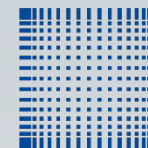




# TASCA 21

GSI, Darmstadt, June 21 - 23, 2021  
18<sup>th</sup> Workshop on  
Recoil Separator for Superheavy Element Chemistry



hochschule mannheim



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UNIVERSITÄT MAINZ

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Omtvedt<sup>5</sup>, A. Yakushev<sup>3</sup> and U. W. Scherer<sup>1</sup>

## Direct coupling of liquid-phase chemical setups for heaviest element studies to a recoil separator



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# Current state of liquid-phase experiments: Categories

- **Motivation**
- ***Design of the Vacuum to Liquid Transfer Chamber (VLTC)***
- ***SRIM simulations***
- ***Fission fragment experiments***
- ***Coupling to ion exchanging alpha detectors***
- ***Conclusion***

[1] Y. Nagame, J. V. Kratz, and M. Schädel, "Chemical studies of elements with  $Z \geq 104$  in liquid phase," *Nuclear Physics A*, vol. 944, pp. 614–639, 2015.

# State of liquid-phase chemistry experiments: Categories

Experimental setups can be categorized in:

➤ **Discontinuous** (cyclic) operating systems e.g. ARCA, AIDA or ALOHA...

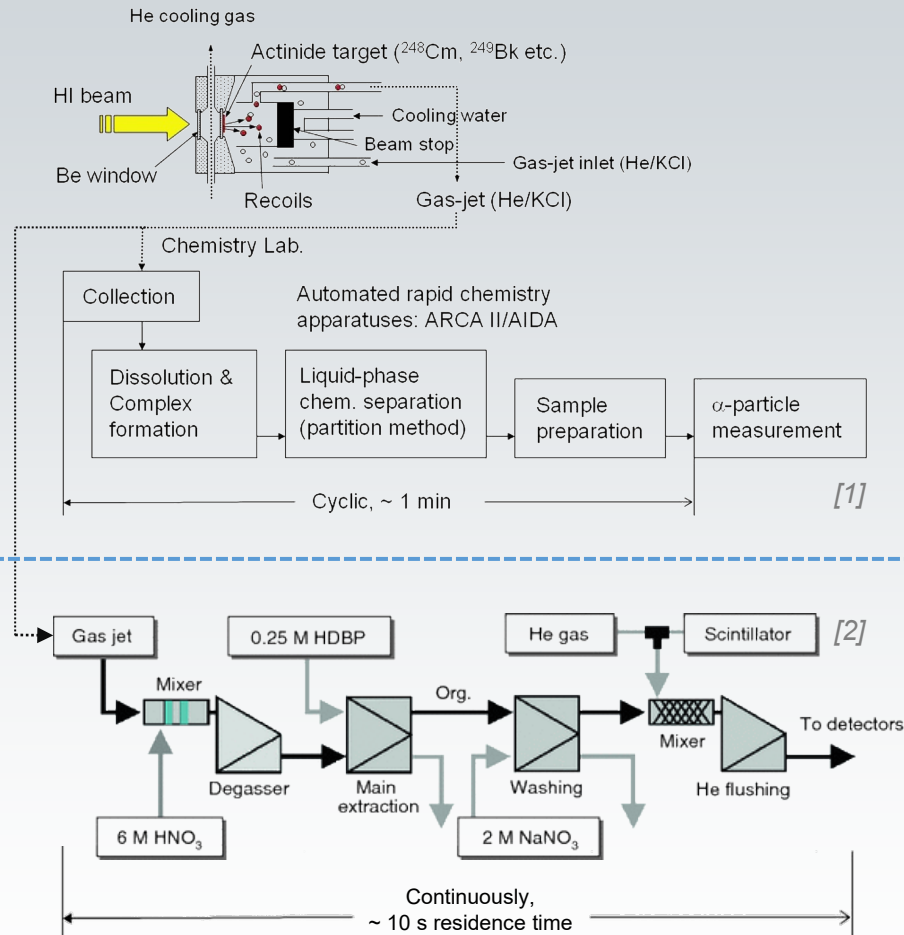
- *Chromatographic experiments*
- *$\alpha$  and SF detection with Si detectors*

➤ **Continuous** operating systems e.g. SISAK

- *Liquid-liquid extraction experiments*
- *$\alpha$ ,  $\beta$  and SF detection with liquid scintillation*

➤ With this systems, the chemical properties of the first three super heavy elements in aq. solution were studied

- *Where  $^{265}\text{Sg}$  ( $t_{1/2}$  14.4 s  $^{+3.7}_{-2.5}$  s) could only be investigated indirectly*



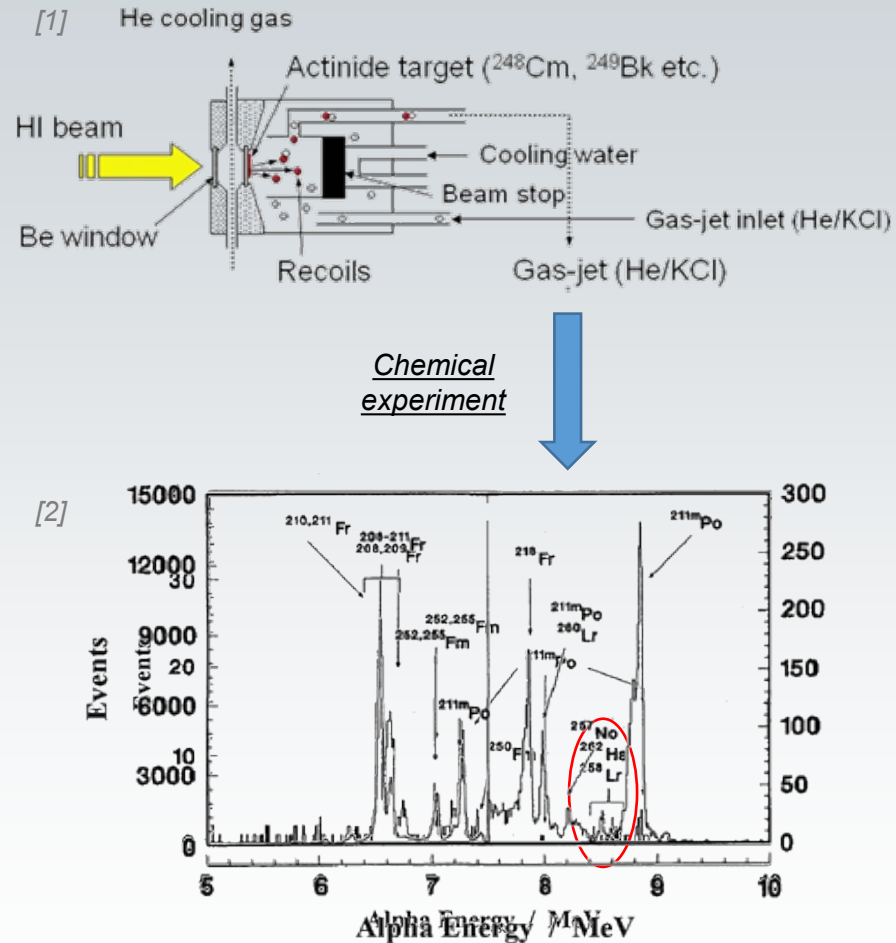
[1] Y. Nagame, J. V. Kratz, and M. Schädel, "Chemical studies of elements with  $Z \geq 104$  in liquid phase," *Nuclear Physics A*, vol. 944, pp. 614–639, 2015.

