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Molten Salt Reactor PROGRAM

Thermochemical and Thermophysical Property Database Development at INL

Dr. Toni Karlsson, Research Scientist
Advanced Technologies of Molten Salts (AToMS)
Idaho National Laboratory, USA

INL Idaho National Laboratory

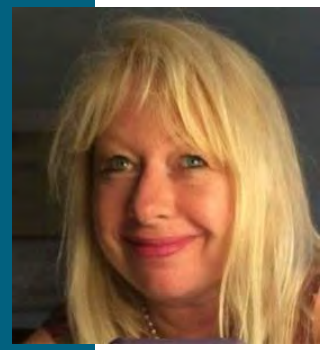
Annual MSR Campaign Review Meeting

Milestones FY23

- M3AT-23IN0705021-Draft Journal Article on NaCl-UCl_3-$PuCl_3$ properties
- M3AT-23IN0705022-Retrieve Salt from FCF Hot Cell Facility and Initiate Chemical/Isotopic Analysis
- M3AT-23IN0705023-Complete Chemical/Isotopic Analysis of FCF Salt

All milestones on track!

- **Teamwork makes the dream work!**
  - Brought together by the MSR Campaign
    - INL, PNNL, ORNL, LANL, U. of South Carolina
- Work as a united, cross-disciplinary team, to collect first-of-a-kind data needed for model validation, database development, and MSR deployment with confidence
- Progress through teamwork - INL


Dr. Paviet


Dr. Karlsson

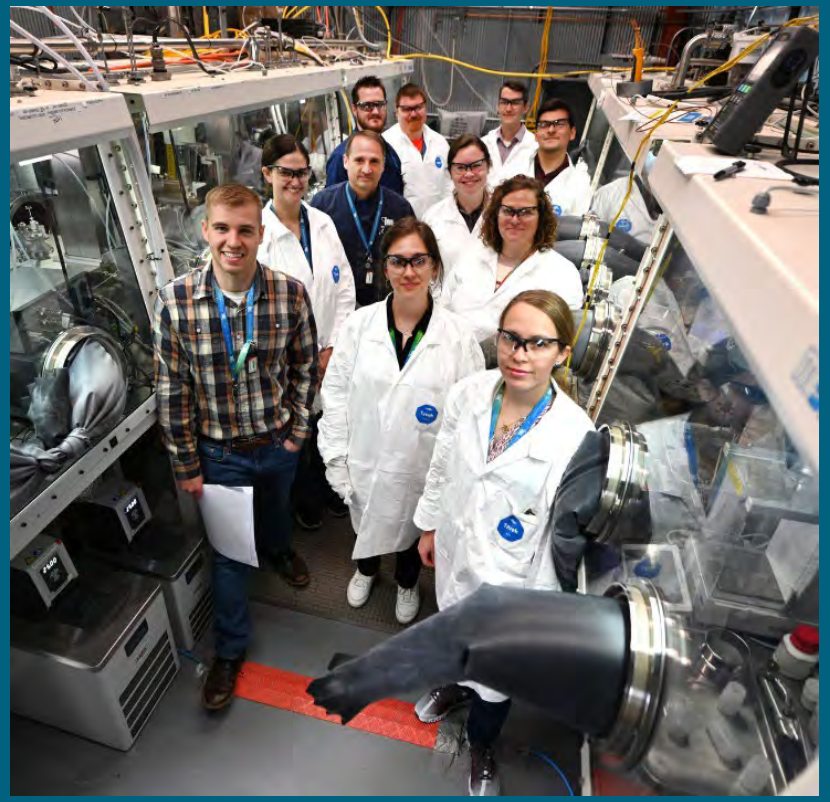

Dr. Nguyen


Dr. Glezakou


Dr. Monreal


Dr. Schorne Pinto

Goals (FY23)



Molten salt interns, post-docs, researchers, and technicians during summer of 2022 at INL

- **Research Goals**
  - Obtain fundamental property data on NaCl-UCl_3-$PuCl_3$
    - Melting temperature, enthalpy of fusion, and density
  - Complement results with “sister” experiments performed at LANL
  - Turn over data to modeling team (ORNL, PNNL) for model improvements and validation
  - Examination of properties from corrosion salt (flowing)
- **Human Performance Goals**
  - Allow INL staff to modify existing and develop new techniques for high temperature property measurements
  - Mentor early career researchers on molten salt thermophysical properties data acquisition, reporting, networking
    - Interns, post-doc, early career researchers
  - Encourage participation in conferences, papers, networking

FY23 Activities

Shawn Reddish, transuranic glovebox operator making salt samples

- $NaCl\text{-}UCl_3\text{-}PuCl_3$ (61-30-9) Fuel Salt
  - Amalgamation and homogenization of salts (FMF)
  - Density determination (FMF)
  - Prepare sample for transfer (to ARL)
  - Melting temperature and enthalpy determination (FFGB)
- $NaCl\text{-}PuCl_3$ (64-36) Used Salt (possible corrosion products)
  - Extraction from flow loop (FCF)
  - Transfer samples (ARL, FFGB, FMF)
  - Elemental, isotopic, and impurity analysis (ARL)
  - Melting temperature and enthalpy determination (FFGB)
  - Density determination (FMF)

NaCl (left)
UCl_3 (middle)
$NaCl\text{-}PuCl_3$ (right)

Note: UCl_3 was made by Hilary Fitzgerald at TerraPower. Thank-you Hilary!

