Nuclear Energy is Promising

Today: 7.9 billion inhabitants

⇒ Around 11 billion by 2100

⇒ An increase of 40%

- Current global power consumption in 2021: 17.7 TW ($10^{12}$), Fossil fuels account for 79% of US energy consumption.
- 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....

COP26 – November 2021
International agreement to limit average temperature rise to <1.5 C

Inspired from C. Poinssot, Atalante 2016
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

...aiming to be ready for industrial deployment by 2030
<table>
<thead>
<tr>
<th>Reactor Type (neutron energy spectrum)</th>
<th>Coolant</th>
<th>Typical Fuels Input at Start of Irradiation Cycle</th>
<th>Fuel cycle</th>
<th>Use</th>
<th>Benefit Relative to Current LWRs</th>
<th>Development stage</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas-cooled fast reactor, GFR (fast)</td>
<td>Helium</td>
<td>U/Pu or U/TRU (MOX, metal, nitride, carbide), and natural U</td>
<td>Closed*</td>
<td>Electricity, Hydrogen**, Process Heat**</td>
<td>Higher thermal efficiency for electricity production (&gt;40% vs. 33%)</td>
<td>Early</td>
</tr>
<tr>
<td>Lead-cooled fast reactor, LFR (fast)</td>
<td>Lead, Lead/Bismuth</td>
<td>U/Pu or U/TRU (MOX, metal, nitride, carbide), and natural U</td>
<td>Closed*</td>
<td>Electricity, Hydrogen**, Process Heat**</td>
<td>Higher thermal efficiency for electricity production (&gt;40% vs. 33%), Low pressure reactor coolant Passive safety capability</td>
<td>Early</td>
</tr>
<tr>
<td>Molten salt reactor, MSR (fast)</td>
<td>Chloride salts</td>
<td>U/Pu or U/TRU in salt, and natural U in salt</td>
<td>Closed*</td>
<td>Electricity, Hydrogen**</td>
<td>Higher thermal efficiency for electricity production (&gt;40% vs. 33%), Low pressure reactor coolant Passive safety capability</td>
<td>Early</td>
</tr>
<tr>
<td>Sodium-cooled fast reactor, SFR (fast)</td>
<td>Sodium</td>
<td>U/Pu or U/TRU (MOX, metal, nitride, carbide), and natural U</td>
<td>Closed*</td>
<td>Electricity</td>
<td>Higher thermal efficiency for electricity production (&gt;40% vs. 33%), Low pressure reactor coolant Passive safety capability</td>
<td>Prototypes exist internationally, currently nothing in U.S.</td>
</tr>
<tr>
<td>Supercritical water-cooled reactor, SCWR (thermal or fast)</td>
<td>Water</td>
<td>Enriched UO₂, U/Pu or U/TRU MOX fuel</td>
<td>Open (thermal) Closed* (fast)</td>
<td>Electricity</td>
<td>Higher thermal efficiency for electricity production (&gt;40% vs. 33%)</td>
<td>Early</td>
</tr>
<tr>
<td>Very high temperature gas reactor, VHTR (thermal)</td>
<td>Helium</td>
<td>Enriched UO₂, prism or pebbles</td>
<td>Open</td>
<td>Electricity, Hydrogen**, Process heat**</td>
<td>Higher outlet temperature, Higher thermal efficiency for electricity production (&gt;40% vs. 33%)</td>
<td>Advanced for lower temperatures, Early for high temperature</td>
</tr>
</tbody>
</table>

*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.
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

**Mission:** 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.

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

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

**MSR Campaign**

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

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

**ARPA-E**

**NRC**

**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

**INTERNATIONAL**
IAEA
NEA/OECD

**MSR TWG**
EPRI
NEI
Campaign Review Meetings
Conferences/MSR workshops

**INDUSTRY**

**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
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.

**NEAMS Overview**

NEAMS is the DOE-NE mod sim program, developing and deploying predictive (multiscale/multiphysics) methods for the analysis and design of LWRs and non-LWRs, in concert with the NE experimental programs.

