UNIVERSITY OF LATVIA
Institute of Chemical Physics

MED-015: Scandium radionuclide production and mass separation.

Edgars Mamis

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02/07/2024

Outline:

1. Results from the research on production and mass separation;
2. Outcome of the project;
3. Proposal of the next project.

Sc radionuclide thermal release

Mamis, E.; Kalnina, P.; Duchemin, C.; Lambert, L.; Conan, N.; Deschamps, M.; Dorsival, A.; Froeschl, R.; Ruiz, F.O.; Theis, C.; Vincke, H.; Crepieux, B.; Rothe, S.; Pajuste, E.; Stora, T. Scandium thermal release from activated natTi and natV target materials in mixed particle fields: Investigation of parameters relevant for isotope mass separation. *Nuclear Instruments and Methods in Physics Research Sec- tion B: Beam Interactions with Materials and Atoms* **2024**, 553, 165400.

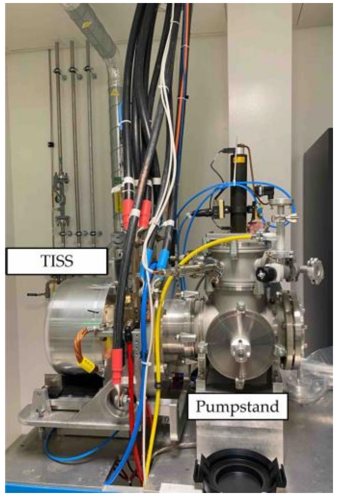

Figure 3. Pumpstand with TISS.

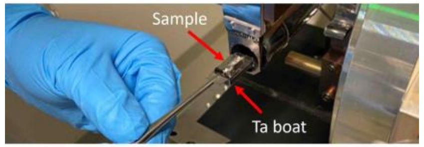

Figure 2. Sample loading in TISS on Ta boat.

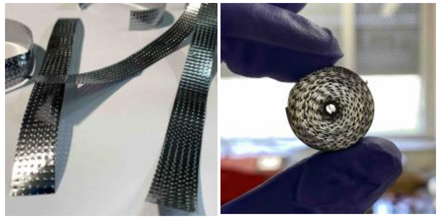

Figure 1. Embossed foils and roll sample.

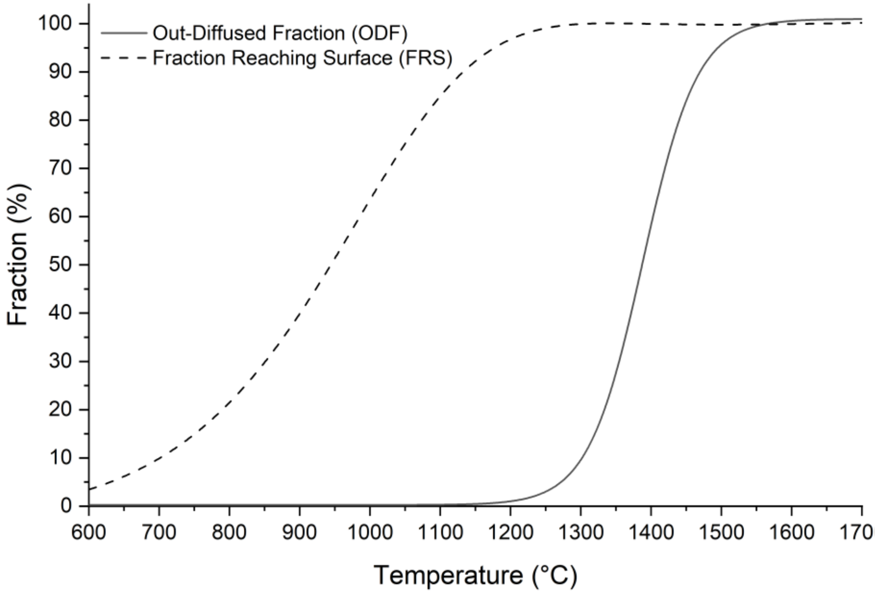

Figure 4. Estimations with the theoretical model formulated and described by F. Ogallar Ruiz [1].

