

Thermal Properties of Molten Salts

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Provide the data necessary to build a FOAK MSR by 2035

Thermophysical property data are needed to design, license and operate an MSR

- Predicting molten salt behavior during normal and transient conditions requires knowledge of thermophysical property values over a range of temperatures and a range of compositions
- Available data are not sufficient or of suitable quality
 - Most developers are considering salt mixtures for which reliable property values must be measured, some data exist but quality standards vary
 - No data exist for actual or simulated irradiated mixtures containing fission products, activation products, impurities, etc. for model validation
- Data quality necessary for licensing a reactor requires use of standardized methods with known precision and accuracy based on measurements with benchmark salts

Our Approach to Meeting MSR Needs

Targeting developer needs to support design, licensing and operation of molten salt reactors

- Thermochemical and thermophysical properties of molten salts
 - Generating quality data to predict the behavior of molten salts during normal and transient operations, particularly systems for which limited or no data exist, including Pu-containing mixtures
 - Measuring property values for use in validating models of thermochemical behavior
 - Developing capabilities to measure salt properties under more extreme conditions (higher temperatures, more corrosive salts)
 - Improving measurement methods to minimize uncertainty in measured values
- Actively engaging with industry, discussing their data and quality needs for MSR development
 - Coordinating GAIN, NEUP, and direct-funded activities with MSR developers

FY22 Chemistry Activities

High Quality Molten Salt Property Measurements

- Performing high-quality measurements with plutonium-bearing salts to expand the database of relevant mixtures available to developers and modelers
 - Composition and impurity contents
 - Phase transition temperatures
 - Specific heats
 - Density
 - volume expansion coefficient
 - Surface tension
 - Thermal diffusivity and thermal conductivity
- Developing sealable sample cells for differential scanning calorimetry (DSC) measurements of high melting mixtures such as NaF-PuF₃.

MSTDB-TP Thermophysical- Chlorides

KCl	A, B								
LiCl	A	A, B, D							
MgCl ₂		A, B, D	A, B						
NaCl	A, B	A, B, D	A, D	A, B					
PuCl ₃					FY22				
ThCl ₄	A				A				
UCl ₃	A, B	A			A, B FY21				
UCl ₄	A, B	A	A	A, B					
ZrCl ₄	A								
	AlCl ₃	KCl	LiCl	MgCl ₂	NaCl	PuCl ₃	ThCl ₄	UCl ₃	UCl ₄

Properties modeled

A: Density

B: Viscosity

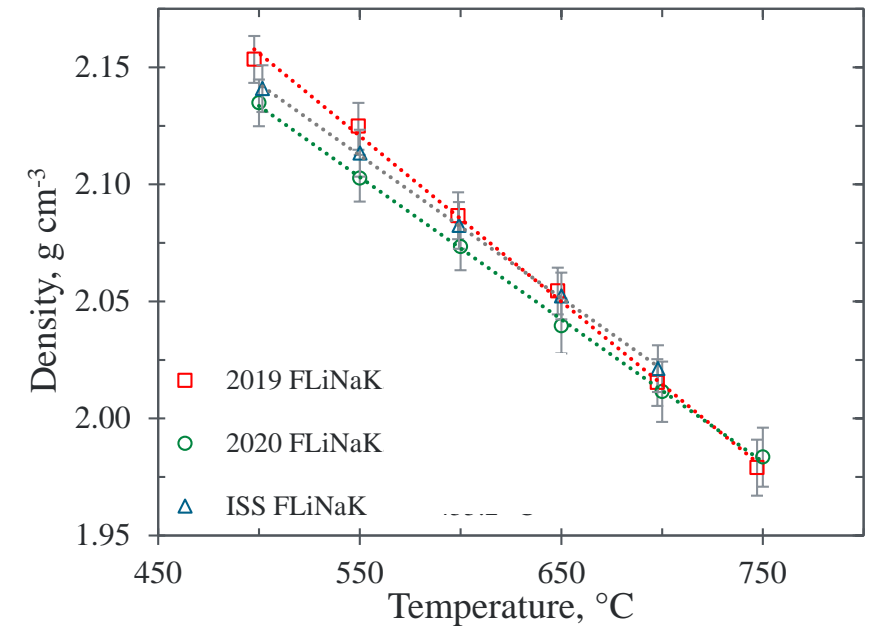
C: Thermal conductivity

D: Heat capacity

Letters in boxes indicate properties for which values are available in the database

