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²²⁵Ac: Getting it out of a ThO₂ target and separating it from ²²⁷Ac for medical purposes at CERN MEDICIS

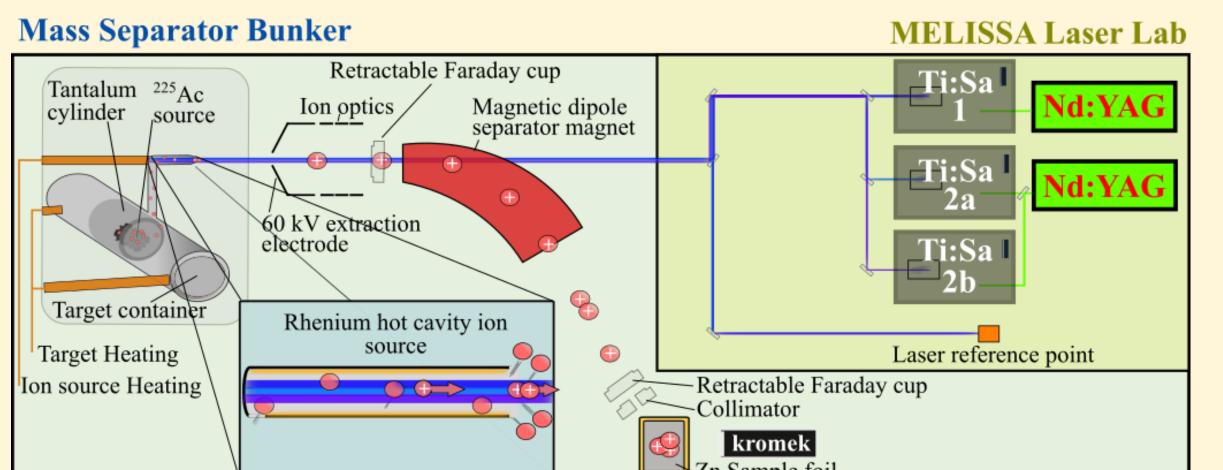
 $^{225}\!\mathrm{AC}$ is a promising candidate medical radio-isotope for the targeted alpha therapy of certain distributed cancers. One of the main pathways $^{\circ}$ that is being explored for its production is the high-energy proton spallation of actinide-based targets. A method that is both isotopically selective and efficient is then required to recover 225 Ac from the hundreds of co-produced reaction products. Of these, the trickiest radioactive contaminant of concern is 227 Ac ($T_{1/2}$ = 22.8 years), that could preclude medical use if not suppressed further than the relative in-target production yield [1].

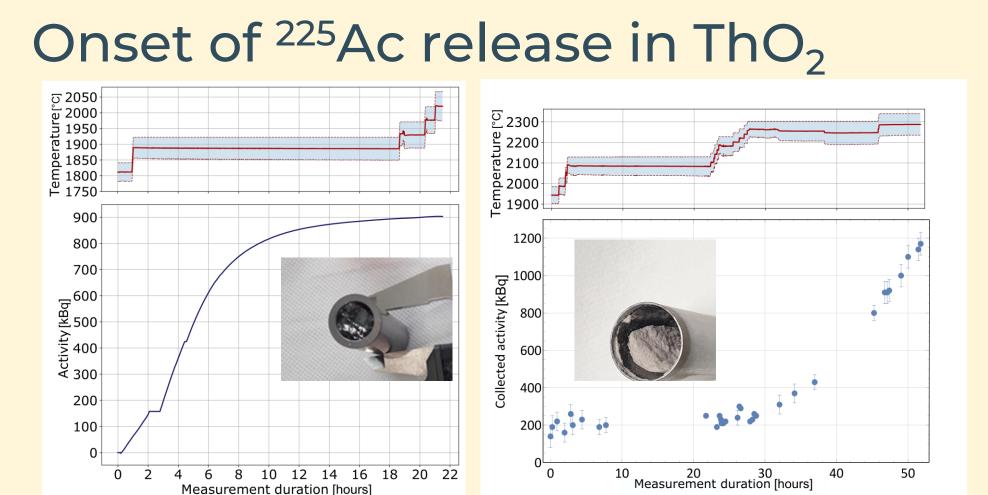
This poster presents three aspects of Laser ionization and Mass Separation (LIMS), a technique that purifies and separates ²²⁵Ac from ²²⁷Ac from an irradiated actinide target. Firstly, we examine the onset of release temperature for ²²⁵Ac from a chemically inert environment and a ThO₂ target matrix respectively. Secondly, we deduce the contamination level of ²²⁷Ac in an ²²⁵Ac sample obtained from an irradiated target on which LIMS was performed. Finally, LIMS is compared to radio-chemical separation. Possible techniques to best balance purity and efficiency are then discussed with consideration to scalability to conclude.

