Extraction of Nh Homologs Using “Designer” Molecules

Cody Folden
Folden@comp.tamu.edu
Cyclotron Institute, Texas A&M University

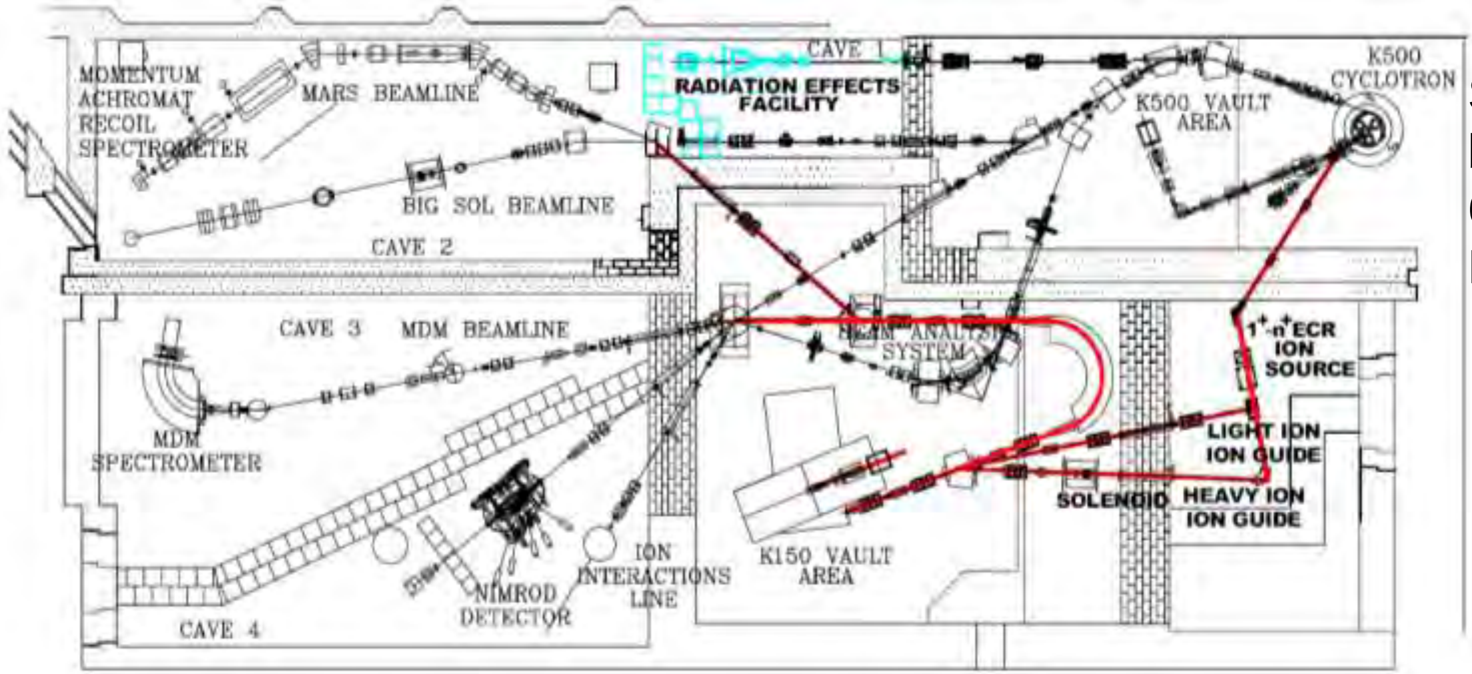
September 1, 2017

Outline

- Facilities Available at Texas A&M University
- New Recoil Transfer Chamber
- Offline Chemistry Experiments with Transactinide Homologs
  - Ionic Liquids
  - Deep Eutectic Solvents
  - Betaine-Based Solvents
- Summary and Future Work

Upgraded Capabilities at Texas A&M

- As part of an upgrade sponsored principally by DOE, the K150 88" cyclotron is being recommissioned.



