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nTOF

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Production of PbSe targets for neutron capture cross section studies

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Overview on the project

Objective

Measurement of the cross section of the **^{79}Se(n, γ)** neutron capture reaction

Motivation

- **Nuclear safety / nuclear waste disposal**

  ^{79}Se is one of the seven long-lived fission products

  Risk of migration from deep geological disposal facilities into the biosphere

  → Transmutation studies of ^{79}Se into the stable isotope ^{80}Se

- **Astrophysics**

  ^{79}Se is a branching point in the slow neutron capture process (s-process)

  → Essential information on the s-process nucleosynthesis

Overview on the project

Methods

- **Time-of-flight (TOF) method**

  Direct cross section measurement of $^{79}Se(n,\gamma)^{80}Se$ @n_TOF facility, CERN

- **Surrogate method**

  Study of the $^{78}Se(^{18}O, ^{16}O\gamma)^{80}Se$ (2n) transfer reaction as a surrogate of $^{79}Se(n,\gamma)^{80}Se$ @Piave-Alti INFN, Legnaro

Targets needed

- Enriched **^{79}Se** target for the TOF method
- Enriched **^{78}Se** for the surrogate method

Characteristics of Se

Se 78	
23.77	
σ 0.38 + 0.05	

Selenium 78

stable nuclide

23.78% natural abundance

Se 79	
3.9 m	$4.8 \cdot 10^5$ y
γ 96 e- β^-	β^- 0.2 no γ g

Selenium 79

produced by high-flux thermal neutrons irradiation of ^{78}Se or by separation of spent nuclear fuel

Problem

Selenium has a **low melting point (217 °C)**, and is a **poor heat conductor**

- ❑ Localized melting at the beam spot
- ❑ Safety concerns during irradiation

Solution

Synthesis of a **selenium compound**

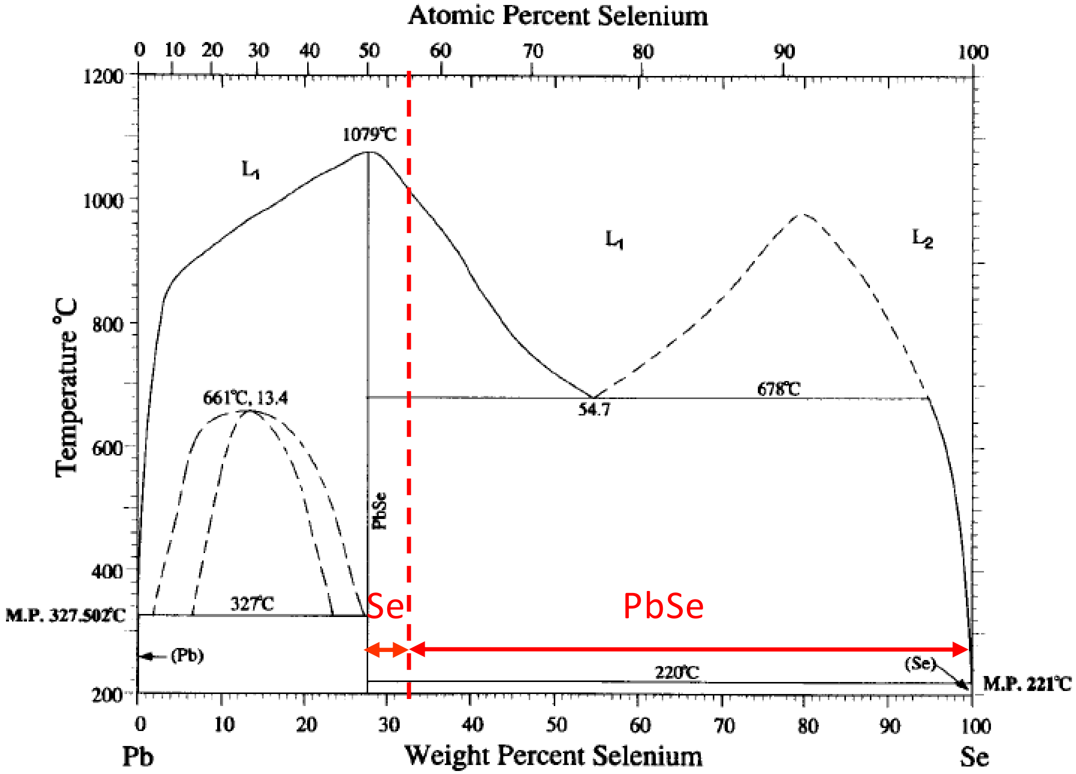
- ❑ High melting point
- ❑ No interference with cross-section studies

