U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Molten Salt Reactor PROGRAM

Molten Salt Thermal Conductivity and Viscosity Characterization at ORNL

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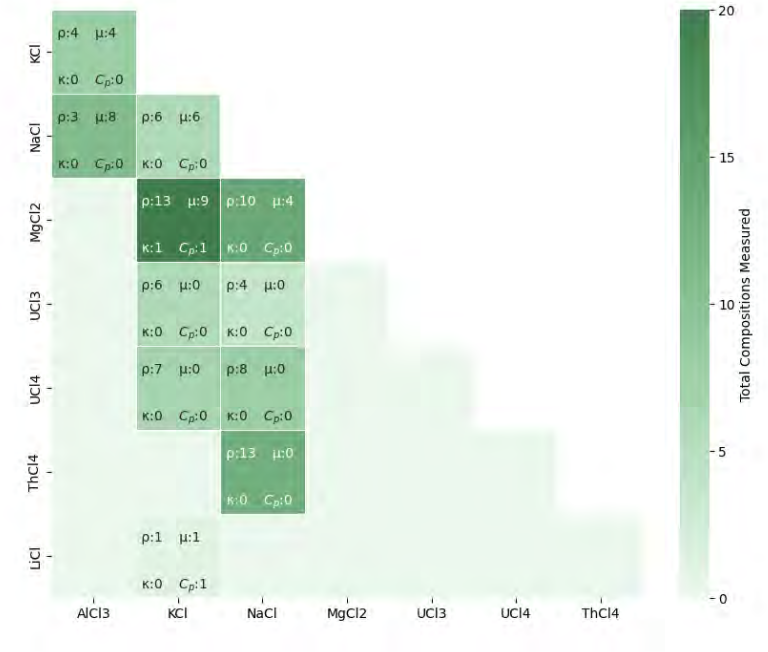
Annual MSR Campaign Review Meeting 2-4 May 2023

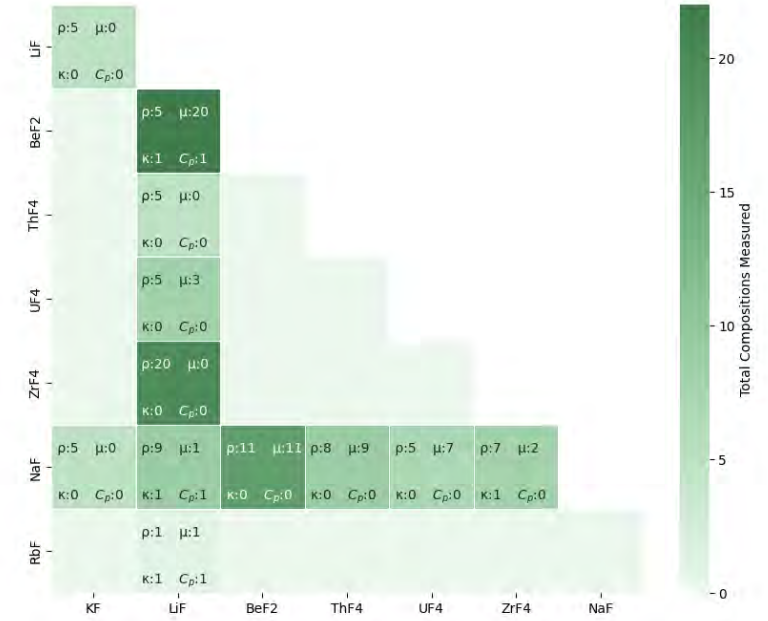
Motivation

- A precise understanding of thermophysical properties of molten salts in MSRs is necessary for developing an accurate understanding of nuclear reactor thermal hydraulics
  - Necessary for understanding the steady-state temperature distribution, thermal response to transient conditions, expected pressure conditions, thermal efficiency of the reactor, etc.
- Developing this understanding for MSR relevant salts is challenging, because:
  - MSRs are typically interested in pseudo-ternary+ mixtures
  - These salts may bear U, Th, and Be
  - Corrosion and fission products may be introduced over the core/reactor lifetime

Chloride Pseudo-Binary Characterizations

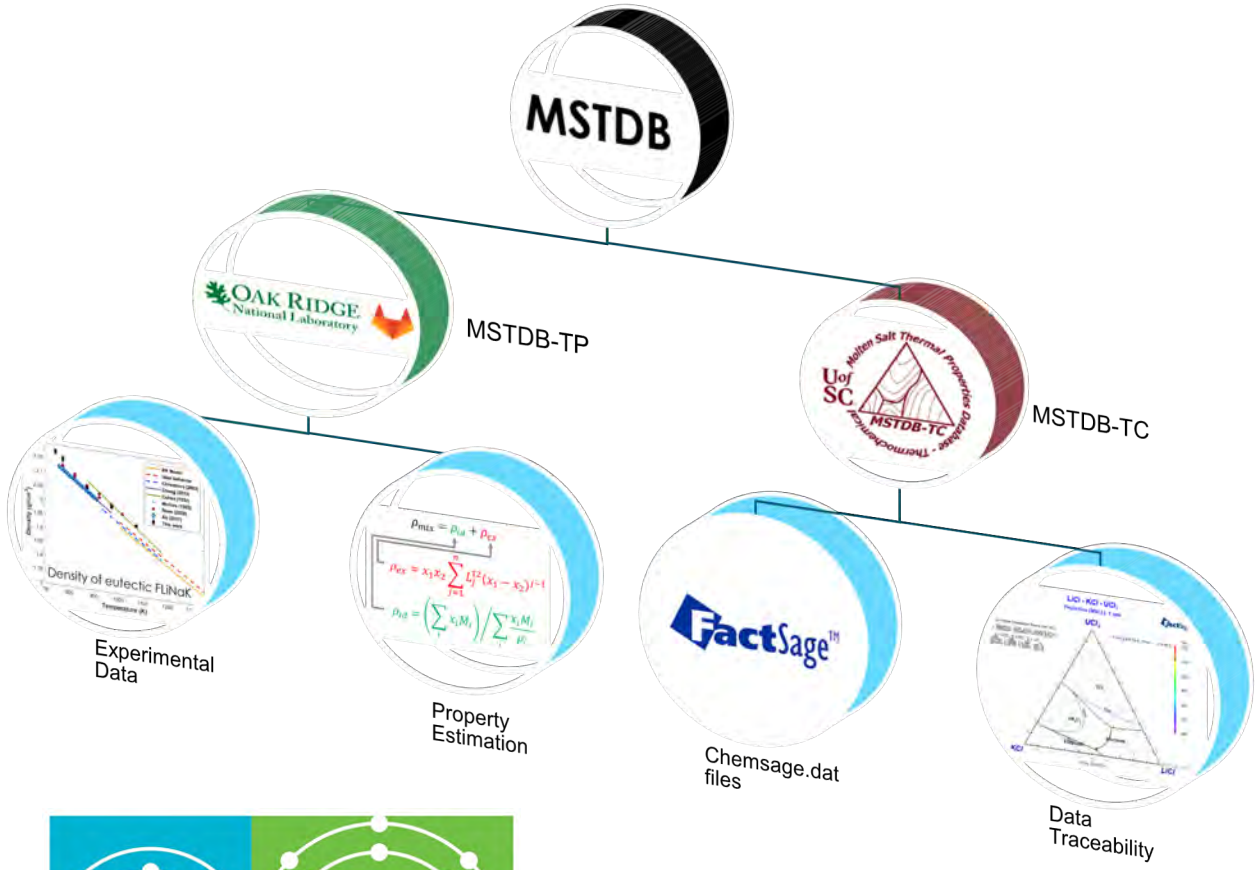


Fluoride Pseudo-Binary Characterizations



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.
- The goal of the MSTDB is to provide thermochemical and thermophysical characterizations of molten salt compounds and mixtures which are relevant to the nuclear industry
- MSTDB-TC is managed by UoSC, MSTDB-TP is managed by ORNL.

