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ACCIDENT TOLERANT NUCLEAR FUEL CLADDING: CONCEPT BACKGROUND, DEVELOPMENT AND CHALLENGES

Introduction

The continuous growth in electricity demand worldwide accompanied with the urgent need for managing atmospheric greenhouse gas emissions simultaneously drives a growing demand for environmentally sustainable electricity generation. Nuclear power generation provides a reliable and economic supply of electricity, with very low carbon emissions and relatively small amounts of waste. Today there are about 440 nuclear power reactors operating in 30 countries with additional 55 under construction, and a combined capacity of about 400 GWe, in 2018 these provided over 10% of the world's electricity [1].

All light water reactors (LWRs) around the world are currently using fuel systems consist of uranium oxide (UO₂) encapsulated within a zirconium-based alloy cladding (fig.1). Some reactors use uranium-plutonium oxide fuels, widely known as mixed oxide (MOX) fuels. The choice of zirconium alloys as the primary cladding material is due to its low neutron absorption,

good corrosion resistance and structural integrity. The oxide fuel-zircaloy system has been optimized over many decades and performs very well under normal operations and anticipated transients. However, because of the highly exothermic nature of zirconium-steam reactions, under some low frequency accidents when core cooling is temporarily lost and part of the core is uncovered – low probability accidents may lead to an excess generation of heat and hydrogen, resulting in undesirable core damage.

Nuclear safety is crucial and a prerequisite for the successful use of nuclear technology. Materials inside the reactor core are exposed to an extremely harsh environment due to the combination of high temperature, high stress, a chemically aggressive coolant, and strong radiation, accordingly; nuclear cladding represents one of the most important components for maintaining fuel integrity and plant safety. Continual improvement of technology, including advanced materials and nuclear fuels, remains central to the industry's success. In the past decades, the emphasis of the R&D of LWR fuel was placed on improving nuclear fuel performance under normal conditions in terms of increased fuel burn up for waste minimization, increased power density for power upgrades, and extended operational service for economic competitiveness.

Enhancing the accident tolerance of LWRs became a topic of serious discussion following the 2011 Fukushima Daiichi nuclear power plant complex accident. The goal of accident-tolerant fuel (ATF) development is to identify alternative fuel system technologies to further enhance the safety, competitiveness and economics of commercial nuclear power.

The complex multi-physics behavior of LWR nuclear fuel makes defining specific material or design improvements difficult. Hence, establishing desirable performance attributes is critical in guiding the design and development of fuels and cladding with enhanced accident tolerance. ATF designs would endure severe accident conditions in the reactor core for a longer period of time than the current fuel system while maintaining or improving fuel performance during normal operations.

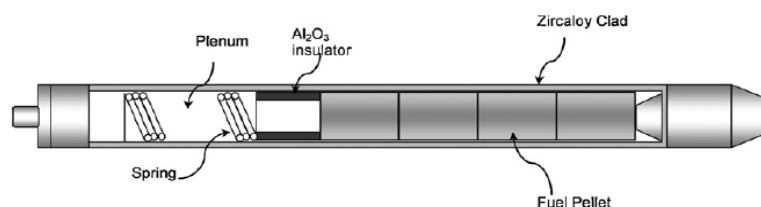


Fig. (1)[2]: (a) Sectional view of clad tube

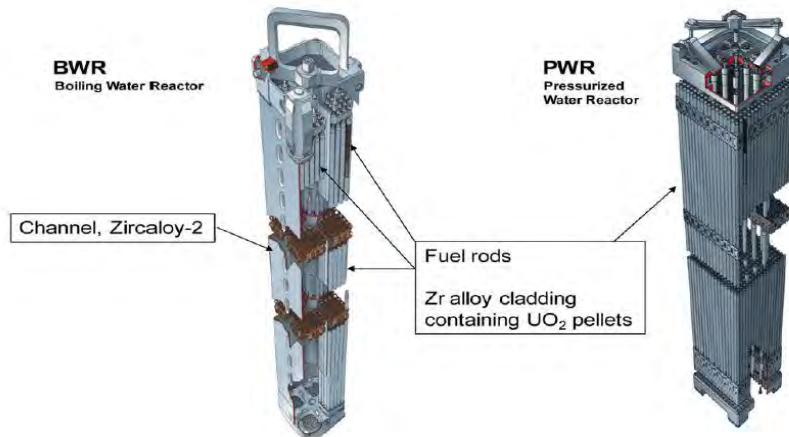


Fig.(1) [3]: (b) Examples of modern LWR fuel

Emergence of the accident tolerant fuel concept

The Fukushima accident in 2011, triggered by a massive 9.0 magnitude earthquake followed by a tsunami, had a devastating effect on the status and prospects of nuclear power worldwide. Alarmed by the incident and the following large release of radioactive material to the atmosphere and the surrounding land and ocean, Japan authorities required a shutdown of its nuclear capacity. Longer term effects included Japan's later announcement of plans to reduce dependency on nuclear power and to revise their Basic Energy Plan. In addition, Germany and Switzerland have announced plans to phase-out nuclear energy in the foreseeable future. As a result of the accident, a renewed interest was generated to address the shortcomings of the traditional zirconium-based (so-called "Zircaloy") claddings under accident conditions. The proposed solutions are called accident tolerant fuels (ATFs) [4].

