

Highlights

Thermophysical Characterization of UFe_3B_2 and USiNi : An Experimental Study

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- Validated ML model's predictions for uranium compounds' thermal conductivity
- First report of UFe_3B_2 and USiNi thermophysical and mechanical properties
- Identified unforeseen experimental challenges beyond ML prediction
- Emphasized need for comprehensive ML framework for advanced nuclear fuel development

Thermophysical Characterization of UFe_3B_2 and USiNi : An Experimental Study

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Abstract

In the development for advanced nuclear fuels for LWRs, the focus has traditionally centered on a limited array of materials like doped- UO_2 , UN, and U_3Si_2 , leaving the vast majority of potential uranium compounds unexplored. To expand the search and expedite the discovery of advanced LWR fuels, we developed a machine learning (ML) classification model dedicated to identifying uranium compounds with excellent thermal conductivity. However, the model's predictive accuracy and the feasibility of such an ML-assisted approach to advanced nuclear fuel discovery have yet to be empirically validated. Therefore, this study examines the thermal properties of two uranium compounds, UFe_3B_2 and USiNi , that are predicted to outperform UO_2 at high temperatures. Our findings confirm that the model precisely predicted the thermal conductivities of both compounds, thereby adding credibility to its reliability. However, the experimental process unveiled several challenges, such as the fabrication of single-phase USiNi and microcrack-free UFe_3B_2 samples, which our ML model, focused primarily on thermal conductivity, could not anticipate. These challenges underscore the need for a more comprehensive ML framework that considers the intricacies of sample fabrication. This advancement is crucial for the more efficient development of practical, accident-tolerant, advanced LWR fuels.

Keywords: accident tolerant fuel, LWR, machine learning, thermal expansion, elastic property, thermal conductivity

1. Introduction

In the aftermath of the Fukushima Daiichi Nuclear Power Plant accident, the development of accident-tolerant fuels (ATFs) has emerged as a crucial aspect of enhancing the safety of LWRs [1, 2]. The purpose of ATFs is to outperform the conventional UO_2 -Zircaloy fuel-cladding system both during normal operations and accidents. A particularly promising approach involves pairing claddings with superior oxidation resistance, such as advanced steel or SiC, with high-density fuels [3]. Compounds like UN, UC, U_3Si_2 , and UB_2 boast high fissile densities, offering a means to offset the neutron absorption penalties of advanced claddings and thus sustain fuel economy without necessitating increased U-235 enrichment. Beyond their high fissile content, these materials are distinguished by thermal conductivities significantly greater than that of UO_2 [4, 5, 6, 7, 8], a critical factor in reducing the fuel's center-line temperature and alleviating associated thermal stress and diffusion of fission products. However, the commercial deployment of these high-density fuels is impeded by significant challenges, notably their reactivity with steam/water in the event of cladding breaches [9, 10, 11, 12, 13]. Additionally, the presence of microcracks in the fabricated high-density UB_2 pellets also raises concerns over their performance [8].

While there have been initiatives to enhance the properties of existing high-density fuel candidates, such as waterproofing

UN [14, 15, 16], the exploration of new uranium compounds as potential advanced fuels has been limited, possibly due to the absence of an effective screening method. To address this issue, we employed machine learning (ML), which has seen tremendous success in the field of materials science [17, 18, 19], to identify uranium compounds with high thermal conductivity that can potentially be used as advanced LWR fuels [20]. Our decision to prioritize thermal conductivity among other defining properties of advanced fuels was informed by the availability of a comprehensive database in Starrydata2 [21], which is imperative to building a robust ML model. Of the assessed 774 stable uranium compounds from the Materials Project [22], our model predicted 146 compounds would exhibit thermal conductivities over $15 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$ between 300 and 1000 K. However, given that the thermophysical properties of most of these compounds have never been reported, it is challenging to evaluate the model's actual performance. As our purpose of incorporating ML to nuclear research is to accelerate the screening for advanced LWR fuels with high thermal conductivity, providing experimental evidence is a crucial next step.

As a continuation of our prior research, we have synthesized and analyzed the thermophysical properties of two uranium compounds, UFe_3B_2 and USiNi , to assess the predictive accuracy of our ML model. Near-single-phase UFe_3B_2 and USiNi were prepared using spark plasma sintering (SPS), and their thermal expansion and thermal conductivity were measured via high-temperature X-ray diffraction (HT-XRD) and laser flash, respectively. Furthermore, we compared the measured thermal

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conductivity of UFe_3B_2 and USiNi with those of existing nuclear fuels candidates and our ML model’s projections. This study provides a first experimental investigation into the viability of an ML-assisted approach for the discovery of novel nuclear fuel candidates. Based on issues revealed during experiments but unforeseen when constructing the ML model, we highlight the challenges that must be addressed for the effective use of ML in the development of practical advanced LWR fuels.

2. Material and methods

2.1. Machine learning model

Complete details of the multiclass classification machine learning model used to predict the thermal conductivity of uranium compounds are available in our prior publication [20]. Additionally, the corresponding machine learning code has been made publicly accessible at the provided GitHub repository: <https://github.com/AzarashiYifan/classification-uranium-thermal-conductivity>. Herein, we provide a quick summary of the ML algorithm employed.

The thermophysical dataset used to train the classification model was extracted from the Starrydata2 database developed by Katsura et al. [21]. Starrydata2 is a database that contains various experimentally-determined thermophysical properties, with a particular emphasis on thermoelectric materials. From the available 372,480 data entries, we processed our dataset to 168,918 records for training. For example, we limited the training data to temperatures from 300 to 1000 K because of data scarcity at higher temperatures. The materials’ thermal conductivities were classified into three distinct categories based on their performance relative to UO_2 : inferior ($0\text{--}5 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$), comparable ($5\text{--}15 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$), and superior (over $15 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$). To minimize data leakage, we grouped the data according to the primary components of the compounds and implemented data augmentation strategies to address class imbalance issues. For the classification model, we incorporated 133 features: 132 from the Magpie feature set [23] and the temperature variable. The Magpie features include various properties and their statistical distribution (e.g. max, min, range, mode) on the constituent atoms of a compound. Subsequently, we trained a random forest classifier and evaluated its performance with 10-fold cross-validation. The trained classification model has a recall of 0.89, 0.49 and 0.71 for classes 0–5, 5–15, and 15+ $\text{W}\cdot\text{m}^{-1}\text{K}^{-1}$, respectively.

