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Accident tolerant fuels for LWRs: A perspective ☆

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ABSTRACT

The motivation for exploring the potential development of accident tolerant fuels in light water reactors to replace existing Zr alloy clad monolithic (U, Pu) oxide fuel is outlined. The evaluation includes a brief review of core degradation processes under design-basis and beyond-design-basis transient conditions. Three general strategies for accident tolerant fuels are being explored: modification of current state-of-the-art zirconium alloy cladding to further improve oxidation resistance (including use of coatings), replacement of Zr alloy cladding with an alternative oxidation-resistant high-performance cladding, and replacement of the monolithic ceramic oxide fuel with alternative fuel forms.

1. Introduction

Nuclear power has proven to be a reliable, environmentally sustainable, and cost-effective source of large-scale electricity. In the interest of continued technological improvement, further improvements in operational reliability, economics, and safety under normal and transient conditions are being pursued worldwide. In regard to safety during anticipated transients and postulated accidents, the safety of the fuel can be affected by numerous fuel system phenomena, including cladding oxidation/hydriding, pellet-cladding interaction, pellet relocation and dispersal, and cladding embrittlement and fragmentation [1].

The high power density that makes nuclear power economical also makes the system susceptible to severe accidents. Typical power densities in light water reactor (LWR) cores are ~50–75 $MW_{th}\ m^{-3}$, which is about a factor of 100 greater than the average power density in fossil energy plant boilers [2]. In a typical loss of coolant accident (LOCA) scenario, a reactor scram dramatically reduces the power generation in the core. However, substantial amounts of heat continue to be generated following the scram: ~7% of full power immediately after the scram, ~1% of full power 4 h after the scram, and ~0.2% of full power 10 days after the scram [3]. Considering that the thermal full power levels in commercial LWRs often exceed 3000 MW, the post-scram decay heat deposited in a reactor core without circulating coolant (i.e., approaching adiabatic conditions) can lead to rapid temperature increases and subsequent core degradation.

Also due to the high power density in nuclear reactor cores, the system has relatively limited margin for accommodating additional energy generation under normal operating conditions. Unlike fossil energy electricity generation sources, fuel, operation, and maintenance of nuclear reactors constitute only a minority fraction (~one quarter) of the electricity generation cost (the rest being the capital costs). Therefore, the economics of nuclear reactor operation improves as the power rating of these units increases, which has led to many of the currently operating units undergoing power uprates. While plant power uprates involve a number of considerations, the thermal limits of the current fuels are one factor that is limiting future power level increases since further local power increases in the core could result in fuel damage under anticipated operational occurrences [1].

In addition to LOCA events as discussed above, another safety design basis is related to the potentially rapid power excursions that can occur from reactivity insertion events, such as control rod ejection in an LWR. Energy deposition resulting from reactivity insertion in the core can be limited by appropriate reactor physics design of the core, balancing the design reactivity worth of the control rods with the fuel temperature feedback. However, the specific fuel system response to any given magnitude of energy deposition varies during its lifetime because of the changing thermal–mechanical properties of the fuel/cladding system.

As discussed in Section 2, numerous operational and retrofit design changes were instituted in LWRs in the 1980s following the LOCA at Three Mile Island, and these changes have substantially reduced the probability of core degradation during a severe accident. These specific design features and upgrades were primarily associated with maintaining adequate core cooling in the event that the primary cooling system is not functional. The recent station

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blackout (SBO) accidents at three of the Japanese Fukushima Daiichi reactors following the devastating 2011 earthquake and tsunami have sparked renewed interest in exploring the possibility of further design and fuel system improvements that could improve the safety of LWRs under design-basis (DB) and beyond-design-basis (BDB) accident scenarios. Section 3 provides a brief overview of core degradation phenomena under DB and BDB accident conditions and possible mitigation by design changes and utilization of new fuel systems. Section 4 reviews the overarching motivation for investigating accident tolerant fuel (ATF) systems, and Section 5 briefly summarizes some of the ATF concepts currently being explored. Several accompanying papers in this special issue provide more detailed information on the current status of some specific ATF systems.

