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URANIUM NITRIDE AND SILICIDE COMPOSITE FUELS USED TO REDUCE FUEL OXIDATION

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Abstract: In the 1960s, the space nuclear program for developing reactor propulsion designs led to uranium mononitride (UN) exploration as a fuel. Since then, the UN has gained traction because of its superior thermal conductivity and uranium density compared to standard UO_2. Reactors using sodium and lead as liquid coolants have a. long history with UN fuel. More recently, a combination of uranium UN and uranium silicide (U_3Si_2) has emerged as a promising fuel option for power units. This composite fuel offers enhanced tolerance and resilience. However, several silicide compounds exist, such as USi, USi_2, USi_3, U_3Si, U_3Si_2, and U_3Si_5. Instead, they use nitride-silicide composites, such as UN-U_3Si_5. The UN-U_3Si_5 fuel, which features a secondary fissile phase with an elevated uranium density, has garnered significant interest. In this study, we compare the performance of UN-U_3Si_2 and UN-U_3Si_5 using Kanthal APMT as cladding.
Keywords: Uranium nitride, uranium silicide, accident tolerant fuel FRAPCON

INTRODUCTION

For the past four decades, uranium dioxide (UO_2) has been the standard fuel in light water reactors (LWRs). However, because of their high thermal conductivity and specific heat, uranium silicides have shown promise as potential substitutes for UO_2. Fuel options that use the U-Si system based on seven intermetallic compounds (USi, USi_2, USi_3, U_3Si, U_3Si_2, U_5Si_4, and U_3Si_5) have numerous benefits [1].

Investigations into ferritic alloys as potential replacements for zirconium alloys have revealed poorer neutronic performance. Furthermore, it has prompted attempts to thin the cladding and increase the amount of ^{235}U enrichment. Composite uranium nitride with uranium silicide is an innovative, more tolerant fuel. Since the space reactor program invented high-temperature and fast-breeder designs in the 1960s, uranium mononitride (UN) fuel has become highly sought-after. Nitride has gained popularity because of its greater melting temperature than UO_2, fissionable density, strong thermal conductivity, and superior melting temperature over UO_2. Fast reactors cooled by lead and sodium, and those using liquid metal coolants frequently use UN fuel.

For the past decade, the accident-tolerant fuel (ATF) plan has been at the forefront of fuel development, exploring uranium silicide mixed with uranium nitride [2]. This plan has focused on finding advanced properties, such as higher uranium density and a thermal conductivity of 52 W/mK. The UN-U_3Si_2-UB_2 composite fuel was a vital component of an earlier, less harmful fuel plan [3]. At 300 °C, the additive UB_2 has a much higher thermal conductivity of 25 W/K than UO_2, 7 W/mK. It also has good corrosion resistance.

Notably, U_3Si and U3Si_2 have higher uranium densities. Compounds like U_3Si_2 will be substituted for U_3Si_5 because irradiation resistance These compounds have a minor effect on the energy interaction with steam and keep the nitride phase safe from water reactions. Compared to U_3Si_2 and U_3Si, U_3Si_5 has a higher silicon content (62.5%) and a lower uranium density. To compare UN-U_3Si_2 and UN-U_3Si_5, we use ferritic alloys, iron-chrome-aluminum (FeCrAl), as cladding [4]. Isotope $^{10.}B$ presents a thermal neutron cross-section of 3800 barns. Today, the UN uses uranium diboride (UB_2) as a second phase for ZrB_2 to suppress initial fuel reactivity [5].

In the last decades, the ATFs plan has tried many different ceramic mixes, including uranium dioxide-diboride (UO_2-UB_2), uranium nitride (UN), uranium silicide (U_3Si_2), and UN-U_3Si_2-UB_2. ATF has tried to find advanced properties, including a higher uranium density and a thermal conductivity of 52 W/mK. Composite uranium nitride

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UN fuel has been in high demand since the space reactor program began with high-temperature and fast-breeder designs in the 1960s. Nitride has gained popularity because of its greater melting temperature than UO_2, fissionable density, strong thermal conductivity, and superior melting temperature over UO_2. Fast reactors cooled by lead and sodium, and those using liquid metal coolants frequently use UN fuel.

MATERIAL AND METHODS

Our investigation aims to enhance the FRAPCON code's physical characteristics to perform advanced simulations based on the material properties library [6]. This research is critical for developing more efficient and safer nuclear fuel systems, and zircaloy cladding is an essential element for standard fuel systems.