ATFs are fuel systems; which in comparison with the standard UO₂-Zr system, can tolerate loss of active cooling in the core for a considerably longer time period (depending on the LWR system and accident scenario) while maintaining or improving the fuel performance during normal operations [5]. These improved properties are mainly focused on reducing the oxidation rate and hydrogen production at high temperatures. Additional, targeted improvements include increasing the cladding melting point and its strength at high temperature in comparison to current Zircaloy cladding. Improvements to the fuel are also under investigation, especially in the areas of improved fission gas retention and higher temperature margin to fuel melt. Today; there are many active programs on ATFs being carried by many companies, institutions and organizations all around the globe.

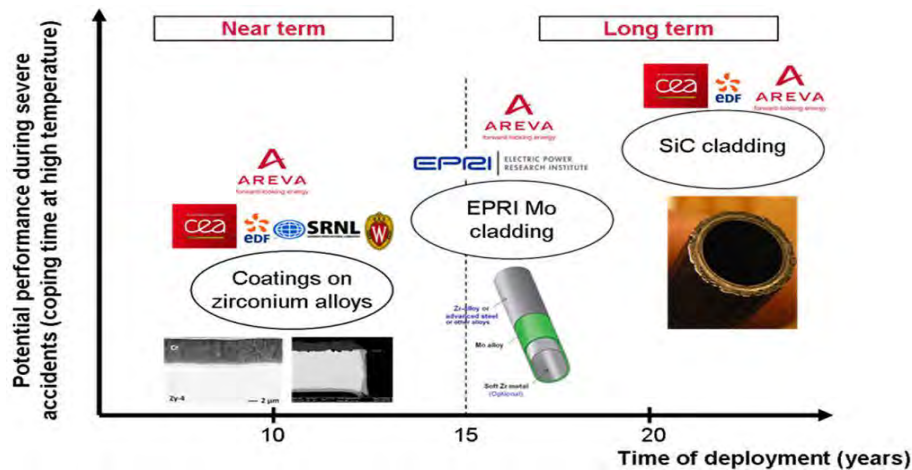


Fig. 2. Potential performance as a function of development time for the various cladding concepts

Investigated by AREVA [6].

Review of Proposed ATF cladding concepts

Coated Zr-based cladding

An immediately obvious and evolutionary approach to ATF cladding is the adoption of a protective coating on the surface of Zr-based alloys. Thin coatings are expected to have a minimal effect on the thermo mechanical behavior of Zr-based cladding, assuming sufficient creep and limited strain mismatch are engineered into the coating. It is necessary that the coating be adherent to and chemically stable with the Zr-based cladding substrate during normal operation and off-normal conditions, protecting it from rapid oxidation during beyond DBAs. Materials that are capable of exhibiting high temperature steam oxidation resistance are Chromia, Alumina, and/or Silica formers. Therefore, any ATF coating technology needs to contain at least one of the elements Cr, Al, or Si [7].

Development status

Several organizations have initiated studies of coatings on monolithic Zr-based alloys over the last five years with the goal of enhanced accident tolerance. The coatings studied thus far broadly fall within two categories [8]:

Metallic coatings:

Pure Cr (AREVA/CEA/EDF, the Korea Atomic Energy Research Institute [KAERI], and University of Illinois Urbana-Champaign [UIUC]);

Cr alloys: Cr-Al binary alloy (KAERI and UIUC);

FeCrAl and Cr/FeCrAl multi-layer (KAERI and UIUC). For FeCrAl or iron-based alloys, a barrier layer is needed at the coating/substrate interface to prevent the formation of Zr-Fe eutectic at around 900°C. In the KAERI concept, a barrier layer of Cr or Cr-Al alloy is considered

Ceramic coatings:

Nitrides: CrN, TiN, TiAlN, CrAlN or multi-layers of different nitrides (IFE/Halden, The Pennsylvania State University [PSU]);

MAX phases: Ti₂AlC, Cr₂AlC, Zr₂AlC, Zr₂SiC (KIT, AREVA).

