Closing the Fuel Cycle with Molten Salt Reactor Technology

Dr. Patricia Paviet, National Technical Director

Joint IAEA/NEA/JRC MSR Fuel Cycle Workshop 3-7 November 2025

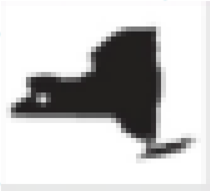
PNNL-SA-217333

History of Reprocessing in the U.S.

Starts as WWII efforts in US and UK

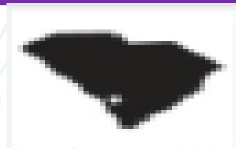
1956
AEC encourages private sector to enter the market

1966
Nuclear Fuels Services licenses the West Valley, NY facility



1967
General Electric licenses the Morris, IL facility

1970
Allied General begins construction of Barnwell, SC facility



1976
Exxon applies for a license

1976
President Ford - reprocessing and recycling of plutonium should not proceed

1977
President Carter - defer any reprocessing due to concerns related to proliferation

1981
President Reagan lifted the ban on reprocessing

1993
President Clinton – Policy statement against reprocessing and recycling

2001
President Bush – US should consider closed fuel cycle for waste minimization and proliferation resistance

2009
President Obama – Blue Ribbon Commission advised to continue long-term R&D on advanced recycling

2017
President Trump - promoting the NE industry through regulatory rollbacks and supporting domestic capabilities

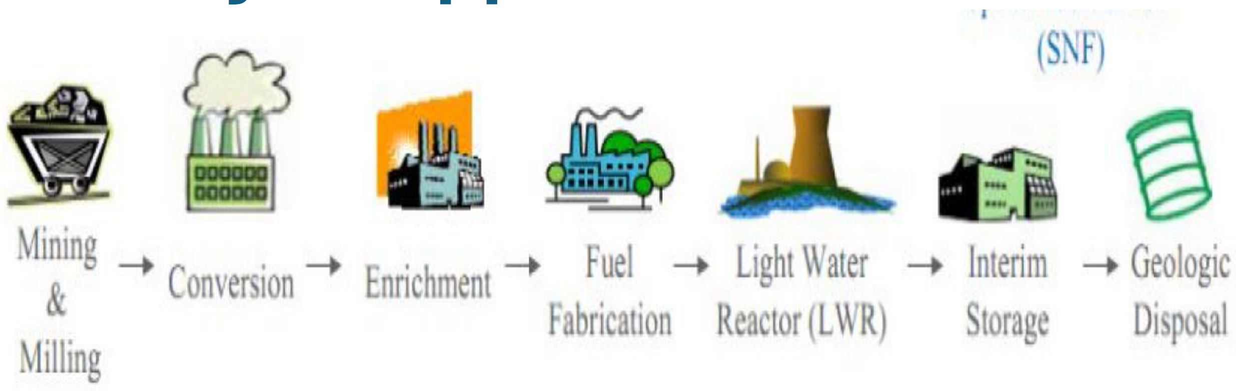
2021
President Biden- nuclear energy is a crucial part of the clean energy agenda

2025
President Trump- Executive orders to accelerate deployment of new nuclear reactor and strengthen the domestic nuclear fuel cycle.

1950 1960 1970 1980 1990 2000 2010 2020

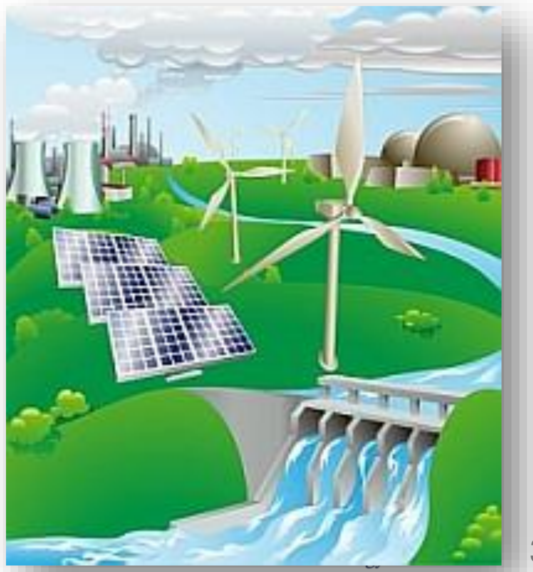
Current U.S. Fuel Cycle Policy

- **Currently, the U.S. is practicing a once-through fuel cycle approach.**
- Used nuclear fuel (UNF) generated @2,000 MT/yr.
- ~94,000 MT of accumulated SNF in the US.



Evaluation of advanced (closed) fuel cycles to support the expansion of nuclear energy and deployment of advanced reactor technologies is needed.

No decisions have been made on future nuclear fuel cycle options in the United States, nor which technologies will be deployed, if a closed fuel cycle is selected.



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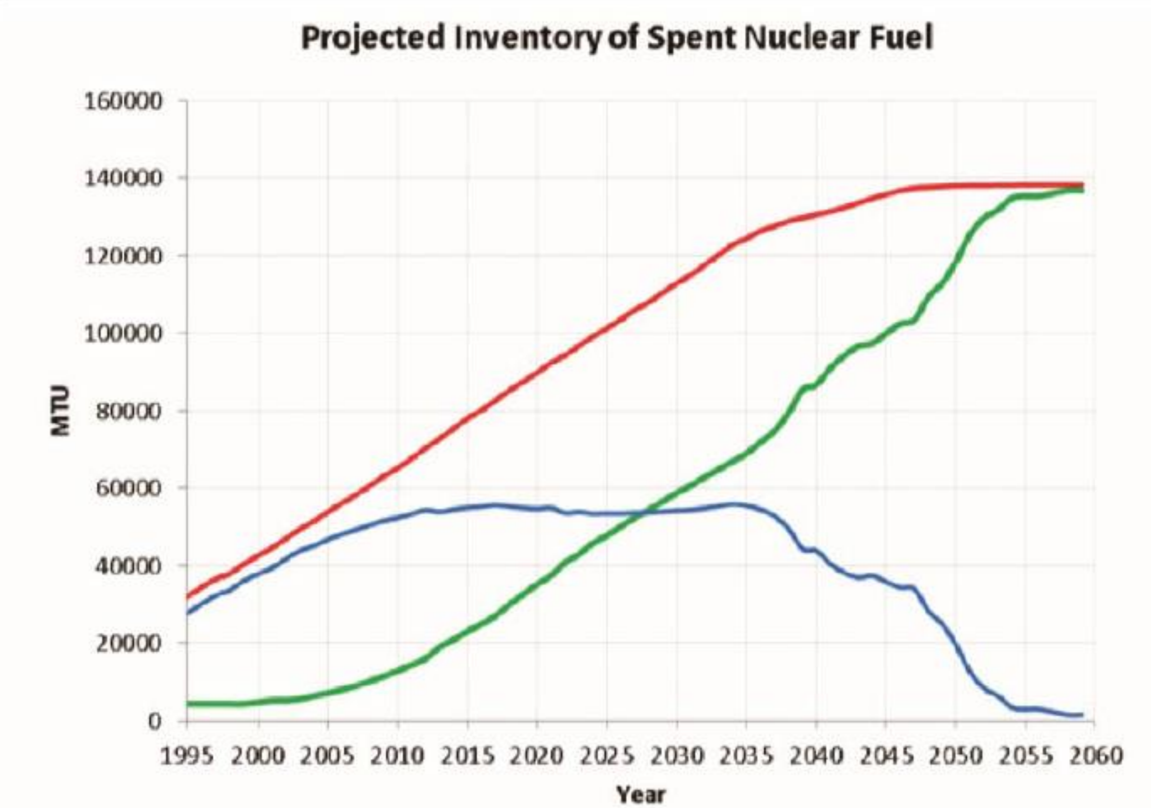
Estimated Number of Geologic Repositories in the U.S. for Different Scenarios of the Cumulative Used Fuel in 2100

Nuclear Futures		Existing License Completion	Extended License Completion	Continuing Level Energy Generation	Continuing Market Share Generation	Growing Market Share Generation
Cumulative Used Fuel in 2100 (MTHM)		90,000	120,000	250,000	600,000	1,500,000
		Existing Reactor Only		Existing and New Reactors		
Fuel Management Approach		Number of Repositories Needed (at 70,000 MT each)				
	Direct Disposal (current policy) Current US policy – June 2025	2	2	4	9	22
	Direct Disposal with Expanded Repository Capacity	1	1	2	5	13
	Limited Thermal recycle With Expanded Repository Capacity	1	1	1	3	7
	Repeated Combined Thermal and Fast Recycle	(Requires	new reactors)	1	1	1

E. Gonzalez-Romero, Euradwaste'08, Luxembourg, 20-22 OCT 08

Projected inventory of SNF until 2060 - Study from Sandia National Lab – March 2018

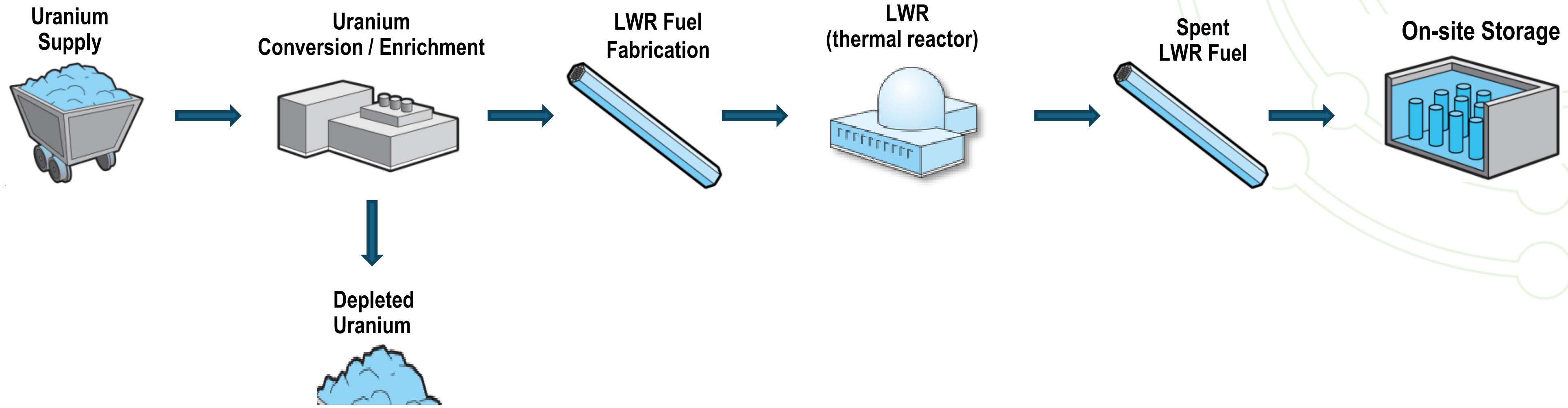
- **In 2018, about 80,150 metric tons heavy metal (MTHM) of SNF—** fuel withdrawn from a nuclear reactor following irradiation, which is referred to as "used" fuel by the nuclear industry because it contains potentially reusable uranium and plutonium—is in temporary storage **at 75 reactor sites scattered across 33 states,** according to the DOE's Sandia National Laboratories.
- Nearly **a third of that SNF—25,400 MTHM—is in dry storage in about 2,080 cask or canister systems**, and the balance is in spent fuel pools, mainly at reactors, or otherwise stranded at independent spent fuel storage installations (ISFSIs) if the reactor has been shut down.
- **Another 2,200 MTHM of SNF is generated nationwide each year**. Because U.S. pools have reached capacity limits, about 160 new dry storage canisters are loaded each year.
- If the Nuclear Regulatory Commission (NRC) grants all license renewals requested by the bulk of the nation's 99-reactor, (extending their operational lives from 60 to 80 years), this would substantially increase the amount of spent fuel requiring long-term storage.



(http://www.powermag.com/wp-content/uploads/2018/03/fig-5_dry-cask-storage_nrc1.jpg)

E. Bonano, E. Kalinina, P. Swift, "The Need for Integrating the Back End of the Nuclear Fuel Cycle in the United States of America", MRS Advances 3(19):1-13 2018

Current U.S. (Open) Fuel Cycle



- The current U.S. fuel cycle is a once-through fuel cycle using enriched uranium fuel in LWRs
- The fuel cycle is not yet complete
  - Disposal paths for spent fuel and depleted uranium are not currently implemented

Courtesy Road Wigeland, retired from INL

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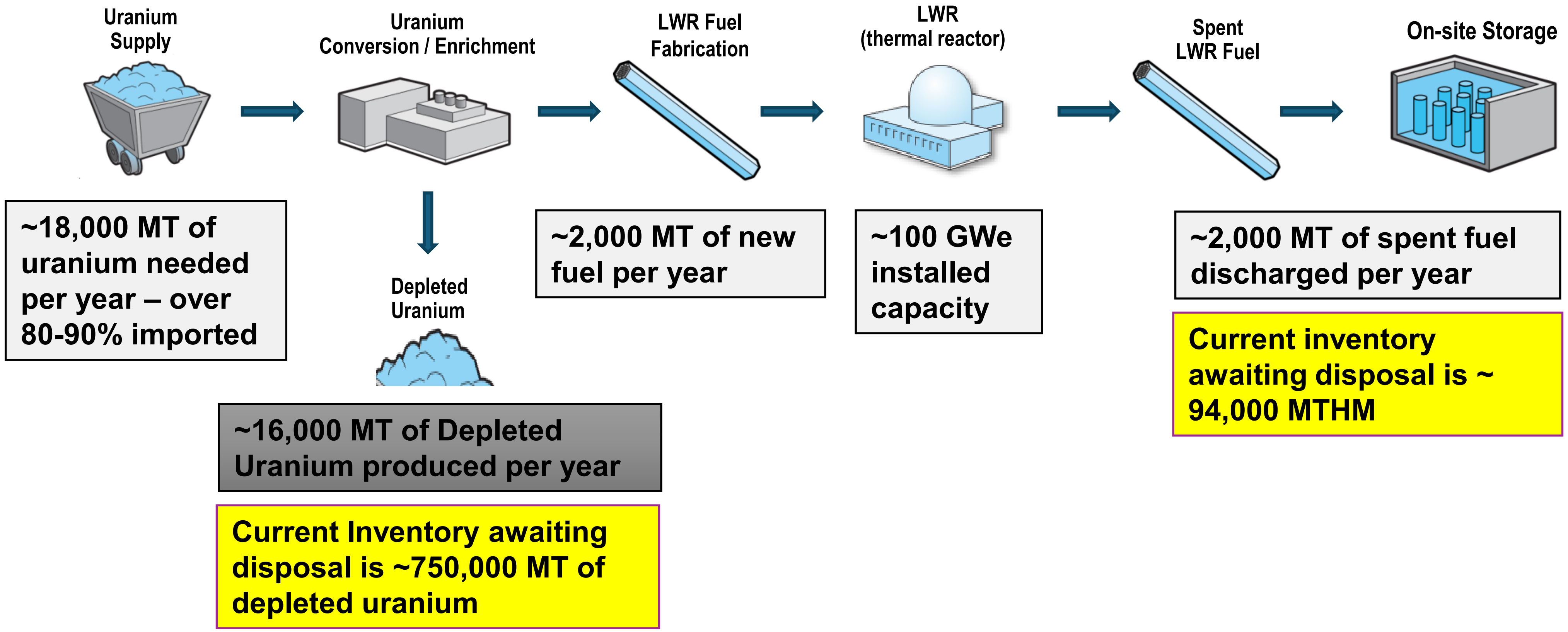
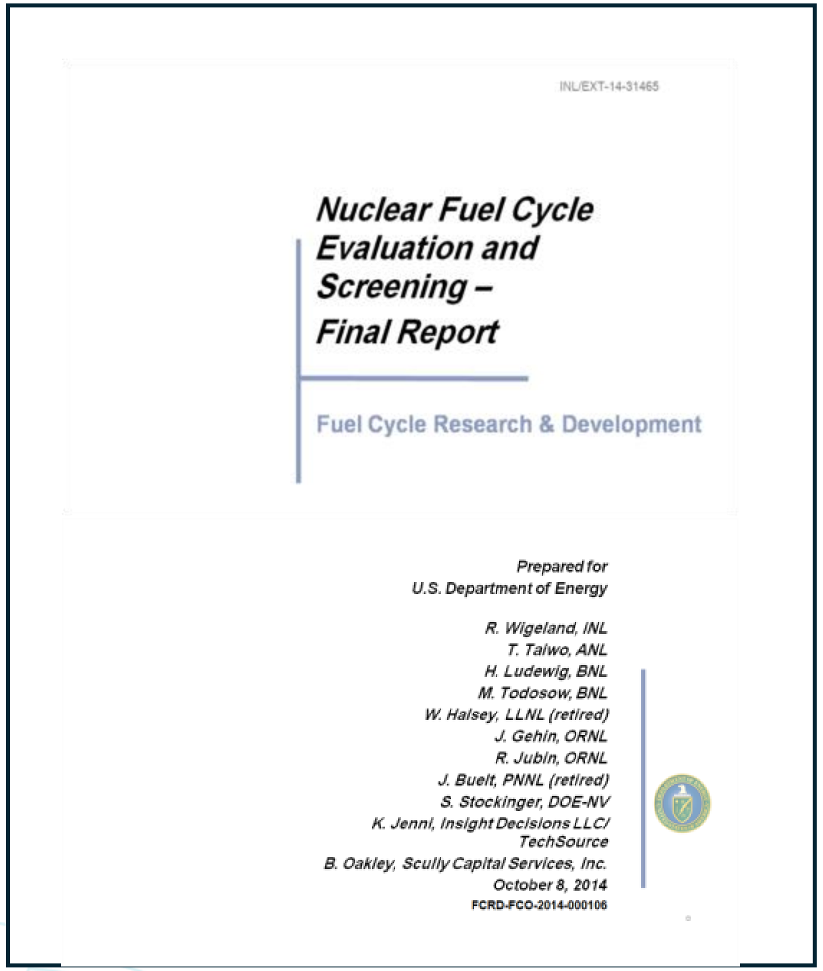
Current U.S. Fuel Cycle Performance



- The current U.S. fuel cycle generates two major waste streams requiring geologic disposal or similar isolation, along with LLW
  - 2,000 MT per year of spent fuel – planned disposal path: geologic repository
  - 16,000 MT per year of depleted uranium – no disposal path identified at this time, but will also require long-term isolation due to buildup of radioactive decay products

Courtesy Road Wigeland, retired from INL


INL/EXT-14-31465

Nuclear Fuel Cycle Evaluation and Screening – Final Report

Fuel Cycle Research & Development

Prepared for U.S. Department of Energy

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K. Jenni, Insight Decisions LLC/ TechSource
B. Oakley, Scully Capital Services, Inc.
October 8, 2014
FCRD-FCO-2014-000106

The study was completed in October 2014

- The study results are based on physics, not specific technologies
- The study was informed by Analysis Examples with specific reactor technologies to calculate fuel cycle performance as needed

The completed study showed that fuel cycles different from the current U.S. once through fuel cycle could achieve substantial improvements

The Evaluation and Screening Study

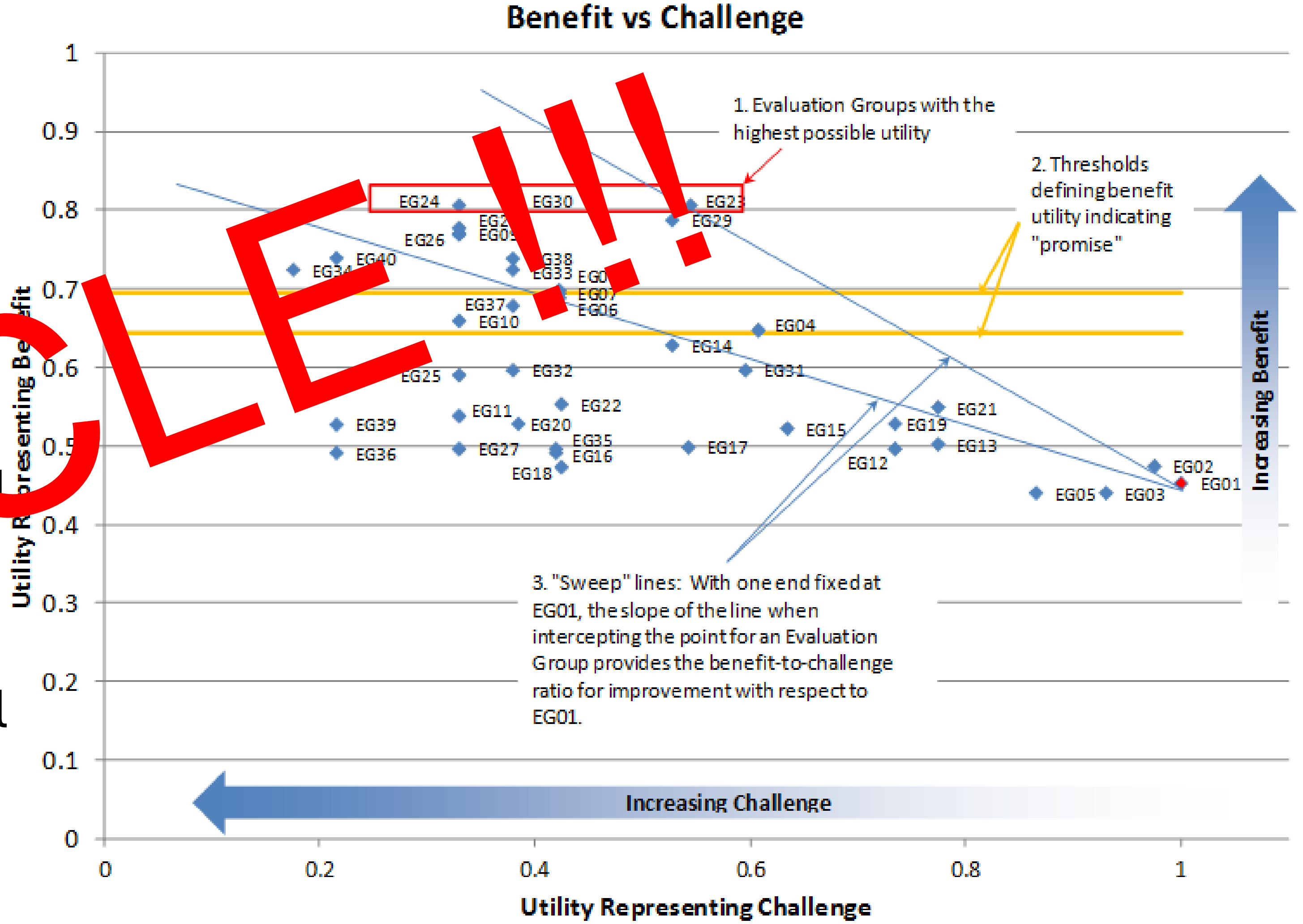
- **DOE/NE chartered the Nuclear Fuel Cycle Evaluation and Screening Study in December 2011**
  - A comprehensive examination of all possible fuel cycles to identify promising fuel cycle options with the potential for substantial improvements compared to the current U.S. fuel cycle
  - DOE/NE specified the Evaluation Criteria for the study, which included safety, proliferation risk, waste management, etc.

https://fuelcycleevaluation.inl.gov/SitePages/Home.aspx

Implementing preferred Fuel cycles while using Capabilities for near-term Applications

- **Most Promising Fuel Cycle Options:**
  - EG23 - Continuous **recycle** of U/Pu with new natural-U fuel in fast critical reactors
  - EG24 - Continuous **recycle** of U/TRU with new natural-U fuel in fast critical reactors
  - EG29 - Continuous **recycle** of U/Pu with new natural-U fuel in both fast and thermal critical reactors
  - EG30 - Continuous **recycle** of U/TRU with new natural-U fuel in both fast and thermal critical reactors

RECYCLE!!!



