

Liquid-Phase Chemistry of SHE Homologues at the CTU in Prague

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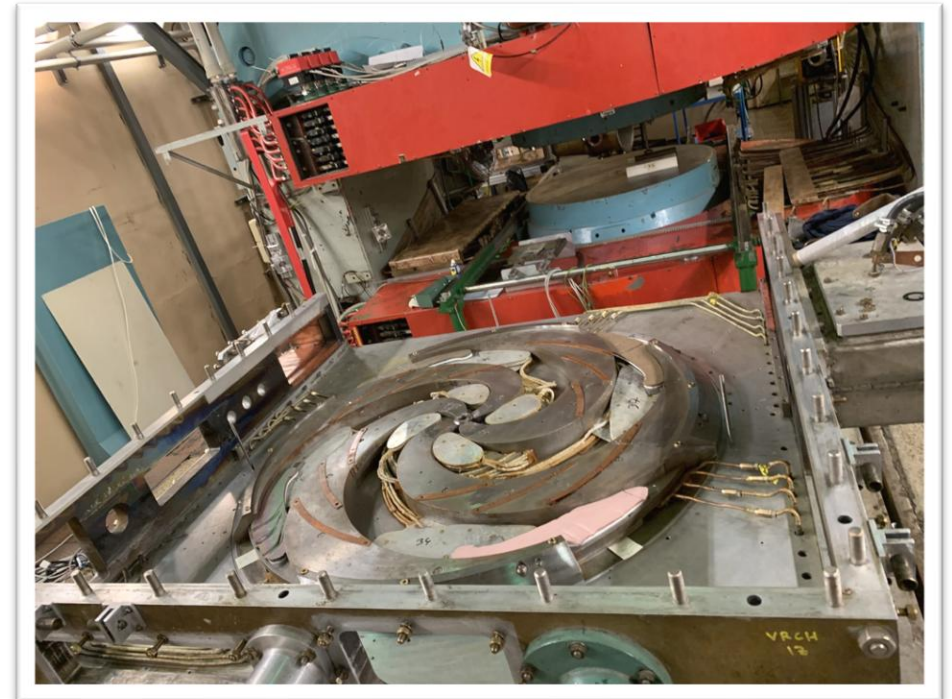
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@GSI, 24.04.2025



Outline

- The Laboratory of Homologues of SHEs in Rez
- Ongoing research work in the ASHES group
 - On-line chemistry apparatus
 - Fast liquid-liquid extraction systems
 - Electrochemistry
 - On-line detection
- Outcome & Future research

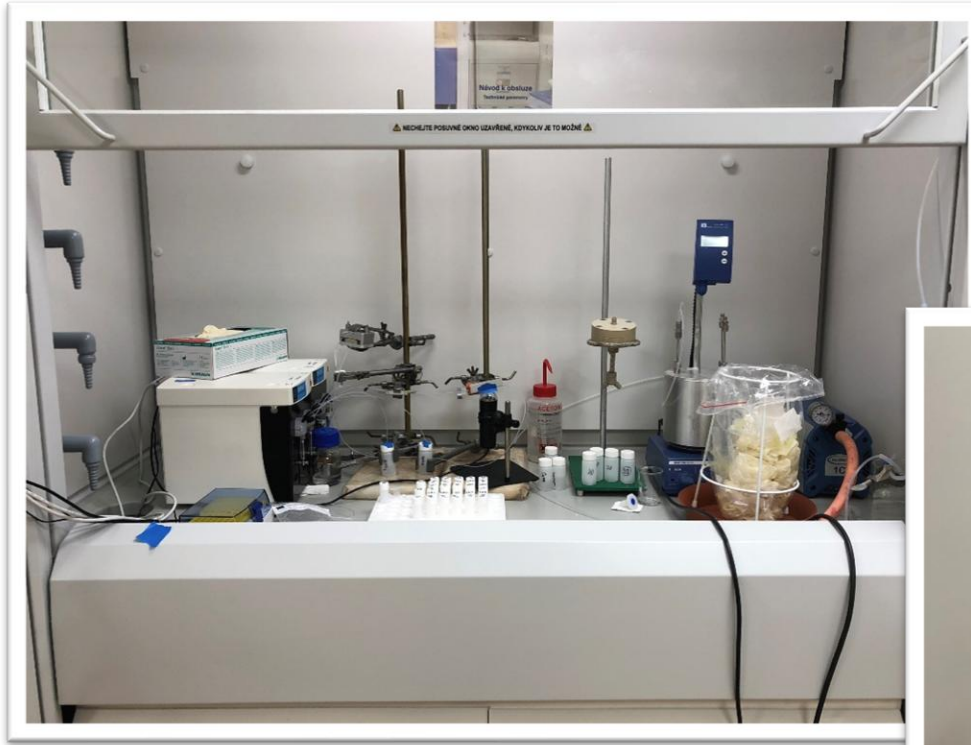


Development of the facility

- Joint **CTU-NPI-UiO laboratory** was set up at the U-120M facility in **2018**
 - UiO and CTU provided experimental and laboratory equipment, furniture, etc.
 - NPI provided dedicated premises in the proximity of the U-120M cyclotron
- New **MARGE target system** commissioned in **2021**
- Research part of the **Centre of Advanced Applied Sciences** project from 2018 to 2023 and the **FAIR-CZ** infrastructure since 2023



Development of the facility



ASHES group until 2025

The MARGE system

Production and transport of
radionuclides in gas phase

10.1016/j.nima.2023.168280



The PILS system

Gas-into-liquid transfer

10.1016/j.nima.2023.168500



The μLLX system

Fast aqueous chemistry processes
(solvent extraction, electrochemistry)

