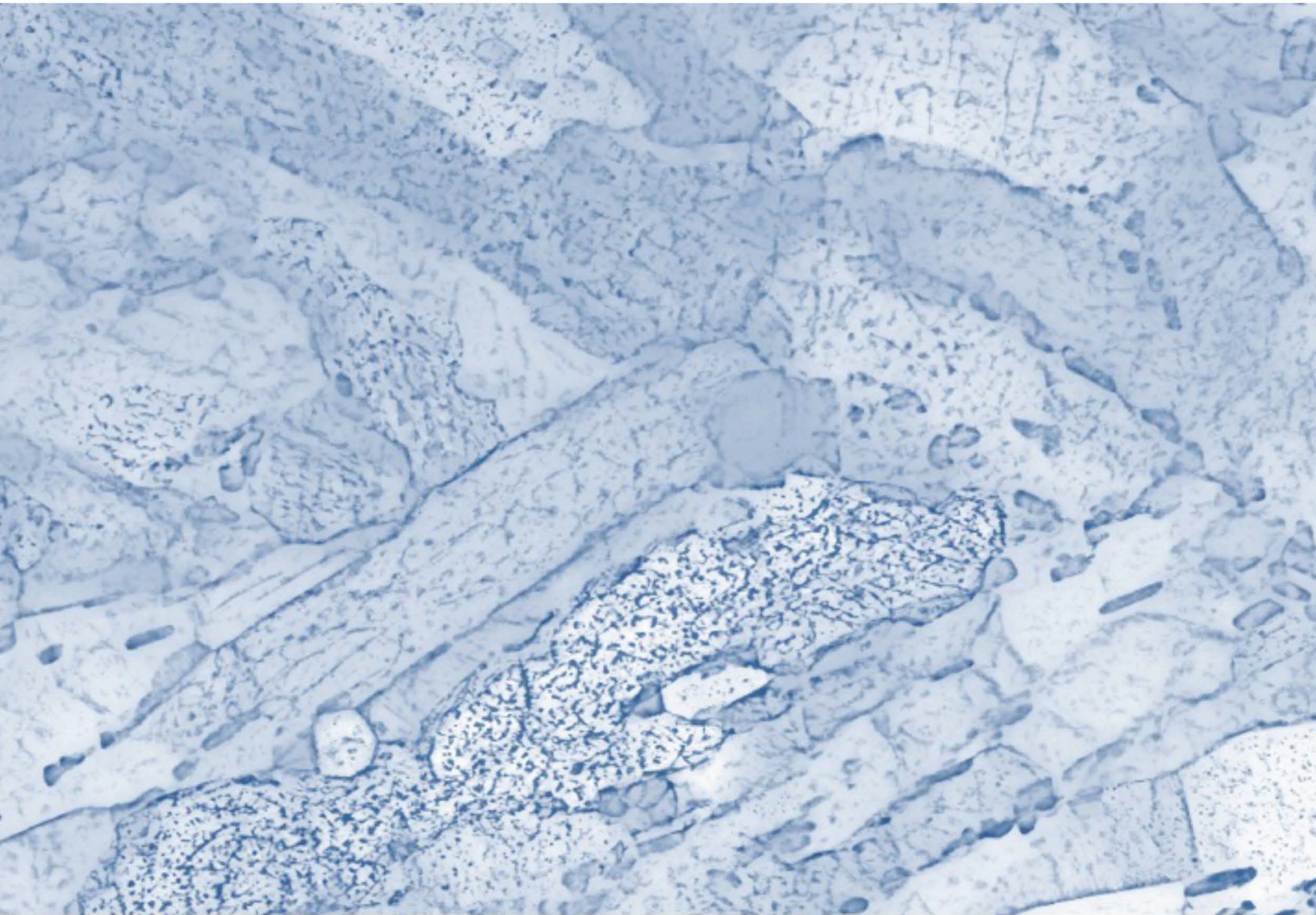


Basic Energy Sciences Roundtable

Foundational Science to Accelerate Nuclear Energy Innovation



*Scientific Breakthroughs to Realize the Full Potential
of Nuclear Energy*

Report of the Basic Energy Sciences Roundtable on Foundational Science to Accelerate Nuclear Energy Innovation

July 20–22, 2022

Workshop Co-Chairs

Marianne Walck	Idaho National Laboratory/National Energy Technology Laboratory
Rebecca Abergel	University of California, Berkeley/Lawrence Berkeley National Laboratory
Blas Uberuaga	Los Alamos National Laboratory

Panel Leads

Janelle Wharry	Purdue University
Jay Laverne	Notre Dame University
Arianna Gleason-Holbrook	SLAC National Accelerator Laboratory
Adrien Couet	University of Wisconsin–Madison
Vanda Glezakou	Oak Ridge National Laboratory

Basic Energy Sciences Team

Teresa Crockett, Linda Horton, Gail McLean, Andrew Schwartz, John Vetrano, Philip Wilk

Other U.S. Department of Energy and Invited Attendees

Listed in Appendix B

Oak Ridge Associated Universities Support

Linda Severs

Oak Ridge National Laboratory Publication Team

Andrea Beatty, Brian Woods, Rachel Brooks, Erica Heinrich, Güneş Özcan

Contents

Figures.....	iii
Sidebars.....	iv
Abbreviations.....	v
Executive Summary	vi
1. Introduction.....	1
2. Priority Research Opportunities.....	7
PRO 1: MASTER COMPLEX ELECTRONIC STRUCTURES TO TAILOR THERMOCHEMICAL REACTIVITY, TRANSPORT, AND MICROSTRUCTURAL EVOLUTION.....	7
1a. Summary	7
1b. Key Scientific Questions	10
1c. Scientific Challenges and Research Opportunities	10
1d. Potential Impacts	14
PRO 2: INTERROGATE AND DIRECT THE PHYSICS AND CHEMISTRY UNDERPINNING NEXT-GENERATION COOLANTS AND SOLVENTS.....	15
2a. Summary	15
2b. Key Scientific Questions	16
2c. Scientific Challenges and Research Opportunities	16
2d. Potential Impacts	19
PRO 3: ELUCIDATE AND CONTROL THE UNDERLYING PHYSICS AND CHEMISTRY OF INTERFACES IN COMPLEX NUCLEAR ENVIRONMENTS.....	20
3a. Summary	21
3b. Key Scientific Questions	21
3c. Scientific Challenges and Research Opportunities	21
PRO 4: BRIDGE MULTIFIDELITY, MULTIREOLUTION EXPERIMENTS, COMPUTATIONAL MODELING, AND DATA SCIENCE TO CONTROL DYNAMIC BEHAVIOR.....	28
4a. Summary	28
4b. Key Scientific Questions	28
4c. Scientific Challenges and Research Opportunities	29
4d. Potential Impacts	32
PRO 5: HARNESS ARTIFICIAL INTELLIGENCE TO DESIGN INHERENTLY RESILIENT CONDENSED PHASES.....	34
5a. Summary	34
5b. Key Scientific Questions	38
5c. Scientific Challenges and Research Opportunities	38
5d. Potential Impacts	41
3. Closing Remarks.....	42
4. References.....	43
Appendix A. Charge.....	A-1
Appendix B. Attendees.....	B-1
Appendix C. Agenda.....	C-1

Figures

Figure 1. Correlation between Fe^{3+} irradiation damage and implantation profiles in pure iron.	8
Figure 2. Advanced experimental techniques to characterize electronic structure evolution.	11
Figure 3. Examples of experiment–theory mismatches in phase diagrams.	12
Figure 4. Distinctly different irradiation-induced microstructures in nickel–titanium depend upon the chemical and structural ordering.	13
Figure 5. Heterogeneity arising from a simple mixture of monovalent and divalent cations in molten $\text{MgCl}_2\text{-KCl}$. ^[51]	15
Figure 6. Actinide-loaded ligands for extraction. ^[53]	17
Figure 7. Novel use of robust optical sensors to probe key characteristics under harsh process conditions. ^[60]	19
Figure 8. Illustration of combined irradiation and corrosion effects adapted from Schmidt et al. ^[65] and Li et al. ^[66] ...	22
Figure 9. Experimental setup used for the measurement of the Raman features of $\text{LiCl-Li}_2\text{O}$ with electrochemically generated lithium.	25
Figure 10. Multitude of processes showcasing the tremendous range in length and time scales needed in experiments and modeling.	30
Figure 11. Schematic representation of the applicable domain of scientifically guided ML.	35
Figure 12. AI models used to discover important features, with the potential to identify novel self-resilient mechanisms.	39
Figure 13. A Gaussian process model can effectively reproduce the grain size dependence of the mechanical strength of an alloy even though it is completely devoid of any knowledge of the effect of the density of grain boundaries for large-grain metals, the effect of grain boundary sliding in nanocrystalline alloys, or even the regime change. ^[117]	40

Sidebars

Sidebar 1. Nuclear Energy in the U.S.....	1
Sidebar 2. Fission and Fusion Reactor Concepts.....	2
Sidebar 3. Extreme Environments.....	3
Sidebar 4. High-Burnup Structure.....	9
Sidebar 5. ML Approaches Elucidate the Dynamic Structure and Coordination of Actinides in Nuclear Environments.....	24
Sidebar 6. Designing Heterostructured Materials and Interfaces with Unprecedented Resilience and Performance under Extreme Conditions.....	27
Sidebar 7. Experimental Steering with ML-Driven Data Fusion.....	33
Sidebar 8. Self-Resilient Mechanisms in Nuclear Materials.....	36
Sidebar 9. High-Throughput Experiment–AI/ML Framework Applied to Materials Discovery.....	37

Abbreviations

AI	artificial intelligence
ASCR	Office of Advanced Scientific Computing Research
BES	Office of Basic Energy Sciences
CASL	Consortium for Advanced Simulation of Light Water Reactors
CCA	chemically complex alloy
CTB	coherent twin boundary
DFT	density functional theory
DOE	U.S. Department of Energy
FES	Office of Fusion Energy Sciences
HBS	high-burnup structure
HEP	Office of High Energy Physics
HLM	heavy liquid metal
HTE	high-throughput experiment
INL	Idaho National Laboratory
ITB	incoherent twin boundary
LANL	Los Alamos National Laboratory
LBNL	Lawrence Berkeley National Laboratory
ML	machine learning
MSR	molten salt reactor
NE	Office of Nuclear Energy
NREL	National Renewable Energy Laboratory
ORNL	Oak Ridge National Laboratory
PDF	pair distribution function
PNNL	Pacific Northwest National Laboratory
PPPL	Princeton Plasma Physics Laboratory
PRD	priority research direction
PRO	priority research opportunity
SC	Office of Science
SLAC	SLAC National Accelerator Laboratory
TEM	transmission electron microscopy
UED	ultrafast electron diffraction

Executive Summary

Energy security, availability, and reliability are among the greatest challenges facing the nation and the planet. An abundant potential source of energy resides in the fundamental atomic building blocks of the universe in the form of nuclear fission and fusion reactions. In fact, energy from nuclear fission currently provides the majority of the world's zero-carbon electricity, and future fusion energy systems offer great promise; carbon-free nuclear energy technologies can be key to the world's decarbonized energy future. Although contemporary fission systems use well-established technologies to supply safe and efficient baseload power, they could be more fuel efficient and less costly. Moving beyond massive light-water fission reactors to a variety of advanced nuclear systems—which will vary in size and operate in extremes of temperature, corrosivity, and other parameters—will place stringent conditions on materials and chemical systems. New demands will be placed on the coolants and solvents, the materials, and the monitoring tools used in these reactors. Fusion-based nuclear energy will require superior materials to withstand extremely high temperatures, plasma exposure, radiation damage, and implanted gases. The advantages associated with these new fission and fusion technologies will be realized only through continued advancements in the fundamental science underpinning our knowledge of the physics and chemistry of nuclear systems gained via improved experimental and computational methods.

In July 2022, the U.S. Department of Energy's Office of Basic Energy Sciences—in coordination with the Offices of Nuclear Energy, Fusion Energy Sciences, and Advanced Scientific Computing Research—held a virtual roundtable titled “Foundational Science to Accelerate Nuclear Energy Innovation” to discuss the scientific and technical barriers for advanced nuclear energy systems. Five priority research opportunities were identified to address these scientific and technical challenges and to accelerate progress toward the realization of next-generation fusion and fission energy systems.

The foundational science gaps inhibiting the advancement of nuclear energy technologies are identified and tackled in five priority research opportunities. These opportunities pave the way to accelerate the development and ultimately the adoption of new nuclear energy systems. They include the fundamental aspects of ion-electron interactions; novel properties of next-generation coolants and solvents; interfacial dynamics, not only in solids, but in other aspects of nuclear reactors; novel operando and in situ monitoring and sensing; and artificial intelligence to accelerate condensed phases discovery. Building on the foundation established by previous Basic Energy Sciences workshops, these opportunities encompass recent advances in fundamental knowledge and focus on the experimental and computational methods needed to resolve major technical challenges for nuclear energy technologies. Through developing fundamental scientific insight as well as pushing the frontiers of modeling complex systems and probing the operation of materials and chemical systems in extreme environments, research motivated by the priorities identified here will further develop the promise, potential, and utilization of nuclear energy for a clean energy future.

Priority Research Opportunities to Accelerate Nuclear Energy Innovation

Master complex electronic structures to tailor thermochemical reactivity, transport, and microstructural evolution

Key question: *How do we elucidate, predict, and harness coupled electron–ion dynamics to enable discovery and deployment of novel materials, coolants, and solvents for future fission and fusion energy?*

Chemical and materials phenomena in nuclear systems have historically been described through atomic and molecular interactions, with limited consideration of sub-atomic electron structures. However, electronic interactions fundamentally govern the nature and energy of bonding, transport, defect energies, and phase stability of solids and liquids. Bridging quantum physical considerations from electron–ion and electron–electron interactions to bulk phenomena like mass transport and mesoscale restructuring is necessary to understand evolution of matter in nuclear environments and to predict phase dynamics and thermophysical properties of liquids and solids in nuclear systems. Harnessing these interactions will subsequently enable tuning of materials and chemical performance under coupled extremes.

Interrogate and direct the physics and chemistry underpinning next-generation coolants and solvents

Key question: *How can we probe and control the physics and chemistry of coolants, solvents, and their solutions in the harsh environments associated with nuclear energy?*

Advanced nuclear technologies will require the deployment of next-generation coolants and solvents. Ionic and organic liquids, molten metals and salts, and gases display fundamentally divergent properties from aqueous solutions, challenging currently established paradigms in actinide and radiation chemistry. We must understand how differences in the fundamental physical and chemical properties of these coolants and solvents influence their behavior under extreme conditions. Mastering these behaviors also requires the ability to dynamically characterize and manipulate the chemistry of solutes, including actinides, fission and corrosion products, and process molecules.

Elucidate and control the underlying physics and chemistry of interfaces in complex nuclear environments

Key question: *How do we harness dynamic interfaces to tailor robust materials and processes for next-generation nuclear reactors?*

Interfaces play a fundamental role in many innovative energy technologies. However, for nuclear energy applications, long-standing research challenges remain and must be overcome to harness the power of interfacial processes. Understanding interfacial properties associated with radiation effects in materials, radiolytic effects in coolants and solvents, plasma-materials interfaces in fusion reactors, and liquid–liquid interfaces in molten salts or liquid metals is essential to predict and mitigate degradation of materials for fission and fusion reactors. Ultimately, the design of new types of compositionally diverse and dynamically evolving interfaces is needed to enable and control the processes occurring in extreme environments.

Bridge multifidelity and multiresolution experiments, computational modeling, and data science to control dynamic behavior

Key question: *Which novel techniques can be coupled to provide operando and in situ measurements to better understand and control dynamical properties, behaviors, and processes for extreme nuclear energy environments?*

The dynamic evolution of nuclear material systems is intrinsically multiscale. Although the creation of radiation damage occurs over picoseconds, subsequent interactions occur over much longer times and lead to microstructural changes over multiple length scales. Similarly, the chemistry of molten salts, solvents, and coolants evolves following complex reaction networks whose elementary steps are realized over vastly different time scales. Clearly, the overall dynamic evolution of complex nuclear systems cannot be captured by isolated experimental probes and modeling approaches that only capture narrow time- and length-scales. Instead, a complete picture can be painted only by correlating and fusing multiple sources of operando experimental and modeling data, simultaneously characterizing the real-time evolution of a target system.

Harness artificial intelligence to design inherently resilient condensed phases







Key question: *What defines self-resilient mechanisms and how can they be discovered in nuclear materials and chemical systems in coupled extreme environments?*

A critical factor in the deployment of advanced nuclear reactors is the development of materials and coolants that are resilient against aggressive environments. However, the discovery, improvement, and assessment of materials resistant to extreme environments is complex and costly. Bottom-up multiscale modeling frameworks promise to aid experimental exploration of new materials. Unfortunately, such scale bridging is plagued by uncertainty propagation across scales and can incorporate only known physics, preventing the effective use of computational approaches for the discovery of novel materials. Breaking this conundrum necessitates the development and use of pioneering predictive methods enabled by data from high-throughput experiments coupled with machine learning and artificial intelligence.

1. Introduction

Clean energy is imperative for the future of the nation and of the world. Despite increased adoption of intermittent renewables such as wind and solar for electricity production, fossil fuels continue to dominate the world energy mix, comprising nearly 80% of overall energy production.^[1] Nuclear energy provides 10% of the world’s electricity and is the second-largest source of carbon-free electric power worldwide.^[2] In the U.S., 92 power reactors produce 18% of the nation’s electricity and half of the nation’s carbon-free power.^[3] In addition to its carbon-free advantage, nuclear power is the most reliable source of electricity available; it has a U.S. capacity factor^(a) of more than 90% (see **Sidebar 1**). Nuclear fission’s firm baseload power is critically important for reliable, economical energy supplies and grid stability today and into the future. Although still in their infancy in comparison with nuclear fission, nuclear fusion concepts further extend the promise of abundant, inexpensive, and carbon-free energy. Simply stated, nuclear energy can play a key role in enabling the future decarbonized energy system.

SIDEBAR 1—NUCLEAR ENERGY IN THE U.S. The U.S. is the world’s largest producer of nuclear power, accounting for more than 30% of worldwide nuclear generation of electricity. Nuclear is also the single largest source of clean power in the U.S., where the resulting emission-free electricity helps avoid more than 470 million metric tons of carbon each year. Although nuclear energy is recognized as a reliable, clean, and safe source of energy,^[4] almost all of the U.S. nuclear-generating capacity comes from reactors built between 1967 and 1990, and the challenges of sustained reliance on nuclear energy are associated with high construction and operation costs as well as public apprehension about safety and nuclear waste disposal. Higher efficiency and enhanced safety characteristics will only be achieved through continued fundamental science advancements. Additionally, fusion energy is seeing ever greater investment and promises to contribute to the U.S. nuclear energy portfolio.

	<p>Most reliable U.S. energy source: 93 nuclear reactors operate in 28 states with a 92.7% capacity factor</p>		<p>Powers over 70M U.S. homes: Nearly 800B kWh per year or 18% of total electricity in the U.S.</p>
	<p>Largest U.S. clean energy source: 50% of total carbon emission-free electricity</p>		<p>Large economic footprint: Nearly 0.5M U.S. jobs and \$60B/yr contribution to U.S. GDP</p>
	<p>Unmatched fuel energy density: 1 uranium pellet equivalent to 17,000 ft³ natural gas, 120 gal oil, or 1 ton of coal</p>		<p>Established safety: 1000x safer operating record relative to conventional energy sources</p>

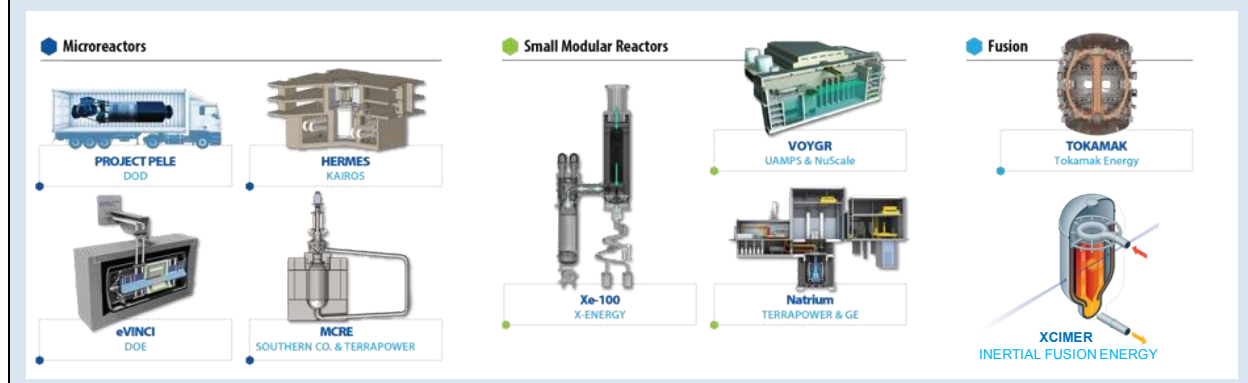
The future of fission nuclear energy will look very different from today’s suite of massive light-water reactor plants. Advances in fundamental science are enabling the development of simpler, smaller, and more nimble nuclear reactor designs, often operating at higher outlet temperatures

^(a) The capacity factor defines how often a given energy source is producing energy at maximum power.

(i.e., higher thermal efficiency) and lower pressures, that will produce either process heat or electricity for a variety of energy applications (see **Sidebar 2**). More-efficient fuel and advanced materials and designs will provide enhanced safety characteristics. The advantages associated with these new technologies will be realized through continued advancements in the fundamental science underpinning the knowledge of the physics and chemistry of nuclear interactions in materials.

In parallel, recent advances^[5] reinforce the potential of nuclear fusion technologies for future clean energy, but with technologies such as inertial confinement fusion, the path to commercial energy production is still long.^[6] Fundamental science advances are essential to build upon decades of R&D and to engineer fusion reactions into operating energy production facilities. Energy yield from controllable fusion technologies has increased significantly in recent years for both magnetic and inertial confinement fusion systems, but engineering energy parity is still an aspirational goal.^[7] The synergies between the fundamental science needed for practical fusion energy and enhanced fission systems are remarkable; enhanced scientific understanding will benefit both fission and fusion.

SIDEBAR 2—FISSION AND FUSION REACTOR CONCEPTS. Industry and government-funded R&D has produced numerous new fission reactor designs that have advanced to the development and demonstration stages. Microreactors are planned to be small (<30 MWe), factory-built, and transportable. Although the current nuclear fleet relies on light-water reactors (LWRs) that use low-enriched uranium (<5% ²³⁵U), most microreactor designs use high-assay low-enriched uranium (HALEU) fuel^(b) and have a variety of non-LWR core designs ranging from high-temperature gas reactors to molten salt reactors (MSRs). Proposed small modular reactors have larger electric output (a few hundred megawatts electric) and include concepts such as LWRs (VOYGR), high-temperature gas-cooled reactors (Xe-100), and sodium fast reactors (Natrium). The majority of commercial fusion concepts focus on magnetic plasma confinement, typically using tokamak designs, with a smaller number of companies exploring inertial confinement and other fusion concepts.



