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Variable Gap Thermal Conductivity Measurements at ORNL

Workshop on Measurement and Analysis of Thermochemical & Thermophysical Properties of Molten Salts

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U.S. DEPARTMENT OF ENERGY

Motivation:

- Thermal physical Properties are pivotal for multi-physics reactor codes, for design and NRC benchmarking of molten salt reactors
  - Accurate understanding of properties informs steady-state temperature distributions, thermal responses to transient conditions, expected pressure distributions, thermal efficiency of reactor etc.
- Thermal Conductivity measurements of MSR relevant salts are incredibly challenging:
  - U, Th, and Be bearing salts are reactive/hazardous
  - Direct standard steady state measurement techniques are notoriously sensitive to experimental parameters(heat loads/losses/resistances) in a changing system
    - Corrosion/salt intrusion/hazards makes repeat measurements challenging
  - Indirect measurement/transient techniques rely on knowledge of other parameters (density/heat capacity) to calculate thermal conductivity

Main Driver for Property Characterization: MSTDB

- The Molten Salt Thermal Property Database (MSTDB) is an effort funded by the DOE-NE funded Molten Salt Reactor (MSR) Campaign and the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program.

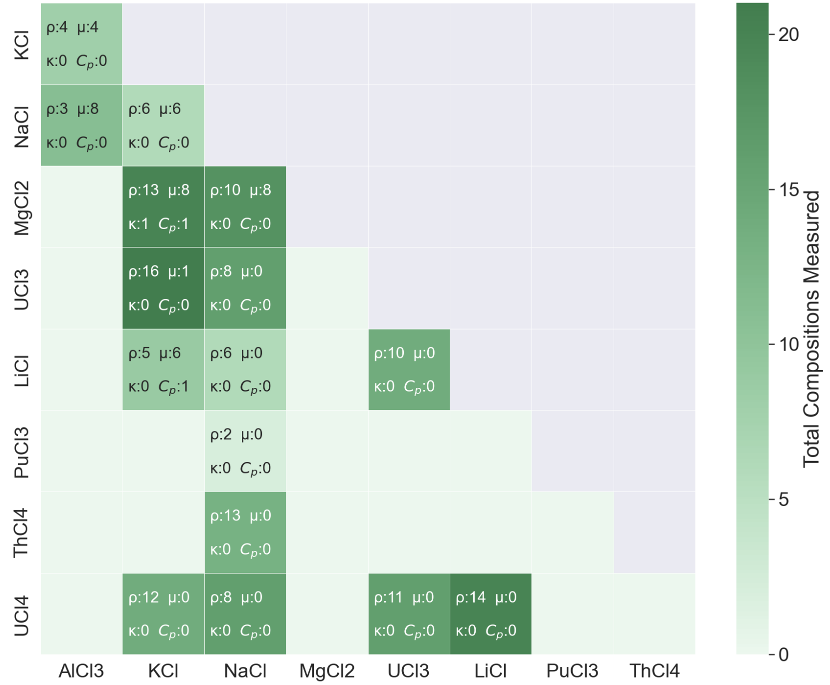
- Thermal Conductivity has the least amount of entries in MSTDB-TP
  - Almost no actinide/Be data.

- MSTDB-TC is managed by UoSC, MSTDB-TP is managed by ORNL.

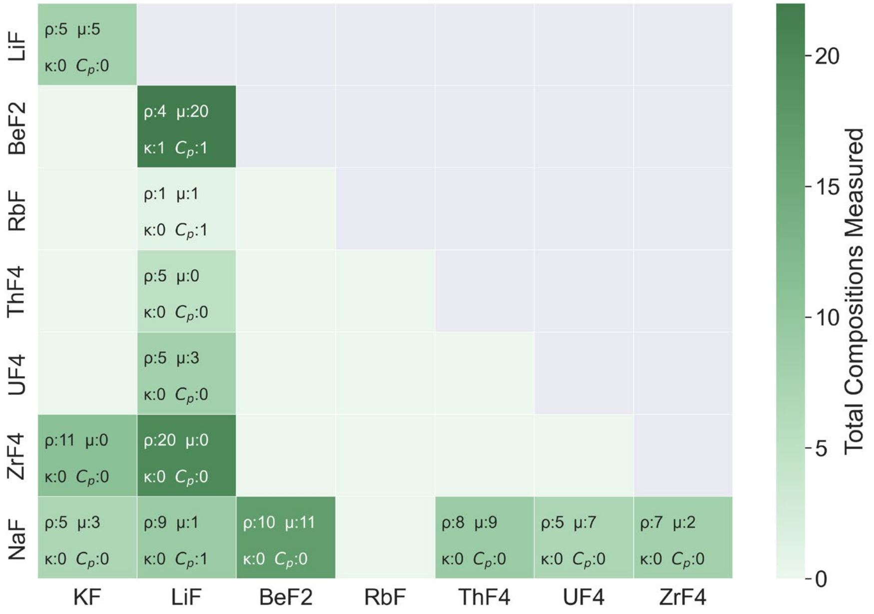
NEAMS
Nuclear Energy Advanced Modeling and Simulation