Density

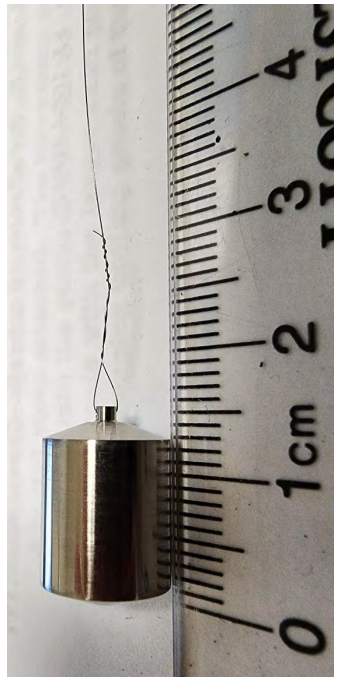
- Archimedes' hydrostatic method

Fuel Manufacturing Facility (FMF) transuranic glove box operators and researcher

Experimental Setup

- Bobbers are fabricated at INL
  - Each calibrated on a calibrated balance
  - Air, water, ethanol
- Load salt into crucible, placed in furnace, heated to desired temperature
- Mass measurements at each temp

Densitometer setup in FMF



Density bobber

Equipment	Description
Furnace	Ventura electromelt with graphite crucible
Bobber	Ni, ESPI Metals 99.99%, 1.0408 ± 0.0006 cm^3
Wire	0.1 mm W, Alfa Aesar, 99.95% metals basis, Batch #10404 Lot #X28G043
Crucible	GAZA2, 25 mL Sigradur cylindrical crucible, HTW Germany
Balance, Bottom-loading	Mettler Toledo WXSS204, INL ID#525063 tolerance 0.8 mg
Thermocouple	Omega TJ36-CAIN-116U-8-SMPW-M, Inconel sheathed K-type ungrounded
Thermocouple Reader	Omega HH502, s/n 21000159, uncertainty 0.05% of reading + 0.3 K

Density

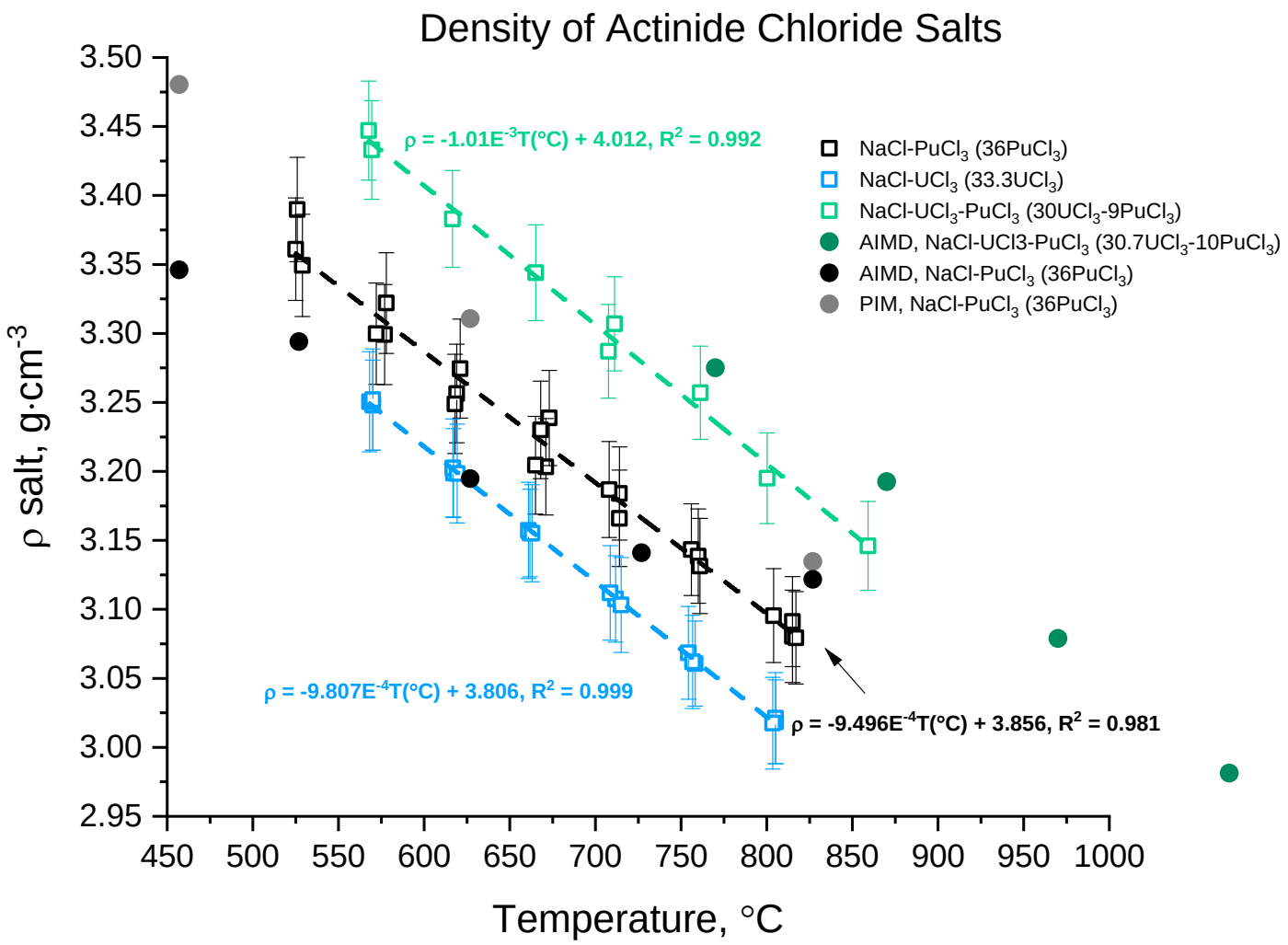
- FY23 we are studying
  - NaCl-UCl_3-9$PuCl_3$ (INL)
  - NaCl-UCl_3-5$PuCl_3$ (LANL)
  - Modeling at PNNL/ORNL
- Goal is collaborative journal publication

