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Review

Materials challenges for nuclear systems

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The safe and economical operation of any nuclear power system relies to a great extent, on the success of the fuel and the materials of construction. During the lifetime of a nuclear power system which currently can be as long as 60 years, the materials are subject to high temperature, a corrosive environment, and damage from high-energy particles released during fission. The fuel which provides the power for the reactor has a much shorter life but is subject to the same types of harsh environments. This article reviews the environments in which fuels and materials from current and proposed nuclear systems operate and then describes how the creation of the Advanced Test Reactor National Scientific User Facility is allowing researchers from across the United States to test their ideas for improved fuels and materials.

Successful operation of current light water reactors and implementation of advanced nuclear energy systems is strongly dependent on the performance of fuels and materials. A typical Light Water Reactor (LWR) contains numerous types of materials (Fig. 1) that must all perform successfully. A majority of the LWRs in the U.S. are extending their operating licenses from a 40 year period to a 60 year period, with initial discussions about 80 year

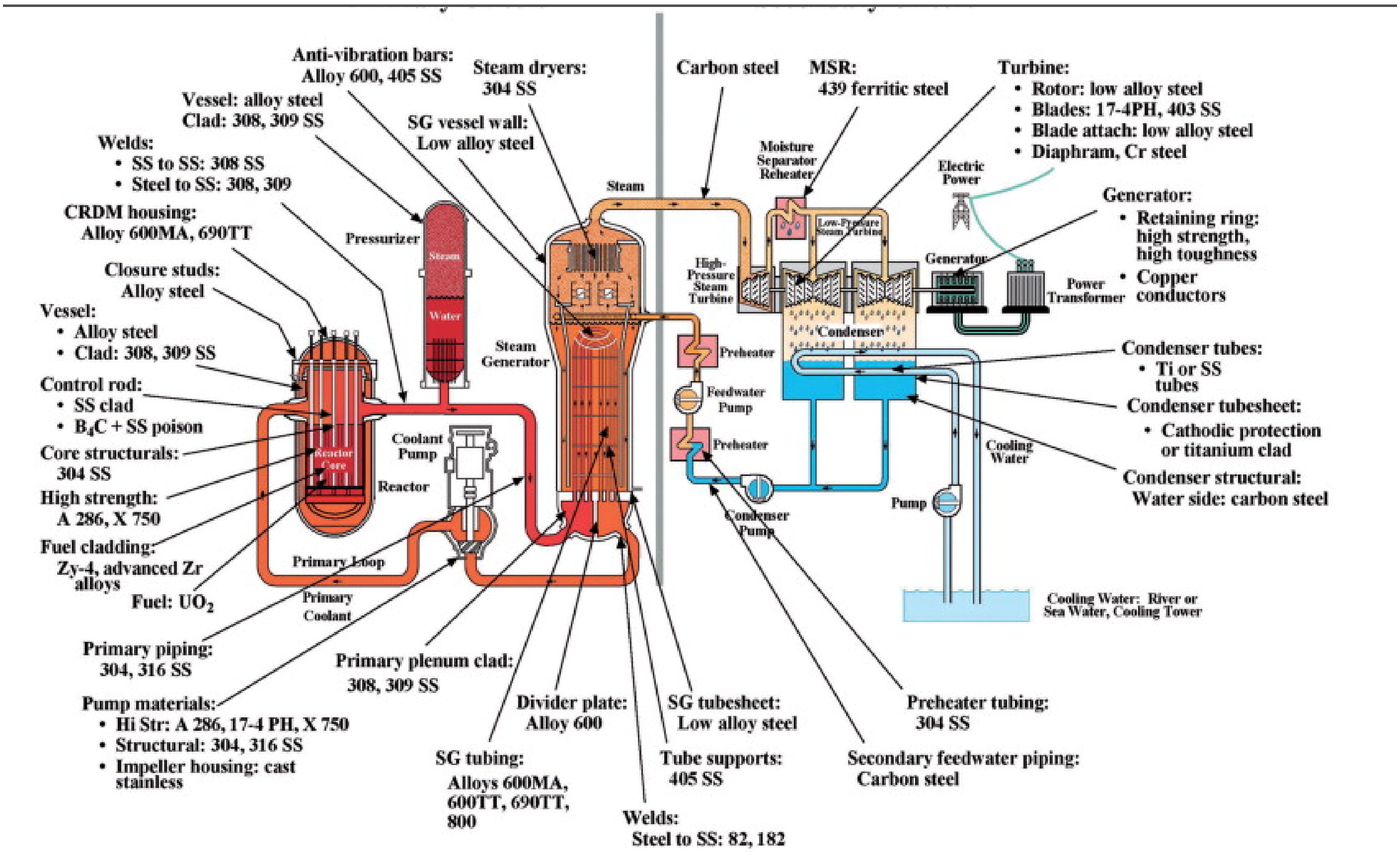


Fig. 1. Outline of PWR Components and Materials.

Courtesy of R. Staehle.

Table 1 summarizes the expected environments during normal operation for the six Generation IV systems. For comparison, the operating conditions for a Pressurized Water Reactor (a type of light water reactor) are also listed. The Generation IV systems are expected to operate at higher temperatures, to higher radiation doses, at higher pressures, and in some cases with coolants that present more challenging corrosion problems than current LWRs. Generation IV systems are expected to operate for at least 60 years.

