

# Extraction of Nh Homologs Using “Designer” Molecules

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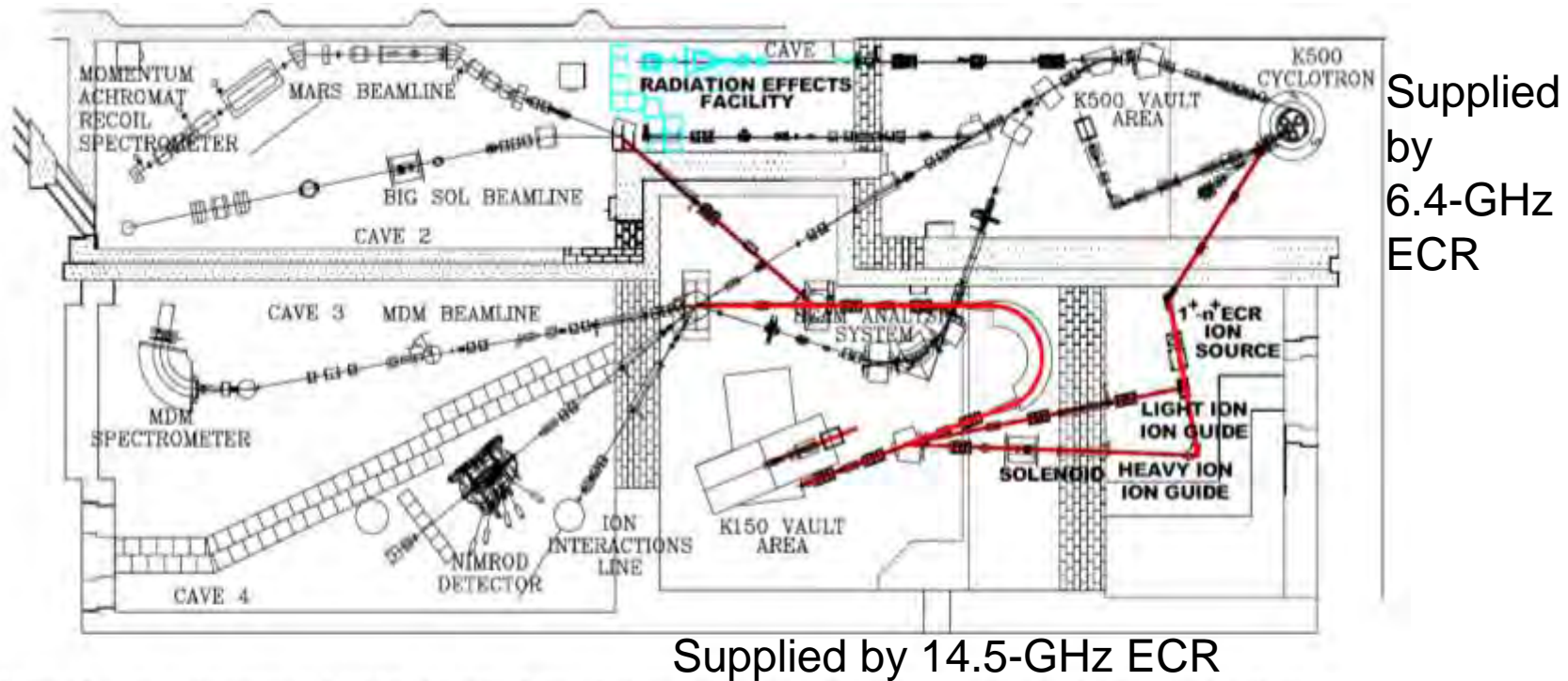
September 1, 2017

