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Accident Tolerant Fuel Cladding Materials for Light Water Reactors: Analysis of Neutronic Characteristics

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Abstract. Nuclear reactor fuel is the basic component of all safety requirements associated with nuclear energy production. The neutronic characteristics of light water reactors with candidate accident-tolerant fuel cladding materials were investigated in this work to improve nuclear safety. The materials were chosen for their high-temperature strength, radiation resistance, and corrosion resistance, which will contribute to nuclear safety. The research was conducted using a deterministic neutronic code for reactor design and analysis. The neutronic properties of a light water reactor system with candidate cladding materials were analyzed and compared to those of the reference. The reference cladding material was Zircaloy-4, and the candidate fuel cladding materials examined were FeCrNi alloy, oxide dispersion strengthened steels of $FeCrY_2O_3$ and $FeCrZrO_2$. The eigenvalues of the reactor were computed at various fuel temperatures and burnup stages. The results of the study revealed that employing candidate cladding materials led to a slightly hardened neutron spectrum, reducing initial excess reactivity and resulting in lower fuel burnup. This can be compensated by adding fuel enrichment up to 6%., which yields burnup value of 42000 MWd/T, which is higher than the reference of 40000 MWd/T with a reduced gap in initial excess reactivity from 28.4% to 25.4%. The candidate fuel cladding materials also showed better than reference reactivity loss properties in case of unwanted temperature increase. This demonstrated that the examined improved tolerant materials have the ability to increase the inherent safety of nuclear reactors.

Keywords: ATF cladding; Doppler coefficient; Neutronic characteristics; Nuclear safety; Oxide dispersed strengthened

1. Introduction

Nuclear energy is the most modern energy source, capable of producing massive amounts of clean energy safely and reliably. This is required to run modern industrial societies economically and sustainably, both environmentally and in terms of the availability of resources. As a result, nuclear energy should play a significant part in the necessary transformation of the 21st-century energy-supply system. Moreover, modern technology can support the advancement of a sustainable energy sector that is capable of efficient production, management, and control. This can help strike a balance between

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economic progress, effective ecosystem management, and environmental conservation (Berawi, 2021; Berawi *et al.*, 2020; Brook *et al.*, 2014).

Following the core melt accident at the Fukushima Daiichi nuclear power plant, there has been a growing interest in novel approaches to improving nuclear safety. Existing knowledge must integrate with modern, environmentally friendly innovation in safety technology (Yadav, Pal, and Karthikeyan, 2023; Rivai *et al.,* 2022). High-temperature water vapor interaction with zirconium alloy cladding employed in the light water reactor (LWR) exacerbated the accident, resulting in the discharge of volatile hydrogen gas. Researchers have since been studying ways to improve reactor safety in the event of a loss of coolant, such as the feasibility of using accident-tolerant nuclear fuel cladding options (Prianka and Prodhan, 2024; Takeda *et al.,* 2016). Different potential cladding materials for light water reactors have been investigated as alternatives to the zirconium alloys widely used in LWRs. Nuclear energy's viability as a clean and safe source of energy for many developing countries will undoubtedly increase as technology advances and delivers reliable and uninterrupted electrical energy systems to the consumers with appropriate technological conditions for all parts of the electric networks (Gracheva *et al.*, 2020, Antariksawan *et al.*, 2017, George *et al.*, 2015, Soentono and Aziz, 2008).

Because of its low parasitic neutron absorption, zirconium alloy fuel cladding has been widely used since the inception of commercial nuclear power plants (Thilagam and Mohapatra, 2023). However, in a design-based accident scenario, a condition may result in a reduction in safety margins. It is widely acknowledged that the outcome of a severe accident scenario in a Light Water Reactor (LWR) is primarily influenced by the operation and availability of safety systems. Given this feature, many researchers are collaborating to investigate new fuel and cladding concepts that provide better safety margins (Chen and Yuan, 2017; Dobuchi, Takeda, and Kitada, 2016; Barrett, Bragg-Sitton and Galicki, 2012).