# State of liquid-phase chemistry experiments: Optimizations

Further, experiments can be categorized in:

## 1. Stopping of evaporation residues **before** separation

- *All nuclear reaction products are transported to the chemical experiment*
- *High selectivity in the chemical experiment is required in order to detect decays of SHE*
- *After successful separation of the nuclear by-products from the SHEs under investigation,  $\alpha$  decay can be clearly detected by high-resolution  $\alpha$  spectrometry with Si detectors...*



[1] Y. Nagame, J. V. Kratz, and M. Schädel, "Chemical studies of elements with  $Z \geq 104$  in liquid phase," *Nuclear Physics A*, vol. 944, pp. 614–639, 2015.



# State of liquid-phase chemistry experiments: Optimizations

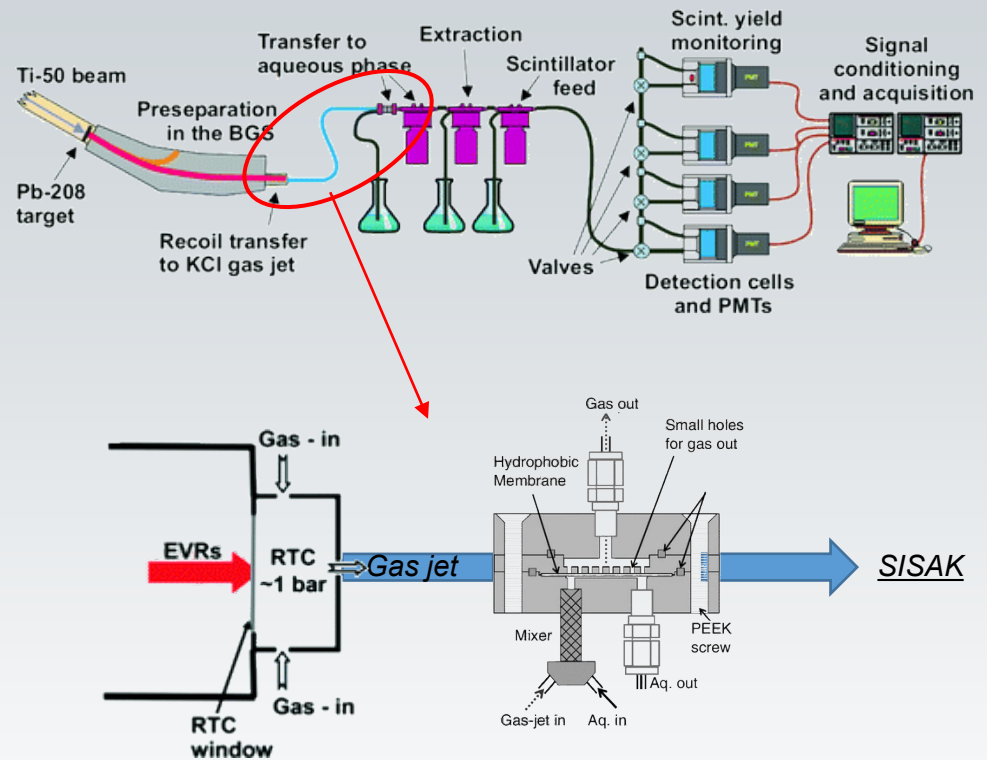
Further, experiments can be categorized in:

## 2. Stopping of evaporation residue **after** separation

- *Unwanted nuclear reaction products and the (unreacted) primary beam are filtered physically...*
- *Coupling a chemical experiment with a physical pre-separator simplifies the chemical experiments & reduces the spectral background*

➤ Pioneer experiment at the **Berkeley Gas-filled Separator (BGS)** coupled through a gas jet system with the **SISAK** system

- *Successful experiments with the  $^{257}\text{Rf}$  ( $t_{1/2} 4.4 \text{ s}^{+0.6}_{-0.5} \text{ s}$ ) were possible due to this coupling*



[1] Y. Nagame, J. V. Kratz, and M. Schädel, "Chemical studies of elements with  $Z \geq 104$  in liquid phase," *Nuclear Physics A*, vol. 944, pp. 614–639, 2015.

# The Vacuum to Liquid Transfer Chamber (VLTC)

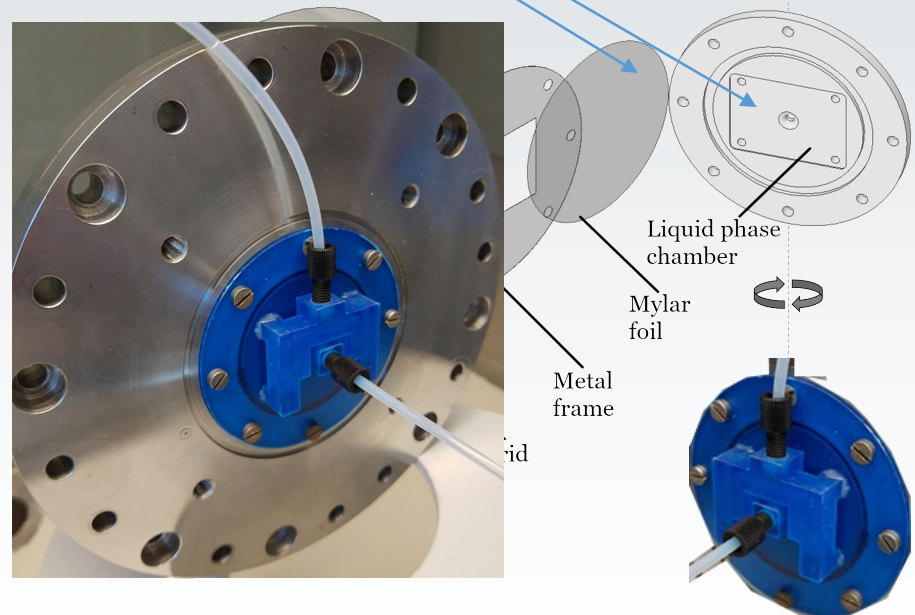
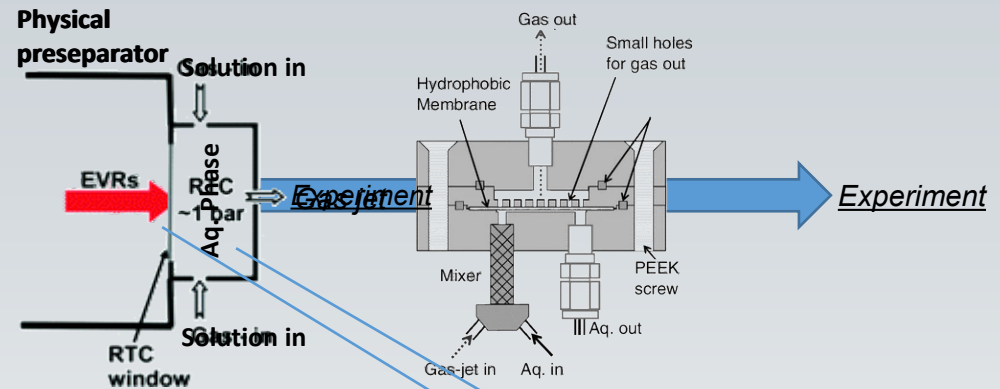
## New developments