Argonne Approach to Generating Quality Data

- Combining use of benchmark salts and calibration standards to establish accuracy and quantify the precision of measured data
- Using differential scanning calorimetry routinely to analyze and verify complex salt mixtures
 - Phase transitions are more sensitive to small variations in salt composition than can be measured using analytical methods.
- Using a combination of DSC and analytical methods to detect preferential volatility during the measurements that may have changed the salt composition
- Proceduralizing our measurement techniques and controls used to achieve NQA-1 data quality in preparation for ASTM standardization



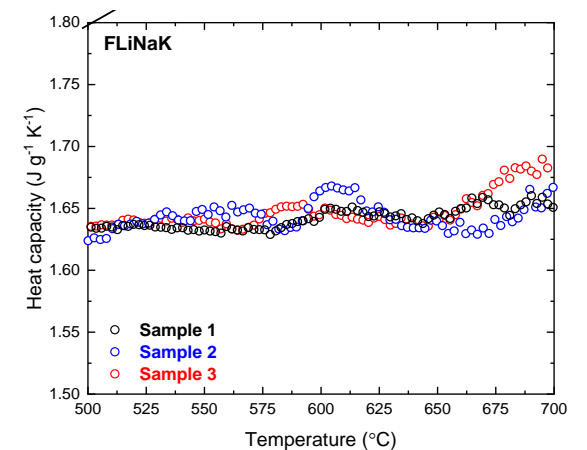
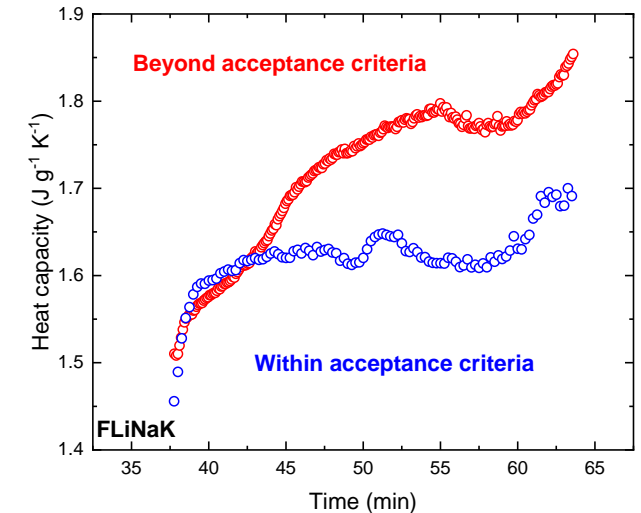
Density measurements of eutectic FLiNaK from several different sources- differentiated by their melting temperatures as measured by DSC.

Continuing FY21 Work: NaCl-KCl- UCl_3 Measurements

- Improved measurement techniques for heat capacity, density, thermal diffusivity and conductivity, viscosity and surface tension to provide high quality property values
 - Defined acceptance criteria linked to instrumental drift
- Performed measurements using improved techniques to expand the database of relevant binary and ternary mixtures
- Measured values for benchmark salts FLiNaK and FLiBe as well as fuel bearing salts NaCl- UCl_3 and NaCl-KCl- UCl_3 .
- Measurements on the ternary salt NaCl-KCl- UCl_3 continue this FY.