Table 1. Approximate operating environments for Gen IV systems

Very High Temperature gas-cooled Reactor (VHTR)	600	1000	1-10	7	Helium
Sodium-cooled Fast Reactor (SFR)	370	550	200	0.1	Sodium
Lead-cooled Fast Reactor (LFR)	600	800	200	0.1	Lead
Gas-cooled Fast Reactor (GFR)	450	850	200	7	Helium/SC CO_2
Molten Salt Reactor (MSR)	700	1000	200	0.1	Molten Salt
Pressurized Water Reactor (PWR)	290	320	100	16	Water

*

dpa is displacement per atom and refers to a unit that radiation material scientists used to normalize radiation damage across different reactor types. For one dpa, on average each atom has been knocked out of its lattice site once.

For existing LWRs, extending the lifetime of each fuel element would improve the energy extraction from the fuel, limit the total amount of unused fuel (approximately 95% of the energy content remains at the end of the current useful life of a typical LWR fuel pin), and improve the overall economics of the plant. For many of the proposed advanced systems, specifically the fast spectrum systems like the Sodium Fast Reactor (SFR), Lead Fast Reactor (LFR), and Gas Fast Reactor (GFR), advanced fuel forms purposefully contain fission products from previously used fuel with the goal of burning these fission products to reduce the long-lived radioactivity associated with the fuel. These fast reactor fuels, in addition to having different compositions, are exposed to different reactor conditions. Since these fast reactor fuels are less technologically developed, a test program is needed to prove the fuels perform as anticipated.

An additional source of uncertainty also exists with extended operation or new operating regimes: the potential for new forms of degradation. For example, in the area of radiation effects, in the past, when new reactor operating conditions (temperature, flux, or fluence) have been established at least one new radiation-induced phenomenon has been found. In the 1960s irradiation-induced hardening was discovered. Swelling was a major concern for fast reactors in the 1970s and high-temperature embrittlement due to helium was a surprise in the 1980s. For new Generation IV systems or the extension of current technology, one should be

Laboratory as a user facility in 2007, allowing access to reactor test space and post-irradiation examination facilities through an open solicitation and project selection based on peer review. The ATR National Scientific User Facility (ATR NSUF) now provides the nuclear energy research community a means of testing concepts with the potential to improve the ability of current and advanced nuclear systems to benefit operating performance, economics, safety, and reliability.

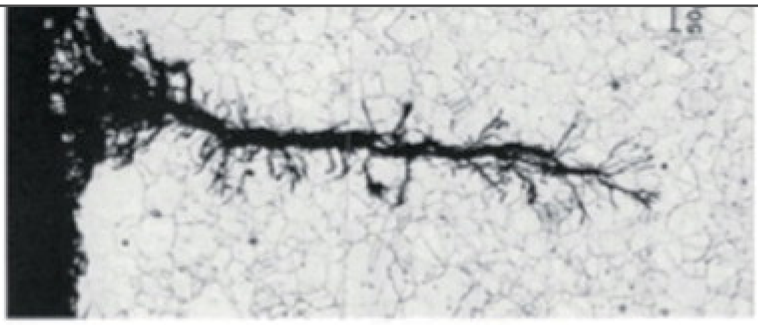
Many countries across the world are working on advanced reactor concepts and while each may use materials with a unique designation system, the fuels and materials used are typically similar and the challenges outlined in this article are common, whether the researcher is from Europe, India, Japan, South Korea, Russia, the United States or any of the other countries researching fuels or materials for nuclear systems. This review article outlines some of the challenges associated with materials and fuels for nuclear systems and describes the ATR NSUF.

Challenges for materials in nuclear power systems

Nuclear reactors present a harsh environment for component service regardless of the type of reactor. Components within a reactor core must tolerate exposure to the coolant (high temperature water, liquid metals, gas, or liquid salts), stress, vibration, an intense field of high-energy neutrons, or gradients in temperature. Degradation of materials in this environment can lead to reduced performance, and in some cases, sudden failure.

Materials degradation in a nuclear power plant is extremely complex due to the various materials, environmental conditions, and stress states. For example, in a modern light water reactor, there are over 25 different metal alloys within the primary and secondary systems (Fig. 1); additional materials exist in concrete, the containment vessel, instrumentation and control equipment, cabling, buried piping, and other support facilities. Dominant forms of degradation may vary greatly between different systems, structures, and components in the reactor and can have an important role in the safe and efficient operation of a nuclear power plant. When this diverse set of materials is placed in the reactor environment, over an extended lifetime, accurately estimating the changing material behaviors and service lifetimes becomes complicated.

Today's fleet of power-producing light water reactors faces a very diverse set of material challenges. For example, core internal structures and supports are subjected to both coolant chemistry and irradiation effects. These stainless steel structures may experience irradiation-induced hardening, radiation-induced segregation and changes to the microstructure. In



(b)

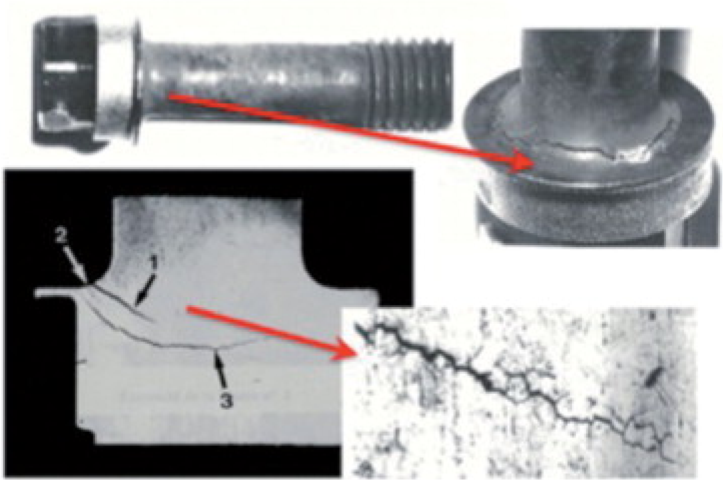


Fig. 2. Examples of stress-corrosion cracking in LWR power plants. (a) Primary water stress corrosion cracking in steam-generator tubing and (b) irradiation-assisted stress corrosion cracking in a PWR baffle bolt.