# 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

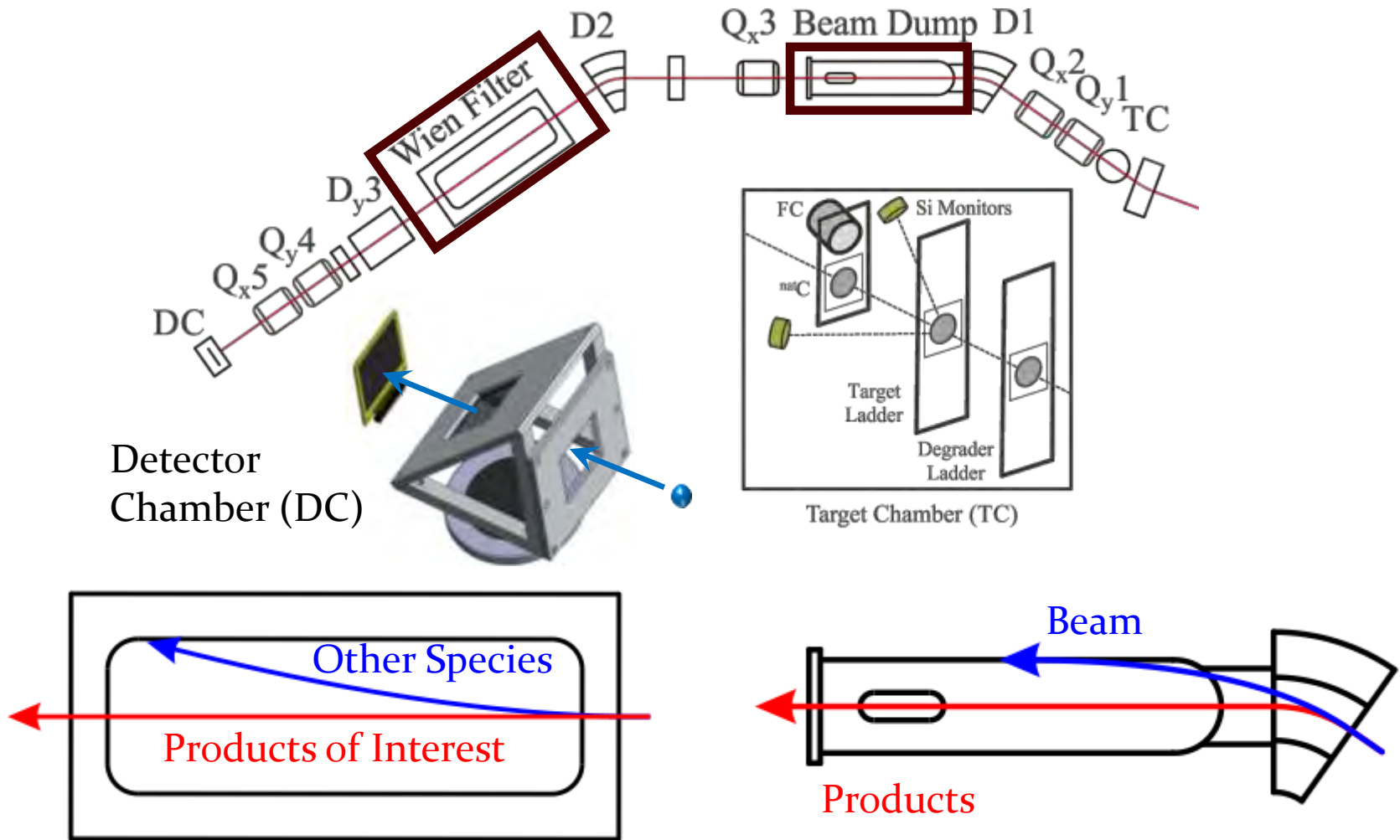
# Upgraded Capabilities at Texas A&M

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



**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.

# 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:  $\pm 50$  mrad horizontally and vertically
- Efficiency: 50-75% for  $^{59}\text{Co} + ^{209}\text{Bi} \rightarrow ^{267}\text{Ds} + n$
- 120 x 40 strips (4,800 pixels) focal plane detector.
- Multiplexed pre-amps require only 16 ADC channels.