Molten Salt Reactor
PROGRAM

Chloride Pseudo-Binary Characterizations

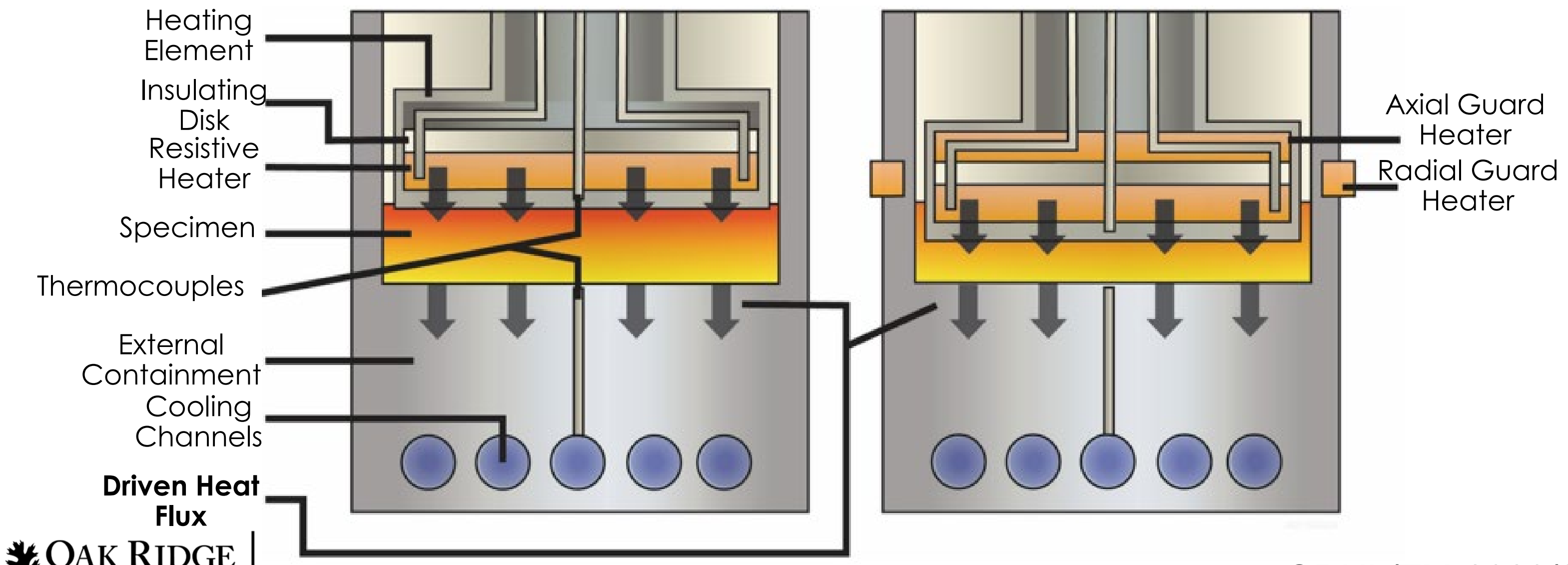


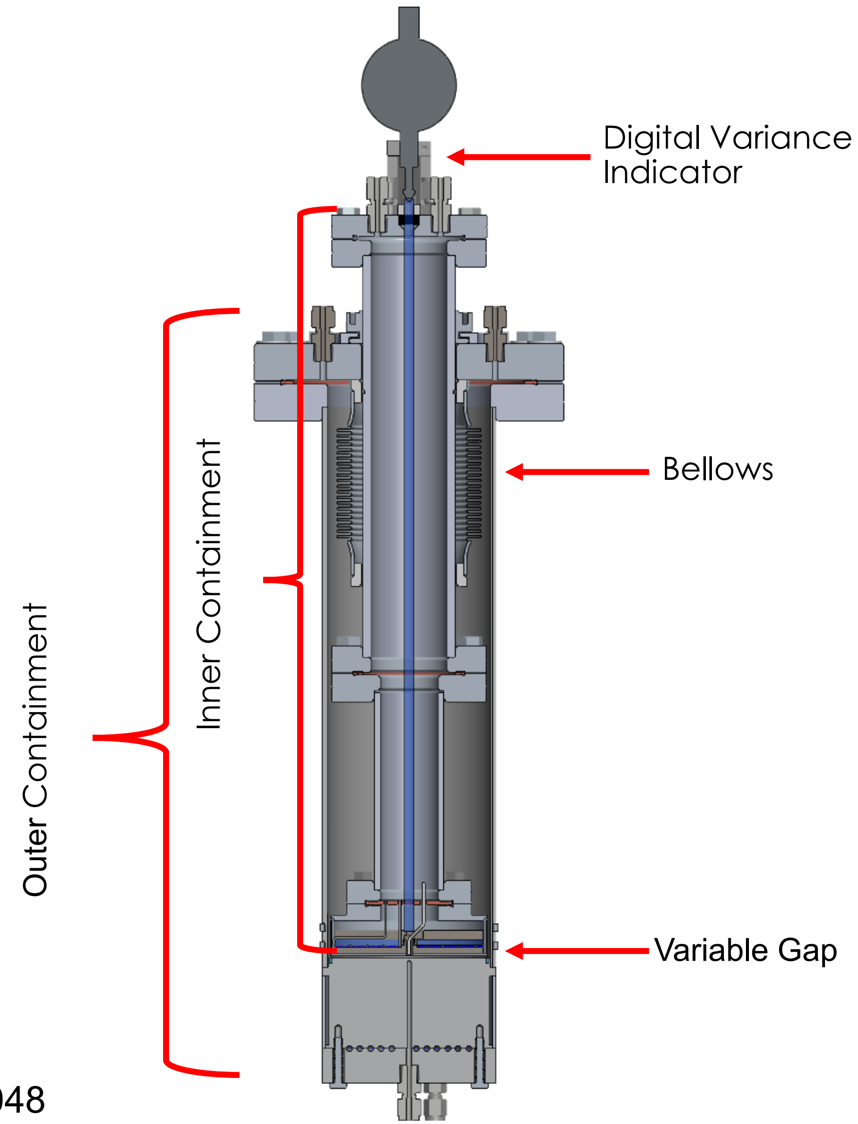
Fluoride Pseudo-Binary Characterizations



ORNL Variable Gap System:

- Steady-State direct thermal conductivity measurement.
- Two sections:
  - Inner Containment contains heater/thermocouples
  - Outer Containment contains salt specimen.
- Inner containment can move vertically to achieve different specimen lengths or "gaps"
- Small gap sizes and large area allows for 1D heat transfer to be used with temperature differences to back out the thermal conductivity

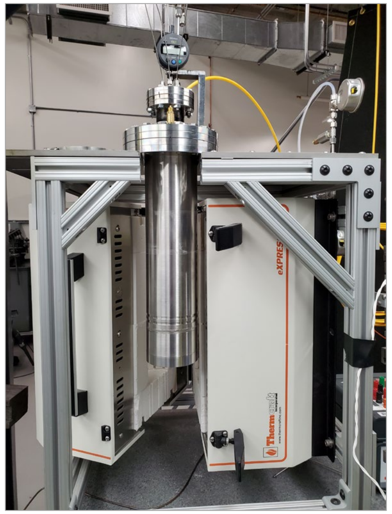


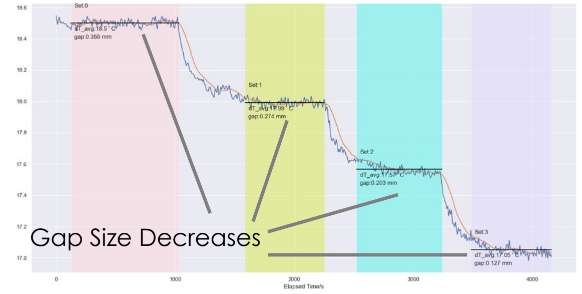


Calibrate with Helium at different experimental parameters

→ Load outer containment with purified salt in glovebox

→ Seal off outer containment to inner containment and transport to furnace stand





→ Turn on internal heaters and furnace to appropriate testing temperature, and let system come to thermal equilibrium over 24 hours

