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.

Keywords: accident tolerant fuel, light water reactors, ceramic coatings, cladding

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 [1], 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

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cladding deformation [2]. 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) [3]. 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 coating candidates since they possess both ceramic (corrosion resistance) and metallic (high thermal conductivity) properties. MAX phases have the general formula $\text{M}_{n+1}\text{AX}_n$ where $n = 1, 2, \text{ or } 3$; M is an early transition metal; A is an A-group element; and X is carbon or nitrogen [4]. 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 [5]. In addition to MAX phase, researchers at The Pennsylvania State University have proposed to use TiAlN as coating material [6]. TiAlN is a nano-composite material of TiN and AlN that improves oxidation resistance, corrosion resistance, and hardness, but exhibits limited thermal conductivity [7]. In a recent study [8], 1 μm thick $\text{Ti}_{0.35}\text{Al}_{0.65}\text{N}$ coatings were deposited onto Zircaloy-4 tubing and exposed to deaerated 500 $^\circ\text{C}$ and 25 MPa water for 48 hours. 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 [9, 10]. 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 [11], neutron irradiation induces a reduction in thermal conductivity [12], and recent hydrothermal corrosion experiments showed significant mass loss of SiC in BWR-like conditions [13]. 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 [1]. Austenitic stainless steels, such as Alloy 33 [14], offer resistance to highly oxidizing media, high yield strength and excellent toughness. TZM is an advanced molybdenum alloy features high thermal conductivity, low coefficient of expansion, and good ductility at low temperatures [15]. Molybdenum has very poor corrosion resistance when exposed to high purity, high temperature steam; therefore, reactor applications will likely require a thin tube molybdenum alloy coated with either zirconium alloy or advanced steels [16].

ATF concepts are expected to work within the framework of current and future LWRs; therefore, their viability besides improvements in BDBA conditions will strongly depend on the performance during normal operation. Among the advantages of zirconium-alloys is the very low neutron absorption. Most ATF concepts suggest replacing zirconium, partially or completely, with materials of higher neutron absorption and will negatively impact fuel cycle length and/or initial fuel enrichment. The purpose of this study is to assess the neutronic performance and evaluate design solutions to enhance such performance of previously described ATF concepts for use in both pressurized water reactors (PWRs) and boiling water reactors (BWRs). Section 2 describes the models used in the analysis. Section 3 illustrates the results obtained in terms of reactivity penalty, cycle length, and changes in fuel geometry and enrichment. Section 4 discusses the significance of these results.

2. Models and Methodology

The performance of ATFs was evaluated in PWRs and BWRs against that of conventional fuel. Neutronic calculations were performed using the Monte Carlo code Serpent [17]. The models used for the comparison are based on a single assembly and are described in this Section.

The PWR reference assembly is based on the AP1000 17×17 fuel element [18]. It contains 264 fuel rods, 24 guide thimbles, and 1 instrumentation thimble (Figure 1). The corresponding core parameters are shown in Table 1. Reflective boundary conditions on four sides generate an infinite radial lattice of finite length. Assembly extremities consist of axial reflectors made of a mix of steel and water representative of those regions (50%-50% volume mix, 100 cm length). ENDF/B-VII cross sections were applied at the following temperatures: 1,057 K for fuel, 637 K for clad, 600 K for coolant, and thermal scattering cross sections were applied to water. Reference UO_2 fuel was assumed enriched to 4.5 at.% and Zircaloy-4 (0.1% Cr, 0.21% Fe, 1.45% Sn, and 98.24% Zr; 6.56 g/cm^3) was chosen as clad material.

Table 1: Properties of the PWR reference design.

Property	Value
Total Power, MW	3,400
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

Figure 1: Reference AP1000 assembly as reproduced in Serpent.