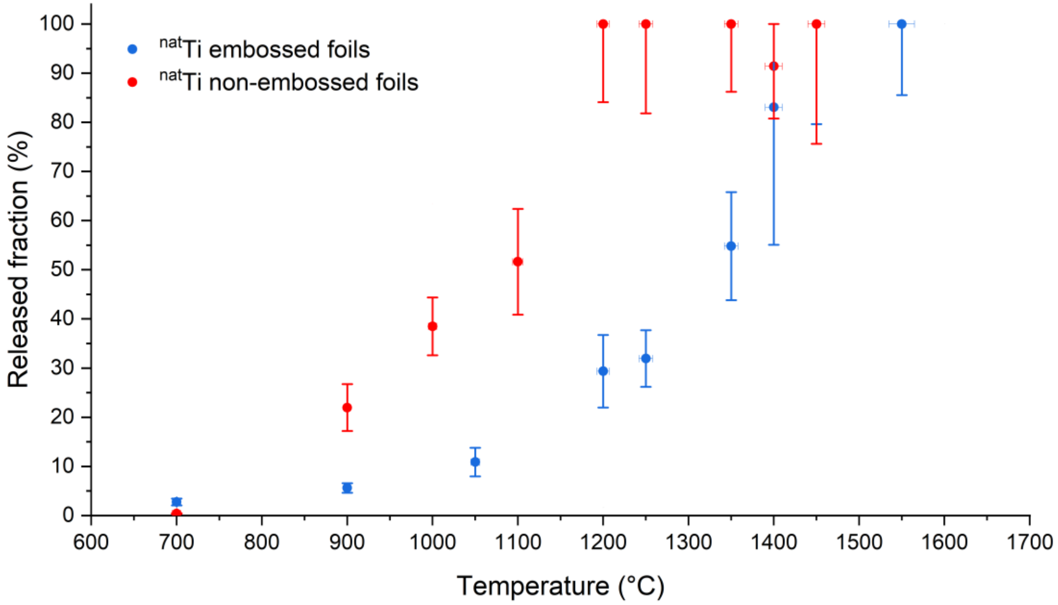

Figure 5. Sc fractional thermal release results from ^{nat}Ti foil roll samples.

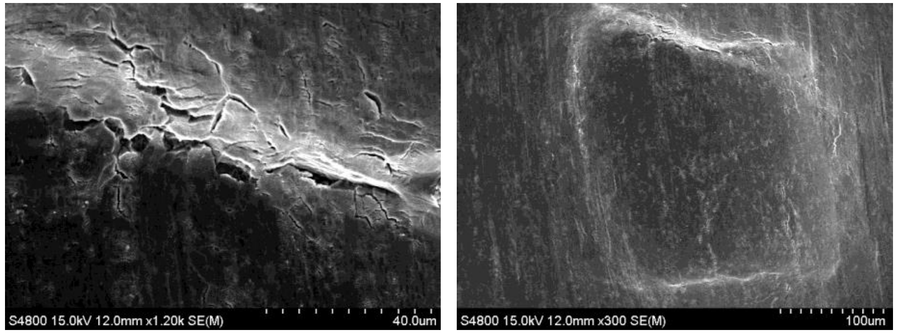

Figure 6. Scanning Electron Microscopy (SEM) images of embossed foil.

- Full Sc radionuclide release in 1 hour of hold time at the peak temperature
  - Embossed sample: 1450 °C;
  - Non-embossed: 1200 °C;

- The impact of heating ramp for ^{nat}Ti foil roll at 1400 °C
  - 0 h hold time: F_{rel} of 51 ± 11 %
  - 1 h hold time: F_{rel} was 91 ± 19 %

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[1] F. Ogallar Ruiz, Ph.D. thesis, *Evaluation and criticality assessment of radiologi- cal source terms to be used for fire risk studies at accelerator facilities.*, University of Granada (2021), https://cds.cern.ch/record/2802879

Sc radionuclide thermal release from ^{nat}V

Mamis, E.; Kalnina, P.; Duchemin, C.; Lambert, L.; Conan, N.; Deschamps, M.; Dorsival, A.; Froeschl, R.; Ruiz, F.O.; Theis, C.; Vincke, H.; Crepieux, B.; Rothe, S.; Pajuste, E.; Stora, T. Scandium thermal release from activated natTi and natV target materials in mixed particle fields: Investigation of parameters relevant for isotope mass separation. *Nuclear Instruments and Methods in Physics Research Sec- tion B: Beam Interactions with Materials and Atoms* **2024**, 553, 165400.