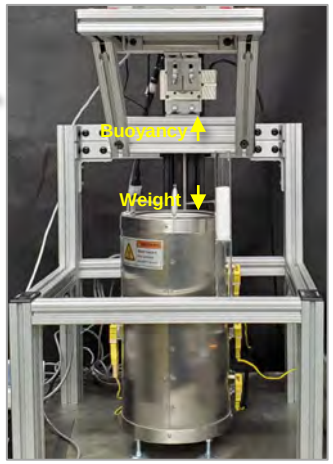


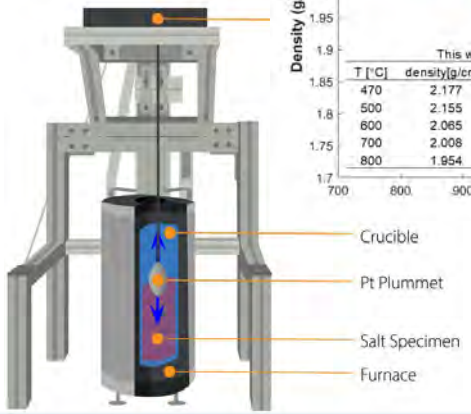
NEAMS
Nuclear Energy Advanced Modeling and Simulation

Molten Salt Reactor
PROGRAM

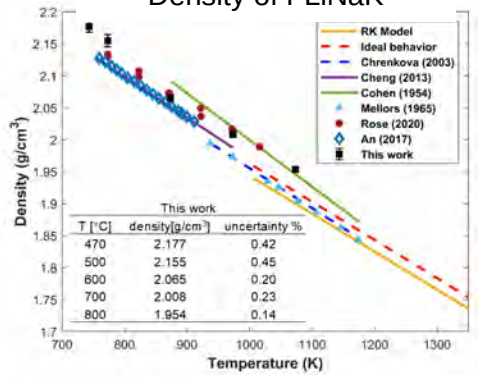
ORNL's Thermophysical Property Systems FY23 measurement focus – driven by MSTDB-TP

Density





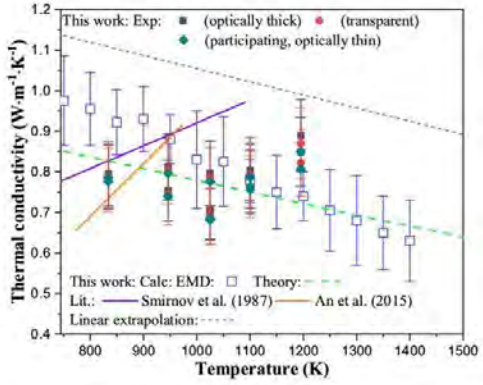
Density of FLiNaK

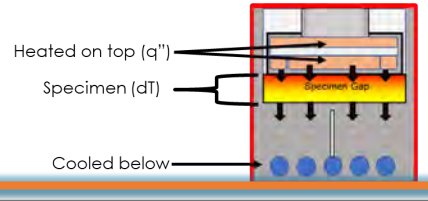


This work

T [°C]	density[g/cm³]	uncertainty %
470	2.177	0.42
500	2.155	0.45
600	2.065	0.20
700	2.008	0.23
800	1.954	0.14