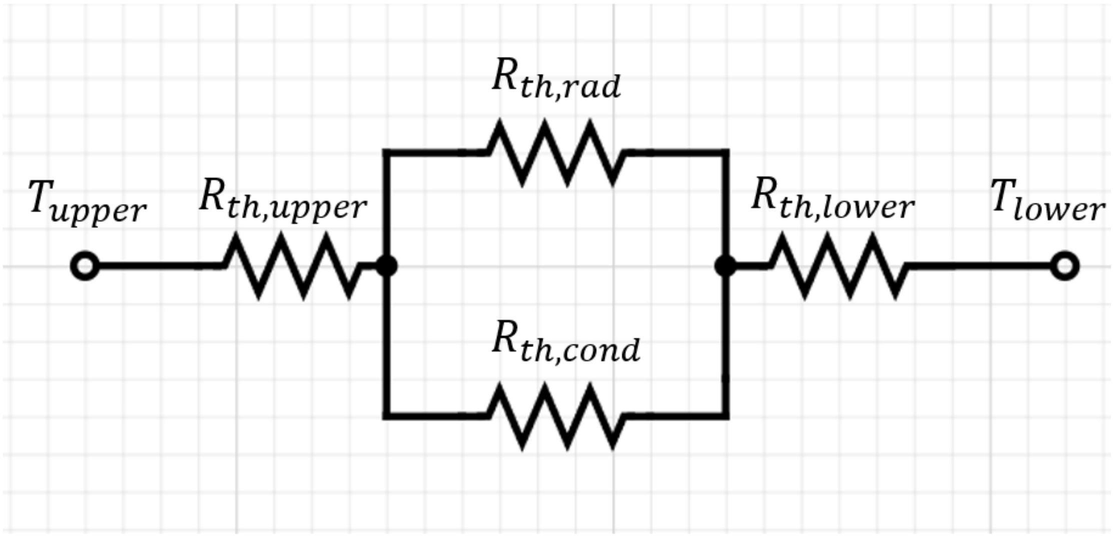
→ Calculate temperature drop through gap at varying gap size

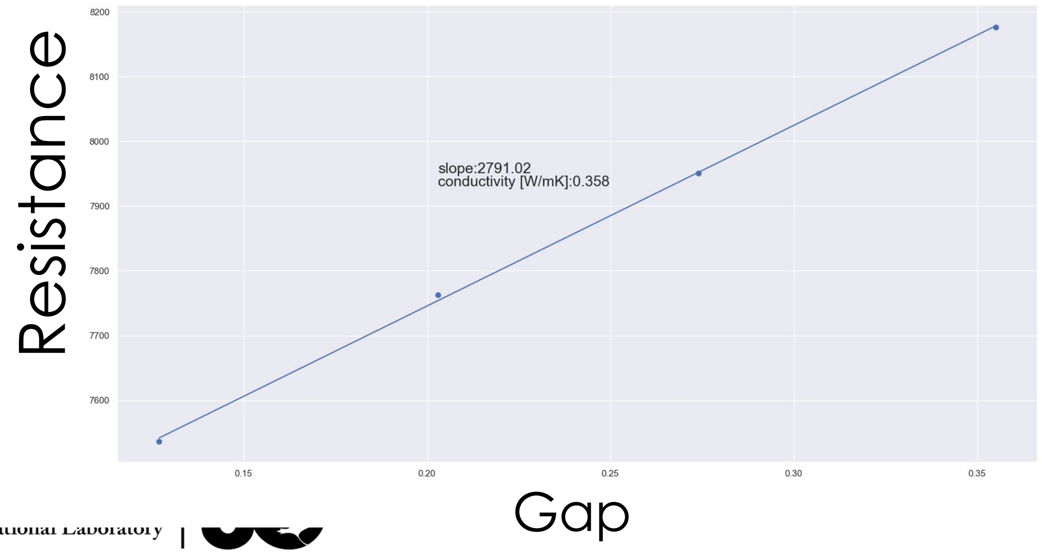
→ Need more trials?

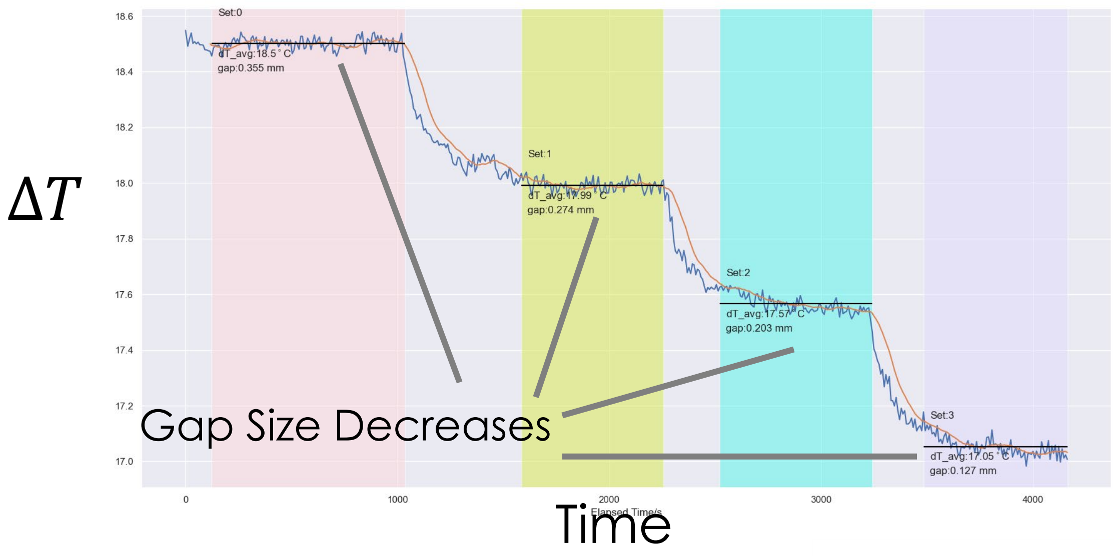
- Reset Gap → Calculate temperature drop through gap at varying gap size
- No → New Temperature
  - Set new furnace temperature → Turn on internal heaters and furnace to appropriate testing temperature, and let system come to thermal equilibrium over 24 hours
  - No → Turn off furnace and cool.