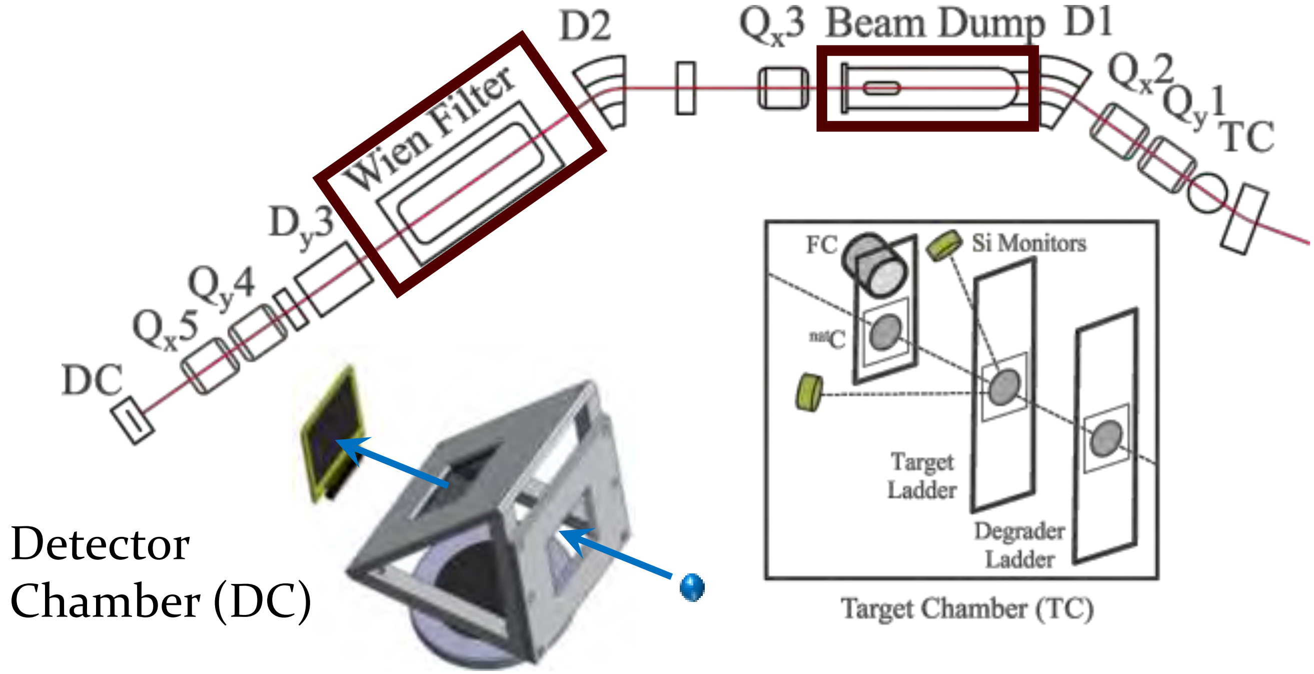
Supplied by 6.4-GHz ECR

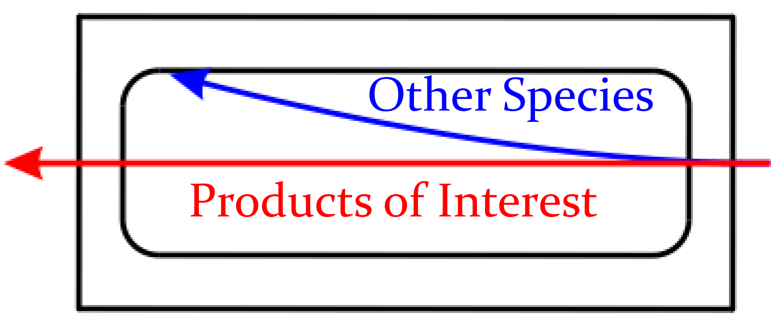
Supplied by 14.5-GHz ECR

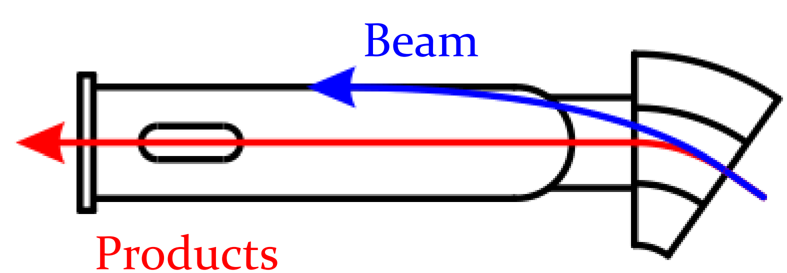
Figure 1. Layout for upgraded TAMU Facility. New additions are shown with red lines. The relocated SEE line is shown in light blue. High-intensity stable beams from the re-commissioned K150 Cyclotron will be used with ion guide techniques to produce high quality reaccelerated rare-ion beams from the K500 Cyclotron. K150 beams will be delivered to existing K500 experimental areas.