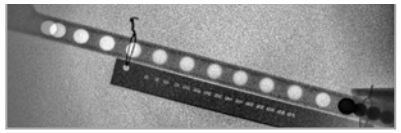
Thermal Conductivity

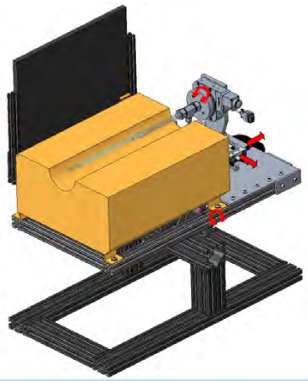


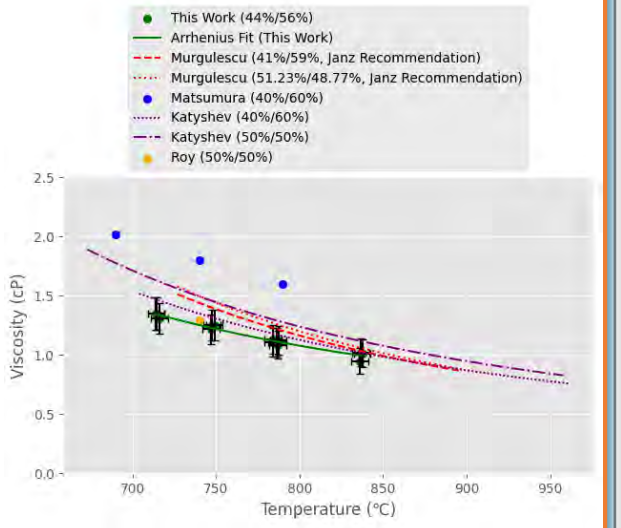


Viscosity

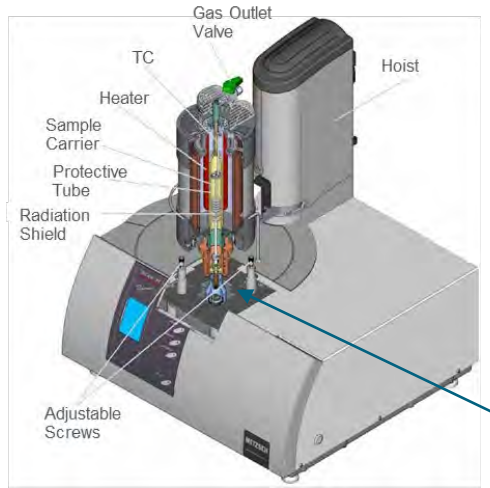
Radiograph of ball decent



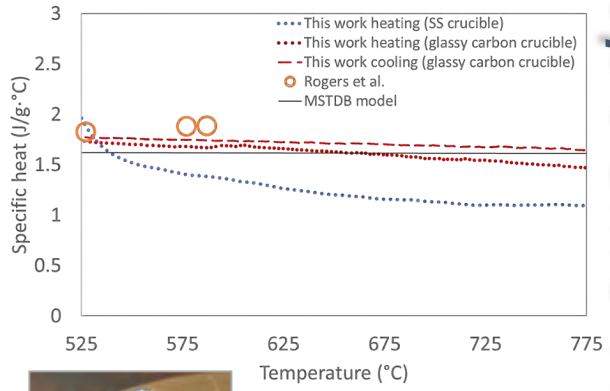




Specific Heat

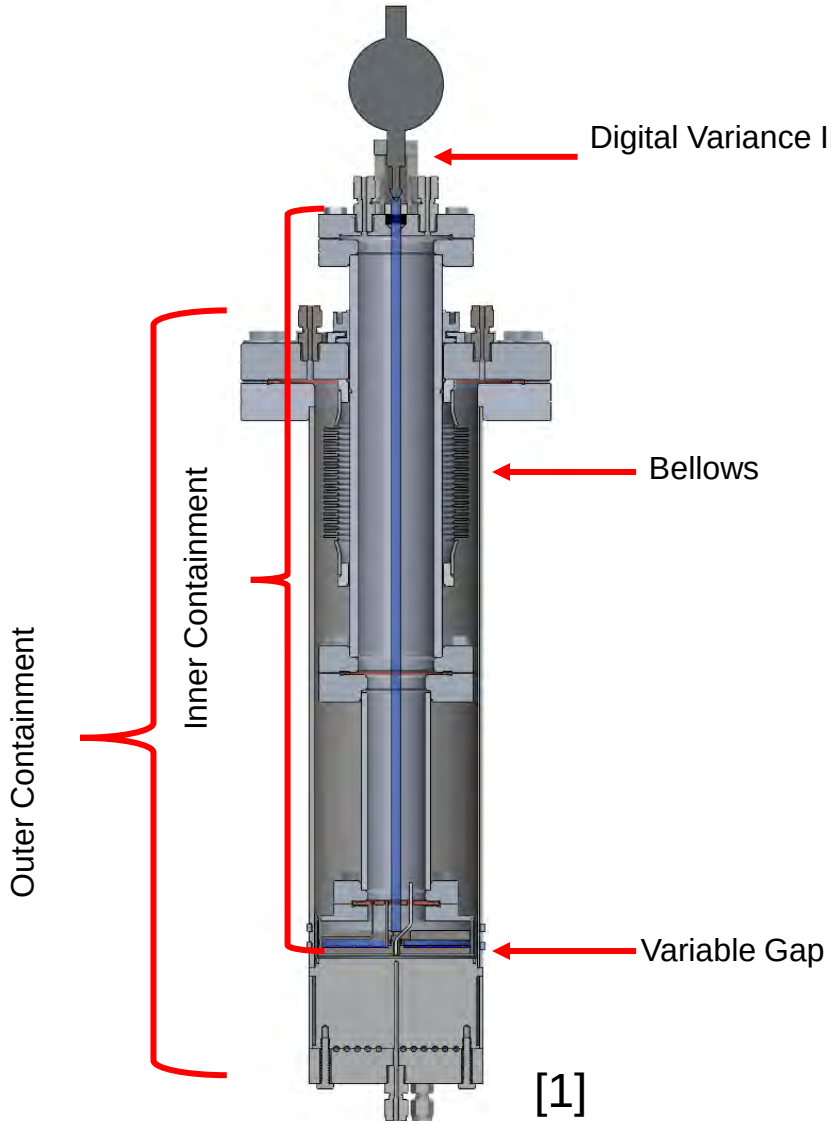


FLiNaK melt



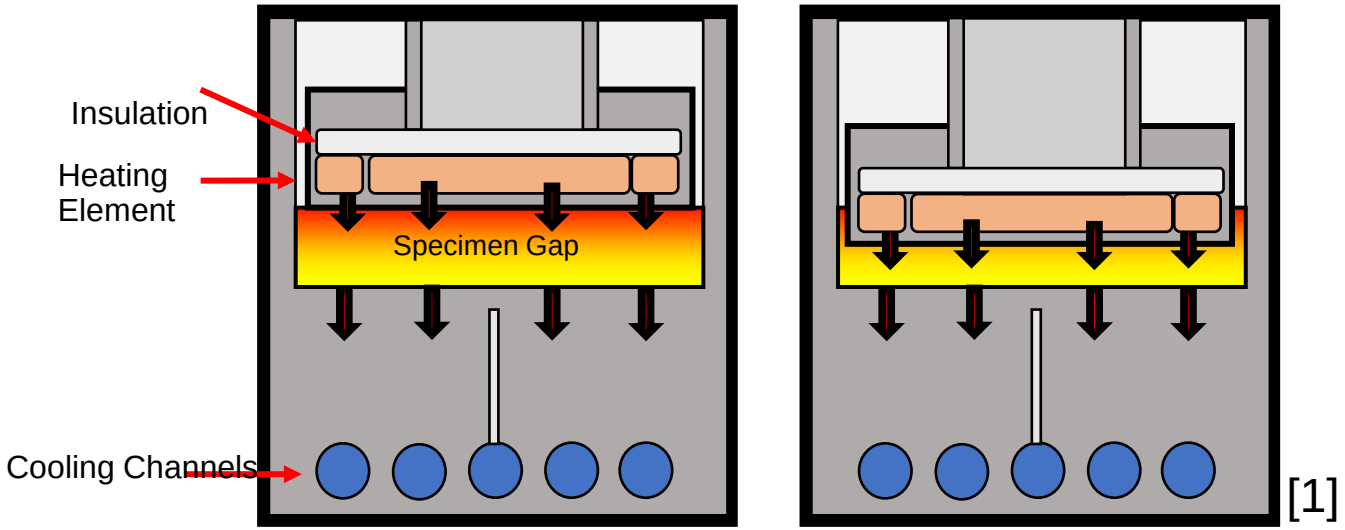
E-beam welded crucibles

FY2023 System Focus 1: Thermal Conductivity



[1]

- This system is based on the variable gap technique
- There is an inner and outer containment
  - Specimen is stored within the outer containment
  - Heating elements are within the inner containment
- A temperature difference is driven across the specimen gap, and based on 1D heat transfer we can back out thermal conductivity
- Equations are modified to account for radiant heat transfer
- Gap is <0.3 mm, making convective heat transfer negligible
- Since this is a differential approach, extra thermal resistances which affect the temperature difference can be effectively canceled out



[1]

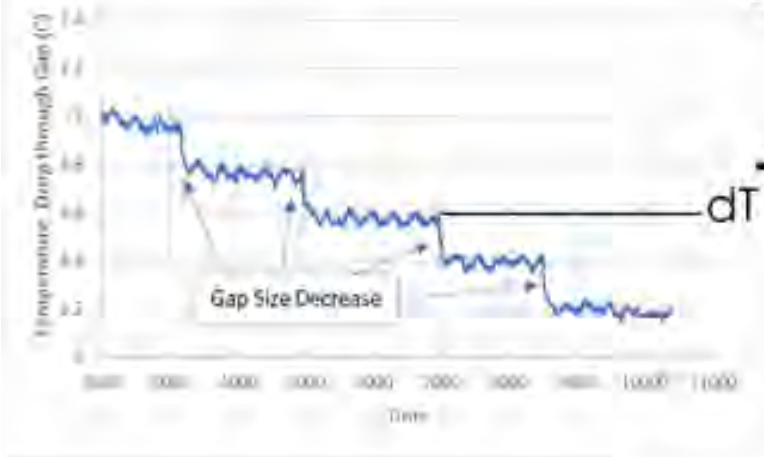
[1] DOI: 10.1016/j.ijheatmasstransfer.2022.122763

Methodology

Calculate temperature drop (*dT*) though gap at varying gap size using top and bottom thermocouples