- **After an exhaustive study, it was concluded that only options with continuous recycle are the most promising**

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https://fuelcycleevaluation.inl.gov/SitePages/Home.aspx

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)	Fluoride and Chloride salts	U/Pu , U/TRU, U/Th in salt, and natural U in salt	Closed*	Electricity, Hydrogen** Desalination	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 >750ºC is assumed for higher-efficiency hydrogen production, and >850ºC 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

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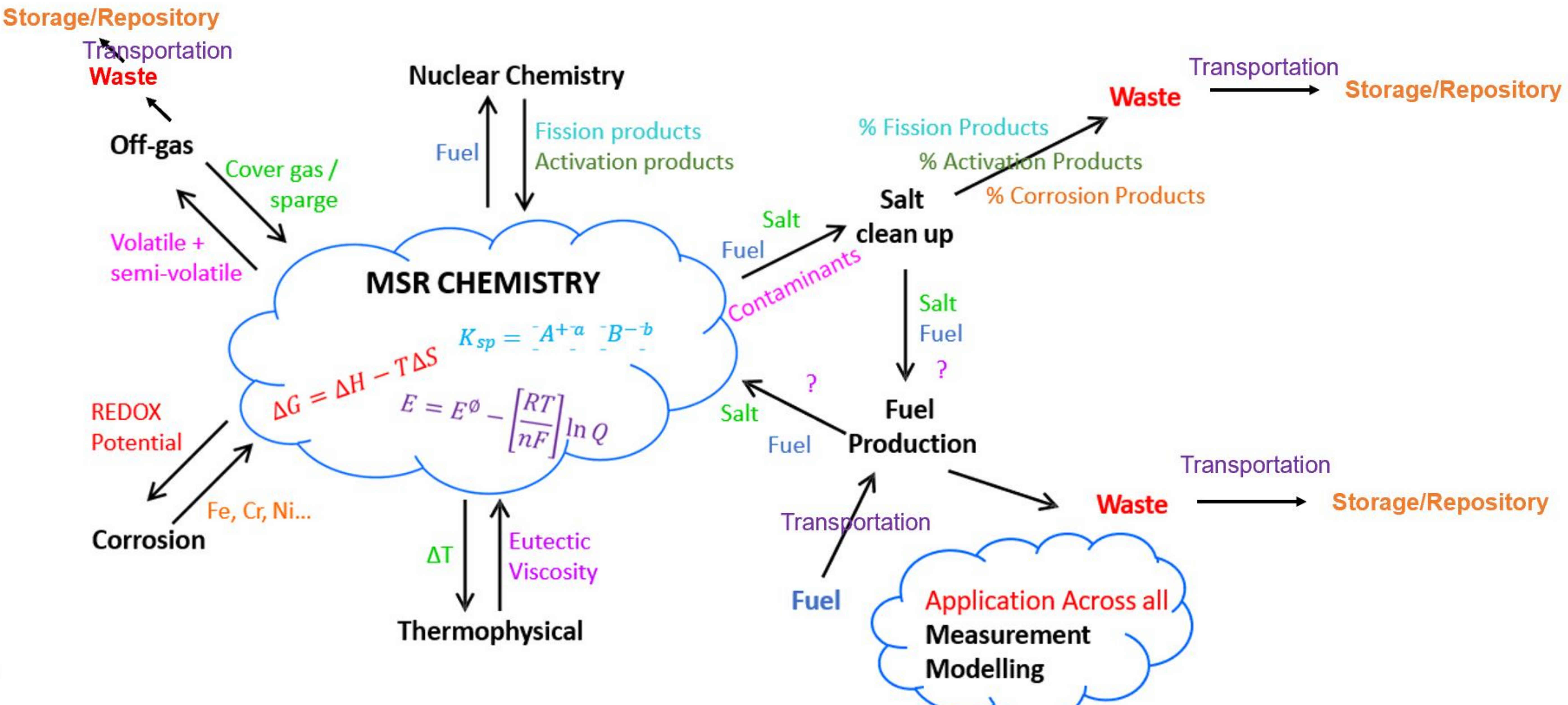
Courtesy R. Wigeland, INL

Closing the fuel cycle with MSR – A dream comes true

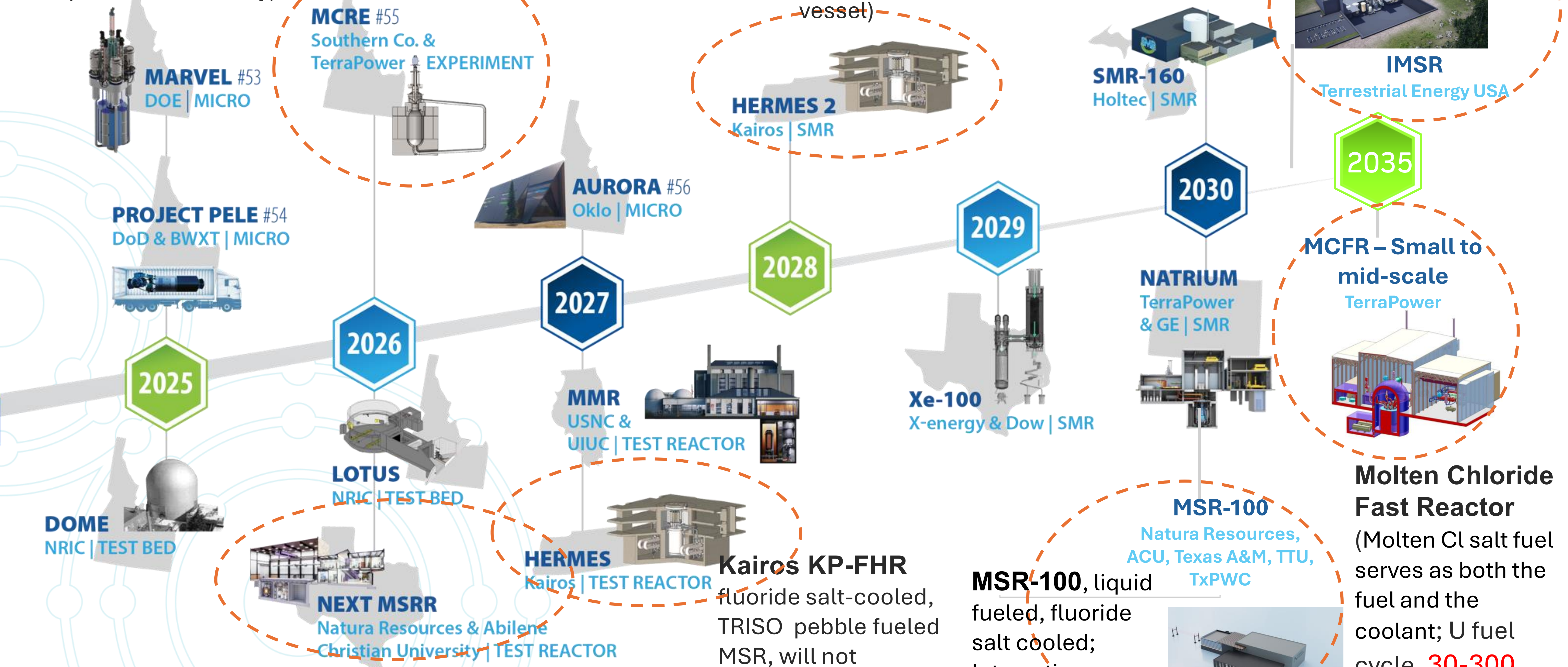


Courtesy Mike Edmonson, NNL, JUN 2023

Accelerating Advanced Reactors Demonstration & Deployment in the U.S.

Molten Chloride Reactor Experiment (only liquid fueled design supported under ARDP – will not produce electricity) - 150 kWth

Integrated Molten Salt Reactor (molten fluoride salt reactor; integration of primary reactor components into sealed replaceable vessel)



MSR-1, Low power (up to 1 MWth, to support academic research

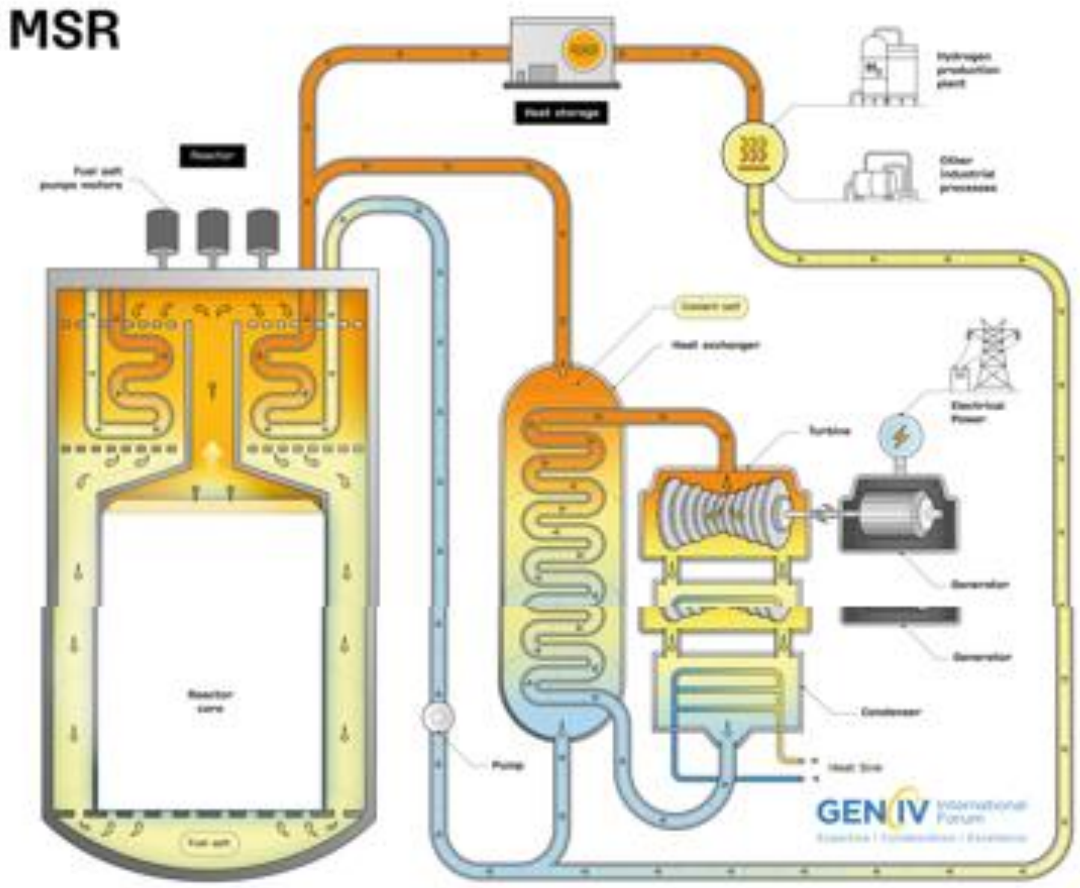
Kairos KP-FHR fluoride salt-cooled, TRISO pebble fueled MSR, will not produce electricity, 35 MWth

MSR-100, liquid fueled, fluoride salt cooled; Integrating desalination with energy production

Molten Chloride Fast Reactor (Molten Cl salt fuel serves as both the fuel and the coolant; U fuel cycle, 30-300 MWe

Some challenging operating conditions

- High Temperatures: Operations temperatures ranging from 600 °C to 750 °C
- Radiation: Exposure to neutron and gamma radiation
- Corrosiveness of Molten Salts increases with impurities and fission products: Interaction with fluoride and chloride mixtures
- Off-gas management
- Salt clean-up/processing



And other challenges

Chemistry Challenges

- Gaps in thermophysical and thermochemical properties of molten salts, Actinide fluoride and chloride bearing mixtures. Limited irradiation studies on chloride fuel salt. Enrichment/Separation .

Monitoring and Chemistry Control of Molten Salts, with potential Online Purification

- Fuel Salt Processing: The online processing of liquid fuel salts to extract fission products and manage the chemistry of the salt mixture is one of the most promising aspects of MSRs. However, this area needs further investigation and demonstration to ensure reliability and efficiency. Some gaseous fission products that are continuously generated in the fuel must be monitored, trapped or released.

Safety Demonstration and Regulation

- Novel Designs and Fuel Cycles: MSRs employ innovative designs and fuel cycles, necessitating thorough safety demonstrations and regulatory approval. Developing safety cases and regulatory frameworks for MSRs is a key condition for its success.

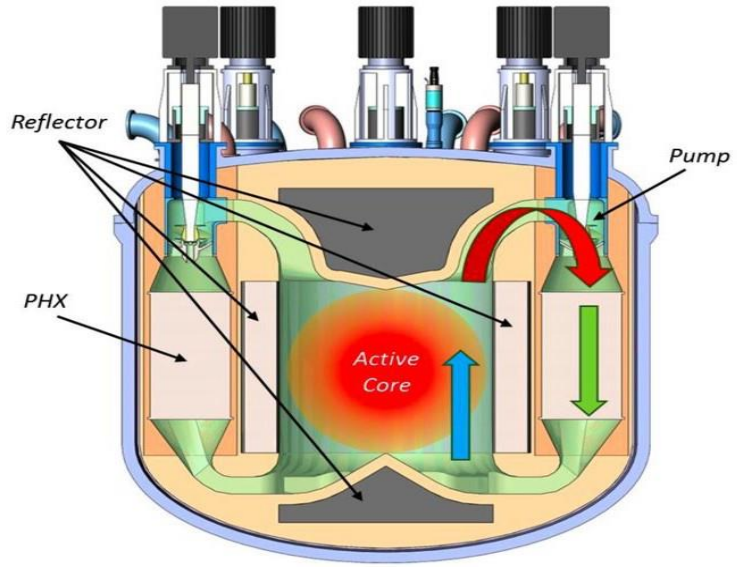
Material Challenges

- MSRs operate at high temperatures and with molten salts for which corrosion can be an issue. Finding materials that can withstand this harsh chemical environment, resist corrosion, endure high temperatures, and tolerate neutron fluxes over long periods remains a significant challenge. Addressing these materials challenges requires extensive research and development. This includes testing and qualifying new alloys, composites, and coatings that can meet the demanding conditions of MSR operation.

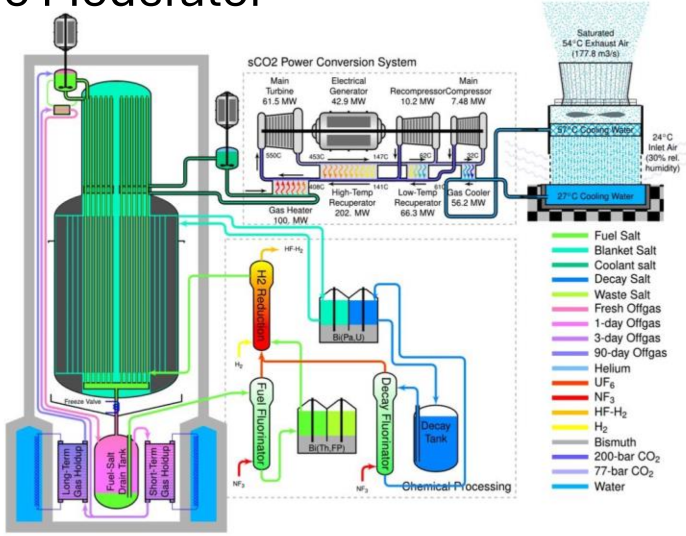
Scale-up and Commercialization

- Scaling Up Technology: Transitioning from experimental or small-scale models to full-scale commercial reactors involves significant challenges in scaling up the technology and demonstrating its economic viability at a commercial level.

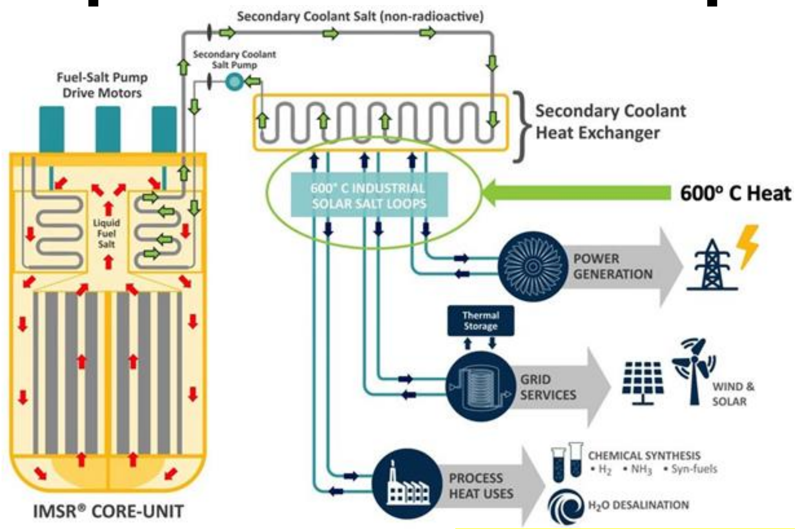
Example of MSR development



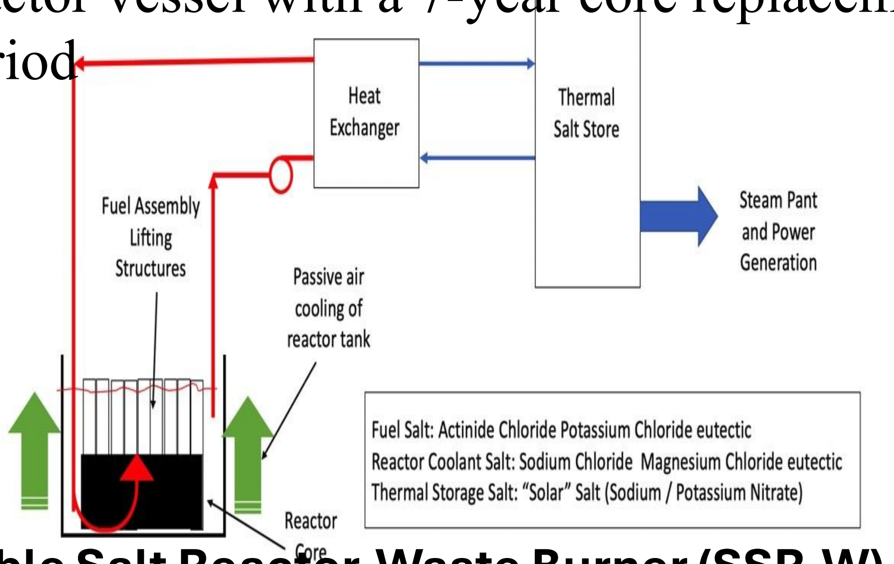
Molten chloride fast reactor (MCFR) -TerraPower – NaCl-UCl3
Primary cooling fluid: NaCl-MgCl2 - No Moderator



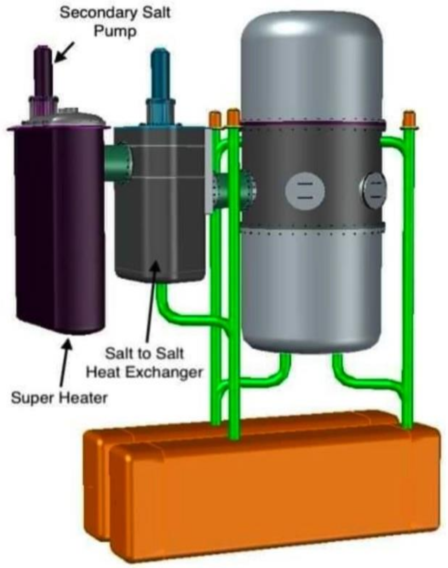
Lithium fluoride thorium reactor (LFTR) (Flibe Energy) - Fuel salt – LiF-BeF2- UF4 (FliBeU), Blanket salt - LiF-ThF4 (FLiTh) – moderator Graphite. Replace every 4-year graphite



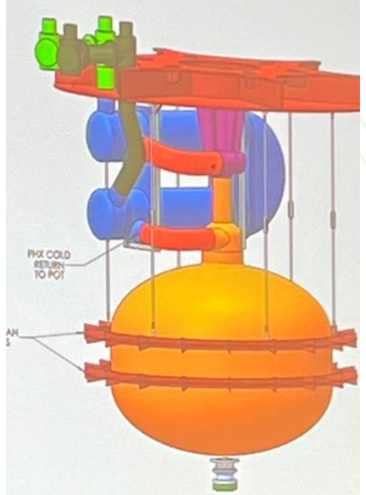
Integral Molten Salt Reactor – Terrestrial USA–
Burner design with 2% LEU at start-up and <5% LEU makeup. Fluoride based fuel salt -sealed reactor vessel with a 7-year core replacement period



Stable Salt Reactor-Waste Burner (SSR-W) (Moltex Energy) - 45% KCl 55% Pu and mixed lanthanide/actinide tri- chlorides in traditional fuel pins, Coolant Salt – NaCl/MgCl2 – No moderator, Lifetime 60 yrs



Molten chloride salt fast reactor (MCSFR) Exodys Energy – Used Nuclear Fuel (U/Pu)O2 converted to UCl3/PuCl3 – Chloride based Fuel Salt – Fuel Purification 40-60 yrs



Thermal molten salt ThorCon US, Inc. - NaF-BeF2-UF4 (72-16-12 mol%), Moderator Graphite, lifetime 80 year for power plant - Replace every 4-year graphite

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

Thermophysical and Thermochemical Properties of Molten Salts –
Experimental and Computational
Salt purification and synthesis

TECHNOLOGY DEVELOPMENT

Off-Gas Management
Radionuclide Release Monitoring,
Sensors & Instrumentation
Salt Loops: LSTL & FASTR; MSTTE; ASL; SAAF

NATIONAL AND INTERNATIONAL MSR SAFETY

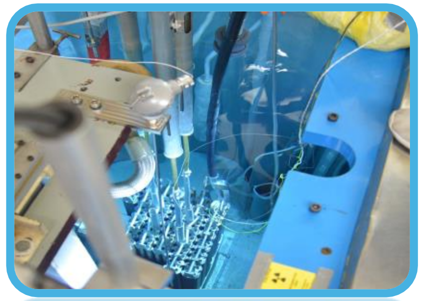
Guidance on reasonable approaches to demonstrating safe and economic commercial prototype molten salt reactors.

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

1) MSRs can provide a substantial portion of the energy needed for the U.S. to have energy security, energy independence.
2) There is a need for an abundant energy for the foreseeable future for data centers, desalination, process heat, hydrogen production...

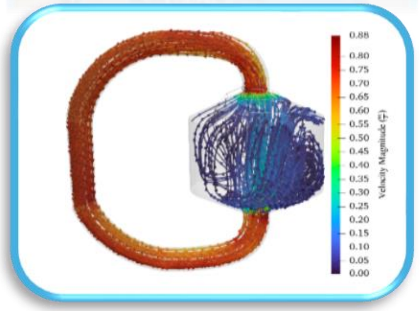
SALT IRRADIATION

Fuel salt irradiation and post irradiation examination capabilities
NRAD, ATR
.



MODELING & SIMULATION

NEAMS Tools & MELCOR utilized for
Species Tracking
Accident Scenario
Mechanistic Source Term...
.



MSR RADIOISOTOPES

Developing new Technologies to separate Radioisotopes of Interest to the MSR Community.

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Molten Salt Chemistry

General Salt Chemistry Consideration

- In general, salts used for high temperature applications, including as fuels and coolants for MSRs, must be of **high purity**
  - Both trace metals impurities and light element impurities are important
    - Affect corrosion, precipitation of oxides, volatility, etc.
- **Moisture is the primary concern** for many salt systems
  - Maintaining anhydrous salts through synthesis, purification and storage is essential
- **Trace element impurities**
  - Affect neutronics, corrosion, etc.
    - Limits are reactor vendor and neutron spectrum dependent
- Control of the redox state governs many chemical properties
  - U^{3+}/U^{4+} ratio control is essential to maintain solubility of various species and minimize corrosion and volatility

Electrochemical Monitoring of Salt Composition[1]

Fuel Salt Synthesis: Feedstock Considerations

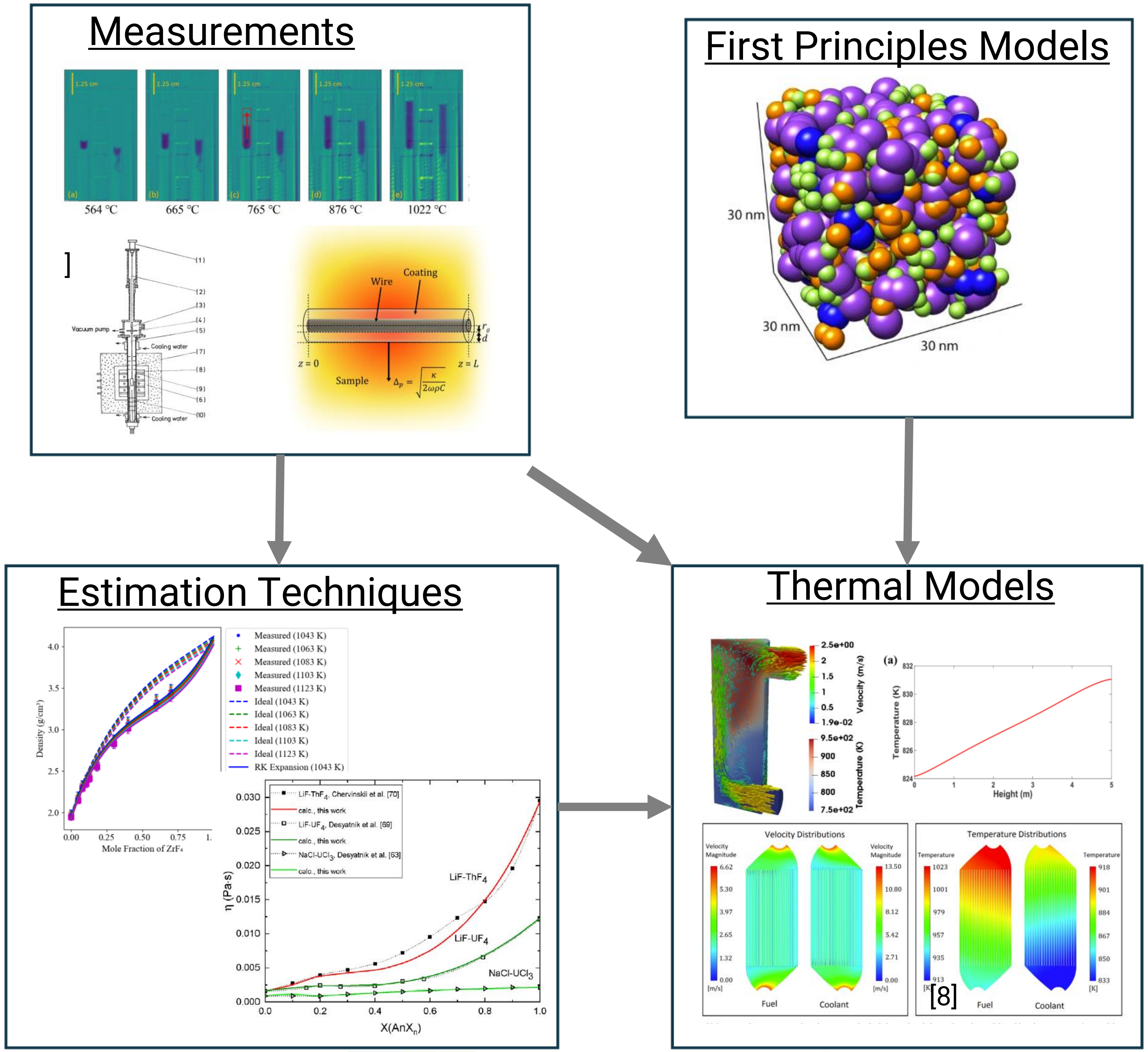
- Synthesis route chosen is dependent on the chemical and physical form of the feedstock to be used
  - Metallic, Oxide, Fluoride, etc.
  - For fresh fuels, minimizing excess steps reduces processing losses, waste, expense, etc.
  - For fluoride fueled reactors, deconversion of UF_6 to UF_4 is relatively straightforward, however, other fuel types are more complicated
- Also dependent on history of feedstock
  - Is the fissile material fresh or irradiated?
  - Incorporating used LWR reactor fuels into an MSR fuel cycle is advantageous from a waste minimization and uranium utilization perspective but introduces significant challenges from a material handling perspective.