3 Master theses
1 Bachelor thesis



An unknown detection system

On-line detection

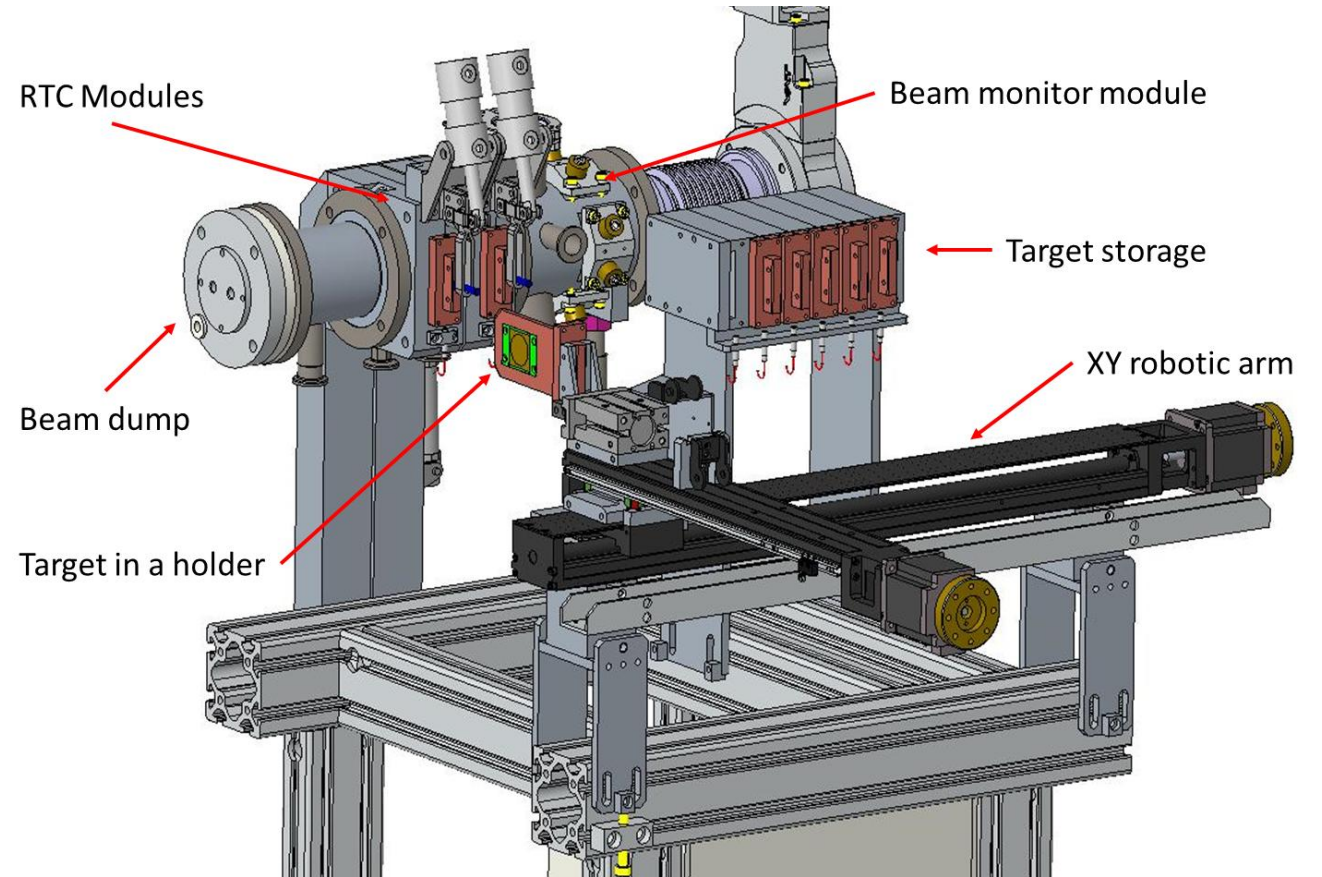
2 Masters theses

1 Doctoral thesis

The MARGE system

MARGE system = **ModulAr Robotic Gas-jet targEt** system

- Remote target switching
 - Radiation protection
 - Beam time management
- Better ion beam monitoring
 - Both shape (focus) and position
- Modular RTCs
 - **Independent experiments -> more efficient beam time**