2. Historical perspective

Reactor designers and regulators during the early decades of commercial nuclear energy established the current safety basis framework for nuclear reactor design. Their understanding of the integral behavior of these power systems at the time led them to envision a number of postulated accident scenarios that aimed to envelope the safety design requirements by considering two separate classes of DB accidents as bounding events: LOCAs and reactivity initiated accidents (RIAs). It was understood at the time, given a deterministic analytical approach, that the likelihood of an accident scenario extending beyond the safety design limits would be negligible. Therefore, the safety design requirements by regulatory authorities did not require mitigation strategies for BDB accident scenarios. It is also important to note that the design safety features of these reactors were based on evaluations of the engineering performance of the fuel/cladding system under accident scenarios. For example, the emergency core cooling system (ECCS) was specifically designed and configured to provide sufficient cooling to prevent cladding and fuel from reaching temperatures that could result in failure under design basis LOCA conditions.

Understanding of the specifics of the behavior of the ECCS, and hence its requirements during DBAs, evolved considerably in the 1970s and 1980s [4]. At the same time, the partial meltdown of the core at Unit 2 of the Three Mile Island power generating station induced two important shifts in reactor safety assessment and implementation approaches. The first was to augment the deterministic approach with probabilistic risk assessment (PRA) to better capture the complexities of system behavior. The second shift was to transition away from active safety systems towards passively safe reactor concepts. While the former was adopted more readily, the latter is just now being realized and only in certain parts of the world; only a few of the 80+ nuclear reactors under construction globally take advantage of at least partially passive safety systems. Specific examples of reactors with passive safety include generation III+ designs such as the Westinghouse AP1000, GE-Hitachi ESBWR, and most small modular reactor concepts. For future reactor designs, passive safety features and ATFs can be combined to provide new concepts that minimize the potential for off-site releases, lessen the necessity for operator actions, and simplify the reactor system overall, thereby reducing capital and operating costs.

The recent events at Fukushima Dai-ichi have once again underscored the importance of adopting passively safe reactor designs and robust safety systems. These BDB accident scenarios have also challenged the outcomes of the current PRA models used to quantify operational risk for the current operating fleet of reactors belonging to older generation designs. The issue is not the PRA approach but rather the specific modeling assumptions regarding event probabilities that impact the overall probabilistic results. Most pertinent to this discussion, these events have also showcased the potential major consequences of the large energy released from LWR cores as they physically and chemically degrade in a high-temperature steam environment under BDB events. Accordingly, a significant focus is now being placed on core degradation processes that are largely governed by fuel behavior and the potential to enhance safety margins by utilizing ATFs to alter these processes.

3. Review of core degradation processes and mitigation by ATFs

Fig. 1 provides a general overview of core degradation processes under coolant-limited conditions and highlights the potential for ATF cladding concepts to alter the rate of accident progression. Essentially, during the first phase of the accident, dubbed Lead Up in Fig. 1, core cooling capability is lost either via loss of water coolant due to a

