



PNNL-27369

# Pacific Northwest National Laboratory Capabilities for Molten Salt Reactor Technologies

**March 2018**

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## Summary of PNNL MSR-Aligned Capabilities

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Reactor Design and Modelling	David Wootan, Ronald Omberg, Ram Devanathan, Thomas Michener	10+
Waste Forms	John Vienna, Brian Riley, Jarrod Crum, Joseph Ryan	10+
Licensing	Bruce McDowell, Steven Short, Ken Geelhood	15+

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## 1.0 Materials Corrosion – Modelling and Performance

### 1.1 Corrosion Modelling

Materials corrosion has been recognized as an issue in structural materials for MSR. 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 development of 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, 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.

### 1.2 Salt/Material Interactions and Corrosion

PNNL has a wide range of experience working on 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, as well as high-pressure autoclaves.

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## 2.0 Fuel and Coolant Salt Processing

### 2.1 Producing fluorinated actinides through process monitoring

To convert existing fuel into a form that could be used in the MSR, the actinides require fluorination using flame fluorination or voloxidation (or volox) followed by fluorination. If volox is used followed by fluorination, then PNNL has the capability to do volox with a thermogravimetric-differential thermal analyzer-mass spectrometer (TG/DTA/MS) apparatus in a modular hot cell in one of the existing facilities (i.e., PDC-1 in RPL). Here, irradiated metal fuel could be combined with a gaseous fluorinating agent while products are monitored with a mass spectrometer. This can also be done with oxide fuels (following an oxidative step).

### 2.2 Removing fission products from salt mixtures

PNNL currently has a cross-flow ultra-filter (CUF) operating in shielded analytical laboratory hot cells. An analogous apparatus could be built for processing MSR fuels in those hot cells with personnel already trained to operate this apparatus. The type of filtration system would be different from the CUF and more applicable to the MSR salts using fluorination technologies established at PNNL to operate an apparatus with surrogate materials outside of the hot cell. Additionally, PNNL has experience with using  $\text{NF}_3$  sparging gas to remove impurities during operation of a molten salt using TG/DTA/MS apparatuses in fume hoods and gloveboxes and these could be adapted for use in hot cells.

### 2.3 Fuel and Coolant Salt Processing References

1. Scheele, R.D., A.M. Casella, and B.K. McNamara, "Use of nitrogen trifluoride to purify molten salt reactor coolant and heat transfer fluoride salts," *Industrial and Engineering Chemistry Research* **56**(19), 5505-5515 (2017).
2. Scheele, R.D. and A.M. Casella, "Assessment of the use of nitrogen trifluoride for purifying coolant and heat transfer salts in the fluoride salt-cooled high-temperature reactor," PNNL-19793. Pacific Northwest National Laboratory, Richland, WA.

### 3.0 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, 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 DOE-NE and USNRC. 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 key recent experience relevant to sensors, instrumentation and control include:

- A joint advanced sensors needs assessment with ORNL and 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 US 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.
- Developing a fundamental understanding of sensor materials behavior under irradiation, with the goal of designing and deploying sensors for monitoring temperature and fuel mechanical properties during irradiation testing. This work, jointly conducted with INL, led to new insights into sensors behavior under in-core and in-vessel conditions relevant to MSRs. Currently, PNNL is designing a sensor and instrumentation package that will be available as part of the I&C technology offered to experiments at the TREAT facility.
- 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 nondestructive evaluation (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 online monitoring of instrument calibration, to provide early warning of sensor failure while avoiding unnecessary recalibration.
- Sensors and algorithms for prognostic health monitoring of components, and predictive risk-informed methods for predicting plant condition 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 safety and economics of operation (avoid unnecessary repair and maintenance actions).

### 3.1 Spectroscopic-Based Process Monitoring Summary at PNNL

PNNL has advanced the use of spectroscopic-based (i.e., ultraviolet-visible, or UV-vis, 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). This process monitoring 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 was demonstrated on actual used commercial fuel samples and on numerous simulated fuel reprocessing solutions. Quantitative measurement of HNO<sub>3</sub> concentration and pH monitoring was also demonstrated. These methods were also effectively miniaturized to allow for monitoring of microfluidic systems, which can reduce both dose to workers and 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 (UV-vis) 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.

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
- Calculation of diffusion coefficients of electroactive metal species
- Determination of redox potentials for metal species

The current alternative electrochemical process monitoring is electrode-based characterization during electrodeposition of bulk metals, where the composition is precisely known only at the electrode surface. This alternative methodology allows for rapid, solution-based measurements



and can be implemented in both stationary and flowing systems. This approach can provide necessary process information by identifying and quantifying the species present. This approach is not limited to any specific process type and can be applied to molten salt, aqueous, and non-aqueous systems.

## 3.2 In-vessel MSR/FHR Monitoring Requirements, Challenges, and Opportunities

### 3.2.1 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 advanced small modular reactor (AdvSMR) or 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 H<sub>2</sub>O, CO<sub>2</sub>, CO, CH<sub>4</sub>, and O<sub>2</sub> 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 <sup>3</sup>H 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.

### 3.2.2 EM Pump Monitoring

Degradation and performance is 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 AdvSMR coolants. Optical vibrometry techniques may be feasible on components above the coolant level.