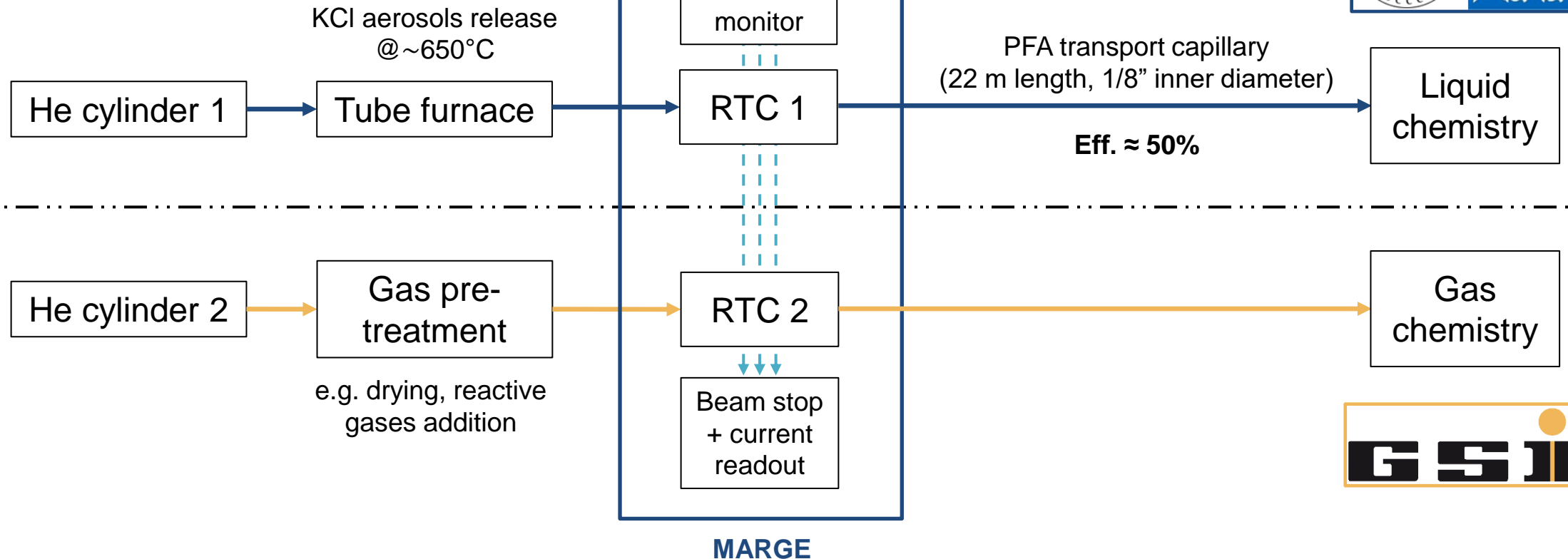


MARGE GJT systems



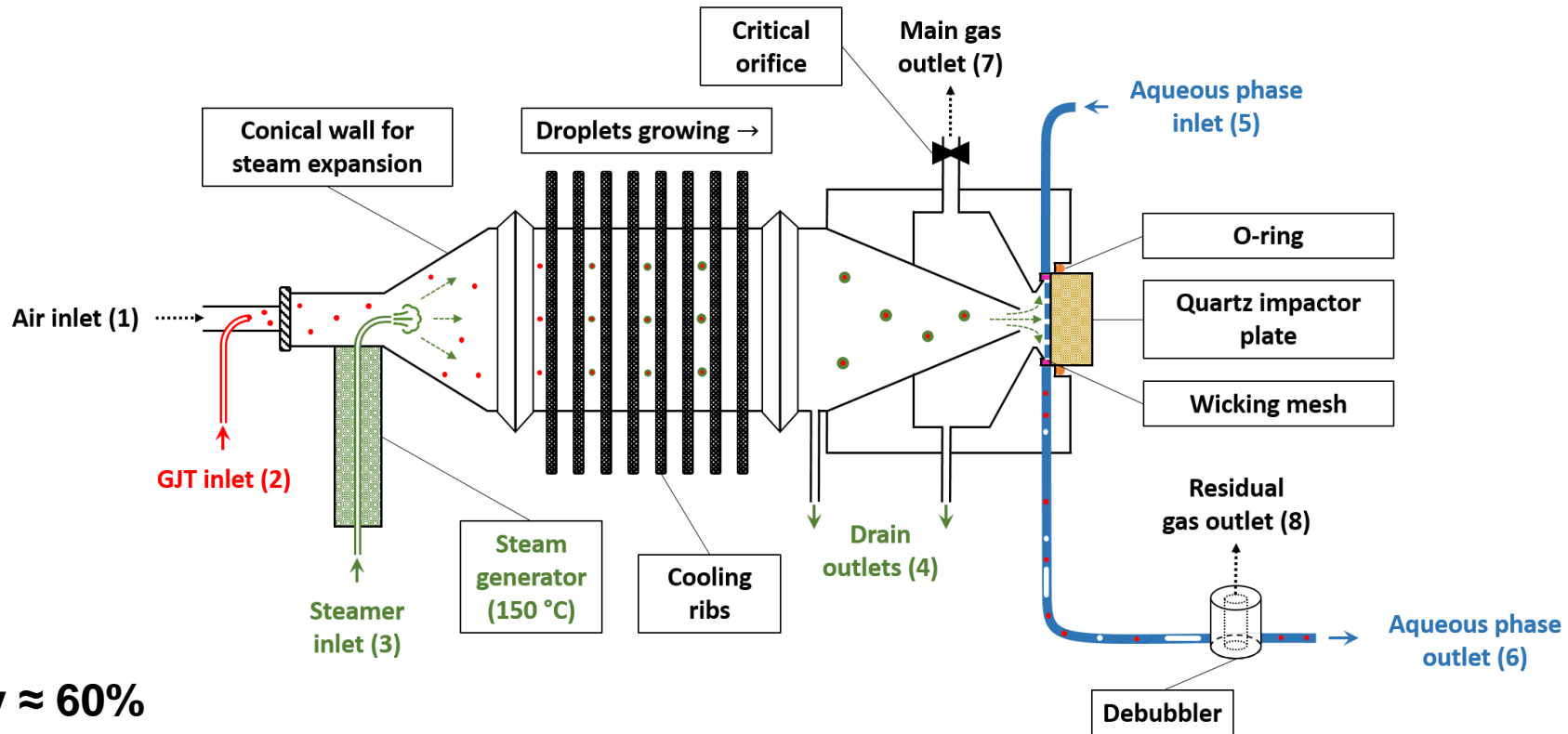
Cyclotron
U-120M

^3He beam
~50 MeV
~300 nA



The PILS system

PILS = **P**article **I**nto **L**iquid **S**ampler



Efficiency \approx 60%

D. Orsini *et al.* Atmos Environ, 37, 1243-1259 (2003)

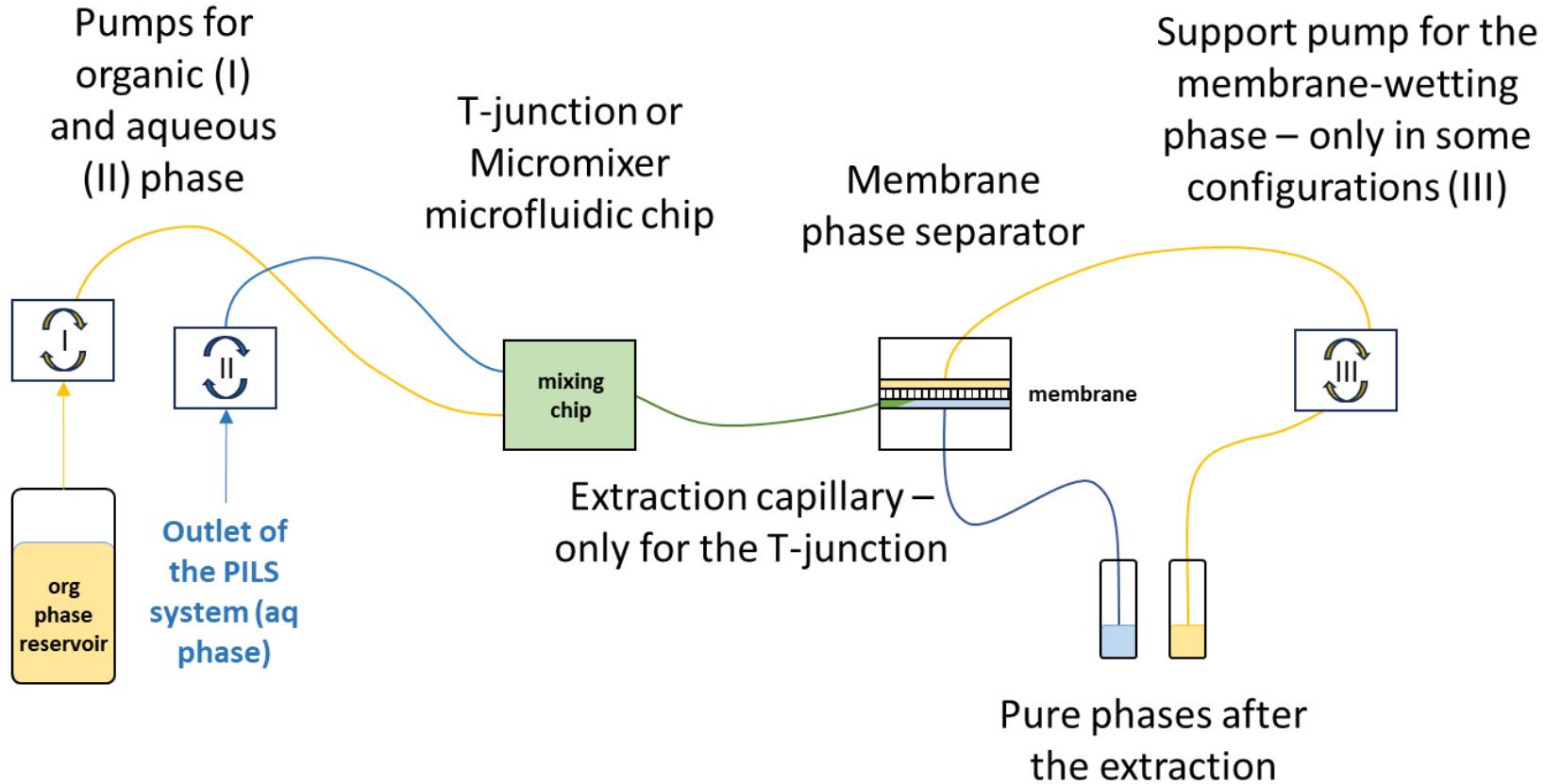
P. Bartl *et al.* Nucl Instrum Methods Phys Res A, 1055, 168500 (2023)

DOI: 10.1016/j.nima.2023.168500



The μ LLX system

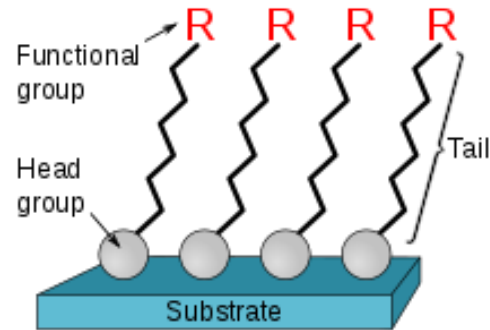
μ LLX = **MICRO**fluidic **Liquid-Liquid** **eXtraction**



Currently undergoing optimization and upgrade (pumps, phase separator, pressure...)