Pb^{79}Se target for the **TOF method**
Pb^{78}Se target for the **surrogate method**

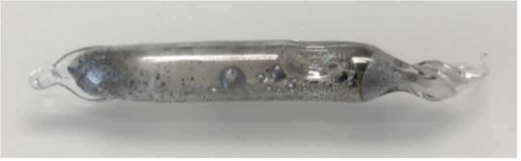
Synthesis of PbSe

Synthesis of PbSe

Reaction of Pb and Se (in slight excess) in an evacuated quartz tube @1250 °C for 24 h

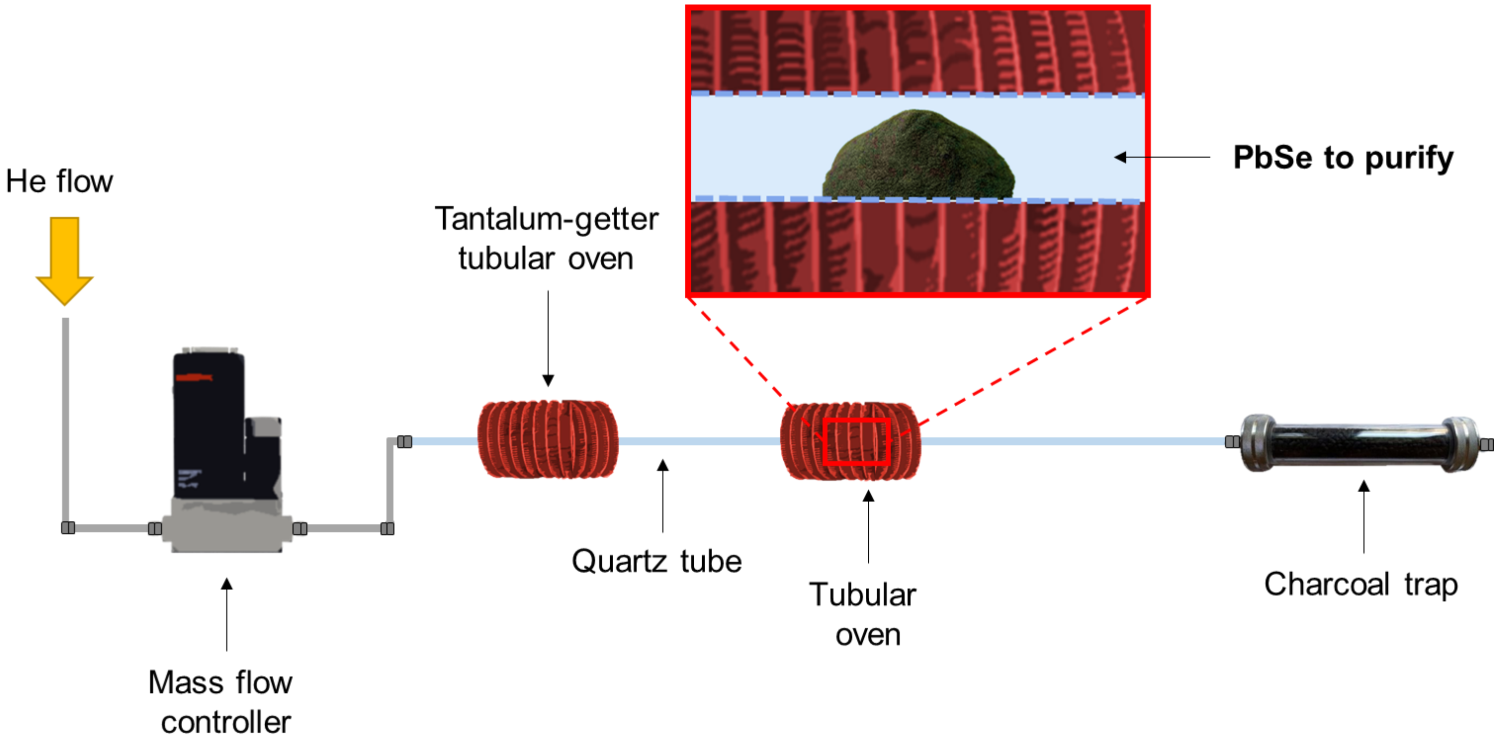


Phase diagram from: Lin J.C., et al., J. Phase Equilib., 17 (1996), pp. 253-260

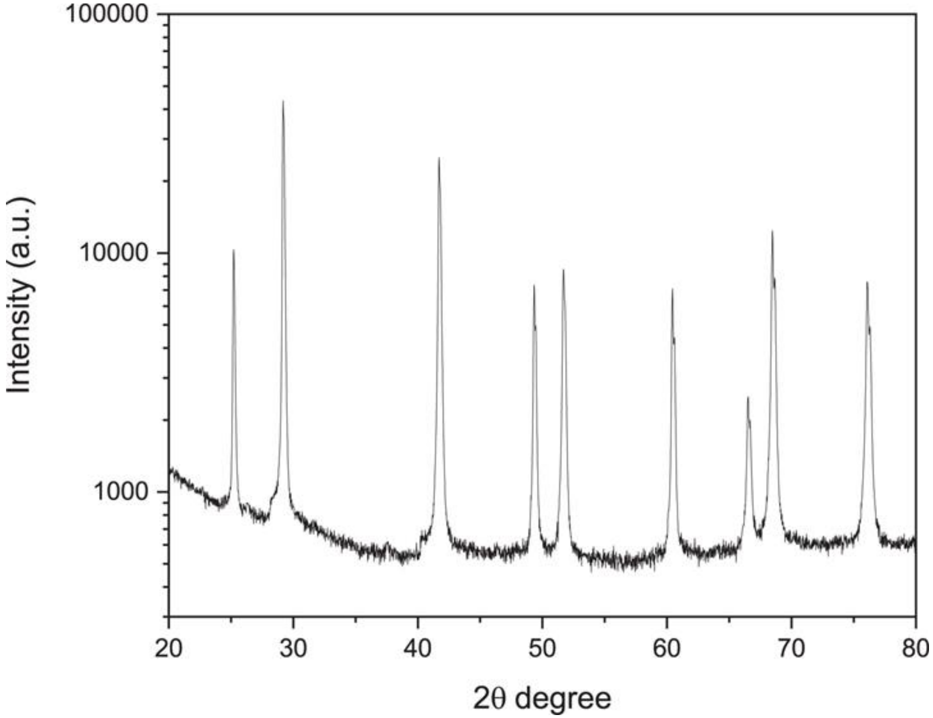


Purification of PbSe

Removal of Se in excess by grinding the synthesized PbSe, and placing it in a quartz tube heated up to 450°C under a He gas flow (25 ml min^{-1}) for 2 hours.



Characterization of the synthesized PbSe: XRD



Lit. * 2θ, °	Exp. 2θ, °	d	h, k, l
25.21	25.2	3.5321	1, 1, 1
29.19	29.18	3.0587	2, 0, 0
41.75	41.72	2.1638	2, 2, 0
49.40	49.33	1.8463	3, 1, 1
51.75	51.69	1.7675	2, 2, 2
60.52	60.48	1.5299	4, 0, 0
66.62	66.52	1.4049	3, 3, 1
68.59	68.47	1.3696	4, 2, 0
76.23	76.2	1.2487	4, 2, 2

