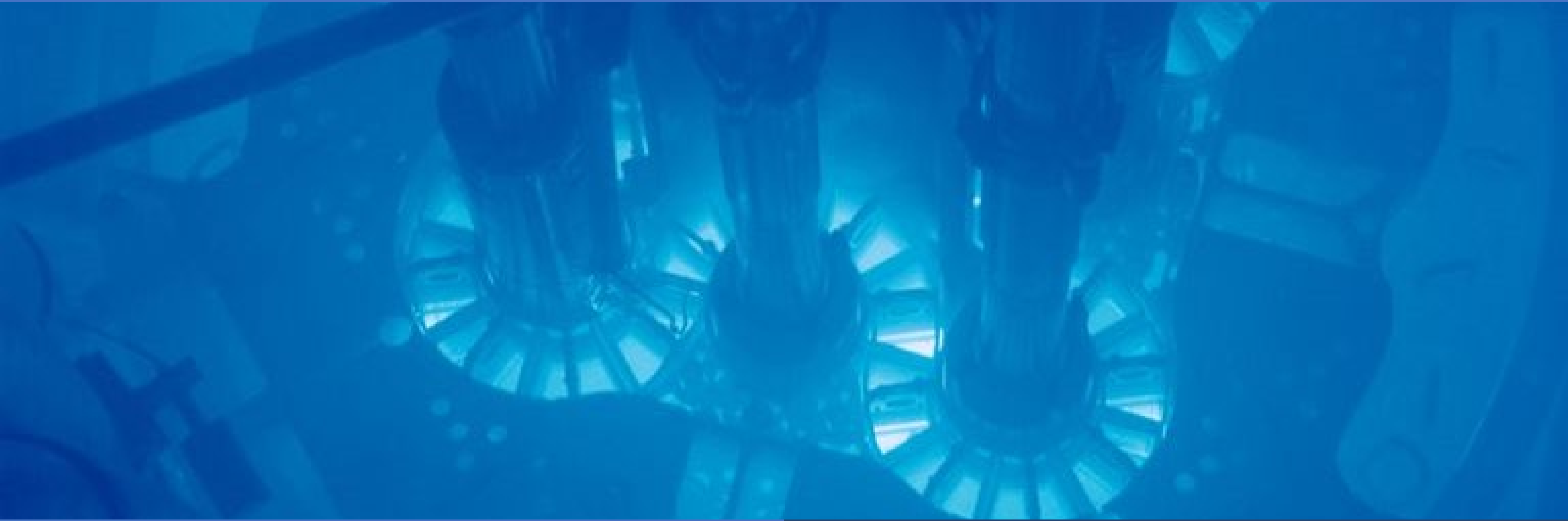
U.S. DEPARTMENT OF ENERGY | Office of NUCLEAR ENERGY

Clean. Reliable. Nuclear.



Molten Salt Reactor Campaign
An Overview

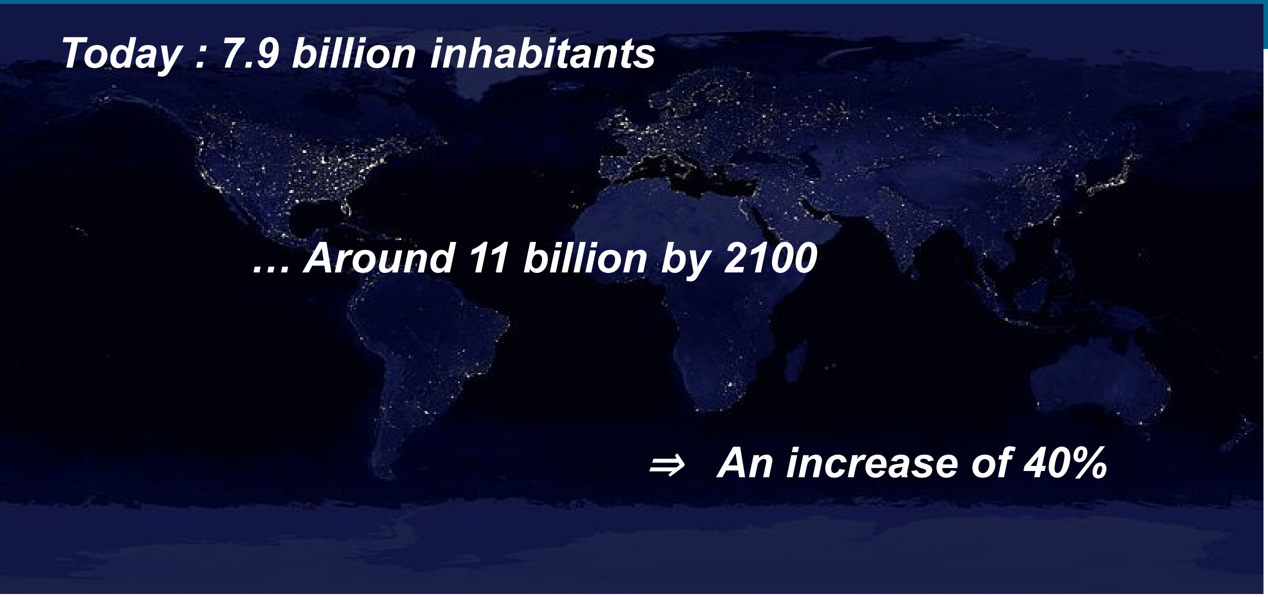
Dr. Patricia Paviet

National Technical Director

November 15-17, 2021

PNNL-SA-168351

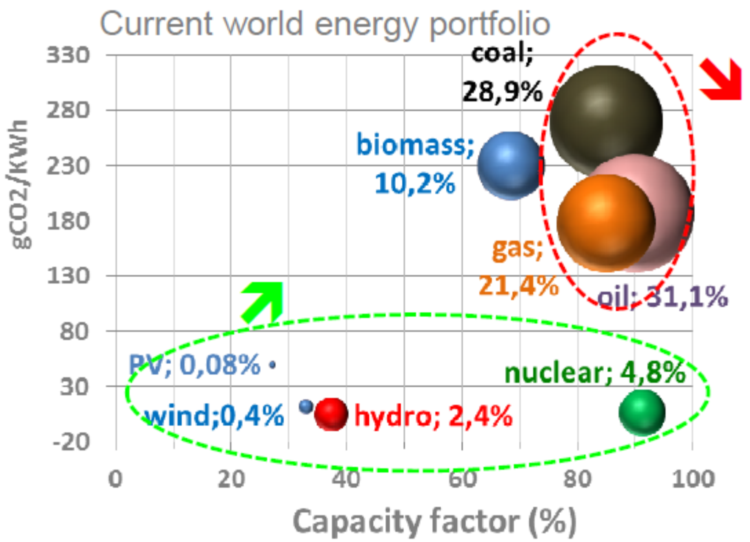
Nuclear Energy is Promising



COP26 – November 2021

International agreement to limit average temperature rise to <1.5 C

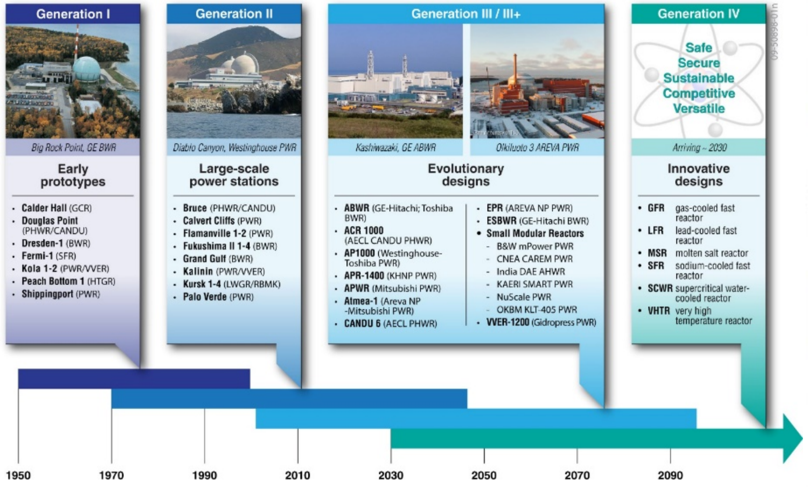
- Current global power consumption in 2021: 17.7 TW (10^{12}), Fossil fuels account for 79% of US energy consumption.
- Atmospheric CO_2: 1900 – 270 ppm, 2000 – 377 ppm, April 2020 – 412.5 ppm, 2100 – 550 ppm.
- To stabilize at 550 ppm, 15 TW of emission free power is needed by 2050.

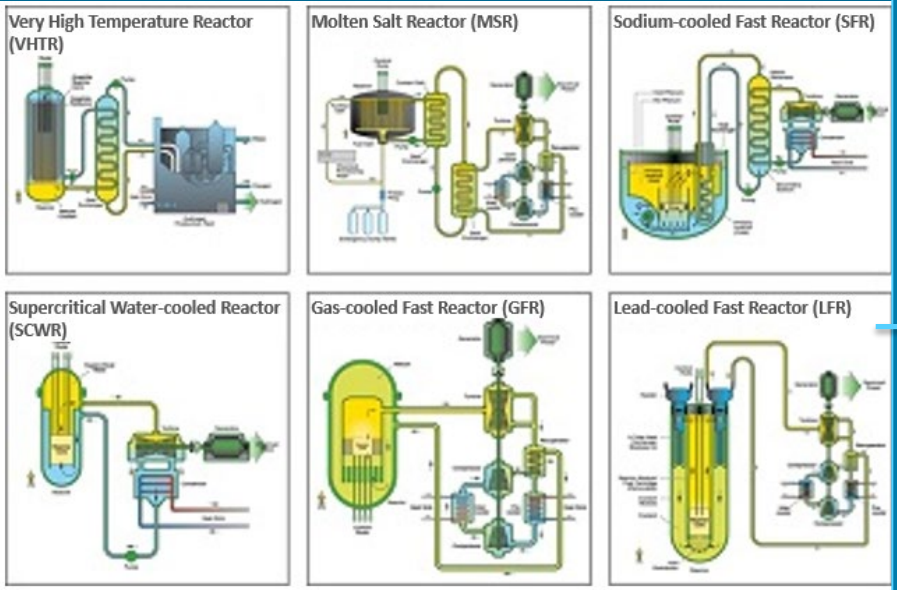


Energy transition… will require technologies that are power dense and capable of scaling of many tens of TWh… Most forms of renewable energy are, unfortunately incapable of doing so… Nuclear fission today represents the only present-day-zero-carbon-technology able to meet….

Ecomodernist Manifesto, 2015

Six Generation IV Reactor Technologies





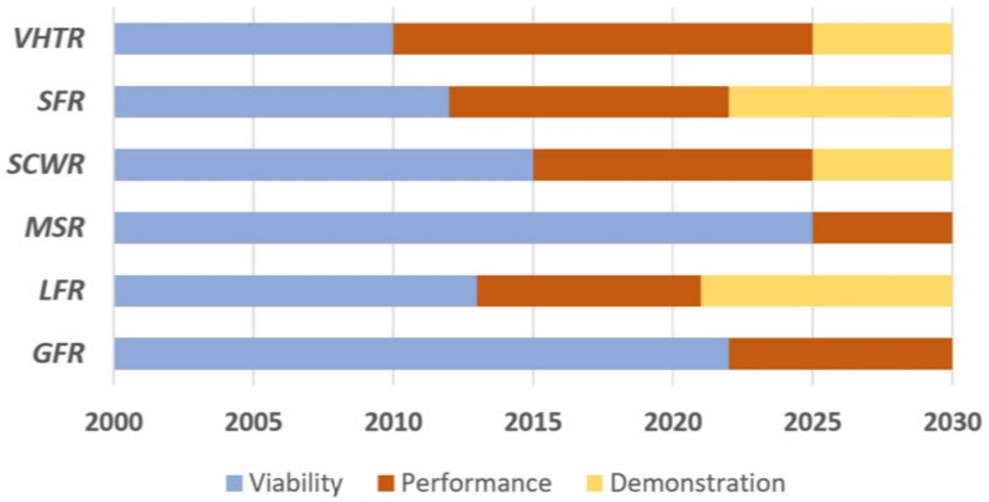
Cross-cutting Collaborations

- Economics & Modelling
- Education & Training
- Proliferation Resistance & Physical Protection
- Risk & Safety
- Safety Design Criteria
- R&D Infrastructure

To achieve goals in four areas:

1. Sustainable energy with minimum waste
2. Life cycle cost advantages
3. Safety and reliability
4. Proliferation resistance & physical protection

GIF Roadmap 2013



…aiming to be ready for industrial deployment by 2030

Reactor Types: GEN IV Concepts

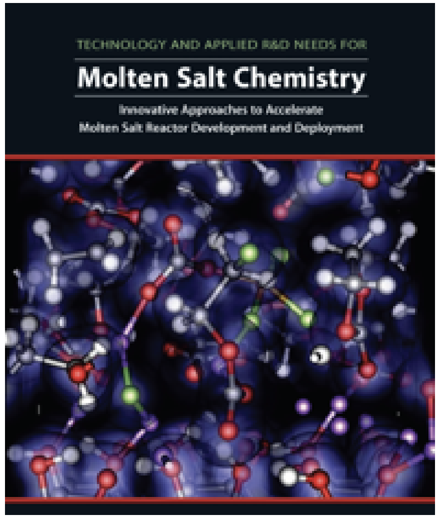
Reactor type (neutron energy spectrum)	Coolant	Typical Fuels Input at Start of Irradiation Cycle	Fuel cycle	Use	Benefit Relative to Current LWRs	Development stage
Gas-cooled fast reactor, GFR (fast)	Helium	U/Pu or U/TRU (MOX, metal, nitride, carbide), and natural U	Closed*	Electricity, Hydrogen**, Process Heat**	Higher thermal efficiency for electricity production (>40% vs. 33%)	Early
Lead-cooled fast reactor, LFR (fast)	Lead, Lead/Bismuth	U/Pu or U/TRU (MOX, metal, nitride, carbide), and natural U	Closed*	Electricity, Hydrogen**, Process Heat**	Higher thermal efficiency for electricity production (>40% vs. 33%), Low pressure reactor coolant Passive safety capability	Early
Molten salt reactor, MSR (fast)	Chloride salts	U/Pu or U/TRU in salt, and natural U in salt	Closed*	Electricity, Hydrogen**	Higher thermal efficiency for electricity production (>40% vs. 33%), Low pressure reactor coolant Passive safety capability	Early
Sodium-cooled fast reactor, SFR (fast)	Sodium	U/Pu or U/TRU (MOX, metal, nitride, carbide), and natural U	Closed*	Electricity	Higher thermal efficiency for electricity production (>40% vs. 33%), Low pressure reactor coolant Passive safety capability	Prototypes exist internationally, currently nothing in U.S.
Supercritical water-cooled reactor, SCWR (thermal or fast)	Water	Enriched UO_2, U/Pu or U/TRU MOX fuel	Open (thermal) Closed* (fast)	Electricity	Higher thermal efficiency for electricity production (>40% vs. 33%)	Early
Very high temperature gas reactor, VHTR (thermal)	Helium	Enriched UO_2, prism or pebbles	Open	Electricity, Hydrogen**, Process heat**	Higher outlet temperature, Higher thermal efficiency for electricity production (>40% vs. 33%)	Advanced for lower temperatures, Early for high temperature

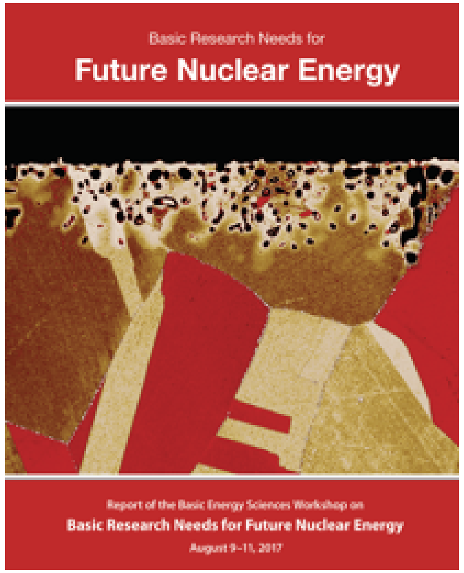
*A "Closed" fuel cycle may be self-sustaining, i.e., not needing new fissile from outside of the fuel cycle
** In this table, a reactor coolant outlet temperature >750°C is assumed for higher-efficiency hydrogen production, and >850°C for process heat
Any of the reactors could be designed for sizes up to at least 1500 MWe, although the initial Gen-IV selection considered specific ranges at the time

Courtesy Dr. Roald Wigeland, retired from INL

DOE Workshops to enable the design of revolutionary Molten Salt Reactors

- **Radionuclide Chemistry**
  - **Gap 1** – Determining chemical speciation and oxidation states of dissolved constituents within molten salts.
  - **Gap 2** – Determining transuranic and fission product solubilities in molten salts.
  - **Gap 3** – Determining salt-phase thermodynamics.
  - **Gap 4** – Determining the radiation effects in molten salt media.
  - **Gap 5** – Separation of impurities and recycling of the salt matrix.
- **Monitoring and signatures**
  - **Gap 1** – Developing and demonstrating sensors for monitoring the oxidation-reduction ratio as well as fission product concentrations in salt systems.
  - **Gap 2** – Building a foundation to monitor operations with observable signatures.
- **Interfacial chemistry**
  - **Gap 1** – Investigations of local interfacial changes induced by corrosion
  - **Gap 2** – Understanding materials degradation driven by coupled phenomena
  - **Gap 3** – Understanding multiscale evolution leading to degradation of fuels and structural materials





Programmatic Goal and Objectives of the MSR Program

<u>Mission:</u> Develop the technological foundations to enable MSRs for safe and economical operations while maintaining a high level of proliferation resistance.

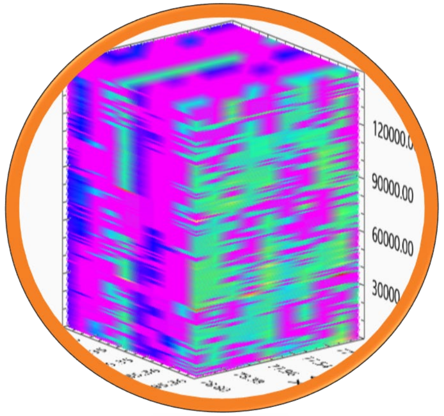
1) a substantial portion of the energy needed for the US to achieve net zero carbon emissions by 2050 and
2) abundant energy worldwide for the foreseeable future.

<u>Vision:</u> The DOE-NE MSR campaign serves as the hub for efficiently and effectively addressing, in partnership with other stakeholders, the technology challenges for MSRs to enter the commercial market.

Salt Chemistry
Determination of the Thermophysical and Thermochemical Properties of Molten Salts – Experimentally and Computationally

Technology Development and Demonstration
Radionuclide Release Monitoring, Sensors & Instrumentation, Liquid Salt Test Loop

Materials
<u>First to Market</u>: Gaps in Codes and Standards for 316H
<u>Near Term Deployment</u>: Use corrosion resistant clad on ASME qualified base metal
<u>Long-Term Solution</u>: Develop and qualify next generation structural materials for MSR
<u>Salt/Graphite Interaction</u>



Modeling
Integral molten salt reactor response to support radionuclide sensor technology development; Integral system analysis to characterize the magnitude and composition of radionuclide transport from a molten salt to different regions of an operating MSR plant.

Involving Scientists and Engineers from 6 US National Labs

US MSR Community = the four Pillars: Government, Industry, National Labs, Universities – Recipe for Success

Nuclear Regulatory Commission is developing a technology-neutral, performance-based, risk-informed regulatory framework

NRC

EERE/NREL
Chloride Salt – Concentrated Solar Power Judith Vidal (POC)

EFRC
Molten Salts in Extreme Environments (MSEE) – 2018-2022

ARPA-E

UNIVERSITIES

University of South Carolina (Prof. Ted Besmann)
UC Berkeley (Prof. Raluca Scarlat)
NEXTRA - Nuclear Energy eXperimental Testing Research Alliance (Prof. Tony Hill)
IRP – MIT (Prof. Charles Forsberg)
NEUP – NEUP review (28-30 JUN 2021) organized by NE-43

MSR Campaign

INDUSTRY

MSR TWG
EPRI
NEI
Campaign Review Meetings
Conferences/MSR workshops

INTERNATIONAL

Gen IV International Forum
IAEA
NEA/OECD

Department of Energy (DOE)-Office of Nuclear Energy (NE) activities focus remains on enabling MSR industry and building supporting infrastructure

Nuclear Energy Advanced Modeling and Simulation (NEAMS) tool development: AMMT, ART, FC, ASI, ARS…
Nuclear Energy University Program (NEUP) (20% of budget)
Small business opportunities: Gateway for Accelerated Innovation in Nuclear (GAIN) vouchers: Direct industry awards.

Cooperation and Collaboration

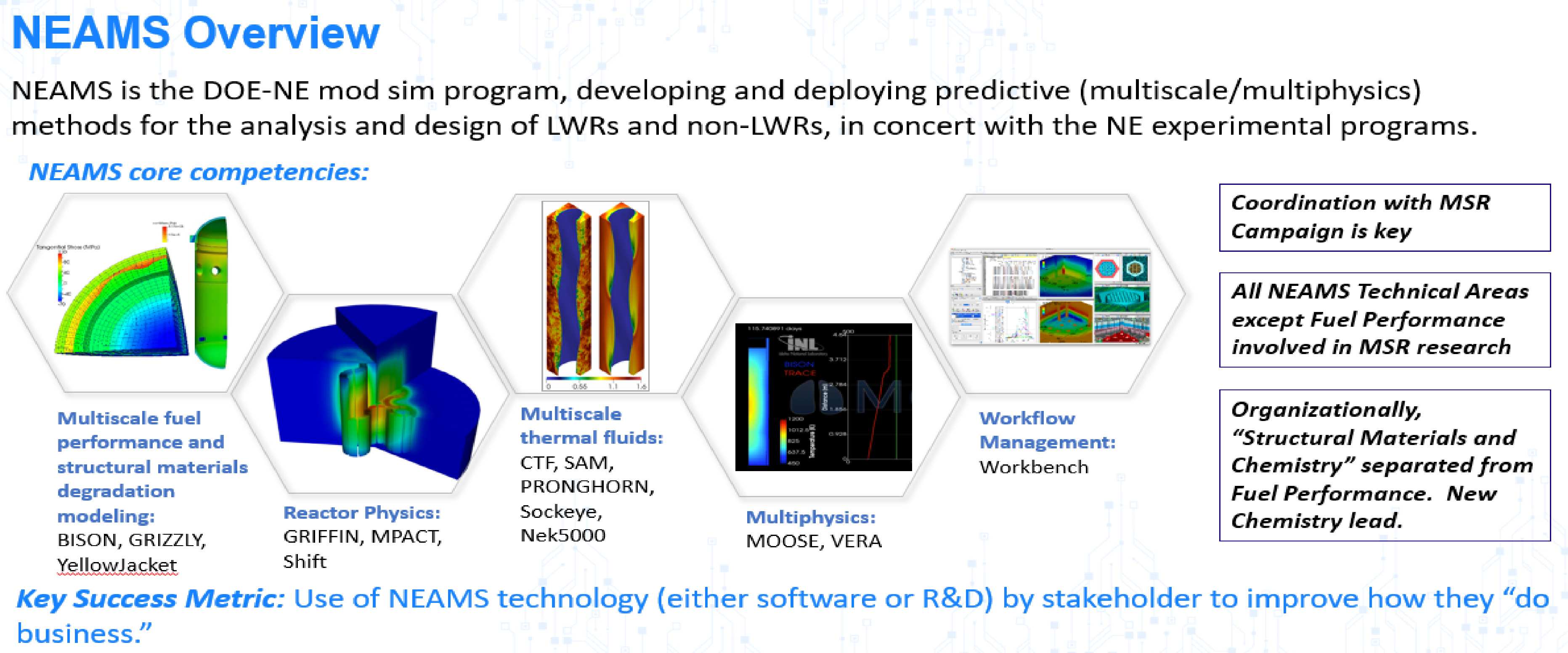
Congress feeds nuclear industry billions to support new reactors and existing fleet

By Jeremy Beaman
November 11, 2021 - 11:00 PM

MSR and NEAMS Campaigns

DOE Molten Salt Reactor (MSR) campaign supports advancing MSR technology by developing technical information and tools for the NRC and industry and performing data and methodology validation experiments under appropriate quality control that are responsive to the data needs of the stakeholders. **Experimentation and modeling and simulation are inextricably linked**.

NEAMS tools are used to rapidly and cost effectively generate property estimates that are subsequently validated by targeted measurements performed within the MSR campaign.