Dr. Woods
Experiment

Dr. Nguyen
Modeling

Dr. Glezakou
Modeling

Experimental and Modeled Density of Actinide Chlorides



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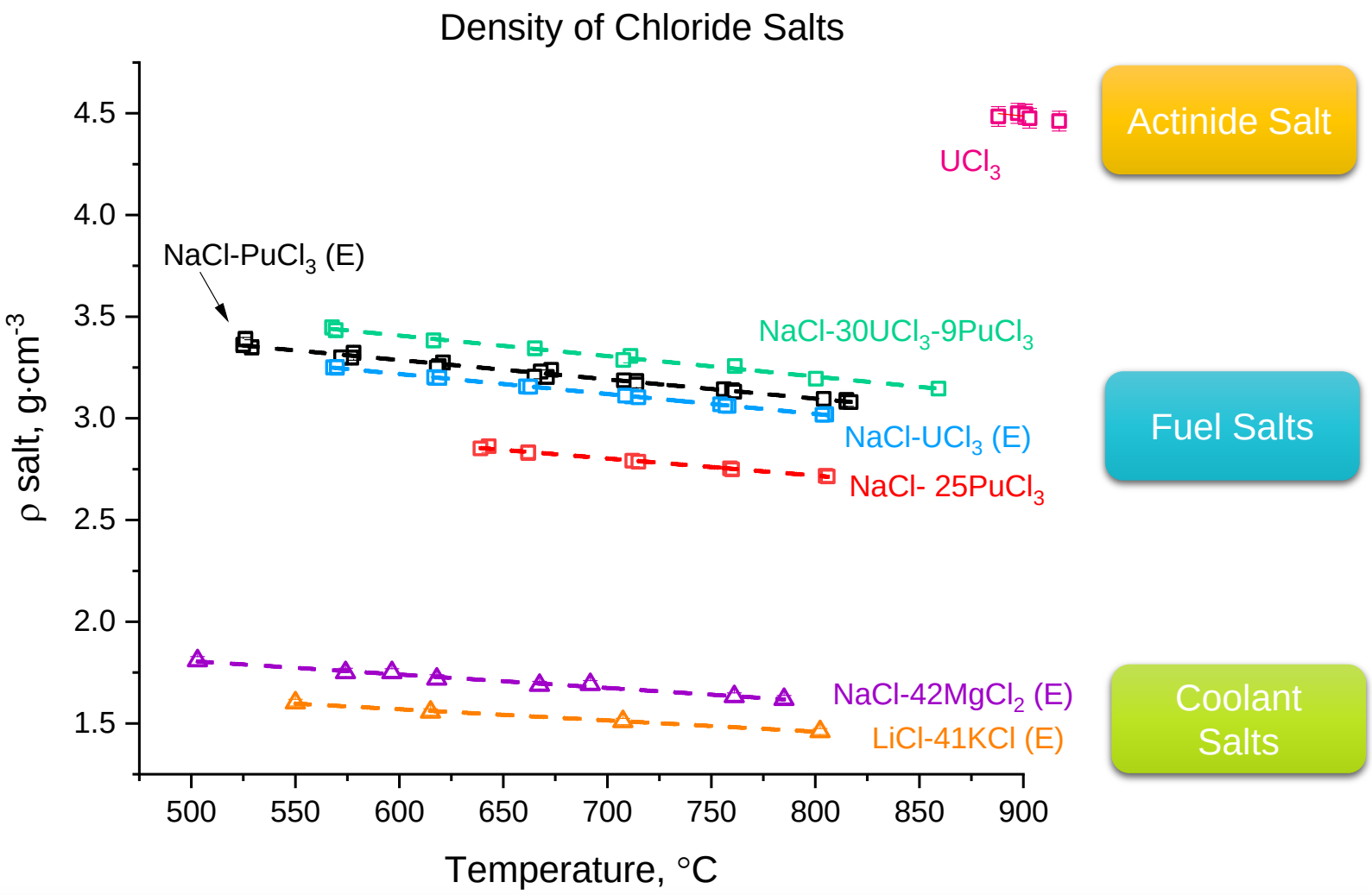
Molten Salt Reactor PROGRAM

Density

Applications of salts

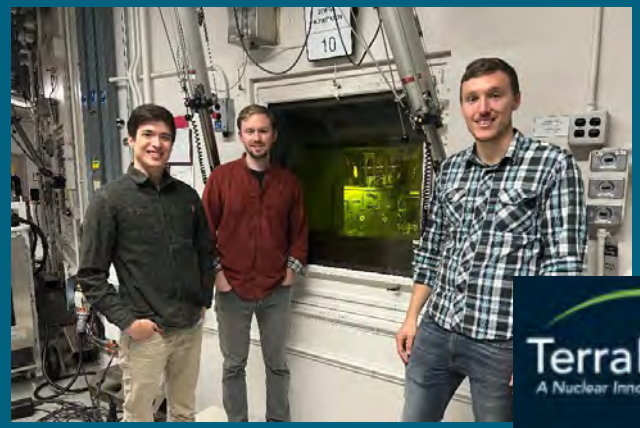
- Metal production
- Energy storage
- Pyrochemical reprocessing
- Fuels and coolants for MSRs
- Many others

