RRMC
Radiobioassay & Radiochemical Measurements Conference

DEVELOPMENT OF A NEW RESIN-BASED MATERIAL FOR THE RADIOCHEMICAL SEPARATION OF SELENIUM

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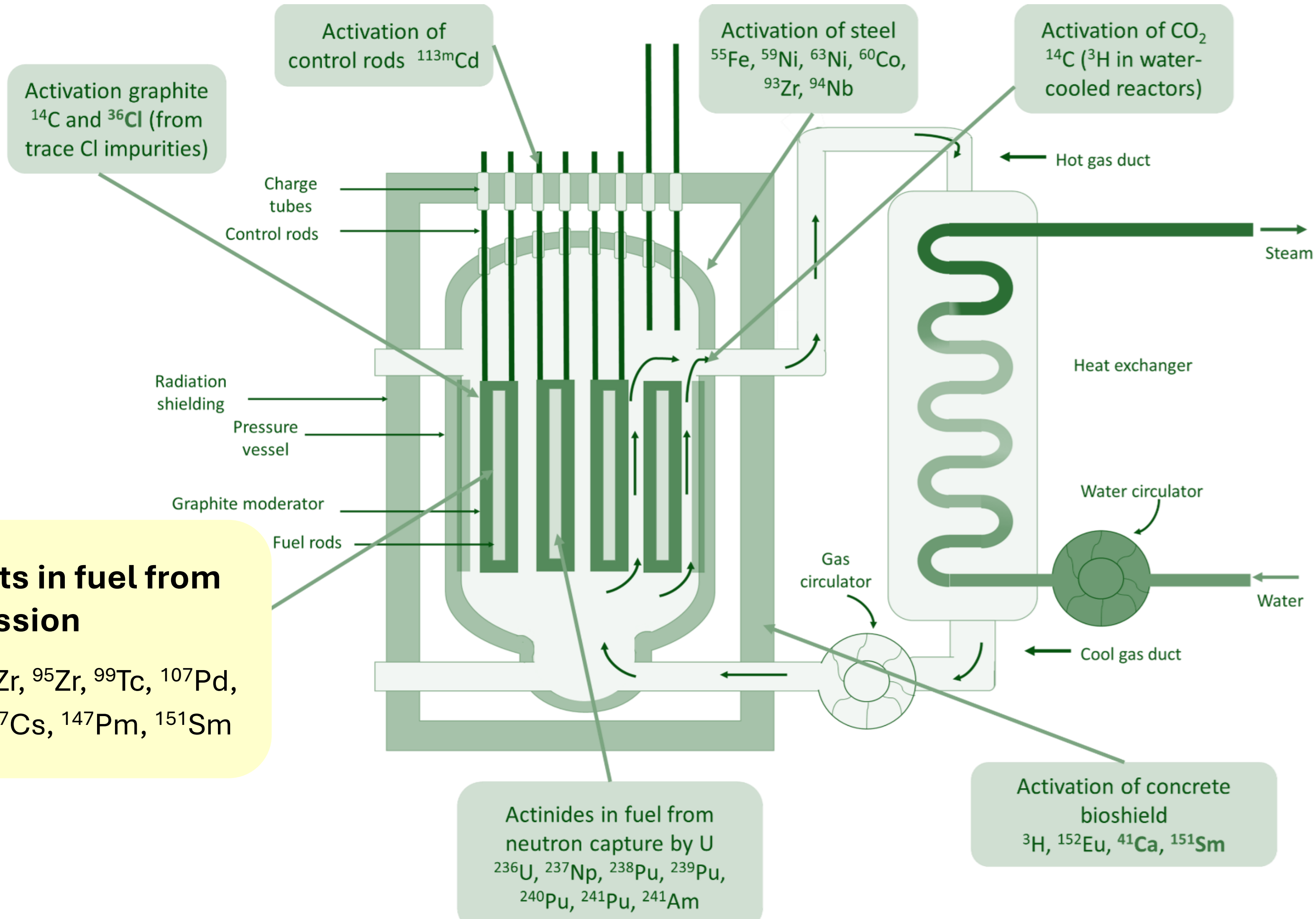
TRISKEM
Expertise in Separation Chemistry

^{79}Se in radioactive waste

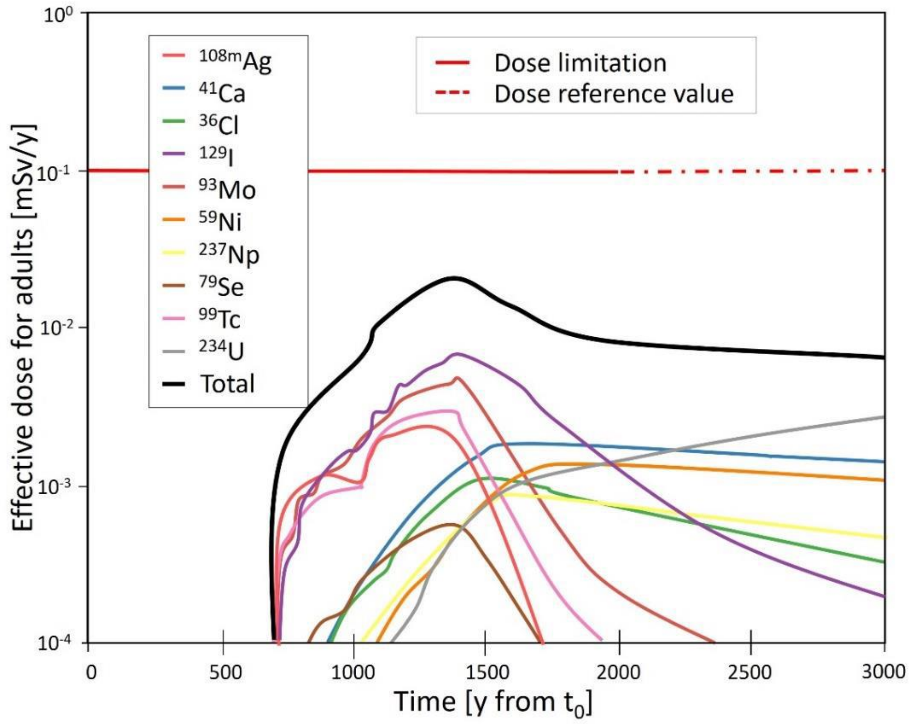


Fission products in fuel from ^{235}U fission

^{3}H, **^{79}Se**, ^{90}Sr, ^{93}Zr, ^{95}Zr, ^{99}Tc, ^{107}Pd, ^{126}Sn, ^{129}I, ^{135}Cs, ^{137}Cs, ^{147}Pm, ^{151}Sm

^{79}Se in radioactive waste

- Long half life (~ 10^5 years)
- Considered in assessment studies of waste storage or disposal and monitoring
- High-level radioactive samples



Impact curves of the most contributing radionuclides in terms of long safety assessment regarding the effective dose to adults (ONDRAF/NIRAS, 2019)

^{79}Se in radioactive waste

- Environmental protection
  - Toxicity of selenium [Se(IV) or Se(VI)]
  - Immobilization
- Present in trace levels in high level waste

Removal of radionuclides occurring at high level activity (e.g. ^{90}Sr, ^{137}Cs or ^{60}Co) to decrease the **global activity** of the sample

^{79}Se determination

- Beta particle emitter → ^{79}Se to ^{79}Br
  - E_{max} → 150 keV
- Selenium oxidation adjustment needed
  - Possible species: Se(IV) as SeO_3^{2-} or Se(VI) as SeO_4^{2-}
- Chemical separation applied

Distillation	**Ion exchange columns**	**Metallic Se**
Br removal	Global activity	Gravimetry
Applied Radiation and Isotopes 148 (2019) 35	Talanta 69 (2006) 565–571	Applied Radiation and Isotopes 148 (2019) 35