➤ Instead of further optimizing the multi-stage process:

1. Thermalizing the EVR in gas
2. Gas jet transport of the aerosols
3. Phase transfer of the SHE to the aqueous phase
4. Subsequent chemical experiment

➤ A direct coupling between the low pressure side ("vacuum") of a physical pre-separator and the liquid phase of a chemical experiment was designed

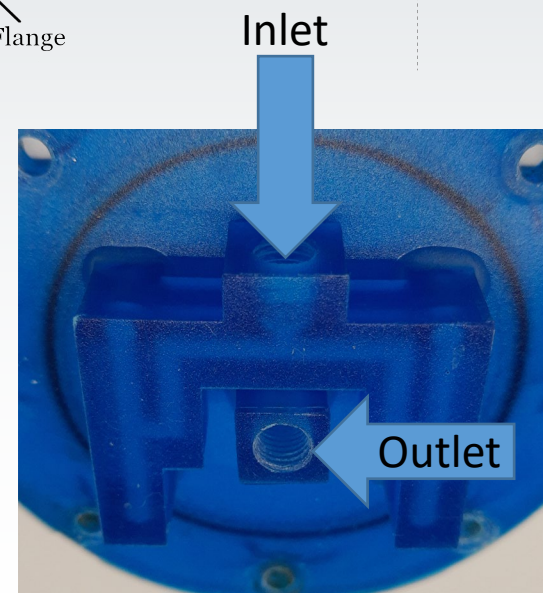
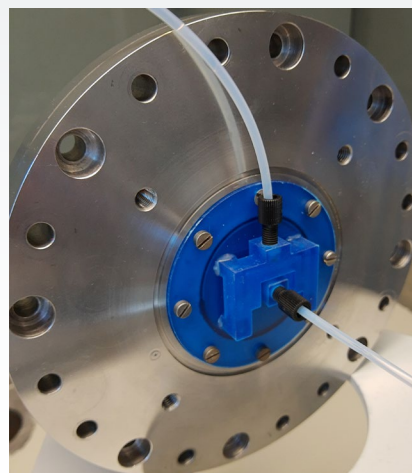
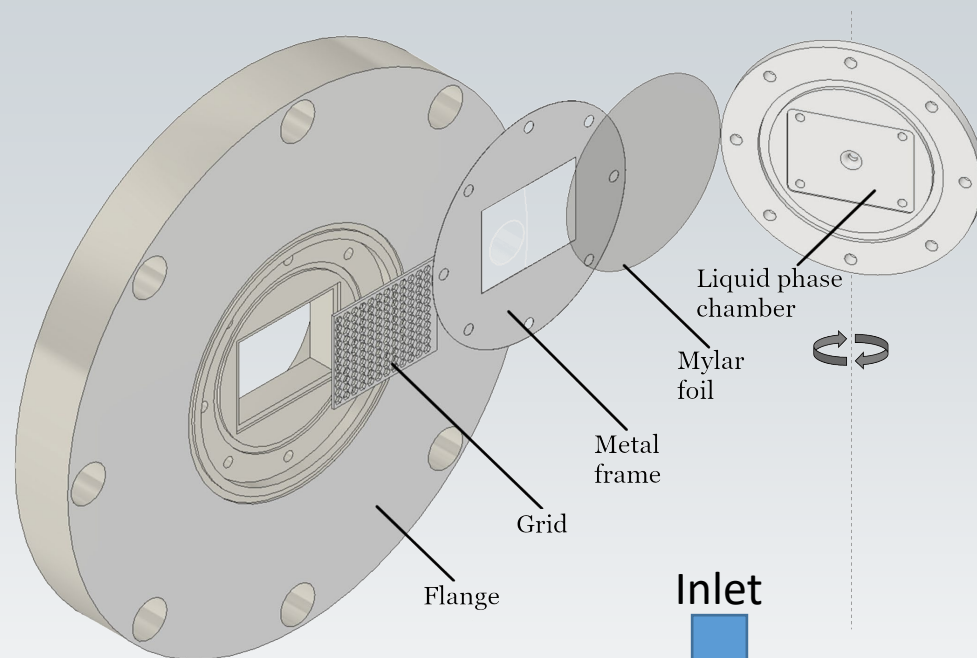
➤ Resulting in the **V**acuum to **L**iquid **T**ransfer **C**hamber (VLTC)



# The Vacuum to Liquid Transfer Chamber (VLTC)

## New developments

- Standard flange
  - Window size: 1270 mm<sup>2</sup>
- Grid
  - 80 % ion transparency
- Metal frame
  - Covered with 3.3 μm or 6.0 μm thin Mylar foil
- Liquid phase chamber
  - 3D printed (PMMA-like)
  - Standard 1/4-28 fitting connection
  - Sealing ring between Mylar Foil and Chamber
  - Chamber depth: 500 μm
  - Total volume 635.5 μl





# SRIM Simulation applicability for SHE experiments

## Proof of concept: 1. Simulations

➤ In advance, simulations were carried out with SRIM on the feasibility of a VLTC...

- ...using as example experiments with  $^{289}\text{Fl}$  at TASCA [1].