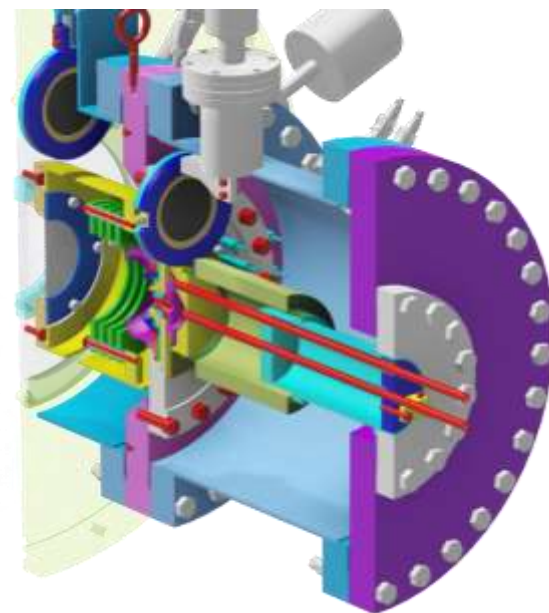
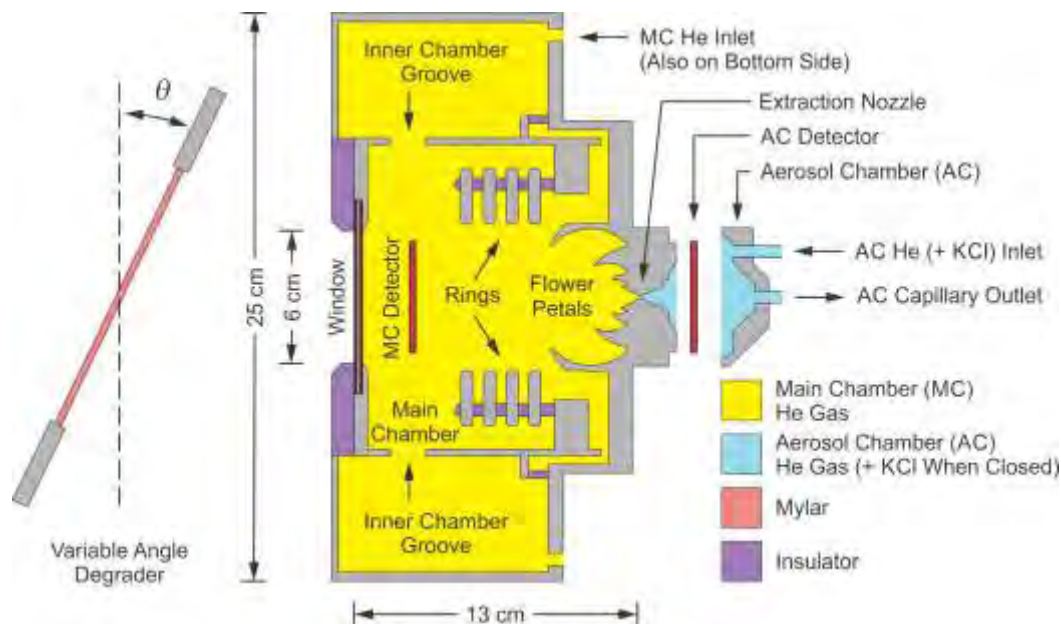


# New Recoil Transfer Chamber (RTC)



Marisa Alfonso

- The RTC is designed to stop heavy elements and transport them to a chemistry laboratory.
- A “Variable Angle Degradator” 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  $^{40}\text{Ar} + ^{118}\text{Sn} \rightarrow ^{152}\text{Er} + 6n$  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  $(70 \pm 9)\%$ .

# Online Radiochemistry Laboratory

- Equipped with:
- 6-ft HF/HClO<sub>4</sub> 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  $^{184}\text{Hg}$ ,  $^{192}\text{Pb}$ , and  $^{199}\text{At}$ . Analysis is ongoing.