2.2. Sample selection

For the experimental phase, we sought to fabricate two compounds whose thermal conductivities were predicted to reside within the $5\text{--}15 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$ or over $15 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$ ranges at temperatures ranging from 300 to 1000 K. Given that compounds with thermal conductivities in the $0\text{--}5 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$ range do not align with the objectives for advanced fuels, they were not considered for fabrication. To ensure the viability of fabrication, we excluded compounds lacking an entry in the Inorganic Crystal Structure Database (ICSD). However, entries in ICSD does not guarantee single-phase can be easily fabricated, such as the

case for such as UN_2 [24] or UBN [25], which requires the element of human judgment during material selection. We further narrowed down the candidates by considering the abundance of constituent elements, excluding rare elements like Re, Ir, and Pt due to scarcity and cost. Additionally, we ruled out elements that pose significant fabrication challenges, such as P, S, and As, due to their toxicity or volatility. However, we did not rule out boron (B) despite the fact that natural boron, which contains approximately $\sim 20\% \text{ }^{10}\text{B}$, can introduce a neutron absorption penalty in the fuel. This is because, by deliberately adjusting the $^{10}\text{B}/^{11}\text{B}$ ratio, it is possible to tailor the fuel’s reactivity, thereby eliminating the need for additional burnable poisons. We also chose not to select simple binary metallic uranium compounds such as UTi_2 and UCo , which are readily inferred to have high thermal conductivity based on their electronic properties. This selection process led us to choose UFe_3B_2 and USiNi as the subjects of our study. Both compounds met all outlined criteria and were deemed suitable for further investigation. It is important to note that the uranium densities of UFe_3B_2 and USiNi are lower than those of traditional fuel candidates such as UC and UN, due to the inclusion of heavier elements. While the primary focus of this study is to evaluate the performance of our machine learning model through experimental investigations, it is crucial to consider uranium density in the selection process for advanced LWR fuels.

2.3. Fabrication

UFe_3B_2 and USiNi samples were fabricated by arc melting stoichiometric amounts of U, Fe (4N, Kojundo Chemical), B (3N, Furuuchi Chemical), semiconductor-grade Si, and Ni (3N, Furuuchi Chemical). The resulting ingots were crushed and hand-milled, and the subsequent powders were densified via SPS under argon flow (200 mL min^{-1} , 6N, Air Liquide). The SPS parameters are detailed in Table 1. Post-SPS, the UFe_3B_2 pellet was wrapped in carbon foil and annealed at 1223 K for 24 hours in a vacuum-sealed silica tube. Similarly, the densified USiNi pellet was warped in carbon foil and heat-treated in a vacuum-sealed silica tube at 1073 K for seven days.

Table 1: Spark plasma sintering conditions

Sample	Temperature, K	Pressure, MPa	Time, min
UFe_3B_2	1173	100	20
USiNi	1073	100	15

2.4. Characterization

2.4.1. XRD and SEM-EDS

To analyze the phase purity and thermal expansion of the fabricated samples, we performed X-ray diffraction (XRD) and HT-XRD measurements (Ultima-IV, Rigaku). During HT-XRD experiments, a He gas flow was maintained to prevent sample oxidation. The 2θ scan ranges for both RT-XRD (room-temperature XRD) and HT-XRD were from 20° to 120° , using $\text{CuK}\alpha$ radiation. To focus on the high-intensity diffraction peaks, we limited the 2θ range to between 20° and 90° in the XRD patterns presented in this study. Peak angle calibration

was performed using NIST silicon powder, and the lattice parameters were calculated using a least-squares method that accounted for the samples surface's vertical displacement [26]. The coefficients of thermal expansion were calculated by performing least-squares fitting on the temperature-dependent lattice parameters and volumes with the following equations:

$$\alpha_l = \frac{1}{l_0} \frac{l - l_0}{T - T_0} \quad (1)$$

$$\alpha_v = \frac{1}{v_0} \frac{v - v_0}{T - T_0} \quad (2)$$

Here, the reference temperature T_0 is 298 K, l_0 is the lattice parameter at T_0 , and v_0 is lattice volume at T_0 . l and v are the lattice parameter and lattice volume at a given temperature T , respectively.

Additional investigations into the microstructure and elemental distribution within the samples were conducted using scanning electron microscopy and energy dispersive X-ray spectroscopy (SEM-EDS, JSM-6500F, JEOL).

2.4.2. Thermal and electrical conductivity

The sample's total thermal conductivity κ_{total} is calculated with the following equation:

$$\kappa_{\text{total}} = \alpha \rho C_p \quad (3)$$

where α is the thermal diffusivity measured by laser flash (LFA457 MicroFlash, Netzsch) under argon gas flow (200 mL/min, Air Liquide, 6N), ρ the sample's density, and C_p the specific heat capacity. Density's temperature-dependences are calculated using the two sample's respective volumetric thermal expansions, while heat capacity is assumed to follow the Dulong-Petit law of $C_p = 3nR$, where n is the number of atoms per molecule and R is the ideal gas constant.

The sample's total thermal conductivity can be further divided into lattice thermal conductivity κ_{lattice} and electronic thermal conductivity $\kappa_{\text{electronic}}$, corresponding to the contributions from phonons and electrons, respectively. The electronic thermal conductivity can be calculated using the Wiedemann-Franz law:

$$\kappa_{\text{electronic}} = L\sigma T \quad (4)$$

Here, L , the Lorenz number, is taken as $2.44 \times 10^{-8} \text{ V}^2 \text{ K}^{-2}$, σ is the electronic conductivity, and T is the temperature. Consequently, the lattice thermal conductivity is expressed as:

$$\kappa_{\text{lattice}} = \kappa_{\text{total}} - \kappa_{\text{electronic}} \quad (5)$$

The sample's electrical conductivity σ was measured under vacuum using an in-house built four-probe measurement device, and the device's accuracy was evaluated using nickel (Figure S1, supporting information).

2.4.3. Mechanical properties

The sample's longitudinal (v_l) and transverse (v_s) sound velocities were measured using the sing-around technique (UVM-2, Ultrasonic Engineering) and used to calculate the sample's

mechanical properties using the following equations [27, 28]:

$$G = \rho v_s^2 \quad (6)$$

$$E = G \left(\frac{3v_l^2 - 4v_s^2}{v_l^2 - v_s^2} \right) \quad (7)$$

$$B = \rho \left(v_l^2 - \frac{4}{3} v_s^2 \right) \quad (8)$$

$$\nu = \frac{1}{2} \frac{v_l^2 - 2v_s^2}{v_l^2 - v_s^2} \quad (9)$$

$$\theta_D = \left(\frac{h}{k_B} \right) \left[\frac{9\rho n N_A}{4\pi M (2v_s^{-3} + v_l^{-3})} \right]^{\frac{1}{3}} \quad (10)$$

Here, G , E , B , ν , and θ_D are the shear modulus, Young's modulus, bulk modulus, Poisson's ratio, and Debye temperature, respectively. ρ , h , k_B , n , N_A , and M are the density, Planck's constant, Boltzmann's constant, number of atoms per molecule, Avogadro number, and molar mass, respectively.