Challenges on ^{79}Se determination

- ^{79}Se measurement

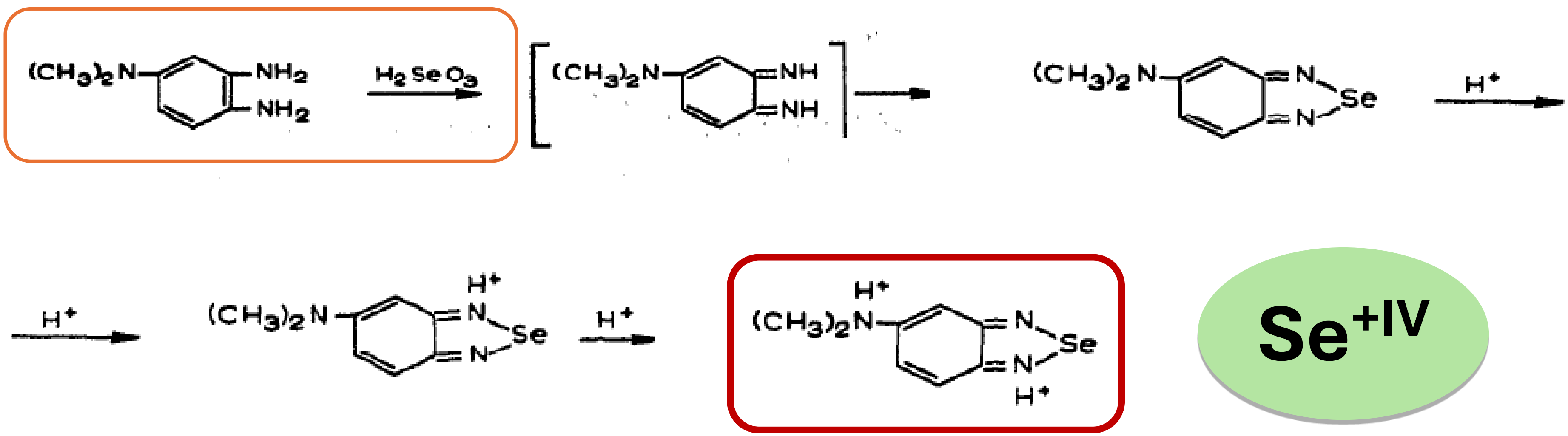
ICP-MS	Liquid scintillation counting	AMS
Isobaric interferences ^{79}Br	Spectrometric interferences ^{99}Tc, ^{147}Pm, ^{151}Sm, ^{93}Zr, ^{63}Ni or ^{241}Pu	Isobaric interferences ^{79}Br

- Different Se oxidation states → Se(IV) / Se(VI)
  - Differences in chemistry
- Lack of ^{79}Se reference material or standard solution

