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IN-SITU CHARGED PARTICLES AND X-RAY DETECTION IN Pd THIN FILM-CATHODES DURING ELECTROLYSIS IN Li_2SO_4/H_2O

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Measurements of energetic charged particle and soft X-ray emissions have been performed using calibrated CR-39 plastic track and LiF/Al_2O_3:C-Thermo-Luminescent (TLD) detectors. It was found that during the electrolysis of thin Pd-film cathodes on the dielectric substrates, the alpha-particles ranging from 11.0-16.0 MeV and protons near 1.7 MeV are emitted. No significant X-ray emission with upper dose limit of ~ 1 mrem (corresponding to ~ 5.0 X-ray photon/s×cm^2 with E_x=10 keV) was detected.

1. Introduction

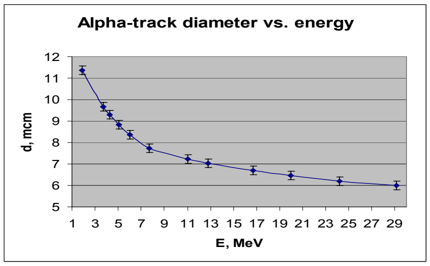
In order to obtain a proof for nuclear reactions [1] in thin-film cathodes (to explain excess heat production) the detection of nuclear radiation accompanying the electrochemical loading of those cathodes is strongly desirable. If massive nuclear reactions in such a type of cathode are really take place then nuclear energy conversion to the lattice excitation has to generate characteristic X-rays of cathode material as well as intensive soft Bremstrahlung. Moreover, generation of energetic charged particles during electrolysis could be also considered as signature of nuclear processes in thin-Pd film cathodes.

Earlier, using various Si surface barrier (SSB) detectors, including dE-E telescope we just studied the emission of charged particles, accompanying exothermic deuterium (hydrogen) desorption in Au/Pd/PdO:D(H) heterostructure [2]. The new phenomenon of long-range alpha particle generation in the energy range of $8.0 \leq E_\alpha \leq 14.0$ MeV for Au/Pd/PdO samples after their electrolytic loading (either by deuterium or by hydrogen one) was discovered.

So far, we studied long-range alpha emission only after the electrochemical loading of Pd with deuterium/hydrogen. Meanwhile, it was a great interest to expand the experiments on charged particle detection to the electrolysis process, where during the loading of Pd metal accompanying by an enormous lattice deformation the stronger nuclear effects should be expected. Unfortunately, it is hard to apply electronic SSB and X-ray detectors directly to the cathode during electrolysis experiment. That is why, in the present study we have designed a new technique, which is allowed in-situ measurement of energetic charged particles and X-rays during the electrochemical loading of the flat-plate Pd-thin film cathodes.

2. Experimental technique

The thin Pd-film cathodes and electrolysis procedure are described in [3]. For charged particle detection the purified "Radtrack" CR-39 plastic track detectors (with the size 2.0×1.0 cm^2) by Landauer Inc. have been used. Especial purification procedure utilized for detector manufacturing as well as hermetic saving condition allow to minimize initial alpha track density of these CR-39 to less than 10 cm^{-2}.



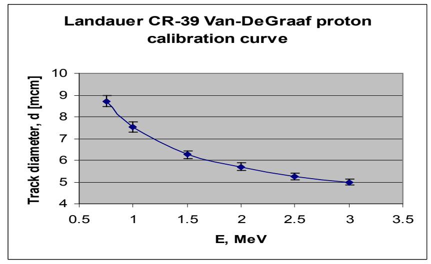


Figure 1 **Figure 2**

These detectors were calibrated with alpha-sources (in the range of 1.6 – 7.7 MeV) and by monoenergetic cyclotron alpha-beams (in the energy range of 10.0 – 30.0 MeV) as well as by proton beams with energy ranging of 2.0-3.0 MeV. (**Fig.1**). For energetic proton detection the detectors were also calibrated with Van DeGraaf accelerator by monoenergetic proton beams (energy ranging from $0.75 \le Ep \le 3.0$ MeV), (Fig.2). After the beam exposure detectors were etched in 6N-NaOH at t=70°C during 7 hrs. and investigated with optic microscope. Typical view of alpha-track picture is presented in Fig.3. As seen, at normal interaction of monoenergetic cyclotron beam with detector, the tracks observed after the etching have almost ideal circle-like shape. These nuclear tracks can be easy distinguished from the defects of CR-39 subsurface structure.