Each sample's hardness was evaluated five times with a Vickers hardness indenter (HVM-G30S, Shimadzu) with an applied load of 9.807 N and a holding time of 10 seconds. The sample's Vickers hardness H_v can be calculated using:

$$H_v = 0.1891 \frac{F}{d^2} \quad (11)$$

where F is the applied load (N) and d is the length of the diagonal (mm).

2.5. Fuel pellet's temperature profile

During operation, a nuclear fuel pellet develops a temperature gradient along its radial direction due to its finite thermal conductivity. This gradient is not only critical for determining the fuel's centerline temperature but also influences the generation of thermal stress, which can lead to cracking. To explore the relationship between the fuel's thermal conductivity and these operational safety concerns, we calculated the temperature and linear expansion profiles for various hypothetical fuel pellets (e.g. UO_2 , U_3Si_2 , UFe_3B_2 , USiNi) operating with a linear power density of $40,000 \text{ W} \cdot \text{m}^{-1}$ and a surface temperature of 900 K [29].

For a cylindrical fuel pellet without any center hole, its surface temperature can be derived with Fourier's law, while considering only radial-direction heat transport [29]:

$$\frac{1}{x} \left(\kappa \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial x} \left(\kappa \frac{\partial T}{\partial x} \right) + \frac{P}{\pi} = 0 \quad (12)$$

Here, x is the normalized radial distance r/r_s , r_s the pellet's radius, κ the pellet's thermal conductivity, T the pellet's temperature, and P the linear power density. The following boundary conditions for heat transport are applied,

$$T(x = 1) = T_s \quad (13)$$

$$\frac{\partial T}{\partial r} = 0 \quad \text{at } x = 0 \quad (14)$$

and the temperature T at a given location x can be obtained via:

$$\int_{T_s}^T \kappa(T) dT = \frac{P}{4\pi} (1 - x^2) \quad (15)$$

Here, T_s is the temperature at the pellet's surface, and factors such as uranium enrichment or neutron absorption are considered to be uniform along the radial direction.

3. Results

3.1. Phase characterizations

3.1.1. UFe_3B_2

The XRD patterns of the UFe_3B_2 sample post SPS and subsequent annealing at 1223 K are presented in Figure 1. The SPS-fabricated UFe_3B_2 sample exhibited a single hexagonal phase, with a very faint UO_2 impurity peak [30] at approximately 28° . However, post-annealing, the intensity of the UO_2 impurities peaks (most notably at 28° and 33°) increased, indicative of oxidation during annealing. The decision to anneal the single-phase UFe_3B_2 sample while risking further oxidation was to eliminate the hysteresis in the measured thermal conductivity, later detailed in Section 3.3.1. Finally, the two peaks marked with triangles in Figure 1 are attributed to tetragonal- Fe_2B [31].

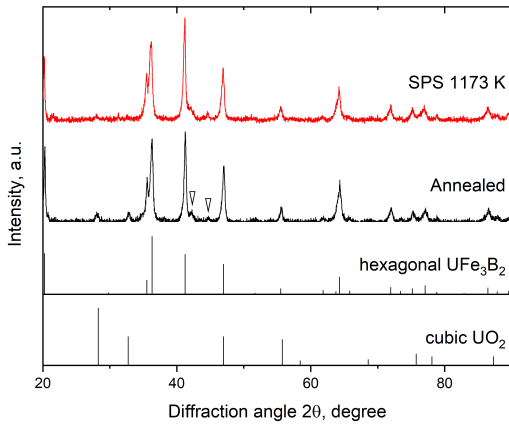


Figure 1: XRD patterns of the UFe_3B_2 sample after SPS and annealing, with reference patterns for UFe_3B_2 [32] and UO_2 [30]

Lattice parameters of UFe_3B_2 remained relatively consistent before and after annealing, aligning well with previously reported values (see Table 2). The sample also retained a high relative density of 95.4% after annealing. SEM-EDS images of the annealed UFe_3B_2 sample, as shown in Figure 2, confirmed a uniform distribution of its constituent elements. Although cubic- UO_2 diffraction peaks were identified in the XRD analysis, EDS did not reveal oxygen-rich regions, suggesting that the sample experienced only limited oxidation. The bright spots observed in the SEM image are artifacts attributed to the uneven surface of the sample after the polishing process.

3.1.2. USiNi

Prokes et al. [33] have previously indexed the crystal structure of as-cast USiNi samples as having a TiNiSi -type orthorhombic structure, a characteristic we also observed in our densified USiNi sample post SPS. However, considering the

Table 2: Lattice parameters of UFe_3B_2 and the sample's relative density.

	Lattice parameters		Relative density, %
	a (nm)	c (nm)	
SPS	0.5046	0.3007	96.7
Annealing	0.5045	0.3004	95.4
Reference [32]	0.5051	0.2997	-

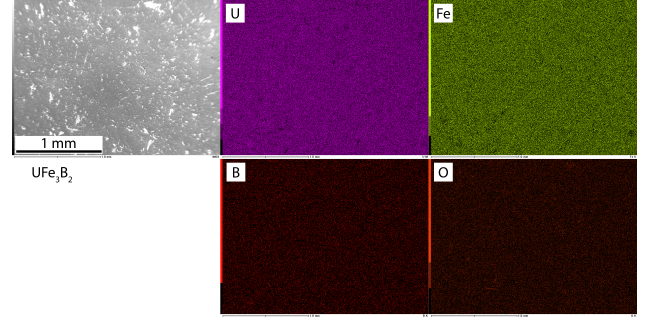


Figure 2: SEM-EDS images of UFe_3B_2 after annealing.

operational conditions of nuclear fuels, our objective was to achieve the hexagonal phase of USiNi , known to emerge after extended heat treatment periods [34, 35]. To investigate the orthorhombic-to-hexagonal phase transition, we annealed the USiNi sample initially for two days, followed by XRD analysis, and subsequently for five days, followed by another XRD measurement. The annealing was discontinued after seven days due to the absence of further changes in the diffraction patterns.