Reports Delivered in FY2021:

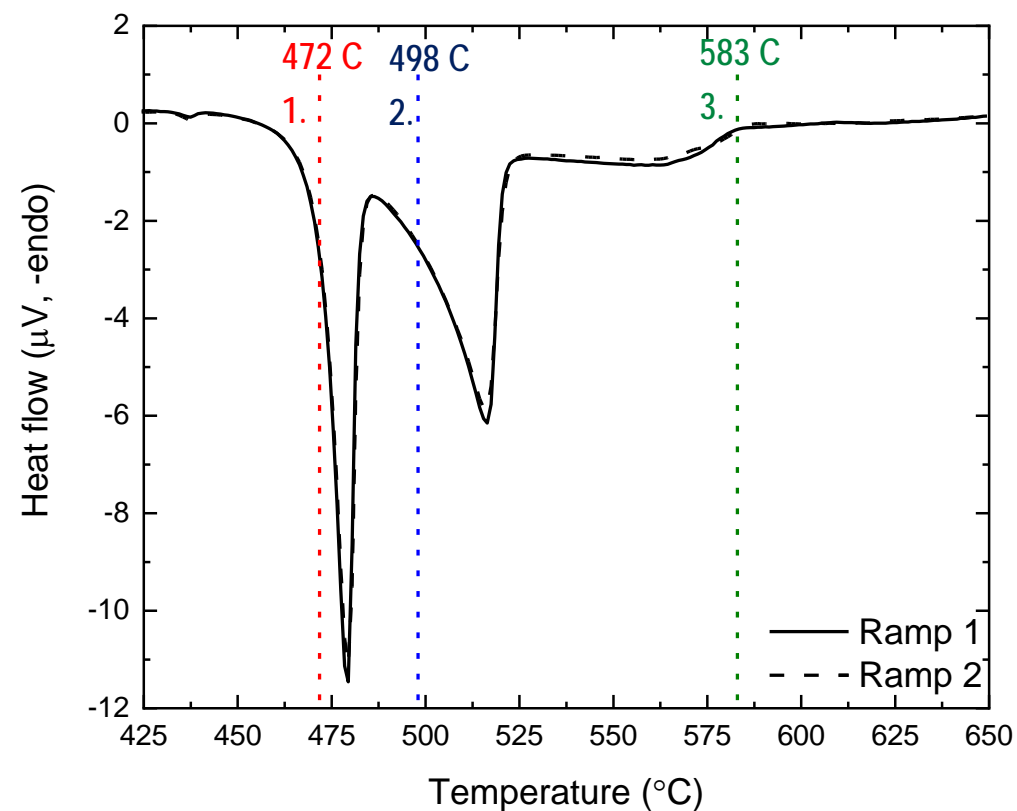
- Production and chemical analysis of NaCl-KCl- UCl_3 salts (M4AT-21AN0705011)
- Precision of Property Measurements with Reference Molten Salts (M3AT-21AN0705017)
- Data Quality of Salt Property Measurements (M3RD-21AN0703021)



Application of acceptance criteria for heat capacity measurements improves precision of property values for benchmark salts