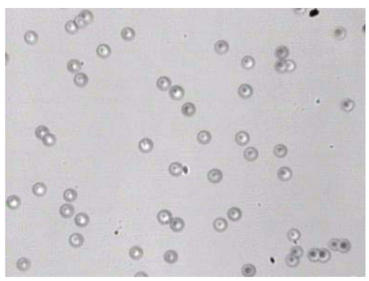


Figure 3

The efficiencies of CR-39 detection with respect to different energy alphas and protons were estimated in accordance with their critical angles θ_c being determined by formula [4]: $\theta_c = \sin^{-1}\{[1-(d_E/2h)^2]/[1+(d_E/2h)^2]\}$, where d_E-is the track diameter produced by charged particle with energy E (Fig 1,2), and h = 9.1μm is the depth of etched layer in CR-39 at our etching condition. Knowledge of the critical angles calculated from above formula allow to determine the efficiency ε of the charged particle detection as:

$$\varepsilon = \tfrac{1}{2}(1-\sin\theta_c) \qquad (1)$$

In electrolysis experiments the freshly opened CR-39 detector chips were attached either to the Pd thin film cathode(Foreground) or to the substrate side or/and immersed in electrolyte in the cell (Background). Background experiments showed proportional growth of track density vs. time for CR-39 immersed in electrolyte (Fig.4). At large Background duration it is possible to observe two separate alpha-peaks with track diameters located at 8.0 and 9.0 μm, respectively (Fig.4). The energy positions of these peaks are in good agreement with conventional alpha-Background and are normally corresponded to about 7.0 MeV radon (8.0 μm) and 5.0 MeV (9.0 μm) thoron series of natural alpha-nuclides.

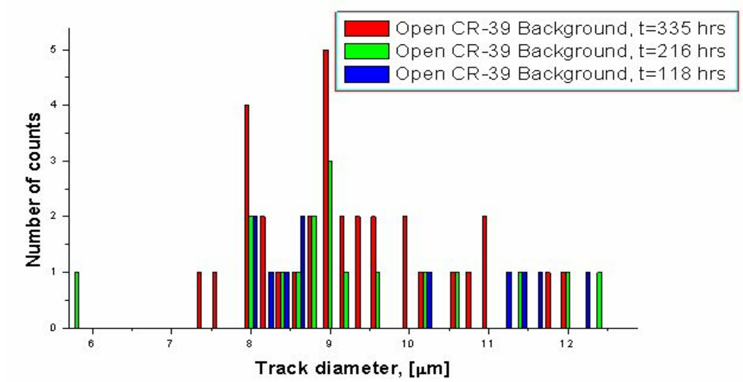


Figure 4

In order to separate high-energy alphas and low-energy protons that could be possibly emitted during electrolysis runs, the thin Cu-foils (25 μm thick) were inserted between the cathode metallic coating and the CR-39 surface. 25 μm Cu coating is completely absorbs all alpha-particles and protons with energies below 9.0 and 2.3 MeV, respectively. Background measurements in experiments with Cu-covered CR-39 were performed similarly to that with open detectors. As expected, these background experiments showed significant reduction (~2 times) in the total track density compared to that obtained with open CR-39 detectors (Fig.5).

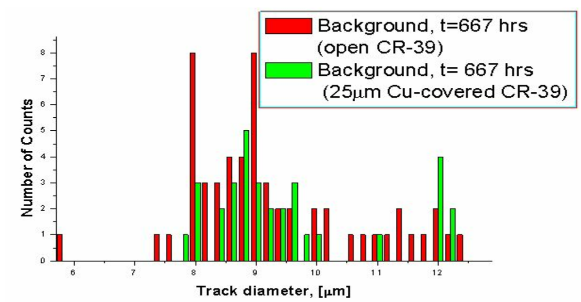


Figure 5

To in-situ detection of the expected soft X-ray radiation during electrolysis, the single-crystal TLD (Landauer Inc.) covered with 50 μm polyethylene film were applied to the Pd surface of electrodes. The Background TLD was immersed at the bottom of the employed electrolytic cell. The TLD reading out procedure was carried out in Landauer Inc. The nominal sensitivities of TLD used (with the size of $S = 2.0\times2.0\ mm^2$) with respect to absorbed dose of the soft X-ray photons (with energy $E_x \geq 2.0$ keV) was in the range of 0.1 (Al_2O_3:C) by the 1.0 (LiF) mrad. However, in fact, due to about 2 months storage of detectors in background environment before experiments the initial background levels of TLD were about 10.0 mrad and 4.0 mrad for Al_2O_3:C and LiF, respectively. Thus, actual sensitivity of TLD in electrolysis experiments did not exceed 1-2 mrad.

