UN Multi-design Irradiation Campaign: A Critical Assessment of Accelerated Burnup and Main Correlations for Mechanistic Fuel Performance Modeling

Nuclear Science User Facilities (NSUF)

Annual Program Review

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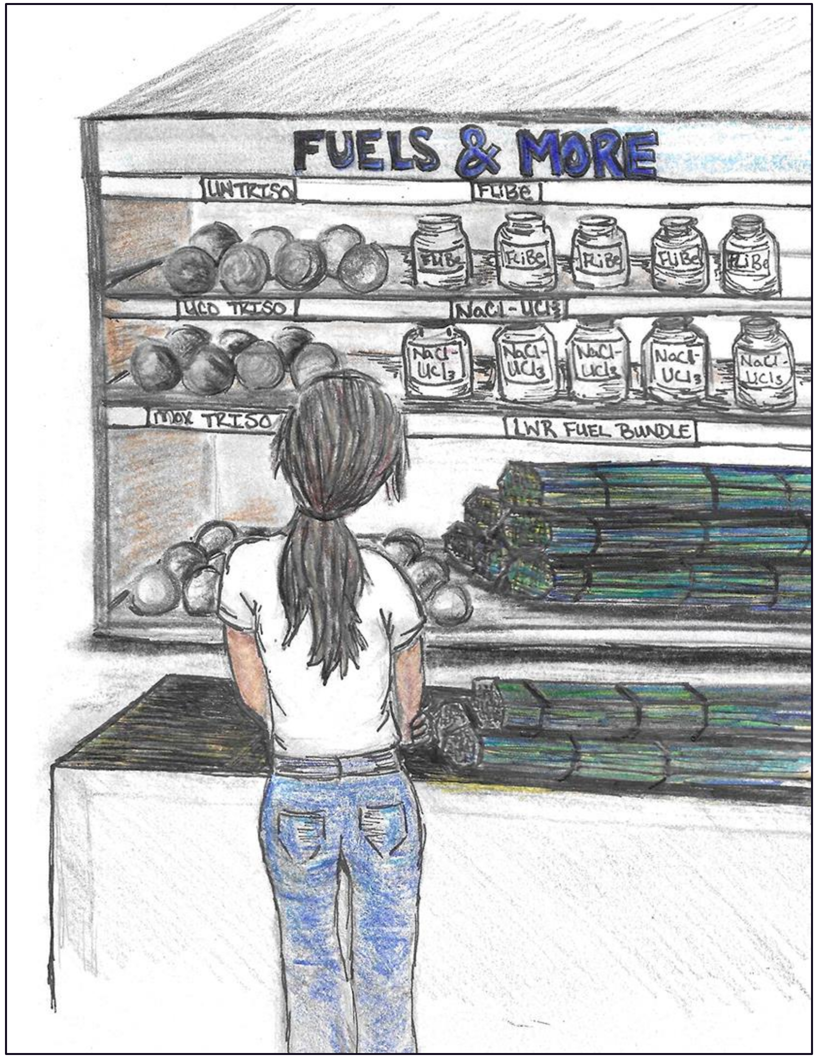
Motivation for fuel development and dynamic testing

