Neutronic analysis for accident tolerant cladding candidates in PWR

Khalid A. Alamri [1,2,3], Meshari M. Alqathani,[4], and Abdullah I. Almarshad [5]

[1] *MS in Nuclear Engineering Program, College of Engineering, King Saud University, Riyadh 12372, Saudi Arabia;*

[2] *K.A. CARE Energy Research and Innovation Center at Riyadh, King Saud University, Riyadh 11421, Saudi Arabia.*

[3] *Engineering and Project Management Sector, King Abdullah City for Renewable and Atomic Energy (K.A.CARE), Riyadh 11451, Saudi Arabia.*

[4] *Nuclear Technologies Institute, King Abdulaziz City for Science and Technology (KACST), Riyadh 11442, Saudi Arabia.*

[5] *Chemical Engineering Department, King Saud University, Riyadh 12372, Saudi Arabia*

442105850@student.ksu.edu.sa, mmalqahtani@kacst.edu.sa, aalmarshad1@ksu.edu.sa.

Abstract –*The nuclear fuel performance during accidents became a critical issue after the Fukushima Daiichi nuclear accident in 2011. Currently, various research and development programs are being carried out to enhance the fuel's reliability and durability under such conditions. These programs are collectively known as the Accident Tolerant Fuel (ATF) R&D program, which involves multiple countries, research institutes, and fuel vendors. ATF is an enhanced fuel that can tolerate longer periods of active cooling system failure, without significant fuel/cladding system degradation. Moreover, it can improve fuel performance in normal operations, transients, as well as design-basis accident (DBA) and beyond design-basis (BDBA) scenarios. This paper presents a preliminary neutronics analysis for Accident Tolerant Fuel (ATF) cladding materials for a standard PWR fuel rod (fuel pellet and dimension). The candidate cladding materials were compared with the original Zircaloy-4 cladding material. To confirm the necessary geometry requirements for achieving end-of-cycle fuel reactivity, a parametric evaluation was conducted on fuel and cladding materials. The findings were then compared with the standard PWR reference fuel-cladding system. A number of reactor safety parameters are evaluated for the candidate cladding materials as reactivity, radial power distribution of fuel pellet, reactivity coefficients, spectral hardening. This study used OpenMC code to model two-dimensional space Standard PWR nuclear fuel rods and the neutronics and burnup analysis of ATF cladding materials.*

Keywords: Accident Tolerant Fuel (ATF), Zircaloy-4 and FeCrAl cladding materials, OpenMC code.

I. Introduction

The Fukushima Daiichi nuclear incident in 2011 underscored the critical importance of nuclear fuel performance during accidents. As a response to this concern, extensive research and development initiatives are presently underway to enhance the reliability and durability of fuel under such challenging circumstances. These collective efforts are referred to as the Accident Tolerant Fuel (ATF) Research and Development (R&D) program, involving numerous countries, research institutions, and fuel suppliers. ATF represents an advanced type of fuel capable of enduring extended periods of active cooling system failures with minimal degradation to the fuel/cladding system [1]. Additionally, it has the potential to enhance fuel performance in routine operations, transient events, as well as design-basis accidents (DBA) and scenarios beyond design-basis (BDBA) [1].

Zirconium alloys are known for their inherent resistance to various environmental conditions, making them a popular choice as cladding materials in light water reactors [2,3,4,5]. Despite their advantages, such as excellent neutron economy and low capture cross sections, these alloys exhibit reduced resistance to oxidation at elevated temperatures during reactor operation. This leads to increased hydrogen absorption, impacting the material's microstructure and causing a loss of ductility over time [2,3,4,5]. Since 2011, extensive research has explored alternative cladding materials capable of replacing Zirconium [6]. These investigations encompass a wide array of studies on mechanical properties, irradiation behavior, corrosion resistance, including reactions with water, and their interaction with fuel [7].

FeCrAl has been proposed as a promising option for fuel cladding due to its favorable thermo-mechanical attributes, reduced reactivity with steam, and lower propensity for hydrogen generation [8]. Due to its substantial aluminum content, FeCrAl forms a robust Al2O3 layer during high-temperature oxidation, characterized by superior strength [9,10]. Al2O3 exhibits reduced permeability in comparison to ZrO2, resulting in significantly improved oxidation properties, especially in high-temperature steam environments [10].