Relative Gap Size (*dX*): recorded on digital variance indicator
Heater Power (*Q*): recorded by power supply
Area of Heater (*A*): measured and adjusted for thermal expansion

heat flux can be corrected for heat losses using radial and axial thermocouples, or directly calculated using the heat flux sensor below the specimen



dT

Thermal Resistance:
$R = dT/(Q/A)$

Calculate Thermal Resistance (R) at each gap size(dX)

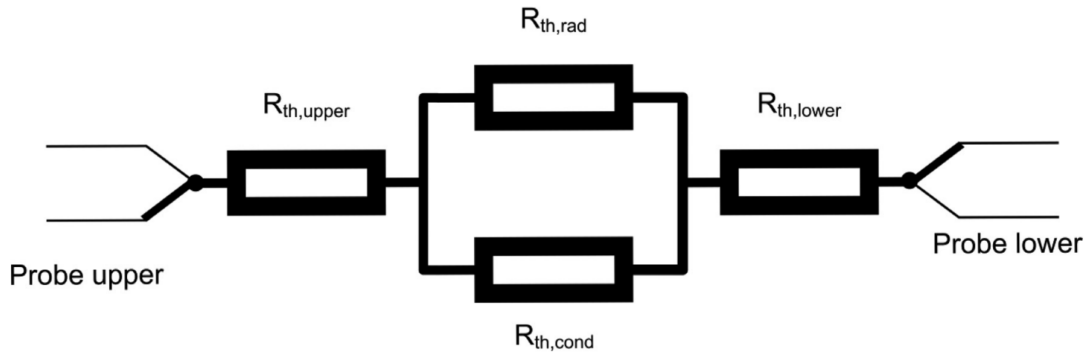
Thermal conductivity (k) is calculated by a least square fit of equations 1) or 2) on resistance versus gap size data

$$R = k^{-1}dx + C \qquad (1)$$

$$R = (k - k_r)^{-1}dx + C \qquad (2)$$

k_r is the apparent radiative conductivity, and is calculated with different methods, depending on the optical properties of the specimen

Repeating at different temperatures yield temperature dependent thermal conductivity



Power/Loss Correction

- Major care has been put into fully characterizing the power input (Q) for the system
- A parametric study using a well-known fluid (Helium) has been conducted to study the effects of:
  - Main power heater
  - Guard power heater
  - Axial power heater
  - Furnace temperature
  - Cooling Air Pressure
- A repeat of the NaCl-KCl near-eutectic salt measurement is currently being conducted to confirm the reliability and accuracy of the system.

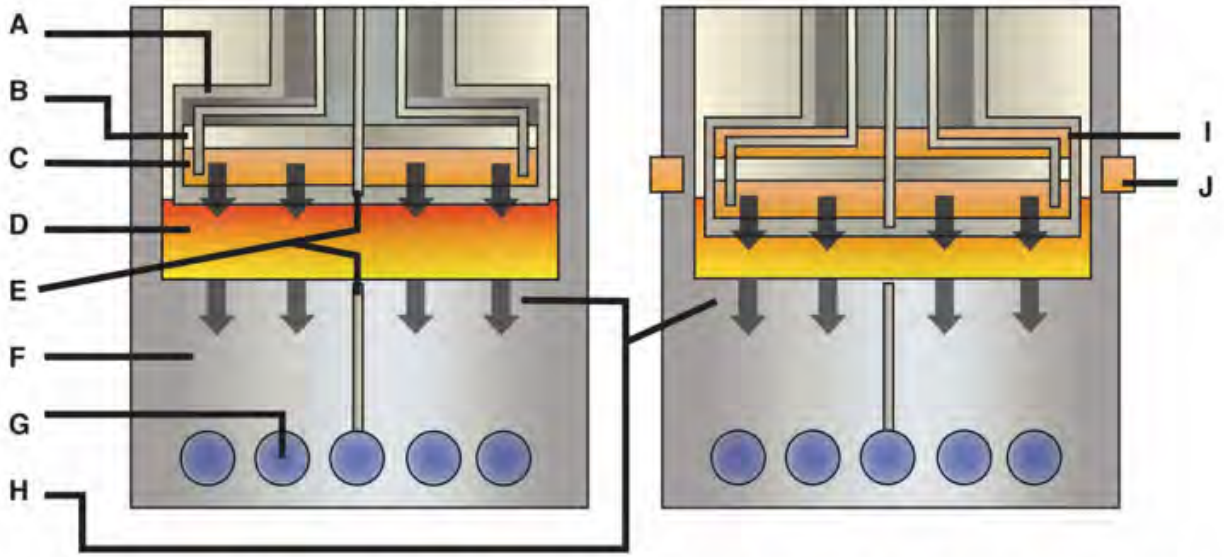
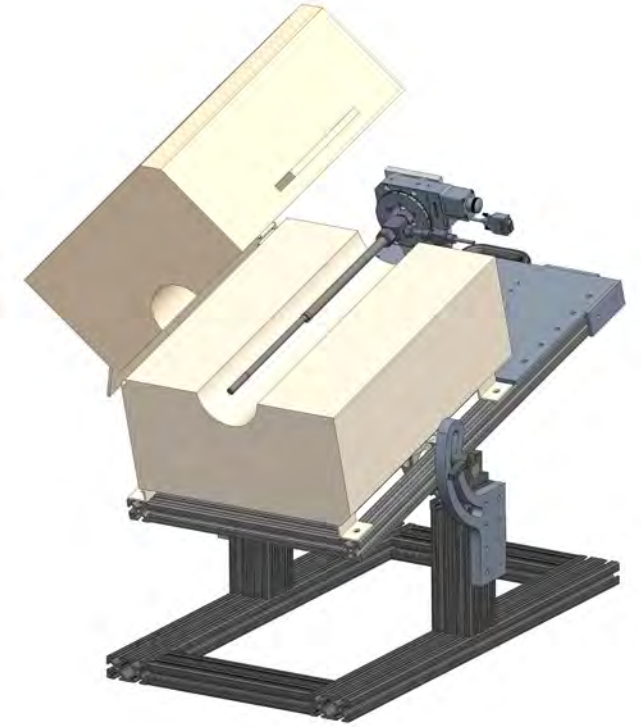


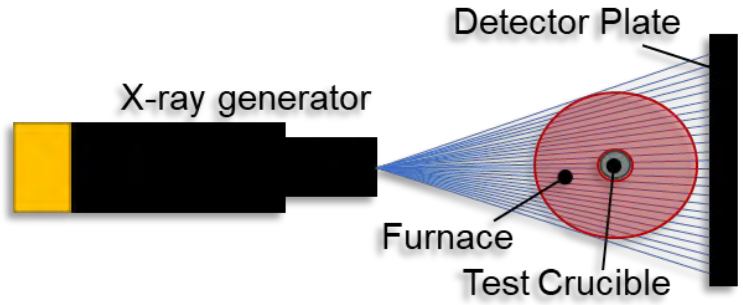
Fig. 1. Depiction of the specimen region of the previous variable gap thermal conductivity system (left) and the modified version used herein (right). (A) heater cell, (B) insulating disk, (C) resistive heater, (D) specimen, (E) thermocouples, (F) external containment chamber, (G) cooling channels, (H) ideal heat flux vectors, (I) the axial guard heater, (J) radial guard heater.