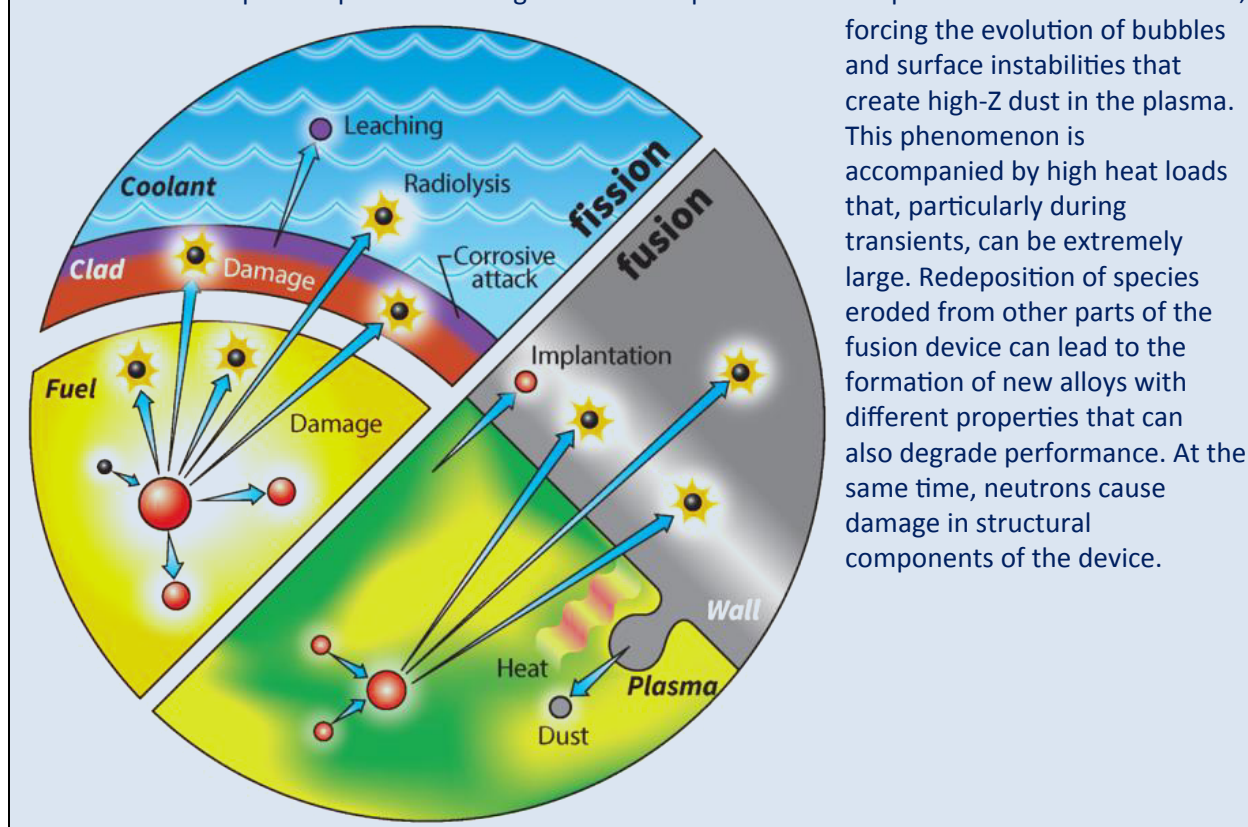
All nuclear energy systems—fission and fusion—are defined by some of the most extreme and hostile environments imaginable (see **Sidebar 3**). High temperatures and temperature gradients, extreme fluxes of energetic particles, radiolysis, corrosive attack, heavy elements, radionuclides, and large concentrations of generated and implanted gas species are just some of the features that define extreme nuclear environments. All components of these systems—the structural and fuel materials; the coolants, solvents, and liquid fuels; and the plasmas in fusion systems—experience dramatic evolution as a consequence of being exposed to these extreme conditions,

^(b) HALEU fuel, by definition, contains, or is enriched by, 5%–20% uranium-235. By contrast, LEU (low-enriched uranium) fuel has less than 4.95% uranium-235.

often to the detriment of performance. Materials exhibit severe changes in microstructure and phase structure upon irradiation, corrosion, and exposure to plasmas, often with macroscopic consequences such as swelling, embrittlement, and fragmentation. Liquids experience radiolysis and changing chemistry as new species are leached. Plasmas degrade as they are contaminated by dust from the structural materials. Understanding these effects presents unique challenges because in situ diagnostics are typically not robust enough to survive hostile environments or are unable to probe relevant regions of the system. Thus, an essential need exists for fundamental science to advance knowledge and capabilities of how these extreme environments affect the component materials and liquids and how researchers can measure that effect.

SIDEBAR 3—EXTREME ENVIRONMENTS. The condensed phases present in both fission and fusion environments are subjected to a number of extreme environments that limit their durability and, thus, their lifetime. In fission environments, radiation damage affects the fuel, structural materials, and coolant. At the same time, the coolant can corrode the solid materials, and these effects can be synergistic with radiation damage effects. Radiation damage produces large concentrations of nonequilibrium defects that not only drive microstructural and chemical evolution in the materials but also radiolysis in the liquids. Furthermore, those defects couple with the mechanisms that drive corrosive attacks. Species can leach from solids into liquids that change the chemical compositions and associated properties of both phases.

Similarly, in fusion systems, the interaction between the plasma and the wall is an extremely hostile environment. The plasma produces a large amount of particles that implant into the wall materials,



forcing the evolution of bubbles and surface instabilities that create high-Z dust in the plasma. This phenomenon is accompanied by high heat loads that, particularly during transients, can be extremely large. Redeposition of species eroded from other parts of the fusion device can lead to the formation of new alloys with different properties that can also degrade performance. At the same time, neutrons cause damage in structural components of the device.

In 2017, the Office of Basic Energy Sciences (BES) of the U.S. Department of Energy (DOE) hosted a workshop on the *Basic Research Needs for Future Nuclear Energy*. Participants identified five priority research directions (PRDs) in the areas of the design of molten salts and

liquid fuels, mastering hierarchical materials for nuclear environments, tailoring interfaces to control the impact of those environments, revealing multiscale processes in coupled extreme environments, and identifying and controlling rare events in extreme environments. Although these PRDs are still very much relevant, as discussed earlier, dramatic advances have occurred in nuclear energy systems since that report that demand a reassessment of basic research needs for the advancement of nuclear energy. At the same time, significant advances have occurred in computing, epitomized by the development of exascale computing platforms, algorithms, and software; in situ and operando sensing and diagnostics; and artificial intelligence (AI)/machine learning (ML) as applied to condensed phase systems to understand fundamental properties as well as discover new materials. Finally, recent years have seen a resurgence in novel materials such as complex concentrated alloys/chemically complex alloys (CCAs)^(c). All of these factors make a refresh of the 2017 report timely. These advances highlight four emerging scientific themes encompassing the many challenges posed by the development of modern nuclear technologies, listed in the following subsections.

Materials discovery and resilience in advanced nuclear systems: Materials in nuclear systems are subjected to extreme and complex environments over their lifetime. This environmental condition requires the development of specific and tailored property responses, for which the underpinning science is lacking. Multi-objective design and functionality of materials for coupled extreme environments will require new scientific approaches, including multimodal characterization of defects in materials. Although nuclear environments present a variety of hostile conditions, most materials have been studied and adopted for use from the point of view of their performance against one metric (e.g. radiation tolerance or corrosion resistance). Many materials that have shown exceptional performance in one metric come with remarkably poor performance in others. With the exception of cases such as Hastelloy,^[8] not many examples exist in which materials are simultaneously optimized against multiple performance metrics. Modern materials such as CCAs and heterostructured composites offer great promise for performance but are even more difficult to optimize against multiple metrics because the chemical and phase spaces are huge. Modern experimental and computational tools can help this issue. Similarly, understanding defects in materials, particularly those that are destined for use in extreme environments, is a grand challenge. Defects ultimately drive the evolution of materials in response to irradiation, corrosion, temperature, and stress. A complete understanding of these defects requires attacking them from multiple perspectives. Enhanced insight can be gained by integrating multiple experimental characterization techniques as well as computational approaches. The use of AI/ML can also bolster the quantity, quality, and speed of data generated through experiments and models. Combining multiple methods to enhance their strengths and minimize their weaknesses to more completely understand defects in materials will advance the ability to predict and design materials.

Physical processes and chemical evolution in nontraditional coolants and solvents: Advanced nuclear fusion and fission technologies propose the use of next-generation coolants and solvents as fuels or coolants in their designs to achieve enhancements in operational safety and efficiency.^[9] The incorporation of nonaqueous coolants and solvents is a transformational shift from the aqueous-based coolants and solvents currently employed. These next-generation coolants and solvents depart significantly from the thermodynamics of ideal solutions, making a description and prediction of their chemistry a scientific challenge. Therefore, this challenge requires fundamental knowledge of their chemical behavior and the structure and energetics of the solutions they form under the extreme conditions of heat, radiation fields, and compositional complexity. Current descriptions of solution chemistry and the understanding of the speciation,

^(c) These types of alloys are alternatively called high-entropy alloys (HEAs) or multiprincipal element alloys (MPEAs) depending on the details and the author.

reactivity, and interactions among solutes such as actinides and fission and corrosion products in these solvents require reevaluation. These nontraditional solvents represent new chemical systems and opportunities in which understanding interactions across multiple energy, length, and temporal scales is needed to sense, predict, and control their behaviors. Understanding the basic science of these complex systems in extreme radiation environments will lead to robust and specialized interrogation techniques that can probe multiple scales of time and length.

Spectroscopic signatures and in situ sensing under extreme conditions: The dynamic evolution of materials and processes within nuclear systems is inherently multiscale, spanning broad energy, length, and temporal landscapes. The overall dynamical evolution of complex nuclear systems cannot, therefore, be elucidated by isolated experimental probes and modeling approaches that only capture narrow time and length scales. Instead, a thorough understanding will only be achieved through the correlation and integration of multiple sources of operando experimental and modeling data, simultaneously characterizing the real-time evolution of a target system. The development of new instrumental probe technologies for in situ—including in-reactor—and ex situ sensing, combined with multimodal analysis frameworks that link these characterization experiments with models, is necessary to describe and predict processes at play and ultimately tune and control the dynamical evolution of nuclear systems.

Computationally-driven discovery and accelerated data generation: A variety of computational advances that accelerate modeling, simulation, and data analysis will improve the understanding of material and chemical properties associated with nuclear energy systems. Advances in AI/ML are particularly important; these fields are opening new opportunities to predict and interpret novel thermodynamics and kinetics phenomena related to condensed phases in extreme environments. Successful use of AI/ML in nuclear materials science requires the fusion of multiple data streams, uncertainty quantification and propagation, high-throughput computation, and scientific guidance of AI/ML algorithms. Nuclear materials science is also too data-poor to apply pure *big data* techniques to predict structure–property relationships, necessitating physics-based guidance for AI/ML application. AI/ML techniques provide more flexible numerical parameterizations compared with physics-based models. Additionally, AI/ML enables data extrapolation from simpler systems to extremely complex systems for which computations using physics-based simulations are daunting. When applied to multiple data types for a specific phenomenon of interest, fusing different types of data streams will allow for the extraction of scientific knowledge. At the same time, high-throughput data generation, whether by simulation or next-generation scattering facilities, is critically needed for simpler relationships that can be harnessed by combined AI/ML and physics-based algorithms. Nuclear systems require systems knowledge across orders-of-magnitude variations in both time and length. AI/ML techniques are useful for bridging those scales and in quantifying uncertainties at each scale and then propagating those uncertainties across scales. Finally, AI/ML techniques will aid in identifying features of interest from both experimental (e.g., transmission electron microscopy [TEM] images) and computational (e.g., molecular dynamics) datasets. At the same time, advances in exascale computing, both in terms of available computing power and the advent of algorithms that can use such resources, provide new opportunities for generating novel simulation data that provide fundamental insight as well as data to train AI/ML models.

The roundtable: The DOE BES Roundtable on “Foundational Science to Accelerate Nuclear Energy Innovation” was held July 20–22, 2022. The roundtable participants were charged with defining the new insights needed from basic research to enable future scientific and technological advances in materials and chemistry that are required for advanced nuclear energy systems, including both fission and fusion. BES led the roundtable, in coordination with the Offices of Advanced Scientific Computing Research (ASCR), Fusion Energy Sciences (FES), and Nuclear Energy (NE), to establish fundamental scientific research objectives related to accelerating

progress in fission and fusion energy systems. Concerted engagement from multiple offices of DOE was instrumental in establishing a focused framework for roundtable participants to discuss how to overcome current scientific and technical barriers in advanced nuclear energy systems, as well as to ensure that the fundamental scientific directions had relevance to the technical concerns of the relevant stakeholders. As a result, five priority research opportunities (PROs) were identified, adding to the PRDs outlined in the 2017 report. Together, these PRDs and PROs define the directions that will establish the foundational science needed to advance innovation in nuclear energy technologies:

- Master complex electronic structures to tailor thermochemical reactivity, transport, and microstructural evolution.
- Interrogate and direct the physics and chemistry underpinning next-generation coolants and solvents.
- Elucidate and control the underlying physics and chemistry of interfaces in complex nuclear environments.
- Bridge multifidelity, multiresolution experiments; computational modeling; and data science to control dynamic behavior.
- Harness AI to design inherently resilient condensed phases.

Since the advent of nuclear energy technologies in the post-World War II era, the U.S. has led the global development and deployment of peaceful nuclear energy systems. Maintaining and enhancing that leadership is critical for energy production and nuclear security around the globe. Investing in the science foundational to nuclear energy technologies, both fission and fusion, cements the nation's leadership position and provides important opportunities to lead innovation on the world stage. Because new nuclear plants have the potential to be *dropped in* locations of retiring fossil fuel facilities, enhanced use of nuclear energy for electricity need not require massive new infrastructure development. Using nuclear as a source of heat for industrial processes and energy storage provides previously unimagined levels of flexibility in future energy systems.

2. Priority Research Opportunities

PRO 1: Master Complex Electronic Structures to Tailor Thermochemical Reactivity, Transport, and Microstructural Evolution

Overarching Question: How do researchers elucidate, predict, and harness coupled electron–ion dynamics to enable the discovery and deployment of novel materials, coolants, and solvents for future fission and fusion energy?

Extreme irradiation and corrosive environments in advanced nuclear systems are well-known to affect chemical and materials phenomena at the nano- through mesoscale. The underlying mechanisms controlling these phenomena have historically been described through atomic and molecular level interactions, with only limited consideration for subatomic electron structures. However, electronic interactions fundamentally govern the nature and energy of bonding, transport, and defect energies; phase stability in solids and liquids; and thermophysical properties. Thus, understanding the bidirectional relationship between electronic structure and microstructure—and their evolution under extremes—is essential to the design and discovery of novel materials, coolants, and solvents for advanced nuclear systems.

Specifically, upon completion of a fission or fusion nuclear reaction, electromagnetic forces are the only fundamental forces actively controlling the fate of reaction products such as defects, decay products, ionization products, primary knock-on atoms, radiolysis products, gases, and fission fragments. Thus, bridging quantum physical considerations of electron–ion and electron–electron interactions to bulk phenomena such as mass and thermal transport, including micro-through mesoscale restructuring, is necessary to enable understanding of material evolution under extreme, inherently nonequilibrium conditions. To achieve this goal, electronic interactions and their relationships to atomic and molecular structures must be elucidated to predict phase dynamics and thermophysical properties of liquids and solids in nuclear systems. In other words, explicit consideration of electronic structure and dynamics is critical for a fully descriptive and predictive understanding of nuclear condensed phases. Harnessing the science of electron–ion and electron–electron interactions will subsequently enable tuning of materials properties and performance under coupled external stimuli.

1a. Summary

Future nuclear fission and fusion energy systems require advances in nuclear materials, coolants, solvents, and fuels designed to perform safely, reliably, and economically through coupled thermal, corrosive, and radiation extremes. Basic research over the past two decades has made tremendous progress toward identifying the physical mechanisms that describe how materials evolve, restructure, and inevitably degrade in nuclear extremes and have consequently pushed the boundaries of practical engineering design limitations. This research has revealed a hierarchical relationship between atomic/molecular arrangements, micro- and mesostructure evolution, and their implications on materials properties and performance. However, many physical phenomena in nuclear chemistry and materials remain unexplained because this length-scale hierarchy has yet to capture subatomic electronic structures and dynamics.

Focusing on electronic structures and transitions presents an opportunity to accurately understand solid- and/or liquid-state evolution by bridging subatomic structural impacts to thermophysical and thermochemical properties under inherently nonequilibrium conditions, moving beyond fundamental equilibrium thermodynamics. This research opportunity motivates investigations of phenomena including mass transport effects that govern the accommodation of

light elements (e.g., hydrogen and helium) and fission products; excited state dynamics and kinetics that influence the development of irradiation damage cascades and defect accumulation; and physical properties (e.g., thermal, mechanical) of many-electron species in solids, solvents, and at complex interfaces over varied length and time scales.

Recent research^[10] has demonstrated numerous examples of multielectron solids and liquids—including metals, intermetallics, ceramics, glasses, polar compounds, and nonpolar compounds—that exhibit varied resilience to reactive environments and irradiation.^[11] These materials hold promise for enabling novel reactor designs and closing the nuclear fuel cycle^(d). However, a unifying explanation does not exist for the evolution of these varied material classes in nuclear extremes. Such a theory, if it exists, should be founded on the fundamental subatomic interactions common across all of these material classes.

A demonstrative example of the underlying subatomic nature of materials' resilience under extremes is the concept of ion irradiation damage in solids. Several key subatomic factors are at play. First, irradiating particles transfer their kinetic energy to the target material through nuclear and electronic energy losses (which itself is typically not treated in any fundamental manner). The relative extent of each is dependent on the incident ion energy at a given point along its trajectory. Additionally, at least some of the injected ions will come to rest within the target material, introducing additional interstitials and electronic charges that must be accommodated. Consequently, complex, multilayered defect microstructures are generated with different layers corresponding to differing levels of nuclear stopping, electronic stopping, and injected ions,^[11] an example of which is shown in **Figure 1**.

In ceramic nuclear fuels, the additional introduction of extreme gradients in temperature and fission density lead to even further microstructural complexity known as the high-burnup structure (HBS), as detailed in **Sidebar 4**. Thus, electron-level physics play a critical role in the creation of irradiation damage, the subsequent mass transport to form irradiated microstructures, and the corresponding thermophysical property evolution. Understanding these foundational physical principles will enable researchers to explain the unique irradiation damage responses—and

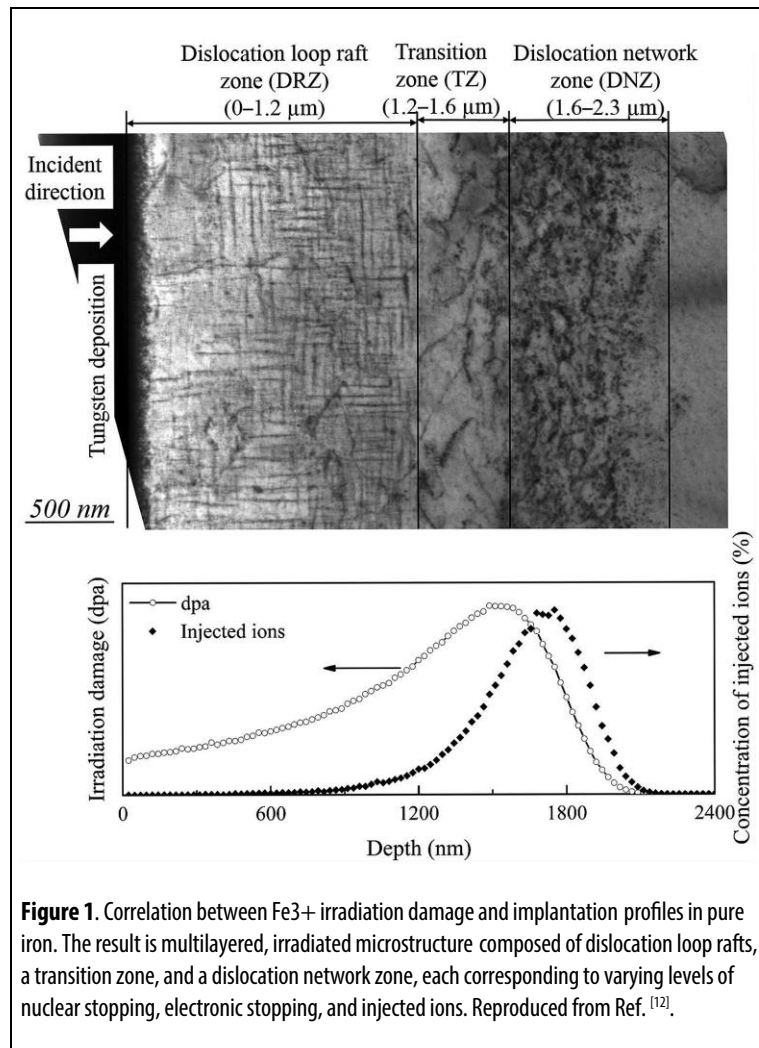
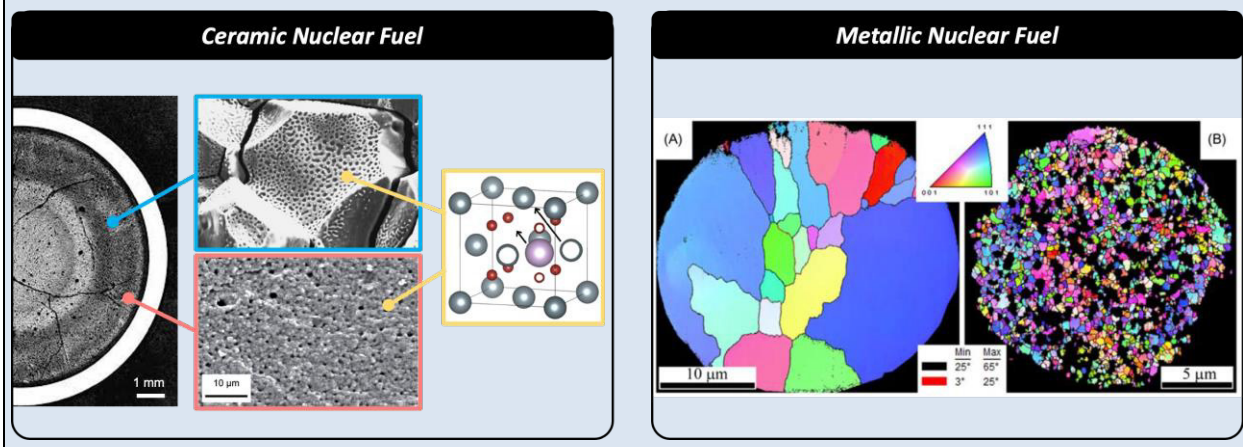


Figure 1. Correlation between Fe³⁺ irradiation damage and implantation profiles in pure iron. The result is multilayered, irradiated microstructure composed of dislocation loop rafts, a transition zone, and a dislocation network zone, each corresponding to varying levels of nuclear stopping, electronic stopping, and injected ions. Reproduced from Ref. ^[12].

^(d) A closed nuclear fuel cycle is one in which the spent fuel is reprocessed and reused.

subsequently design irradiation damage resilience—across structural metals, ordered intermetallic structures, and compounds exhibiting varying degrees of bonding ionicity or covalency.

SIDEBAR 4—HIGH-BURNUP STRUCTURE. The high-burnup structure (HBS) in oxide nuclear fuels is among the most complex materials challenges in nuclear systems because its genesis inherently links multiscale structures (i.e., bridging from the electronic to the mesoscale) to multiparameter operating environments with considerations for fission density, fission gas release, and thermophysical properties. As shown in the blue boxed regions of the ceramic fuel figure,^[13] intergranular porosity occurs because of fission gas bubble nucleation, whereas in the pink boxed regions,^[14] a combination of extreme-grain nanostructuring and gas bubble nucleation occurs. Density functional theory simulations have predicted the equilibrium positions and diffusivities of fission gas atoms in oxide lattices, but these predictions have not yet been comprehensively linked to the fuel restructuring and its implications on fuel pin integrity. Metallic fuels lend some insight to bridge electronic systems and enable the prediction of HBS in which a more uniform void structure is observed, as shown in the bottom right image,^[15] potentially because of a more uniform thermal profile associated with the metallic nature of bonding. Researchers have identified the structures that form in oxide fuels as well as the approximate burnup level at which they begin to be observed. However, there remains no mechanistic understanding for why these structures form and the thresholds for which they appreciably degrade the fuel form. A fundamental understanding is needed to map these phenomena to fuels with varied electronic and microstructures, as well as for reactors with alternative fission spectra.



The length and timescales of electronic interactions have thus far limited researchers' abilities to characterize and predict these behaviors. However, the advent of exascale computing has improved the efficiency and accuracy of relativistic methods. The growth and adoption of AI methods (**PRO 5**) also improve the accuracy of interatomic potentials of many-electron systems and enable extraction of a multitude of data from theory and experiments. Additionally, improvements in length- and timescale resolution in electron microscopy, beamline or scattering techniques, and in situ monitoring enable breakthroughs in experimental evaluation of dynamic behaviors of electronic structures in extreme environments, which will be discussed in **PRO 4**. Collectively, these recent advancements in experimental and computational techniques present a timely opportunity to bridge length and timescales to elucidate and tailor nonequilibrium phenomena under combined extremes.

1b. Key Scientific Questions

- How do varied, possibly nonequilibrium or metastable, electronic structures evolve under the coupled extremes of high temperature, corrosion, and irradiation? Can a single unifying theory capture these electronic effects across all material classes of interest to advanced nuclear systems?
- How can one predict the fate of fission, fusion, and corrosion products in materials as a function of electronic structure?
- What is the interplay and hierarchy among thermal, electronic, and ionic or atomic disorders? What is the effect of this hierarchy on micro- and mesoscale evolution under nonequilibrium dynamics?
- How do the subatomic bond and electronic structure control the formation and evolution of irradiation defects? Conversely, how do defects affect bonding and electronic structure?
- How do subatomic electron structures affect post-irradiation properties and performance, such as deformation mechanisms, order/disorder transformations, mass and thermal transport, or trapping of fission products?

1c. Scientific Challenges and Research Opportunities

- Advance experimental approaches capable of elucidating electronic structures, dynamics, and transitions under fission and fusion extremes.
- Cultivate accurate and efficient computational methods capable of modeling and predicting electronic structures, dynamics, and transitions, accounting for their evolution over length and time scales relevant to fission and fusion environments.
- Devise novel and integrated approaches that can correlate electronic structures to defect structures, and subsequent evolution of thermophysical properties, bond nature, and local environment.

Advance experimental approaches capable of elucidating electronic structures, dynamics, and transitions under fission and fusion extremes.

Direct experimental measurements of the electronic structure of materials are inherently challenging because of resolution limits associated with subatomic length scales. The dynamic evolution of materials at the electronic structure level under dynamic processes, such as irradiation damage or under steep thermal gradients, is even more difficult to capture because of the attendant picosecond timescales.