http://cyclotron.tamu.edu/facility_upgrade.pdf

Momentum Achromat Recoil Spectrometer (MARS)



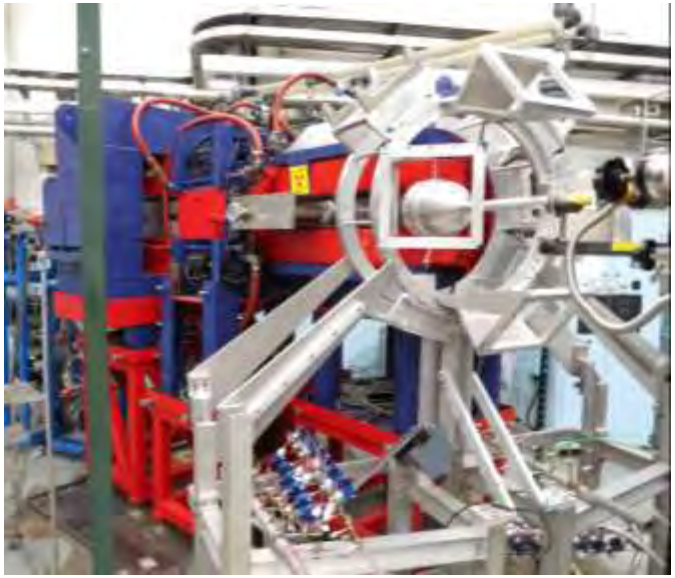




R.E. Tribble, R.H. Burch, and C.A. Gagliardi, Nucl. Instrum. Methods A **285**, 441 (1989).
CMF *et al.*, Nucl. Instrum. Methods A **678**, 1 (2012).

AGGIE

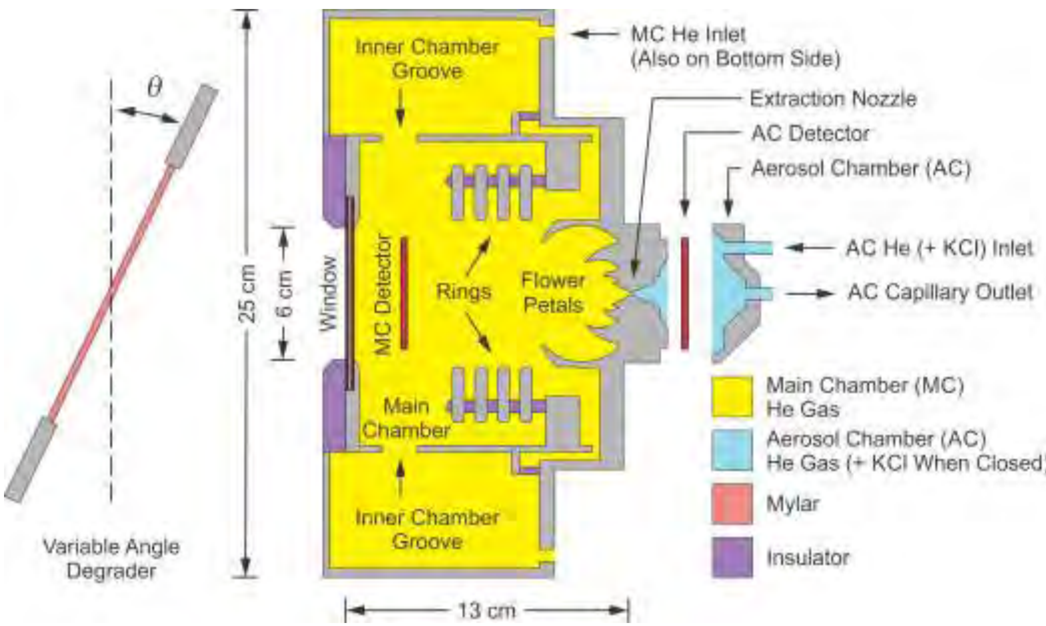
- Gas-filled separator formerly at Yale University.
- Acceptance cone: ±50 mrad horizontally and vertically
- Efficiency: 50-75% for $^{59}Co + ^{209}Bi \rightarrow ^{267}Ds + n$
- 120 x 40 strips (4,800 pixels) focal plane detector.
- Multiplexed pre-amps require only 16 ADC channels.

