ANALYSIS OF THE PHYSICAL MODELS USED FOR NUCLEAR CERAMIC FUEL

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ABSTRACT: Nuclear ceramics resist to radiation exposure and may change their thermal and mechanical properties. Many ceramic fuels, such as UO_2 and mixed oxide (MOX), are the primary fuels used in the current commercial light water reactors. Fusion reactor use of low-activity ceramics, primarily silicon carbide, will eliminate the generation of large quantities of highly radioactive waste. Silicon carbides show desirable mechanical properties and corrosion resistance at high temperatures, making them essential for nuclear fuel cladding intended for a vast range of fuels in LWRs, Gas-cooled reactors use TRISO fuel at high temperatures. Generation IV reactors scheduled for 2035 have defined seven reactor types that will operate at higher temperatures with extended burn cycles, employing innovative fuels focusing on carbide UC and nitride UN. A review of the thermal and mechanical models used in the FRAPCON codes is presented.

KEYWORDS: UN, U_3Si_2, UO_2, FRAPCON.

INTRODUCTION

The international community has widely accepted that global climate change results from greenhouse emissions [1]. Nuclear energy is a zero-emission clean energy source, more sustainable, and has a fundamental role in electric generation. It produces about 10% of the global electricity, with about 440 power reactors in operation.

Nuclear fuel systems have employed ceramic oxides as fuel and metallic cladding in thermal reactors for over six decades, showing more than sixty years of experience. Ceramic oxides, such as uranium dioxide, UO_2, and several more tolerant fuels, such as uranium nitride (UN), uranium silicide (U_3Si_2), thorium dioxide (ThO_2), and mixed oxide (MOX) fuel (U-Pu) O_2, are the most used nuclear fuels [2].

Nuclear ceramics have good resistance to radiation exposure and high temperature. Thus, nuclear energy could solve the natural shortage of resources with no carbon emissions. Since 2001, the Generation IV forum (GIF) has chosen at least six reactor designs to replace the third generation, with over forty years of operation. GIF planned at least six advanced reactor designs to reduce costs, increase design flexibility and thermal efficiency, and reduce radioactive waste and toxicity [3].

After the nuclear accident at Fukushima in 2012, the U.S. Congress directed the pursuit of advanced fuel technologies to enhance the safety of nuclear power reactors. In 2022, through Act-2022, U.S. interagency coordination and prioritization of civil nuclear exports provided new tools for U.S. agencies to fund civil nuclear projects in partner countries.

Since the beginning of nuclear power generation in the 1950s, nuclear ceramics based on the fissionable metals U-235 and Pu-239 have been made into highly reliable fuel pellets for water-cooled and liquid-metal-cooled reactors. Ceramic materials are refractory resources: for instance, the melting point of UO_2 is around 2840°C. Second, the crystal structure allows the retention of released fission gas, and their highly variable oxygen-to-metal ratio can shift to accommodate burnup. They have excellent irradiation resistance and chemical stability in water environments. On the other hand, the ceramic-metallic (Cermet) is an emerging fuel concept for fast nuclear reactor systems designed to combine ceramic and metal beneficial properties. Ceramic fuels are used for various fast-spectrum reactors, light-water reactors (LWRs), and many other designs for the future generation of fast nuclear reactors.

Uranium nitride and carbide do not react with sodium coolant or molten lead. However, the UN and UC show poor stability in water contact. In contrast, UO_2 has good stability in water contact an is commonly used in LWRs. Comparatively, the fuel swelling under irradiation is low for UO_2, medium for UN, and U_3Si_2.

Nuclear ceramic fuel accounts for around 96% of the fuel for water-cooled reactors. In contrast, changing UO_2 to pure UN and UC has been challenging because of water incompatibility. A possible solution is to use additives such as silicide thorium, chromium, or aluminum [4], which could develop a protective corrosion layer on the surface to mitigate the oxidation of the UN.

In this context, we investigate the physical properties of ceramic fuels. We show thermal conductivity, specific heat, thermal expansion, and rapid commentary about the fission gas released.

MATERIALS AND METHODS

FRPACON is a computer code for calculating the steady-state thermal-mechanical behavior of oxide fuel rods for high burnup [5]. The primary purpose is to analyze fuel behavior under normal operational and expected transient conditions in light water reactors (LWRs). Thus, we implemented a new version for U_3Si_2 and UN.