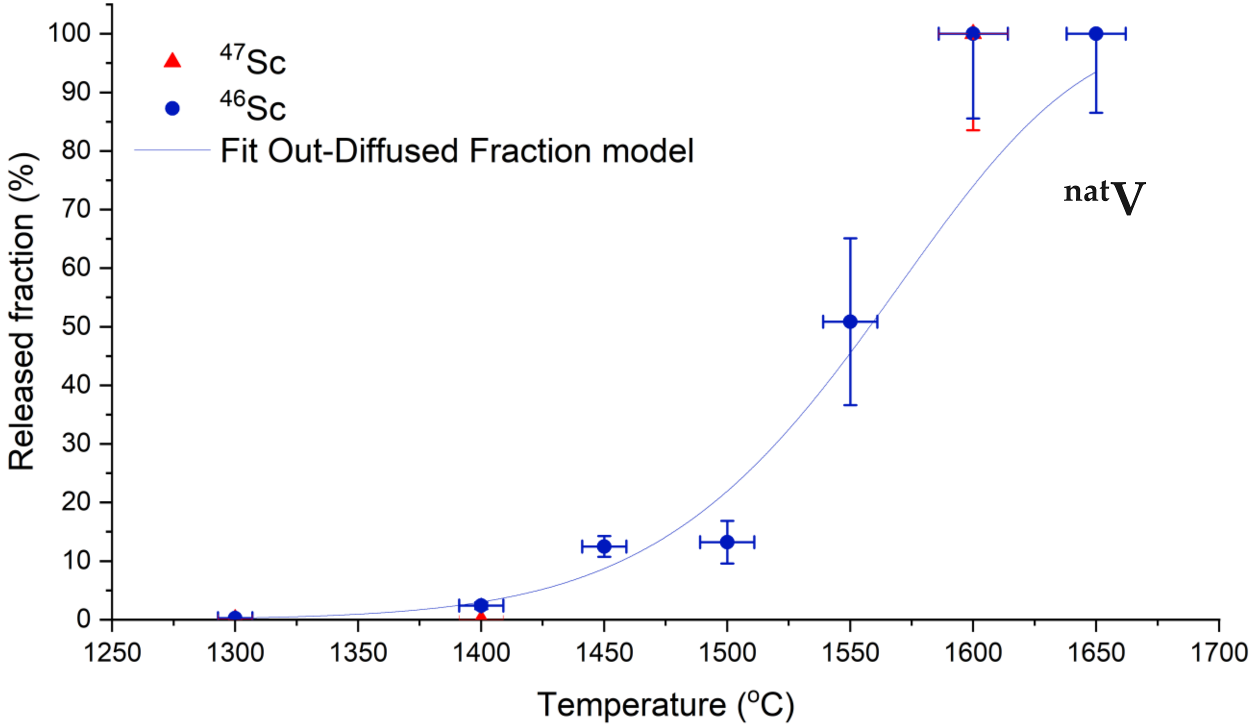


Figure 7. Sc radionuclide fractional thermal release results from ^{nat}V foil roll samples.

- Full Sc radionuclide release in 1 hour of hold time at the peak temperature for non-embossed foil samples: 1600 °C;

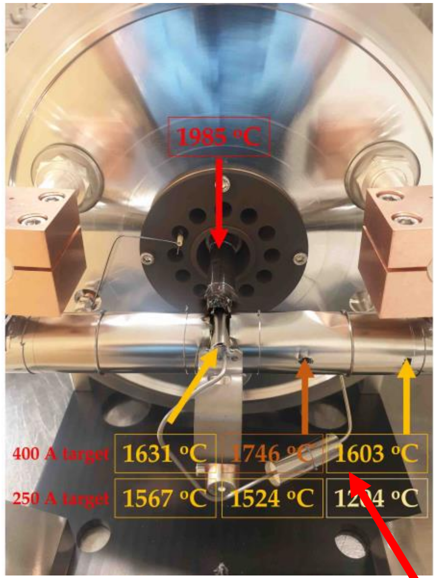
Target and ion source system developments

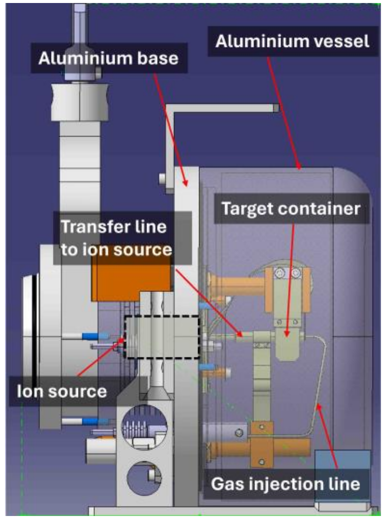
Mamis, E.; Duchemin, C.; Berlin, V.; Bernerd, C.; Bovigny, M.; Chevallay, E.; Crepieux, B.; Gadelshin, V.M.; Heinke, R.; Hernandez, R.M.; Pajuste, E.; Stora, T. et al. Target Development towards First Production of High-Molar-Activity 44*g*Sc and 47Sc by Mass Separation at CERN-MEDICIS. *Pharmaceuticals* **2024**, 17, 390.

Table 1: Target and ion source system configurations for Sc mass separation and collection studies.