The BWR reference assembly model is based on the GE14 10×10 design. Key parameters are summarized in Table 2 [19, 20, 21]. Such assembly contains 78 full length fuel rods, 14 partial length fuel rods (PLFR), and 2 water rods [19]. Four different axial regions were used to model uranium **enrichment** and gadolinium distributions [22]: (1) lower blanket, 15.24 cm long, composed of natural uranium; (2) lower dominant zone, 228.6 cm; (3) upper vanishing zone, 106.68 cm and PLFRs vanish; (4) upper blanket, 17.53 cm, composed of natural uranium blanket. Fuel rod cladding, water rod cladding, and channel box are made of Zircaloy-2 (0.12% O, 0.1% Cr, 0.1% Fe, 0.05% Ni, 1.4% Sn, 98.23% Zr). Water rod and channel box coolant are at inlet conditions, and remaining coolant was separated evenly into 24 axial zones of variable density [23]. Figure 2 shows radial and axial profiles of the reference BWR assembly. Reflective boundary conditions on four sides

create an infinite radial lattice of finite length. Assembly axial extremities contain a bottom reflector made of a water and steel **representative mix** (50%-50% volume mix, 100 cm length), and a top reflector constructed of a water/steam mixture (84% water and 16% steam), steam, and steel (35%-35%-30% volume mix, 100 cm length). ENDF/B-VII cross sections were applied at the following temperatures: 928 K for fuel, 629 K for clad, 600 K for coolant, and thermal scattering cross sections were applied to the water.

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

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

The Monte Carlo code Serpent was used for criticality and depletion calculations. Cycle length and attainable burnup were projected from the depletion data of a single assembly. **Core average reactivity over a cycle was calculated under the assumption of constant power throughout the assembly lifetime and a radial leakage of 3% was applied to correct for the infinite lattice model [23].** A three-batch and four-batch refueling scheme was assumed for the PWR and BWR, respectively. Depletion calculations employed Chebyshev Rational Approximation Method (CRAM) and a high-resolution flux spectrum based on the unified energy-grid structure to calculate one-group cross sections. All material compositions, except fuel composition, were assumed constant during irradiation unless otherwise specified. Reference cycle lengths were found to be 443.2 and 395.6 EFPD (effective full power days) for the reference PWR and BWR models, respectively.

3. Results

3.1. Ceramic coatings in PWR

Ceramic coatings were applied to the cladding exterior, including instrument and guide tubes, at thicknesses of 10, 50 and 100 μm ; clad outer diameter and thickness were kept constant. Coating materials chosen for this study include three MAX phases (Ti_3AlC_2 , Ti_2AlC , and Nb_2AlC), and $\text{Ti}_{0.5}\text{Al}_{0.5}\text{N}$ [24]. A fictitious coating made of Zircaloy-4 was also included in the analysis in order to quantify the affects of reduced moderation due to water displacement by the coating. Properties of candidate coating materials are given in Table 3.

Table 3: Properties of candidate coating materials and Zircaloy-4.

Material	Density, g/cm^3	Thermal conductivity, $\text{W}/(\text{m K})$
Ti_3AlC_2	4.50	40.0
Ti_2AlC	4.11	33.0
Nb_2AlC	6.50	29.0
$\text{Ti}_{0.5}\text{Al}_{0.5}\text{N}$	3.56	5.5
Zircaloy-4	6.56	20.5

Upon the addition of ceramic coatings, parasitic absorption within the coatings at beginning of life (BOL) increases linearly with thickness (Table 4). As a direct result, absorption and hence fission decreases in the fuel generating a reactivity penalty (Table 4). Such a penalty propagates through the fuel lifetime causing shorter fuel cycles (Table 5). Figure 3 shows the enrichment increase necessary to alleviate the reactivity penalty and match the reference cycle length. Coatings 10-30 μm thick are to be preferred to limit reactivity and cycle length loss, but thicker coatings remain acceptable as enrichment increases are minimal.

Table 4: Neutron absorption by the coating and reactivity change in PWR at beginning of life as a function of coating composition and thickness.

Coating material	Neutron absorption, %			Reactivity change, pcm		
	10 μm	50 μm	100 μm	10 μm	50 μm	100 μm
Ti_3AlC_2	0.17	0.86	1.70	-198	-905	-1,887
Ti_2AlC	0.15	0.76	1.51	-164	-832	-1,671
Nb_2AlC	0.12	0.60	1.18	-81	-628	-1,225
TiAlN	0.15	0.77	1.52	-130	-856	-1,704
Zircaloy-4	0.02	0.09	0.18	-10	-176	-428

Table 5: Cycle length penalty (EFPD) in PWR as a function of coating composition and thickness.