The reactor pressure vessel, a low-alloy steel component, also experiences radiation-induced changes and can be susceptible to embrittlement. The last few decades have seen remarkable progress in developing a mechanistic understanding of irradiation embrittlement[7]. This understanding has been exploited in formulating robust, physically-based and statistically-calibrated models of Charpy V-notch (CVN)-indexed transition temperature shifts. The progress notwithstanding, however, there are still significant technical issues that need to be addressed to reduce the uncertainties in regulatory application.

Components in the secondary (steam generator) side of a nuclear reactor power plant are also subject to degradation. While the secondary side of the reactor does not have the added complications of an intense neutron irradiation field, the combined action of conditions leading to a very complex component. Stress-corrosion cracking is found in several different forms and may be the limiting factor for component lifetime. The integrity of these components is critical for reliable power generation in extended lifetimes, and as a result, understanding and mitigating these forms of degradation is very important.

In general, concrete structures can also suffer undesirable changes with time because of improper specifications, a violation of specifications, or adverse performance of its cement paste matrix or aggregate constituents under environmental influences (e.g., physical or chemical attack). Some examples are shown in Fig. 38, 9, 10, 11. Changes to embedded steel reinforcement as well as its interaction with concrete can also be detrimental to concrete's service life. A number of areas of research are needed to assure the long-term integrity of the reactor concrete structures. A database with a compilation of performance data under service conditions is an initial need. An additional requirement is a systematic and mechanistic understanding of the mechanical performance impacts from the long-term effects of elevated temperature and, for some locations, the effects of irradiation.

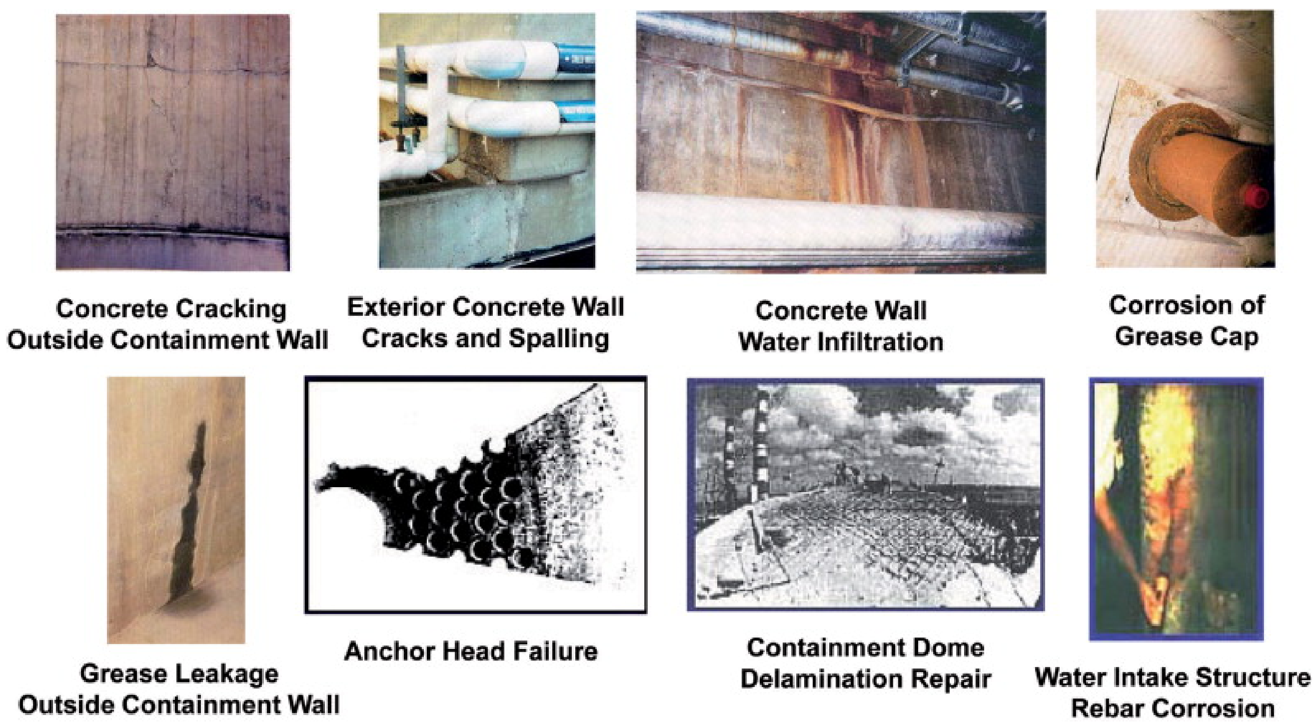


Fig. 3. Examples of degradation in concrete structures.