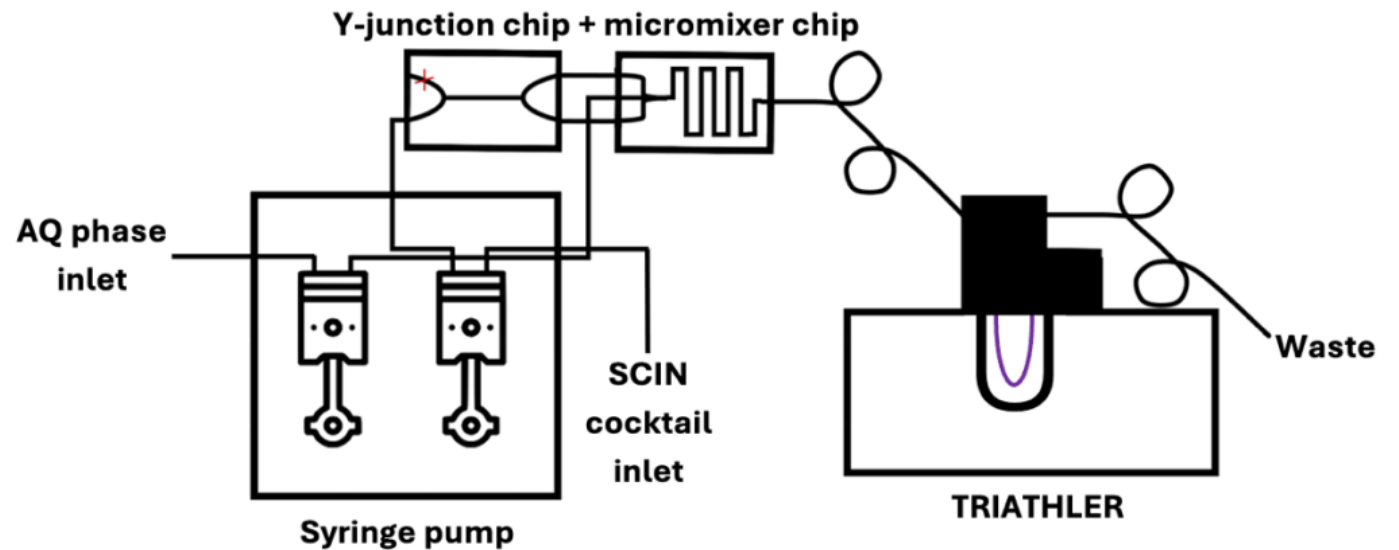


On-line detection



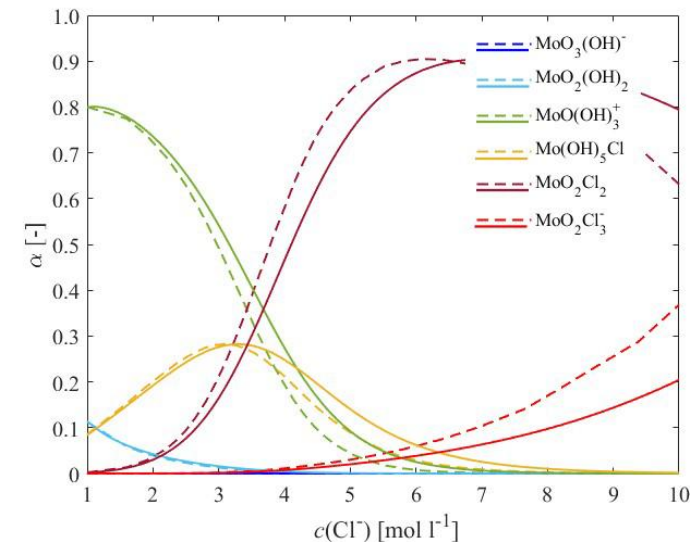
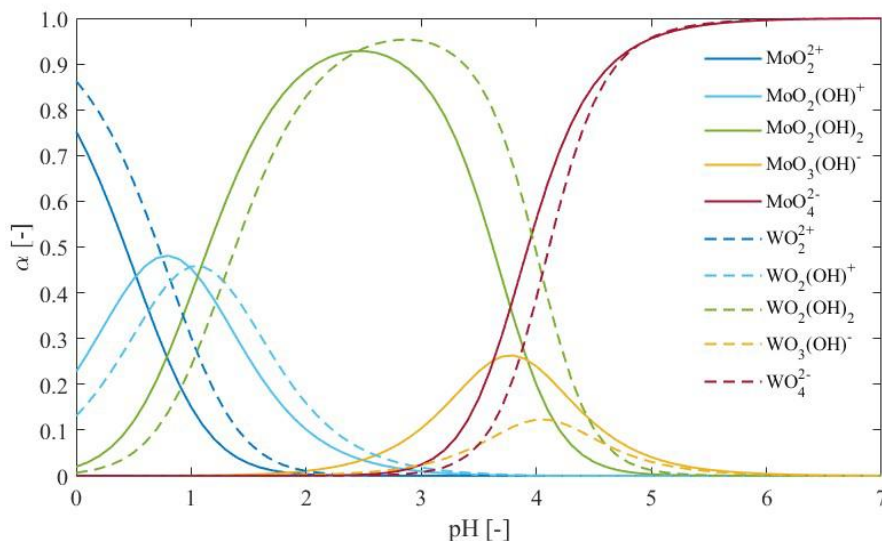
- On-line detection of alpha and beta in a capillary – liquid scintillation counting directly following the μ LLX system

- The use of selfassembled monolayers (SAMs) to functionalize surfaces of alpha detectors is currently under investigation

