Self-sputtering during isotope collections

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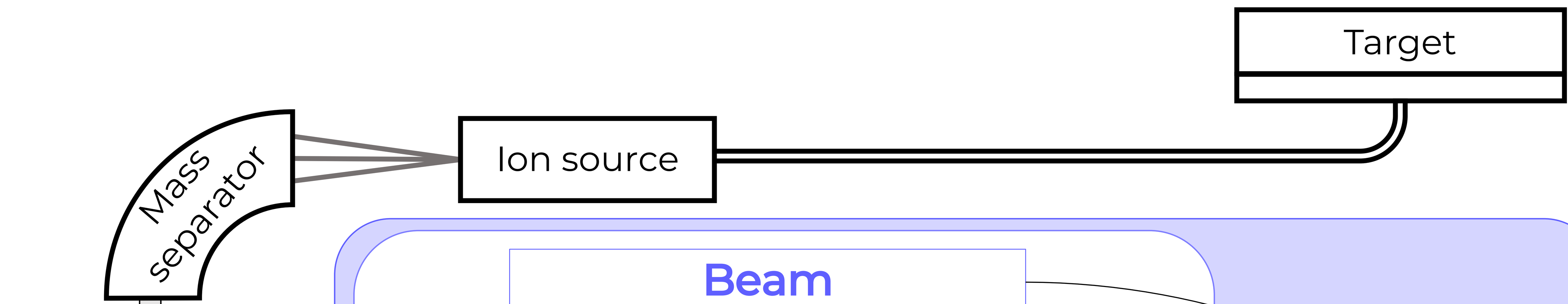
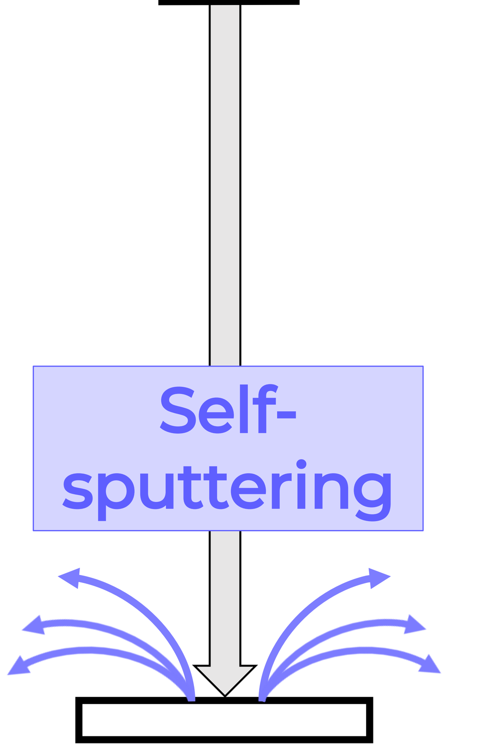
Context

For radionuclide collections the goal is to maximize the amount of collected radioisotopes

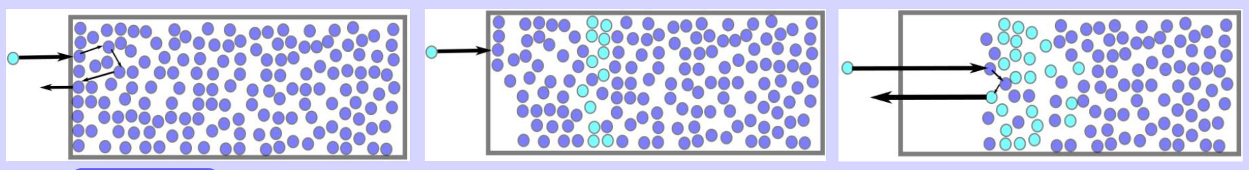
Self-sputtering has limited several collections at CERN-MEDICIS