We conducted analyses to gain initial insights into the neutron-related aspects of employing alternative fuel cladding concepts within pressurized water reactor (PWR) cores. This step is considered essential to provide valuable guidance for broader fuel development and qualification endeavors. While numerous alternative cladding materials are under scrutiny as potential ATF cladding concepts, this paper specifically focuses on a particular FeCrAl option. We compare the FeCrAl cladding material with the reference Zircaloy-4 clad fuel pin, utilizing data from depletion calculations, spectral analyses, reactivity coefficient calculations, and radial fission power assessments. Currently, the study's scope is limited to single fuel rod within a PWR; however, future work aims to expand these analyses to the assembly-level or full-core scale.

II. Methodology and Input Parameters

II.A. Pin Cell Model

The pin cell model was generated using the OpenMC code, which is an open-source Monte Carlo code developed by the MIT Computational Reactor Physics Group (CRPG) [11]. This code has the capability to perform fixed source, k-eigenvalue, and subcritical multiplication calculations for models constructed using either constructive solid geometry or computer-aided design (CAD) representations [12]. Neutronic and burnup calculations were conducted using the OpenMC code [11]. For this analysis, the ENDF/B-VII.1 library [13] was utilized, along with the simplified CASL PWR depletion chain consisting of 255 nuclides [14], owing to its notable accuracy in thermal spectrum reactors. Our analysis utilized a total of 40 million particles, consisting of 400 active cycles and 100 inactive cycles, resulting in a standard deviation of approximately 13 pcm for the multiplication factor k. Figure 1 illustrates the pin cell model representing the reference case, based on a Westinghouse 17 × 17 PWR fuel rod.

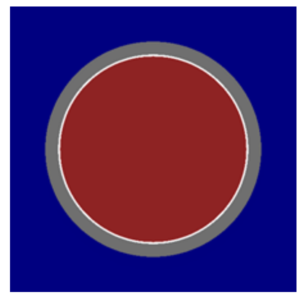


Fig. 1. Westinghouse 17 × 17 PWR pin cell model.

All the simulations are conducted using a single unit cell, with the primary neutronic parameter under investigation being the infinite neutron multiplication factor. To ensure a consistent power transfer, the pitch-to-rod-diameter (P/D) ratio remains fixed at 1.326. Additionally, the gap between the pellet and cladding is held constant at 82.55 μm to preserve the gap's thermal conductivity. Adjustments to the cladding thickness are accomplished by modifying both the pellet radius and the inner diameter of the cladding. An average concentration of 630 ppm boron, equivalent to the concentration at the Middle of Cycle (MOC). Detailed information on the remaining parameters can be found in Table 1

Table 1 The parameters and dimensions of Westinghouse 17 × 17 PWR assembly

Property	Value	Ref.
Pitch-to-rod-diameter ratio	1.326 cm	15
Number of fuel rods per assembly	264	16
Assembly fuel height	365.76 cm	16
Fuel pellet radius	0.409575 cm	16
Gap thickness	82.55 μm	16
Fuel enrichment	4.9%	16

Cladding inner radius	0.41783 cm	17
Cladding thickness	0.05715 cm	17
Cladding outer radius	0.47498 cm	17
Number of guide tubes	25	17
Cladding inner radius of guide tube	0.5624 cm	17
Cladding outer radius of guide tube	0.6032 cm	17
Fuel density UO2	10.47 g/cm^3 (96% theoretical density)	15
Specific power density for reference UO2-Zr	38.33 Watt/gram	15
Coolant density	0.7119 g/cm^3	17
Helium density	1.625 g/L (2.0 MPa)	17
Simulation time	1565 EFPD	
Coolant temperature	580 K	
Fuel temperature	900 K	
Clad and gap temperature	600 K	
Boundary conditions	Reflective	

II.B. *Input Parameters*

Table 2 displays the elemental compositions for both a baseline zirconium alloy (Zircaloy-4) and an iron-chromium aluminum alloy (FeCrAl). Table 3 presents the density and the microscopic thermal neutron absorption cross-section (σ_a) associated with each cladding material.

