Future Nuclear Energy
Factual Status Document
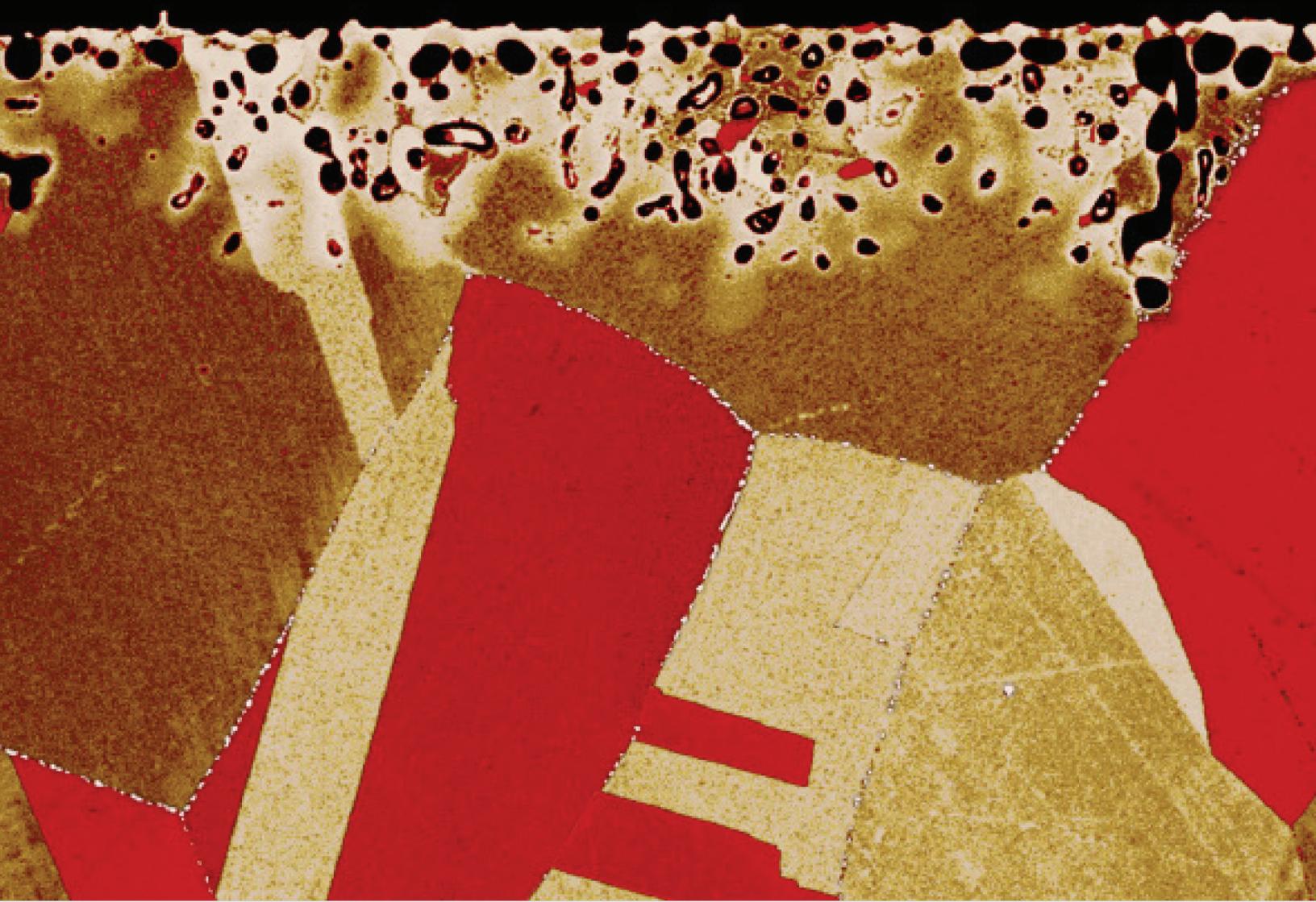
Resource Document for the Workshop on Basic Research Needs for

Future Nuclear Energy

July 2017

Technology and Applied R&D Needs for Future Nuclear Energy

**Resource Document for the Workshop on
Basic Research Needs for Future Nuclear Energy**

J. Busby, Oak Ridge National Laboratory
D. Crawford, Idaho National Laboratory
P. Demkowicz, Idaho National Laboratory
M. Farmer, Argonne National Laboratory
J. Gehin, Oak Ridge National Laboratory
S. Hayes, Idaho National Laboratory
P. Hildebrandt, Idaho National Laboratory
R. Horn, General Electric–Retired
P. Hosemann, University of California–Berkeley
J. Kacher, Georgia Institute of Technology
S. Kalinin, Oak Ridge National Laboratory
M. Li, Argonne National Laboratory
S. Maloy, Los Alamos National Laboratory
E. Marquis, University of Michigan
K. McClellan, Los Alamos National Laboratory
M. Meyer, Idaho National Laboratory
A. Nelson, Los Alamos National Laboratory
C. Parish, Oak Ridge National Laboratory
D. Petti, Idaho National Laboratory
P. Ramuhalli, Pacific Northwest National Laboratory
S. Sham, Argonne National Laboratory
B. Spencer, Idaho National Laboratory
A. Stack, Oak Ridge National Laboratory
K. Terrani, Oak Ridge National Laboratory
G. Was, University of Michigan
B. Wirth, University of Tennessee
R. Wright, Idaho National Laboratory
Y. Yang, Oak Ridge National Laboratory
G. Yoder, Oak Ridge National Laboratory
Y. Zhang, Oak Ridge National Laboratory

July 2017

Contents

FIGURES

TABLES

ACRONYMS AND ABBREVIATIONS

2D	two dimensional
3D	three dimensional
4D	four-dimensional
AGR	Advanced Gas-Cooled Reactor
APT	atom probe tomography
ATR	Advanced Test Reactor
BWR	boiling water reactor
CANDUs	Canada Deuterium Uranium Reactor
CHX	compact heat exchanger
Cr	chromium
DIC	Digital Image Correlation
DOE	Department of Energy
dpa	displacements per atom
EELS	electron-energy-loss spectrometer
FCCI	fuel-cladding chemical interaction
FCRD	Fuel Cycle Research and Development
FFTF	Fast Flux Test Facility
FIB	focused ion beam
GFR	Gas-Cooled Fast Reactor
GIF	Generation IV International Forum
GPU	graphics processing units
GWe	gigawatt electric
HBS	high burnup structure
HEDM	high-energy diffraction microscopy
HFIR	High Flux Isotope Reactor
HPC	high performance computing
HRP	Halden Reactor Project
HTGR	high-temperature gas-cooled reactor
IAC	irradiation accelerated corrosion
IASCC	irradiation assisted stress corrosion cracking
JHR	Jules Horowitz Reactor
JNT	Johnson Noise Thermometry
LAMDA	Low Activation Materials Development and Analysis
LWR	light water reactor
MOX	mixed oxide
MSBR	Molten Salt Breeder Reactor

MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
MWe	megawatt electric
NDE	non-destructive evaluation
NE	Office of Nuclear Energy
NEUP	Nuclear Energy University Programs
ODS	oxide dispersion steels
ORNL	Oak Ridge National Laboratory
PWR	pressurized water reactor
R&D	research and development
RPV	reactor pressure vessel
SCC	stress corrosion cracking
SCWR	supercritical-water-cooled reactor
SDD	silicon drift detectors
SFR	Sodium-Cooled Fast Reactor
SNS	Spallation Neutron Source
TEM	transmission electron microscopy
TREAT	Transient Reactor Test Facility
TRISO	tristructural-isotropic
TRL	technical readiness level
VHTR	Very High Temperature Reactor
VVER	Vodo-Vodyanoi Energetichesky Reactor
Zr	zirconium

EXECUTIVE SUMMARY

Since commercialization in the mid-1950s, nuclear power has been widely deployed and provides an important clean source of energy, primarily through the generation of electricity. There are currently 449 commercial nuclear power reactors operating in 31 countries with over 390 GWe total capacity, with 99 operating units in the United States with a total generating capacity of about 100 GWe. Nuclear power plants provide over 11% of the world's electricity production and, as of 2016, thirteen countries rely on nuclear energy to supply at least one-quarter of their total electricity. Virtually all of that power is generated with water-cooled reactor systems. A number of advanced nuclear energy system concepts have been developed to provide improved efficiency, greater fissile fuel utilization, reduced high level waste generation, and increased margins of safety over today's water-cooled systems. However, there are a number of technical challenges that must be addressed to enable the broad deployment of these systems from concept to the electrical grid.

The development and deployment of advanced reactors requires research and development of materials and understanding the chemistry of coolants, fuels, and interactions. Common areas across the six advanced designs discussed include the development and qualification of advanced fuel forms, high-temperature radiation-tolerant materials, a range of coolant interactions with materials with an emphasis on corrosion, and the development of materials for high-temperature, coolant-compatible instrumentation. In addition, given the desire for commercial deployment starting in 2030, approaches that can accelerate the development and qualification of materials are needed.

The DOE's Offices of Basic Energy Sciences and Office of Nuclear Energy will convene a workshop in August 2017 to engage the scientific community in defining research opportunities that can support the development of next-generation nuclear systems. At this workshop, scientists from the national laboratories, academia, industry, and other organizations will discuss gaps in scientific knowledge that, if filled, could significantly advance deployment of technologies for advanced nuclear systems.

This Technical Document is written to provide workshop participants with an overview of the current status of a number of advanced reactor systems. The focus of this document includes the following technical areas:

- Key issues for advanced nuclear energy, including coolants, materials in extremes (irradiation, coolant, and/or high temperature) and interfaces;
- Nuclear fuel technologies for key reactor types and limitations in testing and design; and
- State of science and understanding for nuclear systems, including characterization, testing, and modeling and simulation tools.

For each of these technical areas, this document describes the technological challenge, current state of the art for the technology, and barriers to advanced nuclear concepts.

NUCLEAR POWER BACKGROUND AND OVERVIEW

Nuclear power has been widely developed and deployed and provides an important clean source of energy, primarily through the generation of electricity. There are currently 449 commercial nuclear power reactors operating in 31 countries with over 390 GWe total capacity, with 99 operating units in the United States with a total generating capacity of about 100 GWe. Nuclear power plants provide over 11% of the world's electricity production and, as of 2016, thirteen countries rely on nuclear energy to supply at least one-quarter of their total electricity. The largest fraction of electricity generation by nuclear power is in France, with nuclear energy providing over 70% of the country's total electricity. [1]

Globally, 60 reactors (approximately 60 GWe total capacity) are under construction, 65% of which are under contract to organizations in Russia and China. The reactors under construction are primarily light water reactors (LWRs), with a few advanced reactor concepts. Of these ongoing activities, four LWR units are under construction in the U.S. (two at Vogtle Electric Generating Plant and two at Virgil C. Summer Nuclear Generating Station). [1]

This section provides an overview of nuclear power reactors including a discussion of the development of currently deployed systems, an overview of reactor technology, and a description of the primary advanced reactor concepts.

U.S. AND INTERNATIONAL STATUS OF NUCLEAR ENERGY

The development of nuclear reactor technology has been characterized in terms of "generations" by the Generation IV International Forum. [2] These generations, summarized in Figure 1, range from the very first reactors, known has Generation I, to the currently deployed systems, predominately Generation II, III, and III+, to the future advanced reactors in Generation IV.
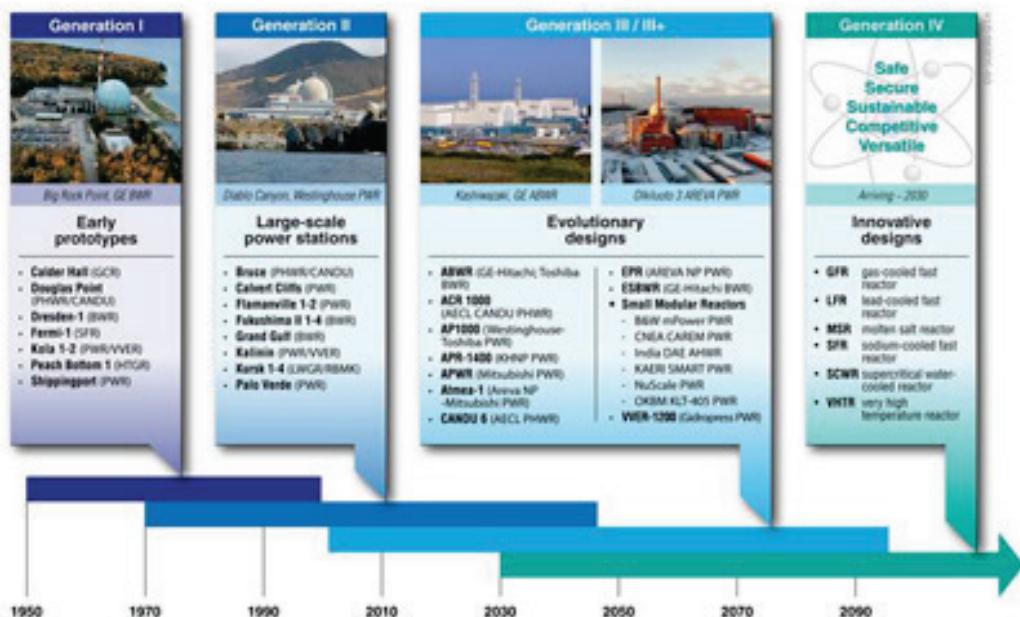
Figure 1.Generation I-IV reactor technology development history. Source: Generation IV
International Forum, www.gen-4.org.

Peaceful and commercial use of nuclear fission began in the 1950s in the U.S. with the adaptation and evolution of LWR technology from the Naval Nuclear Propulsion Program, with the early prototypes including Shippingport pressurized water reactor (PWR) in Pennsylvania, Dresden Unit 1 boiling water reactor (BWR) in Illinois, and the graphite moderated, gas-cooled (carbon dioxide) Magnox concept that was developed in the United Kingdom and initially prototyped at Calder Hall in Wales. These Generation I reactors have all been decommissioned.

Most of the current fleet consists of Generation II and III reactors including PWRs, BWRs, CANada Deuterium Uranium reactors (CANDUs), advanced gas-cooled reactors (AGR), and Vodo-Vodyanoi Energetichesky PWR-type reactors (VVERs). These reactors were developed, designed, and built by several suppliers including Westinghouse, General Electric, B&W, Combustion Engineering (now Westinghouse) [all U.S.], Framatome (now part of AREVA) [France], and Rosatom [Russia]. Variations of these designs have been adapted by Japan, South Korea, and China for their nuclear fleets.

The Generation III+ evolutionary designs include large reactors such as Westinghouse's AP-1000 [3], General Electric-Hitachi's Economic Simplified Boiling Water Reactor [4] and AREVA's EPR [5] as well as several small modular reactor concepts, including the NuScale [6] design. The Generation IV and other advanced nuclear energy system concepts offer the promise of superior economics, improved efficiency, greater fissile fuel utilization, reduced high-level waste generation, and increased margins of safety. [2]

The U.S. took the lead on a version of the very high temperature, gas-cooled reactor system through the Next Generation Nuclear Plant program [7]. That program, now continuing as the very high temperature reactor (VHTR) program, has made important progress in the development and qualification of tristructural-isotropic (TRISO) ceramic coated fuel, characterization of graphite used for structural and moderator purposes, codification of high temperature metals, and the foundational steps in developing a modernized regulatory framework that can be used as the basis for licensing advanced nuclear energy systems (technology inclusive, risk informed, and performance based).

There are a considerable number of U.S.-based and international programs and companies evaluating and in various stages of developing advanced nuclear energy systems ranging from evolutionary LWR designs to variations on the Generation IV concepts. In the U.S. alone, there are several dozen startup ventures that have attracted considerable early investment. It is expected that there will be a natural down selection to the most promising concepts as the realities of overcoming technology hurdles, finding the large investment required for commercialization, and the interests of the future owners are better understood. In the U.S., the potential future owner/operators and industrial energy end-users are only beginning to indicate future interests in deployment of advanced nuclear energy systems – while still working to complete construction of the four AP-1000 units at Vogtle Electric Generating Plant in Georgia, USA and at Virgil C. Summer Nuclear Generating Station in South Carolina, USA.

REACTOR TECHNOLOGY OVERVIEW

A high-level understanding of the design and operation of nuclear reactors can be gained by examining the plant layout and conditions for a typical PWR. Figure 2 depicts a notional configuration for a generic LWR/PWR. PWRs of this general configuration typically operate at reactor outlet temperatures of about 300°C with plant capacity ratings ranging from 400 to 1700 MWe. There are typically multiple loops (two to four) with multiple steam generators (two to four total) that reduce equipment size and provide redundancy. The reactor is cooled during operation by the reactor coolant system via the steam generators with one or more heat sinks. The reactor coolant system is subcooled by the overpressure provided by the pressurizer, which also provides a surge volume for transients. The fuel consists of uranium dioxide

pellets clad in zirconium alloy tube and arranged in fuel assemblies to form the active core region, supported by various structural components and systems. The core is contained in a steel pressure vessel.

The Rankine cycle power conversion system typically includes a single large turbine with high and low-pressure stages driving a single electric generator connected to the grid. The low-pressure turbines exhaust to a condenser maintained at a partial vacuum and cooled in an open cycle by a large body of water (e.g., lake, river, ocean) or by a closed cycle cooling tower system. The efficiency of the Rankine cycle is improved through use of reheat cycles including a moisture separator reheater and several stages of feedwater heating (not shown).
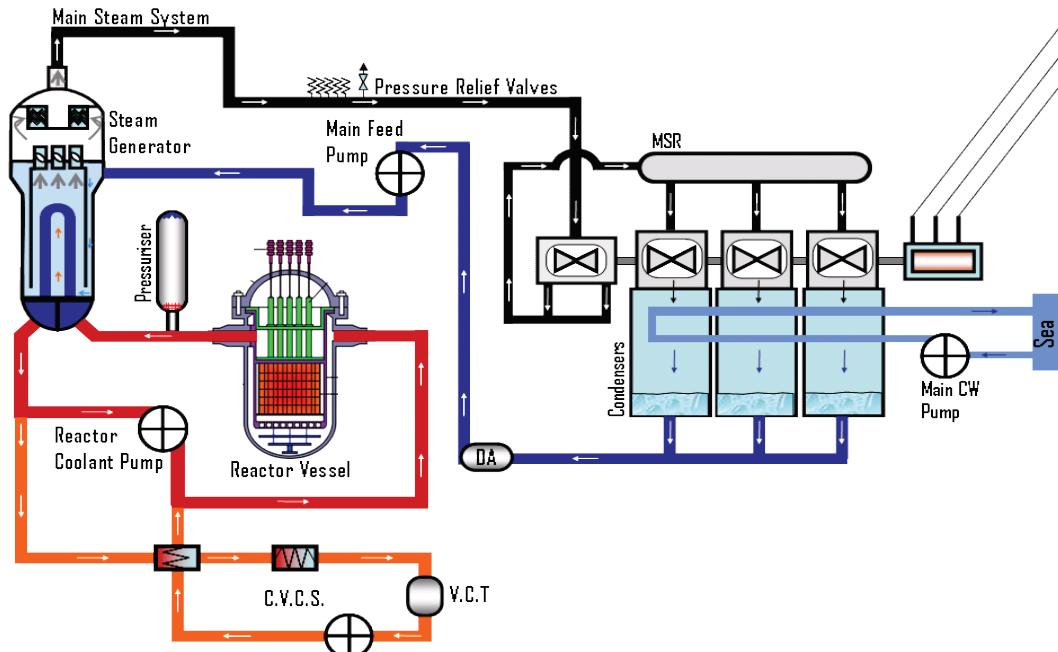
Figure 2. Diagram of a pressurized water reactor (PWR). Source: Nuclear Energy Technology – Knowledge Transfer Workshop, Idaho National Laboratory, Nov. 15-17, 2016.

The safety case for the LWR is based on analyses of the postulated and known hazards and control of three essential safety functions:

- Controlling reactivity of the core (via the control rods and chemical shim [soluble boron burnable neutron poison])

- Controlling cooling of the core (via the reactor coolant system, decay/residual heat removal system, and emergency core cooling systems)

- Controlling reactivity release (via the barriers of the fuel cladding, reactor coolant system, and the containment)

- A typical emergency core cooling system configuration is shown in Figure 3 and includes high, intermediate, and low-pressure injection systems, backup sources of cooling water (accumulators and refueling water tank), and a heat transfer path to the ultimate heat sink via the decay/residual heat exchanger. These systems respond to and mitigate the effects of loss of coolant and loss of heat sink (cooling) accident scenarios

In Figure 3, the containment building is notionally depicted by the shaded area. Containment is also provided within systems that cross the containment building boundary. Multiple safety support (e.g.,

containment cooling) and control systems (e.g., engineered safety features actuation system) are not shown. The specific hazards, the method of analyses, design basis accident scenarios, and evaluation rules are part of the design, analyses, and regulatory process used as the basis for a license application.
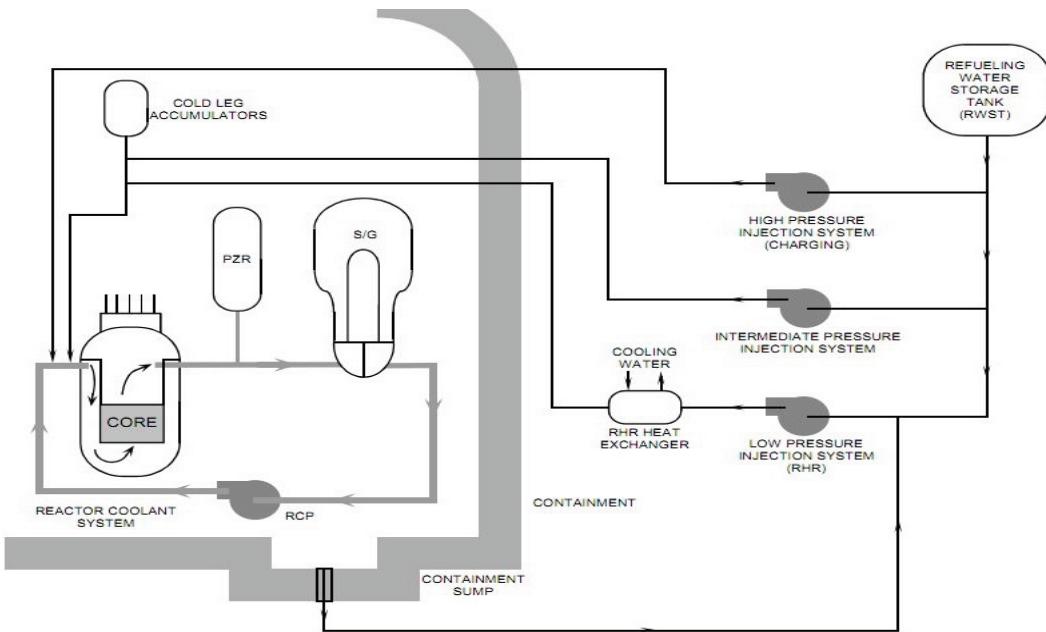
Figure 3. Pressurized water reactor (PWR) high-level layout. Source: Nuclear Energy Technology – Knowledge Transfer Workshop, Idaho National Laboratory, Nov. 15-17, 2016.

As noted above, the vast majority of nuclear power generated in the world today utilizes water reactor technology. These designs are well established and have a long history of performance. However, light water reactor technologies do have limitations, including thermal efficiency, limited upper operating range, and generation of used nuclear fuel. Advanced reactor designs are required to move past these limitations to ensure efficient nuclear power for the future, including applications beyond electricity generation. For example, the higher operating temperature of gas reactors may allow the use of process heat for other missions such as desalination or hydrogen production, which is simply not possible with water reactor technology. Other designs may enable to transmutation or "burning" of used nuclear fuel from the water reactor fleet. Many of the design and safety considerations listed above for water reactors are also a basis for these advanced designs. However, in order to be a viable and economical form of energy, each design has challenges such as overcoming the increasingly harsh environment, identifying optimal fuel forms, and making the design affordable. Each of the key Gen IV advanced reactor concepts is described in more detail below.

OVERVIEW OF GENERATION IV ADVANCED REACTOR CONCEPTS

Over the history of nuclear reactor development, many potential concepts and designs have been developed. To focus on the most promising the designs, the Generation IV International Forum (GIF) has selected six concepts for future research and development. [2] These concepts include:

- Molten salt-cooled reactor system
- Very high temperature gas-cooled reactor system

- Gas-cooled fast reactor system
- Sodium-cooled fast reactor system
- Lead-cooled fast reactor system
- Supercritical water-cooled reactor system

Nearly all of these concepts have previous technology development and some are envisioned for commercial deployment beginning in 2030. Details on the progress in the development of these reactor designs and research and development (R&D) needs can be found in the *Technology Roadmap Update for Generation IV Nuclear Energy Systems* [2] and the associated references in that document. Information on specific designs can be found in the International Atomic Energy Agency *Advanced Reactors Information System [8]*. A brief overview of the Generation IV advanced reactor concepts is provided below, along with a high level summary of technical needs. More detailed discussion of technical needs for materials and fuels are outlined in the sections on key limitations and nuclear fuels, below.