The accident tolerant fuel (ATF) cladding concept refers to approaches to developing new types of fuel cladding materials with improved properties at higher temperatures. In this concept, zirconium alloy is modified or replaced with another high-performance oxidation-resistant material to improve nuclear safety. The potential cladding materials should also have high-temperature stress resistance and low thermal neutron absorption cross-section for neutron economy (Dani *et al.,* 2023). Chromia, alumina, and silica formers are recognized for their resistance to high-temperature steam oxidation and low neutron capture cross-section. Consequently, any new cladding material must incorporate at least one of the elements Cr, Al, or Si (Sakamoto *et al.*, 2018; Terrani, 2018; Eiselt *et al.*, 2016; Pouchon *et al.*, 2005).

Among the many alternatives, FeCrNi alloy and ODS (Oxide Dispersion Strengthened) stainless steels have been found to outperform Zircaloy-4 in mechanical performance and are thus considered among the most promising candidates (Panitra, Rivai and Aziz, 2022). In the event of an accident, the cladding materials can prevent a hydrogen explosion by suppressing hydrogen generation caused by the oxidation of zirconium-based alloys at high temperatures. They can also endure extremely corrosive conditions at high temperatures. Furthermore, they are resistant to void swelling and structural deformation. This study's use of $FeCrY_2O_3$ and $FeCrZrO_2$ as cladding materials is of special relevance because of the current effort to use local rare-earth elements such as yttrium, which may be found in bauxite residue from the aluminum industry and zirconium, which is abundant in Kalimantan (Kusrini *et al.*, 2020; Lu *et al.*, 2017; Younker and Fratoni, 2016; Pint *et al.*, 2015; Li *et al.*, 2013).

We previously deliberated on the synthesis and simulation of advanced materials for use as structures of nuclear reactors, such as for shielding and fuel cladding (Aziz *et al.*, 2020; Silalahi *et al.*, 2020; Aziz, Panitra and Rivai, 2018). In this paper, we investigated the neutronic characteristics of ATF cladding materials and compared their performance with reference material Zircaloy-4 for use in a standard 1000 MWe LWR nuclear power plant. The materials that were examined include FeCrNi alloy, $FeCrY_2O_3$ (ODS-1), and $FeCrZrO_2$ (ODS-2). Because of its lower parasitic capture cross-sections, Cr was preferred over Al or Si in this case. Analyses were carried out to gain insight into the neutronic aspects of their use as fuel cladding in LWRs. Reactor core eigenvalues calculated at various ^{235}U enrichment temperatures and burnup steps were used to study the neutronic characteristics. The goal is to assess the safety of using the candidate ATF cladding materials in LWRs concerning core neutronics. The current study is limited to analyzing a single fuel assembly level in an LWR. Future work should expand this analysis to include a full-core assessment.

2. Methods

The neutronic behavior of an LWR core was investigated using the SRAC (Standard Reactor Analysis Code) system, a deterministic nuclear reactor analysis code. SRAC is a versatile and widely used tool for different reactor core calculations. The nuclear data used was JEFF (Joint Evaluated Fission and Fusion File) produced through a collaboration of NEA Data Bank participating countries (NEA Data Bank, 2021; Okumura, Kugo and Tsuchihashi, 2007).

Figure 1 shows a representative diagram of a fuel assembly in a nuclear reactor. Geometric parameters and model description were based on the cylindrical model of a standard 17×17 fuel assembly of 1000 MW LWR, as shown in Figure 1a. In this investigation, control rods and instruments were assumed to be withdrawn from the reactor core. The simulation was based on a single unit of fuel assembly homogenized into a unit cell depicted in Figure 1b. The pitch-to-rod diameter was kept constant to maintain power transfer. The spacing between the pellet and the cladding was kept constant to maintain the thermal conductivity of the gap. The temperature in the fuel pellet, cladding, and moderator was varied in steps to investigate the effect of temperature on reactivity. The fuel pellet radius in this calculation was 4.12 mm, the homogeneous cladding plus gap thickness was 0.64 mm, the cladding outer radius was 4.76 mm, and the reference fuel enrichment was 4.12 percent. The ^{235}U enrichments in the fuel with the ATF cladding were varied in this calculation at 4.12 percent, 5.35 percent, 5.60 percent, and 6.00 percent to investigate their effect on core neutronics. Under normal operating conditions, the fuel temperature is 900 K, the cladding temperature is 600 K, and the coolant temperature is 562 K.