The most widely explored coating technologies on Zr-based alloys to date are the ones that form chromia. Specifically, Cr metal, CrAl, and CrN coatings have been studied. In the case of a metallic Cr coating with a thickness of a few to tens of micrometers (Figure 3), the resulting chromia that forms under both aqueous and high temperature steam conditions protects the underlying Zr metal. Multiple experiments to further evaluate the performance of this technology are currently ongoing, with preliminary ion irradiation data indicating adequate behavior [7].

Coatings that are meant to form alumina or silica have predominantly manifested as MAX phase compounds or FeCrAl in the case of the former. Ti₂AlC, TiAlN, Ti₃SiC₂, and Cr₂AlC as MAX-phase coatings have been examined, although none of these examinations to date have produced a complete assessment of coating performance under normal operation, DBA, and beyond DBA conditions. The FeCrAl coating, although adequate for normal operating conditions, forms a eutectic with Zr at temperatures <1200°C and is not deemed a useful ATF coating [7].

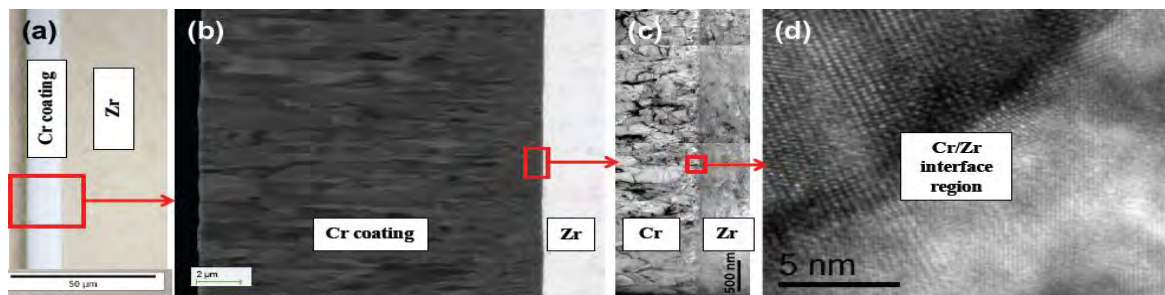


Fig. 3[7]: Metallic Cr coating of thickness 12–15 µm deposited on the surface of Zr-based cladding. The coating appears to be fully dense and homogeneous while the Zr–Cr interface shows good metallurgical bonding without indications of cracks or voids.

Challenges

The same attribute that makes surface coatings on Zr-based alloys the most viable near-term: ATF cladding technology is also its biggest performance challenge, for a LOCA; even a design basis LOCA, rod ballooning and burst occurs at temperatures as low as 700°C. This exposes at least some fraction of the cladding's internal surface to the oxidizing environment, even though the outer surface may be protected by the coating. A recent and ongoing effort aims to tackle this issue by adding an inner surface coating.

Need for elucidation of beyond DBA behavior: Except for a few studies, none of the research groups to date have exceeded the temperature limit of the design basis LOCA scenario (1204°C [9]). Owing to the R&D programs, several emerging coating technologies hold abundant promise for improving fuel performance during normal operation (Cr, CrN, and TiN). However, beyond DBA testing must be conducted on ATF cladding technologies to showcase their improvements in cladding performance.

FeCrAl cladding

Fe-based alloys have been used as nuclear fuel cladding since 1951, when the Experimental Breeder Reactor I (EBR-I) first went critical with austenitic stainless steel-clad Mark-I fuel assemblies. Austenitic stainless steels used as cladding in BWRs were eventually replaced with Zr-based cladding due to the stress corrosion cracking (SCC) failure experienced in high-oxygen-activity coolant environments of the pre-1990s era without water chemistry control. Although austenitic steel clad fuel operated reliably in PWRs [10], the drive to achieve higher burn ups, and by extension better economics, also facilitated their eventual replacement by Zr-based cladding. Ferric steels, having a BCC structure as opposed to the Ni-stabilized FCC (face-centered cubic) structure of austenitic Fe-based steels, are known to exhibit better SCC resistance but were never adopted for use in commercial LWRs. After survey tests examining a variety of candidate Fe-based alloys, re-examination of oxidation-resistant Fe-based alloys for LWR application was proposed [10].

