

PNNL-30446

Pacific Northwest National Laboratory Capabilities for Molten Salt Reactor Technologies

September 2020

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Contents

| | |
|--|-----|
| Contents | iii |
| 1.0 Introduction | 1 |
| 2.0 Molten Salt Physical, Chemical, and Thermodynamic Properties | 3 |
| 2.1 Safe Handling of Molten Salts..... | 3 |
| 2.2 Thermal Expansion and Density | 6 |
| 2.3 Chemistry of Molten Salt Using X-ray Absorption Spectroscopy | 7 |
| 2.4 Thermal Properties & Thermodynamics of Chloride and Fluoride Molten Salts | 8 |
| 2.4.1 Drop Calorimetry | 8 |
| 2.4.2 Differential Scanning Calorimetry..... | 10 |
| 2.4.3 Laser Flash Analysis | 11 |
| 3.0 Materials Corrosion – Modeling and Performance | 12 |
| 3.1 Corrosion Modeling..... | 12 |
| 3.2 Salt/Material Interactions and Corrosion..... | 13 |
| 3.3 Corrosion References | 13 |
| 4.0 Fuel and Coolant Salt Processing | 14 |
| 4.1 Isotopic enrichment and separation of Cl-37 | 14 |
| 4.2 Producing fluoride salt fuels from irradiated fuels | 14 |
| 4.3 Removing fission products from salt mixtures | 15 |
| 4.4 Fuel and Coolant Salt Processing References | 15 |
| 5.0 Instrumentation, Control, and Online Monitoring | 15 |
| 5.1 Spectroscopic-Based Process Monitoring Summary at PNNL | 17 |
| 5.2 On-Line Structural Health Monitoring of Critical Reactor Components..... | 18 |
| 5.3 In-vessel MSR/fluoride salt-cooled high-temperature reactor (FHR) Monitoring Requirements, Challenges, and Opportunities | 19 |
| 5.3.1 Coolant & Cover Gas Chemistry Monitoring | 19 |
| 5.3.2 Electromagnetic Pump Monitoring | 19 |
| 5.3.3 Flow Blockage Detection..... | 19 |
| 5.3.4 Coolant Flow and Pressure | 19 |
| 5.3.5 Coolant Level | 20 |
| 5.3.6 Neutron Flux/Power | 20 |
| 5.4 Instrumentation, Control, and Online Monitoring References | 20 |
| 6.0 Reactor Design and Safety Analysis | 21 |
| 6.1.1 Safety Analysis..... | 22 |
| 6.1.2 Quality Assurance | 22 |
| 6.1.3 Reactor Chemistry | 22 |
| 6.1.4 Reactor Physics, Neutronics, and Nuclear Design | 22 |

| | | |
|--------|--|----|
| 6.1.5 | Mechanical Engineering | 23 |
| 6.1.6 | Component Integrity | 23 |
| 6.1.7 | Probabilistic Risk Assessment (PRA)/Severe Accidents | 23 |
| 6.1.8 | Radiation Protection and Dose Assessment..... | 23 |
| 6.1.9 | Human Factors Engineering | 24 |
| 6.1.10 | Site Characteristics and Site Parameters | 24 |
| 6.1.11 | Operating Reactor Experience Base..... | 24 |
| 6.1.12 | Fire Protection..... | 24 |
| 6.1.13 | Cyber Security..... | 25 |
| 6.1.14 | External Hazards..... | 25 |
| 7.0 | Capture and Immobilization of Actinides and Fission Products in Waste Forms..... | 26 |
| 7.1 | Glass and Material Science | 27 |
| 7.2 | Nuclear Waste Immobilization Technology..... | 27 |
| 7.3 | Waste Form Durability | 28 |
| 7.4 | Immobilization of electrochemical salt wastes | 28 |
| 7.5 | Capture and Immobilization of Volatile Radionuclides..... | 28 |
| 7.5.1 | Development of Sorbents for Volatile Radionuclides | 29 |
| 7.5.2 | Capture and immobilization of volatile radionuclides using metal organic frameworks (MOFs) and off-gas system design..... | 29 |
| 7.5.3 | Capture and Immobilization of Volatile Radionuclides References | 30 |
| 8.0 | Safeguard & Signatures..... | 32 |
| 9.0 | Licensing..... | 32 |
| 9.1 | NRC Licensing References..... | 34 |

Figures

| | |
|---|----|
| Figure 2.1. a) Laser Star welder capable of hermetically sealing metal crucibles. B) Precession of welds on a 6mm diameter silver crucible. | 5 |
| Figure 2.2. Al_2O_3 and BN crucibles for testing thermal expansion and calculating density with a TMA. | 6 |
| Figure 2.3. a) Non-rad Netzsch TMA located at PNNL capable of measuring thermal expansion up to 1500°C and b) density data calculated on the NaCl-KCl binary using the TMA and pycnometer. | 6 |
| Figure 2.4. EasyXAFS 300 for in situ molten salt measurements at PNNL..... | 7 |
| Figure 2.5. Schematic of high-temperature twin Calvet calorimeter and assembly for drop solution calorimetry in a molten oxide solvent | 10 |
| Figure 2.6. Netzsch DSC capable of testing non rad salts to 1500°C and b) TA DSC capable of testign rad salts to 700°C | 11 |
| Figure 2.7. a) Beginning of a phase diagram map showing the melting temperatures of the NaCl-KCl binary and b) the heat capacity from 50°C to 900°C for the NaCl-KCl binary. | 11 |
| Figure 2.8. Netzsch LFA capable of testing thermal difusivity from 25 to 1000°C | 12 |
| Figure 7.1. Silver-functionalized silica aerogel developed at PNNL to capture and immobilize radioiodine from reprocessing off-gas streams..... | 29 |
| Figure 7.2. The PNNL patented MOF technology can be used to separate the noble gases from off gases | 30 |
| Figure 7.3. The two-step approach removes Xe (bed 1) and Kr (bed 2) at room temperature | 30 |

1.0 Introduction

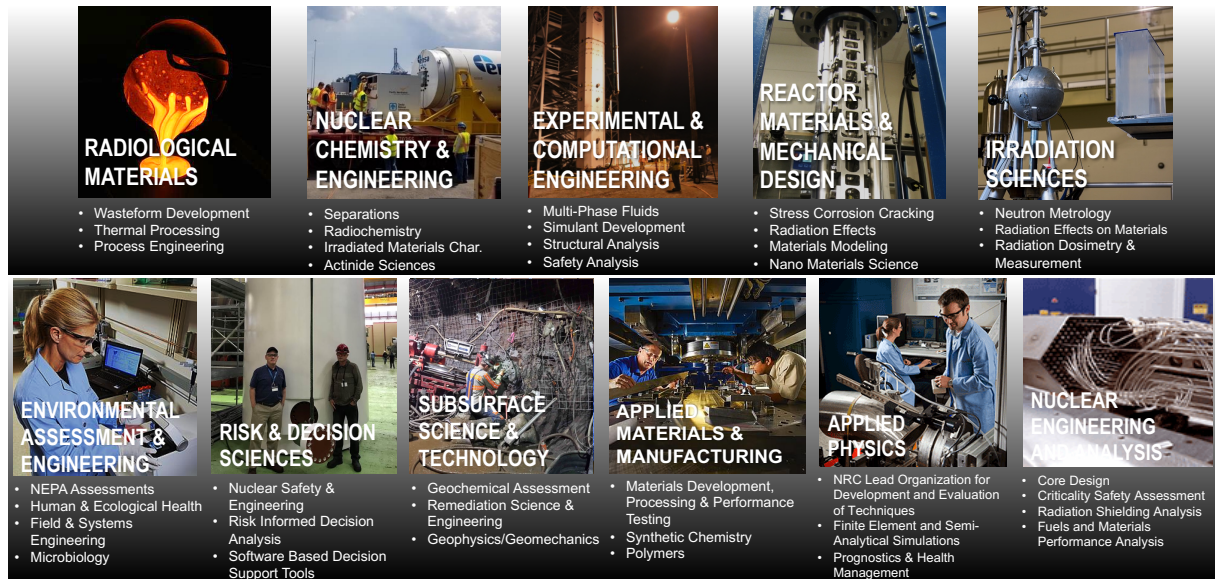
Pacific Northwest National Laboratory (PNNL) has a long heritage supporting our nation's nuclear energy programs by facilitating the regulatory licensing, design, and analysis of conventional and advanced reactors; safely extending the operational lives of the existing fleet; and developing and implementing processes for recycling, storage, transportation, and disposal of spent nuclear fuel.

With more than 250 staff committed to the nuclear energy mission space, complemented by state-of-the-art nuclear facilities—such as the Radiochemical Processing Laboratory (RPL), a Hazard Category II Non-Reactor Nuclear Facility—our diverse expertise and innovative solutions are enabling both federal and commercial partners to achieve a reliable, safe, and secure energy future.

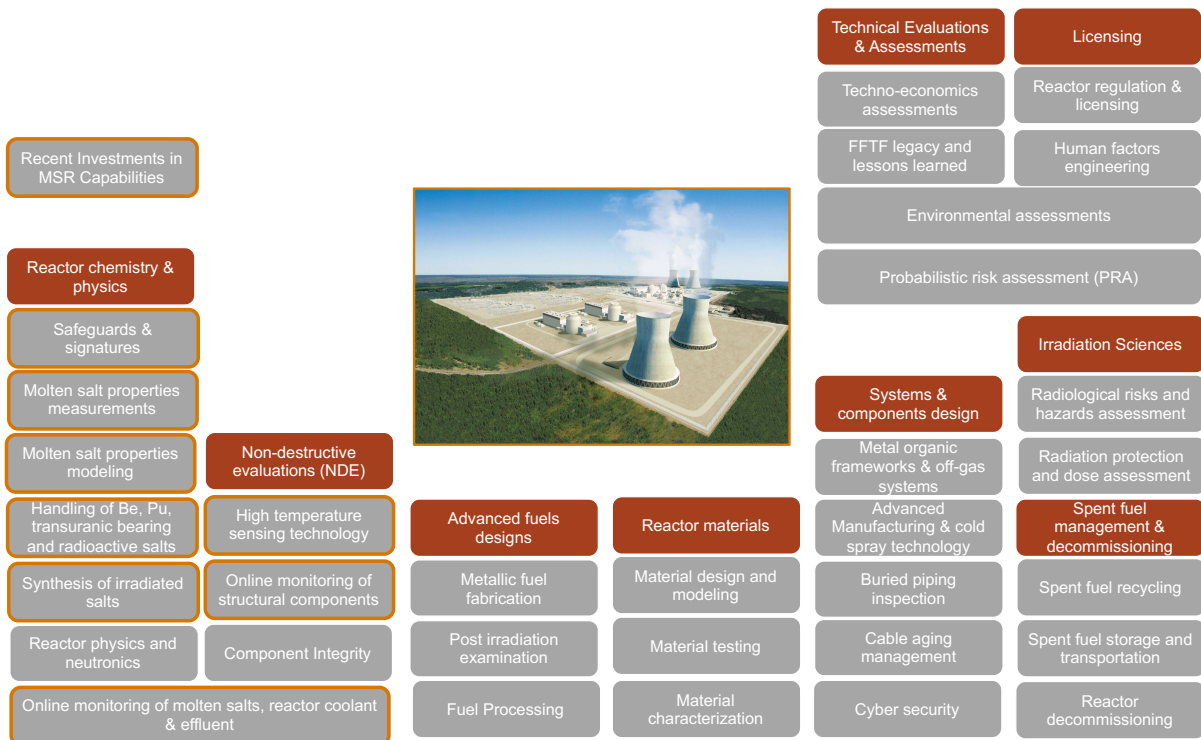
PNNL's capabilities in support of deploying advanced nuclear reactors include:

- Advanced Fuel Concepts
- Advanced Modeling and Simulation
- Cybersecurity
- Material Science
- Nondestructive Evaluation (NDE) and Structural Component Integrity
- Post-Irradiation Examination
- Probabilistic Risk Assessment
- Radiological Risks and Hazards Assessment
- Reactor Decommissioning
- Reactor Regulation and Licensing
- Risk-Informed License Amendment Reviews
- Seawater-Based Critical Materials
- Spent Fuel Recycling
- Spent Fuel Storage and Transportation
- Techno-Economic Assessments
- Advanced Manufacturing Techniques

This document presents detailed information regarding PNNL's capabilities that support the development and ultimate deployment of advanced molten salt reactors (MSRs). Our expertise and experience provide for seamless development and maturation through technology readiness levels to enable the resolution of critical technical challenges in the development and deployment of MSRs. Our facilities where nuclear research and development activities are conducted are comprehensive, modern, and unique.

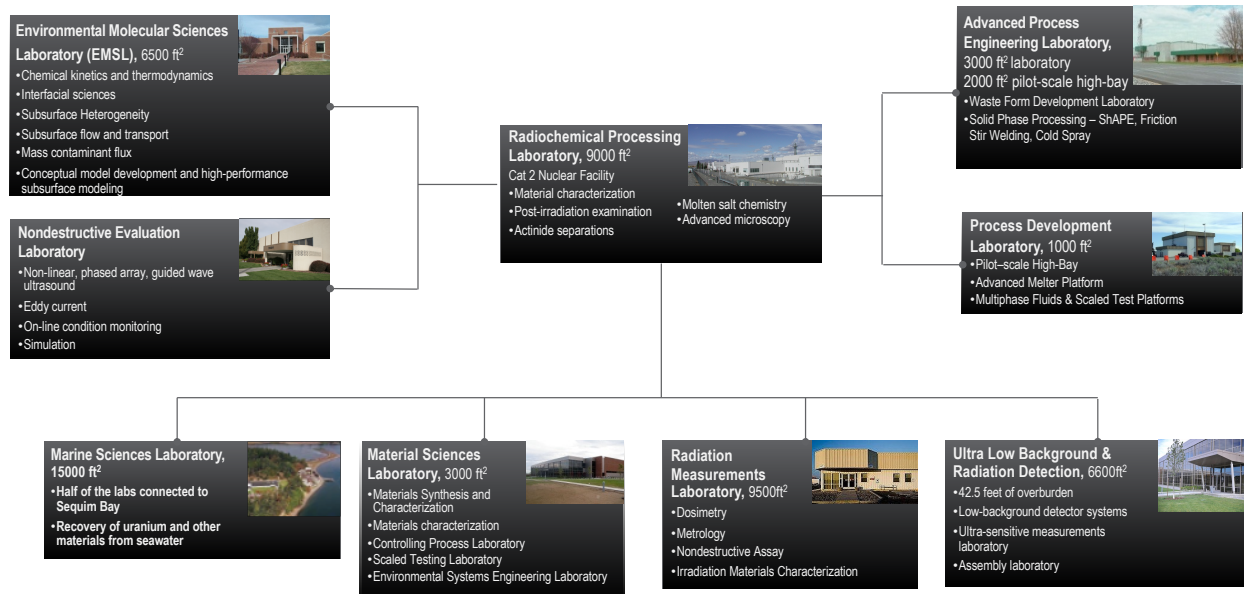


32



5

PNNL Expertise and Capabilities for Molten Salt Reactor Research and Development



PNNL Nuclear Energy Research and Development Laboratories

2.0 Molten Salt Physical, Chemical, and Thermodynamic Properties

During the operation of a molten salt reactor, composition of the fuel salt will continuously vary as fission, activation, and corrosion products are introduced. Coolant salts will undergo similar changes due to the accumulation of activation and corrosion products. This will result in evolution of the thermodynamic properties of fuel and coolant salts, including melting point, heat capacity, density, viscosity, and thermal diffusivity/conductivity. The operation of the reactor will be directly affected by these adjustments in salt properties. Additionally, if the concentration of a given species exceeds its solubility limit in the salt, it could aggregate and collect in regions of reduced flow within the reactor. Understanding these salt properties as a function of temperature and fuel burnup is key to advancing the design of many molten salt reactor concepts. Knowledge regarding chemical species and the oxidation state of actinides and transition metal elements in molten salts is limited, uranium being the nearly exclusively studied element. This problem is compounded by the fact that a variety of different fluoride- and chloride-based salt mixtures have been proposed for application in MSRs. PNNL has capabilities for in situ chemical speciation analysis of actinides and fission products required for a full thermodynamic description of the molten salt systems. We can also test thermophysical properties of salts up to 1000°C under inter gas atmospheres.

2.1 Safe Handling of Molten Salts

Our initial efforts to test thermal properties with commercially available crucibles resulted in the destruction of equipment due to the significant creep and corrosion exhibited by both chloride and fluoride salts in a molten state. Simply put, traditional high temperature crucibles that are available on the market are not enough to safely test molten salts at temperatures past their melting point. Since then, significant work at PNNL has gone into developing true hermetically sealed crucibles for thermophysical properties testing techniques such as differential scanning calorimetry (DSC), high temperature drop calorimetry (HTDC), and laser flash analysis (LFA).

This effort has led to successful tungsten inert gas (TIG) and laser welding approaches, which provide true hermetic seals that are resistant to corrosion and any internal vapor pressures developed due to volatilization or reaction.

We use laser welded silver crucibles for DSC and HTDC. Currently, silver and stainless-steel crucibles have been successfully developed with maximum use temperatures of 900 and 1450°C, respectively. Ongoing work is being performed to develop molybdenum crucibles with maximum use temperatures in excess of 2000°C. In addition, alternate form factor crucibles are currently being developed for LFA in order to measure thermal diffusivity up to 1000°C with molten materials.

Custom Boron Nitride (BN) and Al_2O_3 crucibles have also been developed for use in a thermal mechanical analyzer (TMA) in order to measure thermal expansion coefficients and calculate densities.

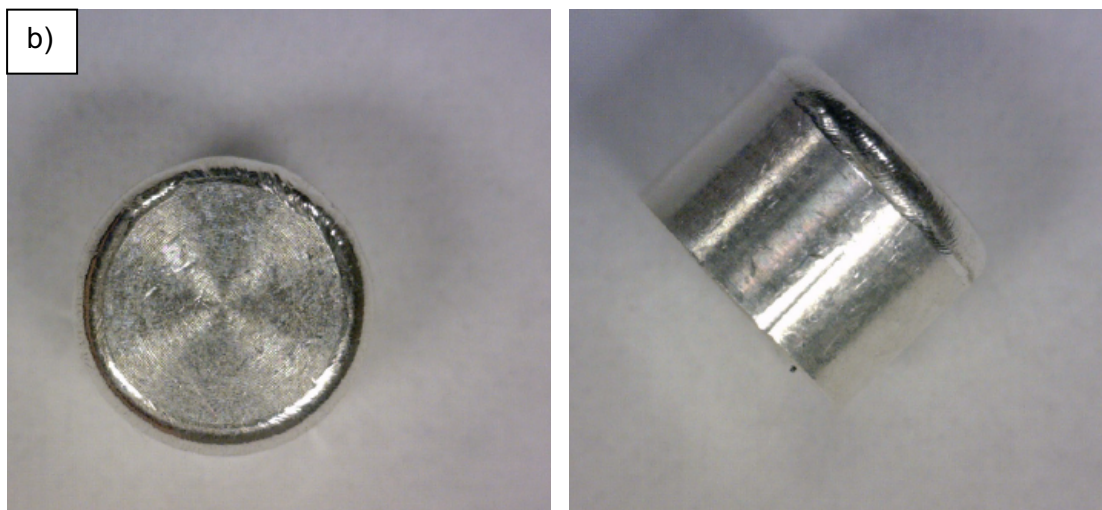


Figure 2.1. a) Laser Star welder capable of hermetically sealing metal crucibles. B) Precession of welds on a 6mm diameter silver crucible.



Figure 2.2. Al_2O_3 and BN crucibles for testing thermal expansion and calculating density with a TMA.

2.2 Thermal Expansion and Density

PNNL uses TMA to measure the linear expansion of the salts with temperature. This can be used to calculate all densities from 25 to 1500°C after an accurate density measurement is made at a single temperature. PNNL is currently utilizing gas pycnometry in order to make initial room temperature density measurements. Data has been collected on NaCl-KCl salts using an Al_2O_3 crucible in the TMA from 25–900°C, coupled with pycnometry data at room temperature, to determine density across the binary.

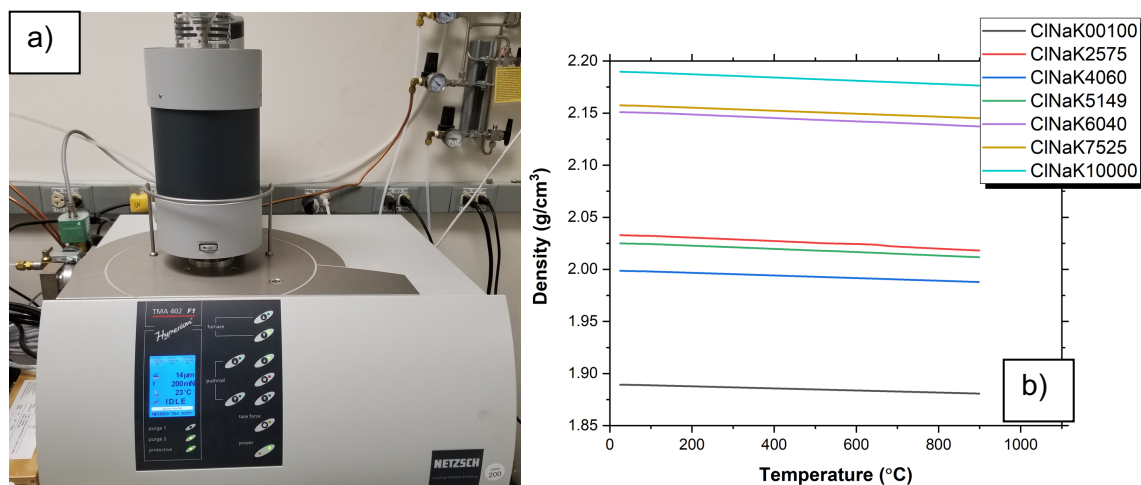


Figure 2.3. a) Non-rad Netzsch TMA located at PNNL capable of measuring thermal expansion up to 1500°C and b) density data calculated on the NaCl-KCl binary using the TMA and pycnometer.