A simple definition of primary nuclear fuel is a naturally occurring uranium fissionable isotope. U-235 is the primary nuclear fuel that permits fission with thermal neutrons and, in fission, liberates one to three neutrons. In contrast, plutonium is an artificial fissionable element obtained through the reaction of neutron capture of U-238 transmuting in Pu-239.

Ceramic fuels used in nuclear reactors

All actinide dioxides also exhibit a fluorite or CaF_2 structure. In CaF_2, eight nearest neighbors surround each metal atom, such as ThO_2, PuO_2, CeO_2, NpO_2, and AmO_2. Thus, the UO_2 reveals a fluorite structure CaF_2 at ambient conditions, which consists of a face-centered cubic arrangement. Besides, the hyper-stoichiometry of $UO_{(2+x)}$ induces point defects and clusters, producing both anion and cation sub-lattices. Crystal defects increase material creep at high temperatures under high stresses and increase accident risks. Since the 1950s, the beginning of nuclear power generation, laboratories have developed nuclear ceramics based on the fissionable materials U-235 and Pu-239 into highly reliable fuel pellets for water-cooled and liquid-metal-cooled reactors and other designs.

Ceramic fuels are refractory materials; for instance, the melting point of UO_2 is around 2800°C. Second, the crystal structure allows the retention of the released fission gas, and their highly variable oxygen-to-metal ratio can shift to accommodate burnup. In the UO_2 matrix, a small addition of impurities can change the fluorite structure, the stoichiometry of UO_2, density, and grain size. Table I shows the thermal properties of ceramic fuels in the temperature range of 25°C, such as uranium densities, thermal conductivity (k), and melting point (MP).

Fuel	MP (°C)	Density ($gU.cm^{-3}$)	*k* ($W.m^{-1\circ}.C^{-1}$)
UO_2	2800	9.6	6.02.5
U_3Si_2	1665	11.3	1527.5
UN	2630	14.3	15.028.0
UC	2768	13.0	12.616.0
ThO_2	3378	8.45	102.45
UN-20PuN	2495	13.5	14.023.0
UC-20PuC	2302	12.9	16.022.0
UO_2-20PuO_2	2745	9.7	8.913.42

Table I. Thermal properties of ceramic oxides

The current nuclear fuel system, which consists of UO_2 fuel and Zr-alloy cladding, can produce successive events similar to those observed in Fukushima Daichi, 2011. During the Fukushima events' timeline, the high fuel temperature and zircalloy oxidation over 900°C in contact with water steam. As a result, oxidation produces a large amount of hydrogen, followed by hydrogen ignition or blasting, releasing radioactive materials into the environment. Uranium silicides, U_3Si_2, is a ceramic fuel considered a good contender for ATF fuels because of their high thermal conductivities and uranium densities compared to UO_2. The thermal conductivity increases with increasing temperature. Figure 1 displays the fuel densities of nuclear ceramics.

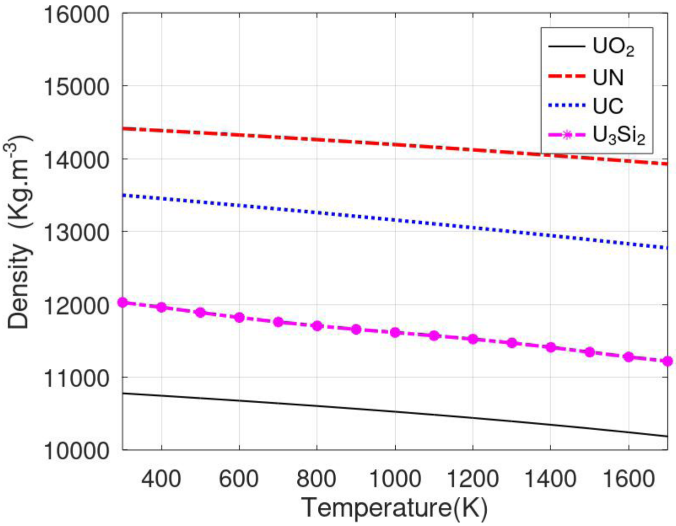

Figure 1: Density of ceramic fuels versus temperature

Higher thermal conductivity generally results in lower fuel centerline temperatures and lower stored thermal energy. The heat transfer equations show three related properties, thermal conductivity (*k*), thermal diffusivity (*α*), and specific heat capacity (*cp*), valid for pure and composite materials with composites. For determinate, composite ceramic fuel properties commonly employ a derivative of the empirical Koop Neuman rule (KNR), known as the mixture rule. Crystallography and metallurgy adopted Vergard's law, which resembles KNR. Equation (A) represents the relationship between thermal diffusivity and thermal conductivity.

$$k = \alpha \times \rho \times C_p \qquad \text{(A)}$$

where α is the thermal diffusivity ($m^2 \cdot s^{-1}$), k represents thermal conductivity ($W.m^1.°C^{-1}$), C_p is the specific heat ($J \cdot kg^{-1} \cdot °C^{-1}$), and ρ expresses the density ($kg \cdot m^{-3}$), and the thermal diffusivity describes the speed at which heat travels through a material.