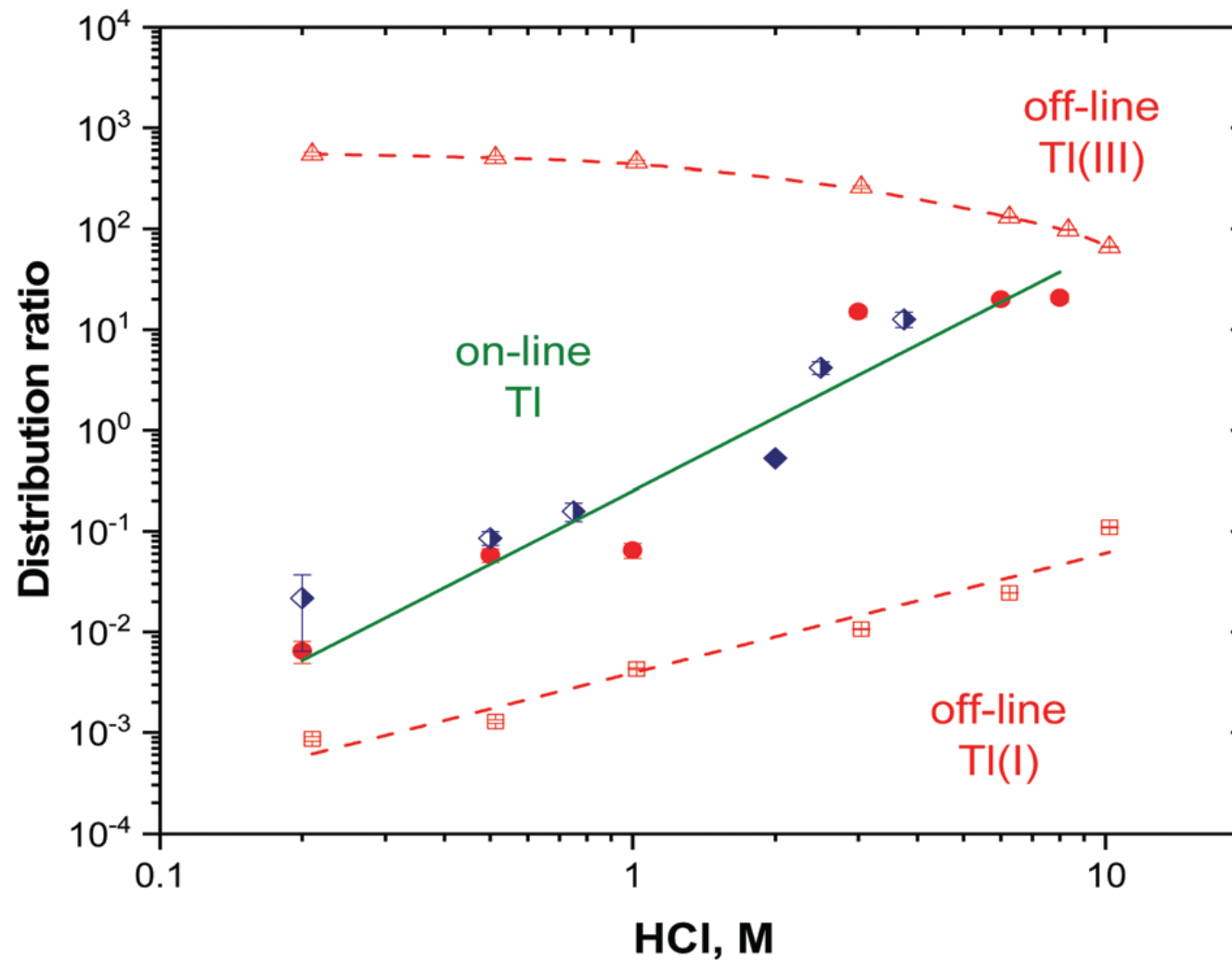


Extraction of Sg and Db homologues

- **Speciation is crucial** -> equilibrium state
- Assessment of systems' suitability for SHE research
- Characterization in equilibrium to precisely determine extraction mechanism
- Kinetics studied via fast extraction in microfluidic LLX system
- **Systems for Mo/W evince fast kinetic, dependent on the mechanism**
- Research on Nb/Ta has started only recently



(In)stability of Tl oxidation states

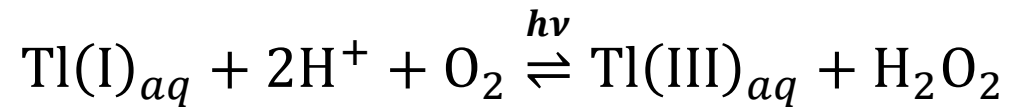


K. Čubová *et al.* J Radioanal Nucl Chem, 318, 2455-2461 (2018)
E. Tereshatov *et al.* New J Chem, 45, 3377-3381 (2021)

(In)stability of Tl oxidation states

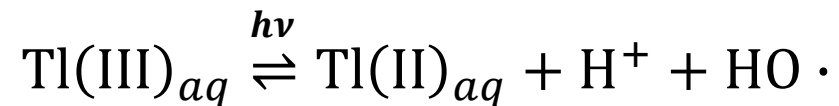
Turns out thallium likes fresh air (O₂) and sunshine (light)

Tl(I) can be oxidized!

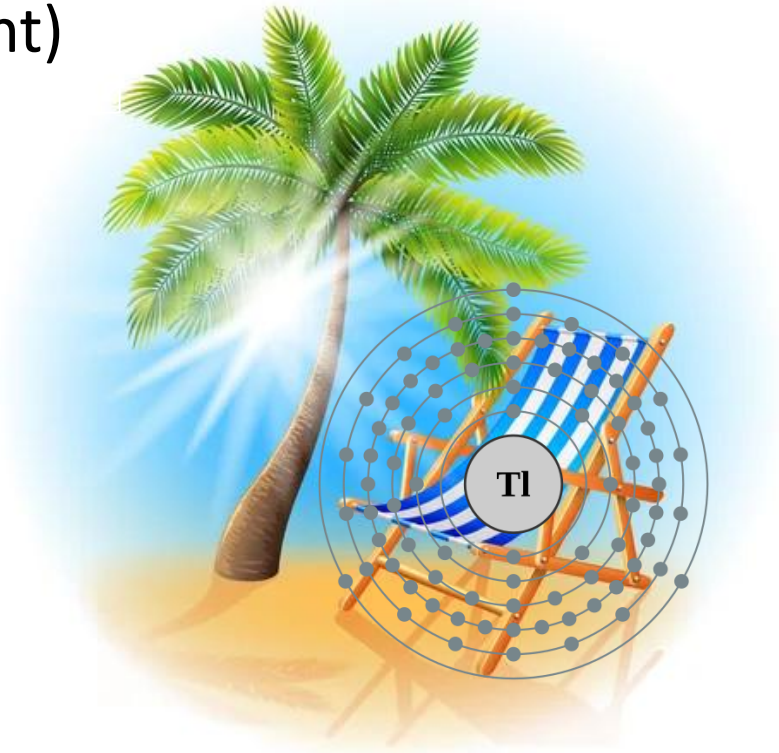


... in acidic solutions containing Cl⁻ ions

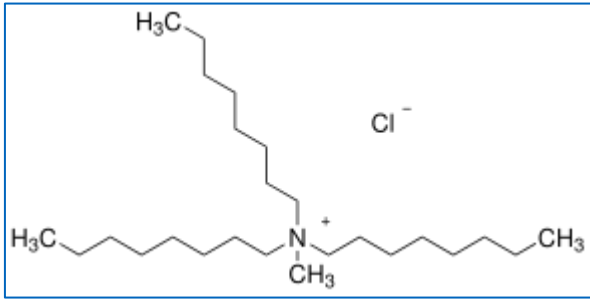
But also, Tl(III) can be reduced!