Coating material	Thickness, μm		
	10	50	100
Ti_3AlC_2	-3.7	-18.9	-39.8
Ti_2AlC	-3.7	-17.5	-35.0
Nb_2AlC	-2.8	-12.8	-28.0
TiAlN	-3.4	-16.8	-35.3
Zircaloy-4	-1.2	-4.4	-8.8

Figure 3: Additional enrichment necessary to match reference cycle length in PWR as a function of coating composition and thickness.

With changes to the fuel assembly geometry and composition its necessary to evaluate the impact on reactivity coefficients. Assuming 50 μm thick coatings, the fuel temperature coefficient (α_{fuel}) was calculated increasing the fuel temperature only by 100 K; the moderator temperature coefficient (α_{mod}) was calculated increasing moderator temperature by 10 K and modifying its density accordingly; coolant void coefficients were calculated decreasing coolant density by 5% (small void, α_{sv}) and 90% (large void, α_{lv}). Tables 6 and 7 show the results at beginning and end of life (EOL). All coefficients remain negative and similar to reference values.

Table 6: Reactivity coefficients in PWR at beginning of life for 50 μm coating as a function of composition.

Coating material	α_{fuel} , pcm/K	α_{mod} , pcm/K	α_{sv} , pcm/void %	α_{lv} , pcm/void %
Ti ₃ AlC ₂	-1.14 $\pm$ 0.07	-30.74 $\pm$ 0.66	-101.33 $\pm$ 1.33	-483.45 $\pm$ 0.18
Ti ₂ AlC	-1.03 $\pm$ 0.07	-30.20 $\pm$ 0.66	-102.04 $\pm$ 1.33	-483.88 $\pm$ 0.18
Nb ₂ AlC	-1.17 $\pm$ 0.07	-32.82 $\pm$ 0.66	-104.95 $\pm$ 1.32	-507.72 $\pm$ 0.18
TiAlN	-1.17 $\pm$ 0.07	-32.80 $\pm$ 0.66	-103.26 $\pm$ 1.33	-485.27 $\pm$ 0.18
Zircaloy-4	-1.08 $\pm$ 0.06	-29.73 $\pm$ 0.65	-99.45 $\pm$ 1.31	-488.43 $\pm$ 0.17
Ref.	-1.12 $\pm$ 0.06	-29.49 $\pm$ 0.65	-96.86 $\pm$ 1.30	-481.80 $\pm$ 0.17

Table 7: Reactivity coefficients in PWR at end of life for 50 μm coating as a function of composition.

Coating material	α_{fuel} , pcm/K	α_{mod} , pcm/K	α_{sv} , pcm/void %	α_{lv} , pcm/void %
Ti ₃ AlC ₂	-2.21 $\pm$ 0.23	-75.80 $\pm$ 2.30	-245.70 $\pm$ 4.61	-976.43 $\pm$ 0.67
Ti ₂ AlC	-2.94 $\pm$ 0.23	-79.52 $\pm$ 2.29	-250.96 $\pm$ 4.61	-976.33 $\pm$ 0.66
Nb ₂ AlC	-2.43 $\pm$ 0.23	-78.58 $\pm$ 2.29	-253.96 $\pm$ 4.60	-1019.62 $\pm$ 0.71
TiAlN	-2.29 $\pm$ 0.23	-71.69 $\pm$ 2.29	-244.17 $\pm$ 4.61	-979.49 $\pm$ 0.67
Zircaloy-4	-2.30 $\pm$ 0.23	-73.04 $\pm$ 2.27	-242.04 $\pm$ 4.55	-990.31 $\pm$ 0.67
Ref.	-2.54 $\pm$ 0.23	-73.46 $\pm$ 2.18	-241.46 $\pm$ 4.54	-987.11 $\pm$ 0.69

To ensure all zirconium-alloy surfaces are protected from high temperature steam, it may become necessary to coat spacer grids in addition to fuel rods. A model was set up to assess the effect of coating twelve grids [25] with 50 μm thick material. For simplicity grids were not modeled explicitly but uniformly mixed with the coolant. Each grid is 21.9 cm long, 21.9 cm wide, 3.81 cm tall, and grid walls are 0.05 cm thick [26, 27, 28]. Decreased moderation and excess absorption associated with coated grids enhances the cycle length penalty by about 25% relative to the case of uncoated grids (Table 8).