Experiments on detection of nuclear products during electrolysis runs were carried out using simultaneously open and Cu-shielded CR-39 chips and Foreground TLD crystal all attached to the same thin film cathode. Two Background CR-39 (open and Cu-shielded) and Background TLD were fixed inside the electrolyte in the employed electrolytic cell. The charged particle and x-ray detection were carried out simultaneously with excess heat measurements in open type calorimeter. The electrolysis current and duration during one Foreground run were normally varied in the range of 50-400 mA and 2.0-30 days, respectively.

3. Experimental Results and Discussion

The Foreground runs with electrolysis of Pd-thin film cathodes the exposed CR-39 detectors (t~2.0-30 days) showed the appearance of unusual diameter tracks that were not observed in Background detectors exposed in the same electrolytic cell. Indeed, in the track diameters distribution N(d), two significant peaks located at 7.0 mm and 6.0 mm observed in the Foreground runs (with electrolysis) with opened CR-39 detectors (Fig.6).

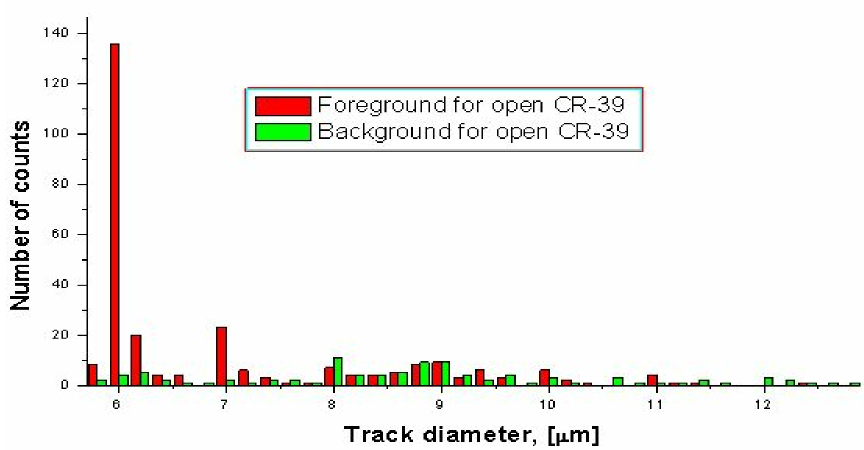


Figure 6

At the same time, almost no counts for tracks with d < 7.5 mm was found in the corresponding Background runs for detectors exposed in the same electrolytic cells. The low diameter tracks appeared to accompany electrolysis of the thin Pd-film and Pd-black cathodes. The intensity of charged particle emissions and ratio between 6.0 and 7.0 μm peaks during electrolysis are strongly depended upon the cathode history and structure (Table 1). It should be noted that generation of charged particle emissions during the electrolysis of thin Pd cathodes has a good reproducibility (in contrast to the irreproducible emission of DD-products in Pd-D systems). The count rates of protons and alphas after Background subtracting, presented in the Table 1, are statistically significant with Background level being close to zero. In the control experiment with CR-39 detector attached to the thin film NiO_x (obtained by annealing of Alumina/Ni(4000A) sample in air atmosphere), where despite of the high voltage applied (U~10.0 V) the electrolysis current in the cell was very low (I~1.0 mA), no tracks with d < 7.8 μm were detected. (Table 1).

Table 1

Charged particle emissions (alphas and protons with background subtraction) during electrolysis of different metal thin film cathodes, accordingly to open CR-39 detectors

Sample	Glass/Pd(2000A)	Alumina/Pd(4000A)	Pt/Pd-black, electroplated	Pd/Cu/Pd, Electroplated	Alumina/NiOx
Time of exposure, [hrs]	48	118	216	802	118
History of sample	Pd-coating fractured for 48 hrs. No excess heat	Survived, Excess heat, $E_{ex} \sim 20$ %	Pd-black partially detached, $E_{ex} \leq 8.0\%$	Survived. No excess heat	Survived. No electrolysis at the Ni-surface. No excess heat
$\langle N_\alpha \rangle \times 10^{-4}$, $[s^{-1} * cm^{-2}]$	4.8 ± 1.0	6.9 ± 1.2	0.80 ± 0.28	0.18 ± 0.08	0.0
$\langle N_p \rangle \times 10^{-4}$, $[s^{-1} * cm^{-2}]$	28.6 ± 4.1	16.0 ± 0.2	5.8 ± 0.9	0.24 ±0.11	0.0
$\langle N_p \rangle / \langle N_\alpha \rangle$ ratio	6.0	2.3	7.2	1.5	-
X-ray, dose [mrad], TLD	2.0 ± 1.0 (LiF) 0.5± 0.5 (Al_2O_3:C)	0.2 ±1.0 (Al_2O_3:C)	No TLD measurement	No TLD measurement	0.1 ± 1.0 (Al_2O_3:C)

Note: In accordance with (1) the mean CR-39 detection efficiencies for 11.0 – 16.0 MeV alphas and 1.5-1.7 MeV protons are calculated as: $\langle \varepsilon_\alpha \rangle = 0.13$ and $\langle \varepsilon_p \rangle = 0.10$, respectively.