Comparison of Density for Different Salts



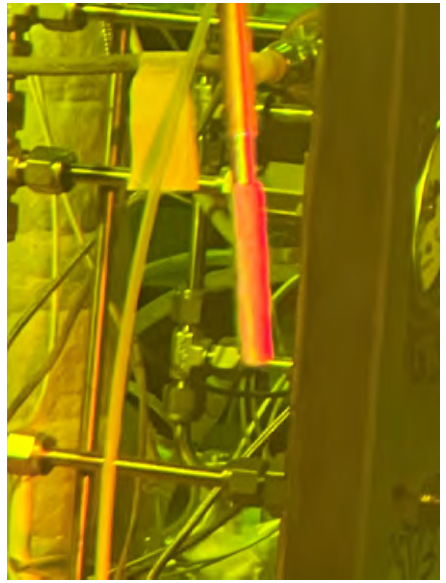
Corrosion Experiment (Salt)

- $NaCl-PuCl_3$ (E) salt in natural convection flow loop for 1000hr
  - TerraPower interested in flow loop material
  - MSR Campaign interested in used salt



TerraPower staff after the salt began circulating
Left to right: Ivan Mitchell, Sean Gagnon, Cody Falconer

Dip sampler removing salt from drain tank



Salt sample on dip sampler

$NaCl-PuCl_3$ salt, post corrosion experiment, sent for analysis

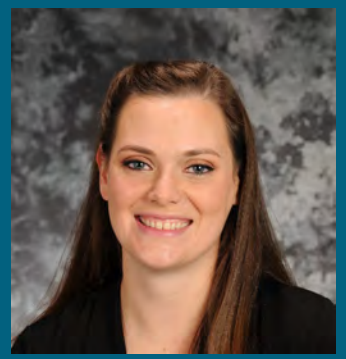
David Tolman, FCF Engineer

Hot cell operators Chase Wren (left) and Tyler Rigolout (right) in FCF

Microloop experiment was performed under a Cooperative Research and Development Agreement (CRADA) between TerraPower and INL designed to examine the corrosion of structural material when exposed to molten flowing salt $PuCl_3$-NaCl

Recovered Flow Loop Salt

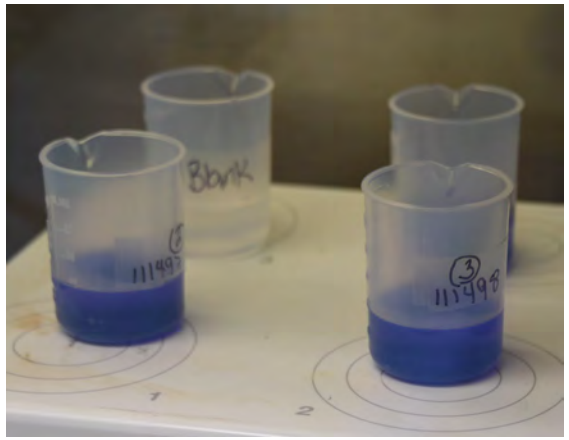
- Recovered 40 grams of salt from the flow loop for analysis and property measurements
  - Elemental/isotopic analysis
  - Impurity analysis
  - Melting temperature
  - Enthalpy
  - Density