The fuel fragmentation process occurred above 1200°C. Nitride fuel could disassociate at lower temperatures. Nitride composite fuel has shown lower swelling under irradiation than UO_2. Initially, a large range of ceramic fuels such as UN and UC does not react with sodium coolant or liquid metals. Figure 2 exhibits ceramic fuel thermal conductivities as a temperature function. Figure 3 illustrates the heat capacity of ceramic fuels.

Since 1960, uranium mononitride (UN) has awakened interest in high-temperature space propulsion programs and advanced fast breeder reactors. International research programs investigated nitride fuels during the 1960s and 1970s, restricting sodium and helium-cooled fast reactors working with UN and (U-20%Pu). UN has been an alternative for fast reactors. Under regular operation, the UN suffers cracking and extensive fuel fragmentation during startup and shutdown. Figure 2 exhibits ceramic fuels' thermal conductivity as a temperature function. Figure 3 illustrates the heat capacity of ceramic fuels.

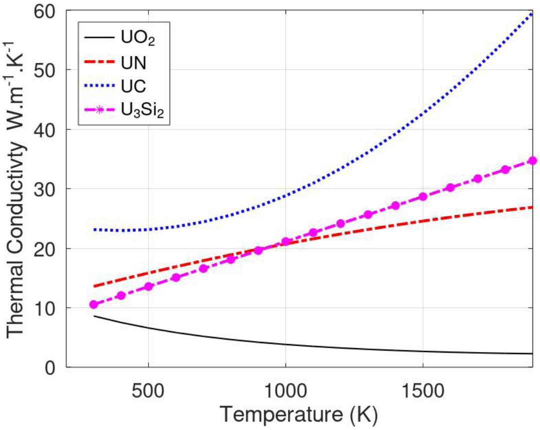

Figure 2: Thermal conductivity of ceramic fuels used in light water reactors.

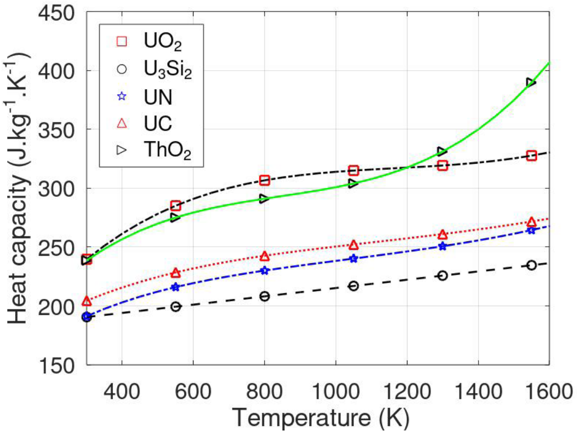

Figure 3: Heat capacity of ceramic fuels usually employed in reactors.

Initially, a large gamma of ceramic fuels, such as UN and UC, does not react with sodium coolant or liquid metals. The fuel fragmentation process occurs above 1200°C. Nitride fuel could disassociate at a lower temperature. Nitride composite fuel has shown lower swelling under irradiation than UO_2.

However, UN and UC show poor water or steam contact stability. In contrast, UO_2 has good stability in water contact, commonly used in LWRs. Comparatively, the fuel swelling under irradiation is low for UO_2, medium for UN, and U_3Si_2 but high for UC. In light water reactors (LWRs), there are problems, including the reactivity of isotope ^{14}N, because of the high neutron capture cross-section. A solution adopted uses enriched nitrogen ^{15}N, which has a lower absorption cross-section [6]. Besides, because of the tendency of UN powders to oxidize in contact with air or water steam environments, this reaction is exothermic with water, resulting in fuel pellet pulverization followed by relocation. The UN powders showed the capacity to self-ignite at about 260°C, producing a mixture of U_3O_8, UO_2, UO_3, U_2N_3, and UN. Figure 4 displays the thermal expansion coefficient versus the temperature of ceramic fuels used in nuclear generation.