TISS Nr.	Ion source	Configuration / modification	Target load
685M	Re surface	Standard Ta target container.	$^{45}Sc_2O_3$ sample.
686M	W surface	Standard Ta target container.	$^{45}Sc_2O_3$ sample.
689M	Re surface	Standard Ta target container.	Irradiated ^{nat}Ti and ^{nat}V rolls.
702M	VADIS	Standard Ta target container.	^{nat}TiC target load.
723M	VADIS	Standard Ta target container.	^{nat}Ti foil target load.
731M	VADIS	No target container. Small Ta oven for sample heating connected to gas injection line and directly into the ion source.	$^{45}Sc_2O_3$ and $^{45}ScF_3$ samples.
741M	VADIS	VD-5 Ta target container. Gas leak cooling structure with a thermocouple.	^{nat}Ti foil target load.
766M	VADIS	VD-5 - Ta target container. Gas leak cooling structure.	^{nat}V foil target load.
790M	VADIS	VD-5 with V foil lining inside target container and W lining for VADIS cathode and transfer line.	^{nat}V foil target load.
801M	VADIS	VD-5 with an increased gap between cathode and anode grid (1.7 mm).	$^{43}ScCl_3/^{44g/m}ScCl_3$ sample externally irradiated at PSI, Switzerland [33]
805M	VADIS	VD-5 with Ta target container that has the transfer line heating connection from the backside. Prototype gradient heat screen assembly.	^{nat}V foil target load.

VD-5 VADIS TISS with hot transfer line (~2000 °C)





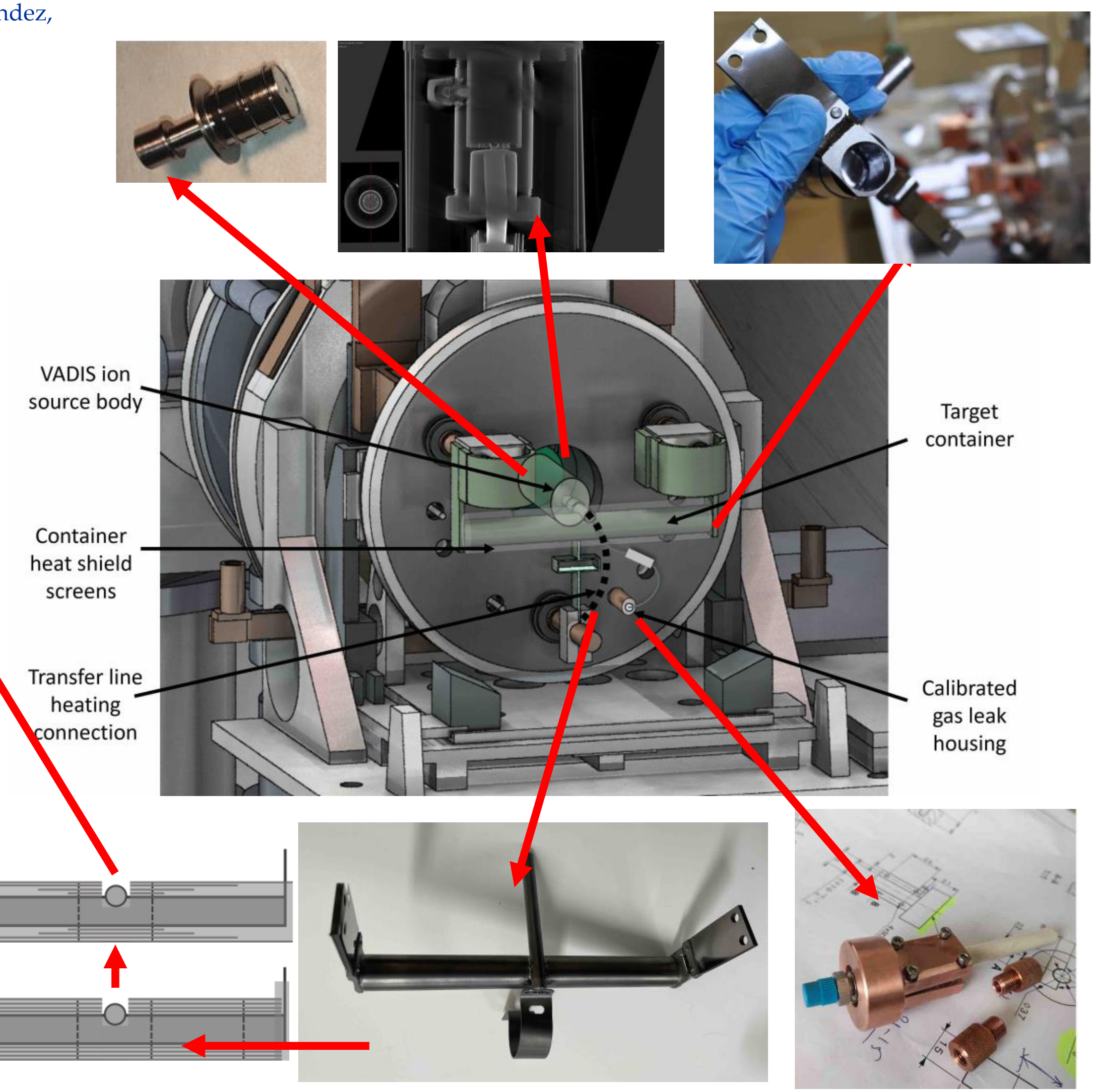


Figure 8. Target and ion source system with its main components and modifications.