Table 8: Comparison of cycle length penalty (EFPD) in PWR with coated and uncoated spacer grids.

Coating material	Uncoated grids	Coated grids
Ti ₃ AlC ₂	-21.4	-25.5
Ti ₂ AlC	-19.2	-22.8
Nb ₂ AlC	-15.6	-20.4
TiAlN	-19.7	-23.1
Zircaloy-4	-5.9	-6.9

3.2. Ceramic coatings in BWR

In BWRs fuel cladding, water rod cladding, and shroud combine to significantly enhance the surface of zirconium-alloy to be protected compared to the PWR assembly. Ceramic coatings are applied to every surface in direct contact with water, i.e. exterior of the fuel cladding, interior and exterior of the water rods, and interior and exterior of the shroud, and have a pronounced effect on neutron economy. Excess absorption from the shroud, water rods, and their coatings (Table 9) produces substantial cycle length penalties (Table 10) approximately a factor of two larger than those found in the PWR (Table 5).

Table 9: Neutron absorption in the coatings of BWR core components at beginning of life as a function of coating composition and thickness.

Coating thickness, μm	Coating material	Coating absorption, %		
		Clad	Shroud	Water rod
10	Ti_3AlC_2	0.14	0.09	0.02
	Ti_2AlC	0.12	0.08	0.01
	Nb_2AlC	0.10	0.04	0.01
	TiAlN	0.12	0.08	0.01
	Zircaloy-2	0.01	0.01	0.00
50	Ti_3AlC_2	0.70	0.43	0.08
	Ti_2AlC	0.62	0.38	0.07
	Nb_2AlC	0.46	0.20	0.05
	TiAlN	0.62	0.37	0.07
	Zircaloy-2	0.07	0.03	0.01
100	Ti_3AlC_2	1.37	0.84	0.15
	Ti_2AlC	1.22	0.75	0.13
	Nb_2AlC	0.88	0.39	0.10
	TiAlN	1.22	0.73	0.14
	Zircaloy-2	0.13	0.06	0.01

Table 10: Cycle length penalty (EFPD) in BWR as a function of coating composition and thickness.

Coating material	Thickness, μm		
	10	50	100
Ti_3AlC_2	-6.2	-29.6	-63.5
Ti_2AlC	-5.5	-27.6	-55.6
Nb_2AlC	-2.8	-14.2	-31.3
TiAlN	-5.3	-27.0	-54.7
Zircaloy-2	-0.9	-3.3	-5.7

3.3. Alternative cladding materials in PWR

In the attempt to cover the variety of proposed alternative cladding solutions, this analysis included four candidate materials: SiC, FeCrAl, TZM, Alloy 33. Table 11 shows the density and composition of each candidate material. For a fair comparison, besides the alternate materials, all calculations were also performed with Zircaloy-4 cladding in PWR and Zircaloy-2 cladding in BWR.

Table 11: Density and composition (wt%) of cladding materials.

Material	Density, g/cm ³	C	N	Al	Si	Ti	V	Cr	Fe	Ni	Cu	Zr	Mo
SiC	3.10	30	-	-	70	-	-	-	-	-	-	-	-
FeCrAl	7.15	-	-	5.3	-	-	-	22.0	72.7	-	-	-	-
TZM	10.22	-	-	-	-	0.5	-	-	-	-	-	0.08	99.42
Alloy 33	7.90	0.01	0.4	-	-	-	-	33.0	33.39	31.0	0.6	-	1.6

To better understand how alternative cladding materials effect neutronic performance, the Zircaloy-4 clad was replaced with SiC, FeCrAl, TZM , and Alloy 33 clads of same thickness (571.5 μm). Among the four materials considered, three show enhanced neutron absorption and large reactivity penalties (Table 12)¹. SiC instead features reduced absorption compared with the reference. A parametric analysis was conducted to explore design options that would eliminate cycle length penalties (Table 12). For each candidate material two routes were considered: (1) changing the clad thickness and keeping the fuel enrichment constant; (2) varying the enrichment level with a constant clad thickness.

