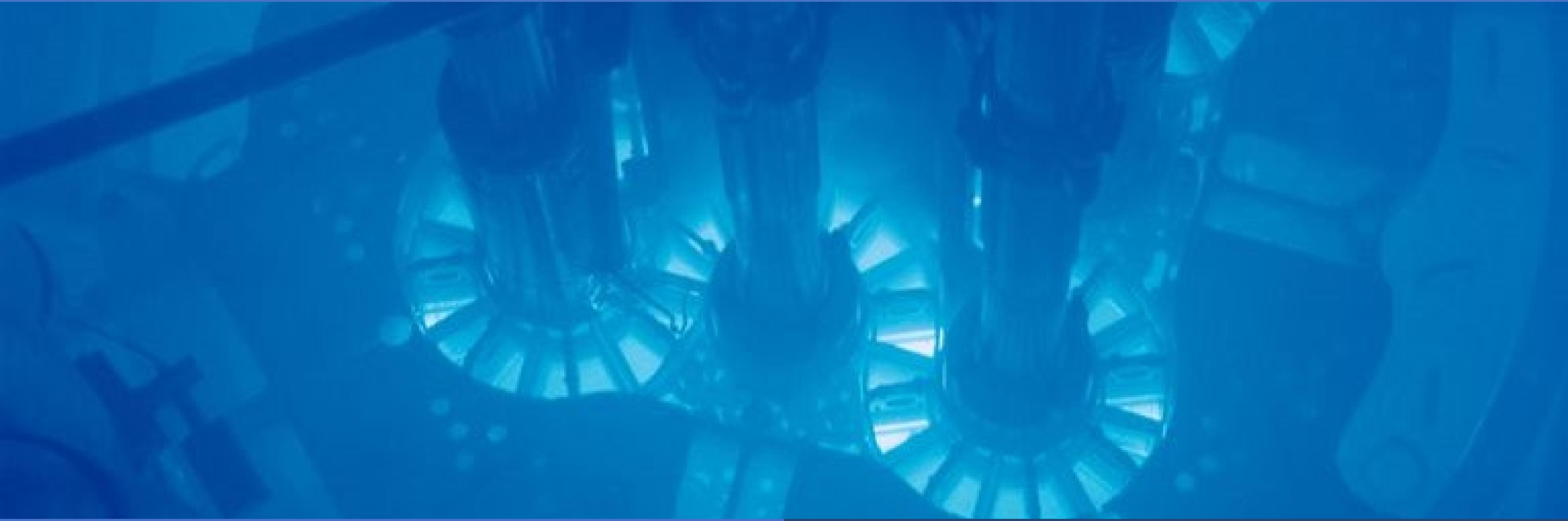


The Fuel Cycle of a Molten Salt Reactor

RIC Conference

March 9, 2022

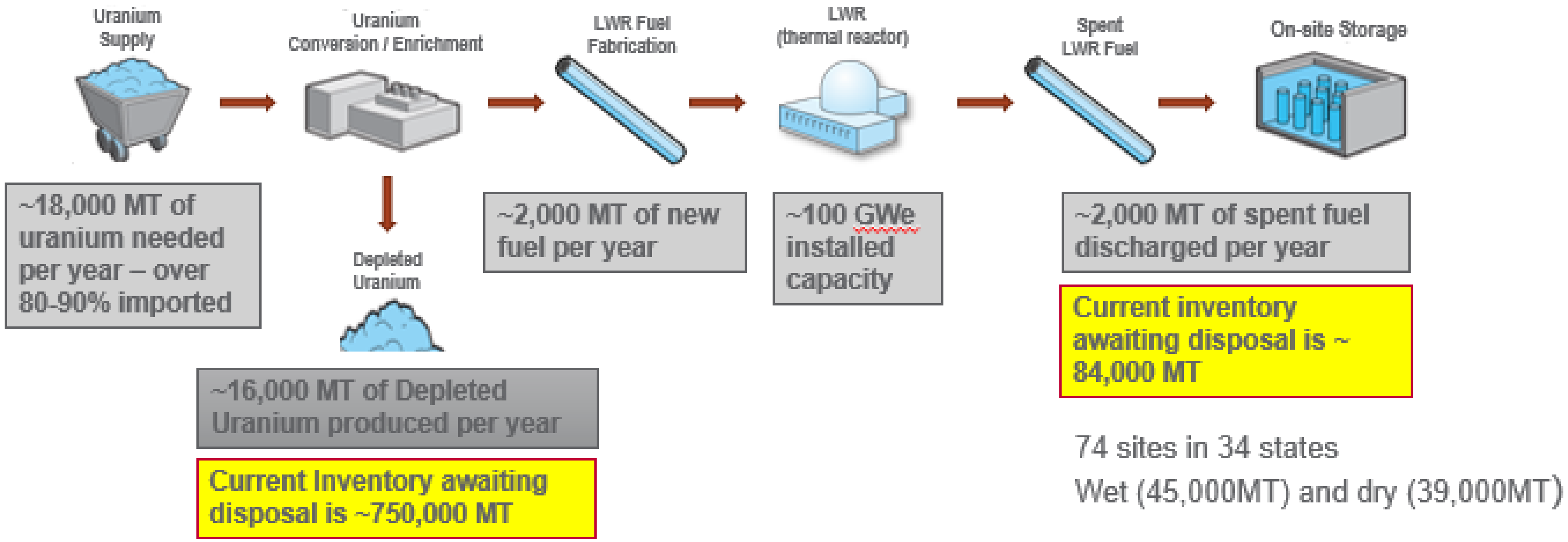
Dr. Patricia Paviet

National Technical Director

Molten Salt Reactor PROGRAM

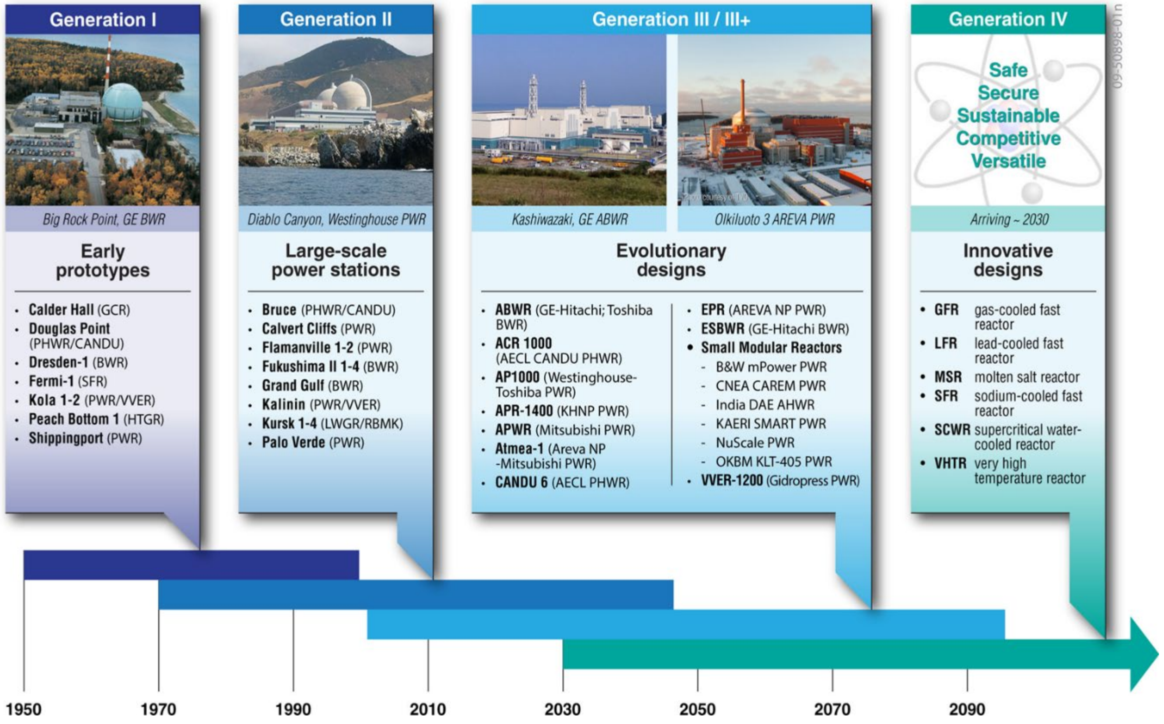