*Downs et al. (1993) American Mineralogist 78, 1104-1107

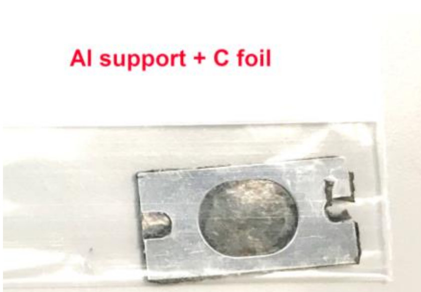
Lattice constant = 6.12 Å

Target for the surrogate method

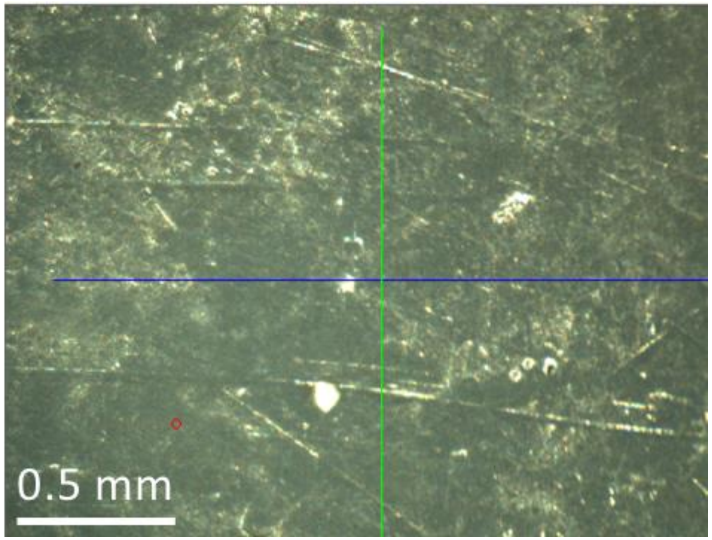
PbSe target for the surrogate method

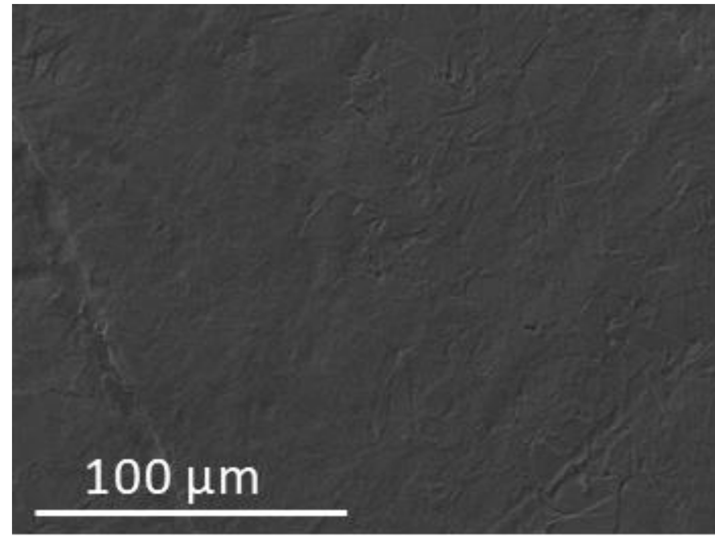
Physical vapor deposition with the thermal evaporation station Univex 450 (Leybold vacuum) equipped with a **mechanical shutter** and a **quartz crystal microbalance**.

- Operational vacuum: 10^{-5} mbar
- Crucible: Ta boat coated with Al_2O_3
- Applied current: 4.5 A
- Temperature (Type-K thermocouple): 220 °C
- Evaporation rate: 0.2 nm s^{-1}.
- **Thickness deposited layer: 400 nm**



Characterization of PbSe layer: XRF and SEM





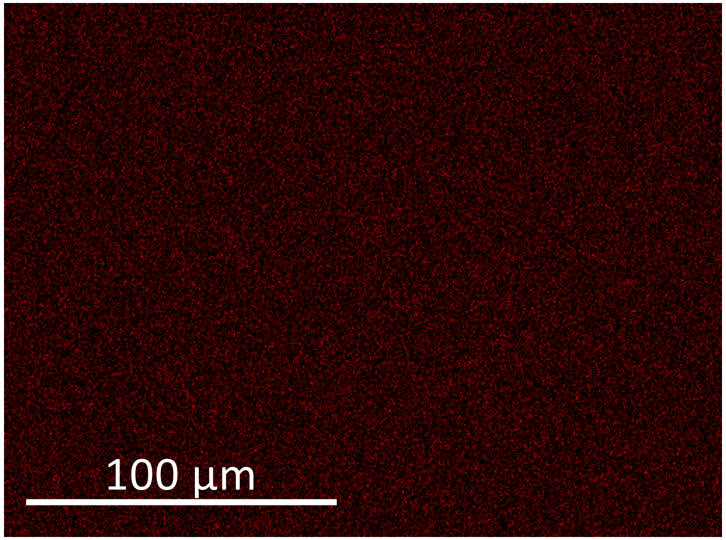
PbSe deposited material (XRF)

