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

Molten Salt Reactor PROGRAM

Overview of MSTDB-TC

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University of South Carolina

Annual MSR Campaign Review Meeting 2-4 May 2023

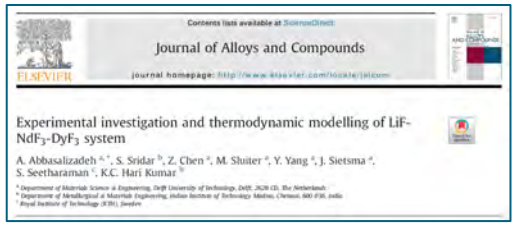
MSTDB-TC Adopts the FactSage™ ASC II Datafile Format (.dat)

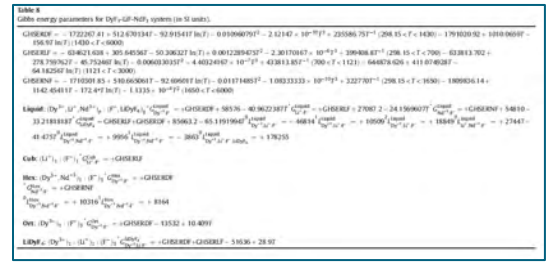
- MSTDB-TC is a library of Gibbs energy functions that incorporate compound/gas species values and models for solid and liquid (salt melt) solutions
- Database provides
  - Thermodynamic functions readable by FactSage™ and open source Thermochimica
  - Pseudo-binary and –ternary subsystem data
    - Reference values from literature with sources
    - Original measurements including uncertainties
    - Details of function fitting and uncertainties
- As systems are validated as they are added to database and new versions made available

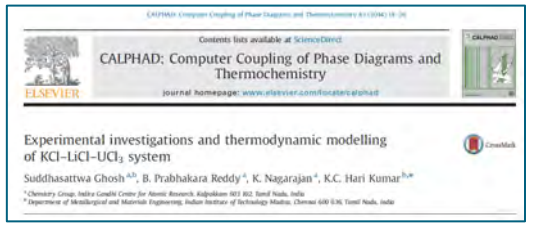
```
System U-F-Li
    3    2    2    3   12
 U                        F                        Li
   238.02891000             18.99840320              6.94100000
   6   1   2   3   4   5   6
   6   1   2   3   4   5   6
 gas_ideal
 IDMX
 LiF
   1  1    0.0    1.0    1.0
  6000.0000     -351581.57      37.443358     -35.397917     -.93533200E-03
 0.27571767E-07 0.00000000
UF4
   1  1    1.0    4.0    0.0
  6000.0000     -1639992.8      343.21185     -103.82600     -.47745000E-02
 0.24183333E-06  510660.00
LIQUsoln
 SUBG
  2.40000
   2   3
 Li//F
   1  1    0.0    1.0    1.0
  2500.0000     -617790.20      386.90980     -64.182999     0.00000000
 0.00000000     0.00000000
  1.00000      1.00000     0.000000     0.000000     0.000000
 U//F
   1  1    1.0    4.0    0.0
  2500.0000     -1966756.7      1054.9383     -174.74000     0.00000000
 0.00000000     0.00000000
  1.00000      4.00000     0.000000     0.000000     0.000000
   2   1
 Li                       U
 F
  1.00000      4.00000
   1   2
  1.00000
   1
   1   2
   1   1
   1   1   3   3  6.0000000      6.0000000      6.0000000      6.0000000
   2   2   3   3  6.0000000      6.0000000      1.5000000      1.5000000
   1   2   3   3  2.0000000      6.0000000      1.7142857      1.7142857
   3
 G   1   2   3   3   0   0   0   0
 0.00000000       1.00 0.00000000       1.00 0.00000000       1.00
 0.00000000       0.00 0.00000000       0.00 0.00000000       0.00
   0   0 -16108.400     0.00000000     0.00000000     0.00000000
```

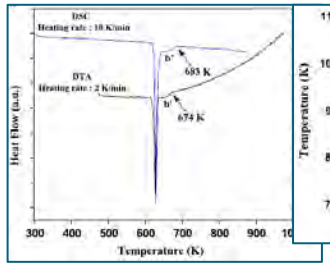
Values Traceable to Original Sources: Information Linked Within the User Database

