PAUL SCHERRER INSTITUT

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Superheavy elements copernicium and flerovium selenides: First Cn-Se bond observation

TASCA 15 – 14th Workshop on Recoil Separator for Superheavy Element Chemistry
GSI, Darmstadt – October 23, 2015

The Superheavy Elements Cn and Fl

IUPAC Periodic Table of the Elements

Key: atomic number / Symbol / name / standard atomic weight

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
1 H hydrogen [1.007, 1.009]																	2 He helium 4.003
3 Li lithium [6.938, 6.997]	4 Be beryllium 9.012											5 B boron [10.80, 10.83]	6 C carbon [12.00, 12.02]	7 N nitrogen [14.00, 14.01]	8 O oxygen [15.99, 16.00]	9 F fluorine 19.00	10 Ne neon 20.18
11 Na sodium 22.99	12 Mg magnesium [24.30, 24.31]											13 Al aluminium 26.98	14 Si silicon [28.08, 28.09]	15 P phosphorus 30.97	16 S sulfur [32.05, 32.08]	17 Cl chlorine [35.44, 35.46]	18 Ar argon 39.95
19 K potassium 39.10	20 Ca calcium 40.08	21 Sc scandium 44.96	22 Ti titanium 47.87	23 V vanadium 50.94	24 Cr chromium 52.00	25 Mn manganese 54.94	26 Fe iron 55.85	27 Co cobalt 58.93	28 Ni nickel 58.69	29 Cu copper 63.55	30 Zn zinc 65.38(2)	31 Ga gallium 69.72	32 Ge germanium 72.63	33 As arsenic 74.92	34 Se selenium 78.96(3)	35 Br bromine [79.90, 79.91]	36 Kr krypton 83.80
37 Rb rubidium 85.47	38 Sr strontium 87.62	39 Y yttrium 88.91	40 Zr zirconium 91.22	41 Nb niobium 92.91	42 Mo molybdenum 95.96(2)	43 Tc technetium	44 Ru ruthenium 101.1	45 Rh rhodium 102.9	46 Pd palladium 106.4	47 Ag silver 107.9	48 Cd cadmium 112.4	49 In indium 114.8	50 Sn tin 118.7	51 Sb antimony 121.8	52 Te tellurium 127.6	53 I iodine 126.9	54 Xe xenon 131.3
55 Cs caesium 132.9	56 Ba barium 137.3	57-71 lanthanoids	72 Hf hafnium 178.5	73 Ta tantalum 180.9	74 W tungsten 183.8	75 Re rhenium 186.2	76 Os osmium 190.2	77 Ir iridium 192.2	78 Pt platinum 195.1	79 Au gold 197.0	80 Hg mercury 200.6	81 Tl thallium [204.3, 204.4]	82 Pb lead 207.2	83 Bi bismuth 209.0	84 Po polonium	85 At astatine	86 Rn radon
87 Fr francium	88 Ra radium	89-103 actinoids	104 Rf rutherfordium	105 Db dubnium	106 Sg seaborgium	107 Bh bohrium	108 Hs hassium	109 Mt meitnerium	110 Ds darmstadtium	111 Rg roentgenium	112 Cn copernicium		114 Fl flerovium		116 Lv livermorium		
		57 La lanthanum 138.9	58 Ce cerium 140.1	59 Pr praseodymium 140.9	60 Nd neodymium 144.2	61 Pm promethium	62 Sm samarium 150.4	63 Eu europium 152.0	64 Gd gadolinium 157.3	65 Tb terbium 158.9	66 Dy dysprosium 162.5	67 Ho holmium 164.9	68 Er erbium 167.3	69 Tm thulium 168.9	70 Yb ytterbium 173.1	71 Lu lutetium 175.0	
		89 Ac actinium	90 Th thorium 232.0	91 Pa protactinium 231.0	92 U uranium 238.0	93 Np neptunium	94 Pu plutonium	95 Am americium	96 Cm curium	97 Bk berkelium	98 Cf californium	99 Es einsteinium	100 Fm fermium	101 Md mendelevium	102 No nobelium	103 Lr lawrencium	

