


# Evidence for Condensed Matter Enhanced Nuclear Reactions in Metals with a High Hydrogen Solubility



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# Typical CMNS effects do not show nuclear signatures

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- *Magnitude of specific energy of Excess Heat (keVs/Pd-atom)?*  
not enough by itself to convince in nuclear origin!!!
- *$^4\text{He}$  and  $T$  accumulation?* Detected in atomic form (not as nuclear particles), may be extracted from surrounded media: still indirect evidence!!! If  $^4\text{He}$  detected is really from giant enhancement of  $d(d,\gamma)^4\text{He}$  channel of DD-reaction (the rate is  $\sim 10^{11} \text{ s}^{-1}$ ), why no sign of intensive (at least  $10^4 \text{ s}^{-1}$  rate) of  $d(d,p)t$  and  $d(d,n)^3\text{He}$  channels (even suggesting their suppression by factor  $10^7$  compared to “main” He-4 channel). No intensive X-ray.
- “*Transmutations*”: Only changes in stable isotope are observed. No signature of accompanying nuclear emissions and X-ray radiation.

# Nuclear signature of CMNS effects

- DD-reaction enhancement during low energy deuteron bombardment of metallic targets (accelerator and glow discharge experiments)
- Low intensity emissions of DD-reaction nuclear species: 3 MeV protons, 1 MeV tritons, 2.45 MeV neutrons
- Energetic alpha emission ( $E_{\alpha} \geq 10$  MeV). These alphas cannot be emitted by natural radionuclides/cosmic background.
- Soft X-ray ( $E_x \leq 1.5$  keV) emission in experiments with pulsed Glow Discharge and during D-desorption from Pd/PdO:D<sub>x</sub>

# I. Enhancement of DD-reaction in metal targets

- Accelerators (J. Kasagi et al): Low energy, high-current accelerators:  $2.5 < (E_d)_{\text{lab}} < 20$  keV, Beam current  $I \sim 10\text{-}300$   $\mu\text{A}$ . Energy spread  $< 1.0$  %. Duo-plasmatron ion source ( $E \sim 50$  keV), decelerating system, magnetic optics to deflect neutrals from target. Target cooling system: (77-300 K). Energy spread  $< 1$  %.
- Pulsed Glow discharge:  $\text{D}_2/\text{H}_2$  GD,  $P(\text{D}_2) = 2.0 - 10.0$  tor. Ti-cathode ( $S=0.64$   $\text{cm}^2$ ), Mo-anode, (cathode-anode distance:  $x \sim 5.0$  mm). Square-shape pulses: duration  $\tau = 200\text{-}400$   $\mu\text{s}$ ; rise time  $\sim 1$   $\mu\text{s}$  pulse; repetition 2.0 kHz; Voltage:  $U = 0.8\text{-}2.5$  kV; Current  $I = 150\text{-}450$  mA. Stability of  $U$  and  $I$ :  $\pm 10$  %.  $E_d$  energy spread  $\sim 15$  %.
- Charged particle detectors: Accelerators and GD produce huge noise. Neutron detection is difficult. Si-surface barrier (SSB) dE-E. Full noise suppression. Detection of 3.0 MeV protons from  $d(d,p)\text{T}$  reaction. Plastic track detectors CR-39 ( $N_{\text{track}} < 20$   $\text{cm}^{-2}$ ) detection of 3.0 MeV protons

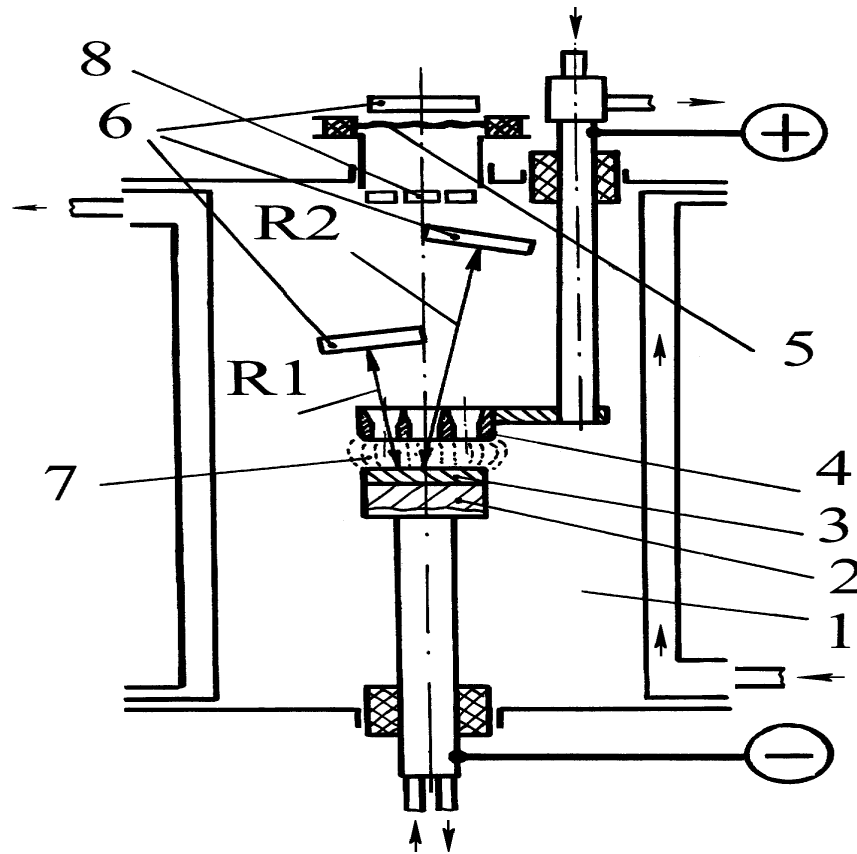


# Enhancement of DD-reaction in metal targets at low deuteron energy ( $1.0 < E_d < 10$ keV)

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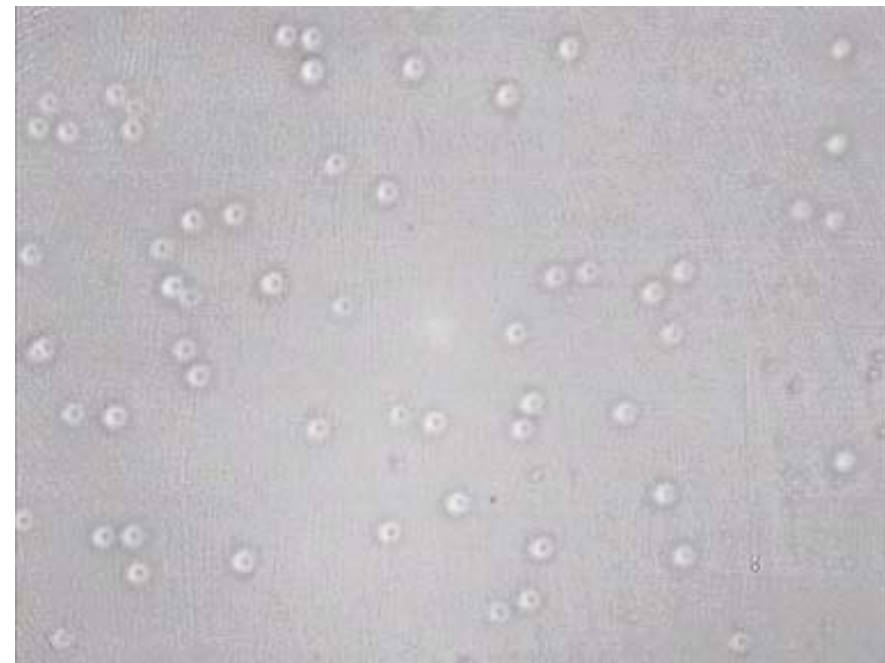
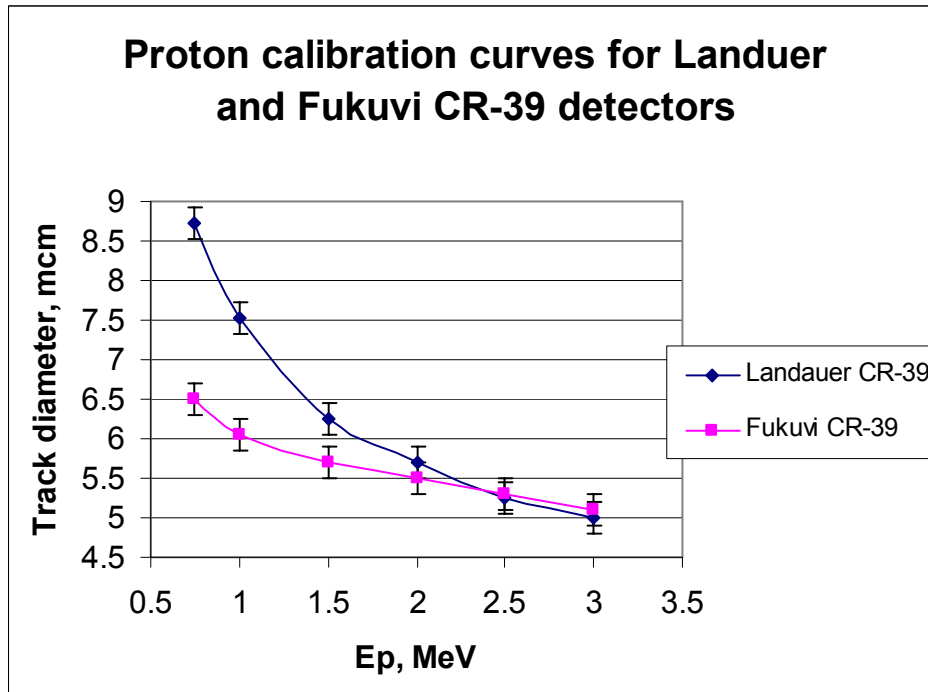
- Most metals show enhancement of DD-reaction yield at  $E_d \ll 10$  keV compared to the standard yield obtained by extrapolation of the DD-reaction cross-section to these  $E_d$  (see accelerator experiments: F. Raiola et al., Nuclear Physics, A719, 61C (2003), J. Kasagi et al., J. Phys. Soc. Jpn., **71**(12), 2881 (2002)).
- Recently, high-current glow discharge measurement showed strong enhancement of DD-yield – about 9 orders of magnitude at  $E_d = 1$  keV in Ti target (A. Lipson et al., JETP, **100**, 1175 (2005)).

# Glow discharge set up



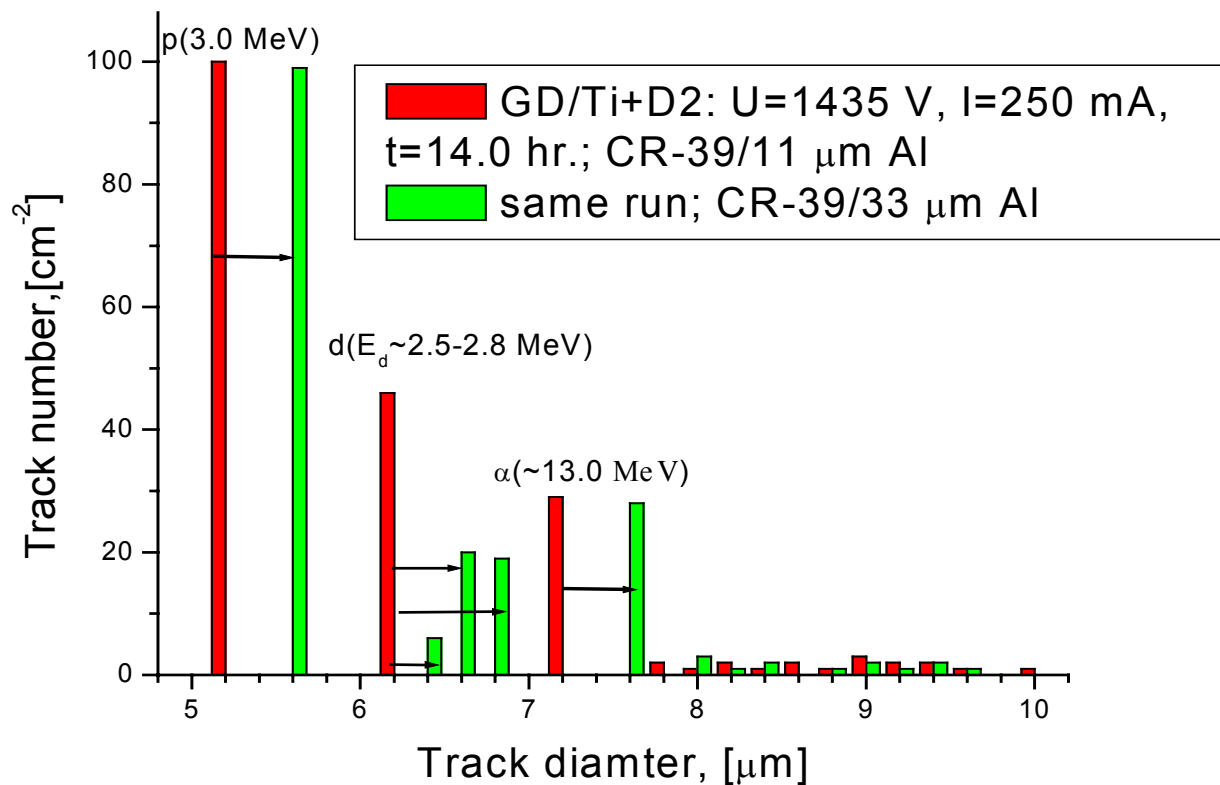
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# CR-39 track detectors: Proton calibration with Van De Graf accelerator $E_p = 2.5$ MeV