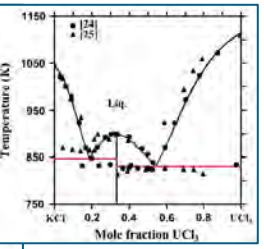
- Pure substance, single cation phase values from tabulated sources, e.g., NIST, SGTE via FactSage™
- Melt, solid solution, or complex compound models and values/parameters are
  - Obtained as is from published papers/reports
  - Determined in assessments/reassessments using values from published papers/reports
  - Computed for compounds obtained using *ab initio* modeling & used in assessments
  - Obtained using original DSC and high temperature XRD measurements at USC





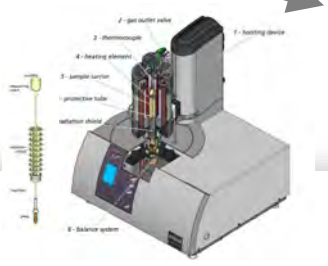


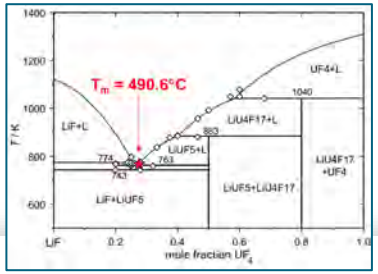


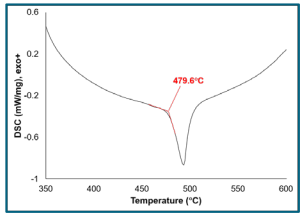


DFT

	H⁰0K (eV)	H⁰0K (J/mol)
K_2UF_6	-3.643	-3163464.3