The Molten Salt Thermal Properties Data Base (MSTPDB)

MSR Campaign

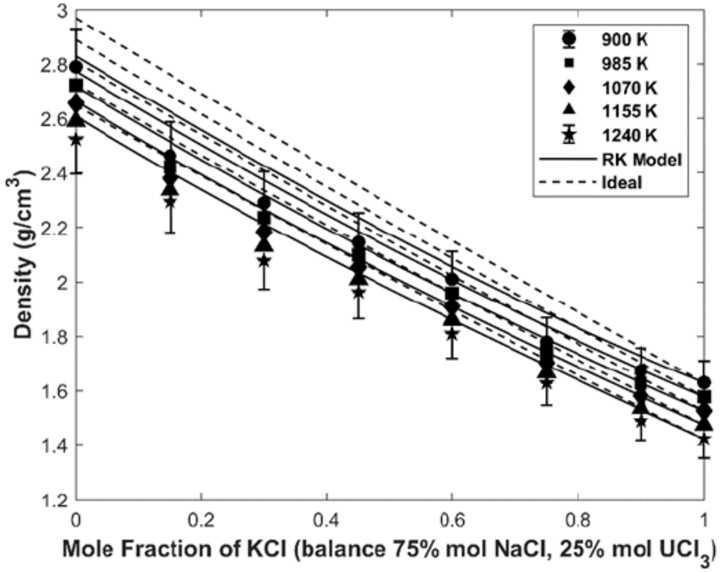
Pure and pseudo binary salts properties as a function of temperature and composition

NEAMS Campaign

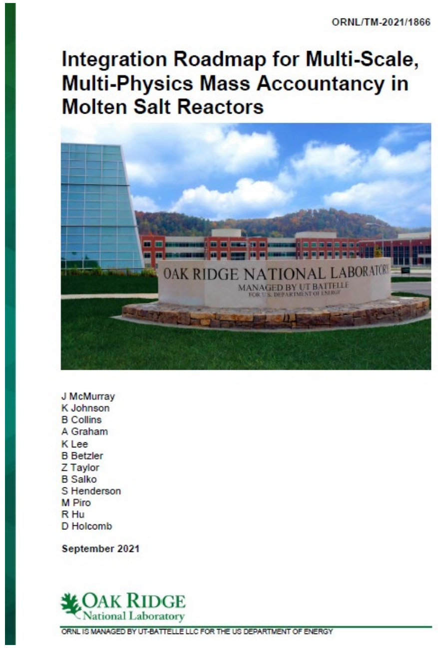
MSTPDB – Estimation of data-based property and phase equilibria of multicomponent salt mixtures
AIMD and MD

MSTDB- TP

MSTDB- TC



Calculated density for the pseudo-ternary NaCl-KCl-UCl_3 system extrapolated from the pseudo-binaries


ORNL/TM-2021/1866

Integration Roadmap for Multi-Scale, Multi-Physics Mass Accountancy in Molten Salt Reactors

J McMurray
K Johnson
B Collins
A Graham
K Lee
B Betzler
Z Taylor
B Salko
S Henderson
M Piro
R Hu
D Holcomb

September 2021

Oak Ridge National Laboratory

ORNL IS MANAGED BY UT-BATTELLE LLC FOR THE US DEPARTMENT OF ENERGY

Integration Roadmap for Multi-Scale, Multi-Physics Mass Accountancy in Molten Salt Reactors/ ORNL/TM-2021/1866

MSTDB – TP – Dr. Dianne Ezell

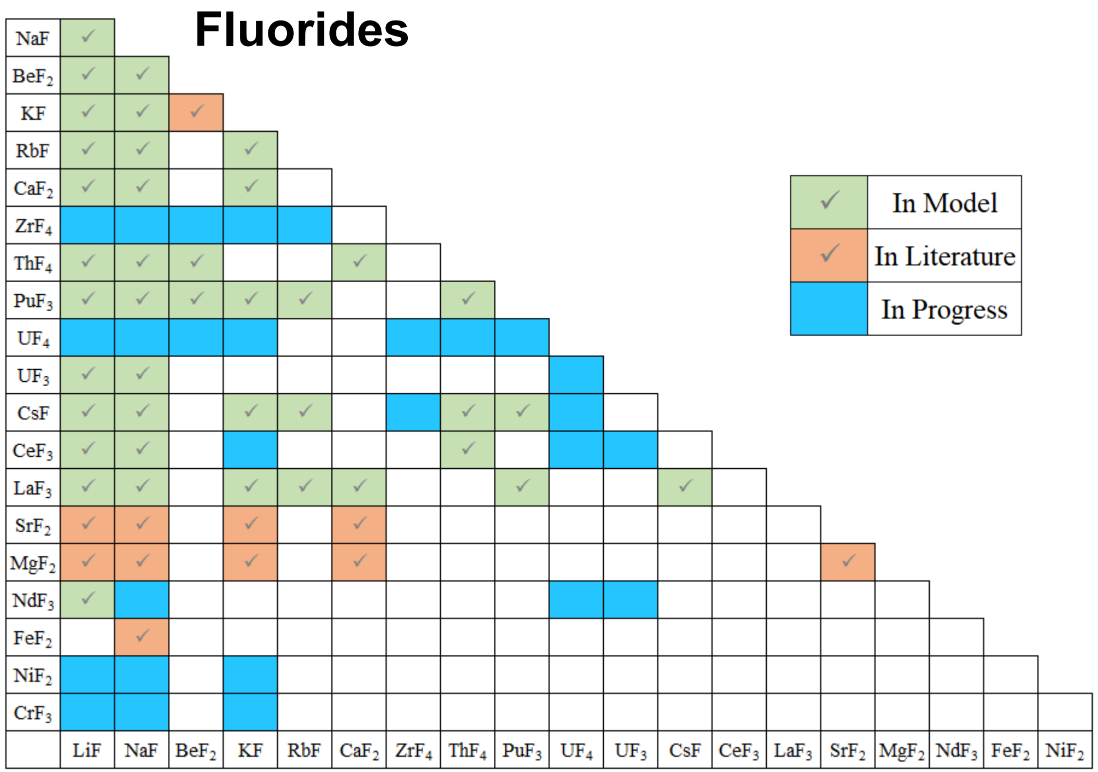
- The MSTDB-TP is a collection of empirical models for representing ρ, ν, κ, and Cp of molten salts as a function of temperature and composition.
- These are required inputs for thermal hydraulics and mass transport models.
- Models are additive based on a mechanical mixture of the pure salt compound constituents with binary interactions only.
- In some cases, when available, ternary or higher order interaction parameters may be included.
- Currently there are 62 entries.
    - 27 are pure compounds (14 fluorides and 13 chlorides),
    - 8 pseudo-binary systems (1 chloride and 7 fluorides),
    - 10 pseudo-ternaries (all of them fluorides)
    - 5 pseudo-quaternaries (all of them fluorides).

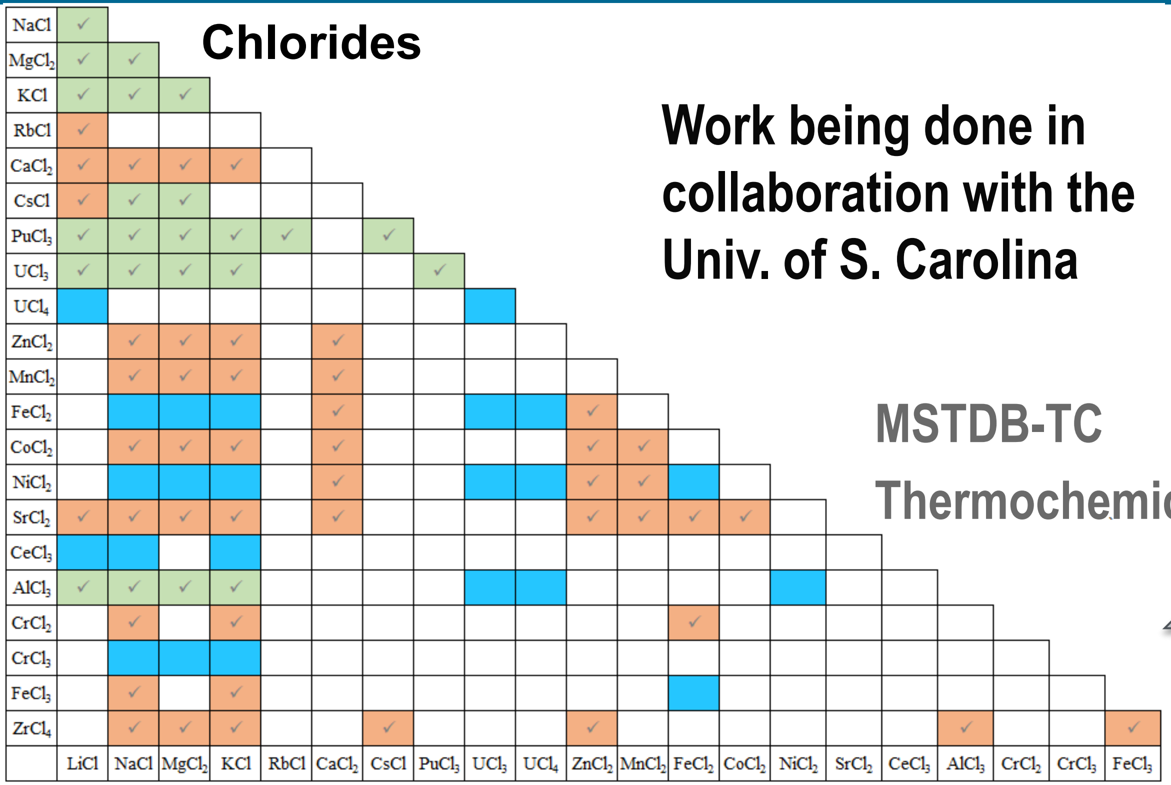
MSTDB – TC – Prof. Ted Besmann

- The Calculation of Phase Diagram (CALPHAD) method is the accepted approach for thermodynamic modeling and database development within the materials community.
- The MSTDB-TC is developed from existing literature, newly generated experimental data primarily from the MSR campaign, and computational derived properties using classical or ab-initio Molecular Dynamics.
- An accompanying data package documenting the source of every model and/or the raw data used for its continuing development is included.
- Currently, it accommodates
    - at least 21 elements and models for 63 pseudo-binary (47 fluoride, 16 chloride)
    - 29 pseudo-ternary (28 fluoride, 1 chloride) molten salt solutions along with 26 solid solutions and 89 stoichiometric compounds.

Cf. to their presentation during the workshop…

Salt Chemistry - Property measurements relevant to industry needs → support through the development of MSTDB (Molten Salt Thermal Properties Database) – ORNL, ANL, INL, PNNL, LANL





Work being done in collaboration with the Univ. of S. Carolina

MSTDB-TC
Thermochemical

MSTDB-TP
Thermophysical

	BeF2	KF	LiF	NaF	ThF4	UF3	UF4
KF	B						
LiF	A, B, D	A, B, C, D					
NaF	A, B	A, B, C	A, B, C, D				
ThF4		A, B	A, B, D	A, B			
UF3							
UF4		A, B	A, B	A, B			
ZrF4			A	A, B, D			

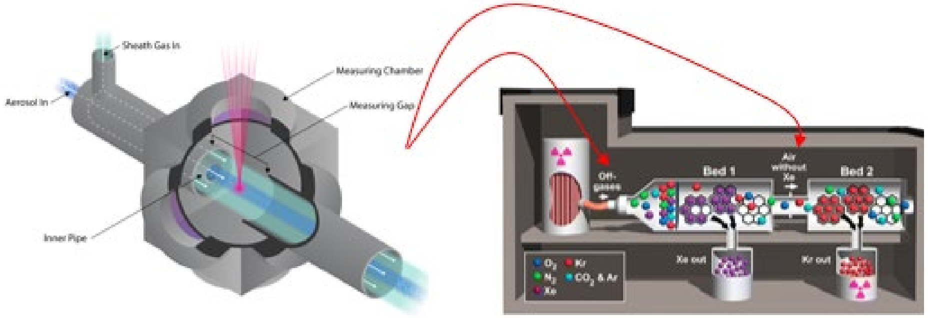
Properties modeled
A: Density
B: Viscosity
C: Thermal conductivity
D: Heat capacity

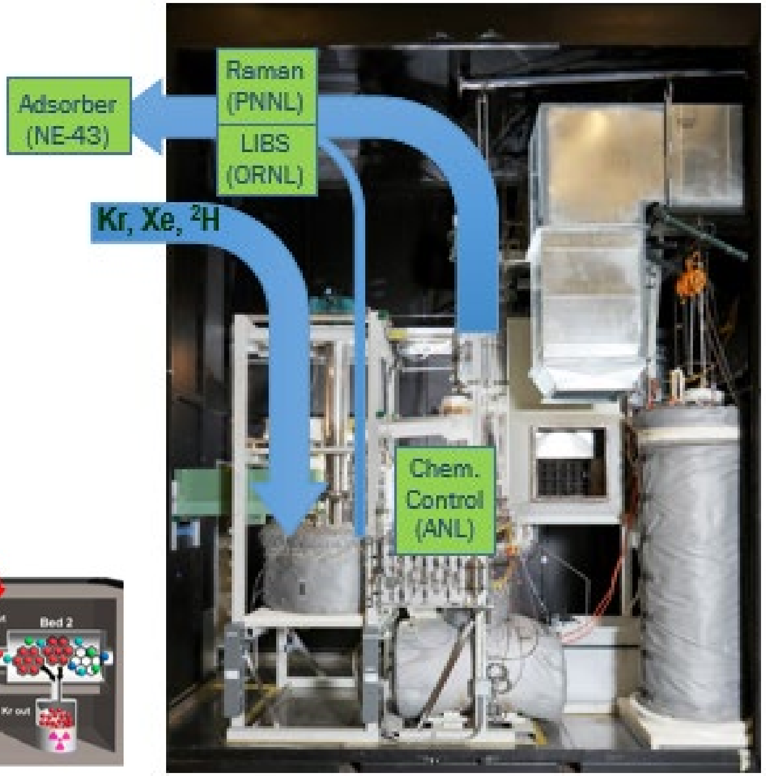
	AlCl3	KCl	LiCl	MgCl2	NaCl	PuCl3	ThCl4	UCl3	UCl4
KCl	A, B								
LiCl	A	A, B, D							
MgCl2		A, B, D	A, B						
NaCl	A, B	A, B, D	A, D	A, B					
PuCl3									
ThCl4		A			A				
UCl3		A, B	A		A, B				
UCl4		A, B	A	A	A, B				
ZrCl4		A							

Technology Development and Demonstration

GOALS: Design and evaluate technologies to mitigate radionuclide release from MSRs.
Assist in the development of analyses and models of fission product release.

PNNL/ORNL Xenon Radionuclide Release and Monitoring using Laser Induced Breakdown Spectroscopy



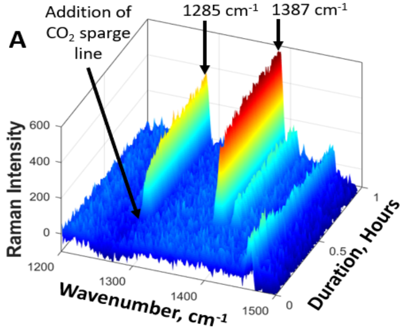


Liquid Salt Test Loop @ ORNL: Existing & operable salt test facility which is unique in the U.S. for technology development and demonstration under MSR-relevant powers, temperatures, and flowrates

ANL - Distributed salt chemistry monitoring and control

Liquid Salt Test Loop at ORNL with installation locations for planned Argonne-developed salt monitoring and control system

PNNL– Raman and FTIR sensor development for iodine species and tritium



Salt spill testing (ANL)

Perform laboratory-scale tests focusing on key processes using salt (F and Cl) bearing U and surrogate fission products to generate insights and data needed to derive accident scenario models

- Develop methods that can be used in large-scale integrated process testing

Four test methods to generate data addressing specific processes:

Spreading and Heat Transfer Tests

- Leading edge vs. time
- Covered area vs. time
- Temperature of steel and salt surfaces vs. time

Flowing and Freezing in Tubing Tests

- Temperature of tubing surface during salt draining
- Locations where tubes of different diameter plug with salt

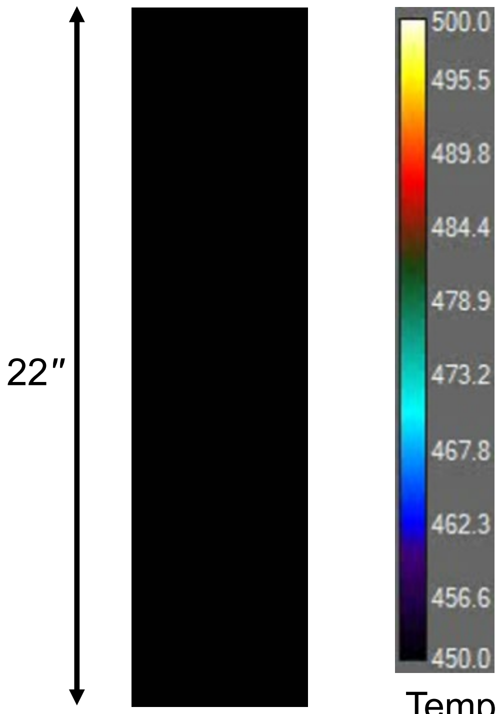
Splashing and Aerosol Generation Tests

- Compositions and sizes of splatter and aerosols generated by splashing

Corrosion Tests in Molten Salt

- Electrochemical corrosion rates at fixed redox, salt chemistry, and temperature

Infrared video



Temp °C

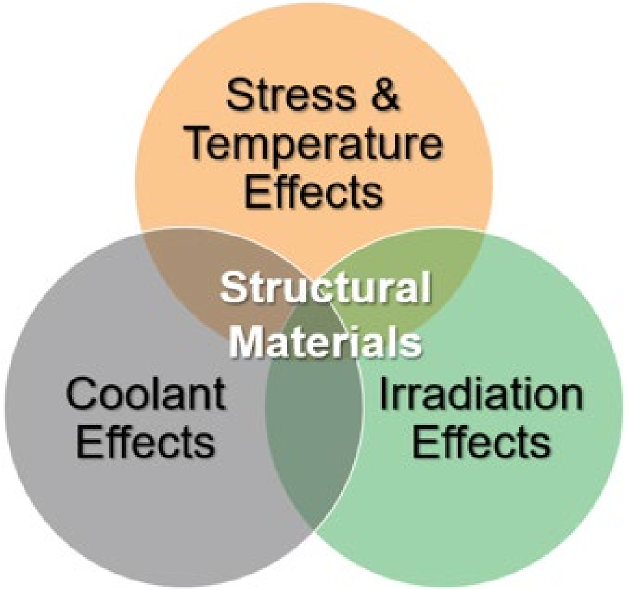
500 °C: FLiNaK (40 g) flowing and freezing on 316 stainless steel tilted by 2.5° in argon atmosphere

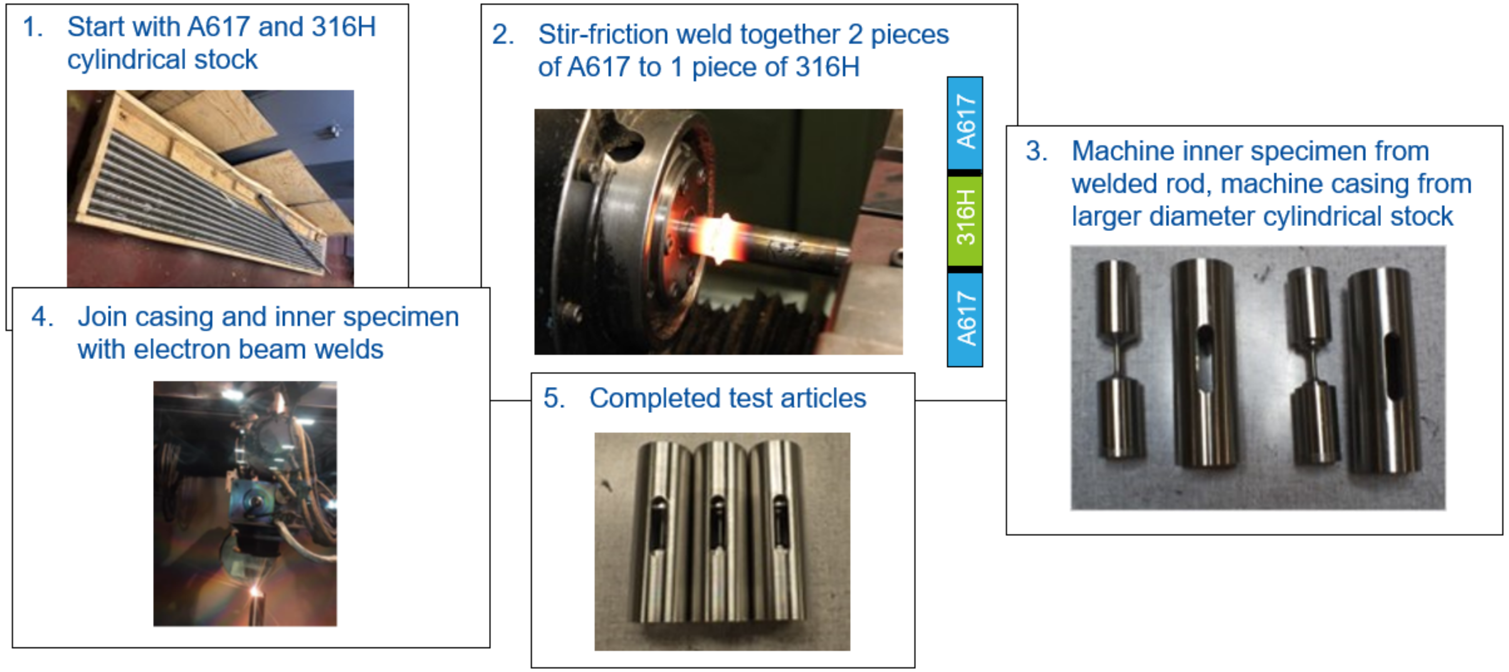
- Tests provide gravity-driven flow and freezing behavior of molten material across surfaces
- Data used in MELTSPREAD model developed for corium spreading and modified for molten salts

Advanced Materials - INL

Materials Degradation During Advanced Reactor Operations

- Information on materials degradations during advanced reactor operations is limited
- Effects of materials degradations during reactor operations are synergistic, involving:
  - Irradiation, corrosion, elevated temperature exposure and stress (creep-fatigue loading)
- Establishment of surrogate materials surveillance program for the management of materials degradations would be an important pathway in support of the timely licensing of advanced reactors





Advanced Materials

Salt and Material Interaction (ORNL)

Conduct 316H thermal convection loop experiments with a peak temperature of ≥700 C for at least 1000 h with flowing salt to demonstrate the applicability of a corrosion model for long-term flowing salt environments based on the results and facilitate modeling the performance of stainless steels in normal and off-normal (higher temperature) conditions.