2.3 Chemistry of Molten Salt Using X-ray Absorption Spectroscopy

To evaluate the chemistry of actinides, lanthanides, and transition metal fission products in molten salt media, PNNL has installed a laboratory instrument for in situ measurements with X-ray absorption spectroscopy (XAS) that probes the local coordination environment and chemical bonding of the interrogated atom; there is no requirement for the surrounding atoms to be formally bonded to the absorbing atom. It is a bulk technique that generates signal averaging of all chemical environments of the analyte present in a sample, and data interpretation relies on the availability of reference compounds. X-ray absorption near edge structure spectroscopy (XANES) provides information on the oxidation state and coordination symmetry of the absorbing atom. X-ray absorption fine structure spectroscopy EasyXAFS (EXAFS) provides information on the coordination number, formation of molecular complexes, and structural order at short and intermediate distances. This information is paramount to gain essential chemical understanding of the molten salts, but to date, the measurements have been limited by the difficulties of performing in situ molten salt studies with radioactive materials at large synchrotron facilities as well as their availability. Review of the available to-date reports indicates that in situ molten salt XAS investigations are extremely hard to realize experimentally and are therefore scarce.

To address this gap, PNNL has installed an EasyXAFS 300 instrument in its RPL. The EasyXAFS300 instrument is designed for the purpose of X-ray absorption spectroscopic measurements with the energy ranges suitable for transition metals, lanthanides, and some actinides (5-19 keV). The instrument operates in the transition mode, and it is equipped with interchangeable molybdenum and tungsten anode X-ray tubes, silicon drift detector (SDD), spherically bent crystal analyzers, i.e., Ge 111, Ge 620, Si 551, and Si 211. A helium-filled flight path box is positioned on the way of X-ray flux to minimize X-ray absorption by air. The collected spectra of the standards (iron and nickel foils) confirmed excellent reproducibility of the XANES and EXAFS spectra with the reference spectral database obtained at the Advanced Photon Source (APS) synchrotron. Similarly, EXAFS data for the series of lanthanide chlorides at room temperature are in excellent agreement with the literature data. Currently, PNNL, in collaboration with the University of Washington, is finalizing development of the sample holder/micro furnace for the molten salt in situ measurements in operando up to 1200°C.

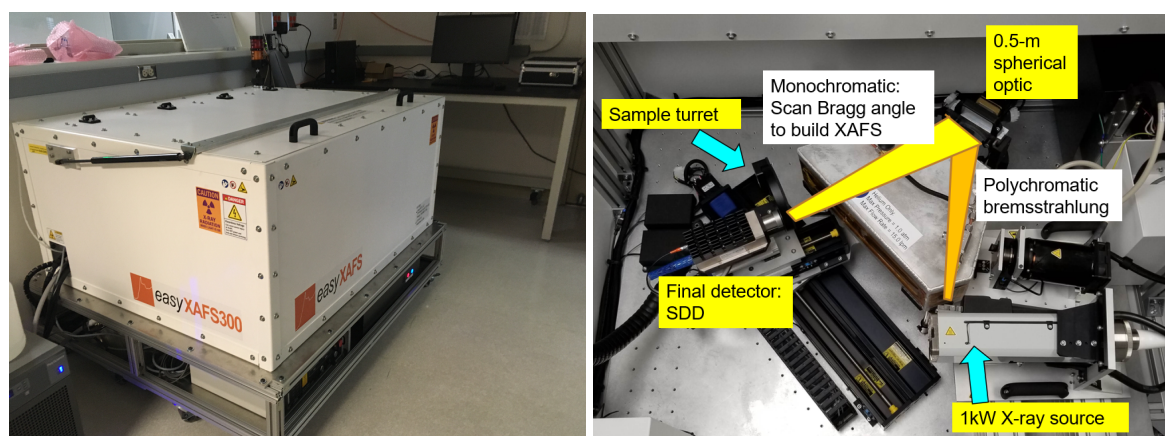


Figure 2.4. EasyXAFS 300 for in situ molten salt measurements at PNNL

2.4 Thermal Properties & Thermodynamics of Chloride and Fluoride Molten Salts

Basic thermophysical properties that are critical for the design of an efficient and reliable heat exchange fluid are unknown or poorly understood for most salt systems that are being proposed for next generation molten salt reactors. Specifically, thermal conductivity is a property that is essential to model and predict the transfer of heat as elucidated by the Fourier Equation.

Equation 1

$$q = -k\nabla T$$

Where q is heat flow, k is thermal conductivity, and ∇T is a three-dimensional thermal gradient. Thermal conductivity is a property that is typically calculated based on the measurement of three sub properties, thermal diffusivity, heat capacity, and density, as shown in Equation 2.

Equation 2

$$k = \alpha\rho C_p$$

Where α is thermal diffusivity, ρ is density, and C_p is heat capacity. PNNL has developed or is adapting several methods to measure the thermal properties that are required to calculate thermal conductivity. In addition, other fundamental thermal and thermodynamic properties, such as melting and volatilization temperature, enthalpies of heat, fusion, and dissolution, are being developed for both fluoride and chloride salts.

The combination of diffusivity, density, and heat capacity measurements from 25 to 1000°C gives PNNL the ability to calculate thermal conductivity, a value essential to the design of a heat transfer system and almost nonexistent in the literature. In addition, phase diagrams of novel salts can be determined.

2.4.1 Drop Calorimetry

The fundamental thermodynamic properties of the molten salt media can be determined by solution drop calorimetry using PNNL's high-temperature twin Calvet AlexSYS calorimeter. It allows the measurement of enthalpies of formation of 5–10 mg materials that are typically hard to dissolve for solution calorimetry (e.g., many oxides, minerals) by dropping them into a molten oxide solvent at 700–800°C. High-temperature oxide melt solution calorimetry has already been applied to the thermochemistry of a number of oxides containing thorium and uranium. We are currently adapting the AlexSYS with suitable sample containment to study the complex actinide/alkali halide chemistry of relevance to MSR at temperatures up to 1200°C. The reaction vessel is a platinum crucible containing 5–10 ml of salt (chloride or fluoride), and a sample is supplied as a 3–10 mg pellet containing the analyte of interest. Sparging with Ag provides inert atmosphere, and a specialty gas can be delivered to investigate chemical reactivity and/or corrosion.

Our calorimeter is located in the RPL and is equipped with inert gloved enclosures that can handle radioactive materials. This gives us the ability to store, batch, and seal sample crucibles without oxygen or water contamination. Currently, we are developing high-purity calibration

standards of known pedigree for thermodynamic measurements, including the following modalities:

- Enthalpy of dissolution
 - effect of the increasing analyte concentration
 - solubility/precipitation
- Specific heat capacity C_p
 - At fixed temperature
 - Dependence of C_p on temperature
- Temperature and enthalpy of phase transition
- Enthalpy of chemical reaction in the molten salt
 - Reactivity of the molten salt matrix toward fission products
 - corrosive behavior of the molten salt
- Enthalpy of formation
 - Relative stability of complex ions formed in the melt.

Drop calorimeter measurements can be coupled with XAFS and other spectroscopic structural measurements to correlate thermal properties with redox and chemical speciation. Although our drop calorimetry system directly measures enthalpy, heat capacity may be obtained by testing at varying temperatures and utilizing equations derived from the first law of thermodynamics.

Equation 3

$$C_p = \left(\frac{dh}{dT} \right)_p$$

Where C_p is heat capacity, dh is the change in enthalpy, and dT is the change in temperature. Therefore, heat capacity can be determined by the difference in enthalpy between any two temperatures that a salt is tested at.

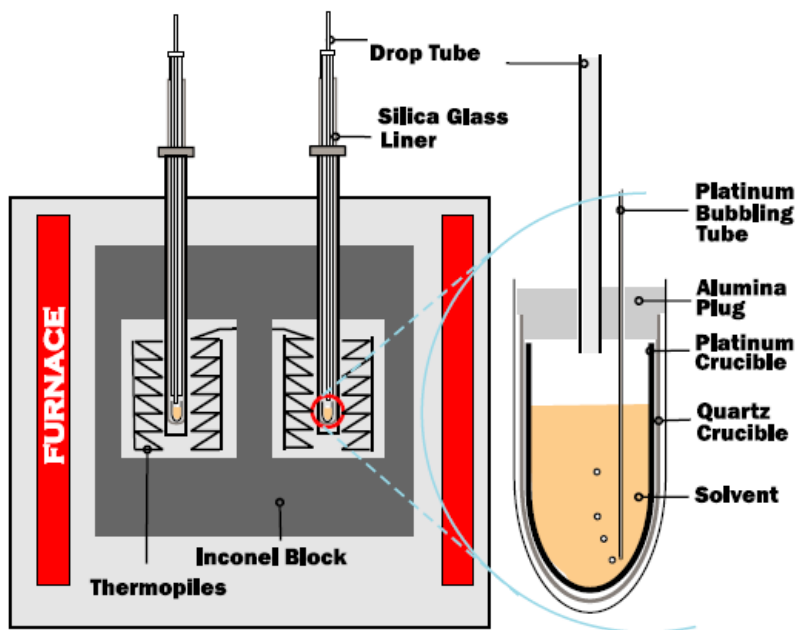


Figure 2.5. Schematic of high-temperature twin Calvet calorimeter and assembly for drop solution calorimetry in a molten oxide solvent

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2.4.2 Differential Scanning Calorimetry

Thermodynamic properties such as the heat of fusion (ΔH_f) and thermal properties such as specific heat (C_p) can be rapidly measured with high temperature differential scanning calorimetry. PNNL currently has temperature capabilities of 1500°C for non rad salts and 700°C for rad salts. Several DSCs are available at PNNL for molten salt testing.