Variable Gap Working Principles: Electrical Circuit analog

- Fixed resistance: containment components
- Gap dependent resistance: Salt
- $\frac{\Delta T}{q''} = R_{Th,Fixed} + \frac{1}{\kappa_{cond}+\kappa_{rad}} x$
- Measuring the thermal resistance at multiple gaps and taking the slope of the line removes fixed resistances







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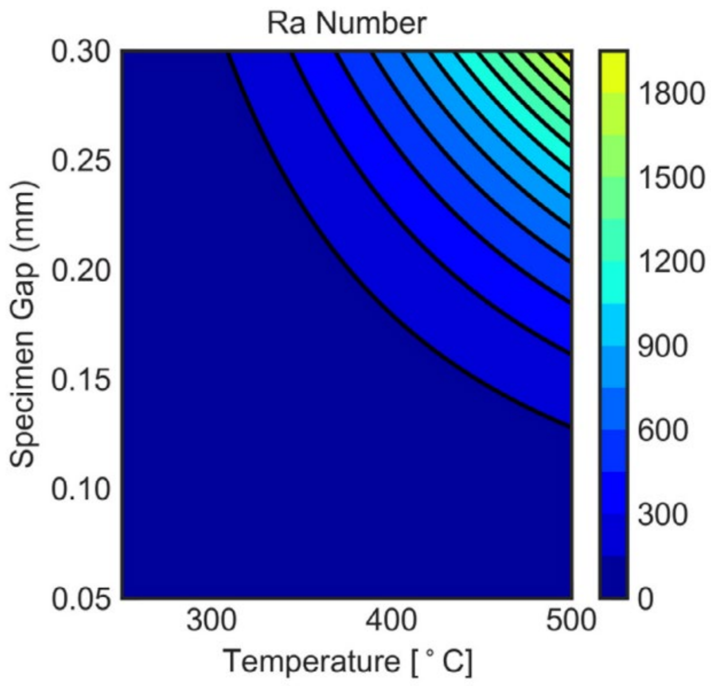
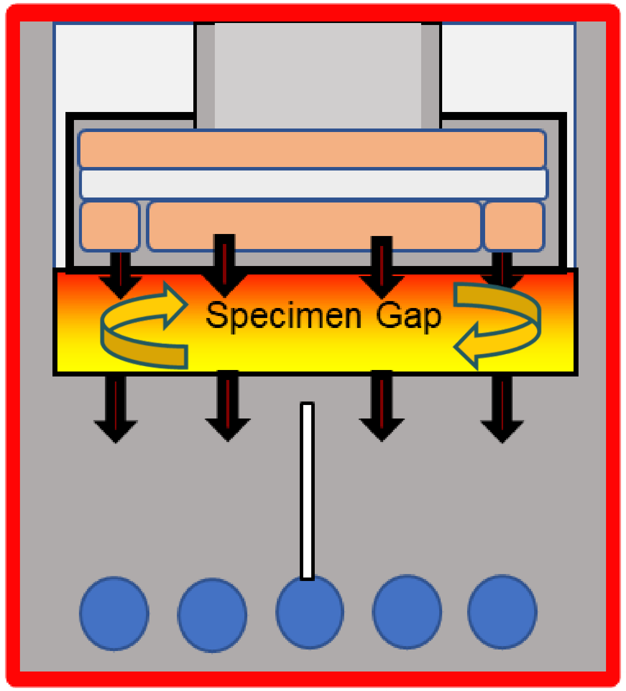
Convection Effects:

- For a fluid under temperature difference, natural convection needs to be considered.
- Very thin gap (<0.3mm) heavily reduces these effects.
- Measured by the Rayleigh number:
  - $Ra = \frac{g\alpha\Delta T x^3 C_p \rho^2}{\kappa\eta}$
- A conservative criterion of <1700 was given to ensure purely conductive flow.
- For gap size, targeted ΔT, and salt properties, convection can be assumed negligible in majority of salt systems measured.