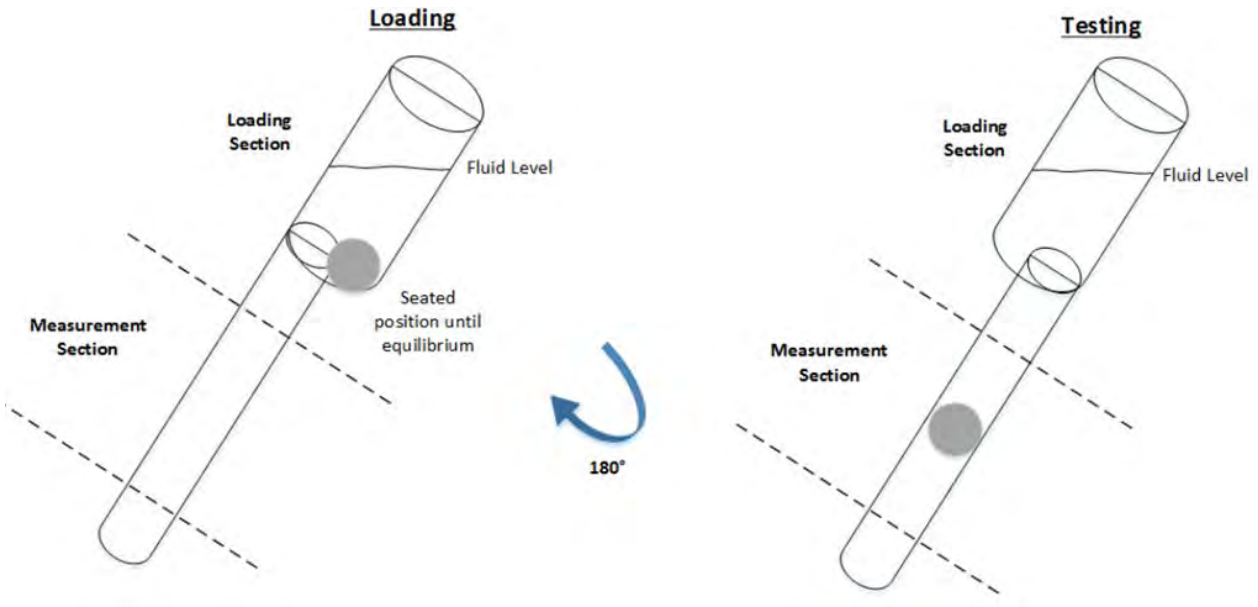
From Doi: https://doi.org/10.1016/j.molliq.2022.119151

FY2023 System Focus 2: Viscosity

- Technique: Falling Ball
- The viscosity of a fluid can be determined based on the terminal velocity of the ball going through that fluid in a tube
- Can track the ball using radiography if a non-transparent crucible is used
- Crucibles are maintained under inert environment
- NIST standard oils with well known viscosities used to calibrate the system