Aim: investigate self-sputtering during radionuclide collections

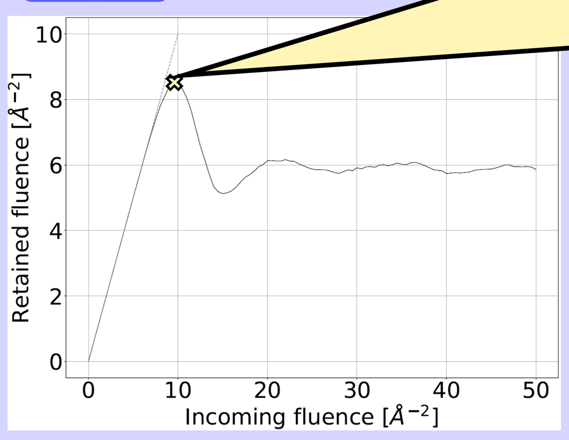




General principle



Scan me

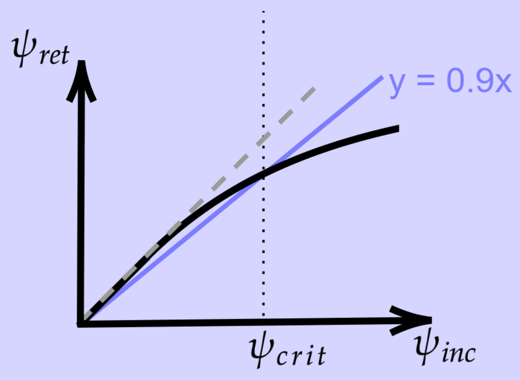


PROBLEM:
Fundamental limit is reached: no matter how much is implanted, the retained amount does not increase