Understanding fundamental irradiation damage physics in materials particularly requires techniques to probe the electronic structure evolution (or infer this evolution from thermal transport measurements) as well as the role of electronic structure on the development of irradiation damage. Until now, researchers have tended to explain irradiation damage somewhat phenomenologically because nuclear and electronic energy losses can have additive effects on the creation of irradiation damage in some materials, and they can have competitive effects in other materials.^[16] However, recent work has suggested that energy dissipation processes must consider the temporal and spatial coupling of the electronic and ionic subsystems^[17] if a unifying, material-agnostic theory is to be realized. In structural metals, for example, irradiation damage via nuclear stopping is generally understood through the binary collision approximation, which is based on simplified Newtonian physical assumptions. Consequently, irradiation damage predictions that are valid for structural alloys are not applicable to intermetallic compounds or concentrated solid-solution alloys exhibiting chemical ordering or complexity,^[18] let alone compounds having varying degrees of ionic and covalent bonding or electronic localization (i.e., bond directionality).^[17, 19]

Electronic structure and its evolution are also critical to understanding mass transport phenomena in advanced nuclear systems. In fusion reactors using deuterium–tritium fuel, for example, considerable generation of solid and gaseous transmutants is expected in materials exposed to 14 MeV neutrons. In particular, iron-based alloys will be subjected to significant accumulation of helium and hydrogen atoms,^[20] leading to far-from-equilibrium precipitation of these species into bubbles and voids, and segregation to grain boundaries, precipitate interfaces, and dislocations.^[21] In tungsten alloys used in the divertor, high concentrations of transmutation rhenium, osmium, and platinum lead to accelerated precipitation of nonequilibrium, brittle intermetallic phases that severely affect materials' performance.^[22] Understanding the link between transmutant accumulation and their interactions both between themselves and with the material microstructure is also an important electronic structure problem with implications for reactor performance, operation, and lifetime. Similarly, in fission reactors, the nonequilibrium mass transport of hydrogen, transmutation products, and fission gases—through fuel and structural materials—is at the most fundamental level also dependent upon the electronic structure and spin-polarization.^[23] Finally, macroscopic phenomena such as silver transport in tristructural-isotropic (TRISO) fuels^[24] and fuel-clad chemical interactions^[25] require experimental linkage to fundamental electronic structures to elucidate the mechanisms for observed, thermodynamically unexpected transport of fission products.^[26]

Advanced experimental techniques are necessary for characterizing electronic structure evolution, particularly in the presence of defects. Pair distribution function (PDF) analysis is a rapidly advancing technique for studying short- to medium-range order in materials.^[28] X-ray PDF analysis can reveal structural evolution under irradiation (**Figure 2a**).^[12] However, electron PDF, which can be conducted within a transmission electron microscope, is especially well-suited for materials having weak Bragg diffraction and higher diffuse scattering, such as disordered or amorphous materials and nanoparticles.^[29] More recently, atomic electron tomographic reconstruction methods have experimentally determined the 3D atomic positions and short-through medium-range order of disordered solids.^[30] Finally, positron annihilation spectroscopy is a powerful technique for determining the population and character of irradiation-induced defects in irradiated materials (**Figure 2b**).^[27, 31]

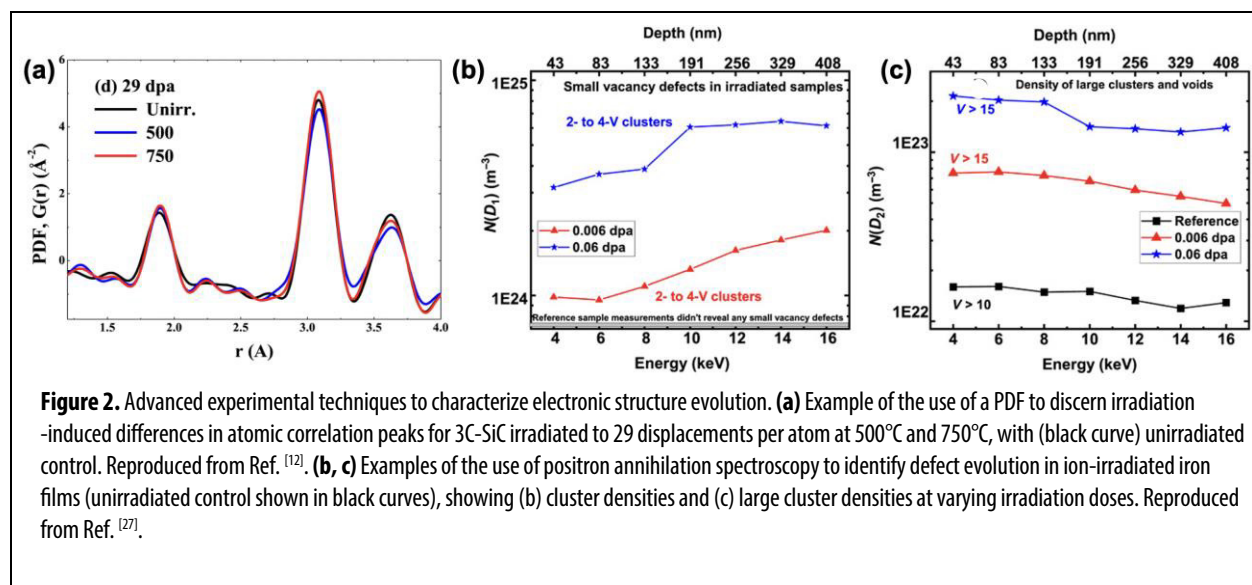


Figure 2. Advanced experimental techniques to characterize electronic structure evolution. **(a)** Example of the use of a PDF to discern irradiation-induced differences in atomic correlation peaks for 3C-SiC irradiated to 29 displacements per atom at 500°C and 750°C, with (black curve) unirradiated control. Reproduced from Ref. ^[12]. **(b, c)** Examples of the use of positron annihilation spectroscopy to identify defect evolution in ion-irradiated iron films (unirradiated control shown in black curves), showing **(b)** cluster densities and **(c)** large cluster densities at varying irradiation doses. Reproduced from Ref. ^[27].

Cultivate accurate and efficient computational methods capable of modeling and predicting electronic structures, dynamics, and transitions, accounting for their evolution over length and timescales relevant to fission and fusion environments.

Accurately and efficiently capturing ion–electron interactions in computational and theoretical space is a persistent challenge that hinders researchers from precisely understanding defect, phase, and compound evolution under extreme environments. Even under thermodynamic equilibrium conditions, high-quality interatomic potentials for many-electron species typically do not exist. Consequently, inconsistencies exist between the phase diagrams and synthesized structures of 5f-electron compounds for solid- and liquid-phase nuclear fuel systems. For example, inaccuracies abound in the phase diagrams of uranium-bearing materials and fluids such as the systems of U–Si,^[32] U–Zr,^[33] and NaCl–UCl₃.^[34] Recent thermodynamic modeling has helped improve the accuracy of phase boundaries and identify new phase regimes (**Figure 3a** and **Figure 3b**).^[33a] Nevertheless, experiments persistently reveal inaccurate temperatures and compositions for phase boundaries, as well as previously unknown miscibility gaps.^[33b, 35] Another relevant example is that of iron and tungsten alloys being considered as structural materials in fusion devices. These materials are characterized, among other things, by the open structure of their crystal lattices, which is created by the nonmetallic bonding of their d- and f-electrons.^[36] This structure questions the use of interatomic potentials based on formulations for close-packed metallic systems and opens the way to electronic structure–augmented theory formulations for these materials such as bond-order potentials,^[37] the tight-binding method,^[38] or Hubbard-corrected density functional theory (DFT) approaches.^[39]

Subsequently subjecting these unpredictable many-electron compounds and complexes to nonequilibrium thermodynamic environments, such as irradiation under steep thermal gradients, further exacerbates and underscores the gaps in understanding of phase stability and evolution, including the effect of metastable phases and states of matter. For example, irradiation of solid actinide compounds accelerates phase transformations in a currently unpredictable manner and with unexpected morphologies.^[40] Fission products and hydrogen also strongly influence phase stability of actinide compounds because of the subatomic interactions and accommodations within the lattice (**Figure 3c**).^[35, 41] Beyond solid-phase stability, there are also gaps in understanding the evolution of liquid-phase fuels and coolants such as molten salts under irradiation extremes. Specifically, including subatomic interactions is necessary for establishing phase diagrams for molten salts, understanding actinide solubility, and accurately predicting the evolution of these systems under irradiation.^[42]

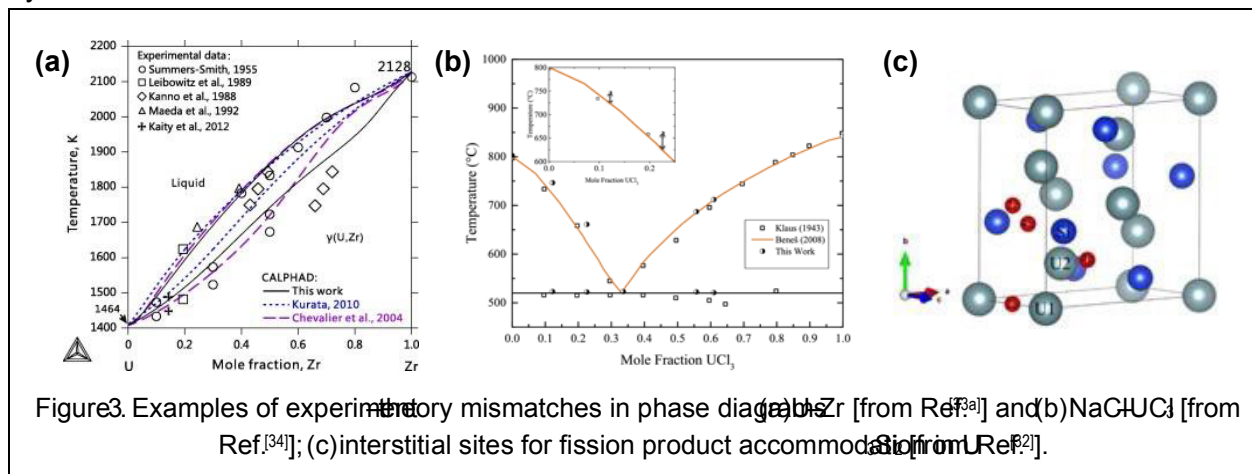


Figure 3. Examples of experimental theory mismatches in phase diagrams (a) U–Zr [from Ref.^[33a]] and (b) NaCl–UCl₃ [from Ref.^[34]]; (c) interstitial sites for fission product accommodation [from Ref.^[42]].

Current approaches include coupling finite difference methods with two-temperature models,^[43] but these approaches are generally used for metals and are less effective for solids having directional bonds and varying degrees of ionicity. Furthermore, they are typically challenging to parameterize for arbitrary material choices. Time-dependent DFT methods are effective and state-of-the-art for predicting electron–ion effects but are computationally expensive for many-electron systems and have limited applicability beyond *ab initio* conditions. Furthermore, extrapolating *ab initio* results to high-temperature is nontrivial. An opportunity exists to develop highly accurate reactive interatomic potentials and integrate them with fast ML methods to efficiently use data from DFT for training.^[44] Accurate low-scaling approaches that go beyond DFT and take into account important relativistic electron effects are crucial,^[45] which may also be useful in providing training data for ML methods. In some cases, accounting for excited electronic behavior may also be important.

Devise novel and integrated approaches that can correlate electronic structures to defect structures and subsequent evolution of thermophysical properties, bond nature, and local environment.

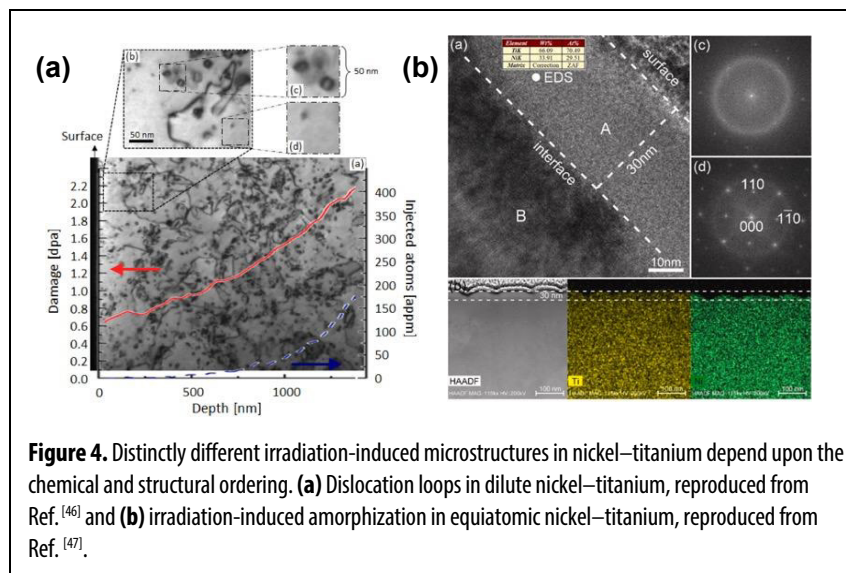
At the most fundamental level, the generation, stability, and mobility of defects in liquids and solids is governed by the electronic structure. Because these defects ultimately determine the evolution of thermophysical properties and microstructure development, researchers must be able to experimentally and computationally correlate electronic structures to larger length- and timescale structures and properties.

Building upon the nonuniversality of irradiation damage creation in materials having different electronic and bond structures discussed previously, the subsequent evolution of irradiated microstructures is also highly variable. The role of chemical and bond ordering on the development of the microstructure can be clearly observed in nickel–titanium alloys (**Figure 4**).

Nickel systems with dilute titanium have a face-centered cubic crystal structure without chemical ordering and are able to develop dislocation loops under irradiation.^[46]

However, increasing titanium concentration forms a B2 ordered crystal structure. Irradiation-induced dislocation loops cannot be stabilized, but instead, amorphous regions form.^[47]

The underlying explanations for these divergent irradiated microstructures likely lies within the subatomic bonding and electronic structures that govern ordering.^[48]



Understanding electron–structure–property relationships will also enable researchers to harness these relationships to tailor the stability and performance of materials and interfaces to enable novel nuclear systems. For example, some uranium compounds demonstrate improved electronic and thermal transport compared with traditional fuels largely because of electronic and phononic structures. Researchers have hypothesized that the increased thermal and electronic transport

will lead to a more stable microstructure under irradiation. A unified methodology for evaluating and predicting electron–structure–property relationships and evolution will enable the design of stable and robust fuel structures. Looking to actinide-bearing compounds with enhanced thermal conductivity, unirradiated intermetallic and metallic compounds notably facilitate improved performance with respect to heat transfer and reduced thermal stresses. However, the evolution of these compounds under irradiation in comparison with oxide fuels is largely unknown at the fundamental electronic structure level.^[49]

In practice, novel material discovery has traditionally faced challenges to implementation because of the timescales and costs required for the collection of tremendous volumes of experimental data, with some applications requiring 5–15 years of experimentation prior to industrial consideration for nuclear applications in the current regulatory environment. The nuclear materials community needs complementary computational and experimental capabilities that shorten the time between materials discovery and deployment.^[50] Advances in AI, lower length-scale modeling, and computational techniques that bridge the mesoscale to the engineering scale could screen promising materials for further experimental interrogation, reducing the overall burden on experimental data collection and accelerating the time to discovery of novel materials. In this way, synergistic maturation of computational, experimental, and regulatory approaches will facilitate the needed, abridged time to implementation for novel, promising material, coolant, and solvent systems.

1d. Potential Impacts

Scientifically, this PRO will enable an understanding of the interplay between varied electronic structures and their transformations under coupled extremes. This opportunity will enable researchers to build physical models of nonequilibrium phenomena that consider electron–ion states while bridging from the short length and time scales of electronic interactions to the longer length and time scales of mesostructural evolution. This opportunity can provide a fundamental understanding of the reactivity of many-electron systems in condensed phases and at interfaces, as well as an understanding of hydrogen and helium mobility and accommodation in fission and fusion systems. The explicit inclusion of electronic behavior and properties into the multiscale paradigm of radiation damage simulation will provide a more physical foundation for the understanding and prediction of radiation damage in nonmetals and allow for improved understanding of, for example, ionization processes and the effect of ionized defects on optical and electronic properties.

Research addressing this opportunity will have far-reaching effects across energy technologies, specifically through the improved insight into the possibility for electronic structure tailoring for material, chemical, and solvent resiliency under extreme environments. This research opportunity will improve the accuracy of service life predictions and maximize the performance of nuclear systems by providing the underpinning science necessary for designing novel and optimized materials, as well as enabling shortened material qualification timelines. Work will advance the state of knowledge of the nuclear fuel chemistry for next-generation reactor technologies and will enable technologies to close the nuclear fuel cycle. Research can be applied even more broadly to correspondingly advance hydrogen sequestration and storage technologies because a greater understanding of hydrogen accommodation and transport in materials will translate to those technologies as well.

PRO 2. Interrogate and Direct the Physics and Chemistry Underpinning Next-Generation Coolants and Solvents

Overarching Question: How can researchers probe and control the physics and chemistry of coolants, solvents, and their solutions in the harsh environments associated with nuclear energy?

Advanced nuclear fission and fusion technologies require next-generation coolants and solvents that possess fundamentally divergent physical and chemical properties from traditional dilute aqueous solutions. The behavior of these next-generation media challenges currently established paradigms in actinide and radiation chemistry. Understanding the properties of these media will have a broad scientific impact, including the ability to determine the efficiency, longevity, and safety of future nuclear energy technologies. Molecular-scale heterogeneity of the media, their chemical complexity, and nonequilibrium conditions owing to multicomponent intense radiation at high temperature and strong electric and/or magnetic fields are expected in advanced nuclear fission and fusion reactors and throughout the nuclear fuel cycle. The chemistry, stability, and transport of actinides, fission and corrosion products, process molecules, and impurities in these next-generation media are unknown. Access to the fundamental physics and chemistry governing the behavior of these next-generation media and their solutions requires new knowledge of their fundamental interactions along with new capabilities to probe these fluids over multiple length and temporal domains. Thus, the basic science underpinning a new generation of sensors as well as novel applications of sensors are additionally needed to interrogate the physics and chemistry underpinning next-generation coolants and solvents.

2a. Summary

Advanced nuclear fission and fusion technologies require next-generation coolants and solvents (e.g., ionic and organic liquids, molten metals and salts, and gases) to take full advantage of technological developments since the initial deployment of water-cooled reactor and fuel management (separations and storage) technologies in the U.S. over 70 years ago.^[52] Next-generation coolants, solvents, and their solutions possess fundamentally divergent physical and chemical properties from aqueous solutions. The chemistry of solutes in dilute aqueous solutions can typically be described by existing equilibrium thermodynamic models for ideal solutions that are homogenous and where the energetic interactions among the components are small or weak. In contrast, liquid metals, molten salts, and ionic liquids are known to be nonideal solutions in which energetic interactions among the solute and solvent are large, and distinct domains emerge on the molecular scale as shown in **Figure 5**.^[51] Organic solvents and highly concentrated aqueous solutions—where water is essentially a solute—also offer unique physics and chemistry. The nonideal behaviors and properties of these fluids as solvents challenge current descriptions of solution chemistry and understanding of the speciation, reactivity, and interactions among solutes such as actinides, fission and corrosion products, and process molecules in these solvents. The efficiency and longevity of nuclear technologies employing these next-generation solvents is predicated on mastering their chemistry. Therefore, researchers must achieve a detailed, multiscale, mechanistic understanding of how differences in the fundamental physical and chemical properties of next-generation coolants and solvents influence solute behavior in

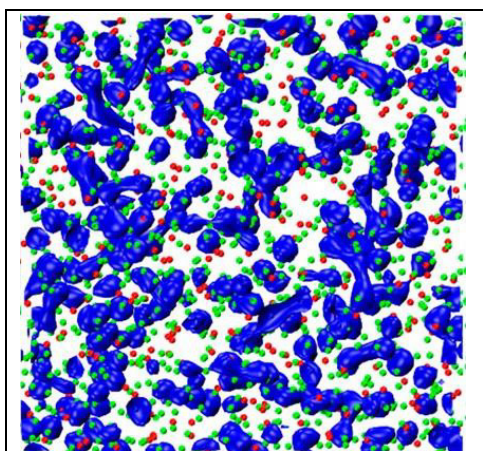


Figure 5. Heterogeneity arising from a simple mixture of monovalent and divalent cations in molten $\text{MgCl}_2\text{-KCl}$.^[51]

extreme environments at high temperatures, in complex chemical environments, and in the presence of intense multicomponent ionizing radiation as well as electric and magnetic fields. New knowledge in this area will enable the *interrogation, prediction, and control* of solution chemistry in nonideal, next-generation media.

Connecting these fundamental knowledge gaps necessitates a systematic investigation into the energetic relationships within these media across multiple length and timescales from the molecular through the mesoscale to the bulk scale. Access to this new knowledge requires innovative in situ and real-time interrogation techniques (which are also discussed in **PRO 4**), the basic science for which is currently lacking in the extreme chemical and ionizing radiation environments encountered in nuclear energy systems.

The effects of ionizing radiation are ubiquitous throughout all aspects of a nuclear fuel cycle or energy system. However, the complexity and intensity of these effects are greatest during reactor operations and in the management of used nuclear fuel owing to the presence of a wide spectrum of radionuclides from neutron capture, fission, and fusion processes. These nonequilibrium, radiation-induced processes are initiated by the ionization and/or excitation of an absorbing species (bulk coolant/solvent and concentrated/solute) at femtosecond timescales, leading to a cascade of chemical reactions that involve a suite of radical, ionic, and/or molecular species. These initial radiolysis products are typically highly reactive and have the capacity to initiate chemical processes in the surrounding medium, often leading to the formation of nonequilibrium metal ion oxidation states and radiolytic degradation that affect the physical and chemical properties of the irradiated medium and, ultimately, the performance and longevity of nuclear fuel cycle technologies. Consequently, the fundamental science underpinning radiation-induced processes in next-generation coolants and solvents must be mastered.

2b. Key Scientific Questions

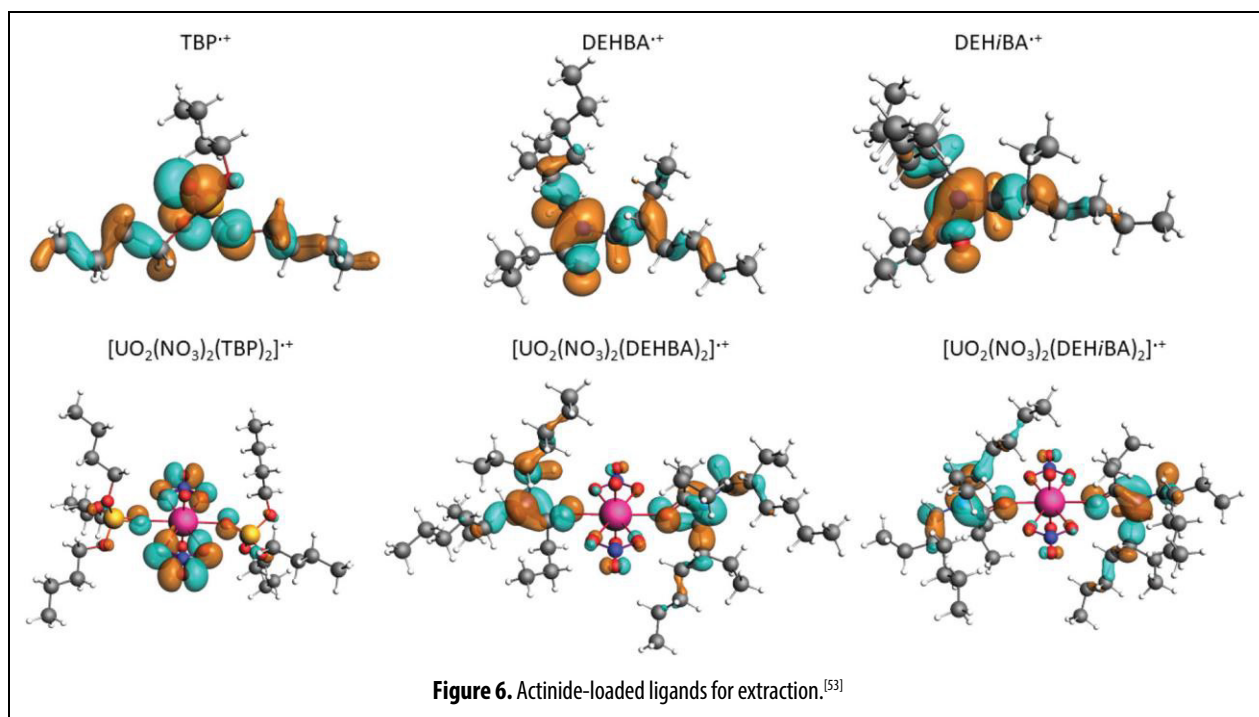
- How do researchers elucidate the molecular structures and thermodynamic properties of next-generation coolants and solvents, and how do these structures and properties evolve in extreme (high-temperature, chemically complex, and intense electric and magnetic fields) ionizing radiation environments?
- Can the speciation, redox chemistry, chemical kinetics, stability, and transport of solutes (actinides, fission and corrosion products, impurities, and process chemicals) be predicted in next-generation coolants and solvents?
- What basic scientific knowledge is required to design and develop sensors to monitor complex, multicomponent media in extreme chemical and ionizing radiation environments over multiple length and timescales?
- Can new theories and computational models be developed to explain observations, guide future experiments, and ultimately predict the evolving physics and chemistry of next-generation media in extreme ionizing radiation environments?
- How do heterogeneous systems such as particles, clusters, and interfaces respond to next-generation coolants and solvents and their solutions exposed to extreme multicomponent radiation fields?