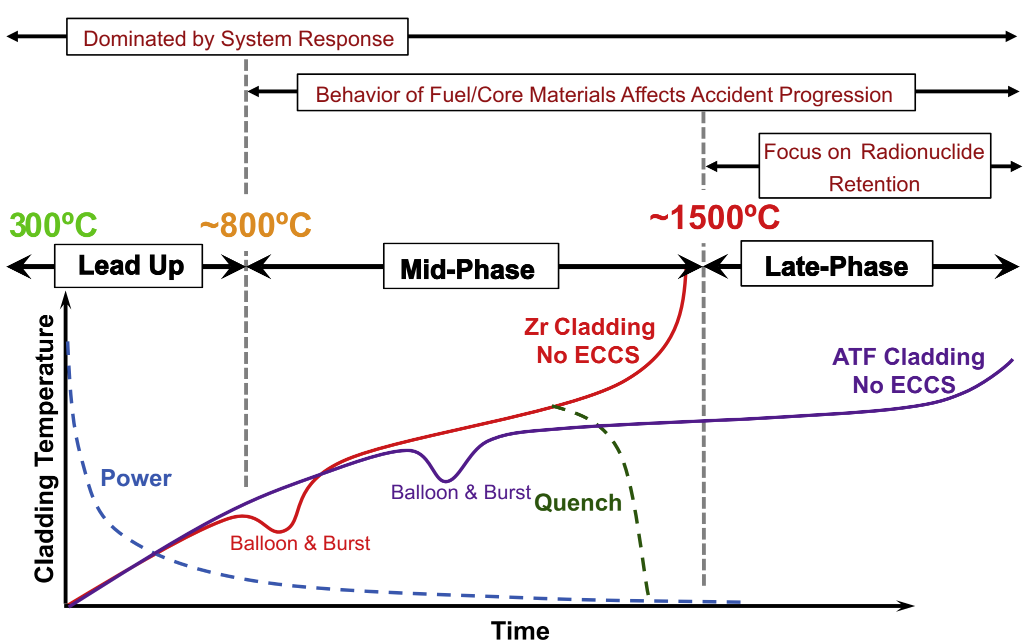

Fig. 1. General overview of coolant-limited accident progression inside an LWR core.

pipe break or loss of cooling capability due to pump failure (eventually leading to depressurization of the core without active core cooling). This phase can span over seconds in the case of a large-break LOCA, minutes in the case of a small break LOCA, or hours in the case of an SBO. During the lead-up phase, the decay heat in the core and the stored energy in the fuel boil off the remaining coolant in the core, subsequently driving the fuel temperature higher.

Once the fuel reaches temperatures close to ~800 °C, a new phase in accident progression is initiated (referred to here as the Mid-Phase), comprising the onset of physical and chemical degradation phenomena in the core. Fuel rod burst for the current UO_2–Zr alloy fuel system is expected at the temperature range 700–1200 °C [5], releasing a fraction of the fuel rod inventory of volatile fission products. Accelerated oxidation and detrimental interactions (e.g., low melting temperature eutectic phase formation) of the zirconium alloy fuel cladding and various other core components is also initiated during the Mid-Phase. These scenarios have been studied extensively for the current UO_2–Zr alloy fuel system [6,7]. In case of a design-basis LOCA, once core depressurization has occurred, or even before that (given the nature of safety systems in place), coolant is injected into the core and the fuel is quenched by the ECCS. However, if the ECCS is not operational, the oxidation reaction between steam and 20–60 tons of Zr alloy in the core along with other reactive constituents (e.g., control rod constituents) results in the generation of appreciable amounts of heat and hydrogen gas. During some periods of the accident, the heat generated from oxidation can be comparable to or even greater than the decay heat [8]. The heat generated from oxidation exacerbates the nature of accident progression by driving the core temperature higher while increasing core cooling requirements. Once the core temperature reaches values above ~1500 °C, severe degradation is expected to have occurred. This is specified as the Late-Phase in Fig. 1. After this point, the focus essentially shifts to radionuclide release management and protection of the larger containment structure. Hydrogen gas, the inevitable by-product of steam oxidation, can contribute to the containment compromise through slow pressurization or explosions.

For the case of reactivity initiated accident scenarios, the chief concern for the current Zr alloy clad UO_2 fuel system is rapid fuel pellet expansion against the cladding (pellet-cladding mechanical interaction) due to prompt fuel melting (with ~10% volumetric expansion) that can cause cladding failure and fuel dispersal [9]. Several additional RIA phenomena could also lead to fuel failure [10]. These issues become progressively important at medium to high burnups due to pellet-clad gap closure, cladding embrittlement due to radiation damage and waterside oxidation (loss of sound metal and formation of hydrides), and other mechanisms [10]. Potential accident tolerant fuel approaches for RIA would include new cladding systems that have improved strength and ductility compared to current Zr alloys, and new fuel forms that decrease the differential thermal expansion of the fuel compared to the cladding under RIA conditions (e.g., lower thermal expansion coefficients and/or increased temperature intervals between normal operation and melting points for the fuel form compared to monolithic uranium-based oxide fuels). Also, fuel forms that contain their fissile portion as discrete regions dispersed inside an inert matrix can potentially offer enhanced tolerance to postulated RIA scenarios since during such an event the energy is selectively deposited in those locations. In this manner the rapid local heating produces a strong negative reactivity feedback due to the Doppler effect, while expansion and melting of the fissile portion can be localized and contained within the inert medium [11].