Table 1 The weight percentage of cladding materials used in this study

Zircaloy-4	Zr-Sn-Fe-Cr (balance, 1.5, 0.3, 0.2)
FeCrNi alloy	Fe-Cr-Ni (balance, 20.0, 10.0)
ODS-1	Fe-Cr-Y_2O_3 (balance, 10.0, 0.5)
ODS-2	Fe-Cr-ZrO_2 (balance, 25.0, 0.5)

The cladding composition in weight percent of Zircaloy-4, FeCrNi alloy, ODS-1, and ODS-2 is shown in Table 1. Table 2 shows the main parameters of the reference LWR under consideration. When evaluating the Doppler coefficient, α_F, of the LWR design, we used the parameters given in Table 3.

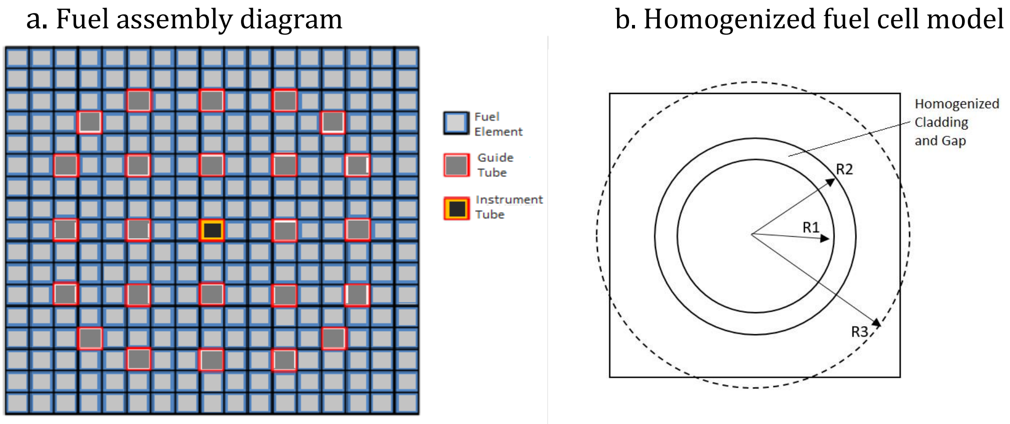

Figure 1 A representative diagram of standard 1000 MWe LWR 17x17 fuel assembly (a), and the model for the cell homogenization where R1, R2, and R3 refer to fuel pellet radius, outer radius of fuel cladding, and equivalent radius of coolant, respectively (b)

Table 2 The LWR main parameters used in this neutronic characteristics analysis

Thermal Power	3,000 MW
Electric Power	1,000 MWe
Fuel enrichment of ^{235}U	4.12 % (reference), 5.35%, 5.60%, and 6.00%.
Fuel type	UO_2
Fuel density	10.28 g/cc
Cladding	Zircaloy-4, FeCrNi alloy, ODS-1, ODS-2
Fuel Burnup	40000 MWd/T
Fuel cycle	3 years
Coolant/moderator	water
Core shape	cylindrical

Table 3 Parameters used in the Doppler coefficient calculation

Parameter	HZP	HFP
Fuel temperature, (K)	551.0	900.0
Cladding temperature, (K)	551.0	600.0
Moderator (coolant) temperature, (K)	551.0	562.0
Moderator (coolant) density (kg/m3)	766.0	748.0

The reactivity, ρ, of the LWR core, can be calculated from Equation 1.

$$\rho=(k-1)/k \tag{1}$$

where k is the neutron multiplication factor. If the value of k is larger than one, the reactivity is called excess reactivity (ρ_{ex}). The value of k, meaning the ratio of the number of neutrons produced in one generation to the number of neutrons absorbed in the preceding one, can be obtained from the core eigenvalue or criticality calculation using SRAC. Hence, reactivity implies a deviation of a multiplication factor from one, and excess reactivity may be used as a measure of a reactor's departure from criticality.