A variation in clad thickness implies an overall adjustment of fuel pin dimensions. A detailed design procedure for cladding made of the selected materials has not been established; therefore, three approaches were followed upon changing clad thickness: (1) the clad inner diameter (ID) was kept constant; (2) the clad outer diameter (OD) was kept constant whereas pellet and gap size were calculated under the assumption that the volume ratio between gap and fuel would remain as in the reference design, i.e. constant pellet-to-inner clad diameter ratio; (3) the clad OD, as well as the gap thickness were kept constant and the pellet diameter was allowed to change.

Detailed results for each material are presented in the following sections. The main findings are summarized in Table 13. SiC does not generate any significant neutron loss and the clad thickness could be increased

¹In order to track the numerous cases that have been considered we assigned to each case an identification code; a case consists of a unique combination of cladding material, pellet diameter, and gap and clad thickness.

Table 12: Neutronics effects of candidate cladding materials in PWR.

Case	Cladding material	Cladding absorption, %	Reactivity change, pcm	Cycle length change, EFPD
S1	SiC	0.36	+397	+11.5
F1	FeCrAl	6.62	-5,831	-115.3
T1	TZM	12.05	-10,532	-225.1
A1	Alloy 33	9.43	-8,780	-174.0
Ref.	Zircaloy-4	0.91	-	-

up to 720 μm and still match the reference fuel economy. All other alloys present enhanced neutron loss requiring a thinner clad in order to match the cycle length constraint.

Table 13: Clad thickness necessary to meet the cycle length constraint as function of clad material.

Material	Thickness, μm	
	Holding constant ID	Holding constant OD*
SiC	720	620
FeCrAl	130	270**
TZM***	160	280
Alloy 33	90	210

*Results are independent of the approach used to determine the gap thickness

**Can be increased to 300 μm enriching chromium

***With enriched molybdenum

Neutron absorption in SiC is smaller than in Zircaloy-4 and a slightly thicker clad could be employed (Table 14). Clad thickness can increase to 620 μm and HM load decrease by about 2% and still achieve acceptable cycle length. If the pellet diameter is kept constant, clad thickness can increase up to 720 μm . When increasing clad thickness, a reduced moderator-to-fuel volume ratio dominates reactivity loss rather than excess cladding absorption. This is evident when comparing case S2 and S3 with S1. In both cases clad thickness was doubled, but for S3 half is filled with SiC and half is void. Reduced moderation generates 76% of the reactivity drop derived from doubling the clad thickness. Figure 4 shows the change in neutron spectrum as a function of SiC clad thickness. **The most noticeable change is the hardening of the spectrum when clad thickness is doubled as in case S2.**

Table 14: Neutronics effects of SiC cladding in PWR.

Case	Compared to reference	Clad thickness, μm	HM load, g/rod	Clad absorption, %	Reactivity change, pcm	Cycle length change, EFPD
S1	no change	571.5	2,357	0.36	+397	+11.5
S2	same ID and gap	1,143.0	2,357	0.75	-2,075	-40.5
S3		1,143.0*	2,357	0.36	-1,492	-25.2
S4		720.0	2,357	0.48	-227	+0.1
S5**	same OD and gap	620.0	2,302	0.42	+505	-0.1
Ref.	-	571.5	2,357	0.91	-	-

*571.5 μm SiC and 571.5 μm void

**Results are independent of the approach used to determine the gap thickness

Figure 4: Neutron spectrum deviation from reference at beginning of life for selected SiC cases.

Replacing Zircaloy-4 with FeCrAl generates a reactivity penalty of 5,831 pcm at BOL and results in a cycle length penalty of 115 EFPD (Table 15). Such penalty is eliminated if the clad thickness can be reduced to 270 μm and the HM load increased by about 15%. If the pellet diameter is kept constant, the clad thickness must be reduced to 130 μm . Figure 5 shows the change in neutron spectrum as a function of FeCrAl clad thickness. **Enhanced thermal absorption results in a slightly harder spectrum for case F1. Smaller diameter pins as in case F5, instead, feature softer spectrum.**

Table 15: Neutronics effects of FeCrAl cladding in PWR.