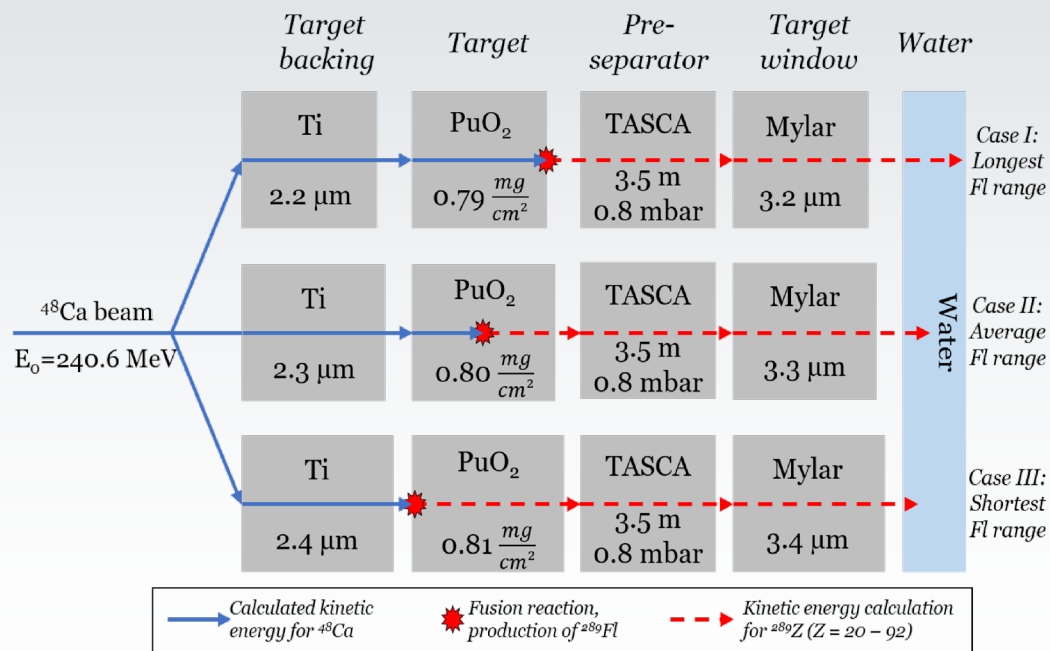
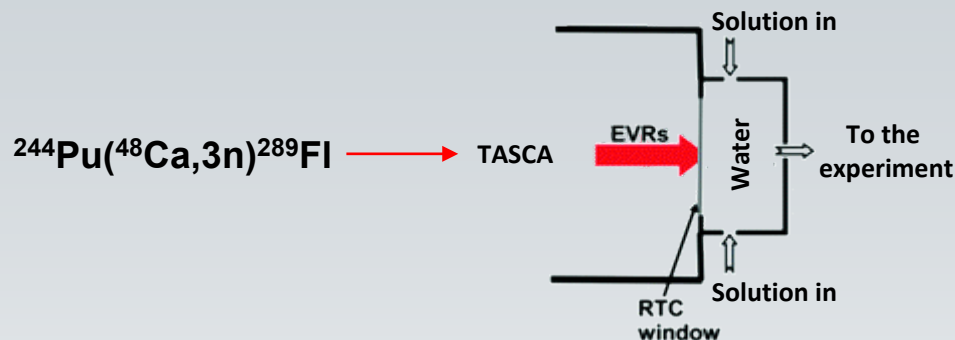
- The aim was to assess the transfer of an EVR directly after TASCA into the aqueous phase with the aid of a VLTC.

➤ Three cases were considered:

Case I: Longest Fl range

Case II: Average Fl range

Case III: Shortest Fl range

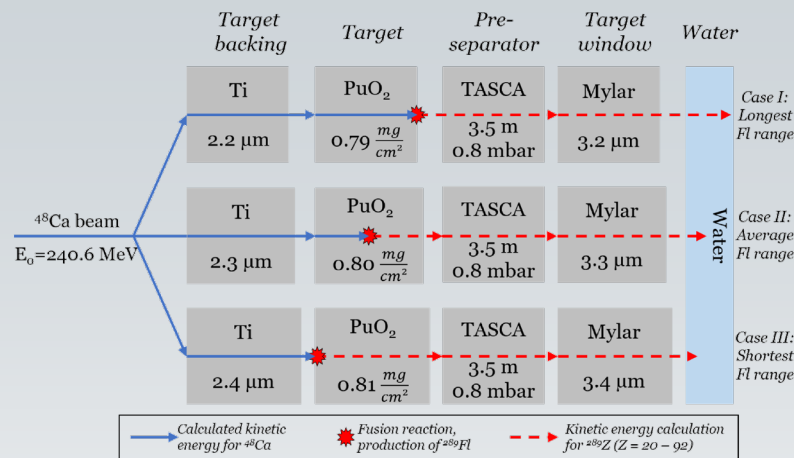


[1] A. Sâmark-Roth, et al., "Spectroscopy along flerovium decay chains: Discovery of Ds 280 and an excited state in Cn 282," *Phy. Rev. Let.*, vol. 126, no. 3, p. 032503, 2021.

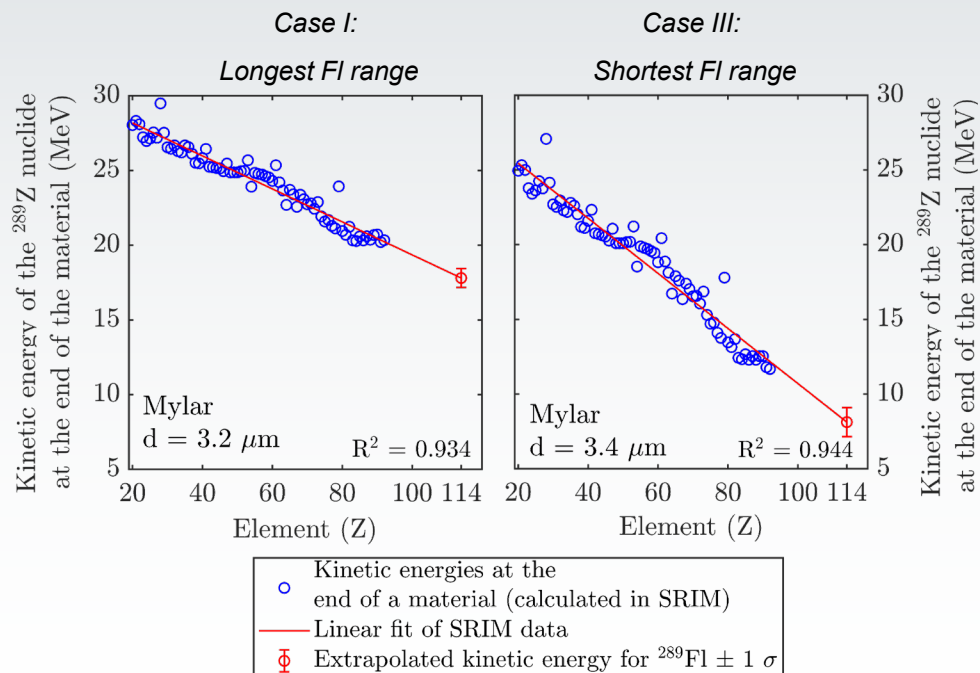
# SRIM Simulation applicability for SHE experiments

## Proof of concept: 1. Simulations

- SRIM calculates only the ranges of elements up to uranium ( $Z = 92$ )
  - Ranges were calculated for  $^{289}Z$  nuclides ( $Z = 20 - 92$ ) in each material and extrapolated to  $^{289}Fl$ .
  - If  $^{289}Fl$  still has kinetic energy when it leaves the Mylar foil, it is stopped in the liquid phase, completely.



- In a VLTC with...
  - ...a  $3.3 \mu\text{m} \pm 0.1 \mu\text{m}$  thin Mylar foil  $^{289}Fl$  enters the liquid phase in all cases
  - ...a  $6.0 \mu\text{m} \pm 0.2 \mu\text{m}$  thin Mylar foil,  $^{289}Fl$  enters the liquid phase in case I and II
- Experimental proof: literature data shows, that SHE can be transferred through  $6 \mu\text{m}$  Mylar foil



# Residence time

## Residence time studies

- Mean residence time (MRT) in the liquid phase chamber was determined with  $^{68}\text{Ga}$  in 0.1 M HCl
  - Vacuum side: 100 mbar and equipped with a shielded NaI(Tl) detector
  - The  $^{68}\text{Ga}$  tracer solution was delivered through the liquid phase chamber with a peristaltic pump at 100 ml/min and 50 ml/min respectively