Molten Salt Reactor

Molten salt reactors (MSRs) represent a class of reactors that involve the use of a salt in liquid state (either a fluoride salt or a chloride salt) either as a coolant with a solid fuel (as in fluoride salt–cooled high-temperature reactors) or with a fuel dissolved in liquid salt that also serves as the coolant material. In dissolved-fuel MSRs, the salt can be processed additionally, either online or in a batch mode, to allow for the removal of fission products and for the introduction of fissile fuel and fertile materials during reactor operation. MSR concepts have been developed with both thermal and fast neutron spectra and with uranium, thorium, and plutonium fuels. Figure 4 provides a diagram of the reactor concept showing the reactor core, intermediate and secondary loops, chemical processing system, and power conversion system.
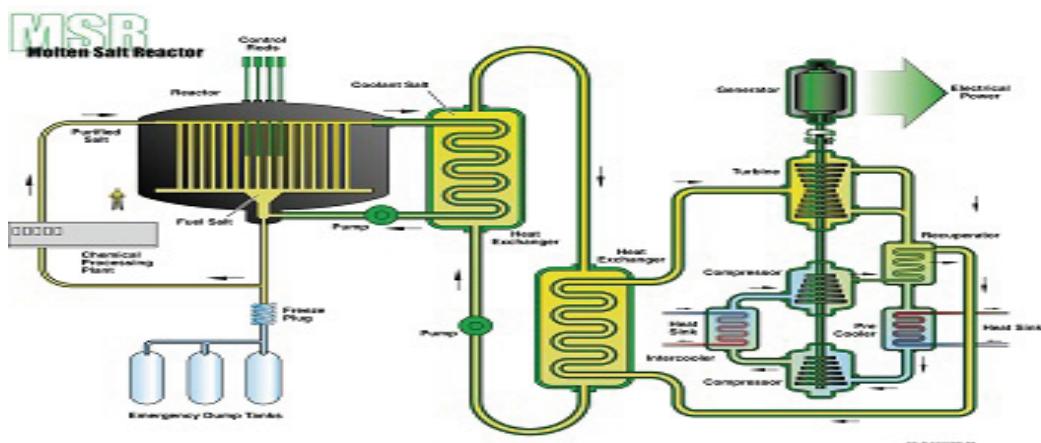
Figure 4. Concept diagram for a liquid-fueled molten salt reactor.
Gen-IV International Forum, https://www.gen-4.org.

Historically, the MSR is most commonly associated with the U-233/Thorium fuel cycle as the nuclear properties of U-233 combined with the online removal of parasitic absorbers results in the ability to design a thermal-spectrum breeder reactor. An extensive program supporting research and development of a thermal-spectrum Molten Salt Breeder Reactor (MSBR) at Oak Ridge National Laboratory (ORNL) in the 1950s – 1970s resulted in reactor designs and a significant amount of technology development in materials, salt technology, and reactor components. This research program included the operation of the Molten Salt Reactor Experiment (MSRE) in the 1960s, providing operating data. Today, several companies are developing this concept including Terrestrial Energy, Transatomic Power, FLIBE Energy,

Terrapower, and Kairos Power. Research is also being performed internationally in Europe, Russia, China, and India.

While there are several design variations, a general summary of the key materials representing the fuel, coolant, moderator, and structures along with operating conditions are provided in Table 1.

Table 1. General materials and operating conditions for Molten Salt Reactor concepts

Parameter	Description
Coolant	Fluoride or chloride salts, most commonly FLiBe, though several different salts are currently being considered. Liquid fuel concepts have coolant containing actinides and fission products, solid fuel concepts have relatively clean salt.
Fuel	Liquid fuel concepts: Uranium, Plutonium thorium dissolved in fluoride or chloride salts.
	Solid fuel concepts: Low enriched uranium in the form of TRISO coated particle fuel in a graphite matrix in pebble or compact form.
Moderator	For thermal spectrum systems, graphite and zirconium hydride. None for fast spectrum systems.
Structural materials	In-core generally ceramic materials including graphite, carbon/carbon and SiC/SiC composites.
	Vessel and piping generally high nickel, low-chrome alloys, lower-cost steel alloys being considered in some designs.
Neutron Spectrum	Thermal and fast
Outlet Temperature	Typically, ~700°C
Operating Pressure	Near atmospheric
Fuel Cycle	Once-through and closed

There are broad development needs to advance MSR concepts to the deployment stage related to materials and chemistry, including

- Development of radiation tolerant, high-temperature materials that are compatible with fluoride and chloride salts, actinides, and fission products for use as moderator (low atomic number), such as graphite and zirconium hydride, and structural components, including ceramics and metals.
- Understanding of liquid salt chemistry and properties, control of salt chemistry during operations, and compatibility of the salts with fuel and structural materials.
- Management and control of tritium produced in the salt from fission and neutron capture to avoid migration and release.
- Development and qualification of TRISO fuels for solid-fuel concepts and qualification of structural materials to support reactor design and licensing.
- Chemical processing and materials recovery from liquid salts for salt purification in solid-fuel concepts and closed fuel cycle applications in liquid fuel concepts.
- Sensors and instrumentation to monitor salt conditions.

Defining the coolant, coolant chemistry, and coolant purity levels are essential to make progress in the broad needs defined above. Development, testing, and qualification of materials (structures, clad, and/or fuels) under prototypic environments all require established coolant conditions.

Very-High-Temperature Reactor

The Very-High-Temperature Reactor (VHTR) uses a gas coolant and fuels and structural materials that can be used under high temperature conditions (>750°C) to provide high efficiency electricity generation or high-temperature heat to be used for industrial processes. The fuel consists of TRISO fuel in graphite that is formed into either pebbles or compacts in graphite blocks. This fuel, along with the reactor design, provides a high degree of safety under a wide range of accident conditions. The current reactor designs will operate with a thermal neutron spectrum with enriched uranium fuel, but concepts are being investigated that could use plutonium and/or thorium fuels (Figure 5).
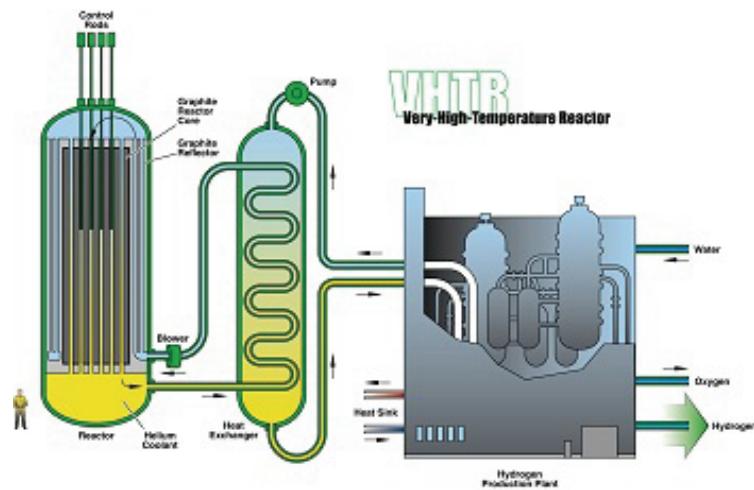
Figure 5. Concept diagram for the Very High Temperature, a Gas-Cooled High-Temperature Reactor. Gen-IV International Forum, https://www.gen-4.org.

Historically, gas-cooled reactors based on this technology have been under development since the 1950s with several operating reactors developed in Germany (AVR and THTR), the US (Peach Bottom II, Fort St. Vrain), Japan (HTTR), and China (HTR-10). Modern concepts have been developed including the GT-MHR, NGNP, PBMR, and the HTR-PM, currently under construction in China. In the U.S designs, a prismatic block core design is being developed by AREVA and pebble-bed design is being developed by X-energy. The goals of the Generation IV VHTR activity are to significantly increase the reactor outlet temperature to support a wider range of applications.

Key material systems and operating conditions typical of VHTR designs are summarized in Table 2.

Table 2. General materials and operating conditions for very-high-temperature reactor concepts

Parameter	Description
Coolant	Helium
Fuel	Low enriched uranium (UO_2, UCO) in TRISO fuel in graphite matrix
Moderator	Graphite
Structural materials	Graphite, ceramic materials in core, metal vessel and piping
Neutron Spectrum	Thermal
Outlet Temperature	>750°C
Operating Pressure	7 MPa
Fuel Cycle	Once-through

The key materials development needs for VHTRs include the following items focusing on technology development to support deployment and extending the reactor operating temperature:

- Qualifications including irradiation testing and examination of TRISO fuel for high burnup, high temperature conditions including normal operations, and accident conditions exceeding 1600°C.
- Improvements in TRISO fuel concepts to support very high temperature concepts, including alternate coating materials, such as ZrC, rather than the standard SiC fission product barrier.
- Radiation-tolerant, high-temperature materials, such as C/C and SiC/SiC composites, for control rod sheaths for higher outlet temperatures.
- Qualification of pressure vessel and piping materials for higher temperature operation, including extending Alloy 800H and Alloy 617.
- Irradiation testing and examination of current grades of nuclear graphite.
- Fuel cycle needs including alternative fuels including Pu and Minor Actinides.
- High-temperature, radiation tolerant sensors and instrumentation.

High-temperature gas-cooled reactors have had considerable study and, as noted above, concepts have been developed and operated in several countries. Resolving these material needs provides an opportunity to extend operation to higher temperatures and optimize performance, making the technology more flexible and attractive.

Gas-Cooled Fast Reactor

The Gas-Cooled Fast Reactor (GFR) is a reactor concept that the uses a gas coolant to enable a high outlet temperature to meet the same objectives as high-temperature gas-cooled reactors (high efficiency electricity production and industry process heat applications). However, the GFR design has a fast neutron spectrum that provides the ability to fuel cycle objectives including a higher utilization of uranium through improved conversion of fertile U^{238} to fissile Pu^{239} and reduction in waste through fuel recycle and reuse. The low-moderating, single phase gas coolant results in a low void reactivity in comparison to other fast reactor concepts, but offers challenges in natural circulation coolant (Figure 6).
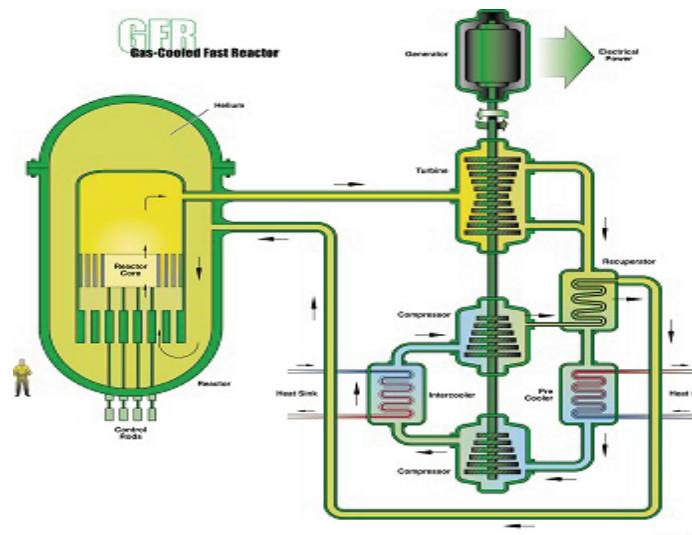
Figure 6. *Concept diagram for the Gas-Cooled Fast Reactor. Gen-IV International Forum, https://www.gen-4.org.*

The GFR has historically been investigated but an operating GFR has never been built. Concepts for a GFR are being developed in the U.S. by General Atomics with the EM2 concept and an experimental demonstration reactor, ALLEGRO, is being developed in Europe. Given the lack of demonstrations, the GFR is at an early stage of development in comparison to other advanced reactor concepts.

Key material systems and operating conditions typical of GFR designs are summarized in Table 3.

Table 3. General materials and operating conditions for gas-cooled fast reactor concepts

Parameter	Description
Coolant	Inert gas, typically helium
Fuel	UC, PuC in SiC matrix or cladding
Moderator	None
Structural materials	Ceramic materials (SiC, ZrC, Zr_3Si_2)
Neutron Spectrum	Fast
Outlet Temperature	850°C
Operating Pressure	7 MPa
Fuel Cycle	Once-through and closed

The primary materials and chemistry development needs for the GFR are as follows:

- Development of fuel system to meet high temperature conditions in normal operations and accident conditions, generally UC, PuC with SiC and ZrC matrix, and/or cladding. Including fuel testing and qualification and manufacturing.
- Development of high-temperature, radiation tolerant structural materials for core components and high-temperature materials for the reactor vessel and system piping.
- Development of high-temperature instrumentation for GFR conditions for monitoring core and structure temperatures.

As described above, this reactor type has been explored previously, but has not been built. This concept is at an earlier stage of technical readiness and development of high temperature materials is essential to move beyond the concept stage.

Sodium-Cooled Fast Reactor

The Sodium-Cooled Fast Reactor (SFR) is one of the most developed of advanced reactor concepts, characterized by the use of liquid sodium coolant whereby providing a system with low pressure and a high-heat removal capability and resulting in high power densities. The reactor is designed to operate with a fast neutron spectrum providing high uranium resource utilization and waste minimization. The reactor operates at a higher temperature than LWRs with an outlet temperature in excess of 500°C, but not as high as other advanced reactor concepts. The continued development of SFRs are focused on the development and use of advance fuels, including those for transmuting actinides, and reduction in costs to enable commercial deployment (Figure 7).
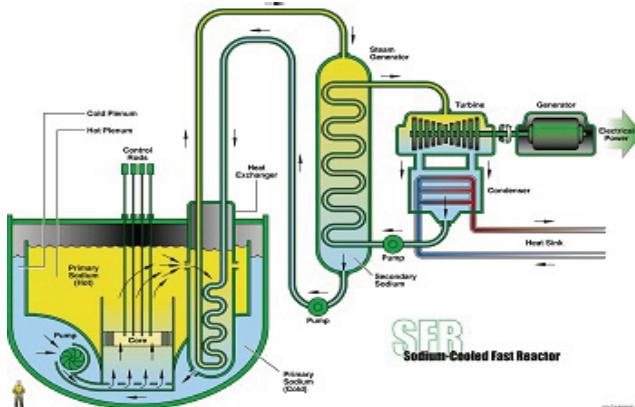
Figure 7. *Concept diagram for the Sodium-Cooled Fast Reactor. Gen-IV International Forum, https://www.gen-4.org.*

The SFR has undergone significant development with several experimental and commercial reactors being developed and operated. The U.S. operated several test reactors, including EBR-I, EBR-II, FFTF and a commercial reactor, Fermi 1, and supported the development of commercial reactors, including the Clinch River Breeder Reactor and the General Electric PRISM design. Other experimental, test, and commercial reactors have been developed and operated world-wide, particularly in France, Russia, and Japan, though there are active programs also in China, India, and South Korea. In the U.S. currently there are several concepts being developed including the General Electric PRISM design, Terrapower traveling wave reactor, the Advanced Reactor Concepts ARC-100, and by Oklo. The Russian Federation is currently operating two SFR power reactors, the BN-600 and BN-800.

Key material systems and operating conditions typical of SFR designs are summarized in Table 4.

Table 4. General materials and operating conditions for sodium-cooled fast reactor concepts

Parameter	Description
Coolant	Sodium
Fuel	Metallic, U-Zr, U-Pu-Zr and oxide, UO_2, UO_2-PuO_2
Moderator	None
Structural materials	Metallic (steel alloys)
Neutron Spectrum	Fast
Outlet Temperature	500-550°C
Operating Pressure	Near atmospheric
Fuel Cycle	Closed

Identified development needs for materials and chemistry include:

- Development of radiation-tolerant fuels and fuel cladding material to support high-burnup applications extending well beyond historical experience (>200 dpa).
- Development of advanced fuels for transmutation applications including those containing minor actinides, including the development of manufacturing processes.
- Development of advanced measurement systems and instrumentation to include under-sodium viewing and monitoring of core and structural temperatures.

Sodium-cooled systems are well-established reactors, with several sodium reactors currently operating around the world. Additional materials development is not required for the viability of this design, but will support improved safety, efficiency, and economics.

Lead-Cooled Fast Reactor

The Lead-Cooled Fast Reactor (LFR) uses lead or lead-bismuth eutectic as a coolant to provide a low neutron absorbing, high-temperature coolant with a fast spectrum reactor. The high temperature operation can support high electrical efficiency and industry heat applications, while the fast neutron spectrum supports sustainable fuel cycle objectives. Molten lead, as a coolant, has a very high boiling point (1749°C) and does not react significantly with air or water providing potential benefits in terms of safety and design, including the elimination of intermediate loops required in other fast reactor concepts. Lead bismuth eutectic has a lower melting point than lead (124°C vs 327°C), potentially leading to a simplified design and operations (Figure 8).
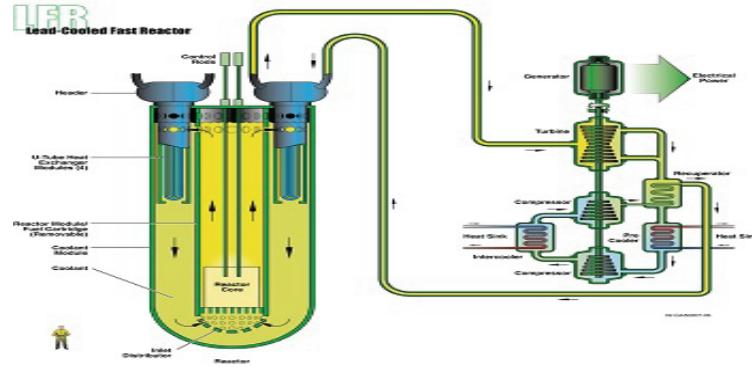
Figure 8. Concept diagram for the Lead-Cooled Fast Reactor. Gen-IV International Forum, https://www.gen-4.org.

LFRs have seen limited actual use, historically in Russian nuclear power submarines. However, several concepts are under development in Russia (BREST, SVBR), the U.S. (Westinghouse), and Europe (ELFR and MYRRAH).

Key material systems and operating conditions typical of LFR designs are summarized in Table 5.

Table 5. General materials and operating conditions for lead-cooled fast reactor concepts.

Parameter	Description
Coolant	Lead or Lead-Bismuth Eutectic
Fuel	UO_2, UO_2-PuO_2 UN, UN-PuN
Moderator	None
Structural materials	Metallic (steel alloys)
Neutron Spectrum	Fast
Outlet Temperature	480-550°C
Operating Pressure	Near atmospheric
Fuel Cycle	Closed

The key materials and chemistry development needs for LFRs include:

- Development and qualification of radiation-tolerant, high temperature structural materials with lead, lead-bismuth salt compatibility.
- Corrosion of materials, particularly metallic cladding and structural materials through oxidation control and/or protective layers/surface treatments.
- Fuels development and testing for use of oxide fuels for near-term lower temperature designs and nitrides for longer-term, higher temperatures (at both normal and accident conditions).

- For fuel cycle applications, development of nitride fuel reprocessing and minor-actinide fuels for transmutation.
- Lead, lead-bismuth chemistry and purification including the understanding of fission product retention for accident source terms.
- High-temperature instrumentation for core, coolant, and structural material monitoring.
- Methods for viewing through lead, lead-bismuth to support operations and maintenance.

Lead-cooled systems have many of the same challenges associated with liquid metal environments as sodium reactors, however there is far less operating experience than with sodium. Material development and degradation understanding will be required to advance this technology beyond the concept stage.

Supercritical-Water-Cooled Reactor

Supercritical-Water-Cooled Reactors (SCWR) use light water as the reactor coolant, but unlike BWRs and PWRs, operate above the critical point of water (374°C, 22.1 MPa). This allows for a higher outlet temperature that increases the plant's thermodynamic efficiency over that of currently operating light water reactors. The reactor can be designed to operate with either thermal for fast neutron spectrum, depending on the type of fuel used (Figure 9). The use of super-critical water potentially offers design simplifications by removing reactor coolant pumps, while the direct use of the outlet superheated steam in the power conversion system eliminates the need for steam generators and steam dryer/separators. The development of SCWRs can leverage experience with existing LWRs, particularly BWRs, as well as supercritical water fossil-fired plants.

Historically, the direct generation of superheated steam was seen as an evolution of the boiling water reactor, which included the construction and limited operation of the Boiling Nuclear Superheater Reactor
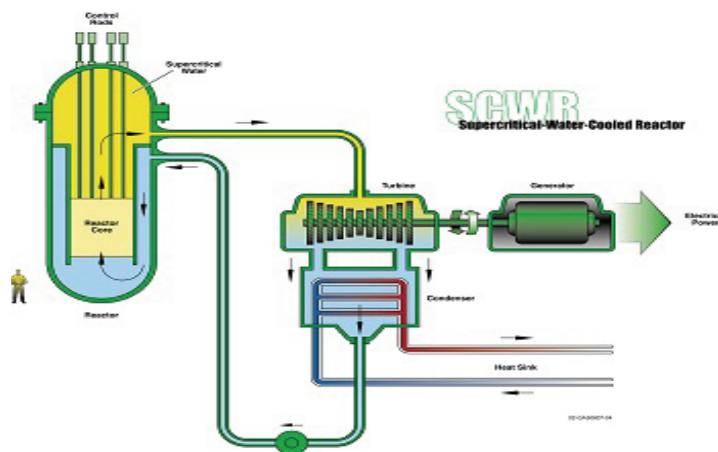
Figure 9. Concept diagram for the Supercritical-Water-Cooled Reactor. Gen-IV International Forum, https://www.gen-4.org.

in Puerto Rico in the 1960s, but not as supercritical water conditions. Design concepts based on pressure-vessel and pressure tube designs are being considered. A pressure-vessel type reactor with a 500°C core outlet temperature has been developed in Japan and a pressure tube SCWR is being developed in Canada with a 625°C outlet temperature. Additional concepts are being developed in Russia and China.

Key material systems and operating conditions typical of SCWR designs are summarized in Table 6.

Table 6. General materials and operating conditions for super-critical water-cooled reactor concepts

Parameter	Description
Coolant	Supercritical light water (>374°C, >22.1 MPa)
Fuel	UO_2, UO_2-PuO_2
Moderator	Light water
Structural materials	Metallic (steel alloys)
Neutron Spectrum	Thermal or Fast
Outlet Temperature	500 - 625°C
Operating Pressure	25MPa
Fuel Cycle	Open/closed

The key materials and chemistry development needs for SCWRs include:

- Development and qualification of alloys for cladding structures in SCWR operating conditions including irradiation, pressure, temperature, water radiolysis, corrosion, and corrosion product deposition.
- For pressure tube designs, long-life pressure tube materials to prevent the need for replacement over the life of the plant.
- Understanding of water chemistry in supercritical conditions corrosion product and fission product transport.
- In core instrumentation for temperature, pressure, and flux/power measurements.

Supercritical systems offer a breakthrough in performance and thermal efficiency. However, solving material performance limitations is essential before SCWR's can move beyond construction of individual components.

The development and deployment of advanced reactors share many similar needs, requiring further development of materials and understanding the chemistry of coolants, fuels, and interactions. Common areas across the six advanced designs discussed include the development and qualification of advanced fuel forms, high-temperature radiation-tolerant materials, a range of coolant interactions with materials with an emphasis on corrosion, and the development of materials for high-temperature, coolant-compatible instrumentation. These technical challenges are outlined in further detail in the next section.

Summary of Key Barriers for Advanced Nuclear Energy Systems

As noted above, each of the advanced reactor concepts offers several advantages over water reactor technologies, including greater thermal efficiency, passive safety systems, or enabling new missions such as process heat or waste transmutation. However, each design also has barriers to development and the ultimate deployment of the technology. There are several common areas for key materials that must be overcome to enable advanced nuclear energy systems. Coolant compatibility is a key limitation for many advanced concepts, although the specifics vary by coolant and material choices. The impacts of high temperature, mechanical stress, and irradiation environments also pose a key challenge to deployment. The mechanistic understanding of these individual extreme elements and combined environment is needed. The mechanisms for interfacial processes are essential for prediction of degradation and design of tolerant materials.

Similar limitations for nuclear fuels exist. There are key degradation issues that must be resolved for both thermal and fast spectrum reactor fuels. Fuel forms for gas and molten salt cooled reactors have unique issues due to the very high operating temperatures and coolant interactions that must be resolved. Interfacial understanding is essential to predict and mitigate key interactions (e.g. clad and coolant; fuel and coolant; and/or fuel and clad).