As depicted in Figure 3, after annealing for seven days, the majority of the sample's diffraction peaks correspond to the hexagonal phase. The impurity peaks, marked with triangles, are indexed as orthorhombic $\text{U}_3\text{Si}_4\text{Ni}_4$ [36]. Additionally, two peaks at 31.3° and 36.3° , marked with star symbols, are attributed to cubic $\text{U}_{0.91}\text{Si}_{0.09}$ [37]. Given that these impurity peaks were not present in the sample post-SPS, it is speculated that the orthorhombic-to-hexagonal phase transformation of USiNi during annealing may have led to the formation of these impurities.

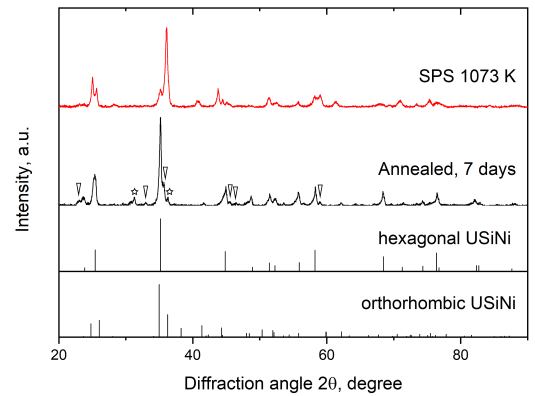


Figure 3: XRD patterns of the USiNi sample after SPS and annealing for seven days, with reference patterns for hexagonal [34] and orthorhombic [33] USiNi .

The lattice parameters of the hexagonal USiNi phase, detailed in Table 3, align well with literature values [34]. Post-annealing, the sample achieved a high relative density of 95.8%, assuming a single hexagonal phase during the calculation. SEM-EDS imaging, as shown in Figure 4, confirmed an overall homogeneous distribution of elements within the sample, suggesting that impurity phases are present in minor quantities. This leads to the conclusion that we have successfully fabricated a near-single-phase hexagonal USiNi sample. For simplicity, hexagonal-phase USiNi is referred to as USiNi in this study. The linear features visible in the SEM image of Figure 4 are attributed to scratches resulting from the polishing process.

Table 3: Lattice parameters of USiNi and the sample's relative density.

	Lattice parameters		Relative density, %
	a (nm)	c (nm)	
Annealing	0.4031	0.3735	95.8
Reference [34]	0.40378	0.37218	-

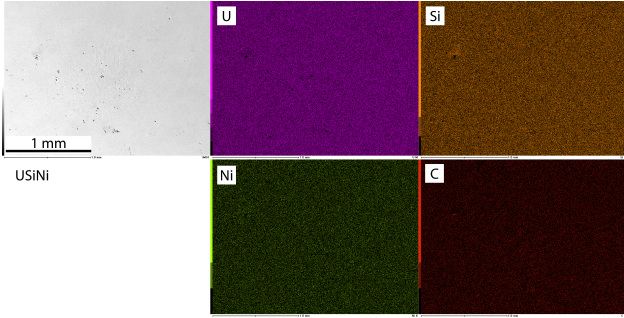


Figure 4: SEM-EDS images of USiNi after annealing.

3.2. Phase stability and thermal expansion

We investigated the high-temperature phase stability and thermal expansion of UFe_3B_2 and USiNi using HT-XRD analysis. The HT-XRD patterns of both materials are provided in the supporting information (Figures S2–S4). Despite conducting the HT-XRD experiments under a constant flow of helium gas, we observed varying levels of sample oxidation at elevated temperatures in both materials.

For UFe_3B_2 , the structure remained relatively stable during heating, with no formation of new impurity phases. At 1073 K, HT-XRD analysis detected only the main hexagonal UFe_3B_2 phase and the two impurity phases, UO_2 and Fe_2B , that were already present in the sample at room temperature. However, the increased intensity of the UO_2 peaks suggests that the UFe_3B_2 sample underwent slight oxidation at elevated temperatures. In contrast, while the USiNi sample remained relatively stable below 773 K, it exhibited poor stability at higher temperatures. Beyond 773 K, a significant increase in the intensity of cubic UO_2 peaks was observed, along with the emergence of new tetragonal USi_2Ni_2 phases [38]. By 1073 K, the hexagonal USiNi phase had completely transformed into UO_2 and USi_2Ni_2 . However, USiNi did not decompose during fabrication when we heated the sample at 1073 K for seven days

in a vacuum-sealed tube. This difference is attributed to the larger surface area of the powdered sample during HT-XRD measurements and the stricter atmospheric control provided by a vacuum-sealed tube compared to the continuous He flow in the sample chamber.

The temperature-dependent lattice parameters of the hexagonal UFe_3B_2 and USiNi phases are illustrated in Figure 5. Due to the significant decrease in peak intensity of hexagonal USiNi after 773 K, its lattice parameters have been calculated only up to this temperature. As shown in Figure 5(a), UFe_3B_2 exhibits highly anisotropic thermal expansion, including a negative thermal expansion along the a -axis, in stark contrast with USiNi. To quantify the thermal expansion coefficients, we performed linear regression analysis on the temperature-dependent lattice parameters and volumes using Equations 1 and 2. A summary of the thermal expansion coefficients along the a and c axes, as well as for the unit cell of both phases, is presented in Table 4.

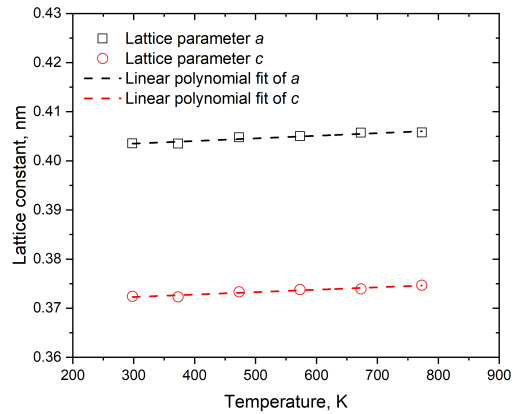
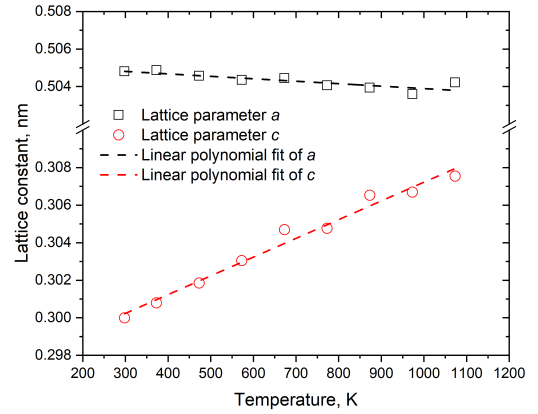


Figure 5: Lattice parameters of (a) hexagonal- UFe_3B_2 from 298 to 1073 K (b) hexagonal-USiNi from 298 to 773 K.