... in acidic solutions, followed by immediate reduction of unstable Tl(II)



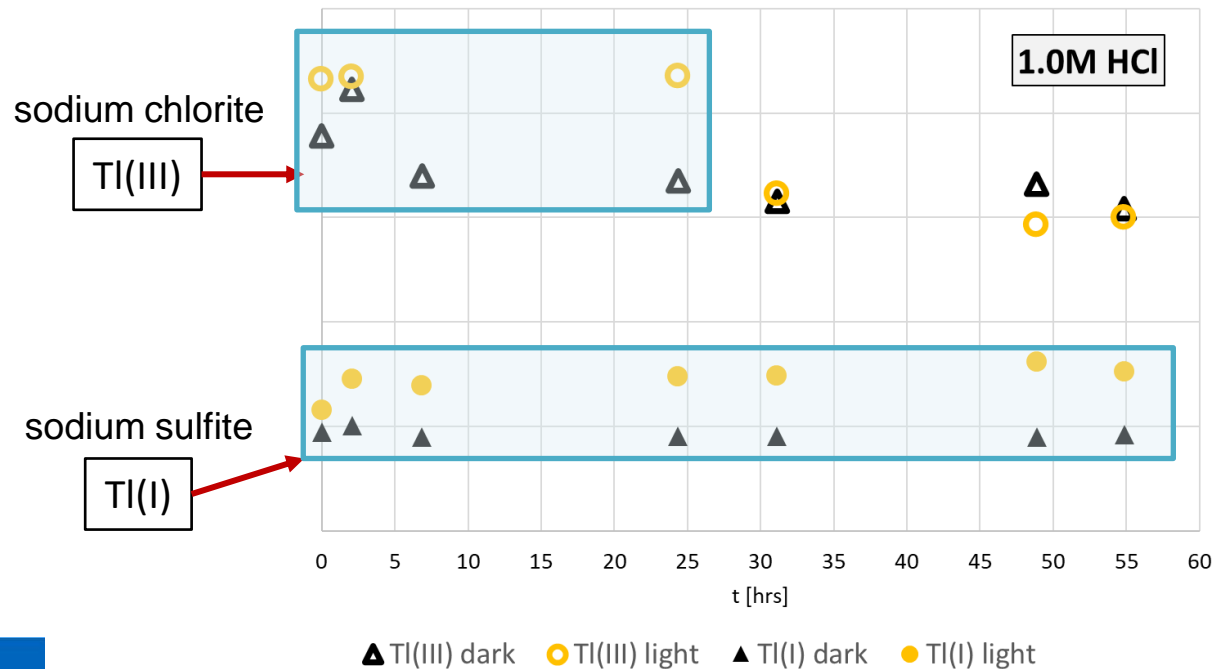
Extraction of Tl by Aliquat 336



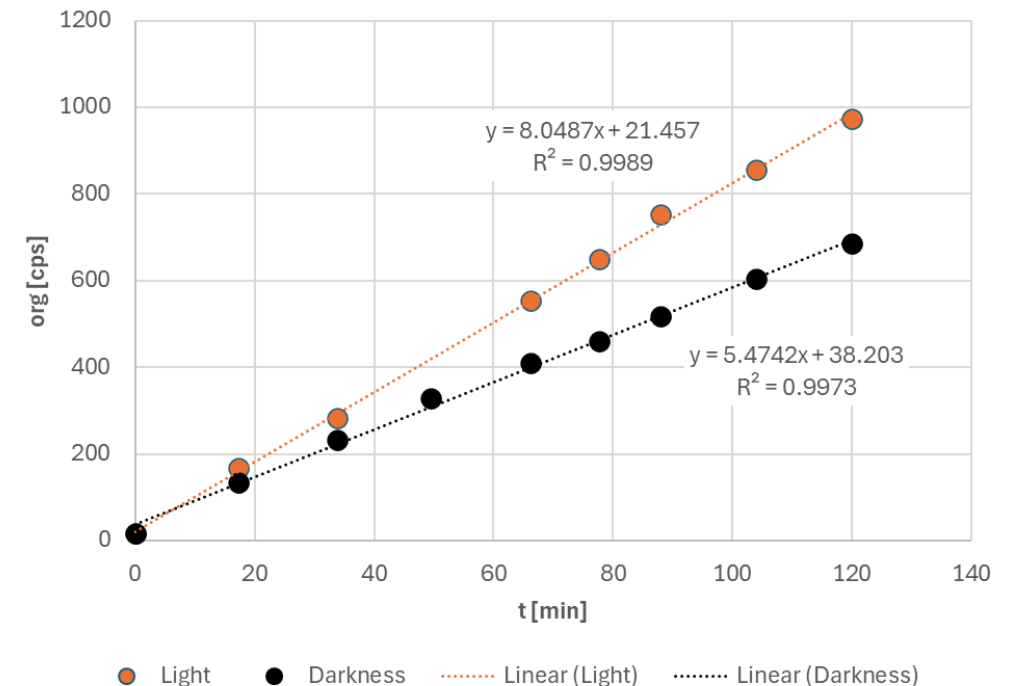
$c(\text{A336}) = 0.2 \text{ mol L}^{-1}$
(in kerosene + 1% octanol)

- Parallel experiments – with and without light
- Beam-induced oxidation not likely
- The instability demonstrated