2c. Scientific Challenges and Research Opportunities

- Characterize and understand the reactivity of next-generation coolants and solvents under extreme conditions.
- Establish the speciation and reactivity of solutes in next-generation solvents and coolants.
- Develop robust and specialized analytical techniques amenable to in situ and real-time sensing.

Characterize and understand the reactivity of next-generation coolants and solvents under extreme conditions.

In advanced nuclear technologies, next-generation coolants and solvents will take advantage of the thermal properties of molten metals (e.g., Pb, Pb and Bi, and Na), salts (e.g., multicomponent fluorides and chlorides), or noble gases (e.g., He and Ar).^[52a, 54] Not only will these next-generation coolants be exposed to intense, multicomponent (combinations of alpha, beta, and gamma radiation; x-rays; neutrons; and fission fragments) ionizing radiation fields, but high temperatures and intense electric and magnetic fields are also envisioned, the latter specifically for fusion reactor concepts. Each next-generation coolant will behave differently under these extreme ionizing radiation environments, where the wealth of practical experience for water obtained in pressurized and boiling water reactors will provide little to no useful insight. For example, heavy liquid metal (HLM) coolants are being considered for fission and fusion energy applications because of their excellent neutronic, thermal hydraulic, and thermophysical properties. However, HLMS can cause significant degradation to structural materials. Corrosion and liquid metal embrittlement are the two major degradation effects caused by structural materials being in contact with HLMS.



The back end of a nuclear fission fuel cycle currently involves a variety of separation systems to reclaim materials for reuse as reactor fuels and to improve waste management. To date, separation technologies have been dominated by hydrometallurgical methods that employ biphasic solvent systems, typically highly concentrated, aqueous HNO_3 in contact with complex organic solutions (**Figure 6**). Pyrometallurgical methods that use molten salt mixtures have also been employed at scale but to a much lesser extent. Innovative development of efficient and economical advanced separation technologies is underway to adapt to next-generation nuclear fuel cycle needs.^[55] This effort aims to expand the use of nonaqueous solutions and solvent systems to other media such as ionic liquids^[56] or organic solutions for direct dissolution concepts.^[55] However, the radiation stability, longevity, and safety of these proposed media are not well known.

Finally, avenues for increasing the utility of advanced reactors through the utilization of *nuclear assets* (i.e., off-peak electricity, high temperatures, and ionizing radiation fields) are also under investigation. Examples include carbon-free production of combustible fuels, steam-methane reforming, and steam-driven electrolytic splitting of water.^[57] These innovative chemical processes may employ nonaqueous coolants and solvents beyond those directly considered for reactor and separations technologies. Thus, advancements in the understanding of next-generation media will have implications beyond direct use in energy technologies.

Establish the speciation and reactivity of solutes in next-generation solvents and coolants.

Current nuclear energy systems operate on aqueous-based chemistries, employing water as coolants, as well as aqueous and biphasic aqueous–organic mixtures in the synthesis of fuel and recycling of used nuclear fuel in the back end of the nuclear fuel cycle. Although the speciation and reactivity of solutes in these systems—actinides, fission products, and corrosion products produced from the interaction of water with fuel cladding and piping—are still being studied, the ability to predict, control, and study this chemistry is based on classic equilibrium thermodynamic models that presume solution ideality or are corrected for nonideal behavior using well-established theories of solution solvation in aqueous systems. This paradigm does not extend to next-generation media. Much work still needs to be done in aqueous systems, but next-generation coolants and solvents deviate significantly away from ideal thermodynamic behavior, where it is known that the solvent cannot be treated as a homogenous continuum nor as a spectator in the speciation and reactivity of the solute. As an example, solutions of molten salts proposed for use as both nuclear fuels and coolants, as well as in pyrochemical separations processes, take on local and intermediate range structures that changes with the chemistry of the solutes and the solvent composition. This change affects the physiochemical properties of the solutions such as viscosity, redox potential, solute solubility, and speciation. Knowledge of the solution chemistry of next-generation solvents at the molecular scale is required for the long-term safety and performance of these next-generation nuclear energy technologies. This scale underpins the chemical processes associated with materials’ performance and actinide, fission, and corrosion product recycling and management, especially under the extreme conditions encountered in nuclear energy systems.

Very little is known about the speciation, structure, and reactivity of actinides at the atomic or molecular level under extreme conditions and outside of aqueous electrolyte solutions. Even in aqueous media, the radiation-induced chemistry of these complex elements has barely been examined.^[58] Virtually nothing is known about their behavior in next-generation coolants and solvents, nor their interplay with other nuclear fuel cycle solutes (fission fragments, impurities, corrosion products, and process chemicals) in extreme ionizing radiation environments.^[53] The emphasis here is on nonaqueous media, but progress in meeting these challenges will require comparison with processes in aqueous media, for which significant fundamental knowledge gaps still exist. Finally, exotic actinide configurations such as colloids, clusters, and metallic frameworks in aqueous and nonaqueous media have only just begun to be systematically studied in ionizing radiation fields and offer new avenues within the nuclear fuel cycle.^[59]

Beyond actinides, fission fragments, process chemicals (e.g., ligands), and corrosion products from the dissolution of structural components and/or fuel elements are additional solutes to be considered in next-generation coolants and solvents. In some cases, these solutes still need to be examined in aqueous solutions. Chemically reactive species produced in these media can attack solutes and interfaces, leading to their radiolysis and extent of corrosion, respectively, ultimately impacting the longevity and performance of nuclear energy technologies. Further, radiation-induced reactions at particle surfaces may change particle-particle interactions affecting nanoparticle formation, aggregation, stability, and rheology. Very little is known about the

mechanisms of particle formation and decomposition induced by ionizing radiation, even in aqueous media. These solutes and interactions will behave differently in next-generation coolants and solvents than what is observed in water-based nuclear fuel cycle technologies.

Develop robust and specialized analytical techniques amenable to in situ and real-time sensing.

Basic science is lacking for the development of sensors for in situ and real-time characterization of advanced nuclear chemical processes. More specifically, the design and integration of robust sensors to deconvolute information from complex chemical and dynamic ionizing radiation environments must be explored. Knowledge gaps must be connected for the development of accurate online sensors for in situ and real-time characterization that span multiple length, energy, and timescales in next-generation coolants and solvents, which is a focus of **PRO 4**. Of particular interest is the novel utilization of optical tools to follow process conditions typically measured by other means (e.g., pH within aqueous solutions, as demonstrated by **Figure 7**).^[60] This technology opens avenues to use robust optical sensors in extreme environments where more-fragile sensors cannot be deployed.

The interplay between empirical data collection, advanced data analysis, and computational modeling must be leveraged to develop predictive tools for in operando chemical behaviors, interactions, and deviations. A key aspect of this goal is to bound the expected uncertainties in the data versus deviations that indicate off-normal reactor and separations behavior. Thus, simultaneous analysis of experimental data and computational predictions for real-time integration and mechanism validation must be explored.

2d. Potential Impacts

Answering the questions motivated by this PRO will lead to significant effects on the development of future nuclear energy systems. In particular, researchers will be able to predict the quantitative relationships between structure and energetics in nonideal, nonequilibrium solutions across multiple length and timescales, a consequence of new, scalable, predictive capabilities for chemistry and structure in nonideal, nonequilibrium systems. This ability will allow for more robust design of new coolants and solvents and can only be accomplished through the integration of experimental, computational, and sensor tools that empower deeper scientific investigation of complex multicomponent systems. Ultimately, this ability will lead to the control of next-generation coolants and solvents for the synthesis of novel materials; innovative separations and recovery; next-generation energy storage systems; and the informative design, innovation, and monitoring of advanced separations for sustainable life cycles of critical materials and nuclear fuels. The tools and knowledge gained will support the deployment of nuclear energy—safe operations, safeguards (rigorous accountancy) for actinides and reactor materials, bounding of measurement uncertainties, process control, process upset avoidance, reliable storage, and more—ultimately supporting cost savings and regulatory needs. Finally, research addressing this PRO will enable chemical knowledge and technologies for next-generation coolants and solvents

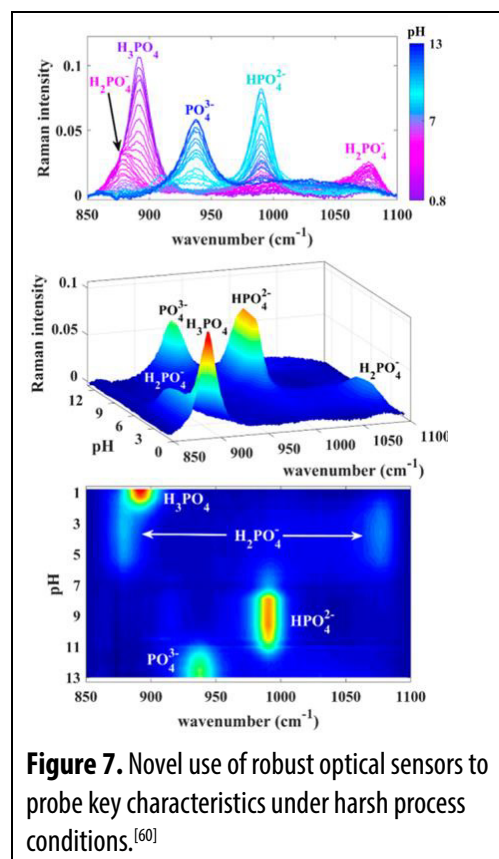


Figure 7. Novel use of robust optical sensors to probe key characteristics under harsh process conditions.^[60]

to be applied in other areas, such as solar and energy storage or hybrid nuclear reactor applications.

PRO 3: Elucidate and Control the Underlying Physics and Chemistry of Interfaces in Complex Nuclear Environments

Overarching Question: How do researchers harness dynamic interfaces to tailor robust materials and processes for next-generation nuclear reactors?

Interfaces have played a fundamental role in energy innovation over the last several decades from hydrogen conversion to electrical energy storage and carbon capture.^[52b, 61] Major advances have been made, with the state of the art evolving from understanding pristine interfaces to probing interfaces under realistic operating conditions and controlling complex interfacial structure and defect density. However, long-standing research challenges remain and must be overcome to harness the power of interfacial processes for nuclear energy applications. In particular, a significant opportunity exists to first understand the dynamic evolution of interfaces under extreme environments and then design systems that leverage that evolution.

Extreme conditions involve radiation, oxidative and corrosive environments, high velocity flow, high energy and charge transfer, or high temperatures that result in morphological changes that need to be quantified and better understood. Materials for nuclear systems or applications are required to exhibit exceptional performance (e.g., radiation tolerance, corrosion resistance, and high-temperature performance). Although such materials are available, they often rely on metastable nanostructures that tend to degrade in these extreme environments.^[62] The influence of temperature, pressure, and radiation on the chemistry occurring at interfaces is extremely important throughout the nuclear industry. Surfaces in real-world conditions are not ordered, flat atomic layers. The role of defects, crystal edges, pores, and other surface structures on the interfacial chemistry must be understood in the nonequilibrium environments that would be experienced in next-generation nuclear systems. Reactions can be initiated at the surface of all materials (e.g., fuel elements, construction materials, electrodes, sensors). Radiolytic reduction of metals leads to the formation of nanoparticles and, ultimately, metallic clusters that are stabilized by surface chemistry. Particle aggregation is controlled by the radiation-induced charges and surface species. Temperature, pressure, and radiation can also lead to the breakup of particles or the dissolution of atoms from solid surfaces.

A central challenge to controlling interface evolution is that key processes span different timescales and length scales and encompass a multitude of physical and chemical changes. Researchers do not have the capacity to subject all materials and interfaces to conditions that would be endured in these nuclear systems; thus, a fundamental mechanistic understanding is needed to connect structure and chemical properties to predicted performance.

Energy, mass, and charge transport through solid and liquid phases control surface chemistry and particle–particle interactions. The atomic structure and chemistry induced at surfaces in contact with extreme environments are not well-characterized or understood. Surface-specific techniques must be exploited to elucidate the effects of energy deposited by the passage of radiation on the subsequent chemical reactivity. Identification of surface species and surface structure is critical for the ultimate determination of specific mechanisms. Radiation effects have the potential to modify surface structures and the resulting interactions; although ion-beam modification of surfaces has a long history of study, how the changes in surfaces induced by irradiation affect interaction with the environment is less studied.

Tracking these dynamic changes, understanding the effect on interfaces, developing capabilities for online monitoring in nuclear environments, and identifying strategies to counter any deleterious

effects of the changing conditions are all crucial. Capabilities must be developed to investigate evolving interfaces and elucidate their dynamic microstructure, especially in operando. An enhanced understanding of dynamic interfaces will allow researchers to control defect behavior and tailor heterogeneous interfaces with increased stability for next-generation nuclear reactors.

3a. Summary

Many material failure modes initiate with surface reactions, and understanding charge/mass transport and reactivity is critical at solid–solid, solid–liquid, liquid–liquid, and solid–gas/plasma interfaces to develop durable materials. Interfacial reactivity can be enhanced by orders of magnitude in the presence of impurities such as moisture, oxygen, or electroactive species. Understanding interfacial properties associated with grain boundaries, precipitates, dispersion matrix effects, and helium-stabilized cavities/bubbles is essential to predict and mitigate radiation effects and tailor radiation-tolerant materials in fission and fusion reactors.^[63] Radiolytic effects can further lead to the formation of new interfaces through precipitation of nanoparticles and metallic clusters. Interfaces between plasma and structural materials in a fusion reactor also need to withstand intense heat fluxes and particle fluences without trapping hydrogen/tritium, which may compromise the material integrity.

In addition to challenges in understanding the liquids themselves (**PRO 2**), new challenges arise when the liquid interfaces with a solid. Molten salts or flowing metal coolants under radiation fields also present unique challenges for the structural components. In these super-concentrated *solutions*, nanostructuring, in some cases driven by the extreme conditions within the reactor, generates dynamic interfaces that drastically affect energy transfer and chemical reactivity, particularly with respect to nucleation and particle interaction processes. The heterogeneous morphological evolution needs to be understood before researchers can control the physical, chemical, and constitutive properties of molten salt reactor (MSR) materials.

Establishing a thorough understanding of the processes that occur in these environments and extreme conditions is therefore critical to developing durable materials and monitoring protocols that will enable fundamental learnings and safe use of next-generation nuclear reactors.^[64] Ultimately, meeting the increasing demands for clean energy will necessitate the design of new types of compositionally diverse and dynamically evolving interfaces that enable and control the processes occurring at extreme environments.

3b. Key Scientific Questions

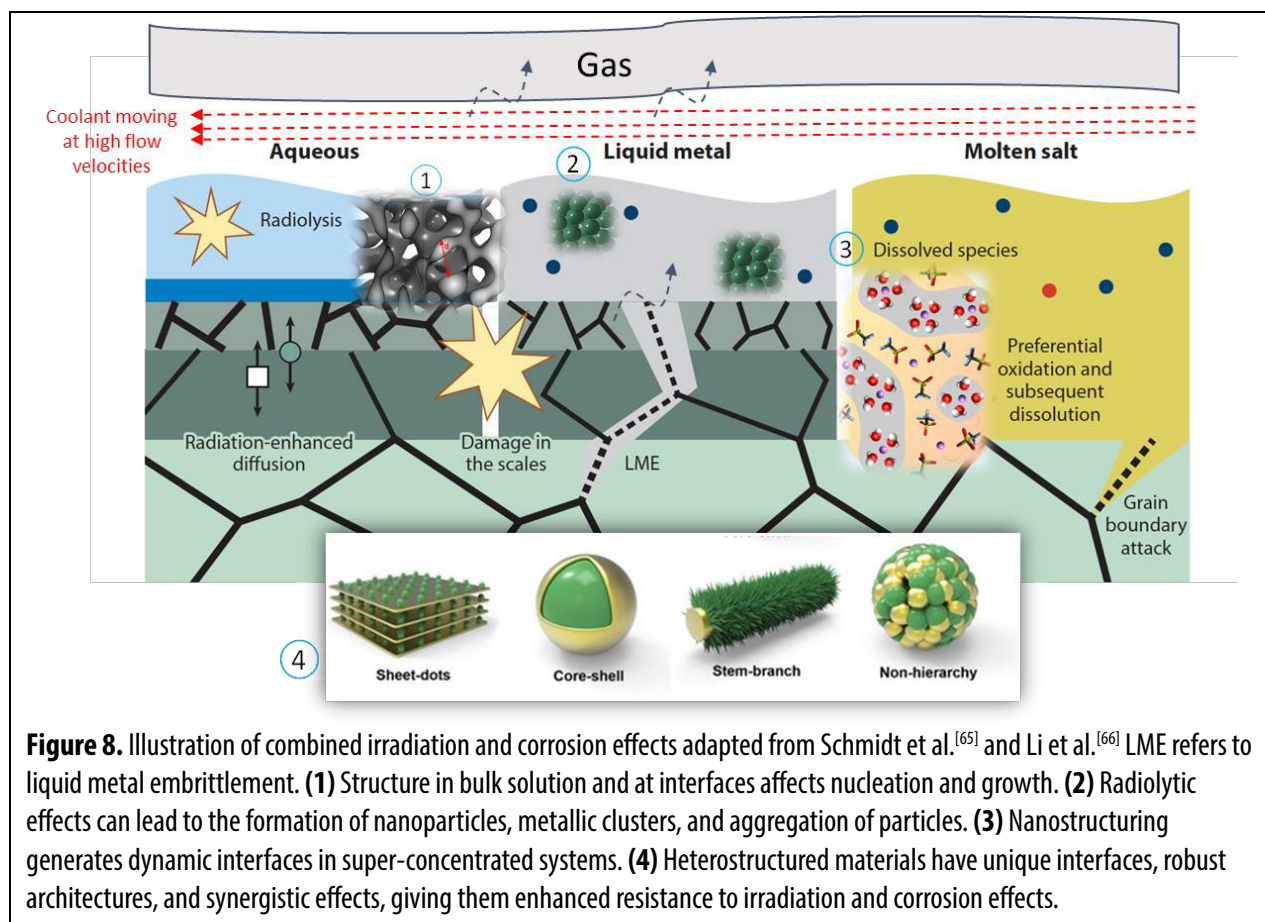
- How do researchers harness dynamic interfaces to tailor robust materials and processes for next-generation nuclear reactors?
- Can researchers characterize, predict, and control the time-dependent evolution of interfaces as they develop in nuclear environments?

3c. Scientific Challenges and Research Opportunities

- Elucidate and control the dynamic interfacial physicochemical phenomena and their evolution in extreme environments of advanced nuclear reactors.
- Bridge the timescale and length scale in the mechanistic understanding of heterogeneities that arise from radiation, temperature, and external fields under extreme conditions.
- Identify and develop the critical enabling capabilities needed to achieve simultaneous imaging and characterization of interfaces in extreme environments.
- Design heterostructured materials with evolving interfaces for unprecedented resilience and performance under extreme conditions.

Elucidate and control the dynamic interfacial physicochemical phenomena and their evolution in extreme environments of advanced nuclear reactors.

Dynamic interfaces between different phases of matter, including solid–liquid, solid–gas, and liquid–gas, have been investigated, but researchers do not understand the relevant properties that are necessary to predict and control the time-dependent evolution of interfaces under extreme conditions. The development of advanced analytical techniques with higher resolution and faster dynamics, coupled with new paradigms in modeling and simulation, will provide opportunities to understand critical interfacial phenomena. Understanding physicochemical reactions at interfaces; in the presence of radiation, electric, and magnetic fields; and in contact with highly corrosive coolants—in some cases, flowing at high velocities—is an important challenge for future nuclear needs.



Interfacial processes are influenced by radiation-induced processes, including defect creation, migration, and aggregation, as well as the formation of bubbles and voids (**Figure 8**).^[62a, 67] Although irradiation accelerates corrosion in light-water reactor conditions, it may decelerate that process in MSR conditions depending on the corrosion mechanism.^[65, 68] Accelerated radiation-induced corrosion was also observed in liquid metal–cooled reactors.^[62a, 69] However, depending on the dominant corrosion mechanism, alloy composition, and temperature, irradiation may also decelerate the corrosion of structural materials in contact with liquid metal coolants similar to recent observations in molten salt.^[70] Furthermore, the alloy composition can be designed to withstand these demanding environments. Additionally, the forces acting on the materials and their environment play a key role (e.g., high-velocity, heavy liquid metal coolants can lead to

erosion at the surface of structural materials).^[71] The evolution of impurities in the coolant as well as vaporization of substances to the cover gas are not well-understood enough at the molecular level, which is necessary to predict behavior.

Radiation-induced processes at and near interfaces contribute to phenomena that affect nuclear energy technologies, including corrosion, dissolution, and catalysis; however, detailed mechanisms of energy transfer are not well-understood. By developing a new understanding of energy transfer and reactive intermediates through bulk solids to the interface, researchers can design functionalized interfaces to harness or disperse this energy and associated reactive intermediate species. Furthermore, by understanding the dynamic evolution of these systems, new interface structures can be designed that exploit the dynamic changes in the system for enhanced performance. Researchers must advance this understanding to encompass the role of the liquid and gas phases, including interfaces created by clustering ions in nontraditional coolants and solvents and gas-phase interactions with liquid metal coolants and impurities/contaminants.

Bridge the timescale and length scale in the mechanistic understanding of heterogeneities that arise from radiation, temperature, and external fields under extreme conditions.

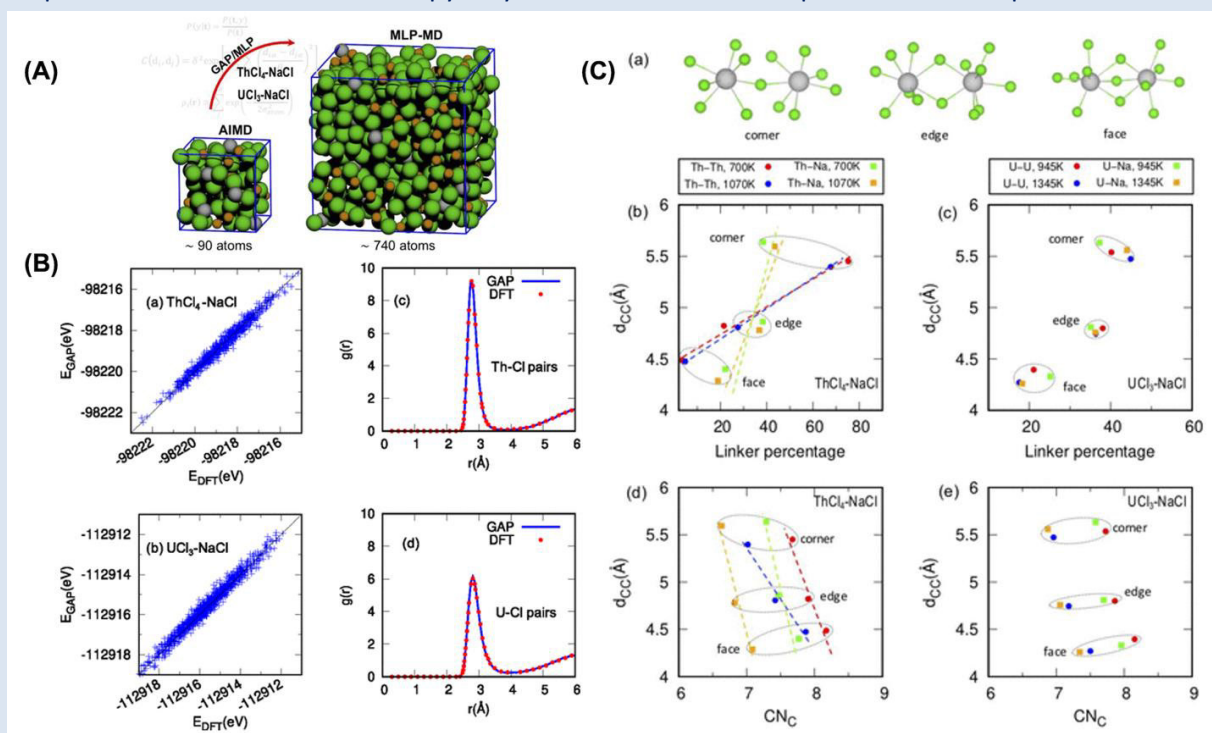
The study of heavy elements found in these environments is extremely challenging. High temperatures are often the root cause of experimental inaccuracies. Additionally, transuranic elements are present in small amounts, making monitoring them difficult and hampering experiments with the hazardous conditions. Computer simulations can circumnavigate these issues and still provide consistent results as long as accurate models are employed and the simulations can capture the underlying physical and chemical events. The combination of atomistic simulations with data science has been gaining momentum for accessing the timescales and length scales with impressive accuracy. Data science has also been used in integrating chemical processing and decision-making (see **Sidebar 5**), converting historical operating data to input for mathematical modeling of nuclear processes.^[72]

This research goal aims to answer critical questions on how, where, and when heterogeneities such as clusters will form, aggregate, or break up in extreme environments. Ideally, researchers wish to understand how the solute/solvent ion network can rearrange dynamically and ultimately harness interfacial processes in condensed phases to control energy transfer and chemical reactivity.

SIDEBAR 5—ML APPROACHES ELUCIDATE THE DYNAMIC STRUCTURE AND COORDINATION OF ACTINIDES IN NUCLEAR ENVIRONMENTS.