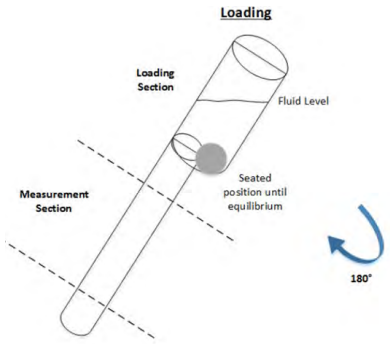


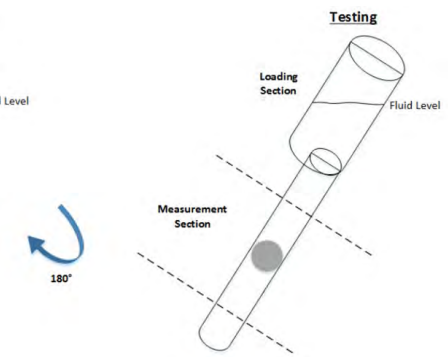


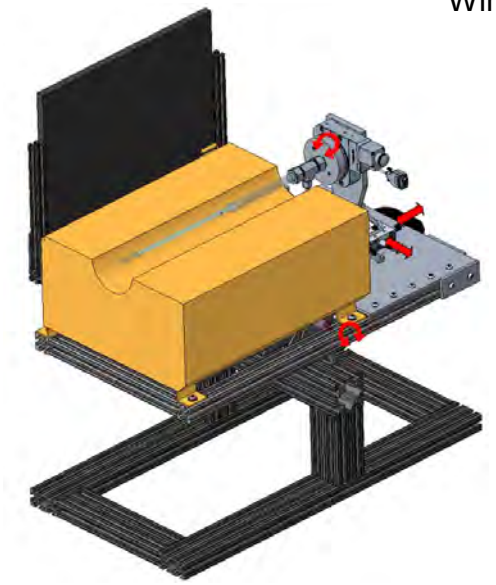


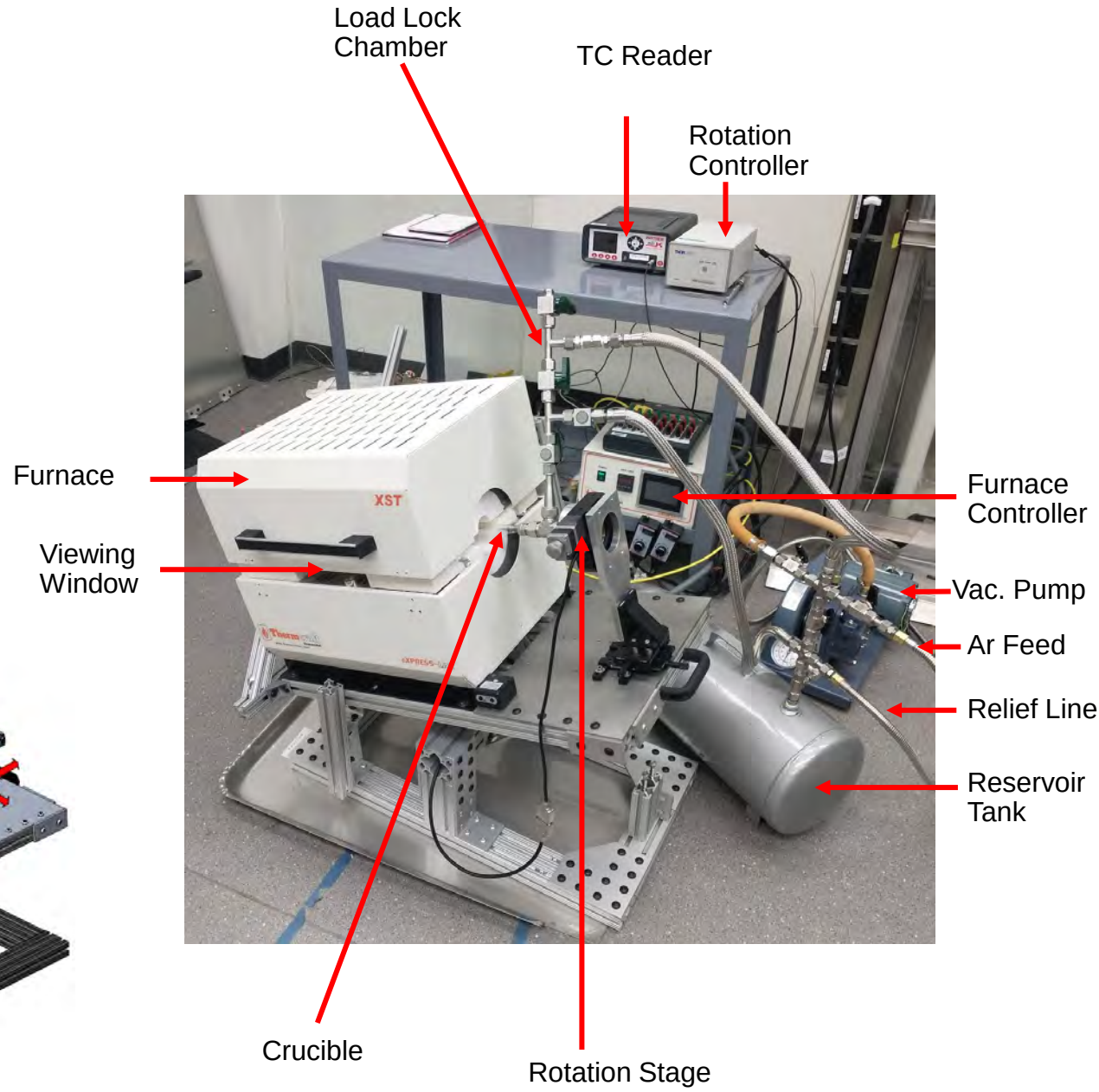
Experimental Configuration

- Custom crucible design allows for ball drop to occur upon axial rotation
- Measurements take place in a furnace on a rotating test stand
- System is backfilled with Ar, new balls are introduced in a mini load-lock chamber
- A viewing window allows tracking of the ball
  - Visual if crucible is glass
  - X-rays if crucible is metal





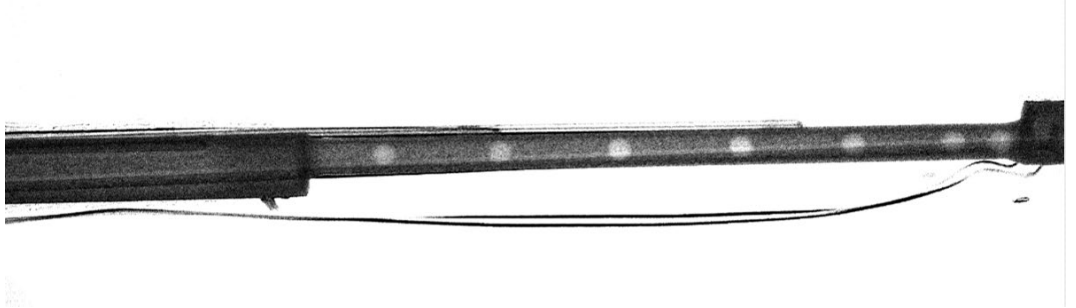


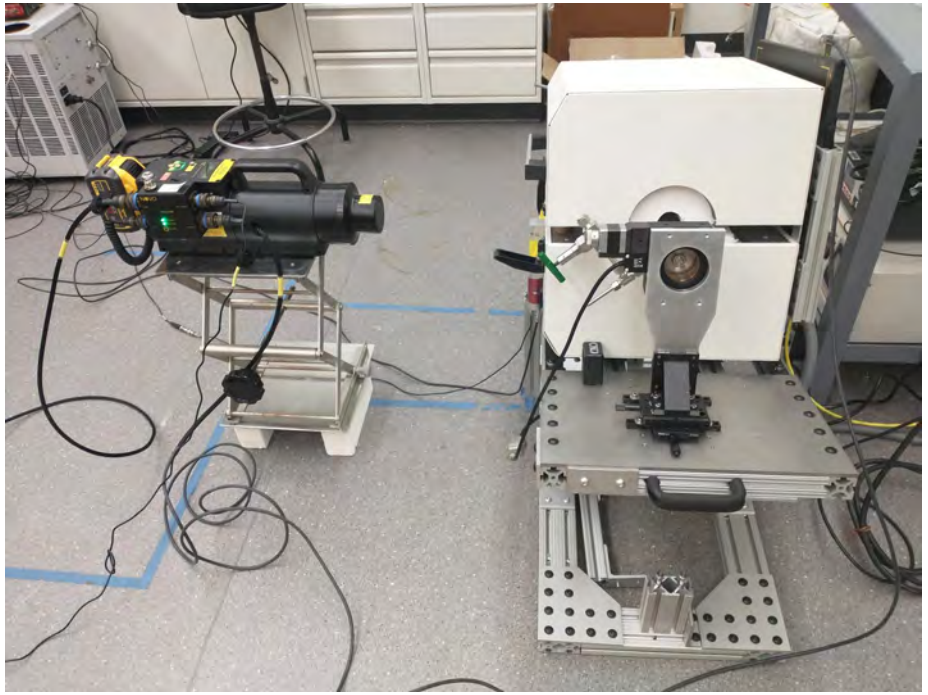


X-ray Compatibility

- Demonstrated x-ray compatibility will NIST oils
- Need to make design changes to crucible to enable more accurate measurements
  - Smaller tube thickness
  - Original design had 3-stages which created air pocket issue
- Glass crucible tests have allowed us to troubleshoot crucible design issues and establish design improvements for the future
- X-rays will be necessary for salts which cannot justifiably be measured in glass crucibles
- A steel crucible is also advantageous because of thermal expansion matching with a steel ball

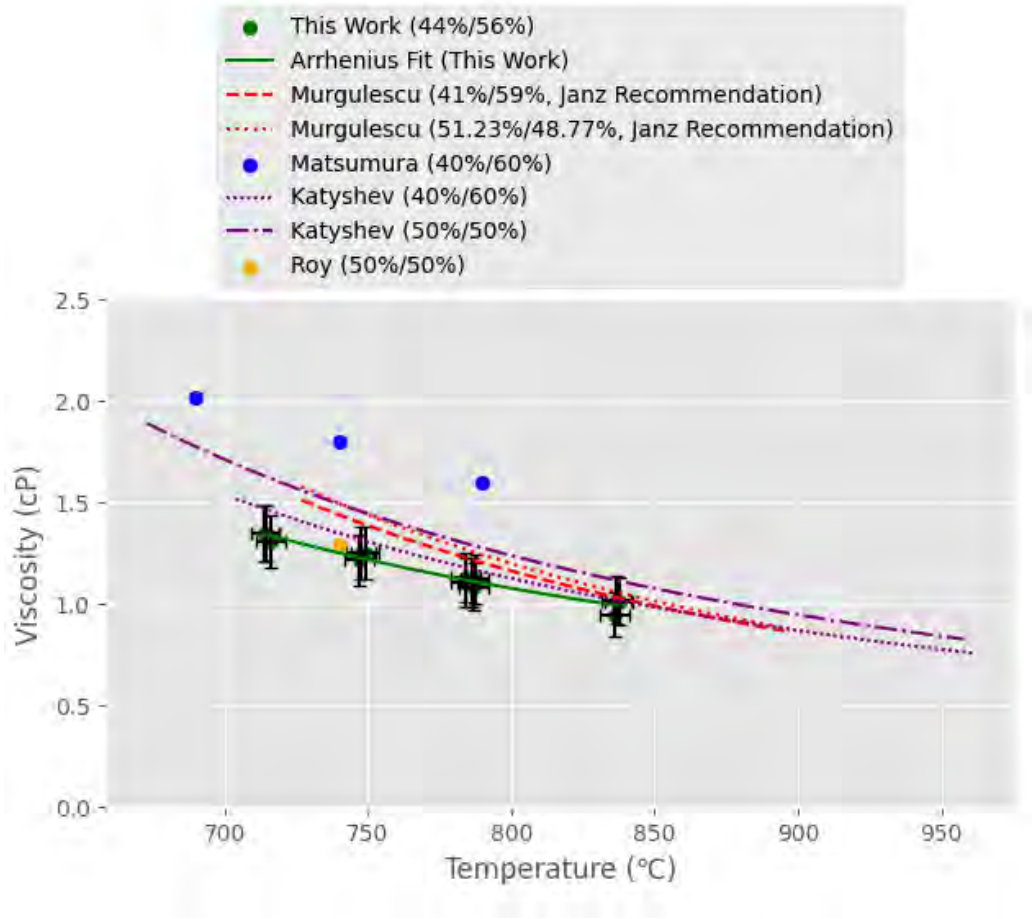
X-ray image of ball rolling through NIST oil