Courtesy Bill Philipps, INL and Bruce McNamara, PNNL

Purpose of Thermophysical Characterization for MSRs

- A precise understanding of thermophysical properties of molten salts is necessary for an accurate understanding of thermal hydraulics
  - Necessary for understanding the steady-state temperature distribution, thermal response to transient conditions, etc.
  - Codes like Mole, SAM, and Griffin will need these properties in the Multiphysics framework

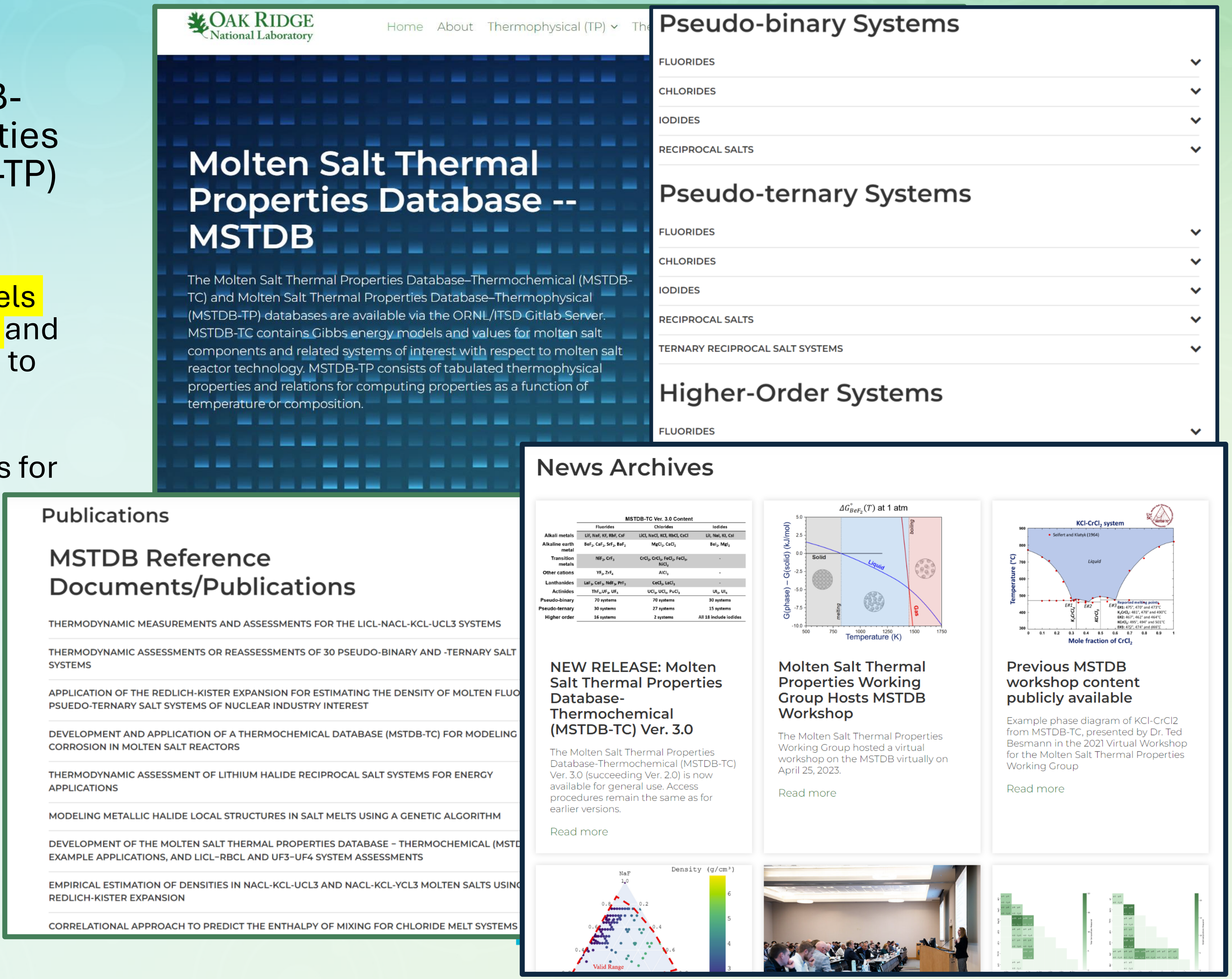
- Developing this understanding for MSR relevant salts is challenging, because:
  - MSRs are typically interested in pseudo-ternary+ mixtures
  - These salts may bear U, Pu, TRU and Be
  - Corrosion and fission products may be introduced over the core/reactor lifetime



Molten Salt Thermal Properties Data Base MSTDB

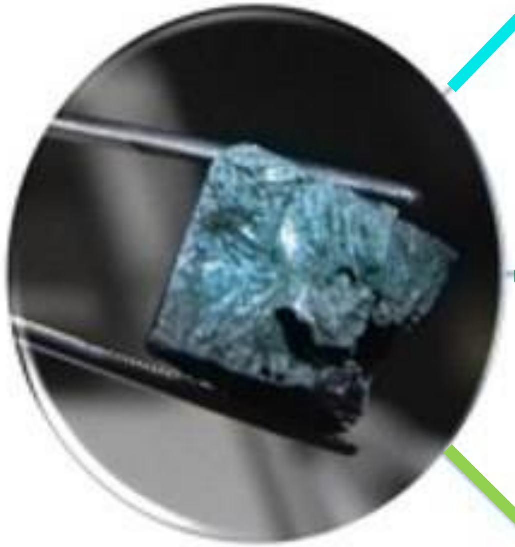
- The Molten Salt Thermal Properties Database–Thermochemical (MSTDB-TC) and Molten Salt Thermal Properties Database–Thermophysical (MSTDB-TP) databases are available via the ORNL/ITSD Gitlab Server.
  - **MSTDB-TC** contains Gibbs energy models and values for molten salt components and related systems of interest with respect to molten salt reactor technology.
  - **MSTDB-TP** consists of tabulated thermophysical properties and relations for computing properties as a function of temperature or composition.
- MSTDB has >320 users currently

Available @ https://mstdb.ornl.gov



OAK RIDGE National Laboratory
NEAMS
UNIVERSITY OF South Carolina
Molten Salt Reactor PROGRAM

MSR Campaign $PuCl_3$-NaCl Collaboration: INL/LANL/ORNL/PNNL



Experimental

- Investigate 3 compositions within binary
- Provide double verified, high fidelity results on thermophysical properties
- Focus: density, melting point and heat capacity

Modeling &Simulation

- Ab Initio Molecular Dynamics (AIMD) method used with solid state parameters
- Validate experimental results
- Extend knowledge into Pu-rich regions

Outcomes

- Peer Reviewed journal article
- Validate data set on eutectic composition
- Experiment – INL and LANL
- Modeling – PNNL and ORNL

Synthesis of $PuCl_3$-NaCl from Pu metal at INL

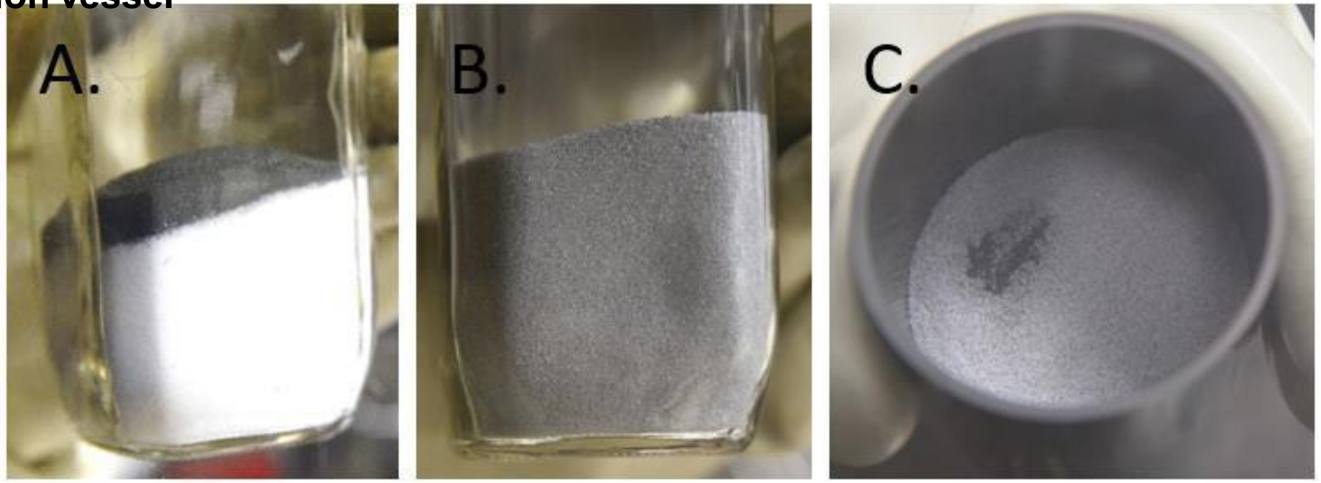
1- Salts

99.99% Pure NaCl
350C, 17.5KPa
3.5 hrs
High purity Ar Atm

99.99% Pure NH_4Cl
100C , 8 hrs

Particle size of NaCl and NH_4Cl was reduced using an agate mortar and pestle to <50 mesh (US sieve).

Homogenization of chemicals for synthesis of NaCl-PuCl3 eutectic composition salt: A. Unmixed NaCl (white), NH4Cl (white), and Pu-metal powders (dark grey), B. Mixed chemicals in glass vial, C. Mixed material in glassy carbon crucible, reaction vessel

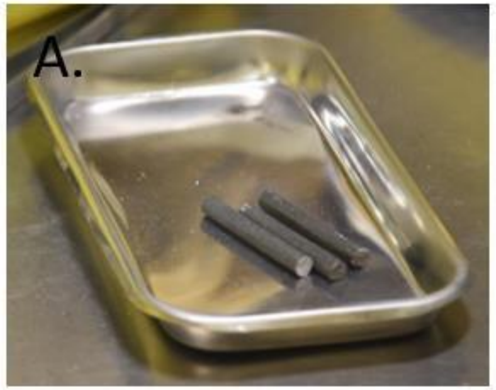


60 hrs
450C to 800C

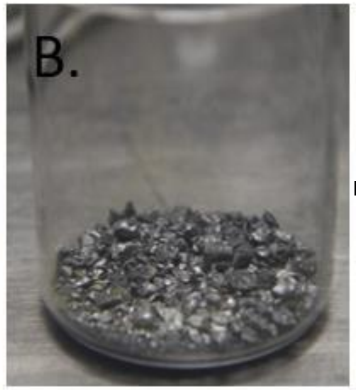
1) De-hydride Process
325C under vacuum
Several hours

2) Chlorination process
NH4Cl/ NaCl

2- Pu metal



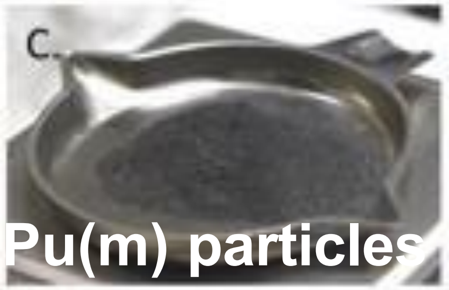
Pu(m)



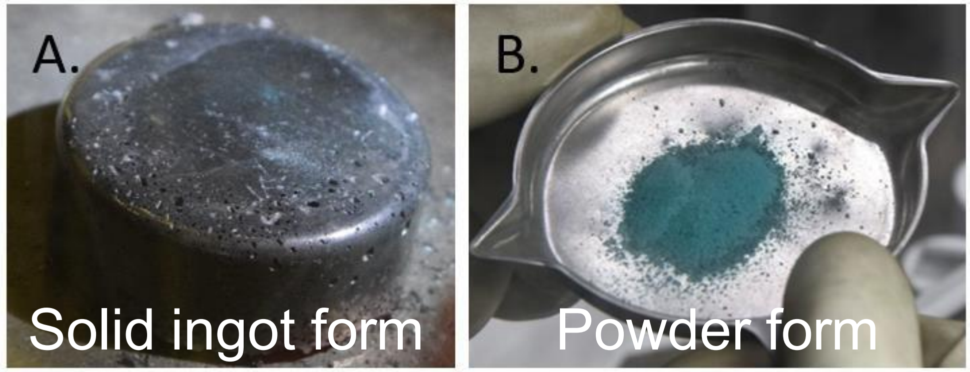
Pu(m) rods

$$Pu + \frac{x}{2} H_2(g) \rightleftharpoons PuH_x$$

Hydride Process



PuH_x



NaCl-$PuCl_3$ eutectic material

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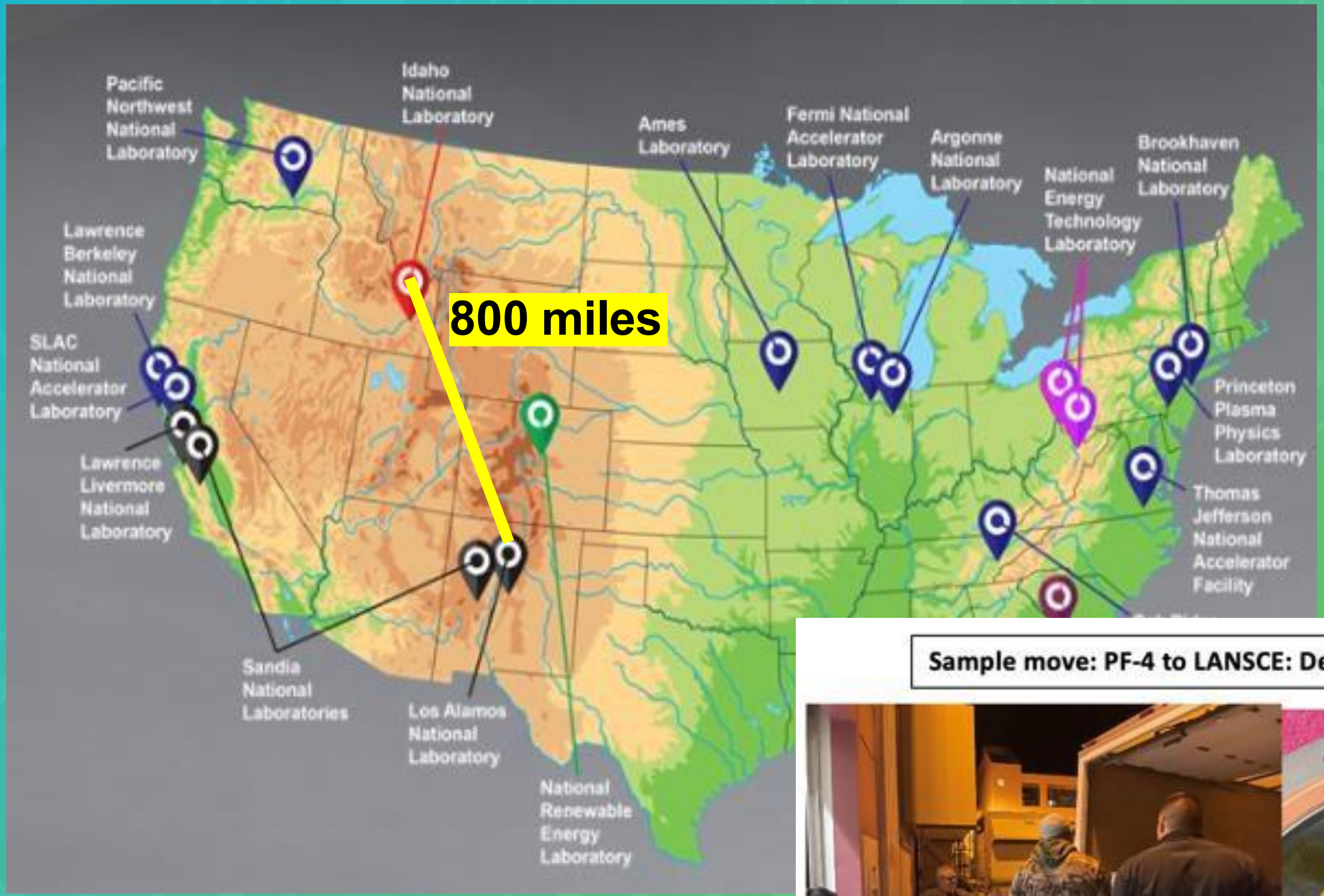
Office of Nuclear Energy

Toni Karlsson et al., J. Mol. Liquids, Vol 387, 122636, (2023)

NaCl-$PuCl_3$ Salt

- Not a commercially available salt
- No research quantities available in DOE complex
- Expensive and difficult to transport
- Limited facilities capable of handling Pu
- So… INL synthesized it and sent some to LANL



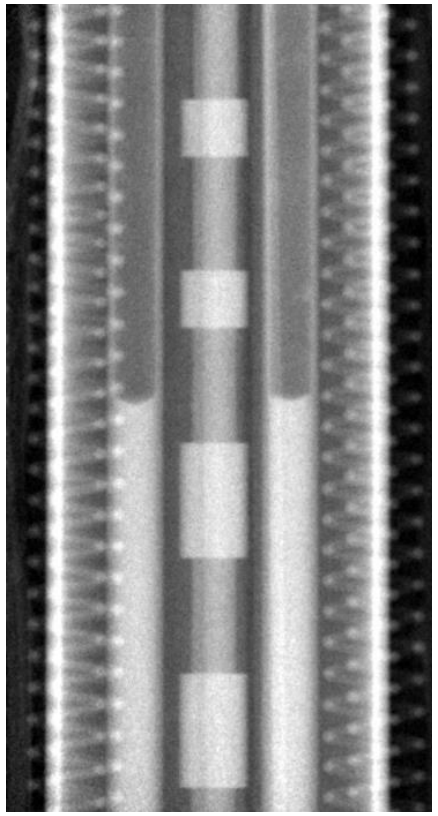
Courtesy Toni Karlsson, INL; Marisa Monreal, LANL

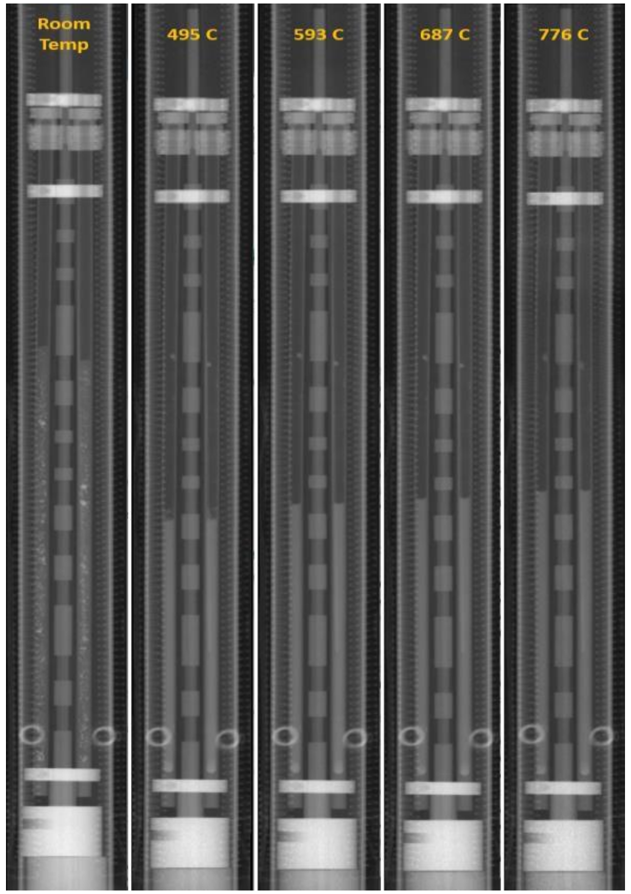
Density Determination - NaCl-$PuCl_3$ Eutectic

Archimedes' hydrostatic method

Experimental density setup at INL showing bottom-loading balance on the stand above a furnace with quartz lid and thermocouple located inside an argon atmosphere glovebox

LANSCE - Neutron Radiography, Preliminary Results (DEC 2022)

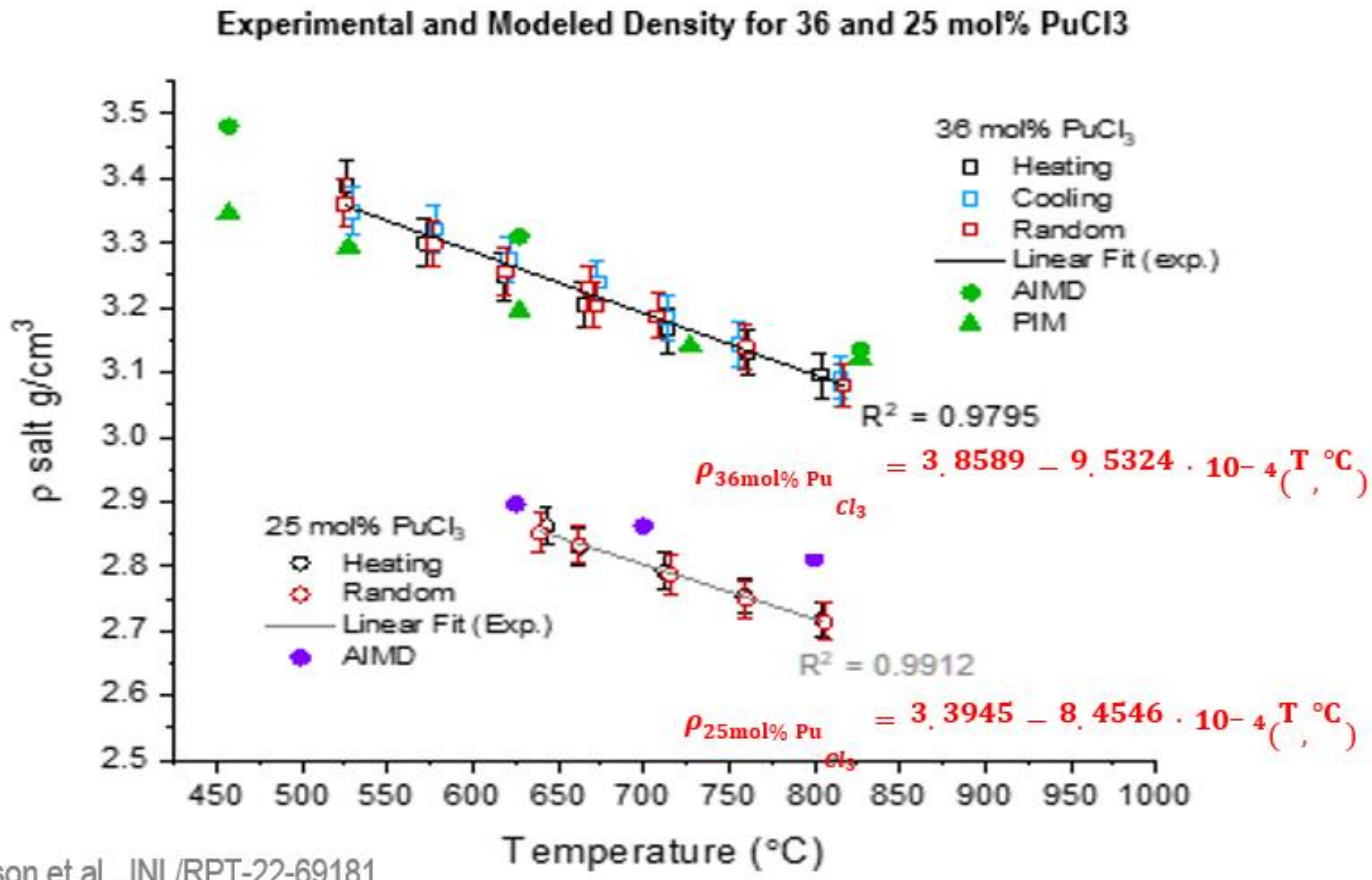




Pu results: neutron images of $PuCl_3$-bearing molten salts at various temp.

State-of-the-art custom furnace
New imaging set-up
Movies of melting and solidification process
Complete automation and remote control
Neutron attenuation images instead of just radiographs

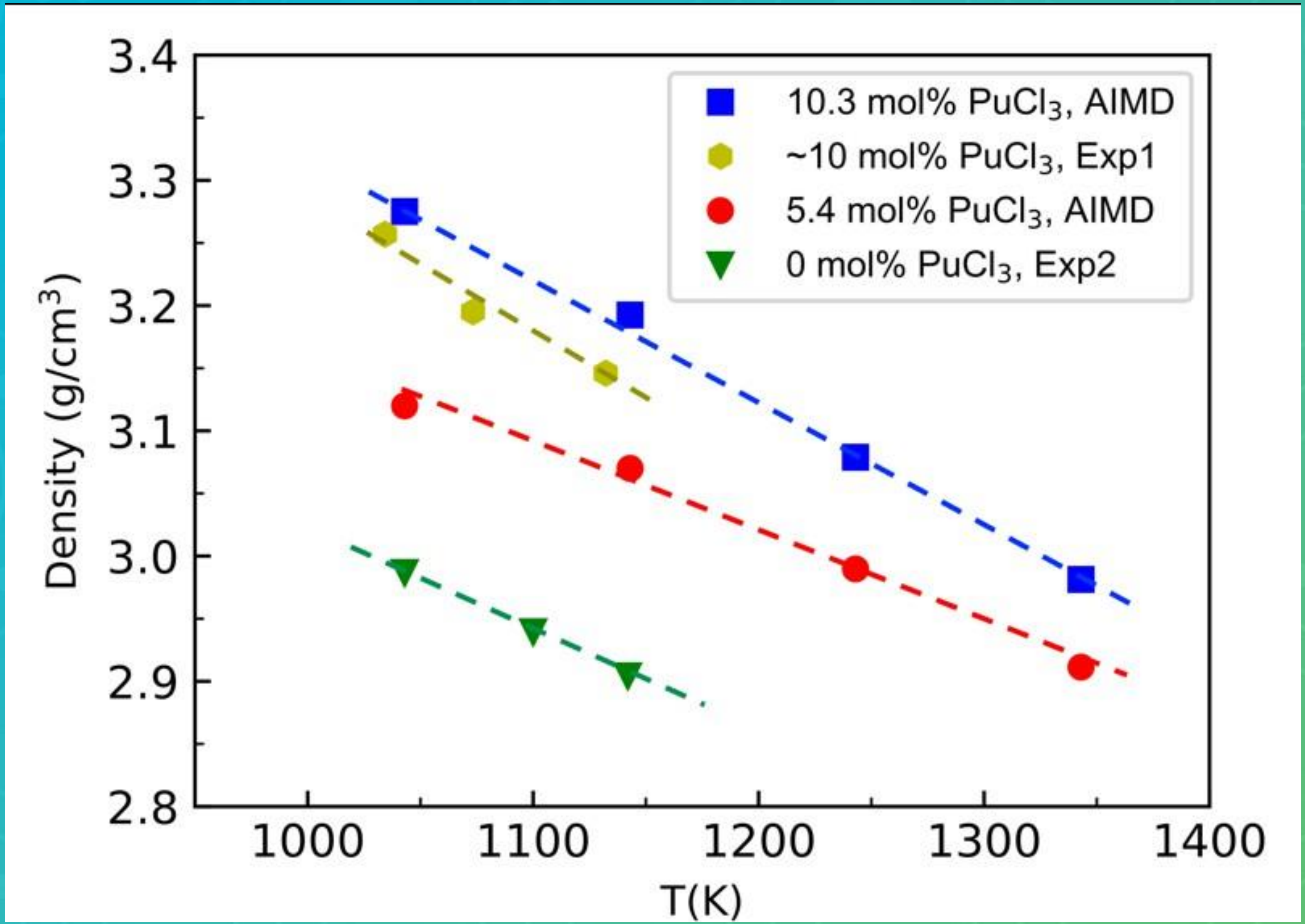
Density determination of NaCl-$PuCl_3$ using Archimedes' hydrostatic method and compared with AIMD

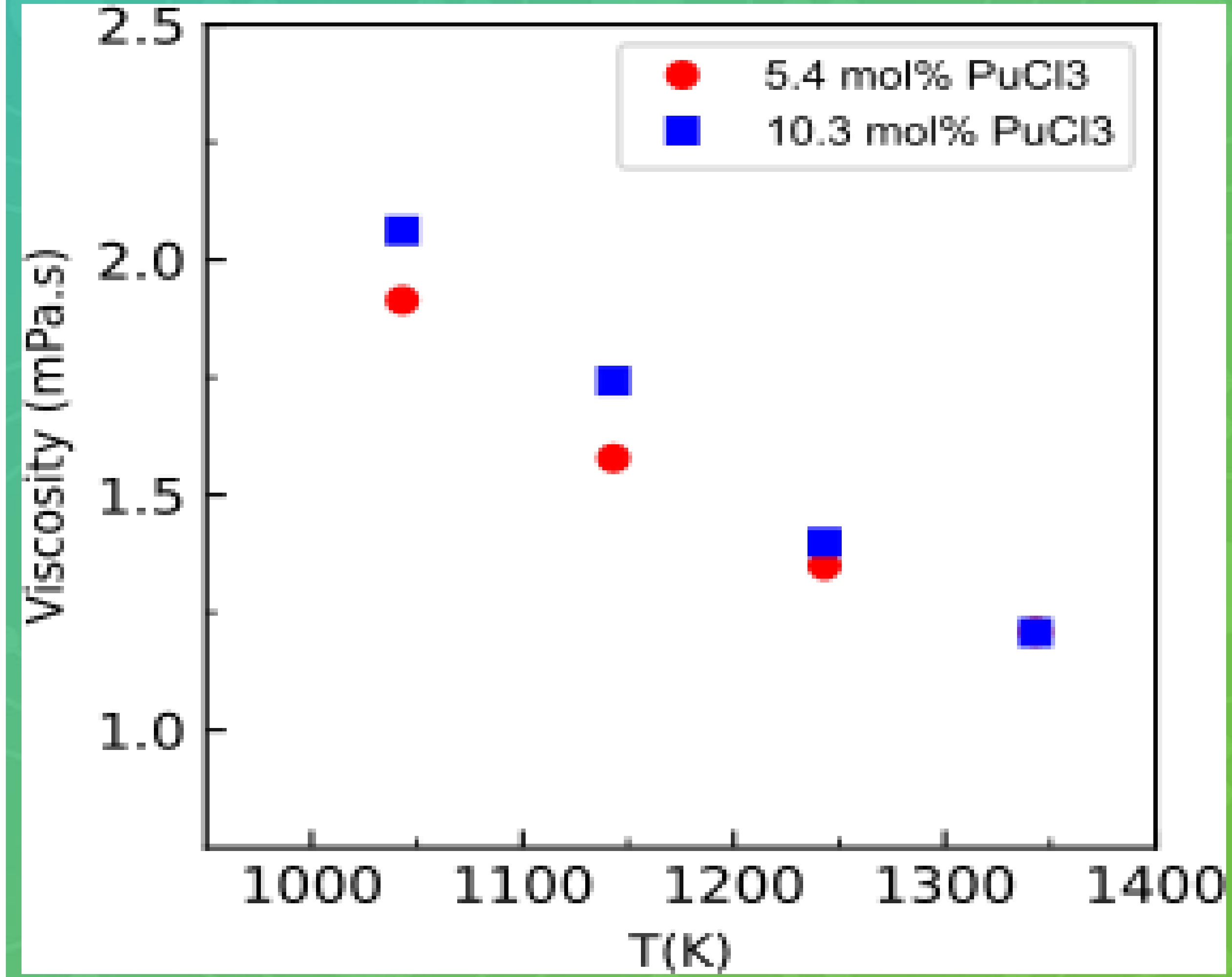


Toni Karlsson et al., INL/RPT-22-69181

Molten Salt Reactor

AIMD and MLIP to determine the Density and Viscosity of NaCl-$PuCl_3$ system at different Pu concentrations