3.3. Thermal and electrical conductivity

3.3.1. UFe_3B_2

The thermal conductivity of annealed UFe_3B_2 , which contains minor UO_2 impurities, alongside predictions from our ma-

Table 4: Thermal expansion coefficients of the a and c lattice axes and lattice volume v of UFe_3B_2 (298–1073 K) and USiNi (298–773 K).

Phase	Thermal expansion coefficients		
	a (10^{-6}K^{-1})	c (10^{-6}K^{-1})	v (10^{-6}K^{-1})
Hexagonal UFe_3B_2	-2.6	33.3	28.0
Hexagonal USiNi	13.2	13.2	39.8

chine learning model, is presented in Figure 6. Predictions are illustrated by the gray areas in the figure because the machine learning model developed classifies thermal conductivity into one of three distinct ranges: [0–5], (5–15], and over 15 $\text{W}\cdot\text{m}^{-1}\text{K}^{-1}$. While the model is trained on data from 300 to 1000 K, it is employed in this work to predict up to 1200 K. This extrapolation to higher temperatures tests whether the model is robust, having truly learned the relationships between input features and the target property. Nonetheless, higher uncertainties in the predicted thermal conductivity above 1000 K should be expected. Overall, the model’s predictions are in excellent agreement with the experimental data in the temperature range of 300 K to 1200 K.

The hysteresis observed in the thermal conductivity of the annealed UFe_3B_2 sample, as shown in Figure 6, was initially evident post-SPS when we conducted laser flash analysis (see supporting information, Figure S5). This observation led us to further anneal the sample in an attempt to eliminate the hysteresis, which proved ineffective, as evident from Figure 6. Consequently, we postulated that the hysteresis is likely due to microcracks that could not be completely healed through annealing. This speculation is supported by the sample’s significantly anisotropic thermal expansion, as detailed in Table 4, suggesting that microcracks formed during sample preparation. In laser flash experiments, these cracks likely healed and reopened as the sample underwent heating and cooling cycles, resulting in the observed hysteresis in thermal conductivity. To ensure a more accurate representation of the thermal conductivity of UFe_3B_2 , we focused on the cooling-phase data (from 1173 K to 300 K), where the sample was presumably in a state with fully healed and not yet reopened microcracks. Consequently, the thermal conductivity of UFe_3B_2 with minor UO_2 impurities is expressed by the following linear equation:

$$\kappa = 9.42 \times 10^{-3}T + 5.20 [\text{W} \cdot \text{m}^{-1}\text{K}^{-1}] \quad (16)$$

Similarly, hysteresis is also observed in the measured electrical conductivity of the annealed UFe_3B_2 sample, as illustrated in Figure 7. Again, the hysteresis is attributed to the closing and reopening of microcracks during the sample’s heating and cooling cycles. During the cooling phase, electrical conductivity data could not be collected below 550 K. This is likely due to deteriorated contact between the sample and the electric probes, potentially resulting from surface oxidation of UFe_3B_2 , as indicated by the blackened sample surface post-measurement. To accurately represent the temperature-dependent electrical conductivity of UFe_3B_2 , we focused exclusively on data obtained during the cooling phase (from 1128 K to 561 K), where the impact of microcracks is minimized. The electrical conductivity

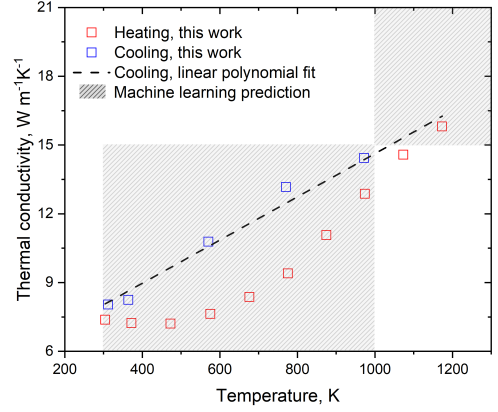


Figure 6: Measured thermal conductivity of annealed UFe_3B_2 sample and predictions by the machine learning model.

of UFe_3B_2 can then be expressed by the following equation:

$$\sigma = 1.20 \times 10^8 T^{-1} + 4.71 \times 10^5 [\text{S} \cdot \text{m}^{-1}] \quad (17)$$

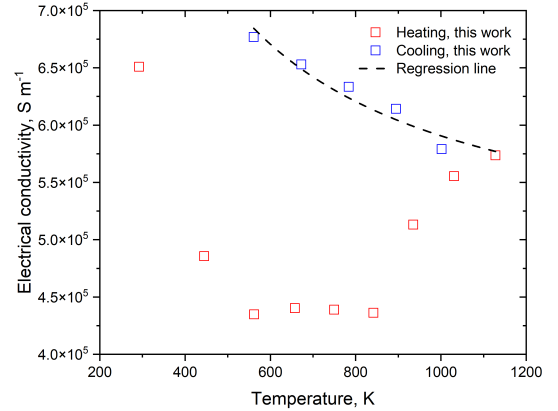


Figure 7: Electrical conductivity of annealed UFe_3B_2 sample measured by the four-point method.

Finally, to calculate the electronic thermal conductivity of UFe_3B_2 , we extrapolated Equation 17 to 300 K. Figure 8 displays the total, electronic, and lattice thermal conductivity of UFe_3B_2 . Consistent with expectations for a metallic compound without a band gap [22], the thermal conductivity of UFe_3B_2 is predominantly influenced by free electron contributions across all temperatures. The dominance of electronic thermal conductivity becomes increasingly apparent at higher temperatures as lattice thermal conductivity diminishes. However, an anomaly is observed starting from approximately 1100 K, where the calculated electronic thermal conductivity surpasses the total thermal conductivity, a result that does not make physical sense. This discrepancy is considered to result from factors such as a Lorenz number for UFe_3B_2 that is lower than the Sommerfeld value of $2.44 \times 10^{-8} \text{V}^2 \text{K}^{-2}$.