# Use of Homologs to Prepare for Transactinide Chemistry

atomic number      atomic weight

14    28.09

**Si**      symbol:

Silicon      black solid  
blue liquid  
red gas

name

- alkali metals
- alkaline earth metals
- transitional metals
- other metals
- non metals
- noble gases

1 1.01 <b>H</b> Hydrogen																	2 4.003 <b>He</b> Helium						
3 6.94 <b>Li</b> Lithium	4 9.01 <b>Be</b> Beryllium																	5 10.81 <b>B</b> Boron	6 12.01 <b>C</b> Carbon	7 14.01 <b>N</b> Nitrogen	8 15.999 <b>O</b> Oxygen	9 18.998 <b>F</b> Fluorine	10 20.18 <b>Ne</b> Neon
11 22.99 <b>Na</b> Sodium	12 24.31 <b>Mg</b> Magnesium																	13 26.98 <b>Al</b> Aluminum	14 28.09 <b>Si</b> Silicon	15 30.97 <b>P</b> Phosphorus	16 32.06 <b>S</b> Sulfur	17 35.45 <b>Cl</b> Chlorine	18 39.95 <b>Ar</b> Argon
19 39.10 <b>K</b> Potassium	20 40.08 <b>Ca</b> Calcium	21 44.96 <b>Sc</b> Scandium	22 47.90 <b>Ti</b> Titanium	23 50.94 <b>V</b> Vanadium	24 51.996 <b>Cr</b> Chromium	25 54.94 <b>Mn</b> Manganese	26 55.85 <b>Fe</b> Iron	27 58.93 <b>Co</b> Cobalt	28 58.70 <b>Ni</b> Nickel	29 63.55 <b>Cu</b> Copper	30 65.37 <b>Zn</b> Zinc	31 69.72 <b>Ga</b> Gallium	32 72.09 <b>Ge</b> Germanium	33 74.92 <b>As</b> Arsenic	34 78.96 <b>Se</b> Selenium	35 79.90 <b>Br</b> Bromine	36 83.80 <b>Kr</b> Krypton						
37 85.47 <b>Rb</b> Rubidium	38 87.62 <b>Sr</b> Strontium	39 88.91 <b>Y</b> Yttrium	40 91.22 <b>Zr</b> Zirconium	41 92.91 <b>Nb</b> Niobium	42 95.94 <b>Mo</b> Molybdenum	43 (98) <b>Tc</b> Technetium	44 101.07 <b>Ru</b> Ruthenium	45 102.91 <b>Rh</b> Rhodium	46 106.40 <b>Pd</b> Palladium	47 107.87 <b>Ag</b> Silver	48 112.41 <b>Cd</b> Cadmium	49 114.82 <b>In</b> Indium	50 118.69 <b>Sn</b> Tin	51 121.75 <b>Sb</b> Antimony	52 127.60 <b>Te</b> Tellurium	53 126.90 <b>I</b> Iodine	54 131.30 <b>Xe</b> Xenon						
55 132.91 <b>Cs</b> Cesium	56 137.33 <b>Ba</b> Barium	57 138.91 <b>La</b> Lanthanum	72 176.49 <b>Hf</b> Hafnium	73 180.95 <b>Ta</b> Tantalum	74 183.85 <b>W</b> Tungsten	75 186.21 <b>Re</b> Rhenium	76 190.20 <b>Os</b> Osmium	77 192.22 <b>Ir</b> Iridium	78 195.09 <b>Pt</b> Platinum	79 196.97 <b>Au</b> Gold	80 200.59 <b>Hg</b> Mercury	81 204.37 <b>Tl</b> Thallium	82 207.19 <b>Pb</b> Lead	83 208.98 <b>Bi</b> Bismuth	84 (209) <b>Po</b> Polonium	85 (210) <b>At</b> Astatine	86 (222) <b>Rn</b> Radon						
87 (223) <b>Fr</b> Francium	88 226.03 <b>Ra</b> Radium	89 227.03 <b>Ac</b> Actinium	104 (261) <b>Rf</b> Rutherfordium	105 (262) <b>Db</b> Dubnium	106 (266) <b>Sg</b> Seaborgium	107 (262) <b>Bh</b> Bohrium	108 (265) <b>Hs</b> Hassium					112 (285) <b>Cn</b> Copernicium	113 (278) <b>Nh</b> Nihonium	114 (288) <b>Fl</b> Flerovium	115 (288) <b>Mc</b> Moscovium	116 (292) <b>Lv</b> Livermorium	117 (293) <b>Ts</b> Tennessine	118 (294) <b>Og</b> Oganesson					
(119)	(120)	(121)	(154)																				