2021 ORNL FLiBe Thermal Convection Loop

Graphite-Salt Interactions (ORNL)

Evaluation of the performance of various graphite grades in molten salt environments and study of

- the graphite-salt chemical interactions that may affect structural or physical properties of graphite
- the wear and erosion behavior of graphite in molten salt
- the potential of graphite for tritium retention

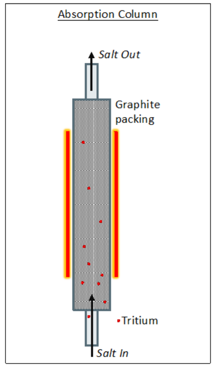
FLINAK Intrusion system

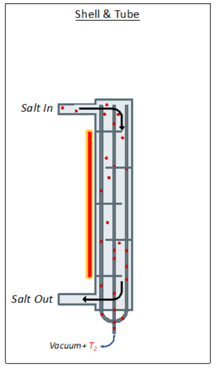
Expanding to handle Be

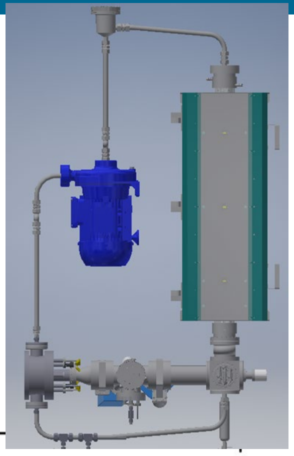
Gloveboxes for Be work

Tritium Transport and Control in MSRs (INL)

Goal: Validate models and provide mitigation strategies for tritium transport in MSRs to support licensing

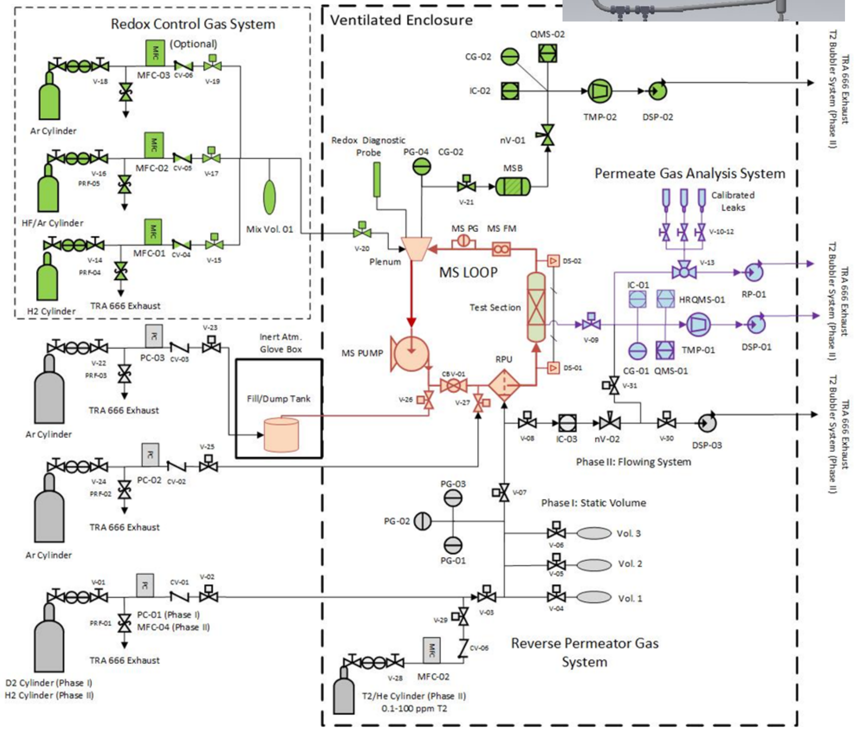






Objectives:

- Develop comprehensive theoretical framework for tritium transport in MSRs
- Measure unknown model parameters in separate-effect tests
- Execute a semi-integral experiment to provide validation data in relevant transport regimes



MSTTE:

- **M**olten **S**alt **T**ritium **T**ransport **E**xperiment (“Misty”)
- Forced-convection FLiBe loop with versatile test section
- Hydrogen isotope (tritium) injection and extraction capability

Courtesy Dr. Paul Humrickhouse and Dr. Thomas Fuerst

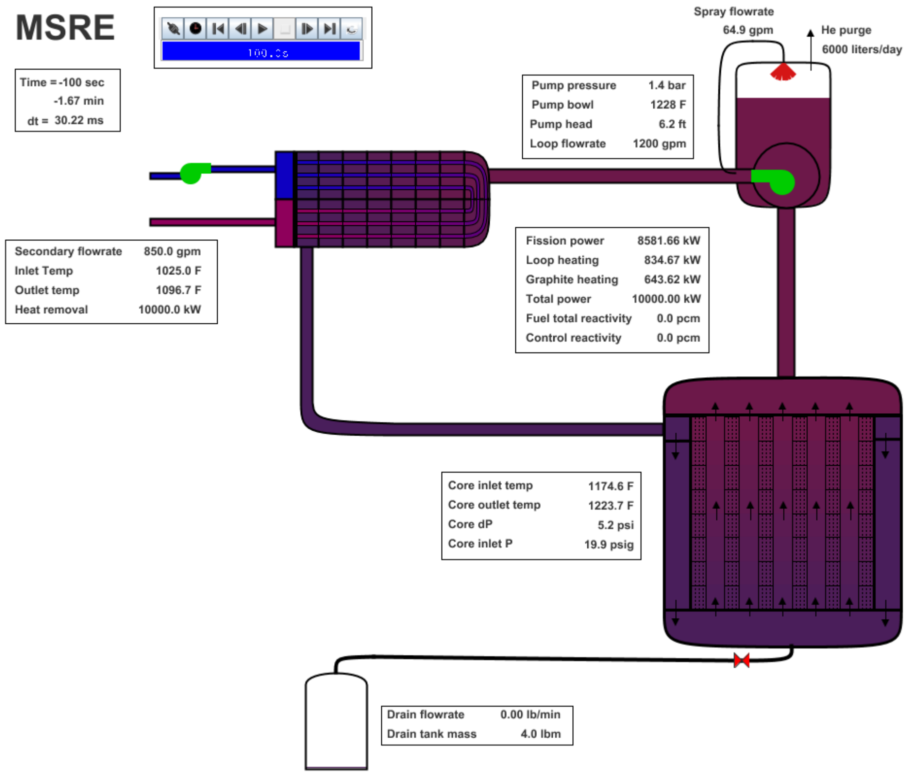
Modeling and Simulation
SNL - MELCOR & MACCS Systems Modeling to Enhance Technology Readiness

Topic 1: Mechanistic Source Term and Consequence Assessments

- Continue to refine the needs of mechanistic source term development and consequence assessment
- Partnering with ANL on MSR salt spill experiments
- Mechanistic source term evaluation for bounding salt spill event, including water release into confinement
- Incorporate model enhancements identified by off-gas system consequence analysis

Topic 2: Modeling and Simulation of Integral Molten Salt Reactor Response to Support Radionuclide Sensor Technology Development

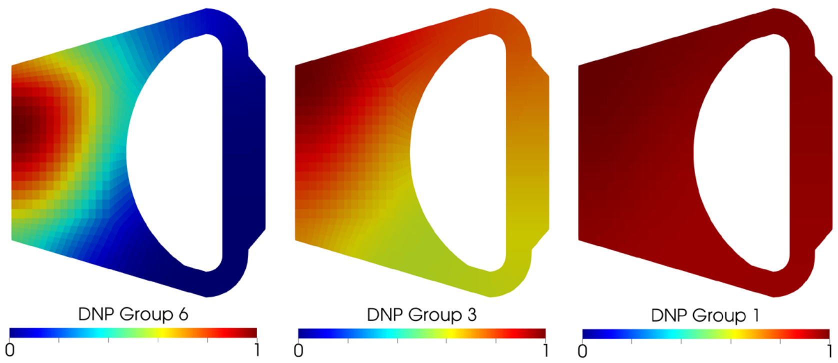
- Apply capabilities recently introduced into MELCOR to characterize radionuclide release and transport in MSRs
- Assumes FLiBe-based salt and prevailing system conditions under operational and anticipated operational transients



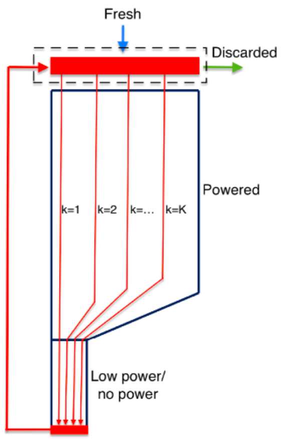
MELCOR MSRE systems model

Modeling And Simulation-MSR Species Tracking Analysis & Prediction - INL

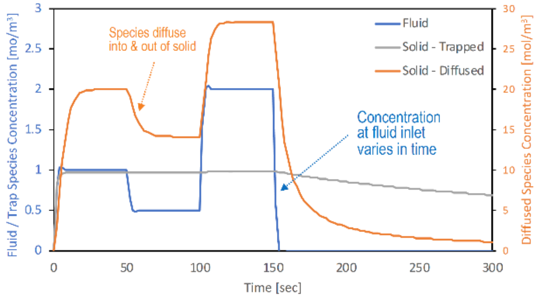
- **Goal**: Generate preliminary evaluation of species transport to off gas vs. heat exchanger vs. vessel vs. salt to guide sensor development work, corrosion assessment, source term models, and decay heat models for MSRs using NEAMS-based tools.
- Leverage 3 ongoing efforts in NEAMS: (1) MSR Griffin+Pronghorn+SAM coupling, (2) MOOSE-based species tracking, (3) Griffin pebble depletion tracking (use same framework but for liquid fuel).
- Activity will collaborate closely with ongoing efforts and focus on high-level application of species tracking + depletion codes to get first order estimate of isotopes in off-gas vs. salt vs. plating
- Initially applied to fast MSR geometry (e.g., MSFR): capability most beneficials to high-burnup designs



Delayed neutron precursor tracking in Griffin + Pronghorn



Pebble depletion tracking in Griffin



SAM tritium transport capability

Relevant References:

- A. Abou-Jaoude *et al.*, “A workflow leveraging MOOSE transient multiphysics simulations to evaluate the impact of thermophysical property uncertainties on molten-salt reactors”, Annals of Nuclear Energy, **163**, 108546, (2021)
- S. Schunert *et al.*, “NRC Multiphysics Analysis Capability Deployment FY 2021 – Part 1”, Idaho National Laboratory, INL/EXT-20-00828, (2020).
- R. Hu *et al.*, “SAM: FY20 Accomplishments”, *NEAMS Thermal Hydraulics Technical Area Deep-Dive*, January 12-13, (2021).
- R. Hu *et al.*, “SAM Developments to Support Transient Safety Analysis of Advanced non-LWRs”, Argonne National Laboratory, ANL/NSE-19/31, (2019)

Fiscal Year 2022 Consolidated Innovative Nuclear Research

PROGRAM DIRECTED: NUCLEAR ENERGY ADVANCED MODELING AND SIMULATION

IRP-NEAMS-1: COMBINED EXPERIMENTAL-MODELING ASSESSMENTS OF IMPURITIES/FISSION PRODUCTS IN MOLTEN SALTS AND FUNDAMENTAL CORROSION MECHANISMS OF RELEVANT STRUCTURAL ALLOYS
(FEDERAL POC – BRIAN ROBINSON & TECHNICAL POC – CHRIS STANEK)
(ELIGIBLE TO LEAD: UNIVERSITIES ONLY)
(UP TO 3 YEARS AND $3,000,000)

During operation of a Molten Salt Reactor (MSR) impurities are present in the salt and furthermore fission products are formed thus affecting the thermophysical properties, corrosion kinetics of structural materials, as well as reactor

PROGRAM DIRECTED: NUCLEAR ENERGY ADVANCED MODELING AND SIMULATION

operations. Proposal are sought to study the impact of halide salt impurities on reactor performance based on experimental and computational methods.

There are several specific topics of interest. First, proposals are sought that address the impact of low impurity concentration on burnup calculations. Specific examples include the impact of low concentrations of impurities on temperature, spectrum and cross-section uncertainties, and reactor operation. Experiments aimed at the identification of specific isotopes are expected be a useful complement to the modeling research. A second topic of interest is salt property alteration with impurity concentrations to ultimately provide recommendations for the allowable concentrations for oxygen, hydroxide, etc. to ensure sound MSR operation, with a focus on heat transfer and containment corrosion. For example, oxide/hydroxide impurities accelerate corrosion rates of halide salt melts in contact with stainless steel and nickel-based alloys. These recommendations generated through the R&D performed should include allowable impurity concentration of the fresh salt melt as well as allowable impurity/fission product concentration during MSR operation.

Vital to the development of impurity limits and guidelines is a better understanding of corrosion mechanisms of austenitic stainless steels, ferritic/martensitic stainless steel, and nickel-based alloys in fluoride and chloride salt melts at 650° C to 750° C. It is currently believed that the corrosion of stainless steel and nickel-based alloys in contact with halide salt melts is derived by the leaching of chromium from the alloy matrix and the formation of stable chromium fluoride or chromium chlorides at the salt-containment interface. The depletion of chromium may weaken the steel structure to further enhance corrosion rates. Overall, corrosion rates of stainless steel and nickel-based alloys could be, as a simplified approach, derived by chromium-self diffusion from the alloy matrices to the surface and the ultimate formation of stable chromium halides. However, overall chromium self-diffusion is a result of matrix and grain-boundary diffusion and their specific ratio is dependent on temperature as well as on microstructure (e.g., phases, precipitates, grain sizes, texture, defects). The proposal should therefore address these and any additional factors to derive an approach for modeling chromium diffusion, and subsequently corrosion of austenitic stainless steels (e.g., Alloy 316H and Alloy 709), ferritic/martensitic stainless steels (e.g., HT9, T91) and nickel-based alloys (e.g., Hastelloy N and Haynes 244). This research will provide a fundamental theoretical and experimental basis for enhancing the current knowledge on corrosion of austenitic stainless steels (fcc) vs. ferritic/martensitic stainless steel (bcc), vs. the more expensive nickel-based alloys in contact with halide salt melts at temperatures applicable to MSR operation.

Proposals are sought that employ integrated experiments and modeling/simulation to address the above problems. Proposals should rely on NEAMS software or at a minimum provide a clear path for model developed in to NEAMS codes.

Since this research is aimed at the deployment of MSR technology, industry partnership is required.

FY21 CINR Awards funded MSR relevant Projects

- **Design and intelligent optimization of the thermal storage and energy distribution for the TerraPower Molten Chloride Fast Reactor in an integrated energy system**, PI: Prof. Brown, University of Tennessee, Co-PI Dr. Kathryn Huff, University of Illinois, Dr. Jamie Coble, University of Tenessee, Dr. Greenwood, ORNL and Mr. Walter TerraPower. NEUP CT-2

- **Total Mass Accounting in Advanced Liquid-Fueled Reactors, PI:** Dr. L. Raymond Cao, The Ohio State University, Co-PI: Dr. Praneeth Kandlakunta – The Ohio State University; Dr. Shelly Li – University of Utah, NEUP CT-4

- **High-Efficiency Electrochemical Test Facility for Corrosion and Hydrodynamic Analysis in Molten Salts, PI: Prof.** Devin Rappleye, Brigham Young University , NEUP General Scientific Infrastructure

- **Real-Time *In Situ* Characterization of Molecular and Complex Ionic Species in Forced-Flow Molten Salt Loops and a Molten Salt Research Reactor, PI:** Kim Pamplin, Ph.D., Abilene Christian University, Co-PI Timothy Head, University of Illinois Urbana-Champaign; Jessie Dowdy, Aaron Robison, and Rusty Towell - Abilene Christian University NEUP General Scientific Infrastructure

- **High-Temperature Molten Salt Irradiation and Examination Facility for the Penn State Breazeale Reactor** , PI: Prof. Amanda Johnsen –Pennsylvania State University , Co-Pi from Pennsylvania State University, NEUP Research Reactor Upgrades Infrastructure Support

The projects would be integrated with the campaign if the campaign had either significant input into the project selections or adequate funding. The efforts are largely separate and not strongly related. However, I am encouraging the PIs to reach out to me and keep me informed.

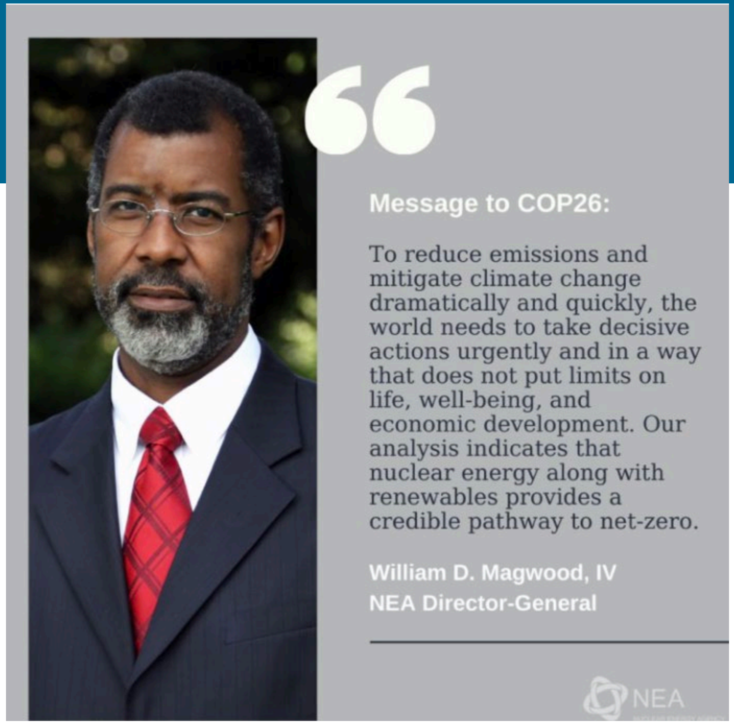
Conclusion

- Despite of a small FY 2022 budget, the MSR campaign is recognized for supporting the MSR community with an increase in collaboration with other DOE NE programs such as NEAMS.
- The MSR campaign serves as the hub for efficiently and effectively addressing, in partnership with other stakeholders, the technology challenges for MSRs to enter the commercial market.
- MSR concepts are on a fast track, and we need to continue with this momentum.
  - MSR Advanced Reactor Demonstration Projects

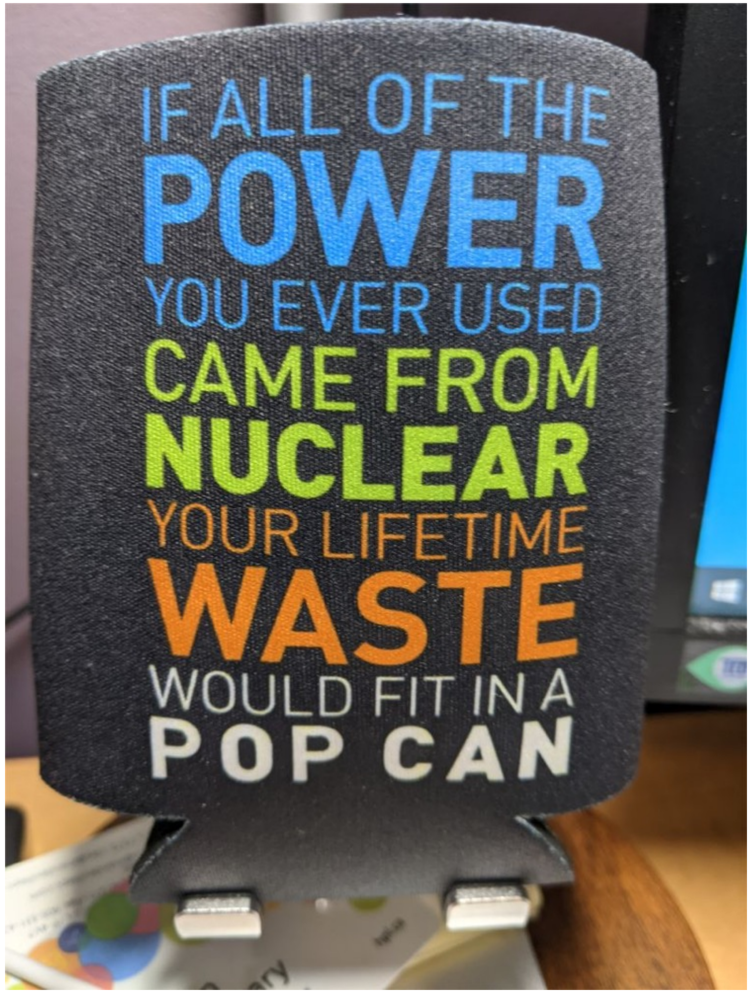
    Kairos Power
    - Hermes test reactor - reduced scale FHR pebble bed test reactor
      - East Tennessee Technology Park (adjacent to ORNL)
      - License Application End 2021 – Construction start 2023 – Operation 2026

    Southern Company Services
    - Molten Chloride Reactor Experiment – fast spectrum
      - Integrated effects test facility anticipated to be operational in 2022
      - Provide data to support the development of TerraPower's molten chloride fast reactor

Questions?



Clean. **Reliable. Nuclear.**



Dr. Patricia Paviet

Patricia.Paviet@pnnl.gov

Overview of the DOE-NE Nuclear Energy Advanced Modeling and Simulation (NEAMS) Program:

(with an emphasis on salt chemistry activities)

Christopher Stanek
Nuclear Energy Advanced Modeling and Simulation Program National Technical Director
Los Alamos National Laboratory
stanek@lanl.gov

Jake McMurray
Oak Ridge National Laboratory

David Andersson
Los Alamos National Laboratory

U.S. DEPARTMENT OF ENERGY

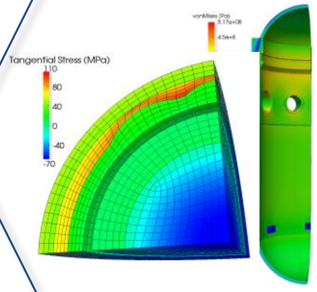
Molten Salt Thermal Properties Workshop
July 14, 2020

DOE-NE NEAMS Program Overview

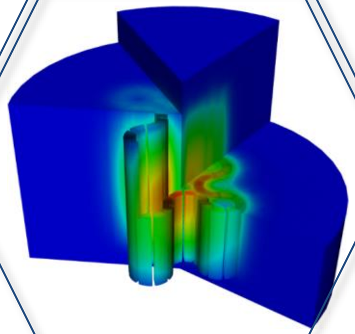
The NEAMS program is a multilab team effort that aims to develop and deploy predictive computer methods for the analysis and design of advanced nuclear reactors. With integration of CASL, LWRs also included in program scope.

NEAMS is the mod-sim relative of the ART-MSR Campaign led by Lou Qualls.

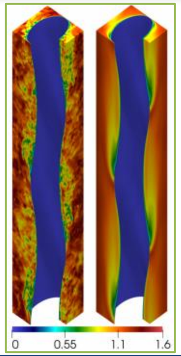
NEAMS core competencies:



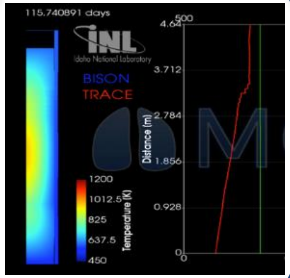
Multiscale fuel performance and structural materials degradation modeling:
BISON, GRIZZLY



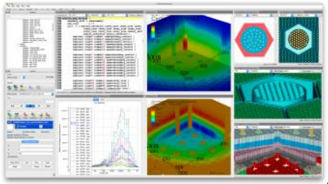
Reactor Physics:
GRIFFIN



Multiscale thermal fluids:
SAM,
PRONGHORN,
Sockeye,
Nek5000



Multiphysics:
MOOSE



Workflow Management:
Workbench

Hallmarks of **advanced mod-sim** are *"multiscale"* and *"multiphysics"* – with the specific goal to be *predictive* through mechanistic insight. Especially important for data poor regimes.

Preferred use of advanced mod-sim is in concert with experiments to solve challenging problems.

DOE-NE Expectation of NEAMS: *Relevance to Industry and NRC*

A key priority (and challenge) for NEAMS is to strike a balance between early stage R&D and industrial relevance.

"I am writing to convey our full support for the Nuclear Regulatory Commission (NRC) adoption and use of the modeling and simulation codes developed by the Department of Energy (DOE), Office of Nuclear Energy (NE) over the last decade."

"Our principal success metric for these programs is the adoption and use of these codes by U.S. industry and NRC for commercial applications."

DOE guidance has catalyzed proactive engagement with the NRC and industry. Continuous exchanges ensure that R&D sweet spots are identified and pursued.

Department of Energy
Washington, DC 20585

April 29, 2019

Dr. Peter Riccardella
Chairman
Advisory Committee on Reactor Safeguards
U.S. Nuclear Regulatory Commission
11545 Rockville Pike
Rockville, MD 20852-2738

Dear Dr. Riccardella:

I am writing to convey our full support for the Nuclear Regulatory Commission (NRC) adoption and use of the modeling and simulation codes developed by the Department of Energy (DOE), Office of Nuclear Energy (NE) over the last decade. The Department has invested significant resources to develop a substantial suite of state-of-the-art multi-physics modeling capabilities for light water reactors under the Energy Innovation Hub for Modeling and Simulation program and for advanced reactors under the Nuclear Energy Advanced Modeling and Simulation program. Our principal success metric for these programs is the adoption and use of these codes by U.S. industry and NRC for commercial applications.

As I publically stated at the August 21, 2018, meeting of the Advisory Committee on Reactor Safeguards Subcommittee on Thermal-Hydraulic Phenomena, I am confident that the DOE NE modeling capabilities are not only useful for reactor design but will ultimately prove useful to NRC in the staff's confirmatory analysis efforts, particularly for advanced reactor designs that do not have the benefit of decades of operational experience and highly refined safety analysis tools.

The Department is very supportive of NRC's Comprehensive Reactor Analysis Bundle (CRAB) vision for advanced reactor design basis event (DBE) analysis (NRC Non-Light Water Reactor (Non-LWR) Vision and Strategy, Volume 1 – Computer Code Suite for Non-LWR Design Basis Event Analysis, ML19093B322). NRC's use of the DOE tools as described in the report is consistent with DOE's vision for use of our codes by NRC. The methodical process by which NRC came to its independent conclusion on CRAB was rigorous and is influencing the prioritization of DOE development efforts going forward.

We understand that any tool used for safety analysis must adhere to strict software quality assurance (SQA) standards and must be validated for the desired application. The Department is committed to rigorous SQA and working with NRC to clearly identify the validation activities needed for each of several advanced reactor design types. We are prioritizing those activities that are

necessary and appropriate for the Department to conduct, recognizing that an applicant will likely conduct additional validation activities using their proprietary models and data specific to their design.

