Novel Synthesis and Examination of Uranium-Zirconium Carbonitride for Nuclear Thermal Propulsion and High-Temperature Gas-Cooled Reactors

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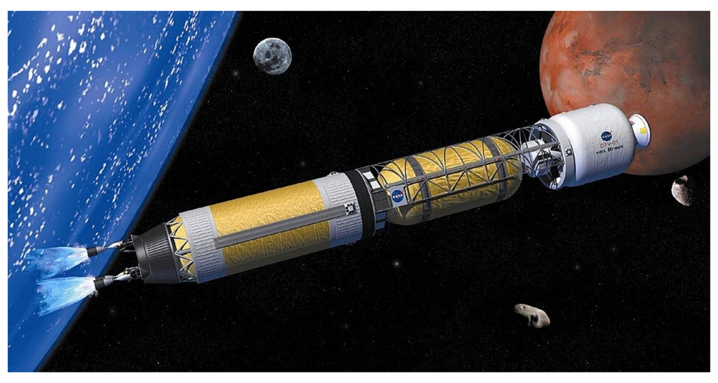
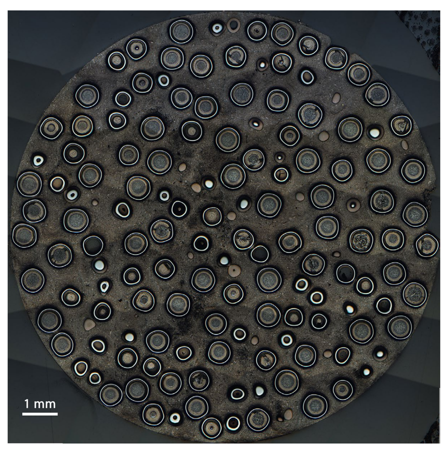
LRS: INL/CON-25-87134

1. What is the problem we're trying to solve?

- Nuclear thermal propulsion is a high-temperature (3000K) reactor that uses hydrogen propellant – there are currently no fuels that can survive the reactor requirements
- We hypothesize that UZrCN can:
  - meet the requirements of modern NTP requirements
  - Improve reactor efficiencies in high-temperature gas-cooled reactors
- UZrCN is highly understudied -- very few studies on the performance of the fuel under prototypic reactor environments
- Currently, the only published way of manufacturing is through carbothermic reduction
- Therefore, the approach of this research is to investigate 3 novel synthesis methods and test UZrCN's viability under prototypic extreme environments/accident tolerance

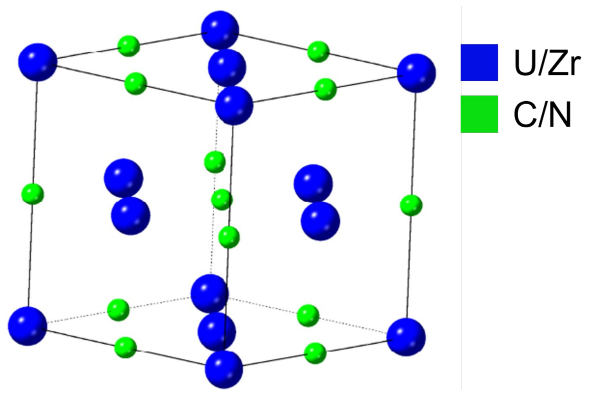




2. Why do we care about UZrCN?

- Uranium-zirconium carbonitride is a promising fuel for advanced reactors
- UZrCN is reported to have:
  - Excellent irradiation behavior [1]
  - Accident tolerant [3]
  - Stable structure, resistant to the amoeba effect [1]
  - Compatible with structural materials [2]
- This fuel is useful for very high temperature gas reactors and nuclear thermal propulsion reactors
- UZrCN has favorable properties such as a high:
  - Thermal conductivity
  - Permissible operation temperature
  - Melting point
  - Chemical resistance