Courtesy of D. Naus.

at 500 °C to ~10 dpa while fuel cladding and duct materials may be required to survive up to 200 dpa in the same coolant[4]. Components in high temperature gas reactors may reach temperatures up to 1000 °C while liquid salt reactors may require even higher temperatures. Lead or lead alloys provide excellent heat transfer leading to inherently safe reactors but typical construction materials made of Fe, Cr, and Ni are soluble in lead so specific high-temperature corrosion protection methods need to be devised to take full advantage of these coolants. Molten salts provide similar heat transfer characteristics to water but would not have to be pressurized, leading to increased safety under a pipe break. The challenge for many candidate molten salts is that they do not form protective oxides with steels, making corrosion protection the critical issue also. These extreme environments demand advanced materials for successful service.

Advanced materials have the potential to improve reactor performance via increased safety margins, design flexibility, and economics and overcome current reactor performance limitations. Increased strength and creep resistance can give greater design margins leading to improved safety margins, longer lifetimes, and higher operating temperatures, thus enabling greater flexibility. Improved mechanical performance may also help reduce the plant capital cost for new reactors both by reducing the required commodities (with concomitant reductions in welding, quality assurance and fabrication costs) and through design simplifications. Successful implementation, however, requires considerable development and licensing effort. Modern materials science tools such as computational thermodynamics and multi-scale radiation damage models, in conjunction with rapid science-guided experimental validation, may offer the potential for a dramatic reduction in the time period to develop and qualify structural materials.

There are many requirements for all nuclear reactor structural materials, regardless of the exact design or purpose. The material must have adequate availability, fabrication and joining properties, as well as favorable neutronic and thermal properties. Further, it must have good mechanical properties, good creep resistance and long-term stability. Sufficient data under the range of in-core operating conditions must be available to support the licensing process. Finally, since the materials will be used in a high-energy and intensity neutron field, it must be tolerant of radiation effects. When selecting structural materials for any fission reactor application, a careful trade-off analysis is needed for each specific reactor design. Reactor characteristics including operating temperature, coolant, neutron flux, neutron spectrum, fuel type, and lifetime must also be considered to select the most suitable structural material.

Another common need regardless of the advanced reactor design being considered is a detailed understanding of compatibility issues between the structural material and the coolant. Compatibility between the structural materials and coolant is a vital c considerable experience with this coolant in fast reactor applications in the U.S. and internationally, there is little recent experience in sodium compatibility and only scarce data on new alloys currently being developed.

Only through careful evaluation of all factors and a thorough trade analysis will the most promising candidate materials be chosen for further development. It is important to note that there is no ideal material that is best for each of the considerations listed. Indeed, all candidate materials have advantages and limitations. The most promising alloys, which allow the best performance, are also the least technically mature and will require the most substantial effort. These trade-offs must all be weighed carefully.

A systematic and science-based approach can reduce both time and expense required for development, validation, and qualification. This approach may also enable improvements in performance by optimizing alloy composition and processing for specific service conditions. Using a combination of computational tools and more advanced analytical techniques will greatly accelerate research over past advanced reactor material development programs.

Challenges in the development of nuclear fuels

Nuclear reactors are built around a core of fuel. The performance of reactor systems is determined by the performance of the fuel. The inherent physical features of the fuel, such as thermal conductivity, diffusion rates of gaseous species, and chemical compatibility of the fuel and cladding, in turn, determine the performance of the fuel system. Enabling significant improvements in nuclear reactor and nuclear fuel cycle technology depends, to a large degree, on the understanding and development of robust new fuel systems.

The development of nuclear fuel presents many technical challenges. In-reactor fuel behavior is complex, affected by steep temperature gradients and changes in fuel chemistry and physical properties that result from nuclear fission. These challenges are compounded by the highly radioactive nature of irradiated fuel, and the necessity of conducting fuel examinations remotely, in a heavily shielded environment.

Light water reactor fuel challenges

The majority of the world's commercial nuclear power plants are light water reactors. These reactors, after more than 50 years of operational experience, have proven to be extremely successful, generating emission free electricity at a cost competitive with that of coal-fired plants. Worldwide, 359 LWRs operate with a generating capacity of 338 GWe; LWR plants produce 87% of all nuclear electricity and a total of 14% of the world's total electricity[12].

loading that result in low neutron leakage has also occurred. These changes in operation have resulted in steady increases in power production, but also placed additional stress on the fuel. Fuel failures are not due to the failure of the fissile material, but of the cladding that encapsulates the fuel and separates it from the reactor coolant. Fuel failures, while not significant to plant safety, negatively affect the economics of nuclear plant operation, often requiring plant power restrictions or plant shutdown to replace the leaking assembly. These failures have been aggressively managed by the nuclear industry[13]. Approximately 70% of fuel LWR failures are caused by vibration induced wear and cladding penetration by foreign matter[14]. The remaining 30% of failures are due to CRUD deposits, pellet cladding interaction, and unknown or unassigned causes. CRUD is a tenacious iron, nickel, chromium oxide deposit that forms as the result of deposition of stainless steel corrosion products on the fuel surface which results in altered heat transfer from the fuel. Pellet cladding interaction failures initiate during fuel power changes at locations where there are defects in fuel pellet surfaces due to a combination of fission product attack and stress concentration. Also of concern, if a loss of coolant event occurred, is hydrogen uptake by the zirconium alloy cladding, which can lead to cladding embrittlement.