References

1. Nuclear Energy Institute, https://www.nei.org/Knowledge-Center/Nuclear-Statistics/World-Statistics, accessed July 17, 2017.
2. Generation IV International Forum, *Technology Roadmap Update for Generation IV Nuclear Energy Systems*, January 2014.
3. Westinghouse Electric Company, AP*1000 Pressurized Water Reactor*, https:// http://www.westinghousenuclear.com/New-Plants/AP1000-PWR, accessed July 17, 2017
4. General Electric Company*, Economic Simplified Boiling Water Reactor,* https://nuclear.gepower.com/build-a-plant/products/nuclear-power-plants-overview/esbwr.html, accessed July 17, 2017.
5. Areva, *EPR™ Reactor*, http://www.areva.com/EN/global-offer-419/epr-reactor-one-of-the-most-powerful-in-the-world.html, accessed July 17, 2017.
6. NuScale Power, *Nuscale Power – Our Technology*, http://www.nuscalepower.com/our-technology, access July 17, 2017.
7. Idaho National Laboratory, NGNP Public Document Archive, https://art.inl.gov/NGNP, accessed July 17, 2017.
8. International Atomic Energy Agency, *Advanced Reactors Information System (ARIS),* https://aris.iaea.org.

STATE OF THE ART AND KEY ISSUES FOR ADVANCED NUCLEAR ENERGY

Advanced reactor concepts have many advantages over modern light water reactor technology as described in the previous section. However, these designs have not yet been deployed at the same scale as water reactors and there are technical gaps, which must still be resolved. In this section, key bottlenecks and challenges that are limiting deployment are discussed. Some of these bottlenecks and challenges cut across multiple designs. Specific areas discussed below include coolants and materials.

COOLANTS

Nuclear reactor materials must survive in a unique environment that includes highly corrosive media, radioactive environment, and high temperatures. In this section, the challenges involved with coolant selection and coolant interaction are described, including supercritical water, sodium, lead, liquid salt, gases. Several key aspects will be presented, including the nature of the coolant, properties, advantages, disadvantages, and key challenges.

In a nuclear reactor, a coolant is a necessity for heat transfer and is therefore essential for both power generation and for maintaining a consistent and safe operating temperature. However, a coolant can also play several other key roles and there are several key properties that must be considered. A coolant in a nuclear reactor must have the desired neutronics properties, typically very low cross-sections to minimize neutron absorption in the coolant rather than the fuel. However, in some reactor designs, such as water systems, the coolant also serves as the moderator. Ideally, the coolant will stay in a single state, either liquid or solid during operation, although in systems like boiling water reactors, steam is generated in the reactor core. Therefore, properties like heat capacity, thermal conductivity, and viscosity are very important. Other factors such as transparency may be important for practical applications in a reactor concept. For instance, one of the key challenges for sodium-cooled reactors is the inability to "see" through the coolant and observe components in the reactor core. Perhaps one of the most important

factors is the potential interaction between the coolant and other reactor structural materials. Corrosion (uniform or local), embrittlement, and integrity must all be considered and managed. As a result, coolant choice in a reactor often involves trade-offs in properties to find the right balance for both the reactor mission and lifetime.

Differences in coolants and in operating temperature regimes give rise to materials challenges as discussed in the sections below. There are general issues that affect all reactor concepts. In most reactors, it is thought that impurities at the part per million level significantly affect the potential for corrosion or embrittlement of reactor components. It is difficult to monitor the chemistry of the coolant in different regions of the reactor. Typically, coolant samples are obtained at operating conditions and brought to ambient conditions for chemical analysis. In gas cooled reactors, for example, the sample is taken from 950°C and a pressure of 7MPa to room temperature and one atmosphere or less for analysis. It is not clear in most cases if the coolant and relevant impurities are in thermodynamic or kinetic equilibrium and if the effects of radiolytic production of chemical species are preserved in such samples.

Supercritical Water

Supercritical water reactors (CWRs) are essentially light water reactors LWRs operating at higher pressure and temperature, generally above the thermodynamic critical point of water (374°C, 22.1 MPa). This feature allows far greater thermal efficiencies over other LWR designs, approaching 40-45% versus the 34-36% in GenIII technologies. As in a boiling water reactor, the superheated steam will be supplied directly to the high-pressure steam turbine and the feed water from the steam cycle will be supplied back to the core. These are simpler plants as the need for many of the traditional LWR components such as the coolant recirculation pumps, pressurizer, steam generators, and steam separators and dryers is eliminated. SCWRs build upon two proven technologies, the LWR and the supercritical coal-fired boiler. The main mission of the SCWR is production of low-cost electricity. Thus, the SCWR is also suited for hydrogen generation with electrolysis, and can support the development of the hydrogen economy in the near term. SCWRs are one of only six reactor technologies currently being studied under the Generation-IV international program.

Such SCWR designs have unique features that offer many advantages compared to state-of-the-art water-cooled reactors:

- SCWRs offer increases in thermal efficiency relative to current-generation water-cooled reactors. The efficiency of a SCWR can approach 44% or more, compared to 34-36% for current reactors.
- Reactor coolant pumps are not required. The only pumps driving the coolant under normal operating conditions are the feed water pumps and the condensate extraction pumps.
- The steam generators used in pressurized water reactors and the steam separators and dryers used in boiling water reactors can be omitted since the coolant is superheated in the core.
- Containment, designed with pressure suppression pools and with emergency cooling and residual heat removal systems, can be significantly smaller than those of current water-cooled reactors.
- The higher steam enthalpy allows a decrease in the size of the turbine system and thus helps to lower the capital costs of the conventional island.

These general features offer the potential of lower capital costs for a given electric power of the plant and of better fuel utilization, and thus, a clear economic advantage compared with current LWRs.

Currently, Canada is developing a pressure-tube-type SCWR concept with a 625°C core outlet temperature at the pressure of 25 MPa, and is designed to generate 1200 MW electric power. It has a modular fuel channel configuration with separate coolant and moderator. A high-efficiency fuel channel is incorporated to house the fuel assembly. The heavy-water moderator directly contacts the pressure tube

and is contained inside a low-pressure calandria vessel. In addition to providing moderation during normal operation, it is designed to remove decay heat from the high-efficiency fuel channel during long-term cooling using a passive moderator cooling system. However, the introduction of the passive moderator cooling system coupled with the high-efficiency channel could significantly reduce the core damage frequency during postulated severe accidents such as large-break loss-of-coolant or station black-out events.

However, there are several technological challenges associated with the development of the SCWR, and particularly, the need to validate transient heat transfer models (for describing the depressurization from supercritical to sub-critical conditions), qualification of materials (namely advanced steels for cladding), and demonstration of the passive safety systems.

Limited corrosion and stress-corrosion testing of traditional stainless steels in high-temperature water have had difficulty in identifying materials that would perform satisfactorily in the SCWR. A number of candidate cladding materials have been tested in capsules, autoclaves, and recirculating loops up to 700°C at a pressure of 25 MPa. Stainless steels with more than 20% Cr are expected to have the required corrosion resistance up to a peak cladding temperature of 650°C. More work is needed to develop alloys suitable for use at the design peak cladding temperatures of 850°C for the Canadian SCWR concept. Further work is also needed to better identify the coolant conditions that lead to stress corrosion cracking. It has been shown that the creep resistance of existing alloys can be improved by adding small amounts of elements, such as Zr. In the longer term, the steel experimental oxide dispersion strengthened (ODS) alloys offer an even higher potential, whereas the nickel-base alloys that are being considered for use in ultrasupercritical fossil fired plants are less favorable for use in SCWRs due to their high neutron absorption and associated swelling and embrittlement.

Key water chemistry issues have also been identified, although predicting and controlling water radiolysis and corrosion product transport (including fission products) remain the major R&D areas. Further, the importance of the loss of feedwater as a key abnormal event has been recognized. The design of a suitable high-pressure high-capacity fast-acting auxiliary feedwater system will also be a major challenge.

Sodium

There is considerable experience with sodium-cooled reactors and minimal materials issues remain unresolved today. Impurity transport in the sodium coolant circuits is a potential issue due to temperature gradients in the circuit, but modeling the process can be and has been done. Materials degradation is largely a chemical engineering issue, based on thermodynamics and kinetics and flow rate. Creep-fatigue mechanisms in Na environments are currently being examined in Department of Energy Office of Nuclear Energy (DOE-NE) projects and in Nuclear Energy University Programs (NEUP) projects, especially for 9Cr-1Mo steel and Alloy 709, prime candidates for sodium reactors. Liquid metal embrittlement does not appear to be an issue in Na systems. Coolant chemistry, especially oxygen, is well controlled in sodium even in reactors.

Lead

Heavy liquid metals have been proposed as a coolant for nuclear applications in advanced nuclear reactors due to their excellent heat transport properties, good neutronic properties, and limited reactivity with water and air. Lead or Lead Bismuth Eutectic for fission and Lead-Lithium for fusion are of special interest to the nuclear materials community. Historically several Lead Bismuth Eutectic and lead nuclear systems have been designed and operated in small reactors for submarines or spallation sources. Currently, a range of activities outside the US are ongoing on heavy liquid metal cooled reactors. The

Belgium MYRRHA project is building an ADS system while lead bismuth and lead cooled reactors are being designed in Russia (SVBR-100 and BREST respectively). Project ELECTRA in the EU, spearheaded in Sweden, also designs a small training reactor where the KTH created a spin off company "LeadCold". Despite these international efforts, only small companies exist in the U.S. where some engineering designs for a lead-bismuth reactor are being developed (e.g. Gen 4 Energy).

The main challenge in heavy liquid metal cooled systems is corrosion and precipitation of corrosion products at colder locations in the reactor system. Most common steel alloying elements, especially nickel, have a relatively high solubility in lead based coolants and therefore steel alloying elements can be dissolved on the hot section of a reactor and precipitate on the cold section. The common way to address this issue is by adding small amounts of oxygen to the liquid metal (10^{-5}-10^{-7} wt%) in order to allow the steel to form a passive layer on its surface making most steels usable to temperatures up to ~500°C. Even stainless steel, which contains significant amount of nickel, is protected from corrosion using this approach to this temperature. However, beyond 500°C, it has been found that most Fe-Cr based oxides break down and protection can no longer be assured leading to excessive corrosion especially on stainless steels. It is believed that the diffusion of the involved species through the passive layer becomes the dominant issue at higher temperatures but, given the corrosion rate, this must be accompanied with fast diffusion paths. In recent studies, Fe-Cr-Al alloys have been found to perform significantly better even at temperatures up to 650 or 700°C. However, data on even higher temperatures is scarce and therefore, it is difficult to say what the maximum limit may be on Fe-Cr-Al alloys. Some limited studies of commercial Fe-Cr-Al alloys have shown that passivation breaks down between 750-800°C but the alloys were not optimized for this application and the tests done are limited.

While an understanding of passivation and corrosion of steels in lead or lead bismuth has been established, corrosion phenomena, especially on the more complex oxide structures, still remain a challenge. It was found that multiple different types of oxides can form on the steels surface providing a rich microstructure. Another engineering limitation on heavy liquid metal cooled reactors is the issue of liquid metal embrittlement. It has been found that especially ferritic steels are susceptible to liquid metal embrittlement at temperatures up to ~400°C. This issue can be considered even more severe but also more difficult to study than the corrosion issue since mechanics, corrosion, and microstructure interact with each other. In addition to the materials related phenomena as described above, there are other coolant related challenges for liquid metal systems that remain open including how to control the oxygen concentrations at a wide temperature range, how does thermal cycling affect the observed phenomena, and how can one remove the transmutation product Po210 from the reactor.
.

Liquid Salts

Extensive studies were carried out through the MSRE, an 8 MW test reactor, operated in the 1960s at ORNL. The salt cooled reactor concept differs from the molten salt reactor concept in that no nuclear fuel is dissolved in the salt. Both concepts are low-pressure, high-temperature reactors. The salt cooled reactor uses clean fluoride salt, most likely LiF-BeF, as its primary coolant while the molten salt reactor employs primary coolant salt that contains dissolved fillies material.

Fluoride salts for nuclear reactor heat transport have liquid temperature ranges starting from about 450°C and boiling points near 1400°C providing a several hundred-degree margin from operating temperature to boiling. Liquid fluoride salts act as fluxes and remove any oxide layer that may be present on the metallic material. As a result, the normal approach to providing environmental resistance by alloying to produce a stable chromia or alumina surface layer is ineffective for these systems. Nickel alloys with low levels of Cr additions have become the choice for use in liquid fluoride salts. Other alloying elements are then added to the composition to increase the mechanical properties and provide resistance to oxidation in air

or other environments. One commercial alloy, Hastelloy N, was developed for the MSRE program that contains less than 7 wt% Cr (compared to typical values of 20-25% in high temperature alloys) and is solid solution strengthened by Mo. In addition to general corrosion, gradients in the chemical activities of constituents caused by temperature differences can result in dissolution of metal in one region of the system with subsequent deposition in other portions of the system. A non-isothermal system could also be subject to corrosion from thermal gradient mass transfer if the chemical potential of the corrosion product fluoride or chloride, at a given concentration, is a strong function of temperature, and deposition of the corrosion species occurs in the cooler regions of the system.

While the corrosion rate of Alloy N measured during operation of the MSRE was relatively low, specimens that were exposed to the fuel salt and then tested to failure showed shallow surface cracks at grain boundaries that connected to the salt-exposed surfaces. These cracks were determined to be due to the fission product tellurium. Modification of Alloy N with approximately 2% Nb significantly reduced the cracking and further studies also demonstrated that the cracking could be controlled by the redox potential of the salt.

High Temperature Gas

A number of very high temperature helium-cooled reactors have been built and operated for extended periods. The helium coolant in the primary circuit has been found to contain low levels of impurities after steady-state operation that can lead to an environmental degradation of the high temperature alloys used for internals and heat exchangers. Depending on the impurity concentration and the temperature, high temperature alloys can undergo oxidation, carburization, or decarburization. The concentration of H_2O and CO is of particular interest because they essentially control the oxygen partial pressure and carbon activity, respectively. The optimum coolant chemistry for long-term stability of high temperature alloys is slightly oxidizing and results in formation of a tenacious and protective Cr_2O_3 scale.

All of the high temperature reactor systems operated to date have had extensive gas clean-up systems associated with the helium coolant flow. These systems are intended to keep the total impurity levels in the helium below typically 10 ppm. Particularly in the early reactors, where the fuel was either not intended to contain the fission products or was ineffective in this function, the clean-up systems were also intended to capture radionuclides. Capture of tritium that is produced (at least in part) by transmutation of lithium impurities in the graphite remains an important function of the clean-up system. In the AVR and THTR reactors, active control was maintained on the H_2O and CO concentrations to reduce oxidation of the graphite reflectors, and the other impurities were routinely found to reach acceptable steady-state levels without active control. It has been noted that the clean-up systems may play a secondary role in maintaining gas chemistry, with the massive amount of graphite at high temperature present in all of the reactor designs playing a dominant role.

Interplay between the alloy surface, temperature, and gas composition determines whether corrosive oxidation, carburization, or decarburization occurs. The corrosion mechanisms of particular significance to mechanical stability are carburization and decarburization. Carburization is associated with low temperature embrittlement, and decarburization is linked to reduced creep rupture strength. Ideally, a continuous self–healing, impermeable passivating oxide layer is needed to establish the most corrosion-resistant alloy. In the case of Inconel 617, its chromia layer (Cr_2O_3) is the most important barrier from the effects of corrosive reactor gases. The modified Ellingham diagram describing alloy stability was based on the most relevant species involved in the corrosion process, namely chromium. Identifying which form—Cr_2O_3 chromium carbide or chromium metal—is most stable in a particular environment will determine the ultimate fate of the alloy. It is important to note that the gas chemistries found in operating reactors and used in the previous test programs are not in thermodynamic equilibrium. A steady-state gas

composition is reached at any temperature based on kinetic considerations. The concentrations of H_2O and CO largely determine the partial pressure of oxygen and activity of carbon, respectively.

MATERIALS IN EXTREMES OF FLUX, TEMPERATURE, CHEMICAL REACTIVITY

Nuclear reactor materials must survive in a unique environment that includes with highly corrosive media, irradiation environment, and high temperatures. In this section, each of the key elements of the extreme environment will be explored. It is difficult to examine each possible environmental and material combination, so discussion will focus on key degradation modes and examples. Several unique forms of degradation are also presented.

Material selection for large energy generating systems is typically governed by some predominant environmental effect, e.g., an oxidizing environment. In advanced nuclear systems, other important considerations come into play, including neutronics, the presence of dissimilar materials, and variable environmental interactions in different parts of the plant. In the gas reactor, for example, the pressure vessel is low alloy steel, the core internals are graphite, and the power generation modules are nickel-based alloys. In the molten salt reactor, the power conversion system may have molten salt at ambient pressure on one side, which requires a low chromium alloy, with steam at high pressure on the other which is highly oxidizing and typically calls for a high chromium material. Under these circumstances, it is difficult to specify a single coolant impurity content that is benign to all of the constituents.

High Temperature Performance

The desire for lifetimes of up to 60 years for structural materials at the high temperatures represented by advanced reactor system designs places severe restrictions on materials even in the absence of irradiation or environmental effects. There are only five materials approved for nuclear construction above 425°C in the American Society of Mechanical Engineers Boiler and Pressure Vessel Code (which is likely necessary for obtaining a commercial license). The qualified materials are 2.25%Cr-1%Mo, 9%Cr-1%Mo, Alloy 800H, Type 304 stainless steel and Type 316 stainless steel. None of these materials is qualified for the desired 60-year lifetime or for temperature up to 950°C that is desirable for high temperature gas reactors for some process heat or hydrogen generation applications. All of the materials depend on formation of a chromia scale for environmental resistance, which, as noted above, precludes their use in molten salt reactors. None of the materials uses sophisticated strengthening strategies common in nickel based superalloys common to aerospace applications because of potential issues with microstructure stability at long time and high temperature, the very large scale of nuclear components, and the need to weld heavy sections and complex geometries.

Creep and creep rupture

Creep and creep rupture are significant issues for high temperature reactor components. Not only is the failure of components unacceptable, the stress redistribution associated with creep creates difficulties with component design that are difficult to address with current high temperature design methods. Even with the massive amount of work on creep of some of the allowed materials (on the order of 1,500 creep tests each for Type 316 stainless steel and 9%Cr-1%Mo) and test times approaching fifteen years, there is doubt that there is sufficient experimental verification of long term behavior to design for 60 years. Due to a lack of understanding of very long-term phase stability and creep deformation mechanisms, the design community is only comfortable with extrapolating to longer times by a factor of 3 to perhaps 5 from experiments. This seriously delays acceptance of new materials for nuclear construction even if they are relatively mature and have wide acceptance in other applications. As a result, new materials are being

much more rapidly taken up for large fossil fired plants where the consequences for failure are not viewed to be as consequential.

It is well known from experience with operating fossil power plants that creep rupture life is dramatically reduced if a cyclic load is superimposed on the steady state load that causes creep. This issue has not been significant with the current fleet of nuclear plants since they operate below the creep range but there is a concern that it may be life limiting for advanced nuclear plants operating at higher temperature. There is very limited understanding of the interaction of creep and fatigue at the microstructure or dislocation substructure scale. As with creep rupture, it is also very difficult and time consuming to test in the laboratory. Several design approaches to predicting component life under combined loading conditions are applied by the engineering community. The most commonly used approach in the U.S. is a linear summation of a damage fraction ascribed to fatigue and a damage fraction from creep. This time fraction model is likely to be overly conservative and limit the potential useful lifetime of materials for advanced nuclear systems. The limited amount of experimental data which are available is also almost exclusive to the materials which are already allowed for nuclear construction.

Dislocation motion and vacancy diffusion are intrinsically linked on the time scale of creep and creep-fatigue deformation in structural materials in elevated temperature service. Non-slip mechanisms like climb are often key features of dislocation-controlled creep and at higher temperatures bulk vacancy diffusion can become an important mechanism competing with dislocation transport. Furthermore, grain boundary cavitation processes are often linked to diffusional mechanisms; meaning creep rupture processes as well as bulk creep are often coupled to vacancy diffusion. Despite the close interrelation between diffusion and dislocation motion there has been relatively little basic research into coupled multiphysics simulations of dislocation-diffusion processes in structural materials. Efforts to date have been simplified two-dimensional (2D) models that do not take advantage of modern high performance computing (HPC) resources or recent experimental techniques such as high-energy diffraction microscopy (HEDM) that can accurately characterize large three-dimensional (3D) volumes of material microstructures. Furthermore, models have yet to adequately address the complicated interaction of cavitation-based creep damage and fatigue mechanisms. In many materials, these are two separate interacting mechanisms. Capturing their interaction will require better understanding of the physics of void growth and fracture on the microscale as well as the development and application of advanced numerical methods to track the growth and interaction of voids and microcracks. Finally, non-monotonic loading introduces a host of material phenomena – Bauschinger and Masing effects – that are difficult to accurately describe with a microstructural model.

The lack of physically based, multi-physics simulations of diffusion-dislocation creep processes on the atomistic, dislocation, and mesoscale continuum scales coupled with methods for initiating and growing fatigue microcracking, both remain a limitation for these systems. Grain boundaries will pose a particular challenge as their effect on slip transfer, void nucleation and cavitation, and trans- versus intergranular microcracking is currently poorly represented in simulations. New experimental methods allowing in-situ examination of microstructures under relevant loading conditions are available but must be utilized as the simulation scale should be vast, requiring petascale HPC resources, aiming to resolve length scales on the order of a grain size (microns) to the structural scale (meters) to simulate substantial volumes of material in order to relate directly to engineering response.

Radiation damage/embrittlement

The core of a nuclear reactor is an extreme environment consisting of high temperature water, imposed service stresses and strains, intense radiation fields, and a corrosive medium. Radiation damage has been examined in many material systems for a wide range of nuclear applications. In general, the neutron irradiation field can produce large property and dimensional changes in materials. This occurs primarily

due to interactions between energetic neutrons (or other particles) with atoms on the crystal-lattice for different materials. Incoming neutrons displace these atoms at very high rates and generate vacancy and interstitial populations orders of magnitude beyond thermal equilibrium. This, in turn, leads to lasting effects on the material. Radiation damage processes are highly dependent on the material system, irradiation temperature, and damage rates.

In general, damage is manifested via one of five radiation damage processes: Radiation-induced hardening and embrittlement, phase instabilities from radiation-induced or -enhanced segregation and precipitation, irradiation creep due to unbalanced absorption of interstitials vs. vacancies at dislocations, volumetric swelling from cavity formation, and high temperature helium embrittlement due to formation of helium-filled cavities on grain boundaries. Further, some material systems (e.g. zirconium or uranium metal, which have anisotropic structures) may also undergo irradiation growth due to unbalanced nucleation of defect clusters into different prism planes.

1. *Radiation-induced hardening, localized deformation, and embrittlement*: Radiation hardening and localized deformation results from radiation damage, and the creation of vacancy and interstitial loops, which both impede dislocation motion. Once a few dislocations move along a given slip plane, they clear the "channel" of most of these barriers and subsequent dislocation occurs primarily in these channels, resulting in localized deformation. Both hardening and deformation changes can lead to radiation embrittlement, which results in an increase in the yield and ultimate tensile strength of the material. This increase in strength comes with a corresponding decrease in ductility. Ultimately, hardening and loss of ductility will result in reduced fracture toughness and resistance to crack growth.

2. *Radiation-induced segregation and phase transformations*: Under irradiation, the large concentrations of radiation-induced defects will diffuse to defect sinks such as grain boundaries and free surfaces. These concentrations are far in excess of thermal-equilibrium values and can lead to coupled-diffusion with particular atoms. In engineering metals such as stainless steel, this results in radiation-induced segregation of elements within the steel. For example, in Type 316 stainless steel, Cr (important for corrosion resistance) can be depleted at areas while elements like nickel and silicon are enriched to levels well above the starting, homogenous composition. While radiation-induced segregation does not directly cause component failure, it can influence corrosion behavior in a water environment. Further, this form of degradation can accelerate the thermally-driven phase transformations mentioned above and also result in phase transformations that are not favorable under thermal aging (such as gamma or gamma-prime phases observed in SSs). Additional fluence may exacerbate radiation-induced phase transformations and should be considered. The wealth of data generated for fast-breeder reactor studies and more recently in LWR-related analysis will be beneficial in this effort.

3. *Radiation-induced swelling*: The diffusion of radiation-induced defects can also result in the clustering of vacancies, creating voids. If gas atoms such as He enter the void, it becomes a bubble. While swelling is typically a greater concern for fast reactor applications where it can be life-limiting, voids have recently been observed at lower temperatures and dose rates than previously observed in components such as baffle bolts. In some components and systems, (notably high Ni-alloys that are susceptible to He-generation), swelling can be significant and is considered to be a major operational issue.