Lanthanides

58 140.12 <b>Ce</b> Cerium	59 140.91 <b>Pr</b> Praseodymium	60 144.24 <b>Nd</b> Neodymium	61 (145) <b>Pm</b> Promethium	62 150.40 <b>Sm</b> Samarium	63 151.96 <b>Eu</b> Europium	64 157.25 <b>Gd</b> Gadolinium	65 158.93 <b>Tb</b> Terbium	66 162.50 <b>Dy</b> Dysprosium	67 164.93 <b>Ho</b> Holmium	68 167.26 <b>Er</b> Erbium	69 168.93 <b>Tm</b> Thulium	70 173.04 <b>Yb</b> Ytterbium	71 174.97 <b>Lu</b> Lutetium
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Actinides

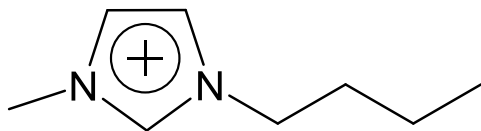
90 232.04 <b>Th</b> Thorium	91 231.04 <b>Pa</b> Protactinium	92 238.03 <b>U</b> Uranium	93 237.05 <b>Np</b> Neptunium	94 (244) <b>Pu</b> Plutonium	95 (243) <b>Am</b> Americium	96 (247) <b>Cm</b> Curium	97 (247) <b>Bk</b> Berkelium	98 (251) <b>Cf</b> Californium	99 (252) <b>Es</b> Einsteinium	100 (257) <b>Fm</b> Fermium	101 (260) <b>Md</b> Mendelevium	102 (259) <b>No</b> Nobelium	103 (262) <b>Lr</b> Lawrencium
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Superactinides      (122-153)

# Separation of In and Tl Using Ionic Liquids

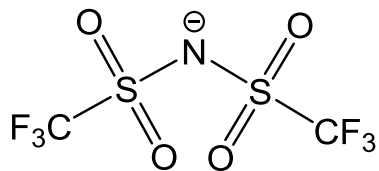


Evgeny Tereshatov



[C<sub>4</sub>mim<sup>+</sup>]

1-butyl-3-methylimidazolium



[Tf<sub>2</sub>N<sup>-</sup>]

bis(trifluoromethanesulfonyl)imide

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

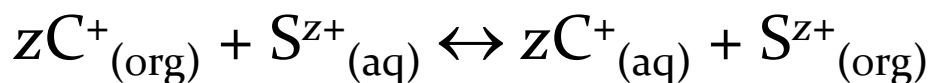
E. E. Tereshatov *et al.*,  
Solvent Extr. Ion Exc. **33**(6), 607 (2015).



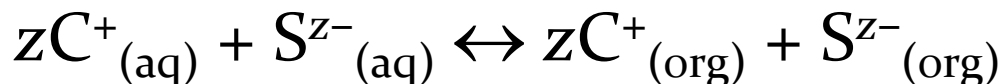
# Mechanism of Extraction of Tl(III) from HCl Using Ionic Liquids

- There are two competing extraction mechanisms.