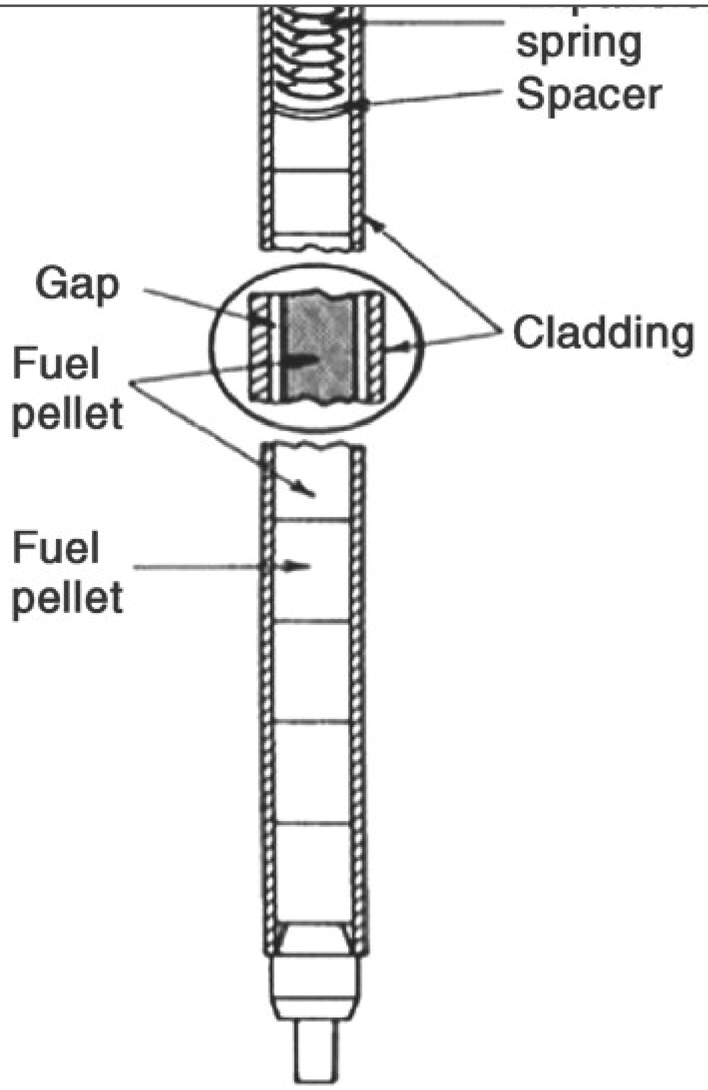


Fig. 4. Schematic of a light water reactor fuel rod.

Given the adverse consequences of fuel failure and commercial limits on uranium enrichment, the practical burn-up limit of current LWR fuels is likely to be in the range of 65 – 75 GWd/MTU[15]. It may be possible to progress beyond this range, either through continued incremental improvements in current fuel technology or by adoption of advanced fuels. Improvements in current fuels would require addressing the primary fuel failure modes discussed above, as well as additional issues that arise at higher burn-up. These additional issues include accelerated irradiation growth of zirconium alloys, management of additional fission gas inventory, the degradation of the mechanical properties of the zirc high conductivity metallic fuel, and composite fuels21, 22. Concepts such as these offer potentially large performance benefits, but may require costly changes to the installed nuclear infrastructure, such as those required for increased enrichment. Advanced fuels will also be required to undergo a long and rigorous licensing process. Based on these factors, deployment of advanced LWR fuels may be possible in the 10–20 year time frame. The journey to deployment of advanced fuels begins with irradiation testing[23] of fuels concepts that have been the subject of careful systems analysis to establish feasibility from a fuel performance perspective.

Beyond electricity: Fuels of high temperature reactors

High Temperature Gas-cooled Reactors (HTGRs) are graphite-moderated nuclear reactors cooled by helium. The high outlet temperatures and high thermal-energy conversion efficiency of HTGRs enable an efficient and cost-effective integration with non-electricity generation applications, such as process heat and/or hydrogen production, for the many petrochemical and other industrial processes that require temperatures between 300 °C and 900 °C. Using HTGRs in this way would supplant the use of premium fossil fuels, such as oil and natural gas, improve overall energy security in the U.S. by reducing dependence on foreign fuels, and reduce CO_2 emissions. Key characteristics of this reactor design are the use of helium as a coolant, graphite as the moderator of neutrons, and ceramic-coated particles as fuel. Helium is chemically inert and neutronically transparent. The graphite core slows down the neutrons and provides high-temperature strength and structural stability for the core and a substantial heat sink during transient conditions. The ceramic-coated particle fuel is extremely robust and retains the radioactive byproducts of the fission reaction under both normal and off-normal conditions.