PNNL-SA-170110

Once Through Fuel Cycle

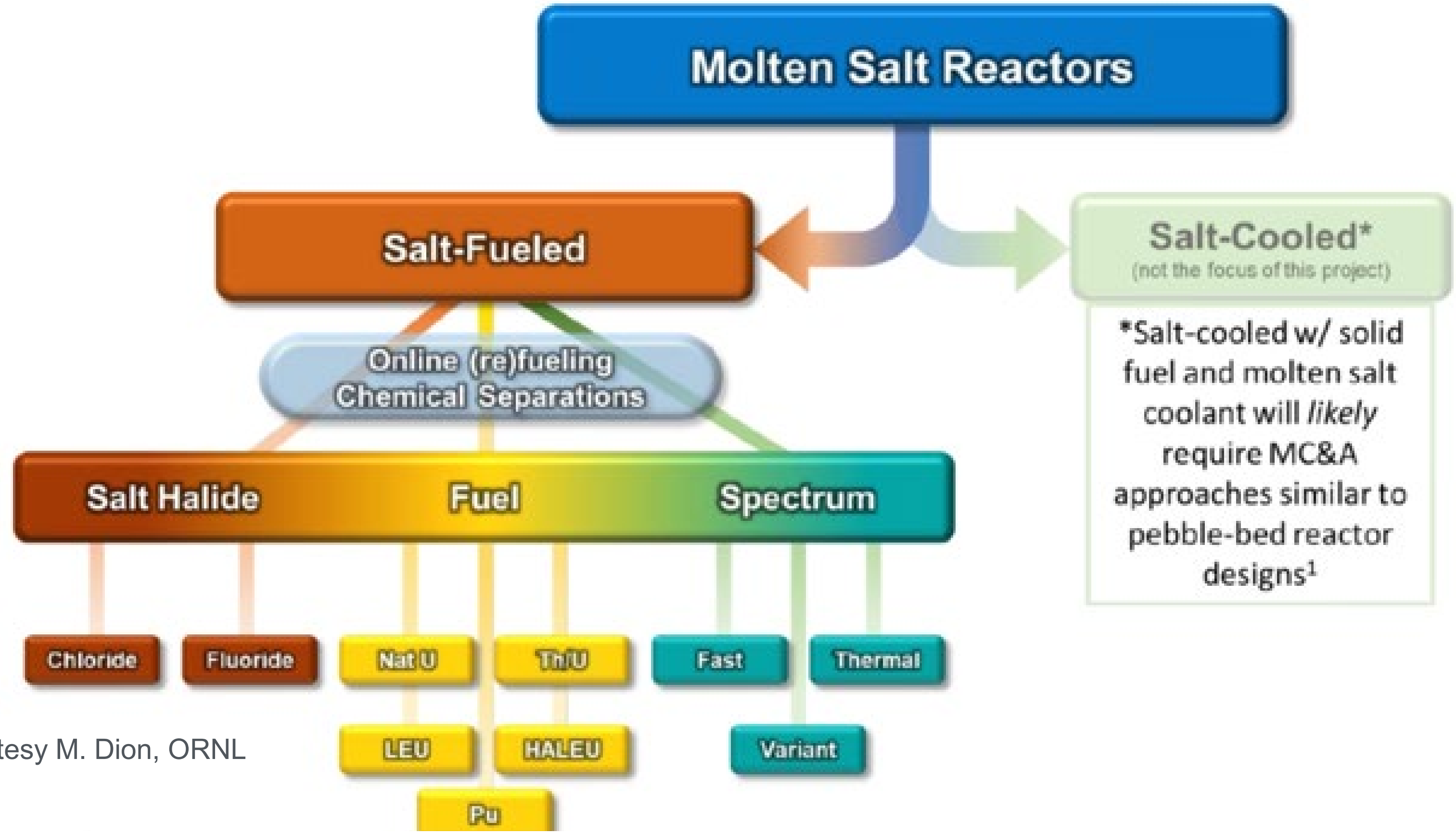


- Currently, the United States operates 94 commercial nuclear power reactors in 29 U.S. states with a combined installed capacity of approximately 100 GWe.

