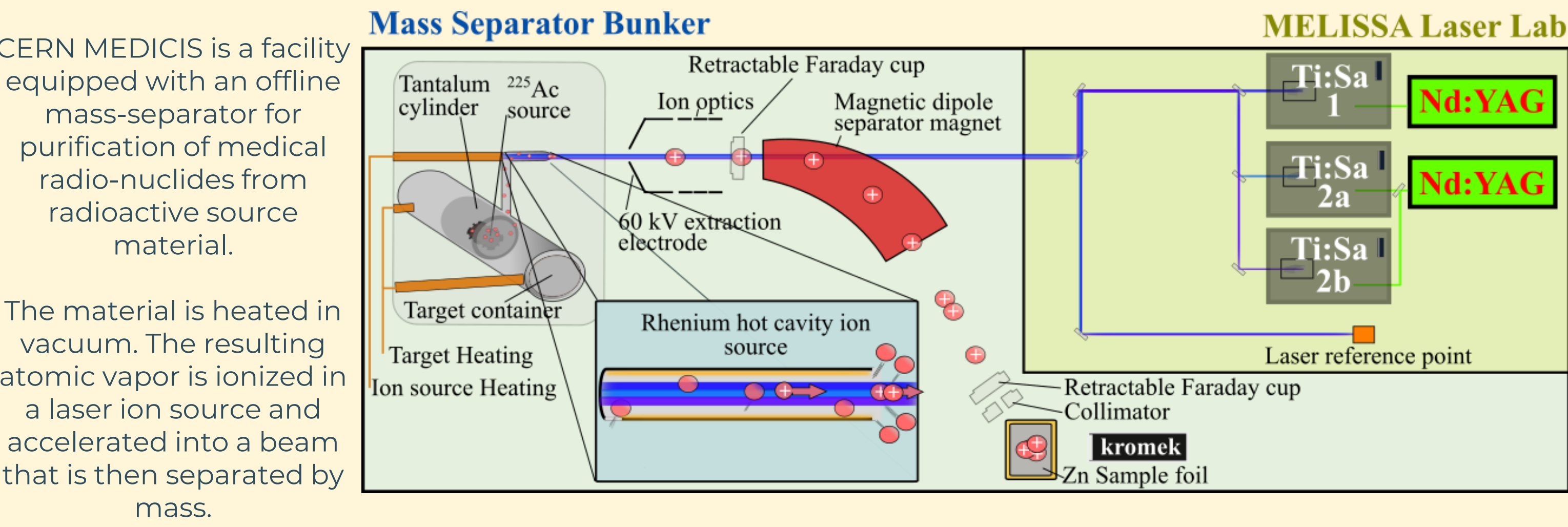


²²⁵Ac : Getting it out of a ThO₂ target and separating it from ²²⁷Ac for medical purposes at CERN MEDICIS

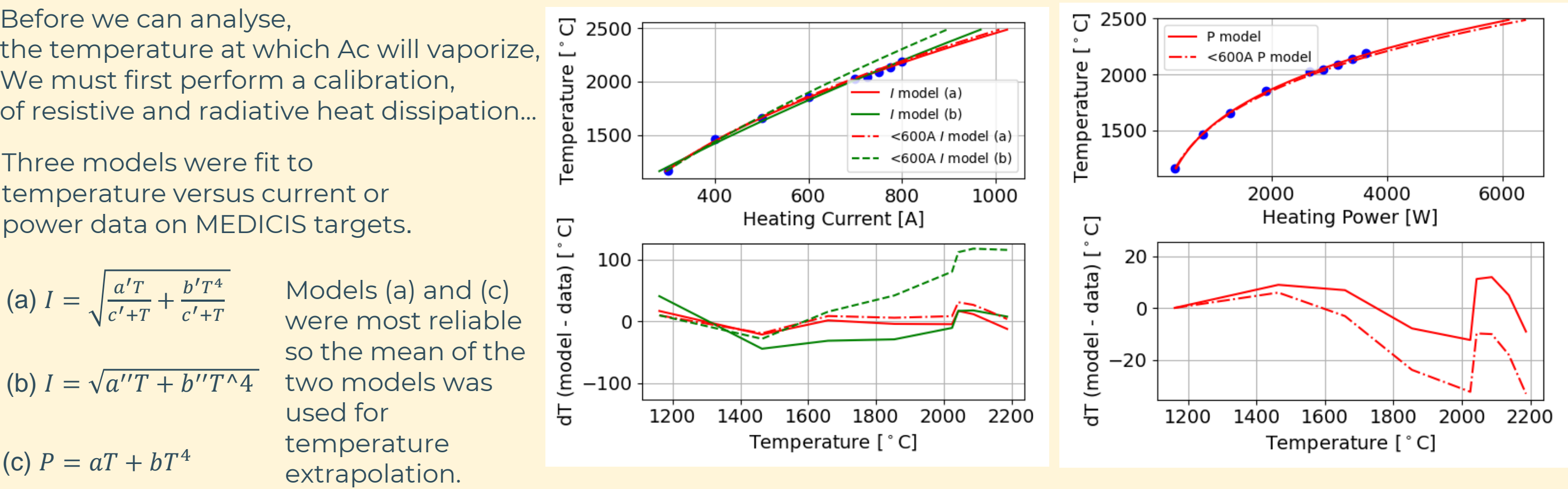
²²⁵Ac is a promising candidate medical radio-isotope for the targeted alpha therapy of certain distributed cancers. One of the main pathways that is being explored for its production is the high-energy proton spallation of actinide-based targets. A method that is both isotopically selective and efficient is then required to recover ²²⁵Ac from the hundreds of co-produced reaction products. Of these, the trickiest radioactive contaminant of concern is ²²⁷Ac (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.