VADIS – Versatile Arc Discharge Ion Discharge

Collection efficiency of Sc at CERN-MEDICIS

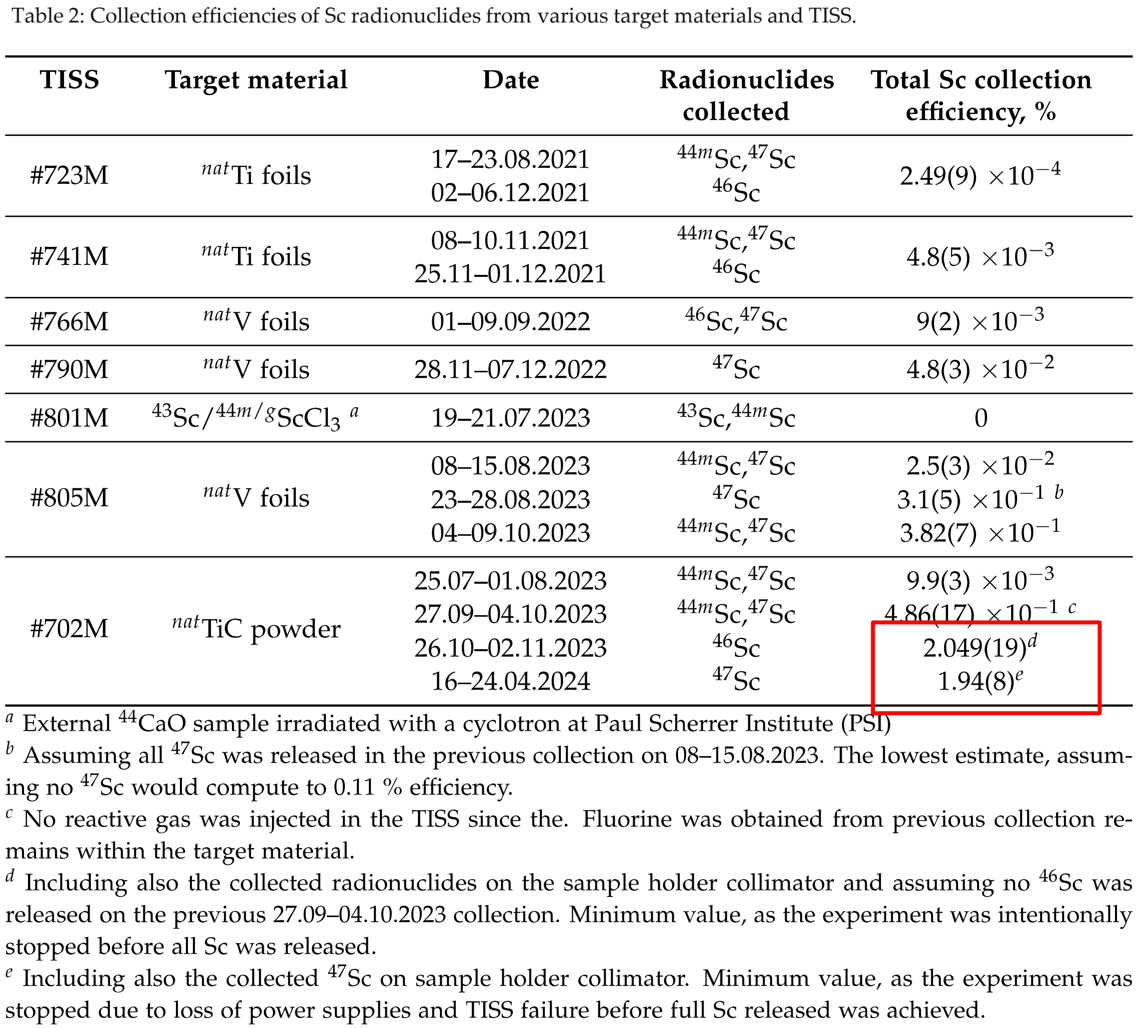


Table 2: Collection efficiencies of Sc radionuclides from various target materials and TISS.