- These pressurized water reactors (PWRs) and boiling water reactors (BWRs) generated approximately **19.7%** of the US electricity demand in 2016.
- The current U.S. once through 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



Molten Salt Reactor – Renaissance?



Courtesy M. Dion, ORNL

A molten salt reactor (MSR) is any nuclear reactor that employs liquid halide salt to perform a significant function in-core. MSRs include a broad spectrum of design options including:

- liquid- and solid-fueled variants,
- chloride- and fluoride-based fuel salts,
- thermal, fast, time variant, and spatially varying neutron spectra,
- wide range of reactor power scales,
- intensive, minimal, or inherent fuel processing,
- multiple different primary system configurations, and compatibility with nearly all fuel cycles

MSRs are a class of reactors in which a molten salt performs a significant function in core

ONE	TWO	THREE	FOUR	FIVE	SIX	SEVEN	EIGHT
TerraPower/ Southern Services	**Thorcon Power**	**Terrestrial Energy**	**Flibe Energy**	**Elysium Industruies**	**Alpha Tech Research Group**	**Muons Inc.**	**MOLTEX**
MCFR, MCRE	Thorcon	IMSR	LFTR, SCIFR	MCSFR	ATRC	Mu*STAR	SSR-W, SSR-U
Fast	Thermal	Thermal	Thermal	Fast	Thermal	Thermal	Fast/Thermal
MSR Fast UCl_3, $PuCl_3$	MSR UF_4	MSR UF_4	MSR UF_4, Th_4, $PuCl_3$	MSR Fast UCl_3, PuCl	MSR ThF_4	MSR UF_4	UCl_3, $PuCl_3$, $AmCl_3$/ UF_4
Breeder	Burner	Burner	Breeder	Breeder	Breeder	Burner	Waste burner/breeder
Salt Cooled	Salt Cooled	Salt Cooled	Salt Cooled	Salt Cooled	Salt Cooled	Salt Cooled	Salt Cooled
Uranium	Thorium	Uranium	Thorium	U/Pu	Thorium	Uranium	
(Could use Th)		(Could use Th)					

Molten Salt Reactor Program: Goal and Objectives

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

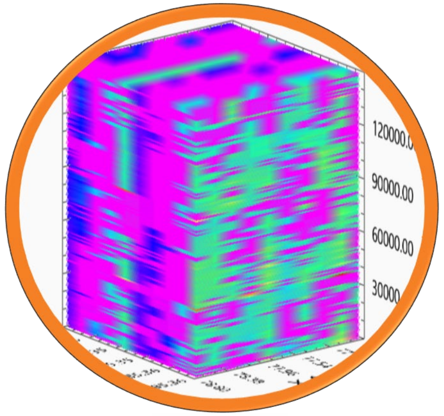
1) a substantial portion of the energy needed for the US to achieve net zero carbon emissions by 2050 and
2) abundant energy worldwide for the foreseeable future.

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 & Computationally

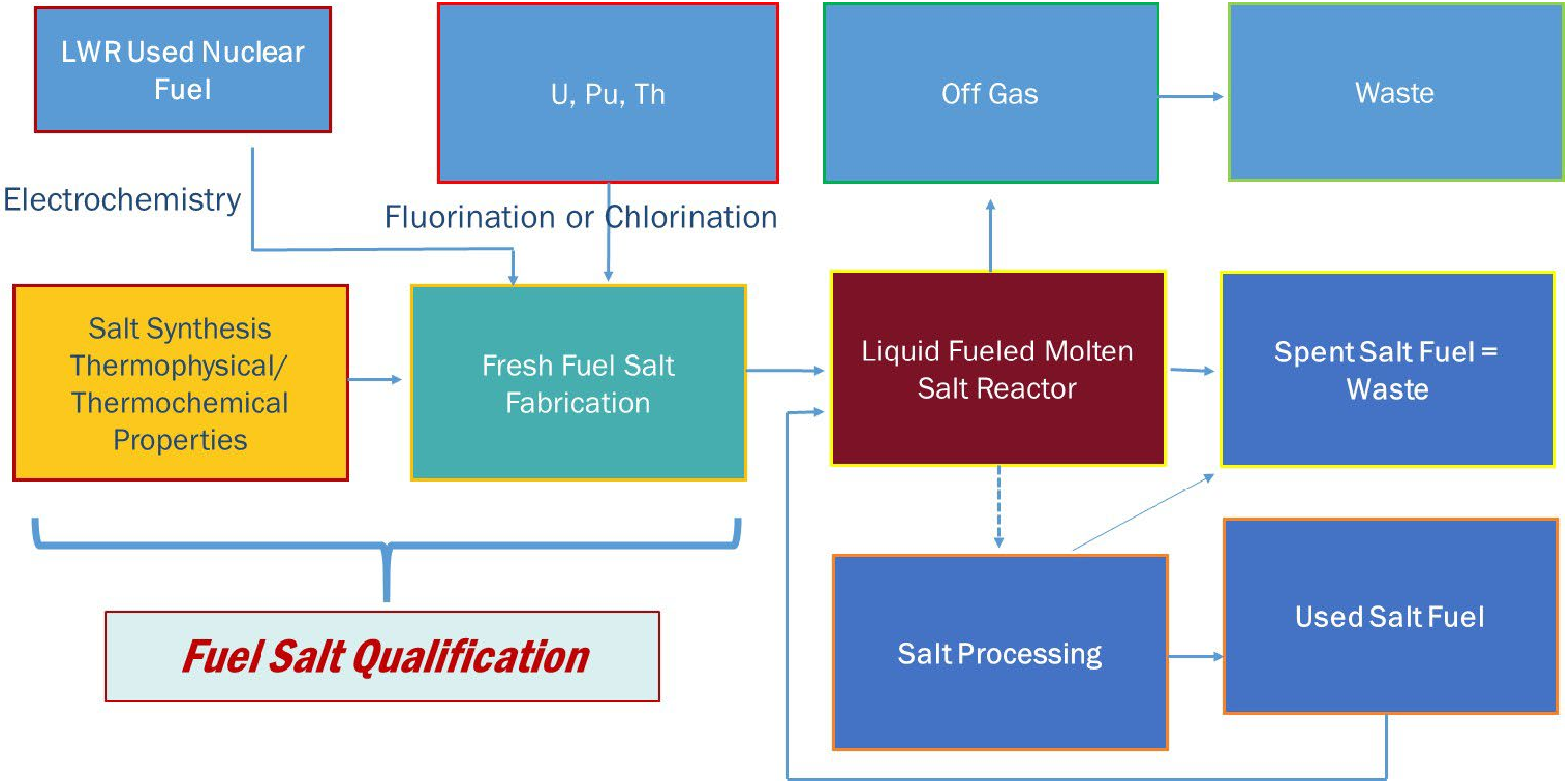
Technology Development and Demonstration
Radionuclide Release Monitoring, Sensors & Instrumentation, Liquid Salt Test Loop