Although experimental techniques often provide a macroscopic view of a process, theoretical investigations are tailored to furnish an atomistic view of structure, reactivity, and transport in these systems. However, these approaches do not come free of problems. For example, a good representation of interfacial processes in MSRs should be able to describe the chemical changes that involve radionuclides and their interaction with the molten salt matrix and other interfaces that are a part of modern nuclear reactors. As a result, the understanding of their chemistry, dynamics, and properties has received only modest attention. In principle, ab initio molecular dynamics can provide the necessary details, but the method becomes very expensive with the system size and simulation length. Recently, neural networks have been used very successfully for the construction of interatomic potentials with near-first principles accuracy at a significantly reduced computational cost.^[73]

In this example, the ion interactions of $\text{ThCl}_4\text{-NaCl}$ and $\text{UCl}_3\text{-NaCl}$ at different temperatures are examined. Understanding the extended structure of actinides is key to interpreting spectroscopic features such as Raman or extended x-ray absorption fine structure signatures. Overall, at higher temperatures, a quasi-octahedral coordination is more likely. The survival time of these complexes is on the order of 10 ps, which implies that they can be traced with scattering methods, even at high temperatures. Pair correlation entropy may also be used as a descriptor of the ionic species.



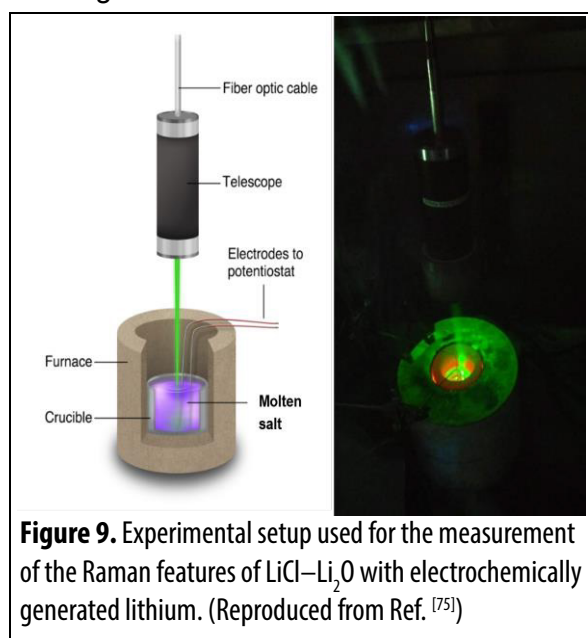
ML potentials based on the Gaussian approximation potential (GAP) were adopted to investigate the structure and ion aggregation in the binary systems $\text{ThCl}_4\text{-NaCl}$ and $\text{UCl}_3\text{-NaCl}$. (A) The potential was trained on a DFT-based atomistic model. (B) The ML-based model (i.e., GAP) shows ab initio molecular dynamics quality. (C) Three different cation–cation associations and their correlations to the coordination environments were identified as a function of temperature. (Reproduced with permission from Nguyen et al. ^[73b])

Identify and develop the critical enabling capabilities needed to achieve simultaneous imaging and characterization of interfaces in extreme environments.

Understanding the operando dynamic evolution of interfaces is an outstanding challenge. Probes used in normal environments simply cannot withstand the extreme conditions associated with nuclear environments and are thus of little utility. This problem places an even larger burden on robust computational approaches to complement experimental interrogation. New experimental and computational techniques must be developed to understand the dynamic restructuring of interfaces under conditions of corrosive environments, high temperatures, radiation, and magnetic fields, as well as the subsequent effects on chemical and physical processes. This knowledge gap is tightly correlated to the development of new materials appropriate for a given measurement probe that can withstand extreme environments and provide reasonable measurement fidelity. Methods need to be developed or optimized for understanding chemistry at both laboratory and operating plant scales. In situ continuous monitoring methods are needed to identify impurities and the evolution of chemistry with time. Methods, such as Raman, infrared, and ultraviolet–visible spectroscopy that can be used through an optical window, need to be developed to maturity. Window materials also need to be developed for use in harsh environments, especially in the presence of lithium, which degrades most known materials. Regarding lithium solutions, Bredig stated, “No insulating material has yet been found which will withstand attack by these solutions.”^[74] Testing of such spectroscopic techniques in a flowing system is also needed for translation to operating plants.

Rapid analytical techniques are needed at the laboratory scale to screen materials, understand the dynamical evolution of interfaces under extreme conditions, and shortlist candidates for synchrotron studies, which often are only available for limited periods of time. Predictive computational methods are needed for rapid prescreening. Although synchrotrons can provide an array of data, no simple experimental setups exist for in situ analysis at the synchrotrons. The existing setups are for very particular systems.^[76] Very few facilities can handle materials such as FLiBe or study accident scenarios, both of which are essential for commercialization of certain MSRs. Studies on accident scenarios—for example, the formation of aerosols or mixing with water—are especially needed to develop mitigation strategies.

Environmental composition affects materials in many ways. The behavior of target analytes in the presence of many other chemical species—and in a radiation field—is unknown. Changes to coordination owing to changes in the surrounding chemical environment can change spectral signatures, making absorbance quantification difficult and altering electrochemical measurements that may be used for quantification. Analytical and electrochemical techniques that can be used in the presence of many species need further development. Additionally, new approaches are required to enable the visualization of radiolytic transformations at interfaces in situ beyond existing ex situ observations and in situ techniques, in which radiation is inextricably tied to the imaging basis (e.g., electron microscopy or synchrotron-based methods).^[77]



Understanding the science behind the degradation of electrodes that are needed to make these

measurements is also critical. Techniques or reference electrodes are needed to determine a thermodynamically defined reference potential, especially for halide salt systems (**Figure 9**). Stable, nonprecious metal-based electrodes are also needed for electrochemical processing of used nuclear fuel.

Basic science studies of fission product and actinide chemical behavior to understand coordination and speciation in the presence of other fission products or actinides will help address the difficulty in interpreting absorbance spectra with multiple varying species (as detailed in **PRO 2**). Continued development of electrochemical and analytical techniques (e.g., optical basicity, cyclic voltammetry) in the presence of many species as well as identifying means to reduce uncertainty in quantification are needed (e.g., electrode shrouds, better determination of diffusion coefficients). Standard methods to characterize and quantify impurities need to be developed so that the behavior of impurities can be linked to fundamental physical and chemical processes.

Design heterostructured materials with evolving interfaces for unprecedented resilience and performance under extreme conditions.

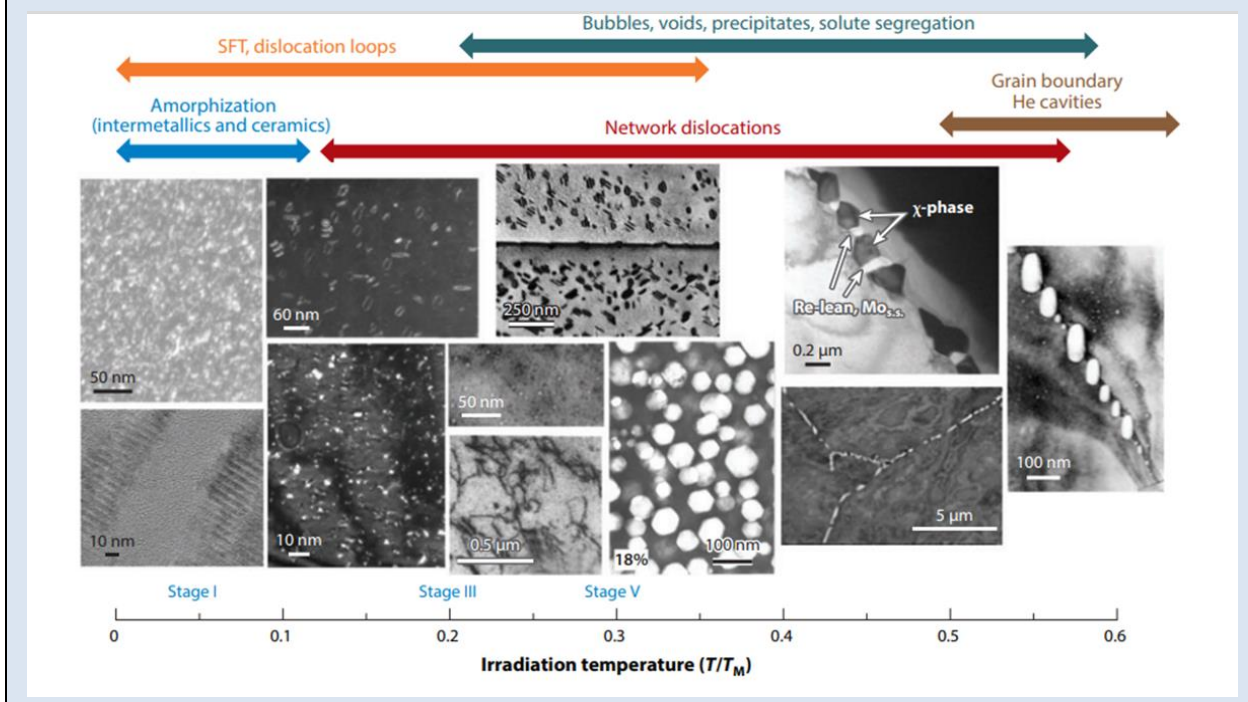
The optimum materials for use in fission and fusion reactors should have a composition and structure that can withstand radiation-induced damage and corrosion.^[65] Currently, no such known material can address all challenges associated with future nuclear needs; therefore, a fundamental understanding of the dominant mechanisms of material degradation in extreme environments encompassing radiation fields, corrosive media, and high temperatures is required for material optimization. The extreme nature of reactor operating conditions necessitates novel, multilevel approaches for material design (e.g., heterostructured materials, glassy materials, metallic glasses, and CCAs beyond regular alloys or common materials with grain boundaries).

Traditional design of materials for extreme environments has centered on monolithic microstructures or materials with periodic structures. Materials that contain heterogeneous zones and interfaces offer novel physical and chemical properties worthy of investigation. Heterostructured materials, such as gradient materials, layered materials, dual-phase materials, core-shell materials, composites, and more, contain interfaces that can be uniquely tailored to enhance behavior in extreme environments (see **Sidebar 6**). Glassy materials do not have grain boundaries and are often more resistant to radiation and corrosion than their crystalline counterparts, though they can be more susceptible to leaching.^[78] CCAs are more corrosion-resistant than traditional alloys, and with multiple components, they can be more easily designed with particular characteristics, such as low activation, which is critical for nuclear applications.^[62c, 79] The elemental toolkit for heterostructures also includes actinides. Incorporation of actinides into hetero- and nanostructures provides new pathways for the control and design of materials based on their properties.^[80]

In addition to heterostructured solids, nanostructuring can also occur in super-concentrated solutions.^[81] Nanoheterogeneities can form in solutions even at lower concentrations, with bulkier ions resulting in a decoupling of ionic mobility from viscosity. This nanostructuring generates dynamic interfaces that affect energy transfer and chemical reactivity, particularly with respect to nucleation and particle interaction processes. Researchers do not know enough about this nanostructuring behavior to predict how these super-concentrated systems will behave in next-generation coolants and solvents, as well as in waste processing and storage, where water will be present as a contaminant.

SIDEBAR 6—DESIGNING HETEROSTRUCTURED MATERIALS AND INTERFACES WITH UNPRECEDENTED RESILIENCE AND PERFORMANCE UNDER EXTREME CONDITIONS.

Heterostructures are created by layering two or more materials with the goal of taking advantage of the unique physical characteristics of the interface between them. The functionality of heterostructure materials is dictated by the interface, which is symbiotic with the nature of the interfacial environment. Since 2017, many new heterostructured materials have been synthesized, but their performance under reactor operating conditions of extreme temperature, pressure, magnetic fields, and radiation has not been demonstrated. These extreme conditions can counteract each other. For example, temperatures exist at which the effects of radiation on corrosion become minimal.^[20] Understanding this interplay between external fields, temperature, and corrosion is essential to predicting the performance of current materials and developing new materials for future nuclear reactor applications.^[65]

**3d. Potential Impacts**

The scientific impact associated with this PRO is far-reaching. Research in this area will lead to the understanding of the formation of interfaces owing to aggregation, nucleation, and transport in unconventional solvents and coolants as well as the processes by which mass and energy are transferred to and away from the interface. This work will provide new avenues for designing interfacial systems for tailored energy transfer or for preventing corrosion. Furthermore, the development of new multimodal capabilities and computational models for studying interfaces at extreme conditions will lead to materials and engineered systems with enhanced safety and efficiency. Opportunities exist to design systems in which the interfaces evolve to preferred structures under those extreme conditions. Beyond nuclear energy systems, these efforts would facilitate the development of novel catalytic and corrosion-resistant materials for other energy technologies. Finally, by improving the general understanding of dynamic interfaces, new materials for separations and energy storage can be developed.

PRO 4: Bridge Multifidelity, Multiresolution Experiments, Computational Modeling, and Data Science to Control Dynamic Behavior

Overarching Question: Which novel techniques can be coupled to provide operando and in situ measurements to better understand and control dynamical properties, behaviors, and processes for extreme nuclear energy environments?

4a. Summary

The dynamical evolution of nuclear material systems and processes is multiscale in nature. Although the creation of radiation damage occurs over picoseconds, other processes, such as chemical species transport and interactions, occur over milliseconds and seconds. This time leads to microstructural changes over multiple length scales that affect the performance of nuclear systems over years and decades. Similarly, the chemistry of molten salts in reactor environments evolves following complex reaction networks whose elementary steps can require vastly different timescales, from very fast radiation-induced reactions to rare thermally activated events.^[82]

Therefore, the overall dynamical evolution of complex nuclear systems cannot be captured by sparse experimental probes and modeling approaches that only capture narrow time scales and length scales or—even worse—only involve post-experiment examination of materials. Instead, a complete picture can only be painted by correlating and fusing multiple sources of experimental and modeling data, simultaneously characterizing the evolution of a target system in operando. This need has been emphasized multiple times in this report.

Although nuclear systems are routinely characterized by multiple approaches, concurrent multiprobe experiments and multiscale simulations offer unique opportunities. This combination can extract fundamental insights that are uniquely revealed by the fusion of multiple data streams (e.g., combinations of embedded, online sensors; neutron/x-ray imaging; scattering and spectra collection for 3D microstructure reconstructions with spatially resolved atomic structure/phase information; and chemical composition piped into physics-based computational modeling). When acquired simultaneously, this fusion of data streams across different length and timescales—each with different yet complementary resolutions and fidelities—will, in turn, offer unique opportunities for finer control and optimization of nuclear systems. For instance, data streams from in situ, multiprobe experiments of structural vessel materials exposure in an MSR; tracking transient chemical speciation and complexation during fuel element-to-cladding interaction; or oxidation sequences in a high-temperature, corrosive environment could be fused with real-time feedback provided by AI-assisted analysis and experiment decision-making. Researchers need to develop experimental capabilities to probe nuclear systems in situ in a laboratory setting and operando in the reactor.

4b. Key Scientific Questions

- What combination and types of novel multiprobe/multiresolution techniques can be coupled together to provide operando and in situ measurements to better understand and control key dynamical properties, behaviors, and processes for fission and fusion materials under extreme environments?
- How can predictive, data-driven models of dynamic evolution in nuclear systems be developed by systematically combining calculations carried out at multiple scales and fidelities?
- How can the information from multiple sources (e.g., traditional and novel multiprobe experiments and multiscale simulations) be integrated into a holistic model, providing sensor–model fusion at scale?^[83]

- Can fused, real-time, sensor-informed models reveal the nature of the fundamental processes that determine the dynamical evolution of nuclear systems so as to enable their tuning and control?

4c. Scientific Challenges and Research Opportunities

- Develop novel multiprobe/multiresolution experimental techniques for operando characterization of nuclear energy systems.
- Integrate exascale-scalable, data-driven, dynamical models of nuclear systems in operando.
- Fuse sensor systems with models for tracking dynamical evolution of nuclear systems.

Develop novel multiprobe/multiresolution experimental techniques for operando characterization of nuclear energy systems.

A holistic, comprehensive understanding of defect behavior and the underlying damage mechanisms in complex and heterogeneous material systems subject to combined extremes of radiation, corrosion, and high-temperatures requires a range of complementary experimental techniques that operate at different time and spatial resolutions. Current techniques for in situ characterization of materials under such coupled extreme environments are limited^[84] or nonexistent, instead relying heavily on posttest characterization techniques.^[85] Key questions posed in **PRO 2** and **PRO 3** also benefit from this focus on operando characterization tools. Novel experimental techniques are needed to concurrently characterize materials across multiple scales (e.g., atomic to mesoscale, angstroms to centimeters, and with temporal sensitivities from electronic structure transitions to material deformation timescales, subpicoseconds to hours). For instance, transient behaviors of solvent systems (particularly the organic phase/extractants) while undergoing irradiation is poorly understood, as emphasized in **PRO 2**. Solvent species break down, and the resulting effects to f-element complexation (and therefore extraction distribution ratios and kinetics) need to be prioritized. Furthermore, radiation effects on behavior at the aqueous–organic interface and subsequent changes to f- and d-element complexation (including possible ties to third phase formation) must be better understood to support larger goals around used nuclear fuel recycling. Monitoring these and other materials' property changes, behavior, and dynamic interactions at combined conditions of corrosion, irradiation, and high temperatures requires concurrent in situ measurements for operando characterization (e.g., neutron and x-ray scattering, positron annihilation spectroscopy, Rutherford backscattering, dynamic-TEM, scanning electron microscopy, electrochemical measurements, and optical spectroscopy).^[86]

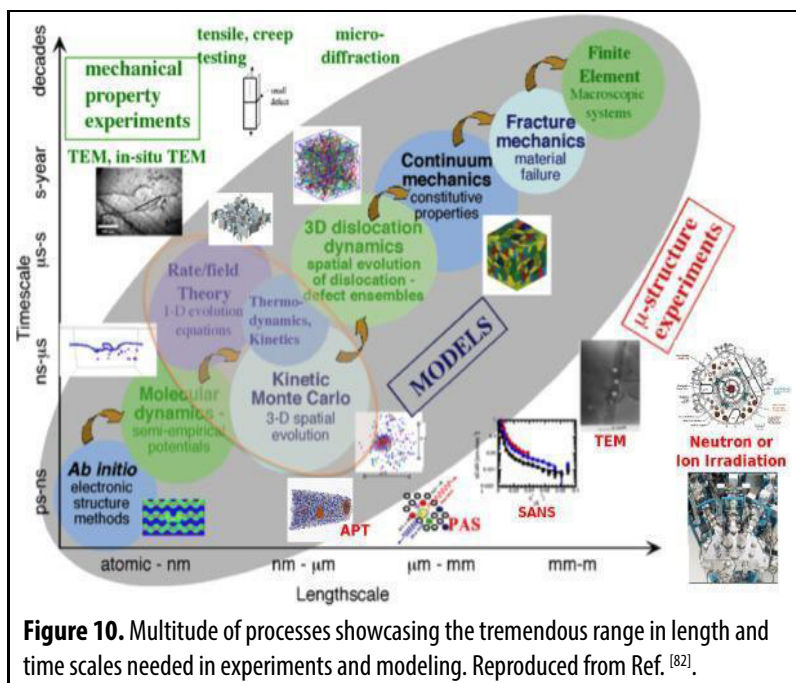
For instance, a multimodal experiment to collect data across needed spatial and temporal scales at combined extreme conditions relevant to nuclear fission and fusion could involve deployment of a microreactor or test vessel to a DOE x-ray light source or neutron facility. Here, a roll-up chamber, which provides the simultaneous extreme states, could be used at a beamline. The use of the unique operando and pump–probe capabilities recently developed at DOE user facilities, such as in situ positron annihilation with ion irradiation and pulsed positron beams,^[87] the creation of radiation damage by accelerated protons or neutrons, or high-power lasers used to generate a secondary ion source for irradiation,^[88] can fill the gap in the understanding of defect formation and evolution on a fundamental level. Capturing ultrafast changes in electronic and lattice structures and processes calls for further development in time-resolved optical, x-ray, and neutron techniques and the use of DOE light sources. For instance, expected upgrades to the Matter in Extreme Conditions facility^[89] at the Linac Coherent Light Source of the SLAC National Accelerator Laboratory (SLAC) will enable 0.1 displacements per atom in 5 μm tungsten in a single shot with a laser-accelerated 10 MeV argon beam. In 10 μm tungsten, several accumulated shots with a laser-accelerated 1 MeV deuteron beam and ion beam focusing components are possible, enabling time-resolved radiation damage mechanism studies across picosecond to

minute timescales with a suite of in situ x-ray and optical probes. Other time-resolved capabilities, such as ultrafast electron diffraction (UED), combined with high-power lasers (e.g., MeV-UED Facility at SLAC) are able to visualize the atomic dynamics during irradiation or corrosion processes and provide direct comparisons with atomistic simulations.^[90] This capability is a unique feature of UED over other conventional techniques, such as time-domain thermoreflectance. Sustained plasma formation on an interface or surface could be examined using time-resolved grazing-incidence small-angle scattering/wide-angle scattering to track the PDF and structure factor change $[S(Q,t)]$ as a function of time and frequency. Furthermore, although in situ capabilities at user facilities will undoubtedly provide great scientific insight, similar capabilities must also be developed that can be deployed directly at reactors to monitor and characterize their evolution under true reactor conditions.

Experiments on thin films and surrogate systems can be a good way to perform and monitor the material behavior and response in real time when reactor-scale measurements are not accessible. However, one must ensure that the sampled microstructures and properties are comparable to bulk forms. For example, the structures and properties of thin films and other small-volume materials are often dominated by strong crystallographic textures (i.e., preferred crystal orientations) and can contain nonequilibrium phases or phase distributions. This material creates an opportunity for researchers to develop rapid screening methods or correlation mechanisms that will allow for rapid extraction of bulk properties from these materials.^[91] These benchtop experiments provide valuable validation of models that can then be extrapolated to reactor conditions.

Integrate exascale-scalable, data-driven dynamical models with nuclear systems characterized in operando.

A key challenge with predicting the dynamical evolution of nuclear systems over application-relevant timescales is that chemically accurate approaches (such as ab initio molecular dynamics) are often difficult to validate with experiments because of very limited spatiotemporal reach, so scale-bridging approaches are essential to predicting performance on engineering scales. In response, the modeling community has developed an arsenal of methods that collectively span length-scales from angstroms to meters and timescales from femtoseconds to decades, as illustrated in **Figure 10**. A key



outstanding challenge is the difficulty of seamlessly bridging scales by coupling/integrating these different models into unified predictive capabilities. Although a number of examples of such coupling have been demonstrated in nuclear energy applications (e.g., using DFT to develop interatomic potentials for molecular dynamics, which then informs kinetic Monte Carlo models that are further upscaled into cluster dynamics simulations),^[82] these almost always involve

considerable human intervention through ad hoc decision-making (e.g., to postulate the key unit reaction steps) and/or manual analysis or interpretation of simulation data.^[73a] This (1) makes the development of multiscale, dynamical models extremely time consuming; (2) introduces hard-to-control uncertainties as information is transferred between scales, and (3) limits the use of leadership-scale computational resources to generate the volume and quality of data required to obtain truly predictive models.

One needed area of improvement is the development of intrinsically data-driven, scale-bridging techniques for dynamical models of chemical or material nuclear systems. These systems need to be rigorously informed by complementary simulation data obtained at various scales and fidelity. Scales range from chemically accurate ab initio calculations through semiempirical and classical molecular dynamics and mesoscale reaction networks and all the way to macroscale reaction-diffusion and microstructural evolution models. Rigorously integrating the results of different simulation methods, which operate at different spatiotemporal scales and possess different intrinsic accuracies, will require the development of rigorous multiscale uncertainty quantification and propagation methods that can optimally correlate, extract, and combine the unique information contained in each data stream. This process would reduce the timescale for developing models while providing uncertainty estimates for key quantities. It would also guide further experimental and computational examination, such as the rapid determination of the impact of dopant/impurity/transmutant effects on transport in complex alloys. Furthermore, efficiently leveraging the capabilities of exascale computing platforms will require algorithms that dynamically balance between different components with vastly different computational demands. For instance, DFT methods are orders of magnitude more computationally demanding than kinetic Monte Carlo models—any approach that integrates two such disparate methods in one simulation framework will need to balance computations between the various components. New algorithms and workflows will be needed to do this efficiently and dynamically as the needs of the simulation evolve during runtime.

The intrinsic and extrinsic chemical complexity of nuclear systems in operating conditions suggests that very large and rich datasets will be required to achieve predictive accuracy. To efficiently leverage upcoming exascale systems,^[92] these models should be equipped with autonomous procedures that can systematically guide their refinement by identifying targeted simulations that would lead to optimal model improvement, including comprehensive in situ simulation setup and analysis (e.g., automated identification of classes of reactions) to scale bridging (e.g., reduction of molecular dynamics data into reaction networks, upscaling of reaction networks into reaction-diffusion models). Modern data science techniques such as Bayesian optimization^[93] and multifidelity/multimodality learning^[94] are likely to be helpful tools in achieving these objectives.

Fuse sensor systems with models for tracking dynamical evolution of nuclear systems.