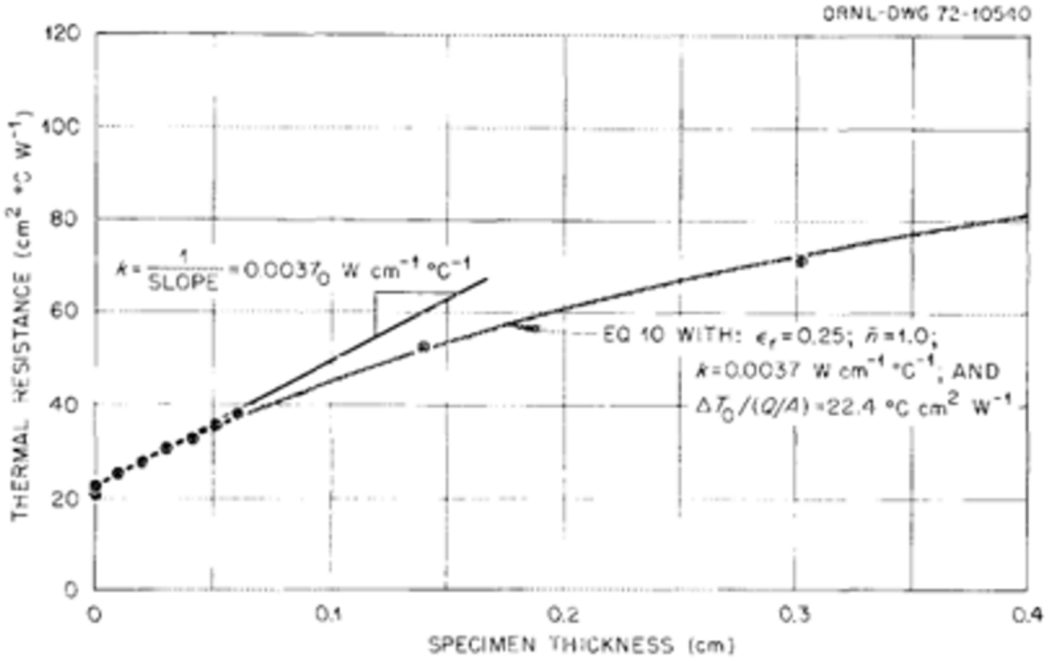




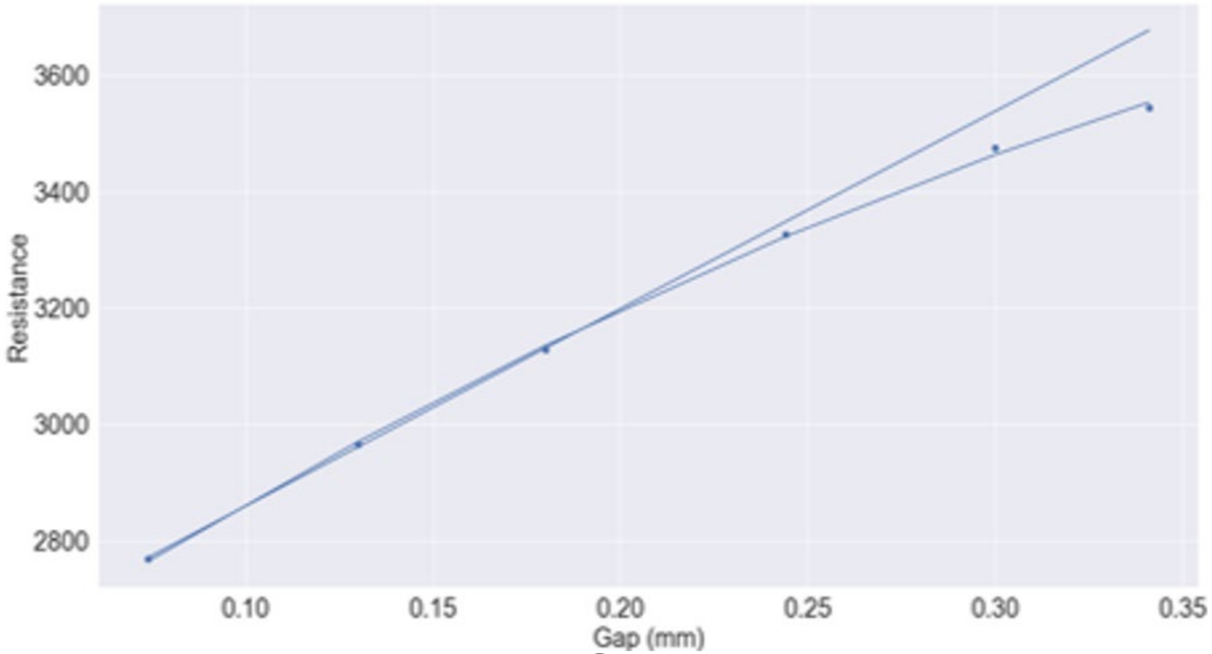
Gallagher R. et al. Design and performance of a variable gap system for thermal conductivity measurements of high temperature, corrosive, and reactive fluids, International Journal of Heat and Mass Transfer, Volume 192, 2022,

Radiative Heat Transfer Effects

- At high temperatures, radiative heat transfer becomes a concern.
- Salt optical properties are not well known (considered optically thin but participating medium)
- For salt, gap sizes are small enough that this portion becomes negligible.
- Experimentally its effects can be seen by deviation from non-linear trend. If present, choose lower gap sizes to fit to.