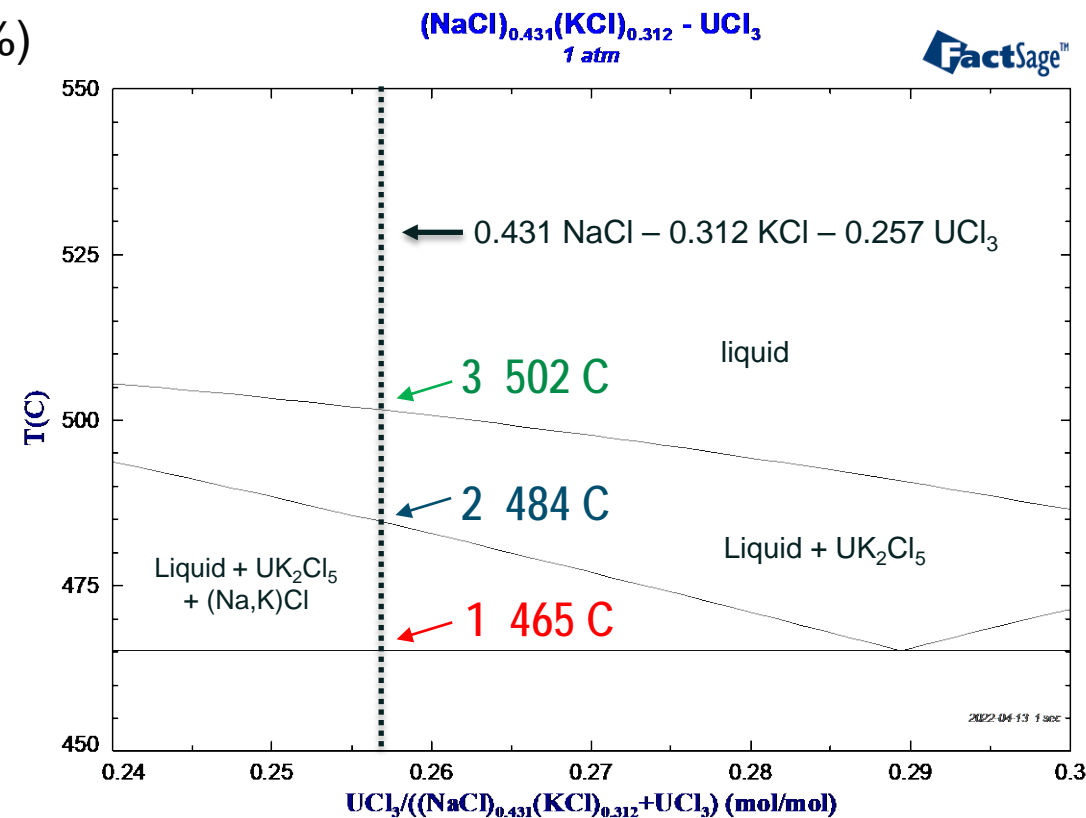
NaCl-KCl- UCl_3 measurement comparison with modeling

- Measured the thermal properties of (43.1-31.2- 25.7 mol%) NaCl-KCl- UCl_3
- Use of values in MSTDB underpredicts liquidus temperature by 82 °C



Right: Phase diagram of NaCl-KCl- UCl_3 system constructed in FactSage™ software using MSTDB-TC data.

Left: Differential Scanning Calorimetry (DSC) of (43.1-31.2-25.7 mol%) NaCl-KCl- UCl_3



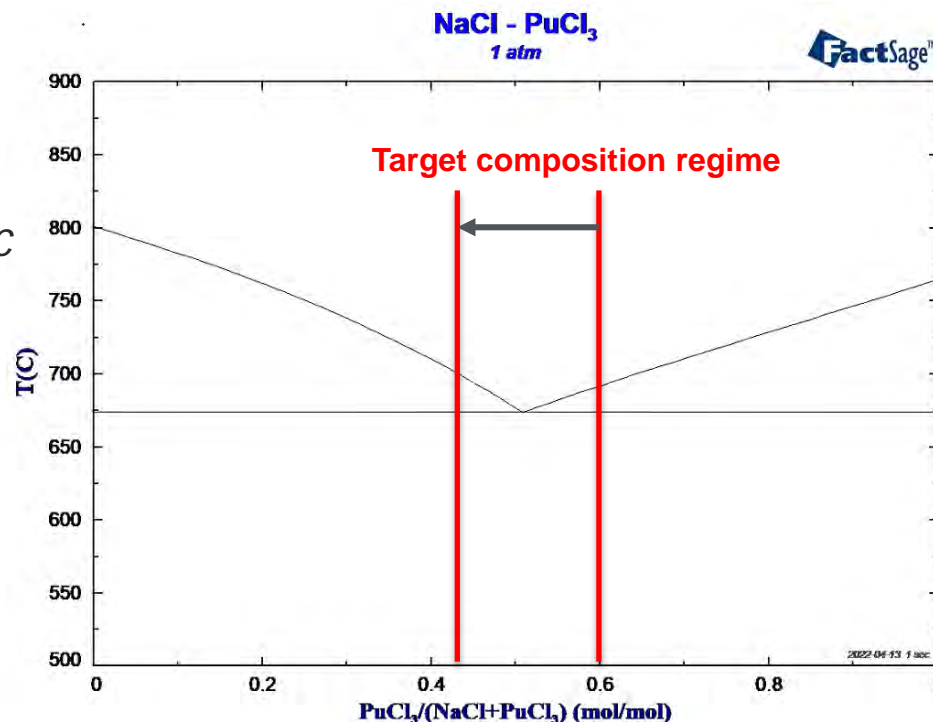
DSC Feature	Reaction	Measured Temp.	Predicted Temp.
1	Solid → Liquid + UK_2Cl_5 + (Na,K)Cl	472 °C	465 °C
2	Melting (Na,K)Cl → Liquid + UK_2Cl_5	498 °C	484 °C
3	Liquidus	583 °C	502 °C

