Analysis of Optimum Accident Tolerant Fuel and Cladding Behavior in Advanced Pressurized Water Reactor

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ABSTRACT

In PWR reactors, a higher temperature than the normal operation rate causes an increase in the oxidation rate between the fuel and the clad (UO$_2$/Zircaloy), and this results in the release of large quantities of hydrogen, which leads to an increase in pressure and temperature inside the reactor core and also on the walls of the pressure vessel, and perhaps partial or total damage to the reactor core. This research examines the development of new types of fuel such as Uranium Nitride (UN), Uranium Silicate (U$_3$Si$_2$). Also, two types of clads such as Silicon Carbide and (Fe-Cr-Al) alloy are tested. The neutronic and thermal properties of these new types have been studied, as they are characterized by the low probability of fuel interaction with cladding, as well as the presence of good neutronic and thermal properties in terms of thermal conductivity and heat capacity, which lead to an improvement in the safety margin during operation and also in the event of nuclear accidents.

INTRODUCTION

In light of the event that occurred at the Fukushima Daiichi nuclear power plant in March 2011, there has been an increased research efforts into accident tolerant fuel (ATF) and cladding materials [1]. Accident Tolerant materials are defined as those that provide significantly increased response time in the event of an accident while providing improved performance than the standard UO$_2$/Zircaloy fuel rods during accident and normal operation [2]. One of the most significant issues in the Fukushima Daiichi accident was the oxidation of Zircaloy that lead to a large inventory of Hydrogen within the core.

The Iron Chromium-Aluminum (Fe-Cr-Al) was proposed to be used as a cladding material due to its low oxidation rate. Oxidation rate for FeCrAl are approximately much lower than the oxidation rate of Zircaloy, The Ferritic alloy (Fe-Cr-Al) has low oxidation behavior with water and steam in the range of normal reactor operations and in the case of severe accident conditions. Also it has better thermodynamic, metallurgical and mechanical properties[3,4]. Silicon Carbide (SiC) which is also used in gas cooled fast reactors has antioxidant capability in steam environment. Uranium Nitride (UN) fuel has higher thermal conductivity and higher content of fissile isotopes than UO$_2$ which implies economic and long fuel cycle. U$_3$Si$_2$ has a number of advantageous thermophysical properties, including; high density (11.3 g/cm$^3$), high thermal conductivity at room temperature (15 W/mK). These properties also support its use as an accident tolerant fuel. Because of its high thermal conductivity, U$_3$Si$_2$ operates at a much lower temperature and experiences lower thermal gradients than UO$_2$. As a result, it is subject to lower thermal stresses, which should mitigate pellet cracking, [5,6,7,9].

The motivation for transitioning away from zirconium-based fuel cladding in light water reactors to significantly more oxidation-resistant materials thereby enhances safety margins during severe accidents. In this study, the neutronic and thermal properties of Fe-Cr-Al and Silicon Carbide alloys are analyzed and compared with Zircaloy. Also, two types of fuel...
namely, Uranium nitride (UN) and Uranium silicide (U₃Si₂) are investigated and compared with the traditional UO₂ fuel.

Gamble K. A. [1] Studied U₃Si₂ fuel and FeCrAl clad under normal and accident conditions both thermophysical and oxidation behavior are analysed. Brown N. R. [2] Studied the Neutronic analysis of UN (Uranium Nitride) under different porosity and additives to UN such as ZrO₂, U₃Si₂, U₃Si₃, UB₄. Rahman M. H. [5] Studied microstructure aspects of SiC particles in Aluminum matrix. Metzeger K. E. [8] model the behavior of U₃Si₂ with the available thermophysical data to predict the cladding temperature and swelling using BISON code. Terrani K. A. [10] studied oxidation behavior, microstructure under burnup for coated Zirconium, Ferritic Alumina forming alloy cladding and Silicon Carbide. Qiu B.[11] reviewed on thermohydraulic and mechanical physical properties of SiC, FeCrAl, and Ti₃SiC₂ as Accident Tolerant Fuel cladding. In the present Study three types of fuel UO₂, UN and U₃Si₂ in addition to three types of cladding Zr-alloy, SiC and FeCrAl are investigated as an Accident Tolerant fuel (ATF) and cladding from neutron and thermalhydraulic point of view. K̅eff, cycle length and discharge burnup are used as neutronic indicator while thermal capacity (k) and thermal capacity (Cp) is used as thermal indicator.