4. *Radiation-induced creep*: The motion of vacancies can also greatly accelerate creep rates, resulting in stress relaxation and deformation. Irradiation-induced swelling and creep effects can be synergistic and their combined influence must be considered, particularly in highly textured materials due to fabrication or even design in components such as fuel cladding. Longer reactor component lifetimes may increase the need for a more thorough evaluation of swelling as a limiting factor in operation. As above, data, theory, and simulations generated for fast reactor and fusion applications can be used to help identify potentially problematic components.

5. *He-induced embrittlement*: Transmutation of various elements in the alloy results in the production of He and especially in high-Ni alloys. He is known to have severe consequences on swelling and mechanical properties. Under high temperatures and stress, He can migrate to grain boundaries or other interfaces. As He accumulates, bubbles can form along these interfaces, greatly reducing the interfacial strength and ductility, sometimes as low as <1%. Embrittlement can be severe.

While not a direct impact on materials, irradiation can also affect the coolant via transmutation, leading to impurities and further complicating compatibility issues. In water-cooled systems, interaction between water molecules and energetic particles leads to radiolysis and a variety of short- and long-lived radicals and species are produced. This can lead to complex changes in corrosion and compatibility.

As noted above, irradiation effects are complex and highly dependent on the material, material state, irradiation temperature, and damage rate. Studies, theory, and qualification experiments must be carefully controlled to provide meaningful data. Furthermore, experiments often require handling of radioactive (and sometimes toxic) specimens. This mandates complex systems for safety of workers and equipment, which significantly complicates research into irradiation effects. Thus, modeling and simulation are increasingly becoming key tools in understanding mechanistic effects and predicting performance under irradiation.

Consider one material system as an example of the complexity of irradiation damage, particularly nuclear graphite. Nuclear graphite is a key material for several high temperature reactor designs providing components for the internal core structure, neutron moderation, and thermal transport and storage. The primary safety function of the graphite components is the protection and retention of the nuclear fuel. Thus, the structural integrity of the graphite components is critically important to the continued safe operation of these reactors and great effort is expended to ensure the components are mechanically sound. While severe degradation from high temperature oxidation or molten salt erosion can compromise the structural integrity of the core components, engineering controls can mitigate or even eliminate the severity of the corrosive effects. Irradiation effects, specifically irradiation induced dimensional change, are the primary life limiting damage mechanisms that can significantly affect the structural integrity of the components lifetime in the core.

During (neutron) irradiation, the macroscopic response of the graphite components is for the material to physically shrink a significant amount (percent change depends upon the graphite grade) up to a crucial dose level termed the Turnaround dose. After Turnaround, the graphite will begin volumetric expansion and continue to expand until complete failure. This macroscopic behavior is in response to neutron damage in the anisotropic atomic crystal structure of graphite where graphite crystallites grow perpendicular to the stacked graphene planes (c-axis direction) and shrink parallel to the graphene planes (a-axis direction) (Figure 10). As expected, during contraction the graphite components are actually stronger and less susceptible to fracture since the strong compressive stresses are built up within the graphite microstructure. However, once Turnaround is achieved, tensile forces build up, cracks begin to form, and the structural integrity can be noticeably compromised. The graphite turnaround dose level thus becomes critical in determining the safety envelope and operating lifetime for the graphite components. Since the Turnaround dose is uniquely dependent upon the accommodation pore volume and crystallite structure/alignment the raw coke source, fabrication method, and graphitization processes for each nuclear graphite grade yields different Turnaround doses. The ability to predict the performance of different grades of graphite is limited by incomplete fundamental understanding of the interrelationship between in-crystal irradiation damage and the out-of-crystal accommodation porosity.
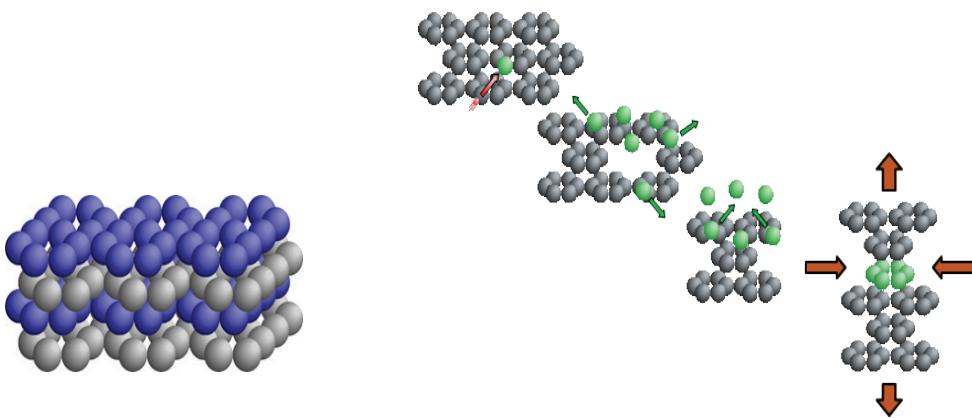
Figure 10. *Sub-plane formation and vacancy collapse in irradiated graphitic structures. Assumed to be the underlying mechanisms for irradiation shrinkage (a-axis direction collapse) and growth (c-axis direction expansion) of irradiated nuclear graphite components. Image courtesy of William E. Windes, Idaho National Laboratory*

References

1. B. Kelly, *Physics of Graphite*, Applied Science Publishers Ltd, London and New Jersey, 1981.
2. R. J. Price, "High-temperature neutron irradiation of highly oriented carbons and graphites", Carbon, Vol. 12, 1974, pp 159-169
3. B. Kelly and A. J. E. Foreman, "The theory of irradiation creep in reactor graphite-the dislocation pinning-unpinning model", Carbon, Vol. 12, 1974, pp 151-158.

Simulation and Study of Radiation Damage

Understanding radiation damage and mechanisms is essential to predicting the response of materials in advanced nuclear systems. Using test reactors and ion beam facilities to study the effects of radiation on nuclear materials has become a widely practiced approach and ion beam facilities are indispensable due to the sparsity and complication of using fast neutron test facilities. The quantification and limitation of using ion irradiation as a surrogate for reactor radiation to evaluate microstructure and property changes of structural materials has been established.

Historically, irradiation effects in materials have been advanced mainly under two extreme conditions: collision cascade processes dominated at low energies and ionization processes dominated at high energies. Research on ion-induced damage in materials is often superficially described in terms of nuclear nergy deposition (displacement cascade events) or electronic energy deposition at much higher energies (hundreds MeV to several GeV), especially from the modeling perspective. In both regimes, it is generally assumed that nuclear and electronic damage processes are fully separable. This generally means that, while ion energy deposition into the electronic and atomic subsystems are coupled in both time and space, the nuclear and electronic energy deposition and dissipation are often simplified as independent and non-correlated processes. In the intermediate ion energy regime (also the most important region for nuclear energy applications), the conventional understanding is that the large amount of energy deposited into the electronic subsystem will ultimately be dissipated by other mechanisms (electron excitation and heat dissipation without significantly affecting defect production), and the energy into the atomic

subsystem will be largely dissipated by the creation of atomic displacements. Electronic energy deposition can cause ionization that produces localized electronic excitations and thermal spikes (highly localized heat in the atomic subsystem along the ion paths, due to electron–phonon coupling), which can modify migration barriers and cause local atomic rearrangements. Recent work has demonstrated that ionization effects in ceramic materials with predominantly covalent bonding or mix covalent-ionic bonding are significant. Despite the fact that energy is thought to dissipate rapidly through the electronic subsystem in metals and conventional alloys (metallic bonding in the form of an electron cloud of delocalized electrons) and consequently have less impact on defect dynamics, new discoveries have shown that extreme compositional complexity in concentrated solid-solution alloys can substantially reduce electron, phonon, and magnon mean–free paths, alter coupling strengths, and therefore greatly affect defect production and microstructural evolution. These recent advances include better understanding of the nonadiabatic processes of the electron–phonon coupling strength on the residual damage, and when such electronic effects should be considered, ignored, or safely removed as heat from the atomic–scale models. Such knowledge is foundational to a better understanding of the interaction between atoms and electrons in the outcome of damage events.

Variation of charge on atomic defects may induce atomic-level stain and modify energy barrier for defect migration and microstructure evolution. Taking oxygen vacancies as examples, similar chemical expansion is found for neutral and charged oxygen vacancies in rock salt, fluorite, perovskite and pyrochlore structures, where neutral vacancies lead to lattice expansion and are stable under lattice expansion (tensile strain) nevertheless positively charged vacancies lead to lattice contraction and are stable under lattice compression (compressive strain). Even though the formation energy of oxygen vacancies decreases in the presence of tensile strain, it is not sufficient to predict vacancy stability. While strain could be used to manipulate defect concentrations, the degree of control largely depends on the material and the interfacial structure. Moreover, the stability of interfaces and grain boundaries is affected by charge redistribution during the non-equilibrium radiation processes. Grain boundaries are conventionally considered as sinks for vacancies, the sink strength of interfaces could depend on the interfacial strain, and the segregation of vacancies to interfaces may not be taken trivially. Local strain can affect vacancy stability and elemental segregation, which provide additional control in materials design for a wide variety of interfacial materials, not limited to nuclear materials, including fast-ion conductors, superconductivity, ferromagnetic, and ferroelectric applications.

Understanding radiation damage and mechanisms is essential to predicting the response of materials in advanced nuclear systems. Studies around the world have identified key mechanisms for a range of radiation-induced processes such as radiation-induced segregation, hardening, and embrittlement among others. However, there are a number of key issues that remain and must be resolved to further the development of radiation tolerant materials. For example, the role of interface character and nano-scale particles must both be understood to further the development of new materials that can resist the effect of irradiation to very long lifetimes. Other limitations include understanding and overcoming the combined effects of stress, coolant environment, and/or irradiation over long lifetimes. Accomplishing these performance goals and yielding a reactor design that is viable and affordable is an overarching limitation to advanced reactor deployment.

INTERFACES

Many of the extreme environmental effects described above rely on interfaces. Understanding interfacial processes is essential to understanding and mitigating degradation. The importance of interfaces is essential and new mechanistic understanding with a focus on key grain boundary issues in high temperature and radiation damage performance and recent research to elucidate underlying mechanisms.

Heat exchangers are an essential part of many aspects of society, not just nuclear power. Interfaces between the material of construction and coolant are essential in safe, reliable operation of these devices, particularly for future nuclear energy systems.

In many nuclear systems, a heat exchanger is required to safely and efficiently transfer heat, entirely dependent on interfaces and reactions between the coolant and materials. More compact reactor design, higher energy conversion efficiency, and smaller plant footprints are significant drivers for capital and operating cost reductions of advanced reactor systems. The use of a compact heat exchanger (CHX) to couple either a sodium fast reactor or a molten salt reactor with a compact, high efficiency, supercritical CO_2 Brayton cycle energy conversion system represents a technological advancement that would allow these potential cost reductions to be realized.

The leading CHX configuration for these applications is the diffusion-bonded micro-channel CHX. It comprises of a series of bonded thin plates or shims with chemically etched micro-channels with integral inlet and outlet flow distribution channels. The liquid sodium or molten salt and supercritical CO_2 flow in the channels in alternate shim plates.

The micro-channel walls are under high-pressure difference, due to the high system pressure of the supercritical CO_2 and the low system pressure of the liquid sodium or molten salt. In addition, the micro-channel geometry causes severe stress and strain concentrations. Thus high-temperature strength of the CHX material is essential. The initial quality and the time at temperature effect on the long-term mechanical reliability of the diffusion bonds are also important considerations. Compatibility of the CHX material with either liquid sodium or molten salt on one side and supercritical CO_2 on the other side of the micro-channels also represents significant challenges.

Thus, before these CHX designs can be deployed reliably in these advanced reactor systems in order to realize the cost reduction potentials, the microstructural underpinning of these phenomena need to be understood in order to down-select, or to develop new CHX material that can meet all of these requirements.

STATE OF THE ART NUCLEAR FUEL TECHNOLOGIES

Nuclear fuels for fission reactors experience profound changes in their chemistry and physical properties during their lifetime [1]. The chemical changes are the direct result of fission, introducing roughly a third of the elements in the period table into a system that originally consisted of two or three elements constituents. The chemistry evolution is often spatially non-uniform and, as a result, chemical potential gradients in the material are complemented with steep temperature and stress gradients [2]. As if the level of complexity was not already overwhelming, another factor, extreme radiation damage, needs to be considered to be able to grasp the phenomenon governing the behavior of fuels. The level of radiation damage far exceeds what is experienced in the most demanding cases for structural materials in fission and fusion reactors and is on the order of thousands of displacements per atom [3].

THERMAL SPECTRUM REACTOR FUELS

Most of our knowledge about nuclear reactor fuel performance is based on the vast experience with water reactor fuels. Many of these observations are relevant to key performance issues and limitations for advanced reactor fuels in thermal reactor designs and are described in more detail below.

High Burnup Effects

The combination of high irradiation dose, fission product buildup, and low temperatures that limit defect mobility, result in formation of a high burnup structure (HBS) on the periphery of the LWR fuel pellets [4]–[6]. This so-called restructuring transforms the original grains of ~$10\mu m$ containing small (few nm) fission product precipitaes and gas bubbles into ~100nm urania grains with large (~$1\mu m$) bubbles without releasing any of the fission gas [7]. Although the properties of HBS are now slowly being understood under normal operating temperatures [8], [9], the origins of its formation and understanding of the basic phenomena governing its behavior under reactor transients remain elusive. On the origins of its formation, multiple theories exist, with the most prominent one being polygonization [10]: buildup of dislocation cell walls to ultimately form low angle grain boundaries. The mechanism of dislocation density buildup and formation of these low angle boundaries and their relationship to the small (few nm) gas bubbles in the grains is not understood. The linear energy transfers of the fission fragments are below the threshold for dislocation loop formation [11], [12], yet high density dislocation networks form that drive the restructuring. It is also not clear whether the small gas bubbles assist in any way in this process and if large bubble manifestation is the cause or the effect of the restructuring.

Although HBS does not result in the release of any significant amount of fission gas (the gas remains in the large bubbles) under normal operation [7], it is prone to pulverization and significant gas release under temperature excursions characteristic of accident scenarios [13]. The mechanism of pulverization is not understood nor is the pressure inside the small and large gas bubbles is quantified in a reliable manner.

Fission Gas Release

Prediction of fuel performance under normal operating conditions is of paramount importance to the reliable and economic operation of the current fleet of nuclear power plants. Amongst the various performance aspects that need to be considered, fission gas release is one of the most impactful and least

understood [14]. The lack of understanding spreads throughout the various processes at high and low length and time scales. Even ignoring some of the engineering aspects (e.g. release due to cracking and under power ramps) [15], some of the most basic processes are still not well-understood. For instance, heterogeneous vs. homogeneous intragranular gas bubble re-solution processes are still debated and no robust mechanism for coarsening of fission gas bubbles under transients exists [16].

Enhanced Accident Tolerant Fuels

Given the strong focus on enhanced safety under LWR beyond design basis accident scenarios, a new host of cladding and fuel systems to replace urania clad in Zr-based alloys are under consideration [17]. The motivation for replacement of Zr is clear: there is 20-40 tons of this metal inside a typical LWR core and under beyond design basis accident conditions where core temperature could exceed beyond 1200°C, it rapidly oxidizes, releasing copious amounts of heat and hydrogen gas (byproduct of oxidation by water) [18], [19]. Therefore, cladding materials that have higher oxidation resistance (e.g. materials that form alumina, chromia, and silica as protective films [20]) have been proposed to be utilized instead. Alternative fuels that either offer higher uranium density, thereby enhancing fuel cycle economics, or barriers to fission product release are also under consideration.

The basic research needs in this area are often specific to the fuel-cladding system of interest. However, there exist key areas that are common amongst the various concepts and are not well understood. Swelling, creep, and fission gas release behavior of many of the non-oxide uranium compounds are not well understood. Often there is sparse data in a limited temperature, burnup, and power-operating regime but there exists no mechanistic and encompassing understanding. Another area, is the oxidation (by water or steam) behavior of the novel fuel and cladding systems [21]–[23]. Although the thermodynamics of these processes is well tabulated, the kinetics and phenomena governing the process are diverse and not well understood. For instance, unlike ZrO_2, the oxide films that form on the surface of the alternative cladding concepts tend to dissolve in high-temperature high-pressure water. Radiolysis and radiation damage also play an important part in this process for some of these materials. Finally, understating how to enhance the oxidation resistance of non-oxide uranium compounds is key in enabling their potential use in LWRs. Otherwise, their incorporation into these cores may compromise the safety of the plants to below today's levels.

There are number of limitations that must be resolved. For example, the high burnup structure formation mechanism in UO2 is not understood. The phenomena governing its behavior under rector transients remain elusive. The theories describing the mechanism of fission gas evolution and release in the current UO2 fuel system are still grappling with many unknowns. Reliable quantification of fission gas bubble pressure and re-solution and coarsening mechanisms are needed to make this form viable. Finally, a mechanistic approach to describe the swelling, creep, and fission gas release behavior of many of the non-oxide uranium compounds is needed. Mechanisms of reaction of water and steam with non-oxide uranium compounds and alternative cladding material to Zr-based alloys are required.

FAST SPECTRUM REACTOR FUELS

Metallic Fuel Forms

Deployment of any nuclear energy system depends, in large part, on a regulator's approval of the associated nuclear safety assessment, such as included with a Design Certification Application submitted to the U.S. Nuclear Regulatory Commission. The relative safety advantages for many fast-spectrum nuclear systems derive from attributes of the selected metallic alloy fuel design, derived from experience such as that gained in the U.S. [D. C. Crawford, D. L. Porter, S. L. Hayes, "Fuels for Sodium-cooled Fast

Reactors: US Perspective", Journal of Nuclear Materials, 371 (2007) 202–231]. Because safety analyses often credit the design of the fuel to ensure containment of radionuclides through normal operation and certain design-basis events (induced by equipment failure or external forces, such as extreme weather or infrastructure disruptions), the regulatory review process typically requires an applicant to demonstrate a sound understanding of the behavior of the fuel design through design-basis and even beyond design-basis conditions. Such understanding establishes confidence in the analytical treatment of phenomena and fuel behavior in the safety analysis and that is often demonstrated by incorporation of predictive models of phenomena into a fuel performance code used to assess by analysis the thermo-mechanical integrity of the fuel design (i.e., the integrity of the cladding that encapsulates the fuel material). Metallic alloy fuel designs have been demonstrated to be quite robust and well-suited for application to the systems in which they are deployed [G. L. Hofman, L. C. Walters, T. H. Bauer, "Metallic Fast Reactor Fuels," Progress in Nuclear Energy, 31 (1997) 83-110]; however, certain scientific phenomena need to be better understood and quantified to inform predictive models that are based on fundamental principles and mechanisms. The phenomena associated with the greatest need are summarized as follows.

Metallic fuel alloys are multi-component systems, as simple as U-Zr or as complex as U-Pu-Am-Np-Cm-Zr, that become increasingly complex in service with the accumulation of fission products and the associated morphological and microstructural changes. The high temperature irradiation environment and the temperature gradients induced across the dimensions of the fuel structures (generally, rods that are optimally designed among requirements for heat transfer and efficient fabrication and fuel handling) lead to localized phase equilibria and behavior. For example, fuel morphology and spatially-dependent composition are observed to evolve from an essentially as-fabricated condition to the condition in which fission gas bubbles exhibit temperature-dependent, and perhaps composition-dependent, accumulation and in which chemical composition varies with radial temperature. The implications for such behavior can be significant in which molten phase formation is observed to have formed after a high-temperature annealing test used to determine limits of acceptable operating temperature. It will be important to demonstrate, based on fundamental principles, the capability to predict localized fuel alloy compositions and phase equilibria under design-basis conditions and postulated-event conditions, to provide confidence for assessments of fuel and cladding integrity and radionuclide behavior. An understanding of metallic fuel phase equilibria for various conditions of local temperature and composition, and under the influence of neutron irradiation, is needed to establish quantitative models of fuel behavior.

During the in-service lifetime of a metallic fuel rod, the extended time at temperature can lead to diffusion of cladding constituents into the fuel alloy and of fuel constituents (including solid fission products) into the cladding – particularly at elevated fuel-cladding interface temperatures [D. D. Keiser, Jr., "The Development of Fuel Cladding Chemical Interaction Zones in Irradiated U-Zr and U-Pu-Zr Fuel Elements with Stainless Steel Cladding," in Nuclear Reactors, Nuclear Fusion and Fusion Engineering, Nova Science Publishers, Inc., Hauppauge, NY, 2009, pp. 163-194]. This phenomenon can lead to formation of phases and compositions with reduced solidus temperatures, similar to the phenomenon that leads to formation of eutectics in binary alloy systems, and can contribute to accelerated diffusion into the cladding, reducing the pressure-bearing wall thickness of the cladding. (Typically, the formation of liquid phases in the fuel region must be avoided to ensure that fuel does not slump or condense into an axial region of the fuel rod in a manner that could add reactivity into the reactor core. That behavior is quite extreme and not typically observed in any fuel system under design-basis events.) These inter-diffusion phenomena have been studied and characterized in terms of phase equilibria information available for binary systems of the multiple constituents, but establishing detailed phase equilibria information has been impractical for complicated mixtures of fuel, cladding, and fission product constituents. However, safety analyses for metallic fuel systems would be greatly enhanced by the incorporation of quantitative, mechanistic inter-diffusion models into fuel performance codes.

Fuel-cladding chemical interaction (FCCI) is primarily a result of lanthanide fission products (primarily La, Ce, and Nd), created in the metallic fuel matrix during irradiation, that migrate through the fuel to the cladding inside surface where they inter-diffuse with the stainless-steel cladding. The resulting interaction product melts at a relatively low temperature, which can become a limiting parameter in SFR operations during off-normal events. There is a desire to find a minor alloying element that will chemically react in situ with the lanthanide fission products and immobilize them, thus mitigating the fuel-cladding chemical interaction they cause by preventing their transport to the cladding surface. Palladium has shown some initial promise in this regard, which necessitates a more complete understanding of the chemistry of Pd with the lanthanides [G.W. Egeland, R.D. Mariani, T. Hartmann, D.L. Porter, S.L. Hayes, J.R. Kennedy, "Reducing Fuel-Cladding Chemical Interaction: The Effect of Palladium on the Reactivity of Neodymium on Iron in Diffusion Couples," **Journal of Nuclear Materials**, **423** (2013) 539-544]. However, there is a need to understand the lanthanide transport mechanism through metallic fuels, which could lead to other insights into mitigating FCCI in metallic fuel systems.

High Density Ceramic Fuel Forms (Oxides, Nitrides, and Carbides)

Metallic fuels present many advantages for fast spectrum reactor applications, primarily owing to their heavy metal densities, reasonable and more stable thermophysical properties, and ease of remote fabrication. However, fission product mobility and subsequent attack of cladding materials remains a key challenge to their deployment. Ceramic fuels have long been considered an alternative to metallic fast reactor fuels as a means of countering the above deficiencies, but demonstrated ceramic fuel forms present their own challenges. The most widely utilized ceramic fuel for fast reactor applications is the mixed oxide (MOX), generally consisting of uranium and plutonium in a solid solution. When transmutation of waste is a priority, plutonium can be displaced by neptunium or americium at levels up to a few atomic percent each without posing significant challenges to the required microstructure. Nitride and carbide ceramic compounds have also seen varying degrees of interest as fast reactor fuels. The mononitride system offers the advantage of broad solubility of both transuranic and key fission product elements. However, fabrication of high-density nitride monolithic fuel pellets, and in particular transuranic nitrides, is a significant challenge. Carbide architectures relieve some of the fabrication challenges of nitrides, but sacrifice many of the thermophysical property and irradiation advantages of the nitride.

MOX fast reactor fuels are at a far higher stage of development than nitrides or carbides. Numerous countries have chosen and utilized MOX over the past several decades to minimize the need for long term nuclear waste storage stemming from commercial light water reactor plants or dispose of plutonium resulting from historic nuclear weapons programs. Conversely, both nitrides and carbides have seen only limited exploration under the nuclear research programs of various countries around the world. The key challenges of ceramic fast reactor fuel forms therefore occupy disparate developmental positions.

The extreme temperature gradients drive mobility of porosity remaining in the fuel pellet following fabrication to the center of the pellet; porosity thus coalesces and forms the central void. While the void and radial cracking are the most obvious features when examined on the millimeter length scale, closer examination reveals further changes on the scale of pellet microstructure. The as-fabricated microstructures quickly give way to formation of columnar grains and radially-oriented porosity at regions of the pellet nearer the central void. Cooler regions of the fuel pellet are initially unaffected by this restructuring. However, at higher burnup the formation of the 'rim,' or 'high burnup structure' is observed at the pellet's outer radius. Here grains that are initially several micrometers in size subdivide and refine until they are tens to hundreds of nanometers in size.