	wt%	wt% error	Literature*
Pb(L)	74.5	1.6	74.3
Se(K)	25.5	0.5	25.7

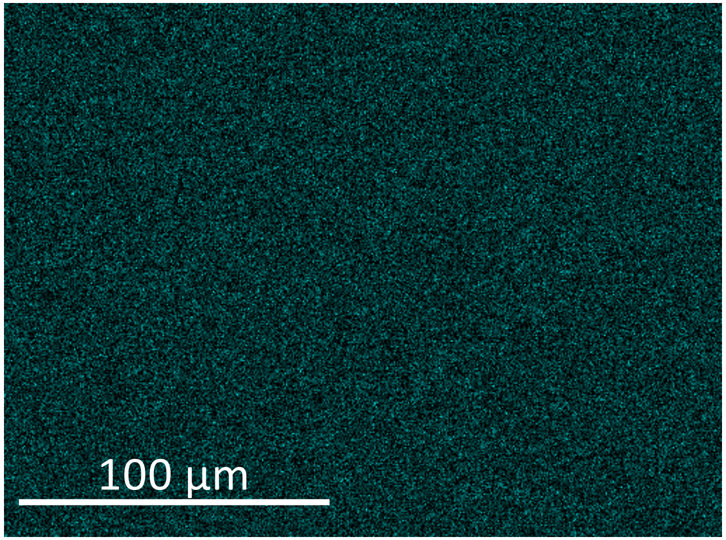
*K. Ravi et. Al., Conf. Ser.: Mater. Sci. Eng. 932, 2020, 0123133.

Characterization of PbSe layer: EDX

Distribution of Se in deposited layer



Distribution of Pb in deposited layer



Composition of deposited PbSe layer (EDX)

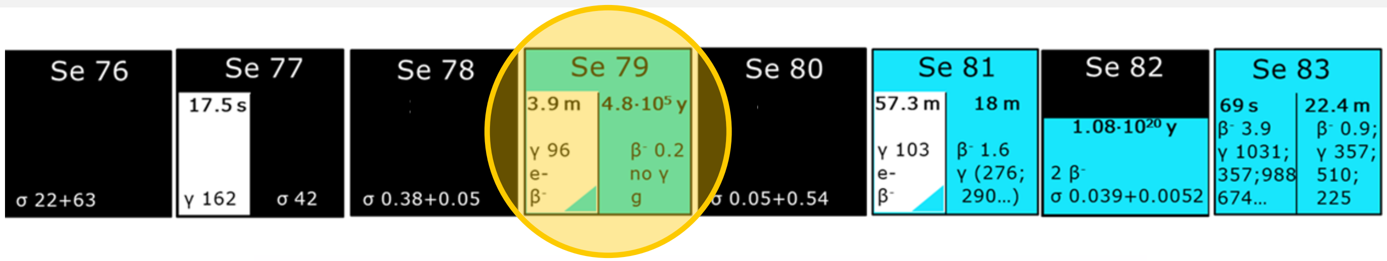
	at%	Literature*
Pb	52.2	52.4
Se	47.7	47.6

*K. Ravi et. Al., Conf. Ser.: Mater. Sci. Eng. 932, 2020, 0123133.