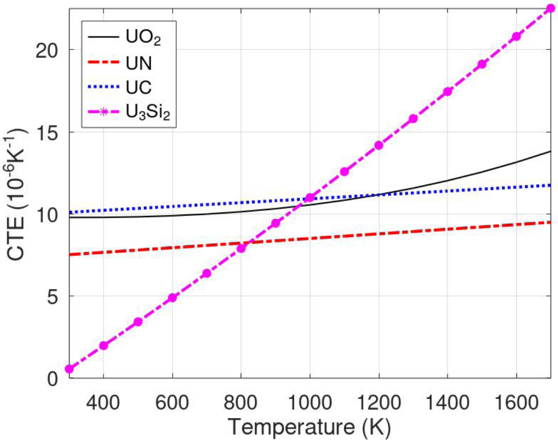

Figure 4: Coefficient of thermal expansion (CTE) of ceramic fuels.

On the other hand, the Russian Federation is developing BREST-300, which uses liquid metal sodium as a coolant. It is a fast reactor of 300 MWe that operates with (PuN–UN) fuel enrichment of about 13.5%. Also, other fast reactors with liquid metal as a coolant must operate using (Pu, Zr)N, (U-20%Pu) fuel, and UN. Moreover, carbide fuels are unsuitable for LWR because of their incompatibility with water coolantsThe energy generated from natural uranium using fast breeder reactors is about 60 times greater than that produced by LWRs, even after allowing for losses in the fuel cycle [7]. In 1968, the American Program developed a liquid metal fast breeder reactor (LMFBR) program using sodium coolant with dominant breeder technology. Into the core, they used fuel pins operated with mixed oxides (U-20% Pu)C in sequence using mixed uranium and plutonium (U-Pu)C carbides. Fuel pins used stainless steel type 316 cladding. The target of the LMFBR Program focused on nuclear power plants with capacities of 1000 MWe in operation in 1970 or early 1980. Uranium carbide is a potential fuel option for some Generation IV reactors, such as the gas-cooled fast reactor, which has been intensively researched.

In the last decade, the ATF plan has tested uranium nitride composed with a secondary phase, based on UN-U_3Si_2 or UN-U_3Si_5, also uranium diborides system UN-UB_2 or UN-UB_4 among other dopant phases, which might shield the nitride and carbide phase from undesirable water corrosion [8]. Thus, new research on silicides, carbides, nitride, diborides, and mixed compositions has arisen for light water reactors (LWRs). In addition, advanced alloys such as iron-chromium-aluminum (FeCrAl) and silicon carbide (SiC) are possible replacements for standard zircalloy used in the last decades [9].

The current U-Si phase diagram is characterized by seven intermetallic compounds (U_3Si, U_3Si_2, U-Si, U_3Si_5, $USi_{1.88}$, USi_2, and USi_3) of which only the U_3Si compound is well understood. However, U_3Si_2 can react with hydrogen, resulting in the degradation of material properties. Uranium silicides have shown advantages as the densities for U_3Si of 15.4 $g{\cdot}cm^{-3}$ and U_3Si_2 of 12.2 $g{\cdot}cm^{-3}$ have maximum uranium loadings for U_3Si and U_3Si_2 of 14.6 $g{\cdot}cm^{-3}$ and 11.3 $g{\cdot}cm^{-3}$ respectively.

The oxidation of UO_2 and U_3Si_2 in air demonstrated that U_3Si_2 has a higher onset oxidation temperature (~450°C) than UO_2 (~410°C). In contrast, U_3Si_2 has a much higher reaction enthalpy (~15 times) than UO_2. Besides, the steam corrosion resistance of U_3Si_2 was inferior to that of UO_2 at high temperatures.

UN and U_3Si_2 composites manufactured by conventional vacuum sintering are subject to microcracks because of a mismatch between silicide and nitride thermal expansion phases. The innovative fuel UN-U_3Si_2 manufactured with a strategic formulation proposed contains 50 wt% UN and 50 wt—% U_3Si_2, showing excellent thermal conductivity and enhanced strength and fracture toughness.

Carbides of uranium and thorium produce mixed carbides such as UC-PuC compared with mixed oxides $(U\text{-}Pu)O_2$, showing higher thermal conductivity and higher heavy metal densities. However, advanced ceramic fuels, including (U-Pu)C carbides and (U-Pu)N nitride have a more expensive and complex sintering route than UO_2.