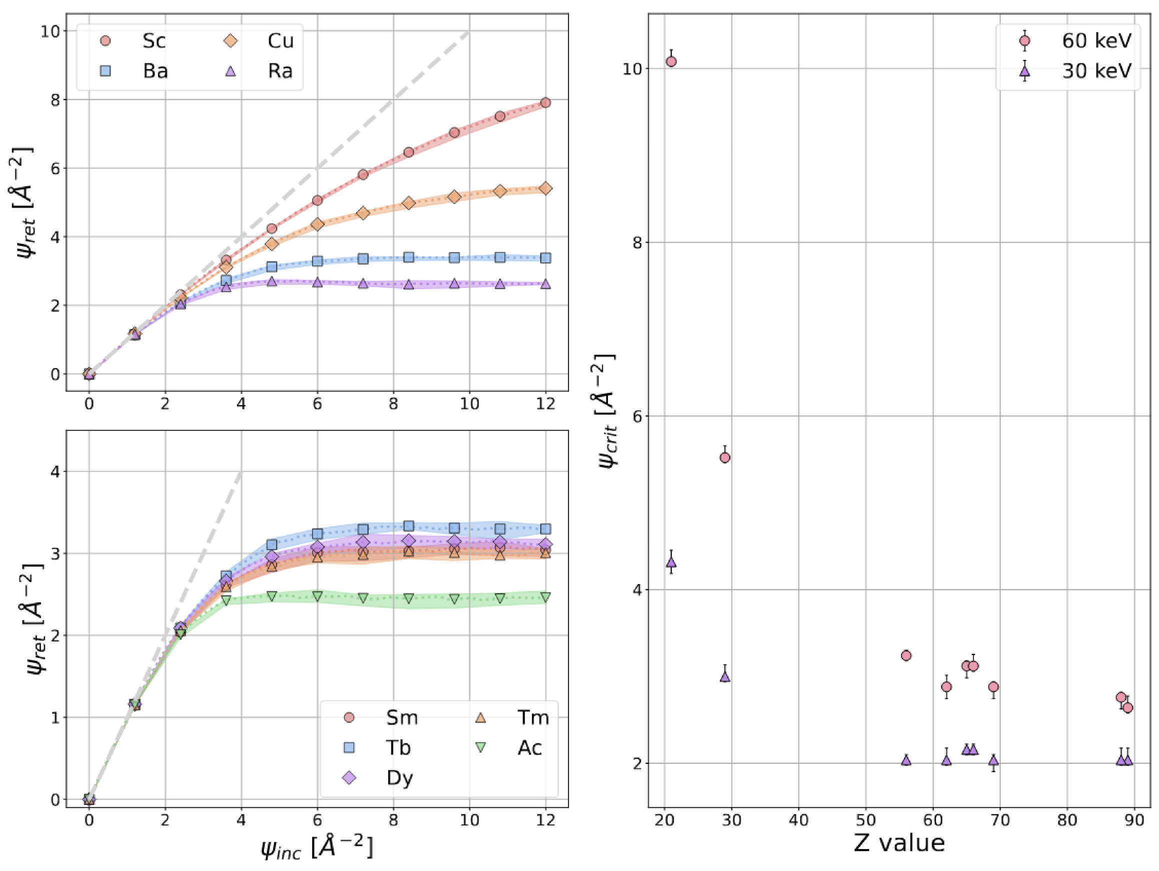
Minimize losses

TRIDYN

- Monte Carlo based simulation package [1, 2]
- **Dynamic** changes to the target ↔ SRIM [3]
- Self sputtering limit:
  $\boldsymbol{\psi_{crit}: \psi_{ret} < 0.9\psi_{inc}}$



Beam



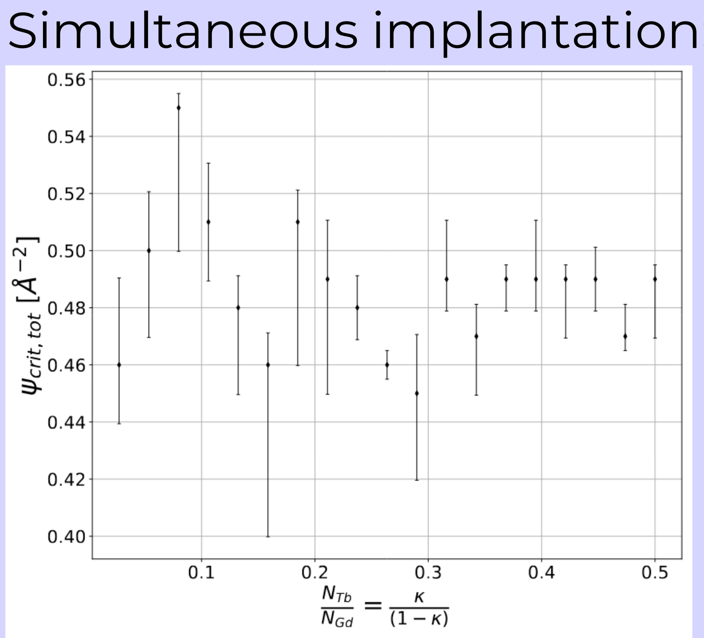
TRIDYN simulations for different incoming beams and beam energies

- Implantation of **low Z elements**
- Implantation in **low Z foils**
- Implantation at **large beam energies**