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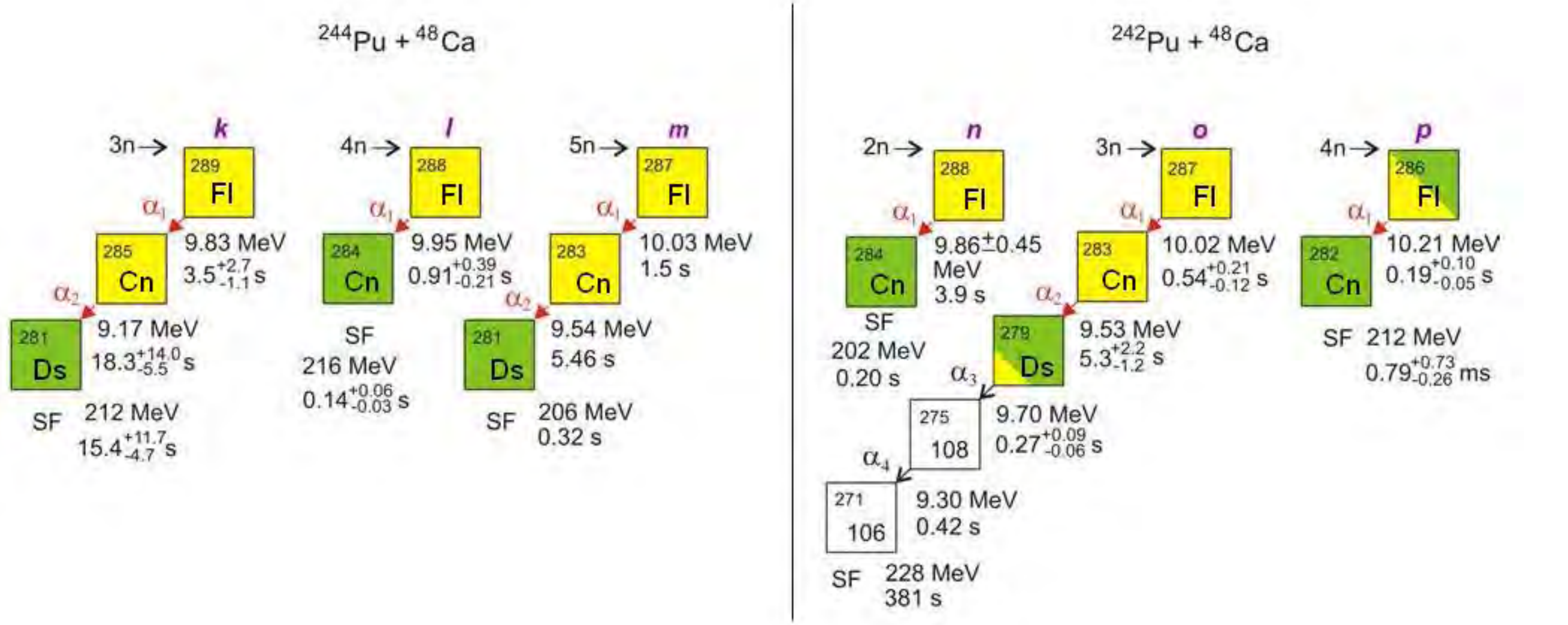
For updates to this table, see iupac.org/reports/periodic_table/. This version is dated 1 May 2013.

Production and identification of Cn and Fl



The possibility of **simultaneous production** of Cn and Fl gives the chance of perform **comparative studies**.

Adapted from OGANESSIAN, Y., et al. "Synthesis of nuclei of the superheavy element 114 in reactions induced by ^{48}Ca". Nature, 1999, 400.6741: 242-245.

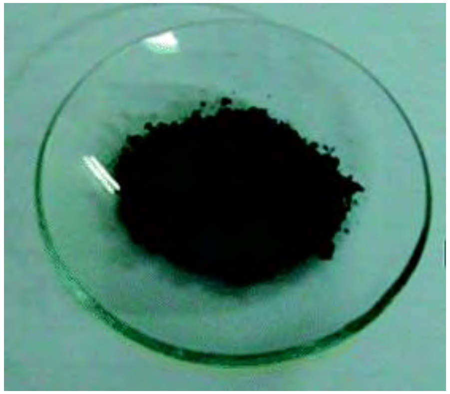
Selenium as stationary surface

VON SZENTPÁLY L., "Predictions of the Sublimation Enthalpies of Group 12 Chalcogenides and the Formation Enthalpies of their Polonides", The Journal of Physical Chemistry A, 2008, 112.49: 12695-12701; HAYNES W.M., LIDE D.R., BRUNO T.J., "CRC Handbook of Chemistry and Physics 2012-2013", CRC press; BOONE S., KLEPPA O.J., "Enthalpies of formation for Group IV selenides ($GeSe_2$, $GeSe_{2(am)}$, SnSe, $SnSe_2$, PbSe) by direct-combination drop calorimetry", Thermochimica Acta, 1992, 197. 1, 109-121; P.A.G. O'HARE, SUSMAN S., VOLIN K.J., "Thermochemistry of germanium monoselenide, and the GeSe bond dissociation enthalpy", The Journal of Chemical Thermodynamics, 1989, 21.8: 827-836.

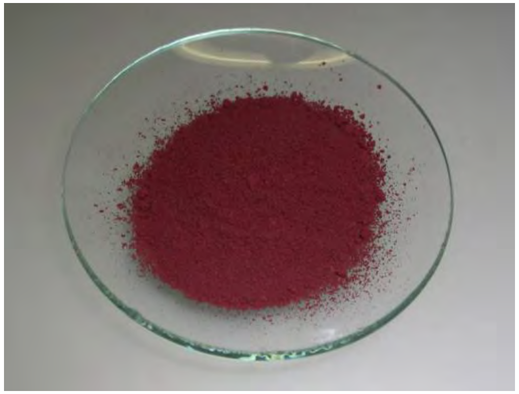
Se major allotropes

trigonal

monoclinic (α-, β-, γ-Se)