The Department has made it a priority to establish and develop strong communication pathways with NRC to help NRC staff familiarize themselves with available modeling and analysis tools, and to facilitate their evaluation of our tools for potential adoption. We understand that adoption of any new modeling and simulation tools will require appropriate training and DOE is committed to providing NRC staff with the necessary training. While future training will certainly be made available to NRC staff, DOE is encouraged by the hands-on involvement to-date of key NRC staff which have already produced key developmental contributions as they become advanced users of DOE codes.

Given the above, I remain confident that NRC will find significant value in the Department's modeling and simulation tools for use in confirmatory and other safety analyses. The Department is unequivocally committed to supporting NRC's adoption and continued use of these codes, and we are prioritizing resources to ensure the codes remain available and responsive to NRC needs.

If you have any further questions or comments, please contact me at your convenience at (301) 903-7808 or shane.johnson@nuclear.energy.gov.

Sincerely,

R. Shane Johnson
Deputy Assistant Secretary for Reactor Fleet and Advanced Reactor Deployment
Office of Nuclear Energy

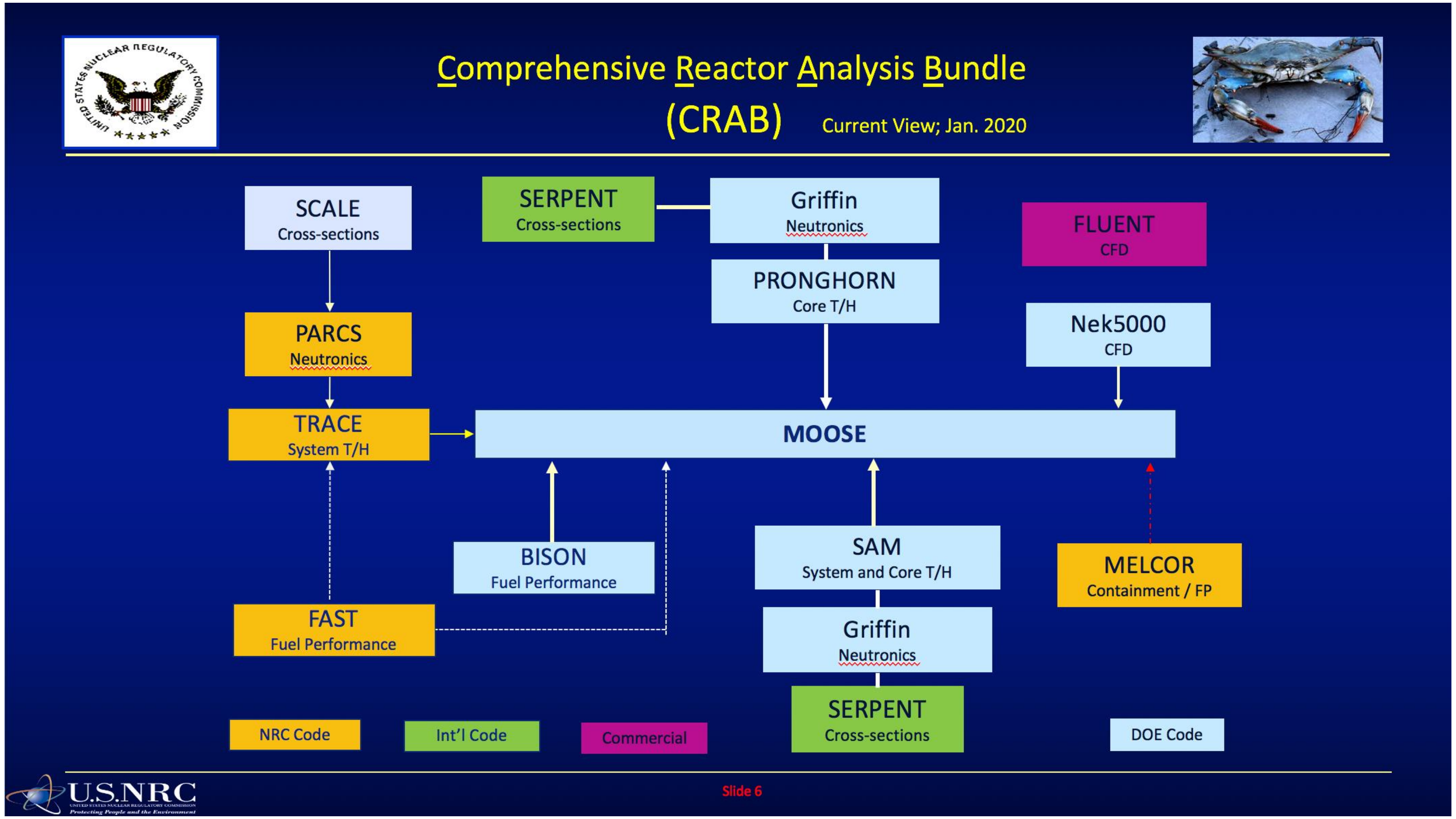
cc: Mr. Matthew Sunseri
Dr. Dennis Bley
Dr. Jose March-Leuba
Dr. Michael Corradini

NEAMS Partnership with NRC: *Comprehensive Reactor Analysis Bundle (CRAB)*

In February, NRC released revised versions of "Non-LWR Vision and Strategy" documents. The Design Basis Events (DBE) volume continues to rely on CRAB concept. **ML20030A176**

NRC-RES has made clear that they can maintain regulatory independence if license applicants use a similar subset of codes. Also, indications of efficiency gains if NRC and vendor use the same set of codes, and NRC has opportunity to do work in advance of license submittal.

NRC has requested a liquid fuel salt modeling capability.



Companies in Pre-Application Discussions with NRC

Three of six non-LWR companies that have notified NRC of their intent to engage in regulatory interactions are "molten salt reactors" (1 FHR and 2 MSR)

Pre-Application Activities

Below is a summary of non-LWR reactor designers that have formally notified the NRC of their intent to engage in regulatory interactions. There are several more potential pre-applicants that participate in various industry activities that could be added to this list once they formally engage with the NRC.

Developer	Pre-application information	RIS response	Technology
General Atomics	General Atomics	N/A	Helium-Cooled Fast Reactor
X-Energy LLC	XE-100	1/16/2018	Modular High Temperature Gas-Cooled Reactor
Kairos Power LLC	Kairos Power Fluoride Salt-Cooled, High Temperature Reactor (KP-FHR)	3/14/2018	Molten Salt Reactor
Terrestrial Energy USA Ltd	Integral Molten Salt Reactor (IMSR)	1/25/2019	Molten Salt Reactor
TerraPower, LLC	Molten Chloride Fast Reactor (MCFR)	6/4/2017	Molten Salt Reactor
Westinghouse Electric Company	eVinci	6/18/2019	Micro Reactor

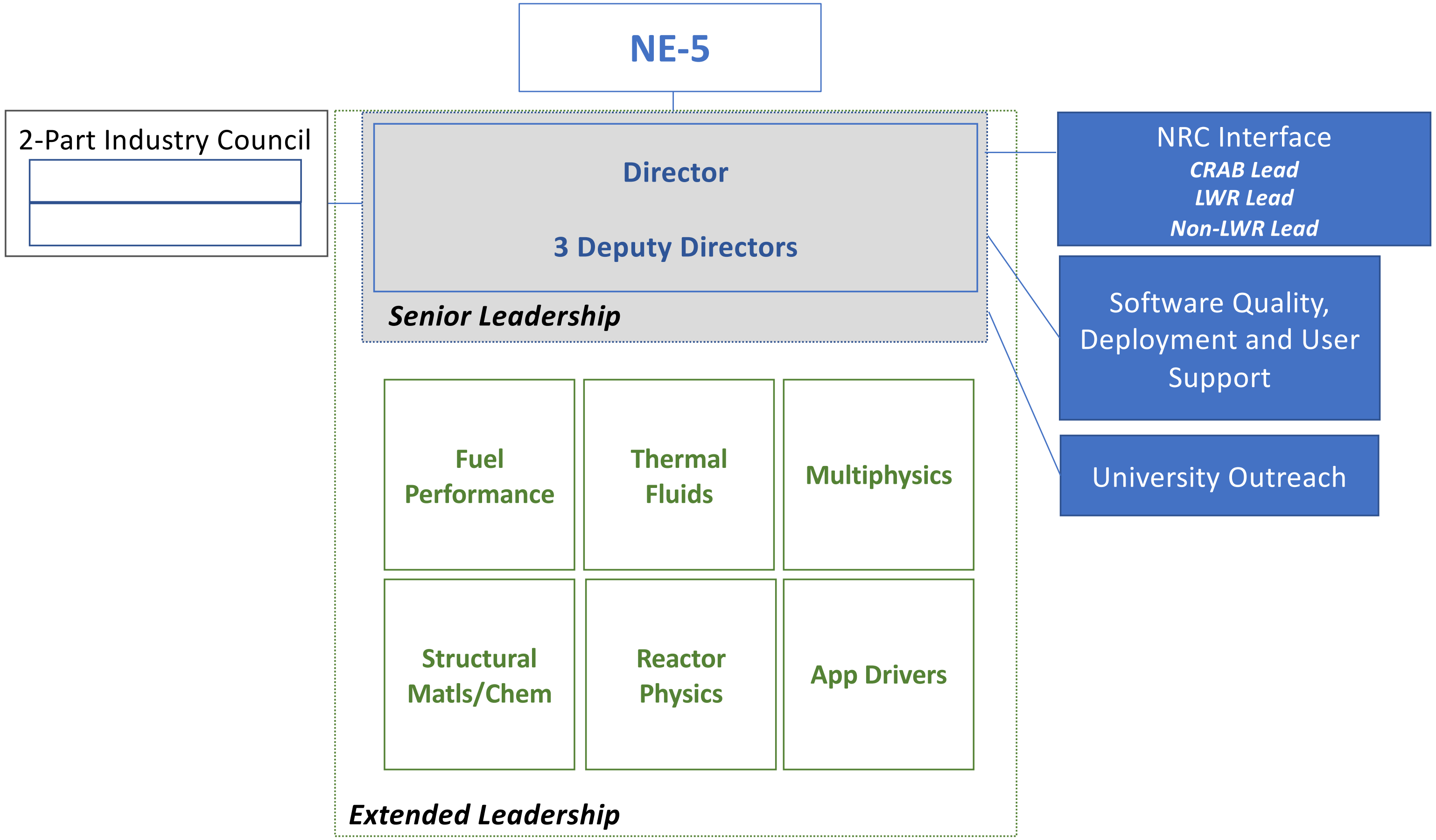
https://www.nrc.gov/reactors/newreactors/advanced.html#preAppAct

NEAMS Interaction with Industry

Primary interaction with advanced reactor industry has been through NEAMS Advanced Reactor Industry Council

Loud and clear guidance for NEAMS to emphasize development of a capability to complement experimental efforts by calculating salt properties (e.g. density, heat capacity, thermal conductivity) and ultimately corrosion.

NEAMS program has responded via effort referred to as YellowJacket. Also integrated CASL and NEAMS program has a new Technical Area: Structural Materials and Chemistry – where Jake McMurray has a leadership role.



Molten Salt Thermodynamic Database (MSTDB)

A particular emphasis of NEAMS program is our contribution to the development to the Molten Salt Thermodynamic Database (MSTDB)

54 pseudo-binary systems			**26 pseudo-ternary systems**	
NaF-LiF	UF_4-NaF	UF_4-ThF_4	LiF-BeF_2-UF_4	NaF-BeF_2-ThF_4
BeF_2-LiF	UF_3-NaF	CsF-ThF_4	LiF-BeF_2-ThF_4	NaF-BeF_2-UF_4
KF-LiF	CsF-NaF	CeF_3-ThF_4	LiF-BeF_2-NaF	NaF-BeF_2-PuF_3
RbF-LiF	LaF_3-NaF	UF_4-PuF_3	LiF-BeF_2-PuF_3	NaF-UF_4-ThF_4
CaF_2-LiF	ThF_4-BeF_2	CsF-PuF_3	LiF-NaF-UF_4	NaF-KF-CaF_2
ThF_4-LiF	PuF_3-BeF_2	LaF_3-PuF_3	LiF-ThF_4-PuF_3	LiF-KF-NaF
PuF_3-LiF	UF_4-BeF_2	RbF-UF_4	LiF-CaF_2-ThF_4	LiF-KF-CsF
UF_4-LiF	RbF-KF	CaF_2-UF_4	LiF-UF_4-PuF_3	LiF-KF-RbF
UF_3-LiF	CaF_2-KF	LaF_3-CsF	LiF-LaF_3-CsF	LiF-KF-CaF_2
CsF-LiF	PuF_3-KF	NaCl-$MgCl_2$	LiF-NaF-LaF_3	LiF-CaF_2-LaF_3
CeF_3-LiF	CsF-KF	NaCl-CsCl	LiF-NaF-RbF	LiF-CeF_3-ThF_4
LaF_3-LiF	LaF_3-KF	NaCl-$PuCl_3$	LiF-NaF-PuF_3	NaF-CaF_2-LaF_3
BeF_2-NaF	PuF_3-RbF	NaCl-UCl_3	LiF-CsF-PuF_3	BeF_2-UF_4-ThF_4
KF-NaF	CsF-RbF	$MgCl_2$-CsCl		
RbF-NaF	LaF_3-RbF	$MgCl_2$-$PuCl_3$		
CaF_2-NaF	ThF_4-CaF_2	$MgCl_2$-UCl_3		
ThF_4-NaF	LaF_3-CaF_2	$MgCl_2$-KCl		
PuF_3-NaF	PuF_3-ThF_4	$PuCl_3$-UCl_3		

NEAMS make a significant investment in university R&D via NEUP/IRP awards (~$6M in FY20), e.g. recent award will expand investment in MSTDB by supporting research at Univ. of South Carolina.

(NEAMS will also closely follow recently awarded IRP "Molten Salt Reactor Test Bed with Neutron Irradiation" at MIT)

NEUP Nuclear Energy University Program
U.S. Department of Energy

Extension of *MSTDB* to Provide a High-Quality, Validated Thermochemical Database for Predicting/Simulating Corrosion in Molten Salt Reactor Systems

PI: Theodore M. Besmann, University of South Carolina

Program: NEAMS

Collaborators: Ming Hu, University of South Carolina
Stephen S. Raiman Oak Ridge National Laboratory
Jacob W. McMurray, Oak Ridge National Laboratory
Ruchi Gakhar, Idaho National Laboratory

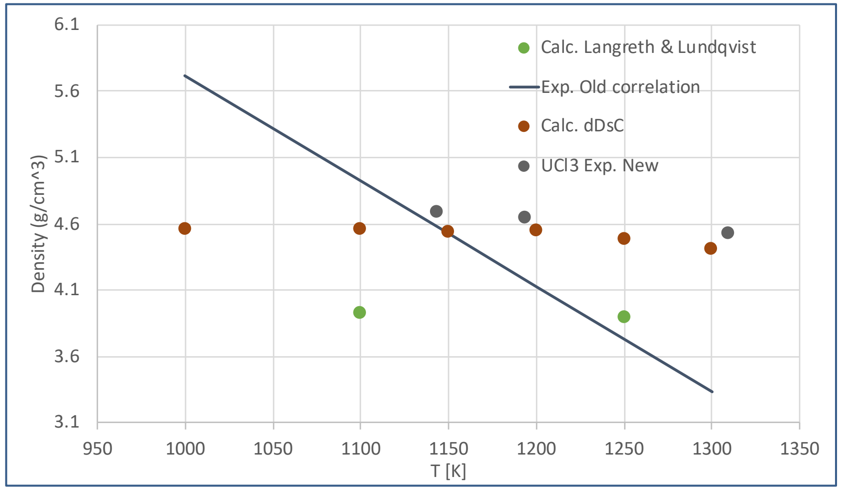
Atomistic simulations for salt properties

DFT employed to simulate chloride fuel salts (near term focus NaCl-UCl_3) and fluoride (e.g. FLiNaK) salts.

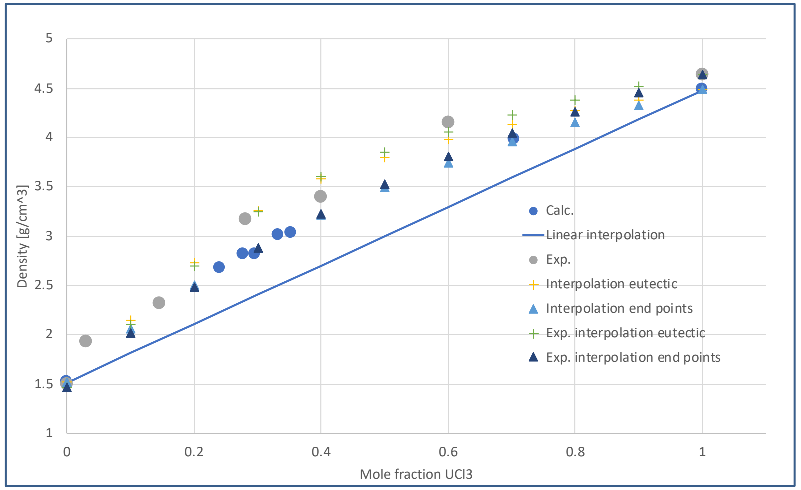
Ab-initio molecular dynamics preferred over classical potential due to ability to treat complex chemistry, but some properties may be computationally too expensive to simulate (e.g. viscosity and thermal conductivity) and empirical potential can be used as a substitute for some cases.

Initial results are encouraging, and provide useful connection to experimental efforts.

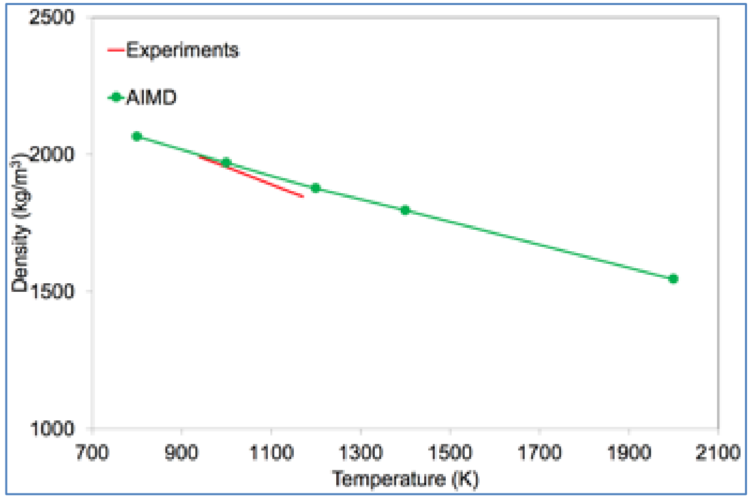
UCl_3 Density



NaCl-UCl_3 Density



FLiNaK (46LiF-12NaF-42KF) Density



David Andersson (andersson@lanl.gov) technical POC for DFT calculations.

Summary

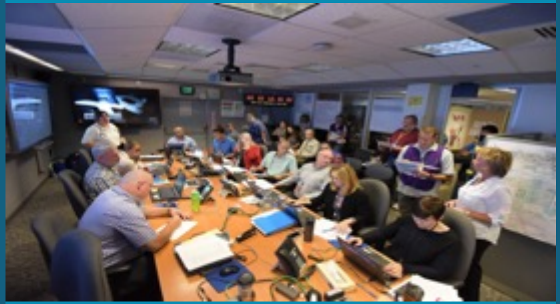
The DOE-NE NEAMS program develops software and performs modeling and simulation R&D for non-LWRs, with an emphasis on performing work of relevance to industry and the NRC.

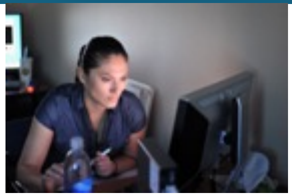
Clear input from NEAMS stakeholders to consider molten salt thermochemistry, thermophysical properties and ultimately corrosion.

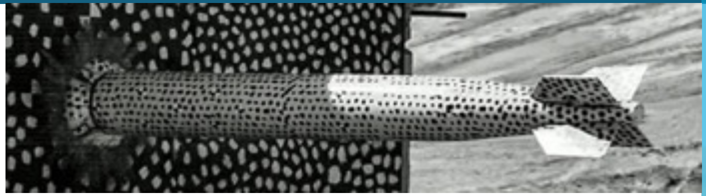
Although the NEAMS program is exclusively mod-sim, the preferred use of mod-sim tools is in collaboration with experimental efforts.

NEAMS is constantly seeking opportunities to optimize the work we perform. One part of a broad research effort to improve our understanding of salt behavior. Very open to ideas for collaboration, specific calculations to be performed, etc.

MELCOR non-LWR Modeling and Simulation Capabilities







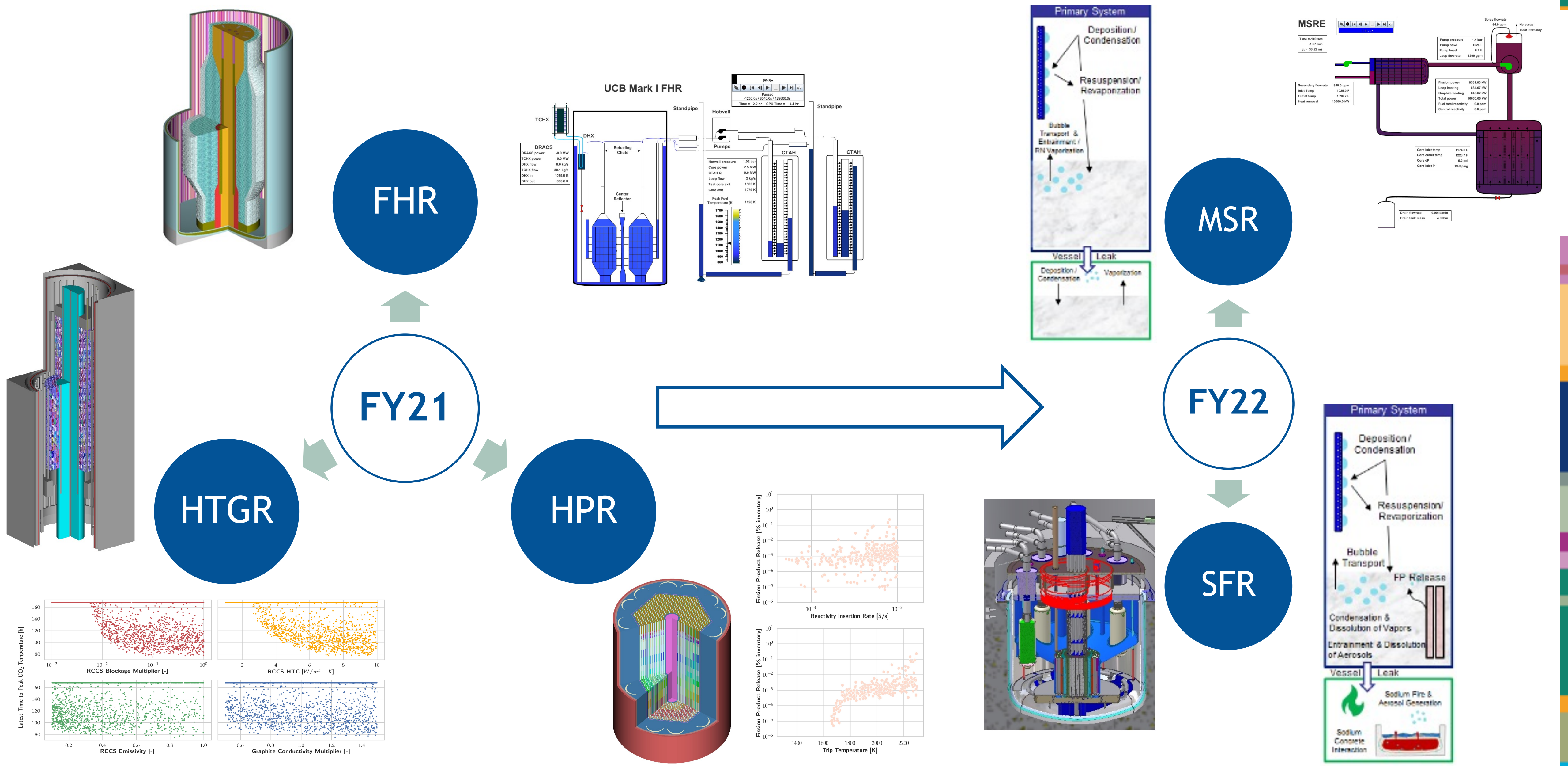
David L. Luxat

Severe Accident Modeling/Analysis Department

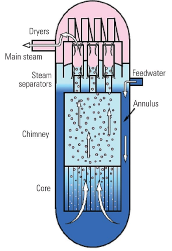
Sandia National Laboratories

Sandia National Laboratories is a multimission laboratory managed and operated by National Technology & Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA0003525.