K_3UF_7	-3.523	-3739095.7
$K_7U_6F_{31}$	-	
KU_2F_9	-3.858	-4466884.5







Each Release of MSTDB-TC Undergoes Third-Party QA

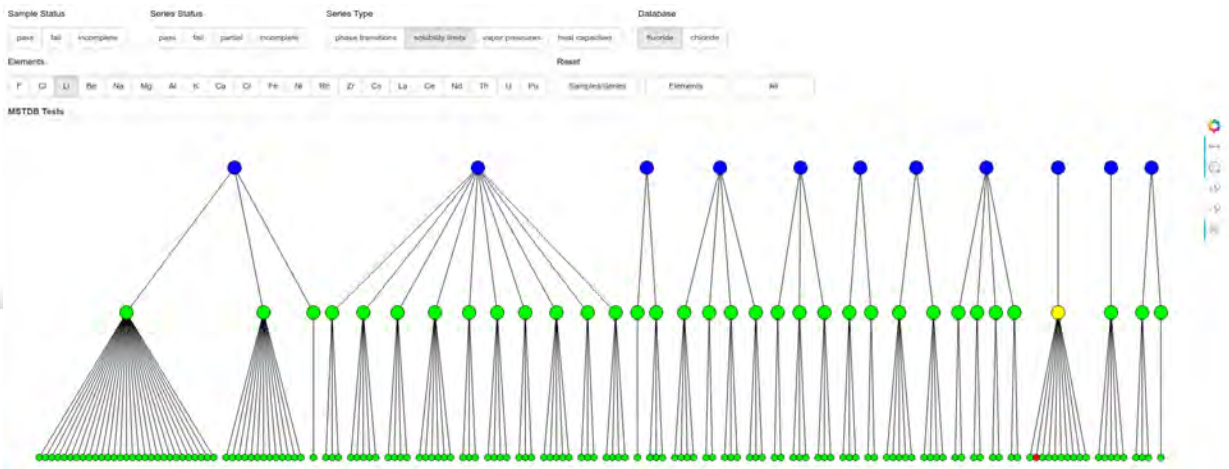
Reproduction of reported phase diagrams

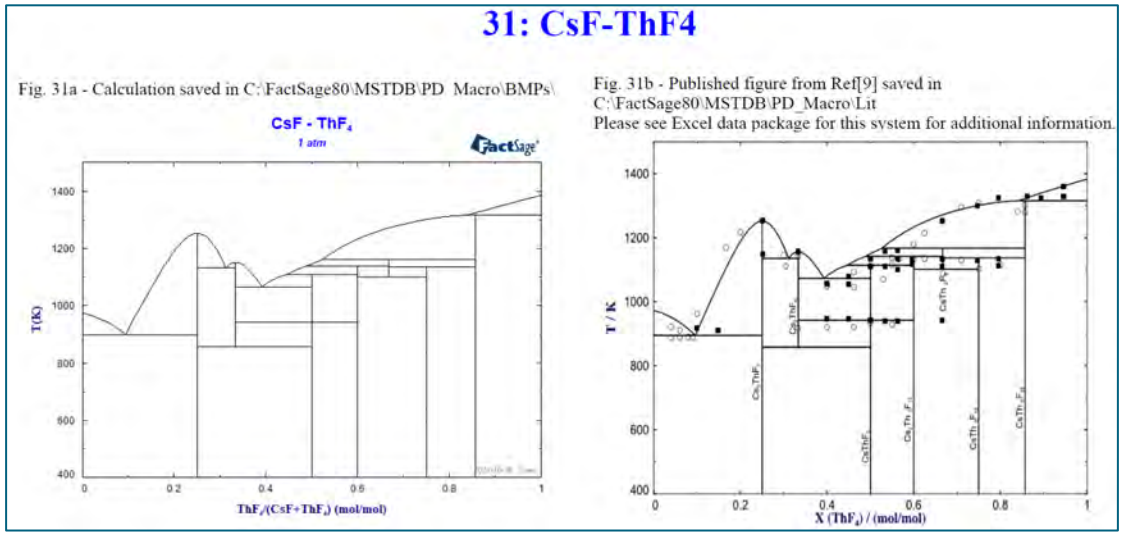
- Documentation files provided in download of MSTDB-TC

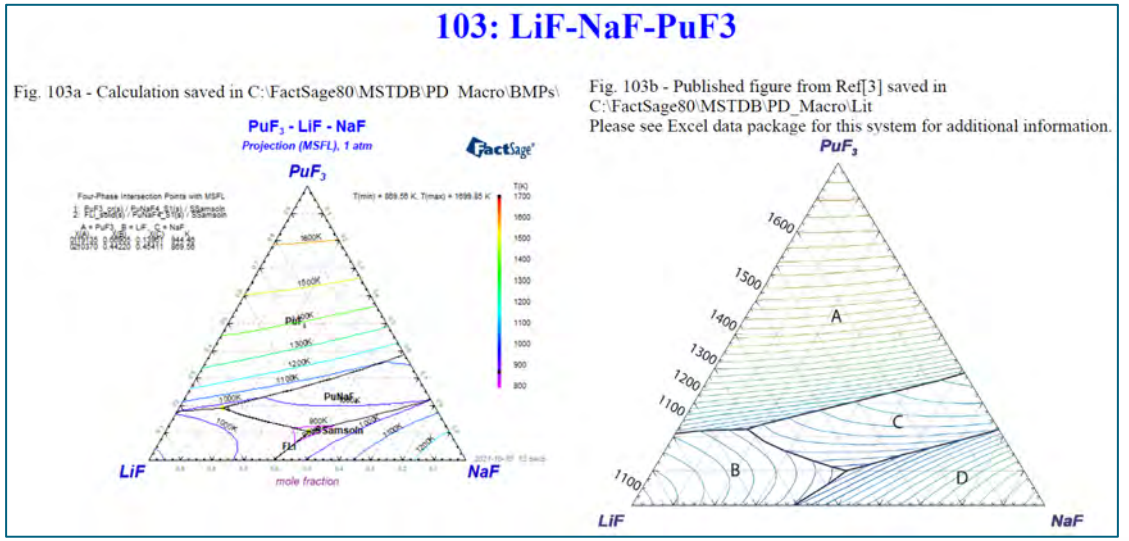
NEAMS effort automating incorporation error limits and propagation to higher order systems in progress at USC

Effort at Ontario Tech University has generated an automated database analysis

Dashboard of validation results from automated QA/QC







Application Examples: MSFR Burnup Behavior and NaCl-$PuCl_3$ Modeling/Phase Diagram