The physics and chemistry of these experiments cannot be fully understood in real time at fission and fusion environments at any scale without the inclusion of data science. Therefore, multisensor datasets must be combined with the state-of-the-art modeling tools to provide experimentalists with active feedback for observation interpretation. To do so, researchers should combine the outcome from complementary techniques into an integrated, multimodal analysis framework that links these characterization experiments with a model. Sensor fusion is the process of combining sensor data or data derived from disparate sources such that the resulting information is richer than what could be inferred if these sources were used individually.^[95] The term *rich* in this case can mean more accurate, more complete, or more dependable or refer to the awareness of emergent phenomena. To produce rich data, novel ML techniques that can synthesize multiple datasets at once will be required to help reveal trends in the chemical and physical temporal

evolution and processes that might be obscured or unseen with a single-modality type of experiment (see **Sidebar 7**). Ultimately, the chemical and physical reactions and processes at play need to be predicted, including temporal behavior and transient species evolution in the ultrafast regime. This ability will enable tuning and control of the dynamical evolution of nuclear systems.

Rigorously leveraging ML for multimodal data streams will likely require physics-informed ML models that are capable of being constrained by multiple experimental and computational measurements taken at different energy, length, and timescales, including both dynamic and static properties. This paradigm significantly deviates from the conventional multiscale modeling approach in which information is typically sequentially upscaled from the bottom up (see **Figure 10**). Instead, all data streams should simultaneously inform the model's functional forms and parameters. Scalable ML training and optimal experimental design approaches will be essential to provide real-time feedback while the measurements are being taken, as will the development of fast communication infrastructure between experimental and computational facilities—in particular, leveraging ESNets.^[96] This integration should enable more-efficient model selection or the optimization of specific figures of merit for the target nuclear systems.

4d. Potential Impacts

Foundational scientific impacts are far-reaching. These effects could include understanding the synergistic interactions of microstructural features at multiple scales (e.g., local chemical composition, interface structure, texture) for materials degradation in complex and heterogeneous systems subject to combined external stimuli. This understanding is particularly important for designing the constituent components of future reactors. Integrated computational simulation models that connect primary damage mechanisms to material property degradation will be inherently more accurate and provide greater predictive capability. The synthesis of operando sensing with active coupling to multiscale models will have a broad effect, allowing for, as an example, the active monitoring of additive manufacturing processes.

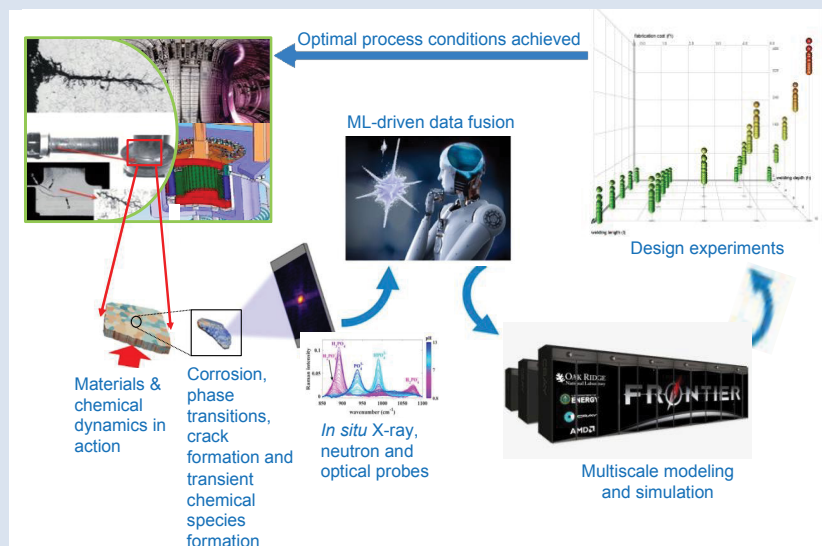
Efforts under this PRO will also provide the ability to integrate active learning strategies to improve experimental data acquisition to ensure experiments are capturing accurate physics. The real-time feedback between these aspects is key. The potential here is to reveal the foundational physics and chemistry dictating the system behavior and have the ability to predict behaviors that are currently inaccessible or extremely time-expensive, such as electronic structure evolution and creep. Additionally, these approaches could generate rich and consistent datasets useful for calibration, benchmarking, and AI/ML across the nuclear energy community.

The effect on energy technologies is the establishment of enabling technology for energy sciences across DOE. This technology is the development of the sensor-fusion architecture for bridging multifidelity, multiresolution experiments; computational modeling; and data science to control the dynamic behavior of *any* complex system. Here, the application space is for nuclear fission and fusion systems, but this idea could be transplanted to tackle other foundational physics challenges (e.g., in additive manufacturing, hydrogen storage, hypersonics, and structural materials).

SIDEBAR 7—EXPERIMENTAL STEERING WITH ML-DRIVEN DATA FUSION. Accelerating materials discovery and addressing knowledge gaps in foundational physical and chemical processes that underpin nuclear fission and fusion will be achieved by bridging multifidelity, multiresolution experiments; multiscale modeling; and data science. A pressing need exists to combine complementary techniques into an integrated, multimodal analysis framework to link characterization experiments with model prediction. Data fusion, bringing together different types of data (from x-ray, neutron, and optical probes) collected across multiple length and timescales (angstroms to centimeters, subpicoseconds to hours) at simultaneous extreme conditions, is challenging. However, by developing new, data-driven, and physics-informed formalisms in ML, efficiently steering high-throughput experiments and providing efficient computational screening tools should be possible. Here, microreactors, advanced small modular reactors, or test vessels, which create the extreme, harsh conditions of interest and are coupled with DOE light sources^[97] and other experimental facilities, are leveraged to enable data–sensor fusion for simultaneous measurements and modeling. For instance, experimental steering could be applied to understanding radiation damage processes in blanket or divertor materials in a tokamak for fusion science^[98] or corrosion processes in MSRs.^[99] While in the extreme environmental state (high-temperature, corrosion, high-intensity laser irradiation for sustained plasma formation/neutron irradiation), in situ measurements of structure, defects, oxidation state, and more are collected. Novel and traditional operando methods—such as electrochemical sensing, microscopy (e.g., dynamic TEM, scanning electron microscopy), time-resolved x-ray diffraction at third- and fourth-generation light sources, neutron and x-ray tomography (e.g., coherent diffractive imaging, phase contrast imaging, and dark-field microscopy), and time-resolved soft/hard x-ray absorption and emission spectroscopies—can be combined to reveal material and chemical dynamics in action, as seen here in corrosion cracking.^[100]

This sensor/data fusion enables simultaneous characterization of processes, which could include transient chemical species formation,^[60] radiation damage cascades influencing microstructures, or the influence of the environment on surface films and chemistry. As data are curated and integrated into a physics-based model, the ML-driven active feedback loop, using large-scale, high-performance computing resources,^[101] is established to aid in steering the experiment. Steering could include what new or next data point should be collected, at what extreme condition, and with what type of measurement to be made over the needed length or timescale where the model is missing data fidelity. Data-driven, multiscale modeling requires high-throughput calculations at various scales and fidelities. This requirement points to the need for leveraging exascale computing to deliver higher simulation and data processing rates.^[92] These

sequences should be automated and uncertainty quantification driven to bridge knowledge gaps across multiple scales. The aim of experimental steering is to provide a holistic picture of materials and chemical challenges facing the fission and fusion energy communities, examining the entirety of advanced fuel cycles or structural materials degradation.



PRO 5: Harness Artificial Intelligence to Design Inherently Resilient Condensed Phases

Overarching Question: What defines self-resilient mechanisms, and how can they be discovered in nuclear materials and chemical systems in coupled extreme environments?

Arguably one of the most critical factors in the fast deployment of advanced nuclear reactors with major improvements in safety and economics is the development of condensed phases, such as materials and coolants, that are resilient to aggressive environments. However, the discovery, improvement, and assessment of materials resistant to extreme environments (see **Sidebar 3**), such as high radiation damage and corrosion rates, in advanced reactors is quite demanding, time-consuming, and costly, which represents a significant barrier to innovation and qualification of novel condensed phases for nuclear energy. Over the past decades, significant effort has been devoted to the development of a bottom-up multiscale modeling framework of nuclear systems (**Figure 10**). This framework is based on first-principles quantum scale modeling at the atomic and picosecond scales to eventually describe macroscale structure–property–performance relationships over decades. Over the past 25 years, the speed of supercomputers has increased roughly tenfold every 5 years. Assuming that same rate in the future, as predicted by Moore’s law, means it would take centuries before one can fully model a nuclear system with fully resolved atomic-scale tools. To circumvent this issue of time, the research community continues to explore scale-bridging methods involving systematically upscaling the lower-scale physics into computationally tractable higher-scale constructs. This time issue has been identified as an ongoing challenge (**PRO 4**). Unfortunately, scale-bridging is plagued by uncertainty propagation across scales. Additionally, this approach is also limited by the simple fact that only known physics can be modeled, and the behavior in coupled extremes is often driven by unknown physics. Finally, this approach hinders the discovery of novel, and potentially groundbreaking, scientific phenomena. Thus, a new modeling paradigm critically needs to be developed to be capable of accelerating prediction of the response of condensed phases to extreme nuclear environments as well as enabling the discovery of novel scientific phenomena.

5a. Summary

Because of the extreme nature of the fission and fusion environments, scientific understanding is quite limited of the responses of condensed phases, such as materials and coolants, to the coupled effects of temperature, irradiation, and environmental degradation. Materials scientists understand well that under irradiation alone, increasing the point defect sink density limits irradiation-induced damage (see **Sidebar 8**). Although manufacturing materials with a high sink density is relatively trivial, the targeted microstructures and microchemistries are often not stable under extreme conditions. Alternatively, if one possesses enough scientific understanding of the degradation processes, a system could be designed that would dynamically evolve toward a metastable state under coupled extremes to become self-resilient to further degradation (see **Sidebar 8**). This strategy is deemed quite promising, as discussed in **PROs 1 and 3**, but is limited and naturally biased by current physical understanding of the degradation mechanisms of materials and coolants. For instance, novel self-resilient mechanisms (i.e. not relying on sink density from manufacturing; see, for instance, driven alloys)^[102] have been identified recently, such as the chemical complexity of CCAs, which would (1) increase irradiation damage cascade-induced phonon scattering, resulting in longer cascade lifetime and less surviving point defects; (2) create a complex vacancy and interstitial migration path distribution, limiting extended defect nucleation and growth kinetics; or (3) form very stable, short-range, ordered structures. Additive manufacturing is also deemed a promising technique to manufacture novel self-resilient alloys. Indeed, the ability to control processing parameters during the build opens up new opportunities such as the design of complex heterostructures (i.e., fuels) or chemical gradients, which could

result in self-resilient systems. Following these two relatively recent examples, it is expected that the identification of additional novel self-resilient mechanisms is within grasp.

However, this identification process is hindered by the vast feature space inherent to these new, promising materials and approaches (see **Sidebar 9**). This hinderance creates an interesting paradigm in which the current approach relying on current physical understanding to discover novel self-resilient mechanisms is inherently limiting, and the search space enabled by novel, promising approaches is deemed too vast to enable such discoveries in a timely fashion. The advent of high-throughput experiments (HTEs) and their coupling to ML and AI could break this conundrum, as evidenced in other materials science fields (see **Sidebar 9**). Interestingly, the dynamic and complex nature of advanced nuclear coolants, such as molten salts, is also facing similar challenges, and HTE coupled to AI/ML could lead to a better chemical and structural understanding of these dynamic ensembles, which is a need detailed under **PROs 1, 2, and 3**.

Thus, HTEs coupled to AI/ML would be a very efficient approach to the identification of novel self-resilient mechanisms for nuclear systems in coupled extremes. However, the success of such approaches often scales with the size and quality of the dataset. Unfortunately, paucity of data is inherent to the nuclear field because of the difficulty in obtaining high-quality and large datasets in coupled extremes, whether experimentally or theoretically. Novel approaches are sought to overcome these challenges, as highlighted under **PRO 4**. Additionally, specifically crafted iterative AI/ML approaches should be designed within those limitations.^[103] In particular, physics-based AI, also known as *scientifically guided AI*, is a promising path to enable the discovery of novel self-resilient mechanisms using data-augmented approaches within limited-size datasets (see schematic in **Figure 11**).

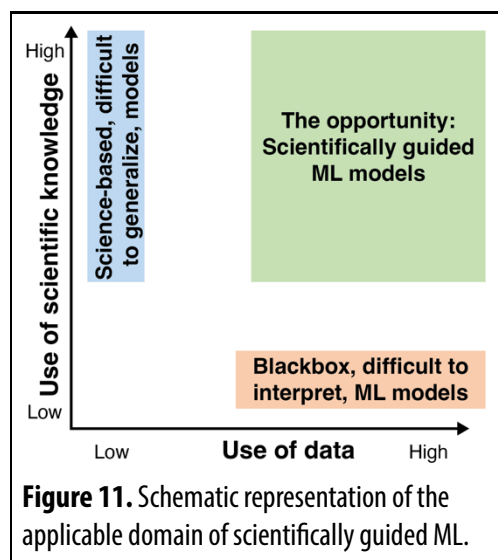
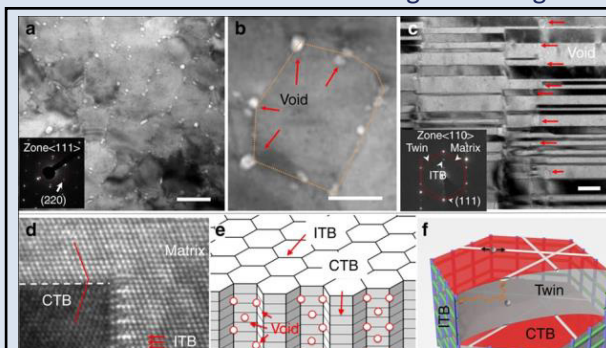


Figure 11. Schematic representation of the applicable domain of scientifically guided ML.

SIDEBAR 8—SELF-RESILIENT MECHANISMS IN NUCLEAR MATERIALS. Radiation damage creates a large population of point defects and small defect clusters. When they recombine and annihilate, they have minimal effects on materials. Self-resilient mechanisms promote recombination and annihilation of defects produced by irradiation. An important scientific and technological challenge is how to design a microstructure to maximize the efficiency of self-resilience and maintain the effectiveness during irradiation or, better yet, evolve toward more resilient states under irradiation. With effective self-resilient mechanisms, radiation effects may no longer be a limiting factor in materials degradation in nuclear reactor environments. Known self-resilient mechanisms rely on various types of defect sinks such as high-angle grain boundaries, immiscible interfaces in nanolayered composites, twin boundaries, free surfaces, and phase boundaries to achieve enhanced radiation tolerance. Nanostructured materials such as nanocrystalline, nanolayered, nanotwinned, and nanoporous materials have generally, but not universally, shown enhanced radiation resistance because of their high-density defect sinks.^[104] Grain boundaries have long been known to act as sinks for all types of defects. Improved radiation resistance in nanocrystalline materials result from their high density of grain boundaries serving as sinks for irradiation-induced defects.^[105] A recent in situ ion irradiation study revealed surprising resilience and self-healing of twin boundaries in response to irradiation.^[106] The migration and removal of various types of defect clusters generated under irradiation was revealed in nanoporous silver by an in situ krypton ion irradiation under TEM, offering new insight into the design of radiation-resistant materials by tailoring nanoscale porosity.^[107] Misra et al. demonstrated that nanolayered niobium–copper composites containing interfaces with controlled sink efficiencies can achieve superior irradiation tolerance, high strength, and high thermal stability.^[108]

Nanoscale particles are also known to act as point defect recombination sites to mitigate damage from irradiation. Nanoparticles of yttrium oxide dispersed in ferritic steels have remarkably improved radiation resistance.^[109] A recent study by Du et al. showed that outstanding radiation resistance in steel can be realized by ultrahigh-density reversible nanoprecipitates.^[110] Defects generated during irradiation were annihilated by the dissolving and renucleation of nearly coherent nanoprecipitates in the steel, a process called reversible disordering-ordering transition. Self-resilience to radiation damage is also influenced by crystal structure and chemical complexity. Face-centered cubic (fcc) crystal structures generally exhibit slightly higher defect production efficiency than body-centered cubic (bcc) crystal structures.^[20] The more finely dispersed distribution of defect clusters observed in bcc crystal systems can also lead to more efficient defect annihilation during subsequent microstructural evolution. The void-swelling of fcc metals is also generally more significant than bcc metals. A recent study has also shown that chemical complexity can be tuned to increase radiation resistance because of the more complex defect diffusion paths in materials with high chemical complexity, suppressing radiation damage accumulation.^[111]

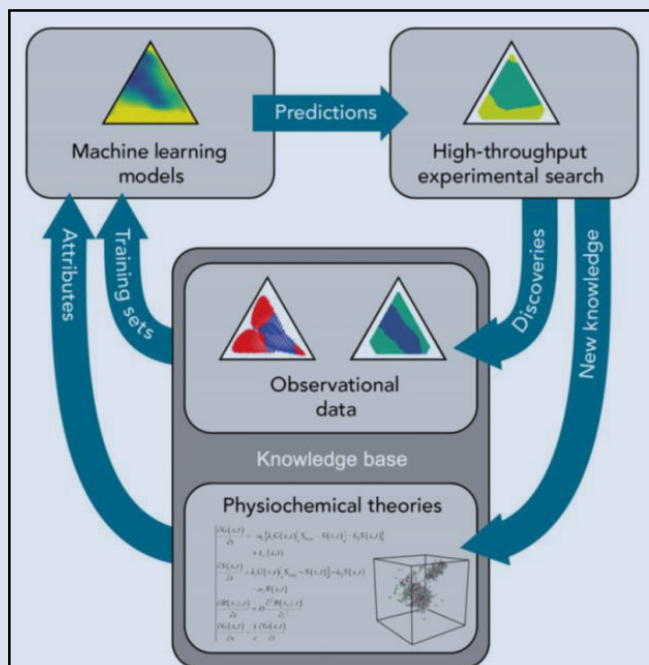


Self-resilient nanostructured materials: (a–b) Plan-view TEM micrograph showing the as-prepared nanovoid-nanotwinned (nv-nt) copper film containing abundant nanovoids primarily surrounding columnar domain boundaries. Scale bar of (a) is 100 nm; scale bar of (b) is 50 nm. (c) Cross-section TEM micrograph shows high-density $\Sigma 3^{[56a]}$ coherent twin boundaries (CTBs) with an average twin thickness of ~ 15 nm and $\Sigma 3^{[110]}$ incoherent twin boundaries (ITBs), which were decorated by a large number of nanovoids with an average diameter of ~ 10 nm. The inserted selected area diffraction pattern confirms the formation of epitaxial nt Cu. Scale bar of (c) is 20 nm. (d) High-resolution TEM image of CTBs and ITBs. (e) A conceptual schematic of metals with CTB and ITB networks and nanovoids. (f) Inside a typical columnar grain radiation-induced interstitials or their loops can rapidly migrate towards ITBs, where they can migrate rapidly to nanovoids.^[104]

SIDEBAR 9—HIGH-THROUGHPUT EXPERIMENT—AI/ML FRAMEWORK APPLIED TO MATERIALS DISCOVERY. As an example, previous sections of this report emphasized the need to investigate CCAs as potential replacements for core internals, radiation shielding, heat exchangers, and more for various nuclear technologies. The chemical landscape of potential new CCAs is as immense as it is exciting. Even restricting the search for new potential five-component alloys to the 30 earth-abundant, nontoxic elements results in billions of possible composition-processing combinations that need to be explored.^[112] This possibility results in far too many compounds to be brute-force calculated or synthesized via traditional high-throughput atomic-scale modeling or experimentation. Coupling these techniques with AI is the only currently existing method that enables the efficient exploration of such a highly dimensional space.^[91c]

To successfully employ AI to discover new materials, a few steps are necessary. For instance, Ren et al. demonstrated that to successfully predict new metallic glasses, one must first encode what is known about the features of materials that predict glass formation into the dataset.^[113] In the case of metallic glasses, possible features are the compositionally averaged melting temperature or the configurational entropy of a given material. Additionally, to train a predictive model, a sufficiently informative dataset is necessary. Depending upon the learning task, the dataset need not be thousands or even hundreds of entries, but it must be sufficiently varied as to capture the expected variation in the observed behavior, and the user should have high confidence in the quality of the labels and values. The AI model can then be trained on the attributes of the training set, and it can be forward-modeled to predict the state of any arbitrary new alloy compositions. Such predictions have to be considered with caution; the model can be wrong for many reasons. Predictions sitting too far away from the training set (called extrapolation), erroneous data points in the training set being too influential for a particular set of predictions, or the

features describing the materials' missing key physics can all lead to incorrect predictions. However, by experimentally (in)validating the AI's predictions, new information can also be provided to the model, improving its overall predictive performance regardless of whether it was correct or incorrect. By including humans in the analysis of the results of the studies, new knowledge is generated that helps to indicate new physicochemical rules that can be included as attributes to boost predictive accuracy of the AI and seed new scientific theory.



Schematic depiction of a paradigm for rapid and guided discovery of materials through iterative combination of ML with HTE.^[113]

5b. Key Scientific Questions

- What are signatures of self-resilient mechanisms, and how can their discovery be facilitated in nuclear condensed phases, such as materials and coolants, considering currently limited scientific understanding of materials' degradation in coupled extreme environments?
- Can a physics-based framework be integrated into AI models to enable AI for accurate prediction and extrapolation, further accelerating discovery of resilient condensed phases?
- How do researchers accurately identify and understand important feature–property relationships in condensed phases for fission and fusion applications?
- How do researchers design materials that will evolve to a favorable, dynamical, metastable state under extreme conditions?

5c. Scientific Challenges and Research Opportunities

- Develop AI models to discover important mechanistic features, or vector of features, with the potential to identify novel self-resilient mechanisms.
- Improve physics-based models with scientifically guided AI models to enable interpolation and extrapolation of complex multiscale feature–property relationships.

Develop AI models to discover important mechanistic features, or vectors of features, with the potential to identify novel self-resilient mechanisms.

The use of data-centric methods to improve predictability in materials science is not new, spanning from the development of the periodic table to novel automated semiconductor searches across thousands of compounds. However, the advent of AI methods has incredibly improved the capacity to analyze large datasets, predict materials' responses in complex environments, and discover features of importance, including within radiative and harsh environments, in driving these responses.^[114] Finding such features of importance using AI computational methods can also help validate physically based models.^[114] For example, the influential features found in the neural network–based prediction of reactor pressure vessel steel embrittlement under irradiation have been found to be consistent with the general physical understanding of the dominant contributors to reactor pressure vessel behavior.^[115] This validation further reinforces the ability of the data-centric model to interpolate, and possibly extrapolate, to other domains. Alternatively, AI approaches can also identify non-expected features of importance, potentially unraveling new science. For instance, it was recently discovered using various ML regression models, such as Random Forest Regressor, that the noble element (nickel) bulk diffusion coefficient was the most important feature governing the corrosion rate of FeCrMnNi CCAs in high-temperature molten salts.^[116] This result was not originally expected and validates that a phenomenon analogous to dealloying occurs in high-temperature systems, paving the way for novel corrosion-resistant alloy design (see **Figure 12**).

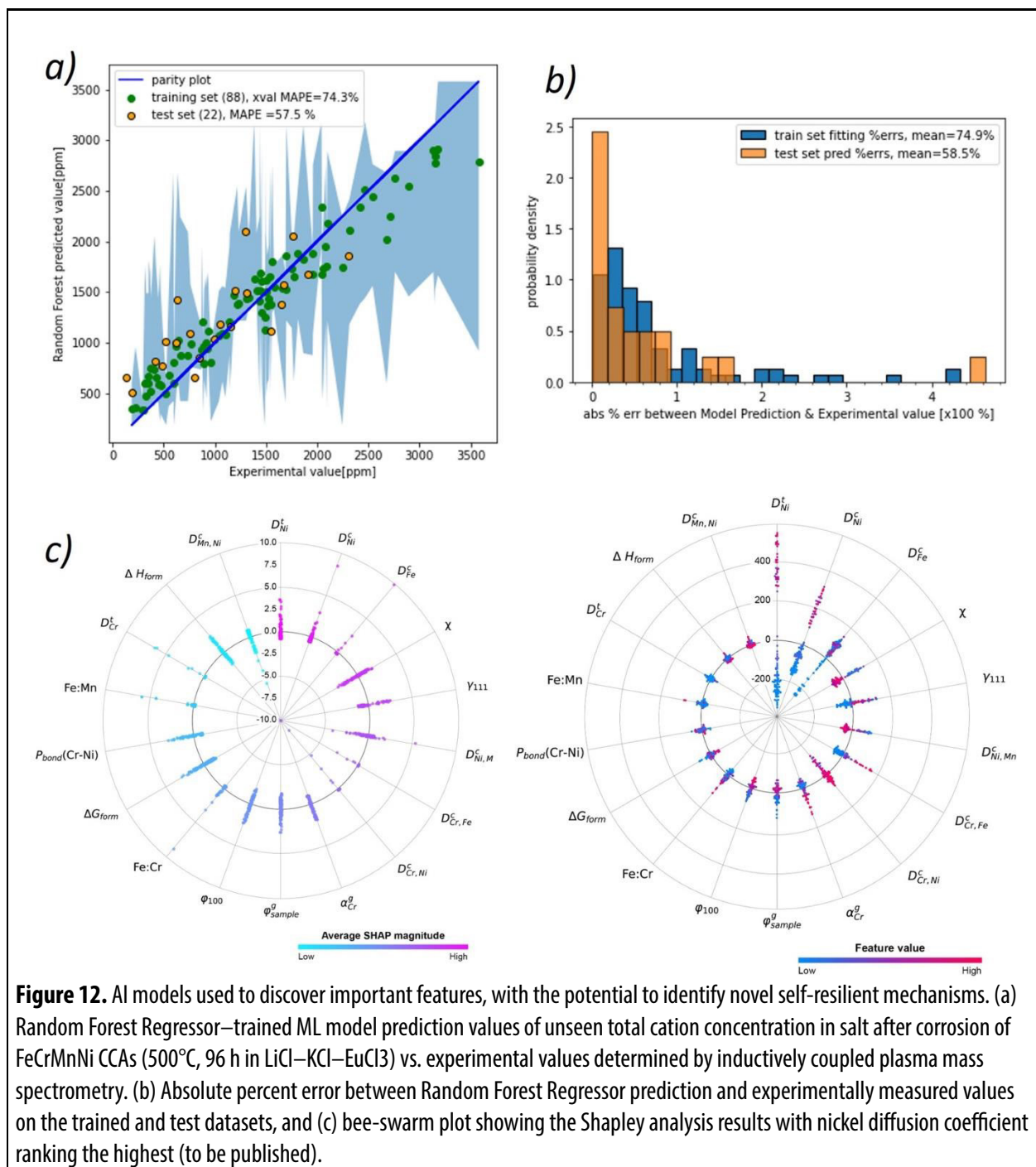


Figure 12. AI models used to discover important features, with the potential to identify novel self-resilient mechanisms. (a) Random Forest Regressor—trained ML model prediction values of unseen total cation concentration in salt after corrosion of FeCrMnNi CCAs (500°C, 96 h in LiCl–KCl–EuCl₃) vs. experimental values determined by inductively coupled plasma mass spectrometry. (b) Absolute percent error between Random Forest Regressor prediction and experimentally measured values on the trained and test datasets, and (c) bee-swarm plot showing the Shapley analysis results with nickel diffusion coefficient ranking the highest (to be published).

