



Advanced Tolerant-Fuels Composites of Uranium Nitride and Silicide

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1. Introduction

During the 1960s, uranium mononitride (UN) manufacturing began to awaken interest in high-temperature space reactors and advanced fast-breeder reactors [1]. The Space Reactor Prototype (SP-100) program adopted a fast spectrum using lithium as a coolant, uranium-nitride (UN) as fuel, and the cladding of (Zr-1% Nb) in 1983. Nitride becomes attractive because of its high thermal conductivity, fissionable density, good chemical tolerance, and higher melting temperature than UO_2 . Thus, it can operate with austenitic steel as cladding. Over the years, reactors based on liquid metal coolants have accumulated sustained experience using UN fuel for sodium-cooled and lead-cooled fast reactors.

The Generation IV International Forum (GIF) started a scientific race to develop six reactor design types. Thus, various advanced fuels have been researched, such as uranium carbide (UC), uranium mononitride (UN), uranium silicide (U_3Si_2), and uranium diboride (UB_2), and mixed compositions like UN- U_3Si_2 and UN- U_3Si_5 . In the next term, accident-tolerant fuel (ATF) plans should replace the standard UO_2/Zr systems for civilian reactors. Besides, there are technological efforts to implement pressurized water reactors (PWRs). It could work with more tolerant fuels, such as UN- U_3Si_2 , followed by UN- U_3Si_5 , employing ferritic alloys (FeCrAl) as cladding [2].

The uranium silicide compounds include USi, USi_2 , USi_3 , U_3Si , U_3Si_2 , U_5Si_4 , U_3Si_5 , and $\text{USi}_{1.88}$. In particular, U_3Si and U_3Si_2 have higher uranium densities. To minimize the energetic interaction with steam and protect the nitride phase from water reactions, silicides such as U_3Si_5 replaced U_3Si_2 [3]. Besides, U_3Si_5 is a brittle and magnetic metal, and its electronic contribution dominates thermal conductivity at elevated temperatures. Compared to U_3Si_2 and U_3Si , U_3Si_5 has a high silicon content (62.5%) and a low uranium density.

Following new trends, we carried out a simulation with the FRAPCON code for the UN- U_3Si_5 fuel, also using Kanthal APMT as a cladding material, showing considerable performance.

2. Methodology

FRAPCON is the U.S. Nuclear Regulatory Commission's (NRC's) steady-state fuel performance code. While the companion code to FRAPTRAN leads to fast transients and severe accidents [5], the updated version is FAST 1.2, the only integrated code. BISON, developed by Idaho National Laboratories, presents options for UN- U_3Si_5 fuel. During the simulation, we inserted all the physical properties of the combined fuel UN-10% U_3Si_5 given in weight (wt%). The Nuclear Material Properties Library (MatLib) contains properties for several nuclear fuels, cladding and structural materials, gases, and coolants. Material libraries like MatLib contain models, typically called correlations, that often use burnup to measure the change in the fuel's properties. The FRAPCON version does not support the UN- U_3Si_5 composition.

The MatLib includes subroutines for fuel density, thermal conductivity, specific heat, enthalpy, mechanical response, and creep rate. The ATF plan has assessed composites with monolithic UB_2 , such as UO_2-UB_2 , $U_3Si_2-UB_2$, and $UN-U_3Si_2-UB_2$. Advanced properties shown by UB_2 include a higher uranium density and a thermal conductivity of 30 W/mK. Isotope ^{10}B presents a thermal neutron cross-section of 3800 barns. Today, UB_2 is a strong option for ZrB_2 , employed as a second phase in the UN to suppress initial fuel reactivity. However, using UB_2 enriched with ^{11}B , which has a much lower cross-section (0.0055 b), using an empirical isotopic ratio of $^{10}B/^{11}B$. Table I slightly compares the properties of silicide, nitride, and diboride with standard uranium dioxide. In this case, the thermal conductivity (k) is given in the temperature range of 25 °C to 1030 °C [4].