Expt 1- Toni Karlsson, INL
Expt 2 - Desyatnik, V., et al., Soviet Atomic Energy, 1975. 39(1): p. 649-651

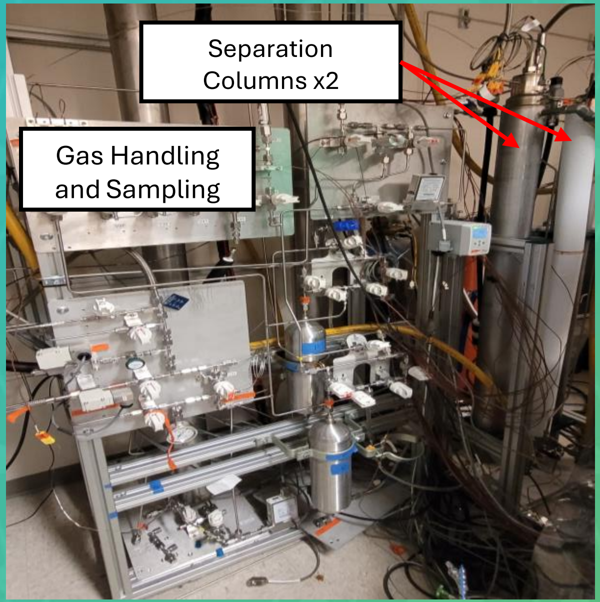
Manh Thuong Nguyen, PNNL report SA-190390

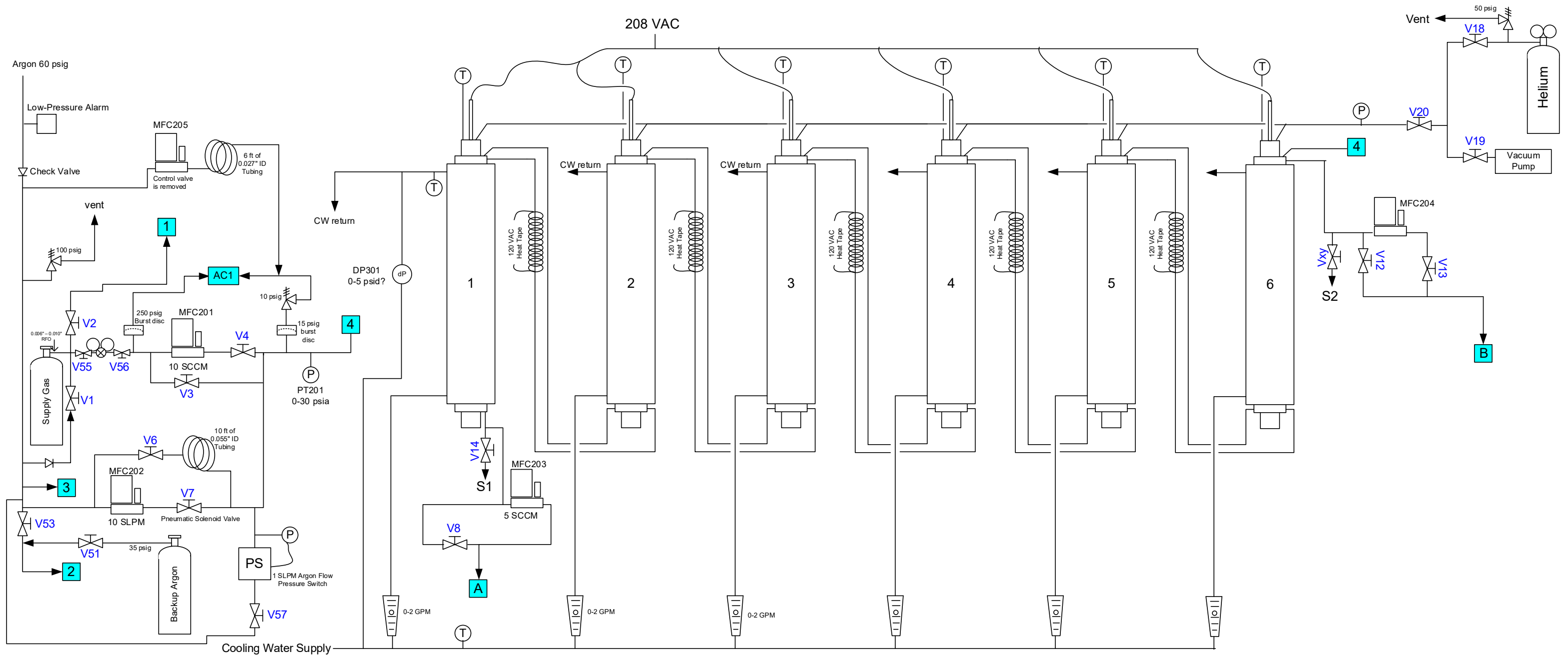
Chlorine Isotope Separation System for Chloride MSR

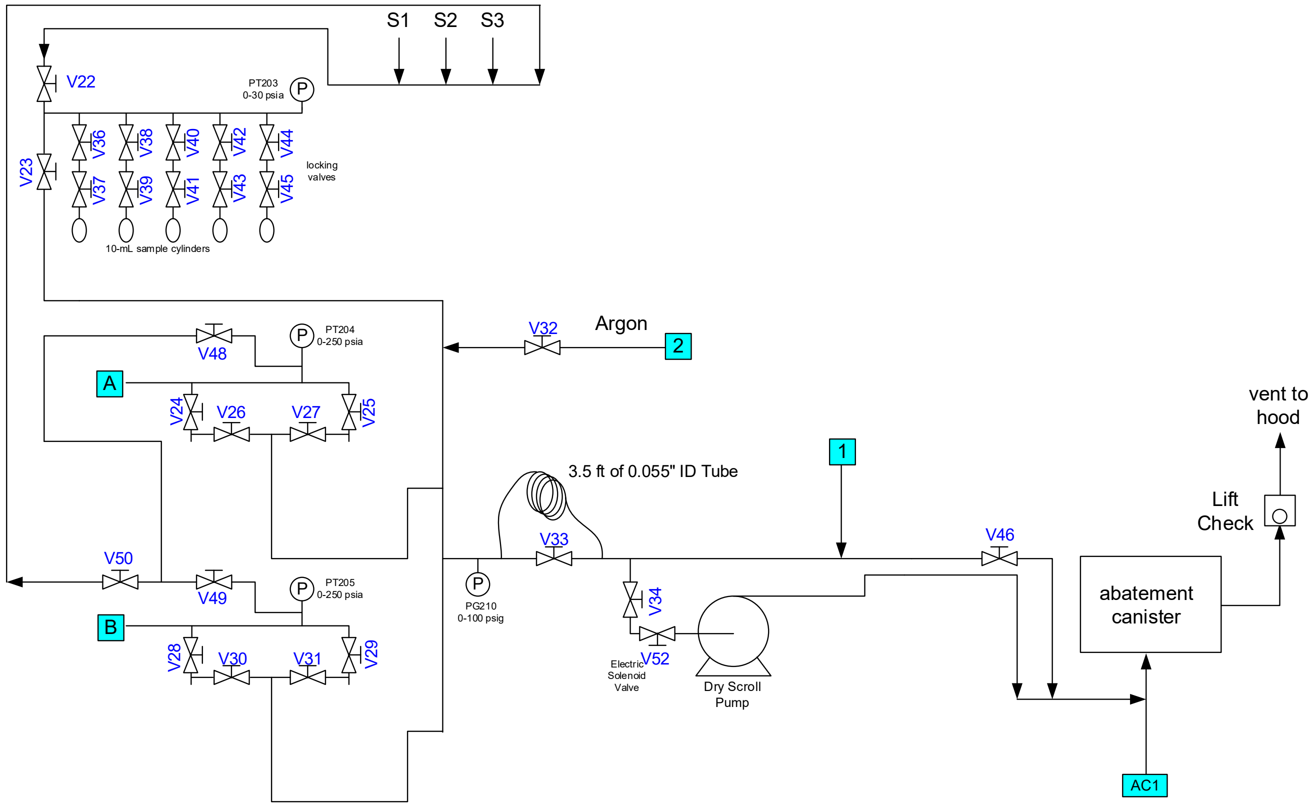
- Thermal diffusion isotope separation system for enrichment of ^{37}Cl.
- Multi-physics model exists to optimize and inform facility designs at multiple scales
- Precise Cl isotope ICP-MS method with HCl(L) – no chemistry needed and >1% accuracy on ^{37}Cl/^{35}Cl ratio

WHY is Cl enrichment needed?

- ^{35}Cl (76% of natural chlorine) has large neutron capture cross section
- ^{36}Cl activation product is long-lived (301,000 years) and energetic (709 keV) beta emitter , highly soluble in water which will complicate waste form and final disposal
- Neutron irradiation of ^{35}Cl (n, p) ^{35}S - ^{35}S is more corrosive than Fe, Cr, Ni







HCl Columns Flow diagram

Courtesy B. McNamara, PNNL

Integrated Off-Gas Management

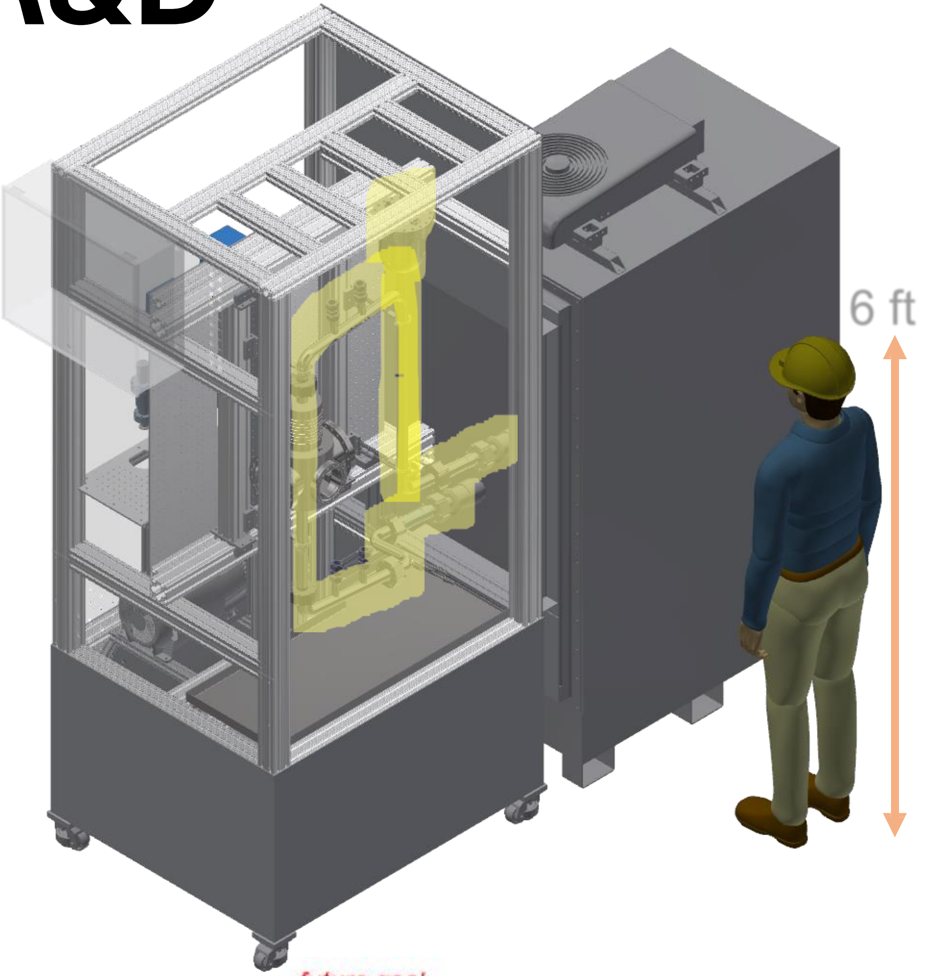
Relevant Regulatory Drivers for U.S. Off-gas Treatment

- **Volatile off-gas components have wide range of half-lives and disposal requirements:**
  - ^{3}H 12.31y
  - ^{14}C 5715 y
  - Xe Stable and very short half-life < 30 days
  - ^{85}Kr 10.76 y
  - ^{129}I 1.57 x 10^{7} y
- **Regulations**
  - Standards for normal operation (40 CFR 190) (Applies to Fuel Cycle Facilities)
    - *25 mrem/y whole body*
    - *75 mrem/y thyroid*
    - *50 000 Ci/GWy ^{85}Kr - (1.85 x 10^{15} Bq / GWy)*
    - *5 mCi/GWy ^{129}I - (1.85 x 10^{8} Bq / GWy)*
    - *0.5 mCi/GWy ^{239}Pu and other alpha-emitters*
  - 40 CFR 61 (Applies to facilities operated by DOE).
    - *Limits equivalent dose to the public to 10 mrem/y.*
- **NRC**
  - Standards for protection from ionizing radiation (10 CFR 20) (Applies to NRC Licensed Facilities)
    - Applies to facilities licensed by NRC.
    - Total equivalent dose not to exceed 0.1 rem/y.
    - Provides a table of air concentration limits for each radionuclide.

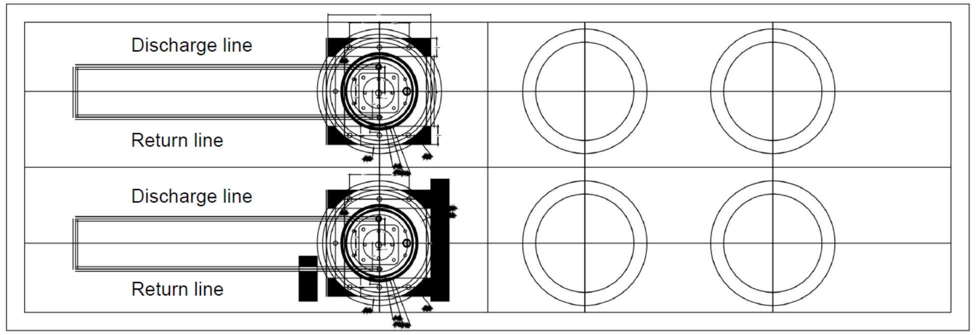
Jubin, Paviet, Bresee, Atalante 2016

Salt Loops supporting MSR R&D

MSTTE – Molten Salt Tritium Transport at INL
FY 2025 Deuterium- FLiNaK salt

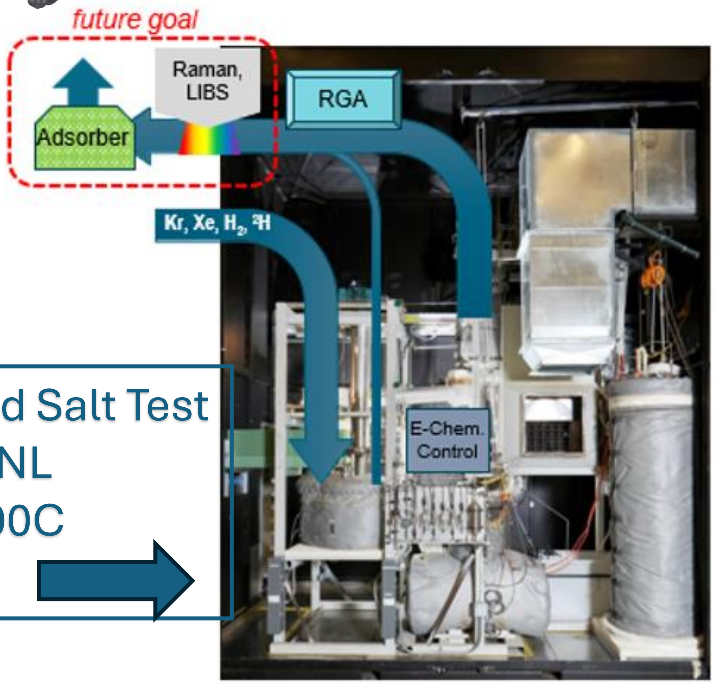


Actinide Salt Loop at ANL



FASTR -Facility to Alleviate Salt Technology Risks – ORNL
NaCl-KCl-MgCl2
725C
154L

LSTL - Liquid Salt Test Loop at ORNL
FLiNaK – 700C
80L

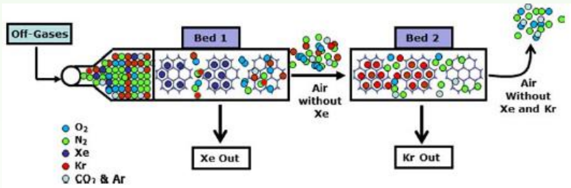


Technology Development and Demonstration
Multi-faceted approach to investigation of technologies for MSR off-gas systems

Component Testing

Large Scale Test Loop

Xe/Kr separation in MOF

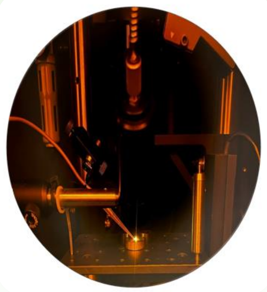


Radionuclide Identification/Speciation

Raman Spectroscopy

405 nm 532 nm 671nm

Laser Induced Breakdown Spectroscopy



Gas/Liquid Interface

Bubble Measurement

Source Term Modeling

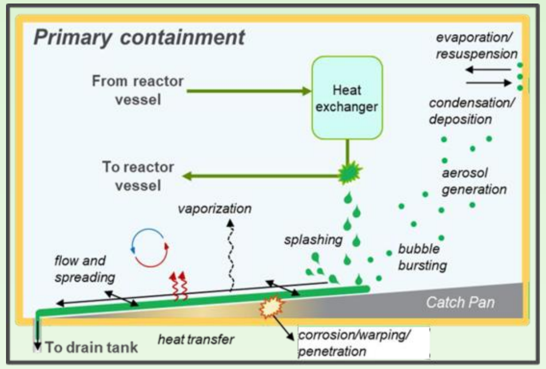
Bulk Gas	Gas Film	Liquid Film	Bulk Liquid
p_i pressure	p_i^*	c_i^*	c_i concentration

$$c_i^* = p_i H$$

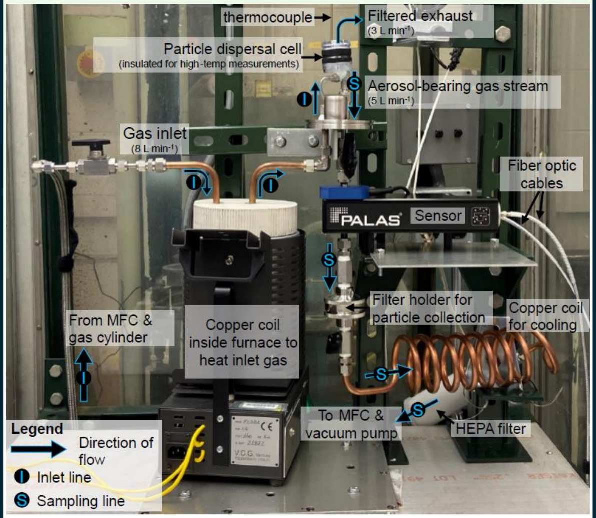
$$p_i^* = c_i / H$$

Spill Test

Engineering Scale Salt Accident Analysis Facility

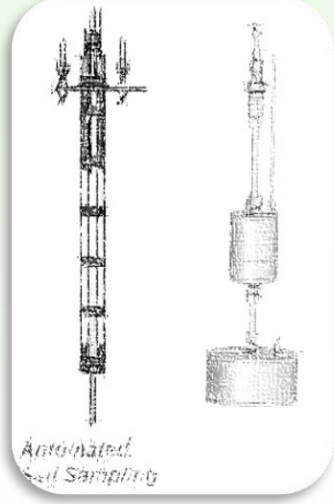


Salt Aerosol Concentration and Size Measurements



Sensors/Salt Chemistry

Salt Composition
Redox
Salt Level

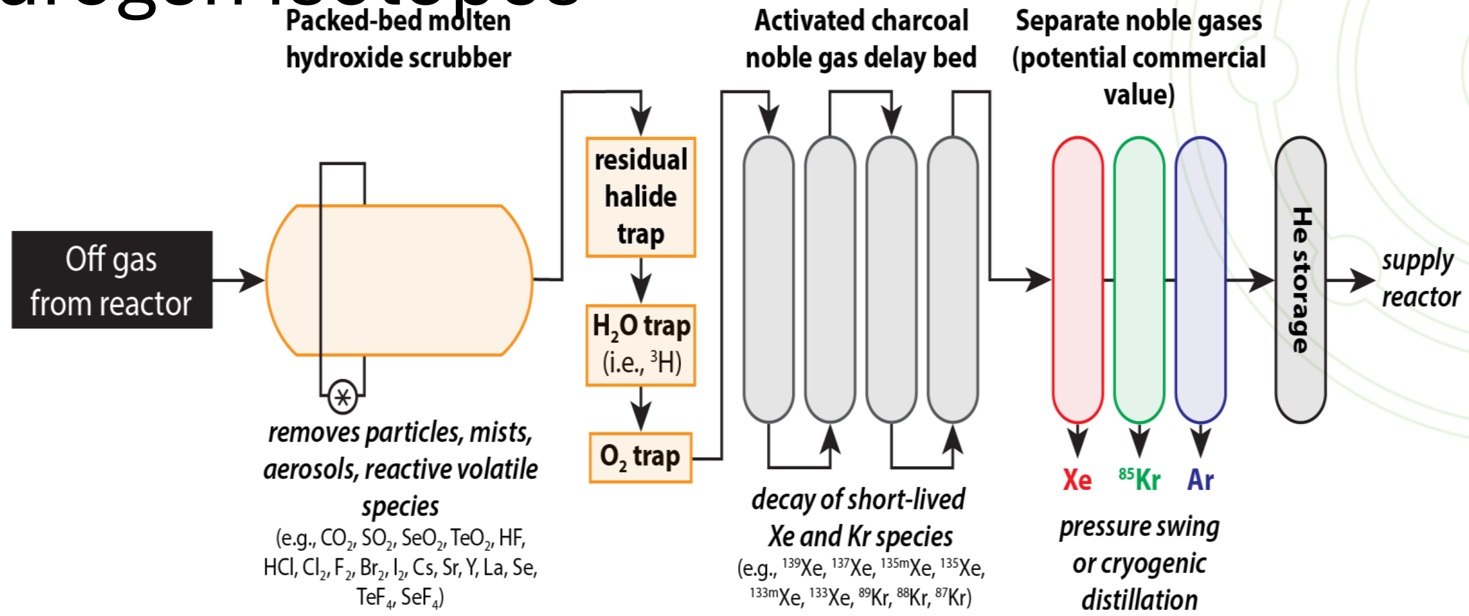


Application of Online Monitoring to MSR off-gas streams – The Molecular approach

- Development of OLM capabilities for chemical characterization of off-gas components
  - Demonstration on I_2, ICl, and hydrogen isotopes

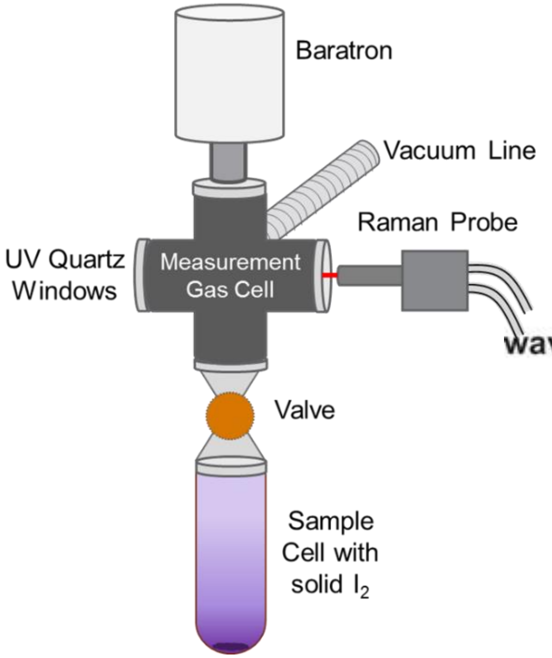
- Focus on Raman and FTIR
  - Ideal for deployment

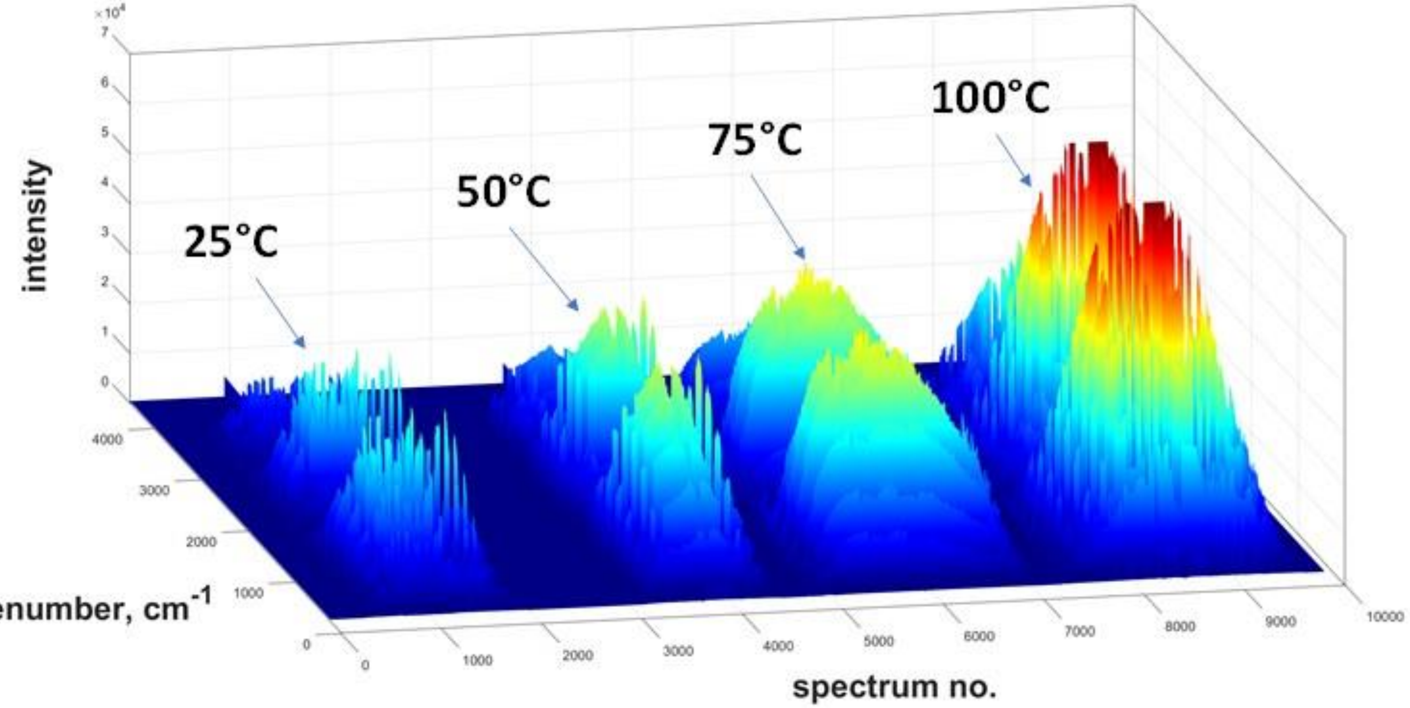
- PNNL collaboration with ORNL

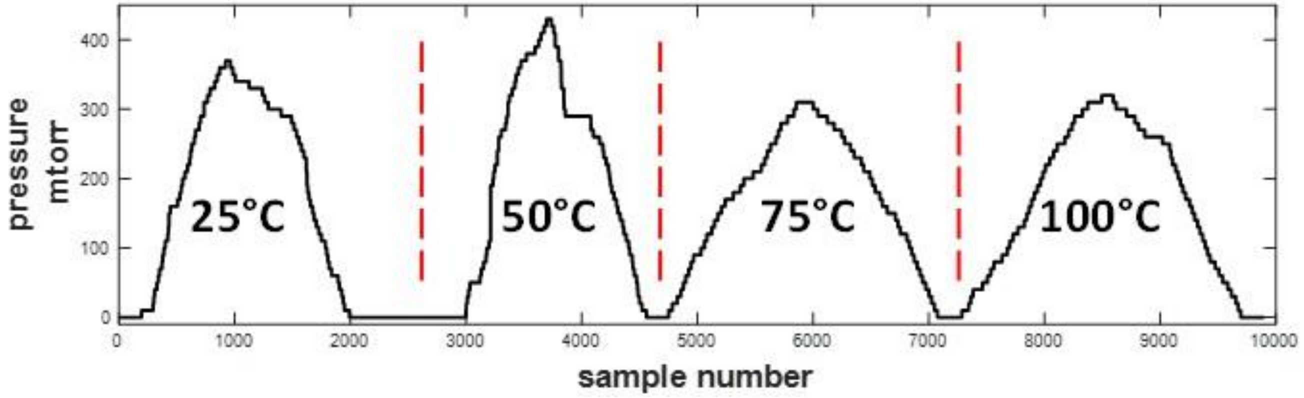


Mcfarlane, J.; Ezell, N.; Del Cul, G.; Holcomb, D. E.; Myhre, K.; Chapel, A.; Lines, A.; Bryan, S.; Felmy, H. M.; Riley, B. *Fission Product Volatility and Off-Gas Systems for Molten Salt Reactors*; Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States): 2019.