Materials
First to Market: Gaps in Codes and Standards for SS316H
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.

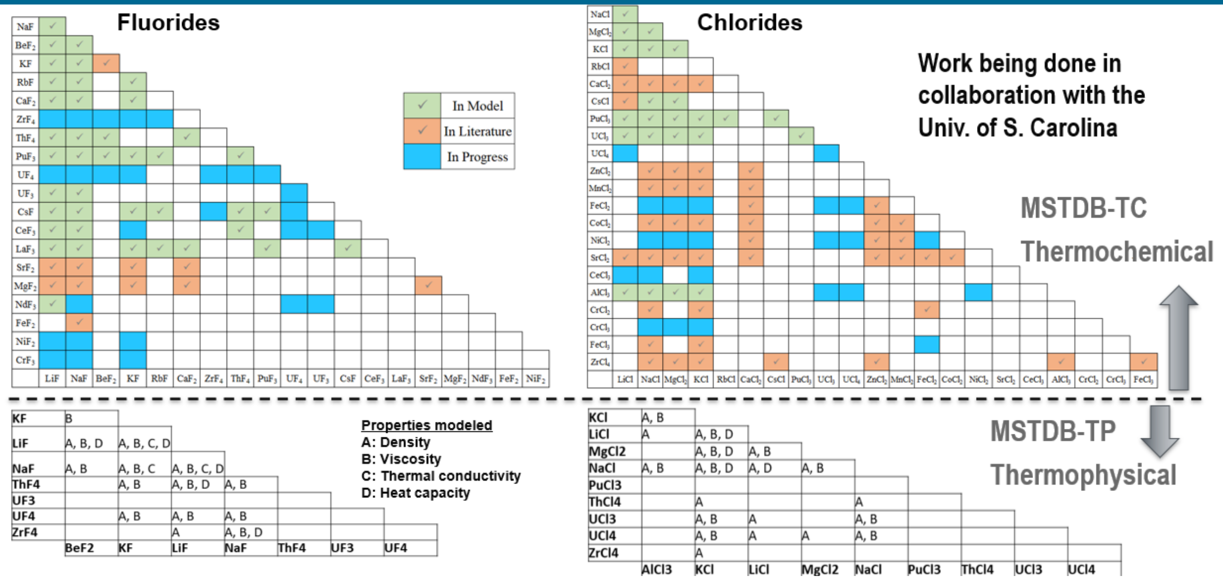
Liquid Fueled MSR – Generic Fuel Cycle



Salt Synthesis – Thermophysical and Thermochemical Properties

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

- Complete table with missing data/database
- Implement QA (graded approach)
- Develop standard methodology (share best practices)
- **No standard metric currently exists on adequate purity for fuel salts** and it is recommended that the MSR campaign work with industry to **develop consensus standards** as to **acceptable fuel salt** and other material characteristics.



Data quality is a key element to assessing technical uncertainties in property values of salt mixtures that are pivotal in molten salt reactor (MSR) design and safety assessments under both normal operating conditions and accident scenarios.

Some key properties for the salt mixtures being evaluated for use in MSRs have not been measured and the **few values** for relevant salt compositions available in the literature **are inconsistent** and **not suitable for use in licensing**.

The applications of established techniques commonly used for property measurements to molten salts are still experimental in nature and vetted consensus standards are not yet available.

Work is in progress to develop and standardize methods through DOE and commercially funded projects.

W. Ebert & M. Rose, ANL/CFCT/21/18, August 2021

Molten Salt Thermochemical Properties Database vs. Molten Salt Thermophysical Properties Database

- **Public version has been completed, tested, and made available at https://code.ornl.gov/neams/mstdb/**
- **Obtainable cost-free after registering an ORNL XCAMS account and access approval**
- **Permits access to *MSTDB-TP* as well**

Fluoride and Chloride Salt Content

96 Pseudo-binary systems

37 Pseudo-ternary systems

42 Solid Solutions

229 Stoichiometric compounds
130 Gas species

- ***MSTDB-TP* Version 1.0 finalized (open access available)**
- **Type of data (along with references and uncertainty)**
  - Melting temperature
  - Boiling temperature
  - Density
  - Thermal Conductivity
  - Heat capacity
  - Viscosity
- **62 entries currently: 27 pure, 8 binaries, 10 ternaries & 5 quaternaries**

https://sc.edu/study/colleges_schools/engineering_and_computing/research/research_centers_and_institutes/general_atomics_center/molten_salt_thermal_properties_databases/index.php