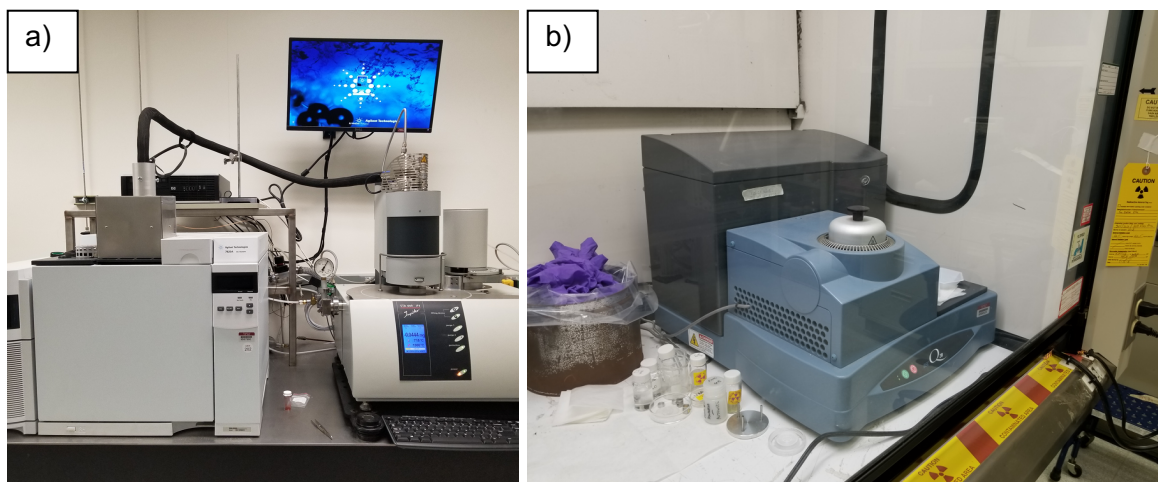


Figure 2.6. Netzsch DSC capable of testing non rad salts to 1500°C and b) TA DSC capable of testign rad salts to 700°C

PNNL has invested significantly in the development of capabilities so that hygroscopic salts can be purified, stored, and hermetically sealed in inert atmospheres. With this setup, PNNL can develop phase diagrams, determine energetics of reactions, and measure the heat capacity of both coolant and fuel salts. We have begun to collect data on Cl salts—mapping melting points and heat capacity across the NaCl-KCl binary.

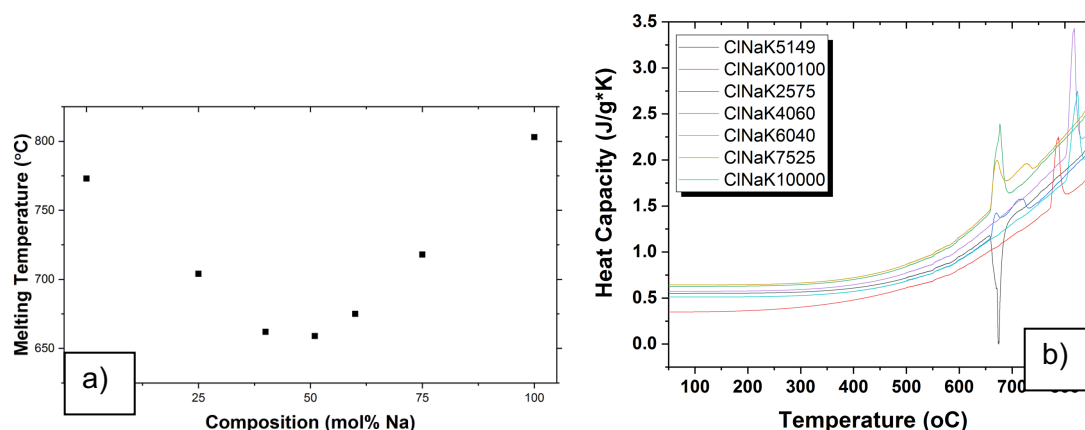


Figure 2.7. a) Beginning of a phase diagram map showing the melting temperatures of the NaCl-KCl binary and b) the heat capacity from 50°C to 900°C for the NaCl-KCl binary.

2.4.3 Laser Flash Analysis

PNNL is establishing LFA capabilities to test molten salts from 25 to 1000°C. Work is ongoing to develop custom metal liquid sample holders that can be laser welded together. In addition to directly measuring diffusivity, LFA can also calculate heat capacity utilizing a comparison method that provides a nice check for the heat capacity values generated through calorimetry.

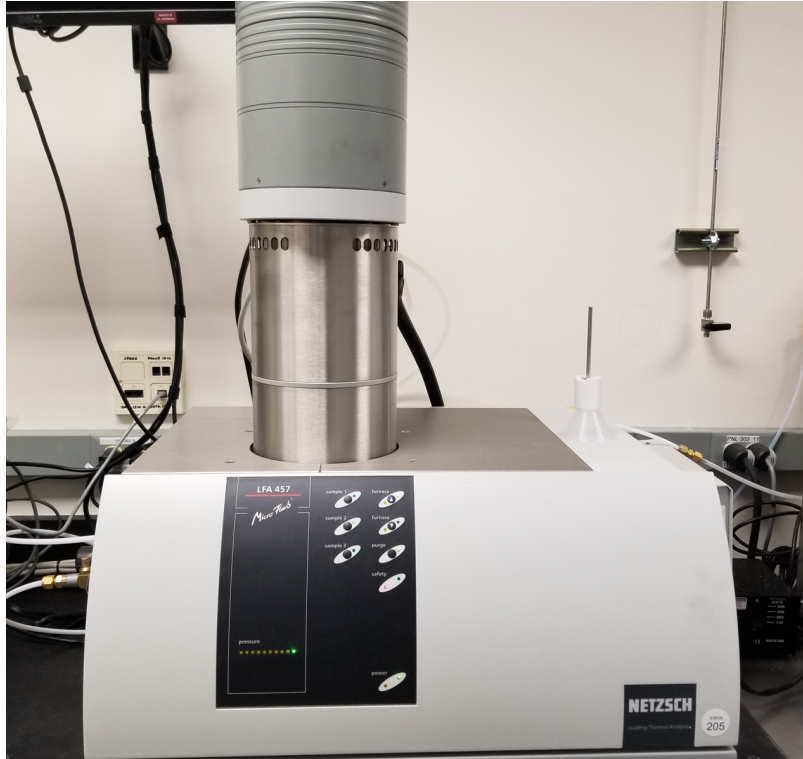


Figure 2.8. Netzsch LFA capable of testing thermal diffusivity from 25 to 1000°C

3.0 Materials Corrosion – Modeling and Performance

3.1 Corrosion Modeling

Materials corrosion has been recognized as an issue in structural materials for MSRs. PNNL has expertise in simulation of the interaction of materials with extreme environments from the density functional theory level to finite element methodology. The focus is on predicting the life of materials subjected to erosion and corrosion from flowing molten salt and on developing advanced alloys that can withstand the combined effects of elevated temperature, radiation, stress, and contact with molten salt.

In molten salts, passive oxide films are chemically unstable, and corrosion is initiated largely by the thermodynamically driven dissolution of alloying elements into the molten salts. Furthermore, the high energy particle irradiation (e.g., neutron and gamma rays) results in high defect concentrations and ionization, which affect mobility and the chemical driving force of defect migration (i.e., microstructural evolution and property degradation kinetics). To predict material performance, it is critical to understand the effect of the reactor environment on the thermodynamic and kinetic properties of alloying elements and the effect of as-fabricated microstructures and evolving microstructures on material property degradation. At PNNL, we have strong modeling capabilities (i.e., ab initio molecular dynamics, object kinetic Monte Carlo, and phase-field methods) for investigating the thermodynamic and kinetic properties of defects, defect accumulation, microstructure evolution, and property degradation from atomic scale to mesoscale modeling. For the application to MSR, these modeling capabilities could be integrated to study the mechanisms behind the corrosion processes and quantitatively predict

the effects of dislocations, grain boundaries, secondary phase inclusion, and radiation conditions on corrosion kinetics in various alloys.

3.2 Salt/Material Interactions and Corrosion

PNNL has a wide range of experience working on the corrosion of metals. For instance, PNNL has the capability to expose irradiated materials to salts at elevated temperatures in autoclaves. Structural materials can be exposed to irradiation during in situ electrochemical testing. Mechanical testing can be performed on the samples before and after exposure, and the materials can be fully characterized for corrosion products. PNNL has excellent facilities to study the effects of aggressive environments on mechanical properties of advanced materials, including environmental mechanical testing up to 1600°C, specimen preparation, advanced characterization, and computational modeling capabilities. Both non-radioactive and radioactive materials testing capabilities exist, including high-pressure autoclave testing.

The materials characterization capabilities include aberration-corrected scanning transmission electron microscopy and microanalysis, scanning electron microscopy, electron backscatter diffraction, 3D-Atom probe tomography to reveal chemical distributions to ~0.5 nm, and preparation of highly radioactive materials from core components or experimental test samples. The integration of these capabilities with multiscale materials modeling creates a unique capability to understand the combined effects of corrosion and radiation.

3.3 Corrosion References

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4.0 Fuel and Coolant Salt Processing

Designs for MSR come in many different forms, with most using either fluoride or chloride-based salts. PNNL is capable of synthesizing and handling a range of clean, fuel-bearing, and irradiated salts, including salts that contain both surrogate and actual fission products and transuranic elements.

4.1 Isotopic enrichment and separation of Cl-37

Designs for MSR come in many different forms, with most using either fluoride or chloride-based salts. Choosing salt components comes with many factors to consider; however, chlorine-37 (Cl-37) based eutectics may offer benefits over current alternatives. The major isotope of chlorine, chlorine-35 (Cl-35), has a strong (n, proton) threshold reaction that acts to poison the reactor and also generates chlorine-36 (Cl-36) as an activation product. Cl-36 is a long-lived energetic beta source that could impact waste management processes and disposition pathways. Cl-37 may be a preferable anionic salt in an MSR. However, there is currently no purification strategy for separating Cl-37 from the more abundant Cl-35 that would be needed for MSRs.

Literature suggests that Cl-37 can be separated by capillary electrophoresis. The electrophoretic mobility difference between Cl-37 and Cl-35 is potentially large enough to allow for isotopic separation using electrophoretic methods. The element itself has also been separated from other anionic elements by a technique known as isotachopheresis. The advantage of isotachopheresis over capillary electrophoresis is that the electric field can be balanced by a counterflow in the opposite direction, a technique known as stationary isotachopheresis, so that the isotachopheresis stack can be maintained for an indefinite period of time to achieve the desired separation.

Numerical simulation using the finite element solver COMSOL Multiphysics®, under optimal conditions, indicates that separation is achievable, and PNNL is developing the technology to test and demonstrate this approach.

4.2 Producing fluoride salt fuels from irradiated fuels

Used oxide fuels from light water reactors can be converted to fluoride salt fuels through the process of voloxidation, followed by flame fluorination. This process aims to first fully oxidize the fuel, which opens up the structure, leading to a large increase in surface area while releasing gases trapped in the fuel matrix. Subsequently, the entire fuel is fluorinated in a single aggressive step in which all volatile fluorides are released. Alternatively, a less aggressive fluorinating agent can be used to perform this conversion, allowing for a selective separations process in which a series of temperature soaks are used to control the conversion and release fluorides that are formed and become volatile at sequentially higher temperatures. In particular,

the fluorinating agent NF_3 has shown promise in this selective volatilization process. The NF_3 fluorination through successive temperature soaks can be performed as a stand-alone process or subsequent to a voloxidation step. Selective fluorination and separation can also be achieved by treating metal fuels with NF_3 . Experiments characterizing these fluorination reactions are typically conducted through the combined use of thermogravimetric analysis, differential thermal analysis, and mass spectrometry (TG/DTA/MS). Standing capability for these experiments exists in fume hoods and glove boxes and has previously been set up and conducted in hot cells.