- 50 mL/min

50 %     $14.7 \text{ s} \pm 1.5 \text{ s}$

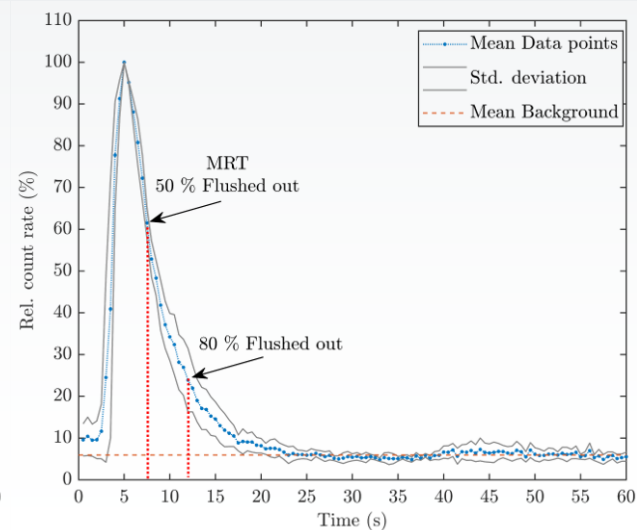
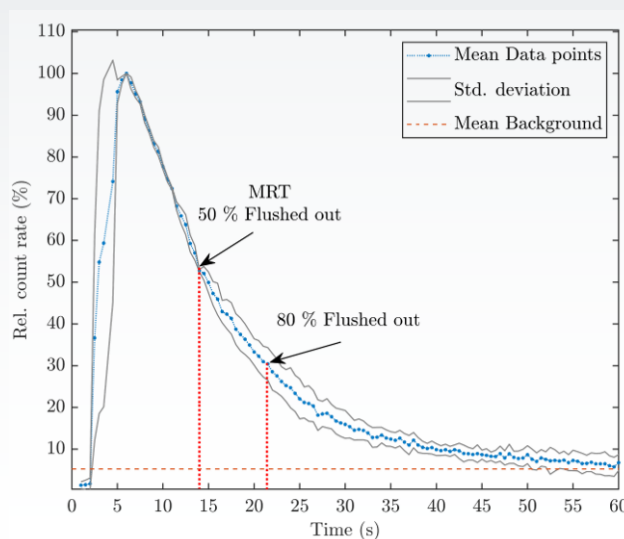
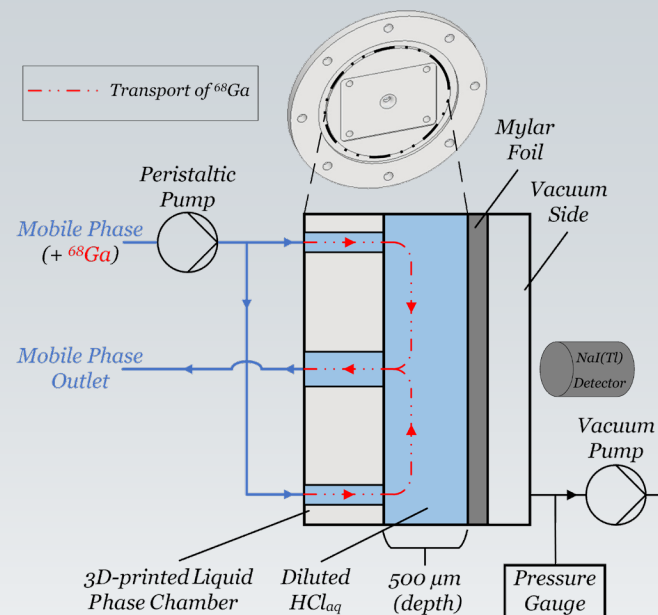
80 %     $21.5 \text{ s} \pm 3.0 \text{ s}$

- 100 mL/min

50 %     $7.7 \text{ s} \pm 1.0 \text{ s}$

80 %     $12.0 \text{ s} \pm 2.5 \text{ s}$

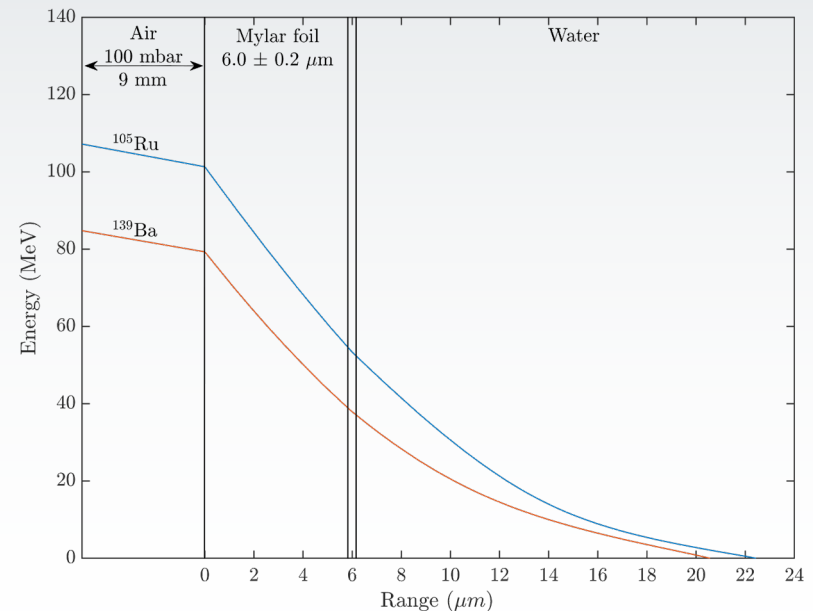
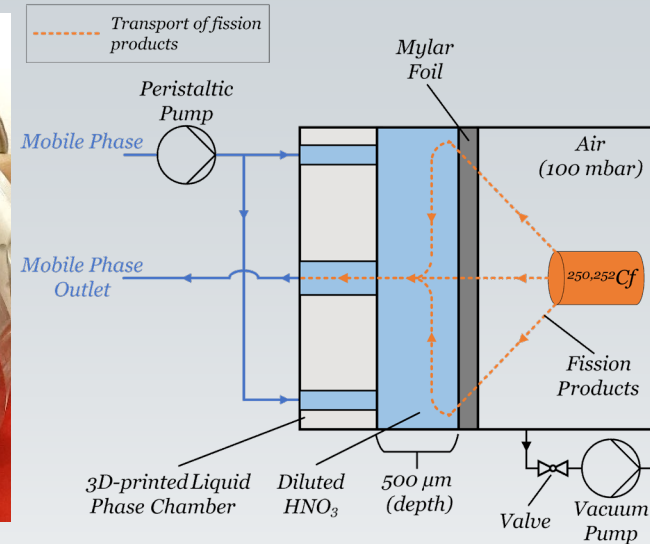
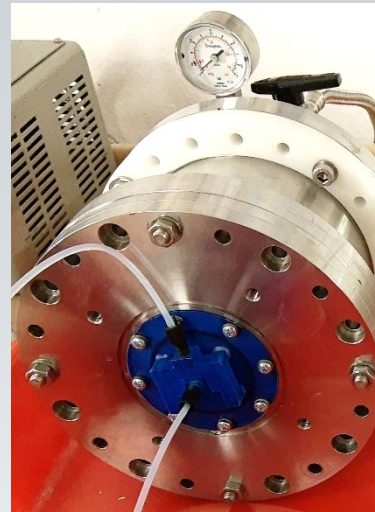
- 50 % were eluted in  $\sim 12.5 \text{ ml}$