Viscosity Data

- Agrees generally well with most literature of measurements of similar NaCl-KCl mixtures
- However, Matsumura and Murgulescu both measured pure NaCl in separate studies, and these measurements were high relative to well known reference values
  - Matsumura ~30% high near the melting point
  - Murgulescu ~10% high near the melting point
  - Both used oscillation damping method
- Unclear what method was used by Katyshev
- Roy used capillary method

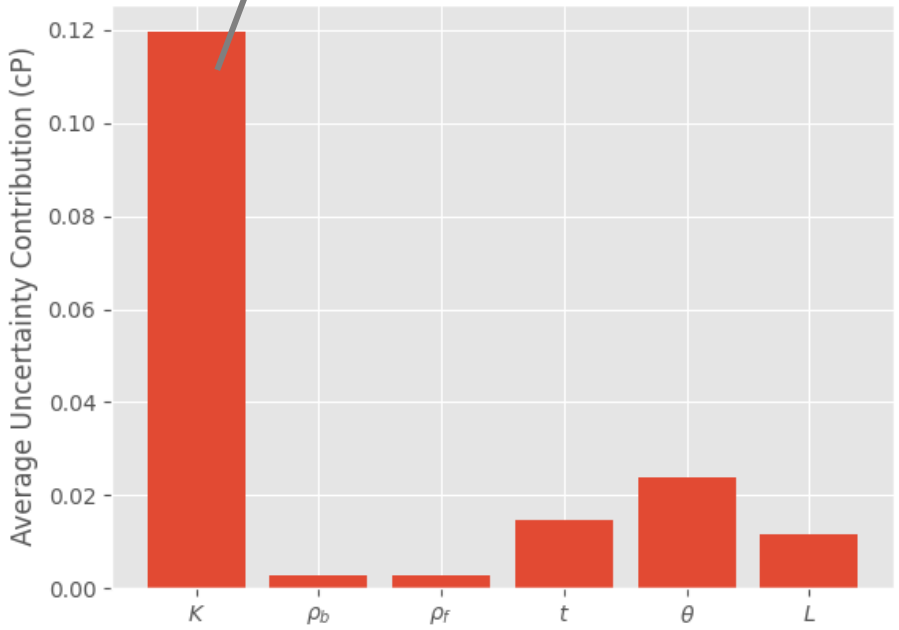


This data will be published in an article currently under review: "*Development and Demonstration of a Rolling Ball Viscometer for Molten Salts with Near-Eutectic NaCl-KCl*" in review with Thermal Science and Engineering Progress

Uncertainty Quantification

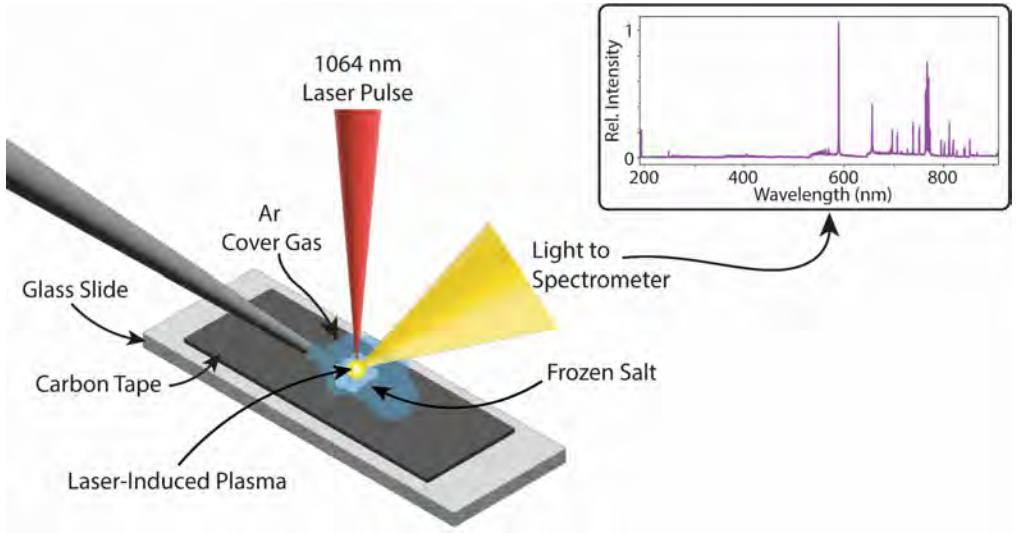
- Used Gaussian error propagation to quantify the experimental uncertainty
- Several sources of uncertainty
  - Ball diameter
  - Crucible inner diameter
  - Measured fall time
  - Angle measurement
  - Measured test section length
  - Material densities
  - Thermal expansion coefficients
  - Temperature
- All these sources of uncertainty feed into equations derived via Gaussian error formalism
- All mathematical details of this will be in upcoming publication
- **Average experimental uncertainty of 10.8% in this study**