NaCl-PuCl₃ Measurements

NaCl-PuCl₃ was chosen due to developer interests in the eutectic composition, but data set is lacking:

- Limited empirical investigations into the phase equilibria of this system
- Eutectic composition is between 36 and 38.3 mol % PuCl₃ based on published studies

Phase diagram constructed in FactSage[®] using data from MSTDB-TC



Thermochemical data generation for NaCl-PuCl₃ this FY:

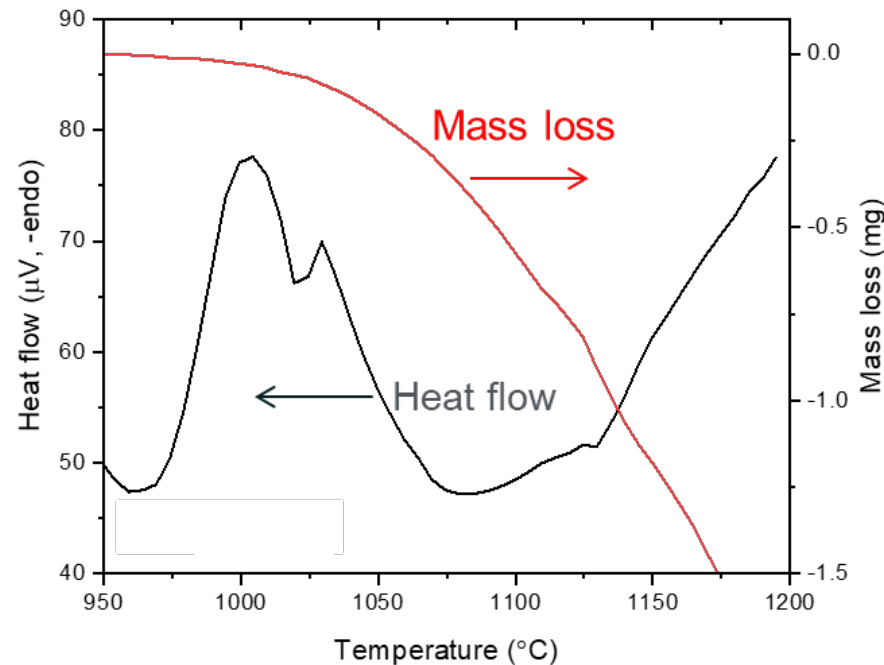
- Phase equilibria being measured for series of near-eutectic compositions by using DSC
 - Began at highest PuCl₃ content in the region of interest (red line on the right)
 - Adding NaCl to generate compositions within the region of interest
 - Data set will identify eutectic composition
- Heat capacity of solid and liquid NaCl-PuCl₃ at all compositions being measured by DSC
- Thermal diffusivity of the identified eutectic composition will be measured by laser flash analysis