Table 2 Cladding material compositions [15]

Material	Wt%				
	Fe	Cr	Al	Zr	Sn
Zircaloy-4	0.15	0.1		98.26	1.49
FeCrAl	75	20	5		

Table 3 Density and microscopic thermal neutron absorption for cladding material [15]

Material	Density (g/cm^3)	σ_a (barns)
Zircaloy-4	6.56	0.200
FeCrAl	7.1	2.43

II.C. Geometric and Enrichment Parameters

Table 4 presents cases that were utilized in reactivity calculations. Case 1 serves as the reference case, while the other cases aim to increase the concentration of heavy metals and fissile materials within the fuel rod. This is achieved by either expanding the pellet diameter at the expense of reducing cladding thickness (Cases 2–4) or by enhancing the uranium enrichment (Cases 5). It's worth emphasizing that the thickness of the FeCrAl cladding material has been conservatively reduced to 350 µm, aligning with historical practices of using iron-based alloys as fuel cladding in LWRs [10]. Additionally, it's important to take note that the specific power, measured in megawatts per metric kilogram of uranium (MW/KgU), corresponds to a constant power level of 0.0682 MW per fuel rod modeled throughout the depletion cycle [15].

Table 4 Various cases used for reactivity calculations

Name	**Case Number**				
	1 (ref)	**2**	**3**	**4**	**5**
Material	Zircaloy-4	FeCrAl	FeCrAl	FeCrAl	FeCrAl
Pellet OD [mm]	8.1915	8.1915	8.5345	8.6345	8.6345
Clad ID [mm]	8.3566	8.3566	8.6996	8.7996	8.7996
Clad OD [mm]	9.4996	9.4996	9.4996	9.4996	9.4996
Clad Thickness [µm]	571.5	571.5	400	350	350
U-Enrichment	4.9	4.9	4.9	4.9	5.5
Specific Power (MW/MTU)	38.33	38.33	35.31	34.5	34.5
Notes: OD = Outer Diameter; ID = Inner Diameter.					

III. Results and Discussion

III.A. Depletion k-infinity Results

Figure 2 displays the relationship between reactivity and Burnup (GWd/ton) in fuel rod for Westinghouse 17 × 17 PWR fuel rods geometry and cladding materials. Specifically, the discharge burnup values for Zircaloy-4 and FeCrAl were 36.52 and 28.52 GWd/ton, respectively. In Figure 3, you can observe the detrimental impact on neutronics associated with the utilization of FeCrAl as the cladding material. At a burnup level of 60 GWd/ton, FeCrAl incurred a reactivity penalty of -4250 pcm, primarily due to its larger neutron absorption cross section.

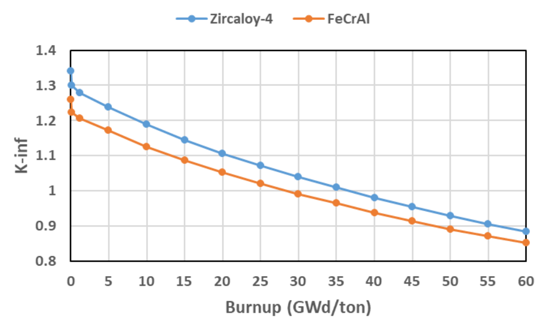


Fig. 2. Infinite multiplication factor for cladding materials.

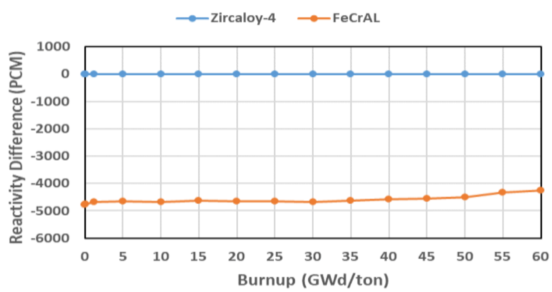
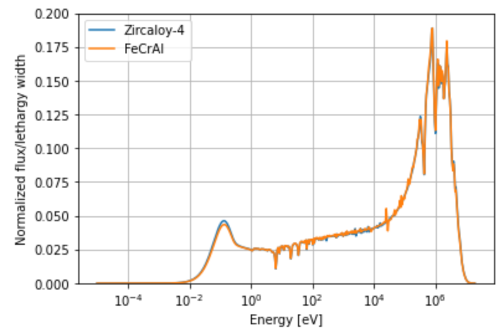


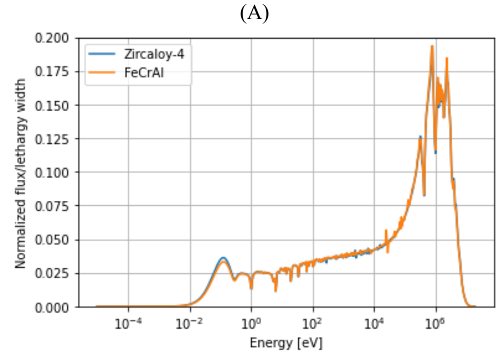
Fig. 3. Reactivity difference from Zircaloy-4 clad fuel versus burnup for FeCrAl cladding material.