MATHEMATICAL AND COMPUTATIONAL MODEL

In the following section, both neutronic and thermodynamic models are established to study the neutronic and thermal properties of different clad and fuel types. The behavior of thermal conductivity and heat capacity with temperature are considered as an indicator of the thermal properties of the clad and fuel materials.

Neutronic Model

MCNPX code, [12], has been used to design a computer model for an assembly of advanced PWR reactor. The assembly is 17 x 17 fuel rods. It contains 264 fuel rods and 24 positions for burnable poison or control rod and one central position for instrumentation [13]. The horizontal and vertical layout of the model is illustrated in Figure 1. The outer dimension is 21.4 x 21.4 cm. The fuel rod pitch is 1.264 cm. fuel rod radius is 0.4096 cm, outer clad radius is 0.475 cm and a thin layer of Helium between fuel and clad with thickness of 0.0083 cm. Figure 2 illustrates the model of the fuel, clad and coolant. Three types of clads are tested, ZircaloY-4 (Zr), silicon Carbide (SiC) and Ferritic steel alloy (Fe-Cr-Al) with density 6.6 g/cm³, 3.21 g/cm³, and 7.15 g/cm³, respectively. The ratio between isotopes in Ferretic steel is 74:21:5, respectively, [9].

Three types of fuel are also considered in the analysis namely, UO₂, UN, and U₃Si₂ with fixed enrichment 4.95 %. The assembly is burned in a typical operating conditions in pressure and temperature for PWR reactor. High fuel burnup with sixteen time steps up to the multiplication factor approach one is allowed for each case. The power assigned to the assembly during burnup is 20 MW. Reflective boundary conditions are considered for all peripheral boundaries and 50 cm of water is considered on the top and bottom of the assembly. ENDF-VI is the neutron cross section library for the materials in the model. Table 1 illustrates the density of clad and fuel used in the paper and Table 2 illustrates Chemical composition of Zr-4.

Table (1): Clad and Fuel density

<table>
<thead>
<tr>
<th>Clad and Fuel type</th>
<th>Zr-4</th>
<th>SiC</th>
<th>FeCrAl</th>
<th>UO₂</th>
<th>U₃Si₂</th>
<th>UN</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density (g/cm³)</td>
<td>6.6</td>
<td>3.21</td>
<td>10.2</td>
<td>11.56</td>
<td>13.5</td>
<td></td>
</tr>
</tbody>
</table>

Table (2): Chemical Composition of Zr-4

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Zr</th>
<th>Sn</th>
<th>Fe</th>
<th>Cr</th>
<th>O</th>
</tr>
</thead>
<tbody>
<tr>
<td>Weight fraction %</td>
<td>98.43</td>
<td>1.2</td>
<td>0.18</td>
<td>0.07</td>
<td>0.12</td>
</tr>
</tbody>
</table>

Several neutronic parameters such as cycle length, discharge burnup and breeding capability are compared for all of the three fuel types. Also thermal properties parameters such as heat capacity (Cp) and thermal conductivity (k) are examined. For cladding material, three cases for Zr, SiC and Fe-Cr-Al, assuming UO₂ fuel in all cases, are compared to analyze the accident tolerant cladding materials.

Thermodynamic Model

Thermal conductivity (k) and heat capacity (Cp) are the key parameters for thermal properties. A higher thermal conductivity results in a lower fuel rod temperature and smaller temperature gradient. The following formula are used to study the behavior of both k and Cp over the entire range of normal and accident conditions in the PWR reactor.

U₃Si₂ Fuel

Thermal conductivity and heat capacity of U₃Si₂ fuel are computed using temperature dependent empirical relations which are given in reference [8].
\[ k \left(\frac{\text{W}}{\text{K.m}}\right) = 7.98 + 0.0051 \times (T - 273.15) \]  
(1)

Where, \( T \) is temperature in K. This expression is valid for temperatures from room temperature to 1473.15 K.