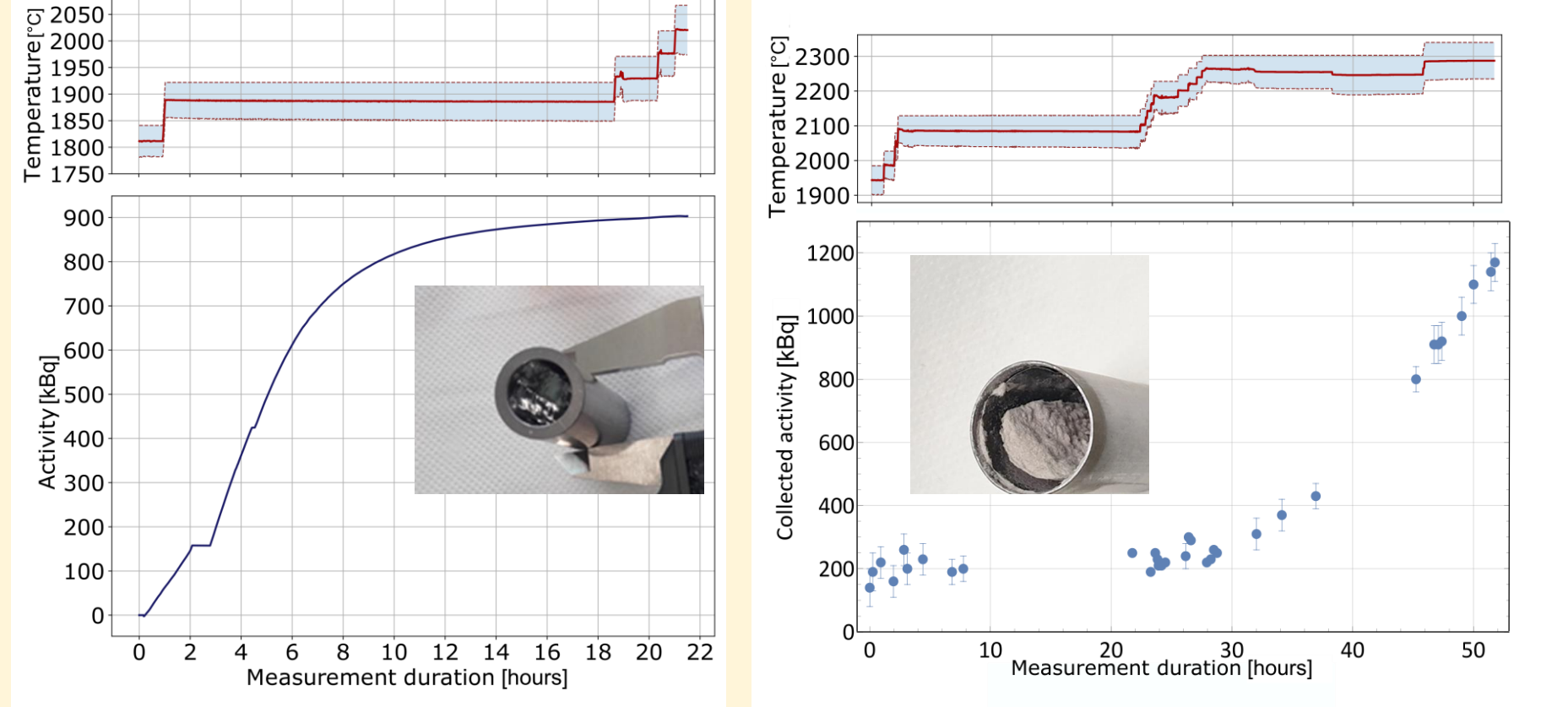
CERN MEDICIS isotope separator overview



Target temperature calibration for resistive heating



Onset of ²²⁵Ac release in ThO₂

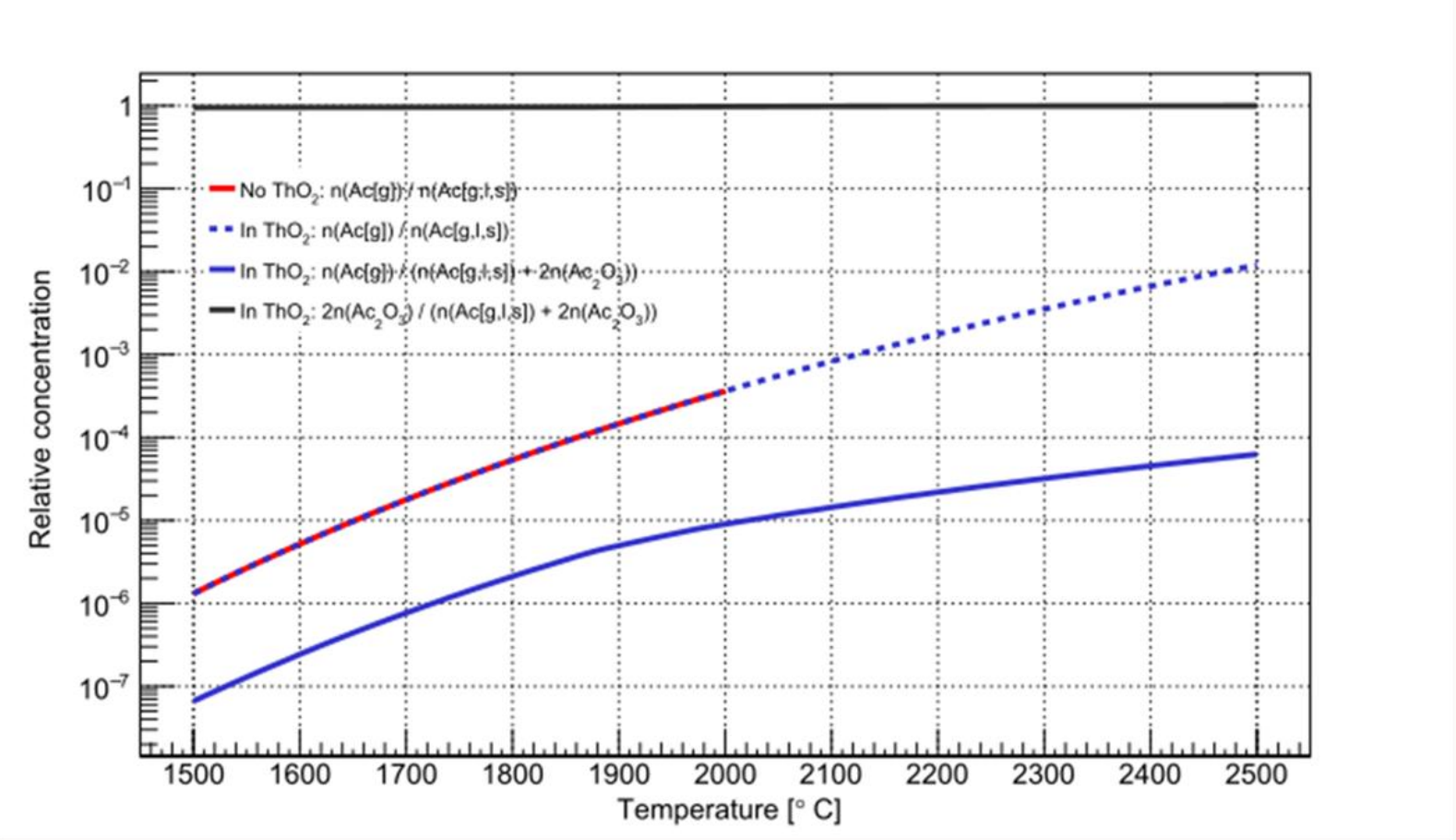


We observed that 1900°C was great, to extract ²²⁵Ac dried on a rhenium substrate, But when it was dried on a felt of thoria, It was seen only at 2250°C, to the operators' euphoria.