Case	Compared to reference	Clad thickness, μm	HM load, g/rod	Clad absorption, %	Reactivity change, pcm	Cycle length change, EFPD
F1	no change	571.5	2,357	6.62	-5,831	-115.3
F2	same ID and gap	400.0	2,357	4.89	-3,510	-71.7
F3		300.0	2,357	3.71	-2,002	-44.8
F4		200.0	2,357	2.49	-639	-17.4
F5		130.0	2,357	1.63	-158	-0.2
F6	same OD and gap*	400.0	2,555	4.61	-4,350	-54.6
F7		300.0	2,673	3.38	-3,458	-12.3
F8		270.0	2,709	3.02	-3,211	+0.8
Ref.	-	571.5	2,357	0.91	-	-

*Results are independent of the approach used to determine the gap thickness

Figure 5: Neutron spectrum deviation from reference at beginning of life for selected FeCrAl cases.

Table 16 breaks down neutron absorption by FeCrAl components: ^{56}Fe (91.75% natural abundance) is the major absorber; ^{53}Cr (9.50% natural abundance) is the second larger absorption. In the attempt to reduce absorption in FeCrAl the effectiveness of eliminating (reducing to 0.1% of Cr) ^{53}Cr was tested. This resulted in a 300 μm thick clad meeting the cycle length requirement as opposed to a 270 μm thick clad. It remains to be evaluated if such improvement is enough to justify the cost of chromium ^{53}Cr depletion.

Table 16: Relative neutron absorption in FeCrAl by constituent isotopes.

Element	Natural abundance, at. %	Relative absorption, %
^{27}Al	100.00	1.01
^{50}Cr	4.35	6.13
^{52}Cr	83.79	5.92
^{53}Cr	9.50	15.17
^{54}Cr	2.37	0.08
^{54}Fe	5.80	3.70
^{56}Fe	91.72	66.11
^{57}Fe	2.20	1.55
^{58}Fe	0.28	0.12

TZM is a strong neutron absorber. More than 12% of neutrons are absorbed in the clad if Zircaloy-4 is replaced with TZM (case T1 in Table 17) resulting in a 225 EFPD cycle length penalty; therefore, it was

assumed that TZM can not be considered a viable LWR cladding material unless molybdenum is depleted of ^{95}Mo . In such case absorption in TZM clad decreases by a factor of 2.3 (case T2 in Table 17). During this analysis it was assumed that ^{95}Mo would be isotopically separated from the other molybdenum isotopes and its concentration reduced to 0.1%. Under these assumptions a TZM clad still generates a reactivity penalty of 3,689 pcm at BOL and a cycle length penalty of 87 EFPD. Such penalty is eliminated if the clad thickness can be reduced to 280 μm and the HM load increased by about 14%. If the pellet diameter is kept constant, the clad thickness must be reduced to 160 μm . Figure 6 shows the change in neutron spectrum as a function of TZM clad thickness. Trends are similar to those observed for FeCrAl.

Table 17: Neutronics effects of TZM cladding in PWR.

Case	Compared to reference	Clad thickness, μm	HM load, g/rod	Clad absorption, %	Reactivity change, pcm	Cycle length change, EFPD
T1	no change, natural Mo	571.5	2,357	12.05	-10,532	-225.1
T2	no change	571.5	2,357	5.23	-3,689	-87.1
T3	same ID and gap	400.0	2,357	3.82	-1,955	-49.8
T4		300.0	2,357	2.91	-958	-28.5
T5		200.0	2,357	2.00	58	-8.4
T6		160.0	2,555	1.63	391	-0.1
T7	same OD and gap*	400.0	2,673	3.94	-3,121	-39.8
T8		300.0	2,697	3.06	-2,705	-6.5
T9		280.0	2,559	2.88	-2,630	+0.3
Ref.	-	571.5		0.91	-	-

*Results are independent of the approach used to determine the gap thickness

Figure 6: Neutron spectrum deviation from reference at beginning of life for selected TZM cases.