**NEAMS core competencies:**

- **Multiscale fuel performance and structural materials degradation modeling:** BISON, GRIZZLY, YellowJacket
- **Reactor Physics:** GRIFIN, MPACT, Shift
- **Multiscale thermal fluids:** CTF, SAM, PRONGHORN, Sockeye, Nek5000
- **Multiphysics:** MOOSE, VERA
- **Workflow Management:** Workbench

**Key Success Metric:** Use of NEAMS technology (either software or R&D) by stakeholder to improve how they “do business.”

*Courtesy Dr. Chris Stanek, NTD NEAMS, MSR ORNL Workshop 12-13 OCT 2022*
The Molten Salt Thermal Properties Data Base (MSTPDB)

- Pure and pseudo binary salts properties as a function of temperature and composition
- MSTPDB – Estimation of data-based property and phase equilibria of multicomponent salt mixtures
  - AIMD and MD

**MSR Campaign**

**NEAMS Campaign**

**MSTDB- TP**

**MSTDB- TC**

Calculated density for the pseudo-ternary NaCl-KCl-UCl$_3$ system extrapolated from the pseudo-binaries
• The MSTDB-TP is a collection of empirical models for representing $\rho$, $\nu$, $\kappa$, and $C_p$ 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).

• 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

**Fluorides**

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

**Chlorides**

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

MSTDB-TC
Thermochemical

MSTDB-TP
Thermophysical
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

**ANL** - Distributed salt chemistry monitoring and control

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

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.
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

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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

Courtesy Dr. Sara Thomas, ANL
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

1. Start with A617 and 316H cylindrical stock
2. Stir-friction weld together 2 pieces of A617 to 1 piece of 316H
3. Machine inner specimen from welded rod, machine casing from larger diameter cylindrical stock
4. Join casing and inner specimen with electron beam welds
5. Completed test articles
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.

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
**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:**
- **Molten Salt Tritium Transport Experiment** (“Misty”)
- Forced-convection FLiBe loop with versatile test section
- Hydrogen isotope (tritium) injection and extraction capability
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

*Courtesy Dr. Jennifer Leute and Dr. David Luxat*
• **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

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**Relevant References:**

Fiscal Year 2022
Consolidated Innovative Nuclear Research

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

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 stoichiometric 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 (e.g., Alloy 316H and 316L), 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 Tennessee, 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.
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?

Message to COP26:
To reduce emissions and mitigate climate change dramatically and quickly, the world needs to take decisive actions urgently and in a way that does not put limits on life, well-being, and economic development. Our analysis indicates that nuclear energy along with renewables provides a credible pathway to net-zero.

William D. Magwood, IV
NEA Director-General


You have an important role in helping to create a better future for the entire world. That’s because, as nuclear innovators and leaders, you are in a position to help our world rise to meet two significant, interwoven challenges: stopping climate change and raising global living standards.

Bill Gates

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*

Molten Salt Thermal Properties Workshop  
July 14, 2020
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.
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.
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.**
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).

https://www.nrc.gov/reactors/newreactors/advanced.html#preAppAct
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.
A particular emphasis of NEAMS program is our contribution to the development to the Molten Salt Thermodynamic Database (MSTDB)

<table>
<thead>
<tr>
<th>54 pseudo-binary systems</th>
<th>26 pseudo-ternary systems</th>
</tr>
</thead>
<tbody>
<tr>
<td>NaF-LiF</td>
<td>UF₆-NaF</td>
</tr>
<tr>
<td>BeF₃-LiF</td>
<td>UF₆-ThF₄</td>
</tr>
<tr>
<td>KF-LiF</td>
<td>CsF-ThF₄</td>
</tr>
<tr>
<td>RbF²-LiF</td>
<td>LaF₃-NaF</td>
</tr>
<tr>
<td>CsF²-LiF</td>
<td>ThF₄-BeF₂</td>
</tr>
<tr>
<td>PuF₃-LiF</td>
<td>ThF₄-BeF₂</td>
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<tr>
<td>CsF²-LiF</td>
<td>NaCl-CaCl₂</td>
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<tr>
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<tr>
<td>CsF²-LiF</td>
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</tr>
</tbody>
</table>

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)

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
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

Program: NEAMS
Atomistic simulations for salt properties

DFT employed to simulate chloride fuel salts (near term focus NaCl-UCl₃) 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.