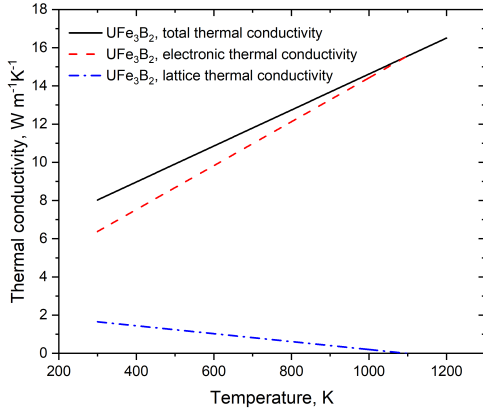


Figure 8: Total, electronic, and lattice thermal conductivity of annealed UFe_3B_2 sample.

3.3.2. USiNi

The calculated thermal conductivity of the annealed USiNi sample, from 300 to 1200 K, aligns well with the predictions of our machine learning model, as shown in Figure 9. In contrast with UFe_3B_2 , the thermal conductivity of USiNi does not exhibit hysteresis and instead demonstrates a linearly increasing trend with temperature. This temperature-dependence can be described by the following equation:

$$\kappa = 6.38 \times 10^{-3}T + 3.25 [\text{W} \cdot \text{m}^{-1}\text{K}^{-1}] \quad (18)$$

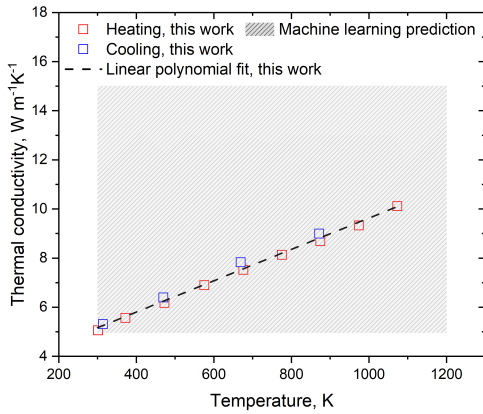


Figure 9: Measured thermal conductivity of annealed USiNi sample and predictions by the machine learning model.

The electrical conductivity of USiNi, measured using an in-house built four-point device, is presented in Figure 10. Due to the relatively poor oxidation resistance of USiNi compared to UFe_3B_2 , as evidenced in our HT-XRD results (Figure S3–S4, supporting information), the obtained electrical conductivity data was limited. During the heating phase, it was not possible to collect data at temperatures exceeding 750 K, likely

due to increased contact resistance. This assumption is supported by the blackened appearance of the sample surface post-measurement. Utilizing the data collected within the temperature range of 294 to 750 K, the temperature-dependent electrical conductivity of USiNi is described by the following equation:

$$\sigma = 1.44 \times 10^7 T^{-1} + 4.50 \times 10^5 [\text{S} \cdot \text{m}^{-1}] \quad (19)$$

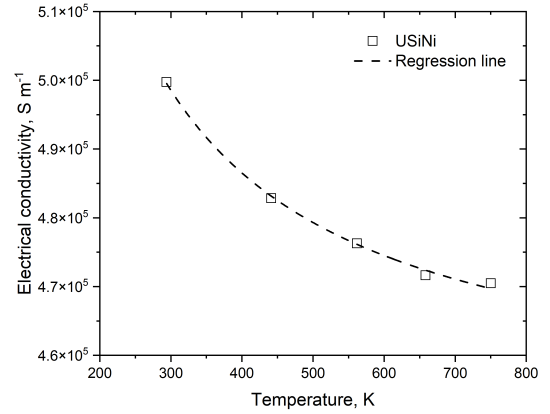


Figure 10: Electrical conductivity of annealed USiNi sample measured by the four-point method.

The electronic and lattice thermal conductivity of USiNi, illustrated in Figure 11, are calculated using Equations 18 and 19. To calculate the electronic thermal conductivity, Equation 19 is extrapolated to 1100 K. Similar to UFe_3B_2 , heat transport in USiNi is also dominated by electrons, especially at higher temperatures. However, at temperatures higher than 630 K, the electronic thermal conductivity calculated using the Wiedemann-Franz law exceeds the total thermal conductivity. This observation suggests that the Lorenz number for USiNi is also likely lower than the Sommerfeld value of $2.44 \times 10^{-8} \text{V}^2 \text{K}^{-2}$.

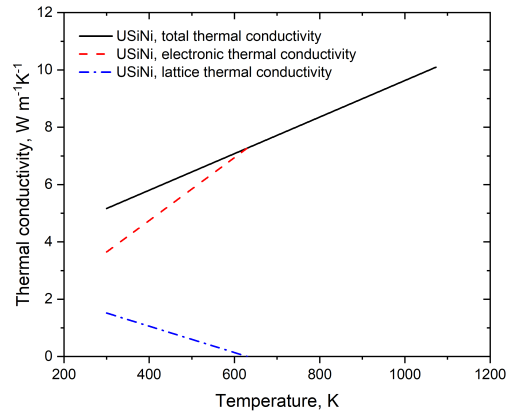


Figure 11: Total, electronic, and lattice thermal conductivity of annealed USiNi sample.

3.4. Mechanical properties

Due to the presence of microcracks in the UFe_3B_2 sample, sound velocity measurements were conducted exclusively on USiNi. The longitudinal and transverse sound velocities of USiNi are determined to be $4271 \text{ m}\cdot\text{s}^{-1}$ and $2359 \text{ m}\cdot\text{s}^{-1}$, respectively. Various derived mechanical properties of USiNi, calculated using Equations 6–10, are summarized in Table 5. This table also includes the average and standard deviation of the Vickers hardness values for both UFe_3B_2 and USiNi. Compared to UO_2 with a 6–7% porosity [39], both UFe_3B_2 and USiNi exhibit superior Vickers hardness at room temperature. Furthermore, the Young's modulus of USiNi is lower than that of UO_2 [40], making it a more pliable pellet. This reduced rigidity can contribute to improving the cladding's integrity by potentially lessening the severity of pellet-cladding mechanical interactions (PCMI).

Table 5: Young's modulus E , bulk modulus B , shear modulus G , Poisson's ratio ν , Debye temperature θ_D , and Vickers hardness H_v of the annealed UFe_3B_2 and USiNi samples, compared with UO_2 .