Development status

Dedicated R&D programs in the United States and Japan are pursuing FeCrAl cladding as an ATF cladding technology. Although the main focus of the former program is on developing wrought oxidation-resistant alloy variants, the Japanese effort intends to also greatly improve on the strength by pursuing oxide dispersion strengthened (ODS) FeCrAl alloys. Accordingly, systematic studies of the critical Al and Cr contents in the alloy system were performed to identify the necessary combination for adequate steam oxidation resistance up to ~1500°C] while minimizing the potential for induced embrittlement or weld-initiated cracking. Furthermore, the effects of alloy composition on its melting point, oxidation beyond melting point, and compatibility with other fuel assembly constituents have been examined. As-fabricated strength and ductility in FeCrAl alloys could be tuned by controlling alloy composition to achieve values comparable to or higher than Zr-based alloys. Mechanical properties after neutron irradiation, for dose re-

gimes relevant to LWR fuel cladding have been quantified. Corrosion behavior of these alloys in LWR coolant environments has been examined [7].

Challenges

Poor thermal neutron utilization factor: The magnitude of neutron absorption cross section of natural Fe and Cr is roughly ~4–6% of thermal neutrons being absorbed in the cladding vs. ~1% absorbed in the Zr-based cladding. To compensate for this absorption, fuel enrichment may be increased and/or cladding thickness reduced and pellet diameter extended, either cases, extra cost will be added; resulting in a ~15- 25% increase in fuel bundle costs [10].

Potential for increased tritium release: Another challenge that requires further understanding and resolution is the permeability of BCC ferric alloys to Hydrogen isotopes, specifically tritium that forms as a result of ternary fission in the fuel. In the case of ferric alloys, permeability roughly two orders of magnitude higher than that of Zr-based alloys and twice that of austenitic Fe alloys has been reported, the full impact of increased release needs to be understood

Need for elucidation of beyond DBA behavior: A final consideration regarding the performance of this cladding material under beyond DBA conditions is crucial. While alumina formation, inherent to the bulk material, offers remarkable oxidation resistance up to ~1500°C, The integral behavior of the FeCrAl-clad pin and fuel assembly beyond this point is not well understood [7].

SiC/SiC cladding

(SiC/SiC) technology has yielded engineering materials used today in highly demanding applications, such as components in commercial jet engines. Remarkably, the technology originated from nuclear-energy-related R&D in 1970s. Application of bulk SiC in fission energy systems dates back even further and persist to this day, with its use as a constituent of coated fuel particles in high temperature gas-cooled reactors. Today; new generations of SiC fiber and methods of composite production that yield nuclear grade SiC/SiC are available. Nuclear-grade SiC/SiC is defined as composites utilizing Generation - III SiC fibers with chemical vapor infiltrated (CVI) or nano-infiltration transient eutectic phase (NITE) SiC matrices. Owing to their exceptional oxidation resistance and high temperature strength, far surpassing Cr-coated Zr-based or FeCrAl cladding materials, SiC/SiC composites are deemed the ideal ATF cladding material [7].

Development status

With the high temperature strength and oxidation resistance of SiC-based materials recognized, their application as LWR fuel cladding was proposed early on. Two distinct production routes for SiC/SiC cladding are in use today, the isothermal CVI methodology is the most common approach for cladding production used in the United States, France, South Korea, and Japan. This method results in a highly pure and crystalline, and therefore highly radiation-stable, composite of relatively low density (with 10–25% porosity). The second route for production of SiC/SiC cladding, primarily pursued in Japan, involves the NITE process, with utilization of hot pressing for production of cladding also demonstrated elsewhere. This methodology delivers a dense material with improved mechanical properties. A combination of the CVI and NITE techniques for nuclear-grade SiC/SiC production has also been suggested [11].

Besides establishing a robust production methodology, the focus of the past decade's development efforts has been fixated on understanding and quantifying the mechanical behavior of SiC/SiC composite tube structures, developing radiation-stable joining methods, and neutron irradiation testing of these composites. Since thermal neutron absorption is even lower in SiC than in Zr, such a transition is accompanied by an immediate advantage towards better fuel utilization in LWRs [7].

Challenges

Two critical issues, namely aqueous corrosion and fuel cladding failure due to micro cracking (Potential for radionuclide release) during normal operating conditions, and the Need for elucidation of beyond DBA behavior were identified as key areas for further examination.

Conclusions

Enhancing tolerability of nuclear fuel cladding to withstand severe accidents will definitely lead to increased nuclear power plants safety, since cladding represents the first barrier against the release of radioactive materials, which in its turn will increase the reliability of using nuclear power and reduce the dependency on fossil fuel. Many organizations and research groups all around the world are currently actively involved in developing ATF systems for near and long term deployments. Coatings technologies are considered as the most promising for the near term deployment. Owing to their exceptional oxidation resistance and high temperature strength, far surpassing Cr-coated Zr-based or FeCrAl cladding materials, SiC/SiC composites are deemed the ideal ATF cladding material in the long term.

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