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Neutronic evaluation of coating and cladding materials for accident tolerant fuels

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ABSTRACT

In severe accident conditions with loss of active cooling in the core, zirconium alloys, used as fuel cladding materials for current light water reactors (LWR), undergo a rapid oxidation by high temperature steam with consequent hydrogen generation. Novel fuel technologies, named accident tolerant fuels (ATF), seek to improve the endurance of severe accident conditions in LWRs by eliminating or at least mitigating such detrimental steam-cladding interaction. Most ATF concepts are expected to work within the design framework of current and future light water reactors, and for that reason they must match or exceed the performance of conventional fuel in normal conditions. This study analyzed the neutronic performance of ATF when employed in both pressurized and boiling water reactors. Two concepts were evaluated: (1) coating the exterior of zirconium-alloy cladding with thin ceramics to limit the zirconium available for reaction with high-temperature steam; (2) replacing zirconium alloys with alternative materials possessing slower oxidation kinetics and reduced hydrogen production. Findings show that ceramic coatings should remain 10–30 μm thick to limit the neutronic penalty. Alternative cladding materials, with the exception of SiC, enhance neutron loss compared to zirconium-alloys. An extensive parametric analysis concluded that reference performance metrics can be met by employing 300-μm or less thick cladding or increasing fuel enrichment by up to 1.74% depending on material and geometry.

1. Introduction

Many years of research and development on light water reactors (LWRs) cladding materials have produced zirconium alloys with slower oxidation kinetics, reduced hydrogen pick-up, and limited irradiation growth and creep (Terrani et al., 2013), but these advancements under normal operating conditions do not translate to beyond design-basis accident (BDBA) conditions. During severe accident conditions with loss of active cooling in the core, similar to those experienced at the Fukushima Daiichi Nuclear Power Plant, concerns surrounding zirconium-alloy cladding arise: embrittlement from hydrogen pick-up leading to possible fracture and fission product release; hydrogen generation from the exothermic reaction with steam; lack of coolable geometry due to cladding deformation (Erbacher and Leistikow, 1985). A large international effort is ongoing to engineer solutions that can enhance the capability of LWR fuel to withstand BDBA conditions by mitigating if not eliminating the detrimental interaction between zirconium alloys and steam, and as a minimum increasing the coping time for the reactor operators. These new solutions, designed for current and future LWRs, are referred to as accident tolerant fuels (ATF) (Zinkle et al., 2014). When compared to conventional UO_2/zirconium-alloy fuel, ATF concepts are expected to provide enhanced performance, reliability, and safety characteristics during off-normal conditions while maintaining or improving performance during normal conditions. Among the many ATF concepts those that can be deployed in existing LWRs are of particular interest: (1) using a thin protective ceramic coating on the zirconium-alloy cladding exterior; (2) replacing zirconium alloys with alternative cladding materials.

Thin ceramic coatings look to provide a protective layer for the zirconium alloy that will reduce oxidation and hydrogen pick-up during normal operating conditions while significantly retarding oxidation kinetics and hydrogen production during BDBA conditions. Under normal operating conditions the requirements for coatings are to: adhere to the zirconium alloy, self-heal if defected, remain stable with respect to spalling and grid-to-rod fretting, and provide stable properties under irradiation. MAX phases are ideal

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coating candidates since they possess both ceramic (corrosion resistance) and metallic (high thermal conductivity) properties. MAX phases have the general formula $M_{n+1}AX_n$ where n = 1, 2, or 3; M is an early transition metal; A is an A-group element; and X is carbon or nitrogen (Barsoum and Radovic, 2011). About 60 different MAX phase compositions exist with various desirable properties such as resistance to chemical attack, low thermal expansion coefficients, and creep resistance. The Industry Advisory Committee to the Idaho National Laboratory Advanced Light Water Reactor Fuel Development Program has proposed applying a 10–20 μm thick MAX phase (e.g. Ti_3AlC_2) layer to zirconium alloys using thermal and cold spray techniques (Bragg-Sitton, 2012). In addition to MAX phase, researchers at The Pennsylvania State University have proposed to use TiAlN as coating material (Liu et al., 2015). TiAlN is a nano-composite material of TiN and AlN that improves oxidation resistance, corrosion resistance, and hardness, but exhibits limited thermal conductivity (Deng et al., 2012). In a recent study (Khatkhatay et al., 2014), 1 μm thick $Ti_{0.35}Al_{0.65}N$ coatings were deposited onto Zircaloy-4 tubing and exposed to deaerated 500 °C and 25 MPa water for 48 h. After exposure, the coated tubes showed reduced oxidation compared to the uncoated tubes because of a robust oxide layer formed on the coating surface.