POC: Ted Besmann, besmann@sc.edu

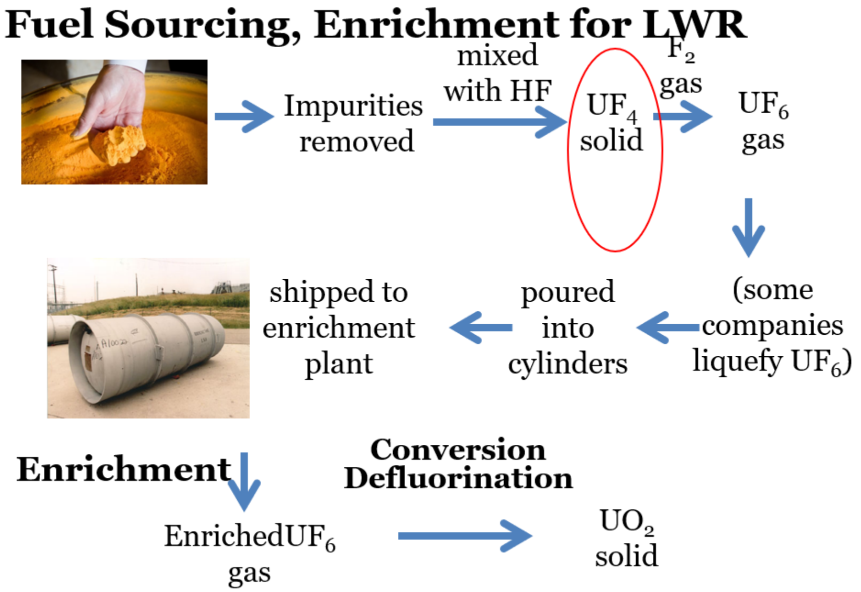
POC: Dianne Ezell, bullnd@ornl.gov

Liquid Fueled MSR - Generic Fuel Cycle

- The fuel salt for an MSR will be a combination of the fissile salt: as an example UF_4 or UCl_3, with nonradioactive diluent or carrier salts, such as LiF, ZrF_4, NaF, and BeF_2 or NaCl, $CaCl_2$ and $MgCl_2$.
- It is likely that the company producing the fuel salt will produce the fissile salt, purchase the non-radioactive salts from commercial sources, and then combine them to produce the fuel salt mixture.
- Depending on the MSR design, the fuel salt may contain other components, such as fertile salts (U or Th) for breeder MSRs.
- Reuse of spent LWR fuel may mean that actinides and fission products are present in the fuel.

Joanna McFarlane, Review of Hazards Associated with Molten Salt Reactor Fuel Processing Operations, ORNL/TM-2019/1195, June 2019

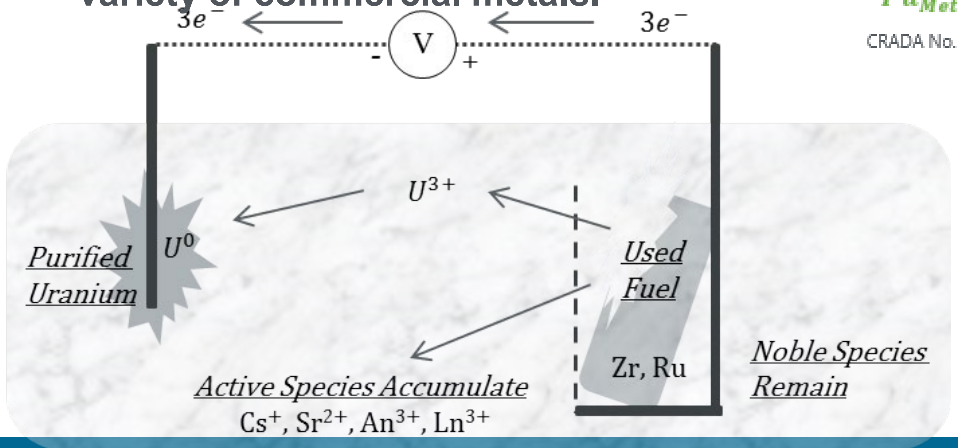
Fluoride System



Chloride System

UCl_3

Heart of modern molten salt-based separations processes is electrorefining, an electrochemical process also used to produce a variety of commercial metals.



Preparation of Starting Material

– Solid : gas synthesis route

$$Pu_{Metal} + 3NH_4Cl = PuCl_3 + 3NH_{3_{gas}} + 1.5H_{2_{gas}}$$

CRADA No. 18-CR-17, "Integrated Effects Testing for a Molten Chloride Fast Reactor."

Fuel Salt Qualification

D. Holcomb, W. Poore, G. Flanagan, USNRC Draft Report, Fuel Qualification for MSR , July 2021, ML 21245A493

“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”

- Reviewed by the ACRS in November 2021 and recommended for publication following resolution of comments.
- Proposed fuel salt qualification process based upon maintaining fuel salt properties within a range that results in plant achievement of fundamental safety functions
  - Both: normal and accident conditions.
- Provides the technical basis for fuel salt property database development.
- Includes rationale for measurement ranges and uncertainty limits.

US Nuclear Regulatory Commission. 2017. Public Meeting on Improvements for Advanced Reactors, August 3, 2017. Washington, DC, https:/www.nrc.gov/docs/ML1722/ML17220A315.pdf

Salt Synthesis

Fresh Fuel Salt Fabrication

GAPS

While the feedstock chemicals for MSR fuel salts have been produced in large quantities, only two relatively small test MSRs have ever been operated: the Aircraft Reactor Experiment (ARE) in 1954, and the MSRE from 1965–69. **Consequently, no large-scale fuel salt production facilities have ever been built.**

Joanna Mc Farlane, Review of Hazards Associated with Molten Salt Reactor Fuel Processing Operations, ORNL/TM-2019/1195, June 2019