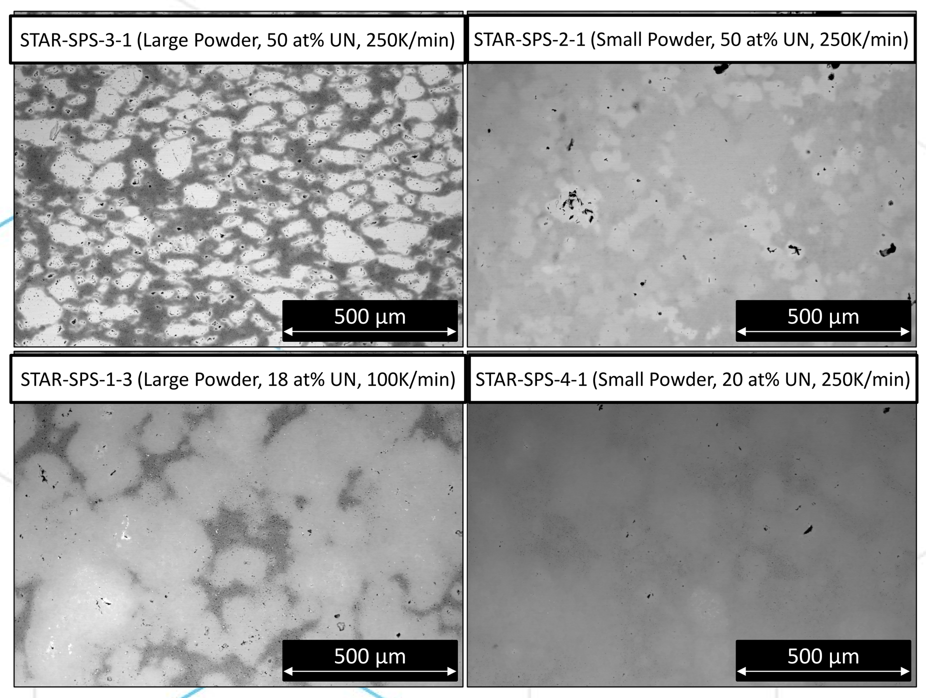
Parameter	UO_2	UN	U-Zr-C-N	Ref
Thermal Conductivity (W/m*K)	3	28	✓ 32	[2]
Permissible Operation Temperature (K)	2200	1700	✓ 2900	[3]
Melting Point (inert medium, K)	3100	3120	✓ 3120	[3]
Uranium Content (g/cm^2)	9.7	13.5	12.5	[2]
Ultimate Strength (MPa)	500	1950	1500	[2]

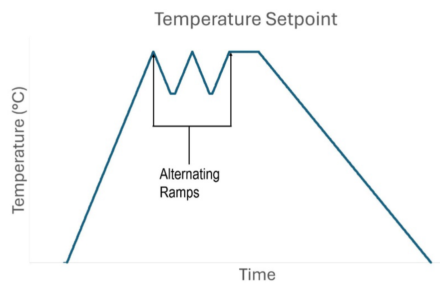


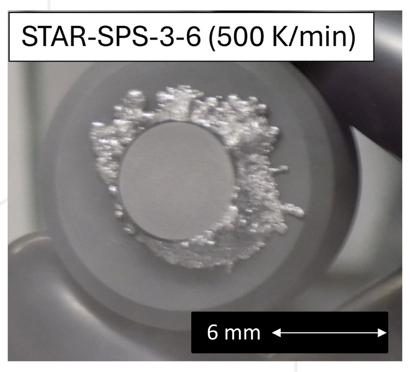
3. Novel Synthesis Methods

Method 1: Electric Field-Assisted Sintering

- This method diffuses uranium mononitride and zirconium carbide powder feedstock into the UZrCN phase which can be done with direct current sintering (DCS) which utilizes an electric field to increase diffusion behavior
- 4 variables were systematically investigated to determine their impact on the diffusion behavior
  - Particle size ✓ Ramp rates ✓ UN Content – Alternating ramps ✗