NRC Analytical Capability Demonstration

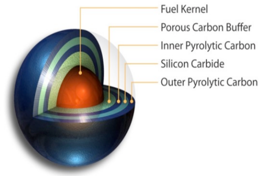


Thermal hydraulics

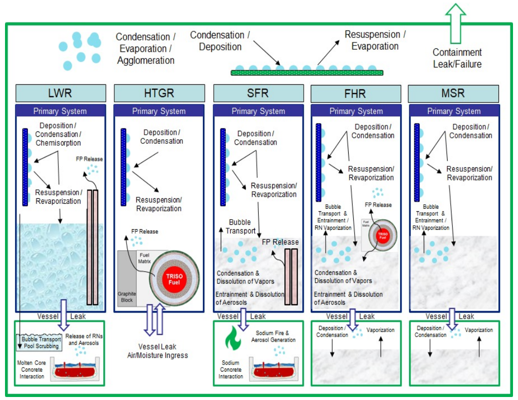


Reactivity Effects

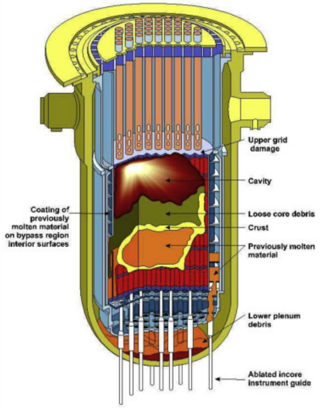
Fuel thermal-mechanical response



Fission product release and transport



Core degradation



Ex-vessel damage progression

MELCOR Molten Salt Reactor Modeling

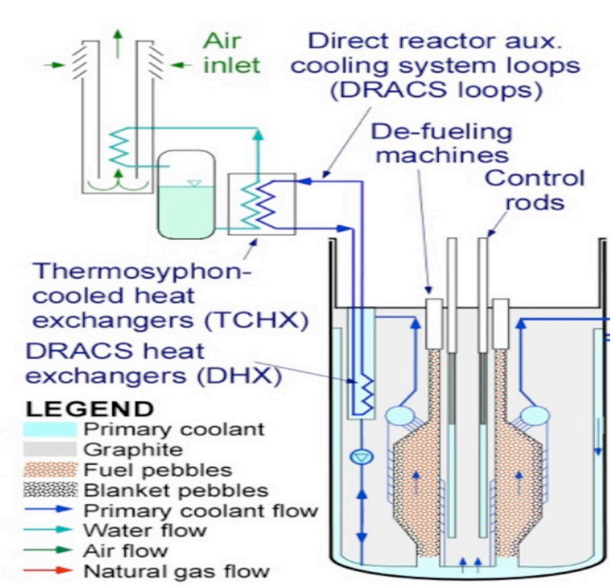
Added molten salt as working fluid

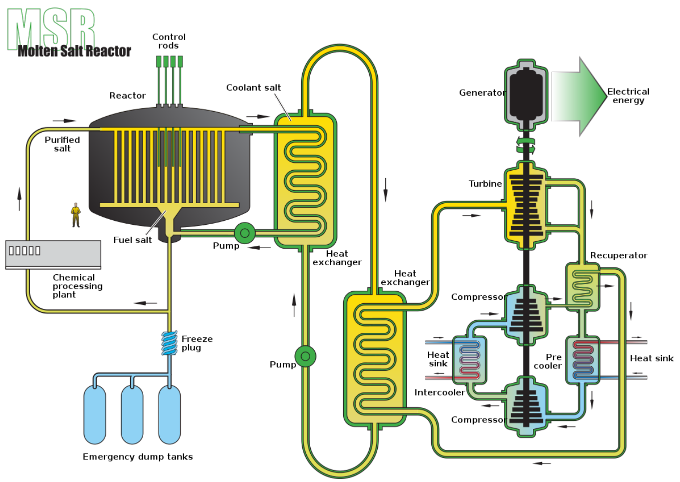
Fission product release
- Release from TRISO kernel
- Radionuclide distributions within the layers in the TRISO particle and compact
- ***Liquid-phase fission product chemistry and transport model***

Additional core models
- Graphite oxidation
- Inter- and intra-cell energy transport
- Convective energy transport
- Fluid mass transport in core

Fluid fuel point kinetics (liquid-fueled molten salt reactors)

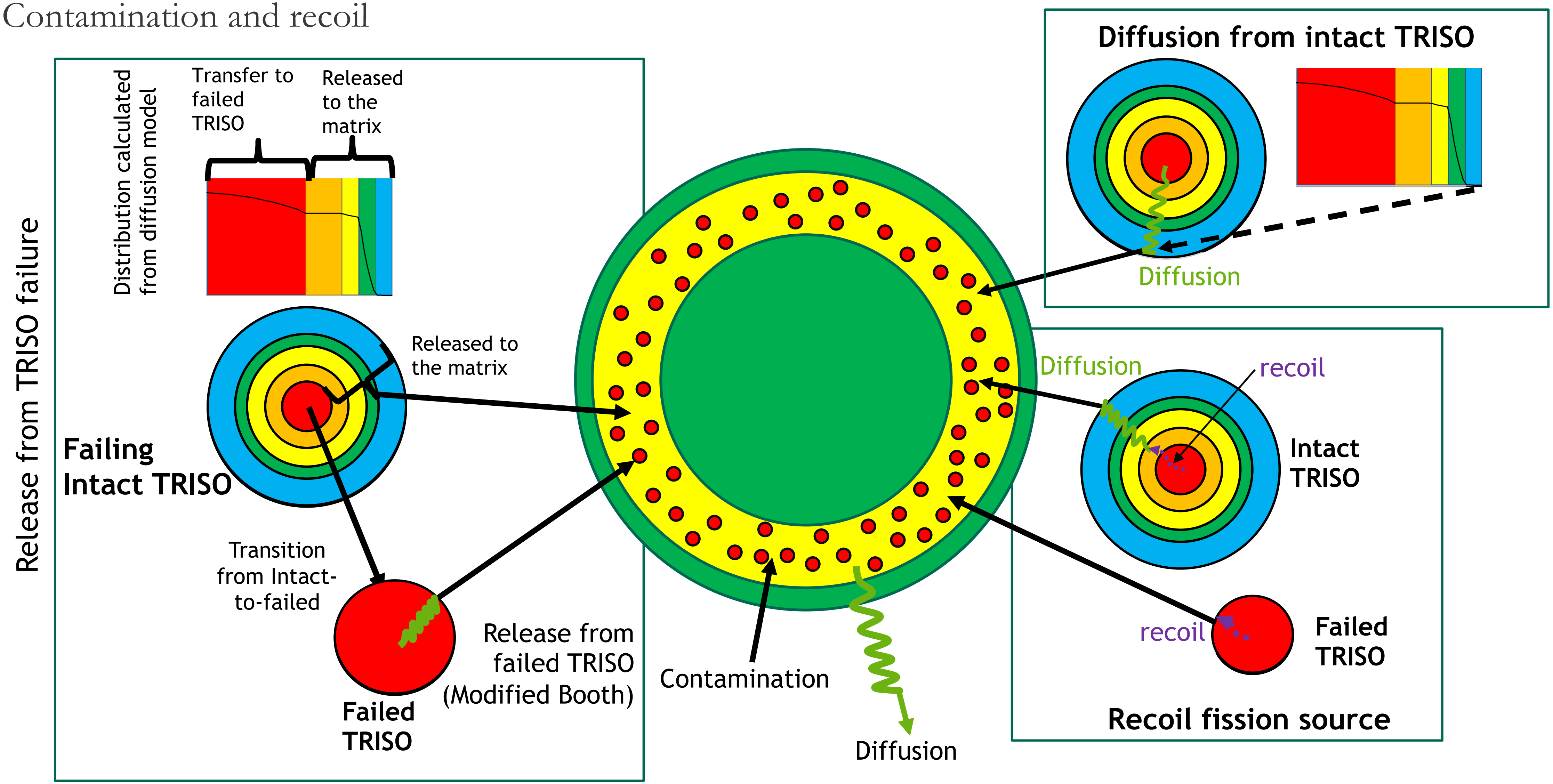
Salt spill modeling





Radionuclide Release Models

Recent failures – particles failing within last time-step (burst release, diffusional release in time-step)
Previous failures – particles failed at a previous time-step (time history of diffusional release)
Contamination and recoil



Molten Salt Chemistry and Radionuclide Release

Model Scope

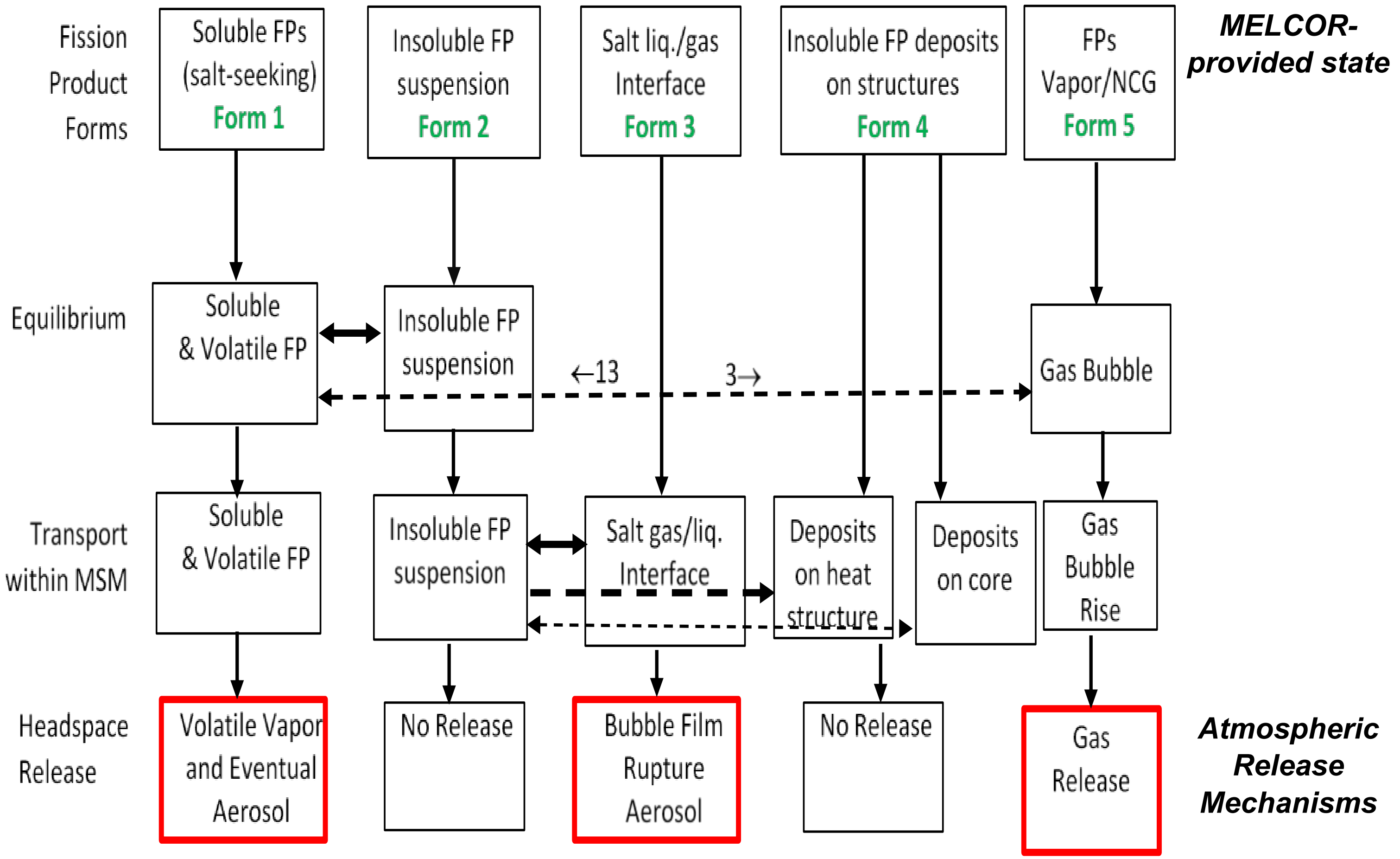
Evaluation of thermochemical state

- Gibbs Energy Minimization with Thermochimica
- Provides solubilities and vapor pressures

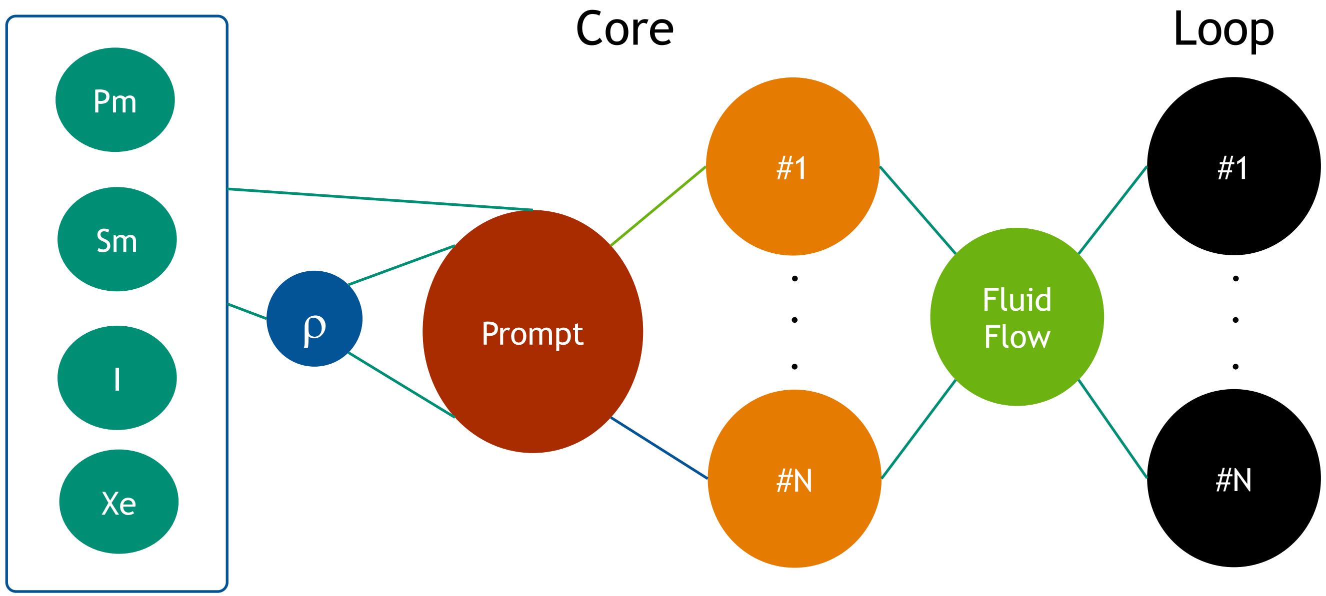
Thermodynamic database

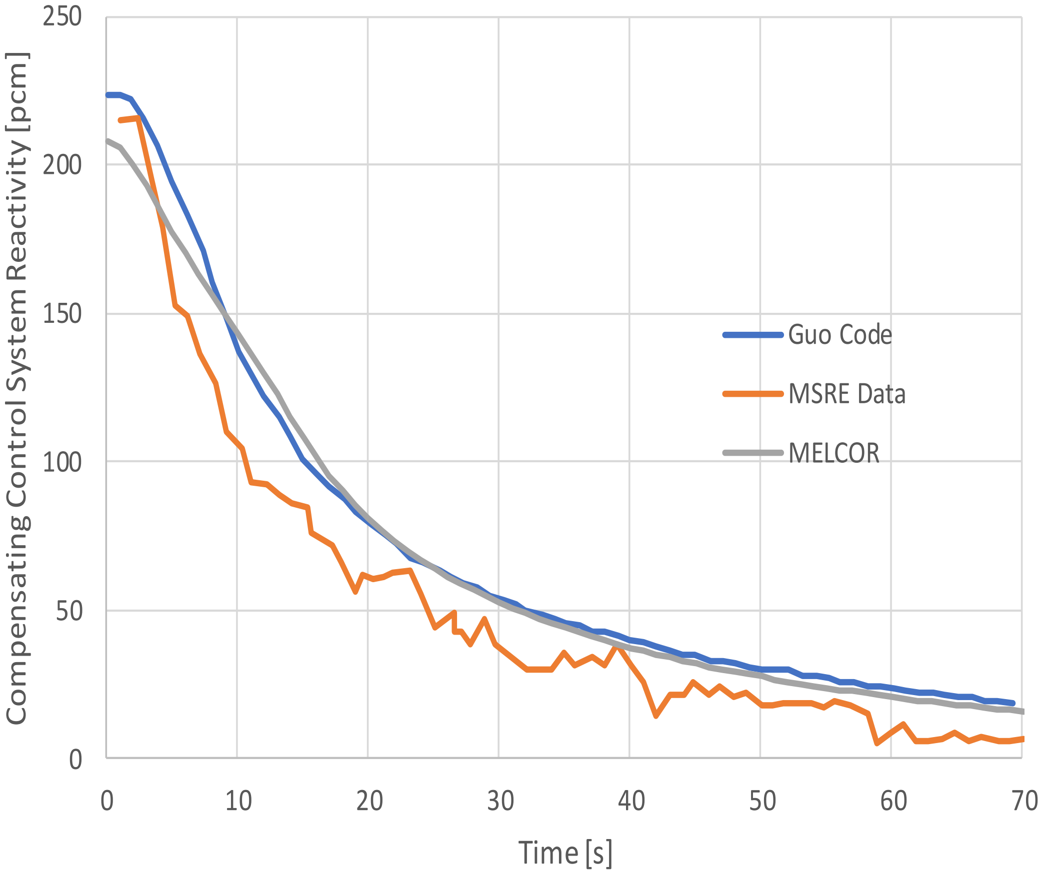
- Generalized approach to utilize any thermodynamic database
- An example is the Molten Salt Thermal Database
  - FLiBe-based systems
  - Chloride-based systems

Radionuclides grouped into forms found in the Molten Salt Reactor Experiment



Point Kinetics with Fluid Fuel Advection





Derived from standard point kinetics equations

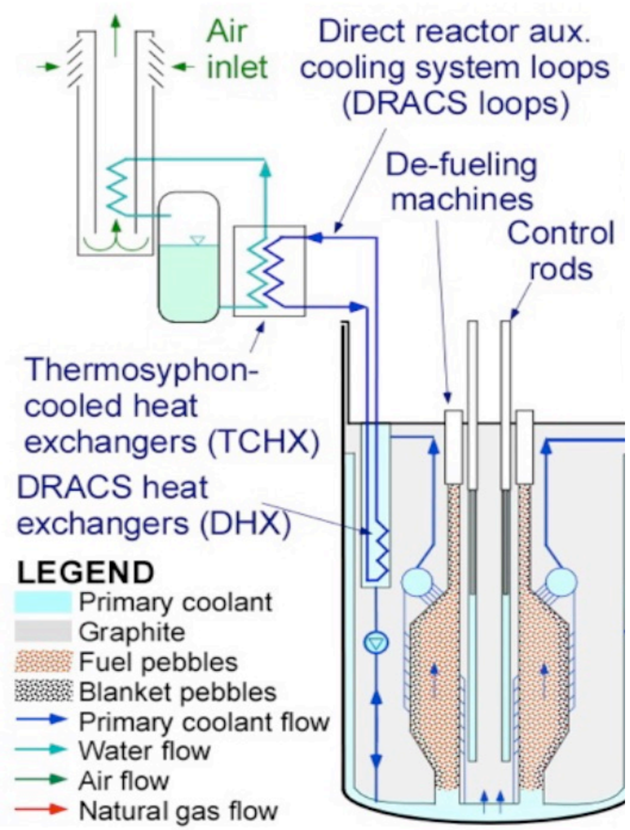
$$\frac{dP}{dt} = \left(\frac{\rho-\beta_{eff}}{\Lambda}\right) P + \sum_{i=1}^{6} \lambda_i C_i^{core} + S_0$$

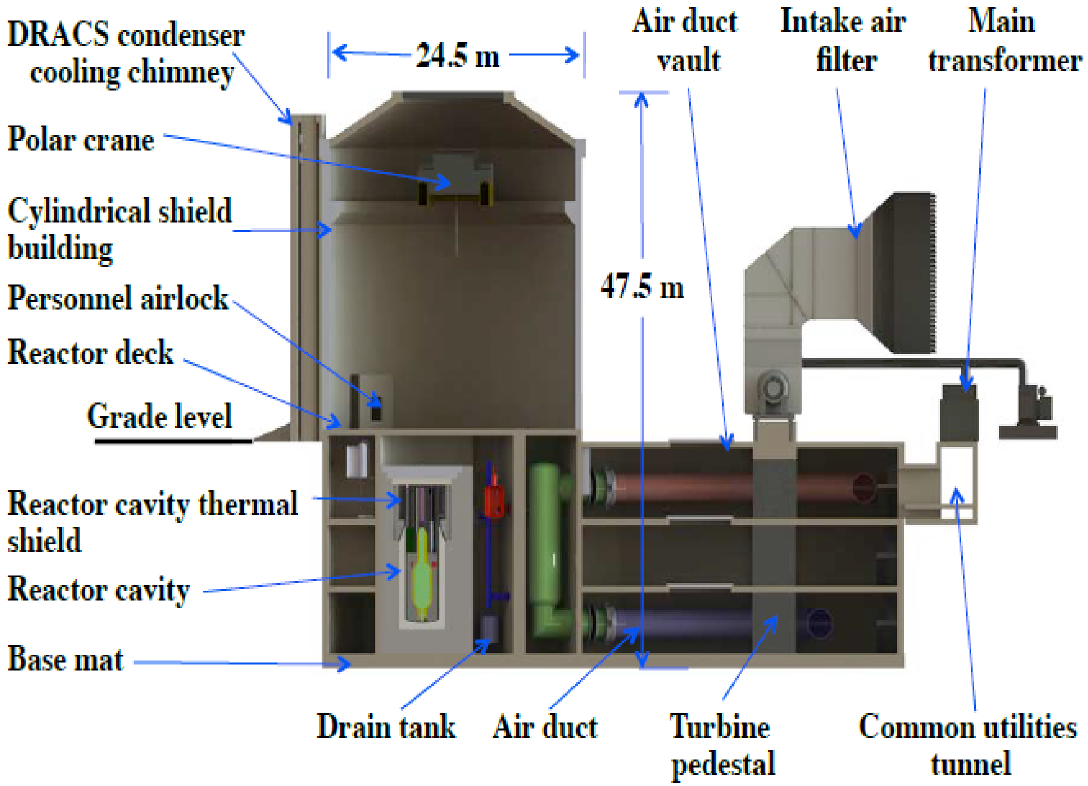
$$\frac{dC_i^{core}}{dt} = \left(\frac{\beta_i}{\Lambda}\right) P - \left(\lambda_i + \frac{2}{\tau_{core}}\right) C_i^{core} + \left(\frac{V_{loop}}{V_{core}}\right)\left(\lambda_i + \frac{2}{\tau_{loop}}\right) C_i^{loop}$$

$$\frac{dC_i^{loop}}{dt} = \left(\frac{V_{core}}{\tau_{core} V_{loop}}\right) C_i^{core} - \left(\lambda_i + {}^{1}/_{\tau_{loop}}\right) C_i^{loop}$$

$$\beta_{eff} = \beta - \beta_{lost} = \beta - \left(\frac{\Lambda}{P}\right) \sum_{i=1}^{6} \lambda_i C_i^{loop}$$