The responses of systems in coupled extremes are unlikely to be driven by a single feature, and clustering approaches, such as K-means, are quite powerful in identifying vectors of features that are driving that response. At the same time, multiobjective optimization, in which multiple functionalities are optimized concurrently, is critical for the development of resilient materials for coupled extremes. Using those AI approaches appears quite promising in the field of nuclear systems to identify novel self-resilient mechanisms based on the concurrent effects of multiple features. For instance, examining the effects of electronic (**PRO 1**) and phonon band structures on irradiation cascade development and point defect diffusion paths using AI-based clustering

approaches could help better understand the specific self-resilient mechanisms of CCAs to irradiation. Of course, to fully enable the power of AI methods in the context of nuclear materials and coolants, the ability to automatically, and possibly autonomously, irradiate, corrode, and/or stress a variety of samples at different temperatures and rates with accelerated experiments and high-throughput online diagnostic tools will become absolutely necessary (see **PRO 4**). Researchers expect that high-throughput experiment irradiation and/or corrosion tests will be routinely performed in extreme environments in the future. However, these accelerated experiments must be correlated quantitatively to actual in-reactor conditions, and AI can help establish such connections. In particular, a mapping between high-fidelity, high-quality (but expensive and slow) experiments and high-throughput experiments could enable high-quality and accelerated predictions for new compositions of interest.

Improve physics-based models with scientifically-guided AI models to enable interpolation and extrapolation of complex multiscale feature-property relationships.

Physics-based modeling is the foundation for advancements in understanding environmental effects and structure–property relationships in fission and fusion materials, as well as for the development of new condensed phases. Physics-based modeling will continue to be a crucial component of materials research for nuclear energy. However, physics-based models face certain limitations (for instance, they are brittle and unreliable outside the bounds of their applicability). Conversely, AI/ML models are infinitely flexible, capable of constantly updating their predictions with the incorporation of new data. But they fail in extrapolation (generalization) and can overestimate their uncertainties outside the bounds of the data with which they were trained. Incorporating physics-based models into AI can be used to leverage the strengths of each approach and expand and accelerate the discovery of new materials and physical laws. Several avenues exist to unite scientifically guided AI and physics-based models, including using ML to improve the numerical description within physics-based models, using ML to extrapolate or interpolate from simpler to more complex systems, and using AI to determine underlying sources of variability for macroscopic material-response function (property) relationships. In the rest of this section, a few generalized examples are discussed.

Physics-based models frequently involve generalized equations that need to be parameterized or even specified with different mathematical forms. Examples of models that are complex to parameterize are the cluster expansion^[118] and the cluster dynamics methods;^[119] an example of a model that requires different mathematical forms to describe different systems is the interatomic potentials used in molecular dynamics.^[120] In these cases, ML can be effectively used to provide a more accurate numerical description of a given system represented in the physics-based model, providing more accurate results of the system behavior or characteristics. Alternative approaches to integrating AI with physics-based models involve

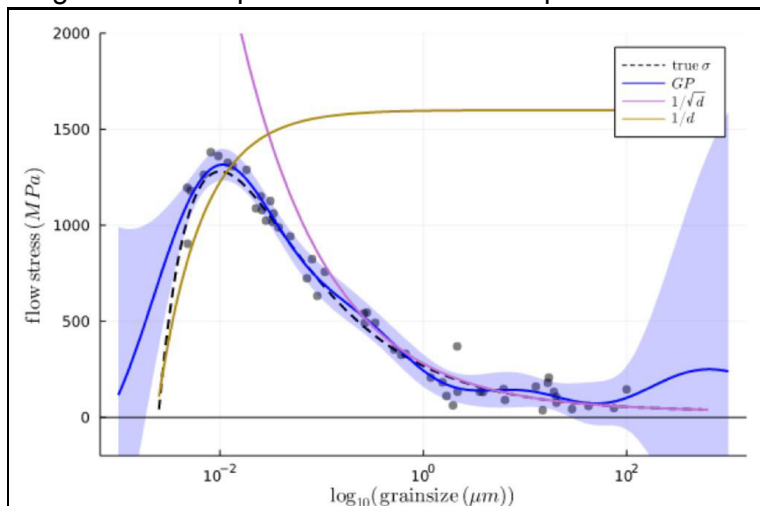


Figure 13. A Gaussian process model can effectively reproduce the grain size dependence of the mechanical strength of an alloy even though it is completely devoid of any knowledge of the effect of the density of grain boundaries for large-grain metals, the effect of grain boundary sliding in nanocrystalline alloys, or even the regime change.^[117]

interpolative or extrapolative methods for determining more complex system properties based on the characteristics of simpler systems. For example, a physics-based model can be used to predict a given property of several two-component systems, and physics-based AI can be used to predict the property for a three- or four-component system based on the two-component systems, for which a physics-based computation would be significantly more expensive (e.g., the elastic constants of multicomponent systems predicted from bicomponent systems). This method is well-suited for relationships that are qualitatively understood but not easily quantifiable. However, the degree of extrapolation and its influence on model performance must be carefully assessed. As an example, DeCost et al. demonstrated the creation of an AI model that combined empirical relationships between grain size and corrosion rate with a nonparametric model that accounts for the compositional contribution to corrosion resistance. Using this model, estimates of the Hall–Petch coefficients for each alloy were extracted, and a grain-size independent corrosion rate was also extracted.^[121] However, one challenge to AI-based extrapolative methods is the possibility of a sharp transition in a property because of, for example, a phase change (see **Figure 13**).^[117]

The use of *big data* for black-box, brute-force number crunching for nuclear materials and coolants or solvents is less likely to occur for macroscale testing to predict properties because of the many causes for variability within materials. This method could be useful for determining sources of variability within a relationship between a measured response function and underlying material properties such as composition or grain structure, and in identification of property transition points. Computer vision is another area in which scientifically guided AI can provide a major impact. Conventional image analysis is laborious, resulting in low data throughput. Improving the quantity, quality, and uncertainty quantification of image statistics will improve the ability of researchers to quantify feature-property relationships.^[122]

5d. Potential Impacts

The rise in the use of AI/ML in the physical sciences demonstrates a strong possibility for using this approach to discover and develop novel, self-resilient materials for extreme environments. Multi-objective, scientifically guided AI frameworks have the potential to improve alloy design in coupled extremes and enable the discovery of new self-resilient mechanisms for materials under extreme environments. This new opportunity in nuclear materials research will have significant scientific impact by enhancing understanding of structure–property–performance relationships and accelerating the discovery of new mechanisms using scientific knowledge *and* data, either from the literature or acquired in operando. Successes in this area would provide novel resilient materials on a timeline conducive to high-temperature nuclear fission and fusion systems deployment. Additionally, using scientifically guided AI methods to design materials resilient to extreme environments would serve as a guide for other scientific and technology fields (e.g., renewable energies, space exploration) and could possibly be adapted for novel and robust, repeatable, predictable processing routes to prequalify high-temperature materials for nuclear applications.

3. Closing Remarks

This report describes existing knowledge gaps, scientific and technical challenges, and, most of all, the fundamental science needs for successfully supporting and implementing innovation in nuclear technologies. These needs are summarized in the five PROs presented herein that are designed to accelerate discovery and set research priorities for the development and deployment of robust, resilient, advanced nuclear systems over the next decade. The PROs build upon but reach further than established research directions outlined in previous BES workshops and reports; they also provide a framework for the basic research needed to develop novel materials, coolants, and solvents that will serve as the foundation of future nuclear energy systems. The last few years have seen the advent of new reactor designs, both for fission and fusion. Further innovation can be expected in the coming years, but it will be accelerated by a solid scientific foundation for understanding, describing, and predicting the behavior and performance of condensed phases in coupled extreme environments. The PROs described here detail advances in understanding materials, coolants, and solvents in extreme conditions as well as the development of novel experimental, modeling, and data science approaches that will aid in the innovative design of new reactor concepts. Furthermore, the science represented by these PROs will have broader effects, aiding in the development of materials and chemical systems that will benefit renewable energy, microelectronics, and other applications in which robust materials are needed for enhanced performance.

4. References

- [1] E. a. E. S. Institute, **2021**.
- [2] L. Varro, B. Wanner, C. Hernández Alva, A. Herzog, P. Fraser, *International Energy Agency*. <https://webstore.iaea.org/nuclear-power-in-a-clean-energy-system> **2019**.
- [3] <https://www.eia.gov> from February 2024.
- [4] Available from: <https://www.energy.gov/articles/infographic-nuclear-energy-numbers>.
- [5] R. Betti, *Nature Reviews Physics* **2023**, *5*, 6-8.
- [6] J. Deutch, *Joule* **2023**, *7*, 603-606.
- [7] O. Hurricane, P. Patel, R. Betti, D. Froula, S. Regan, S. Slutz, M. Gomez, M. Sweeney, *Reviews of Modern Physics* **2023**, *95*, 025005.
- [8] H. McCoy, R. Beatty, W. Cook, R. Gehlbach, C. Kennedy, J. Koger, A. Litman, C. Sessions, J. Weir, *Nuclear applications and technology* **1970**, *8*, 156-169.
- [9] G. Locatelli, M. Mancini, N. Todeschini, *Energy Policy* **2013**, *61*, 1503-1520.
- [10] (a) O. Camara, M. A. Tunes, G. Greaves, A. H. Mir, S. Donnelly, J. A. Hinks, *Ultramicroscopy* **2019**, *207*, 112838; (b) A. Majumdar, K. V. Manukyan, S. Dede, J. M. Roach, D. Robertson, P. C. Burns, A. Aprahamian, *ACS Applied Materials & Interfaces* **2021**, *13*, 35153-35164; (c) S. Xu, C. Zheng, Y. Bi, Q. Mao, H. Qin, X. Li, *Journal of the European Ceramic Society* **2023**, *43*, 1376-1384; (d) L. Zhang, W. Jiang, S. Wang, T. Varga, C. Pan, Z. Wang, L. Chen, B. Li, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **2021**, *507*, 1-6.
- [11] (a) J. Gao, K. Yabuuchi, A. Kimura, *Journal of Nuclear Materials* **2018**, *511*, 304-311; (b) K. A. Smith, A. I. Savva, Y. Wu, D. A. Tenne, D. P. Butt, H. Xiong, J. P. Wharry, *Journal of the American Ceramic Society* **2018**, *101*, 4357-4366.
- [12] D. J. Sprouster, L. L. Snead, E. Dooryhee, S. K. Ghose, T. Koyanagi, Y. Katoh, *Journal of Nuclear Materials* **2019**, *527*, 151798.
- [13] (a) J. Noiro, Y. Pontillon, J. Lamontagne, I. Zacharie-Aubrun, K. Hanifi, P. Bienvenu, L. Desgranges, in *EPJ Web of Conferences, Vol. 115*, EDP Sciences, **2016**, p. 04005; (b) M. Tonks, D. Andersson, R. Devanathan, R. Dubourg, A. El-Azab, M. Freyss, F. Iglesias, K. Kulacsy, G. Pastore, S. R. Phillpot, *Journal of Nuclear Materials* **2018**, *504*, 300-317.
- [14] V. V. Rondinella, T. Wiss, *Materials today* **2010**, *13*, 24-32.
- [15] D. Jadermas, J. Gan, D. Keiser, J. Madden, M. Bachhav, J.-F. Jue, A. Robinson, *Journal of Nuclear Materials* **2018**, *509*, 1-8.
- [16] W. J. Weber, D. M. Duffy, L. Thomé, Y. Zhang, *Current Opinion in Solid State and Materials Science* **2015**, *19*, 1-11.
- [17] Y. Zhang, H. Xue, E. Zarkadoula, R. Sachan, C. Ostrouchov, P. Liu, X.-I. Wang, S. Zhang, T. S. Wang, W. J. Weber, *Current Opinion in Solid State and Materials Science* **2017**, *21*, 285-298.
- [18] Y. Zhang, C. Silva, T. G. Lach, M. A. Tunes, Y. Zhou, L. Nuckols, W. L. Boldman, P. D. Rack, S. E. Donnelly, L. Jiang, *Current Opinion in Solid State and Materials Science* **2022**, *26*, 101001.
- [19] Y. Zhang, T. Varga, M. Ishimaru, P. Edmondson, H. Xue, P. Liu, S. Moll, F. Namavar, C. Hardiman, S. Shannon, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* **2014**, *327*, 33-43.
- [20] S. J. Zinkle, L. L. Snead, *Annual Review of Materials Research* **2014**, *44*, 241-267.
- [21] (a) J. Marian, T. Hoang, M. Fluss, L. L. Hsiung, *Journal of Nuclear Materials* **2015**, *462*, 409-421; (b) R. Sugano, K. Morishita, A. Kimura, *Fusion science and technology* **2003**, *44*, 446-449.

- [22] (a) R. Abernethy, *Materials Science and Technology* **2017**, *33*, 388-399; (b) G. De Temmerman, K. Bystrov, R. Doerner, L. Marot, G. Wright, K. Woller, D. Whyte, J. Zielinski, *Journal of Nuclear Materials* **2013**, *438*, S78-S83; (c) J. Marian, C. S. Becquart, C. Domain, S. L. Dudarev, M. R. Gilbert, R. J. Kurtz, D. R. Mason, K. Nordlund, A. E. Sand, L. L. Snead, *Nuclear Fusion* **2017**, *57*, 092008.
- [23] X.-Y. Liu, D. Andersson, B. Uberuaga, *Journal of Materials Science* **2012**, *47*, 7367-7384.
- [24] (a) T. J. Gerczak, J. D. Hunn, R. A. Lowden, T. R. Allen, *Journal of Nuclear Materials* **2016**, *480*, 257-270; (b) I. Van Rooyen, M. Dunzik-Gougar, P. Van Rooyen, *Nuclear Engineering and Design* **2014**, *271*, 180-188.
- [25] D. D. Keiser Jr, *Journal of Nuclear Materials* **2019**, *514*, 393-398.
- [26] J. Wormald, A. Hawari, *Journal of Nuclear Materials* **2022**, *566*, 153797.
- [27] B. Derby, J. Cooper, T. Lach, E. Martinez, H. Kim, J. Baldwin, D. Kaoumi, D. J. Edwards, D. K. Schreiber, B. P. Uberuaga, *Surface and Coatings Technology* **2020**, *403*, 126346.
- [28] M. W. Terban, S. J. Billinge, *Chemical Reviews* **2021**, *122*, 1208-1272.
- [29] J. B. S. Junior, G. R. Schleder, J. Bettini, I. C. Nogueira, A. Fazzio, E. R. Leite, *Matter* **2021**, *4*, 441-460.
- [30] Y. Yang, J. Zhou, F. Zhu, Y. Yuan, D. J. Chang, D. S. Kim, M. Pham, A. Rana, X. Tian, Y. Yao, *Microscopy and Microanalysis* **2022**, *28*, 224-226.
- [31] (a) R. Auguste, M. Liedke, F. Selim, B. P. Uberuaga, A. Wagner, P. Hosemann, *JOM* **2020**, *72*, 2436-2444; (b) F. Selim, *Materials Characterization* **2021**, *174*, 110952.
- [32] T. L. Wilson, S. C. Vogel, J. T. White, A. D. R. Andersson, E. Sooby-Wood, T. M. Besmann, *Materialia* **2019**, *9*.
- [33] (a) W. Xiong, W. Xie, C. Shen, D. Morgan, *Journal of nuclear materials* **2013**, *443*, 331-341; (b) T. Yao, A. Sen, A. Wagner, F. Teng, M. Bachhav, E.-A. Anter, D. Murray, J. Gan, D. H. Hurley, J. P. Wharry, *Materialia* **2021**, *16*, 101092.
- [34] E. Sooby, A. Nelson, J. White, P. McIntyre, *Journal of Nuclear Materials* **2015**, *466*, 280-285.
- [35] A. P. Shivprasad, V. Kocevski, T. L. Ulrich, J. R. Wermer, D. Andersson, J. T. White, *Journal of Nuclear Materials* **2022**, *558*, 153278.
- [36] J. A. Moriarty, *Physical review letters* **1985**, *55*, 1502.
- [37] D. Pettifor, I. Oleinik, *Physical review B* **1999**, *59*, 8487.
- [38] G. Liu, D. Nguyen-Manh, B.-G. Liu, D. Pettifor, *Physical Review B* **2005**, *71*, 174115.
- [39] (a) B. Himmetoglu, A. Floris, S. De Gironcoli, M. Cococcioni, *International Journal of Quantum Chemistry* **2014**, *114*, 14-49; (b) P. F. Weck, E. Kim, *Physical Chemistry Chemical Physics* **2016**, *18*, 26816-26826.
- [40] A. Moore, C. Deo, M. Baskes, M. Okuniewski, *Acta Materialia* **2016**, *115*, 178-188.
- [41] K. E. Johnson, D. L. Adorno, V. Kocevski, T. L. Ulrich, J. T. White, A. Claisse, J. W. McMurray, T. M. Besmann, *Journal of Nuclear Materials* **2020**, *537*, 152235.
- [42] O. Beneš, R. Konings, *Journal of Fluorine Chemistry* **2009**, *130*, 22-29.
- [43] I. Bogatyrev, D. Grojo, P. Delaporte, S. Leyder, M. Sentis, W. Marine, T. Itina, *Journal of Applied Physics* **2011**, *110*, 103106.
- [44] S. Batzner, A. Musaelian, L. Sun, M. Geiger, J. P. Mailoa, M. Kornbluth, N. Molinari, T. E. Smidt, B. Kozinsky, *Nature communications* **2022**, *13*, 2453.
- [45] L. Lu, H. Hu, A. J. Jenkins, X. Li, *Journal of Chemical Theory and Computation* **2022**, *18*, 2983-2992.
- [46] L. Huang, K. Ma, L. T. Belkacemi, M. Loyer-Prost, E. Meslin, E. Toijer, L. Messina, C. Domain, J. Vidal, M. Nastar, *Acta Materialia* **2022**, *225*, 117531.
- [47] X. Yi, S. Sun, H. Wang, Z. Gao, W. Cai, X. Zhang, *Intermetallics* **2021**, *138*, 107305.

- [48] (a) J. Startt, R. Dingreville, S. Raiman, C. Deo, *Acta Materialia* **2021**, *218*, 117215; (b) H. Bernas, J. P. Attané, K. H. Heinig, D. Halley, D. Ravelosona, A. Marty, P. Auric, C. Chappert, Y. Samson, *Physical Review Letters* **2003**, *91*, 077203.
- [49] (a) L. L. Snead, S. J. Zinkle, D. P. White, *Journal of Nuclear Materials* **2005**, *340*, 187-202; (b) K. Yang, E. Kardoulaki, D. Zhao, A. Broussard, K. Metzger, J. T. White, M. R. Sivack, K. J. McClellan, E. J. Lahoda, J. Lian, *Journal of Nuclear Materials* **2021**, *557*, 153272; (c) S. S. Parker, J. T. White, P. Hosemann, A. T. Nelson, *Journal of Nuclear Materials* **2019**, *526*, 151760; (d) J. T. White, A. W. Travis, J. T. Dunwoody, A. T. Nelson, *Journal of Nuclear Materials* **2017**, *495*, 463-474.
- [50] (a) J. A. Aguiar, A. M. Jokisaari, M. Kerr, R. Allen Roach, *Nature Communications* **2020**, *11*, 2556; (b) K. A. Terrani, N. A. Capps, M. J. Kerr, C. A. Back, A. T. Nelson, B. D. Wirth, S. L. Hayes, C. R. Stanek, *Journal of Nuclear Materials* **2020**, *539*, 152267.
- [51] F. Wu, S. Roy, A. S. Ivanov, S. K. Gill, M. Topsakal, E. Dooryhee, M. Abeykoon, G. Kwon, L. C. Gallington, P. Halstenberg, *The journal of physical chemistry letters* **2019**, *10*, 7603-7610.
- [52] (a) K. Beierschmitt, M. Buchanan, A. Clark, I. Robertson, P. Britt, A. Navrotsky, P. Burns, P. Tortorelli, A. Misra, J. Wishart, P. Fenter, A. Gewirth, B. Wirth, B. Mincher, I. Szlufarska, J. Busby, L. Horton, B. Garrett, J. Vetrano, P. Wilk, K. Runkles, S. Kung, S. Lesica, B. Wyatt, D. Counce, K. Jones, United States, **2017**, p. Medium: ED; Size: 128 p; (b) J. Roberto, T. Diaz de la Rubia, R. Gibala, S. Zinkle, J. R. Miller, S. Pimblott, C. Burns, K. Raymond, R. Grimes, K. Pasamehmetoglu, S. Clark, R. Ewing, A. Wagner, S. Yip, M. Buchanan, G. Crabtree, J. Hemminger, J. Poate, J. C. Miller, N. Edelstein, T. Fitzsimmons, G. Gruzalski, G. Michaels, L. Morss, M. Peters, K. Talamini, United States, **2006**; (c) J. F. Wishart, *Innovative Separations R&D Needs for Advanced Fuel Cycles Workshop* **2022**, Medium: X.
- [53] C. Celis Barros, C. D. Pilgrim, A. R. Cook, S. P. Mezyk, T. S. Grimes, G. P. Horne, *Physical Chemistry Chemical Physics* **2021**, *23*, 24589-24597.
- [54] *High Temperature Gas Cooled Reactor Fuels and Materials*, INTERNATIONAL ATOMIC ENERGY AGENCY, Vienna, **2010**.
- [55] B. A. Moyer, G. J. Lumetta, S. Bruffey, S. Finkeldei, K. Marsden, M. F. Simpson, M. Jensen, P. R. Zalupski, A. E. Clark, P. Yang, Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States), **2022**.
- [56] (a) S. H. Ha, R. N. Menchavez, Y.-M. Koo, *Korean Journal of Chemical Engineering* **2010**, *27*, 1360-1365; (b) J. F. Wishart, I. A. Shkrob, ACS Publications, **2009**.
- [57] (a) M. Jaszczur, M. A. Rosen, T. Śliwa, M. Dudek, L. Pieńkowski, *International Journal of Hydrogen Energy* **2016**, *41*, 7861-7871; (b) R. Pinsky, P. Sabharwall, J. Hartvigsen, J. O'Brien, *Progress in Nuclear Energy* **2020**, *123*, 103317; (c) D. Ryland, H. Li, R. Sadhankar, *International Journal of Energy Research* **2007**, *31*, 1142-1155; (d) S. Şahin, H. M. Şahin, *International Journal of Hydrogen Energy* **2021**, *46*, 28936-28948.
- [58] B. J. Mincher, S. P. Mezyk, *rca-Radiochimica Acta* **2009**, *97*, 519-534.
- [59] (a) M. Fairley, S. E. Gilson, S. L. Hanna, A. Mishra, J. G. Knapp, K. B. Idrees, S. Chheda, H. Traustason, T. Islamoglu, P. C. Burns, *Chemistry of Materials* **2021**, *33*, 9285-9294; (b) M. Fairley, N. M. Myers, J. E. Szymanowski, G. E. Sigmon, P. C. Burns, J. A. LaVerne, *Inorganic Chemistry* **2019**, *58*, 14112-14119; (c) S. E. Gilson, M. Fairley, S. L. Hanna, J. E. Szymanowski, P. Julien, Z. Chen, O. K. Farha, J. A. LaVerne, P. C. Burns, *Journal of the American Chemical Society* **2021**, *143*, 17354-17359; (d) S. L. Hanna, D. X. Rademacher, D. J. Hanson, T. Islamoglu, A. K. Olszewski, T. M. Nenoff, O. K. Farha, *Industrial & Engineering Chemistry Research* **2020**, *59*, 7520-7526.
- [60] H. E. Lackey, G. L. Nelson, A. M. Lines, S. A. Bryan, *Analytical chemistry* **2020**, *92*, 5882-5889.