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

Fuel thermal-mechanical response

Reactivity Effects

Core degradation

Fission product release and transport

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

Diffusion from intact TRISO

Diffusion from failed TRISO

Recoil fission source

Release from Intact TRISO

Failing Intact TRISO

Transition from Intact to failed

Released to the matrix

Released to failed TRISO (Modified Booth)

Recoil

Contamination

Diffusion

Recoil TRISO

Failed TRISO

Distribution calculated from diffusion model
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

- Fission Product Forms
  - Soluble FPs (salt-seeking) Form 1
  - Insoluble FP suspension Form 2
  - Salt liq./gas Interface Form 3
  - Insoluble FP deposits on structures Form 4
  - FPs Vapor/NCG Form 5

- Equilibrium
  - Soluble & Volatile FP

- Transport within MSM
  - Soluble & Volatile FP

- Headspace Release
  - Volatile Vapor and Eventual Aerosol

- No Release
- Bubble Film Rupture Aerosol

- Deposits on heat structure
- Deposits on core

- Gas Bubble Rise
- Gas Release

MELCOR-provided state
Atmospheric Release Mechanisms
Derived from standard point kinetics equations

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

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

\[
\frac{dC_i^{\text{loop}}}{dt} = \left(\frac{V_{\text{core}}}{\tau_{\text{core}} V_{\text{loop}}}\right) C_i^{\text{core}} - \left(\lambda_i + \frac{1}{\tau_{\text{loop}}}\right) C_i^{\text{loop}}
\]

\[
\beta_{\text{eff}} = \beta - \beta_{\text{lost}} = \beta - \left(\frac{\Lambda}{P}\right) \sum_{i=1}^{6} \lambda_i C_i^{\text{loop}}
\]
FHR Demonstration Model – UCB Mark I

Legend:
- Primary coolant
- Graphite
- Fuel pebbles
- Blanket pebbles
- Primary coolant flow
- Water flow
- Air flow
- Natural gas flow

Diagram:
- DRACS condenser
- Cooling chimney
- Polar crane
- Cylindrical shield building
- Personnel airlock
- Reactor deck
- Grade level
- Reactor cavity thermal shield
- Reactor cavity
- Base mat
- Drain tank
- Air duct
- Turbine pedestal
- Common utilities tunnel
- Air duct vault
- Intake air filter
- Main transformer

Fuel to Reactor Core to Reactor System to 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.
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 UO₂ TRISO diffusivity and UO₂ failure data were used. Both are changeable through user input with design-specific data.
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
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

**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

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

Computational Modeling
Rigid and Polarizable Ion Models, AIMD, ML

Phillip Halstenberg  Alexander Ivanov  Shannon Mahurin  Sheng Dai

Matt Emerson  Shobha Sharma  Claudio J. Margulis  Santanu Roy  V. Bryantsev

Anatoly Frenkel  Simerjeet Gill  Ruchi Gakhar  William Phillips  Ryan DeFever  Haimeng Wang  Yong Zhang  Ed Maginn

ORNL  U. Iowa  ORNL  BNL  INL  U. Notre Dame
Transport properties

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

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

Calculating Melting Points: Two Approaches

**Pseudo-Supercritical Path (PSCP)**

Liquid → Weak Liquid (WL) → Dense Weak Fluid (DWF) → Weak Crystal (WC) → Crystal (C)

**Direct Coexistence (DC)**

**Example:** NaCl

\[ \Delta G \approx \Delta A_{i \rightarrow j} = \int_0^1 \left( \frac{\partial U}{\partial \lambda} \right) d\lambda \]