FHR Demonstration Model – UCB Mark I





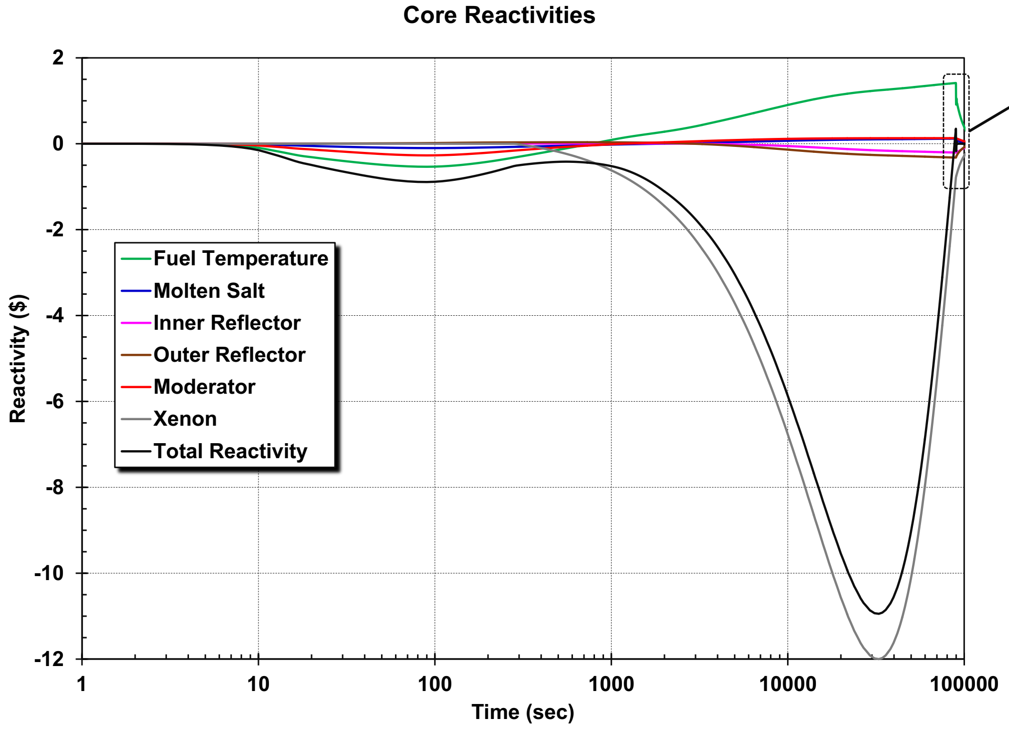
Fuel → Reactor Core → Reactor System → Reactor Containment

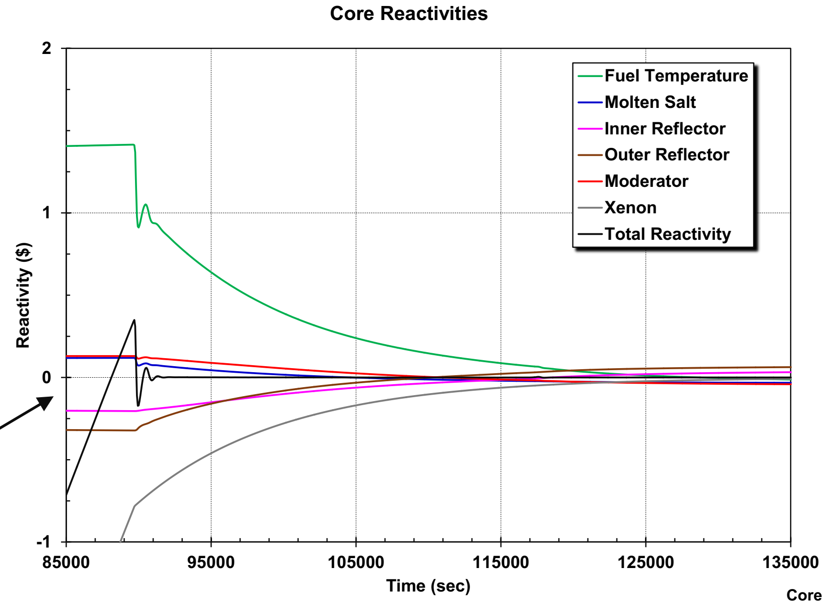
ATWS with 3xDRACS

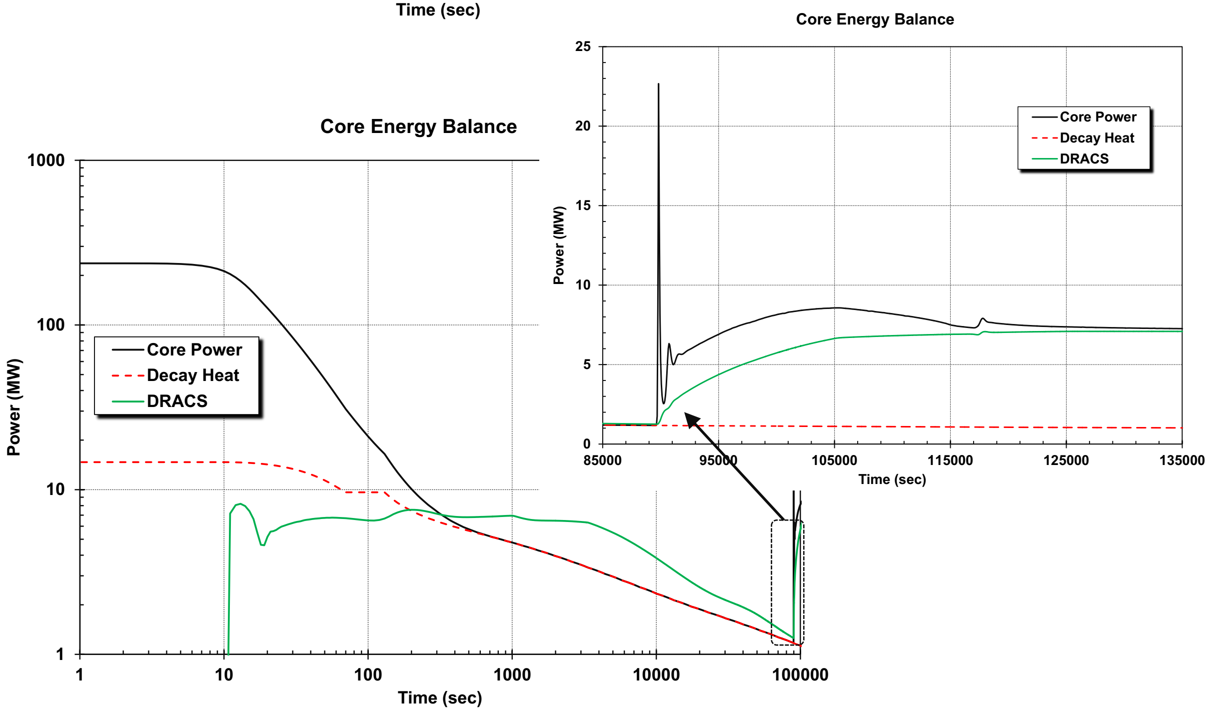
Initial fuel heatup has strong negative fuel and moderator feedback that offsets positive reflector feedbacks

Strong negative Xe transient feedback *

3xDRACS exceeds core power after 330 s





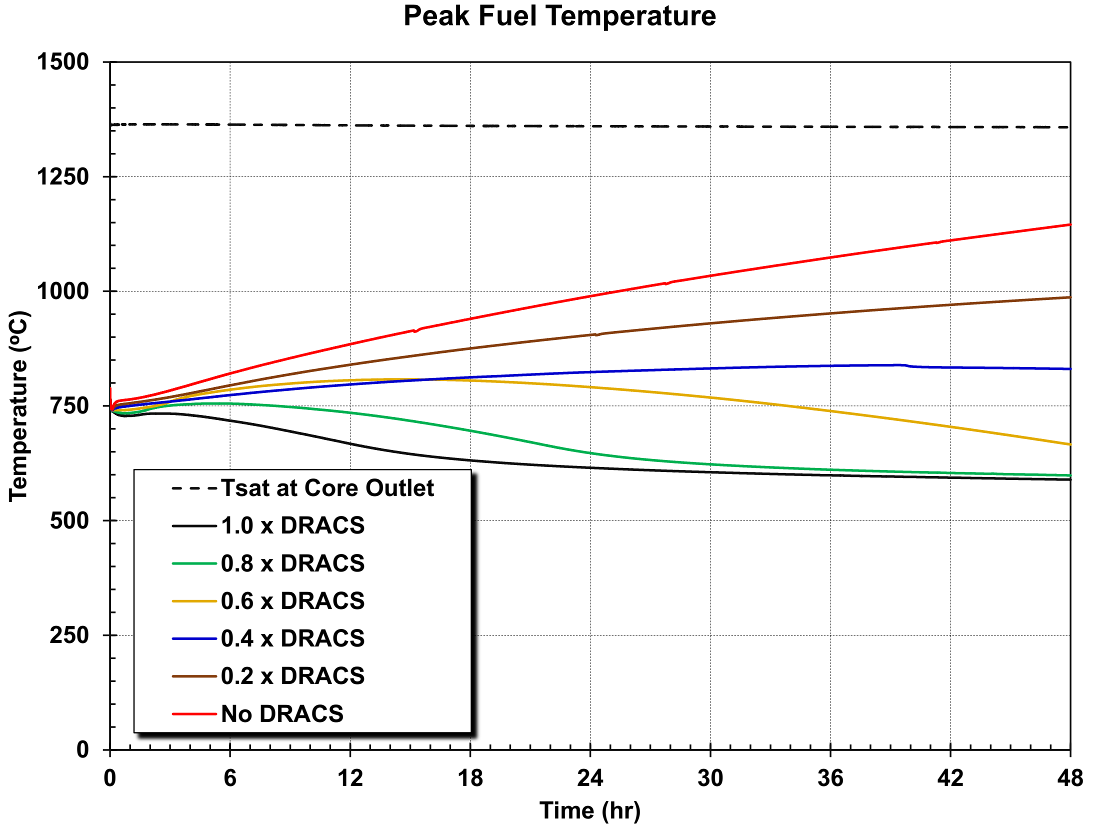


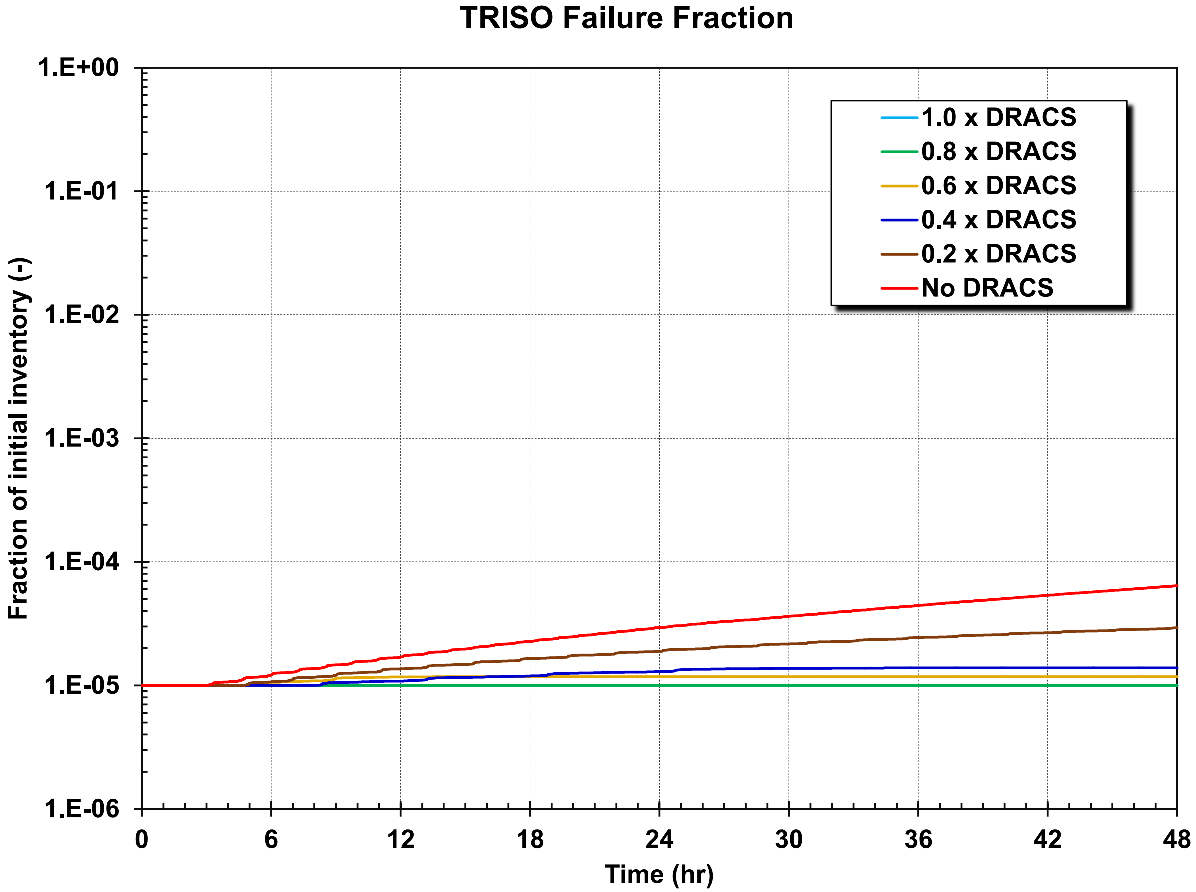
* Xenon transient approximated.

SBO Scenario Simulations

Station Blackout (SBO) scenario simulated with range of DRACS heat removal performance

- ≥40% of one DRACS halts temperature rise within 48 hours





* UCO TRISO thermal failure characteristics were not available, so UO_2 TRISO diffusivity and UO_2 failure data were used. Both are changeable through user input with design-specific data.

Cesium Vaporization from Molten Salt

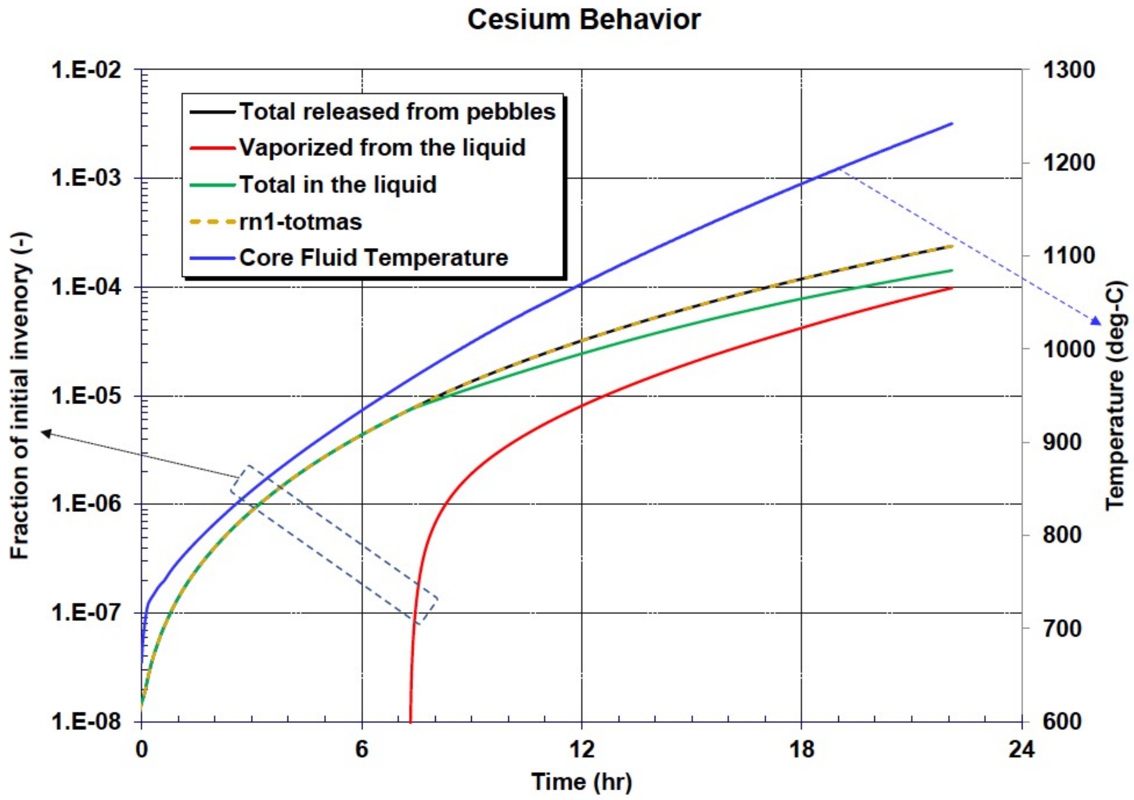
Fission product thermochemistry modeling sample demonstration

- Exercise machinery
- Focuses on Cs and CsF release from salt pool
- Thermochimica Gibbs Energy Minimizer
- Utilizing vapor phase data for CsF*

Demonstration calculation for LOCA sequence

- No core uncovery through 24 hours

Model exhibits Cs and CsF vaporization to gas space at elevated salt temperatures



* With modifications by Ontario Tech.

Thanks for your attention

The Molten Salts in Extreme Environments Energy Frontier Research Center

MSEE
molten salts in extreme environments

James Wishart, MSEE Director
Brookhaven National Laboratory

Brookhaven National Laboratory | INL Idaho National Laboratory | Oak Ridge National Laboratory | University of Notre Dame | Stony Brook University | The University of Iowa | U.S. Department of Energy

MSEE's Mission

To provide fundamental and predictive understanding, based in atomistic/molecular level descriptions, of molten salt bulk and interfacial chemistry, including the effects of solutes, impurities and radiation.

Key Issues and their Impacts

- How does the structure of molten salts control their properties?
  Predicting and controlling MSR fuel properties under normal and abnormal conditions.
- How does salt composition and radiolysis affect speciation, solubility and reactivity?
  Keeping fuel, fission products and corrosion products dissolved in the salt.
- How does the structure of the salt/metal interface affect mass and charge transfer?
  Limiting corrosion and inhibiting processes that compromise materials integrity.

Broader Scientific and Applied Impacts

- Understanding the chemistry and behavior of ionic fluids, in theory and practice
- Energy applications beyond fission reactors
  - Heat transport media (Concentrated solar power and fusion reactor blankets)
  - Pyroprocessing of used nuclear fuel
  - Advanced battery and fuel cell technologies

MSEE is built on two synergistic thrusts

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Thrust 1: Molten Salt Properties and Reactivity

Seeks to understand how molecular-scale interactions, structure and dynamics lead to macroscale behavior of molten salts

Aim 1: Atomic structure, interactions and dynamics that determine molten salt properties.

Aim 2: Molten salt interactions with solutes, including actinides, fission and corrosion products and nanoparticles.

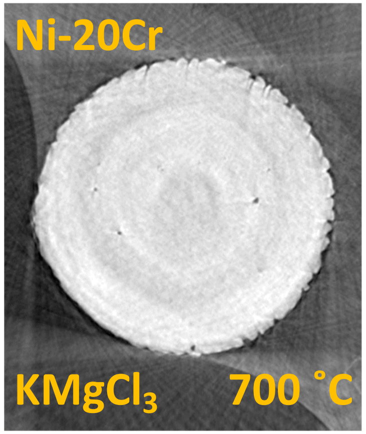
Aim 3: Effects of radiation as a driver of redox processes and metal ion speciation.

Thrust 2: Interfacial and Corrosion Processes in Molten Salt Environments

Seeks to understand atomic-scale structure, dynamics, radiolysis and corrosion at interfaces.

Aim 1: Structure and dynamics of ions at the interface.

Aim 2: Interfacial processes and radiolysis leading to corrosion.



Core activities

Molten salt preparation and handling

Development of **multifunction cells for molten salt experiments**

User facilities are central to MSEE's research

NSLS-II (MSEE uses 9 of 30 beamlines):

- Imaging & Microscopy
- Complex Scattering & Reflectivity
- Diffraction & In Situ Scattering
- Hard X-Ray Spectroscopy

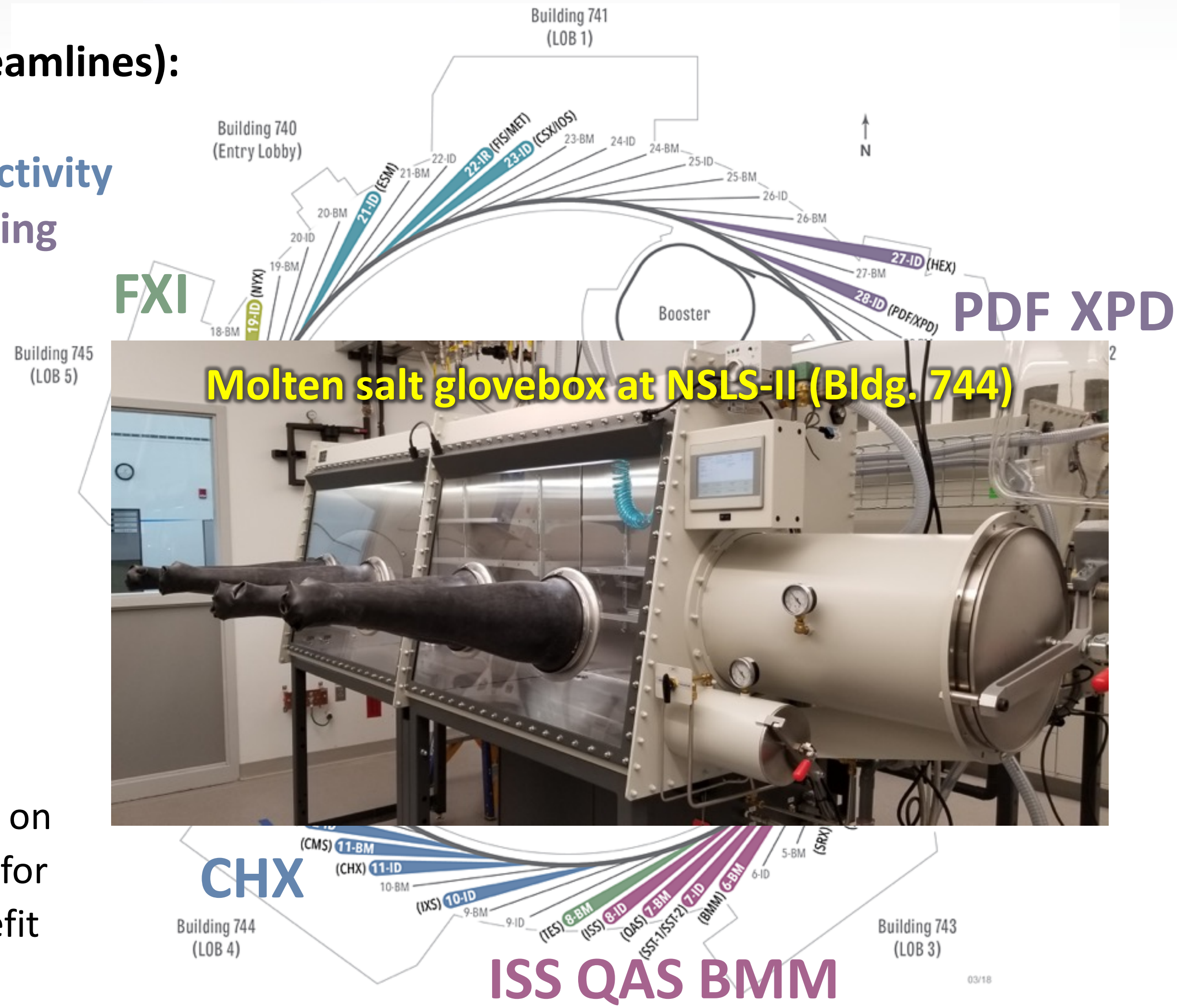
APS:

- X-ray scattering
- XANES and XAFS

Spallation Neutron Source:

- Neutron scattering
- Neutron reflectivity

MSEE works closely with facilities on new instrumentation capabilities for molten salt work, which will benefit other users doing similar work.



Thrust 1, Aim 1:
Molten salt structure and dynamics

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Powerful X-ray, neutron-scattering and optical spectroscopy techniques are coupled with computational approaches to interpret observations and validate predictions in order to assemble a dynamical model of molten salt structure.