In the Foreground runs with the same cathode being carried out with 25 μm Cu-film shielded CR-39 chips, the 7.0 and 6.0 mm peaks disappeared. But the other maximums ranging from 7.5 to 11.4 mm have appeared that were not found in Cu-shielded Background detectors (Fig.7).

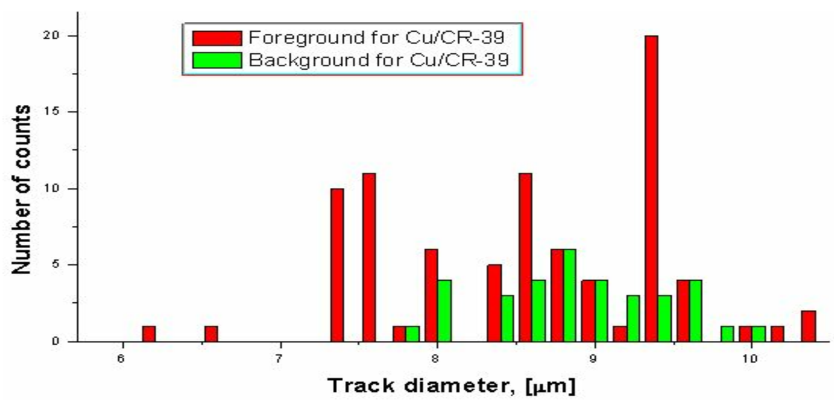


Figure 7

The experiments with Cu-shielded detectors and a knowledge of CR-39 calibration curves (Fig.1,2) allowed to identify the energy and type of particles emitted in the Foreground runs with open CR-39 detectors. Taking into account stopping powers and ranges of 25 μm Cu-film with respect to the alphas and protons with different energies, the initial energies of emitted particles were also calculated. Comparison of pictures obtained with opened and shielded detectors shows that 6 μm peak is completely disappeared while a broad near 7.0 μm peak in Fig. 6 shifted to the larger track diameters and split at least by 3 narrow peaks (Fig.7). Disappearance of 6.0 μm peak in a shielded detector indicates to the low MeV proton nature of this peak. In accordance with our calibration data the estimated proton energy would be within 1.5-1.7 MeV (Fig.8a, b).