# Proof of concept tests

## 2. Fission fragment tests

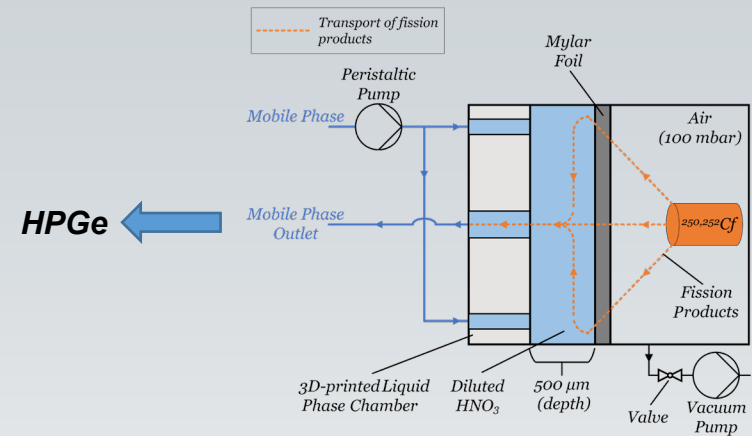
- $^{250/252}\text{Cf}$  fission source
  - Emission of light mass fragments ( $\sim 104 u$ ) with  $E_{kin} \sim 105 \text{ MeV}$
  - and heavy mass fragments ( $\sim 140 u$ ) with  $E_{kin} \sim 80 \text{ MeV}$
- $^{250/252}\text{Cf}$  source was installed on the vacuum side of the VLTC
  - The setup was also simulated in SRIM
  - Required chamber depth:  $23 \mu\text{m}$
  - Total depth (liquid phase chamber):  $500 \mu\text{m}$



# Proof of concept tests

## 2. Fission fragment tests

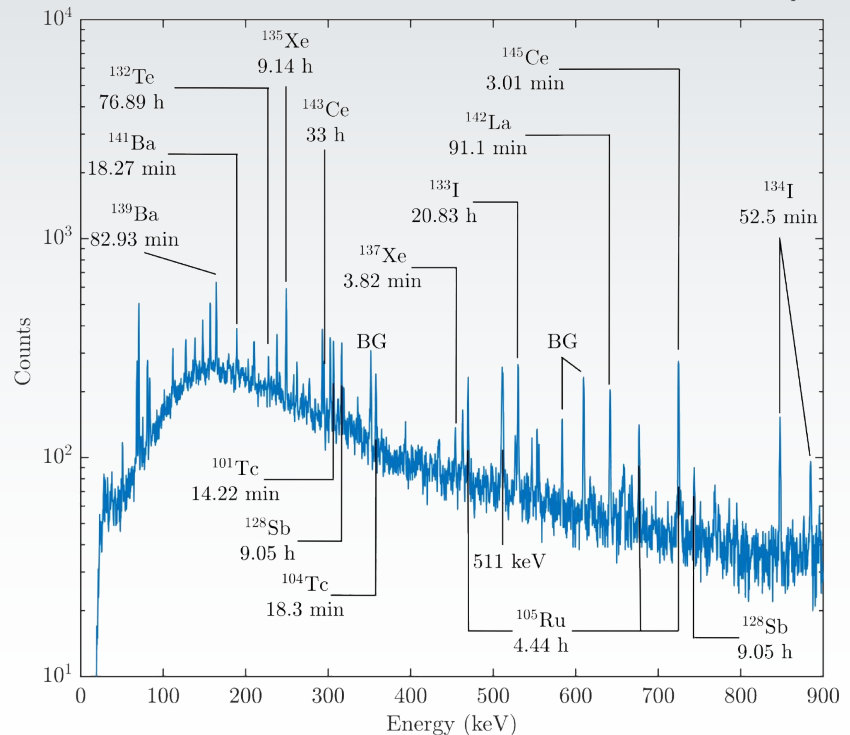
- $^{250/252}\text{Cf}$  fission fragments were collected in the liquid phase chamber over time
- Subsequent measurement of 10 ml samples (HPGe)



### $\gamma$ -spectrometry results for 10 ml samples

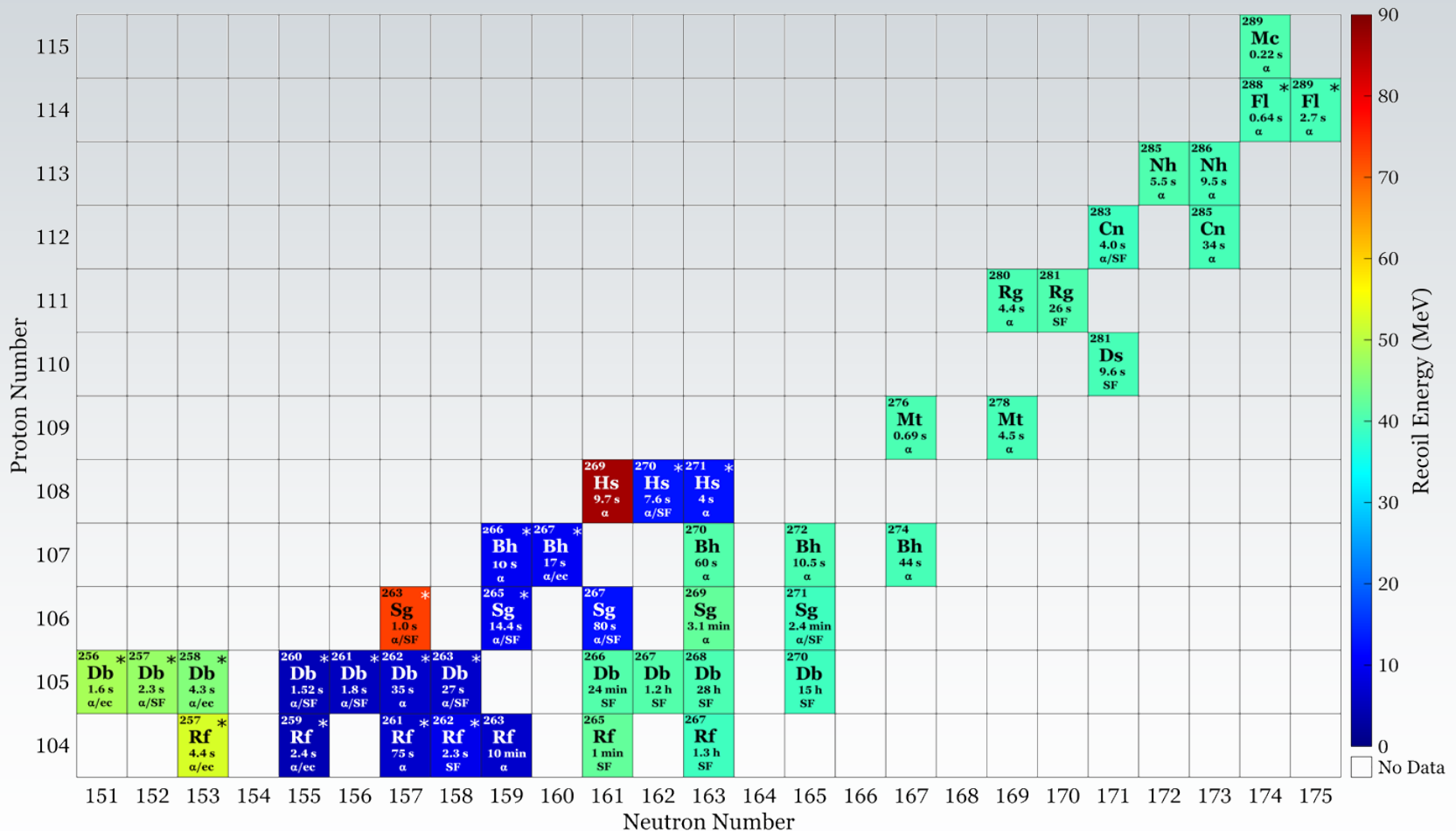
Fission Product	Collected yield [%]
$^{97}\text{Zr}$	$41 \pm 12$
$^{105}\text{Ru}$	$51 \pm 16$
$^{132}\text{Te}$	$34 \pm 9$
$^{139}\text{Ba}$	$35 \pm 9$
$^{143}\text{Ce}$	$40 \pm 7$