The TRISO-coated (TRIstructural-ISOtropic) particle fuel forms the heart of the HTGR concept. Such fuels have been studied extensively over the past four decades around the world including in the United Kingdom, Germany, Japan, the United States, Russia, China, and more recently South Africa[24]. As shown in Fig. 5, the TRISO-coated particle is a spherical-layered composite, about 1 mm in diameter. It consists of a kernel of uranium dioxide (UO_2) or uranium oxycarbide (UCO) surrounded by a porous graphite buffer layer that absorbs radiation damage and allows space for fission gases produced during irradiation. Surrounding the buffer layer is a layer of dense pyrolytic carbon called the Inner Pyrolytic Carbon layer (IPyC), a silicon carbide (SiC) layer, and a dense Outer Pyrolytic Carbon layer (OPyC). The pyrolytic carbon layers shrink under irradiation and create compressive forces that act to protect the SiC layer, which is the primary pressure boundary for the microsphere. This three-

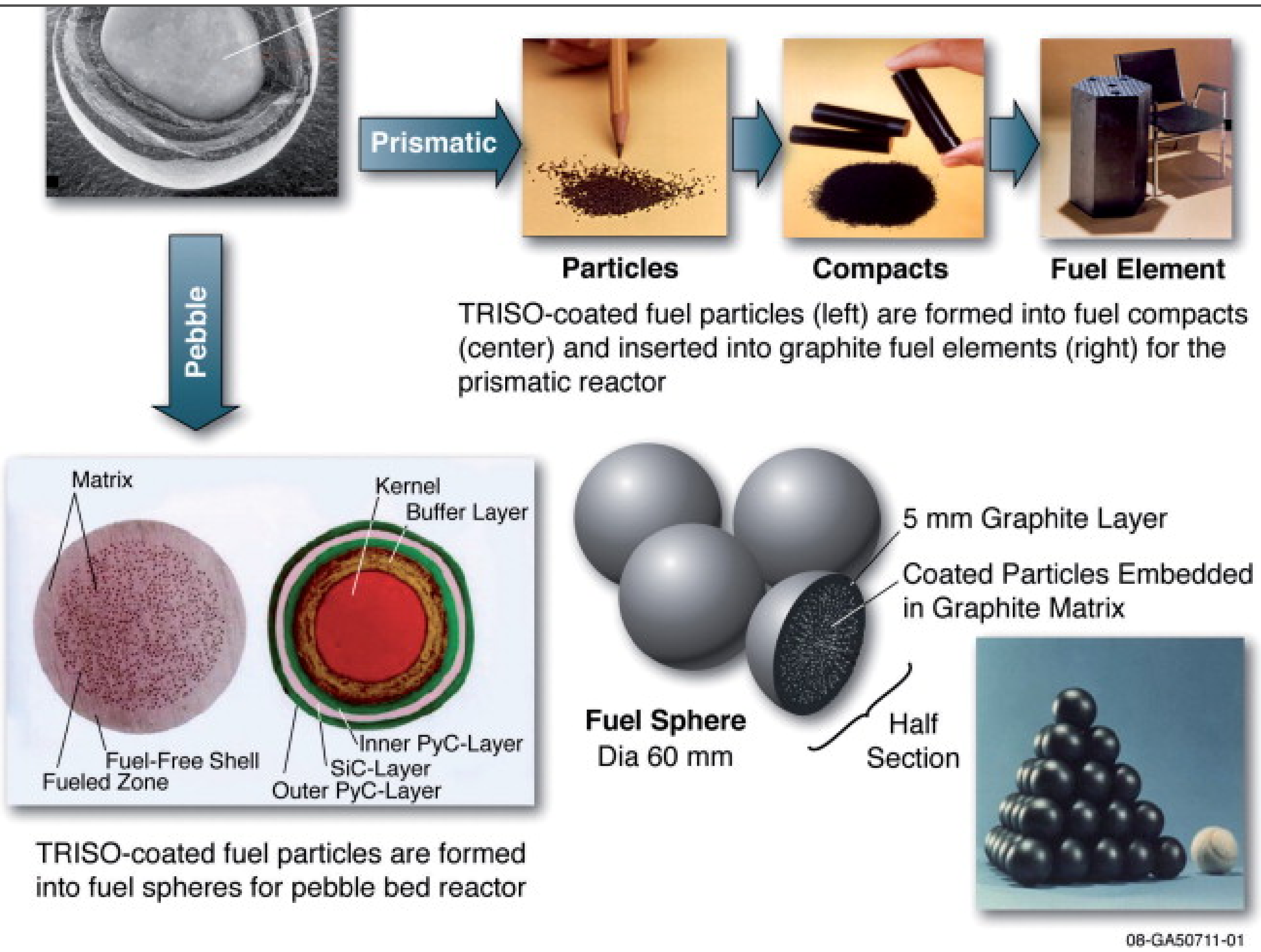


Fig. 5. High temperature gas reactor fuel system, showing TRISO fuel particles consolidated into a graphite matrix as prismatic blocks (upper right) or pebbles (lower right).

An HTGR will contain billions of TRISO-coated particles encased in a graphitic matrix in the form of either small cylinders, called compacts, or tennis-ball-sized spheres, called pebbles (see Fig. 5). Extensive testing has demonstrated the outstanding performance of high-quality low-defect TRISO-coated particle fuels. In the German program in the 1970s and 1980s, over 400 000 TRISO-coated UO_2 particles were irradiated to burn-ups of about 9% at temperatures between 1100 °C and 1150 °C without any failures. Similar results on somewhat smaller particle populations have been obtained with Japanese and Chinese fuels irradiated to low burn-up. About 300 000 TRISO-coated UCO particles have recently completed irradiation in the United States, and no failures have occurred at a peak temperature of 1250 °C up to a peak burn-up of 19%[25]. Testing of German fuel under simulated accident conditior allows these reactors to be located in close proximity to industrial complexes where they can provide heat for high-temperature chemical processes needed for hydrogen production, chemical synthesis, and petrochemical industries.