New resin-based material for selenium radiochemical separation

Principle of piazselenol formation

Anal. Chim. Acta, 27 (1962) 288–294



Se^{+IV}

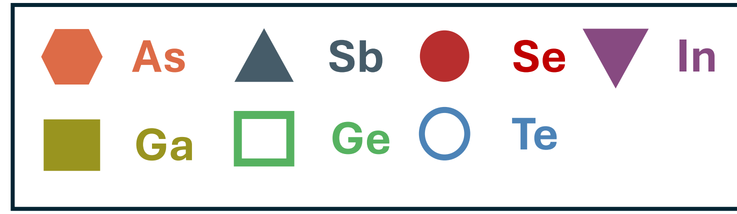
Oxidation state fixation

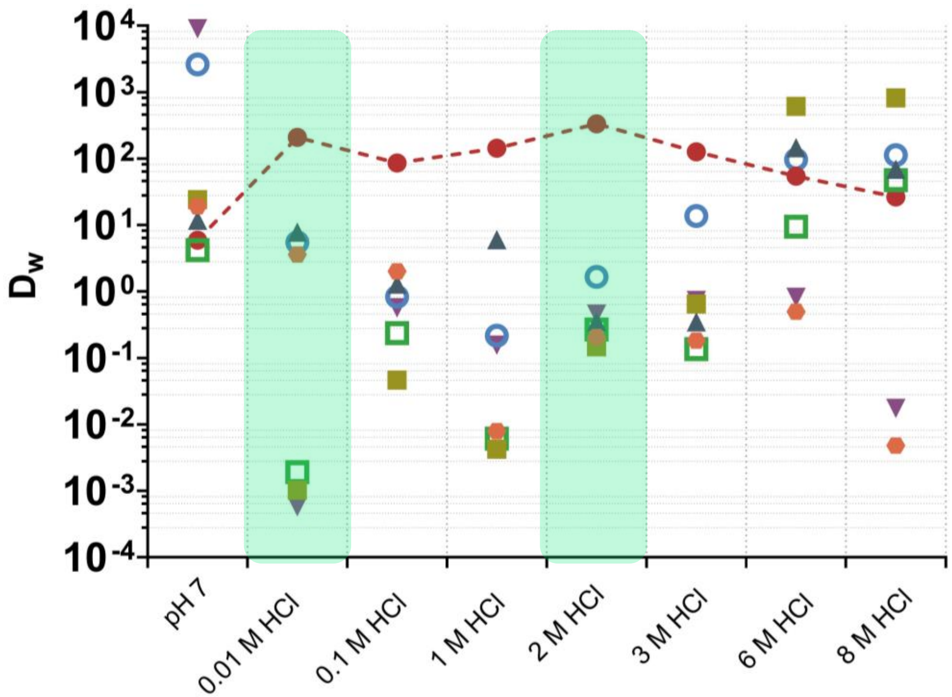
Loading/elution medium

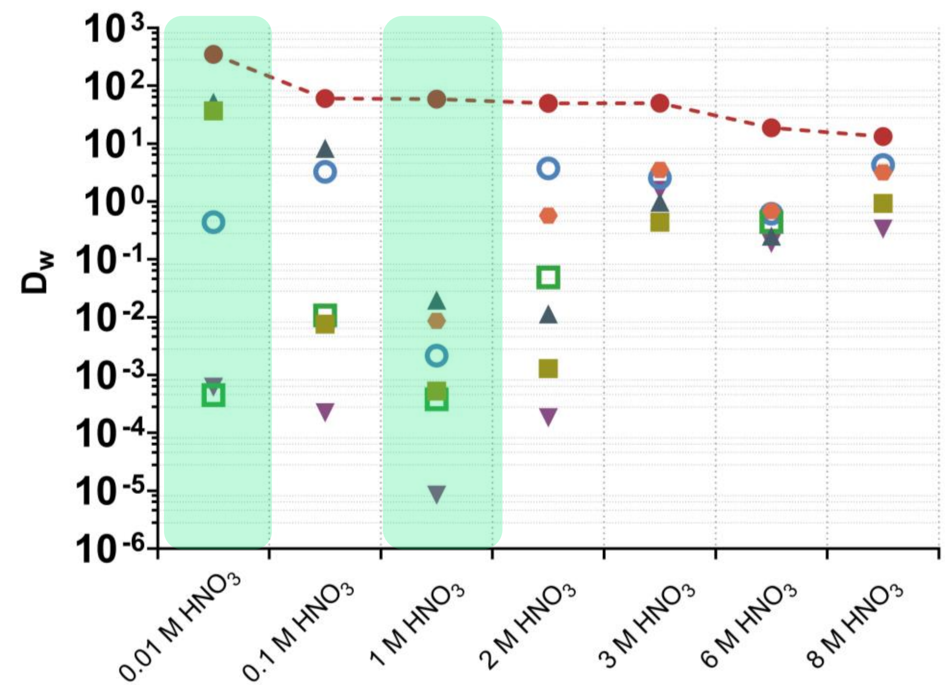
Column bleed

First approach to SE Resin

Previous results reported by Dirks et al. 2016





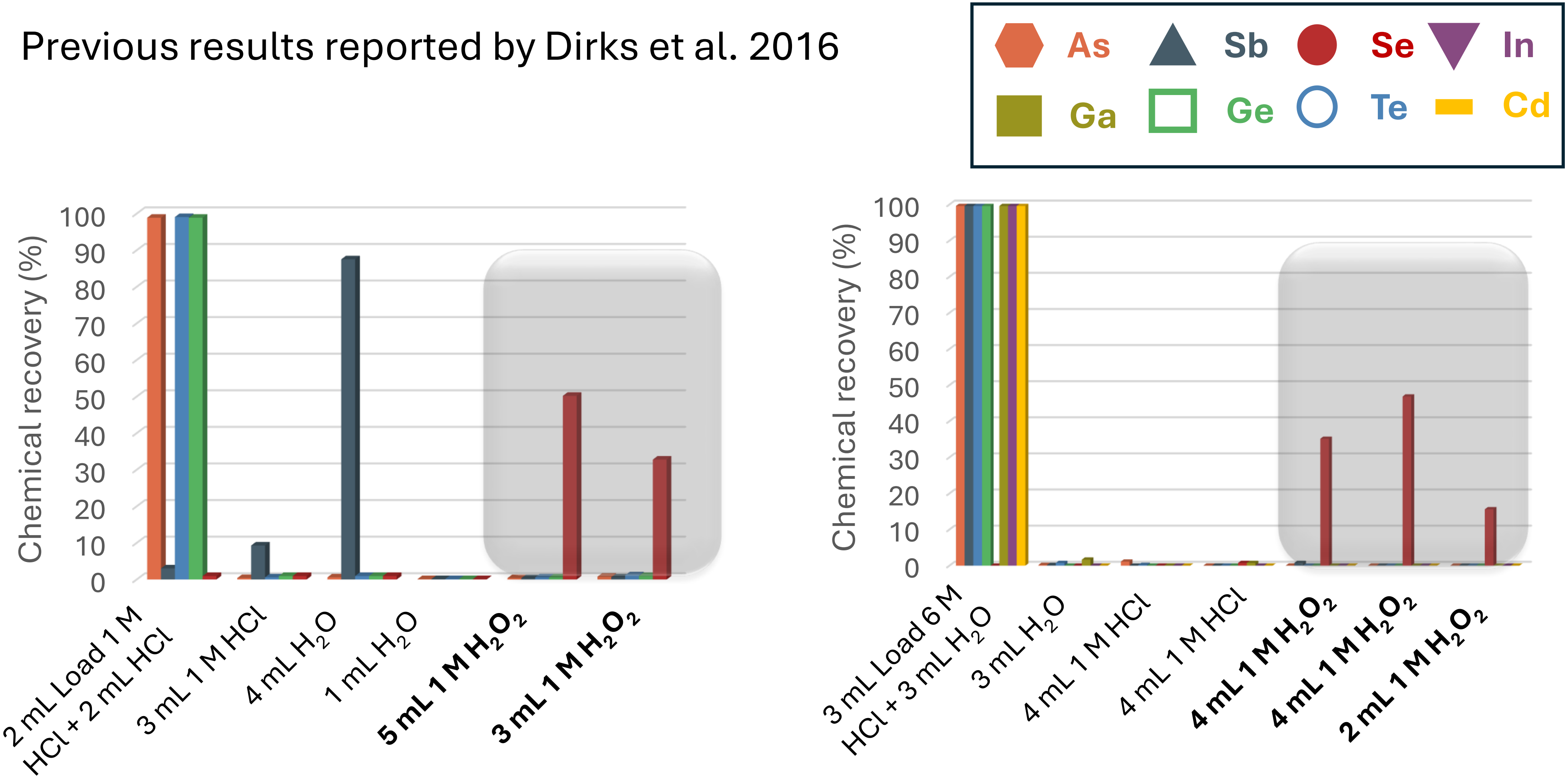


HCl

HNO_3

First approach to SE Resin

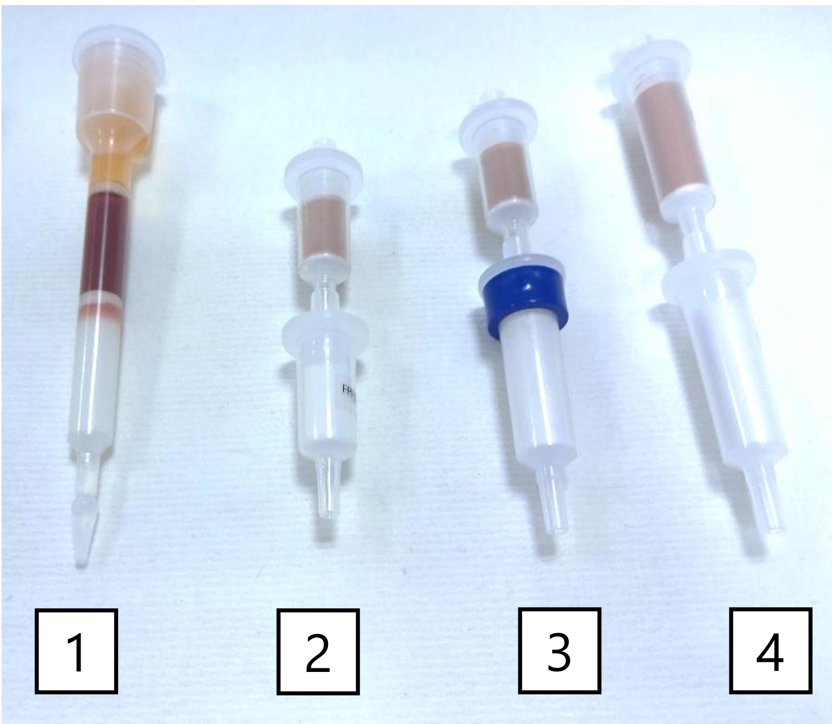
Previous results reported by Dirks et al. 2016



350 mg Se selective Resin **Load 1 M HCl**

300 mg Se selective Resin **Load 6 M HCl**

SE Resin preparation



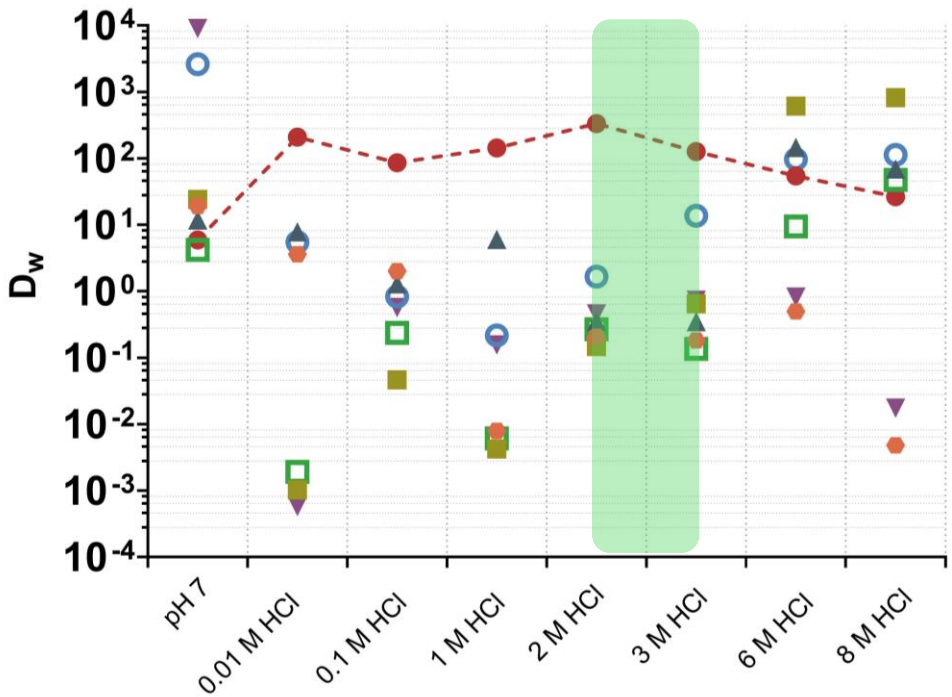
1 mL	1 mL	1 mL	2 mL
1 mL	**1 mL**	**2 mL**	**2 mL**
column	cartridge	cartridge	cartridge