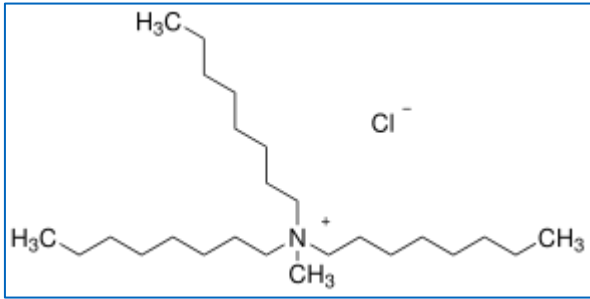
ALL PRELIMINARY



NEW



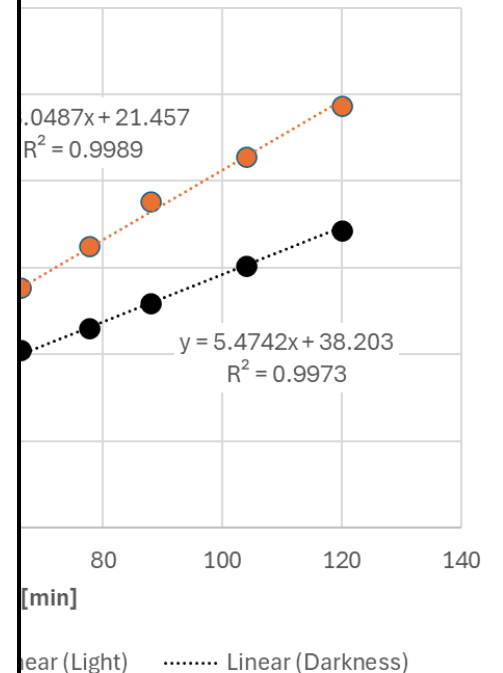
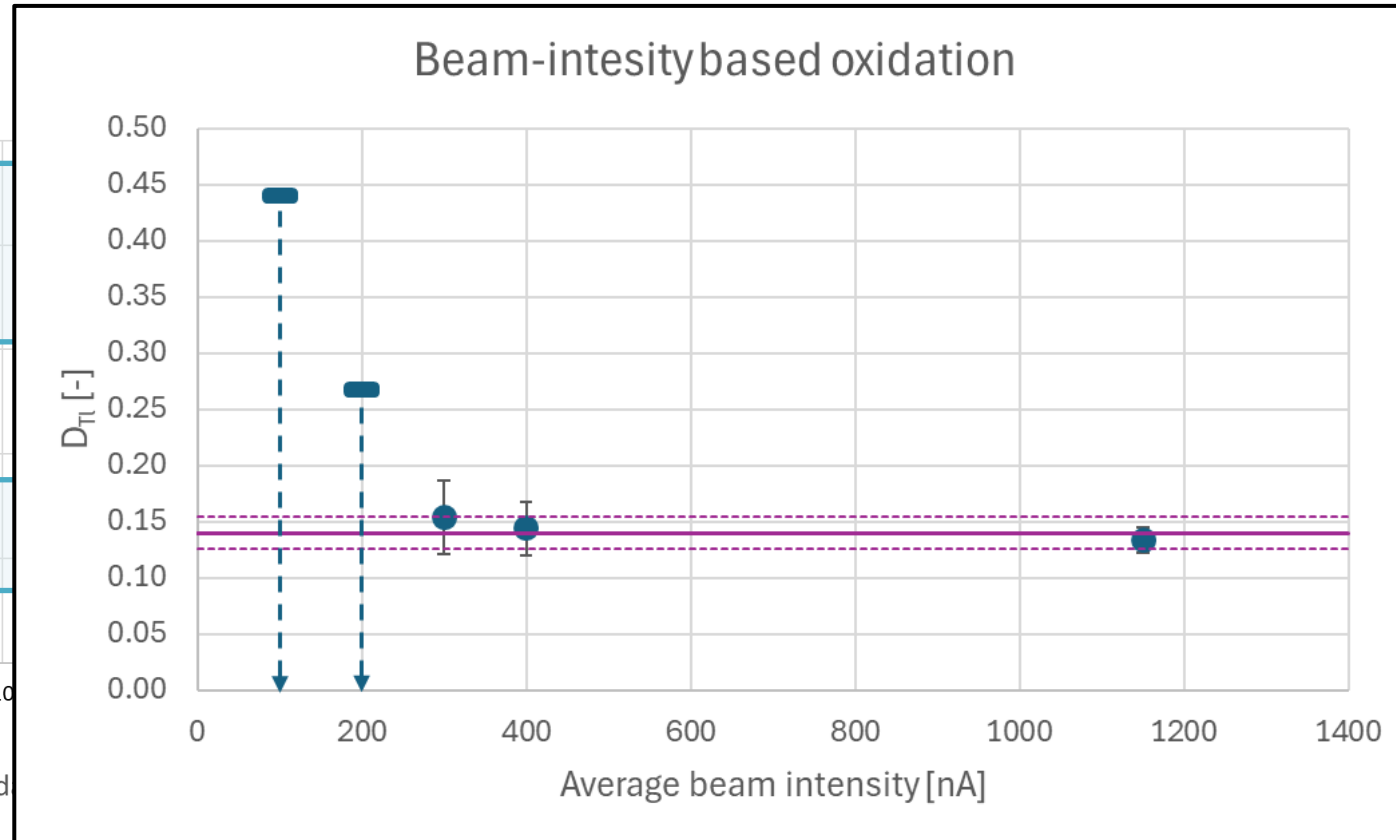
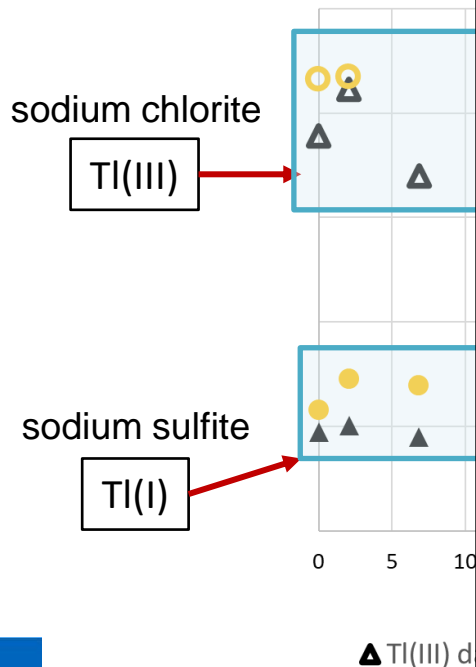
Extraction of Tl by Aliquat 336



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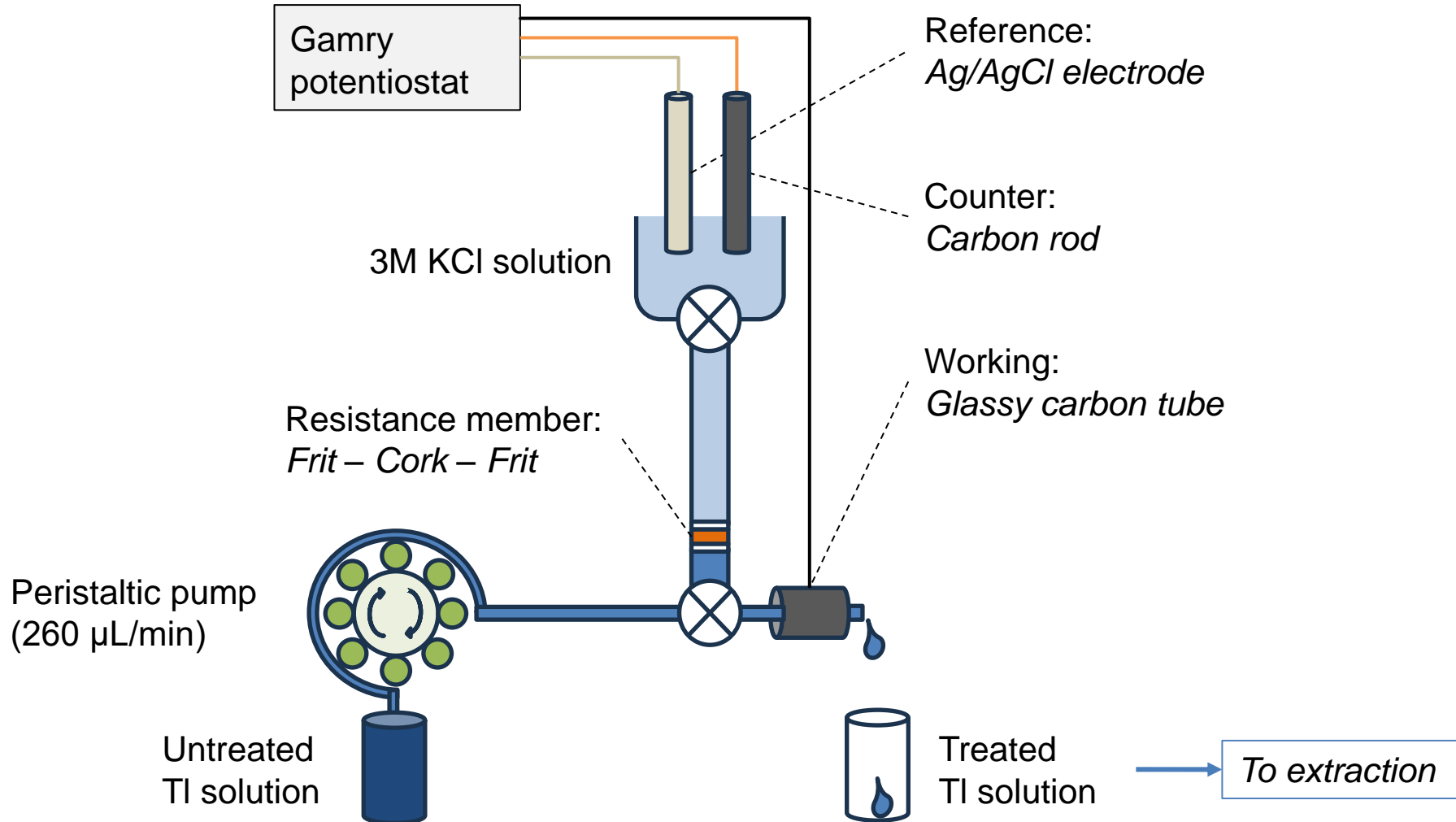
- Parallel experiments – with and without light
- Beam-induced oxidation not likely