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

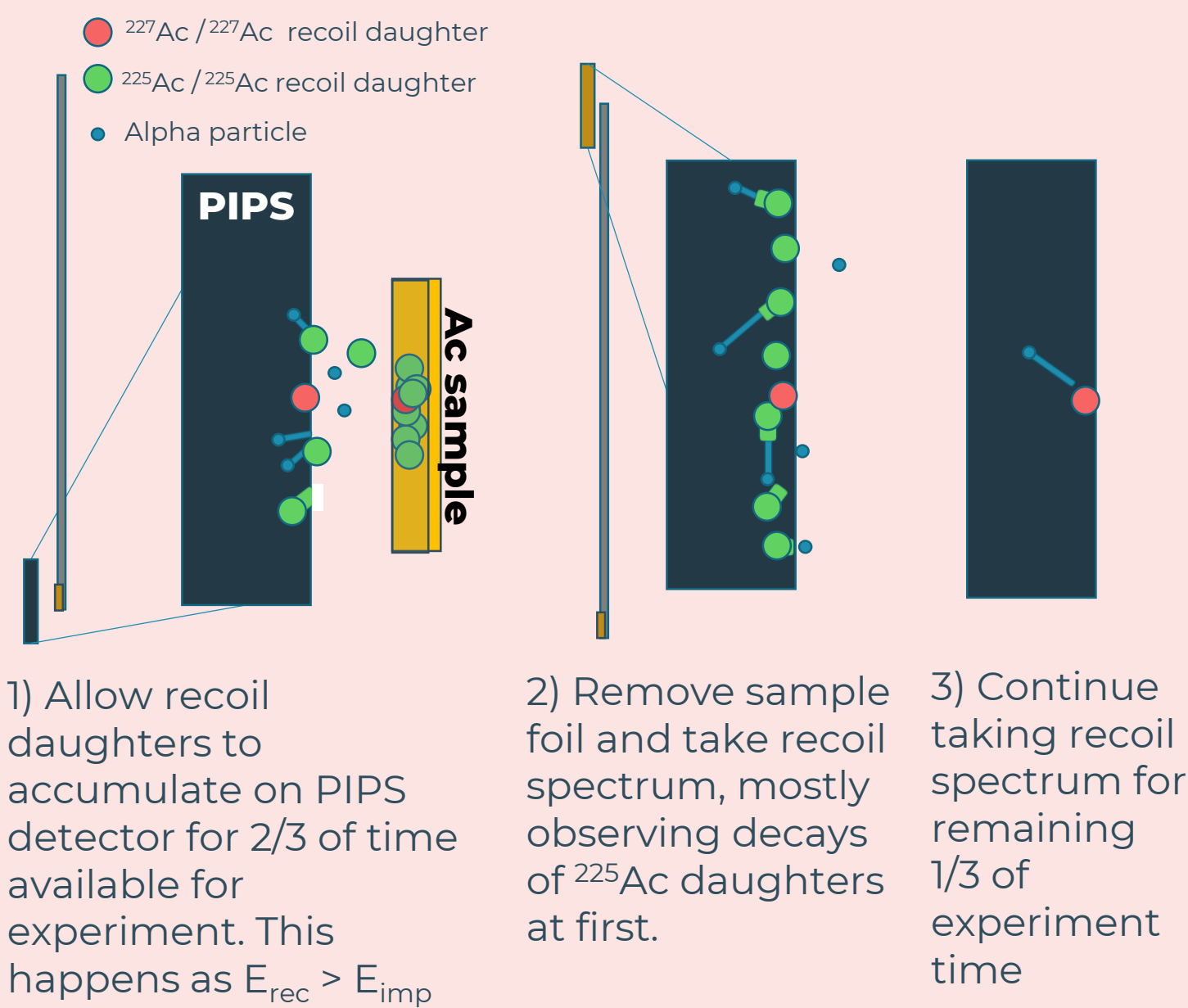
Right fig: The gamma activity of ²¹³Bi during implantation of ²²⁵Ac from ²²⁵Ac dried on ThO₂ matrix.