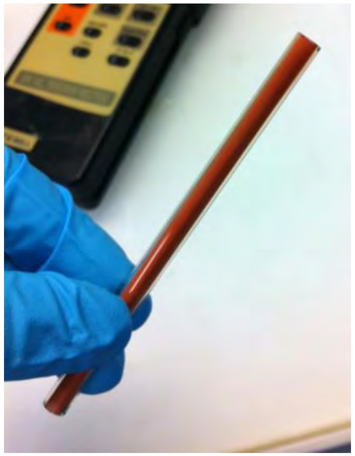


black amorphous Se

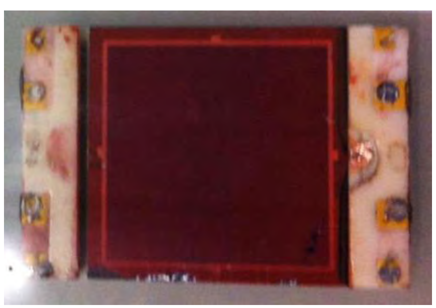


red amorphous Se

Red a-Se covered columns and PIN diodes

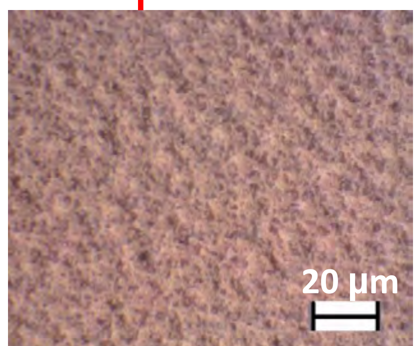


red a-Se coated column

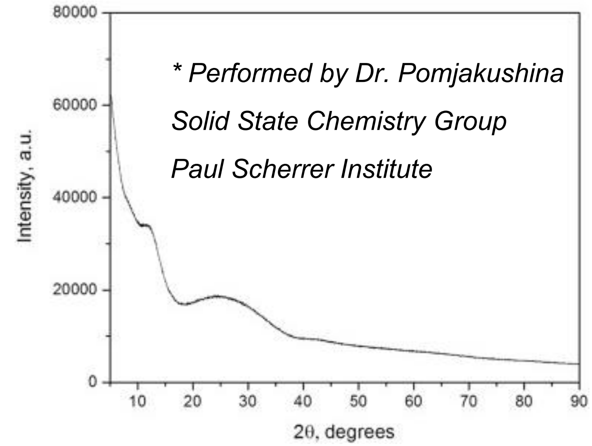


red a-Se coated PIN diode

X-ray diffraction

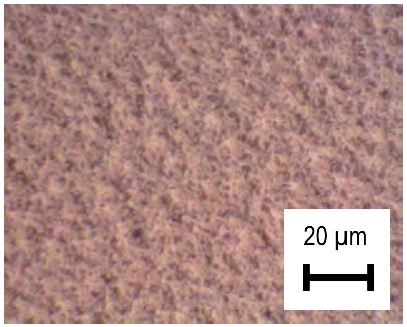


alpha spectroscopy



N.M. Chiera, R. Eichler, A. Vögele, A. Türler, Vapor deposition coating of fused silica tubes with amorphous selenium, Thin Solid Films (2015).