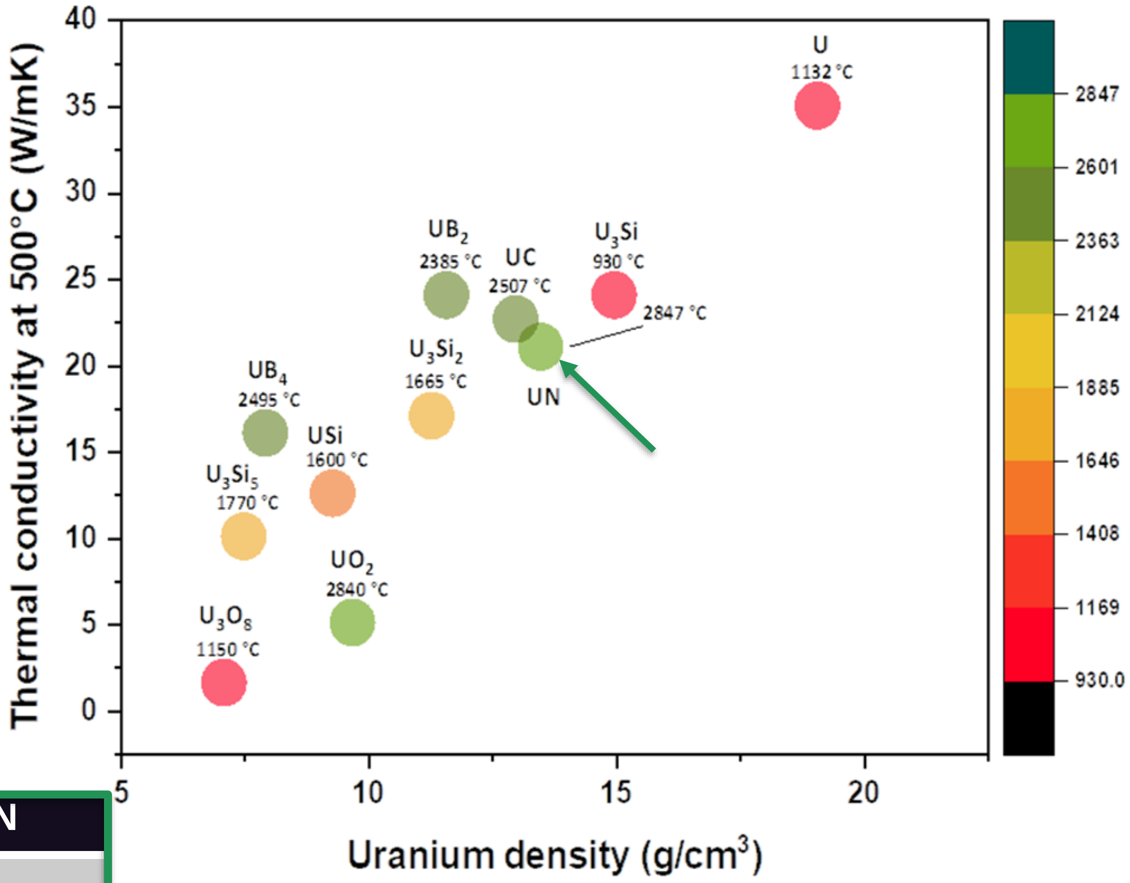
- **There seems to be a new reactor design popping up every other day.**
- **These novel concepts often claim to leverage existing qualified materials.**
- **However, some propose and, in some cases, necessitate the use of unqualified fuels and materials, including but not limited to:**
  - Coolants
  - Structural materials
  - Moderators
  - ***And Fuels***
- **As nuclear material scientists, we try to offer insight to the anticipated performance of these materials.**
  - However, we have data gaps in not only the irradiation performance
  - But also, we lack the fundamental thermodynamic information to even predict equilibrium states
  - The phenomena can span irradiation performance, oxidation, mechanical properties evolution, fuel clad chemical and mechanical interaction, etc.
- **So how do we as a community get to a place where we have multiple qualified, demonstrated materials for the reactor designer to choose from for their optimal design?**



Elizabeth Sooby, *Innovation News Network*, Special Reports, August 2022.

Advantageous Properties of Alternative Fuel Candidates: Focusing on UN

- High uranium atom density
- Low neutron absorption cross section for alloying/compound ion (isotopic separation necessary)
- High melt point and structure stability to melt
- High thermal conductivity
- Inertness to the coolant (depends on the reactor)
- Inertness to the cladding (depends on the fuel form)
- Good mechanical strength
- Stability under irradiation**
  - Several gaps in data, which we aim to address in this NSUF Project