III.B. Spectral Hardening

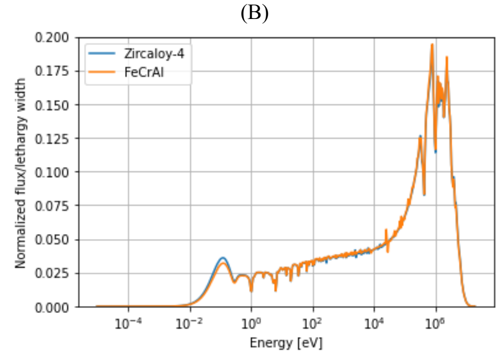
Spectral hardening was investigated for FeCrAl cladding material. Figure 4 illustrates the neutron flux spectrum at beginning of the cycle (BOC), Middle of Cycle (MOC) and End of Cycle (EOC). The increased neutron-absorption cross sections of the FeCrAl cladding material lead to a hardening of the thermal neutron flux spectrum. This spectral hardening effect is also observed during the transition from low burnup to high burnup conditions. In contrast, Zircaloy-4 cladding material, with its lower absorption cross section, leads to a higher inventory of thermal neutrons. On the other hand, FeCrAl cladding exhibits a significantly lower inventory of thermal neutrons due to its higher absorption cross sections. Since thermal neutrons play a critical role in inducing fission, this reduced thermal neutron presence results in a reduction in reactivity when spectral hardening occurs.



(A)



(B)



(C)

Fig. 4. Neutron flux spectrum at BOL (A), MOL (B), and EOL (C).

III.C. Self-shielding on the Fuel Rod

The self-shielding phenomenon within the fuel rod results in an uneven power distribution along its radial axis. This non-uniformity is primarily due to the strong absorption of U-238 near the fuel rod's surface, subsequently influencing both plutonium production and the burnup rate at this specific position. Consequently, it is of paramount importance to investigate the rim effect for FeCrAl. To perform a self-shielding analysis, the fuel region was subdivided into ten concentric rings with equal volumes, while keeping the gap and cladding components positioned externally, maintaining the same dimensions as presented in Figure 5. Figure 6 displays the normalized power distribution concerning the relative

radius. Due to spatial self-shielding effects, the relative fission power is highest near the fuel rod's surface and decreases toward the center of the pellet. At the BOC, although there exists some variation in neutron absorption capabilities between the two cladding materials, but it does not significantly affect the relative power within a fuel rod. During operation, more plutonium accumulates near the surface of the fuel pellet due to spatial self-shielding effects, leading to increased fission reactions in that region. As depicted in Figure 6, fission profiles remain relatively constant in the inner regions, while a sharp increase in power is observed near the fuel rod's boundary.
Notably, FeCrAl exhibits slightly higher relative power near the surface, as it produces more 239Pu in the outer ring due to spectral hardening. This observation underscores the fact that cladding materials with higher neutron absorption capabilities result in reduced reactivity during early life due to the hardened neutron spectrum, but increased reactivity as the EOC approaches due to greater plutonium accumulation.

III.D. Reactivity Coefficients

The study investigated Moderator Temperature Reactivity Feedback Coefficient (MTC), and the Coolant Void Reactivity Coefficient (VRC) for FeCeAl cladding material under the reference zircaloy-4. Meeting the safety standards of operational PWRs requires maintaining negative values for all three coefficients in each cladding material.

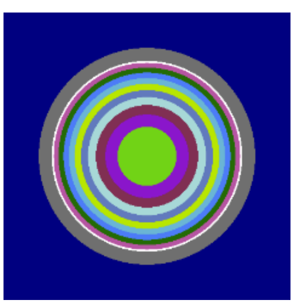


Fig. 5. Radial profile of a fuel-cladding system with fuel region divided into 10 rings.