less self-sputtering

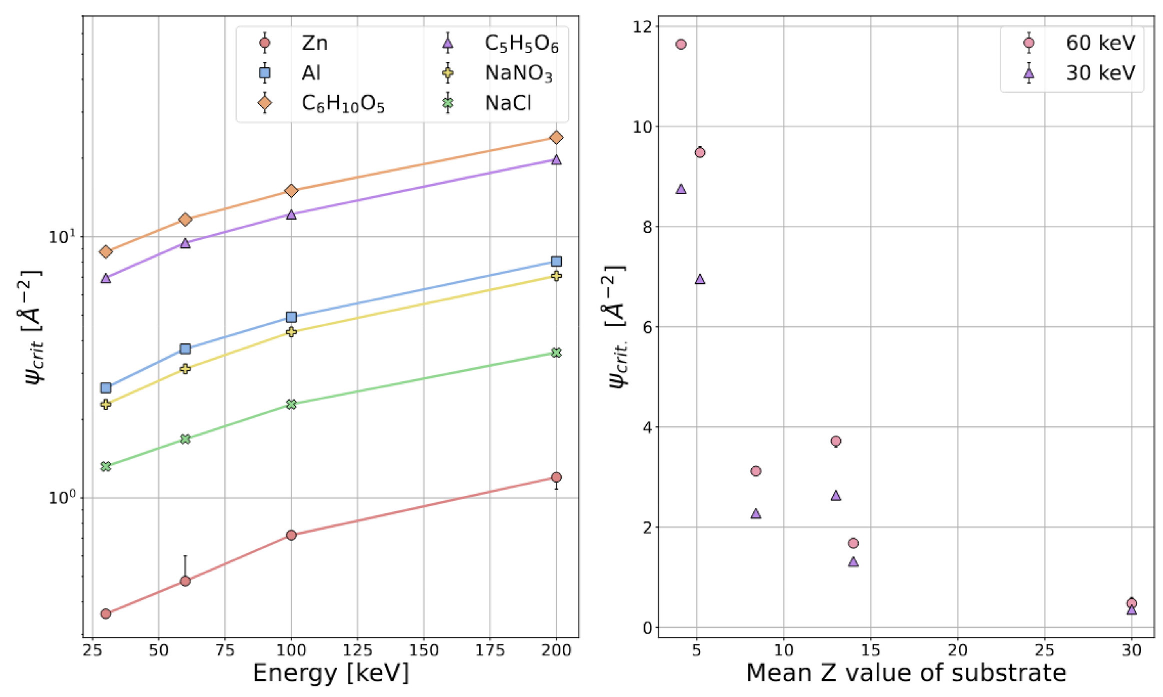
mixed beams

Simultaneous implantation:



Total start fluence of self sputtering ($\psi_{crit,tot}$) = independent of beam purity