For the temperature range where PWRs are steady, UN-U_3Si_2 and UN-U_3Si_5 are about 5 to 7.5 times better at conducting heat than UO_2. Comparative gap closure occurs in about 200 days for UN-U_3Si_2 and UN-U_3Si_5 and about 100 days for UO_2 [7]. For safe operation, the pellet-cladding interaction (PCI) is critical. PCI causes cladding fracturing because the high temperatures accelerate the gap closure in PWR. The dimensional expansion facilitates swelling and PCI.

FUEL SYSTEM PROPERTIES

We used standard UO_2 fuel in this study, which had an enrichment of 3.5%, a pellet length of 11 mm, and an outside diameter of 9.132 mm. Table 1 depicts the physical properties of popular materials used in nuclear fuel systems [8–10].

Materials	Melting point (°C)	Density (g/cm³)	Thermal conductivity (W/m-K)	Heat capacity (J/kg-K)	Thermal expansion (μm/m-K)
UO_2	2850±30	10.96±0.1	8.68±0.5	235±5	9.76±0.5
U_3Si_2	1665±20	12.2±0.1	16.3±0.5	202±5	15.2±0.5
U_3Si_5	1770±10	8.97±0.1	9.32±0.5	195±5	10.6±0.5
UN	2850±30	13.61±0.1	15.0±0.5	239±5	12.5±0.5
Zircaloy	1850±10	6.56±0.1	21.5±0.5	285±5	6.0±0.5
APMT	1450±10	7.25±0.1	11.0±0.5	480±5	12.4±0.5
SS-348	1450±20	8.00±0.1	16.3±0.5	500±5	17.7±0.5

Table 1: Physical properties of advanced fuel materials

Our intended composite fuel was (UN-80%-U_3Si_5-10%), and we used a ferritic alloy (Kanthal APMT). This type of FeCrAlMo alloy is a dispersion-strengthened ferritic iron-chromium-aluminum alloy. It is made up of Fe, Cr (21%), Al (5%), and Mo (3%).

The Spark Plasma Sintering (SPS) type can operate at lower temperatures for shorter processing times. By taking a quicker sintering path, UN-U_3Si_5 can be produced without going through the ternary phase [11]. Compared to U_3Si_2, the U_3Si_5 phase resists oxidation and behaves like UO_2. On the other hand, radiation causes crystal amorphization in U_3Si_2 and U_3Si.

This study used Kanthal APMT as the cladding material because of its superior resistance to oxidation and corrosion, two critical issues when using uranium nitride and silicide composite fuels. The second phase also presents another mechanical aspect of the fuel and cladding. The MatLib includes subroutines for fuel density, thermal conductivity, specific heat, enthalpy, mechanical reaction, and creep rate.

The ATF plan has evaluated composites with monolithic UB_2, such as UO_2-UB_2, U_3Si_2-UB_2, and UN-U_3Si_2-UB_2. UB_2 has advanced features, such as 30 W/mK thermal conductivity and a higher uranium density. The thermal neutron cross-section of isotope ^{10}B is 3800 barns. Figure 1 depicts several

ATF materials' thermal conductivities as temperature functions.

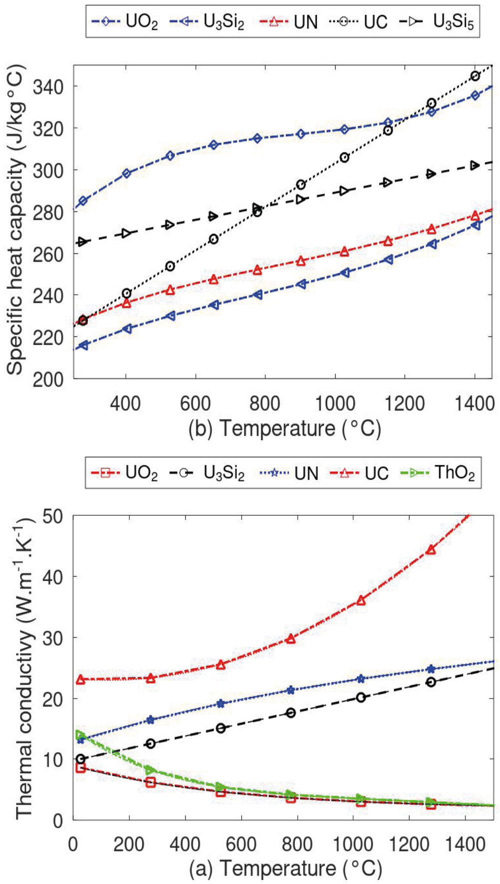


Figure 1. (a) Thermal conductivity of ATF fuel and (b) specific heat capacity.

