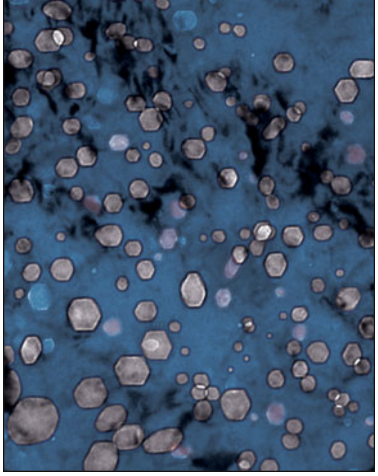

Radiation-induced defects such as voids restrict materials choices for fourth-generation reactors.

Materials hurdles for advanced nuclear reactors

By **Arthur L. Robinson**
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The International Atomic Energy Agency (IAEA) lists 440 operating nuclear plants in 31 countries providing 11% of the world's electricity. Many of these have been in service for 40 years or more, and relicensing to extend their lifetimes another 20 years is ongoing. But to continue providing reliable base-load electricity to satisfy the world's ever-growing energy appetite, the nuclear fleet will eventually have to be replaced and expanded. Generation IV (GEN-IV) nuclear reactors, foreseen to gradually come into service after 2030, promise improved efficiency, safety, and proliferation resistance, along with longer lifetimes and less radioactive waste. There is a hitch, though—many of the materials necessary to build them still need to be identified.

Most reactors in today's nuclear landscape are second- and third-generation designs dominated by light water (pressurized and boiling) reactors (LWRs). While this LWR technology offers a well-developed alternative to fossil fuels and their greenhouse-gas emissions, standing still is not a path to future success. "To continue to be viable, nuclear technology must continuously improve," said Steven Zinkle of the University of Tennessee, Knoxville. "It's just like the auto industry's progression toward better manufacturing efficiency, fuel economy, and safety."

Rudy Konings, of the Institute for Transuranium Elements at the European Union's Joint Research Centre in Karlsruhe, points to the LWR's once-through fuel cycle that taps less than 1% of uranium's energy content, so that depending on the future growth of nuclear power, uranium resources could be depleted by the end of the century. On the public opinion's side, observers mention that rare reactor failures and the need to safely store highly radioactive waste for many millennia can shake consumer confidence.

In the face of the aging reactor fleet, these and other concerns have steered the nuclear industry toward the six GEN-IV reactor designs now under development around the world under the umbrella of the 13-member GEN-IV International Forum (GIF). In 2002, GIF selected six reactor concepts on which to concentrate: Gas-Cooled Fast Reactor (GFR), Lead-Cooled Fast Reactor (LFR), Molten Salt Reactor (MSR), Supercritical Water-Cooled Reactor (SCWR), Sodium-Cooled Fast Reactor (SFR), and Very High Temperature Reactor (VHTR). These are not simply evolutionary versions of advanced LWRs. Of the GEN-IV designs, SFR and VHTR are closest to fruition.

Each GEN-IV reactor has its own set of operating environments and associated challenges, but collectively the materials will face unprecedented combinations of higher radiation levels, higher temperatures, and new corrosion avenues associated with various coolants. Reactor fuels are at risk as well as structural materials. "GEN-IV fuels are operated to high burn-up and at high power, and under these extreme conditions, substantial amounts of fission products aggregate in fuel pellets, causing swelling and possibly leading to breach of the protective cladding," said Konings.

While high neutron radiation is a unique environment for reactors, it is the combination of challenges that are responsible for the emergence of materials degradation modes that are not problematic in LWR materials, such as swelling due to void formation and embrittlement by radiation-induced precipitation in reactor core materials. To make things worse, the effect that radiation damage has on materials properties is also highly temperature dependent. Mix in a long list of metallurgical failure mechanisms that interact with each other in these challenging conditions, and materials selection seems daunting, indeed. Fortunately, "five decades of experience with LWR materials offers a wealth of clues about where to look and not to look," said Zinkle.

Steel alloys occupy a prominent place in any list of candidate materials, except at the highest temperatures where ceramic-based materials such as silicon-carbide composites may be needed. David Petti of Idaho National Laboratory said, "there are some steels that can be used for sodium fast reactor systems and some nickel-based alloys that can be used for high-temperature gas-cooled reactors." But "rather than using the austenitic steels used in today's fast reactors," said Stuart Maloy of the Los Alamos National Laboratory, "ferritic-martensitic stainless steels are strong candidates for the cladding in GEN-IV fast reactors." With the addition of chromium, the material exhibits corrosion resistance and high-temperature stability. It is attractive for its resistance to void swelling, although it is susceptible to radiation hardening and embrittlement.