New SE Resin

TRISKEM

SE Resin	Se retention
Prefilter Resin	↓ extractant bleeding

SE Resin preparation



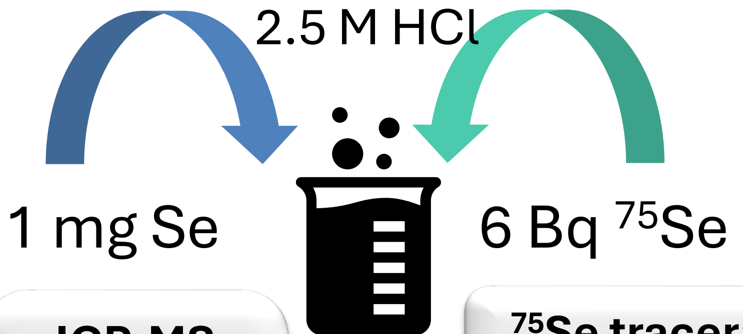
New SE Resin

TRISKEM

SE Resin	Se retention
Prefilter Resin	↓ extractant bleeding

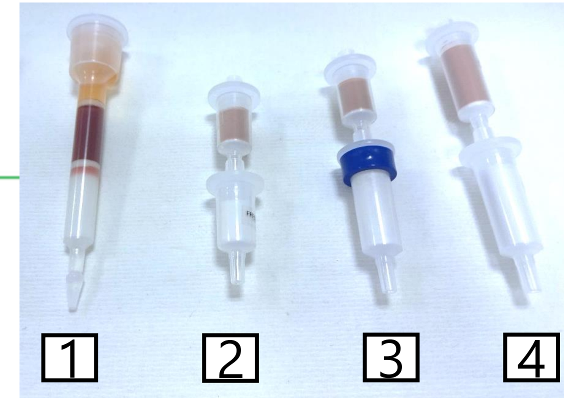
- ✅ ↑ Se retention loading with **2.5 M HCl**
- ✅ With **Prefilter Resin** D_w from 100 to 1000

SE Resin loading

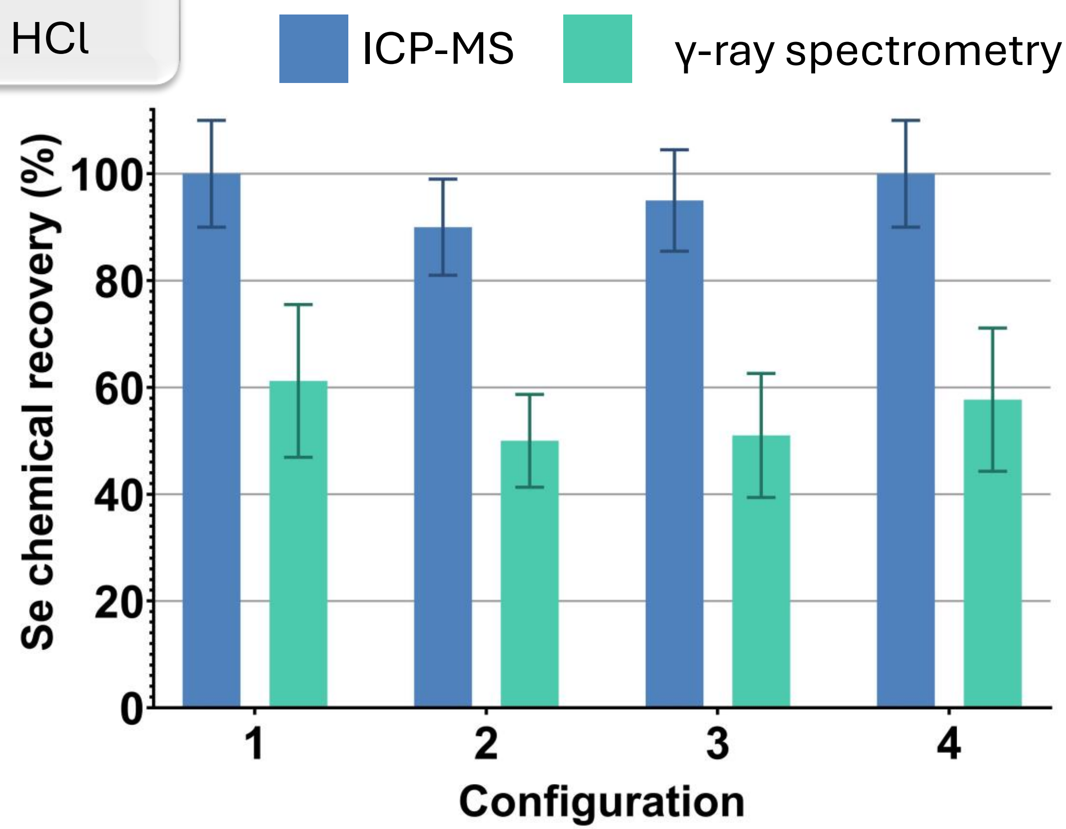


ICP-MS standard: Se in 2% HNO_3

^{75}Se tracer: ^{75}Se in 0,1 M HCl

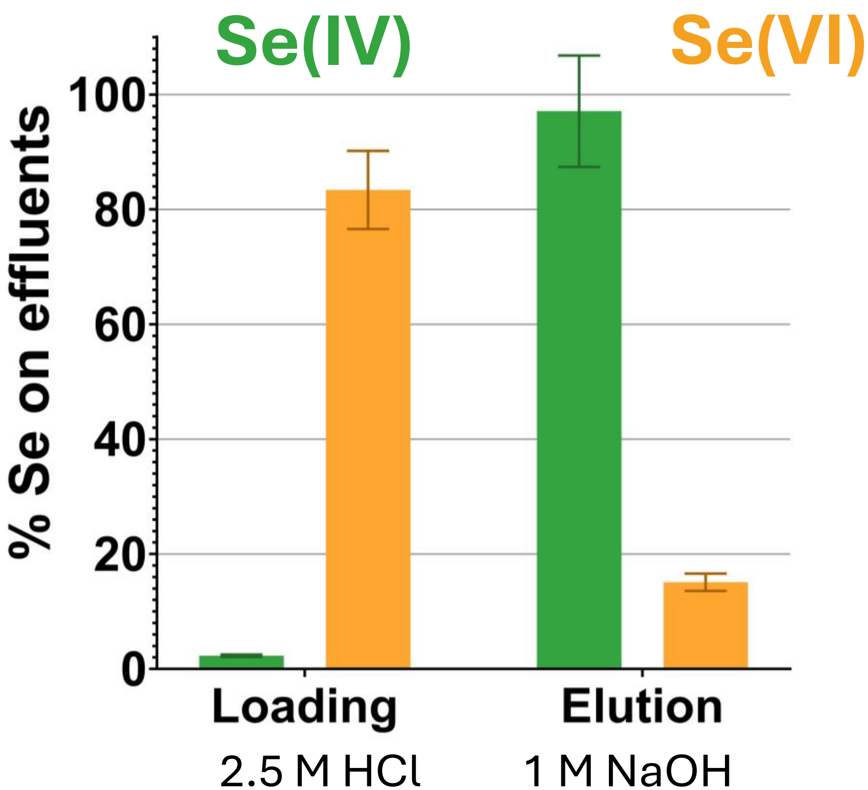


Disagreement of **ICP-MS** and **γ-ray** spectrometry results



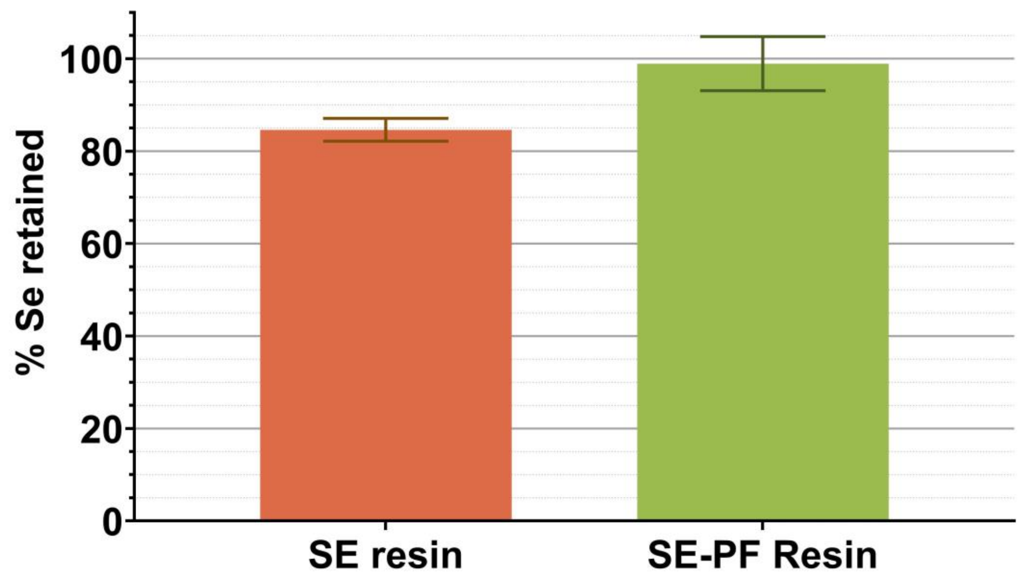
SE Resin loading

Oxidation state



ICP-MS standards:
Se^{+4} (2% HNO_3)
Se^{+6} (in H_2O)