Trigonal Se covered columns and PIN diodes

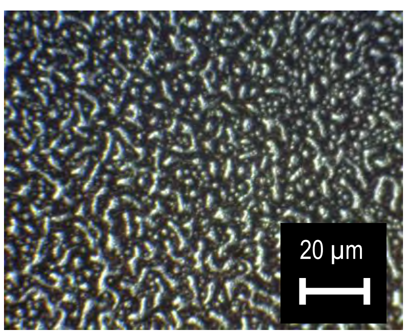


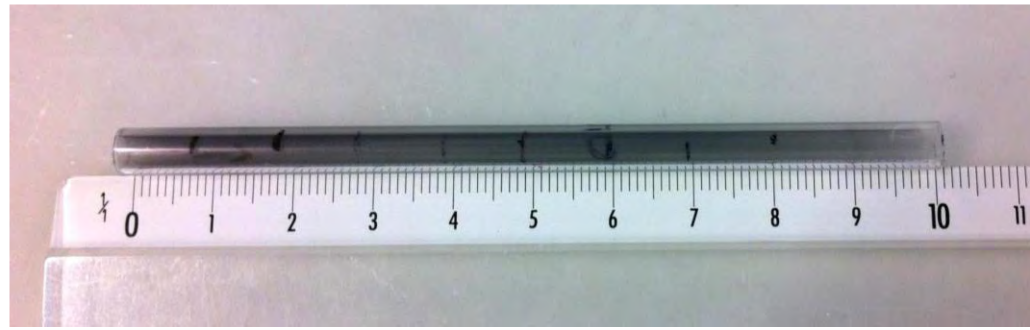
step 1: 100 °C

step 2: 110 °C

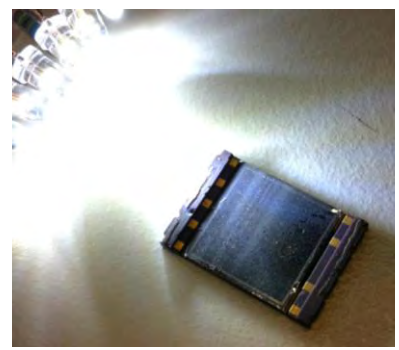
step 3: 150 °C

step 4: CS_2 dissolution





trigonal Se coated column



trigonal Se coated PIN diode

Different allotrope, diverse reactivity

Isothermal chromatography, T = 23 °C

Preliminary results

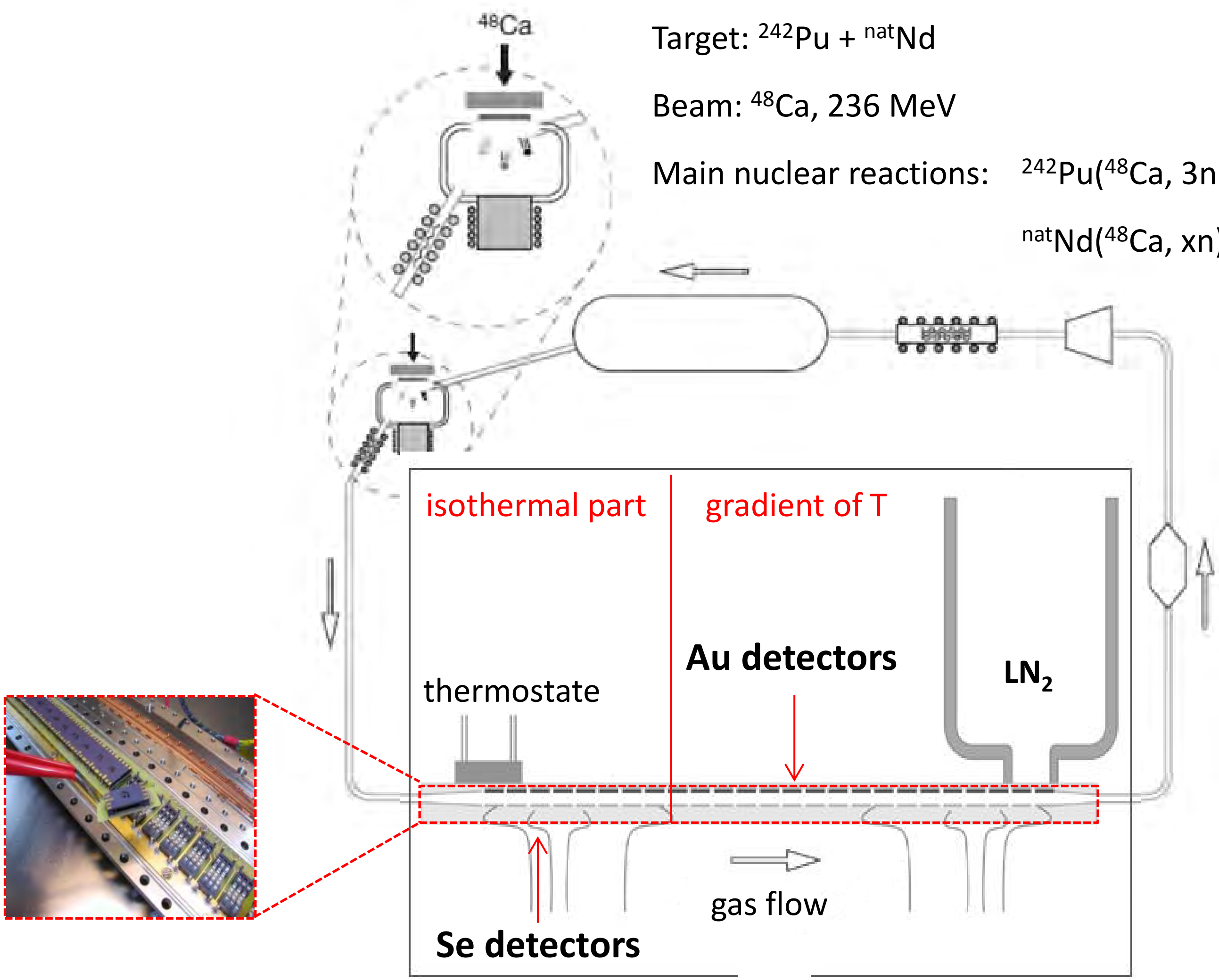
On-line experiments