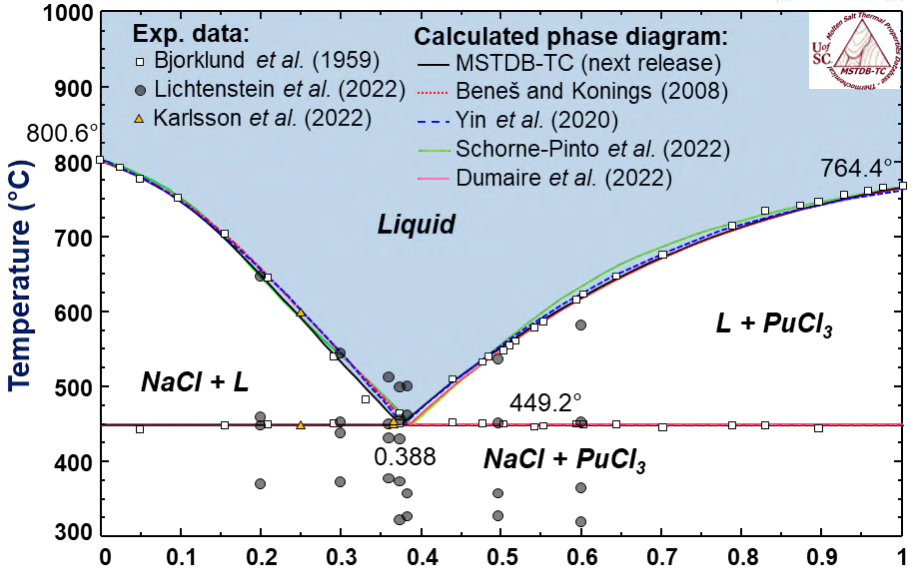
MSFR Serpent depletion calculations for fresh fuel composition 0.775LiF-0.1988ThF_4-0.0263UF_4 (mol fraction)

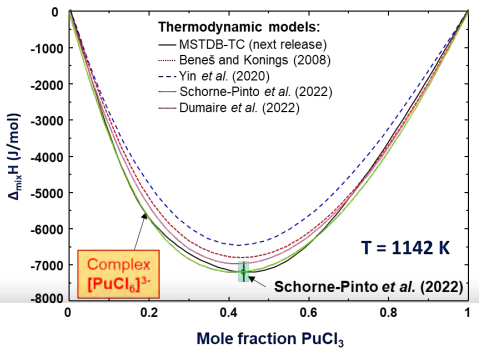
- At 80 GWd/t burnup (surrogate elements used in absence of data in MSTDB-TC)
- For **fresh fuel**, the first solid phase forms from the melt at **790.5K where LiF precipitates**
- For the **burnup composition**, the first solid phase forms from the melt is at **867.9K where the (Li,Rb,Cs)F solid solution phase precipitates**
- <u>Burnup has therefore raised the temperature for the first precipitation of a solid phase by 87K!</u>

80 GWd/t Burnup Model Composition

Element	Atom Fraction
F	62.26528
Li	28.80787
Th	7.40335
U	1.12814
Sr	0.12658
Ba	0.06685
Cs	0.06590
Rb	0.05771
Nd	0.00010
Ce	0.00005
La	0.00004
Pu	0.00002

MSTDB-TC provides for computing accurate phase representation for NaCl-$PuCl_3$



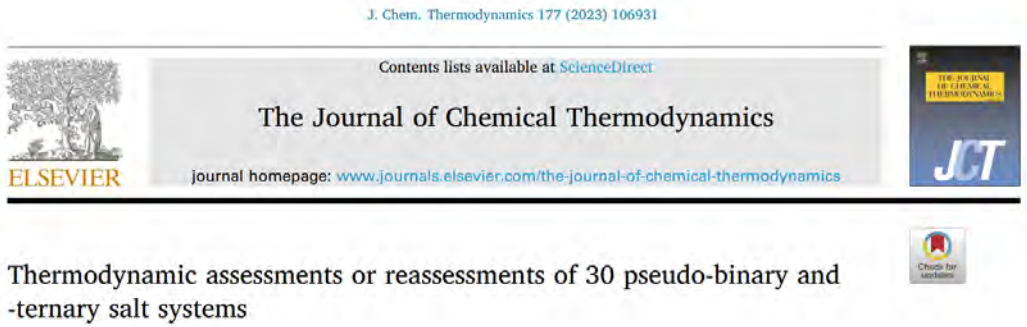


Content of Current Release: MSTDB-TC Ver. 2.0

- **Fluoride-based Systems**
  - 14 cations: Li, Na, K, Be, Rb, Cs, U, Pu, Th, La, Ca, Ce, Ni, Nd
  - 107 stoichiometric compounds
  - 53 pseudo-binary subsystems
  - 25 pseudo-ternary subsystems
  - Higher order systems
    - BeF_2-LiF-ThF_4-UF_4
    - LiF-NaF-ThF_4-UF_4
  - 26 solid solutions
  - 83 vapor species