A relationship for the specific heat \( C_p \) (J/kg.K) of \( \text{U}_3\text{Si}_2 \) was derived in [8] as,

\[ C_p(\text{J/kg.K}) = 199.0 + 0.104 \times (T - 273.15) \]  
(2)

Where, \( T \) is temperature in K.

**UN Fuel**

First, the thermal conductivity for 100% dense UN fuel is calculated from [15], using the following equation:

\[ k \left(\frac{\text{W}}{\text{K.m}}\right) = 1.377T^{0.41} \]  
(3)

For Heat Capacity, \( C_p \), the correlation used in the current analysis is given by [15] as,

\[ C_p(\text{J/kg.K}) = -211.55 + 3.7854T - 6.227 \times 10^{-3}T^2 + 3.63774 \times 10^{-6}T^3 \]  
(4)

Where, \( C_p \) (KJ/mole.k) and \( \theta = 365.7 \) K

**FeCrAl Alloy**

The thermal conductivity of FeCrAl Alloy is given by [11], where \( T \) is temperature in (K)

\[ k \left(\frac{\text{W}}{\text{K.m}}\right) = 3.72 + 0.0427T - 7.2 \times 10^{-6}T^2 + 1.56573 \times 10^{-9}T^3 \]  
(5)

The heat capacity, \( C_p \) (J/kg.k), is also given by:

\[ C_p(\text{J/kg.k}) = -211.55 + 3.7854T - 6.227 \times 10^{-3}T^2 + 3.63774 \times 10^{-6}T^3, \text{if } T \leq 854 \text{ K} \]  
(6)

\[ C_p = 2113.39 - 2.543T + 1.12 \times 10^{-3}T^2, \text{ if } 854 < T \leq 991 \text{ K} \]  
(7)

\[ C_p = 208.78 + T - 6.86 \times 10^{-4}T^2 + 1.7173x10^{-7}T^3, \text{ if } 991 < T \leq 1776 \text{ K} \]  
(8)

**SiC Alloy**

The thermal conductivity is given by:

\[ k = 1.2 + 0.0027T + 0.62 \times 10^{-4}T^2 \text{ (W/m. C)} \]  
(9)

And, the heat capacity (J/Kg.K) is given by:

\[ C_p(\text{J/Kg.K}) = 0.92565 + 0.3772T - 7.9259 \times 10^{-5}T^2 - 3.1946 \times 10^{-7}T^3 \]  
(10)

The thermal conductivities and heat capacities for the traditional \( \text{UO}_2 \) and Zircaloy are extracted from reference [14, 15], respectively.

**RESULTS AND DISCUSSIONS**

Figure 3 illustrates the multiplication factor for an assembly with 3 different cladding materials and \( \text{UO}_2 \) fuel. The results show that \( k_{inf} \) is higher in the case of SiC clad over the entire operation time up to 1000 days. Table 3 shows the value of \( k_{inf} \) at initial time \( (t=0.0) \). The ferretic steel alloy (FeCrAl) shows significant reduction in the value of \( k_{inf} \) because absorption cross section for iron and Chromium are 2.53 and 3.1 barns respectively, [16], in comparisons with those of silicon and Zirconium of 2.33 and 0.18 barns, respectively.
Figure 4 illustrates the multiplication factor for the assembly for 3 different fuel types UO₂, UN and U₃Si₂ with Zircaloy clad. UN is higher in K∞ because it has the higher density of 13.5 g/cm³ as compared to U₃Si₂ and UO₂ which is 11.3 and 10.4 g/cm³, respectively. Higher density implies higher ²³⁵U fissile isotope content and also higher fuel cycle. Uranium fraction in U₃Si₂, UN and UO₂ are 0.962, 0.944 and 0.88, respectively. For this reason, UN and U₃Si₂ have comparable cycle length to each other.

Figure 5 illustrates ²³⁵U and Pu – fissile isotope in atom/barn.cm versus operation times (days) for UO₂ fuel. An operation time of 1050 days is fixed between all 3 fuel types. For example ²³⁵U at initial time and 1050 day is 1.16x10⁻³ and 2.9 x10⁻⁴ atom/barn.cm. The burnup for ²³⁵U during this period is 75 % and the concentration of Pu - fissile approaches to 1.88x10⁻⁴ atom/barn.cm (Pu-fissile isotopes is the summation of both 239Pu and 241Pu).