- Fission fragments of light and heavy mass peaks were observed




# Applicability of the VLTC for SHEs

- Which SHEs can be investigated with the VLTC system? [1]




[1] M. Schädel and D. Shaughnessy, Eds., *The Chemistry of Superheavy Elements*, 2nd ed. Springer, 2014.

# VLTC Paper



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Nuclear Instruments and Methods in Physics  
Research Section A: Accelerators, Spectrometers,  
Detectors and Associated Equipment



Available online 18 June 2021, 165486  
In Press, Journal Pre-proof

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## Speeding up liquid-phase heavy element chemistry: Development of a vacuum to liquid transfer chamber (VLTC)

Dominik Krupp<sup>a</sup>, Christoph E. Düllmann<sup>b, c, d</sup>, Lotte Lens<sup>a</sup>, Jon Petter Omtvedt<sup>e</sup>, Alexander Yakushev<sup>c</sup>, Ulrich W. Scherer<sup>a</sup>

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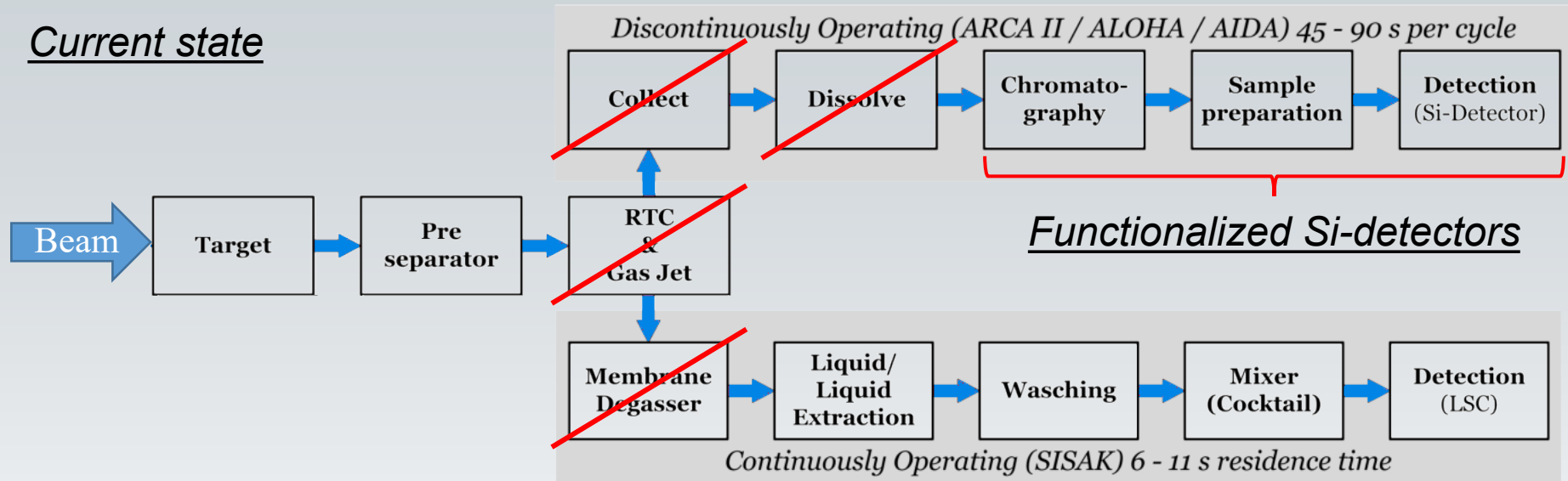
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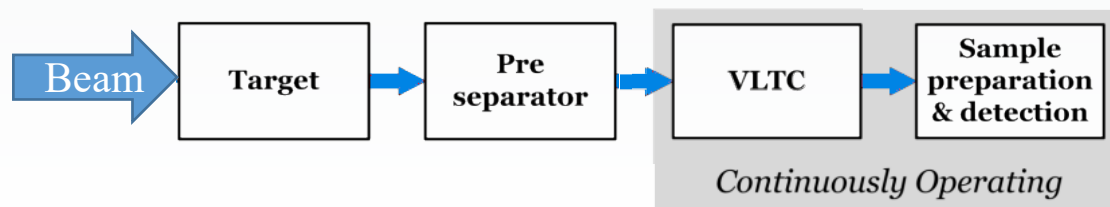
# Direct coupling of liquid-phase chemical setups to a recoil separator

## Current state



## New developments

- Direct coupling of the vacuum side of a physical preseparator with the liquid phase of a chemical experiment → development of a **Vacuum to Liquid Transfer Chamber (VLTC)**
- Followed by continuous operating chemical experiments, e.g. equipped with functionalized Si-detectors [1] TASCA 2019?

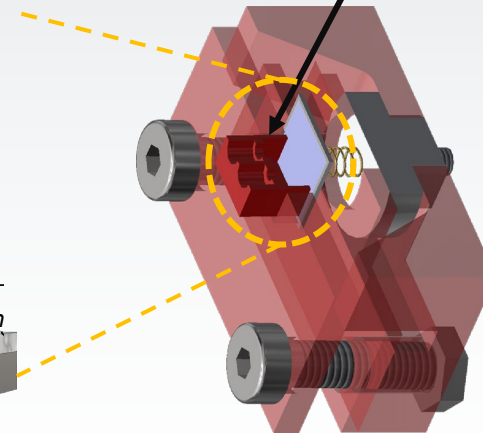
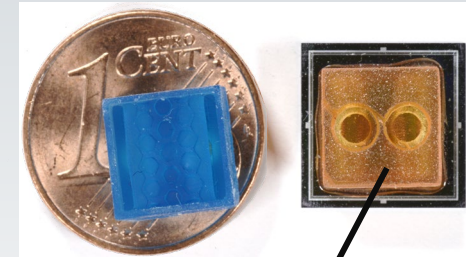
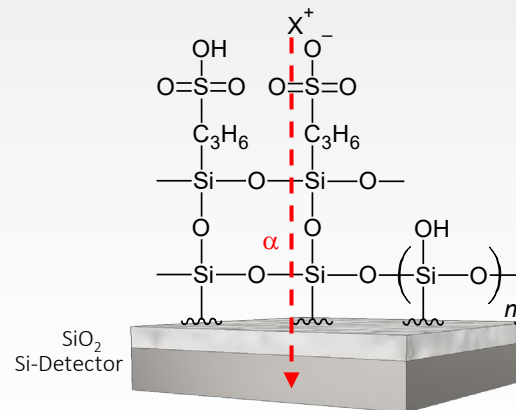