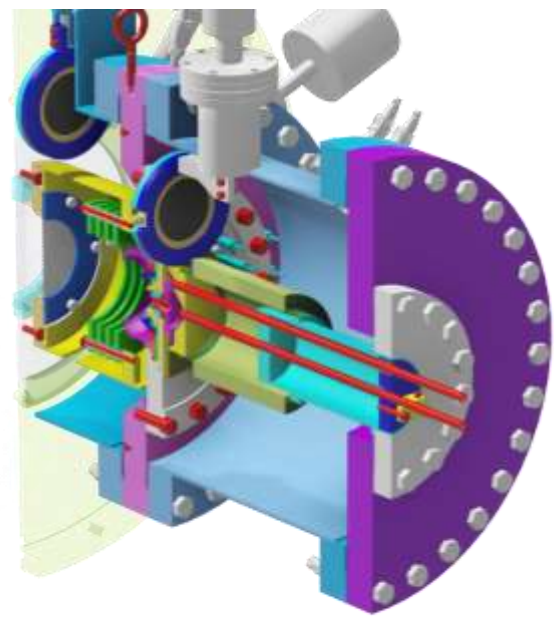


New Recoil Transfer Chamber (RTC)

Marisa Alfonso

- The RTC is designed to stop heavy elements and transport them to a chemistry laboratory.
- A “Variable Angle Degrader” fine tunes the location of stopped ions.
- An electric field focuses the ions into an extraction nozzle.





RTC Efficiency

- We measured the efficiency of the RTC using the $^{40}Ar + ^{118}Sn \rightarrow ^{152}Er + 6n$ reaction.
- The top figure shows the "raw" data. The bottom figure shows the data corrected for "range-out."
- If we correct for the transportation time through the RTC, the efficiency could be as high as (70 ± 9)%.