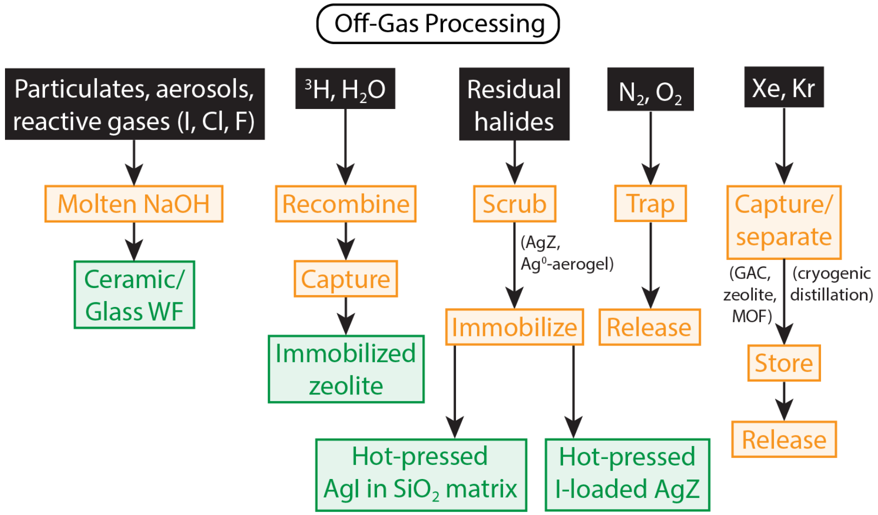
- **Gap 1**: Purification of the initial salt product
  - Production at tonnage scale
- **Gap 2**: Fabrication of the fuel salt at tonnage scale
- **Gap 3**: Fuel qualification
  - Standards do not exist
  - No centralized NQA1 program exists as of today for the determination of thermophysical properties of molten salts
  - Literature is very inconsistent
- **Gap 4**: Transportation of salt/fuel salt to reactor site

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

- **Volatile off-gas components have wide range of half-lives and disposal requirements**:
  - ^{3}H 12.31 y
  - ^{14}C 5715 y
  - Xe-131m 12 d
  - Xe-133 5.27 d
  - Xe-137 3.8 min
  - I-131 8 days
  - I-129 15.7 E6 y
- **EPA 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 - (1.85×10^{15} Bq / GWy)
    - 5 mCi/GWy ^{129}I - (1.85×10^{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.

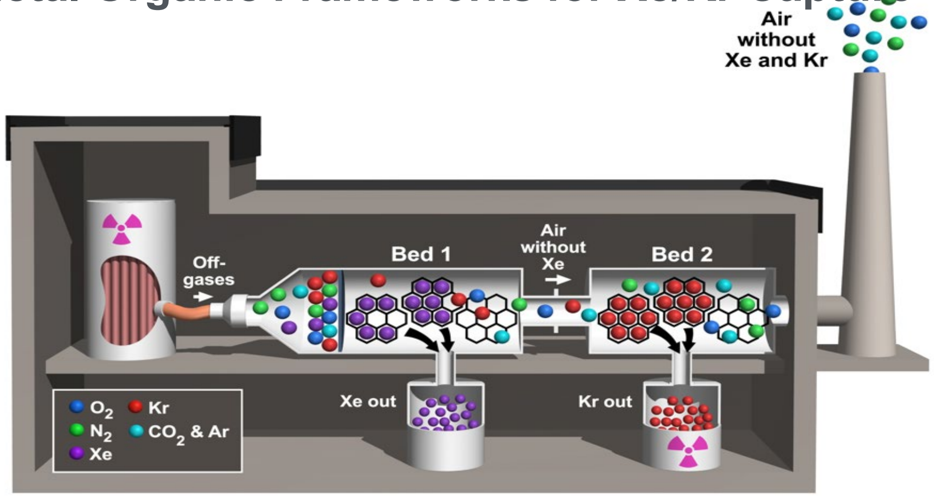
Unlike solid fuel, liquid fuel salt does not retain significant quantities of gaseous fission products, thus increasing the releasable fraction of fission gases from a breach.

Options for Off-Gas Capture and Immobilization for MSR Concept



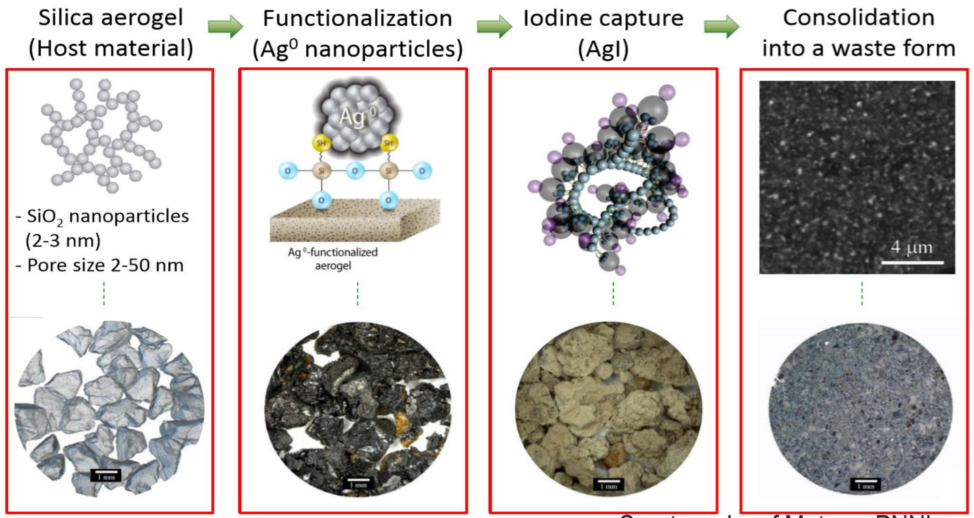
B. J. Riley, J. McFarlane, G. D. DelCul, J. D. Vienna, C. I. Contescu, C. W. Forsberg, "Molten salt reactor waste and effluent management strategies: A review," Nuclear Engineering and Design, Volume 345, 2019, Pages 94-109,

Metal Organic Frameworks for Xe/Kr Capture



Courtesy Praveen Thallapally, PNNL

Silica Aerogel for I-129 capture and immobilization



Courtesy Josef Matyas, PNNL

Proper handling of the off-gas system will be important to reactor operation and site safety.

Configuration of the off-gas system to confine fission gases and trap particulates means that it will be operating continuously and also that on-line maintenance will be required.

The greatest technical challenge for reactor developers will be in assessing off-gas performance during reactor operation.

Gamma spectroscopy is used in current reactor designs, but the background in an MSR off-gas may reduce the effectiveness of gamma analysis. Online sampling methods for FP compounds by optical spectroscopy are considered.