Sample loaded: 2.5 M HCl



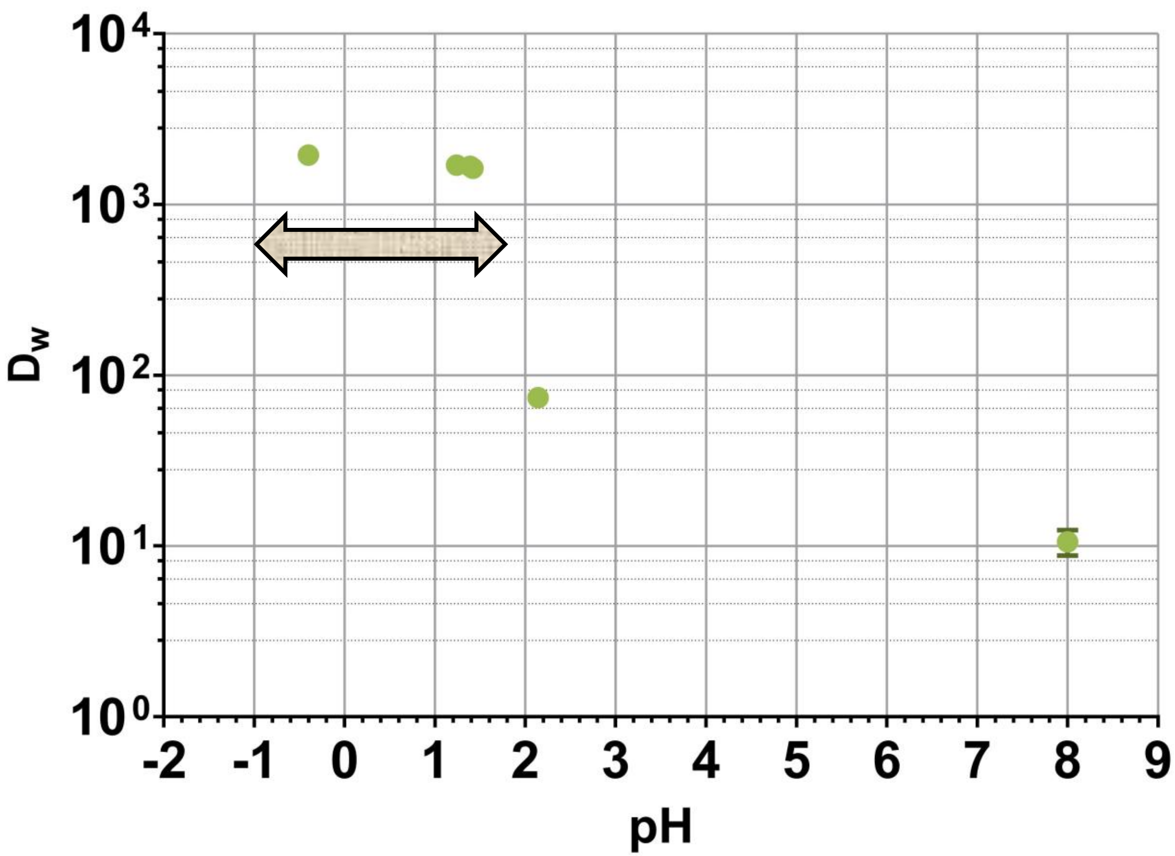
ICP-MS standard:
Se in 2% HNO_3

SE Resin loading

Relevance of pH

- Bosca & Mot, 2021 → relevance of pH on p*iazselenol* formation (**pH~1.3**)

ICP-MS standard:
Se in 2% HNO_3

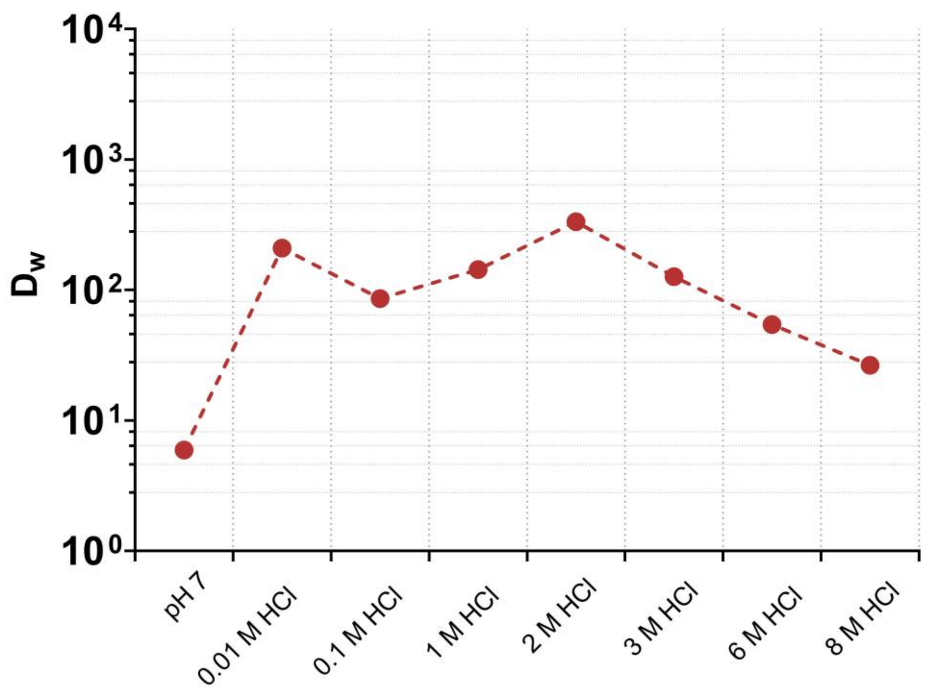


SE Resin loading

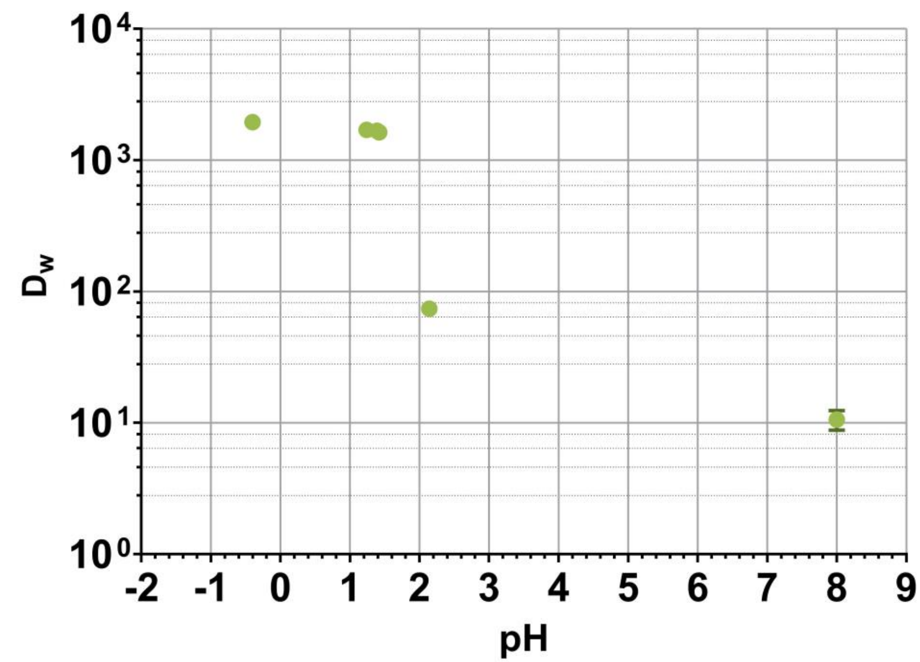
Relevance of pH

- Bosca & Mot, 2021 → relevance of pH on p*iazselenol* formation (**pH~1.3**)