### 3.2.3 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.

### 3.2.4 Coolant Flow and Pressure

Thermal infrared spatial temperature profiling in molten salt coolants may be feasible 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.

### 3.2.5 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.

### 3.2.6 Neutron Flux/Power

Cherenkov radiation monitoring may be very feasible in MSR or fluoride salt-cooled high-temperature reactor (FHR) coolants.

## 3.3 Instrumentation, Control, and Online Monitoring References

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## 4.0 Reactor Design and Modelling

### 4.1 Fast Flux Test Facility (FFTF) Experience

PNNL has capability in the reactor design and modelling area based on staff members with years of experience in the design, operation, and irradiation testing in the Fast Flux Test Facility (FFTF). Although the FFTF was a sodium-cooled reactor, much of the design processes, procedures, and test data apply, or can be modified to apply, to other Advanced Reactors such as the new generation and evolution of Molten Salt Reactors (MSR). PNNL staff currently support advanced reactor designs by expert retrieval of FFTF information. PNNL staff are also able to analyze 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. Below are examples of current PNNL programs in which the FFTF design information and testing data is being used to support the evolution of Advanced Reactors. PNNL would be willing and enthusiastic to employ its staff in similar programs supporting MSR.

- Using applicable FFTF experience to identify more efficient and effective means of designing systems and tests so that experience gained is documented for future use in summary reports designated as lessons learned from FFTF design, operation, and testing,
- Supporting the design of the Versatile Fast Test Reactor (VFTR), which is currently in its design stages, using design information on the major systems in the FFTF,
- Supplying irradiation test data on materials irradiated in the FFTF to a high fluence under controlled temperature conditions to support design of Advanced Reactors so that they will have material test information under the conditions at which they intend to operate,
- Supplying information on FFTF data acquisition systems so that future Advanced Reactors can build upon the plant and test data acquisition systems which performed superbly during the operation of the FFTF.

## 5.0 Capture and Immobilization of Fission Products in Waste Forms

### 5.1 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 (i.e., GNEP, AFCI, FCRD, and NTRD). This work includes the immobilization of used LiCl-KCl and LiCl-Li<sub>2</sub>O 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.

### 5.2 Capture and immobilization of volatile radionuclides

PNNL has decades of experience in studying the capture and immobilization of volatile fission products including (mostly with nonradioactive simulants): <sup>129</sup>I, Xe, Kr, and <sup>14</sup>C. For iodine, Ag<sup>0</sup>-functionalized silica and aluminosilicate aerogels, Ag<sup>+</sup>-impregnated aluminosilicate aerogels, and chalcogenide aerogels were studied at PNNL and show great promise for high-iodine capacities and decontamination factors. Some of the work at PNNL has demonstrated that some of these new sorbents are significantly more effective than silver mordenite (AgZ), the most commonly studied iodine sorbent. Recent work has focused on the capture and immobilization of Xe and Kr using metal-organic frameworks (MOFs). The MOF sorbents have high capacities of these noble gases and different MOFs can be used to selectively capture Kr over Xe or Xe over Kr, depending upon the need. The MOFs also show promise at capturing volatile <sup>14</sup>CO<sub>2</sub> through carbon sequestration efforts. These materials could be used to capture volatile fission products evolved during MSR operations.

### 5.3 Sorbent and Waste Forms References

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## 6.0 Licensing

### 6.1 Licensing Overview

Over the past three decades, the Nuclear Regulatory Commission (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's key recent experience relevant to NRC's advanced reactor regulatory infrastructure development includes:

- PNNL assisted 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 DSRSs provide guidance to NRC staff on risk-informed review procedures for 19 design and siting subject areas, using current knowledge of the proposed designs.
- PNNL participates 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, PNNL supports TerraPower nuclear fuel development programs, including hot cell research into the mechanical properties of irradiated cladding.
- PNNL supported 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).
- PNNL supported 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, prepared "High Temperature Gas Reactors: Assessment of Applicable Codes and Standards" (PNNL-20869, October 2011)
- PNNL is providing NRO the leadership and complete technical team responsible for pre-application activities as well as the preparation of the Environmental Impact Statement for TVA's Early Site Permit for siting one or more SMRs at the Clinch River Site in Tennessee.

### 6.2 Other NRC Licensing Experience

Under the Enterprise Wide Agreement and other contracts, PNNL currently supports NRC more than any other DOE national laboratory. Under the EWA, PNNL currently conducts over 30 active task orders with a total authorized ceiling of > \$85M, covering a broad range of nuclear

reactor safety issues including probabilistic risk assessment (PRA), fuels qualifications, materials degradation, fire protection, seismic analyses, aging management, and siting. Current projects 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

Other NRC Office of New Reactor support has included:

- 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 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.

### 6.3 NRC Licensing References

1. NUREG-0800 SRP/DSRS *Design-Specific Review Standard (DSRS) for the mPower™ Design*. U.S. Nuclear Regulatory Commission.
2. NUREG-0800 SRP/DSRS *Design-Specific Review Standard (DSRS) for the NuScale Design*. U.S. Nuclear Regulatory Commission.
3. COMBINED LICENSE AND EARLY SITE PERMIT COL/ESP-ISG-027 *Specific Environmental Guidance for Light Water Small Modular Reactor Reviews*, Interim Staff Guidance. August 2014



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