Properties	UFe_3B_2	USiNi	UO_2 [39, 40]
E , GPa	-	140.1	219.3
B , GPa	-	106.4	202.1
G , GPa	-	54.7	83.1
ν	-	0.28	0.32
θ_D , K	-	297.1	384
H_v , GPa	6.30 ± 0.45	5.79 ± 0.29	4.90

4. Discussion

This section provides a comparative analysis of UFe_3B_2 , USiNi, UO_2 , and other advanced LWR fuel candidates such as UC, UN, U_3Si_2 , and UB_2 . Properties critical to nuclear fuel design, such as thermal conductivity and thermal expansion coefficient are presented in Table 6, at a representative temperature of 900 K. The mean linear thermal expansion coefficient in the table is calculated based on a reference temperature T_0 of 298 K. While it's premature to classify UFe_3B_2 and USiNi as advanced fuel candidates, we have extended our investigation to their performance under simulated operational conditions, which is a valuable step in assessing their potential in comparison to established nuclear fuels.

4.1. Thermal conductivity and temperature profile

The thermal conductivity of a nuclear fuel is a critical factor that influences its temperature profile. As demonstrated in Equation 15, for a given linear power density P and pellet surface temperatures T_s , higher thermal conductivity correlates with lower centerline temperatures. This property is crucial for mitigating the risk of core meltdown and minimizing the heat stored within the fuel pellet. Figure 12 presents a comparison of the temperature-dependent thermal conductivity for UFe_3B_2 , USiNi, and established nuclear fuels between 300 K and 1200 K. At 300 K, the thermal conductivities of UFe_3B_2 and USiNi are 8.03 and $5.16 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$, respectively, comparable to that

of UO_2 . However, at an elevated temperature of 900 K, their thermal conductivities significantly exceed that of UO_2 , whose heat transport is predominantly facilitated by phonons.

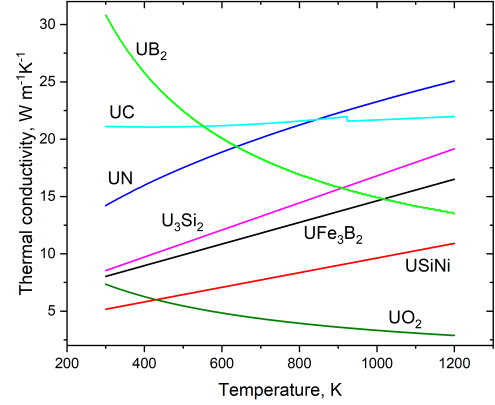


Figure 12: Thermal conductivity of UFe_3B_2 , USiNi, and other established nuclear fuels from 300 to 1200 K.

To quantitatively demonstrate the impact of enhanced thermal conductivity on a fuel pellet's temperature profile, we utilized Equation 15 to simulate the radial temperature distribution, as depicted in Figure 13. Assuming a linear power density of $40,000 \text{ W}\cdot\text{m}^{-1}$ and a surface temperature of 900 K, advanced LWR fuel candidates like UN, UC, U_3Si_2 , and UB_2 show small temperature gradients between the pellet's center and surface, with temperature differentials being 139 K, 147 K, 191 K, and 214 K respectively. The performance of UFe_3B_2 is on par with that of UB_2 , exhibiting a temperature differential of 213 K, due to their similar thermal conductivities at around 1000 K. Although USiNi has a higher temperature differential at 306 K, this is still substantially lower than the 1237 K differential observed for UO_2 , which reaches a centerline temperature surpassing 2000 K. Therefore, the superior thermal conductivity of UFe_3B_2 and USiNi at high temperatures shows promise in enhancing reactor safety by reducing both the peak temperature and the temperature gradient within the fuel pellet.

4.2. Thermal expansion

At a specified temperature, the thermal expansion of a fuel pellet is governed by its thermal expansion coefficient. Lower thermal expansion coefficients are preferred in advanced fuels to prevent PCMIs under all conditions. Moreover, these reduced coefficients help in mitigating the buildup of thermal stress, thereby diminishing the risk of crack formation in the fuel pellets during normal operations.

Among UFe_3B_2 , USiNi, and established advanced LWR fuel candidates listed in Table 6, only UFe_3B_2 and UN exhibit thermal expansion coefficients smaller than that of UO_2 . UC and UB_2 display similar degrees of thermal expansion to that of UO_2 . However, directly comparing these coefficients at a single temperature point, such as 900 K, may lead to misleading interpretations. This is due to the different temperature profiles

Table 6: Thermal conductivity and mean linear thermal expansion coefficients of UFe_3B_2 , USiNi , and other established nuclear fuels at 900 K. Thermal expansion is calculated with a reference temperature of 298 K.

	UFe_3B_2	USiNi	UO_2	UC	UN	U_3Si_2	UB_2
Thermal conductivity, $\text{W}\cdot\text{m}^{-1}\text{K}^{-1}$	13.7	9.0	3.6 [7]	21.9 [5]	22.3 [4]	15.6 [6]	15.8 [8]
Mean linear thermal expansion coefficient, 10^{-6}K^{-1}	9.3	13.2	10.0 [41]	10.8 [42]	8.4 [43]	15.8 [44]	10.2 [8]

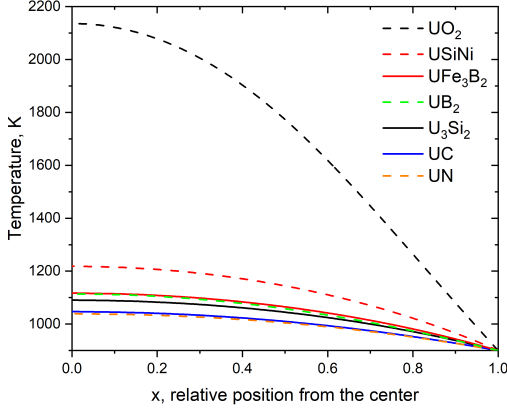


Figure 13: Temperature profiles of the fuel pellets calculated using Equation 15, assuming a linear power density of $40,000 \text{ W m}^{-1}$ and a pellet surface temperature T_s of 900 K.

these materials develop, as illustrated in Figure 13. Therefore, to quantitatively analyze the thermal expansion of these nuclear fuels under simulated operational conditions, we used the temperature distribution from Figure 13 to construct the linear thermal expansion profile, as shown in Figure 14.

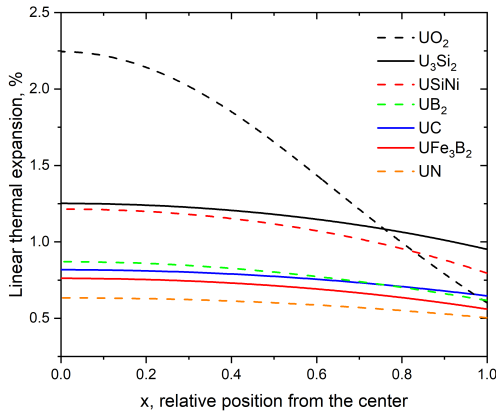


Figure 14: Linear thermal expansion (%) profiles of various fuel pellets along the radial direction. Thermal expansion is calculated with a reference temperature of 298 K.