Target chemistry inhibiting Ac vaporization



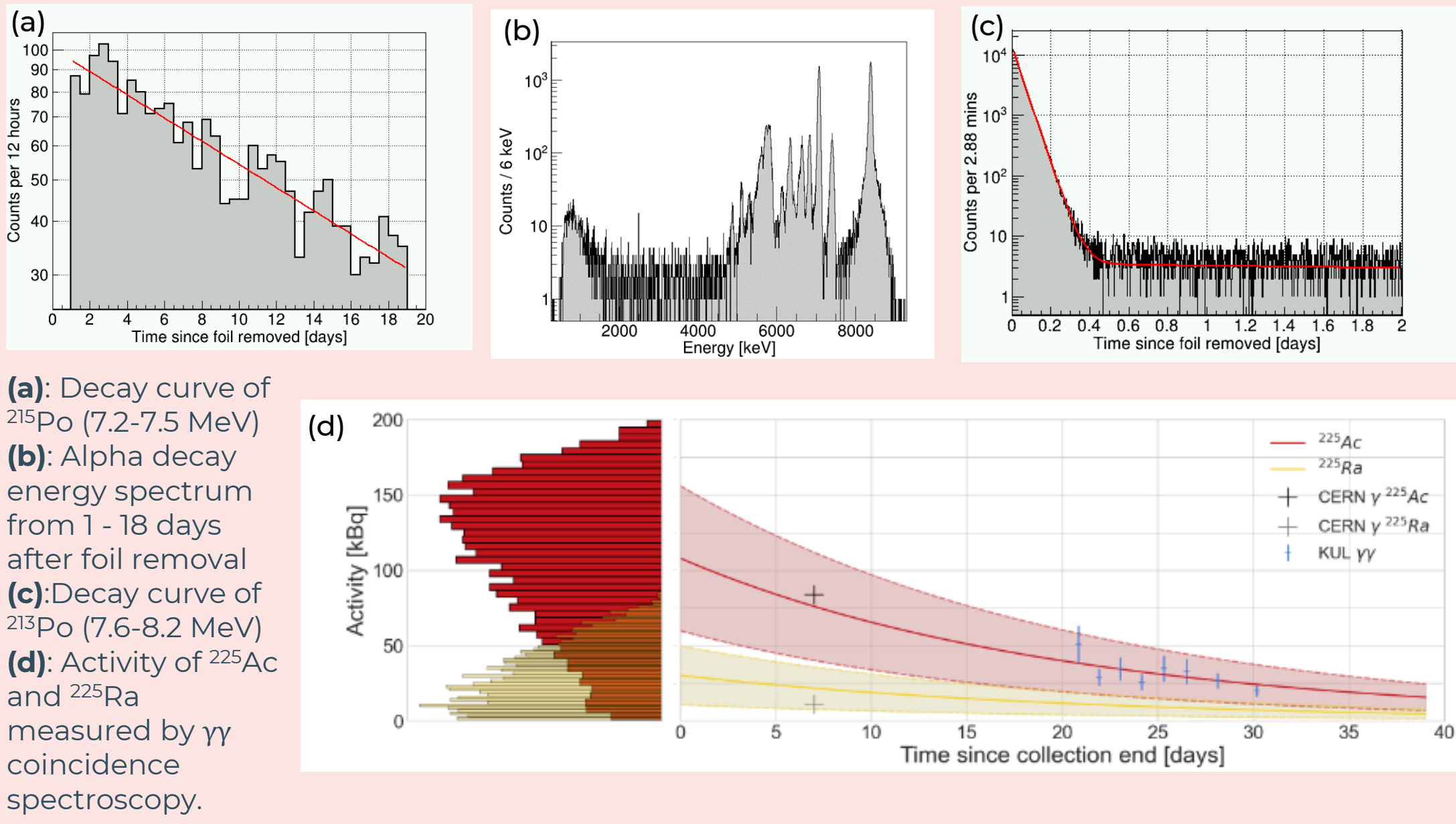
A HSC chemistry simulation was performed, To see what species in the target formed. With an excess of oxygen, there is reason to panic, because Ac readily forms a ceramic. Ac₂O₃ is made instead of Ac vapor, so the target temperature must be made greater.