Implantation foil

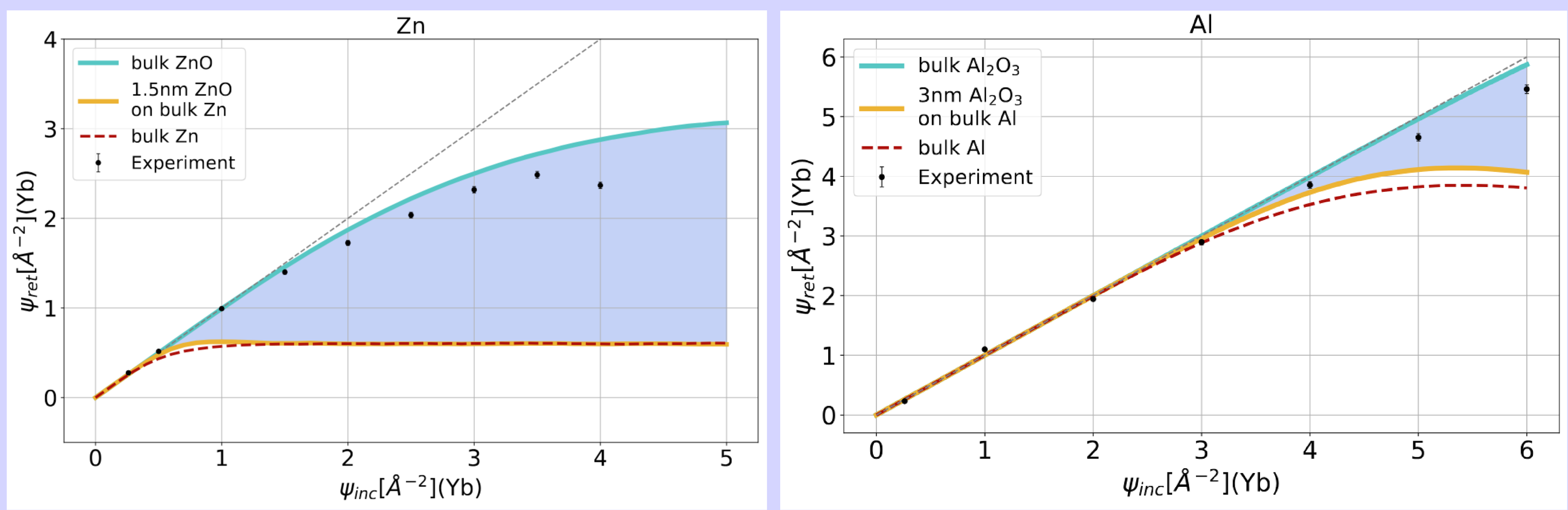


TRIDYN simulations for different substrates and beam energies

Validation of TRIDYN simulations

RADIATE

- Implantation of Yb in Zn and Al foils
- In between implantations: foils subject to air
  → Native layer of oxygen forms on surface
- Expectation within pure Zn/Al with native oxide layer simulation and ZnO/Al_2O_3 simulation



- Test for K implantation in Au and C
  → TRIDYN did not work well, likely due to immiscibility of the compounds

Back-of-the-evelope calculation framework

- Critical fluence of a **pure beam** ($\psi_{crit,tot}^{pure\ X}$) = good estimate for **total incoming critical fluence** ($\psi_{crit,tot}$, including all contaminants) at which **sputtering of the nuclide of interest** starts:

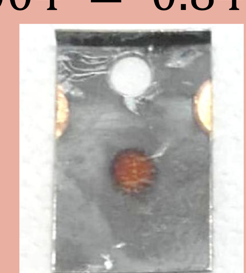
$$\psi_{crit,tot} \approx \psi_{crit,tot}^{pure\ X}$$

- Translated to experimentally accessible quantities:
  - **Critical activity** of isotope of interest:
    $A_{crit} = (2\pi\sigma_r^2)\kappa\lambda\psi_{tot,crit} \approx (2\pi\sigma_r^2)\kappa\lambda\psi_{tot,crit}^{pureX}$
  - Total (isotope of interest + contaminants) **integrated charge** on the foil:
    $Q_{crit}^{int} \approx (2\pi\sigma_r^2) q\, \psi_{tot,crit}^{pureX}$

Example

- 2020 ^{167}Tm collections [4]: 60keV implantation in Zn foil
- Expect self-sputtering effects at an implanted activity of:

$$A_{crit} \approx 2\pi\sigma_r^2\lambda\psi_{tot,crit}^{pureX}\kappa = 2\pi(9\cdot10^6\text{A})^2 0.095\frac{1}{days}0.480\frac{1}{A^2}0.004 = 0.845\text{MBq}$$



- But implantation was continued until ~5MBq ☹
  → Self-sputtering effects were clearly seen
  - Half of implanted activity was 'lost'
  - Au foil is visible through Zn coating because of sputtered Zn

Conclusion and outlook

- **Experimental confirmation** of TRIDYN simulations
- Self-sputtering is optimized by **minimizing the Z of the foil and beam**, **maximizing the energy of the beam** and maximizing the implantation area
- **Total fluence** can be used as indication for when self-sputtering starts to become significant

References

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[3] R. Stadlmayr, PS. Szabo, D. Mayer, C. Cupak, T. Dittmar, L. Bischoff, S. M ̈oller, M. Rasi ̆nski, RA. Wilhelm, W. M ̈oller, et al. Sputtering of nanostructured tungsten and comparison to modelling with TRIDYN. Journal of Nuclear Materials, 532:152019, 2020. doi:10.1016/j.jnucmat.2020.152019.
[4] R. Heinke, E. Chevallay, K. Chrysalidis, T. E. Cocolios, C. Duchemin, V. N. Fedosseev, S. Hurier, L. Lambert, B. Leenders, B. A. Marsh, et al., Efficient production of high specific activity thulium-167 at Paul Scherrer Institute and CERN-MEDICIS, Frontiers in medicine 8 (2021) 712374. doi:10.3389/fmed.2021.712374