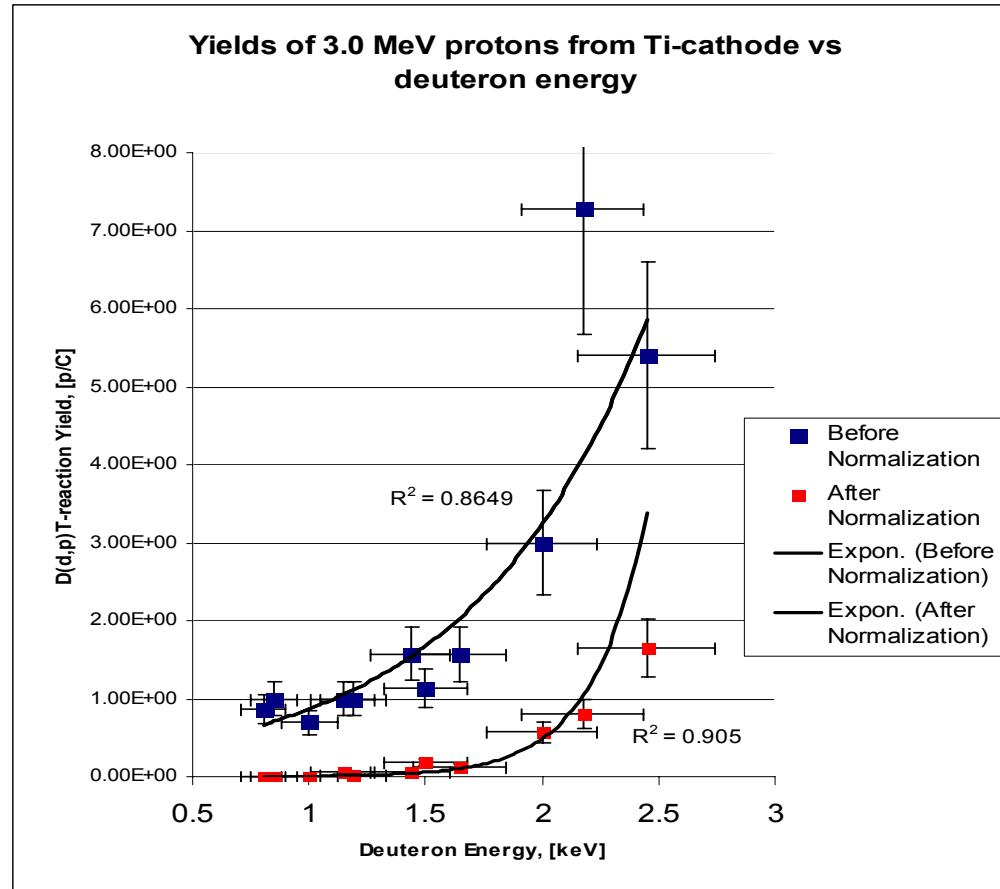
# 3.0 MeV proton energy estimate by absorption in Al-foil

11 $\mu\text{m}$  Al  $\rightarrow$  33 $\mu\text{m}$  Al  $\Rightarrow$   
d(5.2 $\mu\text{m}$ ) $\rightarrow$ d(5.6 $\mu\text{m}$ )  
 $\Rightarrow$  2.9 $\pm$ 0.2  $\rightarrow$  2.8 $\pm$ 0.3  
MeV

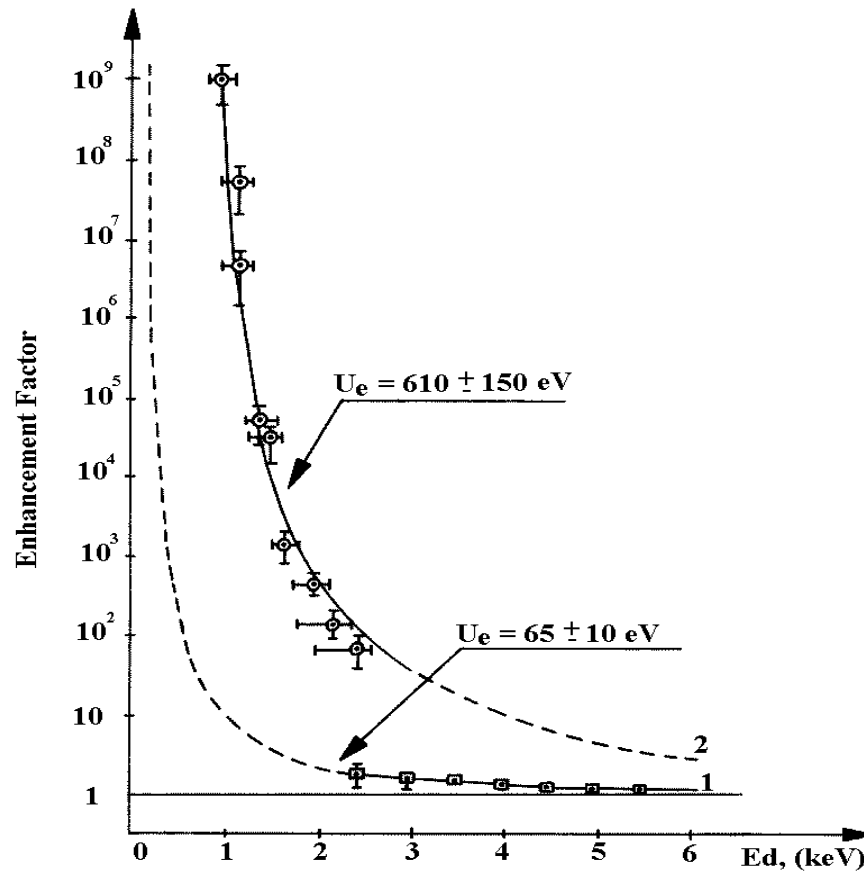




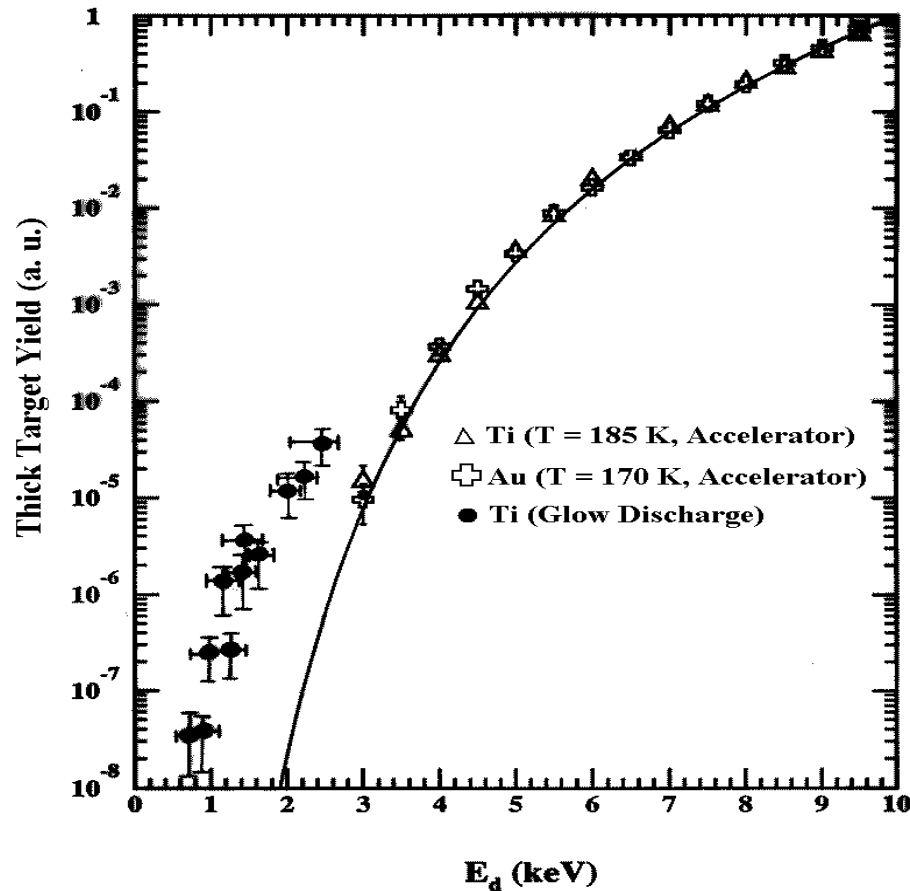
# Yields of 3.0 MeV protons before and after normalization to deuterium concentration



DD-reaction enhancement factor  $f(E) = Y_p(E)/Y_b(E) = \exp[\pi\eta(E)U_s/E]$  for Ti-target:  
(1)-accelerator; (2)-GD

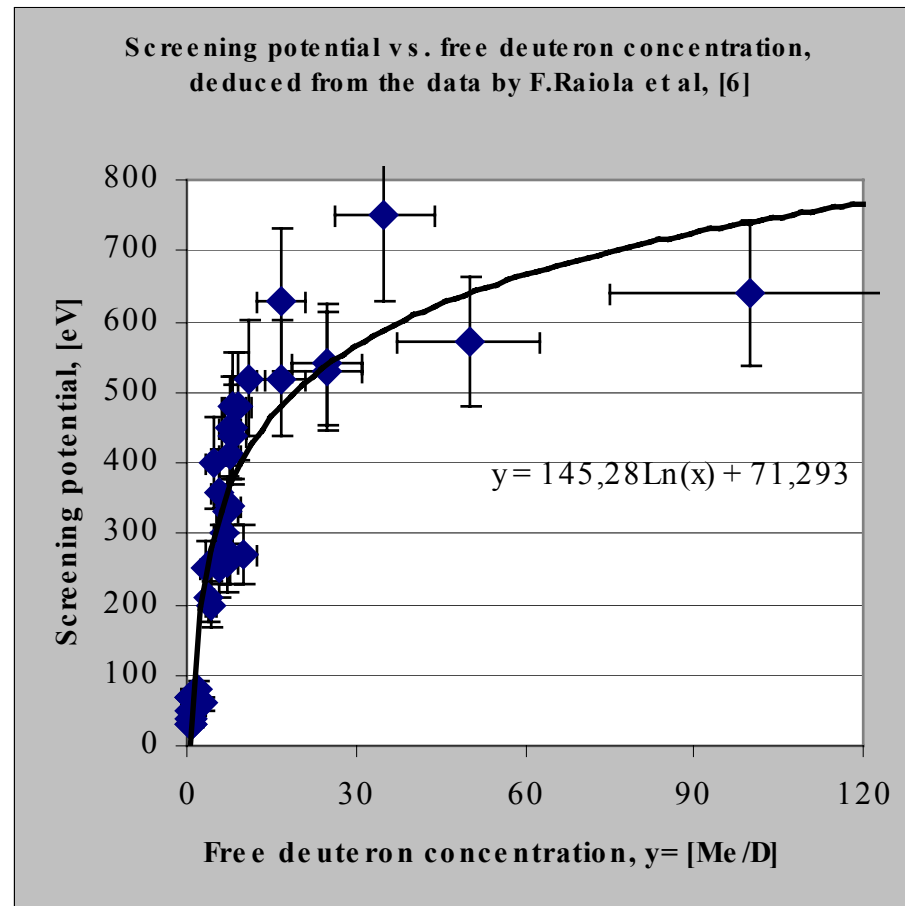


# Thick target yields for accelerator and GD compared to bare yield



Data are taken from F. Raiola et al, Europhys. J.A**19**, 283 (2004) at  
 $T_0=290\text{K}$ ,  $J_0=0.03\text{mA/cm}^2$ ,  $E_d \geq 5\text{keV}$ .

Points are consistent with increase in  $y = \text{Me}/\text{D}$ : Hf, Y, Lu, Sc, Gd, Tm, Ti,  
Ce, Yb, Sm, Zr, Er, Pr, Eu, Ho, La, Ge, C, W, Sr, Ir, Ba, Ru, Au, Ag, Re, Ni,  
Nb, Ta, Zn, Bi, Mo, Mn, Mg, Cu, Rh, Fe, Pt, V, Pb, Pd, In, Tl.