- **Reciprocal Iodide Systems**
  - CsCl-CsI
  - CsF-CsI
  - KCl-KI
  - KF-KI
  - NaCl-NaI
  - NaF-NaI

- **Chloride-based Systems**
  - 14 cations: Li, Na, K, Mg, Cs, U, Pu, Ca, Ce, Rb, Al, Cr, Fe, Ni
  - 82 stoichiometric compounds
  - 60 pseudo-binary subsystems
  - 22 pseudo-ternary subsystem
  - Higher order system
    - AlCl3-KCl-LiCl-NaCl
    - CeCl3-KCl-LiCl-NaCl
    - CrCl2-KCl-MgCl2-NaCl
  - 25 solid solutions
  - 107 vapor species
  - Models for excess K and Mg in solution


J. Chem. Thermodynamics 177 (2023) 106931

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journal homepage: www.journals.elsevier.com/the-journal-of-chemical-thermodynamics

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Thermodynamic assessments or reassessments of 30 pseudo-binary and -ternary salt systems

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MSTDB-TC Ver. 3 Release Expected in May

- **Significant increase in content plus a number of systems revised/updated**
- **New values/models generated from our measurements together with reported properties**

New additions for Ver. 3 over Ver. 2 in **bold**

	Fluorides	Chloride	Iodides
Alkali metals	LiF, NaF, KF, RbF, CsF	LiCl, NaCl, KCl, RbCl, CsCl	LiI, NaI, KI, CsI
Alkaline earth metal	BeF_2, CaF_2, **SrF_2, BaF_2**	$MgCl_2$, $CaCl_2$	BeI_2, MgI_2
Transition metals	NiF_2, **CrF_3**	$CrCl_2$, $CrCl_3$, $FeCl_2$, $FeCl_3$, $NiCl_2$	-
Other metals	**YF_3, ZrF_4**	$AlCl_3$	-
Lanthanides	LaF_3, CeF_3, NdF_3, **PrF_3**	$CeCl_3$, **$LaCl_3$**	-
Actinides	ThF_4,UF_3, UF_4	UCl_3, UCl_4, **$PuCl_3$**	UI_3, UI_4
Pseudo-binary	53 systems (v.2) **70 systems (v.3)**	60 systems (v.2) **70 systems (v.3)**	10 systems (v.2) **30 systems (v.3)**
Pseudo-ternary	25 systems (v.2) **30 systems (v.3)**	22 systems (v.2) **27 systems (v.3)**	None (v.2) **15 systems (v.3)**

New Content

BeF_2 and ZrF_4
- LiF-BeF_2
- NaF-BeF_2
- KF-BeF_2
- CsF-BeF_2
- BeF_2-UF_4
- BeF_2-ThF_4
- BeF_2-ZrF_4
- LiF-ZrF_4
- CsF-ZrF_4

Reciprocal
- LiF-CsI
- LiF-KI
- LiF-NaI
- KI-CsF
- KF-CsI
- NaF-KI
- KF-NaI
- NaF-CsI

Iodides
- KI-CsI
- NaI-LiI
- LiI-KI
- NaI-KI
- NaI-CsI
- LiI-CsI

Higher Order
- LiF-LiI-CsI
- LiF-LiI-NaI
- LiF-LiI-KI
- LiF-CsF-CsI
- LiF-KF-KI
- LiF-NaF-NaI
- LiF-NaF-NaI
- NaI-NaF-KF
- KF-KI-NaF
- NaF-NaI-KF
- LiF-NaF-CsI
- LiF-KF-CsI
- LiF-KF-CsI
- NaF-KF-CsI
- LiF-KF-CsF-CsI
- CsI-LiF-NaF-KF
- $MgCl_2$-NaCl-$UCl_{3,4}$
- $MgCl_2$-KCl-$UCl_{3,4}$