Structuring at the nanoscale may help. Oxide-dispersion-strengthened steels (ODSs) were originally developed in the 1970s, continuing later in Japan, and are now widely studied in the United States and Europe. "They are transformational materials because of their radiation resistance," said G. Robert Odette of the University of California, Santa Barbara. In the radiation-intense environment of a reactor core, helium generation can be a problem by aggregating into bubbles and causing embrittlement. Nano-sized yttrium-oxide precipitates uniformly dispersed

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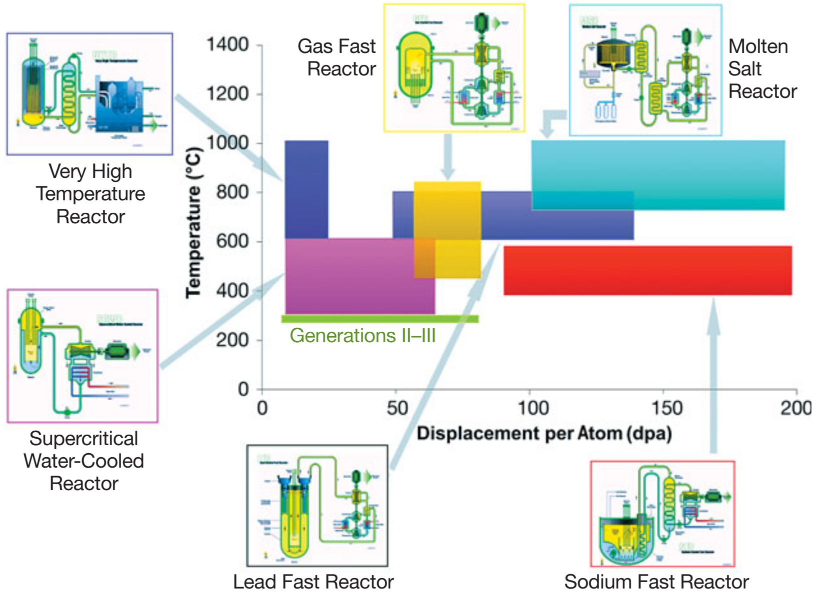
https://doi.org/10.1557/mrs.2015.146 Published online by Cambridge University Press



Range of temperatures and cumulative neutron radiation damage for the six GEN-IV reactor designs. Displacements per atom refers to the number of times an atom will be knocked out of its lattice during its service life. Figure courtesy of G. Was, University of Michigan.

through the matrix of ferritic steels can trap small helium bubbles before they grow large enough to be troublesome.

In the last 15 years, processing improvements such as solute additions and heat-treatment modifications have made the distribution 100 times finer, thereby improving the radiation resistance, raising the temperature tolerance by 200°C, and providing some fracture toughness. But "inexpensive methods to manufacture and weld the thin-walled tubing needed for fuel cladding remain a challenge," said Maloy, who estimated that on a technical-readiness scale, ODSs are almost halfway toward being reactor-ready.

Another way to make steel more radiation-tolerant, while maintaining or improving the temperature- and corrosion-resistant properties, is to increase the grain-boundary area. "Neutron irradiation generates vacancy–interstitial pairs, which are mobile at high temperature, so that vacancy clusters can form voids, except at defects like grain boundaries where they are annihilated before they can cluster," said K.L. Murty of North Carolina State University. "With materials having ultrasmall nano-sized grains, the grain-boundary area is larger, and the radiation resistance is enhanced."

In addition to adding grain boundaries, grain-boundary engineering can help to achieve a structure robust enough to serve in GEN-IV reactors. Todd Allen of the University of Wisconsin–Madison and the Idaho National Laboratory gave a corrosion example: "Diffusion along some kinds of grain boundaries can make stainless steels more susceptible to corrosion, but processing the material to minimize the percentage of the more harmful grain boundaries relative to others can improve corrosion resistance."

Before a regulatory agency will issue an operating license for a reactor, the materials in it must meet standards established by an organization such as the American Society of Mechanical Engineers. "Meeting these standards requires a comprehensive database of test results demonstrating the material can maintain its integrity under the operating conditions it must endure for as long as it must endure them," said Robin Grimes of Imperial College London.

Test facilities are a limiting factor for materials qualification because they are in short supply when it comes to providing realistic environments. "We have only a very few test stands that provide a prototypic coolant environment, such as sodium or helium gas, to study environmental effects on materials," said Petti. "For irradiation testing, fast neutrons are needed for fast reactor fuel cladding and in-core structural materials, and this capability does not exist in the United States." There are very few operating fast reactors, such as BOR-60 in Russia, and using them is a time-consuming process with difficult accessibility.

One alternative is to place materials in existing test reactors that primarily produce a thermal neutron spectrum but include some fast neutrons in the high-energy wing of the distribution. Because the fast-neutron flux is so low, ion-beam irradiation is an alternative. In addition to higher radiation-damage rates, subsequent sample characterization is possible without the need for radiation protection. "We can use dual or triple beams to study helium and hydrogen generation while creating damage with heavy ions, thereby reproducing what happens with neutrons," said Pascal Yvon of the Commissariat à l'Énergie Atomique's Nuclear Energy Division in Saclay, France. On the down side, the radiation effects for heavy ions occur in a thin surface region, making extrapolation to bulk materials more difficult.

Because of the dearth of test facilities, theoretical modeling is a key part of the materials-development process, allowing verification of hypotheses and guiding the development process from the nano- to macroscale. "To go from understanding how damage occurs at the atomic level to understanding the consequences at the component level, we use the output from the lower as input for the next higher level and work our way up the ladder," said Yvon. However, to satisfy the regulators, you still need to do some testing to see if what you predicted to happen actually happens.

While the GEN-IV materials challenges are vast, Grimes summed up, "it is important to remember that the first LWR materials were not perfect, but they improved over time. The first GEN-IV reactor materials will not be exposed to the most extreme conditions, and we won't be building hundreds of reactors at the start. It will be a more gradual process." □

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