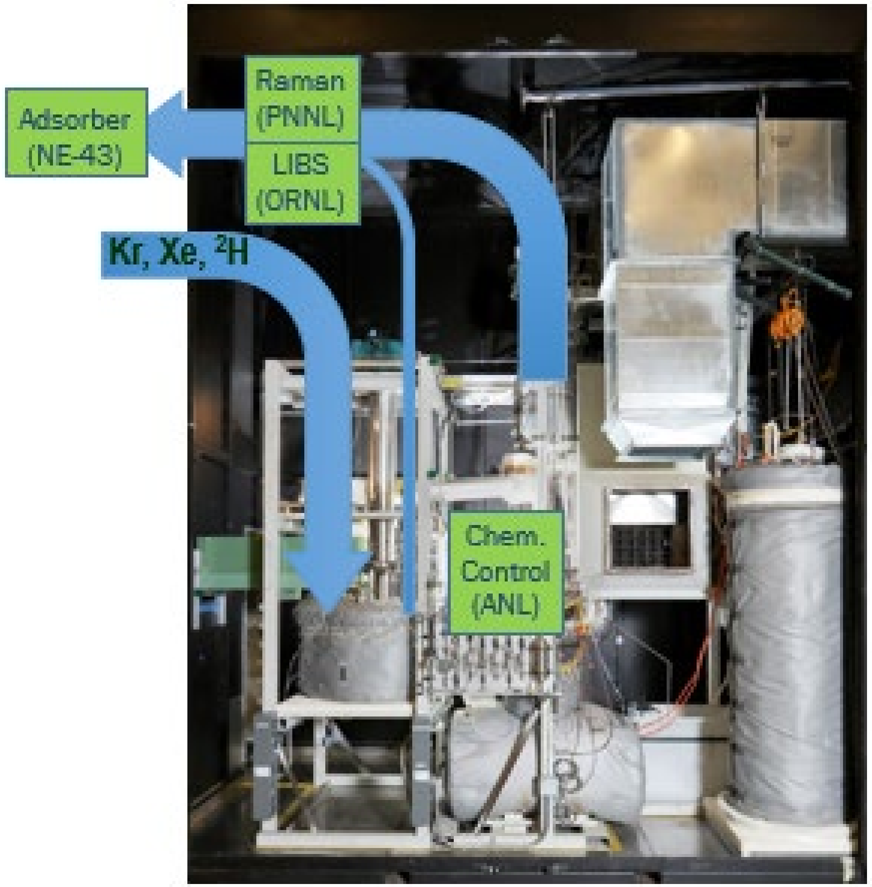
Joanna Mc Farlane, Review of Hazards Associated with Molten Salt Reactor Fuel Processing Operations, ORNL/TM-2019/1195, June 2019

Liquid Salt Test Loop and Sensor Development / Demonstration for MSR monitoring

Existing & operable salt test facility is unique in the U.S. for technology development and demonstration with relevant powers, temperatures, and flowrates

PNNL/ORNL Xenon Radionuclide release and monitoring using laser-induced breakdown spectroscopy

ORNL– Salt loop and capability for testing sensors and off-gas components



ANL - Distributed salt chemistry monitoring and control

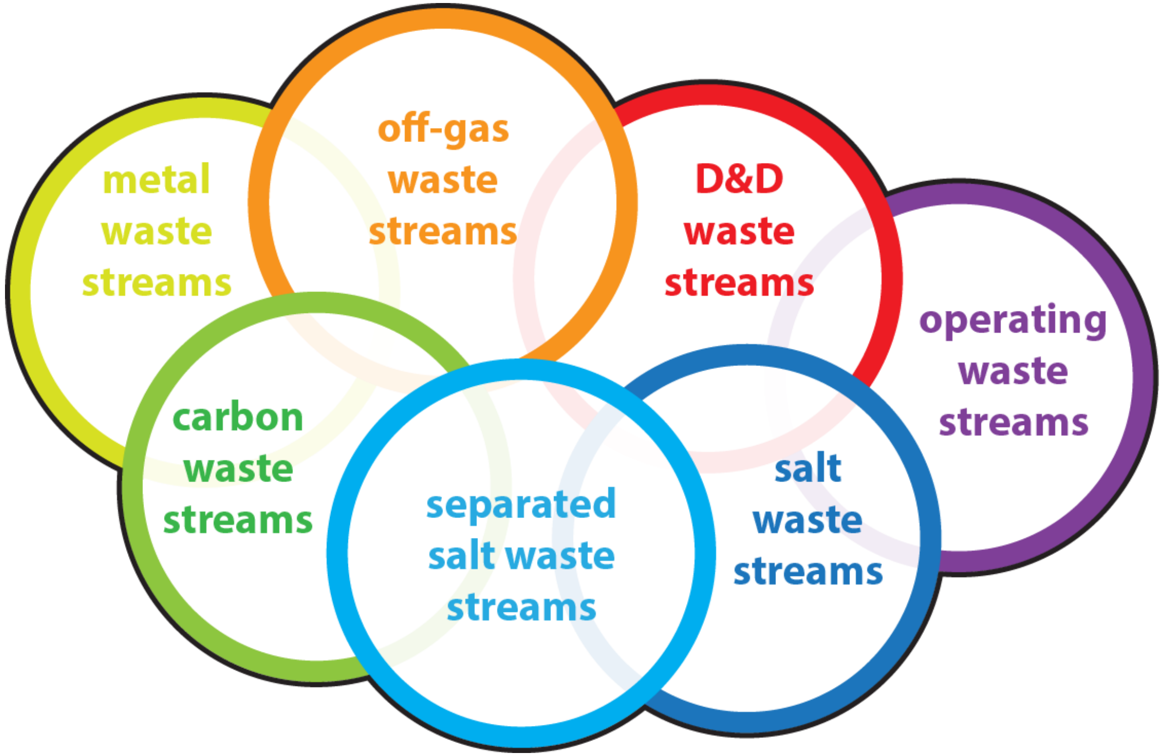
PNNL– Raman and FTIR sensor development for iodine species and tritium