M. C. Alfonso *et al.,* NIMA **798**, 52 (2015).

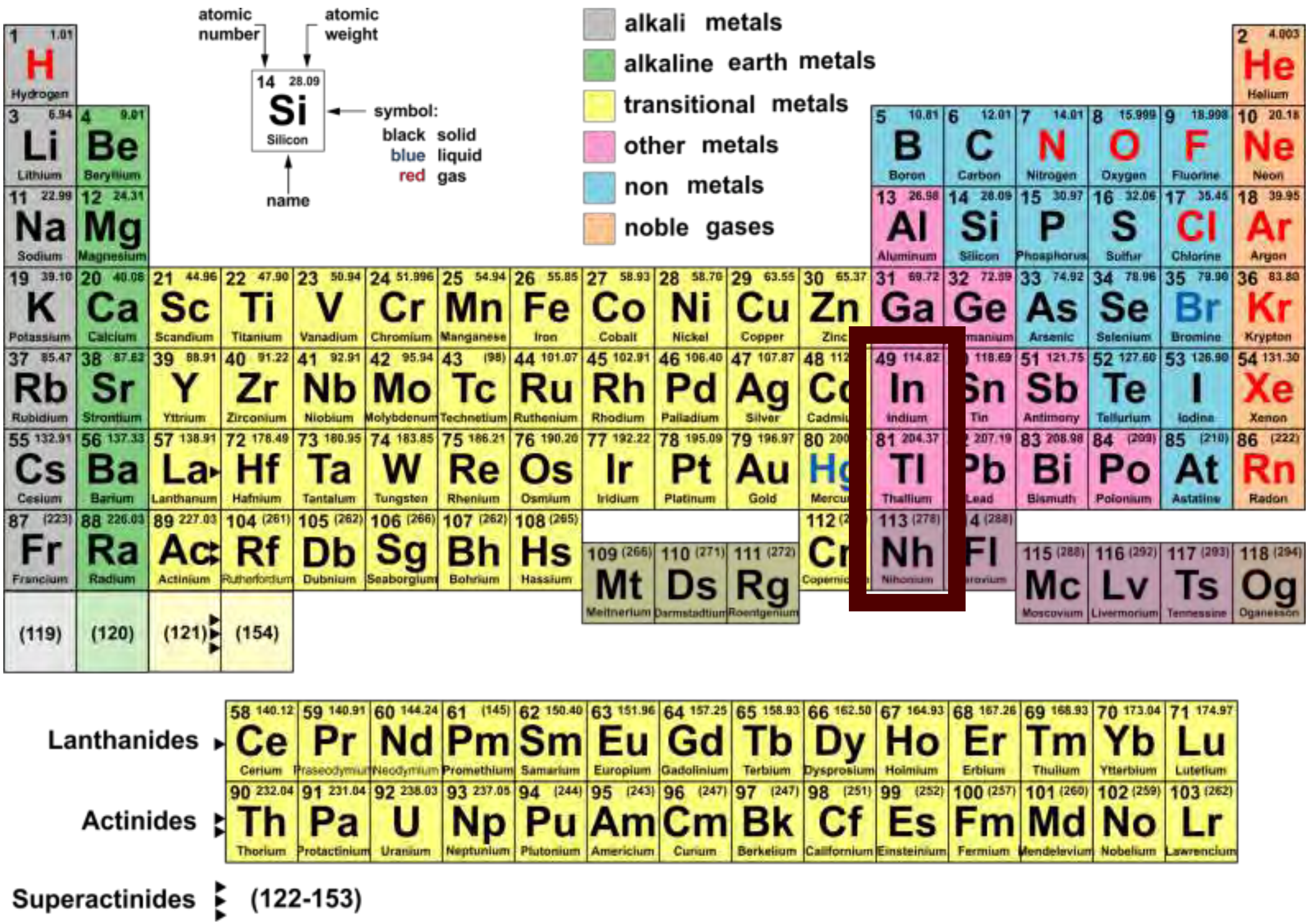
Online Radiochemistry Laboratory

- Equipped with:
- 6-ft HF/$HClO_4$ fume hood.
- Eyewash and safety shower.
- Lab grade sink.
- Isolated waste system.
- Climate controlled.
- Chemical and fire resistant interior.
- Non-slip flooring.
- GFI 110 V/220 V outlets.
- Lab benches.
- Chemical storage cabinets.
- Moveable via crane, forklift, or rolling.
- Track lighting.
- OSHA, State of Texas, and NRC compliant.

COMPACT Experiment at Texas A&M

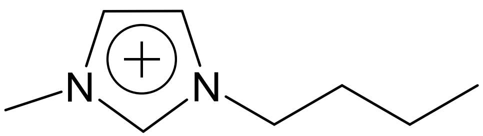
- We conducted an experiment to test improvements to the GSI gas cell and COMPACT using ^{184}Hg, ^{192}Pb, and ^{199}At. Analysis is ongoing.

Use of Homologs to Prepare for Transactinide Chemistry



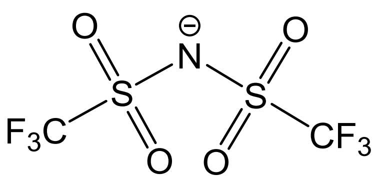
Separation of In and Tl Using Ionic Liquids

Evgeny Tereshatov



$[C_4mim^+]$

1-butyl-3-methylimidazolium



$[Tf_2N^-]$

bis(trifluoromethanesulfonyl)imide

- Separation factors of >10^7 were obtained for In(III) and Tl(III).