TISS	Target material	Date	Radionuclides collected	Total Sc collection efficiency, %
#723M	^{nat}Ti foils	17–23.08.2021 02–06.12.2021	^{44m}Sc,^{47}Sc ^{46}Sc	$2.49(9)\ \times 10^{-4}$
#741M	^{nat}Ti foils	08–10.11.2021 25.11–01.12.2021	^{44m}Sc,^{47}Sc ^{46}Sc	$4.8(5)\ \times 10^{-3}$
#766M	^{nat}V foils	01–09.09.2022	^{46}Sc,^{47}Sc	$9(2)\ \times 10^{-3}$
#790M	^{nat}V foils	28.11–07.12.2022	^{47}Sc	$4.8(3)\ \times 10^{-2}$
#801M	^{43}Sc/$^{44m/g}$ScCl$_3$ [a]	19–21.07.2023	^{43}Sc,^{44m}Sc	0
#805M	^{nat}V foils	08–15.08.2023 23–28.08.2023 04–09.10.2023	^{44m}Sc,^{47}Sc ^{47}Sc ^{44m}Sc,^{47}Sc	$2.5(3)\ \times 10^{-2}$ $3.1(5)\ \times 10^{-1}$ [b] $3.82(7)\ \times 10^{-1}$
#702M	^{nat}TiC powder	25.07–01.08.2023 27.09–04.10.2023 26.10–02.11.2023 16–24.04.2024	^{44m}Sc,^{47}Sc ^{44m}Sc,^{47}Sc ^{46}Sc ^{47}Sc	$9.9(3)\ \times 10^{-3}$ $4.86(17)\ \times 10^{-1}$ [c] 2.049(19)[d] 1.94(8)[e]

[a] External ^{44}CaO sample irradiated with a cyclotron at Paul Scherrer Institute (PSI)

[b] Assuming all ^{47}Sc was released in the previous collection on 08–15.08.2023. The lowest estimate, assuming no ^{47}Sc would compute to 0.11 % efficiency.

[c] No reactive gas was injected in the TISS since the. Fluorine was obtained from previous collection remains within the target material.

[d] Including also the collected radionuclides on the sample holder collimator and assuming no ^{46}Sc was released on the previous 27.09–04.10.2023 collection. Minimum value, as the experiment was intentionally stopped before all Sc was released.

[e] Including also the collected ^{47}Sc on sample holder collimator. Minimum value, as the experiment was stopped due to loss of power supplies and TISS failure before full Sc released was achieved.

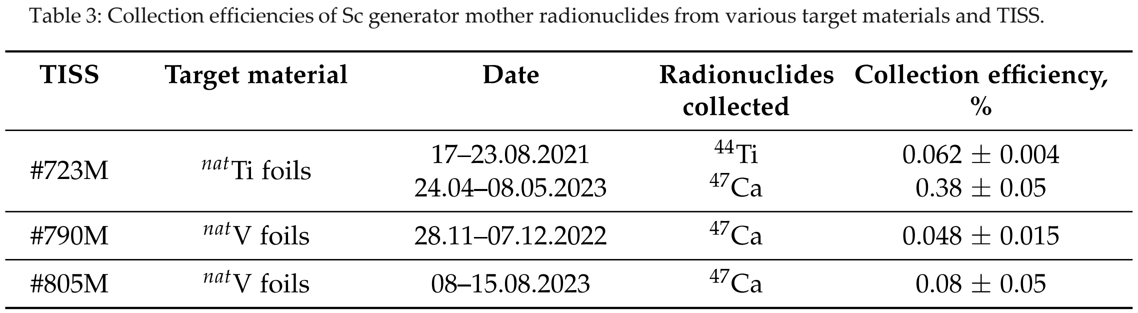


Table 3: Collection efficiencies of Sc generator mother radionuclides from various target materials and TISS.

TISS	Target material	Date	Radionuclides collected	Collection efficiency, %
#723M	^{nat}Ti foils	17–23.08.2021 24.04–08.05.2023	^{44}Ti ^{47}Ca	0.062 ± 0.004 0.38 ± 0.05
#790M	^{nat}V foils	28.11–07.12.2022	^{47}Ca	0.048 ± 0.015
#805M	^{nat}V foils	08–15.08.2023	^{47}Ca	0.08 ± 0.05