CERN MEDICIS isotope separator overview

CERN MEDICIS is a facility equipped with an offline mass-separator for purification of medical radio-nuclides from radioactive source material.

The material is heated in vacuum. The resulting atomic vapor is ionized in a laser ion source and accelerated into a beam that is then separated by mass.





We observed that 1900°C was great, to extract ²²⁵Ac dried on a rhenium substrate, But when it was dried on a felt of thoria, It was seen only at 2250°C, to the operators' euphoria.

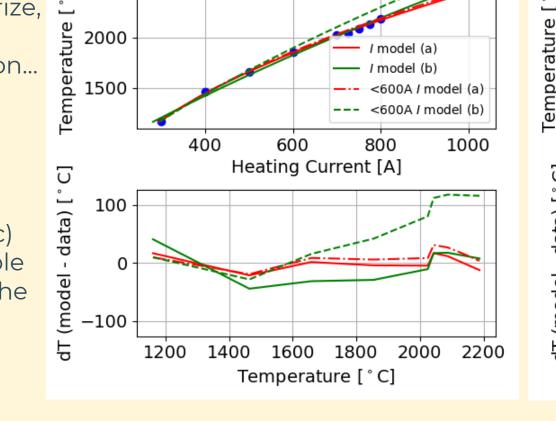
Left fig: The backgroundcorrected integrated ion current converted to equivalent end of collection activity of ²²⁵Ac from inert chemical environment.

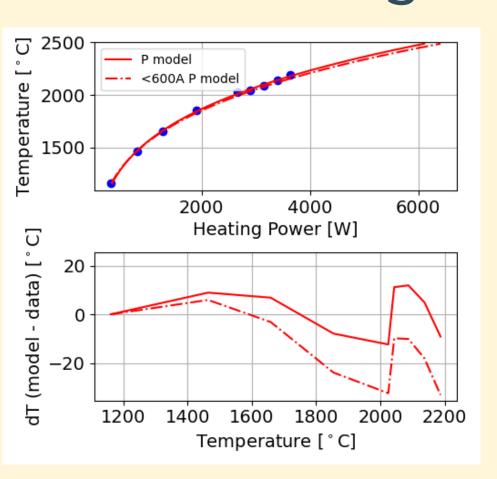
Right fig: The gamma activity of ²¹³Bi during implantation of ²²⁵Ac from ²²⁵Ac dried on ThO₂ matrix.