# Functionalized Si-detectors

## New ways of a faster $\alpha$ spectrometry

- $\mu$ l-flow cells equipped with Si-detectors
- Si-detectors were chemically modified (functionalized)
  - Depending on the functional groups, e.g. cation exchanger ( $R-SO_3H$ )
  - Accumulation of radionuclides on the detector surface
  - Combined chemical separation and detection
- Coupling the functionalized a detectors with the VLTC for continuous fast liquid phase chemistry experiments



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Prototype development of ion exchanging alpha detectors

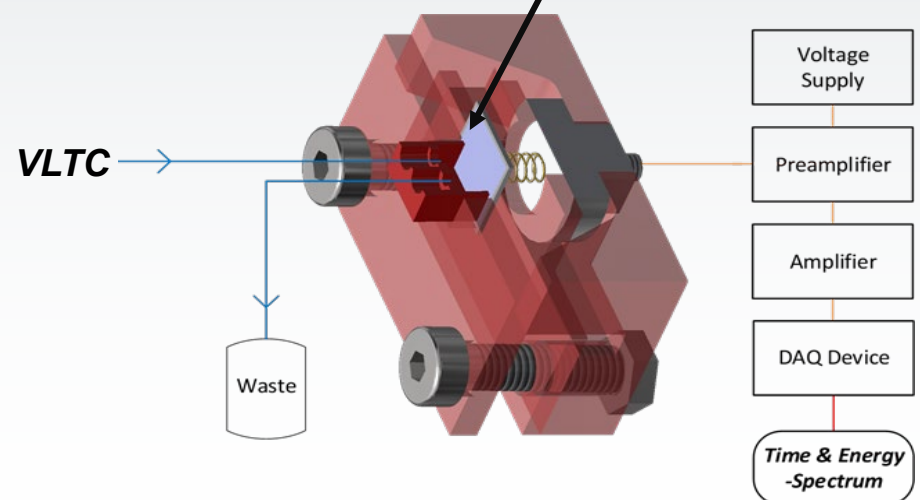
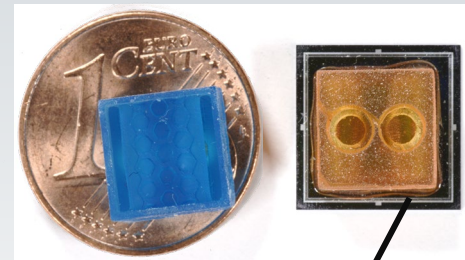
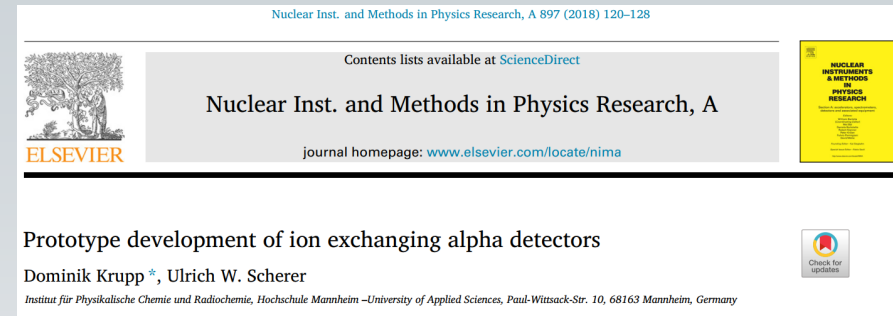
Dominik Krupp\*, Ulrich W. Scherer

Institut für Physikalische Chemie und Radiochemie, Hochschule Mannheim –University of Applied Sciences, Paul-Wittsack-Str. 10, 68163 Mannheim, Germany

# Functionalized Si-detectors

## New ways of a faster $\alpha$ spectrometry

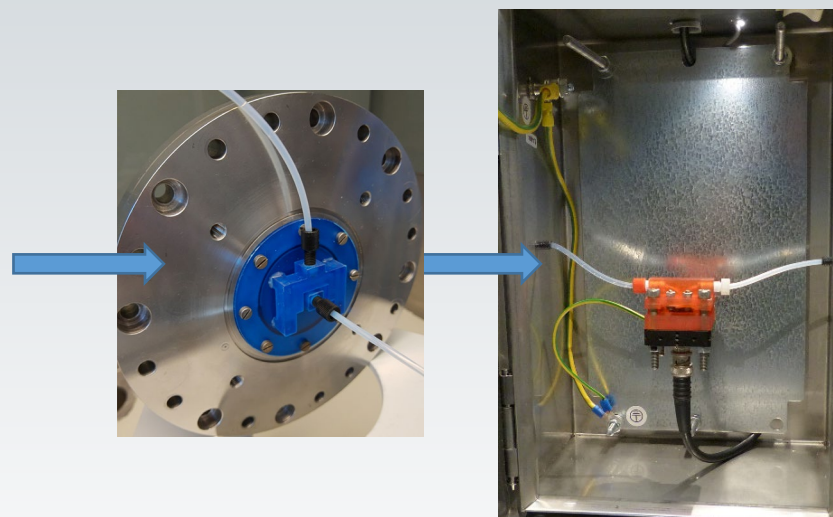
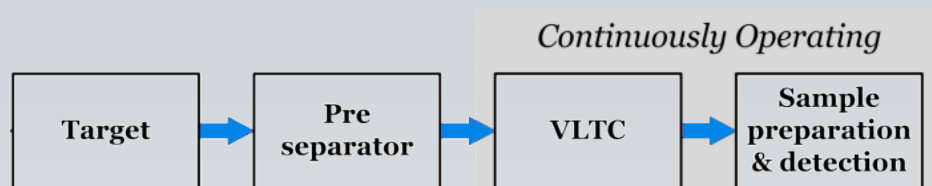
- $\mu$ l-flow cells equipped with Si-detectors
- Si-detectors were chemically modified (functionalized)
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  - Accumulation of radionuclides on the detector surface
  - Combined chemical separation and detection
- Coupling the functionalized a detectors with the VLTC for continuous fast liquid phase chemistry experiments



# Conclusion

## New ways of a faster $\alpha$ spectrometry

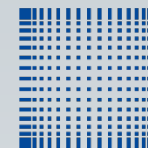
- Conventional liquid phase SHE chemistry experiments consist of several preparation steps, each with its time and yield budget
- With the **V**acuum to **L**iquid **T**ransfer **C**hamber (VLTC) several steps are bypassed
  - *Resulting in a faster and more efficient transport of SHE from a physical preseparator into the liquid phase*
- By coupling the VLTC with flow cells, equipped with functionalized  $\alpha$  detectors continuous experimental runs with Si-detectors can be realized
- This opens the perspective for trans-seaborgium chemistry in the aqueous phase in the future





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Thank you for  
your attention!