Target for the TOF method

PbSe target for the TOF method

- Synthesis of 4.11 g of Pb^{78}Se
- Irradiation for 51 days (total neutron fluence: 5×10^{21} cm^{-2}) @ILL high-flux reactor



Isotope content

Isotope	Se-78	Se-76	Se-77	Se-80	Se-82
Enrichment, (%)	99.30	0.01	0.03	0.64	0.02

Chemical admixture

Element	Al	Na	Ca	Mg	Fe
Content (ppm)	< 20	< 20	< 20	< 42	< 75
Element	**Be**	**Sc**	**Cu**	**Cr**	**As**
Content (ppm)	< 0.2	< 6	< 2	< 10	< 3
Element	**B**	**Mn**	**Ni**	**Si**	**Zn**
Content (ppm)	< 5	< 1	< 2	< 5	40 ± 35
Element	**Ti**				
Content (ppm)	< 4				

PbSe target for the TOF method

Characterization of the irradiated material @PSI, *via* γ-spectrometry.

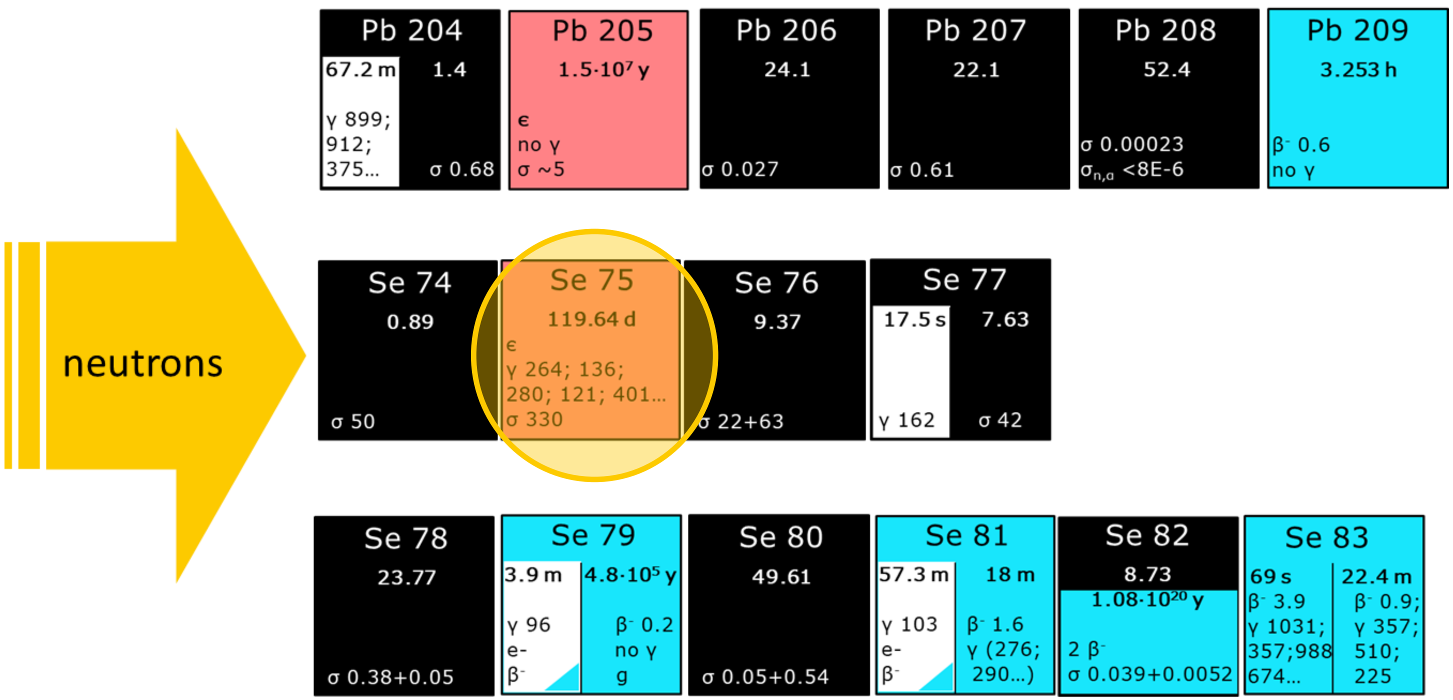
→ Calculated amount of ^{79}Se produced: ~3 mg ←