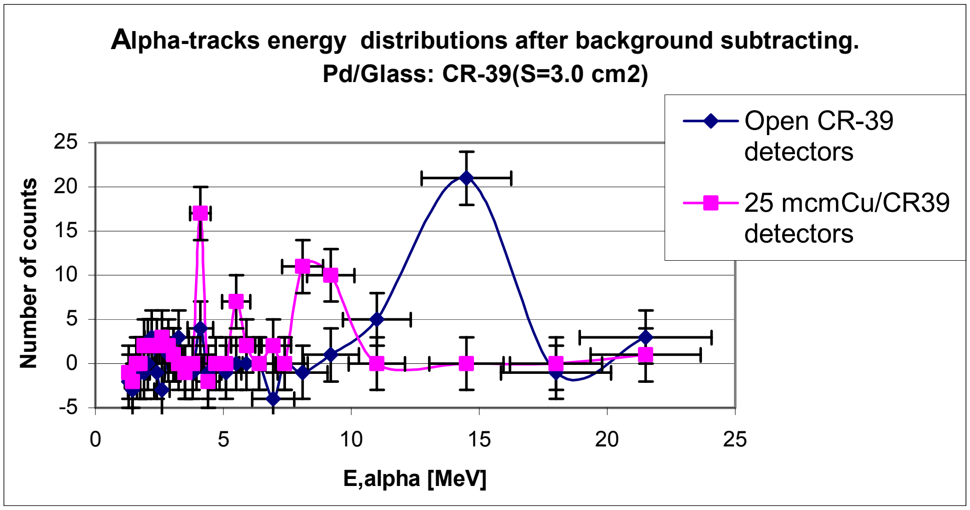


Figure 8a

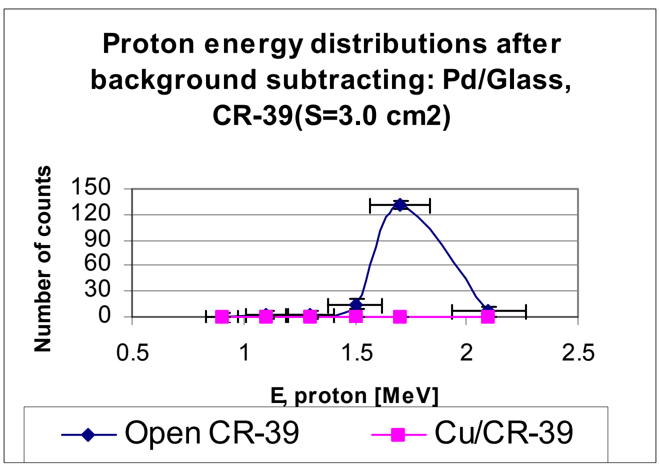


Figure 8b

In contrast to 6.0 μm peak, the 7.0 μm maximum, accordingly to its shift and splitting after crossing the Cu-shield should be ascribed to a broad 11-16 MeV alpha-peak. Indeed, the stopping range of alphas ranging from 11-16 MeV (for open Cr-39) would be consistent with observed narrow bands with energies 11.6, 12.5 and 14-16 MeV, respectively for Cu-covered detectors (Fig.8a). Due to higher resolution of CR-39 alpha-tracks for the lower energy particles (Fig.1) the broad 11-16 MeV alpha band could be observed as the single individual maximums after these particles crossed the Cu-foil. Therefore, we showed that electrochemical loading of Pd thin film cathodes on dielectric substrates unambiguously produce high-energy charged particles: 1.5-1.7 MeV protons and 11-16 MeV alphas.

The remarkable correlation between the p/α ratio and hydrogen mobility in Pd is observed. (Table 1) This p/α ratio for the cathodes with high hydrogen mobility (Pd/Alumina and Pd/Cu/Pd) is 3-4 times lower than for cathodes with low one (Pd/Glass and Pt/Pd-black). Indeed, due to surviving of Pd/Alumina and Cu/Pd samples the hydrogen diffusion in the Pd coating will be much higher that in fractured metal film of the Pd/Alumina or separated Pd-black particles. In the last case, the high loading of Pd-black could be result of some energy conversion caused the accelerating proton emission.

In contrast to charged particle detection results, the statistical significant level and reproducibility of X-ray emission was not satisfied. The best result was obtained for the Pd/Glass sample (that was completely destroyed during 2 days of electrolysis) attached to LiF TLD (the initial background level ~ 4.0 mrad). After the Background subtracting the emitted X-ray fluence was consistent with absorbed dose 2.0 ±1.0 mrad. The experiments with the other cathodes and Al_2O_3:C TLD (Table 1) did not show statistically significant results, obviously, due to higher initial background level (~ 10 mrad) of these TLD compared to that for LiF. In accordance with the 2.0 mrad absorbed dose in LiF TLD, assuming the generation of 10.0 keV X-ray emission, the upper detection limit of X-ray emission yield in the cathodes employed was found to be Y_x = 5.0 X-qunta/s×cm^2. Meanwhile, the expected level of X-ray Bremsstrahlung from emitted charged particles, including both protons and alphas (Fig.8a,b) was estimated as $N_x \sim 0.5$ s^{-1}×cm^{-2}. This level of X-ray emission is one order of magnitude below the detection limit of our TLD.

Thus, the X-ray measurement results indicate the absence of massive nuclear reactions caused by nuclear transmutation in Pd [1]. At the same time, these results still do not rule out the possibility of a weak X-ray emission as a result of the charged particle generation or Pd thin film fracture or detachment from the dielectric substrate.

5. Conclusions

Thus we found that during the electrolysis of thin Pd-film cathodes on the dielectric substrates as well as Pd-black, the alpha-particles ranging from 11.0-16.0 MeV and protons near 1.7 MeV are emitted. At the same time, no significant X-ray emission with upper dose limit of ~ 2 mrem (corresponding to ~ 5.0 X-ray photon/s×cm^2 with E_x=10 keV) was detected.

At the present time, the mechanism of the observed charged particle emission is not clear at all. It probably could be regarded to the strong lattice-nuclear coupling in the Pd thin films under the electrolytic loading. Regardless of the actual mechanism of discovered phenomenon, it should be emphasized that observed effect of long-range alpha and proton emission cannot be described in terms of nor natural alpha-nuclides contaminations in Pd neither the cosmic rays interaction with cathode.

6. References

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