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# Electron Screening in metals

Target/ [Ref.]	$\Delta E_d(\text{lab}), [\text{keV}]$	$\Delta J, [\text{mA}]$	T, [K]	$U_e, [\text{eV}]$	Closest metal-host level	E(level), [eV]
Ti[7]	5-30	0.054	263	$\leq 30$	Ti(M <sub>II</sub> /M <sub>III</sub> )	32.6
Ti[9]	2.5-10.0	0.06-0.25	186	$65 \pm 15$	Ti(M <sub>I</sub> )	58.3
Ti*	0.8-2.45	225-450	$\geq 1800$	$610 \pm 150$	Ti(L <sub>II</sub> )	461
Au[7]	5-30	0.054	263	$61 \pm 20$	Au(O <sub>II</sub> )	71
Au[9]	2.5-10.0	0.06-0.25	180	$70 \pm 10$	Au(O <sub>II</sub> )	71
Pd[7]	5-30	0.054	263	$800 \pm 70$	Pd(M <sub>I</sub> )	670
Pd[9]	2.5-10.0	0.06-0.30	313	$310 \pm 30$	Pd(M <sub>V</sub> )	334
PdO[9]	2.5-10.0	0.06-0.30	193	$600 \pm 20$	Pd(M <sub>II</sub> )	560

# II-III. Low Intensity emissions of DD- reaction products (N) and energetic alphas

## Neutrons emission (references):

- Fracture of deuterated dielectrics, LiD, D<sub>2</sub>O (fractoemission): V.A. Kluyev, A.G. Lipson et al, Sov. Tech. Phys. Lett., 1986 (In  $\sim 10$ -20 n/impact)
- Acoustic cavitation in D<sub>2</sub>O with Ti horn: A.G. Lipson et al, Sov. Tech. Phys. Lett., 1990. (In  $\sim 0.5$  n/s).
- Ferroelectric phase transition through Curie point (T<sub>c</sub> – 220 K) in DKDP single crystal, A.G. Lipson et al, JETP, 1993 (In  $\sim 20$  n/transition).
- Superconducting phase transition (T<sub>c</sub> = 92 K) in deuterated HTSC ceramic YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-x</sub>, A.G. Lipson et al, Tech. Phys., 1995 (I<sub>n</sub>  $\sim 20$ -30 n/transition).
- Exothermic deuterium desorption from Pd/PdO:D<sub>x</sub> heterostructure, A.G. Lipson et al, Sov.Tech. Phys. Lett., 1992 (BF<sub>3</sub>), Fusion Tech., 2000 (NE-213).
- Electrolysis of Pd and Ti in D<sub>2</sub>O: S.E. Jones et al, Nature 1989, A. Takahashi et al, 1989, T. Bressani et al, 1991, etc.

Example: Neutron detection from Au/Pd/PdO:Dx sample (in the cell) with pair of NE-213 low Background/20m underground facility, Hokkaido University (A. Lipson et al . Fusion Tech, 38, 2000),

$$\Sigma\varepsilon(2.45 \text{ MeV } n) = 10 \%$$

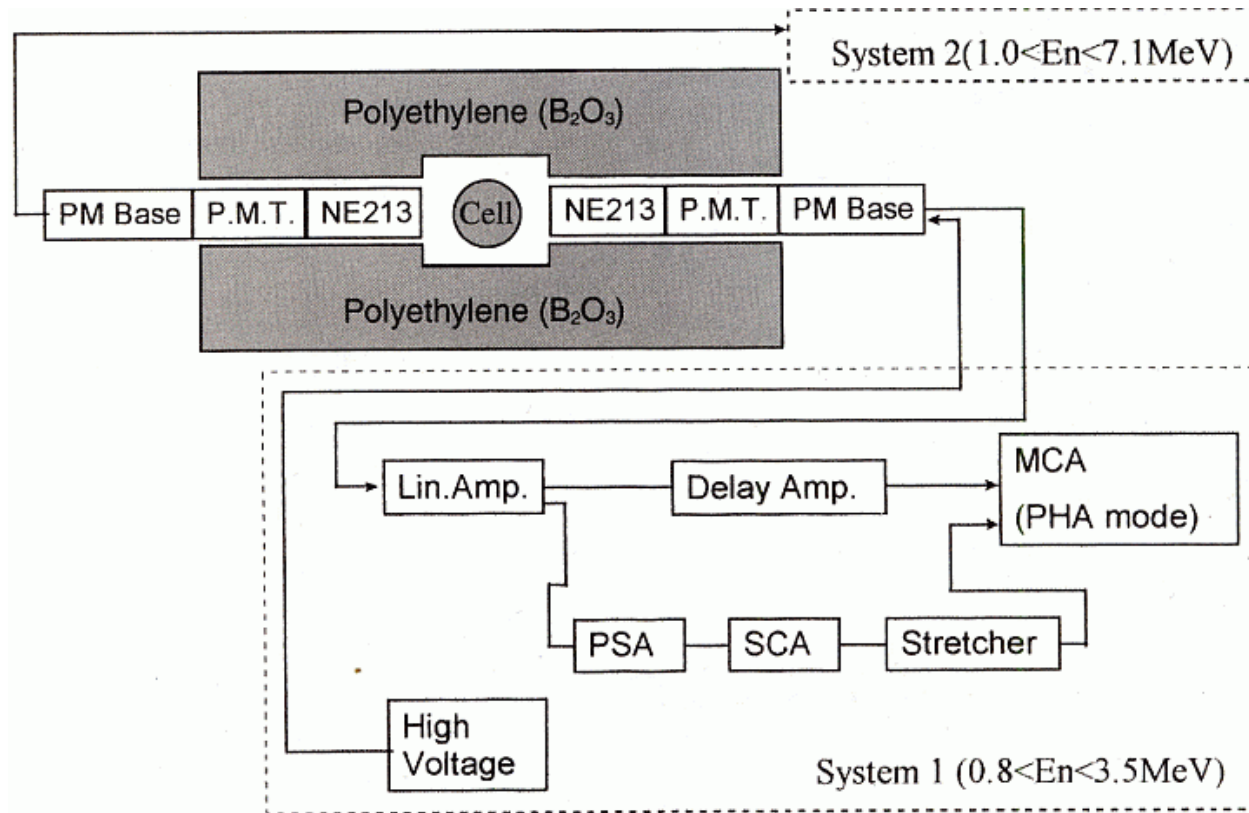
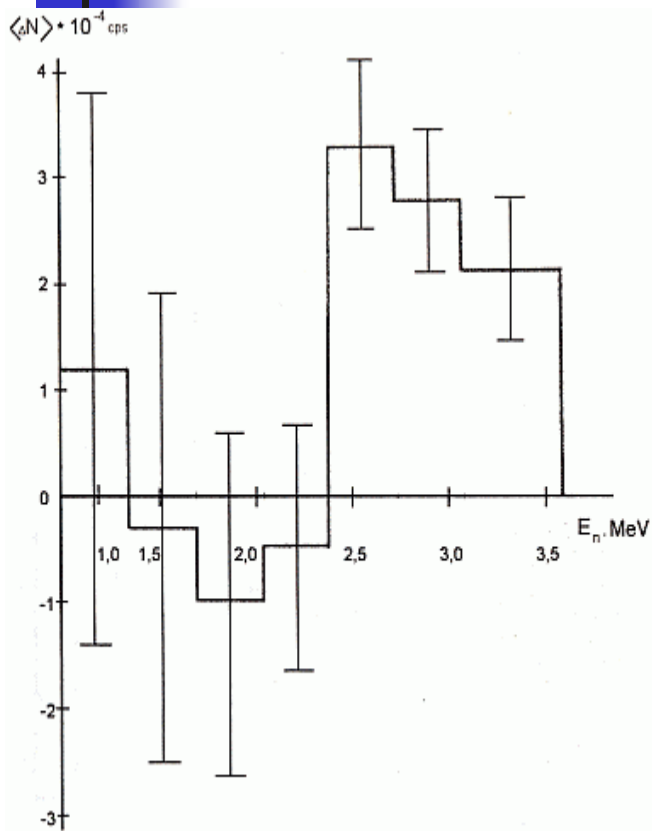
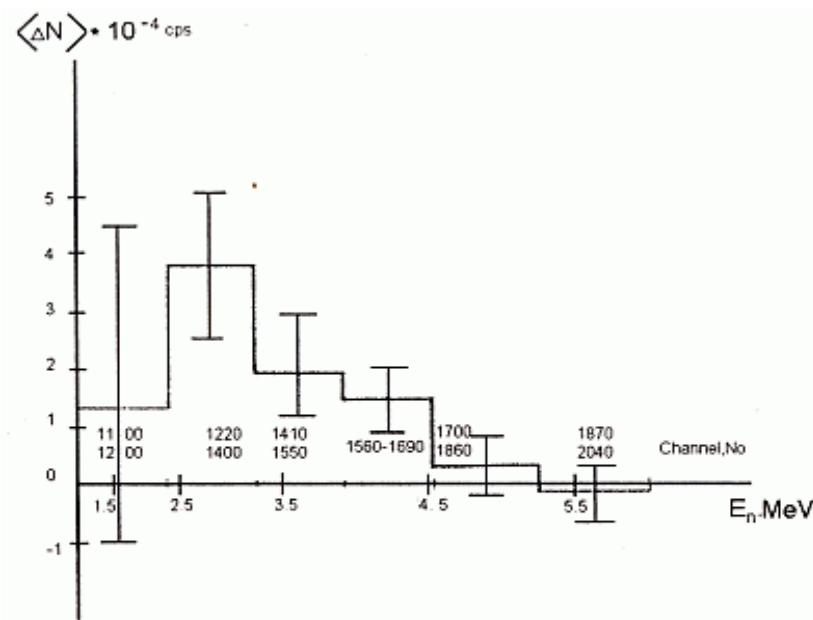


Fig. 1. The NE-213 neutron detection facility.

# Energy distributions of neutron counts from NE-213 detectors: high gain system 1 (a), low gain system 2 (b)



(a)

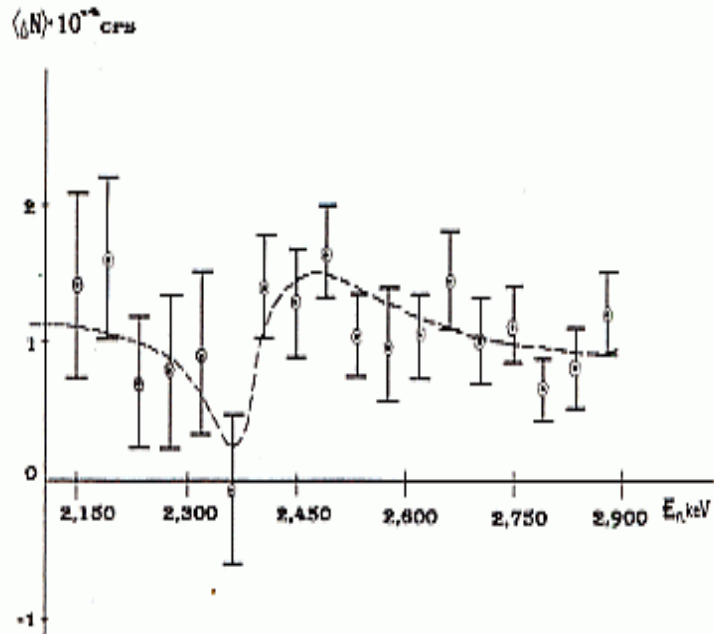


(b)

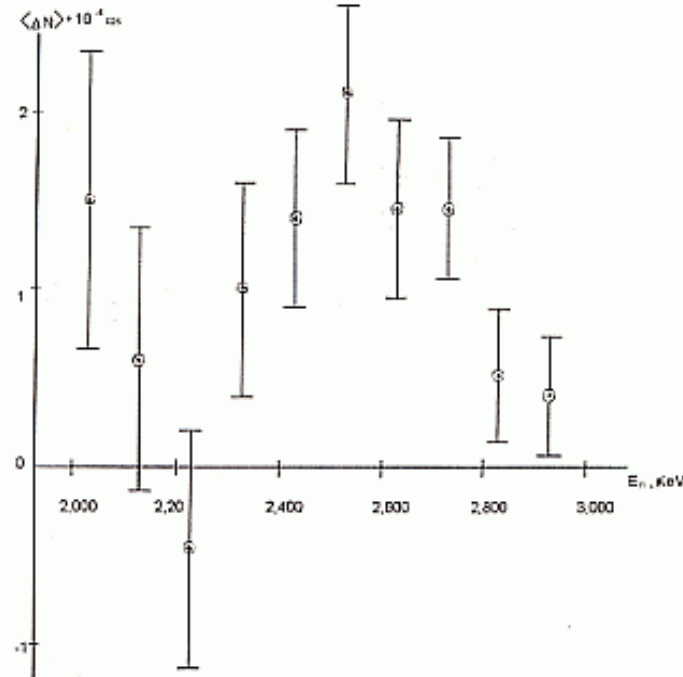


Neutron spectra of Au/Pd/PdO:Dx sample (with background subtracting) in the 2-3 MeV range derived from high gain (a) and low gain (b) NE-213 detectors:

$$I_n = (1.8 \pm 0.19) \times 10^{-2} \text{ n/s}$$



(a)



(b)