As illustrated in Figure 14, UFe_3B_2 , USiNi , and various advanced LWR fuel candidates exhibit minimal variation in linear thermal expansion along the radial direction, which can be partly attributed to the pellet's small temperature gradients. Specifically, the linear expansion differentials between the fuel

pellet's center and surface for UFe_3B_2 and USiNi are only 0.2% and 0.4%, respectively. In contrast, UO_2 experiences a significant centerline linear expansion of 2.2%, despite having a lower thermal expansion coefficient than USiNi , UC, U_3Si_2 , and UB_2 . This results from its higher centerline temperature due to its poor thermal conductivity, leading to a pronounced thermal expansion gradient of 1.6%. While this comparison suggests that UFe_3B_2 and USiNi pellets are less susceptible to the safety concerns brought on by thermal expansion, it's important to recognize that these volumetric changes are reversible upon cooling of the fuel rod. Consequently, to comprehensively evaluate the fuel pellet's safety profile during reactor operations, it is crucial to consider irreversible changes associated with thermal and irradiation creep, as well as swelling due to the accumulation of fission products.

4.3. Machine learning in nuclear fuel discovery: challenges and prospects

As illustrated in Figures 6 and 9, our ML classifier has accurately predicted the thermal conductivity of UFe_3B_2 and USiNi from 300 to 1200 K. The agreement between the model's predictions and the experimental results not only demonstrates the capabilities of our model but also highlights the significant role that ML can play in advancing the search for advanced fuels. Nevertheless, the study has also identified several challenges that call for further investigations.

Firstly, the unexpected accuracy of our ML model in predicting the thermal conductivities of UFe_3B_2 and USiNi , despite the low performance metrics reported in our previous work [20], prompts a re-evaluation of how these metrics reflect the model's true performance, especially considering the significant underrepresentation of uranium compounds in the cross-validation dataset. While a dedicated test set of uranium compounds could more accurately assess the model's predictive capabilities on these materials, the limited thermophysical data available on nuclear materials within databases presents a formidable challenge to this approach. For instance, out of the 362,480 data entries from the Starrydata2 database (January 12, 2023 version) used to train the classification model, only 1,945 were on 74 unique uranium compounds. This lack of a comprehensive dataset specific to uranium compounds is a major barrier to the development and rigorous evaluation of high-performance ML models in nuclear materials research.

In addition, while the ML model accurately identified UFe_3B_2 and USiNi as having superior thermal conductivities compared to UO_2 , it is beyond its capabilities to predict the emerged experimental issues, which preclude these materials from being viable advanced fuel candidates. For example, synthesizing a single-phase USiNi sample proved challenging, and

the UFe_3B_2 sample contained microcracks due to its anisotropic thermal expansion. Moreover, HT-XRD analysis indicated that USiNi has poor oxidation resistance, resulting in its decomposition into UO_2 and USi_2Ni_2 at elevated temperatures. These issues in nuclear fuels could arguably present greater risks to reactor safety than lower thermal conductivity. Therefore, the development of a more comprehensive ML framework that evaluates a broader range of fuel properties is crucial in searching for practical advanced fuels. Specifically, the three challenges identified in this study, synthesizability, thermal expansion, and phase stability, are promising areas for future ML research. Furthermore, models for predicting thermal conductivity should include irradiation conditions as input parameters to provide a comprehensive understanding of a fuel's performance during normal operations. However, developing such a model presents challenges, as data on the effects of irradiation may be scarce. Similarly, ML models should extend beyond predicting thermal expansion to include investigations into swelling and creep, thereby enriching our understanding of the fuel's safety performance during operation.

Finally, significant work is required to extend the previously proposed model for predicting the properties of other types of accident-tolerant fuel designs, such as composites. These efforts would involve integrating more descriptive features and acquiring additional data specific to composites. For example, the current model's reliance solely on composition-based features is inherently limiting, as it overlooks parameters known to affect the thermal conductivity of composites, such as volume fraction or morphology. Moreover, a composite-specific testing set is necessary to rigorously evaluate the trained model's performance.

5. Conclusion

The search for advanced nuclear fuels has traditionally been a time-consuming endeavor, predominantly limited to binary uranium compounds such as carbides, nitrides, silicides, and borides. Expanding on our previously proposed ML model, designed to accelerate the discovery of advanced LWR fuels with enhanced thermal conductivity, this study experimentally investigated two such materials: UFe_3B_2 and USiNi . Both compounds were predicted to demonstrate superior thermal conductivity compared to UO_2 but have not been previously investigated.

At 300 K, the measured thermal conductivities of UFe_3B_2 and USiNi are 8.03 and $5.16 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$, respectively. With the temperature increase to 900 K, their thermal conductivities rise to 13.7 and $9.0 \text{ W}\cdot\text{m}^{-1}\text{K}^{-1}$, outperforming that of UO_2 and contributing to significantly lower maximum temperatures and temperature gradients within the fuel pellet. This experiment result is in excellent agreement with our ML model's prediction. In a simplified simulated scenario with a linear power density of $40,000 \text{ W}\cdot\text{m}^{-1}$ and a surface temperature of 900 K, the temperature differentials between the center and surface of the UFe_3B_2 and USiNi pellets are only 213 K and 306 K, respectively. These smaller temperature differentials could potentially mitigate thermal stress buildup and decrease the likelihood of

crack formation, thereby enhancing the operational safety of nuclear reactors.

However, while this experimental investigation highlights the significant potential of machine learning in facilitating the discovery of advanced LWR fuels, it also reveals that neither UFe_3B_2 nor USiNi can currently be considered viable candidates. Our ML model, which focuses on identifying uranium compounds with high thermal conductivity, was unable to predict various issues that emerged during experiments, such as crack formation in UFe_3B_2 and poor high-temperature stability in USiNi . This realization emphasizes the need for more comprehensive ML frameworks capable of evaluating a wider spectrum of properties, essential for the successful discovery of advanced LWR fuels.

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Data statement

The data that support the findings of this study are available from the corresponding author, YS., upon reasonable request. The ML code for thermal conductivity classification is available at <https://github.com/AzarashiYifan/classification-uranium-thermal-conductivity>.

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