The safety performance of accident-tolerant cladding materials can be expressed in terms of the temperature coefficient of reactivity, α_F, shown in Equation 2. The term α_F represents the change in reactivity per unit change in fuel temperature (Kim and Jo, 2015).

$$\alpha_F = {}^{\Delta\rho_D}/_{\Delta T_F} \tag{2}$$

The $\Delta\rho_D$ is called Doppler defect, which is essentially the reactivity difference from hot zero power (HZP) to hot full power (HFP) conditions, described in Equation 3.

$$\Delta\rho_D = \frac{k_{inf}^{HFP} - k_{inf}^{HZP}}{k_{inf}^{HFP} \times k_{inf}^{HZP}} \tag{3}$$

where k_{inf}^{HFP} and k_{inf}^{HZP} are the effective multiplication factors corresponding to HFP and HZP conditions.

3. Results and Discussion

The SRAC calculations in this work were performed in 16 energy coarse groups, condensed from the original 107 groups. The energy grouping was done to expedite macroscopic constant generation in cell calculation. The structure of neutron energy grouping was emphasized in the area around thermal energy group regions (0.025 eV-1.0 eV). Figure 2 displays the neutron energy spectrum of the considered reactor, acquired from the SRAC cell burnup calculation. This result aligns well with the calculations from the SCALE code (Detkina *et al.*, 2020) and MCNPX calculation. Figure 2a shows the spectrum for the referenced LWR at the beginning of life (BOL) and end of life (EOL). Because the thermal to low-epithermal energy range is of importance in LWR, we compared and magnified the spectrum of the reference cladding with that of candidate cladding materials of FeCrNi alloy displayed in Figure 2b, ODS-1 in Figure 2c, and ODS-2 in Figure 2d.