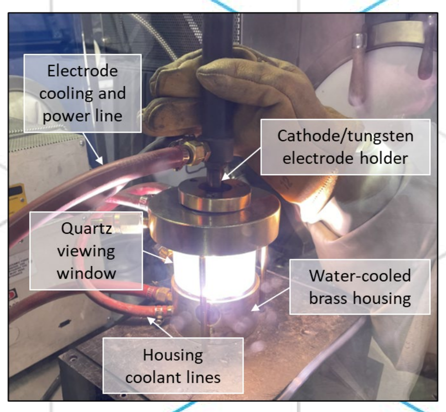


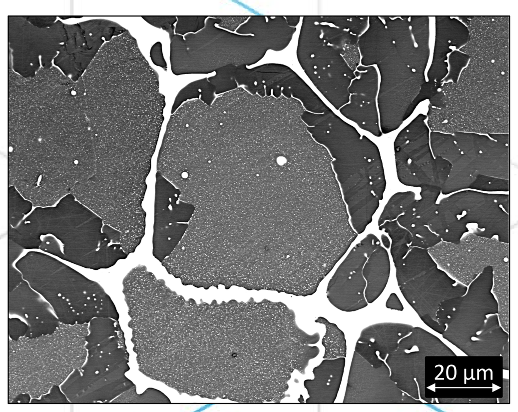




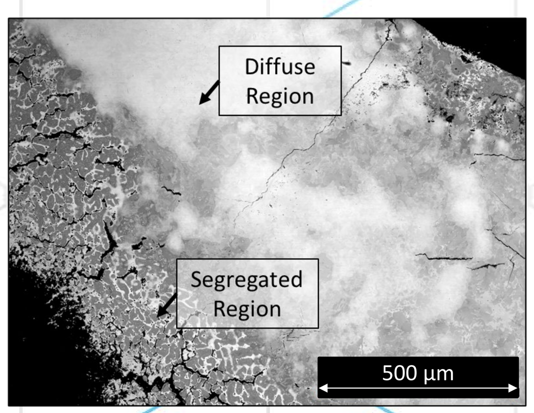
Method 2: Liquid-State Direct Casting

- Alloying uranium, zirconium and carbon under a nitrogen cover gas in an arc-cast melter
- Several remelts are necessary to get an even distribution of the elements with heterogeneity maintained in all samples
- Arc amperage, gas flow rate, alloying order, and feedstock material can all be manipulated to improve outcome
- Annealing under a nitrogen atmosphere has shown to improve homogeneity