- Cation and/or Anion Exchange:



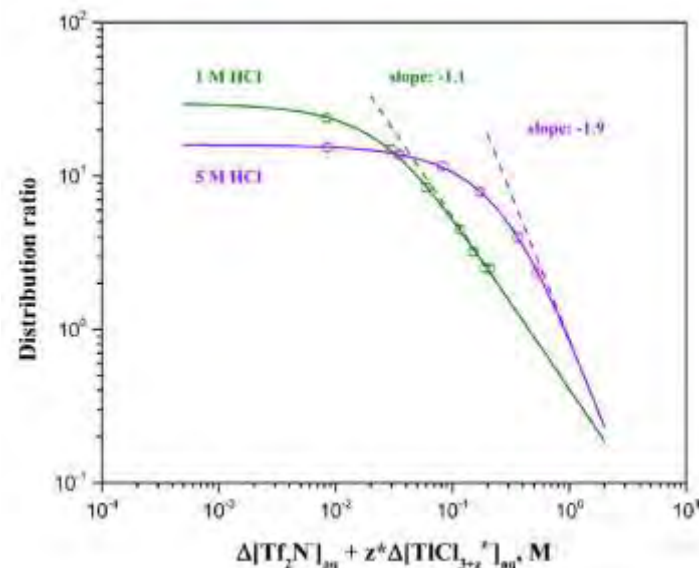
- Ion Pair Exchange:



- It can be shown that:

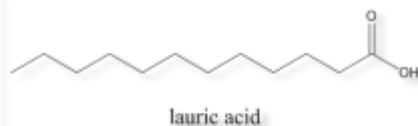
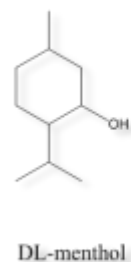
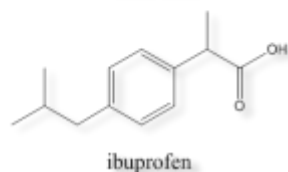
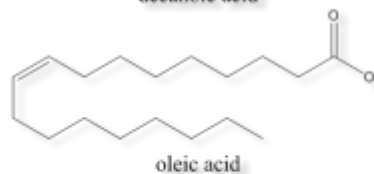
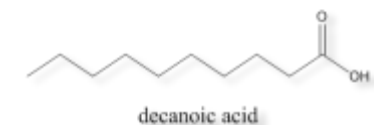
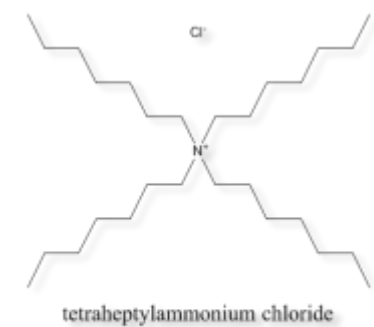
$$K_{IP} = 2^{+z/2} D_{TlCl_{3+z}^{z-}} \left\{ -(z/2) \left( [TlCl_{3+z}^{z-}]_{org} + [Tf_2N^-]_{org} \right) + \left[ (z/2)^2 \left( [TlCl_{3+z}^{z-}]_{org} + [Tf_2N^-]_{org} \right)^2 + 4K_{sp} \right]^{1/2} \right\}^{-z/2}$$

- 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.

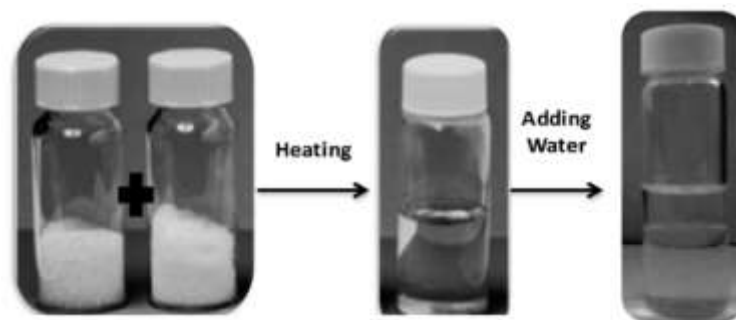


# Hydrophobic DESs

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

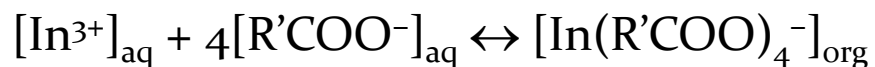


- This effectively creates an ionic liquid from the two components.





# Indium Extraction into DESs



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

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

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

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

# 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.

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