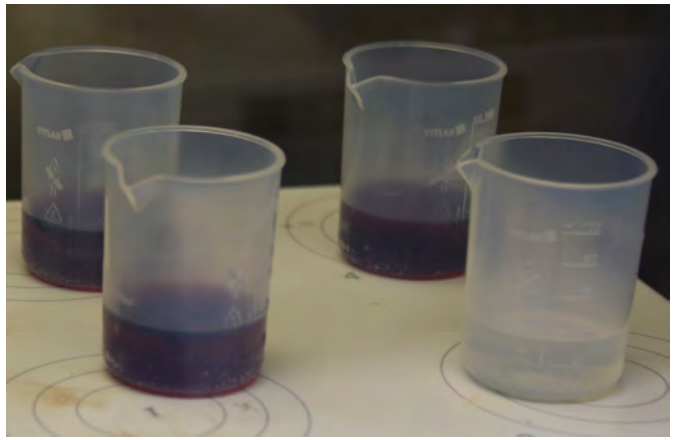
Pamela Wiscaver

Tony Jones

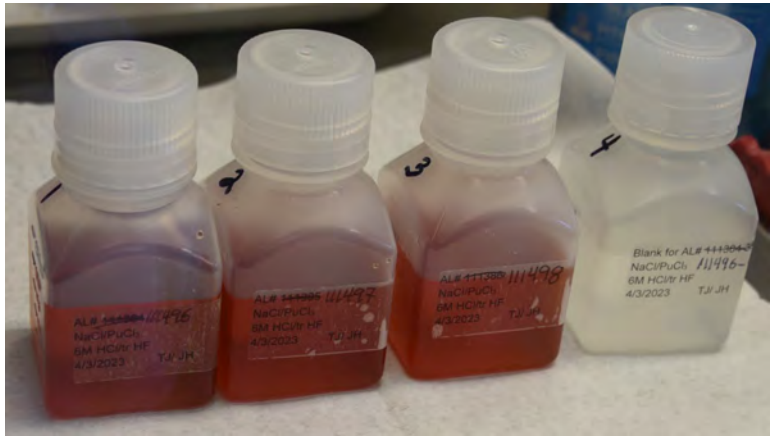
Dr. Lindsey Lecrivain



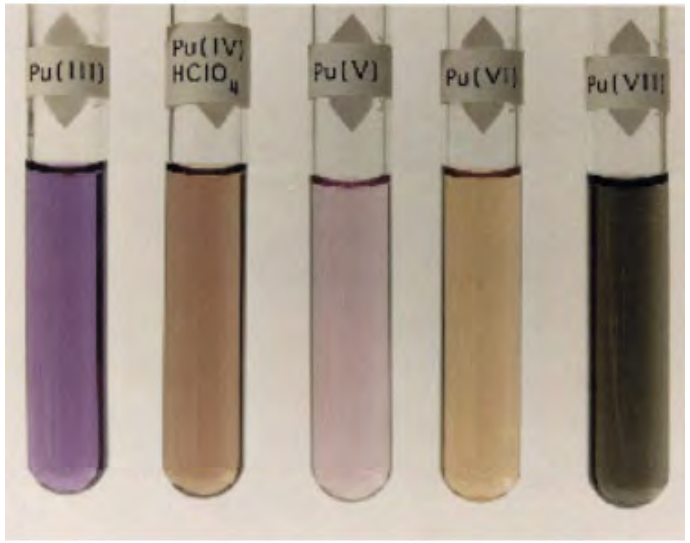
Addition of HCl, Pu^{+3}



Addition of HNO_3 Pu^{+6}



First dilution in sample preparation



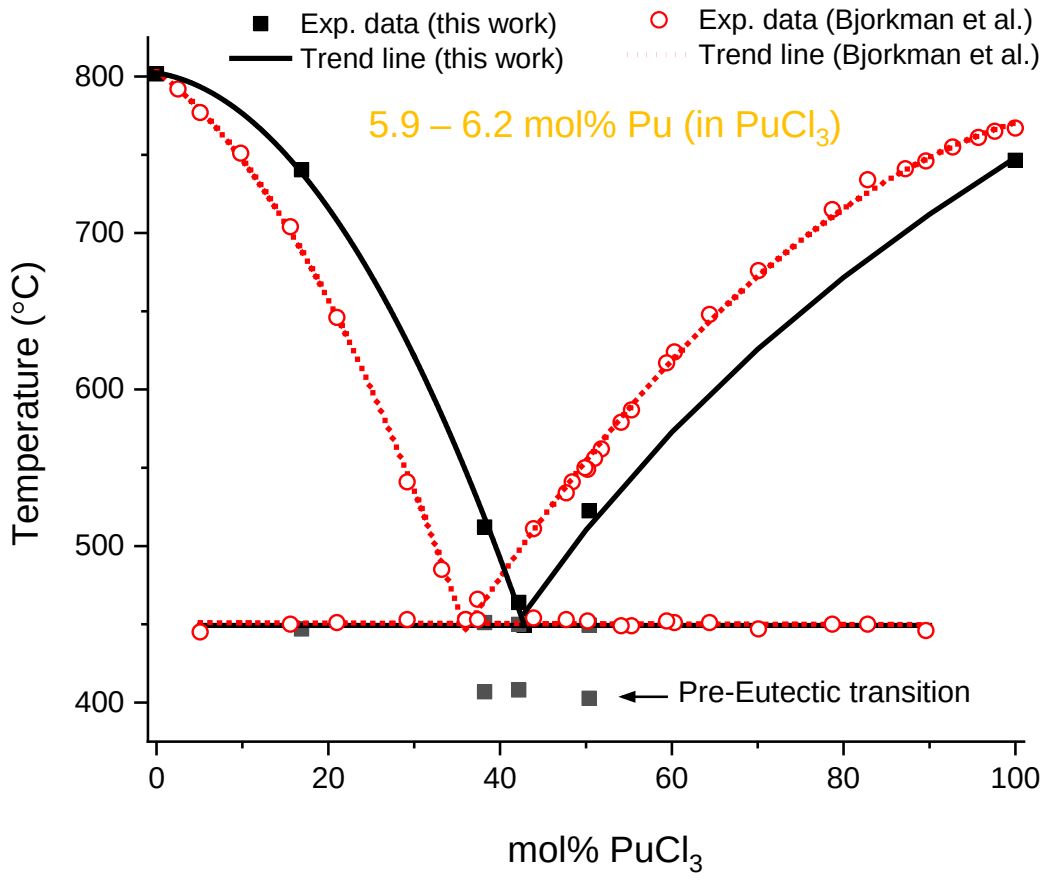
Color of Pu oxidation states

https://www.lanl.gov/orgs/nmt/nmtdo/AQarchive/04spring/XAFS.html

NaCl –$PuCl_3$-Pu Phase Diagram

- Effects of using ~6mol% Pu (in $PuCl_3$) on the NaCl-$PuCl_3$ phase diagram of
  - Shift in the E point from 36 to 43 mol% Pu/$PuCl_3$
  - E transition line still at 449°C
  - NaCl-rich side had increased in liquidus temperature
  - $PuCl_3$-rich side had reduction in liquidus temperature
  - Pre-E transition line at ~406°C, not reported in literature