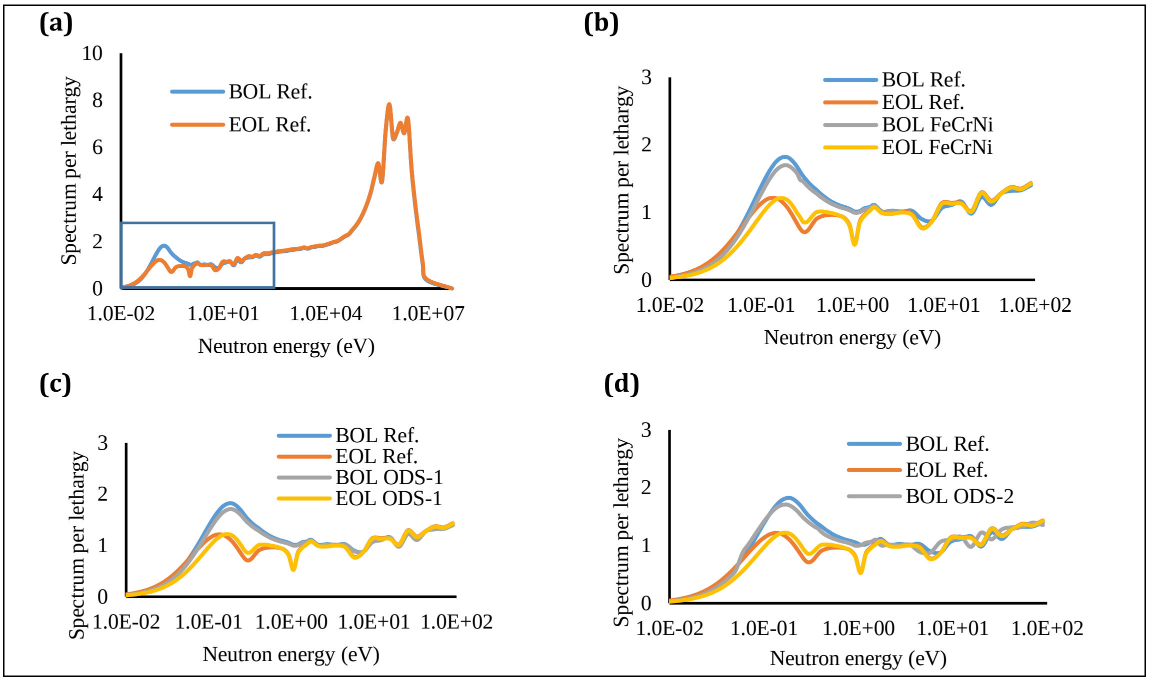

Figure 2 Neutron energy spectrum at BOL and EOL of (a) reactor core with reference fuel cladding, and the zoomed around thermal energy spectrum of that with (b) FeCrNi alloy, (c) ODS-1 and (d) ODS-2

The result showed that the neutron spectra for the FeCrNi alloy, ODS-1, and ODS-2 are slightly shifted to higher energy (hardened) compared to the reference. The shift is due to the increase in the energy of some of the thermal neutrons by way of collision with the new ATF cladding materials, which have much lower atomic mass than the reference. Accumulation of fission products and actinides towards EOL further increases unwanted thermal neutron reactions. Hardened neutrons mean the presence of fewer thermal neutrons needed for the fission reaction to occur (Detkina *et al.*, 2020; Chen and Yuan, 2017).

The effect of fuel burnup on the excess reactivity for ATF cladding under examination is shown in Figure 3, which is computed at hot full power. As a result of spectrum hardening, the excess reactivity was reduced relative to the reference. Figure 3a shows at the BOL, the excess reactivity of the reference 4.12% enrichment fuel is 28.4%dk/k, while the average values for ATF claddings under examination are around 20.6%dk/k and capable of only around 26000MWd/T burnup. Here, it is demonstrated that compared to the reference cladding, all three proposed cladding materials FeCrNi alloy, ODS-1, and ODS-2 - exhibited lower initial excess reactivity values. As a result, they would not supply sufficient excess reactivity to achieve the same final burnup level as the reference (40000 MWd/T).