Measuring ²²⁷Ac activity: Principle



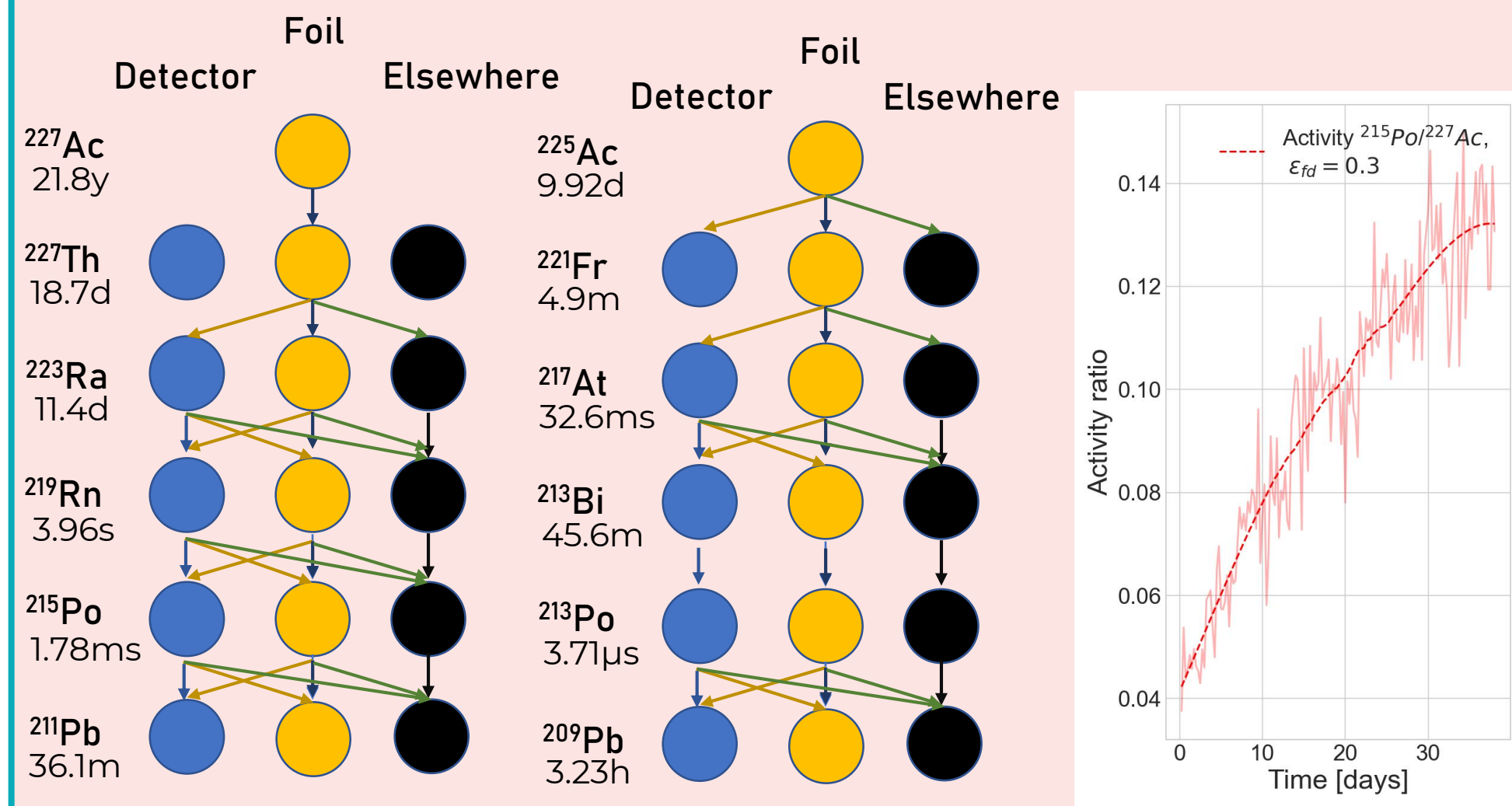
We performed Alpha decay spectroscopy of recoil daughters (E_{rec} ~ 100keV) from an ²²⁵Ac sample collected (at E_{imp} = 60keV) from irradiated ThO₂. The daughters of ²²⁷Ac could be detected thanks to the long half-life of α-decay daughter ²²³Ra (T_{1/2} = 11.4 d). This became visible once the ²²⁵Ac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

Measuring ²²⁷Ac activity: Spectra



keeping track of jumping fleas

Using the Gillespie algorithm to quantify non-equilibrium recoil daughter re-distribution and quantify "r" in equation (1).



To calculate the ²²⁷Ac activity in the sample, we made use of the following ratio: $r = \frac{A^d[^{215}\text{Po}](t_r)}{A^{[^{227}\text{Ac}]}(t_r)} / \frac{A^d[^{213}\text{Po}](t_r)}{A^{[^{225}\text{Ac}]}(t_r)}$. 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[²²⁵Ac](t_r) was determined from "efficiency-free" γγ spectroscopy. The activity of ²²⁷Ac was then calculated from eq. (1).

$$A[^{227}\text{Ac}](t_{\text{eoc}}) = \frac{1}{r} \frac{C^d[^{215}\text{Po}](t_r)}{C^d[^{213}\text{Po}](t_r)} A[^{225}\text{Ac}](t_r) e^{\lambda^{227}(t_r - t_{\text{eoc}})} \quad (1)$$

Preliminary results:

C ^d [²¹⁵ Po](t _r)	C ^d [²¹³ Po](t _r)	A[²²⁵ Ac](t _r)	r	A[²²⁷ Ac](t _{eoc})	A[²²⁷ Ac](t _{eoc})/A[²²⁵ Ac](t _{eoc})
0.00198(8) cps	79.5(2) cps	2720(120) kBq	1.10(6)	0.063(5) Bq	4.3(3) x 10 ⁻⁷

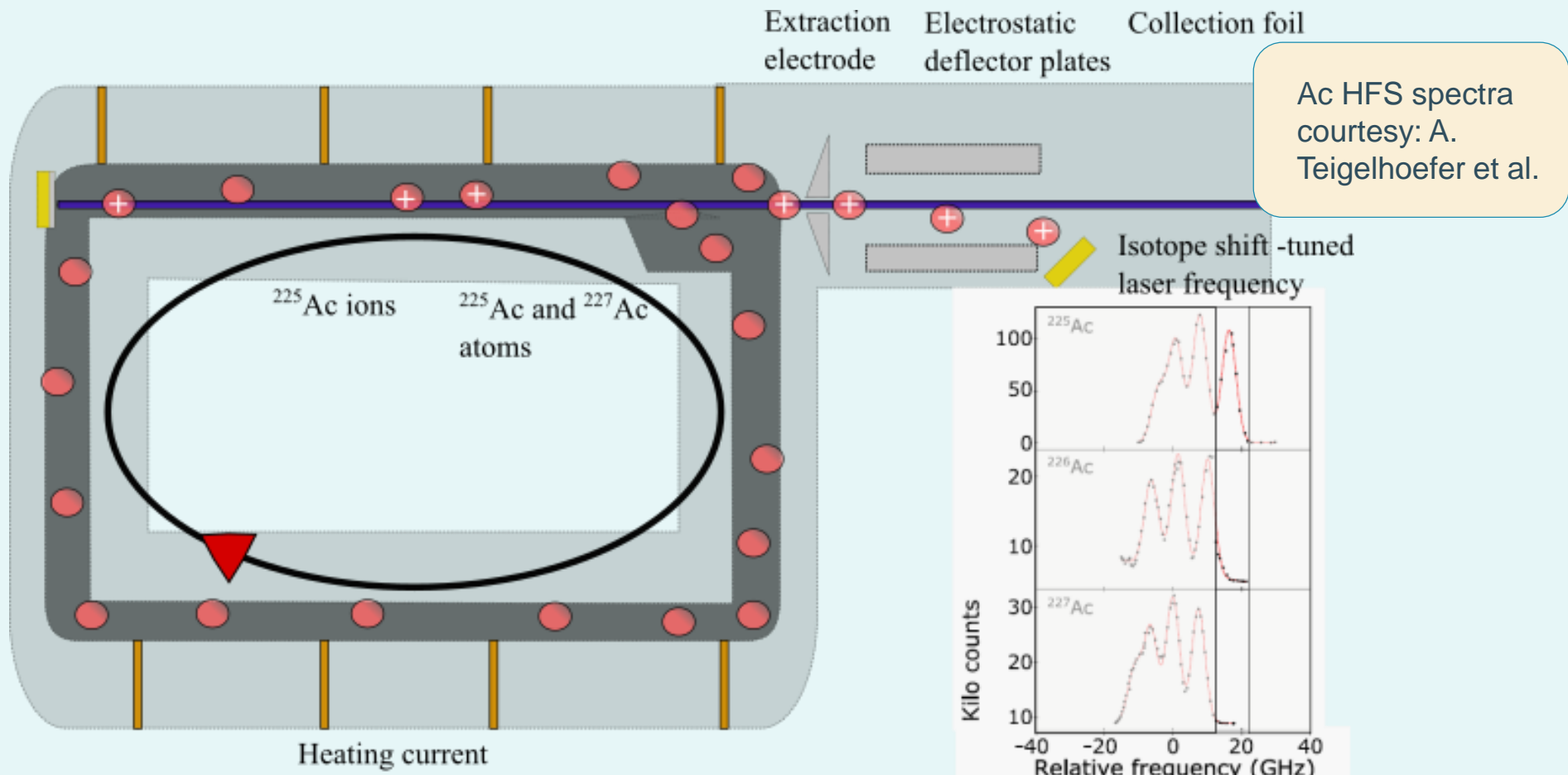
Comparison of separation methods

Single step separation and extraction chromatography [2,3]	Two step separation and extraction chromatography to produce ²²³ Ra/ ²²⁵ Ac and ²²⁵ Ac fractions [3]	Laser ionization and mass separation of nuclides that are sweated out of target
High radio-chemical yields (>85%) demonstrated	High radionuclidic purity of ²²⁵ Ac (99%) with ²²⁷ Ac activity less than 10 ⁻⁴ % of that of ²²⁵ Ac	Super-pure ²²⁵ Ac, with ²²⁷ Ac activity ~10 ⁻⁵ % that of ²²⁵ Ac. Target dissolution not required. Could obtain ²²⁵ Ra/ ²²⁵ Ac generator and pure ²²⁵ Ac from same process
May be unsuitable for routine clinical use due to ²²⁷ Ac contamination	Lower overall ²²⁵ Ac yield compared to ²²⁵ Ac produced in target (15%) due to lower ²²⁵ Ra cross section.	Process limited by ion-source efficiency to ~13%. High temperature needed. Variability and scalability not known