Isotope	Half-life	Activity (MBq) t = 1 y	Mass (ng) t = 1 y	Activity (MBq) t = 5 y
^{75}Se	119.79 d	338	627	0.07
^{110m}Ag	249.76 d	29	165	0.51
^{65}Zn	244.15 d	1.89	6.2	0.03
^{60}Co	5.27 y	2.77	66	1.64

Retrieval of Se from PbSe

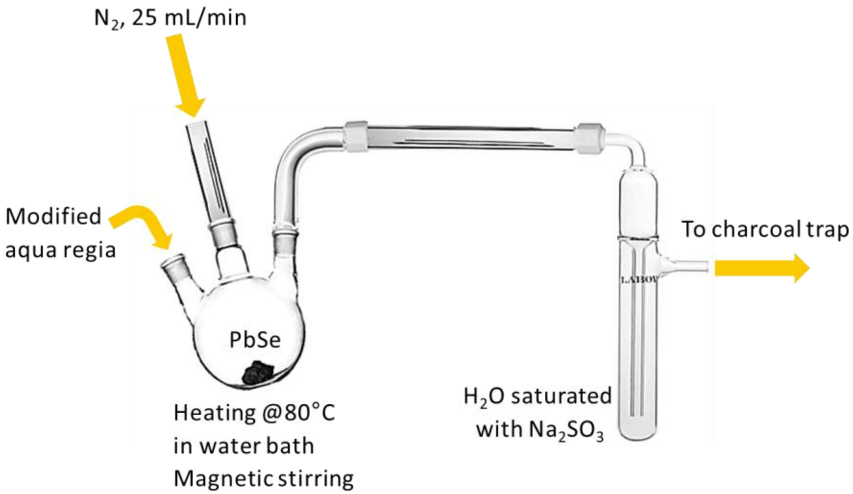
Retrieval of Se from PbSe

- Efficiency of separation tested with Pb^{75}Se
- Irradiation of 0.04 g of PbSe with thermal neutrons @SINQ
- Irradiation time: 1 hour; cooling time: 24 hours