ALL PRELIMINARY



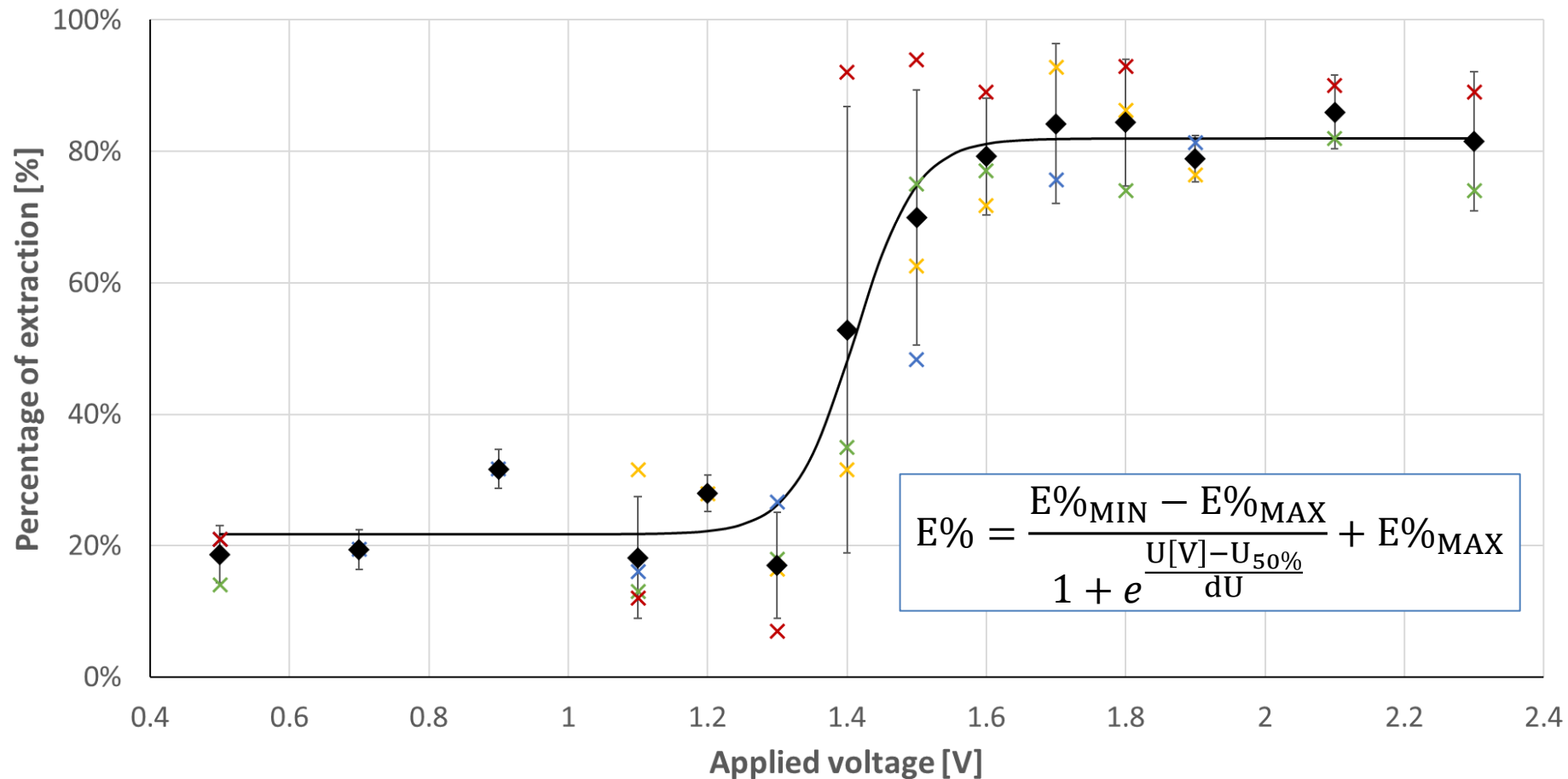
Extraction of Tl by Aliquat 336

Electrochemical pre-treatment step was employed to gain control over oxidation states



Extraction of Tl by Aliquat 336

Tl extraction after electrochemical pretreatment



Fit parameters:

- $E\%_{MAX} = 82\%$
- $E\%_{MIN} = 22\%$
- $U_{50\%} = 1.41 \text{ V}$
- $dU = 0.044$

$$E\% = \frac{E\%_{MIN} - E\%_{MAX}}{1 + e^{\frac{U[V] - U_{50\%}}{dU}}} + E\%_{MAX}$$

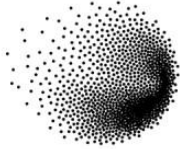


Conclusions

- **The MARGE system routinely running two GJT systems simultaneously** (thus far)
- **The PILS system was tested**, but needs further optimization and needs to be properly connected to the μ LLX system
- Lot of work carried out in the **liquid phase chemistry of Sg, Db and Nh homologues**
- **Tl stability** – photochemistry involved in the redox processes in the liquid phase
- **Electrochemical oxidation** – Tl has been repeatedly successfully oxidized

**Publications coming
soon!**

Two new research projects



P. Steinegger, yesterday

- ELCHOR Project
 - CTU, PSI, JH Inst CAS
 - Electrochemistry of group 11 elements



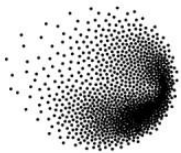
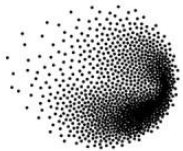
C. Folden, 16:15 today

- SuperDEPO Project
 - CTU, TAMU
 - Self-assembled monolayers for group 12 separation and detection



ASHES group since 2025

The MARGE system



Production and transport of
radionuclides in gas phase



Gas-into-liquid transfer



Fast aqueous chemistry processes
(solvent extraction, electrochemistry)



On-line detection

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1 PhD student
1 MSc student

1 PhD student

Acknowledgments



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JOHANNES GUTENBERG
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