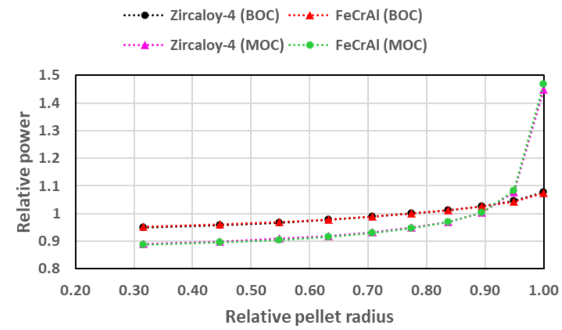


Fig. 6. Relative radial power distribution.

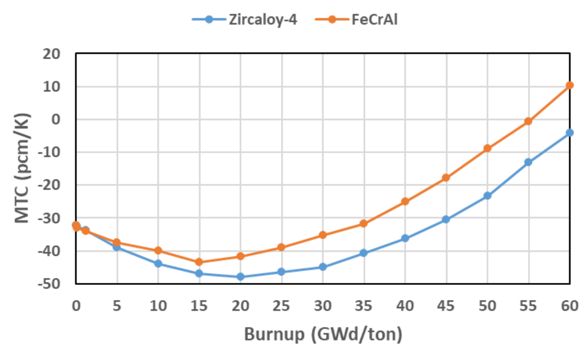


Fig. 7. The moderator temperature coefficient as a function of burnup.

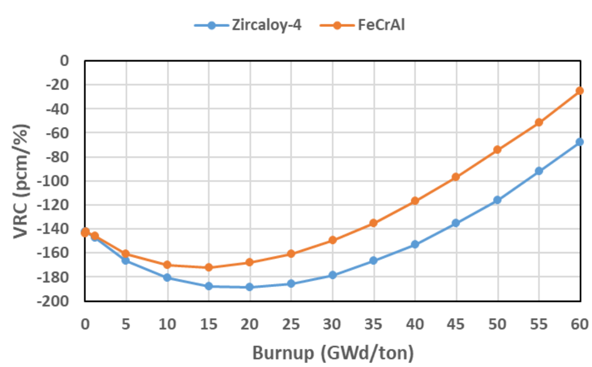


*Fig. **8**. The void reactivity coefficient as a function of burnup.*

The MTC is a parameter that quantifies the impact of changes in reactor coolant temperature on reactivity. It is defined as the change in reactivity per kelvin change in moderator temperature and is calculated using the following formula [18]:

$$\frac{\Delta\rho}{\Delta T} = \frac{k_{inf}(T_2) - k_{inf}(T_1)}{k_{inf}(T_1)\times k_{inf}(T_2)\times(T_2-T_1)} \times 10^5 \left[\frac{pcm}{k}\right] \quad (1)$$

where T_1 and T_2 are two moderator temperature values, k_{inf} (T_1) and k_{inf} (T_2) are corresponding criticality values.

The MTC is expressed in units of pcm per 1K. T_1 and T_2 were set to 580 K and 600 K, respectively, for the calculations. The densities of water at the given temperatures have been derived from Lemmon et al. [19]. The density of water at T_1 was found to be 0.7119 g/cm3, while at T_2 it was 0.66118 g/cm3.

The VRC is defined as the difference in reactivity over the void difference according to the following equation [20]

$$\frac{\Delta\rho}{\%void} = \frac{k_{inf}(V_1) - k_{inf}(V_0)}{k_{inf}(V_0) \times k_{inf}(V_1) \times (V_1 - V_0)} \times 10^5 \left[\frac{pcm}{\%void}\right] \quad (2)$$

V_0 and V_1 refer to the void fraction at nominal condition (%) and varied condition (%), respectively. K_{inf} (V_0) and K_{inf} (V_1) correspond to the criticality values, while the VRC value is measured in terms of pcm per %void. The VRC was calculated for 40% void (density of water = 0.4271 g/cm3).

Figure 7 illustrates how the moderator temperature coefficient (MTC) changes with burnup for cladding materials. Initially, at the BOC, both Zircaloy-4 and FeCrAl materials exhibited negative MTC values, with FeCrAl showing a more pronounced negativity compared to Zircaloy-4. This difference can be attributed to the high absorption cross-sections of the cladding material and the resulting hardening of the neutron spectrum. During operation, both Zircaloy-4 and FeCrAl materials experience a reduction in MTC until they reach values of 60 GWd/ton and 55 GWd/ton, respectively. Moving on to Figure 8, it depicts how the void reactivity coefficient (VRC) changes with burnup for the same cladding materials. Similarly, at the BOC, both Zircaloy-4 and FeCrAl materials displayed negative VRC values, with FeCrAl exhibiting more negative values than Zircaloy-4. Once again, the difference between FeCrAl and Zircaloy-4 is due to the high absorption cross-sections of the cladding material and the accompanying hardening of the neutron spectrum. During operation, both Zircaloy-4 and FeCrAl materials consistently maintain negative VRC values across all burnup phases.