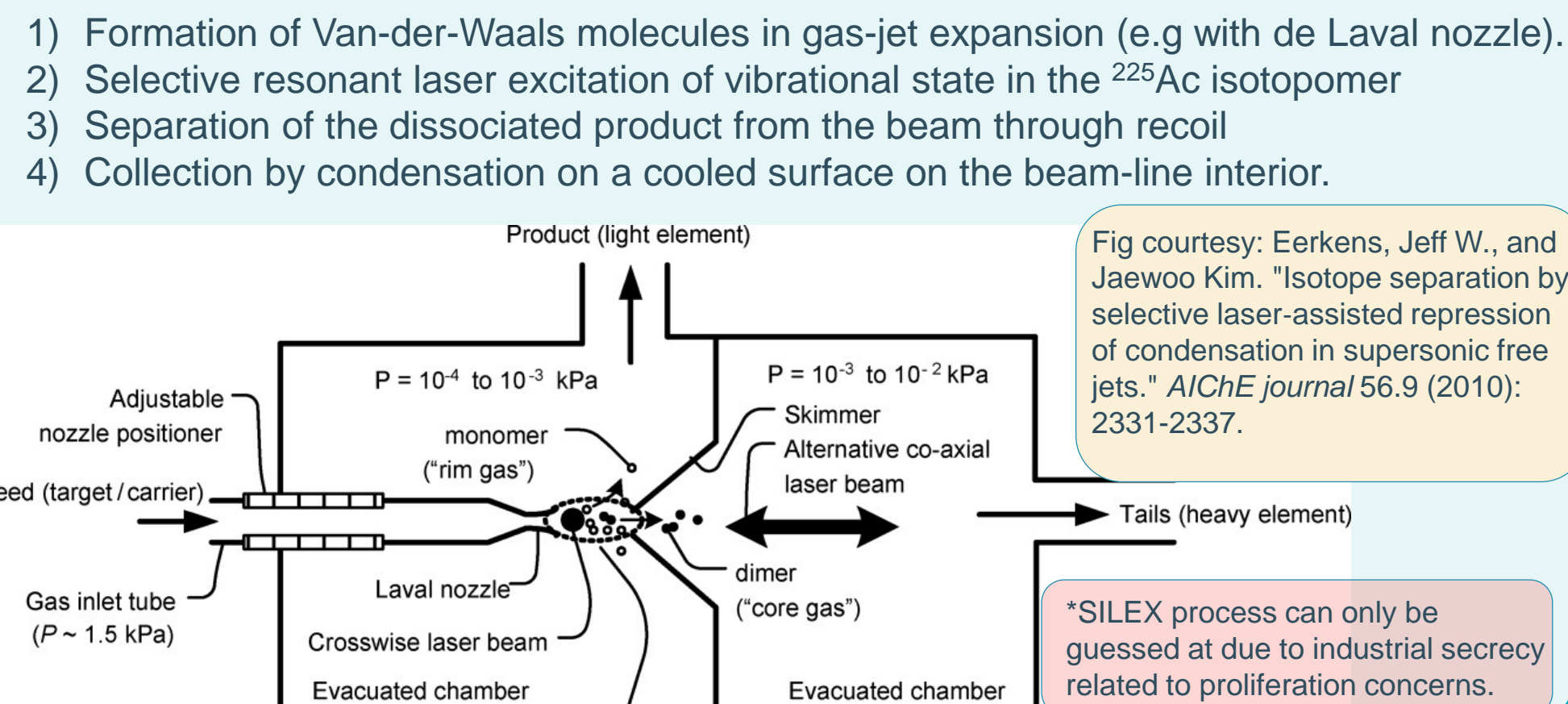
Concept brainstorm: How to make the most of irradiated actinide targets for medical grade ²²⁵Ac production?

An eye towards uranium enrichment methods but applied to medical isotope purification

Atomic Vapor Laser Isotope Separation-type concept?



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



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.
 [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 Waal's molecules." (1977).