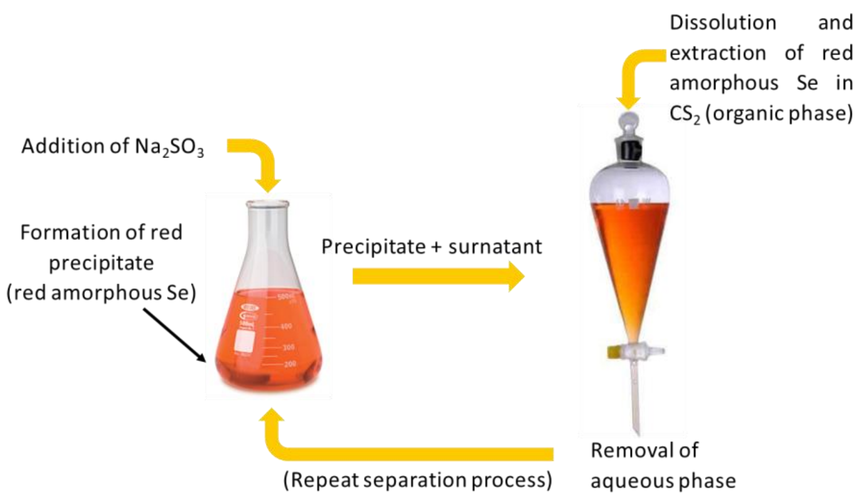


Recovery of Se from PbSe

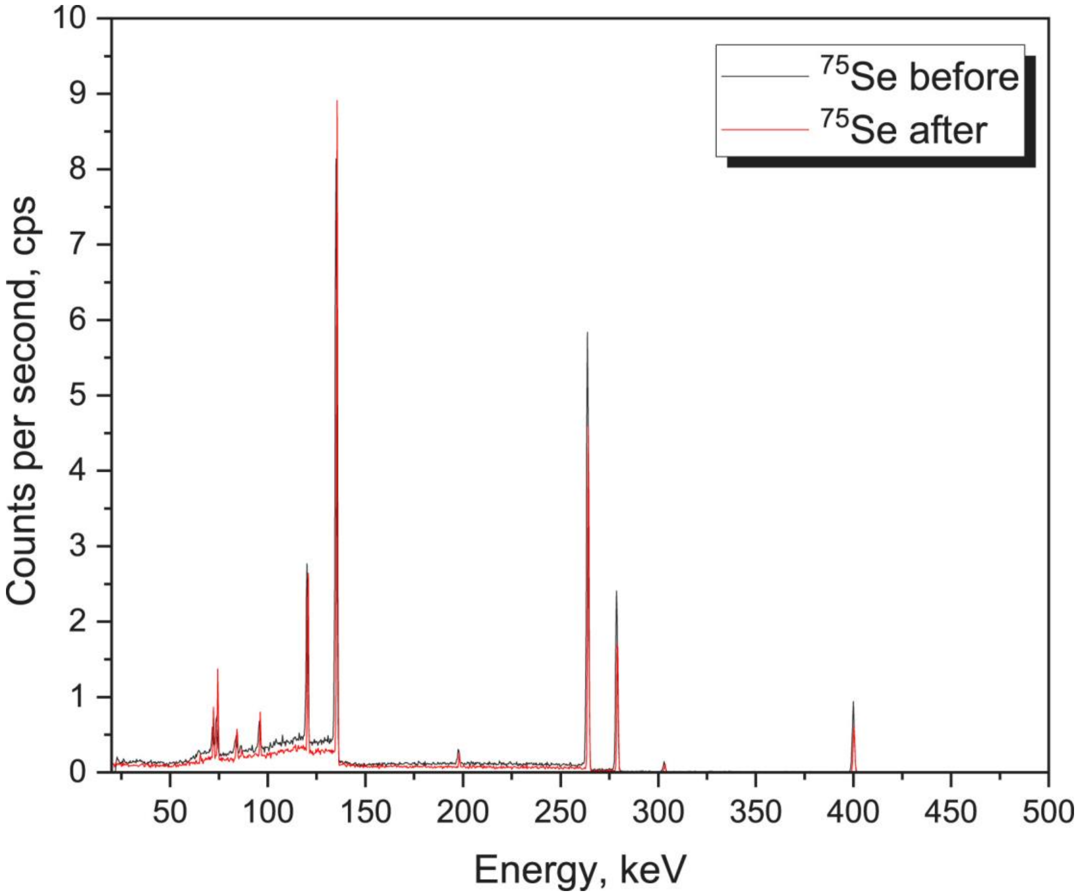
Step 1: Dissolution of PbSe



Step 2: Extraction of Se



Efficiency of separated Se: γ-spectroscopy



Se 75

119.64 d

ϵ

γ 264; 136; 280; 121; 401…

σ 330

Efficiency of the separation process: ~70%

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Purity of separated Se: ICP-OES

- ❑ The separated Se was dissolved in aqua regia
- ❑ Aliquots of 0.01 ml were diluted with 2% HNO_3 (final volume = 14 ml)

Sample	Wavelength (nm)	Concentration (ppm)	SD (ppm)
Se-I	Pb (220.353)	0.003	0.001
	Pb (283.305)	0.005	0.001
	Se (196.026)	3.789	0.040
	Se (203.985)	3.818	0.042
Se-II	Pb (220.353)	0.003	0.001
	Pb (283.305)	0.004	0.001
	Se (196.026)	4.582	0.271
	Se (203.985)	4.606	0.271

Purity of separated Se: ~99%

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Conclusions

- Successfully applied a method for the **synthesis of PbSe**
- Production of **$Pb^{79}Se$** at ILL (**3 mg of ^{79}Se**) for TOF method
- Deposition of thin (**400 nm**) and homogeneous PbSe layers as targets for surrogate method
- Developed a method for the **recovery of Se (99% purity)** from PbSe, with an **efficiency of 70%**.
- The method can also be used to **purify Se prior to the synthesis of PbSe**
- Nadine M. Chiera et al., ***Nucl. Instrum. Methods Phys. Res. A,*** 1029 (2022)

Thank you for your kind attention

Collaborators:

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