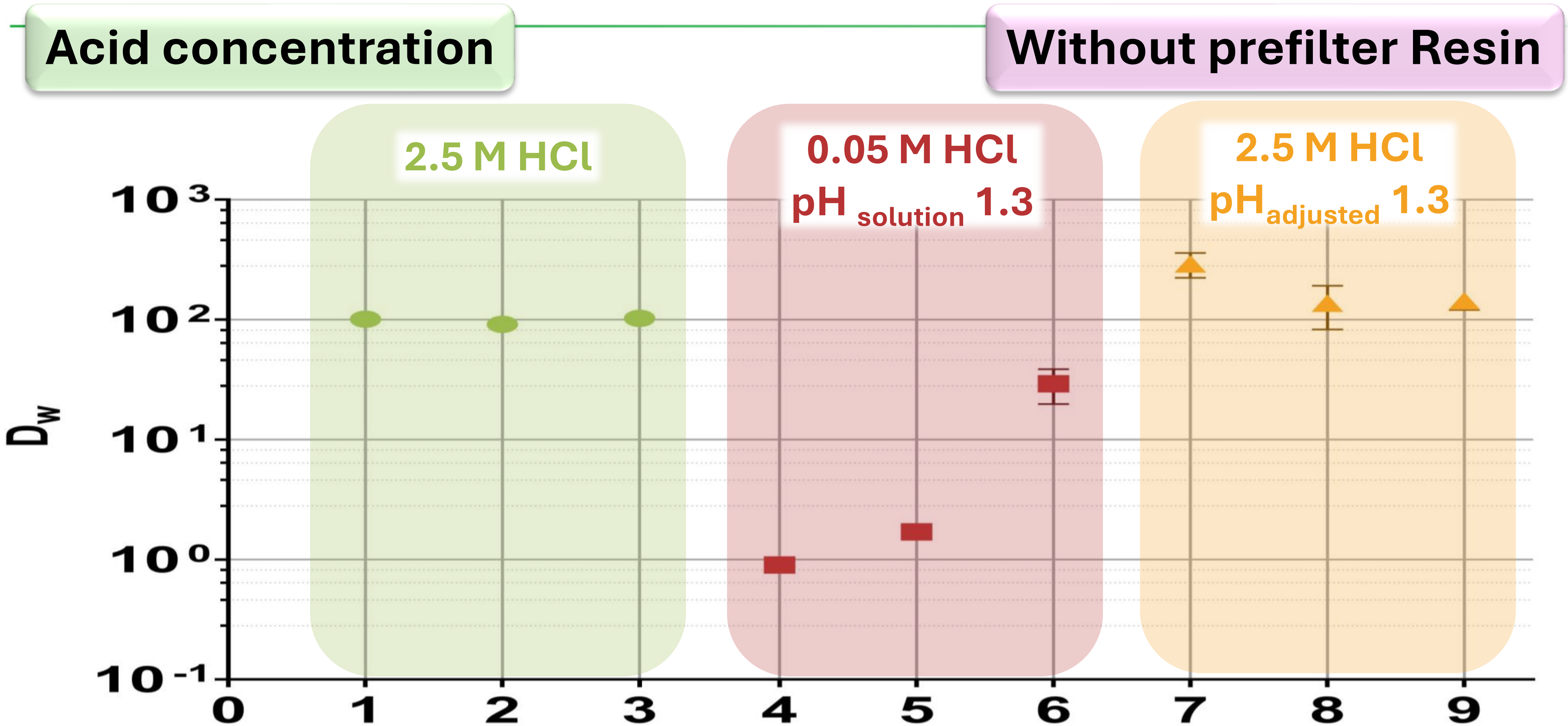
Without prefilter Resin



With prefilter Resin



SE Resin loading

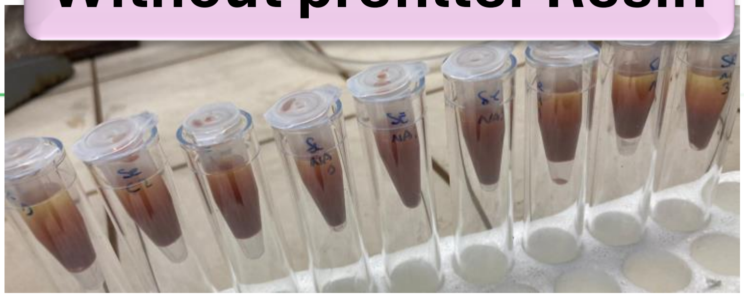


At pH 1,3:

- ↓ acid concentration (**0.05 M HCl**) ↓ D_w
- pH adjusted (with NH_3) ↑ D_w
  - Suitable for Se retention