Given this general and simplified view of accident progression inside an LWR core, it becomes clear that accident progression and mitigation are largely dominated by the availability and nature of safety systems in place (such as the availability and operation of emergency core cooling systems). However, it is also obvious that during the Mid-Phase and thereafter for a coolant-limited transient, steam oxidation along with physical degradation of core components as well as chemical interaction between the various constituents significantly exacerbates the course of accident progression. ATF concepts aim to delay the onset of these detrimental processes (by reducing the high temperature oxidation rate and also by potentially delaying the ballooning and burst of the cladding), thereby reducing the burden on reactor safety systems and the operators managing the accident response, as discussed in the following section.

4. Motivation for exploring accident tolerant fuels

In concert with innovations in reactor design, improvements in fuel materials are being sought to improve accident tolerance. The underlying motivation for exploring accident tolerant fuel systems is twofold: It may be difficult to implement significant design changes in some existing reactors, and, even for cases where reactor design changes can be introduced, further enhancements in safety might be best achieved by utilizing a combination of fuel system innovations along with operational and/or reactor design changes. The idealized goal for ATFs would be to provide substantially improved safety response to a DB or BDB accident while maintaining good operational characteristics under normal conditions. In particular, increased coping time to respond to an accident (and thereby potentially mitigate the consequences by active or passive measures) would be highly beneficial.

The improvements in accident tolerance should preferably accompany simultaneous improvements (or at least only minor hindrances) to normal operations in terms of compatibility with current plants and operations, economics, and impacts on the fuel cycle (fuel fabrication and used fuel disposition, UFD). For example, the ATF system should be designed as a drop-in replacement for existing fuel assemblies. The fuel failure rate should be comparable to or better than that of current state-of-the-art fuel systems during normal operations, and the handling of used fuel should be comparable to (or preferably simplified over) current practices. It is desirable but not mandatory that prospective ATF systems be potentially implemented in a relatively short (decadal) timeframe to provide enhanced safety for both current and future LWRs. Applicability to existing reactors is particularly significant, considering that there are over 400 nuclear reactors in operation worldwide that provide about 13% of the world's electricity, and there are active research programs in several countries to explore the feasibility to safely and economically extend the operating lifetimes of these existing reactors by 20 years or longer.

The easiest ATF system to implement from an operational perspective would be based on slight modifications to the existing Zr alloy clad UO_2 fuel system. However, the performance of fuel systems based on Zr alloy cladding has already been highly optimized over ~60 years of development. Therefore, although further performance improvements may be anticipated, incremental changes in Zr alloy cladding are unlikely to achieve dramatic improvements in accident tolerance. For example, hydride formation that occurs in Zr alloy cladding during operation to high burnups is a major challenge (due to loss of sound metal and toughness degradation) that could ultimately limit fuel lifetime under normal operating conditions as well as strongly affecting LOCA, RIA, and UFD behavior [1,8,12]. UFD behavior is becoming increasingly important in the United States given that the used fuel may need to maintain its integrity over an extended interim storage period and through transportation to an eventual repository for ultimate disposal.