Target temperature calibration for resistive heating

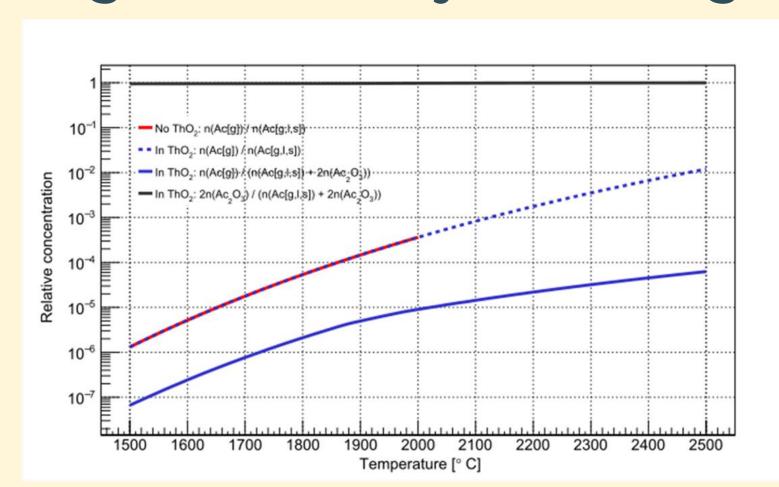
Before we can analyse, the temperature at which Ac will vaporize, We must first perform a calibration, of resistive and radiative heat dissipation... Three models were fit to temperature versus current or power data on MEDICIS targets. (a) $I = \sqrt{\frac{a'T}{c'+T} + \frac{b'T^4}{c'+T}}$ Models (a) and (c) were most reliable so the mean of the (b) $I = \sqrt{a''T + b''T^{4}}$ two models was used for temperature

(c) $P = aT + bT^4$





Target chemistry inhibiting Ac vaporization

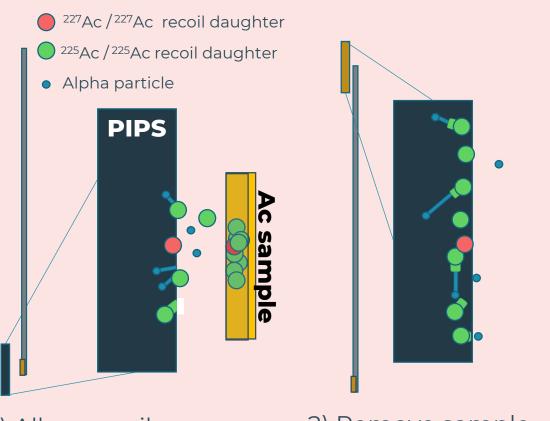


extrapolation.

A HSC chemistry simulation was performed, To see what species in the target formed. With an excess of oxygen, there is reason to panic, because Ac readily forms a ceramic. Ac₂O₃ is made instead of Ac vapor, so the target temperature must

be made greater.

Measuring ²²⁷Ac activity: Principle



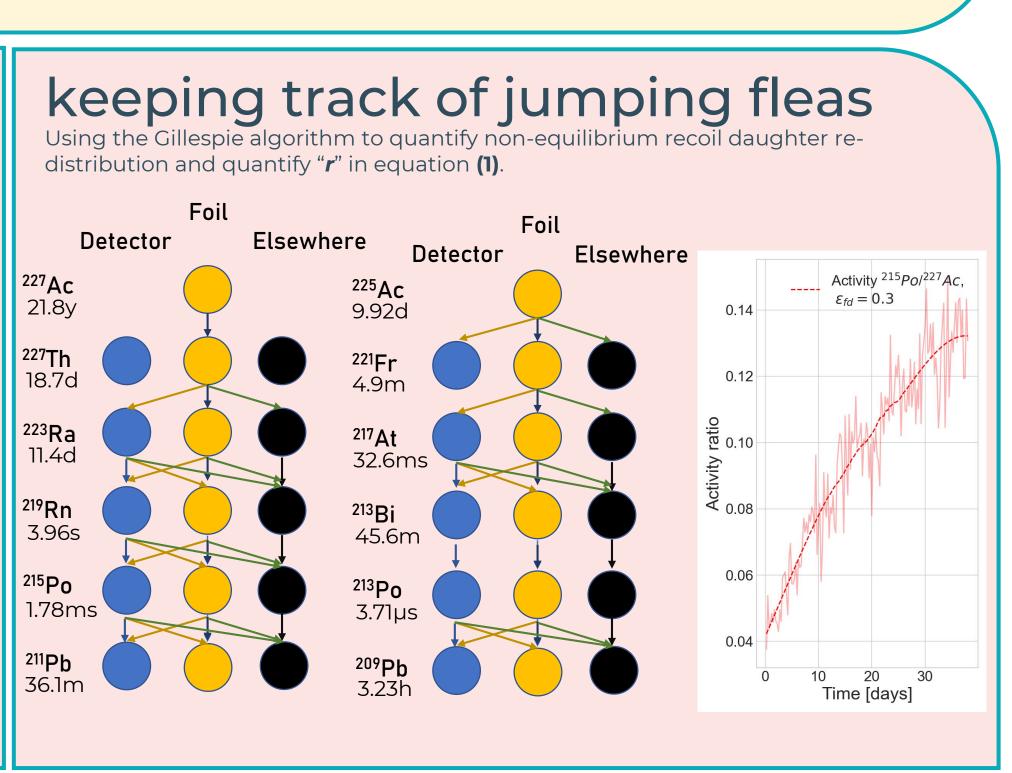
at first.