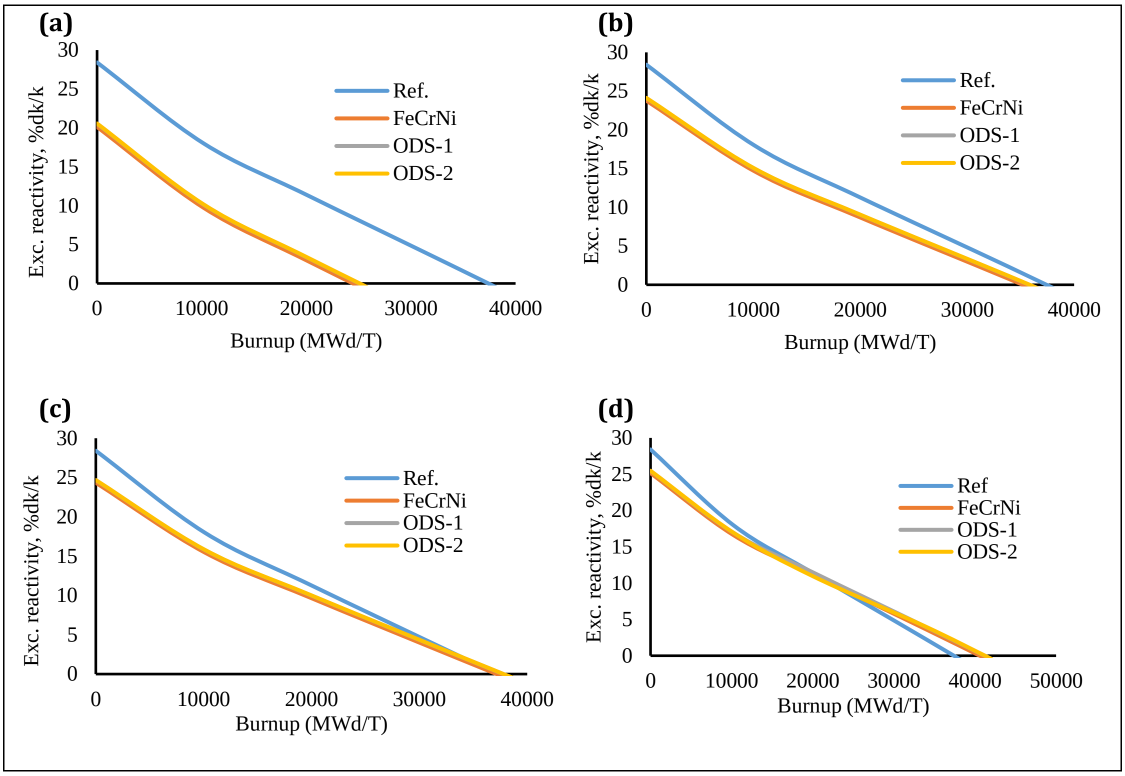

Figure 3 Comparison of the initial excess reactivity behavior of the LWR core using different cladding materials as a function of fuel burnup, where the ^{235}U enrichment in the fuel is (a) 4.12%. (b) 5.35%, (c) 5.60% (c), and (d) 6.00%.

To overcome this, one needs to increase the fuel enrichment to a higher level. In this study, the excess reactivity was improved by raising the fuel enrichment stepwise up to 6%, thereby improving the fuel burnup to approach the reference case. A fuel enrichment of 5.35%, as shown in Figure 3b, improved the fuel burnup to about 38000 MWd/T. A fuel enrichment of 5.60%, shown in Figure 3c, yielded an excess reactivity that was enough to maintain the reactor core to match the burnup of the reference. However, further fuel enrichment up to 6.00%, as shown in Figure 3d exhibited a higher burnup capability of the reactor core up to 42000 MWd/T with a reduced gap in initial excess reactivity from 28.4% to 25.4%. This showed that in general, the new cladding materials give lower initial excess reactivity than the reference material does to the LWR. The lower initial reactivity can be caused by the spectrum hardening in the thermal region, especially as fuel is consumed toward its end-of-life. The deficiency in initial reactivity can be overcome by increasing the fuel enrichment. As the fuel enrichment is increased, the rate of decrease in the excess reactivity becomes less than that of the reference, such that it is possible to have higher than reference fuel burnup at the EOL, as shown in Figure 3d.

In terms of reactivity loss resulting from a change in fuel temperature, Figure 4 illustrates the behavior of the LWR reactor. The figure demonstrates that, in all cases, excess reactivity is consistently lost with any increase in fuel temperature, underscoring the critical importance of this nuclear safety parameter. There are two main contributions to this situation. First, increasing fuel temperature increases resonance capture in ^{238}U. As the temperature rises, the resonance peaks broaden over a wider range of neutron energies, allowing more neutrons to be captured. Second, the ratio of fission to absorption in the fuel changes with fuel temperature, depending on whether the fuel is fresh or in equilibrium fuelling. This ratio decreases as thermal neutrons speed up due to a temperature increase in fresh fuel where ^{235}U is the only fissile nuclide at BOL. With a significant amount of ^{239}Pu created via the transmutation of ^{238}U, the ratio rises at EOL.