Off-gas measurement by Raman Spectroscopy: I_2







Felmy, Clifford, Medina, Cox, Wilson, Lines, Bryan
Environ Sci Technol *2021, 55, 6, 3898–3908.*

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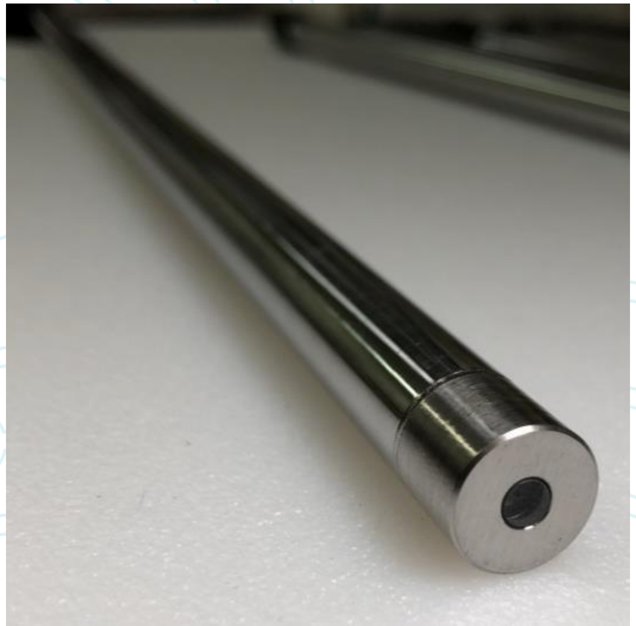
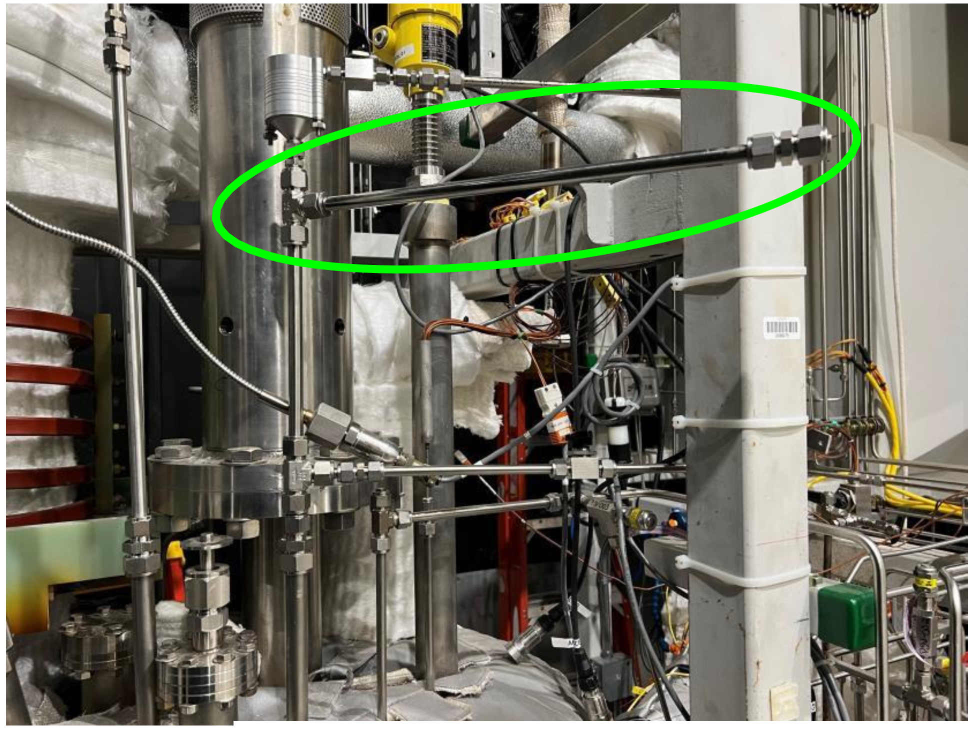
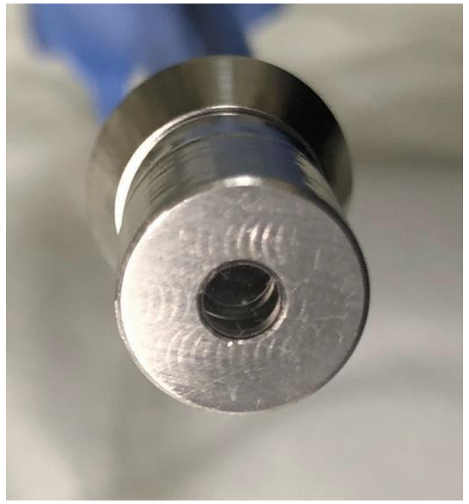
Testing Probe Materials in LSTL

Before incorporation into salt loop



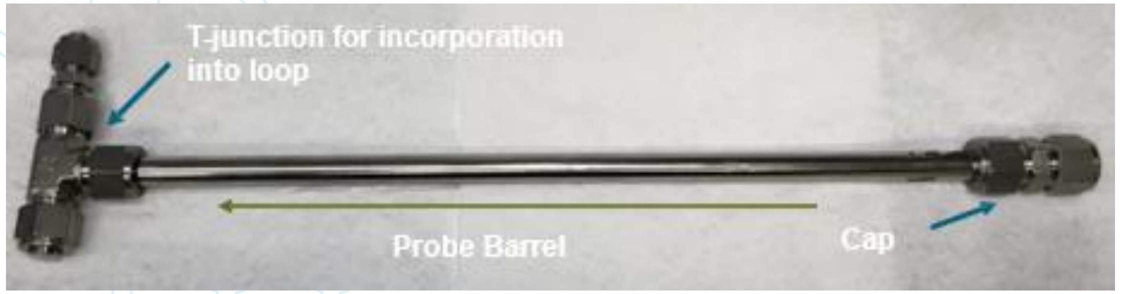
After incorporation into salt loop

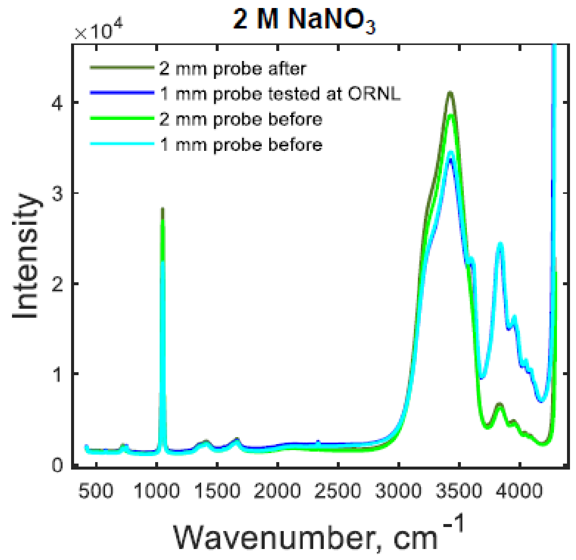


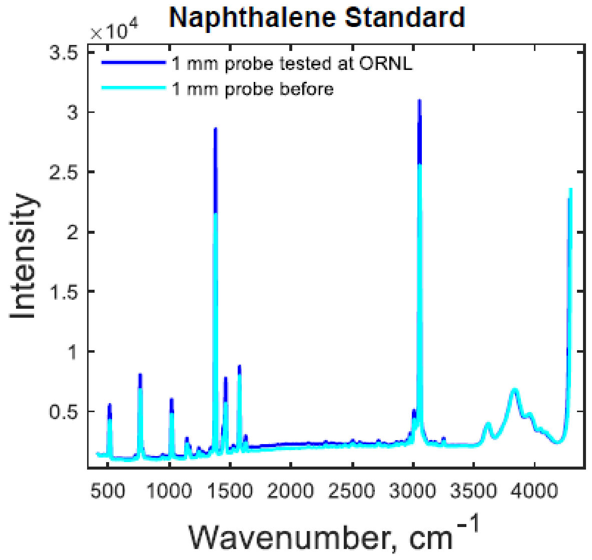


Salt loop testing

- Probe barrel swaged into loop
- **Visually, probe appears in excellent condition after exposure in LSTL**
- **Raman data suggests comparable signal after incorporation into loop**
- **Minimal materials degradation/impacts to performance**





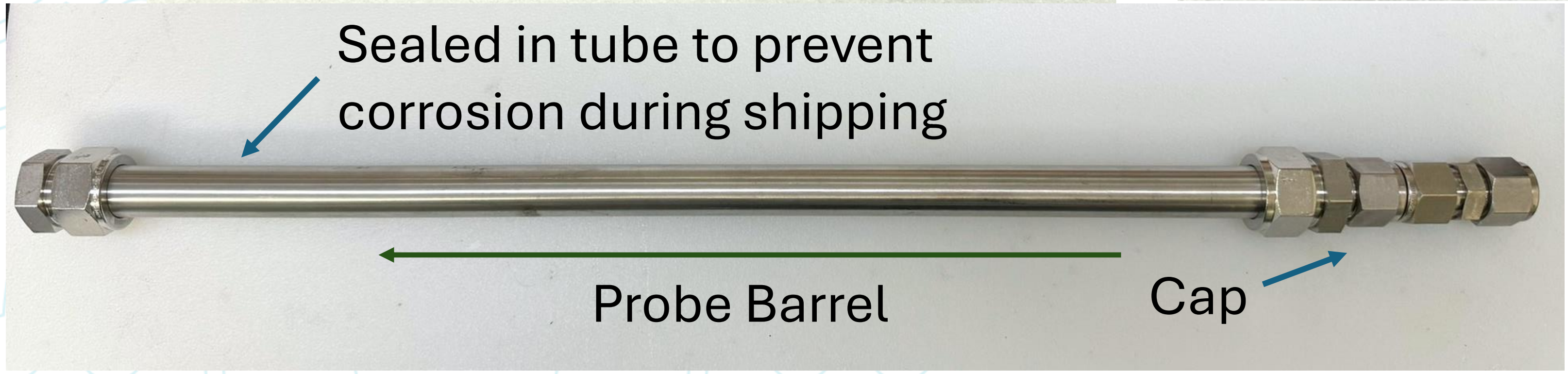


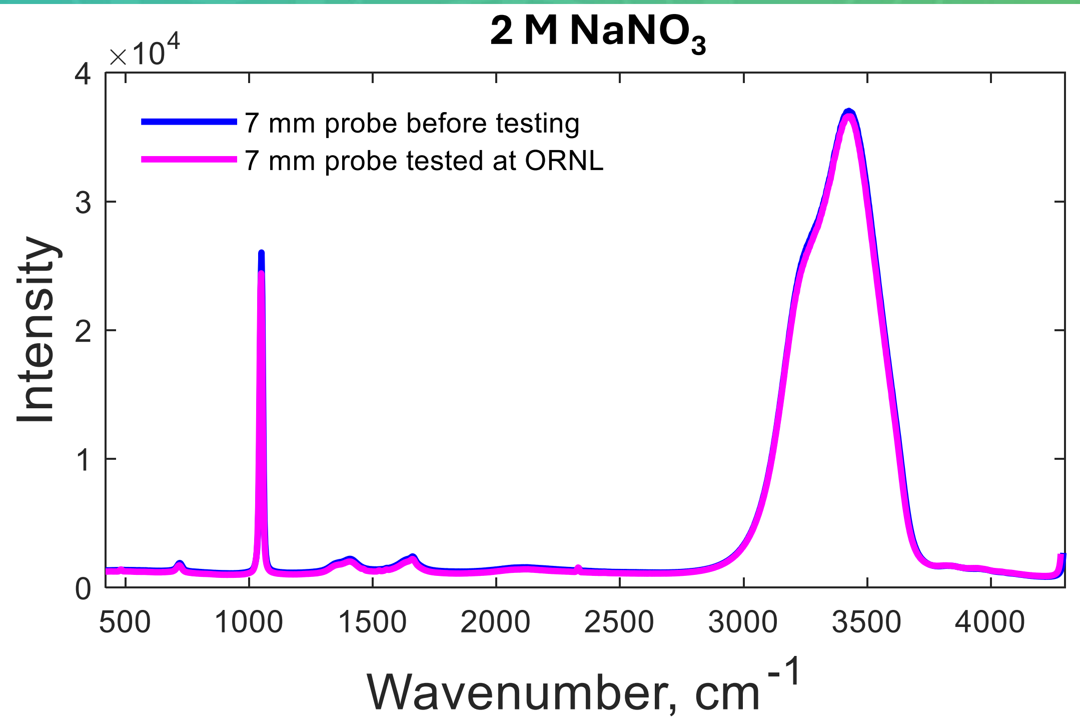
Courtesy Amanda Lines, PNNL

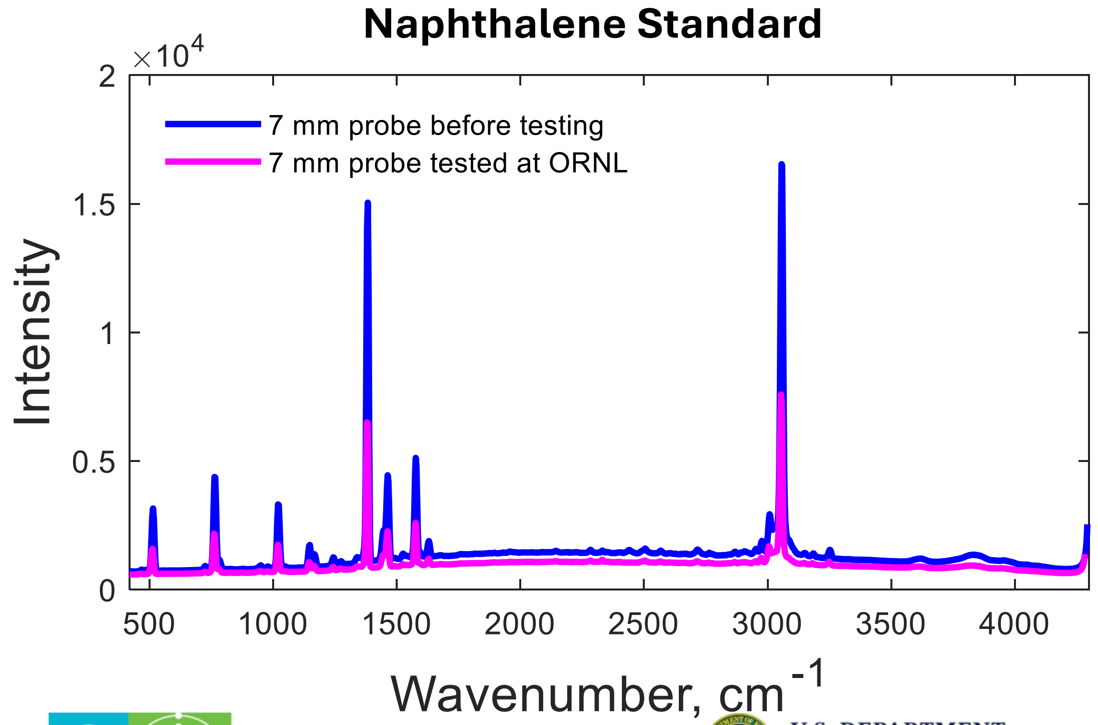
Testing Probe Materials in LSTL

2^{nd} Probe Testing

- **Salt loop testing**
  - Probe barrel Swaged into loop
  - Corrosion and heat damage visible on exterior of probe
  - No significant impact to probe performance

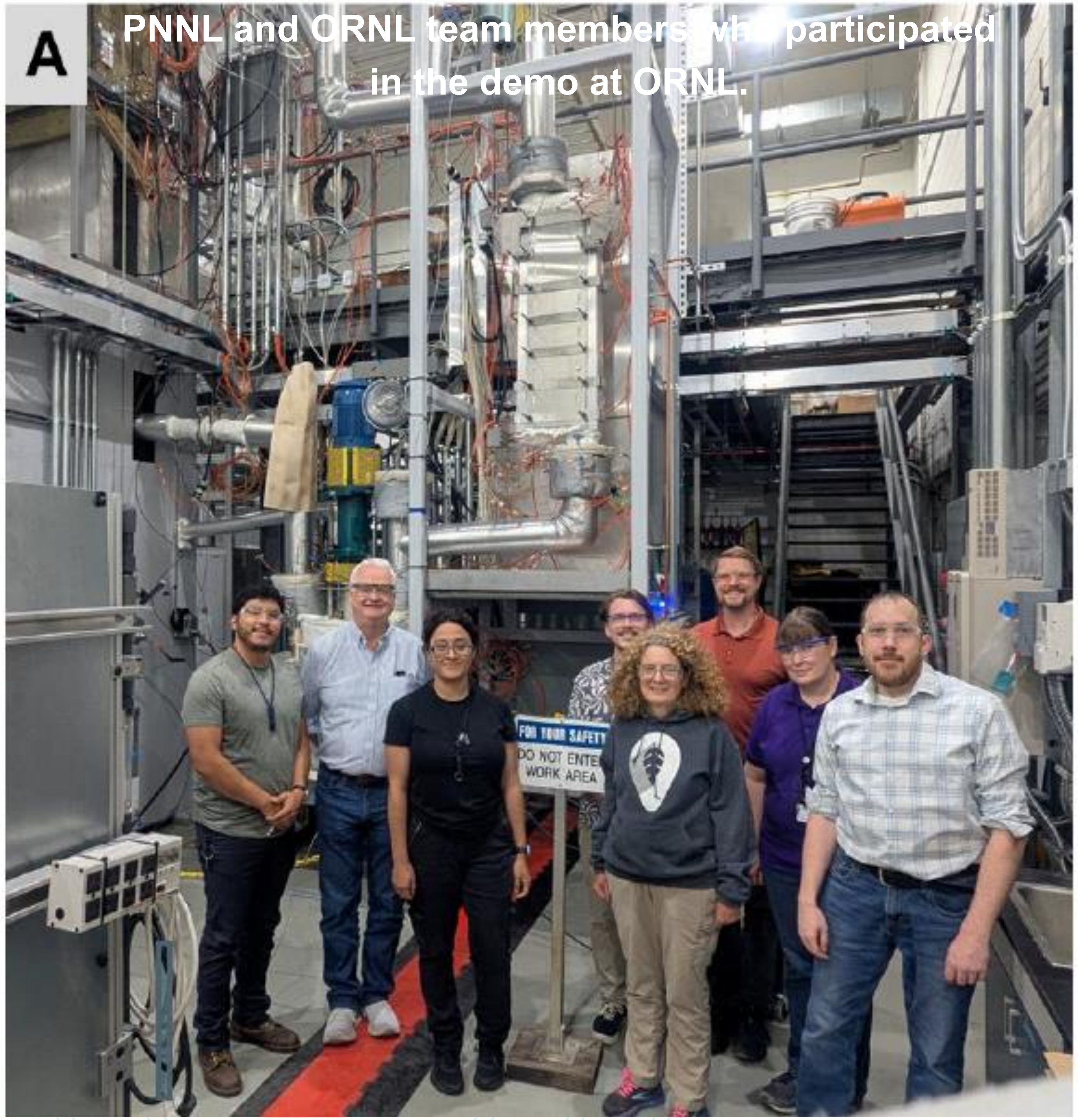






Courtesy H. Felmy, PNNL

Demonstration at FASTR Loop – ORNL – August 2025

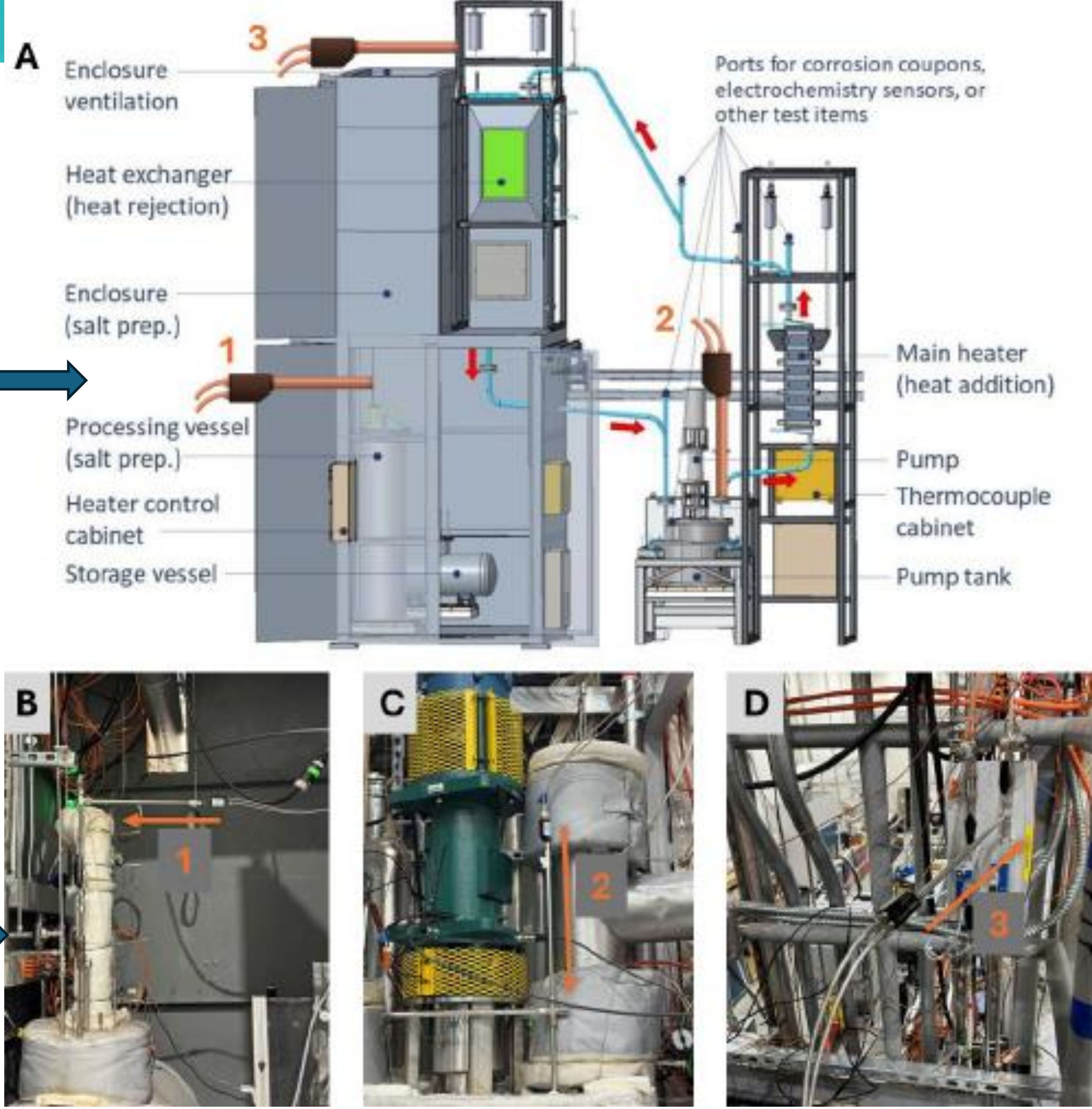


Felmy et al, PNNL-38277, SEP 2025

Molten Salt Reactor PROGRAM

of ENERGY
Office of Nuclear Energy

Schematic of FASTR with Raman probe locations indicated in orange (not to scale)



Photos of locations where Raman probes were incorporated into the FASTR loop are shown in

- **B) Salt Storage Tank**
- **C) pump tank,**
- **D) top of the manifold**

Felmy et al, PNNL-38277, SEP 2025

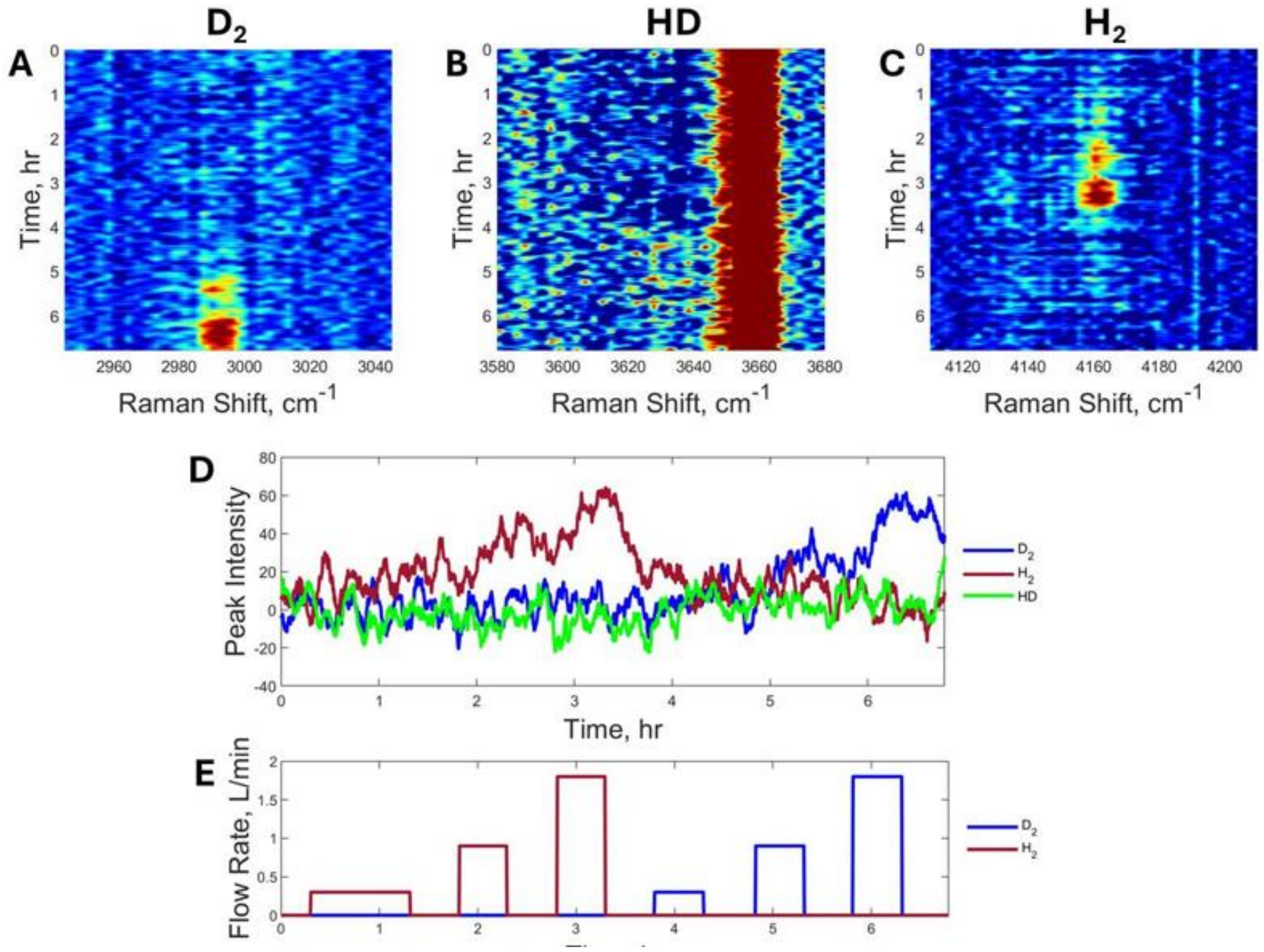
Detection of H_2 and D_2 with Raman probe

Salt storage tank sparged with 4% H2 in Ar followed by 4% D2 in Ar.