Significant research and development related to TRISO-coated fuels is underway worldwide. The fuel system is fairly mature and the current challenge is largely focused on extending the capabilities of the TRISO-coated fuel system for higher burn-ups (10–20%) and higher operating temperatures (1250 °C) to improve the attractiveness of high-temperature gas-cooled reactors as a heat source for large industrial complexes where gas outlet temperatures of the reactor would approach 950 °C[27]. Of greatest concern is the influence of higher fuel temperatures and burn-ups on fission product interactions with the SiC layer leading to degradation of the fuel and the release of fission products. Activities are also underway around the world to examine modern recycling techniques for this fuel and to understand the ability of gas reactors to burn minor actinides.

Closing the cycle: Fuels for transmutation

The total mass of spent fuel generated from nuclear power production in Light Water Reactors is relatively small; approximately 30 tons per 1000 MW electric generating capacity per year. Of this mass, approximately 96% is uranium and an additional 3% are short-lived or stable fission products that do not pose major disposal challenges. Approximately 1 wt.% is composed of transuranic elements; plutonium (0.9%) and minor actinides (0.1%) that pose challenges for disposal. The minor actinides include neptunium, americium, and curium.

Among the possible methods proposed for management of plutonium and the minor actinides is neutron induced fission, during which less problematic fission product elements are formed by the 'splitting' of the heavy transuranic atoms. Neutron transmutation systems also produce fission energy that can be converted into usable electricity or process heat. Implementation of neutron transmutation typically utilizes specialized fuels or targets with high minor actinide content in either a purpose designed minor actinide burner or a more conventional reactor system. Suitable in-reactor performance of these fuels and targets is critical to the operation of neutron transmutation systems. A cross-sectional micrograph of an irradiated minor actinide fuel rod tested in the U.S. is shown in Fig. 6.

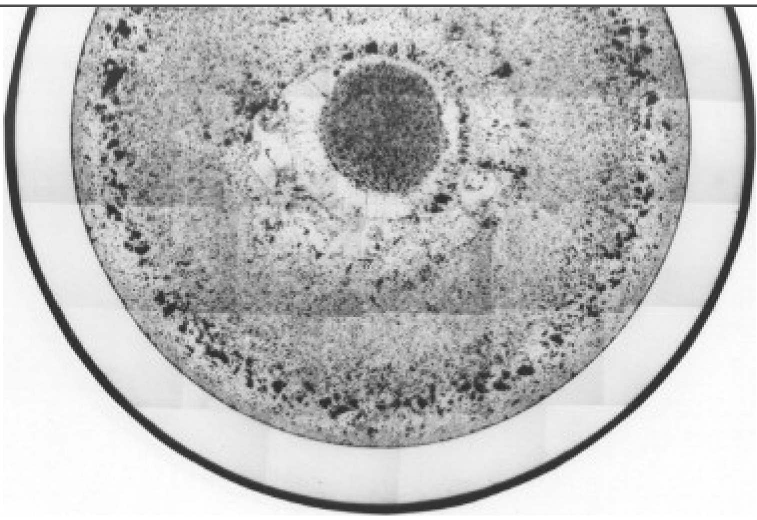


Fig. 6. Cross-sectional micrograph of U-29Pu-4Am-2Np-30Zr metal alloy transmutation test fuel after irradiation to 8.9×10^{20} f/cm^3 (~6 at.% Pu burn-up) in the Advanced Test Reactor.

The fundamental knowledge base of chemical and physical properties of actinide-bearing materials is limited. This includes details of phase equilibrium of multi-component system, fuel microstructure up to reactor operating temperatures; thermophysical properties such thermal conductivity, heat capacity, and thermal expansion coefficients; and chemical properties such as the nature and kinetics of reactions between fuel and cladding material. It is important to understand these properties and kinetic parameters in order to ensure that fuels designed for transmutation meet performance criteria. Modeling of the chemical and physical behavior of these materials is complicated by the presence of the 5f outer shell electrons, and elucidation of properties and parameters has relied heavily on empirical studies. Empirical studies, however, are also complicated by difficulties related to the high activity of these materials. To guide fuel design, continued work utilizing both experimental measurements and computational modeling will be required to provide an understanding of the adequate thermophysical properties.

To provide the highest long-term benefit to reducing radiotoxicity, the quantity of minor actinides placed in a repository should be minimized. The highest potential for material loss occurs during fuel processing to separate the minor actinides from spent fuel and during fuel fabrication. Extending fuel burn-up lifetime, thereby reducing the number of fuel processing cycles is one method of reducing these fabrication losses. The primary candidate fuels for minor actinide transmutation are metal alloy and oxide-based fuels. There is a differ from conventional commercial nuclear fuels31, 32.

Helium production is likely to be the most important fuel design consideration for transmutation scenarios with high minor actinide content. Helium generation is principally due to neutron capture by ^{241}Am to ^{242}Cm and subsequent alpha decay of the ^{242}Cm to ^{238}Pu. A rule-of-thumb for estimating helium production from americium is 50 ml He per gram of transmuted ^{241}Am. The wide range of possible fuel compositions leads to a wide range in the potential for total helium production. There are two possible approaches to dealing with helium. The first is to design and operate the fuel under conditions that promote helium release from the fuel phase to a gas plenum. The second is to design the fuel to effectively retain fission gas and helium while maintaining an acceptable level of gas-driven swelling. Two experiments on americium-bearing oxide fuel effectively demonstrate these divergent approaches. The SUPERFACT experiment[33], which tested two uranium oxide matrix pins containing 20 wt.% americium in the Phénix fast spectrum reactor exhibited gas release of <60%, typical of oxide fast reactor fuel and an acceptable level of fuel swelling at 4.5 at.% burn-up. The EFTTRA-T4 test[34] used a microdispersion of americium oxide in a magnesium-aluminate spinel matrix. Gas release was a fraction of that measured in the SUPERFACT pins. Pellets in this test exhibited volumetric swelling of <18 vol.%, and resulted in excessive cladding strain. It is clear that if gas is to be retained by the fuel, the fuel must be designed to account for large amounts of swelling.