E. E. Tereshatov *et al.*,
Solvent Extr. Ion Exc. **33**(6), 607 (2015).

Mechanism of Extraction of Tl(III) from HCl Using Ionic Liquids

- There are two competing extraction mechanisms.
- Cation and/or Anion Exchange:

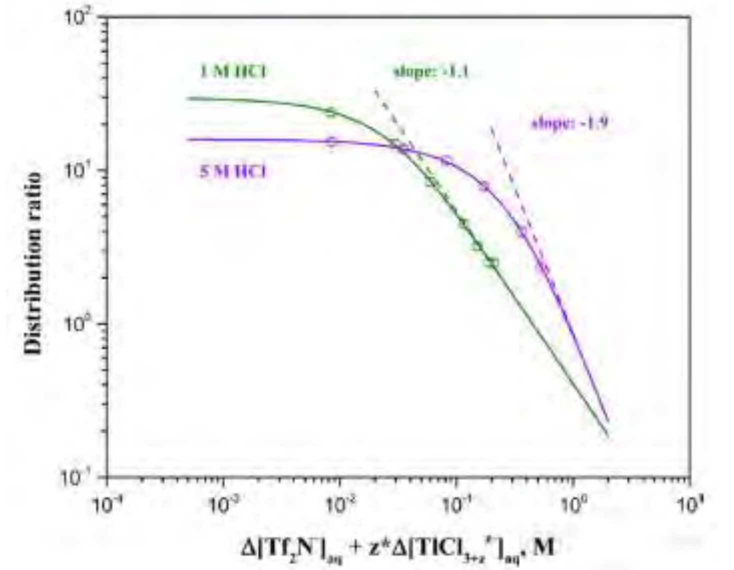
$$zC^{+}_{(org)} + S^{z+}_{(aq)} \leftrightarrow zC^{+}_{(aq)} + S^{z+}_{(org)}$$

$$zA^{-}_{(org)} + S^{z-}_{(aq)} \leftrightarrow zA^{-}_{(aq)} + S^{z-}_{(org)}$$

- Ion Pair Exchange:

$$zC^{+}_{(aq)} + S^{z-}_{(aq)} \leftrightarrow zC^{+}_{(org)} + S^{z-}_{(org)}$$

- It can be shown that:



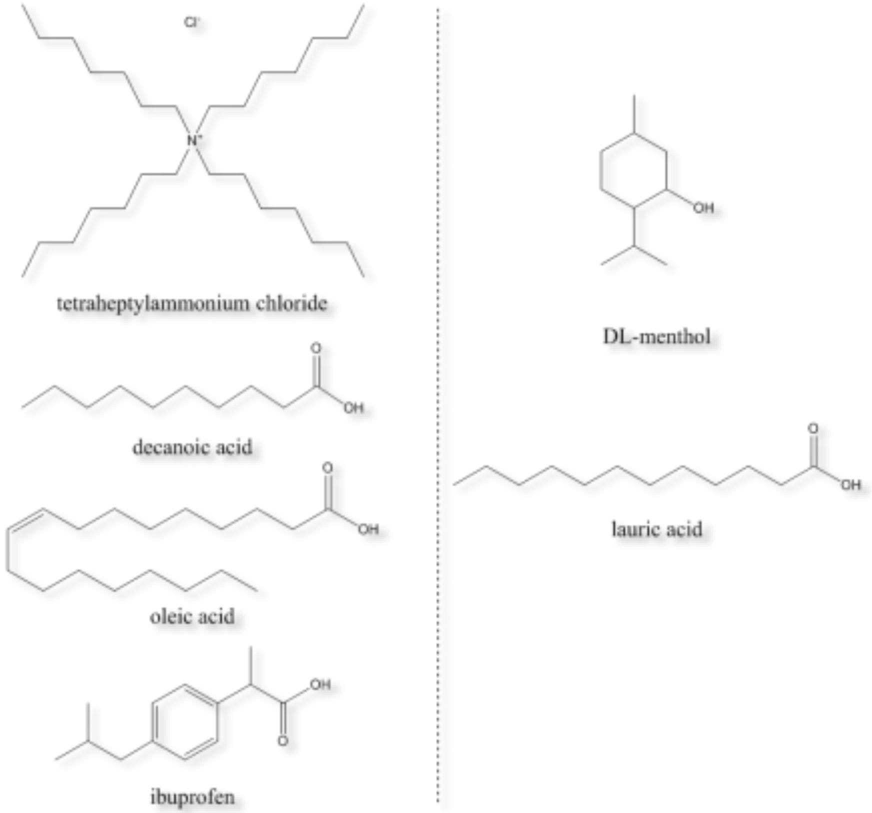
$$K_{IP} = 2^{+z/2} D_{TlCl_{3+z}^{z-}} \left\{ -(z/2)\left([TlCl_{3+z}^{z-}]_{org} + [Tf_2N^-]_{org}\right) + \left[(z/2)^2 \left([TlCl_{3+z}^{z-}]_{org} + [Tf_2N^-]_{org}\right)^2 + 4K_{sp} \right]^{1/2} \right\}^{-z/2}$$