The Accident tolerant fuel (ATF) program has tested advanced materials to substitute UO_2 and zirconium alloys in the future [10]. ATF fuel must tolerate the loss of active cooling in the reactor core for a long time while maintaining or improving fuel performance during normal operations and transient states beyond design basis events.

RESULTS AND DISCUSSION

The Pacific Northwest National Laboratory (PNNL) developed the FRAPCON code for the United States Nuclear Regulatory Commission (US NRC) to license light water reactors in the 1970s. Since 2019, the an upgraded version has added new fuels, uranium metal alloys, and ferritic alloys, such as iron-chromium-aluminum (FeCrAl) alloys. Besides, it is an independent audit tool for licensing nuclear power plants. Thus, a FRAPCON simulation showed that the innovative designs of the AP-1000 are based on simple principles for design-basis accidents. Table II shows a few input parameters for the simulation.

Nuclear Parameter	Values
Pellet outer diameter (mm)	9.50
Fuel density (%)	95.2
Cladding thickness (mm)	0.573
Cold plenum length (mm)	127
Gap thickness (mm)	0.081
Cladding roughness (mm)	0.00050
Fuel roughness (mm)	0.00199
U-235 enrichment (%)	4.8
Pitch, rod to rod (mm)	12.60
Mass flux of coolant ($kg \cdot s^{-1} \cdot m^{-2}$)	3.3906
Primary coolant flow rate ($kg \cdot s^{-1}$)	14300

Table II: Nuclear Parameters

FRAPCON permits the simulation with oxide fuel (UO_2 and $(U\text{-}Pu)O_2$) and zirconium-alloy cladding such as zircaloy-2, zircaloy-4, ZIRLO®, Optimized ZIRLO™ and M5™).

AP1000 uses light water as a primary and secondary coolant, and water is a moderator. The core coolant inlet temperature is around 279.4°C, and the outlet temperature is 324.7°C, with an average increase of 45°C. The unit produces a net power output of 1100 MWe and an average core power density of 109.7 $MW \cdot m^{-3}$.

This study used the adapted fuel code FRAPCON to change the standard fuel system to U_3Si_2/FeCrAl and UN/FeCrAl. Thus, based on PWR 17x17 executed, a simulation as the benchmark calculation for the UO_2/ZIRLO standard fuel system compared with the proposed system, the U_3Si_2/FeCrAl. Figure 4 illustrates the burn cycle behavior, which starts the burn cycle with a high linear power value of 37.5 $kW \cdot m^{-1}$ and decays along the fuel burn cycle.

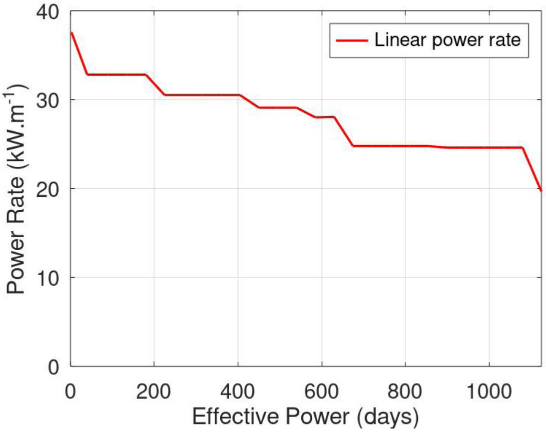

Figure 4: Linear power rate used in burn cycle of 1125 days.

The linear power rate used was (37.5, 32.8, 30.5, 29.1, 28, 24.7, 19.68, and 16.4) kW·m^{-1} in 1125 days of irradiation, achieving 68 MWd·kgU^{-1}. The burn cycle follows safety recommendations, limited to 68 MWd·kgU^{-1}. The fuel temperature must decrease as a function of the reduction in the linear power rate used. When using the other fuels, there is a slight variation in the burning since the uranium density is higher for UN and U_3Si_2 compared with UO_2. Despite the difference behavior between zirconium-based alloys and FeCrAl ferritic alloys, we can see a reduction in fuel temperature. Figure 5 illustrates the average fuel temperature for the fuel system used.