MS structure and dynamics across scales of length and temperature

Salt Preparation
Experimental Scattering

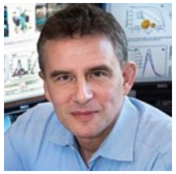

Phillip Halstenberg

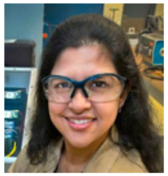

Alexander Ivanov


Shannon Mahurin


Sheng Dai

ORNL


Anatoly Frenkel


Simerjeet Gill

BNL


Ruchi Gakhar


William Phillips

INL

Computational Modeling
Rigid and Polarizable Ion Models, AIMD, ML


Matt Emerson


Shobha Sharma


Claudio J. Margulis

U. Iowa

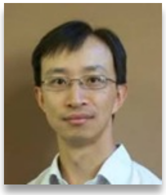

Santanu Roy


V. Bryantsev

ORNL


Ryan DeFever


Haimeng Wang

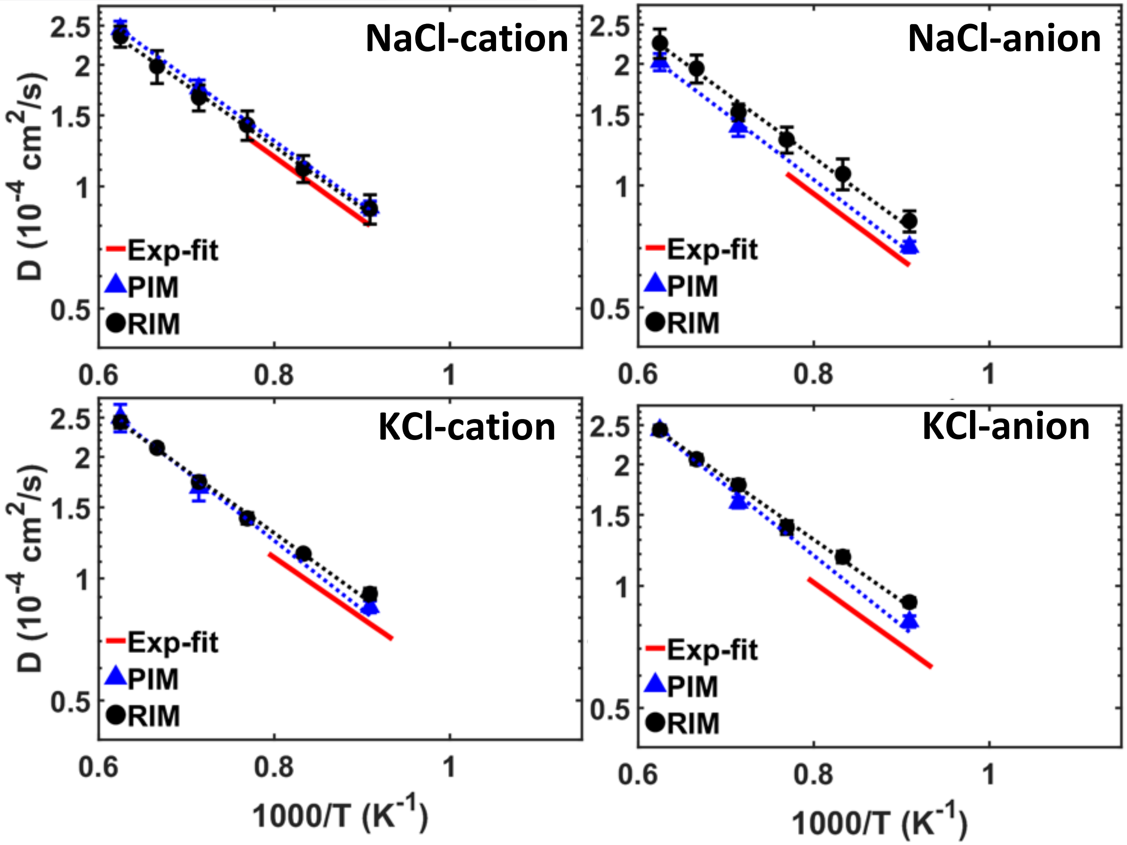

Yong Zhang


Ed Maginn

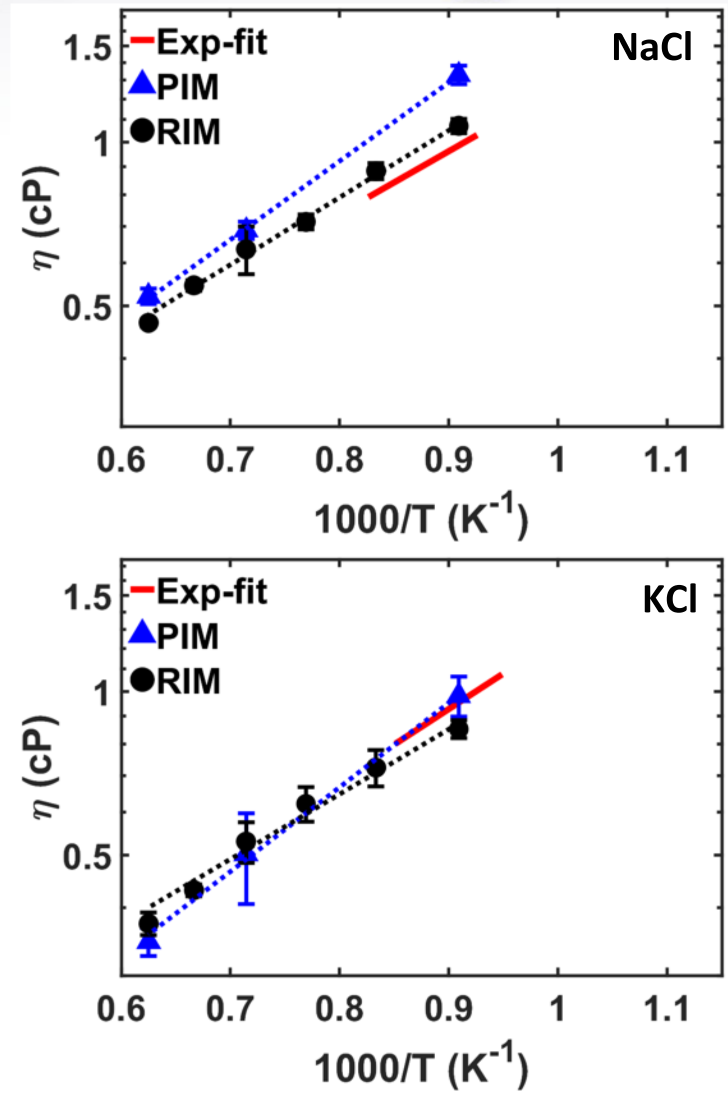
U. Notre Dame

Transport properties

self-diffusivity



shear viscosity



The transport properties of the molten alkali chlorides predicted by both models agree reasonably well with the experimental results.

[1] Wang, H.; *et al. J. Chem. Phys.* **2020**, *153*, 214502.

Simulating solid-liquid phase transitions of alkali chloride salts with non-polarizable and polarizable force fields

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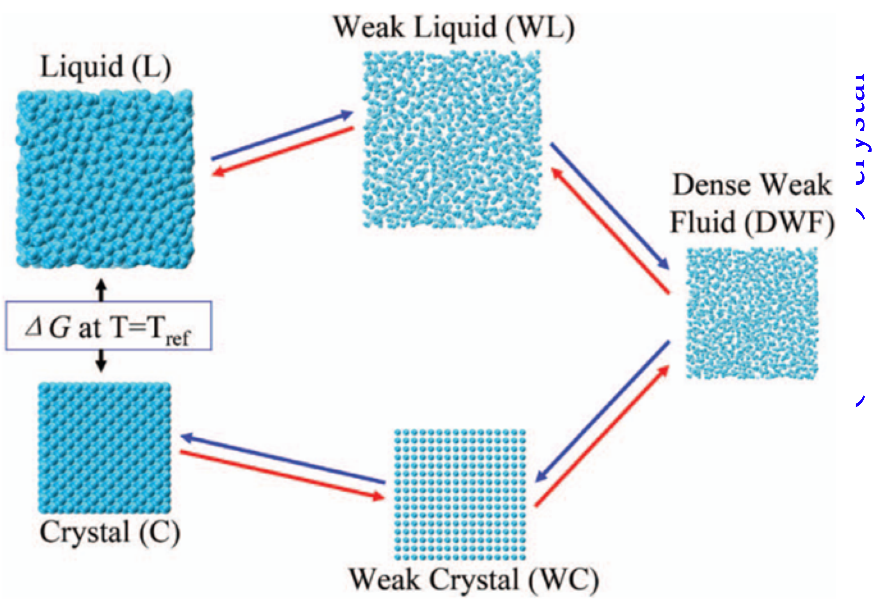
Calculating Melting Points: Two Approaches

Liquid

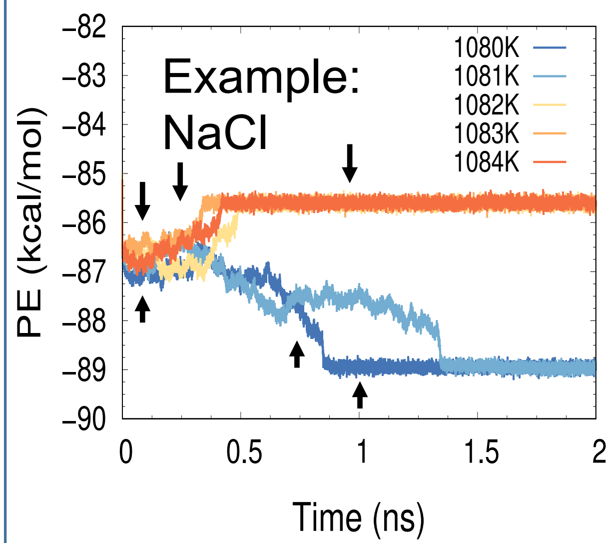
Pseudo-Supercritical Path (PSCP)

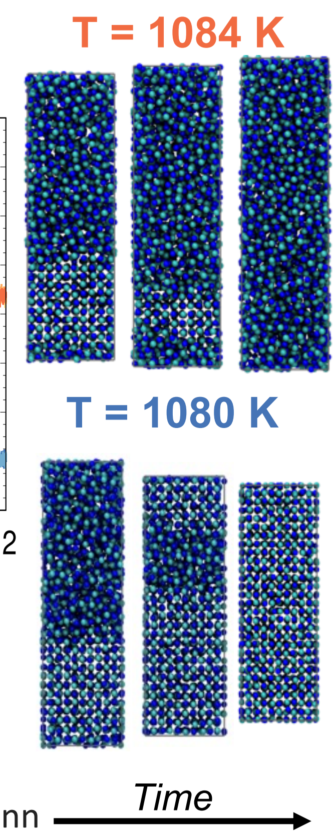


Thermodynamic integration

$$\Delta G \approx \Delta A_{i\rightarrow j} = \int_0^1 \left\langle \frac{\partial U}{\partial \lambda} \right\rangle d\lambda$$

Direct Coexistence (DC)





DeFever R.S.; Wang H.; Zhang Y.; Maginn E.J.; *J. Chem. Phys.* **2020**, *153, 011101*.

Comparison of melting points for RIM and PIM potentials

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Methods comparison

RIM melting temperatures (K) computed with PSCP and direct coexistence methods

	PSCP	Direct coex.
LiCl	782_1	777_3
NaCl	1082_2	1081_2
KCl	1039_2	1038_2
RbCl	1091_1	1091_3

The PSCP and direct coexistence methods very nearly predict the same melting points for all four alkali chlorides.

Models comparison

Comparison of RIM and PIM melting temperatures (K)

	Expt.	RIM	PIM
LiCl	883	777_3	1025_5
NaCl	1073	1081_2	1140_{10}
KCl	1043	1038_2	1053_3
RbCl	988	1091_2	1035_{10}

Neither the RIM nor PIM predicts more accurate melting temperatures across all four alkali chlorides.

DeFever R.S.; Wang H.; Zhang Y.; Maginn E.J.; *J. Chem. Phys*. **2020**, *153, 011101*.

We are now modeling phase diagrams for binary systems.

Simulations Reveal Complexity and Intermediate-Range Structure in a Binary Salt

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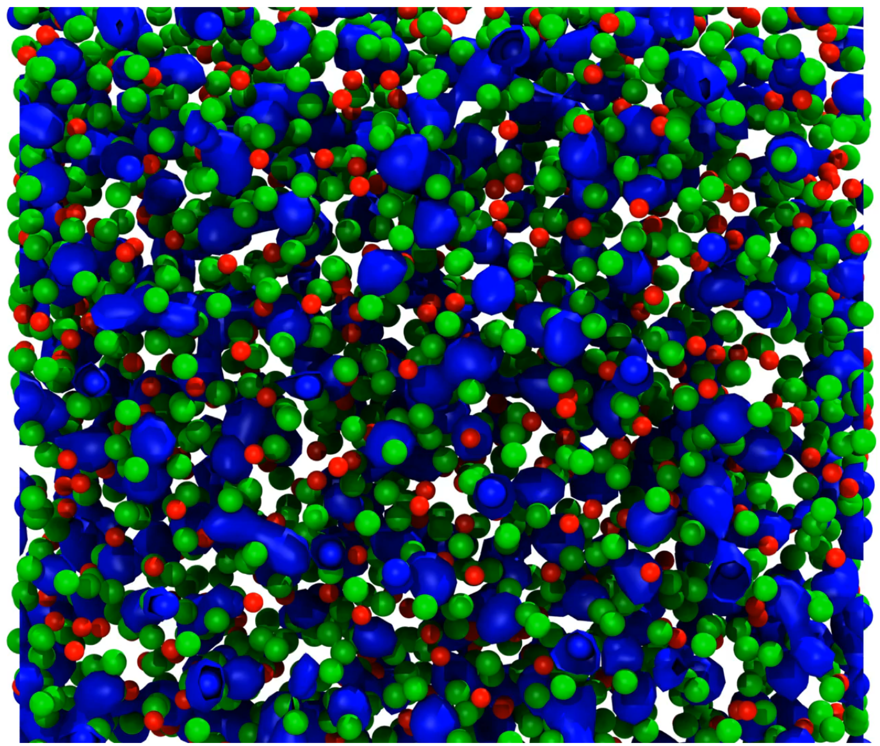
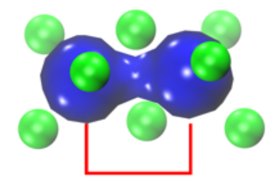
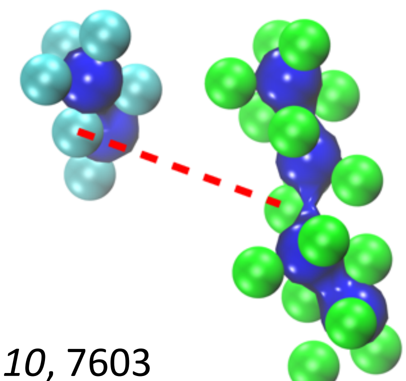
$KMgCl_3$

Blue isosurfaces highlight short transient Mg^{2+} networks.



Blue	Mg^{2+}
Red	K^{+}
Green	Cl^{-}





J. Phys. Chem. Lett. **2019,** *10*, 7603
J. Phys. Chem. B **2020,** *124*, 2892

Slide credit: Claudio Margulis, U. of Iowa

“Anomalous” Temperature Dependence of the Intermediate-Range Order

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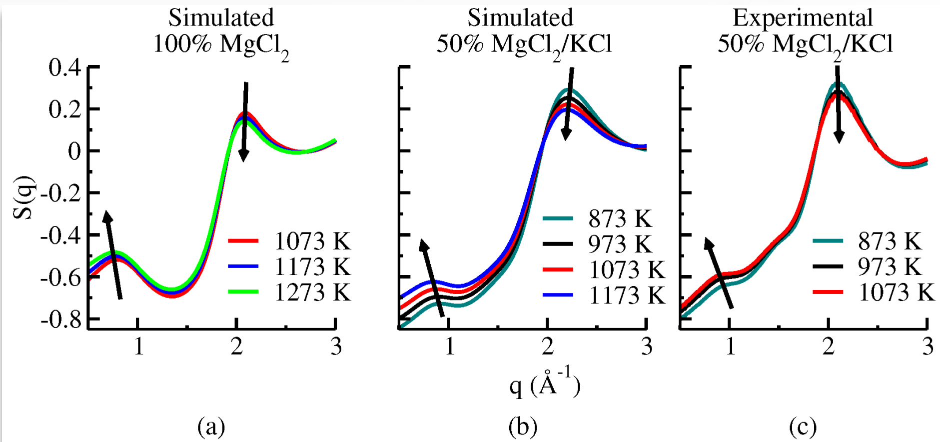

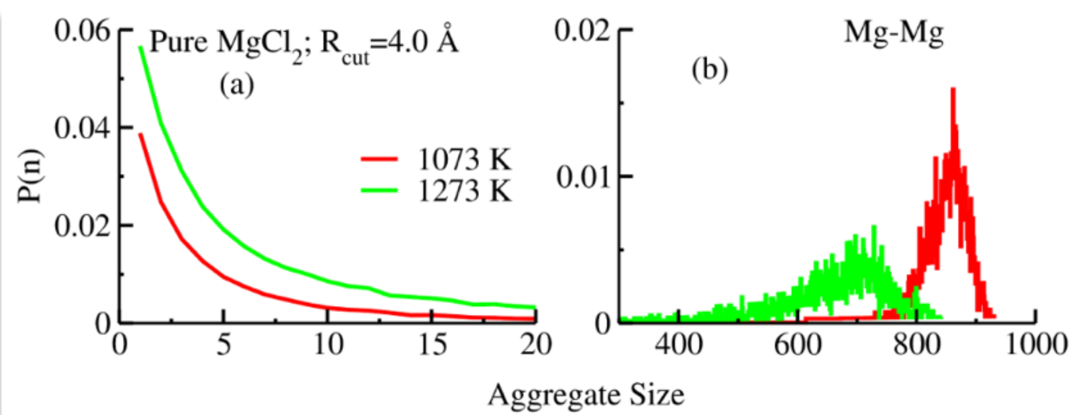

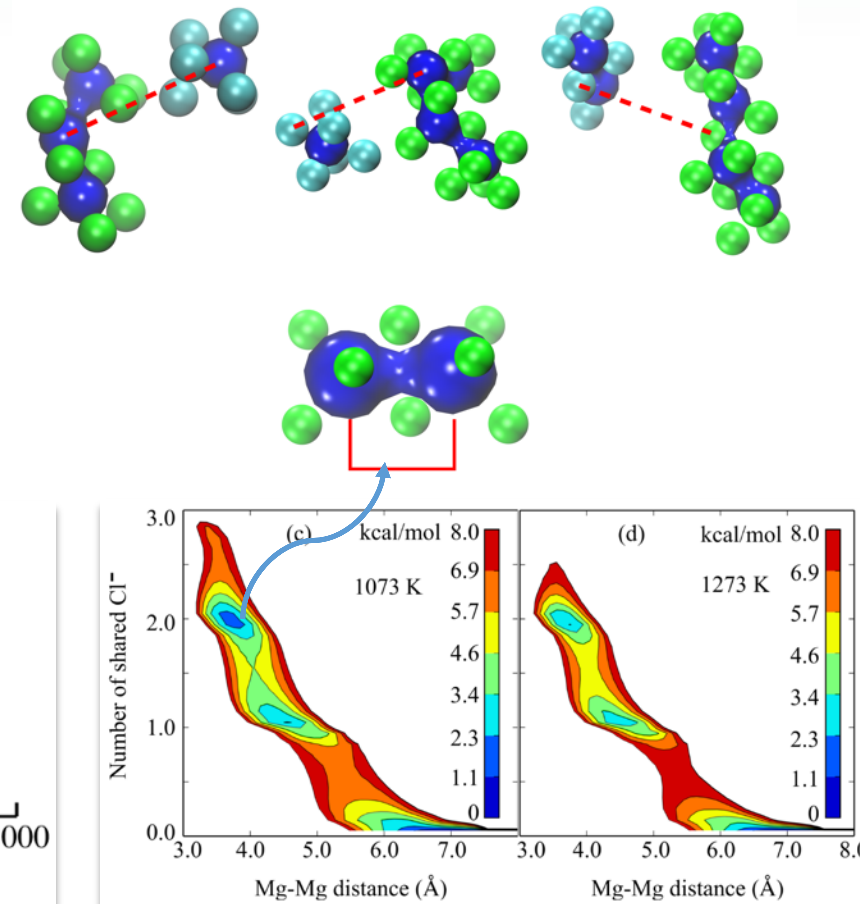

Wu, Sharma *et al., J. Phys. Chem. B*, 2020, **124**, 2892-2899. DOI: 10.1021/acs.jpcb.0c00745

The SEM-Drude model: A fast polarizable force field for the study of neat KCl, neat $MgCl_2$, and their mixtures

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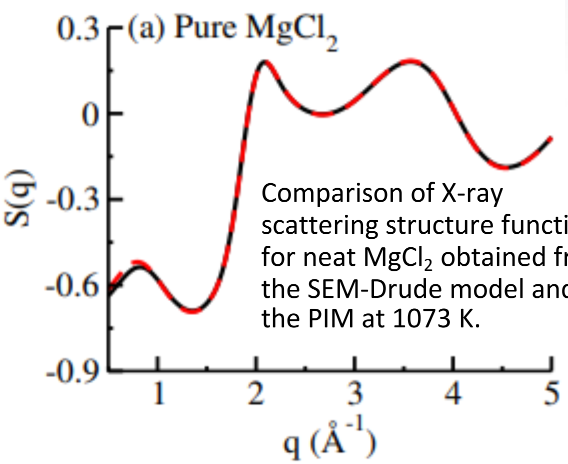
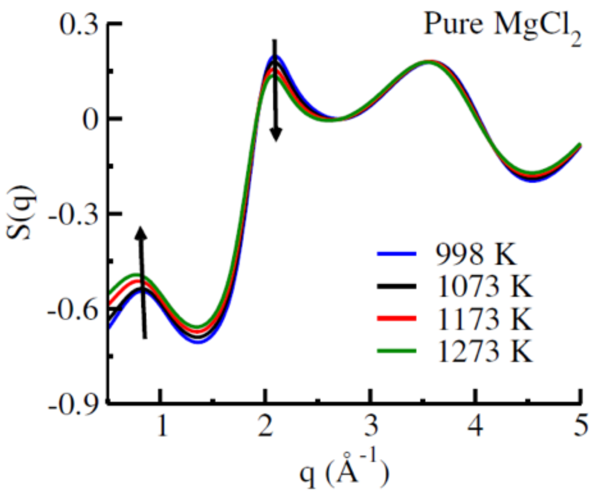


Comparison of X-ray scattering structure functions for neat $MgCl_2$ obtained from the SEM-Drude model and the PIM at 1073 K.



Temperature dependence of X-ray scattering structure functions for neat $MgCl_2$ obtained from the SEM-Drude model.

The Sharma-Emerson-Margulis Drude (SEM-Drude) model

A **computationally-efficient polarizable model** developed to study the structure, transport and themodynamics of KCl/$MgCl_2$ molten salt systems.

Simulations with the new model in the LAMPPS software can be **30 times faster** than using the gold-standard polarizable ion model in the CP2K software.

It allows for simulations of **larger systems for much longer times** providing access to transport, structural and thermodynamic properties that were simply prohibitive to compute otherwise at an essentially identical accuracy.

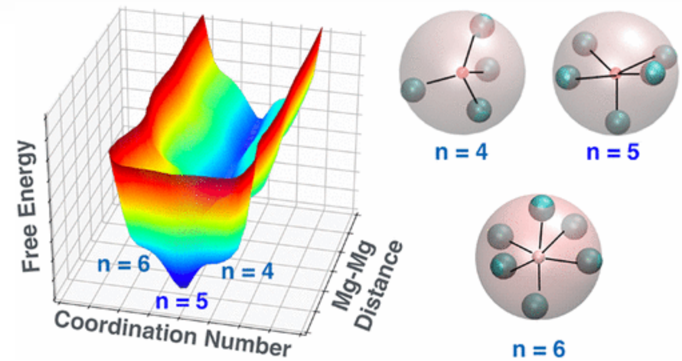
As opposed to other Drude models, this one considers the effect of charge-dipole interaction dumping at short distances. This is important to avoid overpolarization. To the best of our knowledge this may be the first successful Drude model for high-temperature molten salts.

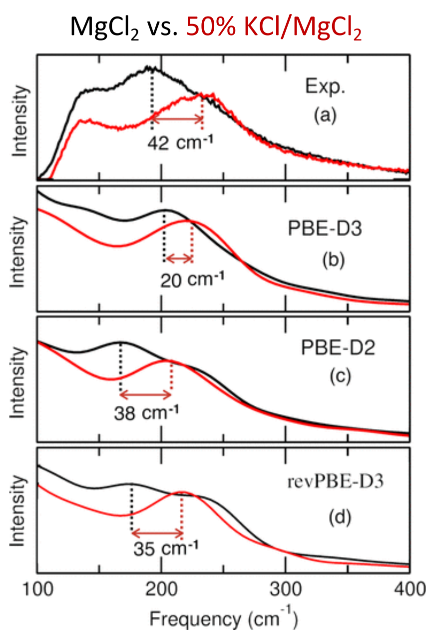
Excellent results from the SEM-Drude model **provide confidence in predictions of quantities never computed before with the PIM due to cost.**

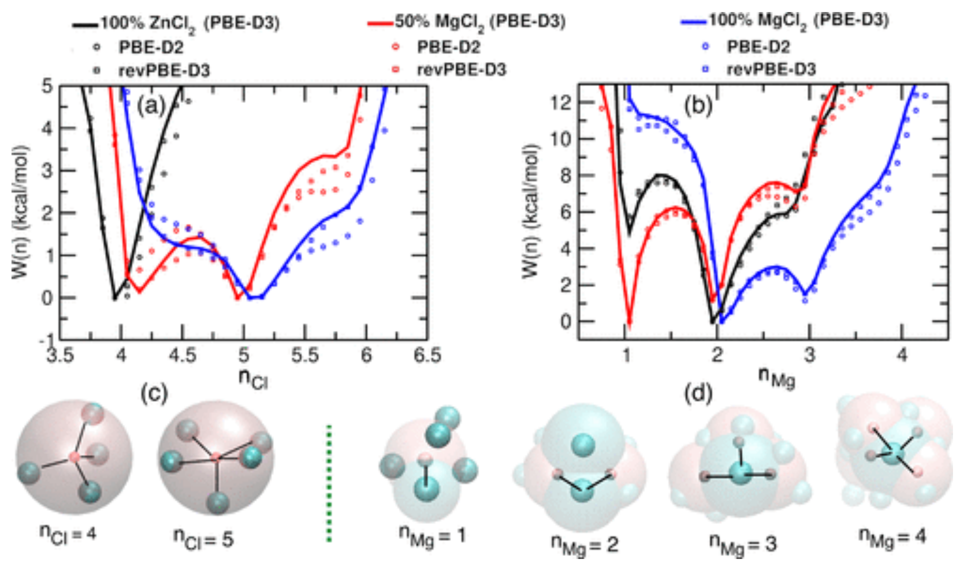
Sharma, S.; Emerson, M. S.; Wu, F.; Wang, H.; Maginn, E. J.; Margulis, C. J., *J. Phys. Chem. A* **2020**, *124*, 7832.