Custom High Temperature DSC cells

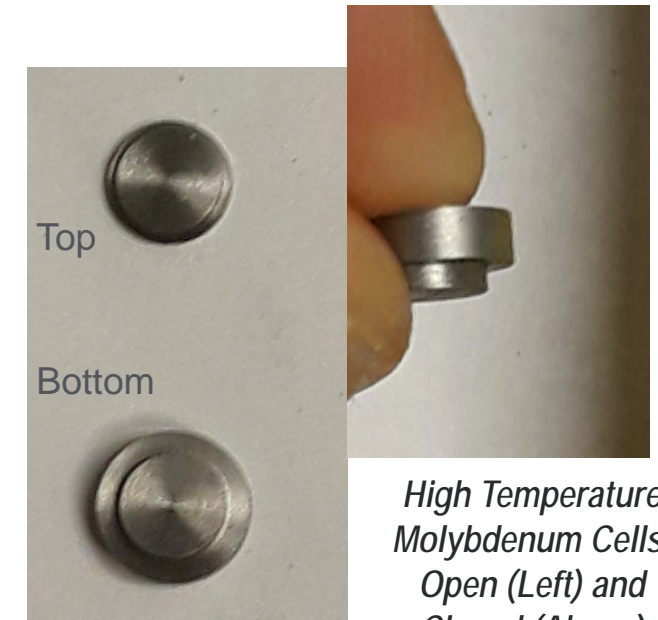
- Developing mechanically sealable cells for use up to 1200 °C .
- Cells made of Ni and Mo are being tested for required shaping and sealing
- Sealed cells are required for molten salts due to salt creep



High Temperature Nickel Cells, Open (Left) and Sealed (Right)



DSC and TG curves of heating UF_4 in a closed but unsealed molybdenum cell



High Temperature Molybdenum Cells, Open (Left) and Closed (Above)

Summary

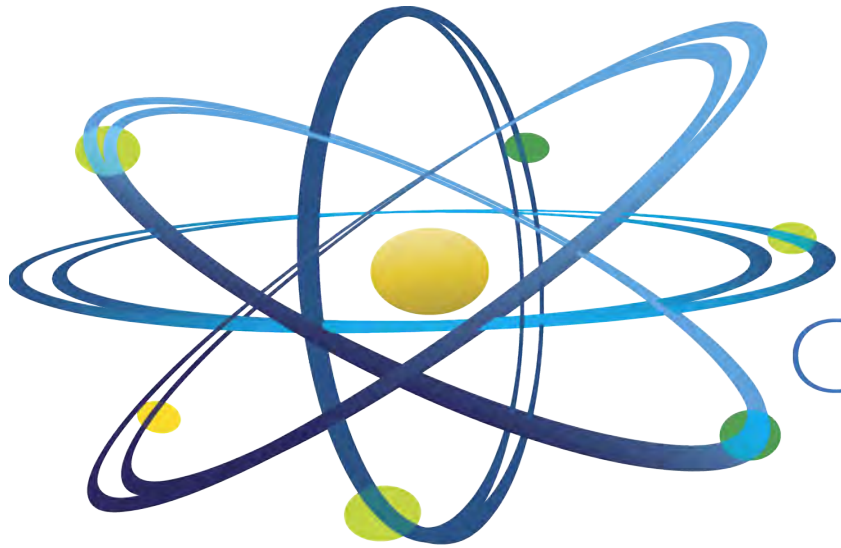
Applying improved methods to provide high quality property value data and fill gaps in the MSTDB

- Comparing measured properties of NaCl-KCl-UCl₃ to simulations made using MSTDB data.
- Measuring NaCl-PuCl₃ compositions bracketing eutectic
- Developing high temperature mechanically sealable cells for use in thermal analysis by differential scanning calorimetry up to 1200° C.

Acknowledgements

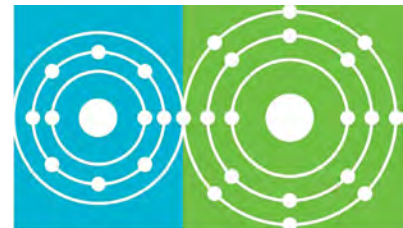
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Questions



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Molten Salt Reactor
PROGRAM