Replacing Zircaloy-4 with Alloy 33 generates a reactivity penalty of 8,780 pcm at BOL and results in a cycle length penalty of 174 EFPD (Table 18). Such penalty is eliminated if the clad thickness can be reduced to 210 μm and the HM load increased by about 18%. If the pellet diameter is kept constant, the clad thickness must be reduced to 90 μm . Isotopic enrichment was not considered for Alloy 33 because it contains multiple high absorbing nuclides. Figure 7 shows the change in neutron spectrum as a function of Alloy 33 clad thickness. Trends are similar to those observed for FeCrAl.

Table 18: Neutronics effects of Alloy 33 cladding in PWR.

Case	Compared to reference	Clad thickness, μm	HM load, g/rod	Clad absorption, %	Reactivity change, pcm	Cycle length change, EFPD
A1	no change	571.5	2,357	7.02	-8,780	-174.0
A2	same ID and gap	400.0	2,357	7.02	-5,587	-114.5
A3		300.0	2,357	5.34	-3,638	-76.0
A4		200.0	2,357	3.62	-1,693	-39.2
A5		90.0	2,357	1.65	+368	+0.8
A6	same OD and gap*	400.0	2,555	6.64	-6,435	-99.9
A7		300.0	2,673	4.91	-5,009	-48.6
A8		210.0	2,782	3.40	-3,757	-0.4
Ref.	-	571.5	2,357	0.91	-	-

*Results are independent of the approach used to determine the gap thickness

Figure 7: Neutron spectrum deviation from reference at beginning of life for selected Alloy 33 cases.

A second parametric analysis was conducted searching for the fuel enrichment necessary to match the reference cycle length as a function of clad thickness. It was found that FeCrAl requires incrementing enrichment by 0.30% for every additional 100 μm starting from the minimum thickness established above; TZM (0.1% ^{95}Mo) requires about 0.25% per 100 μm , and Alloy 33 requires about 0.45% per 100 μm (Figures 8 and 9). Due to increased reactivity and thicker clad it is unnecessary to enrich fuel with SiC cladding.

Figure 8: Additional enrichment necessary to match reference cycle length in PWR as a function of cladding thickness when the ID is held constant.

Figure 9: Additional enrichment necessary to match reference cycle length in PWR as a function of cladding thickness when the OD is held constant.

Reactivity coefficients were also calculated to ensure comparable performance for candidate cladding materials at the reference thickness (571.5 μm). Fuel temperature coefficients are similar to the reference (Table 19). Moderator temperature coefficients, and small and large void coefficients experience larger deviations from the reference with TZM being the most significant (Tables 19 - 20). Molybdenum, specifically ^{92}Mo , ^{94}Mo , and ^{100}Mo , is a strong resonance absorber. Hardening of the spectrum by voiding leads to enhanced capture in the resonances and larger reactivity drop.

Table 19: Reactivity coefficients in PWR at beginning of life as a function of cladding material.

Case	Cladding material	α_{fuel} , pcm/K	α_{mod} , pcm/K	α_{sv} , pcm/void %	α_{lv} , pcm/void %
S1	SiC	-1.10 ± 0.06	-29.15 ± 0.64	-95.04 ± 1.28	-455.87 ± 0.17
F1	FeCrAl	-1.24 ± 0.08	-32.00 ± 0.76	-99.98 ± 1.51	-469.29 ± 0.18
T2	TZM	-1.22 ± 0.07	-41.86 ± 0.76	-132.13 ± 1.52	-643.75 ± 0.25
A1	Alloy 33	-1.55 ± 0.09	-32.58 ± 0.91	-105.64 ± 1.82	-482.26 ± 0.20
Ref.	Zircaloy-4	-1.12 ± 0.06	-29.49 ± 0.65	-96.86 ± 1.30	-481.82 ± 0.17

Table 20: Reactivity coefficients in PWR at end of life as a function of cladding material.