Raman probe inserted above the salt storage tank and on a gas outlet line

Throughout this experiment, H2 and D2 were alternated with Ar gas at increasing sparging gas flow rates

The Raman probe was able to detect H2 and then D2 with peak intensities increasing with gas flow rates as gas concentration in the headspace of the salt storage tank increased.



The Raman region where HD would be expected (3630 cm^{-1}) is plotted but no discernable signal was observed. It is important to note that with this probe, there was a peak at ~3650 cm^{-1} that was present in all spectra. This signal could be caused by probe materials or gas line materials but its presence in all spectra indicates that it is not a gas phase analyte peak.

Felmy et al, PNNL-38277, SEP 2025

Rapid Elemental Analysis using Laser Induced Breakdown Spectroscopy

LIBS is being developed as an analytical monitoring tool for molten salt reactor (MSR) systems.

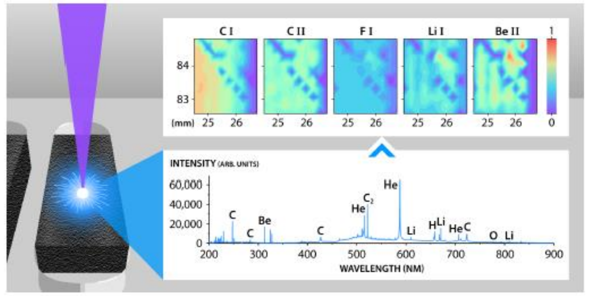
A mobile LIBS platform was configured on a small-scale salt vessel (~100 g of salt) and then transported 0.6 km to successfully monitor an engineering-scale pumped salt loop (~100 kg of salt).

Andrews et al., ORNL/TM-2025/4115, September 2025

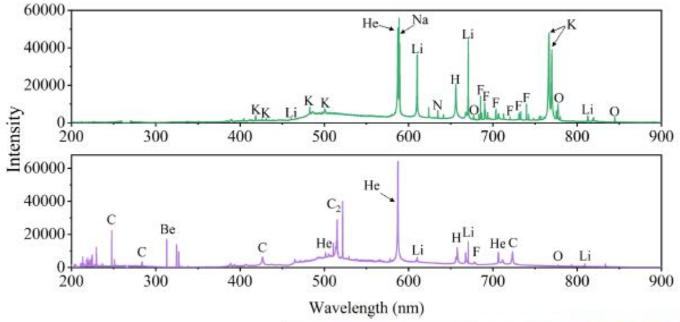
Why LIBS?

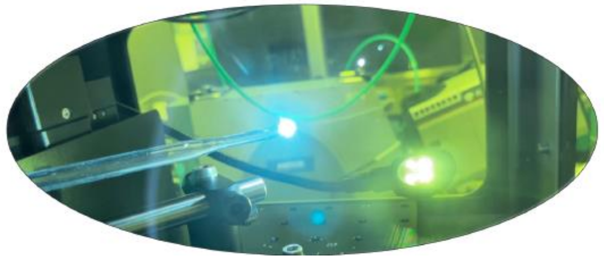
- Sensitivity across the periodic table
- Capable of remote measurements
- Rapid analysis
- Customizable to the application
- Can monitor solids, liquids, gases, and mixtures
- Elemental (occasionally isotopic) technique

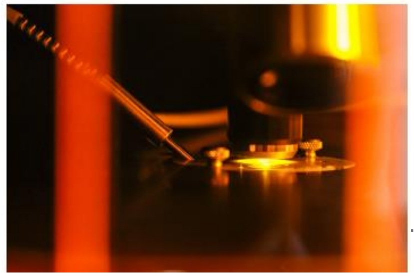
How can LIBS be used?



- Frozen salt analysis
  - As procured, purified, and post testing
- Investigating salt – material interactions
  - Graphite, structural materials
- Online monitoring
  - In-situ salt analysis, off-gas monitoring
- Real-time isotopic composition

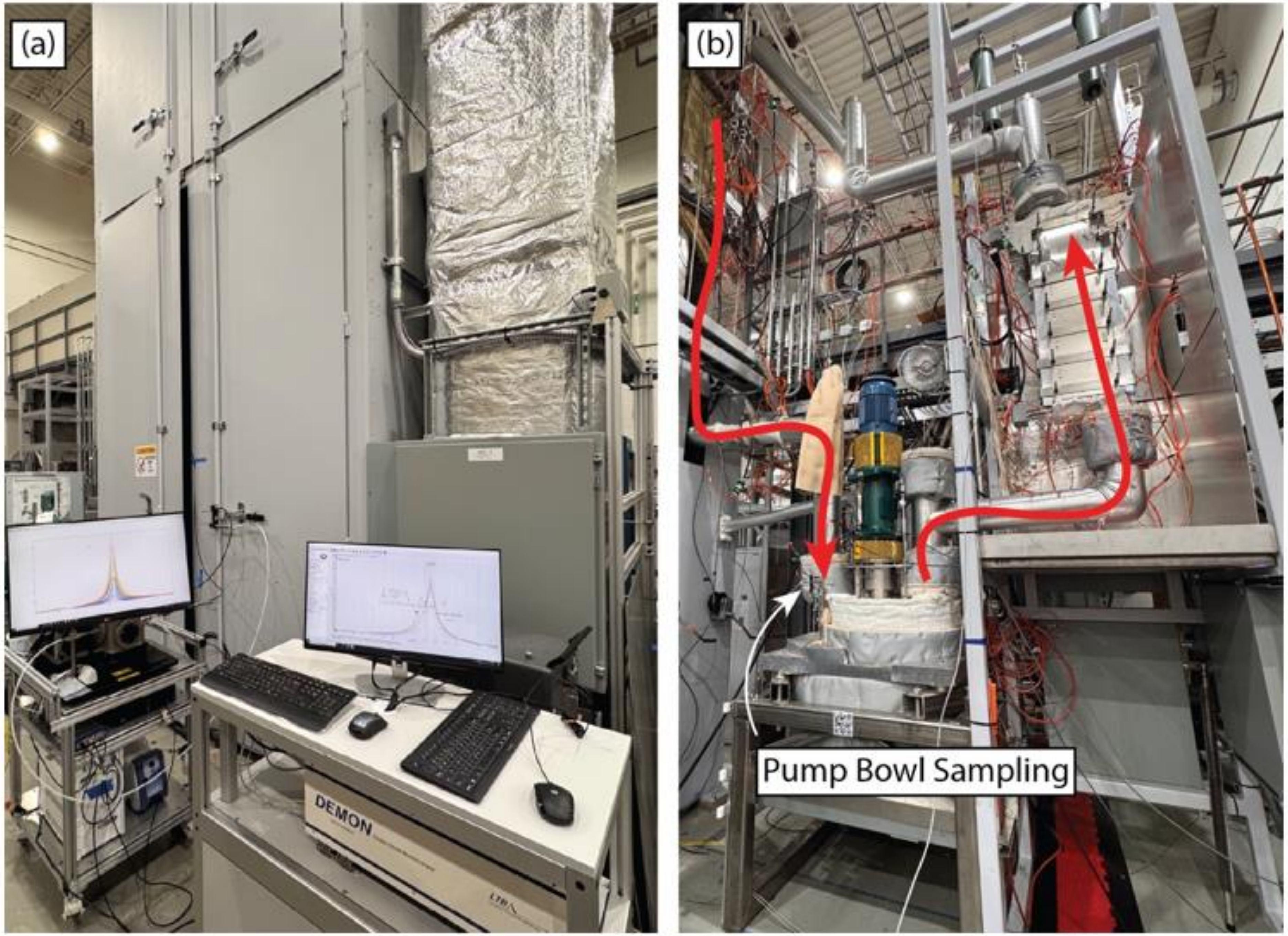






Transportation of the LIBS platform (left) and DEMON spectrometer (right) for tests on FASTR

Andrews et al., ORNL/TM-2025/4115, September 2025



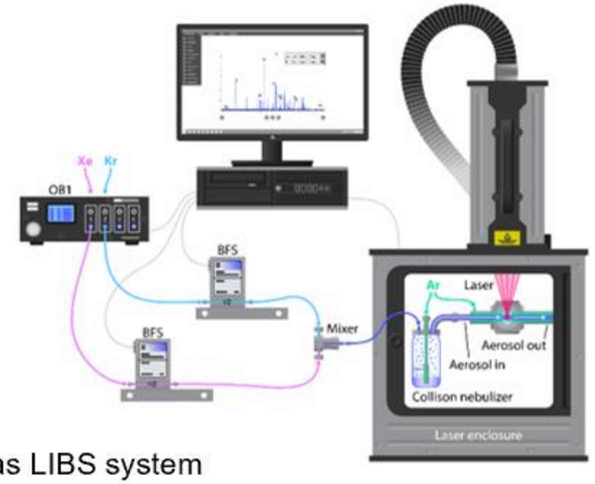
(a) FASTR storage tank with LIBS cart in the front left and DEMON spectrometer in the front right.

(b) FASTR loop with the salt direction shown in red and the pump bowl sampling location indicated.

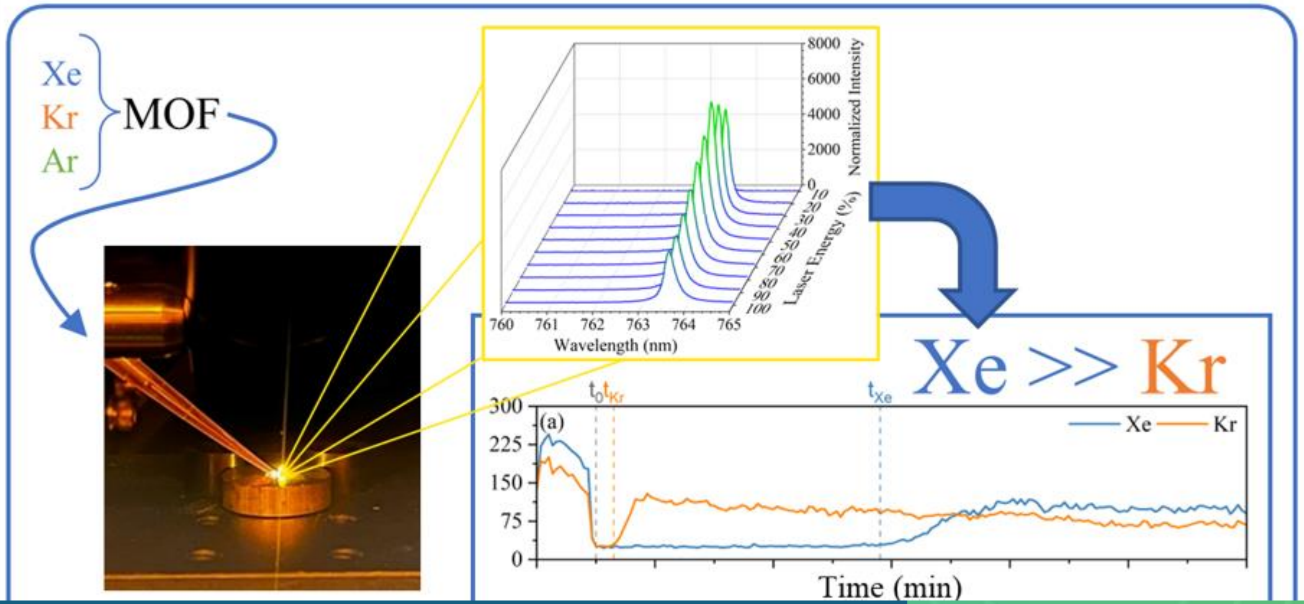
Andrews et al., ORNL/TM-2025/4115, September 2025

Monitoring and Evaluating Xe and Kr Capture

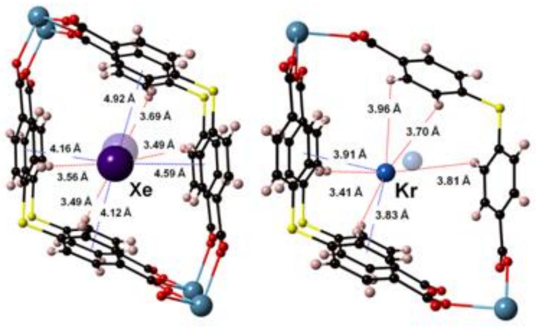
Laser induced breakdown spectroscopy of aerosols and noble gases



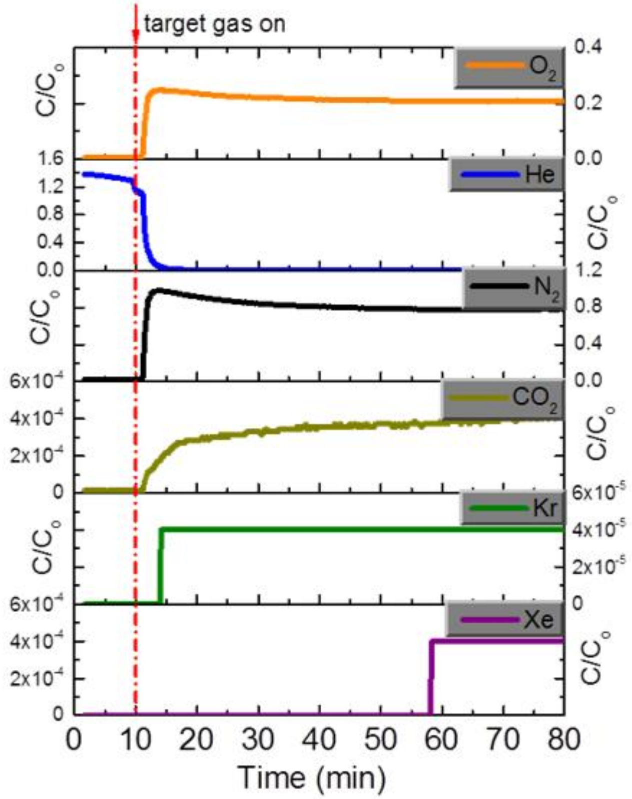
Aerosol/Gas LIBS system



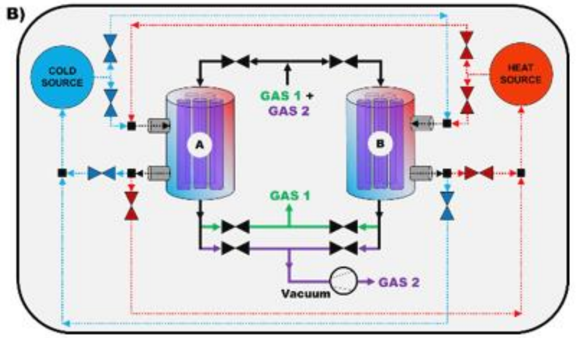
Metal organic framework for Xe and Kr capture



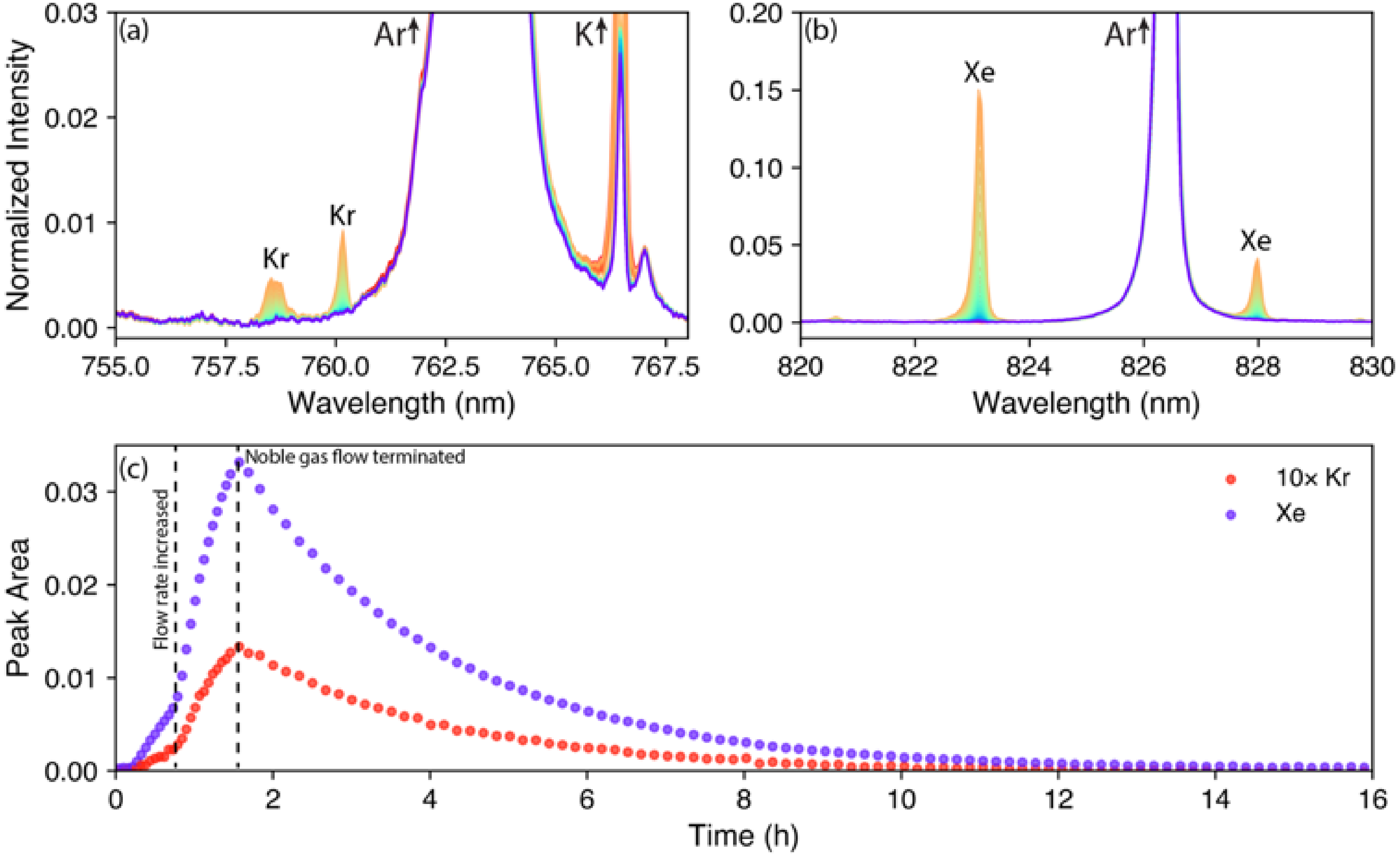
The PNNL patented MOF technology can be used to separate the noble gases from their surroundings.



Wet gas (RH 48%)



Off-gas adsorption and desorption system



Time evolution of (a) Kr and (b) Xe recorded during the noble gas test on the FASTR storage tank as measured on the broadband MCS.

The Xe and Kr peak area trends over time are shown in (c), where the Kr peak area is multiplied by 10 to account for the magnitude of concentration difference.

Andrews et al., ORNL/TM-2025/4115, September 2025

Hydrogen Isotopes Detection by LIBS

LIBS used in tandem with a residual gas analyzer (RGA) and a Raman spectroscopy system to detect hydrogen isotopes as they were sparged through and subsequently depleted from the loop storage tank

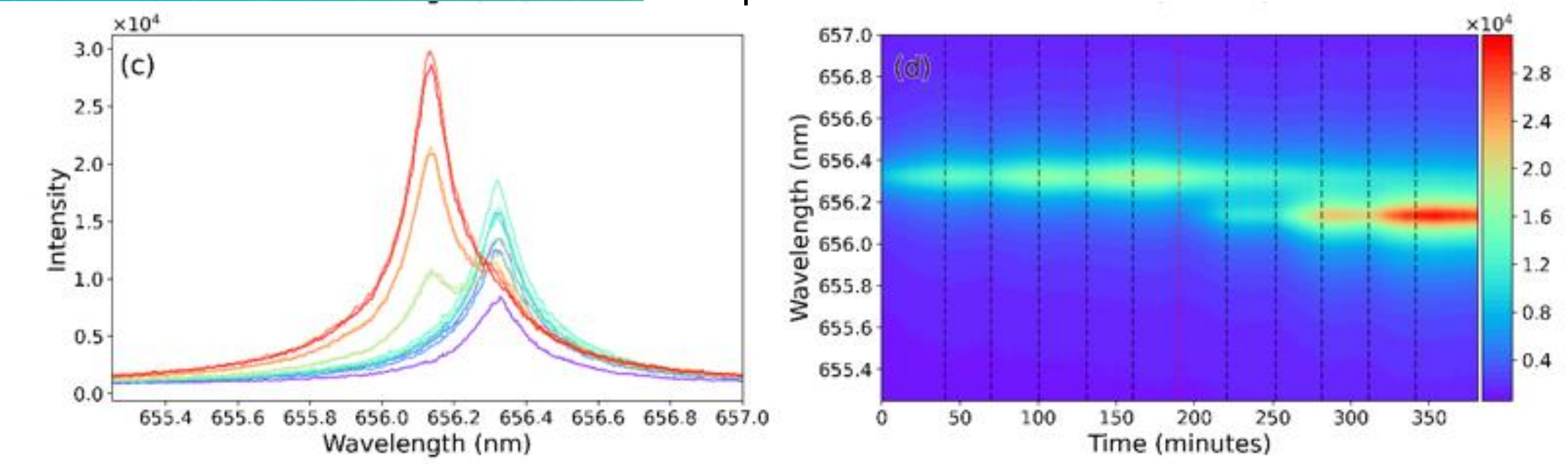
Andrews et al., ORNL/TM-2025/4115 SEP 2025

The corresponding surface map (d) shows the shift in peak position over time as the composition changed from H to D.

The vertical dashed lines (d) show when sparge gas and flow rate were changed and correspond to the times that the spectrum in (b) was recorded.

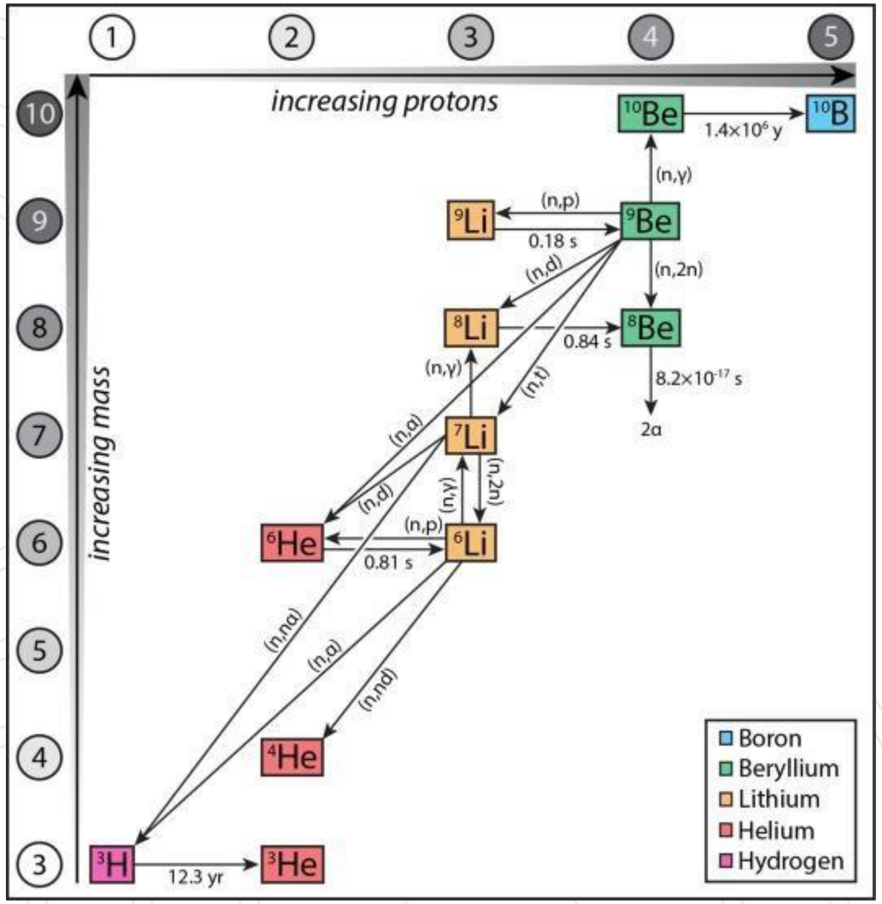
The red vertical dashed line denotes the first addition of D_2.