Changes in chemical state or salt composition (impurities) can affect the liquidus temperature of salt systems




Dr. Karlsson


Dr. Adkins


Dr. Gakhar


Stephen Warmann

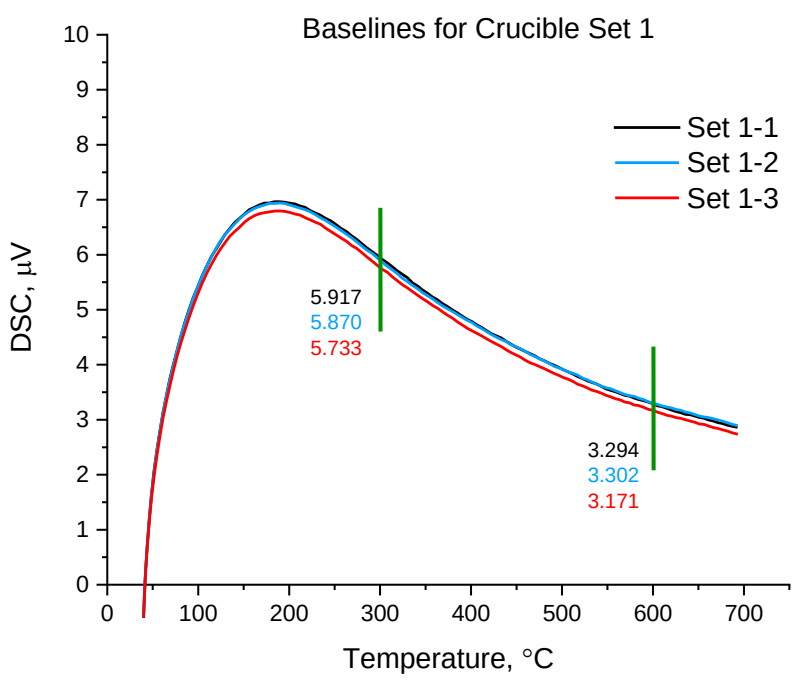
T. Karlsson, C. Adkins, R. Gakhar, J. Newman, S. Monk, S. Warmann. "**Phase Behavior of the Ternary NaCl-$PuCl_3$-Pu Molten Salt**" JNFCWT Vol.21, No.1 (March; 2023)

Heat Capacity Method Development

- What leads to a good heat capacity measurement?
- Where to start?
  - Baseline reproducibility
  - DSC, glassy carbon crucibles
- Experiments done to look at error propagation