1) Allow recoil daughters to accumulate on PIPS detector for 2/3 of time available for experiment. This happens as $E_{rec} > E_{imp}$

3) Continue 2) Remove sample taking recoil foil and take recoil spectrum, mostly remaining observing decays 1/3 of of ²²⁵Ac daughters

We performed Alpha decay spectroscopy of recoil daughters (E_{rec} ~ 100keV) from an ²²⁵Ac sample collected (at E_{imp} = 60keV) from irradiated ThO₂. The daughters of ²²⁷Ac could be detected thanks to the long half-life of α -decay daughter ²²³Ra (T_{1/2} = 11.4 d). This became visible spectrum for once the ²²⁵Ac daughters $(T_{1/2}^{\text{max}} =$ 45.6m) decayed from the detector experiment

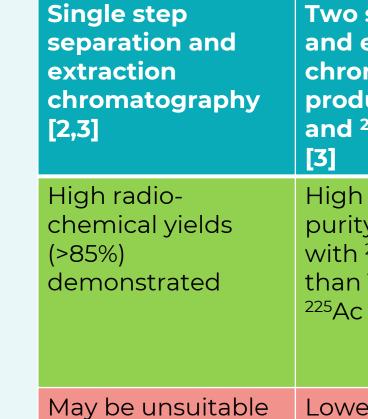
Measuring ²²⁷Ac activity: Spectra (a): Decay curve of ²¹⁵Po (7.2-7.5 MeV) (d) ²⁰⁰ (b): Alpha decay energy spectrum + CERN y ²²⁵Ac from 1 - 18 days + CERN y ²²⁵Ra ¥ 100 ₹ KUL γγ after foil removal (c):Decay curve of ²¹³Po (7.6-8.2 MeV) (d): Activity of ²²⁵Ac and ²²⁵Ra measured by γγ Time since collection end [days] coincidence spectroscopy.



To calculate the 227 Ac activity in the sample, we made use of the following ratio: $r = (\frac{A^d [^{215}Po](t_r)}{A[^{227}Ac](t_r)})/(\frac{A^d [^{213}Po](t_r)}{A[^{225}Ac](t_r)})$. Here A^d refers to the activity from nuclei distributed on the detector surface. The count rates, C^d that were measured from figs. (a) and (c) replace A^d in this ratio as the geometric ratio between C^d and A^d are assumed to be equal for same-generation alpha daughters. r was calculated by simulation. $A^{[225}Ac](t_r)$ was determined from "efficiency-free" $\gamma\gamma$ spectroscopy. The activity of $C^{227}Ac$ was then calculated from eq. (1). $A[^{227}Ac](t_{eoc}) = \frac{1}{r} \frac{C^{d}[^{215}Po](t_r)}{C^{d}[^{213}Po](t_r)} A[^{225}Ac](t_r) e^{\lambda^{227}(t_r - t_{eoc})}$ (1) $C^{d}[^{215}Po](t_r)$ $C^d[^{213}Po](t_r)$ $A[^{225}Ac](t_r)$ $A[^{227}Ac](t_{eoc})$ $A[^{227}Ac](t_{eoc})/A[^{225}Ac](t_{eoc})$ Preliminary results:

Comparison of separation methods

time



for routine clinical

use due to ²²⁷Ac

contamination

Two step separation and extraction chromatography to produce ²²⁵Ra/²²⁵Ac and ²²⁵Ac fractions

High radionuclidic purity of ²²⁵Ac (99%) with ²²⁷Ac activity less than 10-4% of that of

Lower overall ²²⁵Ac yield compared to ²²⁵Ac produced in target (15%) due to lower ²²⁵Ra cross section.

Laser ionization and mass separation of nuclides that are sweated out of target

surface.

Super-pure ²²⁵Ac, with ²²⁷Ac activity ~10⁻⁵ % that of ²²⁵Ac. Target dissolution not required. Could obtain ²²⁵Ra/²²⁵Ac generator and pure ²²⁵Ac from same process Process limited by ionsource efficiency to ~13%. High temperature needed Variability and scalability not known

Concept brainstorm: How to make the most of irradiated actinide

79.5(2) cps

0.00198(8) cps

Atomic Vapor Laser Isotope Separation-type concept?

Extraction Electrostatic Collection foil deflector plates Ac HFS spectra courtesy: A. Teigelhoefer et al. Isotope shift -tuned

Heating current

targets for medical grade ²²⁵Ac production? An eye towards uranium enrichment methods but applied to medical isotope purification

4.3(3) x 10⁻⁷

1.10(6) 0.063(5) Bq

1) Formation of Van-der-Waals molecules in gas-jet expansion (e.g with de Laval nozzle).

Separation of Isotopes by Laser Excitation-type concept?*[4,5]

- 2) Selective resonant laser excitation of vibrational state in the ²²⁵Ac isotopomer 3) Separation of the dissociated product from the beam through recoil
- 4) Collection by condensation on a cooled surface on the beam-line interior.
- Product (light element) Fig courtesy: Eerkens, Jeff W., and Jaewoo Kim. "Isotope separation by selective laser-assisted repression of condensation in supersonic free $P = 10^{-3} \text{ to } 10^{-2} \text{ kPa}$ $P = 10^{-4} \text{ to } 10^{-3} \text{ kPa}$ jets." *AIChE journal* 56.9 (2010): Adjustable -2331-2337. nozzle positioner ("rim gas") Tails (heavy element) Laval nozzle Gas inlet tube ("core gas") *SILEX process can only be $(P \sim 1.5 \text{ kPa})$ Crosswise laser beam guessed at due to industrial secrecy related to proliferation concerns Evacuated chamber Evacuated chamber Supersonic jet contour

Outlook: The most efficient way to produce scalable quantities of ²²⁵Ac is probably a mass-selective or isotope-selective separation method that does not involve chemical selectivity (like a laser ion source) following radio-chemical separation. Potential methods such as AVLIS or SILEX could be looked into. However, the performance of LIMS should also be characterized as a function of atomic flux through the ion source to investigate the scalability of the method.

[1] Morgenstern, A., et al. Supply and clinical application of ²²⁵Ac and ²¹³Bi. Seminars in nuclear medicine. Vol. 50 (2), 2020.

2720(120) kBq

- [2] Aliev, R.et al. Isolation of medicine-applicable ²²⁵Ac from thorium targets irradiated by medium-energy protons. Solvent Extr. Ion Exch. Vol. 32, 2014 [3] Robertson, A. K.et al. ²³²Th-spallation-produced ²²⁵Ac with reduced ²²⁷Ac content. Inorg. Chem. Vol. 59, 2020.
- [4] Makarov, G. Low energy methods of molecular laser isotope separation. Reviews of topical problems. Vol. 58, 2015 [5] Lee, Y. T. "Isotope separation by photodissociation of Van der Wall's molecules." (1977).

Relative frequency (GHz)