Table I: Properties of ATF fuels, uranium nitride, and silicides compared to UO_2 .

Nuclear Fuel	UO_2	UN	U_3Si_2	U_3Si_5	U_3Si	UB_2
Density (gU/cm ³)	9.66	13.5	11.3	7.5	14.6	11.68
Melting Point (°C)	2860 ± 30	2850 ± 30	1665	1770	985	2385
k (W/mK)	7–3	14–24	9–26	4–15	19–33	25–33
Crystal Structure	Cubic	Cubic	Tetragonal	Hexagonal	Tetragonal	Hexagonal

In the last few decades, a mixture of UN and U_3Si_2 powders has produced UN- U_3Si_2 sintered via the arc-melting method. Inconvenient is the unknown ternary phase of the U-Si-N system formed at elevated temperatures. Figure 1 depicts thermal conductivities given as temperature functions for several ATF fuels.

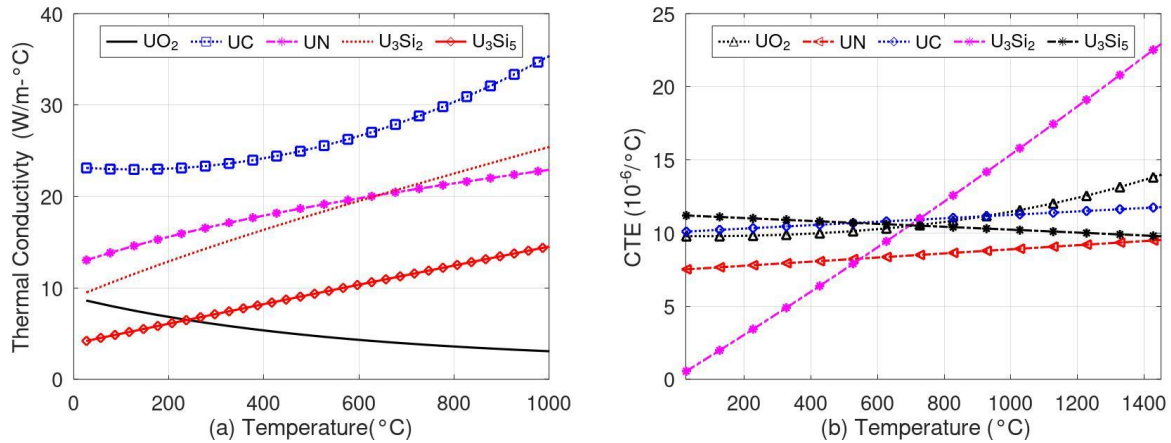


Figure 1: (a) thermal conductivity of ATF (b) thermal expansion coefficients.

The Spark Plasma Sintering (SPS) model's advantage is that it works with shorter processing periods at lower temperatures. This faster-sintering route permits the production of UN- U_3Si_5 , avoiding the ternary phase. The U_3Si_5 phase shields the nitride phase from water due to its better oxidation resistance than U_3Si_2 , and a neutronic response is similar for UO_2 and UN- U_3Si_5 . In contrast, U_3Si_2 and U_3Si under irradiation could produce crystal amorphization.

U_3Si_5 has a poor uranium density (7.5 gU/cm³), lower than U_3Si_2 (11.3 gU/cm³), but it has a high melting point and higher thermal conductivity than UO_2 . Composite UN- U_3Si_5 shows neutronic performance similar to UO_2 . The benefit is that a higher fissile density permits an extended burning cycle. First, work with zircaloy cladding, and the fuel was the typical UO_2 , with a pellet outer diameter of 9.132 mm and a pellet length of 11 mm, with an enrichment of 3.5%. Figure 2 depicts the heat capacity and thermal diffusivity of ATF ceramic fuels.