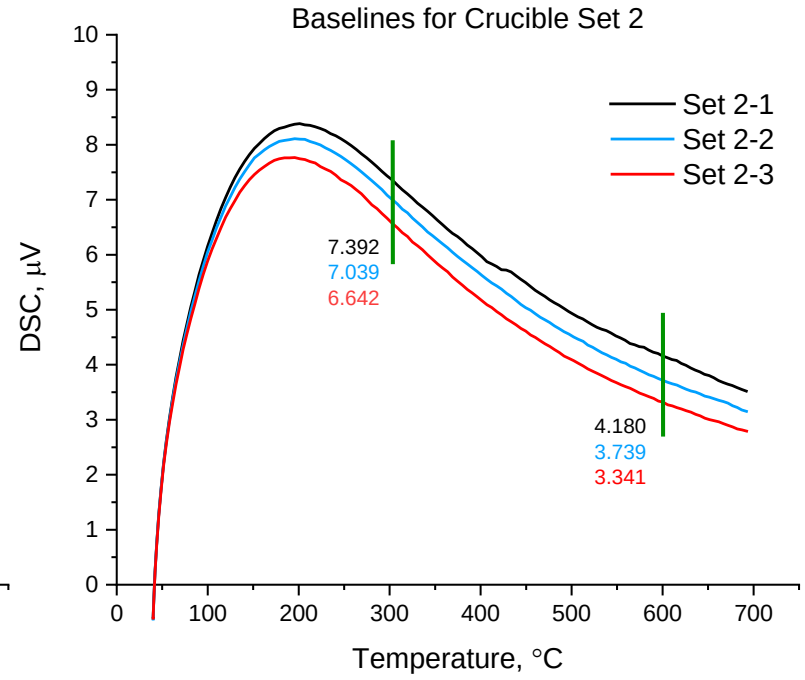
Dr. Robin Roper

Baseline = Reproducible



3.2% RSD @ 300°C
3.8% RSD @ 600°C

Baseline = Not Reproducible



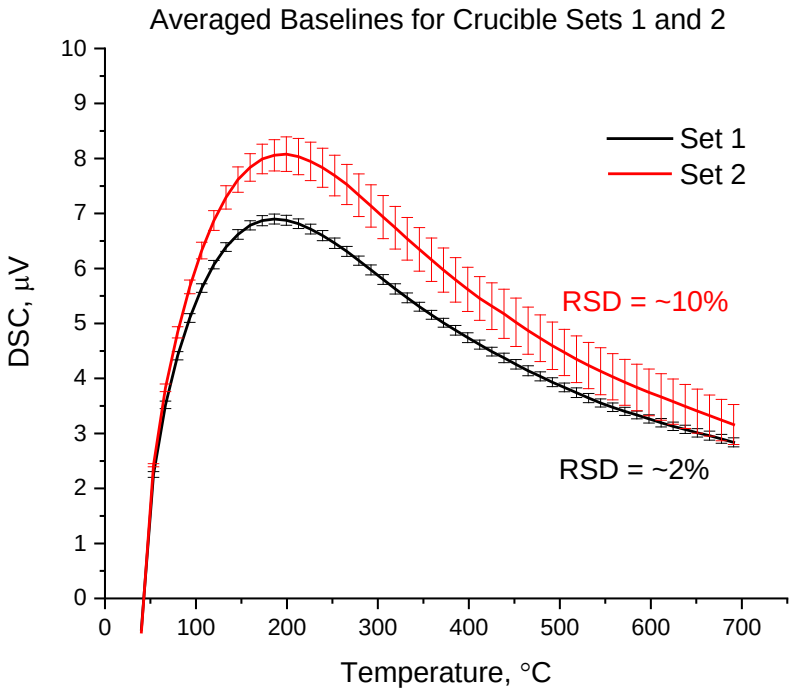
11.3% RSD @ 300°C
25.1% RSD @ 600°C

Heat Capacity Method Development

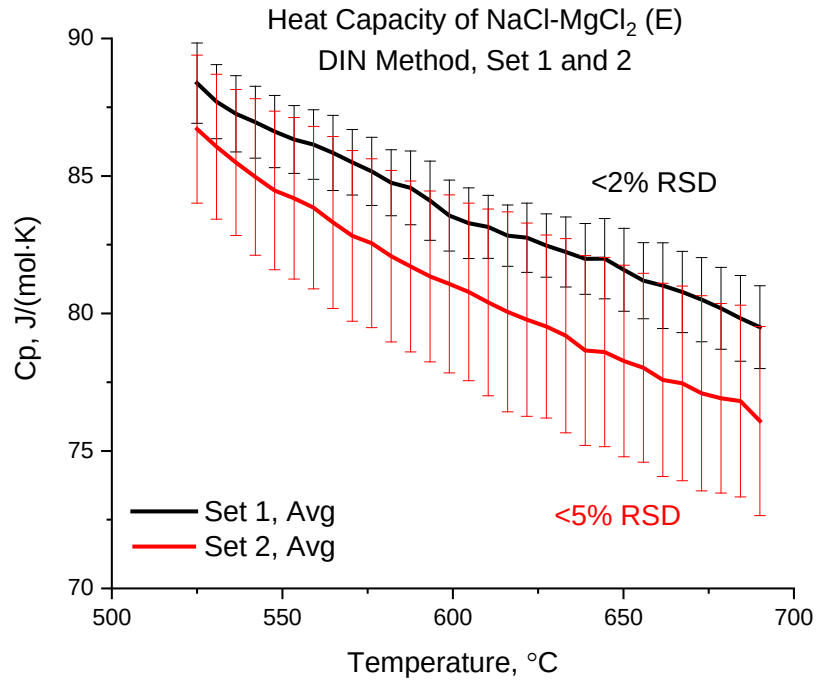
- How does a reproducible or nonreproducible baseline propagate
  - Salt sample
- Sample was heated from 30 to 700°C

Dr. Rocio Rodriguez mentoring interns on thermal property measurements

Baseline Comparison



Sample Comparison



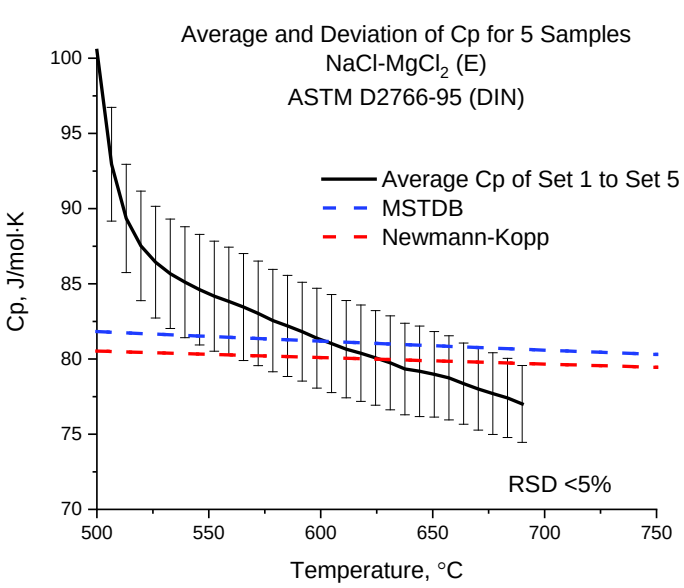
Higher uncertainty for non-reproducible baselines translates to higher uncertainty of measure heat capacity