However, we used an empirical isotopic ratio of $^{10}B/^{11}B$ with UB_2 enriched with ^{11}B, which has a substantially lower cross-section (0.0055 b).

In recent decades, researchers have used the arc-melting process, combining UN and U_3Si_2 powders. However, U-Si-N systems have shown a few unknown ternary phases produced at high temperatures, which is a significant challenge. Understanding the properties and behavior of these unknown phases is crucial, as they can impact the fuel's performance and safety. U_3Si_5 is a magnetic and brittle metal whose electronic contribution dominates thermal conductivity at high temperatures.

Compared to U_3Si_2 (11.3 gU/cm^3), U_3Si_5 has a lower uranium density (7.5 gU/cm^3) but a better melting point and thermal conductivity than UO_2. The composite UN-U_3Si_5's neutron performance is comparable to that of UO_2. A higher fissile density allows for an extended burning cycle, which is beneficial. The Fukushima Daiichi nuclear accident, with its rapid Zr-steam reaction in a high-temperature environment that fully exposed Zr alloys, generating large amounts of hydrogen and causing exothermic heating and blasting, underscores the importance of our research. The fuel used was the standard UO_2, with an enrichment of 3.5%, a pellet length of 11 mm, and an outside diameter of 9.132 mm.

ACCIDENT MORE TOLERANT MATERIALS

Zircaloys, made of zirconium, have a low thermal neutron absorption cross-section, good corrosion resistance, and excellent dimensional stability in an irradiation environment. However, they have several drawbacks, such as cladding embrittlement and undesirable exothermal oxidation above 1204 °C.

Zircaloy replacement shows two options: Next-term, ferritic alloys mixed composed of iron-chromium (20–30%) and aluminum (4–7%), known as FeCrAl alloys; next-term Kanthal APMT, and long-term use of silicon carbide (SiC) fiber-reinforced. Figure 2 shows the creep rate of ATF materials' elastic modulus and uranium silicide.

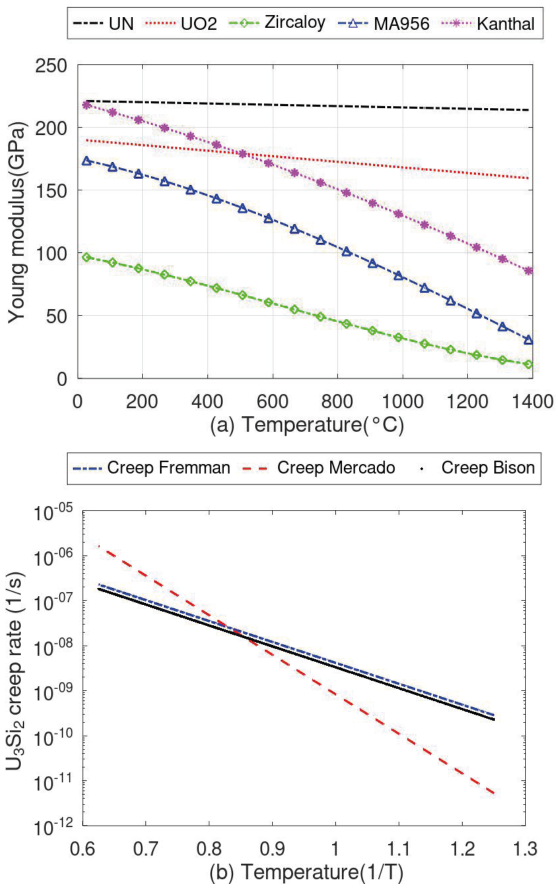


Figure 2. (a) Young modulus of ATF materials, (b) creep rate of U_3Si_2

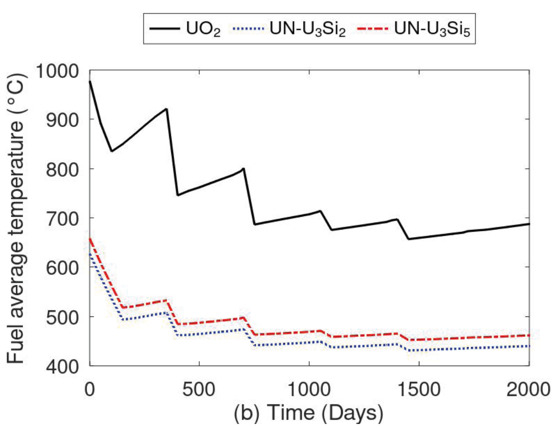
Consequently, it can release an enormous amount of hydrogen, followed by an explosion. Thus, ATF plans have investigated how to shield the cladding materials by adding Cr, Al, and Si elements from deep corrosion above 600 °C in the air using Cr_2O_3, Al_2O_3, or SiO_2 layers, which offer the best corrosion resistance.