Target: $^{242}Pu + {}^{nat}Nd$

Beam: ^{48}Ca, 236 MeV

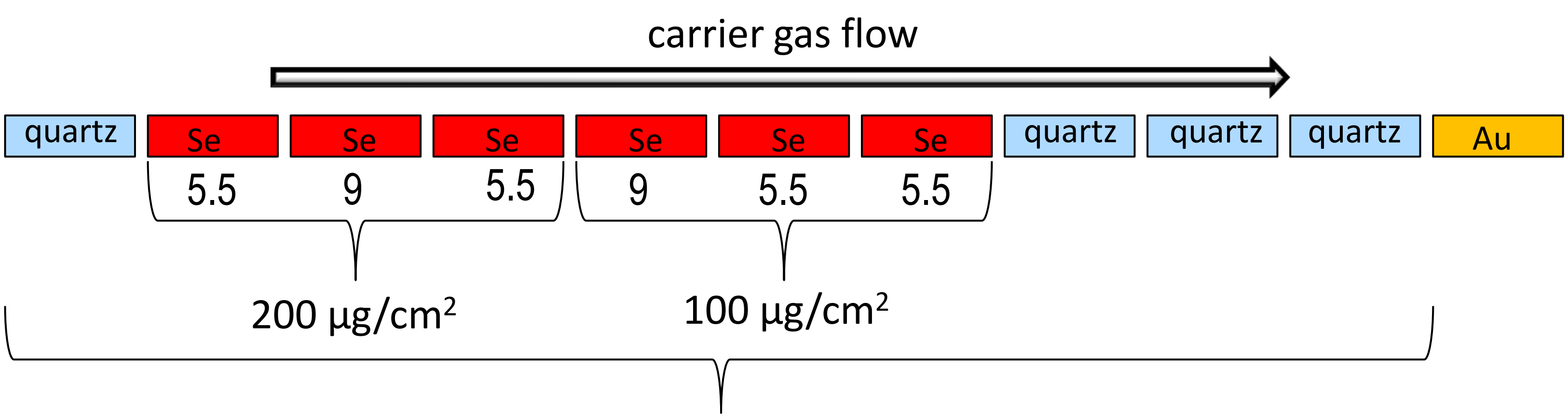
Main nuclear reactions: $^{242}Pu(^{48}Ca, 3n)^{287}Fl \xrightarrow{\alpha} {}^{283}Cn$

$^{nat}Nd(^{48}Ca, xn)^{185}Hg$



Red a-Se covered PIN diodes

- 5 diodes **100 µg/cm²** in thickness
  - 2 diodes aged **9 weeks**
  - 3 diodes aged **5.5 weeks**
- 6 diodes **200 µg/cm²** in thickness
  - 3 diodes aged **9 weeks**
  - 3 diodes aged **5.5 weeks**