# Low Intensity emissions of DD-reaction products (p & T) and energetic alphas

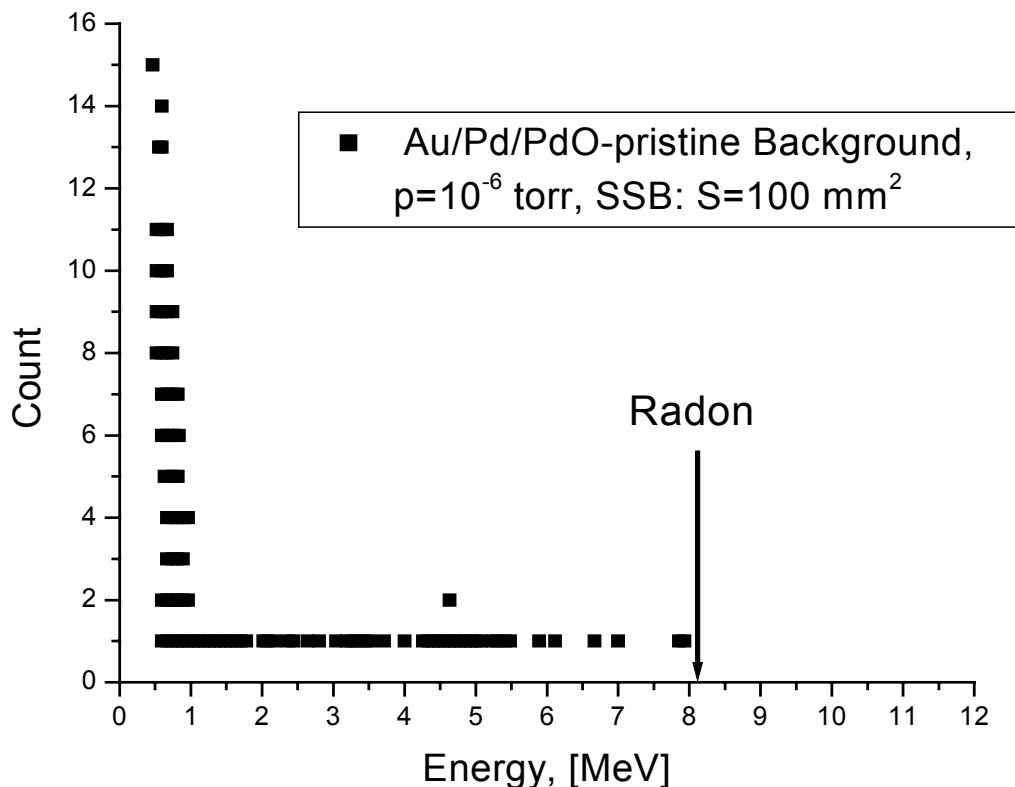
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- p-t coincidence measurements at thermal deuterium desorption from TiDx system (S.E. Jones et al, ICCF-10).
- Random 3 MeV proton emission in Pd/PdO:Dx during exothermic deuterium desorption measured with surface Si barrier detectors (A.G. Lipson et al, Fusion Tech., 2000).
- Reproducible 3 MeV proton and energetic alpha ( $E > 10$  MeV emissions in controllable D-desorption from Pd/PdO:Dx with CR-39 (Lipson et al, ICCF10-12)

# Typical charged particle background in vacuum (t=8 days in a row)

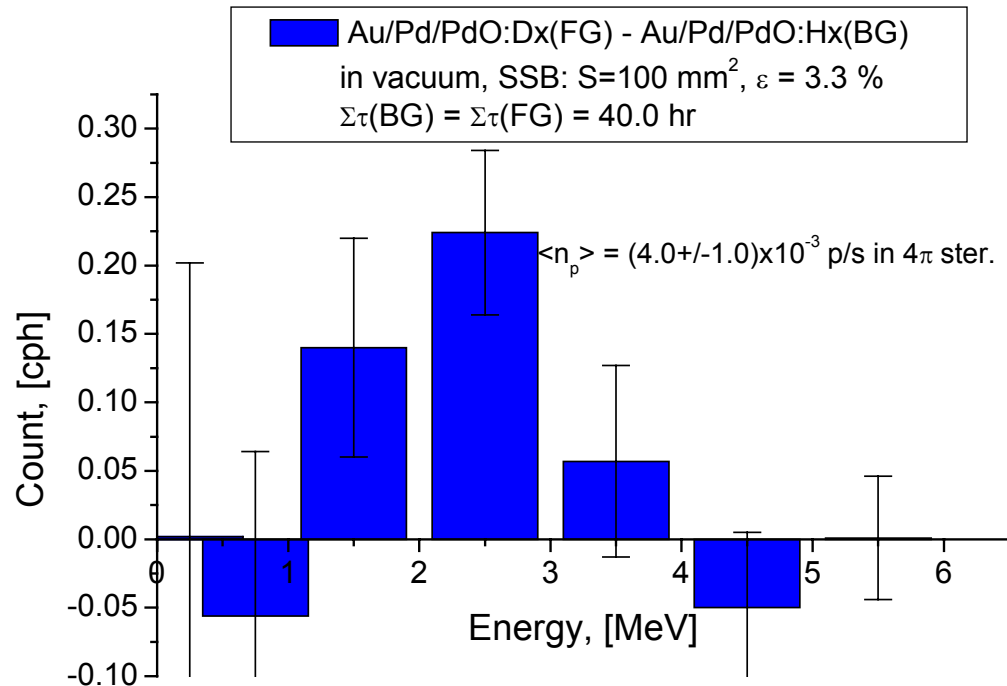
A.G. Lipson et al., Fusion Tech, **38**, 238 (2000)

Au/Pd/PdO pristine sample is in front of the SSB-1 (d=15 mm)

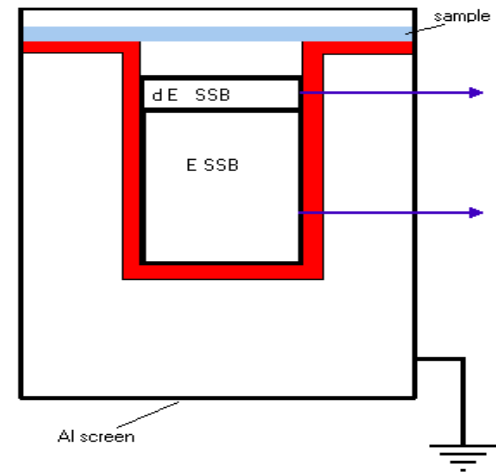
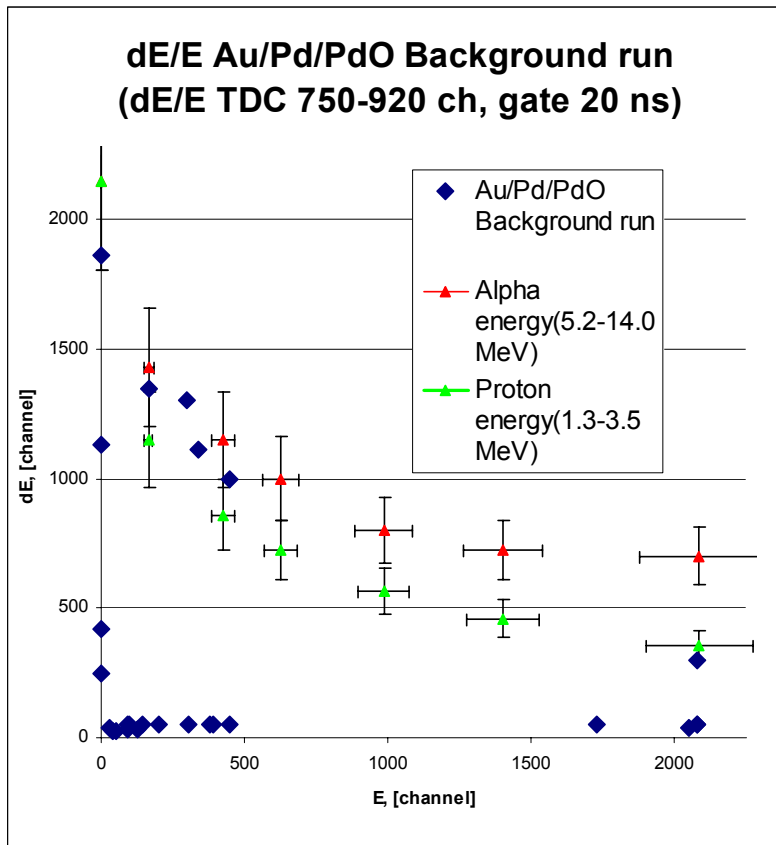


# Typical weak spontaneous DD-proton emission from Au/Pd/PdO:D<sub>x</sub> (after electrolytic D-loading) in vacuum

with Au/Pd/PdO:Hx  
Background subtracting  
SSB- measurement



# dE/E Background spectra of charged particles from pristine Au/Pd/PdO sample



Total measuring time = 500 hr.

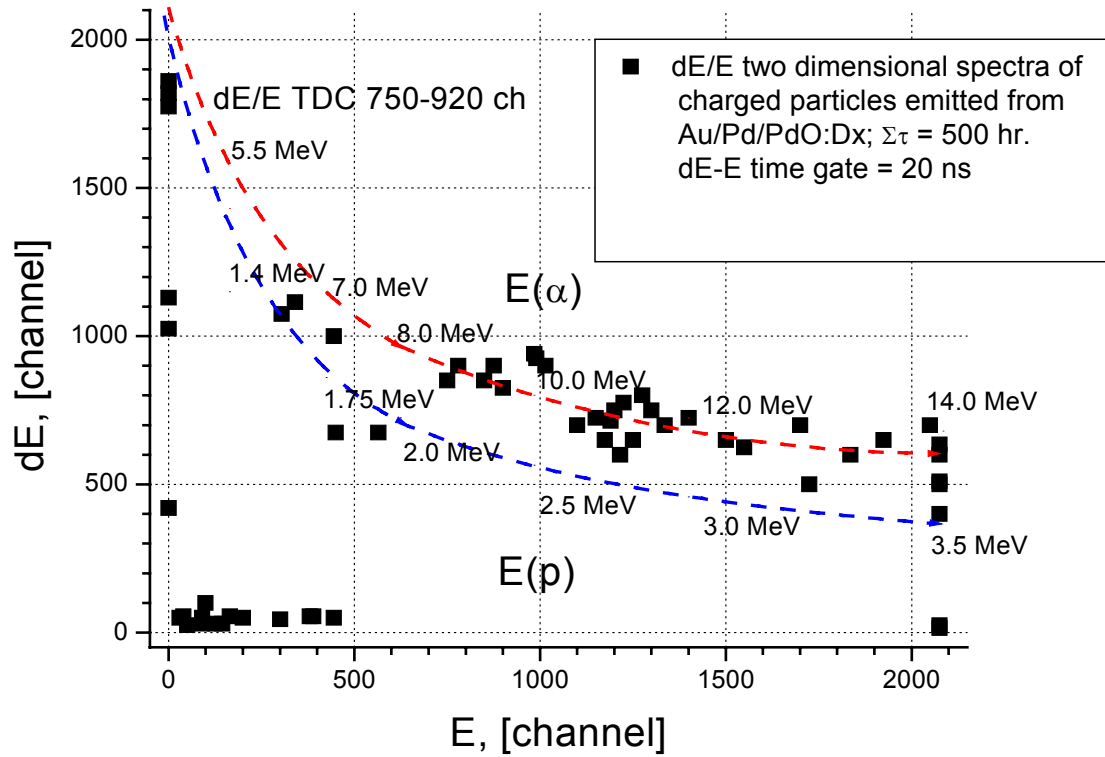
Sample is in front of the dE detector (d=10 mm).