As-Cast Microstructure



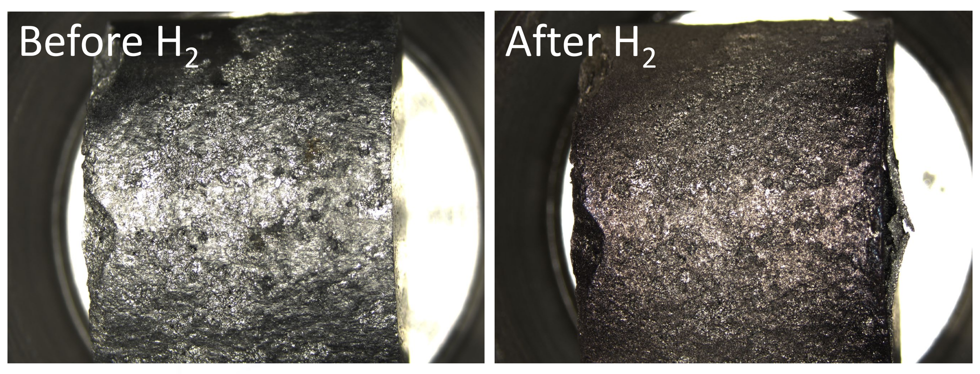
Post-Heat Treatment

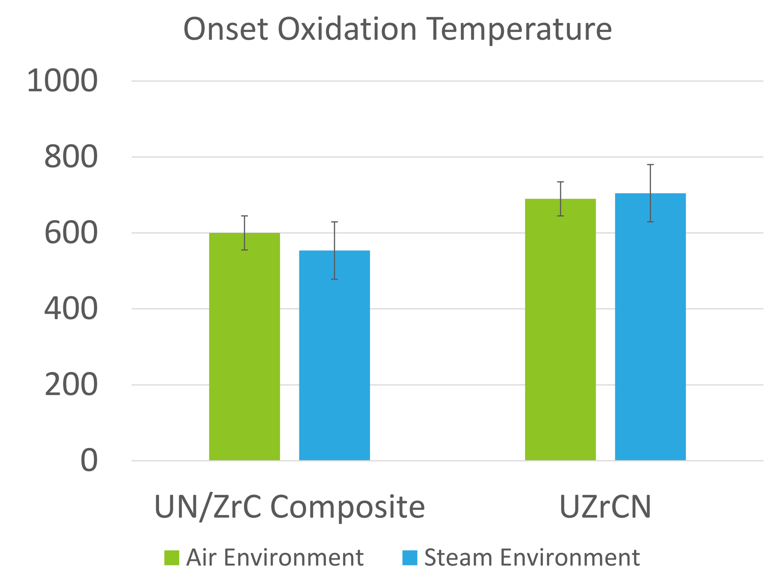
Method 3: Gaseous Cyanide Synthesis

- UZrCN can be synthesized by exposing uranium-zirconium (U-Zr) powder to cyanide gas
- This also allows tight control over the resultant stoichiometry and large batch
- A reaction chamber has been designed and engineered but has been paused due to laboratory constraints

4. Extreme Environment Testing

- To validate the feasibility of this fuel for its intended purposes, the fuel was tested in prototypic environments
- UZrCN was successfully tested in 2600K flowing hydrogen and maintained its physical form.
- For high temperature gas reactors, air and steam ingress are of concern. Samples were tested in air and steam to measure their onset oxidation temperature. UZrCN was found to have an onset oxidation temperature of ~690°C and ~705°C in air and steam, respectively.





Literature Comparison				
Candidate Fuel	Average Onset Oxidation Temp (°C)-Steam		Average Onset Oxidation Temp (°C)-Air	
UZrCN	~700		~660	
UN*	~660	[6]	~390	[9]
UC	<100**	[14]	~260	[15]
$(U_{0.77}Zr_{0.23})N$	~430	[7]	~450***	[16]

*Values are highly density and purity dependent. **Based on non-TGA environmental testing. ***Based on air oxidation of a UN-20ZrN composite.

5. Project Accomplishments

- The technology from this LDRD has been patented in Patent No. 63/503671
- This project has supported 6 students and 2 postdocs
- Research from this project was presented at:
  - American Nuclear Society Winter Conference 2024
  - Pacific Basin Nuclear Conference 2024
  - INL Research and Development Workshop, 2024
  - The Minerals, Metals & Materials Society Conference (TMS) 2025
  - Materials Research Society Conference 2025
  - INL LDRD Symposium 2025
  - Nuclear and Emerging Technologies for Space 2025, 2026
- This project is participating in the Energy Innovation and Commercialization Fellowship to give students an opportunity to learn about technology commercialization
- Customer discovery was performed on this project through the DOE Energy I-Corps Lite program
- Additional funding was won for this project through the NSUF Rapid Turnaround Experiment to do high resolution studies of the UZrCN formation
- UZrCN has been added to the NASA SNP Materials of Interest List to continue the investigation of this fuel
- 3 submitted manuscripts and more to come:
  - "Characterization and Synthesis of Uranium-Zirconium Carbonitride via Liquid Phase Direct Casting"-submitted to Materialia
  - "Point Defects Energetics and Kinetics in ZrC and UN from Density Functional Theory"-submitted to Journal of Nuclear Materials
  - "Synthesis of UZrCN through Field-Assisted Sintering Technique"-submitted to Journal of Nuclear Materials

References

- [1] C. Kouhsen, A. Naoumidis, and H. Nickel, "PREPARATION AND THERMOCHEMICAL STABILTY OF URANIUM-ZIRCONIUM CARBONITRIDES," *J. Nucl. Mater.*, vol. 61, pp. 88–98, 1976.
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- [3] A. N. Bakhin *et al.*, "Preparation for reactor tests of uranium-zirconium carbonitride fuel," *J. Phys. Conf. Ser.*, vol. 1683, no. 3, 2020, doi: 10.1088/1742-6596/1683/3/032040.

Acknowledgements: Work supported through the INL Laboratory Directed Research& Development (LDRD) Program under DOE Idaho Operations Office Contract DE-AC07-05ID14517. We would like to thank the support provided by INL's U100 to conduct this work, the University of Florida's Material Science and Engineering department for their computation contributions, BWXT for their hydrogen testing and the University of Texas-San Antonio for their steam and air testing.

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