The advanced test reactor national scientific user facility: A model for research collaborations

The ultimate performance of a fuel or material in a nuclear system is determined through in-reactor testing. Thus, the availability of test reactors, hot cells, and examination equipment that can handle radioactive materials is required to prove the principle of any advanced concept. The Advanced Test Reactor (ATR) has been in operation since 1967 and mainly used to support U.S. Department of Energy (U.S. DOE) materials and fuels research programs. Irradiation capabilities of the ATR and post-irradiation examination capabilities of the Idaho National Laboratory (INL) were generally not being utilized by universities and other potential users due largely the high cost of using these facilities relative to typical research grant awards. While materials and fuels testing programs using the ATR continue to be needed for U.S. DOE programs such as the Fuel Cycle Research and Development Program and Reactor Concept Research Development, & Deployment programs, the U.S. DOE recognized there was a national need to make these capabilities available to a broader user base.

ATR NSUF, the post-irradiation examination equipment was that available at the INL hot cells, analytical chemistry laboratories, and electron microscopy laboratory.

Since opening up the ATR, the NSUF has expanded the ways potential users can interact with the facility. Specifically:

- Additional capability at INL was opened to potential users, specifically the ATR Critical Facility which is a low power, geometrically identical version of ATR which can be used to test radiation detection systems or validate reactor neutronics codes.

- The post-irradiation examination capability at the INL has been significantly upgraded with the addition of analytical equipment such as an electron microprobe, a field emission gun scanning transmission electron microscope, an atom probe, a scanning Raman system, an atomic force microscope, and dual beam focused ion beam systems.

- A sample library was created that allows potential users to propose specific experiments against materials and fuels previously irradiated in other DOE or industry programs

- Connections are being made with other national user facilities such as the Advanced Photon Source at Argonne National Laboratory that allows experimenters to analyze samples that are transported to these complimentary user facilities.

- A network of university partners has joined the NSUF, providing additional capability for irradiation and post-irradiation testing (Fig. 7 (a) indicates the partner facilities).

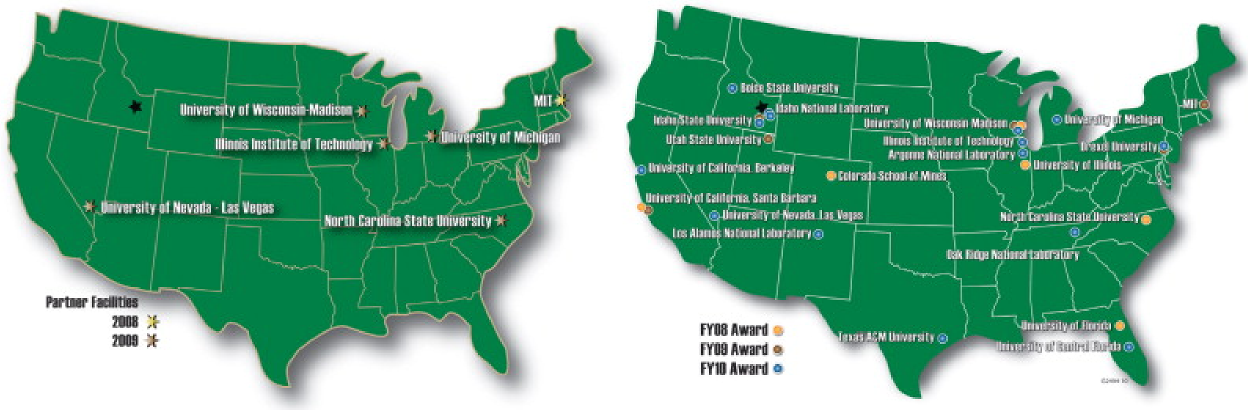


Fig. 7. University partners of the ATR NSUF who are part of an integral national irradiation and post-irradiation testing capability. (b) Universities leading projects currently being supported at the ATR NSUF.

The first full year of implementing the user facility concept was 2008 and since that time the NSUF has initiated work on 23 user-proposed projects. These projects are listed in Table 2. The projects break into two classes of experiment:

- New reactor-based projects. These projects involve designing and inserting new fuels and materials into either the ATR or the MITR or using the ATR Critical Facility. These proposing institutions for these projects are shown in Fig. 7 (b).

- Post-irradiation only projects. These projects analyze previously irradiated material that is held in the sample library against which potential users can propose examinations.

Continuing improvements in nuclear energy technology rely on the development of improved materials and fuels for advanced reactor systems. Continued life extension of current LWR plants relies on a thorough understanding of the effects of the reactor environment on long-term material degradation. Both of these research areas require access to a specialized nuclear research infrastructure, including high flux test reactors, radiation shielded research laboratories, and high-end materials characterization tools dedicated for use on radioactive materials. The establishment of the ATR NSUF has provided an effective mechanism for research teams to access this specialized infrastructure for testing advanced materials and fuel concepts and for better understanding the degradation of materials and fuels in the existing reactor fleet.

Acknowledgements

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