Spectra have not been normalized.



Tritium Generation in MSRs

Tritium generated by neutron reactions with Li, Be, and F.

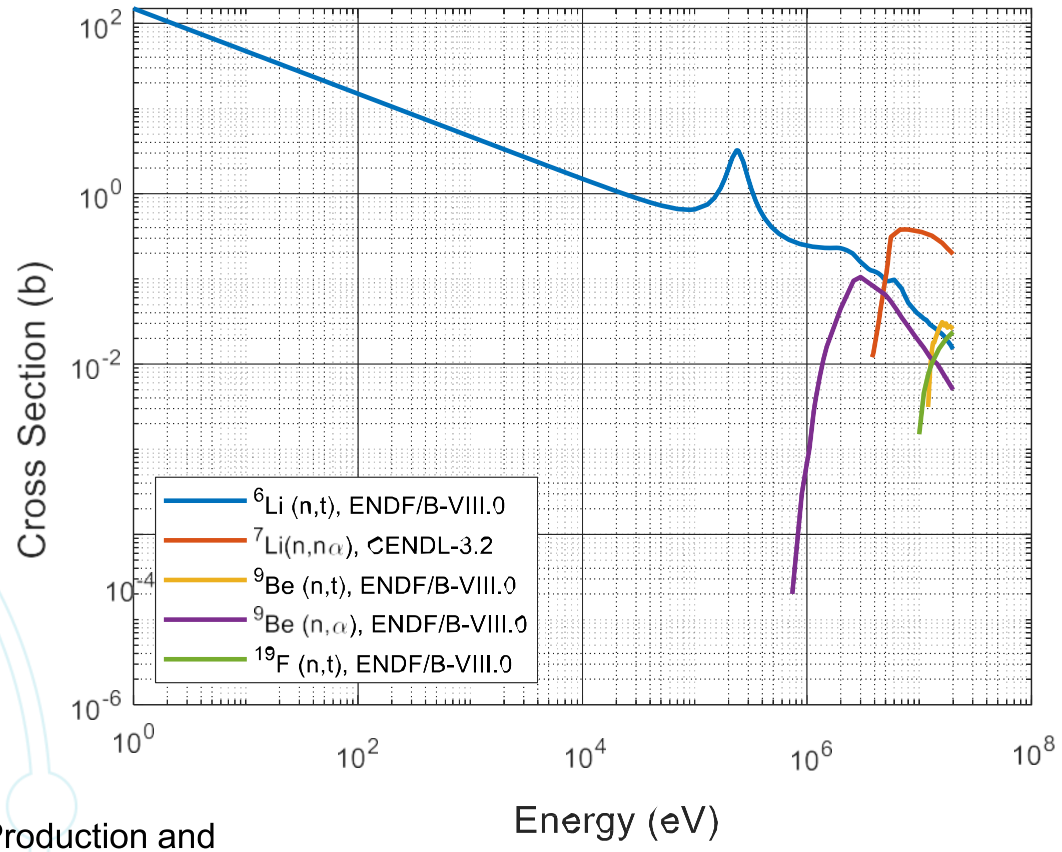


Sabharwall, P.; Schmutz, H.; Stoots, C.; Griffith, G. Tritium Production and Permeation in High- Temperature Reactor Systems;, 2013. https://doi.org/10.1115/HT2013-17036.

^{6}Li (7.5%) large thermal cross-section.

^{7}Li (92.5%) moderate cross-section in fast-spectrum.

^{9}Be and ^{19}F tritium in fast-spectrum.



Courtesy of Thomas Fuerst, INL

Tritium generation rates in fluoride salt reactors are similar to CANDU reactors.

CANDUs produce world's commercial supply of tritium.

Tritium is a valuable byproduct of MSRs.

Reactor Type	Tritium Formation Rate 1000 MWe (Ci/day) [1]
MSR	2400*
CANDU	2700
HTGR	50
PWR	2

*MSBR enriched in ^{7}Li (99.992%).

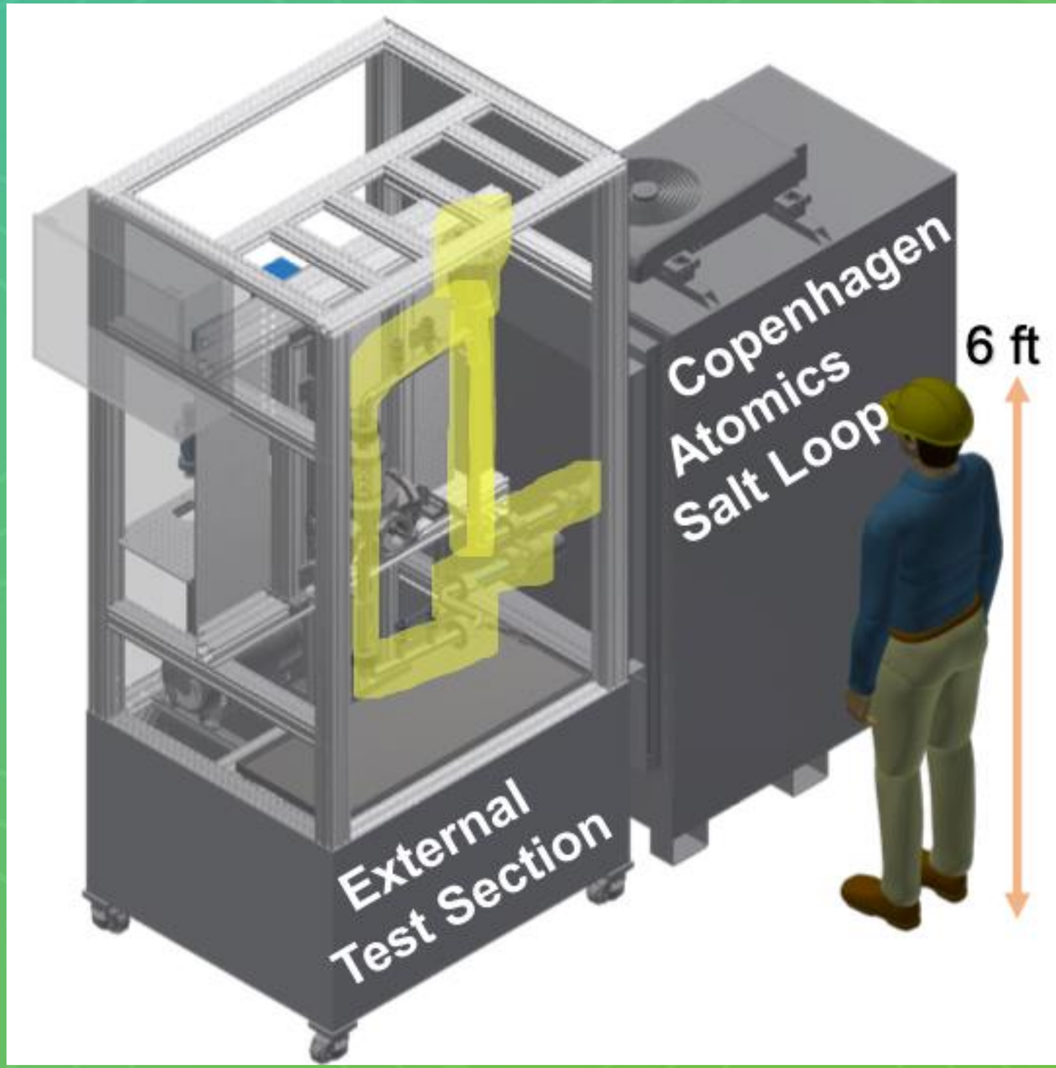
Andrews, Hunter B., et al. "Review of molten salt reactor off-gas management considerations." Nucl. Eng. Des. 385 (2021): 111529.

Molten Salt Reactor PROGRAM

U.S. DEPARTMENT of ENERGY
Office of Nuclear Energy

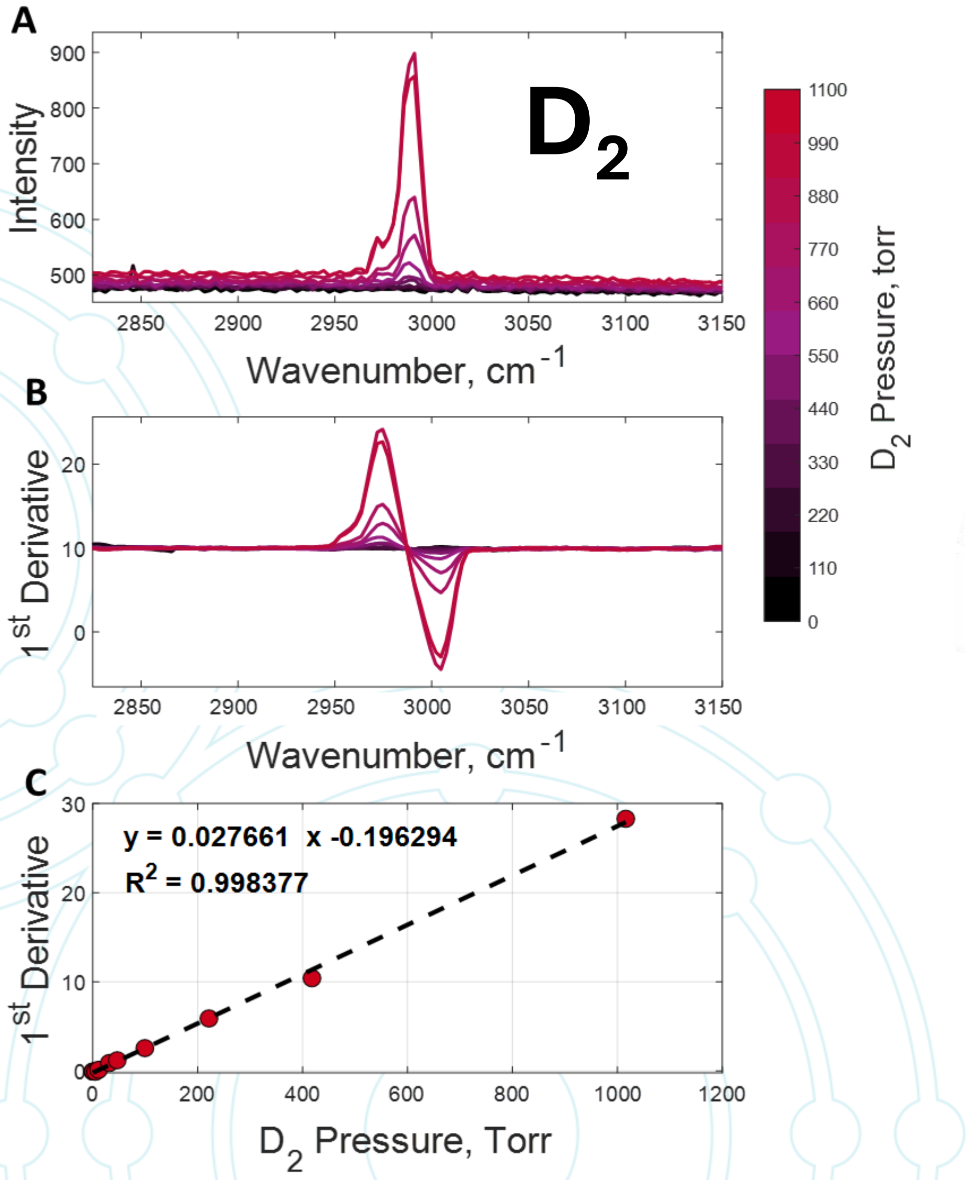
Molten Salt Tritium Transport Experiment

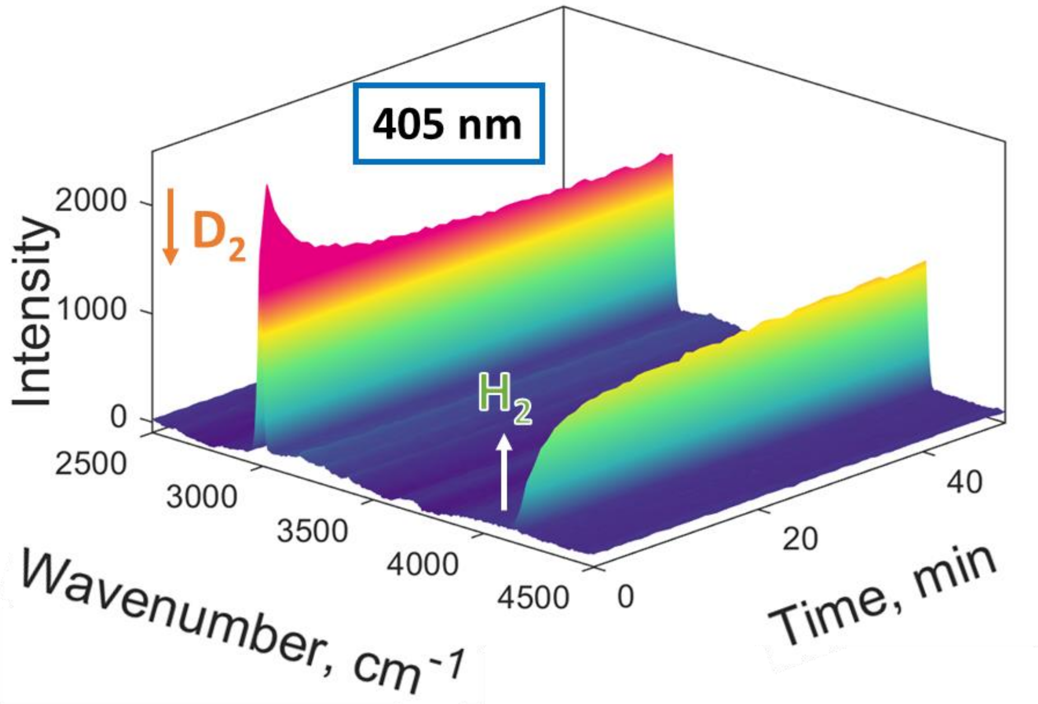
- MSTTE is a semi-integral tritium transport experiment for flowing fluoride salt systems
- Location: Safety and Tritium Applied Research facility
- **Objectives**
  - **(1) Safety code validation data**
  - **(2) Test stand for tritium control technology**
- Major equipment
  - **Copenhagen Atomics Salt Loop**: salt tank, pump, & flow meter
  - **External Test Section:** hydrogen injection, permeation, & plenum
- Phased approach
  - **Phase I:** FLiNaK and D_2 – 2025/2026
  - **Phase II:** FLiBe and D_2 – 2026/2027
  - **Phase III:** FLiBe and T_2 – 2027/2028

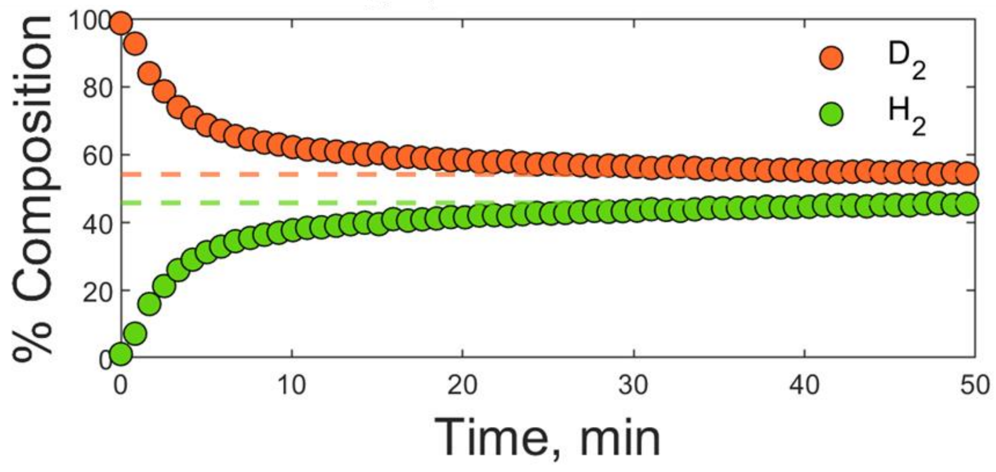


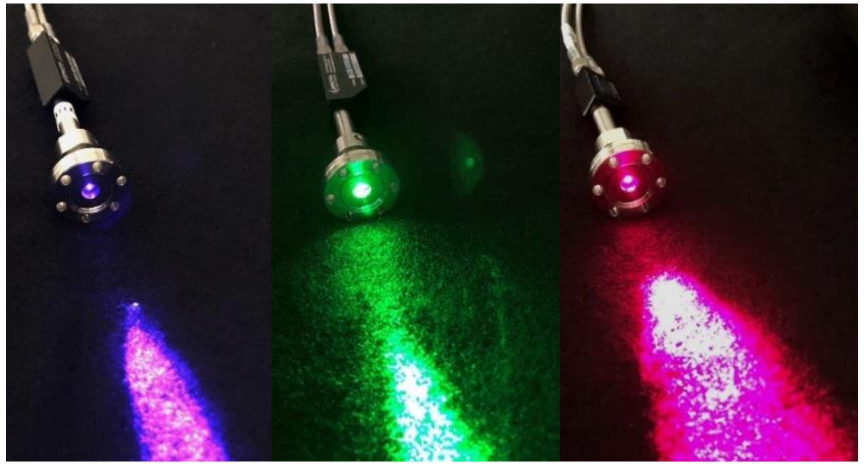
Courtesy Thomas Fuerst, INL

Integration of probes into systems for H isotope analysis









405 nm 532 nm 671 nm

Courtesy Amanda Lines, PNNL

Safety of MSR

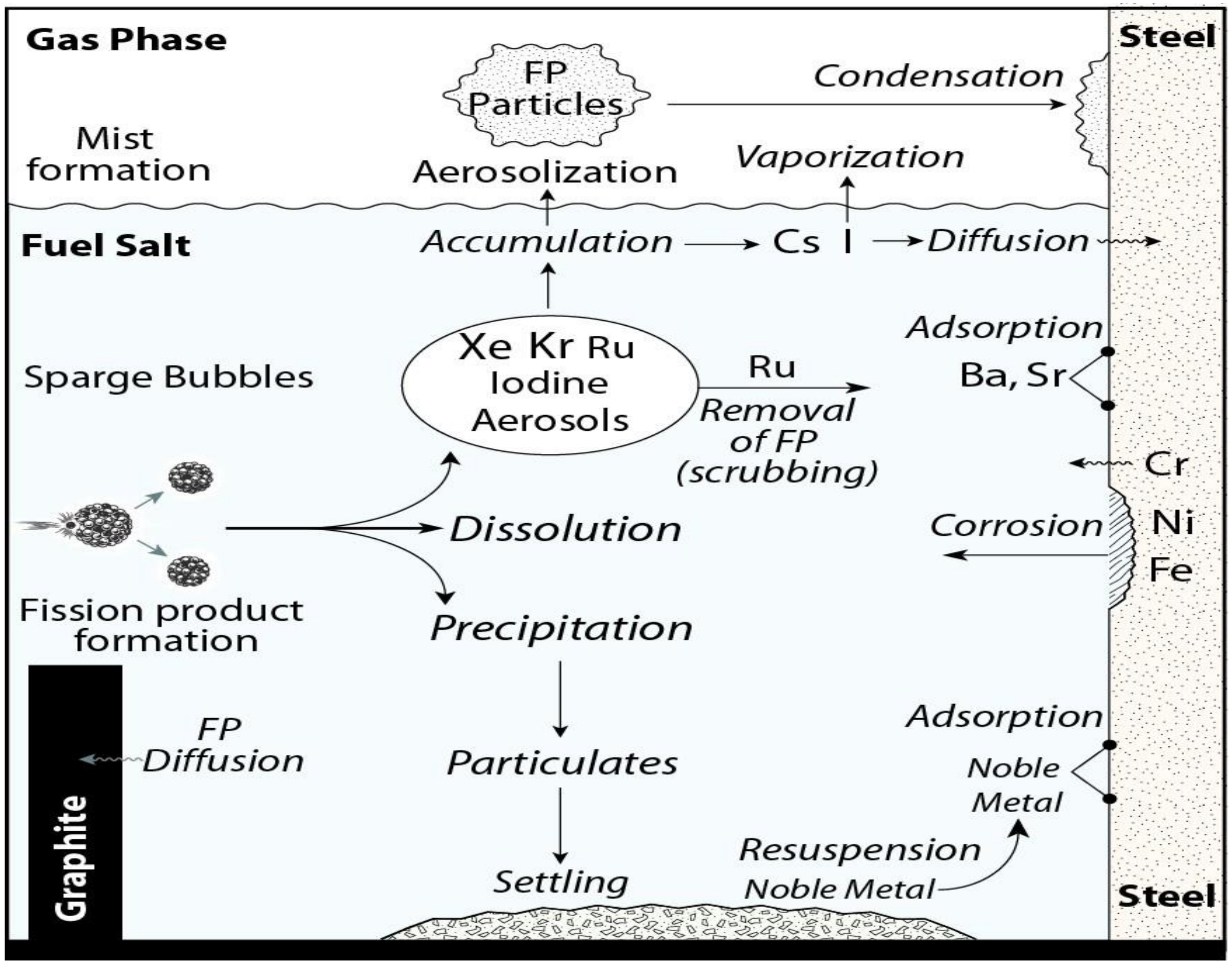
Fundamental MSR Safety Functions

The primary safety criteria that are targeted by fuel qualification activities typically include:

- Maintaining margins for radiological releases
- Ensuring adequate heat removal for reactor shutdowns
- Maintenance of proper reactivity control

All these criteria involve a broad array of reaction mechanisms and phenomena.

Comprehensive understanding of the chemistry is needed to make meaningful predictions.



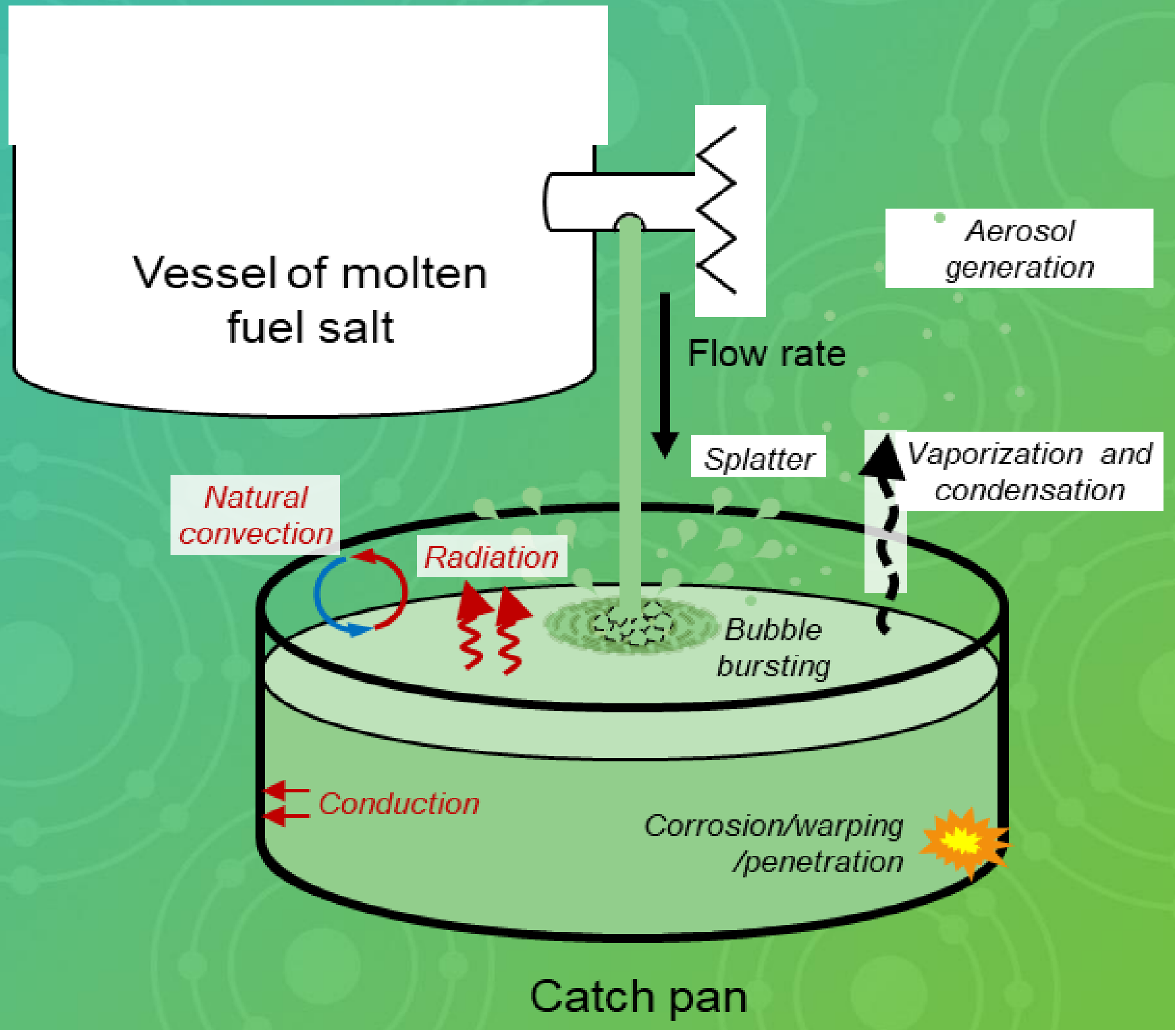
Reactions pathways for radiological releases[1]

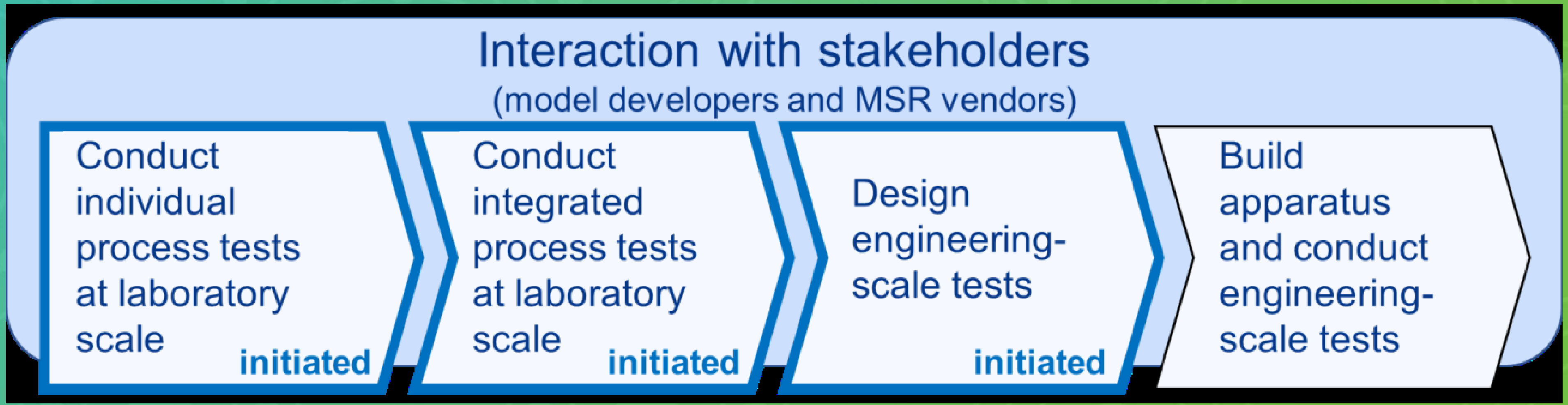
Salt properties relevant to radiological releases[2]