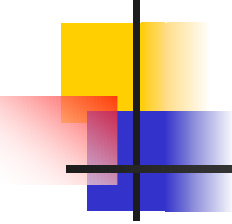
# dE/E (SSB: dE=20 $\mu$ m, E=100 $\mu$ m) 2-dimensional spectra of charged particles from Au/Pd/PdO:D<sub>x</sub> (after electrolysis)

Total measure time = 550 hr

$\langle N_{\alpha} \rangle = (6.4 \pm 1.2) \times 10^{-4} [s^{-1}]$  in  $4\pi$

Alphas:  $8 < E_{\alpha} < 14$  MeV





## Example of reproducible nuclear emissions in controlled conditions of exothermic D-desorption from PdO/Pd/PdO:Dx hetero-structure. Objectives

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- To obtain reproducible nuclear emissions in controlled conditions of exothermic D-desorption from PdO/Pd/PdO:Dx hetero-structure.
- DD-reaction yield (3.0 MeV protons)
- Energetic alphas
- Soft X-rays ?
- Control of sample temperature and D-desorption rate on-line.
- To figure out how these emissions could be linked via D-desorption



# Sample preparation

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- The samples of Pd/PdO were synthesized by thermal growing of thin oxide layer ( $\text{PdO}_y$ ) of  $\sim 20$  nm thick on top of  $110 \mu\text{m}$  thick annealed cold worked Pd foils (area  $5 \times 2 \text{ cm}^2$ ) using an oxygen-propane torch.
- Electrochemical loading of Pd/PdO cathodes by  $x = D(\text{H})/\text{Pd} = 0.7$ ;  $j \sim 20 \text{ mA/cm}^2$  in  $1\text{M-LiOD/D}_2\text{O}$  using a special cell with divided cathode and anodic spaces.
- In control experiments a similar Pd/PdO sample was electrochemically loaded with hydrogen in  $1\text{-M NaOH/H}_2\text{O}$
- The Pd/PdO:Dx sample with attached CR-39 or thermal luminescent (TLD) detectors is placed under mechanical loading ( $m=150 \text{ g}$ ) for one hour at  $T = 20 \text{ }^\circ\text{C}$





# Nuclear Detection

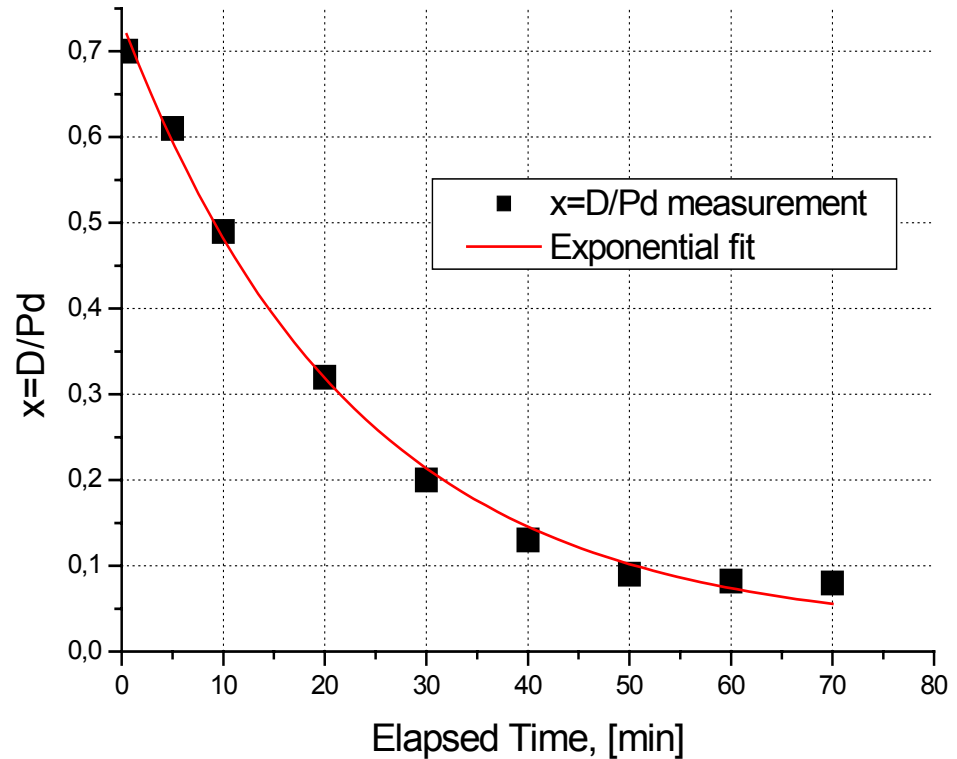
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- Charged particle detection: CR-39 (Landauer inc) with set of metal filters (Al 11-44  $\mu\text{m}$  , Cu 25  $\mu\text{m}$  thick) in order to estimate type and energy distributions of emitted particles: Time of exposure  $t = 12 \times 3600 \text{ s}$ , accordingly to maximal D-desorption rate.
- Identification of individual tracks by in-depth etching of CR-39 (etching depth in the range of 9.2 - 46  $\mu\text{m}$  (7-35 h etch in 6N-NaOH at 70° C) – 3D analysis. Comparison of track diameters with that of similarly etched calibration detectors irradiated with proton or alphas of various energy.

# Integral deuterium desorption from 110 $\mu\text{m}$ Pd/PdO heterostructure vs. elapsed time

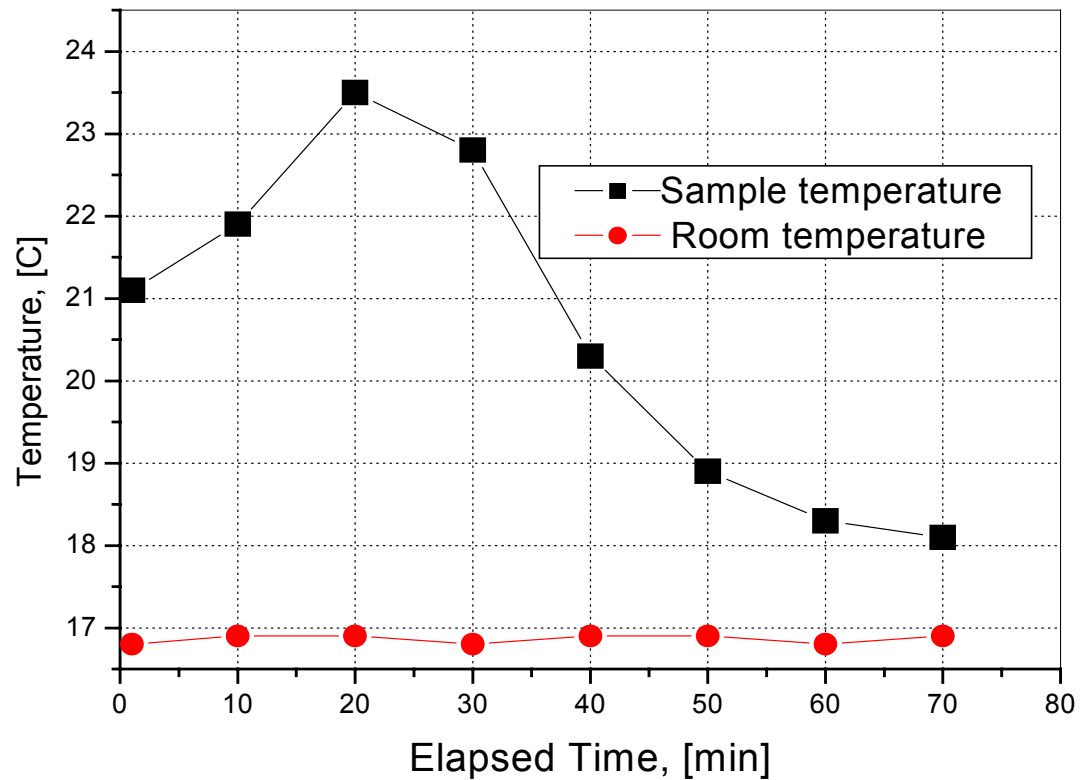
90% of  $\text{D}_2$  escape the sample in 1 hr

$$\langle dN/dt \rangle = 1 \times 10^{17} \text{ D/s-cm}^2$$

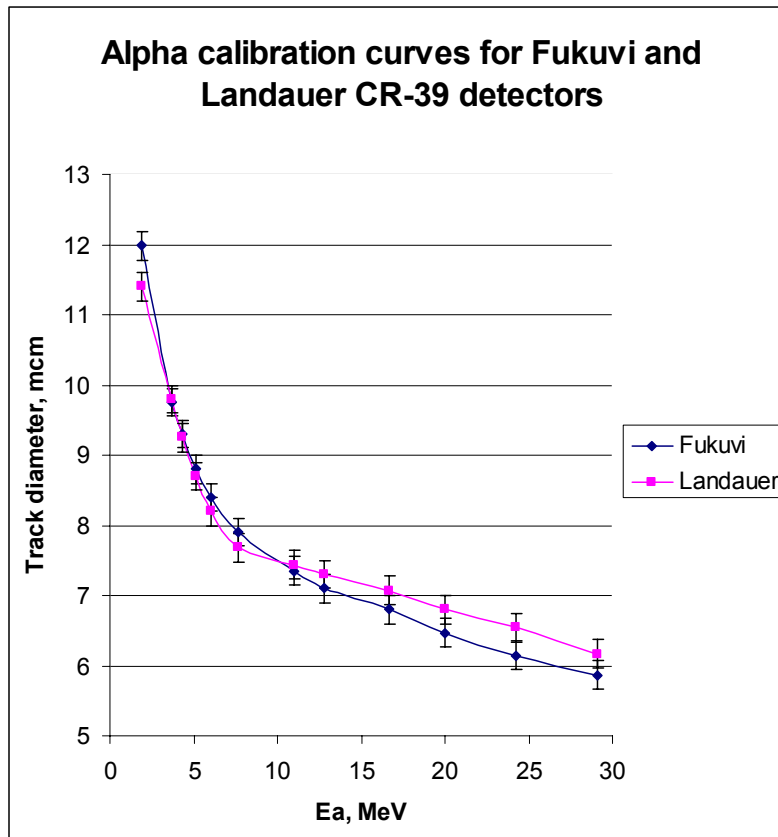


# Pd/PdO:Dx temperature vs. elapsed time

The temperature at the surface is  $\sim 7^{\circ}\text{K}$  higher than the surrounding T

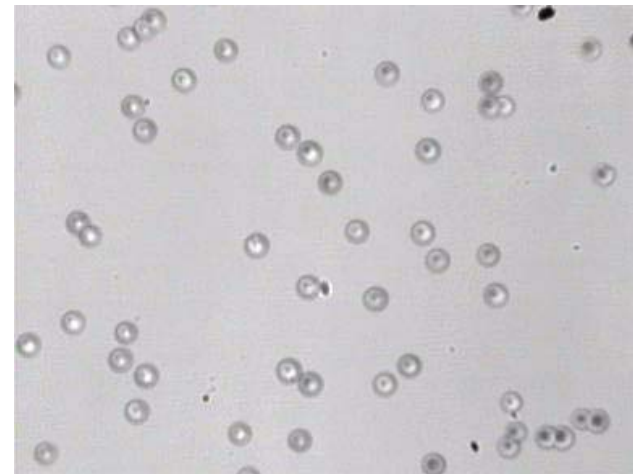


# Alpha-sources and Cyclotron alpha beam calibration (2-30 MeV) of CR-39



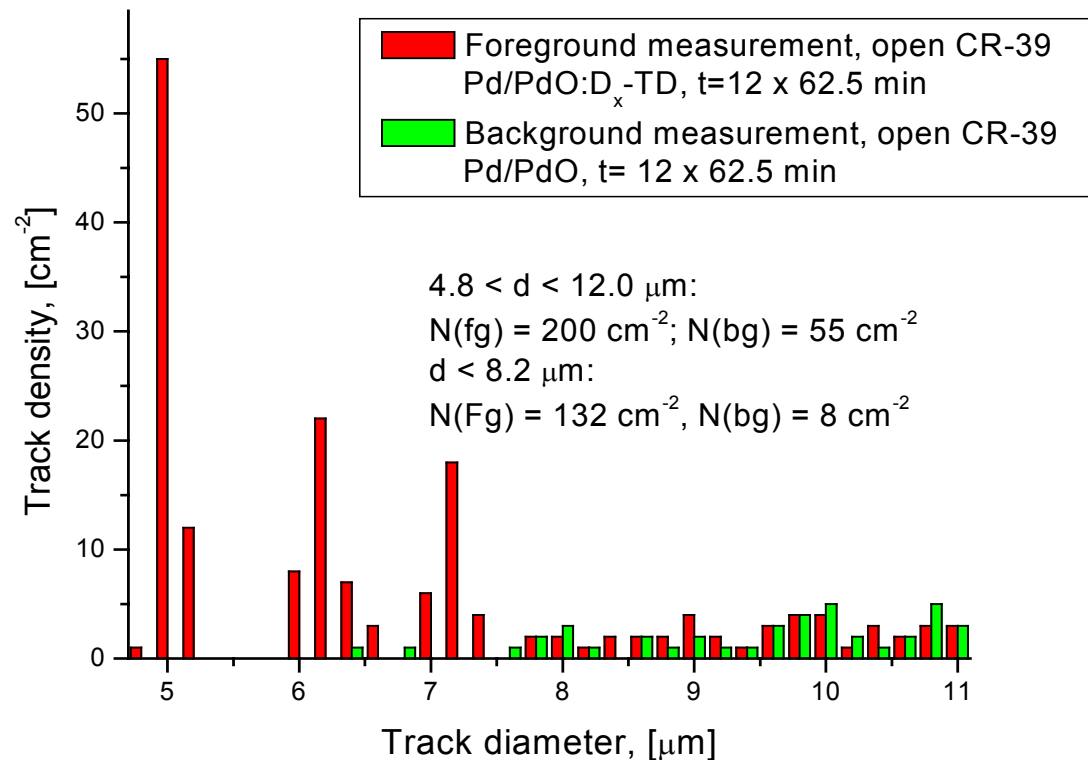
Example: Tracks from 12.0 MeV  $\alpha$ -beam @ normal incidence on CR-39 (Landauer) target