At present, observations of these phenomena are limited to post irradiation examination of MOX fuels. This significantly restricts the quantity and diversity of data available. While these limitations have been

factors for the entirety of the nuclear era, recent developments in modeling and simulation capabilities provide the ability to utilize mechanistic approaches to understand and predict the evolution of MOX structure during irradiation. Improved means of collecting data and statistical quantification of microstructures of existing samples as well as novel out-of-pile methods are likely required to both inform and benchmark modeling and simulation efforts. In turn, new modeling approaches are likely necessary to fully capture porosity mobility and restructuring on the nanometer to millimeter length scales experienced by MOX.

The extreme temperature gradients that give rise to the gross restructuring of MOX fuels also contribute an additional evolution that governs fuel behavior both during normal operation and during design-basis accidents. Plutonium and americium are observed to segregate to the hottest portions of fuel. This behavior poses a challenge to fuel performance models, as local fission density can be impacted by actinide distribution. Oxygen is believed to behave similarly, but experimental observations of light element distribution representative of those experienced in pile following reactor shutdown, sample storage, and processing have so far limited experimental analyses to out of pile investigations. It is well known that the oxygen-to-metal ratio of MOX significantly affects many properties critical to fuel performance (e.g. J Nuc Sci Tech 48 (2006) 646). However, little is known of the rate and extent of oxygen diffusion within an operating fuel pellet.

Modeling of both cation and oxygen diffusion has advanced significantly in recent years (e.g. Scientific Reports 6 (2016) 36024) but experimental approaches are far less mature (e.g. J Nuc Mat 389 (2009) 416). Experimental means of better measuring the distribution of these elements in-pile or under representative conditions are necessary. Further, targeted experimental approaches to address specific data deficiencies of the modeling community are likely required to better understand the effect of Soret diffusion on specie mobility in MOX systems.

Fuels for Sodium Cooled Reactors

Fuels in sodium fast reactors need to be irradiated to at least 20% burnup before discharge and recycling for the process to be economically feasible. Presently, achieving a burnup of 20% in fast reactor fuels is a significant challenge. The cladding materials used in such high dose irradiations will experience at least 200 dpa of exposure. The requirements are that the cladding must contain the fuel until discharge and the duct must maintain ductility and mechanical strength to direct the sodium coolant and allow handling of fuel subassembly during and after discharge.

One major material property need for cladding is radiation resistance (physical and dimensional changes), meaning that cladding dimensional and physical properties must be resistant to radiation-induced degradation. In the core midplane where flux is the highest, the cladding must be resistant up to at least 200 dpa. The corresponding dose at the top and bottom of the core will be substantially lower, but irradiation temperature at these locations often poses additional challenges in maintaining irradiation resistance, especially at the bottom of the core where irradiation temperatures typically drop below 400°C.In addition, tensile properties (strength, ductility), creep resistance, fracture toughness, and fatigue resistance must all be sufficient to contain the fuel during irradiation, discharge, and other fuel handling operations.

Fuel clad interactions with the fuel must also be resistant during irradiation to prevent clad failure. In addition, the cladding materials must be able to be fabricated into component parts and joined to contain the fuel during the irradiation and during discharge. In addition, there are a number of other considerations, such as corrosion resistance (the material must be tolerant of the sodium coolant, with minimal corrosion rate), reprocessing (the cladding must be dissolvable to enable fuel reprocessing), thermal properties (high thermal conductivity and low thermal expansion are important to maintain

desired fuel and clad temperatures during irradiation), neutronic properties (low neutron absorption is required), cladding material availability (sufficient raw material must be readily available), and economics (the cost of the cladding material must be considered but is not a significant driver).

Material performance needs for the duct (structures to define coolant flow) are similar but with some changes to the ranking of the desired material properties. The purpose of the duct material is to control coolant flow distribution through the core, provide a means to insert and remove fuel pins, and provide another layer of protection from fuel pin breach during fuel handling operations. The duct will operate at lower temperature than the fuel, and thus the temperature envelope for the duct will be lower than the fuel. Unlike the fuel pins where stresses can build due to fission gas release, stresses on the duct material will be due primarily to structural support of the fuel pins and from handling of the duct. Tensile strength and toughness will be the properties of highest importance while creep will become less important. High thermal conductivity, low thermal expansion, low neutron absorption cross-section, and high swelling resistance are still important. FCCI and reprocessing requirements are of course a non-issue with the duct.

Sodium Fast Reactor Fuel Clad and Duct Materials

Presently, the cladding material that has been widely used to the highest exposure in U.S. fast reactors in coupon and component form is HT-9 (a 12Cr ferritic/martensitic steel). Swelling was measured to be less than or equal to 2% after irradiating to 200 dpa at 390°C. Swelling at higher temperatures was less. Creep measurements have also been performed after irradiation to 200 dpa at 400-600°C showing stable creep response to temperatures up to 550°C. At 600°C an increase in the magnitude of the transient creep response was observed and creep rate per unit stress began to strongly increase. One of the longest running fuel experiments (the ACO-3 experimental fuel subassembly) in Fast Flux Test Facility (FFTF) consisted of HT-9 clad and duct with oxide fuel. This subassembly ran without issue to a peak dose on the duct of 155 dpa and the plan was to continue to a higher burnup. Laboratory measurements of post-irradiation toughness show a significant reduction for irradiation temperatures below 400°C, but there have been no instances of fracture of HT-9 clad and duct during post-irradiation handling operations at FFTF. Thus, the near-term primary candidate clad and duct material for Fuel Cycle Research and Development is HT-9. The two main limitations of this material are a reduction in toughness at irradiation temperatures below 400°C and a reduction in creep strength as irradiation temperatures approach 600°C. Thus, advanced materials being considered include next generation ferritic/martensitic steels, ODS-strengthened ferritic steels, and other advanced alloys that have the potential to offer better high temperature irradiation creep resistance while maintaining adequate fracture toughness at lower irradiation temperatures. The next generation ferritic/martensitic steels show significant increases in creep resistance at temperatures up to 650°C. These alloys include Mod 9Cr-1Mo, HCM12A, and NF616 (also being investigated as an advanced reactor core structural material). ODS strengthened ferritic steels offer further improvements in creep resistance beyond 650°C and may provide signification radiation tolerance and helium management from the nanometer scale precipitate dispersion. Alloys being investigated include MA-957, 12YWT, and 14YWT along with international collaborations investigating the leading ODS alloys being produced in Europe and Japan. All of these materials still require a great deal of further investigation as they have yet to undergo significant irradiation testing to understand the effect of irradiation on fracture toughness and other key properties.

HT-9 is a key alloy for sodium reactors and has been studied extensively in the past. This ferritic/martensitic steel has several key limitations. For example, radiation resistance (physical and dimensional changes) requires further study. Irradiation tests have been performed on HT-9 in coupon form to greater than 200 dpa and in component form to 155 dpa. Extensive testing has been performed including fracture toughness, Charpy, and tensile testing. Mechanical properties (before and after irradiation) must also be examined. The tensile properties (strength, ductility), creep resistance, fracture toughness, and fatigue resistance have been tested before irradiation. Presently, HT-9 is not available commercially. So, some additional testing will be required on new heats. Irradiation data is presently available to 100 dpa. Previously irradiated samples are presently being irradiated in a fast reactor to obtain data at doses up to and greater than 200 dpa. Significant changes in thermal conductivity are not observed until swelling levels exceed 1%. The coefficient of thermal expansion is nearly independent of irradiation dose/damage.

While, significant data exists showing excellent resistance to corrosion when oxygen levels are controlled to below 10 ppm, fuel-clad-coolant interaction tests will be required. FCCI tests using HT-9 as cladding have been performed with U-10Zr and MOX fuel to burnups approaching 20%. More testing is underway for transmutation fuels containing Pu, Am, Np, and Cm.

Fabrication and joining must also be considered. Fabrication of tubing and duct materials has been successfully performed by industry. Welding parameters are well established and irradiation data is available on welded joints. This cladding is dissolvable to enable fuel reprocessing.

Advanced ferritic/martensitic and ODS-strengthened ferritic alloys are also being explored. There are limitations that must be resolved. Radiation resistance (physical and dimensional changes) is a major need. Previous irradiations were performed in FFTF and MOTA experiments on T91 and MA-957 to doses up to 110 dpa. Most of these samples have been tested. Additional samples were irradiated in the Phenix MATRIX irradiations to doses up to 70 dpa and include T91, HCM12A, NF616, MA-957, 12YWT, and 14YWT at 400 and 500ºC. These samples are being shipped back for testing and will provide some needed data on a coupon scale but additional data on component as well is coupon scale is still required to test the irradiation resistance up to and greater than 200 dpa.

Mechanical properties (before and after irradiation), like HT-9, also requires further consideration. The tensile properties (strength, ductility), creep resistance, fracture toughness and fatigue resistance have been tested for T91 before irradiation. Post irradiation data on T91 are limited to a small number tensile, fracture toughness, and creep tests from specimens with most of the specimens irradiated to less than 100 dpa. There is a small but growing database on the properties of unirradiated ODS strengthened steels (MA-957, 12YWT, 14YWT). There is a very limited database on the mechanical properties of irradiated ODS steels. Further pre-irradiation and post-irradiation testing of these ODS steels is either planned or in-progress.

Most advanced alloys will require compatibility testing. Some testing has been performed on the corrosion resistance of T91 in sodium showing some decarburization at temperatures above 600ºC. Additional sodium corrosion data is needed on advanced ferritic martensitic steels and ODS alloys. FCCI must also be evaluated. Some coupon level tests have been performed to analyze FCCI interactions with T91, HCM12A and some ODS alloys. More testing at coupon level is needed followed by component level testing.

Fabrication and joining of tubes and ducts of T91 and advanced ferritic steels is possible. MA-957 has been produced in tube form and weld methods to seal tubes with endcaps have been developed. More development is in progress to produce large heats of advanced ODS alloys such as 14YWT. Data shows

that reprocessing becomes more difficult with lower chrome alloys. More data is needed on these advanced alloys.

GAS-COOLED REACTOR FUELS

Modular high-temperature gas-cooled reactor (HTGR) designs were developed to be safe through natural processes that prevent significant coated-particle fuel degradation and failure under all licensing basis events. The principle that guides modular HTGR design is to passively maintain core temperatures below fission product release thresholds under all accident scenarios. The required level of fuel performance and fission product retention reduces the radioactive source term by many orders of magnitude relative to source terms for other reactor types and allows a graded approach to emergency planning and the potential elimination of the need for evacuation and sheltering. Achieving this level, however, is predicated on exceptionally high coated-particle fuel fabrication quality and excellent performance under normal operation and accident conditions.

TRISO coated particle fuels are at the heart of the modular HTGR concept. Such fuels have been extensively studied over the past four decades. Layers of carbon and silicon carbide surround a uranium kernel to form a TRISO-coated fuel particle of approximately one millimeter in diameter. A typical modular HTGR will contain billions of TRISO-coated particles. The particles are overcoated with a graphite material and packed together as either right circular cylinders or tennis-ball-sized spheres called pebbles. Rigorous control is applied at every step of the fabrication process to produce high-quality, very-low-defect fuel. Defect levels are typically on the order of one to two defects per 100,000 particles. The TRISO layers provide robust protection for the uranium kernels and superb retention of the radioactive material produced during fission. Extensive testing in Germany in the 1970s and 1980s and recent U.S. DOE tests have demonstrated the outstanding performance of TRISO-coated particle fuels under both normal operation and accident conditions.

Qualification of fuel for use in a licensed reactor involves experiments and examinations that will allow an understanding of the behavior of TRISO-coated fuel under the radiation and temperature environment expected in a modular HTGR. It also involves experiments to allow for an understanding of how well the fission products (i.e., the elements produced when uranium fissions) stay inside or move outside of the coated fuel particles and through the graphite reactor core. Development of modeling and simulation tools to analyze and predict this behavior is also important to the design and safety analysis for the modular HTGR.

Over the past 15 years, the Advanced Gas Reactor TRISO fuel program re-established the capability to fabricate and characterize TRISO-coated particle fuel in the U.S. This was a significant effort that required the development and modernization of the fabrication processes and characterization approaches used in historical TRISO-coated fuel made in the 1970s and 1980s. Many of the procedures and recipes used in the past were still available but were supplemented or replaced with newer technology to take advantage of improvements in materials processing and measurement science over the past 25 years. The result has been better fabrication control and much more accurate and precise characterization of this fuel form. The project is now fabricating TRISO-coated fuel particles with industrial-scale equipment. The first fuel experiment, called AGR-1, completed approximately three years of radiation exposure at the high temperatures expected under normal operation in an HTGR. About 300,000 TRISO fuel particles were tested to a peak burnup of 19 percent without a measurable indication of any fuel kernel exposure as a result of TRISO-coating failure during irradiation. Careful post-irradiation examination revealed that only four particles exhibited moderately elevated fission product release from compromised SiC layers. Subsequent testing of this fuel in high temperature furnaces to simulate postulated accident conditions has

demonstrated continued robustness of the TRISO-coated particle fuel form for hundreds of hours at 1600, 1700 and 1800°C.

The fission product releases measured from this fuel are, except for Ag isotopes, quite small. Key radioactive fission products to be considered are ^{137}Cs and [90]Sr (due to the significant role in offsite dose if released from the core during accidents) and [110m]Ag and [154]Eu (due to the ability of these elements to be released to varying degrees through intact SiC). However, the scientific understanding of the retention and transport mechanisms in the kernel and through the coatings is still incomplete. Recent work has provided some evidence of transport via specific types of grain boundaries in the SiC. Less is known about the behavior in the kernel and buffer layers. Limited data suggest significant retention in the kernel and/or buffer and detailed microstructural examination continues to provide more quantitative results. A better understanding of the phenomena that govern this retention and transport would support the development of improved fission product transport computer codes used by industry to calculate the reactor source term under normal operation and postulated accident conditions. This includes analysis of fission product chemistry inside the particles, which may have a significant effect on transport behavior. Testing samples that did not adequately represent actual irradiated nuclear fuel plagued previous efforts in this area. As a result, experimentation must be focused on using samples that adequately simulate the actual fuel environment from a radiation damage, temperature and chemical potential perspective.

While all the components of the TRISO particle work together as a system to ensure good fission product retention throughout the life of the fuel, most fission product retention occurs within the kernel itself due to the formation of stable compounds. Kernel chemistry is complex due to the generation of numerous fission products, but can be understood through thermochemical modeling. While thermochemical modeling of kernel chemistry as a function of starting composition and burnup has been performed previously (e.g., Homan et al., Nuclear Technology 35, 1977, 428-441), experimental data to support kernel microstructure and chemistry evolution during irradiation—including the chemical state of specific fission products—would help to better understand fission product behavior in the kernels.

Various fissile kernels have been proposed and tested historically around the world as part of the development of different fuel cycles for HTGRs. Plutonium oxide and uranium dioxide kernels have been studied in the past, with most effort on UO_2. A key failure mechanism for this fuel is over-pressurization and corrosion of the SiC layer by CO formed during irradiation. Under irradiation, free oxygen is formed as a result of fission and the valence of the resulting mix of fission products is insufficient to chemically recombine with all the oxygen produced. The extra oxygen reacts with carbon in the buffer to form CO. If there is a crack in inner layers, the CO can attack the SiC layer and cause it to degrade. Chemical buffers are a key technique to engineer around this problem by adding oxygen getters to the kernel.

Uranium carbide is an attractive choice for limiting CO production from UO_2 fission because it does not reduce the density of fissile material. The mixture of uranium carbide and uranium oxide in a UCO fuel kernel has been a focus of the current AGR program. However, one drawback of using uranium carbide as an oxygen getter is that some fission products (e.g., Eu and Sr) have a lower oxygen affinity and therefore do not reside as stable oxides in the UCO kernel. Other oxygen getters (e.g. SiC or ZrC) have been considered for control of CO content while retaining more metal fission product as oxides. These alternate getters may also offer possible retention solutions for fission products, such as palladium, that do not form simple stable oxides. Research is needed to determine the optimal method to introduce getters to the sol-gel kernel fabrication process and to determine the optimal type and quantity of chemical buffering agent for the ranges of burnup and temperature expected for these fuels.
In addition to adding buffering agents to the well-known oxide systems, other uranium compounds may offer solutions for different reactor designs. Uranium carbide and uranium nitride provide the benefit of possibly higher fuel density, but also have limitations and drawbacks that need to be understood.

It has been shown that TRISO particle performance under irradiation can be affected by the interaction of the individual layer at the layer interfaces. Strong bonding is preferred for the IPyC/SiC interface while weaker bonding appears to be beneficial at the OPyC/SiC and Buffer/IPyC interfaces. Based on emerging post-irradiation examination data from the AGR-1 and AGR-2 irradiation experiments, the ease with which the buffer separates from the IPyC layer appears to have important implications for eventual SiC failure; therefore, the strength of the buffer-IPyC interface is of particular importance. Reliable techniques are needed for the measurement of the interface strength of TRISO coated particles and parametric studies to quantify the effect of interface strength on irradiation performance would assist in advancing irradiation performance modeling efforts.

The pyrolytic carbon layers in TRISO particle fuel have a complex microstructure since carbon structures are formed both at the growth surface and in free structures suspended in the reactant gas that later adhere to the growth surface. The balance and combination of these two mechanisms can impact the physical properties of the layer, such as density, permeability, and anisotropy. While it is known how to control the deposition process to achieve the basic structure and required properties of the PyC layers, additional study could further elucidate the direct connection between microstructure, bulk properties, and irradiation performance and broaden the allowable ranges for these properties.

SALT-COOLED REACTOR FUELS

Nuclear Energy applications of molten salts, and complex ionic liquids which can be used for nuclear fuel processing in these systems, will result in these liquids being exposed to extreme conditions. The molten salts must survive in harsh radiation fields and the materials that interact with them must withstand be compatible. Therefore, fundamental science investigations into the key factors that govern the behavior of molten salts with and without dissolved fuel and fission products and the materials they are in contact with under the extreme environmental conditions they would experience in nuclear energy-related applications.

Prior experience with these liquids includes LiF-BeF2 salts, FLiNaK-based salts, and NaF-ZrF4 based salts that were utilized with fuel in very intense fissioning irradiation capsules and while molten, no notable radiolysis effects were reported. Radiolysis, however, has been reported for frozen salts at lower temperatures. Limited experience during reprocessing activities exists regarding radiation stability of molten chlorides salts, intended to be used for fast spectrum reactors, of the U.S. and Russia using various chloride salts ("No data exists" is just not a true statement).

While engineering experience exists, to different extents for the different salt systems, a number of key basic questions remain that are worth exploring.

There is widespread debate whether molten salts experience anything similar to the radiolysis effect observed in water in LWRs. As noted earlier, prior experience implies that in the molten state such a phenomenon seems to be absent. In any case, explaining why there is no radiolysis effect in molten, or if there is, how it takes place is worthwhile from a basic science perspective. This is particularly important since fast spectrum salt reactor designs experience significantly higher levels of radiation dose.

Another important question is the effect of impurities in these salts. The salts may or may not contain U or Pu (fissile elements) and fission products. Also, water and metal halide impurities are worth consideration. Robust mechanistic understanding of tritium transport, gettering, and recovery processes are essential. Understanding of what compounds do the impurities form with the salt and with container materials is important. Does the presence of fuel or fission product exacerbate or alleviate the corrosivity of the salts?

In some of molten salt reactor design, application of discrete TRISO-based fuel has been proposed. Given the higher power density of these systems, the dose and temperatures associated with the fuel operating regime will differ (higher dose, lower temperatures) from the gas-cooled reactor systems. In that case, behavior of the coated fuel particles and their constituents needs to be understood. Understanding the mechanism for interaction of graphite and/or pyrocarbon with molten salt under irradiation is a key question. Also, owing to their low-dose application, swelling and creep coupling for the pyrocarbon layer in the TRISO fuel particles has been ignored in HTGR designs. It is prudent to further investigate and understand this coupling that will have a great impact on thermo-mechanical evolution of the fuel particles under high dose.

Basic thermodynamic data, interatomic potentials, and phase diagrams for salt solutions are needed, especially for chloride salts and salts containing fuel and fission products (the concentration of which may evolve throughout the reactor fuel cycle). In many cases, these can't be made because thermodynamic parameters are lacking (Gibbs free energies).

There are several key limitations that must be resolved for liquid salt fuel systems. For example, basic thermodynamic information on the various salts must (e.g. phase diagrams, interatomic potentials, etc.) be codified for common design and safety analysis. Further, the impact of fuel dissolution into the coolant and the resulting effects on salt chemistry must be understood. The influence and chemistry of impurities (with or without radiolysis) must also be determined to resolve questions about coolant chemistry during operation.

PHYSICAL AND CHEMICAL PROPERTIES AND PROCESSES AT INTERFACES

Many of the coolant systems described above rely on or contain many interfaces. These interfaces range from those separating various phases inside the fuel, in the micrometer or nanometer scale, or more expansive ones separating the fuel from the cladding or structural materials. Understanding interfacial properties and processes is essential to understanding fuel behavior and mitigating potential system degradation modes. This section will describe the importance of interfaces and recent understanding.

Coolant Clad Interfaces

Coolant cladding interfaces are key for reliable and safe operation of fuel in many fission systems. Given the high corrosivity of high-pressure, high temperature aqueous environments, this has been a topic of much attention in LWRs for the past few decades. The basic processes governing corrosion of Zr-based alloys is still being studied [24] and understood. The corrosion behavior of Fe-based alloys in these systems is well understood and many guidelines for optimal operation (e.g. alloy composition and environment control) are now well codified. However, similar to Zr-based alloys, we are not able to pinpoint the exact nature of the oxidizing species and the specific rate limiting processes for cladding oxidation. Utilization of alternative cladding materials, such as SiC, that do not produce inherently protective films and readily dissolve in high-temperature aqueous environments, present a different paradigm [25]. In these cases, the effects of radiolysis and radiation damage in the underlying material are paramount [26].

Studies on gas-solid reactions between graphite and SiC in HTGRs are prevalent [27], [28], however, as they mechanically change when the activity of the oxidizing species in the He coolant changes, some data gaps exists (e.g. when a high partial pressure of oxidizing species is present). As noted earlier,

interactions between discrete fuel constituents (i.e. C and SiC) with molten slat under irradiation is less understood, although some applied studies are underway.

The SFRs appear to offer the least challenging environments when considering cladding coolant interactions. Long-time experience with Fe-based alloys and some recent results on SiC-based material in these environments do not show any evidence of any significant chemical evolution at these interfaces. This is quite different for other liquid metals (e.g. Pb-Bi, Pb-Li) where many factors such as oxygen activity greatly affects the evolution of these interfaces.

Fuel-Cladding Interfaces

The evolution of chemistry (due to fission) under high heat flux and intimate contact between the fuel and the cladding has a significant impact on fuel behavior. In LWR fuel pins, the fully bonded interface, consisting of mostly (Zr, U) O2 affects the thermo-mechanical evolution of the pin. In the earlier designs with limited toughness, stress corrosion cracking due to corrosive fission products (I and Cs) lead to fuel pin failure.

In SFRs, the fission products challenge the integrity of the fuel pin and engineering the interface to withstand attack by corrosive elements is key in reliable operation of the fuel. For HTGR fuel, coated TRISO particles, intimate contact between the fuel and the coating layers coupled with high temperatures results in an environment that can readily facilitate fuel and fission product attack of those layers. Although the coated particle fuel design has been greatly improved to mitigate failure (exceptional reliability has been achieved), a specific set of radionuclide fission products (e.g. Ag) manage to escape from these particles. The transport mechanism and the role of fuel and coating layer interfaces are not fully understood and require attention.

For molten salt systems, the interaction between molten fuel and the reactor primary loop materials is of great significance. It is important to understand the reactions that are taking place at the salt-material interface and the exact nature of the species that are present at the interface. Furthermore, the effect of radiation on these reactions remains to be understood.

Fuel to Fuel Interfaces

Within the fuel itself, many interfaces are present. These could be interfaces between multiple phases that are present in the as-fabricated fuel such as chromia-alumina-silica additives that segregate at the grain boundaries or the oxide and carbide phases that constitute the spherical fuel kernel in some of the TRISO fuel particle designs. As burnup accumulates, some of the fission products may precipitate new phases such as metallic (noble metals and Mo) fission product precipitates and fission gas bubbles. The presence of these phases affects the interfacial energy and cohesive strength of the grain boundaries. In some cases, presence of one interface may result in precipitation of other phase (e.g. gas bubbles and metal precipitates are often co-located.