Ab initio MD, XRS and Raman spectroscopy resolve earlier misconceptions about Mg^{2+} coordination in chloride salts

AIMD simulations provided convergent results that a five-coordinate $MgCl_5^{3-}$ complex is a dominant form in pure $MgCl_2$, a species that has not been considered before in the interpretation of the Raman spectra.







S. Roy *et al. J. Phys. Chem. B*, 2021, 125, 5971 DOI: 10.1021/acs.jpcb.1c03786

Thrust 1, Aim 2: Molten salt interactions with solutes

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X-ray Absorption Spectroscopy
X-ray Scattering

A. Ivanov
(ORNL)

S. Gill
(BNL)

A. Frenkel
(BNL)

Ab-initio Molecular Dynamics

V. Bryantsev
(ORNL)

S. Roy
(ORNL)

C. Margulis
(Iowa)

Optical spectroscopy

R.Gakhar
(INL)

Core Activity: Molten salt preparation

Thrust I. MS Properties and Reactivity

S. Mahurin

S. Dai

Thrust I – Aim 3
Radiation effects

S. Pimblott
(INL)

J. Wishart
(BNL)

J. LaVerne
(Notre Dame)

G. Horne
(INL)

Atomic Insight into Metal Ion Solvation Structure in High-Temperature Molten Ionic Medium

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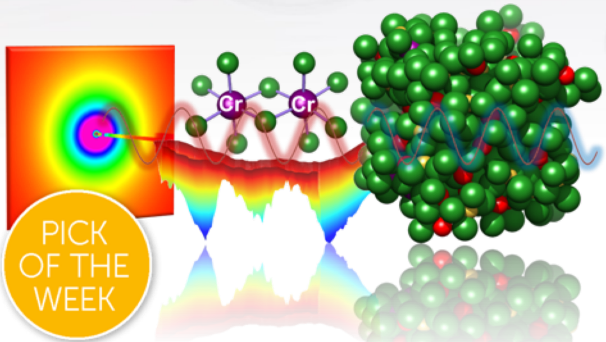
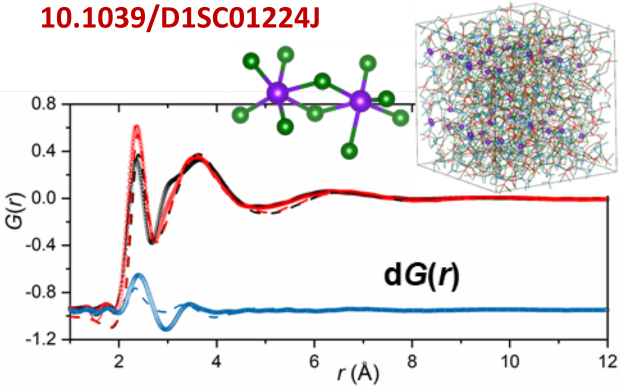


Chemical Science, 2021, https://doi.org/10.1039/D1SC01224J



X-ray pair distribution functions *G*(r) for the KCl-$MgCl_2$ (black) and $CrCl_3$-KCl-$MgCl_2$ (red) molten salt mixtures at 1073K, and the derived differential PDF d*G*(r) (blue).

Inset: A snapshot from the RMC modeling highlighting ionic clustering of Cr^{3+} species (enlarged purple spheres) and representative chromium chloride dimers present in the melt.

Scientific Achievement

The atomic-scale behavior of dilute Cr^{3+} metal ions in a molten $MgCl_2$-KCl salt was investigated by a comprehensive study integrating synchrotron X-ray scattering experiments, *ab-initio* molecular dynamics simulations and rate theory concepts.

Significance and Impact

Clustering of Cr^{3+} ions was unexpectedly revealed.
The results challenge the paradigm of molten salts being a "sea of cations in a sea of anions", where aggregation of dilute species was not expected due to high temperature conditions.

Research Details

- High-energy X-ray scattering measurements for $CrCl_3$-$MgCl_2$-KCl molten salts were performed at the PDF beamline of NSLS-II.
- A differential pair distribution function approach was applied to gain insights into the local structure of Cr^{3+} in a $MgCl_2$-KCl molten salt.
- Reverse Monte-Carlo fitting and *ab-initio* MD simulations assisted by a hybrid transition state-Marcus theory model indicate the formation of **dinuclear chloride-shared Cr-Cr clusters with a relatively long (~30 ps) lifetime** in the melt even under high temperature conditions.

Work was performed at Oak Ridge National Laboratory, Brookhaven National Laboratory, Idaho National Laboratory, and the University of Iowa

Brookhaven National Laboratory · INL Idaho National Laboratory · Oak Ridge National Laboratory · University of Notre Dame · Stony Brook University · The University of Iowa · U.S. Department of Energy

Correlated spectroscopic and simulation studies explain local structures of Ni^{2+} and Co^{2+} in $ZnCl_2$

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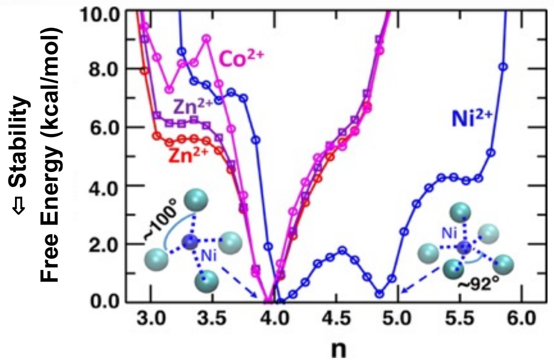
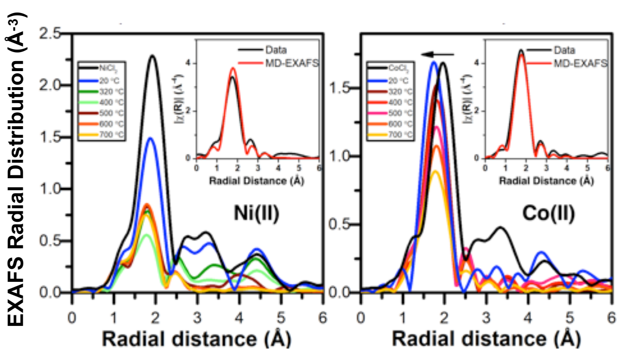


Above: AIMD simulations show that Co(II) and Zn(II) are 4-coordinate in molten $ZnCl_2$, but Ni(II) is distributed between 4- and 5-coordinate forms. This disorder results in low apparent coordination numbers for Ni(II), below left.



J. Phys. Chem. B **124,** 1253-1258 (2020)
DOI: 10.1021/acs.jpcb.0c00195

Scientific Achievement

Complexity of metal ion coordination in molten salts was elucidated by combining X-ray and optical absorption spectroscopy and *ab-initio* molecular dynamics simulations.

Significance and Impact

Strong dynamic heterogeneity in the coordination environment of Ni(II) was revealed by experiment and simulation, providing new insight into the factors controlling the low solubility of Ni(II) in molten $ZnCl_2$. The combined approach is a powerful pathway to understanding speciation and solubility of metal ions in molten salts relevant to molten salt reactors and other applications.

Research Details

- X-Ray absorption spectroscopy of Ni(II) and Co(II) in molten $ZnCl_2$ was performed at the NSLS-II ISS beamline.
- *Ab-initio* MD simulations were essential to interpret the X-ray spectroscopy of Ni(II) and Co(II) in terms of the local structures.
- Simulations at OLCS and NERSC showed the coexistence of 4- and 5-coordinate Ni(II) structures in $ZnCl_2$, resulting in disorder that explains the oddly low coordination number from XAS analysis.

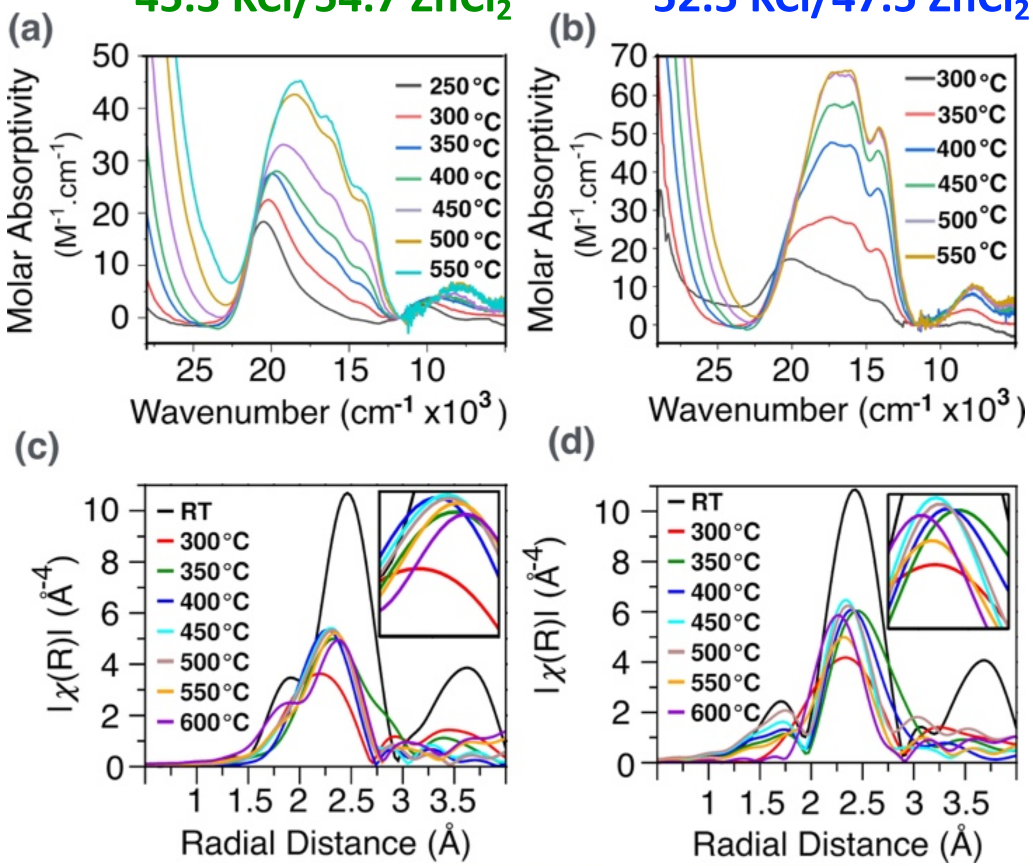
Brookhaven National Laboratory · INL Idaho National Laboratory · Oak Ridge National Laboratory · University of Notre Dame · Stony Brook University · The University of Iowa · U.S. Department of Energy

Ni^{2+} coordination is highly sensitive to the molten salt environment and temperature

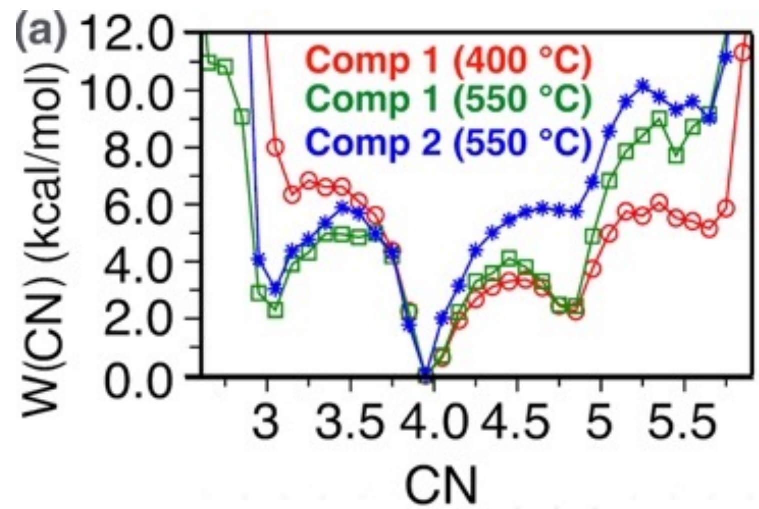
$KCl – ZnCl_2$ mixtures

Composition 1
45.3 KCl/54.7 $ZnCl_2$

Composition 2
52.5 KCl/47.5 $ZnCl_2$



Free energy as a function of Ni^{2+} coordination number (CN)



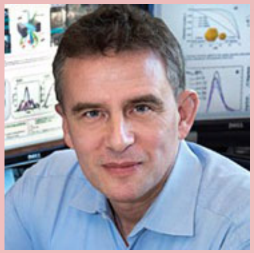
J. Am. Chem. Soc. **2021,** *143*, 15298
Dedicated to the memory of Austen Angell

MSEE's Principal Investigators

MSEE
molten salts in extreme environments

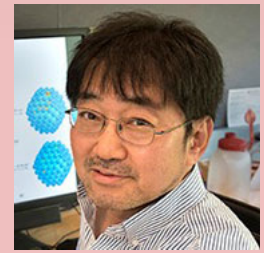
Brookhaven | **Director**


Eric Dooryhee


Anatoly Frenkel


Simerjeet Gill


Benjamin Ocko


Kotaro Sasaki


James Wishart

Idaho | **Deputy Director**


Ruchi Gakhar


Lingfeng He


Gregory Horne


Simon Pimblott

Notre Dame


Jay LaVerne

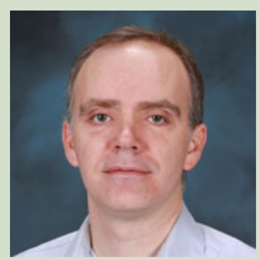

Edward Maginn

Oak Ridge | **Thrust 1 Leader**

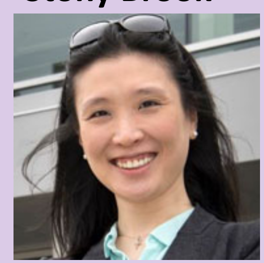

Vyacheslav Bryantsev


Sheng Dai


Alexander Ivanov


Shannon Mahurin

Stony Brook


Karen Chen-Wiegart

Iowa


Claudio Margulis

Brookhaven National Laboratory | INL Idaho National Laboratory | Oak Ridge National Laboratory | University of Notre Dame | Stony Brook University | The University of Iowa | U.S. Department of Energy

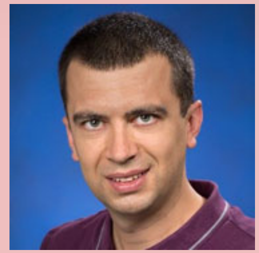
MSEE's Postdocs, Grad Students and Staff

MSEE
molten salts in extreme environments

Brookhaven


Kazuhiro Iwamatsu (PD)


Bobby Layne (TS)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Luis Betancourt (PD)

Idaho


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Kaustubh Bawane (PD)

Iowa


Shobha Sharma (PD)


Matt Emerson (GS)

Oak Ridge


Phillip Halstenberg (GS)


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Notre Dame


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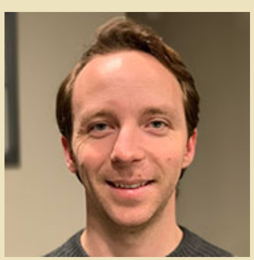
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All MSEE PIs, Staff, Postdocs and Graduate Students (shown earlier)

Bobby Layne, BNL, multiple high temperature sample configurations for NSLS and Chemistry

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MSR Fuel Salt Qualification Method

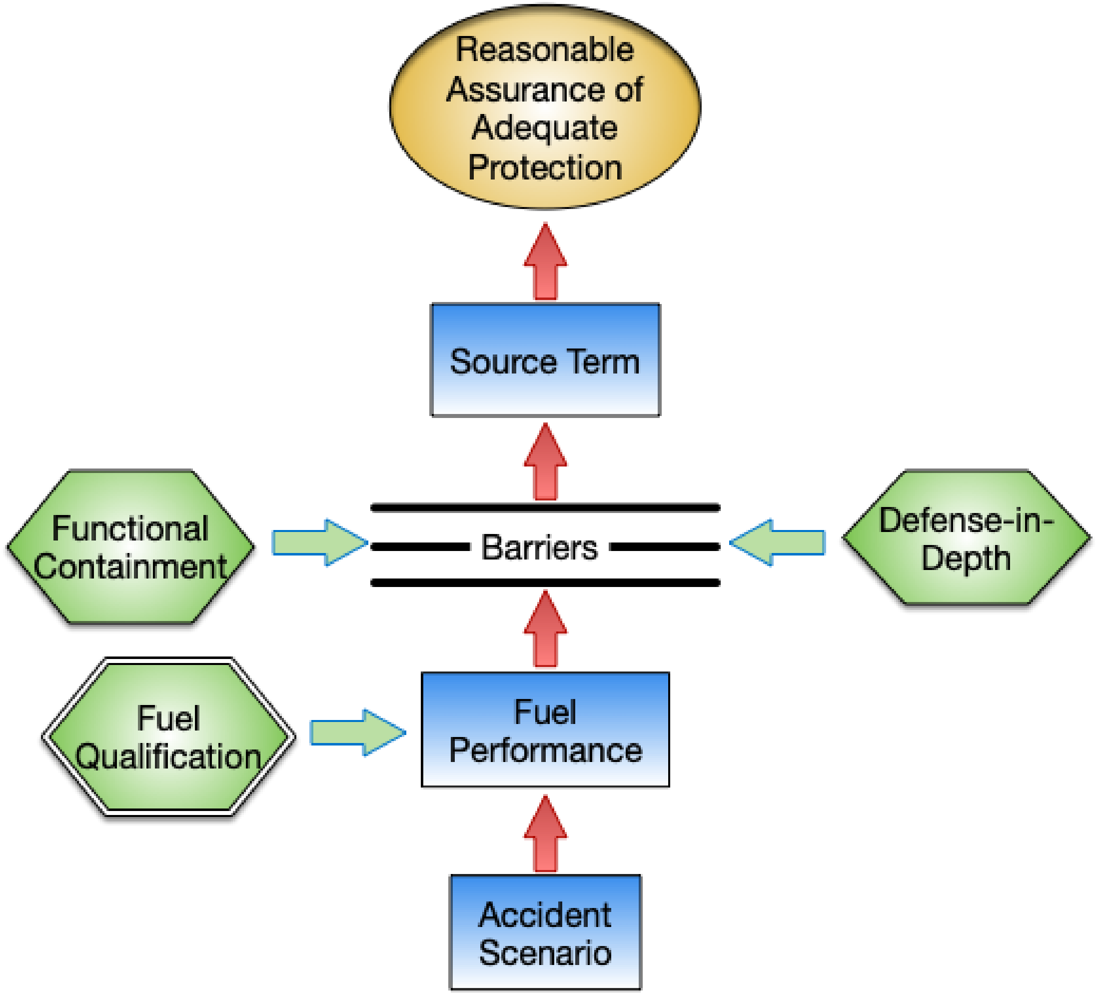
Workshop on Molten Salt Thermal Properties

David Holcomb, George Flanagan, and Mike Poore

November 15th, 2021

Fuel Qualification is an Element in Achieving Sufficient Understanding of Fuel Behavior

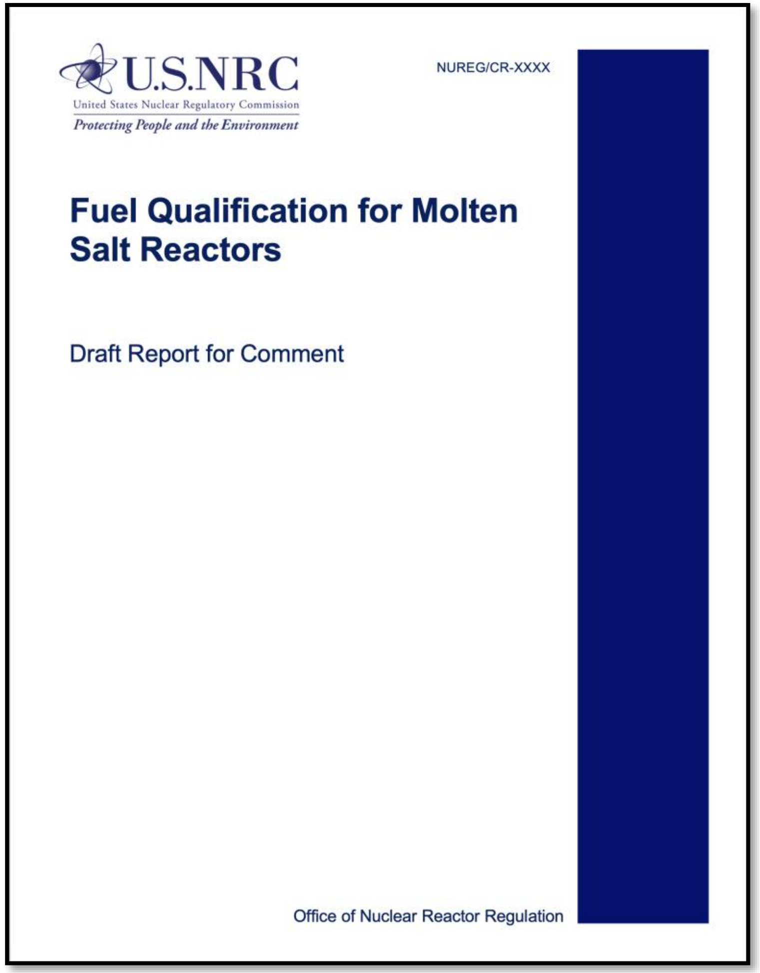
"***Fuel qualification*** *is a process which provides high confidence that physical and chemical behavior of fuel is sufficiently understood so that it can be adequately modeled for both normal and accident conditions, reflecting the role of the fuel design in the overall safety of the facility. Uncertainties are defined so that calculated fission product releases include the appropriate margins to ensure conservative calculation of radiological dose consequences.*"



- NRC Presentation on Possible Regulatory Process Improvements for Advanced Reactor Designs, August 3rd, 2017 (ML17220A315)

DRAFT NUREG/CR on Fuel Qualification for Molten Salt Reactors Currently Under Review

- Fuel salt qualification is a primary end use for fuel salt property data and models
- Proposed fuel salt qualification process based upon maintaining fuel salt properties within an acceptable range that results in plant achievement of fundamental safety functions
  - Under both normal and accident conditions
- Significant departure from solid fuel qualification process
- Provides the technical basis for fuel salt property database development
  - Includes rationale for measurement ranges and uncertainty limits


U.S.NRC
United States Nuclear Regulatory Commission
Protecting People and the Environment

NUREG/CR-XXXX

Fuel Qualification for Molten Salt Reactors

Draft Report for Comment

Office of Nuclear Reactor Regulation

https://adamswebsearch2.nrc.gov/webSearch2/main.jsp?AccessionNumber=ML21245A493

Liquid Fuel Has Substantial, Fundamental Differences From Solid Fuel

- Liquid salt fuel
  - Serves as nuclear fuel and primary heat transfer media
  - Must meet requirements for both purposes

Liquid Fuel	Solid Fuel
Chemically damageable - may be reparable during use	Mechanically damageable
Composition may be adjustable during use	Composition set prior to use
Properties depend on composition and state	Properties depend on fabrication process
Container breach could release nearly all radionuclides	

Qualification is Based Upon Understanding the Chemical and Physical Properties of Representative Fuel Samples

- Liquid state significantly changes the physical behavior of fuel
  - Liquids do not accumulate internal stresses
    - No history dependent properties
  - Flow homogenizes fluid properties
    - No position dependent properties
    - No size dependent properties
- Chemical and physical properties are set by elemental composition and temperature
  - Independent of isotopic content

Small minimally-radioactive liquid fuel salt samples provide representative physical and chemical properties

Fuel Qualification Links the Chemical and Physical Behavior of Fuel to Overall Facility Safety

- Proposed methodology recently presented to advisory committee on reactor safeguards (ACRS)
  - Endorsement letter with comments generated (not yet released)
- Incorporation into regulatory guide anticipated as next step
- Stakeholder review requested to assure method acceptability and minimize potential for either excessive conservatism or inadequate protection