image area  $S = 0.2 \times 0.2$  mm, (X 700)- from the right



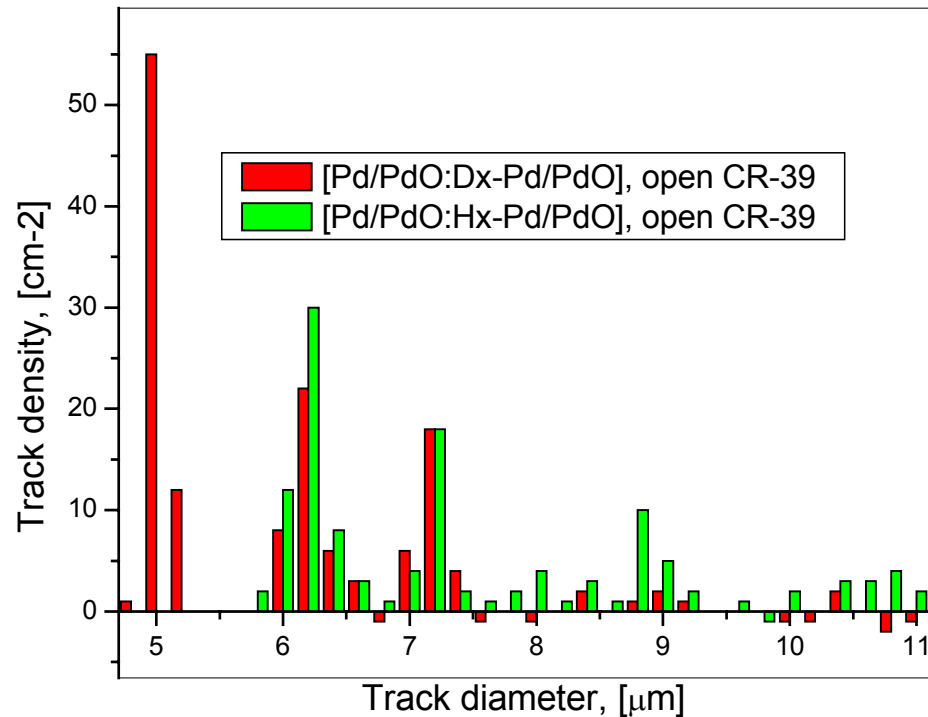
# Foreground and Background counts from Pd/PdO:Dx and Pd/PdO; open CR-39 detector

High Fg/Bg ratio,  
especially below  
 $d = 8 \mu\text{m}$



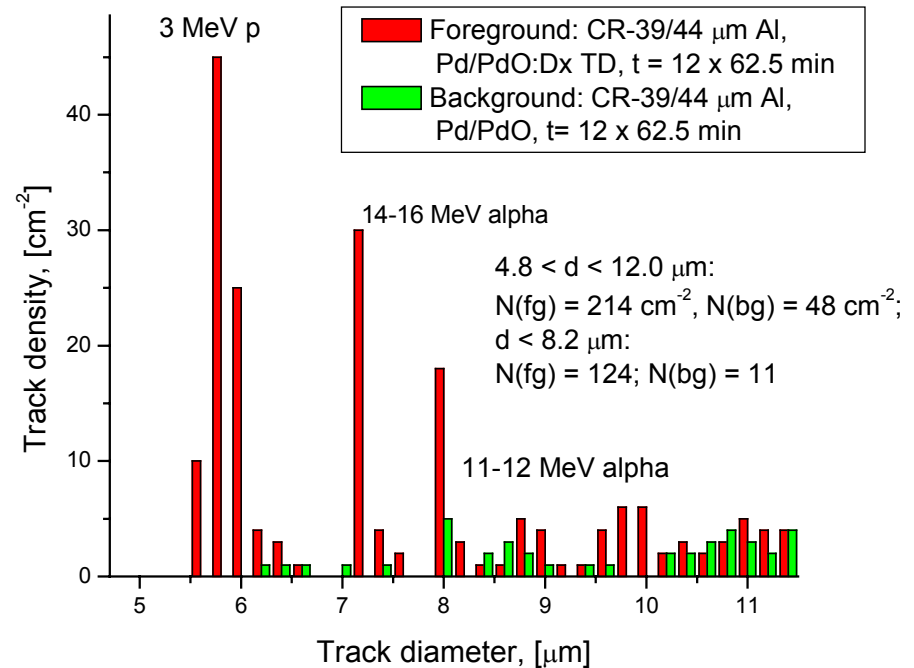
# Comparison of Pd/PdO:Dx and Pd/PdO:Hx track distribution (with Background Pd/PdO subtraction)

- Main difference is the  $\sim 5 \mu\text{m}$  peak in Pd/PdO:Dx
- No such peak in Pd/PdO:Hx



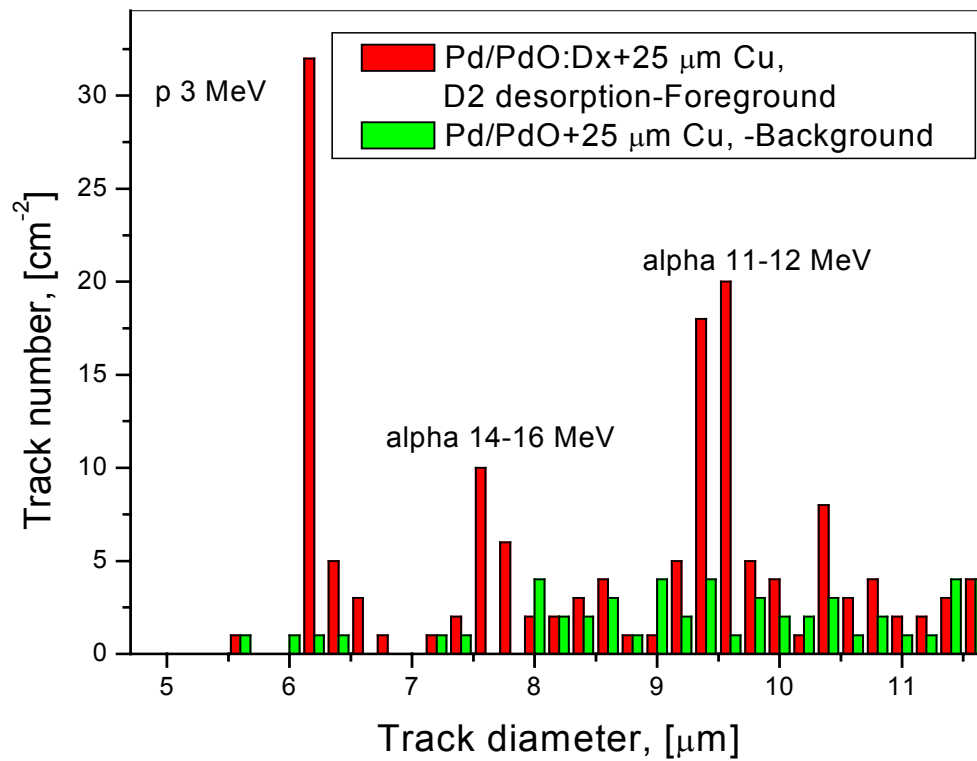
# Charged particle spectra of Pd/PdO:Dx with 44 $\mu\text{m}$ Al filter

- shift of DD-proton track band from 5 to 5.6-6.2  $\mu\text{m}$  diameters
- splitting of alpha track band from 7.2 to 7.3-8  $\mu\text{m}$  diameters
- Note large Fg/Bg ratio at  $d < 8 \mu\text{m}$



# Pd/PdO:Dx spectra with 25 $\mu\text{m}$ Cu filtered CR-39

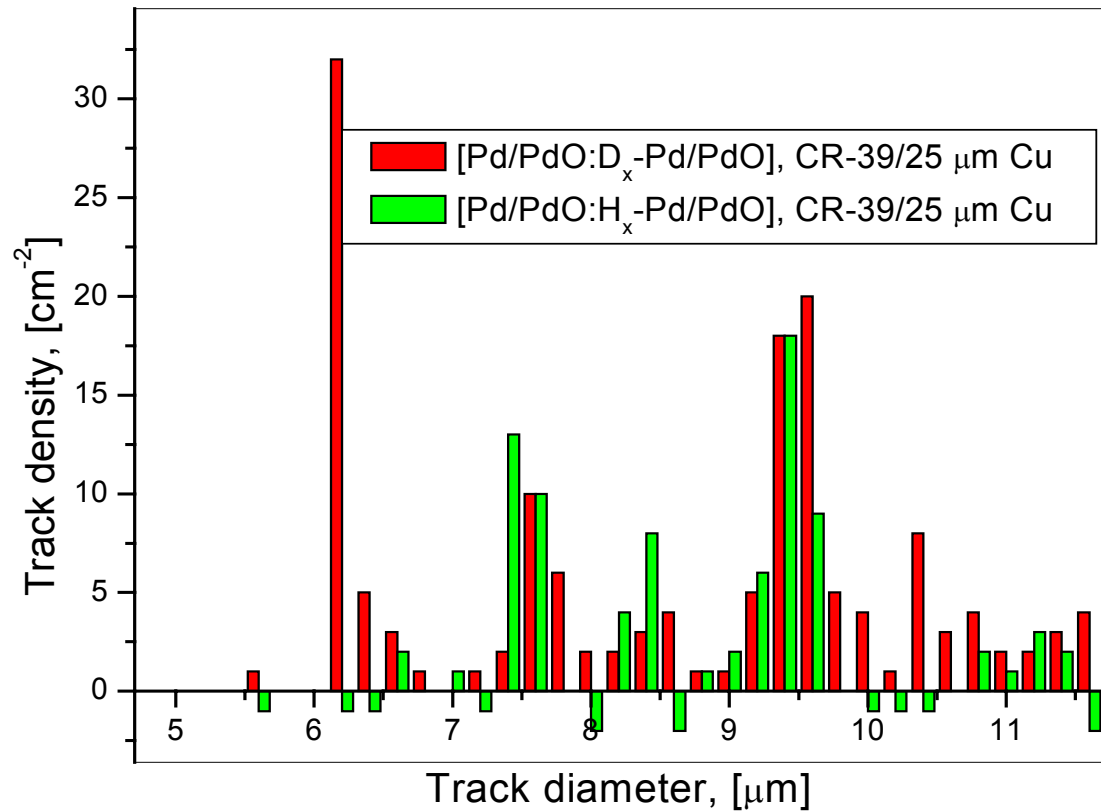
Shifts of track diameters are consistent with 3 MeV protons and 11-16 MeV alphas





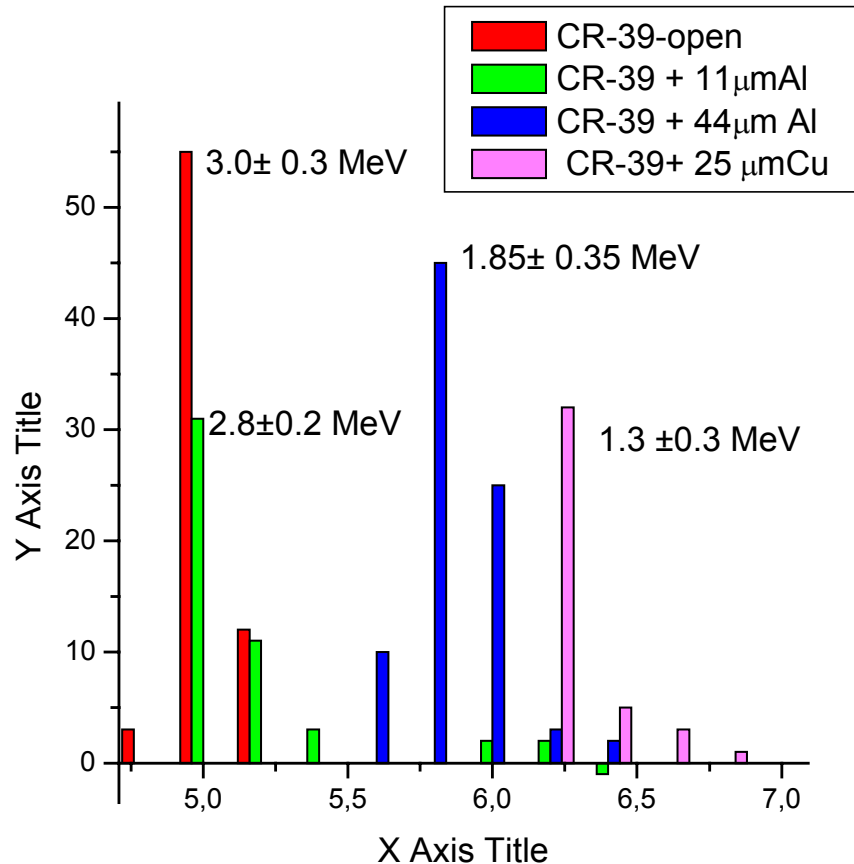
# Charged particle spectra for Pd/PdO:D<sub>x</sub> and Pd/PdO:H<sub>x</sub> filtered with 25 μm Cu (with background subtraction)

