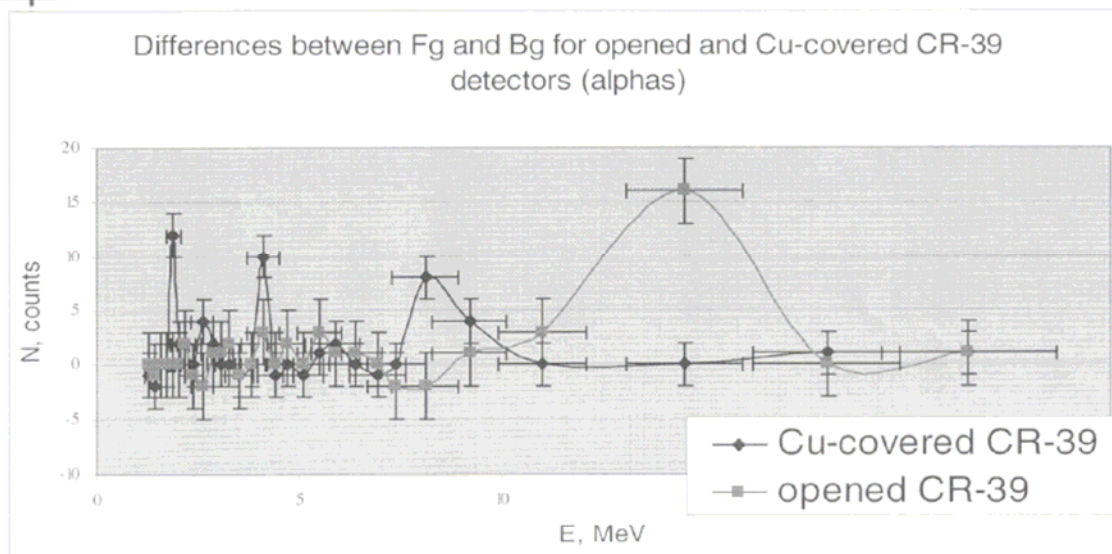
Discussion

- Tracks with $d=7.0\ \mu\text{m}$ could be either 13.5 MeV alphas or 1.25 MeV protons
- Tracks with $d=6.0\ \mu\text{m}$ could be either ~ 29 MeV alphas or 1.7 MeV protons
- $25\ \mu\text{m}$ Cu-foil totally absorbs all alphas with $E < 9.0$ MeV and protons with $E < 2.4$ MeV.

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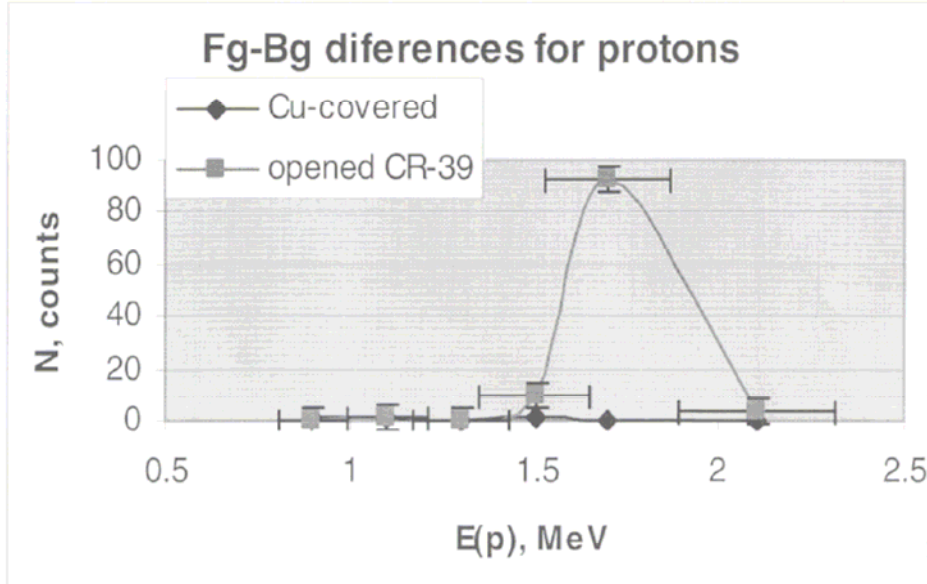
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Alpha-tracks with Background subtracting



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Discussion II

- Disappearance of 6.0 μm peak for CR-39 covered with Cu-foil indicates to 1.5-2.0 MeV protons. $\langle n_p \rangle = (1.15 \pm 0.10) * 10^{-3} \text{ s}^{-1} \text{ cm}^{-2}$
- Shift in 7.0 μm peak (opened detector) to 9.0-11.0 μm is a proof of $13.5 \pm 2.5 \text{ MeV}$ alphas in Foreground.
- Yield of 13.5 MeV alphas at efficiency:
 $\varepsilon = \frac{1}{2}(1 - \sin\theta) = 0.13$; ($\theta = 48^\circ$)
 $\langle n_\alpha \rangle = (2.2 \pm 0.3) * 10^{-4} \text{ s}^{-1} \text{ cm}^{-2}$.

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Conclusions

- During the electrolysis of Pd/Ni thin films in $\text{Li}_2\text{SO}_4/\text{H}_2\text{O}$ electrolyte alpha particles with estimated energy 13.5 ± 2.5 MeV and protons ~ 1.7 MeV were detected.
- Excess heat production does not always correlate with charged particle emissions
- Upper limit of X-ray emission rate during electrolysis doesn't exceed several $5\text{-}10 \text{ keV s}^{-1} \times \text{cm}^{-2}$. No massive nuclear reactions.