[1]Jerden, J, et al. ANL/CFCT-19/4 (2019)
[2]Holcomb, et al. NUREG/CR-7299 (2022)

Advanced Reactor Licensing and Accident Analysis

- U.S. Nuclear Regulatory Commission (NRC) Regulatory Guide 1.233 provides a modernized licensing framework for advanced reactors
  - Identify and evaluate the consequences of postulated accidents
  - Use validated models to predict accident progression and the mechanistic source term for expected radiological release
- All MSR developers will likely evaluate an unintended release of fuel salt
- Experimental data on the key processes that influence the safety-affecting outcomes of fuel salt release accidents are limited





Courtesy S. Thomas, ANL

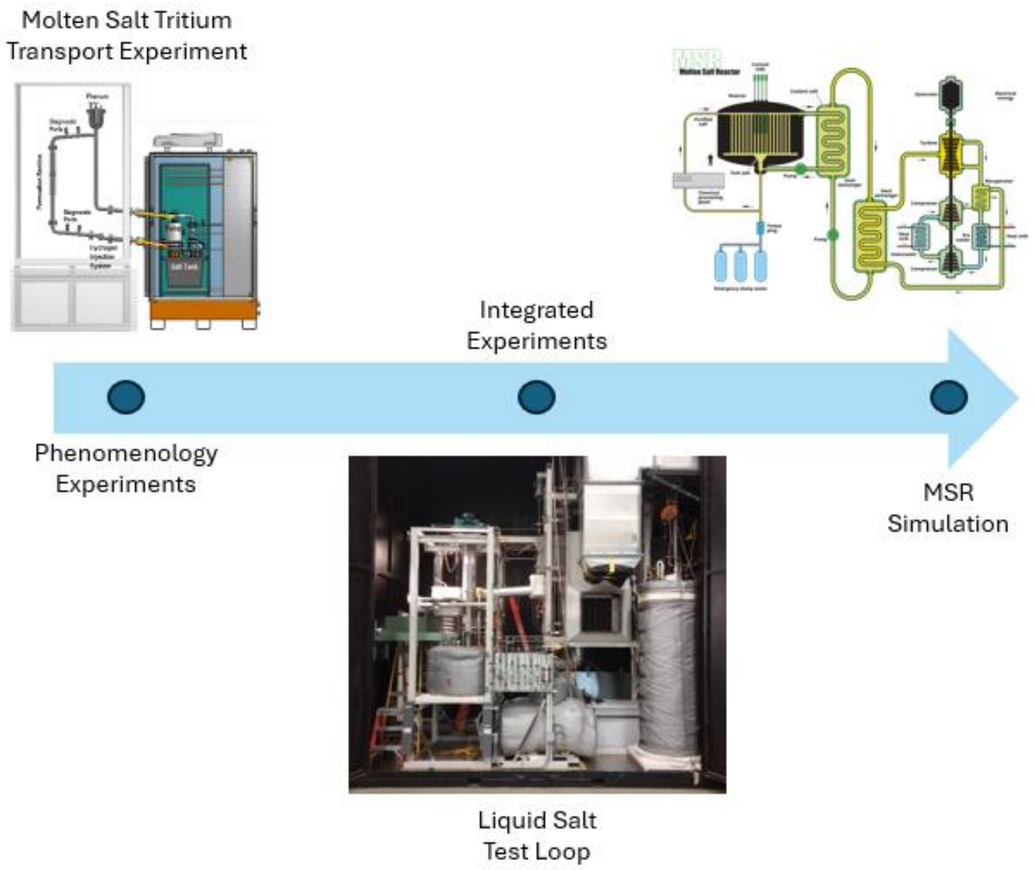
Modeling and Simulation

Modeling and Simulation

MELCOR: nuclear accident simulation code at SNL

Significant strides within the last few years to increase model accuracy for molten salt reactors (MSRs). It is the primary code developed for the

- Assessment and evaluation of safety for a broad range of reactor concepts, notably including MSRs.
- MSR model development has been focusing on improving chemistry and fission product transport mechanisms
- Working with MSR campaign to fill knowledge gaps



NEAMS Tools at INL

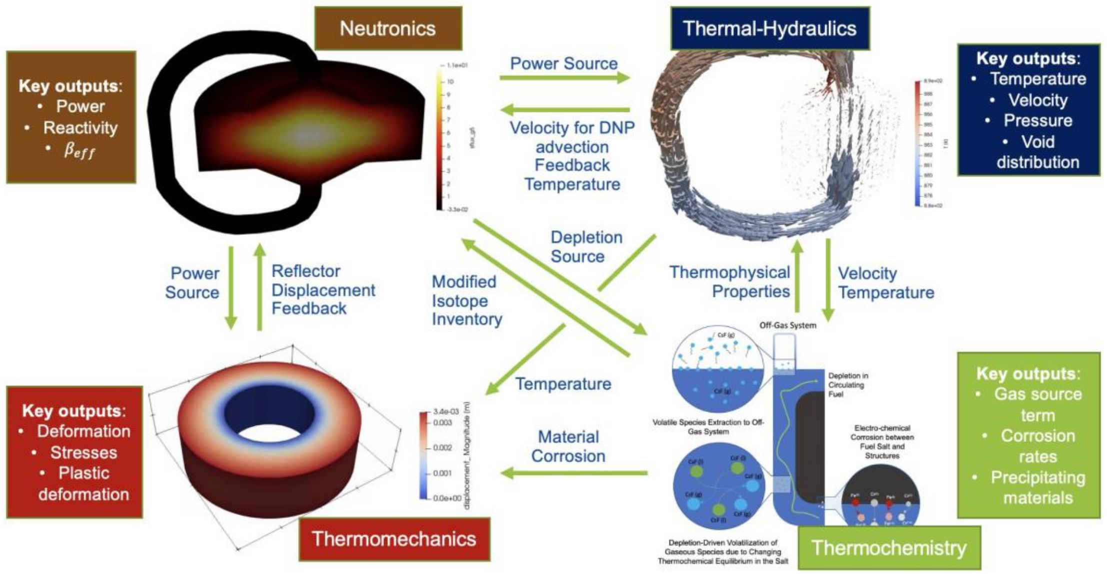
Coupling between:

- Neutronics (multi-fidelity + spatial depletion),
- Thermal-Hydraulics (coarse-mesh CFD),
- Thermomechanics (thermo-elasto-plasticity with irradiation and thermal swelling and creep),
- Thermochemistry (speciation, mass accountancy, and corrosion)

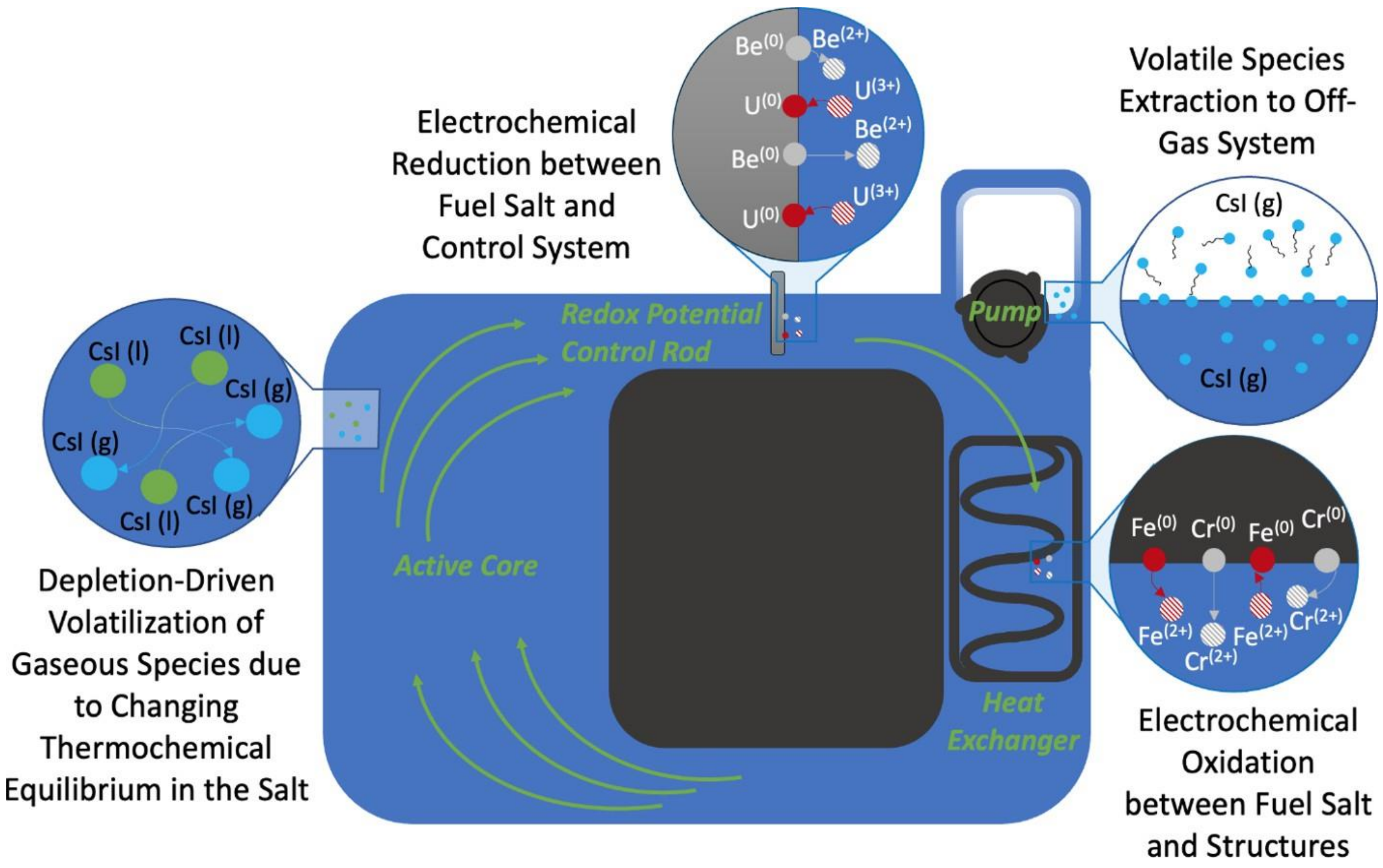
- Species tracking in MSRs
  - Tracking the depletion chain of ${}^{91}Br$

$$ {}^{91}Br \xrightarrow{t_{1/2} = 0.543s} {}^{91}Kr \xrightarrow{t_{1/2} = 8.57s} {}^{91}Rb \xrightarrow{t_{1/2} = 58.2s} {}^{91}Sr \xrightarrow{t_{1/2} = 9.65h} {}^{91}Y \xrightarrow{t_{1/2} = 58.51d} {}^{91}Zr $$

Fission: $y = 6.3954E-4$; Fission: $y = 1.178E-2$; Fission: $y = 2.93761E-2$; Fission: $y = 9.669884E-3$; Fission: $y = 1.88654e-4$; Fission: $y = 1.68E-6$



Why Species Tracking in MSRs?



Chemical species transport examples in MSR system.

- **Containment**: Where do the radionuclides go? What is the reactor source term?
- **Heat removal**: Where do radionuclides plate out? How do we cool the reactor?
- **Reactivity**: Where do the neutron precursors go? What is the reactor beta-eff?
- **Corrosion**: How do fission products interact with the wall? How long will a barrier last?
- **Safeguards**: Where do the fissile nuclides go? How do we monitor where they are?

Courtesy Mauricio Tano-Retamales, INL

Molten Salt Reactor PROGRAM

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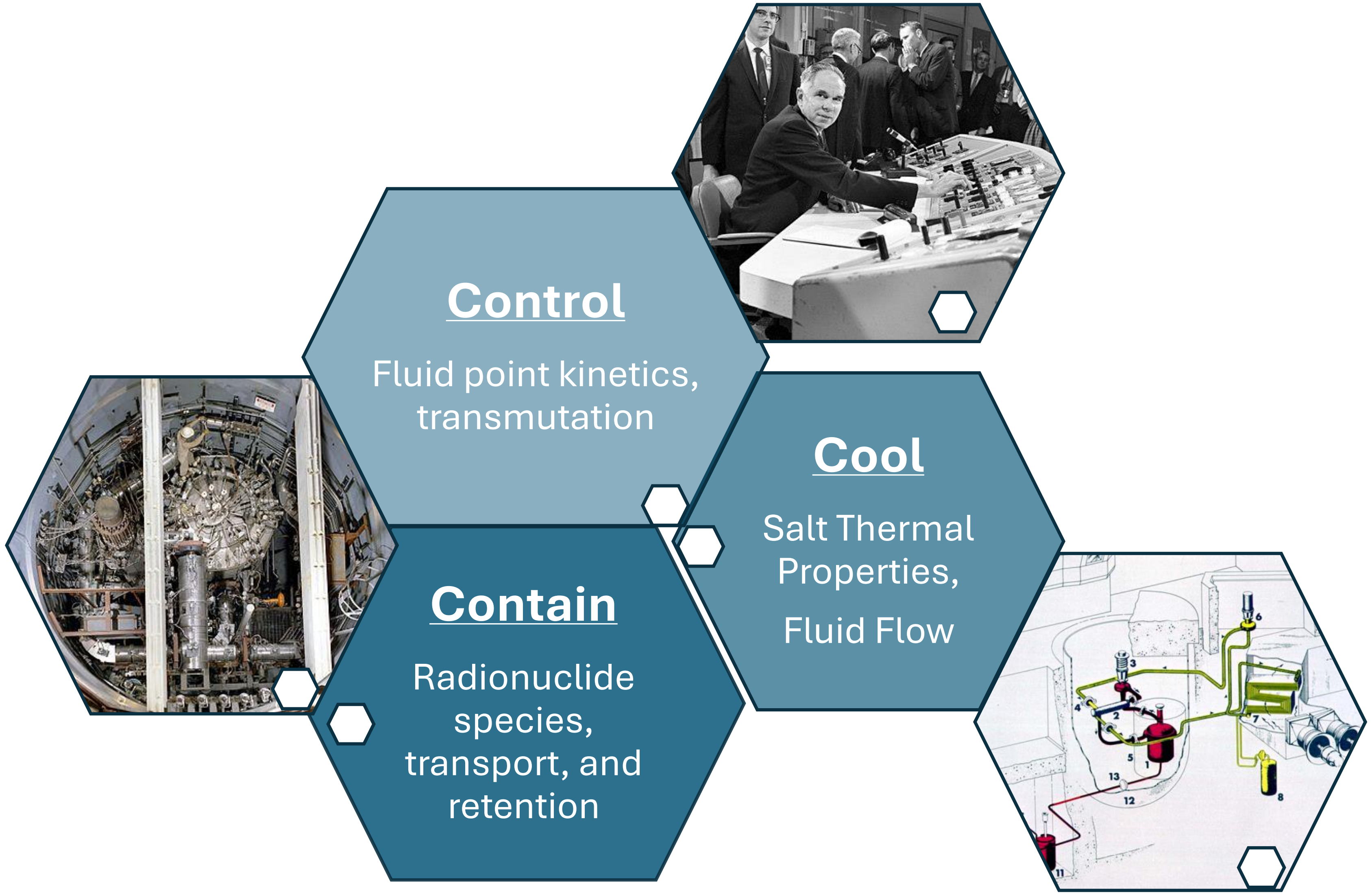
MSR SAFETY Modeling WITH MELCOR (SNL)

- MELCOR MSR source term modeling evaluates efficacy of 3 critical safety functions:
  1. Fission **control**
  2. Reactor **cool**ing
  3. Radionuclide **contain**ment

- MELCOR capabilities have been tested for Molten Salt
  - Additional validation comes from modern reported experiments
  - Incorporation of MSR Campaign results essential to extend validation basis for safety evaluation



Images from https://en.wikipedia.org/wiki/Molten-Salt_Reactor_Experiment

MSR Program helps improve MELCOR MSR models

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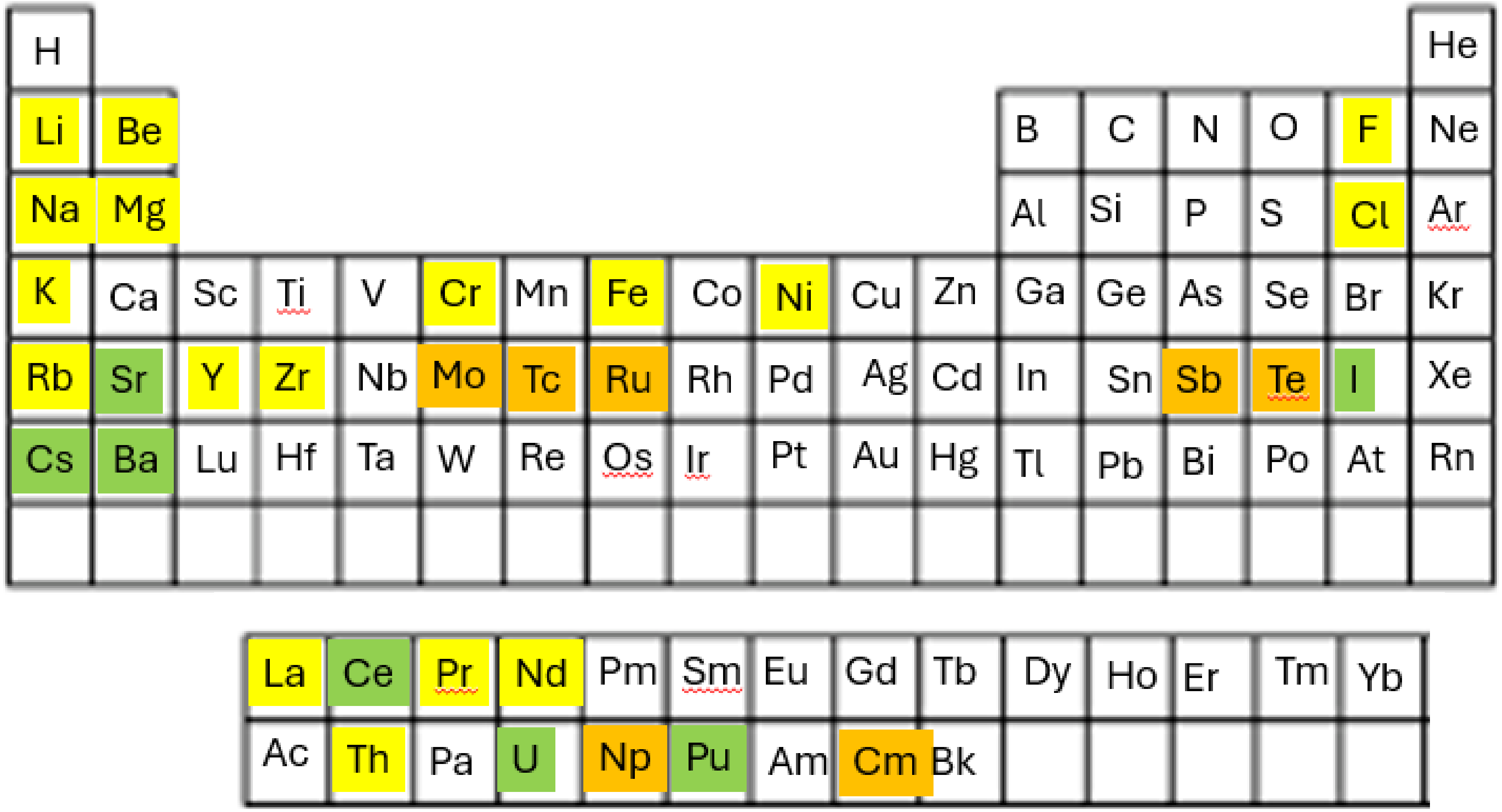
Courtesy Matthew Christian and David Luxat, SNL

Many Health-Consequence Systems Still Need Investigation

- Focus has been on fuel salts and corrosion products
- Many fission product systems remain to be investigated
- Knowing fission product reactivity is important to model source term release

Broader Salt Research Needed

Courtesy Matthew Christian and David Luxat, SNL



MSTDB Fluoride

Toxic RN

MSTDB+Toxic RN

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U.S. MSR Development Includes Government, Industry, and Regulatory Activities as well as International Engagement

U.S. MSR* Development Includes Government, Industry, and Regulatory Activities

- Department of Energy (DOE)-Office of Nuclear Energy (NE) activities remain focused on enabling MSR industry and building supporting infrastructure
  - Largest fraction of DOE support is via the Advanced Reactor Demonstration Program (ARDP)
- DOE-NE continues to support MSRs via multiple mechanisms
  - MSR technical campaign, regulatory development activities, advanced materials and manufacturing campaign
  - Nuclear Energy Advanced Modeling and Simulation (NEAMS) tool development
  - Nuclear Energy University Program (NEUP), small business opportunities, Gateway for Accelerated Innovation in Nuclear (GAIN) vouchers, and direct industry awards
- Advanced Research Projects Agency (ARPA-E) reactor initiatives include MSRs
- Nuclear Regulatory Commission is developing a technology-neutral, performance-based, risk-informed regulatory framework
- DOE- SC, Energy Frontier Research Center – Molten Salt in Extreme Environment
- NNSA- Multiple offices supporting MSR safeguard by design
- CREAMM Initiative (NEA-OECD); CEA-DOE bilateral agreement; IAEA

*MSR support includes both solid (aka FHRs) and liquid fueled concepts

Conclusion

- Nuclear strategic development is key to any long-term success of industrial innovation and important decisions have to be made on the way, which are typically based on current external drivers like political, economic or technical boundary conditions
- For nuclear power to be sustainable as a global source of emission-free energy, the fuel cycle should be
  - Integrated
  - Economically viable
  - Safe
  - Environmentally friendly
  - Proliferation resistant
  - Flexible to adapt to any policy or societal evolution
- MSR concepts are on a fast track, and we need to continue this momentum.

https://gain.inl.gov/doe-molten-salt-reactor-program/

Resources

Save the date
Annual MSR Workshop, Knoxville, TN - 18-20 NOV 2025

Next MSR Program Review in New Mexico, hosted by LANL and SNL – 20-24 April 2026

GIF WEBINARS : https://www.gen-4.org/resources/webinars

Webinar #9 https://www.gen-4.org/resources/webinars/education-and-training-series-9-molten-salt-reactors-msr
Webinar #8 https://www.gen-4.org/resources/webinars/education-and-training-series-8-fluoride-salt-cooled-high-temperature-reactors
Webinar #35 https://www.gen-4.org/resources/webinars/education-and-training-series-35-czech-experimental-program-msr-technology
Webinar #42 https://www.gen-4.org/resources/webinars/education-and-training-series-42-comparison-16-reactors-neutronic-performance
Webinar #44 https://www.gen-4.org/resources/webinars/education-and-training-series-44-molten-salt-reactor-safety-evaluation-us
Webinar #73 https://www.gen-4.org/resources/webinars/education-and-training-series-73-molten-salt-reactors-taxonomy-and-fuel-cycle
Webinar #88 https://www.gen-4.org/resources/webinars/education-and-training-series-88-multiphysics-depletion-chemical-analyses-molten
Webinar #97 https://www.gen-4.org/resources/webinars/education-and-training-series-97-overview-and-update-msr-activities-within-gif

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Samuel Walker, Mauricio Tano, INL, Complete Initial Engineering Framework for Species Tracking in MSRs involving Fuel Salt and Structures

D. Zhang, T. Birri, B. Smith, M. Williamson, V. Glezakou , ORNL, Accelerating Property Prediction and Interfacial Understanding in Molten Salt Reactors Using AI and Advanced Simulations

T.M. Besmann, J. Schorne-Pinto, et al, ORNL, ORNL, Optimization of the thermochemical models for MgCl2-BaCl2, -SrCl2; BaCl2-BaI2; SrCl2-SrI2

N. Shaheen, A. Polke, D. Johnson, N. Hoyt, ANL, Installation of Molten Salt Flow Loop with Chemistry Monitoring and Control System

K. Makovsky, M. Harris, J. Seo, K. Detrick, et al, PNNL, Proposed Strategy for Mitigation of Fluid Inclusions in Molten Salt Precursor Materials

K. Makovsky, M. Harris, J. Seo, K. Detrick, et al, PNNL, Experimental Investigation into Select Thermophysical Properties of the Potassium-Magnesium Chloride Salt System for Molten Salt Reactors

S. Thomas and A. O'Brien,NL, Real-Time Characterization of Salt Aerosols Generated from Static and Sparged Molten Salt ANL/CFCT-25/18

M. Christian, T. Haskin, D. Luxat, SNL, MELCOR Deposition Models Applied to Noble Metals

T. Karlsson, T. Trowbridge, A. Poole, et al, INL, Preliminary Report Summarizing the Initial Investigations into an Irradiated Fuel Salt and Capsule (INL/RPT-25-85239)

C. Downey, T. Karlsson, F. Gleigher, et al, INL, Initial Design of SABRE Fueled Molten Salt Experiment Irradiation Vehicle (INL/RPT-25-87142)

Reports FY 2025 – MSR Program (Cont.)

Bruce McNamara et al, PNNL, Chlorine isotope separations using thermal diffusion (PNNL-38411)
Bryn Merrill, Marisa Monreal et al., LANL, Drop Calorimetry of Actinide-Bearing Chloride Salts
Heather Felmy et al., PNNL, Demonstration of Optical Spectroscopic Flow Cell Design for Molten Salt Reactor Off-Gas Streams (PNNL-38277)
Hunter Andrews et al, ORNL, Analysis of Off-Gas Streams from Small- and Large-Scale Sparging Salt Vessels using Laser-Induced Breakdown Spectroscopy (ORNL/TM-2025/4115)
L Gardner and M Rose, ANL, Uncertainty Analyses of Molten Salt Property Measurements (ANL/CFCT-25/5)
L. Gardner, M Rose et al., ANL, Property Measurements of LiF-NaF-KF Molten Salts Doped with Corrosion Products and Oxygen (ANL/CFCT-25/22)
Manh Thuong nGuyen, PNNL, Computational investigation of thermophysical and structural properties of molten NaCl-PuCl3-AmCl3 (PNNL.38386)
Keerthana Krishnan, Praveen Thallapally et al, PNNL, Radiation Stability of MOFs for Noble Gas Capture and Separation - Gamma radiation study on CaSDB and CuBTC MOFs (PNNL-37418)
Scott Parker, Marisa Monreal et al, LANL, Analysis of Retained Voids in Molten Salts by Novel Image Processing Techniques (LAUR 25-23337)

Thank You

Patricia.Paviet@pnnl.gov

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https://gain.inl.gov/doe-molten-salt-reactor-program/