Alpha particle spectra of both H and D-loaded samples look similar



# Protons of $3\pm 0.3$ MeV incident energy from Pd/PdO:D<sub>x</sub> are consistent with their stopping power in Al and Cu filters

see shift in position of 5  $\mu\text{m}$  peak (open CR-39) to larger diameters



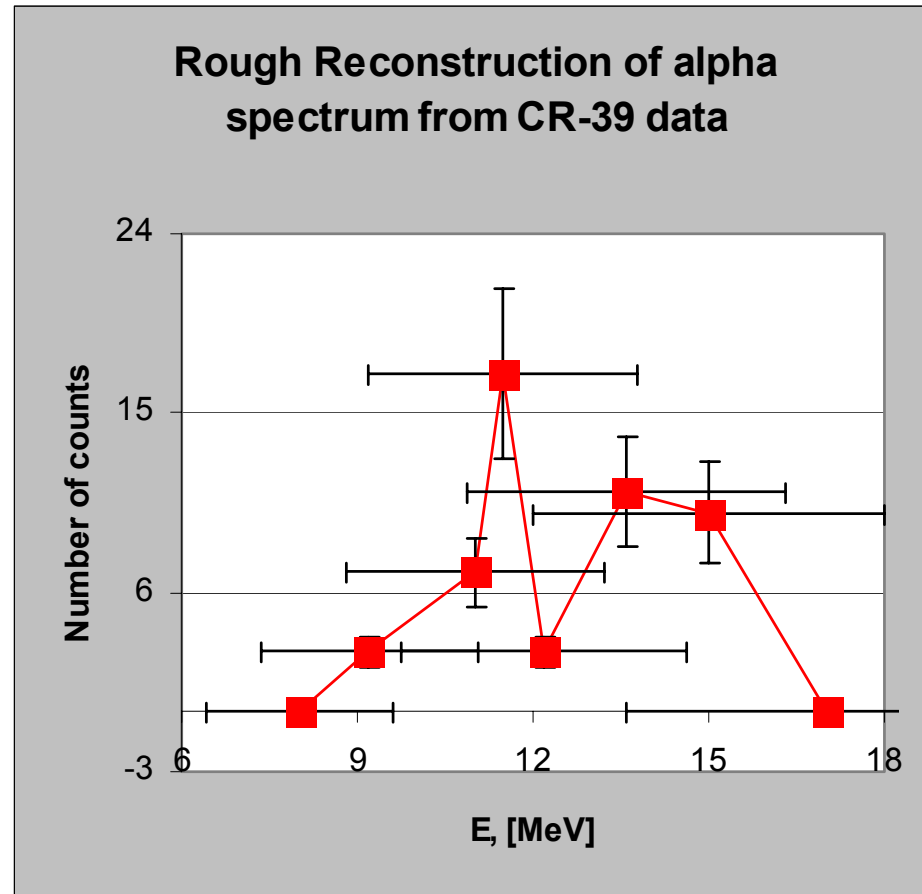


## Results on DD-reaction yield and rate

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- Exothermic D-desorption from Pd/PdO:Dx generate reproducible DD-reaction yield. Taking into account detection efficiency  $\varepsilon = 1/2(1 - \cos\Theta_c)$ , where  $\Theta_c$  – is the critical angle for 3 MeV protons (32-37°) we obtain this yield as  $Y_p(DD) = (1.15 \pm 0.13) \times 10^{-2}$  p/s in  $4\pi$  steradian.
- The yield of 3 MeV proton is in good agreement with 2.45 MeV neutron yield obtained with NE-213 detector pair for similar samples during D-desorption:  $Y_n(DD) = (1.8 \pm 0.19) \times 10^{-2}$  n/s (A. Lipson et al, Fusion Tech., . **38**, 257 (2000)).
- Calculated mean D-desorption rate in Pd/PdO:Dx or deuteron current from the sample found to be  $dN/dt = 17$  mA/cm<sup>2</sup>. The DD-reaction yield in that case was  $Y(DD) = 0.6$  p/C(D). This result suggests very high screening potential  $U_e > 1.0$  keV in Pd/PdO:Dx

# Reconstructed Energetic alpha spectra for Pd/PdO:Dx samples:



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## Energetic alpha emission results

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- Using critical angles for 11-16 MeV alphas  $\Theta_c = 42^\circ$  we obtain the yield of energetic alpha particles in Pd/PdO:Dx during exothermic D-desorption  $Y_\alpha = (5.5 \pm 0.9) \times 10^{-3} \alpha/s$  in  $4\pi$  ster.
- Pd/PdO:Hx samples during H-desorption demonstrate 11-16 MeV spectra that are very similar to that of Pd/PdO:Dx. The yield of alphas at H-desorption is also similar to D-desorption case.
- Reproducibility of both DD-reaction and energetic alpha emissions during D-desorption is close to 100%.

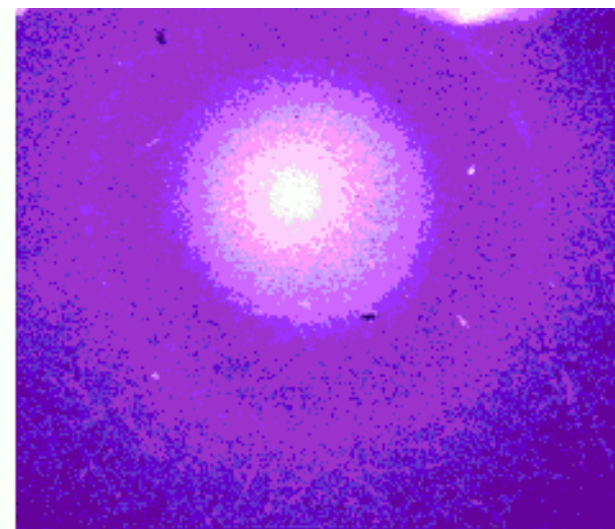
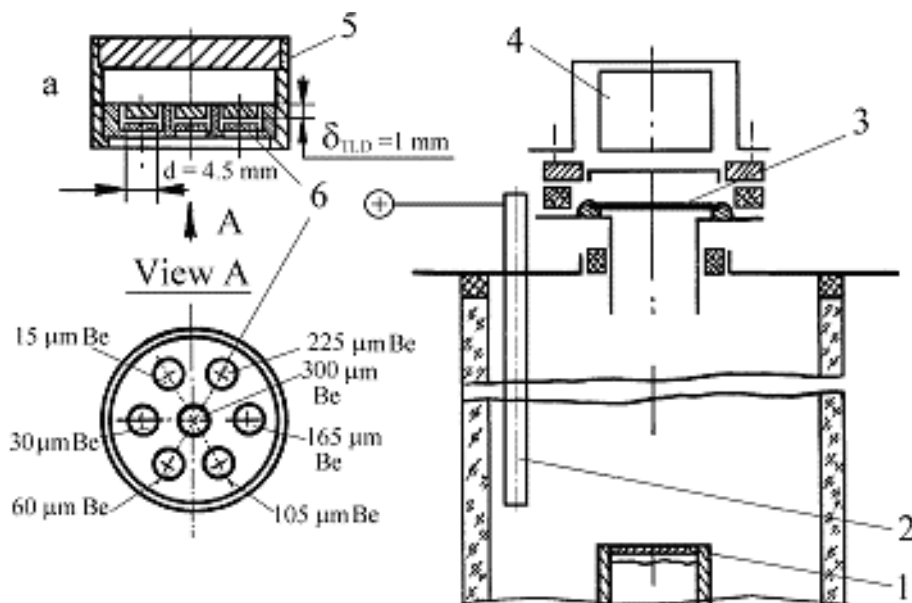


## IV. X-ray emission measurements in pulsed GD

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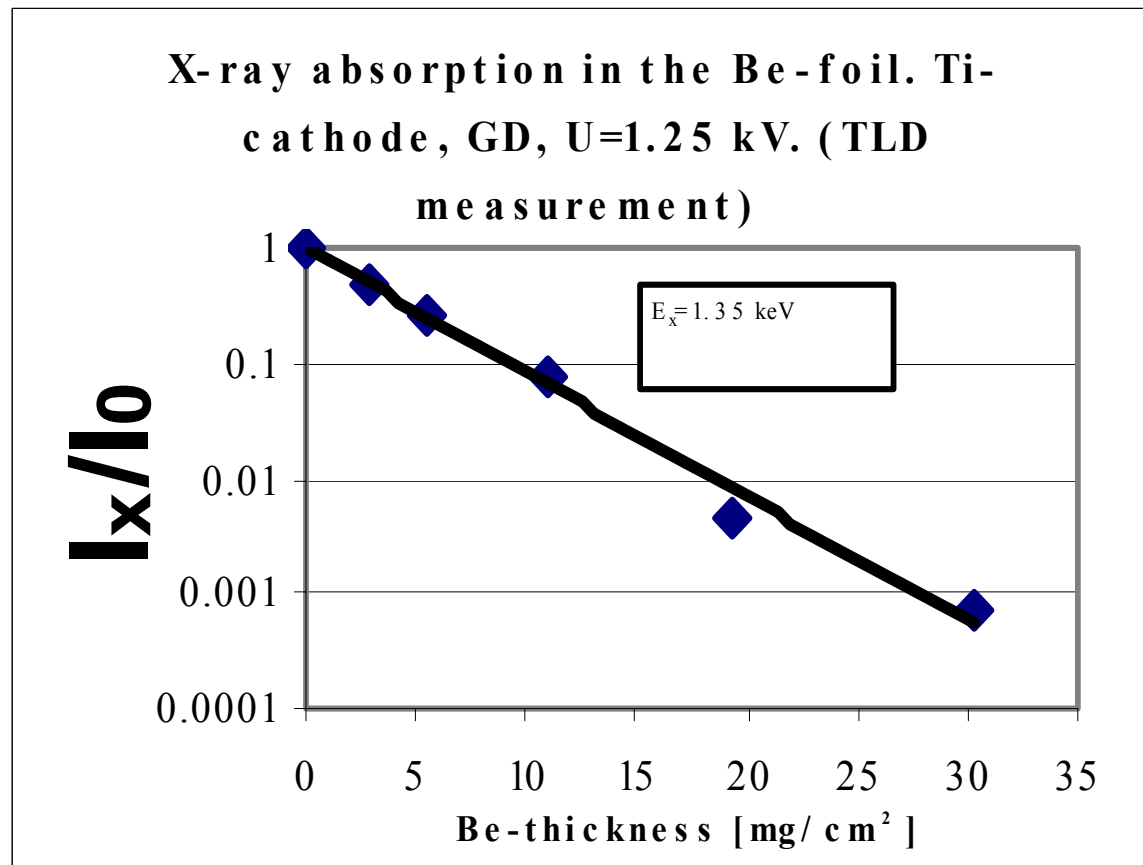
- Sensitive  $\text{Al}_2\text{O}_3:\text{C}$  TLD with a set of 15-300  $\mu\text{m}$  (2.8-55.5  $\text{mg}/\text{cm}^2$ ) thick Be-foils, pin-hole camera; plastic scintillator
- Seven TLD, 5 mm diameter each were located 70 mm from the back side of anode, outside of discharge zone.
- In special experiments to obtain the exact position of X-ray source in GD, Mo-anode was shifted 20 mm with respect to cathode.
- TLD calibration with a standard  $\text{Cs}^{137}$   $\gamma$ -source

using the pin-hole camera. The objective of 0.3 mm diameter is narrowed by use of a 15  $\mu\text{m}$  Be shield in front of the camera. (The image is a positive imprint)