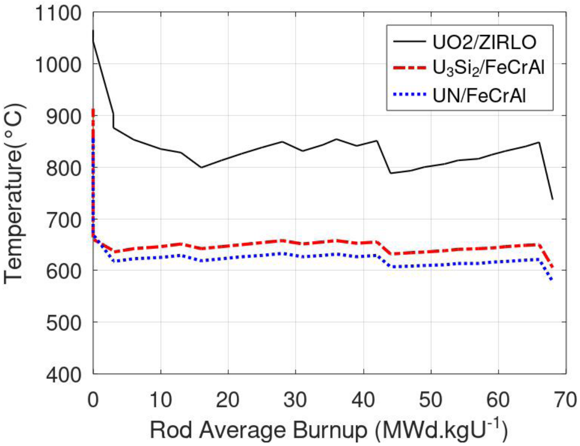

Figure 5: Average fuel temperature for fuel system used in simulations.

Figure 6 shows the fission gas release in each case, UO_2, $U_3Si_{2,}$ and UN, using FeCrAl.

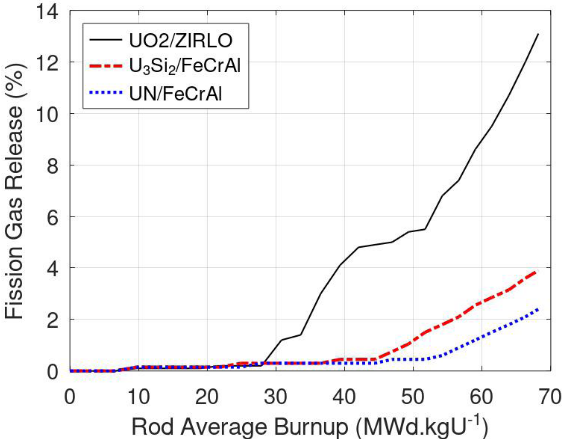

Figure 6: Fission gas release in each case UO_2, U_3SI_2, and UN, using FeCrAl.

The fuel temperature showed a significant reduction because of the higher thermal conductivity of UN and U_3Si_2, over five times the thermal conductivity of UO_2, in a range of temperature operation between 280°C and 325°C. Comparatively, the thermal conductivity of UN is higher than U_3Si_2 by 13% at the same temperature range.

An interesting factor is that while UO_2 has its thermal conductivity reduced with increasing temperature, the other options act, on the contrary, increasing the thermal conductivity of U_3Si_2 and UN. However, the results from the simulation are compatible with the theoretical values.

The plenum of the fuel rod is designed to contain part of the xenon and krypton generated in fission in a restricted volume of each rod. However, the case generation process strongly depends on temperature. Therefore, we can deduce a drop in pressure for fuels with better thermal conductivity. Figure 7 illustrates the plenum pressure versus burn cycle, achieving a 68 MWd·KgU-1 limit.

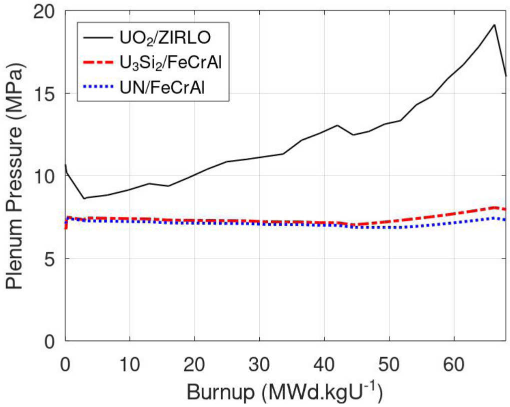

Figure 7: Plenum pressure compared with U3Si2 and UN.

CONCLUSIONS

Ceramic fuel has offered a clean and efficient means of producing electricity through various oxides and metallic fuels. The analysis of simulation results shows a strong unity with the thermal properties of the fuels.

The temperature of fuel suffers a significant reduction. It also reduces fission gases because of generation, which depends on temperature, and increases the production with increasing temperature. However, the great advantage of using U_3Si_2 or UN is the high thermal conductivity coupled with the higher uranium density. The counterpart of the used system also has a problem or penalty because of the more extensive cross-section for thermal neutron flux.

Comparatively, the properties of zirconium base alloys show benefits and drawbacks. The cross-section of zircaloy is ten orders of magnitude less than iron-chromium-aluminum FeCrAl, like the commercial KANTHAL® APMT.

In the simulation, the execution does not correct the cladding thickness but can reduce the clad thickness and increase the pellet radius. However, U3Si2 contains a higher uranium density than UO2 fuel, which provides a potential solution for all alternative cladding with a more extensive thermal neutron absorption cross-section than zirconium alloys. Besides, the big problem with the UN comprises the exothermic reaction between the UN and water. In this context, proposals are being made to use the UN with additives such as UN-U_3Si_2 or UN-U_3Si_5-UB_2.

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