References

[1]   D. Olander, "Nuclear fuels – Present and future," *J. Nucl. Mater.*, vol. 389, pp. 1–22, 2009.
[2]   D. R. Olander, *Fundamental Aspects Of Nuclear Reactor Fuel Elements*. Springfield, VA: Technical Information Center, 1976.
[3]   S. J. Zinkle and J. T. Busby, "Structural Materials for Fission and Fusion Energy," *Mater. Today*, vol. 12, no. 11, pp. 12–19, 2009.
[4]   M. L. Bleiberg, R. M. Berman, and B. Lustman, "Effects of high burn-up on oxide ceramic fuels,"

in *Radiation Damage in Reactor Materials. Part of the Proceedings of the Symposium on Radiation Damage in Solids and Reactor Materials*, 1963, pp. 319–428.

[5] V. V Rondinella and T. Wiss, "The high burn-up structure in nuclear fuel," *Mater. Today*, vol. 13, no. 12, pp. 24–32, 2010.

[6] H. Matzke and M. Kinoshita, "Polygonization and high burnup structure in nuclear fuels," *J. Nucl. Mater.*, vol. 247, pp. 108–115, 1997.

[7] C. T. Walker, S. Bremier, S. Portier, R. Hasnaoui, and W. Goll, "SIMS analysis of an UO 2 fuel irradiated at low temperature to 65MWd/kgHM," *J. Nucl. Mater.*, vol. 393, no. 2, pp. 212–223, 2009.

[8] C. T. Walker, D. Staicu, M. Sheindlin, D. Papaioannou, W. Goll, and F. Sontheimer, "On the thermal conductivity of UO 2 nuclear fuel at a high burn-up of around 100MWd/kgHM," *J. Nucl. Mater.*, vol. 350, no. 1, pp. 19–39, 2006.

[9] J. Spino, J. Cobos-Sabate, and F. Rousseau, "Room-temperature microindentation behaviour of LWR-fuels, part 1: fuel microhardness," *J. Nucl. Mater.*, vol. 322, no. 2, pp. 204–216, 2003.

[10] H. Matzke, A. Turos, and G. Linker, "Polygonization of single crystals of the fluorite-type oxide UO2 due to high dose ion implantation+," *Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms*, vol. 91, no. 1–4, pp. 294–300, 1994.

[11] T. Wiss, H. Matzke, C. Trautmann, M. Toulemonde, and S. Klaumünzer, "Radiation damage in UO2 by swift heavy ions," *Nucl. Instruments Methods Phys. Res. B*, vol. 122, pp. 583–588, 1997.

[12] H. Matzke, P. G. Lucuta, and T. Wiss, "Swift heavy ion and fission damage effects in UO2," *Nucl. Instruments Methods Phys. Res. B*, vol. 166–167, pp. 920–926, 2000.

[13] J. Turnbull, S. Yagnik, J. Noirot, C. Walker, L. Hallstadius, N. Waeckel, and P. Blanpain, "An investigation into fuel pulverization with specific reference to high burnup LOCA," in *Enlarged Halden Program Meeting*, 2014, p. F1.7.

[14] G. Pastore, L. Luzzi, V. Di Marcello, and P. Van Uffelen, "Physics-based modelling of fission gas swelling and release in UO 2 applied to integral fuel rod analysis," *Nucl. Eng. Des.*, vol. 256, pp. 75–86, 2013.

[15] G. Pastore, L. P. Swiler, J. D. Hales, S. R. Novascone, D. M. Perez, B. W. Spencer, L. Luzzi, P. Van Uffelen, and R. L. Williamson, "Uncertainty and sensitivity analysis of fission gas behavior in engineering-scale fuel modeling," *J. Nucl. Mater.*, vol. 456, pp. 398–408, 2015.

[16] D. R. Olander and D. Wongsawaeng, "Re-solution of fission gas–A review: Part I. Intragranular bubbles," *J. Nucl. Mater.*, vol. 354, no. 1, pp. 94–109, 2006.

[17] S. J. Zinkle, K. A. Terrani, J. C. Gehin, L. J. Ott, and L. L. Snead, "Accident tolerant fuels for LWRs: A perspective," *J. Nucl. Mater.*, vol. 448, pp. 374–379, 2014.

[18] M. T. Farmer, L. Leibowitz, K. A. Terrani, and K. R. Robb, "Scoping assessments of ATF impact on late-stage accident progression including molten core–concrete interaction," *J. Nucl. Mater.*, vol. 448, pp. 534–540, 2014.

[19] L. J. Ott, K. R. Robb, and D. Wang, "Preliminary assessment of accident-tolerant fuels on LWR performance during normal operation and under DB and BDB accident conditions," *J. Nucl. Mater.*, vol. 448, pp. 520–533, 2014.

[20] B. A. Pint, K. A. Terrani, Y. Yamamoto, and L. L. Snead, "Material Selection for Accident Tolerant Fuel Cladding," *Metall. Mater. Trans. E*, vol. 2(3), pp. 190–196, 2015.

[21] A. T. Nelson, E. S. Sooby, Y.-J. Kim, and B. Cheng, "High temperature oxidation of molybdenum in water vapor environments," *J. Nucl. Mater.*, vol. 448, pp. 441–447, 2014.

[22] E. S. Wood, J. T. White, and A. T. Nelson, "Oxidation behavior of U-Si compounds in air from 25 to 1000 C," *J. Nucl. Mater.*, vol. 484, pp. 245–257, 2017.

[23] B. A. Pint, K. A. Terrani, M. P. Brady, T. Cheng, and J. R. Keiser, "High Temperature Oxidation of Fuel Cladding Candidate Materials in Steam-Hydrogen Environments," *J. Nucl. Mater.*, vol. 440, pp. 420–427, 2013.

[24] A. T. Motta, A. Couet, and R. J. Comstock, "Corrosion of Zirconium Alloys Used for Nuclear Fuel Cladding," *Annu. Rev. Mater. Res.*, vol. 45, pp. 311–343, 2015.

[25] K. A. Terrani, Y. Yang, Y.-J. J. Kim, R. Rebak, H. M. Meyer, and T. J. Gerczak, "Hydrothermal corrosion of SiC in LWR coolant environments in the absence of irradiation," *J. Nucl. Mater.*, vol. 465, pp. 488–498, 2015.

[26] S. Kondo, M. Lee, T. Hinoki, Y. Hyodo, and F. Kano, "Effect of Irradiation Damage on Hydrothermal Corrosion of SiC," *J. Nucl. Mater.*, vol. 464, pp. 36–42, 2015.

[27] C. I. Contescu, R. W. Mee, P. Wang, A. V Romanova, and T. D. Burchell, "Oxidation of PCEA nuclear graphite by low water concentrations in helium," *J. Nucl. Mater.*, vol. 453, pp. 225–232, 2014.

[28] V. Presser and K. G. Nickel, "Silica on silicon carbide," *Crit. Rev. Solid State Mater. Sci.*, vol. 33, no. 1, pp. 1–99, 2008.

STATE OF THE ART IN SCIENCE TOOLS AND TECHNIQUES

Over the last decade, there are been substantial gains in the tools and techniques available to all of materials and chemistry science. For example, electron microscopy has made significant advances with new detectors and aberration corrected systems, providing spatial and elemental resolution well beyond what was achievable in recent years. These new capabilities and techniques will also enable new advances in understanding and technology for nuclear energy.

In the last ten years, there have been considerable advances in instrumentation, technique, and simulation/processing. These have enabled new understanding in fundamental mechanisms and performance. For example, atom probe tomography (APT) has enabled unique investigations and provided unique insights into the development of microstructures under irradiation. APT complements other commonly used techniques to characterize irradiated microstructures, e.g. transmission electron microscopy, neutron scattering, x-ray scattering, and other techniques by providing three-dimensional information and high-resolution chemical information. Currently, these advanced characterization techniques are commonly used to probe nuclear energy materials-related phenomena such as radiation-induced clustering, segregation, radiation-enhanced diffusion driven transformations in metals and ceramics, and corrosion behavior. However, no single technique or single experiment can provide sufficient information to paint a complete picture of material responses to complex nuclear environments. Each technique, such as high-resolution electron microscopy and atom probe tomography have their unique advantages in probing structural and chemical changes at the atomic resolution currently unachievable through X-ray and neutron beams. Integrating X-rays, neutrons, electrons, and ions into a coordinate tool set that enables characterization of the same location of a sample at different length scales is still needed.

Nuclear reactor research has unique demands in dealing with highly corrosive media, radioactive samples, and high temperatures. Developments in testing and approaches to this work are enabling new science. Materials in reactors are subjected to an extreme environment with multiple components that must be accounted for to understand and predict their behavior. In all reactor designs and concepts, core components are subjected to irradiation, mechanical stress, and a corrosive environment, all at elevated temperature. Thus, understanding the effects of irradiation alone is insufficient to predict the behavior of materials in such a multi-dimensional environment. The challenge is then to devise strategies and techniques to address this multi-faceted environment. All of this must be completed in an environment that is safe for researchers, mandating the use of hot cells and remote handling capabilities. This adds a complexity to testing and analysis that is unique to nuclear energy.

Finally, the rapid development of computational capabilities has provided new opportunities to explore mechanisms of performance over a wide range of scale, both time and space. For example, fundamental physical understanding of radiation-induced microstructural evolution, multiscale materials modeling has experienced many recent advances. These tools are also advancing alloy development and component level analysis in both normal operation and under accident scenarios.

BASIC TO APPLIED TRANSITIONS

Advances in science with respect to the connection between materials R&D and application have been made over past decade that will enable advances in nuclear energy. The transition from fundamental science to system-level applications must also be considered.

Advances in understanding materials to system transition are key to enabling progress in Nuclear Energy. The advanced characterization and computational tools need to be coupled and focused on their application to the real fabricated structures. To do this, the researchers need to characterize interfaces/regions produced by material processing, such as the weld/HAZ region of welded components. Recent advances in modeling of welding processes are being used to drive the design of the joint as well as help set the welding parameters and composition of the weld material. For example, the Ni-base Alloy A690 composition tends to have lower residual strains due to its composition, which in turn affects the SCC behavior. Other system related areas that are employing characterization and simulation are the new advanced processes, such as additive manufacturing and 3D printing, where the fundamental studies are being used to characterize microstructure as well as surface effects. These efforts require collaboration with vendors. New categories of materials such as those used for accident tolerant fuel are also making use of advance characterization studies to establish processing/fabrication approaches needed to achieve optimized final properties in the product forms. Finally, surface processing, critical to the actual performance of structural materials in real systems, is being explored. The understanding of cobalt free wear coating behavior has been used in the development of new alloys. The fundamental behavior of surface changes over time at operating temperature is also being assessed to achieve expected lifetime goals. In the future, collaboration of researchers with system designers, particularly as SMRs are developed, will be needed to learn where there is a need and how to use fundamental material research to advance nuclear energy.

CHARACTERIZATION TOOLS AND TECHNIQUES

There have been considerable advances in instrumentation, technique, and simulation/processing. These have enabled new understanding in fundamental mechanisms and performance. This section will highlight both advances in traditional techniques and new, novel approaches. In each subtopic, several key questions will be answered. *What are the latest advances? How have they been used for nuclear energy? What will future and upcoming advances bring?*

Atom Probe Tomography and Nuclear Energy Materials

Atom probe tomography (APT) has enabled unique investigations and provided unique insights into the development of microstructures under irradiation. APT complements other commonly used techniques to characterize irradiated microstructures, e.g. transmission electron microscopy, by providing three-dimensional information and high-resolution chemical information. Currently, APT is commonly used to probe nuclear energy materials-related phenomena such as radiation-induced clustering, segregation, radiation-enhanced diffusion driven transformations in metals and ceramics, and corrosion behavior.

Recent decisive technological advances in sample preparation and data collection have made the application of the APT technique easier, broader, and more efficient. When combined with sample preparation methods using focused ion beam (FIB) liftout and milling, APT data can be acquired relatively quickly (within one day) and only requires small amounts of material. As such, APT may be used as a screening technique. The use of FIB tools for sample preparation has opened for study new classes of materials, such as ceramics, and is poised to answer a wide range of microstructural questions by enabling the selection of the analysis location, such specific grain boundaries or the interface between a metal and its oxide. For example, the entire depth of an ion-irradiated sample was recently analyzed. FIB tools also have simplified the investigation of radioactive samples through the extraction of volumes that are small enough not to be considered radioactive. Recently introduced, higher efficiency detector technologies now enable the collection of not only larger but also more complete datasets, thereby increasing the amount of useful information collected.

A major challenge in the use of APT to characterize nuclear energy materials is to move from generating generalized qualitative observations to performing quantitative microscopy. A large gap separates the observation, e.g. the presence of solute clusters, from its quantitative description in terms of number density, size distribution, spatial distribution, composition, and all associated uncertainties. The notion of moving towards quantitative microscopy implies several aspects: a physical understanding of and quantitative model for data generation, a cultural shift regarding data analysis, and close collaborations with theoretical efforts.

While recent developments have made APT easier and more attractive, the physical principles and limitations of the technique remain unchanged. The same artifacts observed several decades ago are still visible in recently collected data. These artifacts resulting from the physics of field evaporation have been described previously in detail (however neither quantified nor corrected for). Unlike transmission electron microscopy (TEM), the APT technique does not benefit from a background in modeling or simulation. Consequently, artifacts remain not fully explained through hand-waving arguments and the influence of artifacts on local concentration or feature size measurements remains unclear in most cases.

Collecting quantitative information responds to the need to develop long-term predictive models, e.g. microstructural or mechanical property evolution. However, the direct comparison or use of APT data to benchmark models is rarely trivial: APT analyzes a small proportion of the atoms leading to uncertainties, microstructures may be complex, and the experimental and simulations communities often use dissimilar definitions or analytical tools.

Quantitative microscopy also presumes the reproducibility of data and its analysis. However, there are no established protocols for data reconstruction or data analysis and quantitative data is very rarely, if at all, accompanied by meaningful error bars and uncertainty ranges. A handful of round robin experiments have attempted to bring awareness to this issue and bring the community together to discuss and agree on best practices. Finally, to close on the challenges of quantitative APT, whereas scientific focus may have shifted to significantly more complex microstructures, most APT data analysis methods still remain relatively simplistic and their use is sometimes misguided from lack of more appropriate options.

Electron Microscopy

Since the early 2000s, many exciting advances in electron microscopy technology have allowed rapid improvement in the quality and scientific utility of the data obtained in electron microscopy of materials. Specifically, (1) electron optical aberration correction, (2) high-efficiency, high-speed analytical detectors, (3) efficient codes for simulating electron micrographs, and (4) efficient codes for data-mining analytical electron microscopy data have allowed many advances. When these advances are applied (either individually or in combination) to materials for nuclear energy applications, it is possible to determine with nanometer or sub-nanometer precision the structure and chemistry of defects produced under irradiation in almost any nuclear-relevant materials system. Future advances in nuclear materials science and chemistry will depend upon the application, and further improvement, of these new methods to allow experiments to probe the fine scales and chemical sensitivities needed for quantitative comparisons to modern theory and simulation methods.

Aberration correction makes it now routine for both TEM and STEM to achieve sub-0.1 nm spatial resolution when imaging a thin crystalline specimen tilted to a low-order zone axis[1,2]. This makes, for instance, the examination of dislocation loop cores to determine vacancy- or interstitial-nature fast and unambiguous[3], a tremendous advance over the diffraction-contrast TEM methods used since the 1960s[4]. New high-brightness electron sources, even on an uncorrected electron column, provide large ($\approx$2-5$\times$) factors of improvement in spatial resolution. High-efficiency, high-speed analytical detectors, particularly silicon drift detectors for X-ray spectroscopy[5] and dual-range electron-energy-loss spectrometers (EELS),

can now provide single-atom or near-single-atom sensitivity at nm or sub-nm resolution on a high-quality specimen[7]. These methods can be used to evaluate the chemical state of transition metals (such as Ti oxidation state in Y-Ti-O nanoclusters), measure small clusters and precipitates and low-level segregation to dislocations or grain boundaries, and other high-resolution, high-sensitivity experiments that will enable future experimental results to close the gap toward modeling and simulation results.

The influence of irradiation damage on the mechanical properties of metals is well documented, including the increase in yield stress, decrease in ductility, and increased susceptibility to stress corrosion cracking, and has been experimentally associated with the interactions of dislocations with irradiation-induced defects. These interactions include the localized pinning of glissile dislocations, the concentration dislocation flow into discrete deformation channels, and appearance of internal-grain pileups. However, many competing theories exist into the interaction of dislocations with irradiation-induced defects and the subsequent formation and evolution of deformation channels.

In situ ion irradiation in the TEM is a powerful approach to directly characterize dislocation interactions and observe the evolution of defect structures with time. *In situ* TEM ion-irradiation studies have focused primarily on three key areas: 1) the structural and chemical changes to the microstructure induced by ion irradiation, 2) the interaction of glissile dislocations with irradiation-induced defects, including the formation of deformation channels, and 3) the interaction of dislocations with other defects, such as grain boundaries, in irradiated materials. Results from these studies have proven instrumental in relating the grain and phase boundary state to self-healing properties, guiding the development of irradiation-resistant nanostructured materials, and have contributed to the fundamental understanding of the role of deformation channel/grain boundary interactions in irradiation-assisted embrittlement and stress corrosion cracking.

Future challenges and opportunities for *in situ* TEM irradiation studies include the incorporation of advanced scanning nano-beam diffraction techniques to resolve the local stress state during *in situ* interactions, increasing the integration of electron tomography with *in situ* studies for four-dimensional analysis, and further exploring the influence of non-ambient liquid and gas environments on irradiation damage.

Computational methods, which have advanced strongly in the last 10-15 years, are an indispensable adjunct to electron microscopy in radiation materials science. Simulation methods allow the unambiguous identification of defects by TEM and STEM and frozen atomic structures of radiation damage obtained by simulation methods can be used as input for electron microscopy image simulation[5]. Additionally, simulation of analytical signals (e.g. X-rays [6,7] and EELS[8]) have advanced and reached atomic resolution levels, allowing direct comparison between hypothesized structures and experimental data. Because of the copious (many gigabytes/hour) generation of spectroscopic data, either images or tomograms, produced by modern analytical methods (X-ray mapping, EELS), datasets can no longer be tractably analyzed by a human. Modern data-mining and multivariate statistical analysis methods provide signal aggregation, increasing insight from the noisy high-resolution data most relevant to matching with theory and modeling results, and provide qualitative insight and rank reduction for deeper understanding of the underlying processes captured by the analytical data.

These advances in electron microscopy are also matched by advances in sample preparation. FIB micro-sampling allows great site-specificity (e.g., grain boundaries with a particular coincident site lattice (CSL Σ value) and greatly reduces the radioactivity of the S/TEM specimen when the mother sample is neutron-irradiated. Low energy ion cleaning methods greatly ameliorate damage from the Ga^+ FIB beam, creating near-pristine specimens for S/TEM.

Combining advances in sample preparation with advances in optics, detectors, simulation codes, and data mining codes will allow unprecedented comparisons between future experimental results and modeling and simulation results.

References

1. D. A. Muller, et al., Science **319** (5866), 1073-1076 (2008).
2. P. E. Batson, N. Dellby and O. L. Krivanek, Nature **418** (6898), 617-620 (2002).
3. Y. Lin, et al., Applied Physics Letters **104** (12), 121909 (2014).
4. M. L. Jenkins and M. A. Kirk, *Characterization of Radiation Damage by Transmission Electron Microscopy*. (Intitute of Physics Publishing, Bristol, 2001).
5. R. Schäublin, Microscopy Research and Technique **69** (5), 305-316 (2006).
6. M. W. Chu, et al., Physical Review Letters **104** (19) (2010).
7. C. M. Parish, Microscopy and Microanalysis **21** (03), 706-724 (2015).
8. M. P. Oxley, et al., Physical Review B **76** (6) (2007).

X-ray and Neutron-Scattering

Recent developments in X-ray and neutron characterization tools have provided us unprecedented time and spatial resolution and *in situ* capability with extremely complex sample environments, shedding new light into the mechanisms and pathways of microstructural evolution and its correlation with material's macroscopic behavior.

The high penetrating power of high-energy X-rays and neutron beams allow for construction of complex but robust sample chambers that provide a variety of sample environments involving temperature, load/pressure, and corrosive conditions that are significantly difficult or impossible to achieve with other techniques and studies of lab-scale bulk specimens and nuclear fuels (containing heavy elements), providing measurements of real materials under real conditions in real time. Key processes controlling material behavior can be rapidly identified through *in situ* measurements on dynamic phenomena, e.g. mechanical deformation, nuclear fuel melting, and additive manufacturing.

Rapid advance of 3D X-ray and neutron characterization techniques is paving the way to a more complete understanding of microstructural heterogeneity and localized deformation of irradiated materials, critical to the prediction of material aging and degradation and the design of more irradiation-resistant new materials. A recent study of neutron-irradiated Fe-9Cr model alloy by high-energy X-ray diffraction microscopy (HEDM, also called 3D-XRD) revealed broader size distribution of subgrains formed during tensile deformation, with smaller subgrains preferentially distributed near the center and large subgrains close to the periphery of the parent grain, in contrast to more uniform size distribution of subgrains in the unirradiated specimen (Figure 11). The heterogeneous subgrain structure suggests that the grain-scale homogeneity of plastic deformation is significantly reduced in the irradiated material.

Combining a suite of X-ray or neutron scattering and imaging techniques to analyze the material behavior at multiple length scales in a single experiment has become an extremely powerful way for developing an integrated and accurate view of the structural interactions across the scales.

While the non-destructive nature of X-ray and neutron techniques makes them invaluable for *in situ* characterization, adding complex environments that simulate nuclear reactor conditions, e.g. radiation field, corrosive environments, remains a major challenge. These non-destructive methods are also attractive and promising for *in situ* four-dimensional (4D) characterization by including the fourth dimension of time to follow microstructural evolution, particularly when combined with external stimuli

of radiation field, temperature, load, and corrosive environments. While technically challenging, once overcome, these capabilities can bring revolutionary progress in our scientific understanding of nuclear materials.

As powerful as it can be, no single technique, single experiment can provide sufficient information to paint a complete picture of material responses to complex nuclear environments. Even though synchrotron X-ray and neutron techniques are fast approaching the spatial resolution towards electron microscopy, techniques such as high-resolution electron microscopy and atom probe tomography have their unique advantages in probing structural and chemical changes at the atomic resolution currently unachievable through X-ray and neutron beams. Integrating X-rays, neutrons, electrons, and ions into a coordinate tool set that enables characterization of the same location of a sample at different length scales is still needed.

Most importantly, *in situ* experiments must go hand-in-hand with computer modeling to maximize the impact of *in situ* characterization. Not only does the close coordination of *in situ* characterization and theory hold the promise for understanding, predicting, and controlling the full range of damage production, evolution, and materials performance over all time and length scales, but the measurement-analysis-feedback loop enabled by simultaneous simulation of the experiment will add tremendous value to *in situ* X-ray and neutron experiments of nuclear materials.
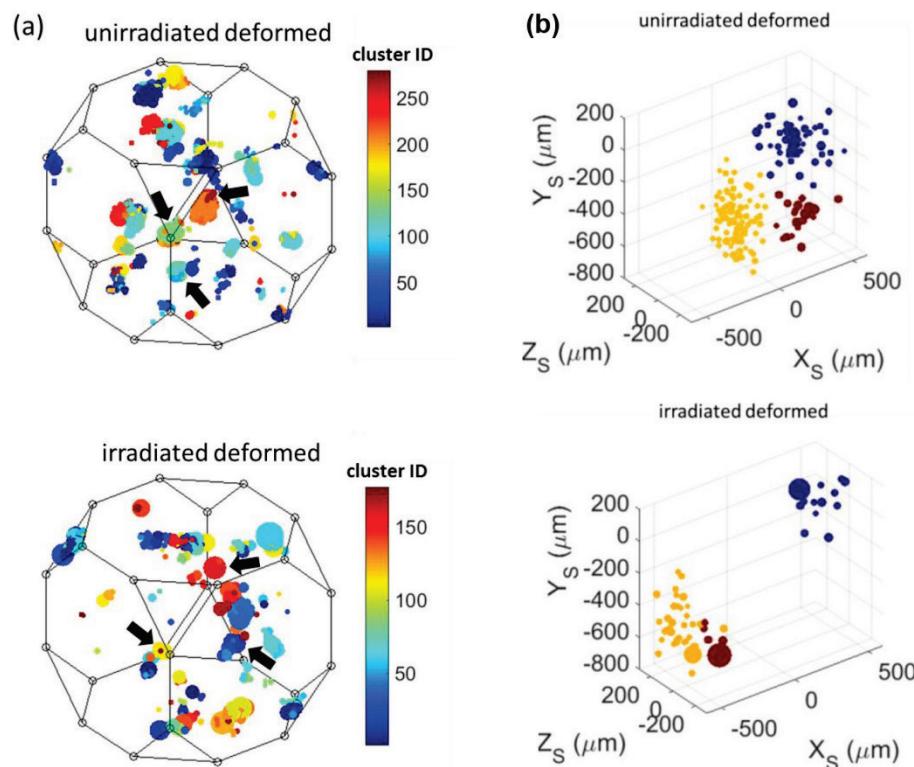
Figure 11. (a) Grain mapping in the Rodrigues-Frank orientation space and (b) maps of three representative subgrains (pointed by arrows in (a)) plotted in the sample coordinates, for the deformed regions of the unirradiated and neutron-irradiated (0.01 dpa/450°C). Image courtesy of Meimei Li, Argonne National Labboratory

Advances in Data Processing/Imaging

As described in the sections above, there have been significant advances in characterization techniques and tools. These are essential to overcome limitations in material performance under coolant and/or irradiation. This, in turn, has led to a need to analyze increasingly large and complex datasets. Development of new materials synthesis and optimization of existing materials systems for structural and functional nuclear materials necessitate the development of integrated synthesis and predictive tools. On the experimental side, the development of synthesis tools based on high throughput automatized synthesis or combinatorial libraries presents a natural venue for development. However, these tools necessitate development of methods for rapid and quantitative application specific functionality probing and data integration. The latter are now being developed in the context of other combi-methods and more broadly big data applications in experimental sciences and are generally available. However, rapid and quantitative assessment of the functional behaviors is a required task for development of imaging tools addressing specific functionalities and the workflows integrating these functionalities with local structural, functional, and chemical imaging.