Figure 6 illustrates ²³⁵U and Pu –fissile isotope in atom/barn.cm versus operation times (days) for UN fuel. At 1050 days of cycle length, ²³⁵U concentration at initial time and 1050 day is 1.617x10⁻³ and 6.78 x10⁻⁴ atom/barn.cm. The burnup for ²³⁵U during this period is 58 % and the concentration of Pu- fissile approaches to 2.77x10⁻⁴ atom/barn.cm.

Figure 7 illustrates ²³⁵U and Pu –fissile isotope in atom/barn.cm versus operation times (days) for U₃Si₂ fuel. At 1050 days of cycle length, ²³⁵U concentration at initial time and 1050 day is 1.435x10⁻³ and 2.434 x10⁻⁴ atom/barn.cm. The burnup for ²³⁵U during this period is 64 % and the concentration of Pu- fissile approaches to 2.65x10⁻⁴ atom/barn.cm.

Figures 4, 5 and 6 compare between burnup of ²³⁵U and breeding of Pu-fissile isotopes for all three fuel type UO₂, UN and U₃Si₂. The results indicate that UO₂ is the higher fuel burnup 75 % while UN fuel is the higher initial ²³⁵U concentration. UN fuel is the higher fuel breeding with Pu-fissile concentration of 2.77 x10⁻⁴ atom/barn.cm.
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Figure 6 illustrates the fuel burnup (GWd/T) versus operation time (days) for the three fuel types; i.e., UO₂, UN and U₃Si₂ fuel. The results indicate that UO₂ have highest burnup (GWd/T) and UN is the lowest between the three types.

Figure 9 illustrates Thermal Conductivity (W/m. K) versus fuel temperature (K) for typical fuel materials namely UO₂, U₃Si₂ and UN. The results indicate that UO₂ thermal conductivity decreases with increasing temperature, while it increases for both U₃Si₂ and UN. Increasing thermal conductivity with temperature rise during accident conditions reduces the fuel centerline temperature and improves the safety margin of the fuel. UN has the superiority for thermal conductivity.

Figure 10 illustrates thermal conductivity (W/m. K) versus fuel temperature (K) for clad materials namely Zircalloy-4, FeCrAl Alloy and SiC. During normal operating conditions of the reactor (600 K), Zircalloy and FrCrAl alloy have higher thermal conductivities than that of SiC. But, at higher temperatures, thermal conductivity of SiC increases dramatically with temperature to values much higher than thermal conductivities of the other two cladding types.
Figure 11 illustrates heat capacity (J/Kg. K) for Zircalloy-4, FeCrAl Alloy and SiC cladding materials versus temperature (K). Zircalloy-4 has discontinuity at temperature 1100 K due to phase transition from α phase to β phase [15]. SiC heat capacity is an increasing function of temperature and has the highest values between the three cladding types.

For Cladding materials, the FeCrAl, Ferretic steel alloy, which is an oxidation resistant, has good thermodynamic properties (high thermal conductivity in the temperature range of normal operation and accident conditions) but, has higher neutron absorption (low neutron economy).

- Silicon Carbide, SiC, clad has better thermal and neutron economy.
- UN and U₃Si₂ fuels have higher cycle length than UO₂, and better thermodynamic properties. Higher cycle length implies more economic utilization of the fuel and less nuclear spent fuel.
- UN fuel has the higher ²³⁵U initial fissile concentration and more breeding than U₃Si₂ and UO₂ for the same cycle length. Pu-fissile at the end of the cycle for UN, U₃Si₂ and UO₂ are 2.77 x10⁻⁴, 2.65 x10⁻⁴ and 1.88 x10⁻⁴ atom/barn.cm, respectively.

**REFERENCES**


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**CONCLUSION**

- MCNPX computer code is used to design a computer model for an assembly of advanced PWR design to study the neutronic and thermal properties of three types of cladding Zr, SiC and FeCrAl and also three different types of fuel that are UO₂, UN and U₃Si₂.