Resistance

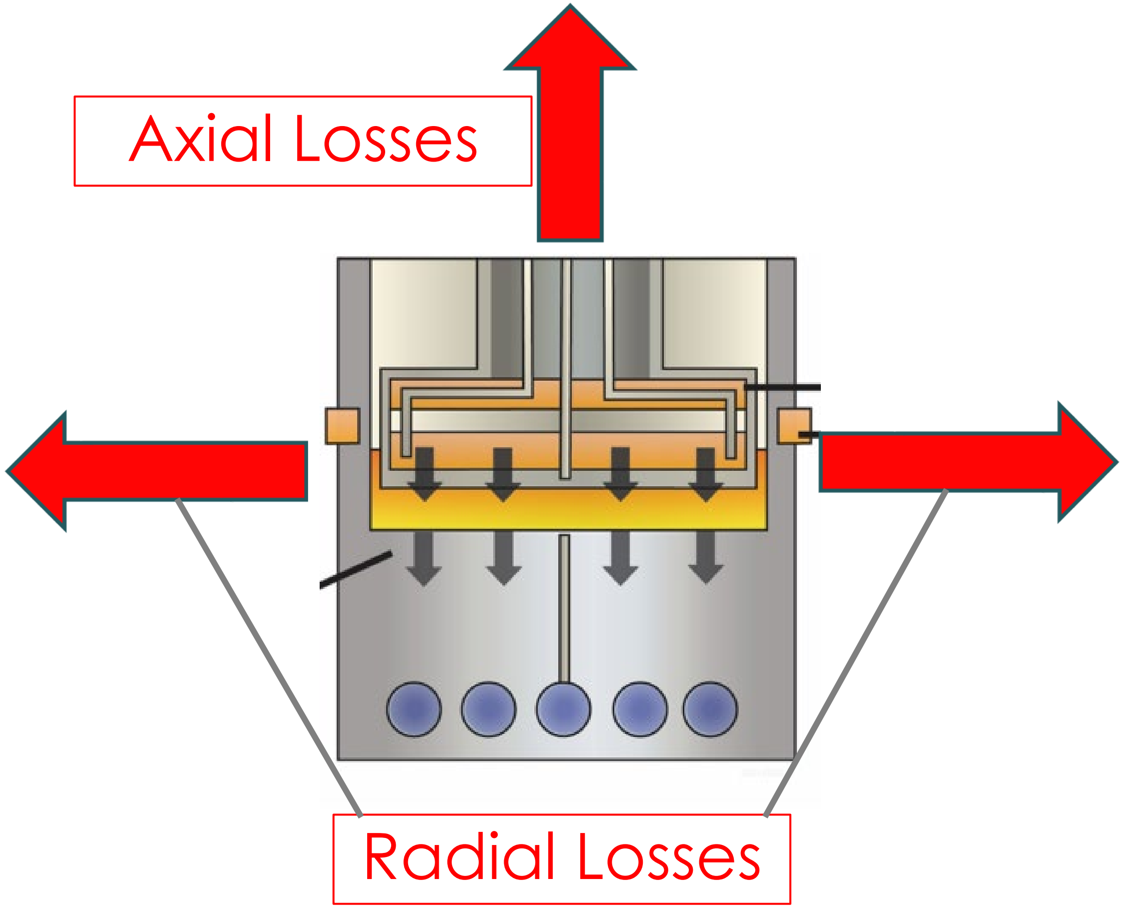


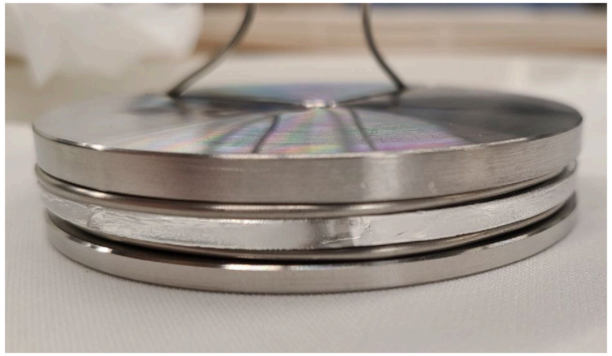
Gap

J. Cooke. Development of the variable-gap technique for measuring the thermal conductivity of fluoride salt mixtures. Technical report, OAK RIDGE NATIONAL LAB., TENN., 1973.

Heat Shunting Effects

- Two resistive heaters ("guard" and "main") apply a heat load to the salt, with active air cooling below the sample.
- While the power output of these heaters can be determined by $P = IV$ through the actively heating length of wire, the amount of this power truly driven through the salt is difficult to calculate.
- The % power lost through axially and radially is called heat shunting.
- This heat shunting effect can change based on furnace temperature/working fluid properties/active cooling/insulation etc.





Calibration and Correction Approach

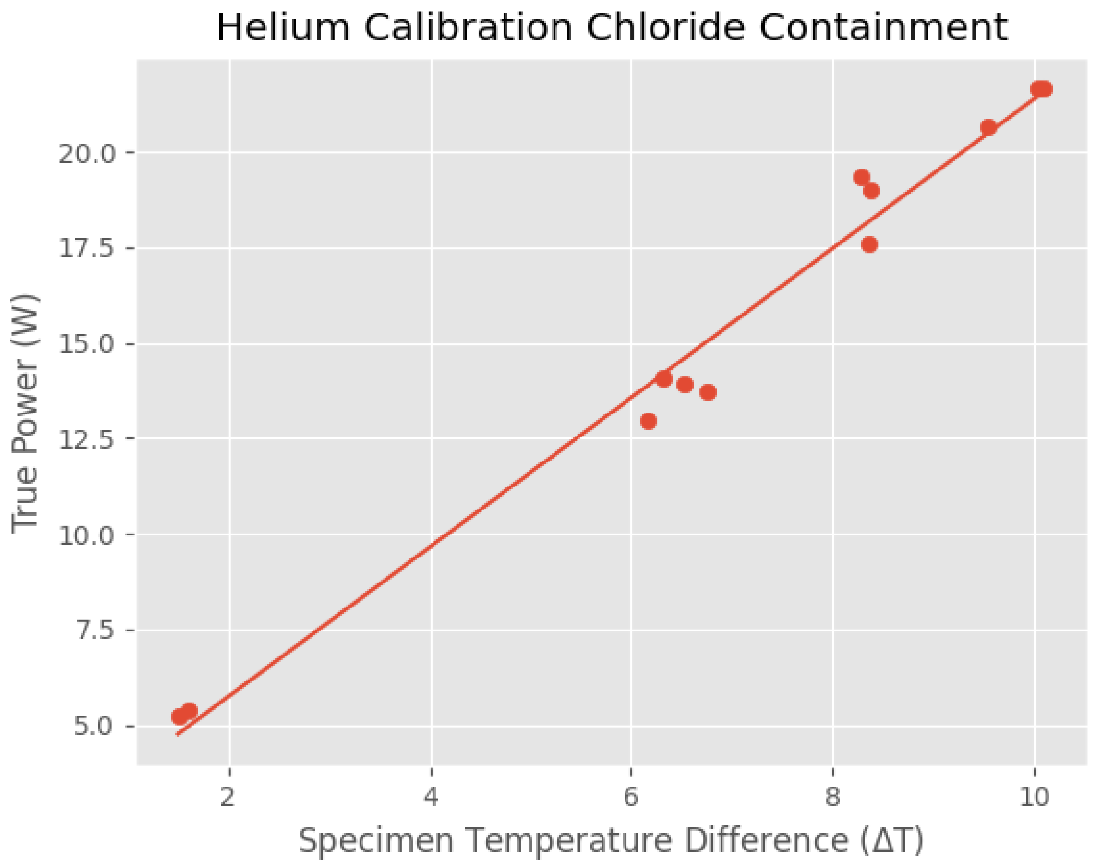
- Major effort has been put forth to fully characterize the power input (Q) for the system
- Determine the true value of Q for varied experimental variables can be challenging due to multiple heat sources and changing experimental conditions:
  - Main power heater
  - Guard power heater
  - Axial power heater
  - Furnace temperature
  - Cooling Air Pressure
  - Material Degradation
  - Insulation
- Therefore, a parametric study was done with a known fluid (He) to determine the effects of these conditions on the experiment

Calibration with Helium Concerns

- To ensure that a proper calibration factor can be applied for the heat shunting, the most important factor is matching the two experiments as closely as possible
- Salt Vs Helium differences
  - Helium is transparent medium vs participating but optically thin for salt (radiative)
    - Low Gap sizes makes negligible.
  - Ra $\rightarrow 0$ for gap sizes chosen for Helium, slightly higher Ra for salt (1400s)
    - Still considered negligible
  - Containment backfill originally with Ar for salt. Significantly lower thermal losses!
    - Both experiments will be done with Helium to keep similarities even if thermal losses are higher