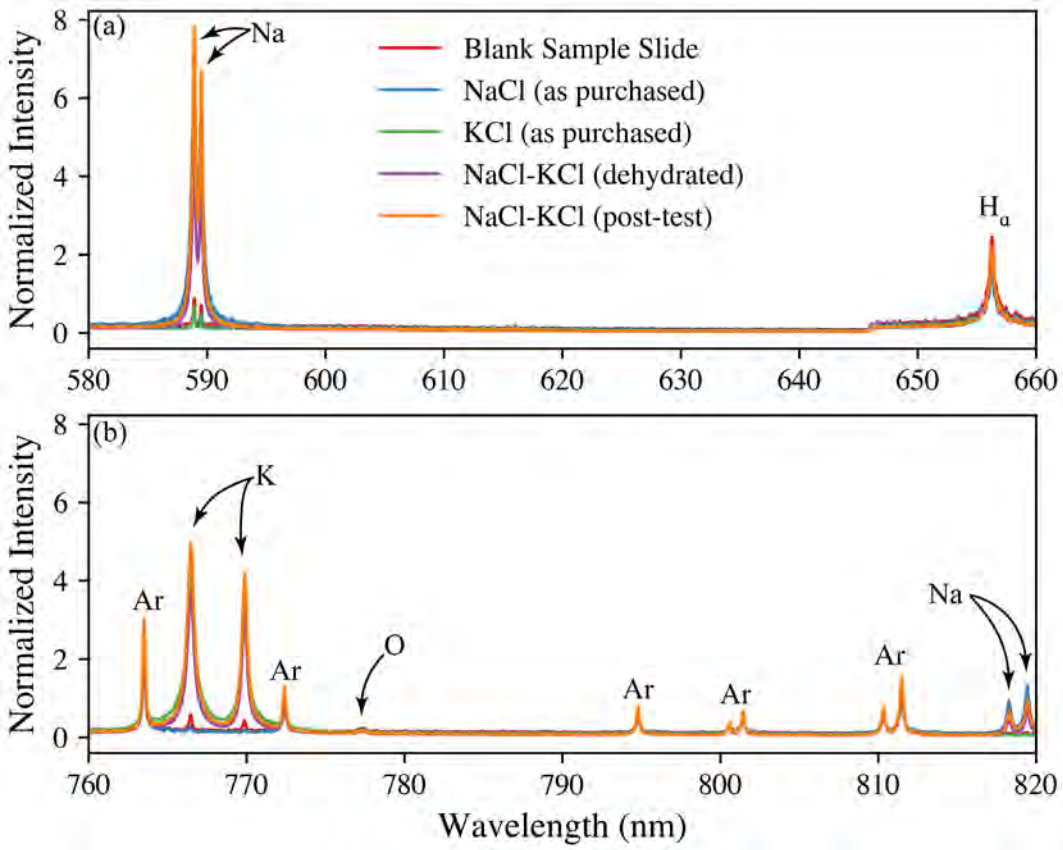
- K is the significant source of uncertainty
- This is primarily due to the sensitivity to d/D



Moisture Content Analysis

- Spectral intensities captured for:
  - Blank sample slide (reference)
  - As bought NaCl
  - As bought KCl
  - Dehydrated NaCl-KCl
  - Post-test NaCl-KCl
- No noticeable increase in O or H from the reference for any of the salts
- Previous studies using similar settings report 8 ppm limit of detection for O(doi: 10.1016/j.talanta.2018.09.078)
- Previous studies on purified FLiNaK showed greater O intensity (doi: 10.1039/d2ja00168c)





Future Plans with Thermal Conductivity

- Measure NaCl-KCl-LiCl in a compositional analysis as well as the pseudo-binary subsystems
- Repeat Thermal Calibration for Inconel Containment (Fluoride Salts)
- Demonstrate an actinide bearing salt
  - Ventilation now in place to handle U-bearing salts
  - Additional concerns for uranium waste streams still need to be worked out

Future Plans with Viscosity

- Measure NaCl-KCl-LiCl in a compositional analysis as well as the pseudo-binary subsystems
  - There is questionability about existing experimental data for NaCl-KCl and NaCl-KCl-LiCl; we would like to compare with our data
- Demonstrate x-ray measurements with a fluoride salt (FLiNaK most likely)
- Demonstrate an actinide bearing salt
  - Ventilation now in place to handle U-bearing salts
  - Could test chloride or fluoride, may be able to use glass

Existing data …

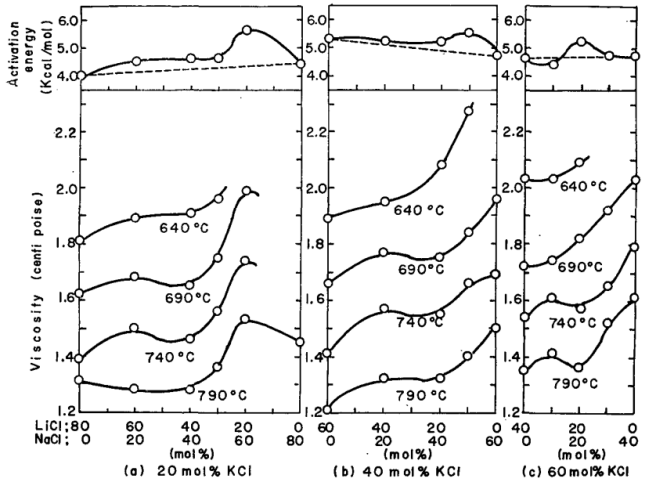


Fig. 2 Viscosity and activation energy of KCl-NaCl-LiCl system

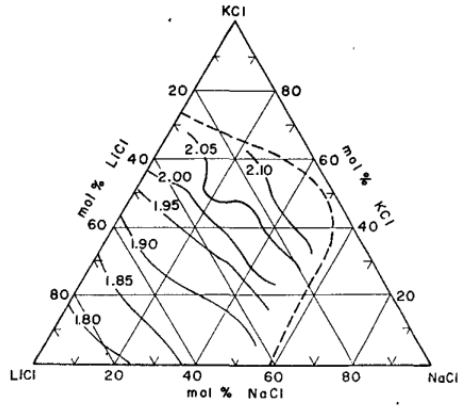


Fig. 3 Viscosity of KCl-NaCl-LiCl system at 690°C

From doi: https://doi.org/10.2207/qjjws1943.36.1211

Why we question it

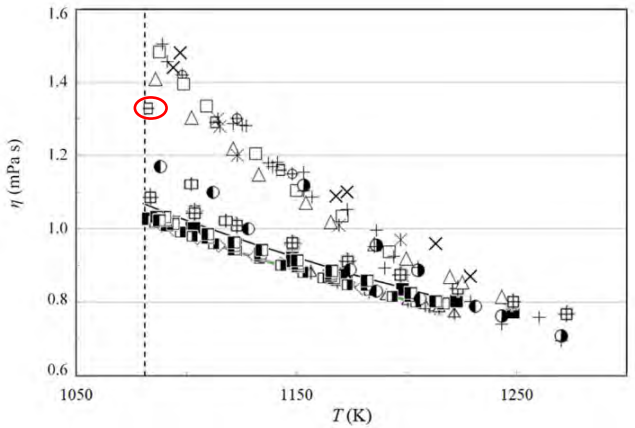


FIG. 1. Viscosity of molten NaCl at 0.1 MPa as a function of the temperature. Tolbaru *et al.*[11] (△), Ito *et al.*[12] (■), Ejima *et al.*[13] (◇), Tørklep and Øye[14] (◨), Abe *et al.*[15] (⊞), Antonov and Khokhlov[16] (—), Brockner *et al.*[17] (◧), Matsumura *et al.*[18] (⊟), Bondarenko and Strelets[19] (◐), Nishihara *et al.*[20] (◫), Murgulescu and Zuca[21] (△), Vershchetina and Luzhnaya[22] (⊕), Ogawa[23] (□), Berenblit[24] (×), Karpachev[25] (◑), Dantuma[26] (+), Fawsitt[27] (✱). Reference correlation of Janz[10] (green solid line), melting temperature (- -).

From Doi: https://doi.org/10.1063/1.5091511

Acknowledgements

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  - Bob Sitterson and David Bryant for assistance with system component fabrication
  - Callie Goetz for working on automation of the rotation stage for the viscosity system
- This work has been driven in part thanks to development of MSTDB, which is directly funded by NEAMS

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Thank you

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