4.3 Removing fission products from salt mixtures

Volatile fluoride separations can also be used to remove components of molten salt mixtures. Fluorinating gases are sparged through the molten fluoride salt and volatile fluorides that are formed are removed. For instance, uranium existing as UF_4 in a molten fluoride salt can be converted to volatile UF_6 , which then leaves the salt. When excess fluorine is added to the molten salt mixture at elevated temperatures, the result can be a very aggressive fluoride attack on the vessel holding the salt. This attack can again be controlled by the use of less aggressive fluorinating agents, such as NF_3 . PNNL is currently developing the capability to make fluoride salts and separate components through volatile fluoride separation in collaborative work with Flibe Energy, Inc.

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5.0 Instrumentation, Control, and Online Monitoring

PNNL has been at the forefront of research, development, and deployment (RD&D) in the area of advanced sensors; instrumentation and control for monitoring processes; and systems, structures, and components (SSC) in light water reactors, small modular reactors, and advanced reactors. Activities in this area have spanned concept development to functional testing and technology transfer and have been supported by a number of sponsoring organizations, including Department of Energy (DOE) Offices of Nuclear Energy (NE) and U.S. Nuclear Regulatory Commission (NRC). Several research needs documents were developed by PNNL (either alone or in collaboration with other laboratories) as a result of the activities supporting advances in nuclear power, and RD&D efforts have resulted in novel sensors, instrumentation, control, and deployment approaches. PNNL's recent key experience relevant to sensors, instrumentation, and control include:

- A joint advanced sensor needs assessment with Oak Ridge National Laboratory (ORNL) and Argonne National Laboratory (ANL) for advanced reactors, including high temperature reactor and fast reactor concepts. This assessment was performed by leveraging information and operational experience from previously operated advanced reactors, such as the Fast Flux Test Facility (FFTF) and the Enhanced Breeder Reactor (EBR-I and EBR-II), the Molten Salt Reactor Experiment (MSRE), Ft. St. Vrain High Temperature Gas Reactor, and numerous other research and commercial advanced reactors in the U.S. and worldwide. A number of R&D needs were identified and covered measurements ranging from neutron and gamma probes, to temperature and flow sensors, to component condition monitoring systems.
- This initial assessment has been continued through a PNNL hosted sensors focused web page (<https://nes.energy.gov/>) to capture needs, commercial offerings, and nascent laboratory/university technologies under development.
- With the restart of the Transient Reactor Test (TREAT) facility, there is interest in developing the ability to obtain as much data as possible from individual transient tests. In particular, one measurement of interest is the change in the diameter of a fuel rod as it experiences a rapid reactivity insertion transient. PNNL is collaborating with Idaho National Laboratory (INL) and ORNL to develop sensors that can provide this measurement. The approach is to develop an arrangement of ultrasonic sensors that can fit within a TREAT sample test holder and that are robust to thermal cycling. This effort involves identifying sensor components such as transducer elements, backing, face plate, casing, bonding agents and cabling; all with the desired thermal, electrical, and bonding properties to allow useful measurements to be transmitted for the longest time possible during a transient test.
- A new immersion ultrasonic phased array sensor and associated instrumentation for under-sodium viewing in sodium fast reactors. This capability, originally developed at the Hanford Engineering Development Laboratory in the 1970s, was resurrected and updated to incorporate recent advances in ultrasonic sensors. The research has resulted in a technology transfer package for this sensor system that is currently in licensing discussions with commercial vendors. Similar technologies have been applied for a number of other applications, including NDE of SSCs in the current nuclear power fleet, and proposed for NDE/condition monitoring of SSCs of components in advanced reactors. In particular, such techniques are useful for monitoring hard-to-access MSR components for corrosion and cracking.
- A state-of-technology assessment of optical technologies for monitoring plant parameters in advanced reactors. The assessment covered both optical materials (for windows and optical waveguides) as well as technologies for optical measurements and data interpretation.
- Sensors for monitoring fluid flow, fluid temperature, and fluid chemistry. Developments in this area have focused generally on challenging environments (high temperatures, pressure extremes, two-phase fluids and slurries with radiological materials). Capabilities in multiphysics modeling along with laboratory-scale testbeds have been applied to design, fabricate, and evaluate these sensor and instrumentation systems, with potential applications including monitoring of MSR coolants for corrosion byproducts.
- Methods for reliability assessment of sensors, instrumentation, and associated measurement techniques. This includes methods for monitoring the condition of sensors and instrumentation when operated over extended time periods. Nondestructive techniques for evaluating instrumentation and power cabling have been developed and assessed at PNNL, and PNNL is participating in a collaborative research program between DOE, NRC,

and industry to address open questions in cable integrity assessment over extended operation periods. PNNL has also developed algorithms (includes uncertainty quantification) for the online monitoring of instrument calibration to provide early warning of sensor failure while avoiding unnecessary recalibration.

- Research in radiation resistant optical materials and corrosion resistant protective coatings. Novel non-oxide ionic bonding chalcogenide glass materials were developed and underwent gamma irradiation tests, showing radiation resistant properties. Continuing studies on neutron irradiation tests were proposed. Protective coatings such as aluminum nitride and nickel-based alloys deposited on commercial optical materials were studied for corrosion resistance in molten salts.
- Sensors and algorithms for prognostic health monitoring of components and predictive risk-informed methods for predicting plant conditions to enable autonomous control of advanced reactors. Technologies in this area were developed to enable proactive control of advanced reactors in response to changing plant and component conditions over time while maintaining the safety and economics of the operation (avoid unnecessary repair and maintenance actions).

5.1 Spectroscopic-Based Process Monitoring Summary at PNNL

PNNL is advancing the use of spectroscopic-based (e.g., ultraviolet-visible absorbance and Raman) process monitoring related to the nuclear fuel cycle, including applications in fuel reprocessing and nuclear safeguards. This work has been primarily funded through DOE-NE (i.e., GNEP, AFCI, FCRD; 2007 to present) and experience spans the spectroscopic-based measurement of actinide and lanthanide metals, as well as inorganic and organic molecules within the PUREX and TALSPEAK aqueous extraction systems. The quantitative measurement of U, Pu, Np, and lanthanide metals has been demonstrated on actual used commercial fuel samples and on numerous simulated fuel reprocessing solutions. Quantitative measurement of HNO_3 concentration and pH monitoring has also been demonstrated. PNNL is building upon these capabilities to apply them to miniaturized or microfluidic systems, which can reduce both the dose to workers and the material quantities required to make quantitative measurements.

This methodology has also been extended to the molten salt environment (i.e., DOE-NE work; 2010–2013). Spectroelectrochemical methods, a combination of spectroscopic and electrochemical techniques, were effectively used to characterize lanthanide metals under pyroprocessing conditions. Target mixtures were comprised of various molten salt eutectics, including LiCl/KCl, LiCl/NaCl, LiCl/RbCl, and LiCl/CsCl systems. Accurate quantification of lanthanides was demonstrated in complex melts containing up to six lanthanides at different concentration ratios. PNNL has been working to apply these techniques to actinides and corrosion products within salt melts as well as extending capabilities into MSR off-gas treatment systems.

The application of spectroelectrochemical methods as on-line, in-situ monitoring techniques for bulk solution analysis within a molten salt media allow for:

- Simultaneous quantitative measurements of multiple metal species within the molten salt
 - Enables process and corrosion control
 - Potential to support materials accountancy
 - Supports informed process design during R&D phases

- Provides unique insight into the chemistry of salt systems for more fundamental characterization of system behavior
 - Calculation of diffusion coefficients of electroactive metal species
 - Determination of redox potentials for metal species.

On-line monitoring provides an in situ and real-time approach to characterize chemical systems. This information often cannot be obtained through traditional grab sample analysis. Furthermore, the optical and spectroelectrochemical methods developed at PNNL provide complementary information to other forms of monitoring that enable unprecedented understanding and control of chemical processes. The techniques being developed at PNNL allow for robust characterization of chemical processes across a wide range of scale (commercial to microfluidic) and in a variety of process matrices (including aqueous, molten salt, and gaseous systems).

5.2 On-Line Structural Health Monitoring of Critical Reactor Components

Advanced MSR design concepts envision operating for extended periods of time without having to enter outages for refueling or maintenance. Monitoring the structural health of systems, structures, and components important to safety and to the efficient operation of the plants will be necessary. As an example, heat exchangers for both light water reactors and sodium fast reactors have historically been the sites for the highest incidents of cracks and leaks of reactor coolant. The long-term performance of heat exchangers is critical for the safe operation and economics of MSRs.

PNNL has an initiative to develop high temperature sensors for on-line monitoring of these components. Building on work performed to monitor fuel rods in the transient reactor test facility (Ramuhalli et.al. 2018), tube outer diameter (OD) sensors have been demonstrated to detect 50% through wall crack-like and pit-like flaws more than 10 ft. away from the sensor. This work is being applied to demonstrate feasibility of an on-line monitoring system for both salt- and lead-cooled reactors (Glass et.al. 2020).

PNNL also has expertise in the use of guided wave Electromagnetic Acoustic Transducers (EMAT). The design of a guided wave EMAT sensor based on the principles of magnetorestriction could allow for the detection of large flaws or geometric changes (e.g., several % creep), from a distance (perhaps as much as 5m). Designing such a system would build on PNNL's experience with waste tank inspections and power harvesting ultrasonic modems. Our expertise could be used to select technologies (sensors, power harvesting, piezoelectric transducer driving circuitry, data transmittal circuitry), validate component performance at 100°C and testing in simulated operational environments, and down selecting and delivering a system that would operate in an MSR environment.

Building on PNNL's experience in ultrasonic power harvesting and communication devices, our engineers and scientists could also develop an approach to instrumenting reactor machinery using thermal energy harvesting, ultrasonic power transfer and communication, and energy storage methods suitable for harsh environments. Power and data would be transmitted ultrasonically between sensors separated by barriers where penetrations are undesirable, or where conduit runs are impractical. Conventional approaches to energy storage fail above 85°C and alternative energy storage approaches will need to be explored and developed.

The development of laser ultrasonic testing, a non-contact method, for sizing of flaws in advanced reactors structural components is another on-line structural health monitoring technique that could be used for MSR components. PNNL's experts in non-destructive evaluation technologies and sensor design could advance the design and application of this method.