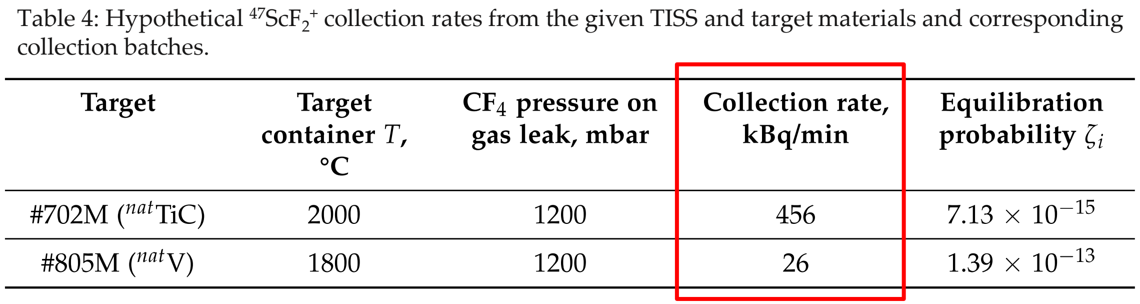


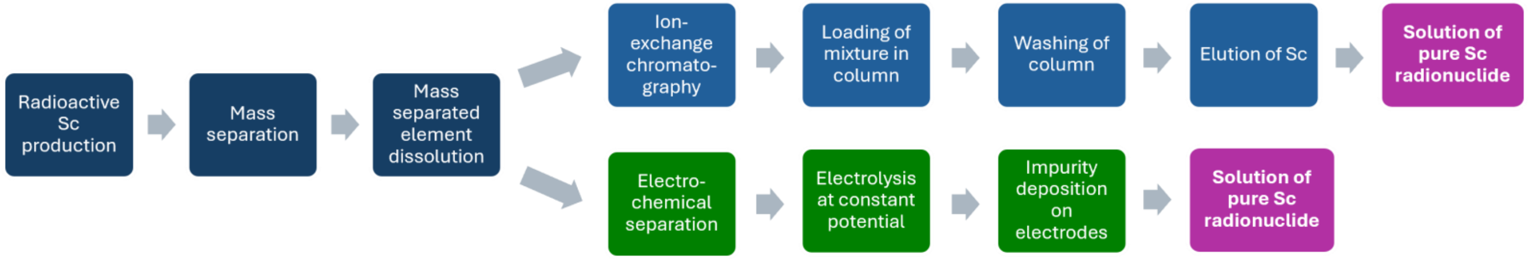
Table 4: Hypothetical ^{47}ScF$_2^+$ collection rates from the given TISS and target materials and corresponding collection batches.

Target	Target container T, °C	CF$_4$ pressure on gas leak, mbar	Collection rate, kBq/min	Equilibration probability ζ_i
#702M (^{nat}TiC)	2000	1200	456	7.13×10^{-15}
#805M (^{nat}V)	1800	1200	26	1.39×10^{-13}

- ➢ The developments in this study resulted in the first mass separation and collection of isotopically pure $^{44g/m}$Sc, ^{46}Sc and ^{47}Sc radionuclides with collected activity ranging from few kBq up to ~10 MBq.
- ➢ The obtained efficiencies in most cases are not the maximum achievable due to stopped collections from TISS failures

Stora, T.;; **Mamis, E.**; et. al. CERN-MEDICIS: Operational indicators to support the production of new med- ical radionuclides by mass separation. *Journal of Physics: Conference Series* **2024**, 2687, 8, 082039, IOP Publishing.

Radiochemical separation



Radioactive Sc production → Mass separation → Mass separated element dissolution

→ Ion-exchange chromatography → Loading of mixture in column → Washing of column → Elution of Sc → **Solution of pure Sc radionuclide**

→ Electro-chemical separation → Electrolysis at constant potential → Impurity deposition on electrodes → **Solution of pure Sc radionuclide**

Figure 9. Full Sc radionuclide separation and purification process flowchart.

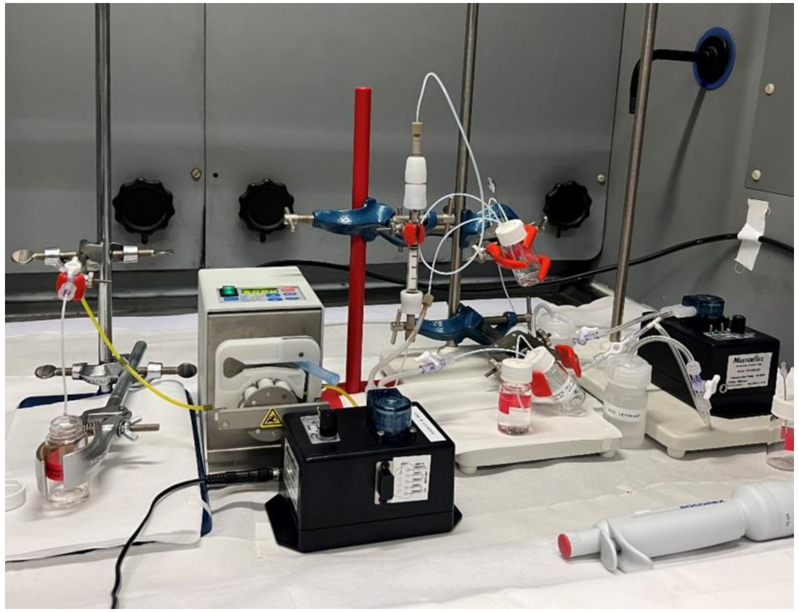


Figure 10. Automated radiochemical separation test setup.

- No ^{47}Sc was trapped within the column or capillaries of the system, indicating a high recovery yield (>99%).
- The radiochemical yield:
  - **81 ± 9 %** with Diba Omnifit Benchmark Microbore 3 mm column
  - **76 ± 12 %** with Diba Omnifit EZ 5 mm column
- The automated setup assembly with the radiochemical separation took ~2.5 hours in total;
- The electrochemical method - for largescale isobar contaminant removal before ion exchange separation.
- The results from the electrochemical separation confirmed that Sc cannot be deposited on an electrode in aqueous electrolysis, which becomes very useful for collection on a Cu foil.