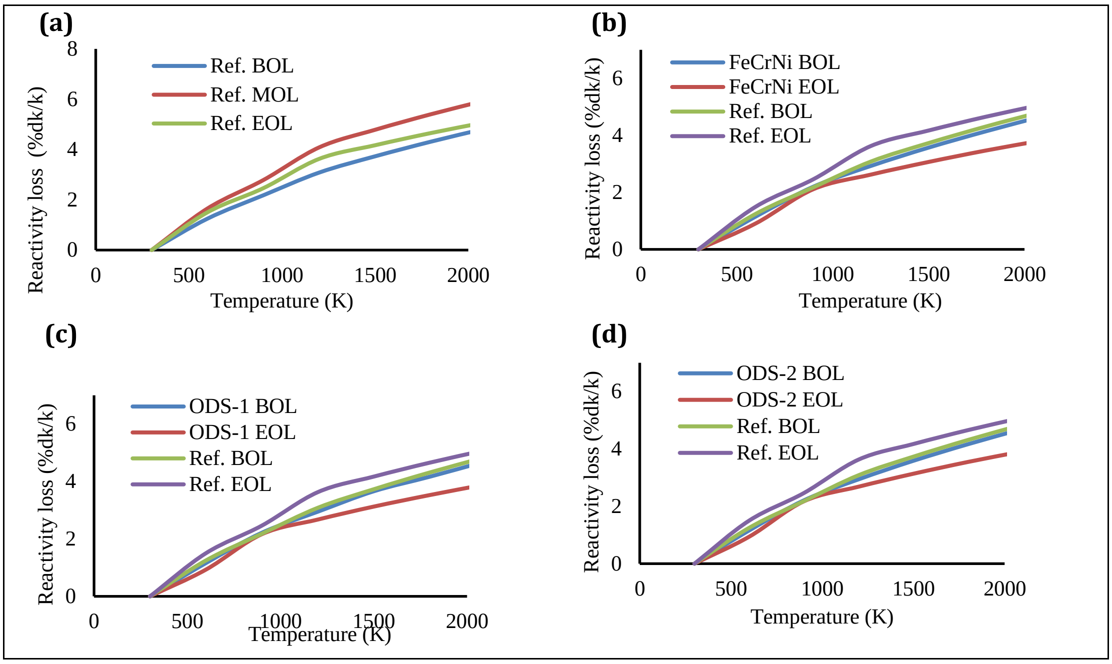

Figure 4 Reactivity loss due to fuel temperature rise at 4.12% fuel enrichment, (a) the reference; (b) FeCrNi alloy cladding; (c) ODS-1 cladding; and (d) ODS-2 cladding

At BOL, as the temperature of thermal neutrons increases, less of the neutrons absorbed in the fuel generate fission. Atom ^{235}U prefers thermal neutrons as opposed to ^{239}Pu, which prefers “hotter” neutrons. As a result, increasing the thermal neutron temperature reduces the reactivity of fresh fuel. At EOL, due to the presence of ^{239}Pu, the reactivity increases with increasing thermal neutron temperature.

The overall change in reactivity is a combination of both effects. The effect of resonance absorption, however, is always bigger than that of hot neutrons. In the reference case, reactivity loss due to temp increase in BOL was always lower than in EOL, as shown in Figure 4a, which indicates the loss in reactivity due to ^{239}Pu buildup was minimal under the reference neutron spectrum. In Figures 4b, 4c, and 4d at EOL, the ATF showed slightly less reactivity loss than the reference. At BOL the reactivity loss was always higher than that in EOL and showed similar features to the reference, indicating a better safety feature in general than the reference.

4. Conclusions

The results of a neutronic analysis of LWR using candidate tolerant fuel cladding materials of FeCrNi alloy, ODS-1, and ODS-2 were compared to the reference material of

Zircaloy-4. The calculations were carried out using the SRAC Code and the JEFF nuclear data file. It was shown that neutron spectrum hardening occurred when the candidate cladding materials were used. The hardening was responsible for the lower excess reactivity value when compared to the reference. As a result, increasing fuel enrichment to compensate for the reduction in initial reactivity is recommended to match the reference reactor fuel cycle. The reactivity loss due to temperature increase in BOL was slightly higher than in EOL, indicating that ^{239}Pu buildup reduced the reactivity loss in the hardened spectrum environment. The Doppler coefficients in the candidate cladding materials were found to be comparable to the reference. Our study also confirmed that the candidate accident-tolerant fuel cladding materials of FeCrNi alloy, ODS-1, and ODS-2 demonstrate the negative temperature coefficient of reactivity of the reactor core examined, which is an important feature in the safety LWR.

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