5.3 In-vessel MSR/Fluoride Salt-Cooled High-Temperature Reactor (FHR) Monitoring Requirements, Challenges, and Opportunities

5.3.1 Coolant & Cover Gas Chemistry Monitoring

Coolant monitoring and purification is generally conducted using process loops external to the reactor vessel; however, in-vessel monitoring could be required for future MSR concepts. Impurity analysis is conducted to monitor corrosion indicators and detect component or fuel failures. Regular coolant chemistry monitoring is required because of the corrosive nature of the salt coolants. Although the chemical behavior of salt changes slowly, continuous in situ measurement techniques will likely be required. Currently, commercially available monitoring instrumentation is unavailable for detailed monitoring of fluoride salt coolants. The standard technique for monitoring the redox condition of salt components is based on electrochemical methods. Absorption spectroscopy may be a potentially useful methodology for identifying trace chemical constituents and their valence states. It may be possible to measure Bi and Cs concentrations, impurities, and corrosion byproducts (e.g., Fe, Ni, Cr) directly in the molten coolant using laser-induced breakdown spectroscopy (LIBS). The species of H₂O, CO₂, CO, CH₄, and O₂ can be monitored in the cover gas area using optical spectroscopy methods. Cesium deposits could be detected using LIBS. Fission gases and fuel pin taggant gases are noble gases without strong optical absorption spectra at usable wavelengths. Optical emission spectroscopy may be effective at detecting noble gases and ³H either by active ionization (e.g., laser, spark gap) or possibly radiation-induced autoionization. Further study will be required to determine if optical detection and monitoring is feasible.

5.3.2 Electromagnetic Pump Monitoring

Degradation and performance are determined by monitoring vibration, temperature, voltage, and current. Vibration monitoring is used to detect impending failure. Commercially available accelerometers may not be feasible to use in high-temperature advanced molten-salt coolants. Optical vibrometry techniques may be feasible on components above the coolant level.

5.3.3 Flow Blockage Detection

Detecting flow blockage of the coolant through the core is a challenging problem. Imaging and processing methods could be developed to monitor changes in coolant surface ripples, which may provide information about changes in coolant flow properties. Thermal infrared spatial temperature profiling in molten salt coolants also may be feasible to infer coolant flow blockage. The camera challenges and opportunities are the same as in-vessel viewing/inspection.

5.3.4 Coolant Flow and Pressure

Thermal infrared spatial temperature profiling in molten salt coolants may be used to infer coolant flow. Laser Doppler velocimetry is used to measure flow in transparent liquids, but this technique requires particles to scatter laser light back to the receiver. This technique may have some potential in molten salt coolants containing dissolved fuel. Reflective spinning rotor

techniques with optical readouts may also be feasible for transparent coolants. Pressure measurements may be made using strain gauge instrumented diaphragm transducers connected to NaK impulse lines. Strain gauges have well-known and undesired drift. Optical Bragg grating sensors are commonly used as a direct replacement for electronic strain gauges.

5.3.5 Coolant Level

In-vessel coolant levels can be measured using ultrasonic transducers and other conventional techniques. It is unclear whether optical-based monitoring techniques have a decided advantage over current methods. If required, noncontact laser ranging, or other optical methods may be feasible. Further study will be required to determine the benefits and performance of each optical monitoring approach.

5.3.6 Neutron Flux/Power

Cherenkov radiation monitoring may be very feasible in MSR or FHR coolants.

5.4 Instrumentation, Control, and Online Monitoring References

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6.0 Reactor Design and Safety Analysis

PNNL's reactor design and safety analysis are well positioned to support the design and safety analysis for advanced MSRs. Our capabilities in this area include:

- Reactor Core Design and Analyses
- Nuclear Fuel and Materials Performance
- Criticality Safety Analyses
- Occupational/Environmental Health Physics, Safety, and Risk Assessment
- Reactor Dosimetry
- Risk-Based Decision Making and Management, Systems Engineering, and Analysis
- Deterministic and Probabilistic Safety Analysis
- Nuclear Plant Fire Protection
- Emergency Management
- Nuclear Plant Aging and Maintenance Analysis

- Nuclear Facility Maintenance
- Safeguards and Security Inventory and Accountability Systems
- Fate, Pathway, and Exposure Modeling
- Human Factors and Human-Machine Interface
- Cybersecurity

PNNL has experience in the design and safety analysis of nuclear systems, much of it gained in the commercial nuclear industry. Recent advances in non-light water reactor (non-LWR) technologies will pose novel challenges in reactor safety reviews under DOE's Documented Safety Analysis process and under NRC's NUREG-0800, *Standard Review Plan for the Review of Safety Analysis Reports for Nuclear Power Plants*. PNNL's experienced staff are uniquely qualified to understand these technical innovations and can apply our core nuclear capabilities in the following key technical disciplines:

6.1.1 Safety Analysis

PNNL has extensive knowledge of NRC regulations and performing reviews in accordance with NUREG-800. This includes reviewing portions of construction permit (CP) or operating license (OL) applications under 10 CFR Part 50 and early site permit (ESP), design certification (DC), combined license (COL), standard design approval (SDA), or manufacturing license (ML) applications under 10 CFR Part 52. Additionally, PNNL has reviewed numerous Final Safety Analysis Report chapters and underlying methodology documentation for operating and DC applications, including documentation associated with fuel and thermal-hydraulic design, neutronics design, and nuclear safety analysis.

6.1.2 Quality Assurance

If required, PNNL's NQA-1-compliant Nuclear Quality Assurance Program (NQAP) ensures that the work performed by trained and qualified staff members and documentation is complete, correct, practical, satisfy the applicable requirements, and include the appropriate quality control (QC) requirements.

6.1.3 Reactor Chemistry

PNNL stewards radiochemistry capabilities and facilities focused on designing materials, sensors, and systems that will perform in the extreme environments expected in the next generation of reactors. For example, we are developing approaches to controlling materials corrosion in molten salt reactors by developing real-time spectroscopic sensing tools. Combining spectroscopic and electrochemical methods to probe speciation and redox states of uranium and fission products in a molten salt reactor will enable the monitoring and control of materials corrosion. This work is conducted in laboratories and test beds at PNNL's RPL, a Category 2 nuclear facility housing hot cells and glove boxes to support bench- and pilot-scale testing of chemical systems.

6.1.4 Reactor Physics, Neutronics, and Nuclear Design

PNNL's expert staff are familiar with the nuclear design of the fuel assemblies, control systems, and advanced reactor cores. PNNL's capabilities in these areas include expertise in criticality safety, shielding, fuel system design, thermal hydraulics, and reactor internal and core support

structural materials. Our staff is proficient with a number of nuclear design codes, including the SCALE simulation suite; the Monte Carlo tools MCNP, Serpent, and Keno; the fuel performance codes FRAPCON/FRAPTRAN and FAST; and Westinghouse and Studsvik core design software.

6.1.5 Mechanical Engineering

PNNL's expert staff familiarity with the design and engineering of structures, components, and equipment is directly applicable to non-LWR designs. These capabilities include application of the American Society of Mechanical Engineers (ASME) codes to non-LWR plant components, seismic qualification and wind loadings, design of containment structures and foundations, fracture mechanics, leak-before-break, fitness for service, seismic fragility analysis, and structural mechanics.

6.1.6 Component Integrity

Our materials scientists are improving our understanding of the mechanisms by which materials degrade, thereby helping ensure that the life of our existing fleet can be safely extended. This research also leads to advanced alloys and materials for use in the harsh environment of the next generation of reactors. PNNL has expert staff familiar with the unique reactor coolant systems anticipated in non-LWR designs, including molten salt, high-temperature gas, and liquid metal. PNNL's capabilities in these areas include the evaluation and testing of reactor vessel materials and coolant system components, the application of applicable ASME codes and standards, and the evaluation of steam generator and other primary and secondary system components. PNNL has expertise in the simulation of the material interactions in extreme environments from the density functional theory level to finite element methodology.

6.1.7 Probabilistic Risk Assessment (PRA)/Severe Accidents

PNNL's institutional expertise draws on practitioners of PRA, licensing application reviews, systems reliability analysis, severe accident modeling, offsite consequence analysis, human factors, uncertainty analysis, and plant safeguards. PNNL has been highly engaged in supporting the Nuclear Regulatory Commission in the establishment of the process for License Amendment Requests (LARs) for conversion to risk-informed, performance-based compliance programs. We developed safety evaluation templates for risk-informed programs and led efforts to assess LARs for fire protection programs, technical specifications, and seismic qualification.

6.1.8 Radiation Protection and Dose Assessment

PNNL's expert staff is familiar with evaluating radiation sources during normal operations and accident conditions in a nuclear power plant. PNNL has developed and maintains several nationally and internationally known health physics codes used for radiological environmental monitoring and emergency response, including the RASCAL Emergency Response Code and the GENII Environmental Dosimetry System. PNNL staff have decades of experience in the field of health physics and the use of health physics codes in the expert assessment of difficult radiological protection issues for the Nuclear Regulatory Commission, the Department of Energy, and other national and international agencies. PNNL led the development of Chapter 12 of the Design-Specific Review Standard for the mPower™ design.

6.1.9 Human Factors Engineering

PNNL supports the NRC in addressing issues related to human performance, human safety, human reliability, and learning and training. PNNL researchers have developed methods to explore the effects of human factors on training and qualification on manual ultrasonic NDE in the nuclear industry, provided ongoing support in human reliability analysis to support safe and reliable nuclear operations, examined environmental factors' impact on operator action in emergency response at nuclear power plants, and leveraged the latest scientific discoveries in human health and performance to inform nuclear regulatory analysis. In addition, PNNL's growing capabilities in human factors enable more expansive technical support in human-machine teaming and cognitive systems engineering that could help modernize operations and decision making in the nuclear domain.

6.1.10 Site Characteristics and Site Parameters

PNNL's expert staff in meteorology, geology, and hydrology apply their capabilities to the unique evaluations of site parameters required in a non-LWR design certification. PNNL is the lead laboratory assisting NRC with the development of the Advanced Reactor Generic Environmental Impact Statement. PNNL assisted NRC with the development of COL/ESP-ISG-027 "Specific Environmental Guidance for Light Water Small Modular Reactor Reviews." PNNL led the development of Chapter 2 of the Design-Specific Review Standard for the mPower™ and NuScale Small Modular Reactor Designs, and PNNL recently led the Environmental Impact Statement for siting an SMR at the Clinch River site in Tennessee.

6.1.11 Operating Reactor Experience Base

For more than two decades, PNNL has stewarded the design information, operating experience, and safety data from the FFTF to support industry and DOE-NE in resolving core and plant performance questions for new reactor concepts. This includes expert retrieval of FFTF information and analysis of metal fuel experiments performed in FFTF so that the results are applicable to—and can be interpreted in terms of—designs being developed by advanced reactor vendors.

PNNL has retained senior staff who worked on FFTF during its design and operation who bring rare, firsthand experience. PNNL has also retained senior staff who have first-hand experience operating Fort Saint Vrain, one of two high-temperature gas-cooled commercial power reactors in the United States.

6.1.12 Fire Protection

PNNL supports the NRC and DOE in developing or reviewing risk-informed and deterministic methods to protect nuclear power plants and nuclear facilities from the consequences of a fire. The support for fire protection programs at nuclear power plants focus on the protection of equipment and systems required to maintain the reactor fuel in a safe and stable condition (during operation, shutdown, and non-power operating conditions). The fire protection programs for DOE nuclear facilities focus on minimizing fire losses to the facility and protecting equipment and systems needed to mitigate the release of radiological consequences to the facility worker and the public. The fire protection methods incorporate National Fire Protection Association (NFPA) codes and standards and other guidance documents developed for the nuclear industry. Risk-informed performance-based methods used in fire protection programs for nuclear power plants and nuclear facilities incorporate a plant's PRA or fire models to determine the required

fire protection systems and features to protect specific equipment and systems that are needed to maintain nuclear safety and mitigate radiological consequences.