SE Resin loading

Without prefilter Resin

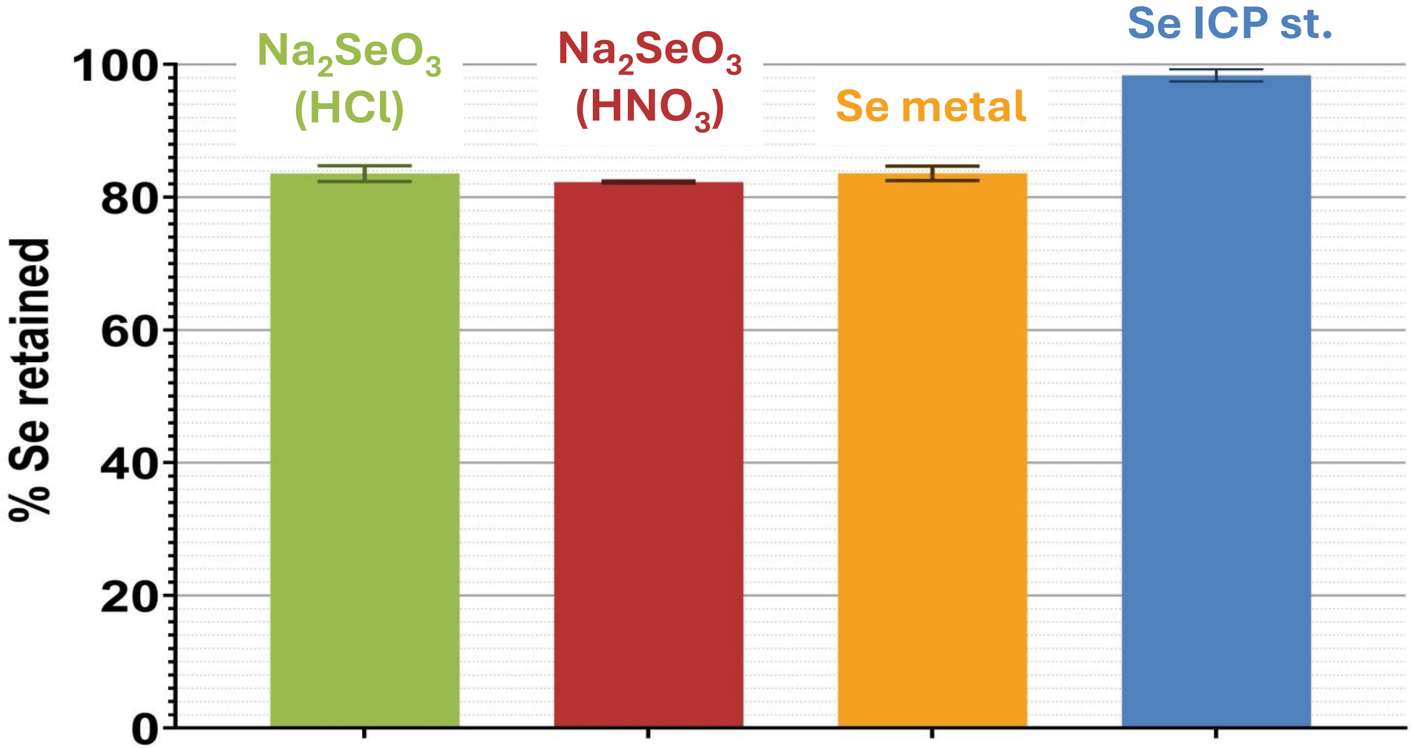


Different Se standards

Batch studies → 50 mg **SE resin** + 1 mL solution

Na_2SeO_3 → 1 mL HCl in 100 mL
1 mL HNO_3 in 100 mL

Metallic Se → digestion with
5 mL HNO_3 in 100 mL



No Prefilter Resin used

Sample preparation for SE Resin

Suitable conditions for Se retention

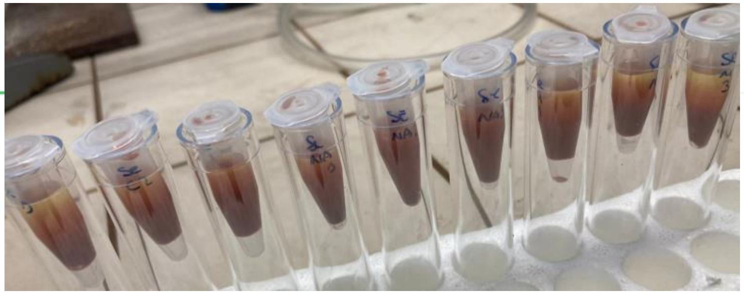
- Loading medium: 2.5 M HCl
- pH adjustment suggested → pH 1.3
- Se volatility
- Need of PF Resin for reducing extractant bleeding

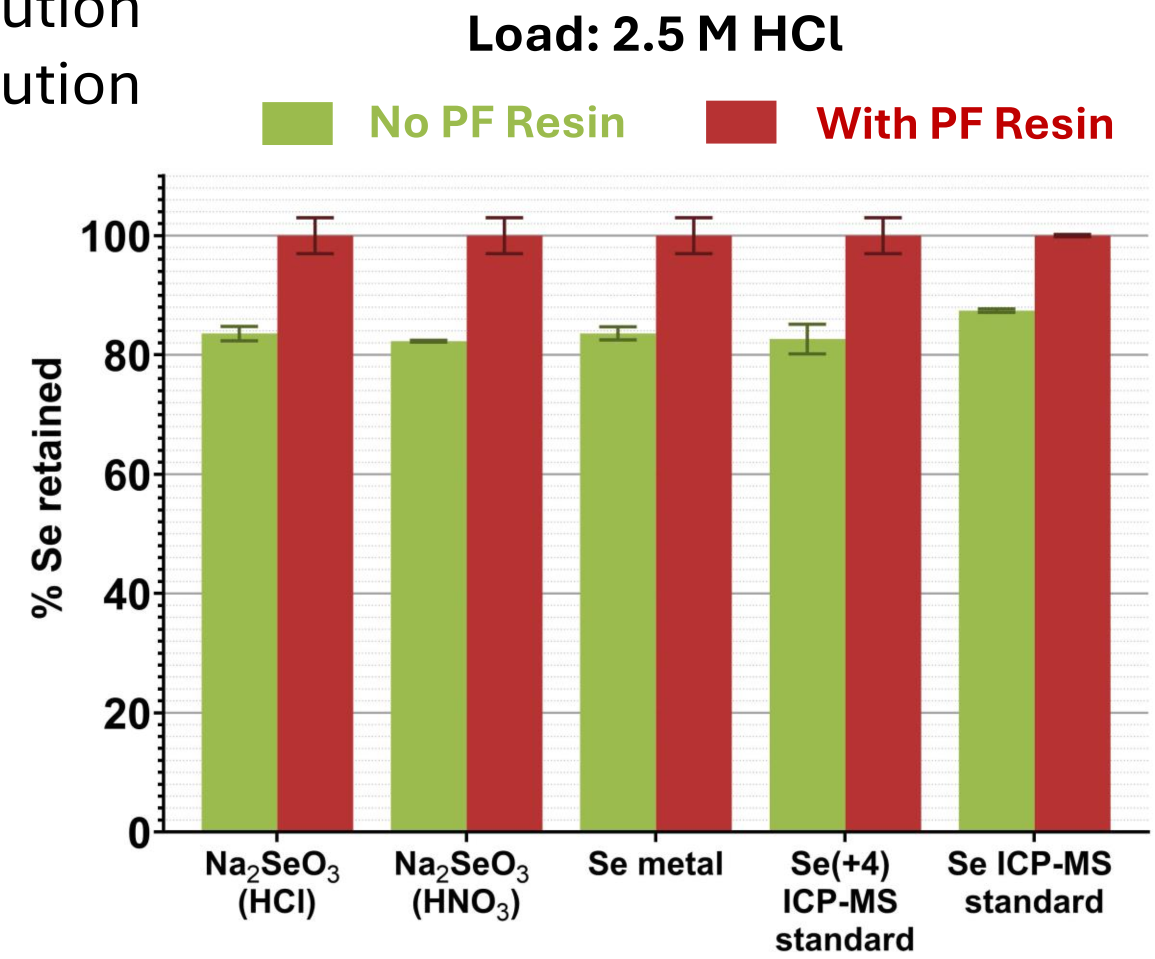
Need of Prefilter Resin

Different Se standards

Batch study:

- 50 mg resin + 1 mL solution
- 50 mg resin + 1 mL solution
  + 0,5 mL PF Resin





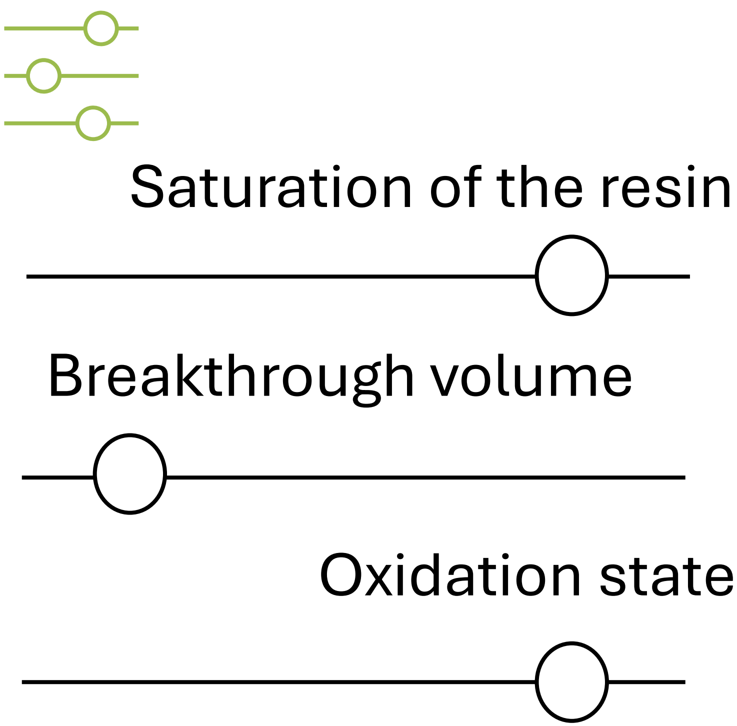
On-going tests

Capacity

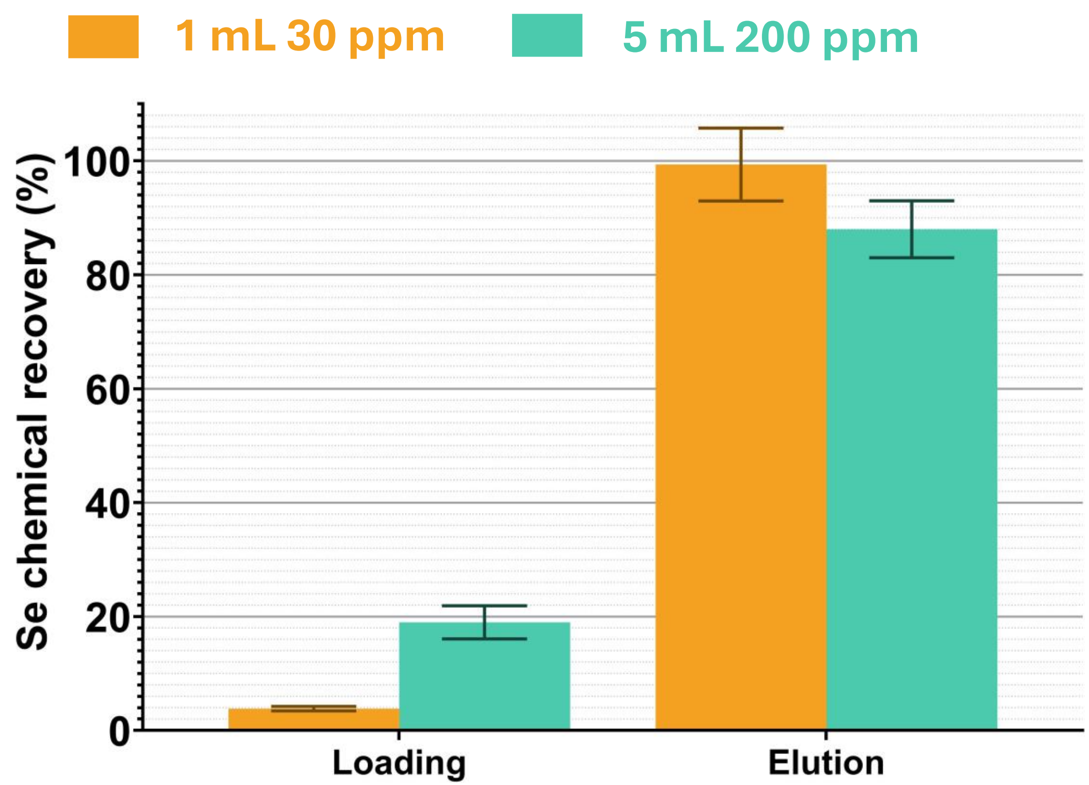
Loaded on 0.5 mL SE Resin:0.5 mL PF Resin

- 0,03 mg Se
- 1 mg Se

Low capacity



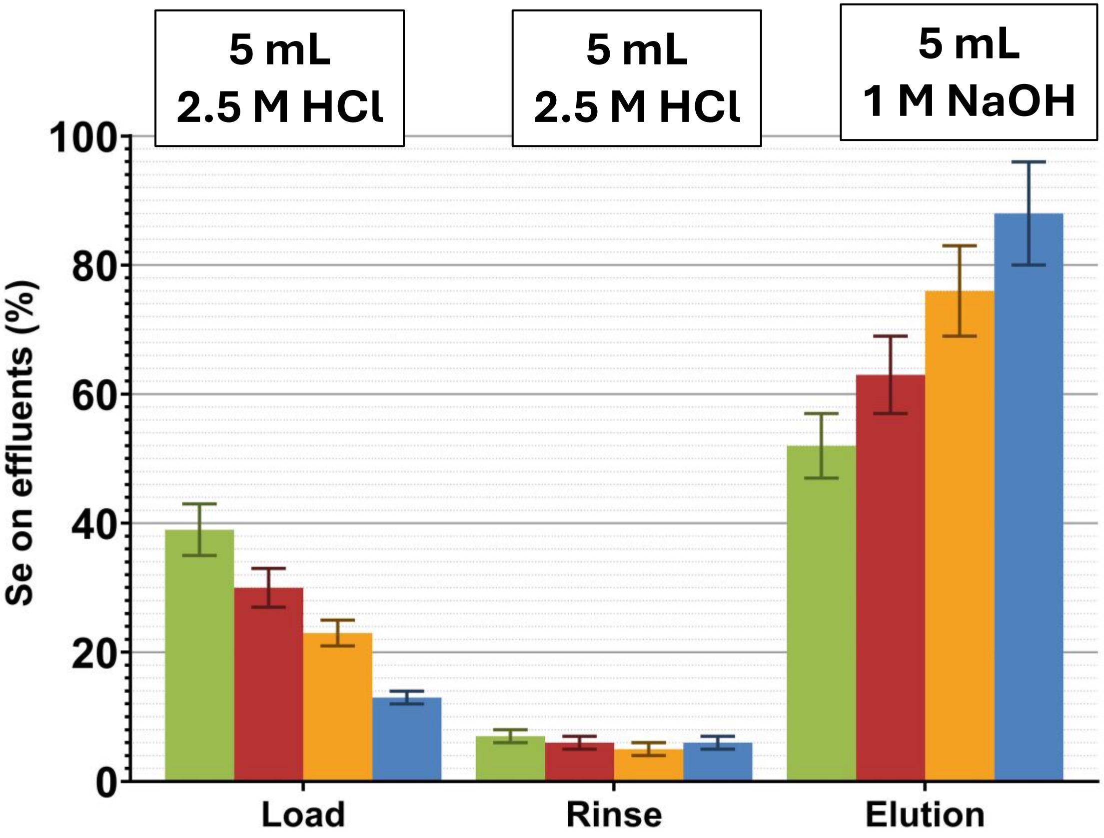
Load: 2.5 M HCl



On-going tests

Comparison with different standards

Na_2SeO_3 (HCl)	Na_2SeO_3 (HNO_3)	Se metal	Se +4 ICP st.
52%	63%	76%	88%

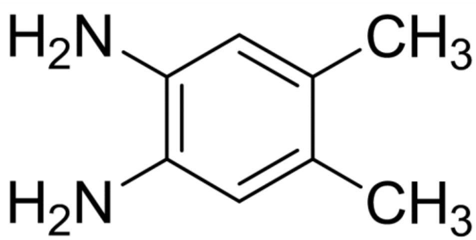


On-going tests

Oxidation state via colorimetric tests

UV-VIS spectrometry

Chromogenic reagent

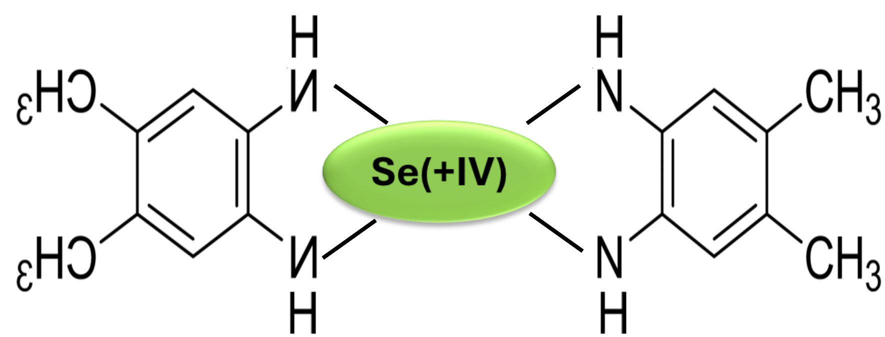


Se containing solution

Se(+IV)

Work on-going in DTU/SCK courtesy of Matic Dokl

Measurement of the absorbance



Based on: Kalaitzidou K., et. al. 2001

Work on-going

Batch experiments + elution profiles with specific Se oxidation state standards

Se(+IV) **Se(+VI)**

- pH
- Acid concentration
- Volume

Capacity tests, colorimetric test to ensure Se oxidation state, durability of SE Resin

Investigation of interferences and development of a method for Se determination in different samples

Thank you for your attention!!

Questions?

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Expertise in Separation Chemistry