$d_{\text{cathode}}$

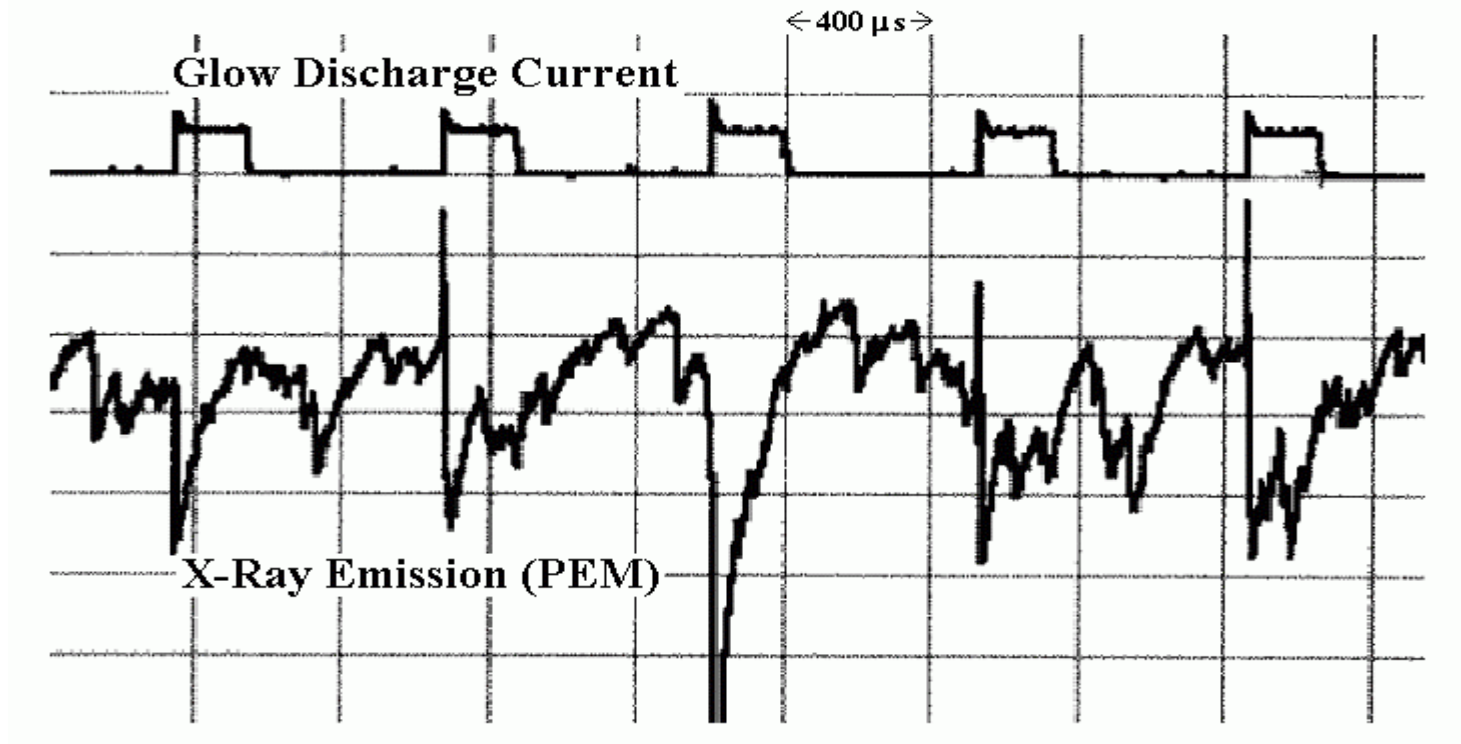
# X-ray energy measurement in Ti/D<sub>2</sub> PGD Karabut's type (A. Lipson et al, JETP, 2005)



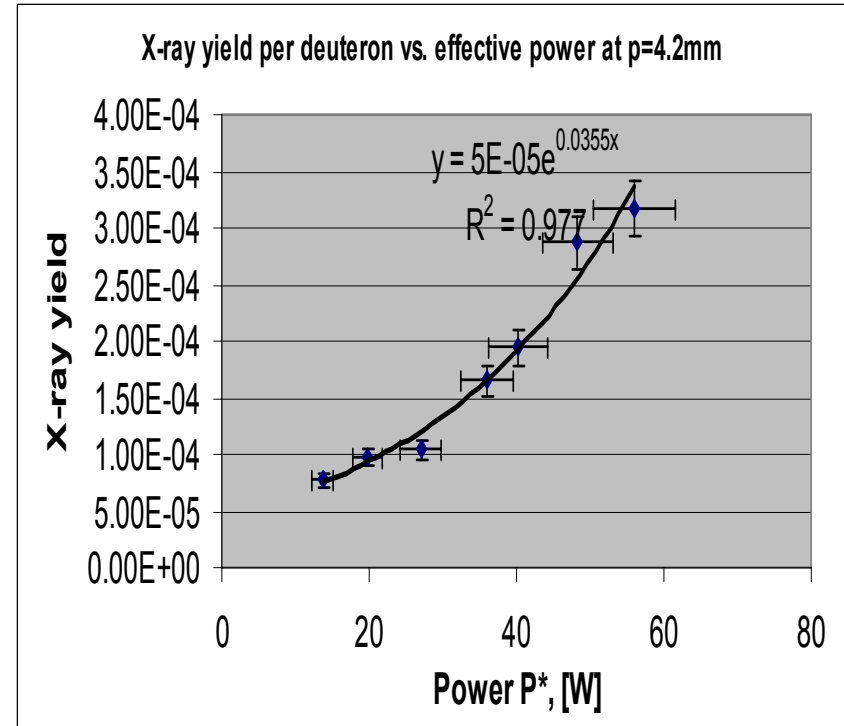
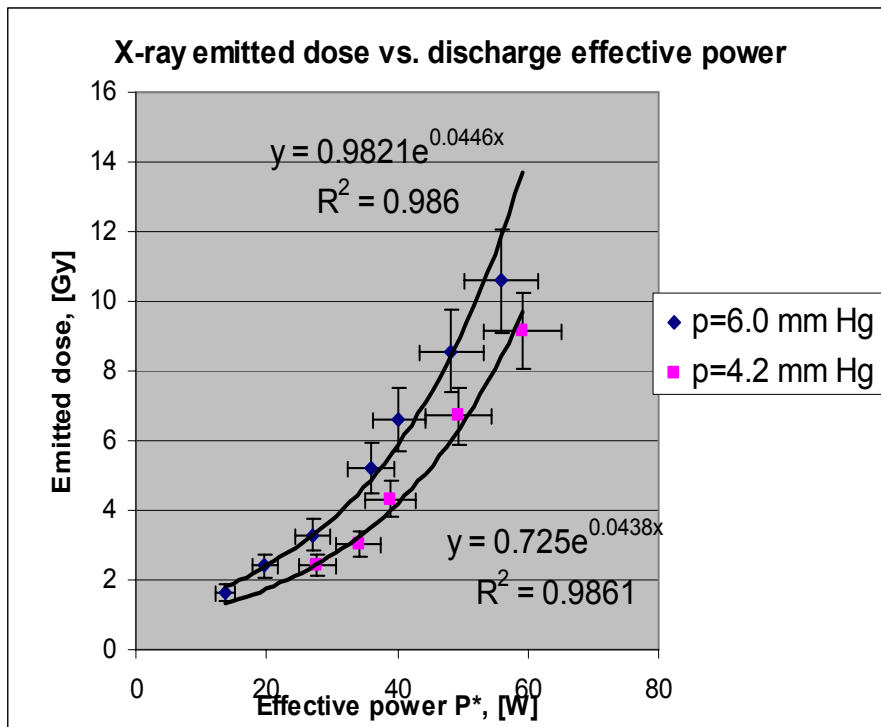
ICCF-13, Sochi, 06/25-07/01,  
2007



# Correlation between X-ray and current pulses in GD

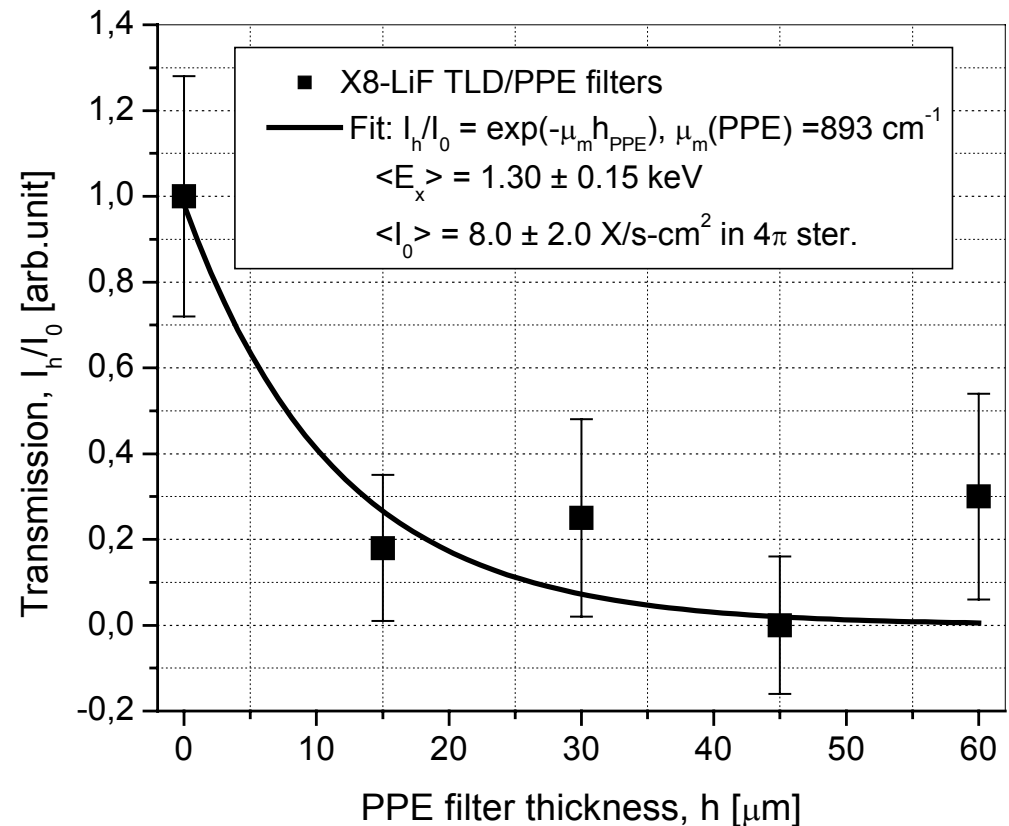


# Emitted doses and X-ray yield from Ti-cathode vs. Effective power $P^* = UIQ$ ; Yield is described by a formula: $I_x = I_0 \exp[(\varepsilon/kT_m)P^*_x/P^*_0]$



# X- ray quanta energy in Pd/PdO:D<sub>x</sub> foils after electrolysis estimated with 5 pairs of X8 TLD with 0-60 μm PPE filters

- X-ray detection: photo-insensitive X8 LiF TLD (Landauer) units (S= 2x2 mm<sup>2</sup>, h=1 mm) open and filtered by 1-4 layers of 15 μm polypropylene (PPE)
- 100 runs of 1-48 hr duration
- 50 μm Pd/PdO:D sample
- TLD reading was carried out in Landauer inc.



# X-ray dosimetry and flux estimate

- The open TLD showed dose  $D = 12 \pm 2$  mrad = 120  $\mu$ Gy. In SI system the equation connecting the dose  $D_x$  [Gy] absorbed by LiF TLD with area  $s = 0.04$  cm<sup>2</sup> and mean flux  $\langle \Phi_x \rangle$  would be written:

$$\langle \Phi_x \rangle = 2D_x \rho(\text{LiF}) / 1.6 \times 10^{-13} [\text{J/MeV}] \times E_x \times \mu_m \times (s/1\text{cm}^2) \times \tau \cong (8 \pm 2) \text{ X-quanta/cm}^2\text{-s,}$$

where  $\rho(\text{LiF})$  – is the density of LiF,  $E_x = 1.3$  keV is the X-ray quanta energy and  $\tau = 7 \times 10^5$  s – is the total time of TLD exposure with Pd/PdO:Dx sample.

The estimated dose obtained from charged particle absorption by LiF TLD is less than 1 mrad (below detection limit).

# Conclusions for controlled D-desorption from Pd/PdO:D<sub>x</sub> experiment

- Good reproducibility of charged particle emissions during GD operation and controlled exothermic D-desorption from Pd/PdO:D<sub>x</sub>
- Relatively high 3 MeV proton yield indicating large DD-reaction enhancement during D-desorption
- No direct sign of 1 MeV tritons: means that effective depth where DD-reaction originates is  $\sim 2 \mu\text{m}$  from the surface. This estimate is in agreement with energy losses for 3 MeV protons
- Identity between energetic alpha spectra during D and H-desorption from Pd/PdOD(H)

# Conclusions II

- The energy of X-ray quanta emitted from Pd/PdO:Dx is in good agreement with Karabut's glow discharge results. Suggests similar mechanism caused by D-desorption
- Observed nuclear effects in metals cannot be explained by fracto-fusion because the electric field in the cracks would be too low. Phonon energy of D-desorption focusing or/and concentration in some specific lattice sites near surface (the sites of a high internal strain ?)
- DD-reaction, energetic alphas and X-ray emissions suggest anomalous energy release via "active" lattice sites of non-equilibrium metal deuterides, indicating intra-atomic electric fields of  $E \sim 10^{10}$  V/cm

Entire results showed that Exothermic D-desorption is that obvious link between DD-screening, soft X-ray emission and high energy alpha generation from the surface of Pd/PdO:Dx

# Possible mechanism - speculations



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- Applied energy focusing/concentration in some specific lattice sites near surface (the sites of a high internal strain ?)
- Coherent energy transfer from DD-reaction sites to the light nuclei (P.L. Hagelstein)
- Effective acceleration of these nuclei (p, d and  $^4\text{He}$ ) by intra-atomic electric fields
- ECP emissions suggest anomalous energy release via the "active" lattice sites of non-equilibrium metal deuterides/hydrides

# How to convince physicists in nuclear origin of Excess Heat effect ?

- Complete experiment with simultaneous detection of excess heat, atomic  $^4\text{He}$ ,  $^3\text{T}$ , charged particles (DD-products +energetic alphas) and neutron emissions, as well as soft X-rays ( $E_x \leq 2.0$  keV), not characteristic  $K_\alpha$  of Pd)
- Search for correlations between excess heat events and emissions of atomic, nuclear species and X-rays
- Special electrolytic cells and appropriate state-of-the-art calorimetric and nuclear detection equipment.
- To use novel (nanostructured/nanolayered) samples as the cathodes (Pd-SWCNT-Pd, Pd-Re-Pd and PdO-Pd-PdO) with enhanced charged particle yield.