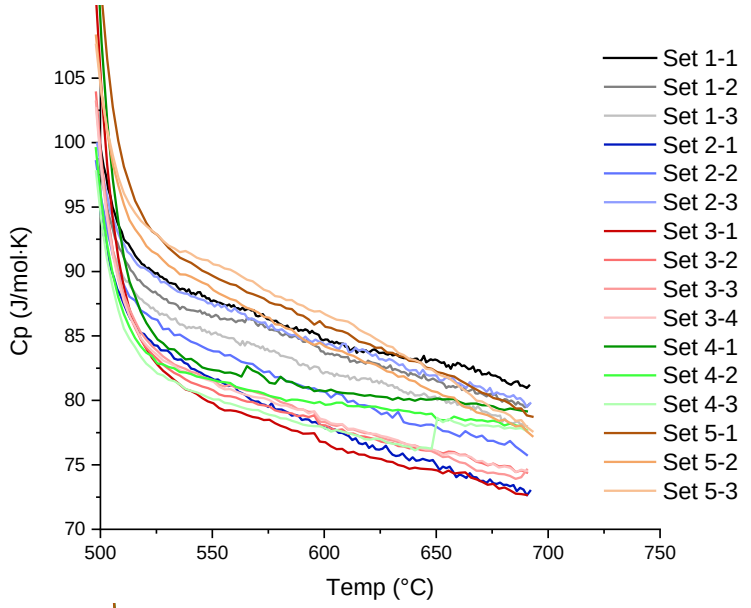
Ratio Method vs ASTM Method

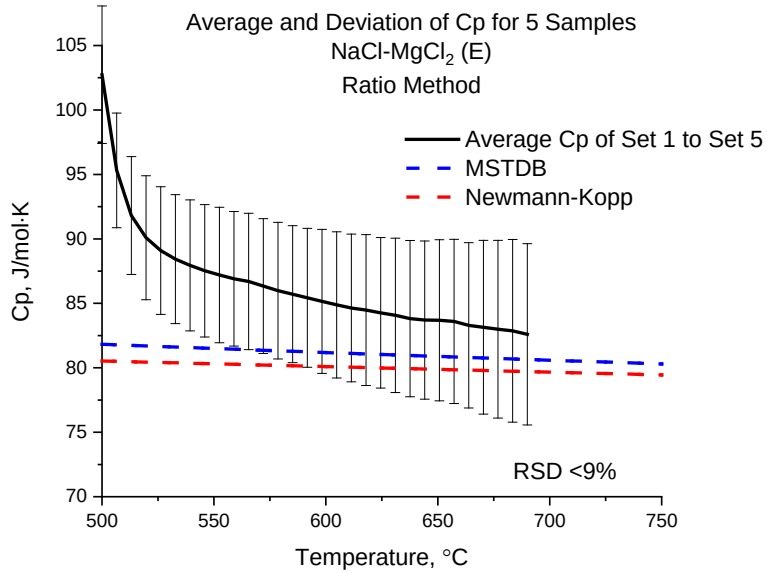
- Using the same data sets
  - Glassy carbon crucibles, lids
  - Eutectic NaCl-$MgCl_2$
  - Sample was heated from 30 to 700°C
- Interested in statistical errors not necessarily precise values

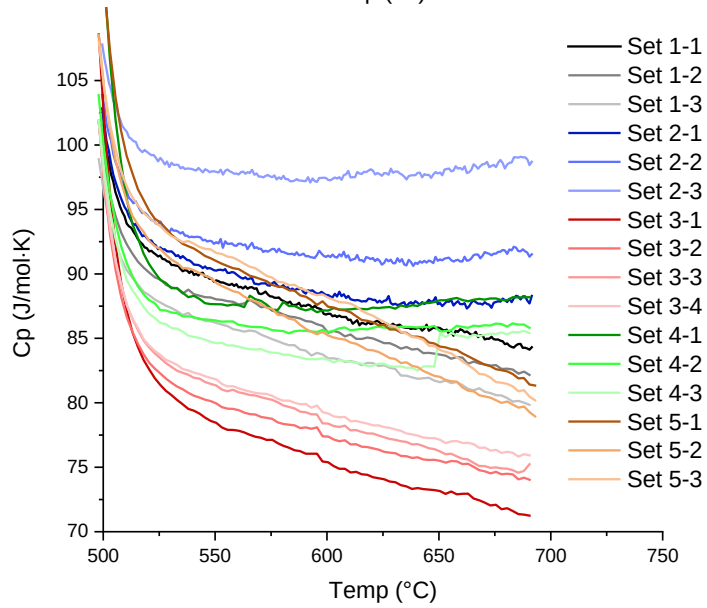
Dr. Robin Roper

Dr. Schorne Pinto









FY23 Activities

Teamwork makes the dream work!

- $NaCl\text{-}UCl_3\text{-}PuCl_3$ (61-30-9) Fuel Salt
  - Amalgamation and homogenization of salts (FMF) ☑
  - Density determination (FMF) ☑
  - Prepare sample for transfer (to ARL) ☑
  - Melting temperature and enthalpy determination (FFGB)
- $NaCl\text{-}PuCl_3$ (64-36) Used Salt (possible corrosion products)
  - Extraction from flow loop (FCF) ☑
  - Transfer samples (ARL, FFGB, FMF) ARL ☑
  - Elemental, isotopic, and impurity analysis (ARL)
  - Melting temperature and enthalpy determination (FFGB)
  - Density determination (FMF)
- Continue mentoring and Method development with early career staff
- Dispose of waste generated in FY22/23

Collaborations

- Conversations, data for mod/sim, lessons learned, method development, etc…
  Lets Talk!!!

Toni.karlsson@inl.gov

- Conferences/Presentation
  - **2022 ORNL MSR Workshop**
    - Thermal Properties and Related Activities at INL (Presentation)
  - **2023 TMS**
    - Dr. Karlsson - Melting Temperature Method for Determining the Concentration of Pu-metal in $PuCl_3$ Salt (Presentation)
    - Dr. Woods - Methodology and Density of $PuCl_3$-NaCl Mixtures
- Papers
  - **Phase Behaviour of the Ternary NaCl-$PuCl_3$-Pu Molten Salt**, Accepted to Journal of Nuclear Fuel Cycle and Waste Technology
  - **Synthesis and Thermophysical Property Determination of NaCl-$PuCl_3$ Salts,** Submitted to Journal of Molecular Liquids

Thank you

Email Address

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