In-situ Experiments for Multiple Environmental Conditions

Materials in reactors are subjected to an extreme environment with multiple components that must be accounted for to understand and predict their behavior. Figure 12 captures the various components of a reactor environment. In all reactor designs and concepts, core components are subjected to irradiation, mechanical stress, and a corrosive environment, all at elevated temperature. Thus, understanding the effects of irradiation alone is insufficient to predict the behavior of materials in such a multi-dimensional environment. For example, creep occurs at high temperature and under stress, but is also strongly affected by irradiation. Corrosion rates are dictated by the environment and temperature, but they can be accelerated by irradiation of the component itself and by the production of reaction products in the coolant. Stress corrosion cracking (SCC) is a consequence of stress, temperature, and the environment. In reactor cores, irradiation assisted stress corrosion cracking (IASCC) emerges and it involves all four of the environmental components. Thus, understanding the behavior of reactor core materials *always* involves multiple components of the environment. The challenge is then to devise strategies and techniques to address this multi-faceted environment.

Experiments have been devised to study the various combinations of the environments in a reactor. Simple, gas-filled capsules containing TEM discs have been used for years to study microstructure effects and irradiation creep has been studied using pressurized tubes. These are simple designs that are generally easy to implement and have focused on the simultaneous effects of radiation, temperature, and stress. Data from these types of samples can span a wide dpa range with rather large intervals between sampling points due to reactor operating schedules. They are also subject to variations in parameters during operation. More sophisticated experiments that include the environment have been devised using flow loops through the core. Experiments have been conducted to measure the time to failure by SCC of samples under either constant load or dynamic straining. Experiments have also been constructed to measure crack growth rates in reactor. These experiments include the effects of radiation, temperature, stress, and the environment. The major difficulty of in-reactor experiments is that in-situ, real-time interrogation of the sample is generally not possible. The one exception is the measurement of crack growth rate during irradiation.
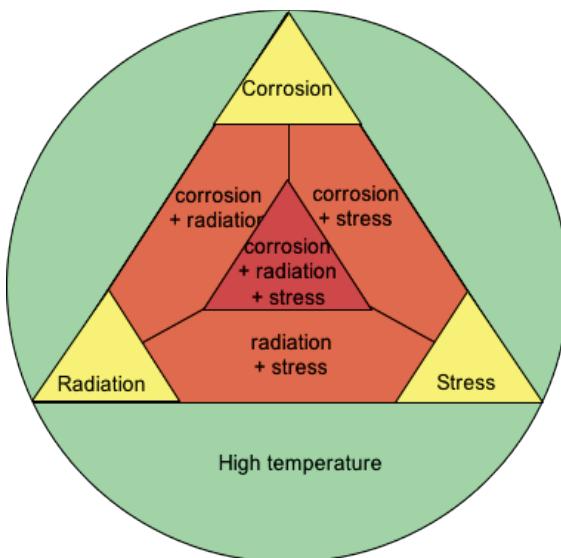
Figure 12. *The reactor environment includes four primary components that affect performance of materials; radiation, corrosion, stress and high temperature. Courtesy of Gary Was, University of Michigan.*

The use of ion irradiation as a surrogate for reactor irradiation experiments has undergone significant development over the past few years. This irradiation technique is more amenable to in-situ analysis of samples in a reactor-type environment. Accelerator-based irradiation experiments also enjoy a high degree of control of the environmental conditions that are not achievable in a reactor. For example, temperature, environment, and stress can be relatively easily maintained to within only a few percent of each parameter over the duration of the experiment. Such experiments have been used to study irradiation creep over a wide range of dpa, temperature, and stress [1]. More sophisticated experiments include the effect of the environment. For example, test cells have been constructed to study irradiation accelerated corrosion (IAC) in either a liquid metal [2] or high temperature water [3].

The coupling of an ion accelerator to a TEM has been used to study irradiation effects in-situ at high temperature [4]. Such techniques have been extremely useful in tracking the evolution of the irradiated microstructure with damage in real time. Dual ion irradiation systems (injection of He from one ion source simultaneously with damage from another) have been developed to visualize the effect of gas injection during irradiation. The possibility also exists to study corrosion and perhaps IASCC in-situ in the TEM using small, heated environmental cells with thin silicon nitride windows to provide a corrosive environment for a sample.

References

1. C. Xu, G. S. Was, "Proton Irradiation Creep of FM Steel T91," J. Nucl. Mater. 459 (2015) 183-193.
2. P. Hosemann, R. R. Greco, I. Usov, Y. Wang, N. Li, "The design, setup and operational testing of the irradiation and corrosion experiment (ICE)," J. Nucl. Mater. 376 (2008)392-395.
3. S. S. Raiman, A. Flick, O. Toader, P. Wang, N. A. Samad, Z. Jiao, G. S. Was, "A Facility for Studying Irradiation Accelerated Corrosion in High Temperature Water," J. Nucl. Mater. 451 (2014) 40-47.
4. M. L. Jenkins, Z. Yao, M. Hernandez-Mayoral, M. A. Kirk, "Dynamic Observations of Heavy-Ion Damage in Fe and Fe-Cr Alloys," J. Nucl. Mater. 389 (2009) 197-202.

Micromechanical Testing

Indentation based testing on materials of interest to the nuclear community has been carried out for decades. With the development of nanoindentation for the general material science community and the introduction of microcompression testing in 2004 by M. Uchic more insight about a material's properties on the micron scale has been obtained beyond conventional hardness testing. A large amount of work has been performed on non-nuclear materials bringing insight into the physics of deformation on metals, ceramics, and plastics. Today's advanced techniques allow one to understand the scaling phenomena on a basic scientific level as well as to obtain information on creep, plastic zone size, stress exponents, etc. While the conventional materials community made great progress utilizing these advances, it was not until ~2008 that these advances gained traction in the nuclear materials community.

Despite the short time that these techniques have been of interest to the nuclear materials community, great progress in science has been carried out by utilizing them. Today, techniques featuring micro-compression testing, micro-bend testing, and micro-tensile testing have been established and are beginning to be utilized on neutron and ion beam irradiated materials.

The driving force for deploying small scale mechanical testing in the nuclear materials area can be placed in two categories, direct engineering benefits and basic scientific advances. From a practical engineering point of view, small-scale mechanical testing enables one to handle more radioactive samples outside of the traditional hot cell environment while still obtaining insight into the mechanical performance of a material. In addition, to date these techniques are the only way one can obtain direct mechanical properties from an ion beam irradiated sample. Furthermore, having small scale probing available allows one to evaluate specific regions of interest in a material (e.g. welds) and enhance the statistics on the same volume of sample. From a basic materials science aspect, it has the potential to probe the radiation induced defect- dislocation interaction and isolate the individual phenomena so one can understand the true contribution to a mechanical property change associated with a specific defect. However, a true understanding of the materials mechanical property changes with specific radiation induced damage and the combination with specific and detailed modeling efforts has not been undertaken yet.

While great progress has been made in the last 5 years developing better and more accurate techniques and tools even enabling measurements at non-ambient conditions, several gaps still exist today. At this date, one can infer bulk property changes in a quantitative fashion using small scale mechanical testing and work is ongoing to correlate data with microstructure and ultimately, large scale mechanical test results. A full procedure and model bridging all relevant length-scales together does not exist at this point in time. Furthermore, the effects of different microstructural features such as grain boundaries, precipitates, dislocation density, etc. and their overall contribution to a full description of the materials behavior also does not exist. Surprisingly few studies have been conducted targeting individual mesoscale microstructural features, such as grain boundaries, which would be needed to understand the behavior of a bulk polycrystalline material. While efforts are ongoing, a significant amount of experimental and modeling work in this area is needed to truly understand the behavior of a large-scale polycrystalline irradiated material.

Another area that only recently has been studied is the effect of non-ambient conditions on a) scaling effects and b) the defect-dislocation interactions. Only two studies exist today hinting that the small scale mechanical testing scaling effects are a function of temperature and therefore the same parameters established for room temperature may not apply at elevated temperatures. It is hopeful that the emergence of new advanced tools will be able to address this complex issue in the future. Once these effects are understood, the community can start to understand how temperature effects dislocation/defect interactions produced from radiation damage. To fully understand the contributions that individual radiation damage

phenomena have on the mechanical properties of a material, fundamental separate effects experiments and a unifying model are needed on well-characterized sample materials.

An area that has not been addressed using small scale mechanical testing is the effect of a harsh corrosion environment. Since most nuclear components are immersed in water, steam, gasses, liquid metals or salts, this increases chance of phenomena like stress corrosion cracking, liquid metal embrittlement etc. These phenomena are not understood on a true fundamental level due to the complexity of the materials in question and due to the bulk approach. Small scale mechanical testing has the potential to contribute to the understanding of these phenomena due to in-situ capabilities as well as confined volume testing making modeling efforts simpler. However, due to experimental difficulties, these areas have not been addressed widely using these techniques and are now open to a wide range of opportunities.

In summary, small scale mechanical testing has been contributing to the fundamental understanding of materials mechanical properties and holds the promise to obtain fundamental insight into the detailed radiation induced defects behavior under deformation. Bridging overall length-scales in a combined modeling and experimental effort that addresses all relevant mesoscale microstructural features and incorporates non-ambient environments is still an area of research that needs to be addressed.

Analysis of Coolant and Material Chemistry

Combined experimental and computational probes have yielded unprecedented advances in our understanding of the atomic-scale controls on interfacial structure, dynamics, and reactivity. Yet, with few exceptions, our atomic-level understanding has not been extended to the extreme environments present in nuclear reactors and associated systems and there is reason to expect that these environments will display properties and reactivity not predictable using the current state of knowledge. Nuclear reactors and associated environments can contain high levels of reactivity in addition to elevated temperatures and pressures and extreme gradients in these that make it problematic to optimize material properties and predict their longevity in the reactor environment. Obtaining a fundamental understanding of the interfacial region under these conditions is critical in order to create a predictive capability that ultimately forms the locale where materials degradation occurs during corrosion and dissolution and is also where scale forms, i.e. the unintentional heterogeneous precipitation of solid phases. Probing the fluid-water interface is a substantial challenge for researchers and many of the classical or status quo assumptions about material properties start to break down at the nanometer scales that comprise typical interfacial region thicknesses. For example, the dielectric constant of fluids present at an interface is not represented well by the bulk fluid value.

Recently developed atomic-scale probes of fluid-solid interface behavior include synchrotron-based crystal truncation rod measurements that have been made of the three-dimensional structure of the mineral-water interface that have revealed coordination environments and adsorption behavior of dissolved species. Quasi-elastic neutron scattering measurements at the Spallation Neutron Source (SNS) at ORNL have revealed that the diffusional dynamics of water at the interfacial region are orders of magnitude slower than in bulk. Studies on nucleation of sparingly soluble salts using Grazing Incidence X-ray Scattering have allowed researchers to link specific solution and system properties to heterogeneous nucleation rates, important for predicting scale formation. Similarly, advances in atomic-scale simulations (both classical ab initio and molecular dynamics) have allowed the determination of reaction mechanisms, rate, and equilibrium constants of reactions occurring at interfaces, especially those too slow to be observed with direct simulation. A critical link to the experimental probes have been the improved abilities to calculate precisely what the molecular model predicts the results of the scattering experiments described above should be. This capability serves two purposes: 1) allows peak assignment and even uncertainty quantification in the experimental data; and 2) validation of the atomic-scale simulation. Overall, the combined experimental and computational toolsets are drastically improving our

knowledge of structure dynamics and reactivity at fluid-solid interfaces under ambient conditions. This in turn is leading to a more robust and accurate quantitative predictive capability for how the interfaces drive overall reactivity of the system.

A key knowledge gap, however, is how the extreme environments present in nuclear reactors affect fluid-solid interfaces and their influence on large-scale phenomena. Ionizing radiation may affect mineral-water interfaces by simply increasing the local temperature and hence the kinetic energy that drives reactions or by creating a host of highly reactive particles such as low-energy or solvated electrons and radicals that can all drive reaction chemistry, such as acidification. Bond breakage in solid phases, and in impurities in those phases, can lead to brittleness that opens new reactive surface area within the solid, which may be a self-reinforcing process. While radiation damage to bulk phases is well known, less understood is what role this plays at interface, but limited evidence exists that high radiation fields can drive substantial instabilities at an interface.

Non-Destructive Evaluation of Materials and Components

Non-destructive evaluation (NDE) is the science of materials characterizing for detecting and quantifying defects. Broadly speaking, most NDE techniques apply energy in one form or another (electromagnetic, acoustic, etc.) to the material under test and analyze the resulting response to determine the material condition.

While several NDE techniques (ultrasonic, eddy current, visual) are currently used to inspect materials and components in nuclear power systems and provide an assessment of materials reliability, they are generally applicable for detecting macroscopic cracks and flaws (inhomogeneities in the material), with the smallest reliably detectable flaw sizes on the order of ~1 mm. Recent advances in this area include the availability of simulation tools for understanding the inspection physics (interaction of the applied energy with the crack), new sensor designs (such as ultrasonic phased array sensors, laser ultrasonic systems, high frequency eddy current arrays, and high-sensitivity thermal imagers), new measurement parameters (such as nonlinear ultrasonic parameter), the application of machine learning and deep learning technologies for NDE data processing to improve detection capability, and model-based inversion techniques for flaw characterization (size and shape estimation). Techniques for uncertainty propagation that provide error bounds on simulation and inverse analysis results are also becoming common. Despite the advances, many technical gaps still exist in this area. These include several key areas listed below.

Measurement techniques and sensor concepts for in-situ microstructural characterization are a prime need. The development of such techniques will serve two purposes. First, increasing sensitivity to microstructural changes (such as segregation, precipitation, and grain growth) will improve the ability to detect precursors to cracks and other flaws, driving the detection threshold lower, and increasing the time available for mitigation actions. Second, such changes are a key element of several forms of materials degradation and aging, including void swelling, irradiation-induced hardening (and reduction of fracture toughness), and high-temperature creep and creep-fatigue. Currently, NDE techniques for these forms of degradation are still in the research phase and while several measurements have been proposed, such as ultrasonic wave speed and attenuation or electrical resistivity, the selectivity of these techniques to the material changes of interest is lacking. The underlying challenges include: increasing selectivity and sensitivity of NDE measurements to the changes of interest (for instance, void formation) while reducing sensitivity to other changes (for instance, carbide precipitate formation at grain boundaries, or residual stress in weld regions); identifying new properties/measurement targets for NDE measurements; generalizing correlative relationships to material classes irrespective of minor compositional variations; baseline-free NDE measurements (i.e., measurements that can be made and interpreted without the need for reference measurements, especially in a field environment); and compensation techniques to

compensate for the effects of common environmental variables (temperature, etc.) affecting measurement data.

NDE techniques for new materials and fabrication methods are also important. As current nuclear plants age, and advanced reactors are designed, new materials are seeing increased attention to their aging behavior. These include not only newer alloys (for instance, 9Cr-1Mo) but also materials such as concrete and electrical cable insulation, as well as fuel and cladding. The latter may require NDE techniques to assure its integrity in intermediate and final storage facilities. The behavior of these materials with temperature, dose, and in some cases coolant chemistry (singly or in combination) is unlike the behavior of classically studied nuclear structural materials and may require adapting existing techniques or the development of new techniques for in-situ assessment. New fabrication methods, such as advanced manufacturing, are also being investigated. Given the current state of technology and anticipated near term (5-10 years) developments, it is likely that inhomogeneities in the manufactured component will remain and drive materials performance. There is therefore a need for quality assurance of nuclear materials and components fabricated using these methods. There are still underlying challenges in addition to those identified in the previous bullet including techniques for post-fabrication examination of AM materials. Methods for in-situ monitoring of materials under fabrication would be ideal and would provide a mechanism for real-time control of the fabrication process.

Model-based interpretive inverse analysis must be considered. Simulation models for NDE typically are macroscopic in nature (length scales of mm and higher) and require as input information the material properties (elastic moduli, electrical conductivity, etc.). Sensor response models that operate on lower length scales (~100s of nm to 10s of um) will be needed to provide an interpretive understanding of the relationship between the microstructural changes (precursors) and the measured sensor responses. Such models may also act as a bridge between the micron-scale phenomena that drive the material property changes and NDE measurements that typically operate at larger length scales. Remaining challenges include: defining the physics coupling the measurement to the microstructure; fundamental data on material properties as a function of material phase (input to models); lab- and field-measurement systems that can monitor microstructure evolution with applied stressor (for verifying and validating models); uncertainty propagation in these models; transformation across length and time scales to connect NDE measurements at engineering scales to microstructural changes; and reconstruction algorithms that can estimate (with uncertainty bounds) microstructural features from measurements.

SPECIALIZED FACILITIES/RESOURCES

Nuclear reactor research has unique demands in dealing with highly corrosive media, radioactive samples, and high temperatures. Developments in testing and approaches to this work are enabling new science. This section will address these key facilities and recent advances with several key questions. *What is the state of the art in specialized facilities? How have they been used for nuclear energy? What will future and upcoming advances bring?*

Coolant Testing and Performance

The heat removal capability of a reactor coolant often dictates core design and reactor safety system features. For the existing generation of commercial nuclear reactors, large-scale non-nuclear experiments were often used to prove the design and safety of the existing commercial reactor fleet. Facilities were built both to directly simulate what would happen during certain accident events and to benchmark thermal hydraulic code systems used to prove the design and safety of these reactors. Advances in thermal hydraulic modeling and simulation techniques and the exponential increase in computational power will

help to minimize the number and scale of experiments needed to design and license the next generation of reactor systems. This should help minimize the time and costs associated with large experimentation. However, these simulations typically attempt to model phenomena at smaller and smaller length and time scales so benchmark experiments will require significantly more sophisticated and detailed experimental measurements in order to provide relevant data.

Advanced reactor designs operate at significantly higher temperatures than existing reactor systems and utilize coolants other than water, making many existing experimental facilities that were developed for light water reactors non-relevant for performing experimentation to support these new designs. The use of simulant fluids to perform scaled experiments should provide a lower cost alternative to building experiments that operate with prototypic coolants at prototypic system temperatures. However, selected experimentation will have to be performed using prototypic fluids and at prototypic operating conditions to provide sufficient confidence in predictions, and, in some cases (such as corrosion), are the only way to characterize coolant performance.

Experiments that incorporate prototypic fluids at typical operating conditions will require instrumentation and materials that can operate at high temperatures (>600°C) and survive in harsh environments. Advances in fluid and thermal measurement techniques such as optical temperature and strain measurements, ultrasonic flow and temperature measurement, laser based measurement techniques such as LDV and PIV, etc. provide technologies that can support the need for detailed benchmarking data. However, their use in advanced reactor environments will require additional effort to extend them to applicable operating temperatures and improve their survivability. Instrumentation capable of operating at high temperatures in general (pressure measurement, flow measurement, etc.) will need to be identified and proven in prototypic environments.

Advances in both static and dynamic seal technologies provide additional opportunities to develop experiments that are more reliable and versatile. However, these new seal designs will need to be proven under prototypic operating conditions to provide assurance that they are reliable under prolonged use. Similarly, advances in deep learning and artificial intelligence methodologies provide opportunities to better couple experimental results with thermal hydraulic code development. Advanced learning techniques should allow much more rapid code validation and more detailed quantification of code uncertainties.

Hot Cell Testing

Scientific understanding of nuclear material behavior requires that response to irradiation be measured and understood on both the macroscale and the microscale. The radiation emitted by these materials requires specialized hot cell facilities.

Measuring the irradiation-induced response of nuclear materials on the macroscale is essential for quantifying fuel swelling response, corrosion behavior, fission product transport, and identifying failure locations and failure modes. Measurements of these parameters have traditionally been made serially, in two dimensions, using contact measurements. Traditional measurements include visual examination, radiography, gamma scanning, corrosion layer thickness measurement, dimensional measurement, geometrical changes (e.g., swelling, bowing, and blistering), mechanical properties, and gas pressure measurement and analysis. Hot cell facilities include the Hot Fuel Examination Facility at Idaho National Laboratory (INL), the Irradiated Fuels Examination Laboratory at ORNL, and the Radiochemical Processing Laboratory at Pacific Northwest National Laboratory. These hot cells utilize manually operated remote manipulators and thick shield windows to perform sample manipulations. The measurement tools used are simple and purpose built for a specific specimen geometry (e.g., a fuel rod or

a fuel plate) because of the difficulty in engineering, reconfiguring, and maintaining complex systems remotely in a high radiation environment.

Current commercially available non-contact measurement technology and advances in tomographic data acquisition and image processing provide the opportunity to transition to new nondestructive examination methods that use parallel acquisition of multiple data types in three dimensions. Data acquired simultaneously from multiple sensors in three dimensions provide a much richer data stream for visualization, analysis, and for use in validating models. Noncontact methods do not require use of geometry-specific measurement systems; plates, rods, and cylinders can be measured with no change in configuration. Measurements can also extend to chemical analysis using techniques such as laser-induced breakdown spectroscopy. Material and fuel behavior under irradiation originates in events that occur at the atomic scale and it is important that atomic-scale damage processes be well understood. Revolutionary advances in materials characterization tools over the last decade now produce rich microstructural information across six orders of magnitude in length scale, allowing the microstructure and materials chemistry to be probed at the atomic scale. Routine use of these methods for the analysis of irradiated nuclear materials requires that they be coupled with hot cell facilities.

Neutron and photon-based scattering methods that probe the atomic structure of matter are also key materials science tools. Major national user facilities such as the SNS, High-Flux Isotope Reactor (HFIR), the Advanced Photon Source, National Synchrotron Light Source–II, and facilities at Stanford National Accelerator Laboratory provide top-level capabilities for x-ray scattering and imaging that accept very small quantities of irradiated materials. Photon and neutron-based characterization tools provide an opportunity to facilitate advances in the nuclear materials arena consistent with the rapid advances facilitated by these tools in other areas of material science.

Fuel Testing

Like irradiated structural materials, nuclear fuel also requires specialized facilities for testing. The irradiation behavior of fuels involves large changes in chemistry and microstructure caused by the production of fission products, in addition to the massive damage produced by heavy atoms at high energy. Although ion irradiation has been used to simulate aspects of displacement damage and gas bubble formation, there is currently no methodology for broadly assessing fuel behavior using ions.

Fuel testing and associated facilities, equipment, and techniques have not changed appreciably in the past 10 to 15 years, with some exception, although approaches to testing continue to evolve. The upcoming restart of the Transient Reactor Test Facility (TREAT) at INL, the Cabri reactor in France, and the startup of the Jules Horowitz Reactor (JHR) in France will make additional capability available for fuel irradiation testing with short, intense bursts of neutrons (TREAT, Cabri) and long-term irradiation testing in a modern research reactor (JHR). Otherwise, fuel irradiation testing is provided in the U.S. at the Advanced Test Reactor (ATR) and the HFIR and at any of several research reactors on college campuses, primarily the Massachusetts Institute of Technology Reactor. These facilities offer a range of irradiation testing configurations, from simple drop-in capsules to larger and more expensive tests in pressurized, flowing water loops, but testing is typically limited to neutron spectra dominated by thermal neutron energies. Fuel testing is also available, in varying degrees, at non-U.S. Facilities, most notably the Halden Reactor Project (HRP) in Norway (a thermal-spectrum, water-cooled reactor) and the BOR-60 sodium-cooled fast reactor in Russia. These irradiation facilities are used in conjunction with fuel fabrication facilities where test fuel is fabricated and shielded hot cell facilities where fuel test vehicles are disassembled after irradiation, examined non-destructively, and fuel samples prepared for post-irradiation characterization.