III.E. U-235 enrichment and cladding thickness matching cycle length of Zircaloy-4

In this section, we conducted calculations to determine the required enrichment level and cladding thickness necessary for FeCrAl cladding to achieve a fuel cycle length equivalent to the reference case (Zircaloy-4). Table 5 presents the end-of-burnup values for different cases, including Case 1 for Zircaloy-4 and Cases 2 to 5 for FeCrAl. In Case 1, the end-of-burnup value for Zircaloy-4 was 36.52 GWd/ton, which FeCrAl must achieve to have an equivalent the cycle length of Zircaloy-4. Cases 2 to 4 involved maintaining the quantity of enriched uranium fuel at 4.9% while reducing cladding thickness from 571.5 to 350 μm and increasing the volume of UO2 fuel pellets. However, FeCrAl cladding still did not achieve the cycle length of Zircaloy-4. In Case 5, the thickness of FeCrAl cladding remained fixed at 350 μm, while the quantity of enriched uranium fuel was increased from 4.9% to 5.5%. This resulted in an end-of-burnup value of 36.45 GWd/ton for FeCrAl, which was close to the end-of-burnup value of Zircaloy-4. After numerous simulation iterations to match the end-of-burnup value of Zircaloy-4, the optimal enrichment level was determined to be 5.5084%.

Table 5 End of burnup values for various cases

Name	Case Number				
	1 (ref)	2	3	4	5
Material	Zircaloy-4	FeCrAl	FeCrAl	FeCrAl	FeCrAl
Clad Thickness [μm]	571.5	571.5	400	350	350
U-enrichment	4.9	4.9	4.9	4.9	5.5
Specific Power (MW/MTU)	38.33	38.33	35.31	34.5	34.5
End of burnup value (GWd/ton)	36.52	28.52	30.89	31.45	36.45
Effective full power days (EFPD)	952.88	744.03	805.96	820.53	950.87

IV. Conclusions

This study has presented a preliminary analysis of the neutron-related aspects of utilizing FeCrAl as an alternative fuel cladding material within pressurized water reactor (PWR). The study aimed to provide valuable insights for the Accident Tolerant Fuel (ATF) by comparing FeCrAl with the reference Zircaloy-4 clad fuel pin. Several key findings have emerged from this study:

Firstly, in terms of depletion k-infinity results, it was observed that FeCrAl exhibited a reactivity penalty of -4250 pcm at a burnup level of 60 GWd/ton when compared to Zircaloy-4. This penalty was primarily attributed to FeCrAl's larger neutron absorption cross section.

Secondly, the study investigated spectral hardening effects and found that the increased neutron-absorption cross sections of FeCrAl led to a hardening of the thermal neutron flux spectrum. This spectral hardening effect reduced reactivity, as thermal neutrons play a crucial role in inducing fission.

Thirdly, an analysis of self-shielding on the radial power distribution revealed that FeCrAl exhibited slightly higher relative power near the fuel rod's surface due to spectral hardening effects. This observation underscores the fact that cladding materials with higher neutron absorption capabilities result in reduced reactivity during early life due to the hardened neutron spectrum, but increased reactivity as the EOC approaches due to greater plutonium accumulation.

Finally, the study determined the required enrichment level and cladding thickness for FeCrAl to achieve a fuel cycle length equivalent to Zircaloy-4. After numerous simulation iterations, the optimal enrichment level for a cladding thickness of 350 μm was found to be 5.5084%

This study provides valuable insights into the feasibility and challenges of adopting FeCrAl as an alternative fuel cladding material for PWRs. Future work in this area should aim to expand these analyses to the assembly-level or full-core scale to further evaluate the feasibility and performance of FeCrAl cladding in practical reactor systems.

Acknowledgments

I would like to extend my appreciation to the Master Degree in Nuclear Engineering program at the College of Engineering, King Saud University, as well as the generous support provided by King Abdulaziz City for Science and Technology (KACST) and King Abdullah City for Atomic and Renewable Energy (K.A.CARE) for their invaluable contributions to this research.