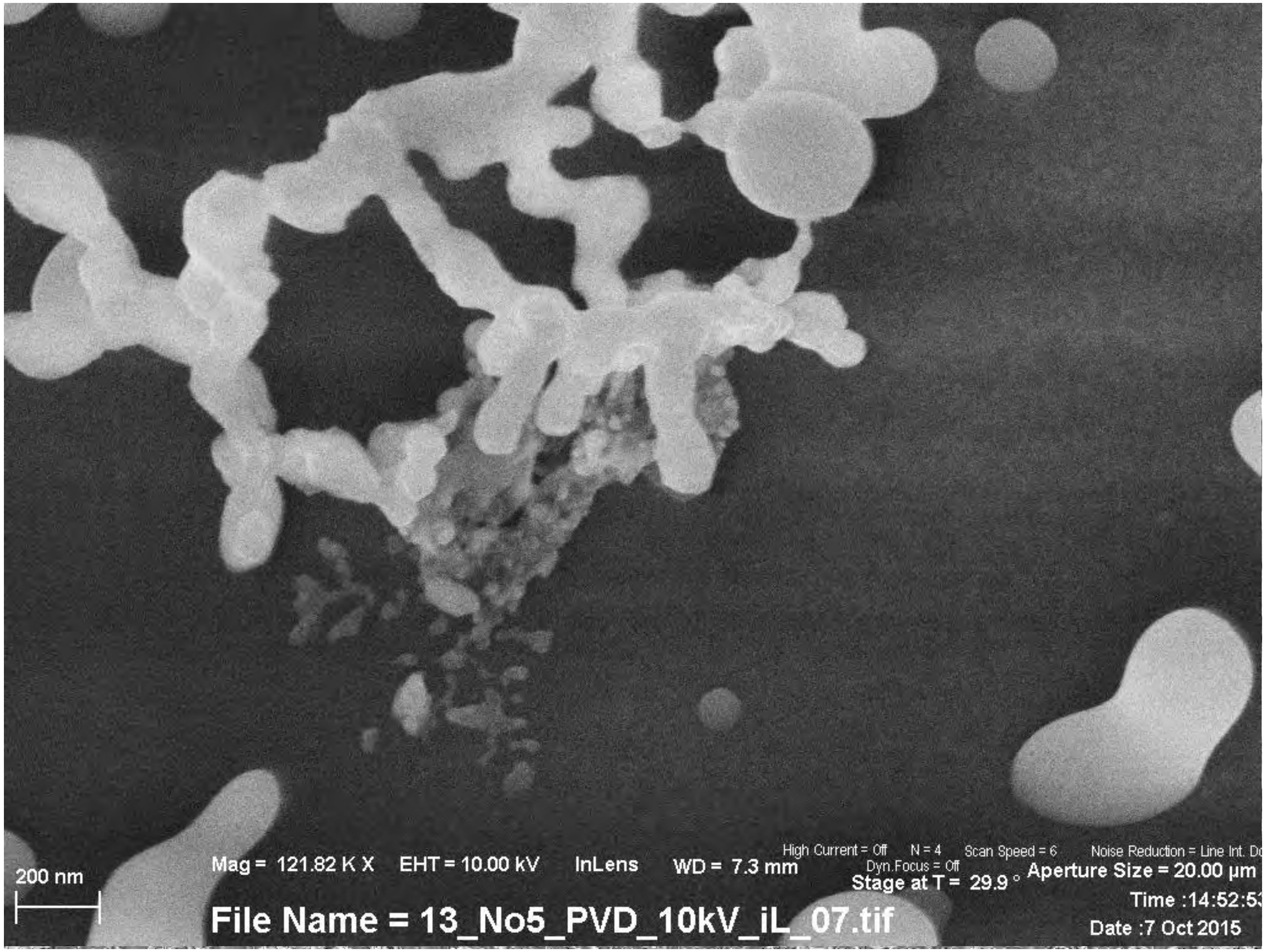


First 10 detectors, isothermal part of the COLD array

^{185}Hg deposition pattern on Se and Au detectors

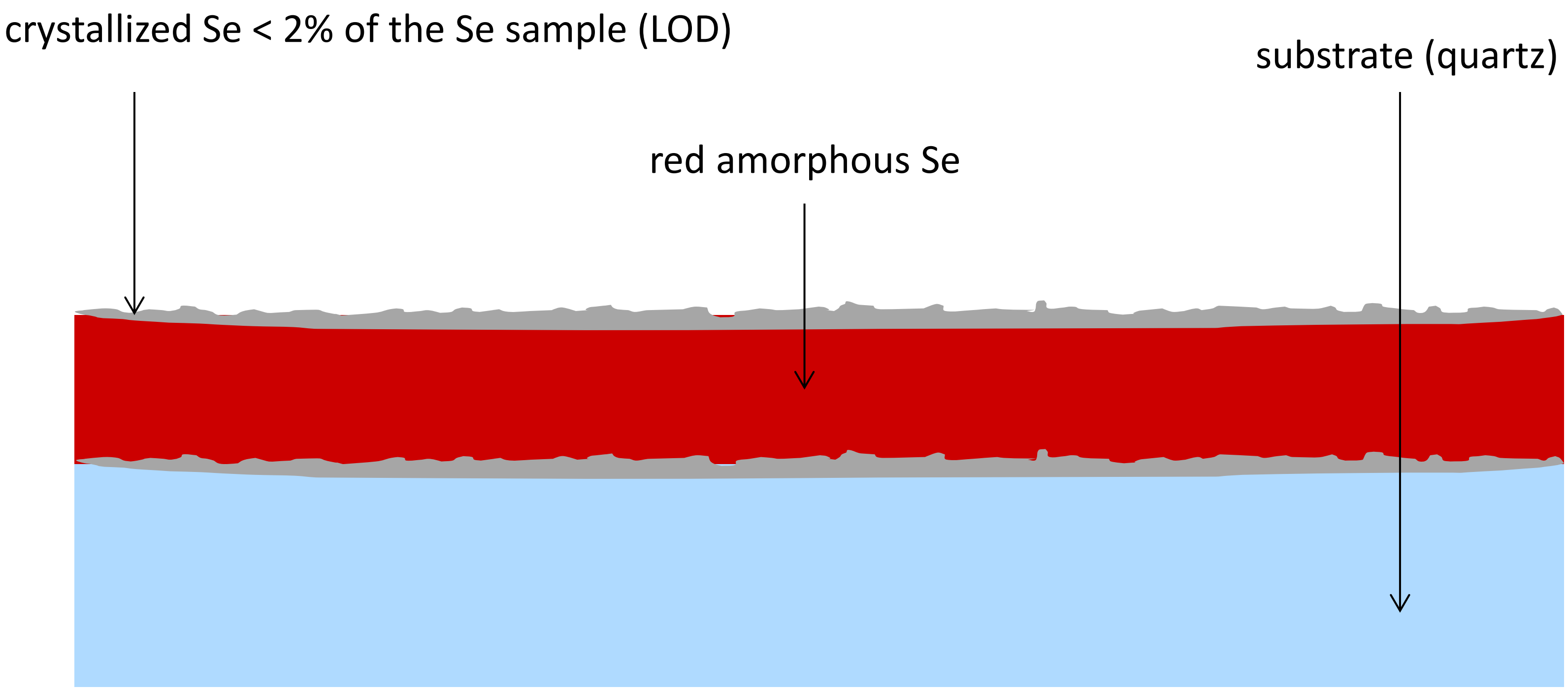
Preliminary results

Se surface scanning electron microscopy (SEM)



Se surface X-ray diffraction (XRD)