6.1.13 Cyber Security

PNNL supports DOE, Department of Defense (DoD), and the NRC in developing risk-based cybersecurity programs to protect energy generating facilities (i.e., nuclear power plants, photovoltaic solar arrays, wind power, marine renewable energy, hydropower plants) from a potential cyberattack. The development of cybersecurity programs utilize industry developed risk management frameworks to identify security controls to protect digital assets, processes, and operators needed to maintain safe operation of the facilities, to implement safe shutdown of nuclear reactors, and to support emergency response plans. Risk-based methods incorporate threat modeling, vulnerability assessments, and consequences analysis to determine the appropriate security controls to protect assets from a cyberattack.

Our team represents a rare combination of expertise in cybersecurity and nuclear power plant operations, including policy development, innovative research, assessments and inspections, and capacity development. Our experience includes:

- Planning and supporting NRC's cybersecurity inspections of all licensee facilities
- Developing and teaching computer security training programs for the International Atomic Energy Agency (IAEA) on topics such as computer security awareness, security assessments, computer security for industrial control systems, security for nuclear and radiological materials facilities, and web-based eLearning
- Co-authoring the NRC inspection manual Inspection for the Cyber Security Program, drafting Reg Guide Cyber Security for Nuclear Power Plants and Authoring Cyber Security Self-Assessment Method for U.S. Nuclear Power Plants (NUREG/CR-6847)
- Developing the PACRAT software tool for integrated vulnerability/risk assessment of coordinated cyber/physical attacks
- Developing an interactive virtual reality training tool for DOE/NNSA to use in training for computer security at nuclear facilities
- Creating accelerated innovation programs focused on cyber ops and intelligence, visualization tools, bio-inspired cybersecurity systems, mobile device security, and asymmetrically resilient cyber systems
- Developing computer security policy and guidance documents for the IAEA on topics such as computer security techniques for nuclear facilities, computer security threat assessment, information sharing and incident response, and computer security for physical protection systems.

6.1.14 External Hazards

6.1.14.1 Flood Hazard Assessments

PNNL has extensive flood hazard assessment capabilities. These capabilities include the full range of hydrologic (local, watershed, and regional scale rainfall/snowmelt modeling and simulation), hydraulic (one, two, and three-dimensional modeling and simulation), and hydrodynamic (nearshore and oceanic modeling and simulation) assessments. PNNL's researchers have developed and currently maintain several of these models that are employed

to solve a range of flood hazard related issues for critical infrastructure and environmental protection and management. PNNL staff support several federal agencies (e.g., DOE, DoD, Department of Homeland Security [DHS], NRC, Environmental Protection Agency [EPA], National Oceanic and Atmospheric Administration [NOAA]) in assessing flood hazards that affect these agencies' missions. PNNL is currently developing procedures for systematically conducting probabilistic flood hazard assessment for critical infrastructure from low-frequency, high-impact hydrometeorological events that can be used to assess risks to both the infrastructure and human health and safety. PNNL's flood-related research also includes effects of flood conditions on human performance as it relates to emergency preparedness and response. Insights on human performance under flood conditions can help prevent loss of life and increase emergency response effectiveness. PNNL's climate change research is also leveraged to provide additional insights into future flooding scenarios and associated uncertainties for decision making.

6.1.14.2 Seismic Hazard Assessments

PNNL has key technical expertise in probabilistic and deterministic seismic hazard analysis, helping to provide end-to-end evaluations. With more than 30 years of professional seismic expertise and 15 years of industry experience related to the evaluation of sites and structures, PNNL is uniquely positioned to advance the state-of-the-art and provide trusted solutions. Our team has specifically led R&D projects involving: 1) site characterization using a wide variety of seismic techniques (e.g., Spectral Analysis of Surface Waves, Refraction Microtremor, Cross Borehole Tomographic Imaging), 2) development of strong motion acceleration estimates for shock mounting and structural evaluations in the National Security space, 3) development of cutting-edge laboratory experiments to evaluate moduli of geologic materials under dynamic conditions, and 4) development and deployment of seismic arrays used for local to regional earthquake monitoring and ambient noise imaging campaigns. Additionally, in recent years, we have focused on providing real-time computational modeling informed by experimental measurements to help stakeholders make critical operational decisions.

7.0 Capture and Immobilization of Actinides and Fission Products in Waste Forms

Molten salt reactors will generate a unique waste stream that will require novel processes for safe and secure management and disposal. The wide variety of potential waste streams presents a complex materials design challenge, requiring the development of many different types of materials specifically tailored to efficiently immobilize specific streams.

PNNL's waste form development capabilities are key to the nation's quest for the safe, long-term storage of nuclear waste. Specifically, our researchers investigate a range of solutions to isolate actinides and radionuclides generated during energy production (including past plutonium production at the Hanford site), used nuclear fuel recycling, and legacy waste from other remediation sites. PNNL's waste form development experience is approaching 50 years; we helped pioneer nuclear waste vitrification efforts in the 1960s, we developed the technology for the ceramic joule-heated slurry-fed melters used at the West Valley and Savannah River sites, and we have since expanded our research capabilities to include glass, glass-ceramic, grout, metal, metal-ceramic, and other waste forms that will withstand corrosion over geologic time.

7.1 Glass and Material Science

PNNL's glass and materials science experts develop innovative materials and fabrication processes for a wide range of nuclear applications. Currently, integrated programs from the DOE Offices of NE and Environmental Management (EM) aim to develop durable forms for radioactive waste both from the legacy weapons programs and from future processing of used civilian nuclear power rods.

PNNL has expertise in bulk and film processing of glasses and ceramics as well as material characterization by diffraction, microscopy, and spectroscopy. The team has particular strengths in synthesis, characterization, and application of glass and ceramic materials.

Our work includes basic and applied research in glass and ceramic chemistry and thermodynamic modeling; photonic, structural, and refractory materials design and development; materials compatibility and integration; microstructural and microchemical design, fabrication, characterization, and analysis; and process flowsheet development for scaled-up industrial applications.

Our applied research has historically involved a large amount of work in development waste forms for nuclear waste immobilization. Recently, however, we have had an increasing amount of work in functional materials design and synthesis.

In addition to our work in applied research, our staff maintains basic theoretical research and development interests, including:

- the nature and structure in the non-crystalline, amorphous, and glassy state
- property evolution in very complex (10+ constituent) glass/ceramic systems
- melting and crystallization thermodynamics and kinetics
- glass corrosion and chemical durability
- crystal chemistry and optical, electrical, and magnetic properties of inorganic materials
- the effects of ionizing radiation on materials.

7.2 Nuclear Waste Immobilization Technology

As leaders in waste glass technology since the late 1960s, PNNL has had a major role in developing and advancing the vitrification technology being employed at the Hanford Site Waste Treatment Plant (Richland, Washington), the Savannah River Site Defense Waste Processing Facility (Aiken, South Carolina), and the West Valley Demonstration Project (West Valley, New York). We provide technical support to melter operations in areas of technology development and demonstration, process and equipment design, and advanced process diagnostics.

Members of our team are involved in current projects studying various aspects of waste forms for existing nuclear defense wastes and proposed wastes for a commercial nuclear fuel cycle. These waste forms are used for immobilizing high level waste (HLW), low activity waste (LAW), and various secondary waste from intermediate processes in radioactive waste treatment.

7.3 Waste Form Durability

One critical aspect to designing applicable waste forms is the assessment of durability, often for geologic time scales. PNNL's expertise in waste form durability and performance assessment is unmatched in the DOE complex and is sought out whenever a new waste form design must be vetted. PNNL's experts have taken the lead in establishing and running an international effort into understanding the long-term corrosion of high-level waste glasses. In this collaborative program, six nations coordinate their activities to minimize duplication and increase confidence in the resultant studies, while focusing on each nation's specific research priorities. This approach is expected to increase national and international confidence in the robustness of the understanding of glass corrosion, leading to cost savings in disposal because of reduced uncertainty. Specific areas of excellence in this collaborative group include:

- Pioneering the use of many standard dissolution tests
- Using rare stable isotopes to accurately track dissolution
- Developing and refining glass corrosion models
- Studying the corrosion of ancient man-made and natural analogue glasses

7.4 Immobilization of electrochemical salt wastes

PNNL has been working in the area of waste forms development and testing for chloride-based electrochemical salt wastes since 2008 through funding from DOE-NE. This work includes the immobilization of used LiCl-KCl and LiCl-Li₂O salts or separated salt wastes in glass-bonded sodalite, tellurite glass, and lanthanide aluminoborosilicate glass. Different waste form options include glasses (e.g., tellurite, lanthanide borosilicate), glass-bonded sodalite, and phosphate matrices. Experiences in these areas of research could be leveraged to look into waste form options for fluoride-based and other chloride-based salt compositions to support the MSR research.

7.5 Capture and Immobilization of Volatile Radionuclides

MSRs that use "liquid" fuels must be able to manage the volatile fission product gases generated, in particular iodine, krypton, and xenon. These radionuclides will be generated continuously during the reactor operation and must be efficiently captured to meet the regulatory limits and subsequently immobilized into either a waste form for subsequent disposition or for distribution as subsequent valuable products. PNNL has decades of expertise in sorbent material and metal organic framework design, development, synthesis, and characterization. In addition, there has been a long-standing history of developing waste forms for these waste streams.

7.5.1 Development of Sorbents for Volatile Radionuclides

Various mesoporous materials were developed and applied to remove contaminants from hazardous waste streams. These materials can be functionalized to target specific species at different environments and conditions. They are highly efficient sorbents, whose rigid, open pore structure allows for rapid and efficient sorption kinetics. Their interfacial chemistry can be fine-tuned to selectively sequester a specific target species. These materials can be used to capture volatile fission products and non-radioactive gases that evolved during operation of molten salt reactors. Subsequently, contaminant-loaded sorbents can be consolidated into durable waste forms. For example, iodine-loaded silver-functionalized silica aerogel with iodine capacities ~480 mg/g can be consolidated by simultaneous application of fast heating rates to temperatures above 1000°C and pressures up to 210 MPa to fully dense silica-based waste form containing ~39 mass% of iodine.

Silver-functionalized silica aerogels and silver-functionalized aluminosilicate aerogels, Ag⁺-impregnated aluminosilicate aerogels, and chalcogenide aerogels were developed at PNNL and show great promise for high-iodine removal efficiency from nuclear reprocessing off-gas streams. These new sorbents are significantly more effective than silver mordenite, the most commonly studied sorbent for iodine.