Cladding materials alternative to zirconium-alloys that exhibit lower oxidation kinetics in high temperature steam environment include ceramics, stainless steels, and advanced metallic alloys. SiC is a ceramic that provides a higher melting temperature, reduced oxidation and heat of oxidation, reduced hydrogen generation under off-normal conditions, low chemical activity, and a lower neutron absorption cross section (Griffith, 2011; George et al., 2015). SiC cladding is a relatively new concept; manufacturing technologies are still under development, a proven solution to hermetically seal the fuel rod end plugs is yet to be found (Yueh and Terrani, 2014), neutron irradiation induces a reduction in thermal conductivity (Katoh et al., 2014), and recent hydrothermal corrosion experiments showed significant mass loss of SiC in BWR-like conditions (Terrani et al., 2015). Ferritic stainless steel alloys, such as FeCrAl, have the ability to enhance oxidation resistance by creating a protective oxide layer; for instance, the aluminum found in FeCrAl combines with oxygen to produce an oxide layer (Al_2O_3) capable of reducing oxidation kinetics in high temperature steam environments (Terrani et al., 2013). Austenitic stainless steels, such as Alloy 33 (Pint et al., 2013), offer resistance to highly oxidizing media, high yield strength and excellent toughness. TZM is an advanced molybdenum alloy that features high thermal conductivity, low coefficient of expansion, and good ductility at low temperatures (Goldstein et al., 1988). Molybdenum has very poor corrosion resistance when exposed to high purity, high

Table 1
Properties of the PWR reference design.

Property	Value
Total Power, MW	3400
Assemblies	157
Core average coolant temperature, °C	303
Pressure, bar	155
Core average coolant density, °C	0.719
Active length, cm	427
Pellet diameter, cm	0.82
Clad thickness, cm	0.057
Fuel rod outer diameter, cm	0.95
Pitch-to-diameter ratio	1.326

Table 2
Properties of the BWR reference design.

Property	Value
Pellet diameter, cm	1.026
Clad thickness, cm	0.076
Gap thickness, cm	0.007
Pitch-to-diameter ratio	1.262
Bundle unit total width, cm	15.24
Inter-bundle gap width, cm	1.219
Water rod diameter, cm	2.590
Water rod cladding thickness, cm	0.076
Shroud thickness, cm	0.254
Full active length, cm	368.9
Partial active length, cm	243.8
Average linear heat rate, W/m	0.133

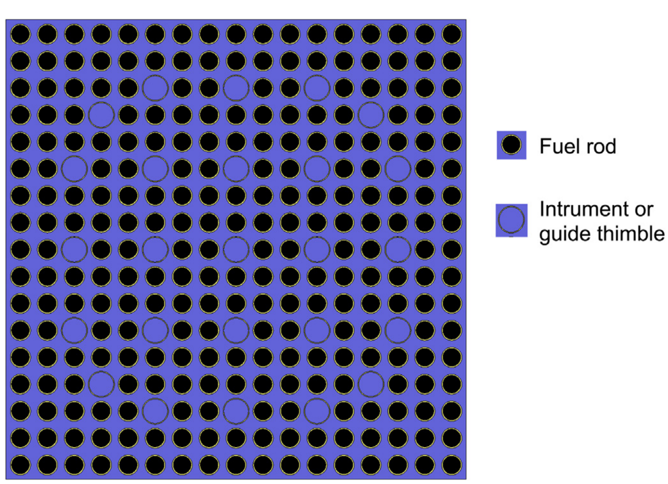

Fig. 1. Reference AP1000 assembly as reproduced in Serpent.

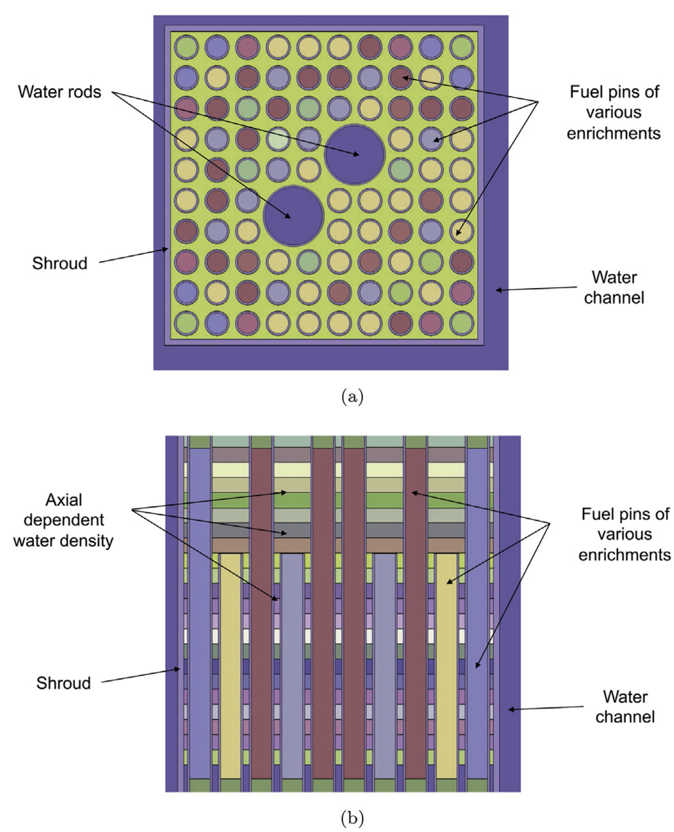

Fig. 2. BWR radial (a) and axial (b) profile as reproduced in Serpent.