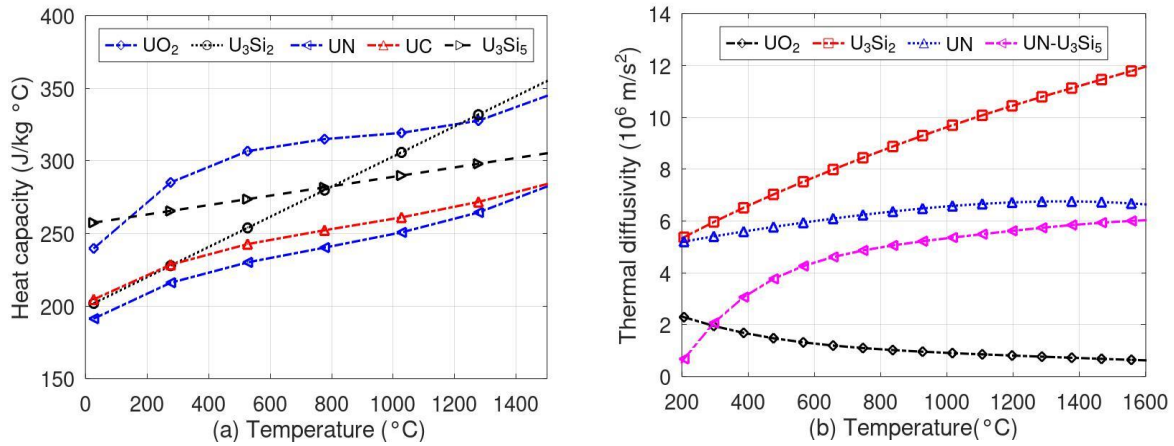


Figure 2: (a) heat capacity of ATF and (b) thermal diffusivity.

3. Results and Discussion

Therefore, the planned composite fuel was UN-10%U₃Si₅ fuel, with ferritic (Kanthal APMT) replacing zircaloy-4. The second phase holds another mechanical dimension of fuel and cladding. However, the enrichment suffers a slight reduction because of the excess uranium density, increasing the neutron multiplication factor. Figure 3 depicts the fuel average temperature and the fission gas release of UO₂ compared with silicide and nitride. The thermal conductivity of pure UN-U₃Si₂ and UN-U₃Si₅ is about 5 to 7.5 times greater than that of UO₂ in the temperature range of the steady-state operation.

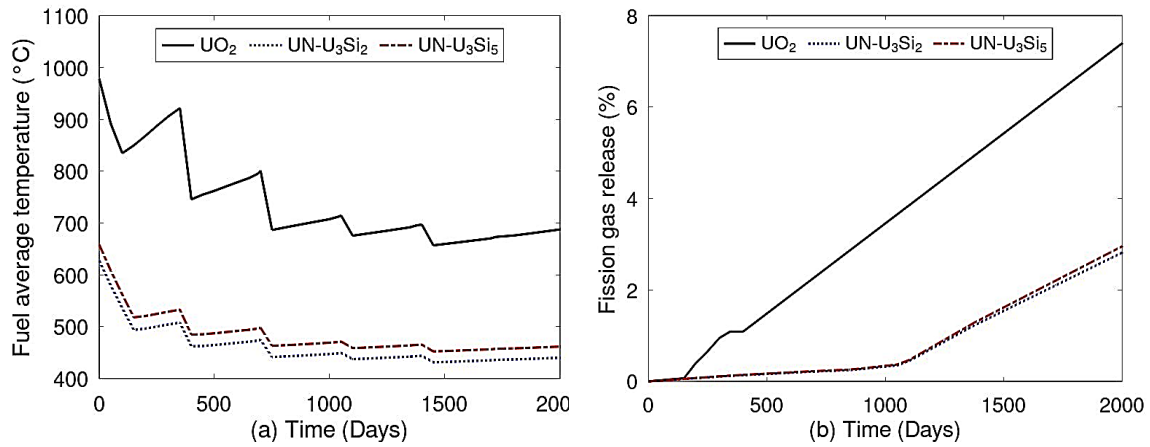


Figure 3: Steady-state response; (a) fuel average temperature; and (b) fission gas release.