The key performance features that constitute accident-tolerant behavior are dependent on the specific accident scenario. Fig. 2

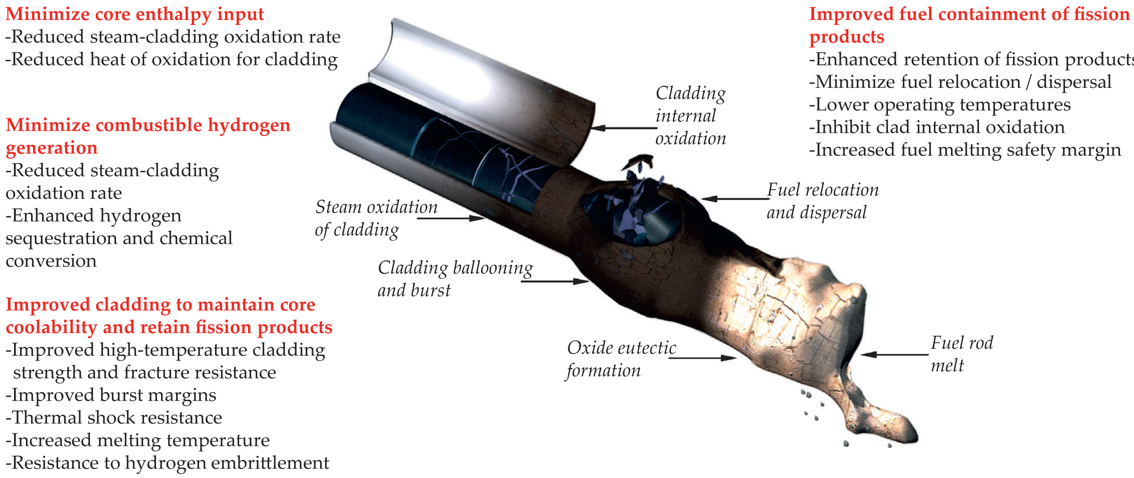

Fig. 2. Evolution of a fuel rod under LOCA and SBO conditions extending well into the beyond design basis space along with four key performance features of ATF concepts for improved safety margins.

shows the evolution of the fuel rod under a LOCA scenario extending well beyond the design basis limit. The fuel segments in the upper portion of the figure are intended to schematically convey the evolution of the fuel system with increasing burn up (fuel swelling and cracking, fission gas buildup and release, partial oxidation of the clad inner and outer surfaces). There are four main desired attributes of ATFs (in addition to maintaining good performance under normal operating conditions), as outlined in Fig. 2. For LOCAs and SBOs, suppressed oxidation compared to Zr alloy cladding in the presence of hot steam (with nonvolatile reaction products) would simultaneously minimize hydrogen generation and enthalpy input. In addition, the accident tolerant fuel system should exhibit good dimensional stability (i.e., good tensile and thermal creep strength of cladding) and fission product retention up to temperatures in excess of 1200 °C in order to maintain a controllable and coolable core geometry without large release of radioactive materials. For RIA scenarios that produce a relatively rapid temperature increase of short duration, a key attribute is resistance to cladding rupture via pellet–cladding mechanical interaction [1,9,10], which implies cladding materials with improved ductility and toughness at moderate to high temperatures (along with improved oxidation resistance at normal operating temperatures) compared to Zr alloys. Finally, fuel forms that enhance confinement of fission products under a variety of accident scenarios are desirable.

5. Overview of potential ATF concepts

Taking into consideration the desirable attributes of ATF systems summarized in Section 4, a wide variety of different potential fuel system concepts can be envisioned. Underlying the selection of any of these ATF systems is the importance of maintaining good performance under normal LWR operating conditions (good neutron economy and burnup limits, resistance to stress corrosion cracking and grid to rod fretting, etc.). The approaches can be grouped into three general categories: improved high temperature oxidation resistance and/or strength of Zr alloy cladding, non-zirconium cladding with high strength and oxidation resistance, and alternative fuel forms with improved performance and fission product retention compared to monolithic UO_2.

5.1. Zr alloy cladding with improved oxidation resistance and strength

The rates of both hydrogen generation and heating in the reactor core due to oxidation are directly proportional to the rate of steam-driven oxidation of the core materials. The oxidation of core materials in existing LWRs during LOCA conditions is dominated by oxidation of the Zr alloy components (fuel cladding, along with channel boxes in boiling water reactors). At temperatures above ~1200 °C, the heating associated with oxidation of Zr alloy core components can dominate the overall heat generation rate in the core for a BDB LOCA or SBO scenario [8,13]. Therefore, a reduction in the high temperature oxidation rate of Zr alloys would provide multiple benefits in terms of improved accident tolerance during LOCA or SBO conditions.