The Se crystallization occurs at the interfaces

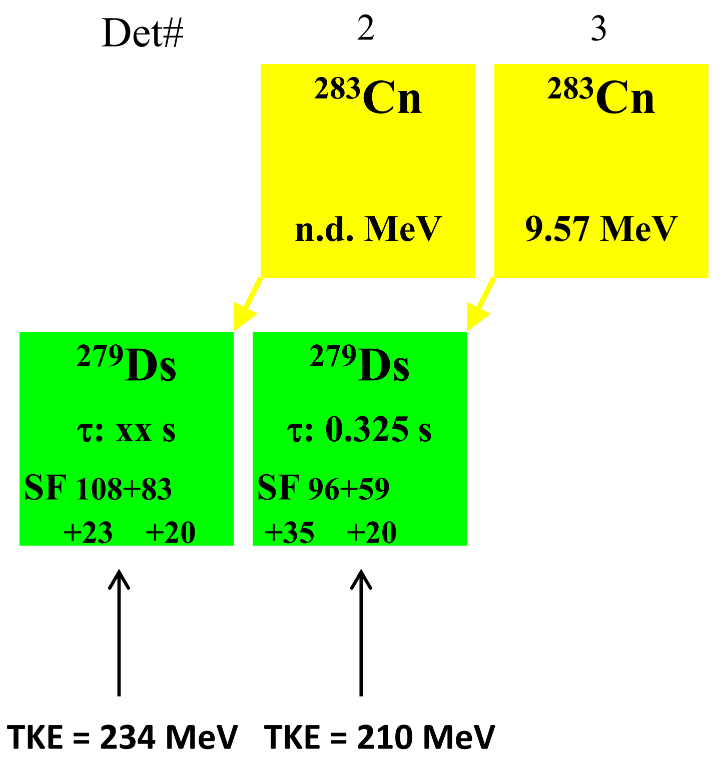


XRD is not enough sensitive to analyze the surface morphology.

Grazing Incidence X-ray diffraction (**GIXRD**)?

KIM, K.S., et al. "Crystallization of amorphous selenium films. I. Morphology and kinetics". J. Appl. Phys., 1973, 44.12: 5237-5244; CLEMENT, R., et al. "The photo-crystallization of amorphous selenium thin films" J. Non-Cryst. Solids, 1974, 15.3: 505-516; LEGROS, A., et al. "Effect on water impurity on the crystallization of vacuum evaporated Se", J. Appl. Phys., 1995, 78.5: 3048-3051

Cn events on Se detectors



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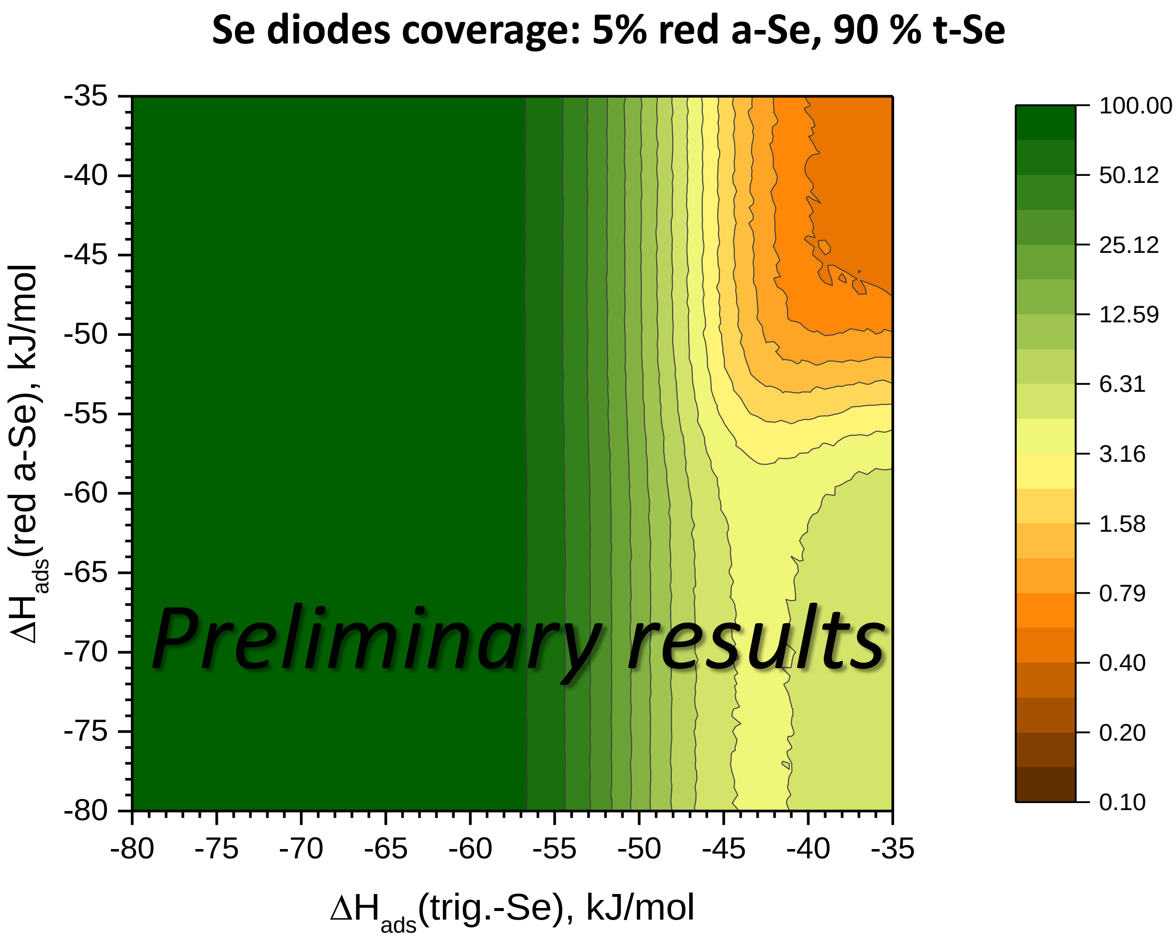
Cn events on Se detectors