MSTDB-TC Thermochemical (Experimental) Data Needs for the MSR Program

- **Selective data needs for current system assessments**
  - **LiF-NiF_2** system: Need enthalpy of mixing, Cp for intermediate compound
  - **NaF-NiF_2** system: Need enthalpy of mixing, Cp for intermediate compounds
  - **KF--NiF_2** system: Need enthalpy of mixing, Cp for intermediate compounds
  - **$PuCl_3$** systems with **LiCl, NaCl, KCl, $MgCl_2$**: MSTDB-TC improved with phase equilibria, enthalpies of mixing, Cp for the intermediate compounds
- **System information and/or assessments needed for new reciprocal salt models**
  - **Ul-$UF_{3,4}$**
  - **Ul-$UCl_{3,4}$**
  - **Bel-BeF_2**
- **Phase Equilibria for Be-containing Systems Requiring Experimental Determination**
  - **BeF_2-CrF_2, BeF_2-FeF_2**, and **BeF_2-NiF_2**
  - **LiF-BeF_2-CrF_2, LiF-BeF_2-FeF_2**, and **LiF-BeF_2-NiF_2**

Institutions Registering to Access MSTDB Reveal Demand for Databases

Companies

AlphaTech Research Corp
Anorganische Chemie/AG Salzchemie
Columbia Basin Consulting Group, LLC
Copenhagen Atomics
Creare LLC
Flibe Energy
Kairos Power
Moltex Energy
NAAREA
ORANO
Orano Group
Plasma Processes
Seaborg Technologies ApS
SGS
SIEMENS
Terrestrial Energy
ThermoCalc
THORIZON
Virtus Solis Technologies Inc

US National Labs

ANL
INL
LANL
NIST
NREL
ORNL
PNNL
SNL

Foreign National Labs

CEA
Chinese Acad. of Sciences
CNL
KAIST
Nifty.NE.JP
Kurchatov Institute
RWTH-Aachen

Universities

Abilene Christian University	University of Alabama
Brigham Young University	University of Idaho
Georgia Tech	University of Iowa
Hamburg University of Technology	University of Massachusetts-Lowell
KYUSHU UNIVERSITY	University of Michigan
Linköping University	University of Nevada
MIT	University of Notre Dame Dame
North Carolina State University	University of Pittsburgh
Ohio State University	University of Tennessee
Politecnico di Torino	University of Texas
Purdue University	University of Wisconsin
Rensselaer Polytechnic Institute.	Virginia Commonwealth University
Texas A&M University	Virginia Tech
Pennsylvania State University	Washington State University
UC-Berkeley	

Other Accomplishments

- **Workshop/Training on MSTDB-TC and –TP (April 25, >90 participants)**
  - Meeting recording and presenter slides will be available on a dedicated USC website
- **USC Publications**
  - Thermodynamic Assessments or Reassessments of 30 Pseudo-Binary and -Ternary Salt Systems, Johnathan C. Ard, Juliano Schorne-Pinto, Mina Aziziha, Jacob A. Yingling, Kaitlin E. Johnson, Clara M. Dixon, Theodore M. Besmann, J. Chem. Thermo. 177 (2023) 106931 https://doi.org/10.1016/j.jct.2022.106931
  - Thermodynamic Assessment of Lithium Halide Reciprocal Salt Systems for Energy Applications, Mina Aziziha, Juliano Schorne-Pinto, Jacob A. Yingling, Clara M. Dixon, Johnathan C. Ard, Mahmoud A. A. Aslani, Amir M. Mofrad, and Theodore M. Besmann, J. Mol. Liq., 364 (2022) 119973. https://doi.org/10.1016/j.molliq.2022.119973
  - Development of the Molten Salt Thermal Properties Database − Thermochemical (MSTDB−TC), example applications, and LiCl−RbCl and UF3−UF4 system assessments Johnathon C. Ard, Jacob A. Yingling, Kaitlin E. Johnson, Juliano Schorne-Pinto, Mina Aziziha, Clara M. Dixon, Matthew S. Christian, Jacob W. McMurray, Theodore M. Besmann, J. Nucl. Matls. 563 (2022) 153631. https://doi.org/10.1016/j.jnucmat.2022.153631