Potential approaches to reduce the oxidation rate include development of new oxidation-resistant Zr alloys and application of coatings (self-passivating or applied prior to service). Pronounced improvement in the oxidation resistance of zirconium alloys up to high burnups at normal operating conditions (~300 °C) has been achieved over the past few decades as a result of continuous alloy development efforts [8]. However, it is considered unlikely that the relatively small (<2% solute) alloying additions used to date in Zr alloy cladding can be further modified beyond the impressive improvements already achieved in order to achieve the needed additional ~100-fold reduction in high temperature oxidation resistance; a markedly different alloy composition may have to be developed.

An alternative approach to bulk compositional changes would be to pursue the development of highly adherent oxidation-resistant coatings. It would be particularly convenient if the coating could be easily regenerated if a small portion spalled off during fuel assembly handling or during normal reactor operation. Major challenges for coating development include careful matching of the coefficient of thermal expansion in order to minimize interfacial stresses and delamination during cycling from room temperature to ~300 °C (normal operations) or from ~300 °C to high temperature (off-normal conditions), including consideration of the highly anisotropic (textured) microstructure of Zr alloy tubing in the axial versus circumferential directions. Moreover, the coating must be compliant enough to withstand the diametrical compression that results from reactor pressurization, making the use of brittle ceramics problematic (generally requiring relatively thin and highly adherent coatings). Differential volumetric and microstructural evolution between the coating and underlying cladding under neutron irradiation (e.g., anisotropic growth) can also lead to coating spallation. The viability of coatings to protect the exposed cladding to a sufficient extent after burst also needs to be considered; that is, after burst, the inner, uncoated surface of the cladding is exposed to the steam, leading to rapid oxidation, hydrogen pickup in the cladding, and cladding embrittlement.

In addition to oxidation resistance, it would be desirable for the Zr alloy-based cladding to exhibit improved resistance to creep deformation and crack growth at high temperatures in the aggressive nuclear fuel environment, in order to retard ballooning and rupture of the cladding during LOCA or SBO scenarios, and to also have improved resistance to pellet-cladding mechanical interaction deformation and fracture processes associated with RIA scenarios. The improved high temperature strength would also be beneficial for minimizing spalling of coatings during high temperature excursions.

5.2. Alternative (non-Zr) cladding materials

Materials research performed over the past several decades has led to the development of several high-performance materials systems that have very high resistance to oxidation in high temperature steam or air. A variety of approaches are being considered as potential accident-tolerant alternatives to Zr alloy cladding, including oxidation-resistant structural alloys, silicon carbide fiber-reinforced SiC ceramic composites, and refractory alloys. In all cases, the desired accident-tolerant attributes are improved high temperature oxidation resistance (>100 $\times$ improvement compared to current Zr alloy cladding) and comparable or superior high temperature strength.

Oxidation-resistant structural alloys such as high-performance stainless steels offer the potential for improved strength and oxidation resistance compared to existing Zr alloys over a broad temperature range [8]. Early commercial reactors successfully used austenitic stainless steel cladding in pressurized water reactors, whereas their performance in boiling water reactors was poor due to stress corrosion cracking problems [8]. Issues to be resolved include the effect of higher thermal neutron absorption cross section on fuel enrichment and other operational requirements, potential enhanced susceptibility to stress corrosion cracking in LWR primary coolant water chemistries (particularly for austenitic alloys), the impact of radiation hardening and embrittlement on cladding performance under normal operating conditions, mitigation/management of the increased tritium burden associated with bare steel clad, and pellet-cladding mechanical interaction behavior under RIA conditions (potential impact of radiation embrittlement).