References

1. Development of Light Water Reactor Fuels with Enhanced Accident Tolerance. Report to Congress April 2015.
2. O. N. Pierron, D. Koss, A. T. Motta, and K. Chan, "The influence of hydride blisters on the fracture of Zircaloy-4" Journal of Nuclear Materials, 322, 21-35 (2003).
3. S. J. Zinkle, and G. S. Was, "Materials challenges in nuclear energy" Acta Materialia, 61, 735-758 (2013).
4. Z. Duan, H. Yang, Y. Satoh, K. Murakami, S. Kano, Z. Zhao, J. Shen, and H. Abe, "Current status of materials development of nuclear fuel cladding tubes for light water reactors" Nuclear Engineering and Design, 316, 131-150 (2017).
5. D. Diniasi, F. Golgovici, A. H. Marin, A. D. Negrea, M. Fulger, and I. Demetrescu, "Long-term corrosion testing of Zy-4 in a LiOH solution under high pressure and temperature conditions" Materials, 14, 4586 (2021).
6. L. J. Ott, K. R. Robb, and D. Wang, "Preliminary assessment of accident-tolerant fuels on LWR performance during normal operation and under DB and BDB accident conditions" Journal of Nuclear Materials, 448, 520-533 (2014).
7. Z.-X. Tan, and J.-J. Cai, "Neutronic analysis of silicon carbide cladding accident-tolerant fuel assemblies in pressurized water reactors" Nuclear Science and Techniques, 30, 48 (2019).
8. A. S. Alomari, P. Joshi, and K. L. Murty, "High-Temperature Burst Creep Properties of Nuclear-Grade FeCrAl Fuel Cladding" Conference Proceedings of the Society for Experimental Mechanics Series. Volume 2, Springer, Cham (2022).
9. B. A. Pint, et al. "High Temperature Oxidation of Fuel Cladding Candidate Materials in Steam-Hydrogen Environments" Journal of Nuclear Materials, 440, 420–427 (2013).
10. N. George, "Assessment of Reactivity Equivalence for Enhanced Accident Tolerant Fuels in Light Water Reactors" Ph.D. thesis, University of Tennessee–Knoxville. Knoxville, TN, (2015).
11. The OpenMC Monte Carlo Code, Available online: https://docs.openmc.org/en/stable/ (accessed on 8 November 2022).
12. P. K. Romano, et al. "OpenMC: A state-of-the-art Monte Carlo code for research and development. Annals of Nuclear Energy" 82, 90-97 (2015).
13. M. B. Chadwick, et al. "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data" Nuclear Data Sheets, 112, pp. 2887-2996 (2011).

14. K. S. Kim, "Specification for the VERA Depletion Benchmark Suite," Consortium for Advanced Simulation of LWRs, (2015).
15. N. George, K. Terrani, J. Powers, A. Worrall, and I. Maldonado, "Neutronic analysis of candidate accident-tolerant cladding concepts in pressurized water reactors" Annals of Nuclear Energy, 75, pp. 703-712 (2015).
16. Xu. Wu, T. Kozlowski, and J. D. Hales, "Neutronics and fuel performance evaluation of accident tolerant FeCrAl cladding under normal operation conditions" Annals of Nuclear Energy, 85, pp. 763- 775 (2015).
17. S. Chen, and C. Yuan, "Neutronic analysis on potential accident tolerant fuel-cladding combination U3Si2-FeCrAl" Science and Technology of Nuclear Installations, Volume (2017).
18. Z. Benrhnia, et al. "Analysis of reactivity control coefficients and the stability of an AP1000 reactor assembly fueled with (Th-233 U) O2 using DRAGON code". Annals of the University of Craiova, 32, P. 88-102 (2023).
19. Eric W. Lemmon, Ian H. Bell, Marcia L. Huber, Mark O. McLinden, in: P.J. Linstrom, W.G. Mallard (Eds.), NIST Chemistry WebBook, NIST Standard Reference Database Number 69, National Institute of Standards and Technology, Gaithersburg MD, 20899, 2022, https://doi.org/10.18434/T4D303, Chapter Thermophysical Properties of Fluid Systems. (Accessed 20 July 2022) from.
20. Y. FUKAYA, Y. NAKANO, and T. OKUBO, "Study on Characteristics of Void Reactivity Coefficients for High-Conversion-Type Core of FLWR for MA Recycling" Nuclear Science and Technology, 46, pp.819-830 (2009).