^{283}Cn

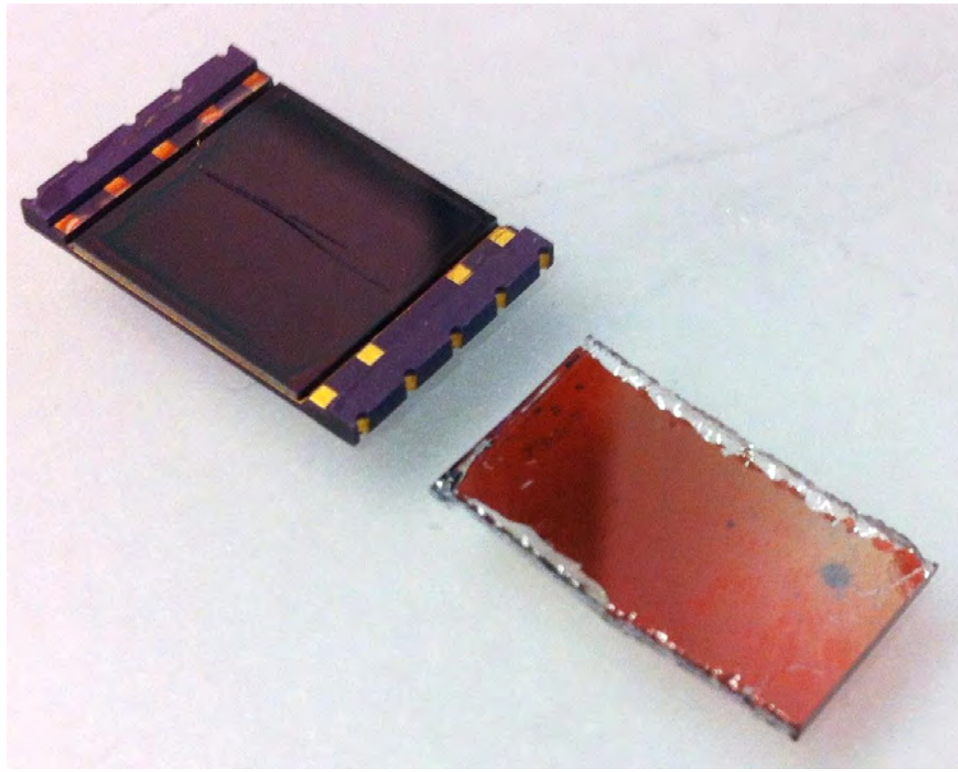
Preliminary results

Probability to observe Cn on Se covered detectors



Cn reacted with trigonal Se with a probability >95%, with a **-ΔH_{ads} > 48 kJ/mol**

Se vacuum evaporation PIN diodes coverage



Next steps:

- crystallization studies (**storage time**) through ^{197}Hg deposition
- **resolution** and layer thickness calculation through **alpha spectroscopy** and **AASIfit simulations**

Conclusions

- The **Se surface crystallization** can be **monitored** by **Hg deposition**

- A severe **kinetic hindrance** of the **Hg / trigonal Se** reaction is confirmed

- First indication that the **interaction of Cn with Se is more favored**, if compared to the Hg / trigonal Se system

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Outlook

- **Inverse thermochromatographic studies** will be performed to understand the **kinetic** process of the **Hg/Se interaction**. Hopefully an activation energy will be derived.

- Future **on-line experiments** are envisaged to:

  - confirm the Cn chemical behavior on trigonal Se surfaces
  - assess the chemical behavior of Cn on red amorphous surfaces
  - assess the chemical behavior of Fl on both selenium allotropic surfaces

Thanks to:

N.V. Aksenov, Y.V. Albin, G.A. Bozhikov, V.I. Chepigin, S.N. Dmitriev, R. Dressler, R. Eichler, V.Ya. Lebedev, A.B. Liechti, O.N. Malyshev, E.A. Maugeri, O.V. Petrushkin, D. Piguet, Y.A. Popov, A.V. Sabel'nikov, P. Steinegger, A.I. Svirikhin, A. Türler, I. Usoltsev, G.K. Vostokin, A. Vögele, A.V. Yeremin

Thank you for your kind attention