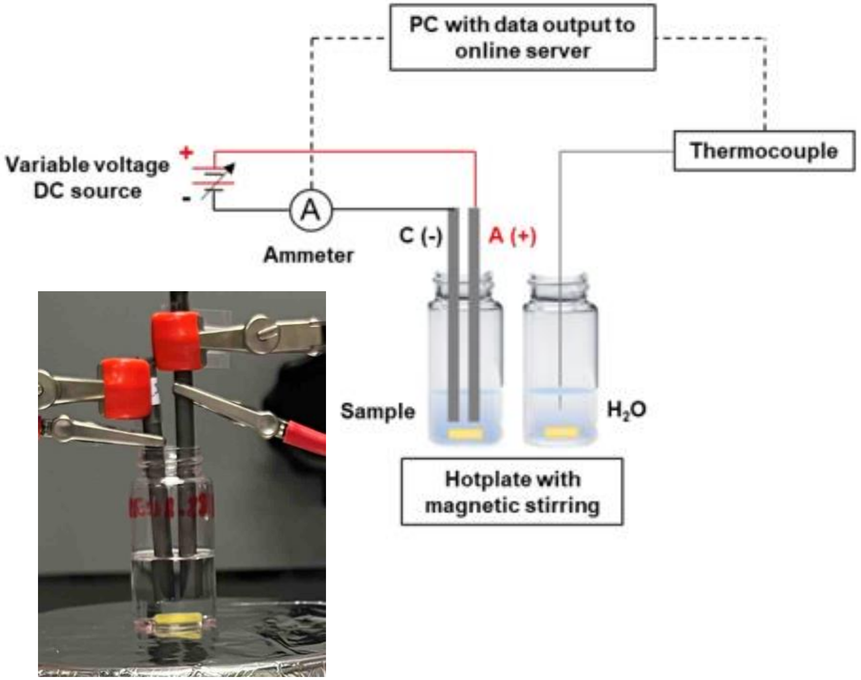


Figure 11. Schematic of electro-radiochemical separation setup.

Special thanks to R. J. Zabolockis and L.D. Pakalniete

Outcome

- Thermal release studies:
  - Publication;
  - Master’s thesis defended by P. Kalniņa;
  - Set-up employed for other radionuclide release studies.

- Target and ion source system developments:
  - Publication;
  - Two-step laser resonant ionization scheme employed at MELISSA;
  - Outlook for other target material investigations on Sc production and mass separation.

- Radiochemical and electrochemical separation:
  - Bachelor’s thesis defended by L.D. Pakalniete,

- Molecular radioactive ion beam studies:
  - Acquired data which will be used for publication;
  - Plan for another Sc external sample mass separation;
  - PhD thesis submitted by E.Mamis.

MEDICIS CERN

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MED-039: Tb and Sc radionuclide separation and purification from solid targets and liquids for theranostic radiopharmaceutical development.

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Maija Radziņa, Toms Torims

02/07/2024

Main tasks:

1. **Obtain bulk target material, suitable for ^{149}Tb, ^{152}Tb and ^{155}Tb radionuclide production at CERN-MEDICIS and cyclotrons, coupled with mass separation and/or chemical purification:**
    1. Investigate composite target materials with resistance to sintering, good mechanical stability and high specific surface area.
    2. Material sample characterization prior and after the thermal treatment and preparation for irradiation and mass separation by the means of Scanning Electron Microscopy (SEM), Energy Dispersive X-ray analysis EDX, Brunauer, Emmett and Teller (BET) method, pycnometry, Fourier Transform Infra-Red (FT-IR) spectroscopy.
    3. Experiments on Tb release from the TaC matrix by doping target material with stable Tb compounds can be made to test the suitability of the proposed composite materials.

2. **Perform mass separation of Tb radionuclides from irradiated Ta target materials:**
    1. Produce ^{149}Tb, ^{152}Tb and ^{155}Tb radionuclides with proton irradiation and perform mass separation and radionuclide collection at CERN-MEDICIS facility.

3. **Perform obtained Tb radionuclide separation and purification by conventional radiochemical methods and ion exchange resins, which includes developing a semi-automated radionuclide separation prototype system (also electrochemical methods).**
    1. Evaluate the separation efficiency, yield and radiochemical purity for method optimization.

Main tasks:

4. **Prepare, characterize and investigate target materials such as VC, VB, TiC (nanoscale) for the Sc production and mass separation with collected Sc sample post purification using the already developed chemical separation methods.**

5. **Test the separation of Tb and Sc radionuclide from mass separation collection foils with electrochemical methods.**
    1. Investigate non-aqueous electrolysis as means of the radionuclide separation from isobaric contaminants.

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