- Fitting the *D*-values shows that $K_{IP}/K_{IE} \approx 200 - 3{,}000$ and $z = 1.12 \pm 0.06$ in 1 M HCl and $z = 1.97 \pm 0.16$ in 5 M HCl.
- The speciation is $TlCl_4^-$ and $TlCl_5^{2-}$ in 1 and 5 M HCl, respectively.

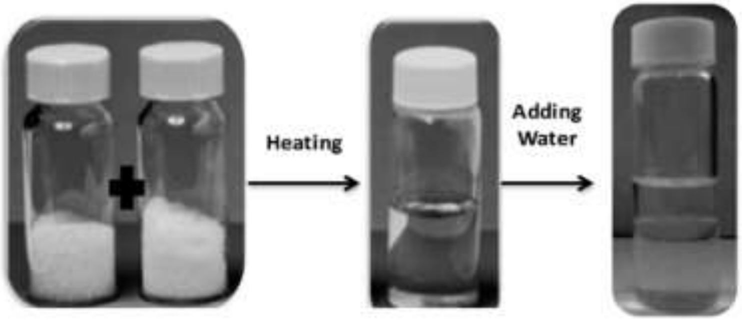
E. E. Tereshatov *et al.*, J. Phys. Chem. B **120**, 2311 (2016).

Hydrophobic DESs

- Deep eutectic solvents are a mixture of two compounds that have a much lower melting point than either compound individually.



- This effectively creates an ionic liquid from the two components.



Indium Extraction into DESs

$[\mathrm{In}^{3+}]_{aq} + 4[\mathrm{R'COO}^-]_{aq} \leftrightarrow [\mathrm{In(R'COO)}_4^-]_{org}$

$K_{\mathrm{ext}} = \frac{[\mathrm{In(R'COO)}_4^-]}{[\mathrm{In}^{3+}][\mathrm{R'COO}^-]^4}$

$[\mathrm{R'COOH}]_{aq} \leftrightarrow [\mathrm{R'COO}^-]_{aq} + [\mathrm{H}^+]_{aq}$

$K_{\mathrm{a}} = \frac{[\mathrm{R'COO}^-][\mathrm{H}^+]}{[\mathrm{R'COOH}]}$

$\log D = \log C - 4\log\,[\mathrm{H}^+]$

$K_{\mathrm{ext}} = \frac{D}{[\mathrm{R'COO}^-]^4} = \frac{D[\mathrm{H}^+]^4}{K_{\mathrm{a}}{}^4[\mathrm{R'COOH}]^4}$

E. E. Tereshatov *et al.*, Green Chem. **18**, 4616 (2016).

Summary and Future Work

- We have extensive facilities for developing online chemistry experiments.
- We have designed, fabricated, and characterized a new Recoil Transfer Chamber with high efficiency.
- We have developed an IL-based method for separations of In and Tl with extremely high separation factors.
- We are looking at other options for extractions using novel materials.
- We are considering new ideas for how the chemical properties of nihonium might be studied.

Acknowledgements: Coworkers and Funding Agencies

- M. C. Alfonso
- M. E. Bennett
- M. Block
- M. Yu. Boltoeva
- M. J. DeVanzo
- Ch. E. Düllmann
- K. J. Glennon
- S. Götz
- D. A. Mayorov
- V. Mazan
- S. Raeder
- J. A. Sefcik
- E. E. Tereshatov
- K. A. Tran
- M. F. Volia
- T. A. Werke
- A. B. Yakushev