Case	Cladding material	α_{fuel} , pcm/K	α_{mod} , pcm/K	α_{sv} , pcm/void %	α_{lv} , pcm/void %
S1	SiC	-2.29 ± 0.22	-72.38 ± 2.23	-238.51 ± 4.49	-950.62 ± 0.64
F1	FeCrAl	-2.79 ± 0.25	-77.56 ± 2.51	-252.67 ± 5.05	-932.65 ± 0.68
T2	TZM	-2.46 ± 0.26	-93.90 ± 2.48	-295.28 ± 5.18	-1272.97 ± 0.94
A1	Alloy 33	-2.57 ± 0.26	-79.64 ± 2.64	-258.72 ± 5.51	-928.35 ± 0.68
Ref.	Zircaloy-4	-2.48 ± 0.23	-73.96 ± 2.18	-241.09 ± 4.54	-987.07 ± 0.69

3.4. Alternative cladding materials in BWR

The inclusion of large water rods and shroud drastically reduces reactivity and shortens cycle lengths for BWRs (Table 21) with the exception of SiC that experiences 18 EFPD longer fuel cycle. A BWR with TZM-made (0.1% ^{95}Mo) core components encounters a 50% reduction in cycle length. The cycle length for FeCrAl and Alloy 33 could not be calculated since criticality was never reached. Although these data are provided for completeness, extremely large penalties are unreasonable for power reactors and will require multiple alterations to fuel rod geometry and average assembly enrichment, but such tasks were beyond the scope of this work.

Table 21: Neutronics effects of candidate cladding materials in BWR.

Cladding material	Neutron absorption, %					Reactivity change, pcm	Cycle length change, EFPD
	Fuel	Clad	Water Rod	Shroud	Coolant		
SiC	92.92	0.39	0.02	0.41	5.86	+1,055	+18.3
FeCrAl	81.03	6.30	0.42	7.10	4.94	-16,388	-
TZM*	85.37	4.82	0.32	3.59	5.49	-7,757	-196.7
Alloy 33	76.43	8.66	0.60	9.50	4.64	-24,318	-
Ref.	92.06	0.90	0.06	0.75	5.80	-	-

*With 0.1% ^{95}Mo

4. Conclusions

Multiple design approaches for accident tolerant fuels are available, ranging from minimal cladding additions to complete fuel redesign. This study reviewed the neutronic impact of protecting conventional LWR cladding with thin ceramic coatings or replacing zirconium-alloy with alternative materials. It was found that when adding thin ($\leq 100 \mu\text{m}$) ceramic coatings, the corresponding reactivity penalties are small and proportional to the coating thickness, and can be alleviated if fuel enrichment is increased by 0.5% or less. Introducing coated spacer grids increases cycle length penalty by an additional 25%. Coatings 50 μm thick

show no major impact on reactivity coefficients as all values remain negative and similar to reference. Due to larger quantities of zirconium alloy and ceramic coatings, BWR reactivity and cycle length losses are a factor of two greater than in PWR. For materials considered in this study it is necessary to minimize the coating thickness (10-30 μm) to limit neutronic penalties.

Substituting FeCrAl, TZM, and Alloy 33 for zirconium-alloy cladding generates fuel cycles 115 to 225 EFPD shorter, whereas SiC improves neutron economy. A parametric analysis showed that FeCrAl, TZM, and Alloy 33 can match the reference cycle length when the cladding ID remains constant if the clad thickness is reduced to 130 μm , 160 μm , and 90 μm respectively. Meanwhile the clad thickness can be increased to 720 μm for SiC. When cladding OD remains at reference value, cladding thickness for FeCrAl, TZM, and Alloy 33 must be reduced to 270 μm , 280 μm , and 210 μm respectively and the HM load increased by 14-18%. SiC thickness, instead, can be increased to 620 μm and the HM load decreased by about 2%. A second parametric analysis determined that an increase of enrichment equal to 0.30% for FeCrAl, 0.25% for TZM, and 0.45% for Alloy 33 is necessary for every 100 μm of additional cladding thickness starting from the minimum values listed above. Although such increase can be considered small, it might lead to exceed the current commercial enrichment limit of 5%. When replacing BWR cladding, SiC lengthens the fuel cycle by 18 EFPD and TZM with 0.1% ^{95}Mo reduces it by a factor of two. Conversely, severe neutronic penalties stemming from FeCrAl and Alloy 33 prevented criticality.

This study assessed the requirements for ceramic coatings and cladding materials necessary to meet the neutronics performance of current claddings. The feasibility of manufacturing coatings or thin cladding and their mechanical performance remain to be established in future work. Furthermore, the improvements on current cladding in beyond design basis accident need to be carefully addressed.

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