- [61] (a) A. T. Bell, B. C. Gates, D. Ray, M. R. Thompson, Pacific Northwest National Lab.(PNNL), Richland, WA (United States), **2008**; (b) J. B. Goodenough, H. D. Abruna, M. V. Buchanan, DOESC (USDOE Office of Science (SC)), **2007**; (c) P. Alivisatos, P. Cummings, J. De Yoreo, K. Fishthorn, B. Gates, R. Hwang, D. Lowndes, A. Majumdar, L. Makowski, T. Michalske, **2005**; (d) J. Stringer, L. Horton, *Report by the DOE-BES–Advisory Committee, ORNL Report 2003*; (e) M. Dresselhaus, G. Crabtree, M. Buchanan, T. Mallouk, Technical report, Argonne National Laboratory, Basic Energy Sciences, US DOE ..., **2003**; (f) S. B. Clark, M. Buchanan, B. Wilmarth, Pacific Northwest National Lab.(PNNL), Richland, WA (United States), **2016**; (g) P. Alivisatos, M. Buchanan, USDOE Office of Science (SC)(United States), **2010**; (h) J. C. Hemminger, J. Sarrao, G. Crabtree, G. Flemming, M. Ratner, USDOE Office of Science (SC)(United States), **2015**; (i) J. Wadsworth, G. Crabtree, R. Hemley, R. Falcone, I. Robertson, J. Stringer, P. Tortorelli, G. Gray, M. Nicol, J. Lehr, DOESC (USDOE Office of Science (SC)), **2008**.
- [62] (a) G. Was, D. Petti, S. Ukai, S. Zinkle, *Journal of Nuclear Materials* **2019**, 527, 151837; (b) J. Čížek, J. Kalivodová, M. Janeček, J. Stráský, O. Srba, A. Macková, *Metals* **2021**, 11, 76; (c) A. O. Moghaddam, A. Cabot, E. A. Trofimov, *International Journal of Refractory Metals and Hard Materials* **2021**, 97, 105504.
- [63] G. Baldacchino, B. Hickel, in *Radiation Chemistry*, EDP Sciences, **2021**, pp. 53-64.
- [64] (a) D. E. Holcomb, R. A. Kisner, S. M. Cetiner, Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States), **2018**; (b) G. Lu, A. J. Haes, T. Z. Forbes, *Coordination chemistry reviews* **2018**, 374, 314-344; (c) C. A. Dennett, K. So, A. Kushima, D. Buller, K. Hattar, M. Short, *Acta Materialia* **2018**, 145, 496-503.
- [65] F. Schmidt, P. Hosemann, R. O. Scarlat, D. K. Schreiber, J. R. Scully, B. P. Uberuaga, *Annual Review of Materials Research* **2021**, 51, 293-328.
- [66] Y. Li, J. Zhang, Q. Chen, X. Xia, M. Chen, *Advanced Materials* **2021**, 33, 2100855.
- [67] (a) Y. Katoh, L. L. Snead, *Journal of Nuclear Materials* **2019**, 526, 151849; (b) Z. Su, H. Jiang, H. Li, Y. Zhang, J. Chen, J. Zhao, Y. Ma, *ChemNanoMat* **2023**, 9, e202200477.
- [68] S. Guo, J. Zhang, W. Wu, W. Zhou, *Progress in Materials Science* **2018**, 97, 448-487.
- [69] (a) C. Fazio, F. Balbaud, in *Structural Materials for Generation IV Nuclear Reactors*, Elsevier, **2017**, pp. 23-74; (b) G. S. Was, T. R. Allen, *Structural Alloys for Nuclear Energy Applications* **2019**, 211-246.
- [70] W. Zhou, Y. Yang, G. Zheng, K. B. Woller, P. W. Stahle, A. M. Minor, M. P. Short, *Nature communications* **2020**, 11, 3430.
- [71] C. Si, W. Lu, J. Ji, S. Zhang, X. Yao, W. Wang, D. Chu, in *Journal of Physics: Conference Series, Vol. 1637*, IOP Publishing, **2020**, p. 012037.
- [72] C. Tsay, M. Baldea, *Industrial & Engineering Chemistry Research* **2019**, 58, 16696-16708.
- [73] (a) Q.-J. Li, E. Küçükbenli, S. Lam, B. Khaykovich, E. Kaxiras, J. Li, *Cell Reports Physical Science* **2021**, 2, 100359; (b) M.-T. Nguyen, R. Rousseau, P. D. Paviet, V.-A. Glezakou, *ACS Applied Materials & Interfaces* **2021**, 13, 53398-53408; (c) G. Sivaraman, J. Guo, L. Ward, N. Hoyt, M. Williamson, I. Foster, C. Benmore, N. Jackson, *The Journal of Physical Chemistry Letters* **2021**, 12, 4278-4285; (d) T. Porter, M. M. Vaka, P. Steenblik, D. Della Corte, *Communications Chemistry* **2022**, 5, 69.
- [74] A. Dworkin, H. Bronstein, M. Bredig, *The Journal of Physical Chemistry* **1962**, 66, 572-573.
- [75] A. Merwin, W. C. Phillips, M. A. Williamson, J. L. Willit, P. N. Motsegood, D. Chidambaram, *Scientific Reports* **2016**, 6, 25435.
- [76] J. Guo, A. Merwin, C. J. Benmore, Z.-G. Mei, N. C. Hoyt, M. A. Williamson, *The Journal of Physical Chemistry B* **2019**, 123, 10036-10043.

- [77] S. L. Riechers, N. Petrik, J. S. Loring, M. K. Murphy, C. I. Pearce, G. A. Kimmel, K. M. Rosso, *Review of Scientific Instruments* **2021**, *92*, 113701.
- [78] S. A. McMaster, R. Ram, N. Faris, M. I. Pownceby, *Journal of hazardous materials* **2018**, *360*, 257-269.
- [79] (a) N. Birbilis, S. Choudhary, J. Scully, M. Taheri, *npj Materials Degradation* **2021**, *5*, 14; (b) A. Kareer, J. Waite, B. Li, A. Couet, D. Armstrong, A. Wilkinson, *Journal of Nuclear Materials* **2019**, *526*, 151744.
- [80] C. A. Dennett, N. Poudel, P. J. Simmonds, A. Tiwari, D. H. Hurley, K. Gofryk, *Nature Communications* **2022**, *13*, 2221.
- [81] G. Horwitz, E. Härk, P. Y. Steinberg, L. P. Cavalcanti, S. Risse, H. R. Corti, *ACS nano* **2021**, *15*, 11564-11572.
- [82] M. R. Gilbert, K. Arakawa, Z. Bergstrom, M. J. Caturla, S. L. Dudarev, F. Gao, A. Goryaeva, S. Hu, X. Hu, R. J. Kurtz, *Journal of Nuclear Materials* **2021**, *554*, 153113.
- [83] D. Ramachandram, G. W. Taylor, *IEEE signal processing magazine* **2017**, *34*, 96-108.
- [84] N. D. Bull Ezell, R. A. Kisner, N. G. Russell, F. K. Reed, J. R. Keiser, P. A. Champlin, A. J. Martin, D. E. Holcomb, Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States), **2019**.
- [85] S. R. Spurgeon, C. Ophus, L. Jones, A. Petford-Long, S. V. Kalinin, M. J. Olszta, R. E. Dunin-Borkowski, N. Salmon, K. Hattar, W.-C. D. Yang, *Nature materials* **2021**, *20*, 274-279.
- [86] (a) H. Andrews, J. McFarlane, D. E. Holcomb, N. Ezell, K. Myhre, A. Lines, S. Bryan, H. M. Felmy, Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States), **2021**; (b) M. Yamashita, H. Konishi, T. Kozakura, J. Mizuki, H. Uchida, *Corrosion Science* **2005**, *47*, 2492-2498.
- [87] A. Jones, R. Greaves, C. Coddling, F. Selim, *Review of Scientific Instruments* **2022**, *93*, 043903.
- [88] M. Passoni, F. Arioli, L. Cialfi, D. Dellasega, L. Fedeli, A. Formenti, A. C. Giovannelli, A. Maffini, F. Mirani, A. Pazzaglia, *Plasma Physics and Controlled Fusion* **2019**, *62*, 014022.
- [89] G. Dyer, S. Green, A. Fry, *Report No. SLAC-R-1152* **2021**.
- [90] M. Mo, S. Murphy, Z. Chen, P. Fossati, R. Li, Y. Wang, X. Wang, S. Glenzer, *Science Advances* **2019**, *5*, eaaw0392.
- [91] (a) L. Malerba, A. Al Mazouzi, M. Bertolus, M. Cologna, P. Efsing, A. Jianu, P. Kinnunen, K.-F. Nilsson, M. Rabung, M. Tarantino, *Energies* **2022**, *15*, 1845; (b) A. King, *Critical materials*, Elsevier, **2020**; (c) D. B. Miracle, M. Li, Z. Zhang, R. Mishra, K. M. Flores, *Annual Review of Materials Research* **2021**, *51*, 131-164.
- [92] F. Alexander, A. Almgren, J. Bell, A. Bhattacharjee, J. Chen, P. Colella, D. Daniel, J. DeSlippe, L. Diachin, E. Draeger, *Philosophical Transactions of the Royal Society A* **2020**, *378*, 20190056.
- [93] B. Shahriari, K. Swersky, Z. Wang, R. P. Adams, N. De Freitas, *Proceedings of the IEEE* **2015**, *104*, 148-175.
- [94] (a) G. Pilania, J. E. Gubernatis, T. Lookman, *Computational Materials Science* **2017**, *129*, 156-163; (b) C. Chen, Y. Zuo, W. Ye, X. Li, S. P. Ong, *Nature Computational Science* **2021**, *1*, 46-53.
- [95] J. Z. Sasiadek, *Annual Reviews in Control* **2002**, *26*, 203-228.
- [96] J. Thayer, D. Damiani, M. Dubrovin, C. Ford, W. Kroeger, C. P. O'Grady, A. Perazzo, M. Shankar, M. Weaver, C. Weninger, in *2019 IEEE/ACM 1st Annual Workshop on Large-scale Experiment-in-the-Loop Computing (XLOOP)*, IEEE, **2019**, pp. 32-37.
- [97] U. S. DOE.
- [98] E. Gibney, *Nature* **2021**, *591*, 15-16.
- [99] C. Renault, S. Delpech, **2014**.

- [100] T. Allen, J. Busby, M. Meyer, D. Petti, *Materials today* **2010**, *13*, 14-23.
- [101] O. R. N. Laboratory.
- [102] R. Averback, P. Bellon, S. J. Dillon, *Journal of Nuclear Materials* **2021**, *553*, 153015.
- [103] (a) A. Pilia, *Machine learning bandgaps of double perovskites*,” *Scientific Reports* **2016**, *6*, 19375; (b) B. Meredig, E. Antono, C. Church, M. Hutchinson, J. Ling, S. Paradiso, B. Blaiszik, I. Foster, B. Gibbons, J. Hattrick-Simpers, A. Mehta, L. Ward, *Molecular Systems Design & Engineering* **2018**, *3*, 819-825.
- [104] Y. Chen, K. Y. Yu, Y. Liu, S. Shao, H. Wang, M. Kirk, J. Wang, X. Zhang, *Nature communications* **2015**, *6*, 7036.
- [105] X.-M. Bai, A. F. Voter, R. G. Hoagland, M. Nastasi, B. P. Uberuaga, *Science* **2010**, *327*, 1631-1634.
- [106] J. Li, K. Yu, Y. Chen, M. Song, H. Wang, M. Kirk, M. Li, X. Zhang, *Nano letters* **2015**, *15*, 2922-2927.
- [107] C. Sun, D. Bufford, Y. Chen, M. Kirk, Y. Wang, M. Li, H. Wang, S. Maloy, X. Zhang, *Scientific reports* **2014**, *4*, 3737.
- [108] A. Misra, M. Demkowicz, X. Zhang, R. Hoagland, *Jom* **2007**, *59*, 62-65.
- [109] G. Odette, M. Alinger, B. Wirth, *Annu. Rev. Mater. Res.* **2008**, *38*, 471-503.
- [110] J. Du, S. Jiang, P. Cao, C. Xu, Y. Wu, H. Chen, E. Fu, Z. Lu, *Nature Materials* **2022**, 1-8.
- [111] Y. Zhang, Y. N. Osetsky, W. J. Weber, *Chemical Reviews* **2021**, *122*, 789-829.
- [112] D. B. Miracle, O. N. Senkov, *Acta Materialia* **2017**, *122*, 448-511.
- [113] F. Ren, L. Ward, T. Williams, K. J. Laws, C. Wolverton, J. Hattrick-Simpers, A. Mehta, *Science advances* **2018**, *4*, eaaq1566.
- [114] D. Morgan, G. Pilia, A. Couet, B. P. Uberuaga, C. Sun, J. Li, *Current Opinion in Solid State and Materials Science* **2022**, *26*, 100975.
- [115] N. Castin, L. Malerba, R. Chaouadi, *Journal of nuclear materials* **2011**, *408*, 30-39.
- [116] Y. Wang, B. Goh, P. Nelaturu, T. Duong, N. Hassan, R. David, M. Moorehead, S. Chaudhuri, A. Creuziger, J. Hattrick-Simpers, *Advanced Science* **2022**, *9*, 2200370.
- [117] N. Fujinuma, B. L. DeCost, J. Hattrick-Simpers, S. E. Lofland, *arXiv preprint arXiv:2112.09764* **2021**.
- [118] D. B. Laks, L. Ferreira, S. Froyen, A. Zunger, *Physical Review B* **1992**, *46*, 12587.
- [119] A. A. Kohnert, B. D. Wirth, L. Capolungo, *Computational Materials Science* **2018**, *149*, 442-459.
- [120] T. Mueller, A. Hernandez, C. Wang, *The Journal of chemical physics* **2020**, *152*, 050902.
- [121] B. DeCost, H. Joress, S. Sarker, A. Mehta, J. Hattrick-Simpers, *JOM* **2022**, *74*, 2941-2950.
- [122] (a) H. Kim, J. Inoue, T. Kasuya, *Scientific Reports* **2020**, *10*, 17835; (b) E. A. Holm, R. Cohn, N. Gao, A. R. Kitahara, T. P. Matson, B. Lei, S. R. Yarasi, *Metallurgical and Materials Transactions A* **2020**, *51*, 5985-5999; (c) J. Simmons, D. Dimiduk, M. Degraef, *Microscopy and Microanalysis* **2005**, *11*, 1634-1635.

Appendix A. Charge

PURPOSE AND OBJECTIVE(S): This roundtable, organized by the Office of Basic Energy Sciences in coordination with the Offices of Advanced Scientific Computing Research, Fusion Energy Sciences, and Nuclear Energy, will assess the status of the field and identify the fundamental science bottlenecks and gaps in the fundamental understanding that limit innovation in current nuclear technologies. The outcome of the roundtable will be identification of priority research opportunities (PROs) for fundamental science to accelerate progress in nuclear energy innovation. The aims are to define the new insights needed from basic research to enable future scientific and technological advances in materials and chemistry that are required for advanced nuclear energy systems, including both fission and fusion. The roundtable will focus on four topics that are critical to sustainable nuclear technologies—materials that can endure the harsh environments encountered in the new reactor designs as well as fusion systems, physical and chemical evolution in coolants, in situ sensing under extreme conditions, and computationally-driven discovery and accelerated data generation. A key aspect to this roundtable will be the use of artificial intelligence and machine learning to accelerate discovery. The roundtable will engage participants in discussions of technological and scientific challenges and fundamental science opportunities. Participants in the roundtable will contribute to a report that describes the scientific challenges and the PROs, which will inform future Basic Energy Sciences research directions in this important area.

The initial discussion of scientific challenges will focus on the following focus areas.

- **Panel 1: Materials Discovery and Resilience in Advanced Nuclear Systems**
Materials in nuclear systems are subjected to extreme and complex environments over their lifetime. This requires the development of specific and tailored property responses for which the underpinning science is lacking. Multi-objective design and functionality of materials for coupled extreme environments will require new scientific approaches, including multi-modal characterization of defects in materials.
- **Panel 2: Physical Processes and Chemical Evolution in Nontraditional Coolants and Solvents**
Opportunities exist to exploit the versatile coordination features of f-elements and fission products in nontraditional fuel and coolant environments to facilitate system improvements as well as chemical and physical separations. In addition, fundamental understanding of extreme fluids that act as coolants in advanced reactor concepts is critical for future innovation.
- **Panel 3: Spectroscopic Signatures and In Situ Sensing under Extreme Conditions**
It is critical to understand the changes that occur due to exposure to extreme environments over the lifetime of a reactor in order to enhance efficiency and extend lifetimes. This will require fundamental science to predict the evolving changes in the systems, but also new approaches for inducing and leveraging spectroscopic signatures in in situ, online monitoring of transient chemical speciation. The science and innovation in techniques/instrumentation for in situ, real-time sensing are central to user facilities.

The initial discussion will also consider cross-cutting opportunities that can provide synergy among these three areas.

- **Panel 4: Cross-Cutting: Computationally-Driven Discovery and Accelerated Data Generation**
Advanced machine learning and artificial intelligence can be used to enhance predictability of materials performance and evolution of chemical speciation, including automated integration of data from multiscale modeling and experiment. This could include advanced methods for

prediction and computational science for data collection and interpretation specific to extreme environments that include radiation.

Appendix B. Attendees

Roundtable Co-Chairs

Rebecca Abergel, University of California, Berkeley/Lawrence Berkeley National Laboratory
Blas Uberuaga, Los Alamos National Laboratory
Marianne Walck, Idaho National Laboratory

Plenary Speakers

Doug Kothe, Oak Ridge National Laboratory, “Opportunities for Advanced Computing in Accelerating Nuclear Energy Innovation: Reflections from CASL and Exascale”
Christine King, Idaho National Laboratory, “Commercial Perspective on Future Research Needs”

Invited Roundtable Participants

Materials Discovery and Resilience in Advanced Nuclear Systems Panel (Panel Lead: Janelle Wharry, Purdue University)

Meimei Li, Argonne National Laboratory
Jaime Marian, University of California, Los Angeles
Suveen Mathaudhu, Colorado School of Mines/Pacific Northwest National Laboratory
Elizabeth Sooby, University of Texas at San Antonio
Mark Weaver, University of Alabama

Physical Processes and Chemical Evolution in Nontraditional Coolants and Solvents Panel (Panel Lead: Jay Laverne, Notre Dame University)

Osman Anderoglu, University of New Mexico
Greg Horne, Idaho National Laboratory
Karah Knope, Georgetown University
Carolyn Pearce, Pacific Northwest National Laboratory
Richard Wilson, Argonne National Laboratory
Jim Wishart, Brookhaven National Laboratory

Spectroscopic Signatures and In Situ Sensing under Extreme Conditions Panel (Panel Lead: Arianna Gleason-Holbrook, SLAC National Accelerator Laboratory)

Ricardo Castro, University of California, Davis
Dev Chidambaram, University of Nevada, Reno
Mike Furlanetto, Los Alamos National Laboratory
Amanda Lines, Pacific Northwest National Laboratory
Joanna McFarlane, Oak Ridge National Laboratory

***Cross-Cutting: Computationally-Driven Discovery and Accelerated Data Generation
Panel (Panel Leads: Adrien Couet, University of Wisconsin–Madison; Vanda Glezakou,
Oak Ridge National Laboratory)***

Jason Hattrick-Simpers, National Resources Canada–CanmetMATERIALS

De-en Jiang, University of California, Riverside

Andrea Jokisaari, Idaho National Laboratory

Danny Perez, Los Alamos National Laboratory

Kirk Peterson, Washington State University

U.S. Department of Energy Participants

Dirk Cairns-Gallimore, NE

Jon Carmack, NE

Daniel Clark, FES

Linda Horton, BES

Stephen Kung, NE

Gail McLean, BES

Gene Nardella, FES

Daniel Nichols, NE

Andrew Schwartz, BES

John Vetrano, BES

Philip Wilk, BES

Invited Observers

Nicolas Argibay, Ames

Mark Asta, LBNL

Sage Bauers, NREL

Luke Branscum, NE

Grace Burke, ORNL

Lali Chatterjee, ASCR

Arun Devaraj, PNNL

Ahmed Diallo, ARPA-E

Leora Dresselhaus-Marais, SLAC

Chris Fecko, BES

Michael Ford, PPPL

Aura Gimm, BES

Athans Hatzikoutelis, HEP

Barbara Helland, ASCR

Scott Hsu, S4

Kevin John, LANL

Colin Judge, INL

Harriet Kung, SC

Ted Lavine, HEP

John Mandrekas, FES

Natalia Melcer, SC

Jonathan Menard, PPPL

Raul Miranda, BES

Brahim Mustapha, Argonne

Jon Neuhoff, Isotopes

Simon M. Pimblott, INL

Jennifer Roizen, BES

Tom Russell, BES

Paul Sammak, BER

Sam Wurzel, ARPA-E

Eva Zarkadoula, ORNL

Mikhail Zhernenkov, BES

Appendix C. Agenda

Virtual Meeting
Platforms: Zoom and Google Drive

July 20–22, 2022

****All times indicated are Eastern****

Day 1: Wednesday, July 20, 2022

- 10:45–11:00 AM **Log-In**
- 11:00–11:20 AM Welcome Comments
Dr. Asmeret Berhe—Director, Office of Science (10 min)
Dr. Kathryn Huff—Assistant Secretary for Nuclear Energy (10 min)
- 11:20–11:30 AM Welcome, Roundtable Charge, Goals, and Logistics (10 min)
Dr. Marianne Walck, Idaho National Laboratory
Dr. Blas Uberuaga, Los Alamos National Laboratory
Dr. Rebecca Abergel, UC Berkeley / Lawrence Berkeley National Laboratory
- 11:30–12:05 PM ***Opportunities for Advanced Computing in Accelerating Nuclear Energy Innovation: Reflections from CASL and Exascale***
Dr. Doug Kothe, Oak Ridge National Laboratory (25 min presentation + 10 min Q&A)
- 12:05–12:40 PM ***Commercial Perspective on Future Research Needs***
Ms. Christine King, Idaho National Laboratory (25 min presentation + 10 min Q&A)
- 12:40–1:05 PM Logistics and Summary of Homework Assignments (Co-Chairs, 25 min)
- 1:05–1:30 PM **Break** (25 min)
- 1:30–4:00 PM ***Breakout Session I: Science Focus***
Panel 1: *Materials Discovery and Resilience*
Panel 2: *Physical Processes and Chemical Evolution*
Panel 3: *Spectroscopic Signatures and In Situ Sensing*
Panel 4: *Cross-cutting Computational Research—panelists will directly participate and interact on other panels:*
1:30–2:00 Panelists start in cross-cutting panel for role assignment
2:00–3:30 Panelists participate in specific/assigned panels (1, 2, or 3)
3:30–4:00 Panelists return to cross-cutting panel with notes to coordinate ideas across panels
- 4:00–4:30 PM **Break** (30 min)
- 4:30–6:00 PM Plenary Session I: Panel Reports: Report out—Discussion and Identification of Potential PROs
- 6:00 PM **Adjourn**

Day 2: Thursday, July 21, 2022

- 10:45–11:00 AM **Log-In**
- 11:00–1:00 PM *Breakout Session II: Science and Gap/Synergy Crosscut Discussion*
Panelists from Panel 4 may be re-assigned for this session:
11:00–12:00 Panelists start in specific/assigned panels (1, 2, or 3)
12:00–1:00 Panelists return to cross-cutting panel
- 1:00–1:30 PM **Break** (30 min)
- 1:30–3:30 PM Plenary Session II: Panel Reports: Report out from Breakout Session II Discussion and Identification of Potential Cross-cut PROs for inclusion in report
- 3:30–4:30 PM **Break** (60 min) **** time for Chairs to collate feedback from Plenary Session II****
- 4:30–6:00 PM Plenary III: Presentation of PRO's/Closing Remarks/Adjourn
Roundtable Chairs (M. Walck, B. Uberuaga, R. Abergel)
- 6:00 PM **Adjourn**

Day 3: Friday, July 22, 2022 *Roundtable chairs, panel leads and designated writers only*****

- 10:45–11:00 AM **Log-In**
- 11:00–11:30 AM Plenary III: Introduction of writing assignments/writing groups
Roundtable Chairs (M. Walck, B. Uberuaga, R. Abergel)
- 11:30–1:00 PM **Writing/editing session 1**
- 1:00–2:00 PM **Break** (60 min)
- 2:00–6:00 PM **Writing/editing session 2**
- 6:00 PM **Adjourn**

Day 4: Friday, August 12, 2022 *Roundtable chairs, panel leads and designated writers only*****

- 8:45–9:00 AM **Log-In**
- 9:00–11:00 AM Plenary IV: Overview of writing progress and writing/editing session 3
Roundtable Chairs (M. Walck, B. Uberuaga, R. Abergel)
- 11:00 AM **Adjourn**

DISCLAIMER: This report was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government.

DOI: 10.2172/2481526



U.S. DEPARTMENT OF
ENERGY

Office of
Science