Silicon carbide fiber-reinforced SiC matrix ceramic composites (SiC/SiC) have been proposed as a potential cladding material [14] (and also as channel boxes for boiling water reactors) [15] due to their low thermal neutron absorption cross section, retention of strength up to very high temperatures, good radiation resistance, and good oxidation resistance in air and steam up to temperatures of at least 1600 °C. This class of materials, while potentially very attractive, has yet to find application in nuclear systems, though SiC composites are now finding industrial application in turbine components. A generic issue with the application of this class of structural materials is the current lack of engineering familiarity in design and application, exacerbated by a significant lack of industry codes and testing standards. Issues to be resolved specific to clad application include defining a path forward for significant reductions in the cost for fabrication of tubing and other geometries (development of low cost, high performance SiC fibers), development of robust joining methods, demonstration of hermetic retention of fission products under normal and transient conditions, development of QA methods and standards, and investigation of pellet-cladding mechanical interaction behavior under RIA conditions. Some of these items are discussed in great detail in accompanying papers in this special issue of the journal.

Refractory alloys based on Nb, Ta, Mo, or W offer the potential for significant improvements in high temperature strength compared to Zr alloys. These refractory alloys generally exhibit poor behavior in high temperature oxidizing environments, although exploratory research has identified some alloys (e.g., Mo_2Si-based) with adequate short-term oxidation resistance [16], and techniques for applying adherent oxidation-resistant coatings to refractory alloys are under development [17]. It is noted that steam attack in these materials has only recently been explored [18] though it is widely known that high temperature oxidation can be significantly different for steam and air. Potential cladding designs for refractory alloys include monolithic (possibly with oxidation-resistant coatings) and multilayered “sandwich” designs where thin oxidation-resistant layers are applied to the inner and outer surfaces of the refractory alloy. Issues to be resolved include investigation of the potential for low-cost fabrication of tubing, the effect of higher thermal neutron absorption cross section for some of these refractories on fuel enrichment and other operational requirements, the impact of radiation hardening and embrittlement on cladding performance under normal operating conditions [19–21], oxidation behavior in steam at high temperature, and investigation of pellet-cladding mechanical interaction behavior under RIA conditions.

5.3. Alternative fuel forms

Consideration of alternative fuel forms to the current monolithic ceramic uranium-based oxide fuel pellets could provide improved accident tolerance via improved fission product retention at high temperatures and improved thermal/mechanical physical properties. Heat capacity of the overall fuel pin significantly affects the heat-up rate during transients [13], and in certain short-term accident scenarios, the stored energy in the fuel that is governed by thermal conductivity can affect the course of accident progression [22]. Alternative fuel systems under consideration include high density and high thermal conductivity fuels (e.g., monolithic nitrides or oxides containing high-conductivity second phases [23]) and dispersed/inert matrix fuel forms where the fission products would be contained within discrete, isolated fuel particles embedded within a nonfissile matrix (e.g., fully ceramic microencapsulated fuel [FCM] forms [24]). Potential benefits for dispersed particle fuel forms include improved retention of fission products via introduction of one or more additional safety barriers for fission product release (at the potential expense of increased fuel isotopic enrichment). Issues to be resolved include fabrication cost, reactor physics design, optimized fuel enrichment level, irradiation behavior, and investigation of behavior under LOCA, RIA, and BDB scenarios. The relative lack of practical operating experience with alternative fuel forms compared to monolithic UO_2-based ceramic fuel is also a barrier to their development, due to potential unknown issues.

6. Conclusions

Several potential approaches for development of ATFs exist that could lead to improved accident tolerance in LWR fuel systems, while not sacrificing the impressive performance features of current fuel systems under normal operating conditions. This special issue of *Journal of Nuclear Materials* is dedicated to exploring these various options and offering basic guidelines on the attributes of ATF concepts. Each of these ATF options has known or potential shortcomings; no panacea is in sight.

In general, accident progression in LWRs is dominated by the integral system response during the course of such events. ATFs can affect the sequence and rate of this progression, thereby minimizing the burden on safety systems and providing enhanced coping time during such events. In this manner, the safety margins for nuclear power systems can be enhanced.

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