Both UN-U₃Si₂ and UN-U₃Si₅ show cycles of around 100 days for UO₂. The dimensional expansion contributes to swelling and pellet cladding interaction (PCI), critical to safe operation. In contrast, we adopted 10% of the weight of U₃Si₅ in this study. FRAPCON should produce a slight fission gas release and thermal expansion because of the decreasing temperature of over 250 °C. The UN-U₃Si₂ fuel system performed well at low burnup, inducing limited pellet swelling and fission gas release (FGR), according to the initial post-irradiation examination (PIE).

This review focused on a marked U-silicide phase, like U_3Si_5 , because U_3Si_5 has a lower U-density and requires a higher enrichment to match the ^{235}U -loading achieved with UO_2 . The ATF program analyzed UN- U_3Si_5 with two volume contents of the UN phase, 55% and 80%, with 35% and 10% of the weight of the U_3Si_5 . Besides, U_3Si_5 is more resistant to irradiation, showing a crystalline form when exposed to a high dose of 8.8 dpa. In contrast, U_3Si_2 loses crystalline properties around 0.3 dpa under the same conditions [6]. When switching from oxide to high-density, the UN- U_3Si_5 composite could minimize operational changes because it displays a thermal neutron response near UO_2 . Besides, combining the UN and silicide phases will improve the thermal conductivity.

4. Conclusions

The aim is to verify the fuel's thermal and mechanical behavior, assuming regular operation. Although the tests carried out during the ATF program presented cycles below 20 GWd/tU, we present a simulation with a cycle of 2000 days. The FRAPCON code was updated to support U_3Si_5 properties. Composite fuel based on UN, U_3Si_2 , U_3Si_5 , or UB_2 for PWRs has many advantages. It will improve nuclear fuel performance by enabling higher burnup, leading to lower waste volumes and longer cycle lengths. The advantages of the UN- U_3Si_5 fuel are its high uranium density, higher thermal conductivity, and reduced degradation of UN in contact with water. The Kanthal APMT with composition [Fe (Bal.) Cr (20–23%) Al (5.8%)] will be the change for zirconium alloys as cladding. Nitride fuels will produce less stored energy under accident scenarios. Kanthal has more resistance to coolant accidents and does not produce hydrogen by oxidation, avoiding zirconium results like hydrogen blasts.

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References

- [1] Matthews, R. B., et al., "Fabrication and testing of uranium nitride fuel for space power reactors." *Journal of Nuclear Materials*, vol. 151, pp. 33–45 (1988).
- [2] Wilson, Tashiema L., et al., "Uranium nitride-silicide advanced nuclear fuel: higher efficiency and greater safety." *Advances in Applied Ceramics*, vol. 117, pp. 76–81 (2018).
- [3] Brown, Nicholas R., Michael Todosow, and Arantxa Cuadra. "Screening of advanced cladding materials and UN- U_3Si_5 fuel." *Journal of Nuclear Materials*, vol. 462, pp. 26–42 (2015).
- [4] White, J. T. et al., "Thermophysical Properties of U_3Si_5 to 1773 K." *Journal of Nuclear Materials*, vol. 456, pp: 442-448 (2015).
- [5] Geelhood, Kenneth J., and Walter G. Luscher. *New Release of Fuel Performance Code, FRAPCON-3.5 and FRAPTRAN-1.5*. No. PNNL-SA-103936. Pacific Northwest National Laboratory. (PNNL), Richland, WA (United States) (2014).
- [6] Hanson, William A., et al., "Post-irradiation examination of low-burnup U_3Si_5 and UN- U_3Si_5 composite fuels." *Journal of Nuclear Materials*, vol. 578, pp. 1543–46 (2023).