The nature of fuel irradiation testing is evolving as experimenters use focused tests to investigate specific details of fuel behavior and effects. Such approaches are increasingly used to obtain selected data to inform the development and validation of high-fidelity models of fuel behavior.

Other developments in fuel testing include the use of modern and evolving materials characterization techniques, as described above for detailed micro-structural and micro-chemical characterization of as-fabricated (i.e., representing the pre-irradiated condition) and irradiated fuel test samples. The radioactivity of actinide-bearing, as-fabricated samples and of irradiated samples requires the use of facilities designed and dedicated to preparation and characterization of radioactive materials, providing containment and shielding, such as available at INL (at the Hot Fuel Examination Facility, Electron Microscopy Laboratory, and the Irradiated Materials Characterization Laboratory) and ORNL (IFEL and LAMDA). Increasingly, the techniques applied make use of very small samples, which have lower radiation fields to require less shielding, but which are sufficient for characterization with modern micro-beam and nano-beam instruments.

Accident Scenario Testing and Analysis

Over the last decade, there have been considerable advances in accident scenario testing and analysis with a particular focus on LWR technology. R&D in this area accelerated following the reactor accidents at Fukushima Daiichi on March 11, 2011. A recent DOE gap analysis on LWR accident progression was conducted with the goal of identifying any data and/or knowledge gaps that may exist given the current state of severe accident research and augmented by insights gained from the Fukushima accidents. This study identified thirteen gaps in the areas of severe accident tolerant components and accident modeling. The results clustered in three main areas; namely, i) modeling and analysis of in-vessel melt progression phenomena, ii) Emergency Core Cooling System equipment performance under beyond-design-basis accident conditions, and iii) ex-vessel debris coolability and core-concrete interaction behavior relevant to accident management actions.

Based on these findings, DOE-NE launched a coordinated R&D effort to address some of the highest-level knowledge gaps. One key area relates to the performance of turbines used to drive emergency core cooling pumps under two-phase water ingestion conditions. This work has focused on developing and refining advanced computational models to better understand this process and to derive simpler correlations form turbine performance that can then be used in system-level severe accident analysis codes. Another key area relates the interaction of molten core debris with materials and structure within the reactor containment one the reactor pressure vessel has failed. Large-scale experiments are being conducted to provide phenomenological insights into ex-vessel core debris relocation and cooling behavior. This work has enabled advances in the understanding of materials, chemistry, and thermal hydraulics related to severe accident behavior and supports theory, modeling, and simulation in this area.

In-Situ Sensor Development

In-situ sensors may be used for monitoring processes or materials/components. An In-situ sensor measures the parameter of interest without interrupting the process or removing the material/component to the lab. Condition measurements are of materials being irradiated – structural materials, fuel.

In this section, we focus on sensors (for processes and materials assessment) for use in specialized facilities (such as test reactors), noting that advances in this area may be applied eventually in operational plants. Consequently, the need is for sensors that can be placed in or near the core and used for determining process and materials condition (generally in real-time) during a test campaign.

From a process measurement perspective, a challenge in test reactors is obtaining the necessary information on dynamics of system performance change (i.e., change in process parameters over time) due to irradiation/temperature/coolant exposure over longer test durations. These types of measurements require fast response times for sensors (to capture changes especially during the early portion of irradiation testing) and performance over extended lifetimes for the sensor (long-lived sensors are necessary to obtain data in longer test durations).

Currently, process monitoring uses many generally available sensors (thermocouples, resistance temperature detectors, pressure transmitters, ultrasonic and magnetic flow meters, flux wires, etc.). These sensors are generally able to meet measurement needs in specialized facilities such as test reactors. However, a key need is that the reliability of these sensors needs to be improved to meet increasingly challenging measurement needs (longer duration, higher temperature, higher flux/fluence tests).

Efforts in this area have generally focused on applying different measurement physics for measuring process parameters. For instance, ultrasonic and optical (fiber optic) methods are being developed for temperature measurement in high-temperature applications. Johnson Noise Thermometry (JNT) is another approach that has been investigated for this purpose. Ultrasonic, optical, and electromagnetic transduction methods have been also investigated for measuring pressure and flow. While generally applicable, these methods can suffer from lower sensitivity in certain domains, the potential for sensor drift, and interference from other factors (such as electromagnetic interference for JNT). For some of the sensor types (such as selected fiber optic materials or piezoelectric materials), radiation tolerance in an LWR spectrum has been demonstrated; it remains to be seen if these results hold when dealing with a fast neutron spectrum and higher fluences.

There are many other aspects with in-situ sensing that are seeing R&D currently but which still have gaps. Sensor calibration may drift over time and online monitoring techniques are of interest not only for detecting such drift but also for online recalibration. Monitoring the chemistry (and change in chemistry) of the coolant or test component is also a need. While fiber optic sensors can be used for sensing hydrogen gas, monitoring dissolved hydrogen and oxygen concentration in the coolant is still a gap and electrochemical sensors may be one approach to addressing this need.

With respect to in-situ materials monitoring, there is much research in non-nuclear applications. In nuclear environments, sensor survivability is the key challenge to obtaining interpretable data. The section on NDE explains the measurement needs for such sensors and includes microstructural variations, selectivity and sensitivity.

In this space, sensors for condition monitoring mostly rely on ultrasonic sensing mechanisms, with high temperature and rad tolerant sensors being researched. Sensor materials that can tolerate these environments have typically low sensitivity (whether they be piezoelectric or magnetostrictive materials).

General challenges in in-situ sensing in specialized facilities include:

- Sensor materials selection/design for improving reliability and survivability especially in fast spectrum scenarios.
- Improved sensitivity and response times to process and test material condition changes.
- Online calibration (monitoring and recalibration of drifting sensors).
- Process measurements – limited access issues for flow, temp, pressure. New sensors that are being developed are sensitive to multiple factors – how to deconvolute multiple effects?
- Condition monitoring of test samples – Same issues as with NDE, for the most part. The major problem is one of interpreting the measured data and quantifying the uncertainty in the measurement.

- Condition monitoring of components/facility. While process measurements are often a reasonable approach for identifying when the facility shifts into an off-normal operational condition, other methods of assessing when the facility is moving into an off-normal situation may be useful. For instance, monitoring equipment vibration and current/voltage may provide early warning of potential problems that can be mitigated easily, instead of waiting for issues to manifest themselves as changes in process data.
- Data and power requirements for in-situ sensing are a challenge. As more sensors are developed and deployed, especially in areas where physical access is limited, methods for conveying power to sensors and data back from the sensors without using cables will be needed. A related challenge is whether parameters in regions where sensor access is not possible can be inferred from other measurements (virtual sensors), perhaps using techniques leveraged from machine learning/deep learning.
- Uncertainty quantification in the measured and in inferred parameters is a challenge and will need to be addressed if the measured data is to be used for any form of decision-making.

COMPUTATIONAL TOOLS

Computational tools have made remarkable gains in recent years. High performance computing has enabled higher resolution simulations on increasing larger scales, both time and spatial. These outcomes are providing new means of exploring fundamental mechanisms, but also enabling evaluation of performance scenarios. In each subtopic, several key questions will be answered. *What are the latest advances? How have they been used for nuclear energy? What will future and upcoming advances bring?*

Radiation effects in fuels and materials

To provide fundamental physical understanding of radiation-induced microstructural evolution, multiscale materials modeling has experienced many recent advances. For example, an impressive ability to capture the multiscale nature of radiation effects on body-centered cubic structural materials, at least in terms of the evolution of the interstitial defect clusters and sub-visible vacancy cluster distribution in neutron irradiated iron, has been recently articulated [1-6]. Additionally, impressive modeling results have demonstrated the ability to predict radiation-induced precipitation in both steels and binary alloys, using atomistically-informed phase field models [7-12]. Opportunities exist to combine in situ irradiation experiments with such modeling which provide a unique opportunity to examine the fine details of defect evolution under continuous irradiation and to verify fundamental assumptions/uncertainties with respect to damage production and kinetics/energetics of irradiation defects.

While the aforementioned results provide a good agreement with the experimental observations, there are a number of assumptions inherent in these modeling results which merit further investigation. First and foremost is the assumption of the isotropic three-dimensional reaction kinetics that underpin the Smoluchowski reaction rate constants of defect interactions. Several groups have begun to investigate this assumption in detail [3,4,13]. However, the mathematical formulations rapidly become quite complex within models that have multiple diffusing species, as is necessary to treat the prismatic interstitial loop behavior in real-world materials containing impurities and for which the resulting migration behavior is presumably determined by trapping and de-trapping interactions. Thus, there is a need for additional, high-fidelity experimental characterization of the defect cluster microstructure in irradiated materials with irradiation temperatures ranging from below Stage III to near Stage V in order to validate the defect cluster diffusion kinetics and reaction rate constants that are inherent to the reaction-diffusion cluster dynamics, or object kinetic Monte Carlo modeling approaches.

Future modeling and experimental studies are also needed to further understanding of the interactions amongst defect clusters and with extended microstructural sinks. For example, there remain numerous unresolved questions about defect cluster evolution as the result of cascade overlap and about the quantitative details in how we treat the reaction rates between defect clusters and various microstructural sinks, including dislocations, grain boundaries, nanometer-sized precipitate interfaces, as well as reactions amongst the defect clusters themselves. The quantification of the sink efficiency of dislocations, free surfaces or grain boundaries with radiation dose and how variations of defect structures over time may relate to the absorption of vacancy versus interstitial defects, is another example of an outstanding question that has been postulated to be responsible for the swelling response of irradiated materials. It is possible that many aspects of this research could benefit from the use of machine learning techniques, as recently used for assessing solute transport in FCC alloys [14].

Most of the recent modeling successes are limited to rather small radiation damage levels below a few displacements per atom. As noted by Li and co-workers in describing the experimental observations in Mo [15], and by Kaoumi and co-workers in the ferritic-martensitic alloys [16], at higher doses a pronounced rafting or spatial arrangement was observed in the visible defect cluster distributions. This has not been captured within the current models, which are limited to one spatial dimension, and for which elastic interactions amongst the defect clusters and other microstructural features are not included. Clearly this is another need for coordinated modeling and experimental studies in the future. It can be further supposed that the observed dislocation loop rafting behavior may be responsible for the saturation of the mechanical properties that is observed at higher doses in structural materials. Unraveling this will require a combination of spatially resolved, e.g., tomographic, studies of the evolution of the defect cluster distributions across a range of radiation exposures and temperatures, coordinated with extensions of the multiscale modeling methodology to specifically investigate the role of elastic interactions amongst defect populations to describe such higher dose behavior.

Finally, the radiation defect evolution modeling results have focused almost entirely on the evolution of vacancy and interstitial type defect clusters in irradiated materials, without any consideration of the coupling of the defect and solute/impurity fluxes. There is long-standing literature on the radiation-induced segregation in austenitic alloys, as well as more recent work on radiation-induced segregation in ferritic-martensitic materials. The coupling the defect and solute fluxes that occur under irradiation to thermodynamic predictions of radiation-induced phase formation is an area which is receiving increasing attention. Seamlessly integrating the evolving defect physics with the radiation-induced segregation and changes to the second phase precipitates represents a true grand challenge within the radiation effects community.

References

1. B.D. Wirth, X. Hu, A. Kohnert, and D. Xu, "Modeling Defect Cluster Evolution in Irradiated Structural Materials: Focus on comparing to high-resolution experimental characterization studies", Journal of Materials Research 30 (2015) 1440-1455.
2. A.A. Kohnert and B.D. Wirth, "Cluster dynamics models of irradiation damage accumulation in ferritic iron. II. Effects of reaction dimensionality", Journal of Applied Physics 117 (2015) 154306
3. A.A. Kohnert and B.D. Wirth, "Cluster dynamics models of irradiation damage accumulation in ferritic iron. I. Trap mediated cluster diffusion", Journal of Applied Physics 117 (2015) 154305
4. X. Hu, D. Xu, T.S. Byun, and B.D. Wirth, "Modeling of Irradiation Hardening of Iron after Low Dose and Low Temperature Neutron Irradiation", Modeling and Simulation in Materials Science & Engineering 22 (2014) 0655002
5. D. Xu, G. VanCoevering and B.D. Wirth, "Defect Microstructural Equivalence in Molybdenum Under Different Irradiation Conditions at Low Temperatures and Low Doses", Computational Materials Science 114 (2016) 47-53

6. A.A. Kohnert and B.D. Wirth, "Grouping techniques for large-scale cluster dynamics simulations of reaction diffusion processes", Modelling and Simulations in Materials Science & Engineering 25 (2017) 015008

7. D. Xu, A. Certain, H.-J. Lee Voigt, T. Allen, and B.D. Wirth, "Ballistic effects on the copper precipitation and re-dissolution kinetics in an ion irradiated and thermally annealed Fe-Cu Alloy", Journal of Chemical Physics 145 (2016) 104704

8. J.H. Shim, E. Povoden-Karadeniz, E. Kozeshnik, and B.D. Wirth, "Modeling precipitation thermodynamics and kinetics in type 316 austenitic stainless steels with varying compositions as an initial step toward predicting phase stability during irradiation", Journal of Nuclear Materials 462 (2015) 250-257

9. H. Ke, P. Wells, P.D. Edmondson, N. Almirall, L. Barnard, G.R. Odette, D. Morgan, Thermodynamic and kinetic modeling of Mn-Ni-Si-rich precipitates evolution in low-Cu RPV steels, (Submitted to Acta Materialia) (2017).

10. J. Gan, G.S. Was, R.E. Stoller, Modeling of microstructure evolution in austenitic stainless steels irradiated under light water reactor condition, Journal of Nuclear Materials 299(1) (2001) 53-67.

11. M. Mamivand, Y. Yang, J. Busby, and D. Morgan, Acta Mat 130 (2017) 94-110.

12. X. Zhang, J.A. Beach, M. Wang, P. Bellon, and R.S. Averback, Acta Materialia 120 (2016) 46-55.

13. A.V. Barashev, S.I. Golubov, Y.N. Osetsky, and R.E. Stoller: Reaction kinetics of non-localized particle–trap complexes. Phil. Mag. 90, 897 (2010).

14. H. Wu, A. Lorenson, B. Anderson, L. Witteman, H.T. Wu, B. Meredig, and D. Morgan, Computational Materials Science 134 (2017) 160-165.

15. M. Li, M.A. Kirk, P.M. Baldo, D. Xu, and B.D. Wirth, Phil. Mag. 92(16), 2048 (2012).

16. D. Kaoumi, J. Adamson, and M. Kirk, J. Nucl. Mater. 445, 12 (2014).

Computational tools for material design

Materials with ultrahigh radiation resistance are desired for next generation nuclear reactors. The leading approach to enhance material radiation resistance is to produce high density of point defect recombination centers (sinks) in microstructure such as the oxide nanoclusters in the ODS. Recent advances of computational thermodynamics and kinetics modeling tools on phases enabled the accelerated design and development of naturally formed nanoprecipitate-dispersed steels. These pre-existing nanoprecipitates will be subjected to complex chemical and structural changes during irradiation. The precipitation evolution is a concern not only to newly developed nanoprecipitates-strengthened steels and alloys, but also to traditional structural materials such as austenitic stainless and reactor pressure vessel (RPV) steels. The precipitates either formed before or after irradiation greatly impact not only radiation resistance but also the mechanical properties.

To understand precipitate evolution in irradiated materials, we need to understand the baseline thermodynamic stability and kinetics of the system under thermal conditions. The current computational thermodynamics and kinetics modeling tools are well suited for this purpose. Irradiation effect comes into the picture by changing local chemistry and diffusivity, resulting in the change of local thermodynamic stability and kinetics. A computational framework that takes both the thermal effect and irradiation effect is schematically shown in Figure 13. In this framework, the data structure is divided into three levels. The first level is the database, which stores the Gibbs energy of phases, the elemental diffusivity in phases and defect-solute interaction energy. The second level is calculation engine, which is integration of modular modeling of different phenomena, including thermodynamics and kinetic modeling of phase under thermal conditions, radiation induced defect evolution, radiation enhanced diffusion, radiation induced segregation and radiation induced phase transformation such as precipitation or dissolution. The third level is the output results on the essential information about irradiated materials microstructure such as microchemistry at defect and in bulk, and phase type, amount, size, distribution and composition. These outputs can be used for future environmental and mechanical property simulation. Recent work showed early promising application of such integrated framework to simulate microchemistry evolution at grain boundary. Key challenges identified from this work include the lack of critical parameters, such as solute-defect interaction energy, etc. The challenge to future work would be the in-situ integration of modular

modeling to handle different phenomena, such as defect evolution, solute/defect diffusion, radiation induced segregation and phase transformation such as precipitation and dissolution, etc.
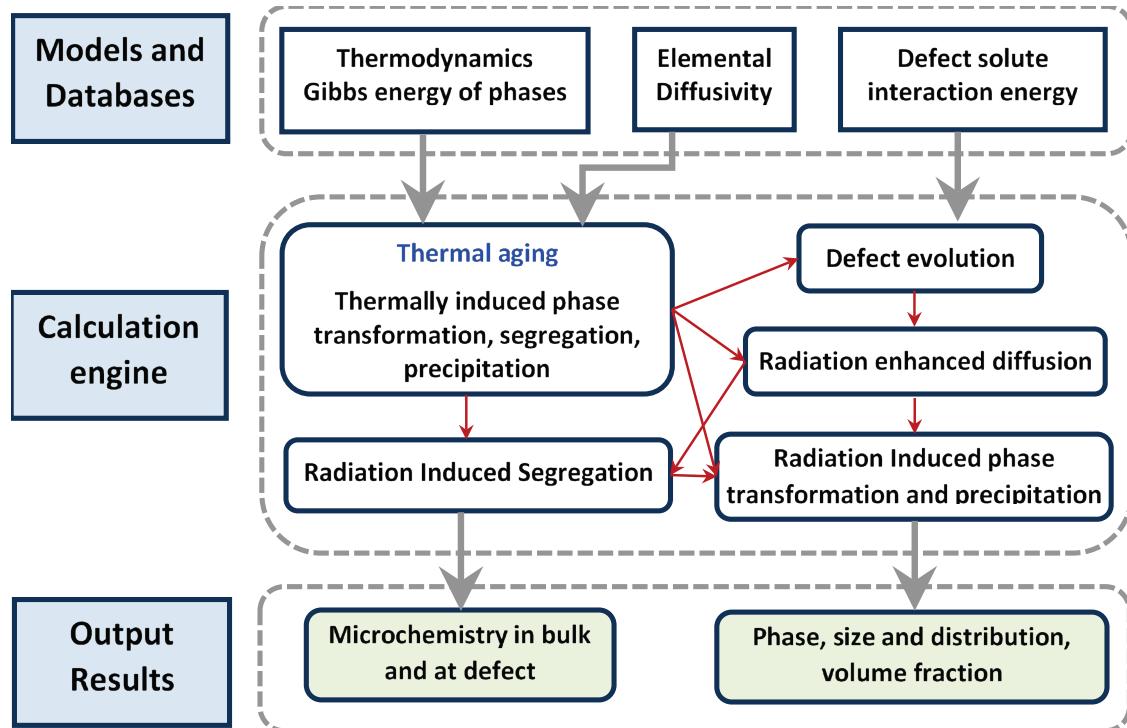
Figure 13. Schematic diagram showing integrated modeling framework on precipitate evolution in irradiated materials. Courtesy of Ying Yang, Oak Ridge National Laboratory.

References

1. Y. Etoh and S. Shimada, , JNM, 200(1993) 59-69
2. L. Tan, Y. Katoh, L. Snead, JNM, 445(2014) 104-110
3. Z. Jiao, G.S. Was, Acta Mater., 59 (2011)1220-1238

Computational tools for component performance design

Developing usable computational tools for nuclear energy applications requires leveraging advances in computer hardware, modern software frameworks, computational algorithms, and advances in understanding of the fundamental mechanisms driving the phenomena being simulated by these tools. Significant advances have been, and continue to be made in all of these areas individually, as well as in the integration of these advances to provide usable computational tools with improved capability to predict complex phenomena of interest for nuclear applications.

Over the last decade, there has been a steady increase in the use of multicore architectures, both at the high end, where the most powerful supercomputers are driven by hybrid architectures that combine multicore and graphics processing units (GPUs), to the low end, where machines with parallel computing capabilities are becoming ubiquitous and far more accessible than they were just a few years ago. As of 2017, there are well over 100 computers in the world with speeds over 1 petaFLOP, and there are significant efforts in multiple countries to develop the infrastructure necessary for exaFLOP scale computers.

Taking advantage of this ever-increasing computational capability in both high-end capability machines, as well as in production-oriented capacity machines requires significant software infrastructure. Modern clusters of multicore processors require a hybrid of message passing and thread-based parallelism for maximum efficiency. Systems based on GPUs require the use of architecture-specific programming, which is a significant obstacle in the way of more widespread adoption of this technology. At the lower end, it is increasingly necessary to take advantage of parallel processing for increased application speed. Recent developments in open source multiphysics simulation frameworks and solvers are making this modern hardware far more accessible to physics tool developers, allowing them to focus on their problem domain. Rather than developing an entire infrastructure for a specific application, developers can use shared libraries that deal with the complexities of these systems.

A wide variety of modeling approaches are needed to take into account the multitude of physical phenomena that ultimately affect problems of interest, such as nuclear fuel performance or structural component degradation. Significant algorithmic improvements are being made at all levels, from atomistic to continuum scales. Much progress has also been made to integrate these advances in specific fields into unified simulation platforms that allow making use of them nuclear applications. However, there is still much to be done toward the goal of developing simulation tools that connect bottom-up models of the fundamental physical phenomena at lower length and time scales to engineering models that can be used with confidence to improve the safety, reliability, and efficiency of nuclear reactors.

For a model to be used with confidence, it must faithfully represent the physical phenomenon it is intended to represent. Computational model development requires experimental data, both to provide a better understanding of the mechanisms of interest, and to validate the models once they are developed. As computational hardware and software enable simulations with ever increasing fidelity, care needs to be taken to ensure that these simulations accurately reflect physical reality. There is a wealth of useful validation data available for nuclear materials and systems. However, as simulation tools increase in their ability to predict the behavior of these systems, they are often able to predict details that are not adequately captured in existing experimental data. To improve confidence in these high fidelity simulation tools, experimental data with similar fidelity, both in the form of integral experiments and separate-effects experiments designed for validation of specific aspects of computational models, are very much in demand.

SUMMARY

Nuclear power represents a significant portion of the world's non-carbon emitting power generation. Most of this generating capacity is based on water-cooled reactor designs. A number of advanced nuclear energy system concepts have been developed to provide improved efficiency, greater fissile fuel utilization, reduced high level waste generation, and increased margins of safety over today's water-cooled systems. However, there are a number of technical challenges that must be addressed to enable the broad deployment of these systems from concept to the electrical grid. Given the desire for commercial deployment starting in 2030, approaches that can accelerate the development and qualification of materials are needed.

The development and deployment of advanced reactors requires research and development of materials and understanding the chemistry of coolants, fuels, and interactions. There are several common areas for key materials that must be overcome to enable advanced nuclear energy systems. Coolant compatibility is a key limitation for many advanced concepts, although the specifics vary by coolant and material choices. The impacts of high temperature, mechanical stress, and irradiation environments also pose a key challenge to deployment. The mechanistic understanding of these individual extreme elements and combined environment is needed. The mechanisms for interfacial processes are essential for prediction of degradation and design of tolerant materials.

Similar limitations for nuclear fuels exist. There are key degradation issues that must be resolved for both thermal and fast spectrum reactor fuels. Fuel forms for gas and molten salt cooled reactors have unique issues due to the very high operating temperatures and coolant interactions that must be resolved. Interfacial understanding is essential to predict and mitigate key interactions (e.g. clad and coolant; fuel and coolant; and/or fuel and clad).

Fortunately, in the last decade, there have been substantial advances in the tools and techniques available to resolve these key limitations. Fuels and materials characterization is being enabled by new developments in techniques such as atom-probe tomography, electron microscopy, and neutron scattering. Unraveling the combined effects of irradiation, coolant interaction, and/or temperature has been a challenge for decades, but new understanding and testing approaches are providing new paths to develop and qualify new materials. Non-destructive testing techniques are proving to be very valuable in translating operational and laboratory experience into fundamental understanding. Finally, computational techniques and tools provide new opportunities to couple experimental and theoretical understanding and to predict performance in service. All of these tools will be required to enable advanced nuclear energy systems.

Disclaimer

Image courtesy of Oak Ridge National Laboratory.

U.S. DEPARTMENT OF **ENERGY** | Office of Science