High-end capabilities are available to simultaneously image morphological and structural changes that occur on the surface of sorbent materials. Three-dimensional tomographic energy dispersive spectroscopy imaging can provide distribution of species. Electron energy loss spectroscopy and diffraction can be used to examine the possible chemical changes that have occurred during sorption. These tools shed light on understanding the fundamental changes that occur as the volatile species sorb onto the solid phase.

Capabilities are available to evaluate real-time sorption performance of sorbents. The sorbents can be tested at static and dynamic conditions, at different concentrations of contaminants, and under prototypical off-gas conditions.

7.5.2 Capture and Immobilization of Volatile Radionuclides Using Metal Organic Frameworks (MOFs) and Off-Gas System Design

The off-gas generated during the MSR operation includes noble gases (Xe and Kr) that must be efficiently captured. Xe isotopes, in particular, are strong neutron absorbers and, as a result, not suitable for recirculating through the reactor core. A mature process used to remove these gases is a cryogenic distillation. In this process, off-gases are first cleaned by removing H₂O, O₂, and N₂, then the resulting gas is cooled to liquid N₂ temperatures (77 K) to separate the noble gases using large distillation column(s). Installing and operating a cryogenic process is expensive and energy intensive. Another major concern with this technology is the accumulation of ozone due to the radiolysis of O₂ at cryogenic temperatures.

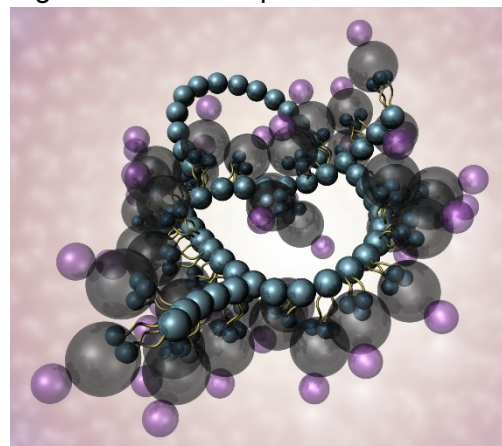


Figure 7.1. Silver-functionalized silica aerogel developed at PNNL to capture and immobilize radioiodine from reprocessing off-gas streams

To overcome the cryogenic distillation approach, PNNL developed a novel class of porous materials known as MOFs for selective removal of Xe and Kr at near room temperature. Among all the MOFs tested, a calcium-based MOF, CaSDB (SDB = 4,4'-sulfonyldibenzoate, with a pore diameter of 4.5 Å) has been shown to outperform all the materials tested at room temperature. Due to high polarizability of Xe, most of the MOFs capture Xe selectively over other gases, including Kr. Therefore, a two-column approach using CaSDB has been demonstrated, where in bed 1 Xe is selectively removed at room temperature from a synthesis gas mixture consisting of 1300 ppm Xe, 130 ppm Kr, 300 ppm CO₂, 0.9% Ar, 78.2% N₂, and 21% of O₂. The off-gas mixture (without Xe) exiting bed 1 was passed through bed 2, also containing CaSDB to remove Kr at room temperature. At this, the adsorption capacity for Xe and Kr was not affected even at 48% relative humidity. Similarly, MOFs can be modified to target gases of specific interest in MSR technology. Therefore, these materials could be used to capture volatile fission products and non-radioactive gases evolving during MSR operations.

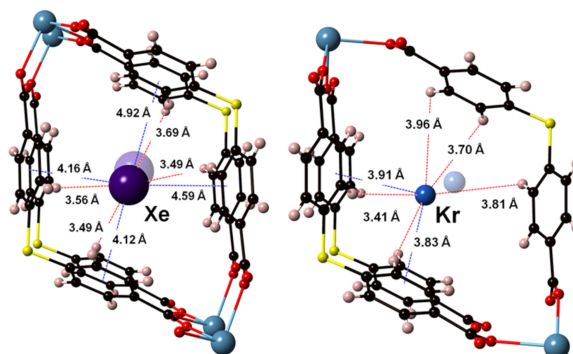


Figure 7.2. The PNNL patented MOF technology can be used to separate the noble gases from off gases

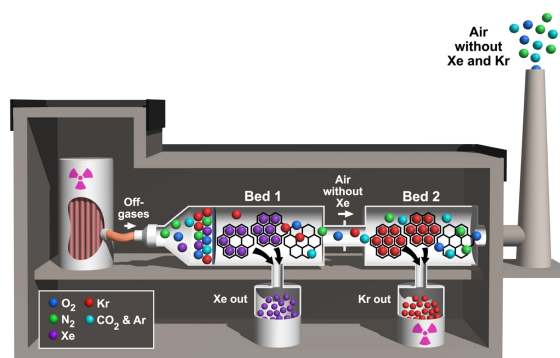


Figure 7.3. The two-step approach removes Xe (bed 1) and Kr (bed 2) at room temperature

7.5.3 Capture and Immobilization of Volatile Radionuclides

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8.0 Safeguard & Signatures

The radionuclide signatures of an MSR are of relevance to nuclear non-proliferation activities, including nuclear explosion monitoring and safeguards activities. PNNL can produce a range of fission and activation products and coupled with high-sensitivity gamma-spectrometry facilities allow for the evaluation of these signatures. This includes the use of a DT neutron generator at PNNL, the Washington State University Reactor (WSUR), and the Shallow Underground Laboratory (SUL) at PNNL. Using these facilities, different actinide (^{235}U , ^{238}U , ^{233}U) and salt (NaCl , KCl , NaF , RbF , NaBF_4) samples have been irradiated and characterized. These experiments have also established a capability to perform long-term radionuclide measurements, including refinements to nuclear data such as the half-life of ^{43}K . Specific to safeguards research, and mitigation against fuel diversion, a technique has been developed to enable the direct determination of ^{233}U using coincidence-based gamma-spectrometry techniques. To evaluate the impact of radionuclide signatures, an atmospheric transport modelling capability has been developed and used to assess the impact of future MSR facilities. Notable discoveries have included that the radioxenon signature of some MSR designs could potentially act as an interference to nuclear explosion monitoring measurements. Collaborative links have also been established with industry, academia and government organizations to support future work.

9.0 Licensing

PNNL has extensive experience supporting regulatory activities for advanced reactor deployment through our support of the NRC. Additionally, numerous staff bring recent first-hand experience from their time working for commercial nuclear vendors in the design, analysis, and licensing of the current fleet and advanced reactors, including fast reactor concepts. With these experiences, PNNL is uniquely qualified to provide independent technical assistance and support to assist in the regulatory aspects of advanced reactors for disciplines, including: environmental reviews, nuclear core performance, nuclear fuel design, plant systems design and performance, nuclear safety analysis, probabilistic risk analysis, materials and waste safety licensing, and others.

PNNL has extensive knowledge of NRC regulations and performing reviews in accordance with NUREG-800. This includes reviewing portions of CP or OL applications under 10 CFR Part 50 and ESP, design certification (DC), combined license (COL), SDA, or ML applications under 10 CFR Part 52. Additionally, PNNL has reviewed numerous Final Safety Analysis Report chapters and underlying methodology documentation for operating DC applications, including documentation associated with fuel and thermal-hydraulic design, neutronics design, and nuclear safety analysis.

PNNL staff are qualified, competent, and fully trained to perform the required technical assistance and support services. This includes having access to, and experience with, a range of mechanical, thermal-hydraulic, and nuclear simulation software used for performing independent confirmatory calculations. If required, PNNL's NQA-1-compliant NQAP ensures calculations and independent reviews are performed by trained and qualified staff members and documentation is complete, correct, practical, satisfies the applicable requirements, and includes the appropriate QC requirements.

Over the past three decades, the NRC has reached out to PNNL at critical stages of nuclear plant design for assistance in developing new standards for safety reviews and in evaluating the safety of new plant designs against those standards.

PNNL support to the NRC encompasses a broad range of nuclear reactor safety issues, including PRA, fuels qualifications, materials degradation, fire protection, seismic analyses, aging management, and siting.

Projects performed under the NRC EWA include:

- APR1400 Fuel System Topical Report Review
- APR1400 Seismic Topical Report Review
- NuScale AREVA Fuel Application Review
- Flood Hazard Re-evaluation
- Materials Degradation Assessments
- Fuel Seismic LOCA design
- Seismic PRA support.

PNNL's recent key experience relevant to advanced reactor regulatory infrastructure development and licensing includes:

- Assisting NRC's Advanced Reactor Program in developing their licensing infrastructure for planned SMR applications, including developing Design-Specific Review Standards (DSRSs) and Safety Evaluation Report templates for the mPower and NuScale designs. These DSRs provide guidance to NRC staff on risk-informed review procedures for 19 design and siting subject areas, using current knowledge of the proposed designs.
- Participating in DOE's Gateway for Accelerated Innovation in Nuclear (GAIN) Program. PNNL and the Columbia Basin Consulting Group (Kennewick, WA) were awarded a voucher to conduct a regulatory gap analysis for their Lead-Bismuth SMR design.
- Under the auspices of DOE-NE, supporting TerraPower nuclear fuel development programs, including hot cell research into the mechanical properties of irradiated cladding.
- Supporting the development of the NGNP High Priority Regulatory Topical Reports related to NGNP Licensing (see SECY-10-0034, Potential Policy, Licensing, and Key Technical Issues for Small Modular Nuclear Reactor Designs).
- Supporting development of ANS 53.1, "Nuclear Safety Criteria and Safety Design Process for Modular Helium-Cooled Reactor Plants," and ASME/ANS S1.4, "Standard for Probabilistic Risk Assessment for Advanced Non-Light Water Reactor Nuclear Power Plants."

- In support of NRC's Advanced Reactor Program, preparing "High Temperature Gas Reactors: Assessment of Applicable Codes and Standards" (PNNL-20869, October 2011)
- Providing NRC leadership and a complete technical team responsible for pre-application activities, as well as the completing the Environmental Impact Statement for Tennessee Valley Authority's (TVA) Early Site Permit for siting one or more SMRs at the Clinch River Site in Tennessee.
- Revising NRC's Standard Review Plan (NUREG-0800) for current new reactor design reviews. NUREG-0800 provides guidance to NRC staff in performing safety reviews of applications for licenses to construct or operate nuclear power plants.
- Assisting NRC in New Reactor Design Certification Reviews. Providing assistance to NRC technical branches in their review of the AP1000 and Advanced Pressurized-Water Reactor (APWR) reactor design certification applications.
- Developing COL/ESP-ISG-027, Specific Environmental Guidance for iPWRLight Water Small Modular Reactor Reviews.
- Revising Regulatory Guidance 4.2, "Preparation of Environmental Reports for Nuclear Power Stations" (in progress). RG 4.2 provides guidance to applicants regarding the preparation of Environmental Reports, with new guidance for SMR applications.
- Assisting with maintaining and updating radiation protection and health physics codes such as GENII, RASCAL, and PAVAN through NRC's Office of Research RAMP program. This program included updating codes such as the GALE code for advanced reactor designs.

9.1 NRC Licensing References

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