ATF envisions implementing SiC fiber-reinforced SiC matrix (SiC/SiC) composites with two or three layers to replace zircaloys in the long term. SiC cladding reacts with water and steam 10,000 times more slowly than zirconium alloys, avoiding hydrogen and accidents.

On the other hand, austenitic stainless steels (SS), such as SS-316, have superior thermal conductivity and mechanical strength compared with zirconium-based alloys. Despite their low melting point of approximately 1450 °C, they outperform zircaloys with a melting point of roughly 1850 °C and UO_2 with a melting point of 2800 °C. The ferritic alloys show good features, such as thermal conductivity, which is somewhat better than zircaloys' in the temperature range of 373 °C to 1165 °C. One of FeCrAL's superior mechanical properties is that it shows a linear curve representing elastic moduli as a function of temperature, which has been 220 GPa at 20 °C and reduced to 130 GPa at 1000 °C. By contrast, zircaloys show an ultimate strength (UTS) of 413 MPa, a yield strength (YS) of 241 MPa, and a 20% elongation break. Meanwhile, Kanthal remains constant between 400 and 500 °C, showing YS of 540 MPa and UTS of 740 MPa. Early, U_3Si_2/FeCrAl and U_3Si_2/SiC-SiC composite fuel-cladding systems have become promising ATF fuel systems.

RESULTS AND DISCUSSION

Consequently, UN-80%U_3Si_5-10% and 10% of porosity was the composite fuel, and ferritic (Kanthal APMT) was intended to replace zircaloy-4. The average temperature of the fuel and the release of UO_2 fission gas compared to silicide and nitride are shown in Figure 3, which shows fuel temperature and stack axial extension.



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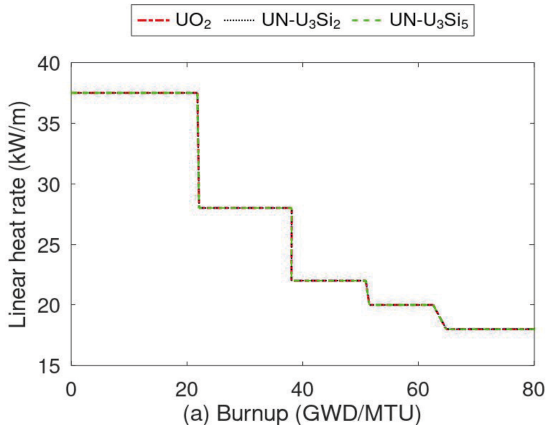

Figure 3. (a) Fuel temperature in function of burnup, (b) fuel stack axial extension

Another mechanical aspect of the fuel and cladding is present in the second phase. Nevertheless, the additional uranium density causes a slight drop in enrichment, which raises the neutron multiplication factor. The study concentrated on certain U-silicide phases because U_3Si_5 has a lower U-density and requires more enrichment to match the ^{235}U-loading that can be achieved with UO_2. UN-U_3Si_5 with two volume contents of the UN phase, 55% and 80%, and 35% and 10% of the U_3Si_5 weight were examined using the ATF program. In the temperature range of steady-state operation, the thermal conductivity of pure UN-U_3Si_2 and UN-U_3Si_5 is approximately 5 to 7.5. For UO_2, UN-U_3Si_2 and UN-U_3Si_5 both display cycles of about 100 days. However, we used 10% of the weight of U_3Si_5. FRAPCON's decreasing temperature of more than 250 °C should result in a modest release of fission gas and thermal expansion. According to the first post-irradiation evaluation, the UN-U_3Si_2 fuel system operated effectively at low burnup, causing minor pellet swelling and fission gas release (FGR). Figure 4 depicts FGR and fuel axial extension.