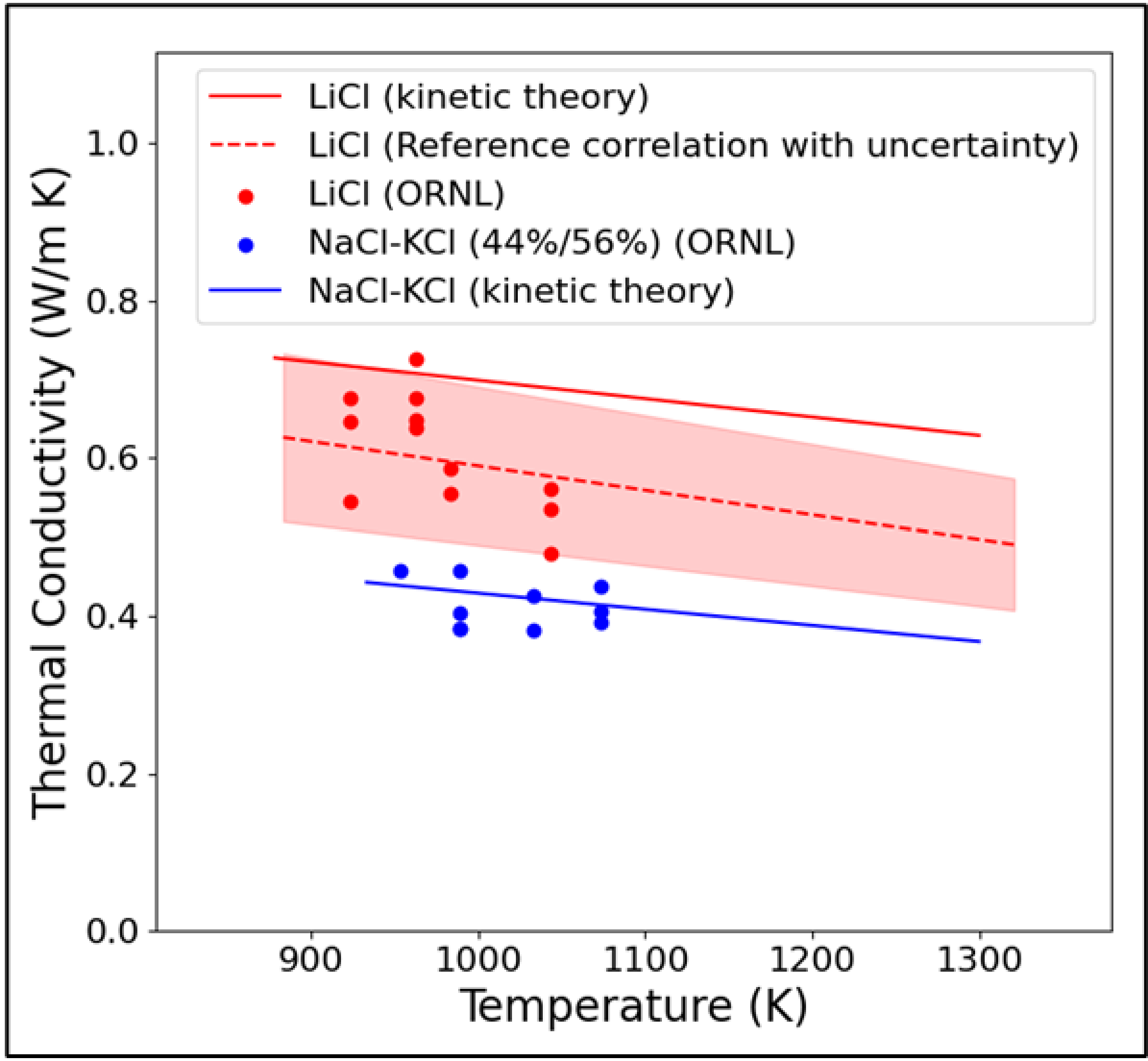
Too much variability from experiment to experiment…

- Need an anchor point:
  - ΔT
- It was found that across different experimental configurations that matching ΔT resulted in the same power correction factor (ie specific heat flux across sample).
- Therefore, the system was calibrated using constant electrical power across a variety of cooling/furnace temperatures using Helium.



Benchmarking New Method: LiCl/ NaCl-KCl Measurements

- To benchmark the new calibration method, LiCl/NaCl-KCl (44-56) salt compositions were measured
- Data shows expected negative trend with temperature
- Trends follow kinetic theory well [1], and reference correlations in MSTDB [2].



[1] Gheribi et al 2024 https://doi.org/10.1016/j.molliq.2024.125239.
[2] Nagasaka, Y et al 1992. https://doi.org/10.1007/BF00501941.

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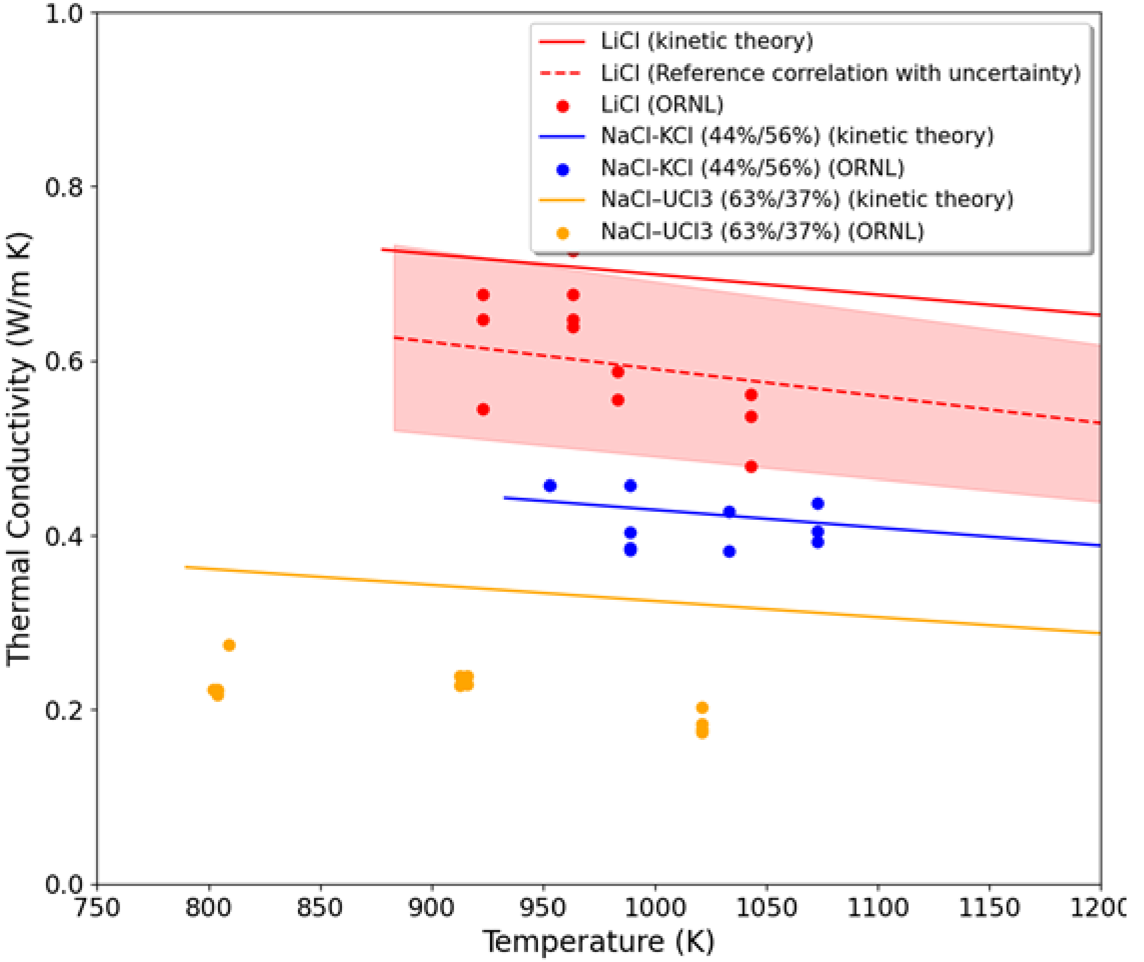
Actinide Measurement (UCl3-NaCl)