INL– Tritium transport

Waste Forms Development

Wastes from an MSR will include:

- those generated during salt preparation or purification prior to irradiation
- those generated during operation such as through sampling, analysis, online processing, filter change-out, or off-gas management
- those generated at the end of a fueling cycle
- those generated at the end of reactor operation

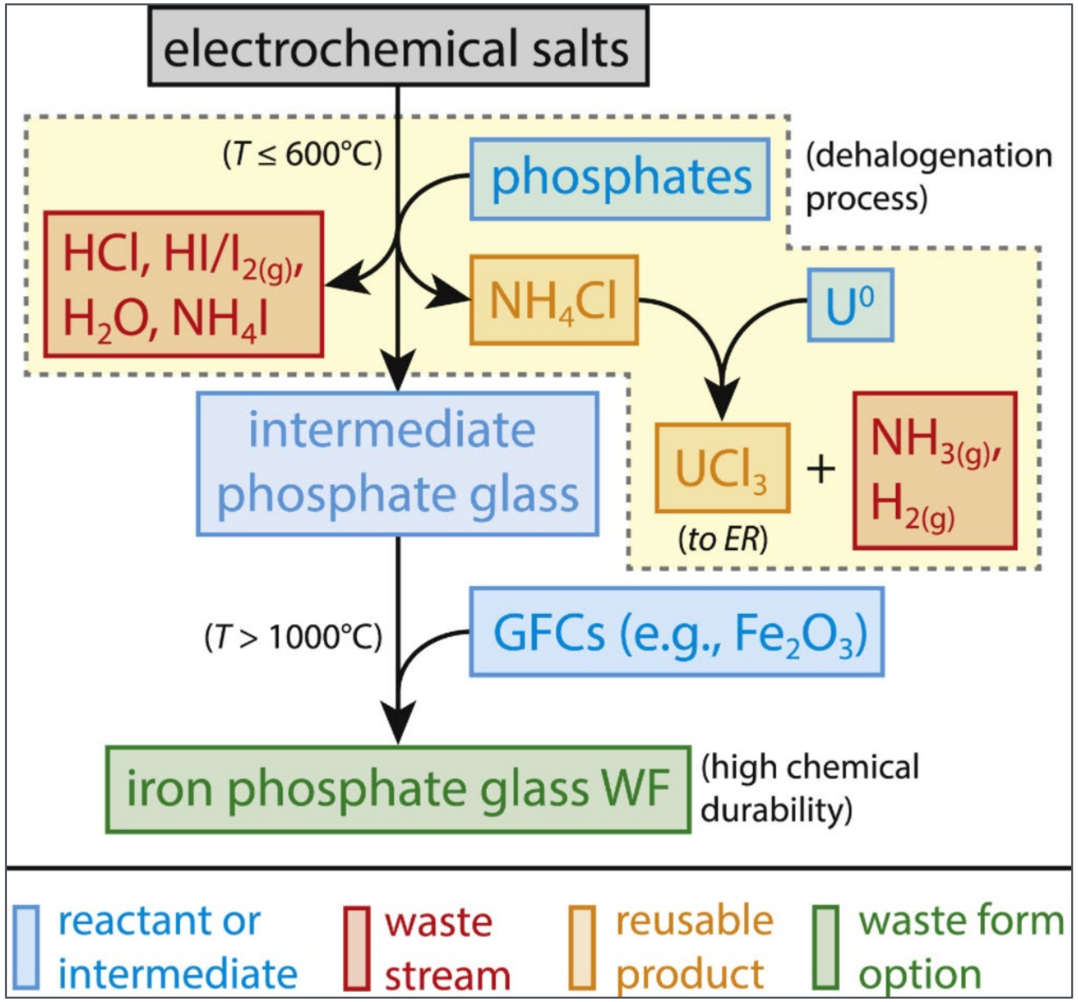


B. J. Riley, J. McFarlane, G. D. DelCul, J. D. Vienna, C. I. Contescu, C. W. Forsberg, "Molten salt reactor waste and effluent management strategies: A review," Nuclear Engineering and Design, Volume 345, 2019, Pages 94-109,

Many of the radiological hazards will be similar to those for operation of other nuclear power plants

Storage

- One distinctive aspect of the MSR fuel cycle is that used fuel salts becomes more difficult to store over time while LWR fuel storage becomes easier.
- Current U.S. regulations require the ability to store used fuel on site indefinitely to accommodate the possibility that a geological repository never becomes available.
- Halogen gas release from used fuel salts during cooling is problematic.
- The high temperature tolerance of fuel salt will allow it to be transferred to air cooled containers likely without ever using a used fuel pool, which significantly ease design basis accident requirements - 10CFR50 Part 155
- Radiolysis in fluoride-based fuel salts results in the formation of fluorine gas (F_2) and the potentially of uranium hexafluoride (UF_6).
- Chloride-based fuel salts lack an equivalent gaseous uranium species, however, upon cooling, radiolytically generate chlorine gas (Cl_2) will form. Chlorine-36 is a long-lived, energetic beta emitter to require containment.



DOE-NE has sponsored development of a dehalogenation method for electrochemically processing chloride salts to allow for stabilization in an iron phosphate glass matrix, and UCl_3 to be suitable for incorporation into fresh fuel salt

B. J. Riley, J. A. Peterson, J. D. Vienna, W. L. Ebert, and S. M. Frank, Dehalogenation of electrochemical processing salt simulants with ammonium phosphates and immobilization of salt cations in an iron phosphate glass waste form, Journal of Nuclear Materials, 529, 2020, DOI: 10.1016/j.jnucmat.2019.151949

Conclusion

- The MSR program serves as the hub for efficiently and effectively addressing, in partnership with other stakeholders, the technology challenges for MSRs to support their entrance to the commercial market.
- MSR concepts are on a fast track
  - 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 development of TerraPower's molten chloride fast reactor system

Molten Salt Reactor
PROGRAM

Clean. **Reliable. Nuclear.**

Patricia.Paviet@pnnl.gov