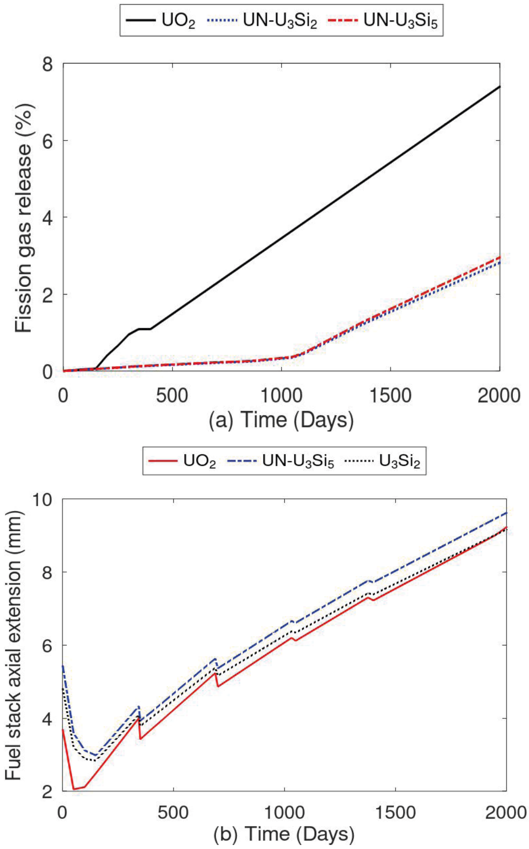

Figure 4. (a) fission gas release and (b) fuel axial extension

The UN-U_3Si_5 composite is a promising, more tolerant accident material for nuclear reactors that reacts to thermal neutrons, which is very similar to the UO_2 response. This exciting discovery suggests that it could easily switch from oxide operations to high-density ones, opening up new possibilities for its application.

When the UN and silicide phases come together, the thermal conductivity will increase, and the uranium loading will improve, making it even more helpful. In the UN-U_3Si_5 fuel concept, the U_3Si_5 phase must protect the nitride from water contaminants. The U_3Si_2, U_3Si_5, and UN are the best-mixed fuel options among the candidates. ATF fuel options have a high uranium density and good thermal conductivity.

The FeCrAl alloy and the SiC/SiC composite material are the most promising

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options for cladding because of their excellent strength and resistance to oxidation.

During a significant break-loss of coolant accident (LOCA), the FeCrAl cladding had meager oxidation and hydrogen production rates. However, U_3Si_2 can keep its crystal structure stable at 350 °C even when exposed to very high doses of radiation that cause amorphization (64 displacements per atom). The U_3Si_2 grain subdivision occurs at a relatively low dose. The higher pellet fuel thermal conductivity would reduce the stored thermal energy in the fuel, which is a critical factor during a LOCA.

Ferritic alloys produce a high neutron penalty, which results in a reduction in cladding thickness and an increase in pellet diameter. Thus, FeCrAl alloys use a cladding thickness of 0.0419 cm, while zircaloys use a thickness of 0.0572 cm. The fuel pellet exhibits an increased diameter of 0.4249 cm. Also, UN-U_3Si_5 has a low power density because it has a lot of heavy metals. It needs to be enriched with ^{235}U by 4.9% and ^{15}N by the same amount.

However, silicides like U_3Si_5 have significant effects. For example, they have a lower uranium density than U_3Si and U_3Si_2. It has a higher melting point (1770 °C) than U_3Si (925 °C) and U_3Si_2 (1665 °C). U_3Si_5 exhibits resistance to rapid oxidative pulverization at elevated temperatures. The starting temperatures for oxidation in U_3Si_5 and U_3Si_2 are about the same. However, the oxidation process in U_3Si_2 is faster, with a steeper slope of oxidative weight gain. Researchers in a different study found that the U_3Si_5 sample had microcracking, rapid oxidation, and a lower onset temperature than the U_3Si_2 and USi samples, which did not have microcracking as bad. This finding has significant implications for materials science and nuclear engineering.

CONCLUSION

The ATF idea shows a significant delay in fuel-cladding contact compared to traditional fuels of equal radial geometry and operational history. Sensitivity studies on fuel thermal creep rate, cladding thermal conductivity, cladding irradiation creep, cladding gap size, and cladding thickness indicate that research priorities for this concept in ATF should revolve around reducing cladding thickness, improving uranium densities and silicide-mixed nitride content, and increasing plasticity.

The goal is to verify the fuel's mechanical and thermal properties under normal operating conditions. The tests conducted during the ATF program saw cycles of less than 20 GWd/tU. Still, we offer a simulation with a period of 2000 days. We modified the FRAPCON code to accommodate U_3Si_5 characteristics.

For PWRs, composite fuel based on UN, U_3Si_2, U_3Si_5, or UB_2 offers several benefits. Permitting greater burnup will enhance nuclear fuel performance, reduce waste quantities, and extend cycle times. UN-U_3Si_5 fuel has the following benefits: high uranium density, increased thermal conductivity, and reduced UN degradation in water contact. Instead, we will use the Kanthal APMT composition [Fe (Bal.) Cr (20–23%) Al (5.8%)] for zirconium alloys used as cladding. Nitride fuels will produce less stored energy if an accident occurs. Because Kanthal does not oxidize to form hydrogen, it is more resistant to coolant mishaps. It does not produce zirconium byproducts like hydrogen bursts.

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