Other Accomplishments

- **USC Conference presentations**
  - **Invited:** Development and Application of the Molten Salt Thermal Properties Database-Thermochemical (MSTDB-TC) in the Simulation of Molten Salt Reactor Behavior, T. M. Besmann, J. Schorne-Pinto, J. A. Yingling, J. C. Ard, M. Aziziha, C. M. Dixon, A. M. Mofrad, J. P. S. Palma, R. E. Booth, J. Wermers, NuMat 2022
  - **Invited:** Practical Approach to Modeling the Complex Thermochemistry of Actinide-Containing Molten Salts, T. M. Besmann, J. A. Yingling, J. Schorne-Pinto, J. C. Ard, M. Aziziha, C. M. Dixon, J. P. S. Palma, R. E. Booth, A. M. Mofrad, J. Wermers, TMS 2023
  - Purification of UF4 for Preparation of Simulated Irradiated Fuel Salt B.W.N. Fitzpatrick1, K. Foster2, T.M. Besmann2, M. Short3, M.H.A. Piro1, Canadian Nuclear Society Annual Meeting
  - Melting point, enthalpy of fusion, and excess heat capacity of a FLiNaK determined by the CALPHAD method, J. Schorne−Pinto, J. C. Ard, M. Aziziha, K. Foster, J. A. Yingling, A. M. Mofrad, M. S. Christian, and T. M. Besmann, MS&T 2022
  - Effective Assessment and Thermodynamic Database Development for Potential Nuclear Reactor Molten Salt Systems, Theodore M. Besmann, Juliano-Schorne Pinto, Jacob A. Yingling, Johnathon C. Ard, Mina Aziziha, Matthew S. Christian, Amir Mofrad, Mahmut A. A. Aslani, Jake W. McMurray, MS&T 2022

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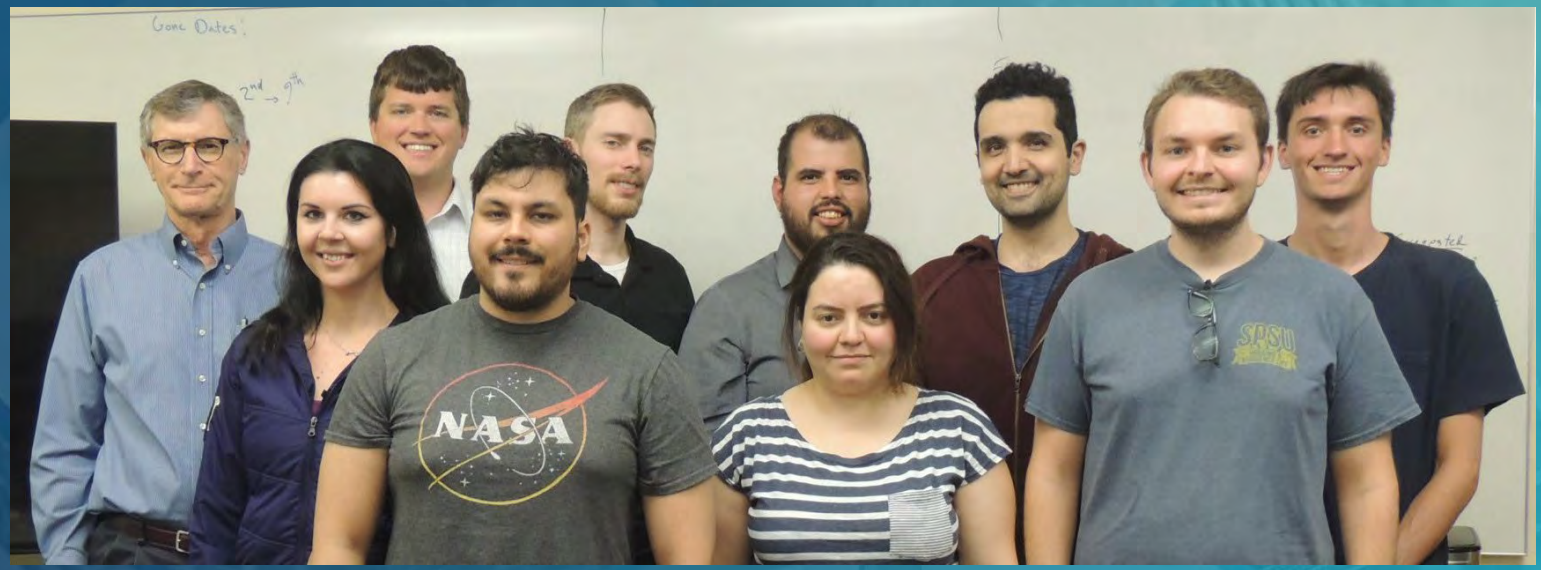
Thank you

Molten Salt Reactor PROGRAM



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