Comparison of melting points for RIM and PIM potentials

Methods comparison

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

<table>
<thead>
<tr>
<th></th>
<th>PSCP</th>
<th>Direct coex.</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiCl</td>
<td>782</td>
<td>777</td>
</tr>
<tr>
<td>NaCl</td>
<td>1082</td>
<td>1081</td>
</tr>
<tr>
<td>KCl</td>
<td>1039</td>
<td>1038</td>
</tr>
<tr>
<td>RbCl</td>
<td>1091</td>
<td>1091</td>
</tr>
</tbody>
</table>

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)

<table>
<thead>
<tr>
<th></th>
<th>Expt.</th>
<th>RIM</th>
<th>PIM</th>
</tr>
</thead>
<tbody>
<tr>
<td>LiCl</td>
<td>883</td>
<td>777</td>
<td>1025</td>
</tr>
<tr>
<td>NaCl</td>
<td>1073</td>
<td>1081</td>
<td>1140</td>
</tr>
<tr>
<td>KCl</td>
<td>1043</td>
<td>1038</td>
<td>1053</td>
</tr>
<tr>
<td>RbCl</td>
<td>988</td>
<td>1091</td>
<td>1035</td>
</tr>
</tbody>
</table>

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


We are now modeling phase diagrams for binary systems.
Simulations Reveal Complexity and Intermediate-Range Structure in a Binary Salt

$\text{KMgCl}_3$

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

Slide credit: Claudio Margulis, U. of Iowa

“Anomalous” Temperature Dependence of the Intermediate-Range Order

The SEM-Drude model: A fast polarizable force field for the study of neat KCl, neat MgCl$_2$, and their mixtures

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

A **computationally-efficient polarizable model** developed to study the structure, transport and thermodynamics 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.**

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.

Thrust 1, Aim 2: Molten salt interactions with solutes

X-ray Absorption Spectroscopy
X-ray Scattering

A. Ivanov (ORNL)
S. Gill (BNL)
A. Frenkel (BNL)
V. Bryantsev (ORNL)
S. Roy (ORNL)
C. Margulis (Iowa)
R. Gakhar (INL)

Optical spectroscopy

Ab-initio Molecular Dynamics

Core Activity: Molten salt preparation
Thrust 1. MS Properties and Reactivity

S. Mahurin
S. Dai

Thrust 1 – Aim 3 Radiation effects

S. Pimblott (INL)
J. Wishart (BNL)
G. Horne (INL)
J. LaVerne (Notre Dame)
Atomic Insight into Metal Ion Solvation Structure in High-Temperature Molten Ionic Medium

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, \textit{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 \textit{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.

Correlated spectroscopic and simulation studies explain local structures of Ni\(^{2+}\) and Co\(^{2+}\) in ZnCl\(_2\)

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.

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.

DOI: 10.1021/acs.jpcb.0c00195
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

**Brookhaven**
- Eric Dooryhee
- Anatoly Frenkel
- Simerjeet Gill
- Benjamin Ocko
- Kotaro Sasaki
- James Wishart

**Idaho**
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- Lingfeng He
- Gregory Horne
- Simon Pimblott

**Deputy Director**

**Notre Dame**
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- Edward Maginn

**Oak Ridge**
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- Sheng Dai
- Alexander Ivanov
- Shannon Mahurin

**Thrust 1 Leader**

**Stony Brook**
- Karen Chen-Wiegart

**Iowa**
- Claudio Margulis
Acknowledgments

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

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

This work was supported as part of the Molten Salts in Extreme Environments Energy Frontier Research Center, funded by the U.S. Department of Energy, Office of Science.
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)
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.

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

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**
- Chemically damageable - may be reparable during use
- Composition may be adjustable during use
- Properties depend on composition and state
- Container breach could release nearly all radionuclides

**Solid Fuel**
- Mechanically damageable
- Composition set prior to use
- Properties depend on fabrication process
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