- UCl3-NaCl (37/63)
  - Synthesized by Bill Phillips (INL)
- UCl3-NaCl shows expected negative trend
- Deviates from kinetic theory in valuation
  - Actinides have complicated structures/parameters less known [1]
- Potential volatilization of the salt



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Kinetic Theory Actinide Deviations

- Compositional Mass Variation
  - Performs well with ions of similar molar mass. Struggles with high disparity (ie actinides)[1]
- Ionic Complexity
  - Production of more complex ions causes kinetic theory to deviate

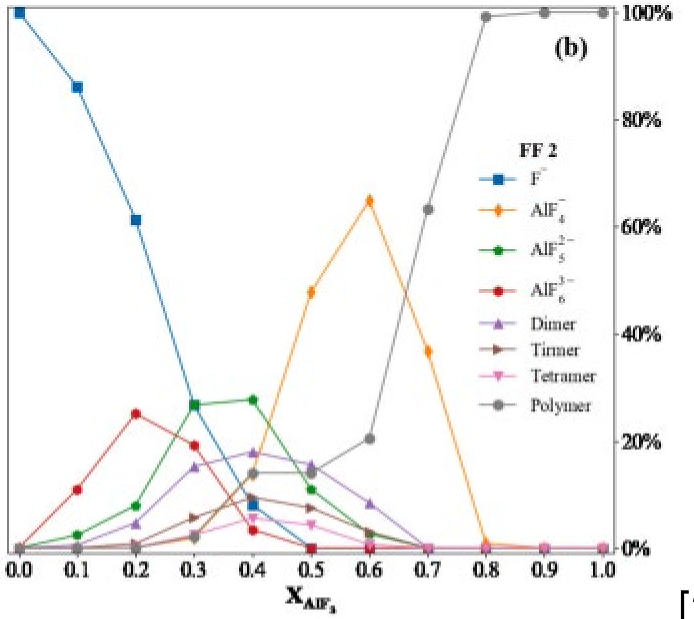
$$\lambda(\boldsymbol{X},T) = \lambda_\sigma(\boldsymbol{X},T)\left[1 - \delta_M^\lambda(\boldsymbol{X},T)\right] \text{ [2]}$$

$$g_{mass} = \sum_{i=0}^{N} X_i * \left[\frac{m_i}{\overline{m}} - 1\right]^2$$

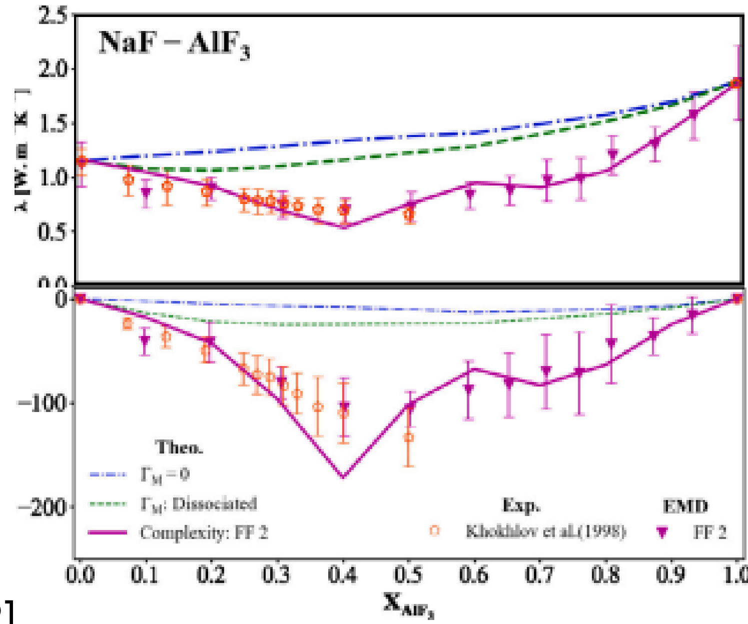
FLiBe → $g = 0.09$

FLiBe+2%UF_4 → $g = 1.45$

Ion Formation



Ion Complexity Vs Deviation



[2]

[1] Bobrova, K. et al 2023.https://doi.org/10.1134/S0036029523020039.
[2] Gheribi et al 2024 https://doi.org/10.1016/j.molliq.2024.125239.

Sources of uncertainty in the variable gap system

- Temperature Measurement:
  - Thermocouple placement and measurement accuracy
- Gap measurement
- Cooling air pressure
- Helium calibration vs salt measurement matching
- Bubble formation in Salt
- Salt degradation/corrosion products

Future Work

- Post Analysis of salt
  - Corrosion products
  - Salt samples taken from different points in containment
  - Confirm composition measured
- Further Measurement of actinide salts
  - UCl_3-NaCl (Compositional)
    - Additional species can be added
  - NaF-UF_4 Eutectic (PNNL)
  - NaF-KF-UF_4 (57/16/27) (Virginia Tech)
- Further reduction in uncertainty in optimization of setup
  - Streamline calibration

Questions?

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