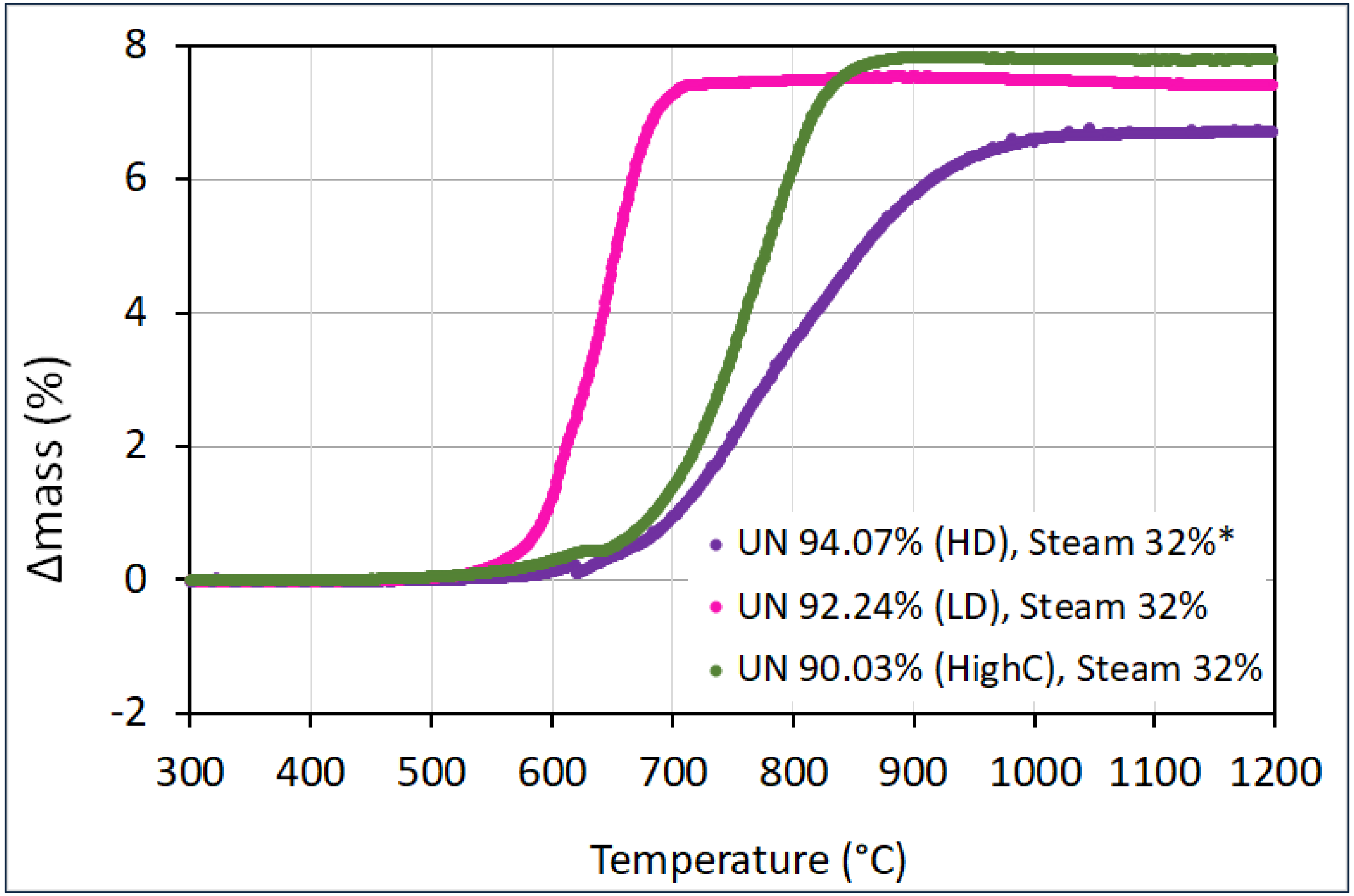


J. Watkins, et al, Journal of Nuclear Materials, 2021.

Material Properties	UO_2	U_3Si_2	UB_2	UN
Uranium density (g-U/cm³)	9.7	11.3	11.7	13.5
Thermal conductivity (W/m·K at 300°C)	6.5 (95% TD)	14.7 (98% TD)	25.1 (95% TD)	16.6 (95% TD)
Melting temperature (°C)	2840	1665	2385	2847

Variation in oxidative performance of UN as a function of sample variability

- **Inertness to the coolant- addressing fuel behavior during a leaker**
  - Clear sensitivity to small % changes in density observed throughout the experimental effort.
  - Inconsistent results with respect to C content, however these were our smallest sample set.
  - Reaction is pulverizing, though less energetic than what was observed with U_3Si_2.
  - Further research is ongoing in this space.
- **This project will address irradiation performance**



E. Sooby, et al*, "Steam Oxidation of Uranium Mononitride in Pure and Reducing Steam Atmospheres to 1200 °C," *JNM*, 560, March 2022.

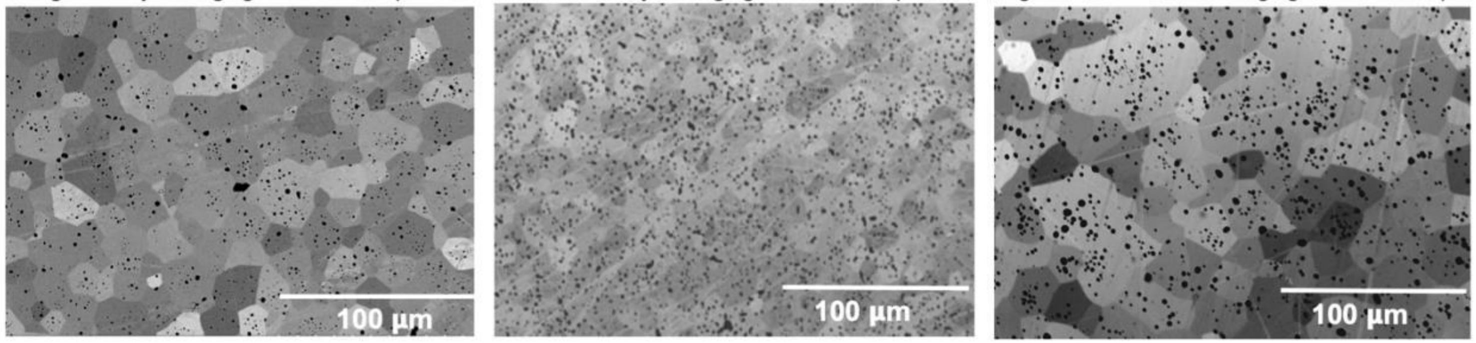
Pellet Before

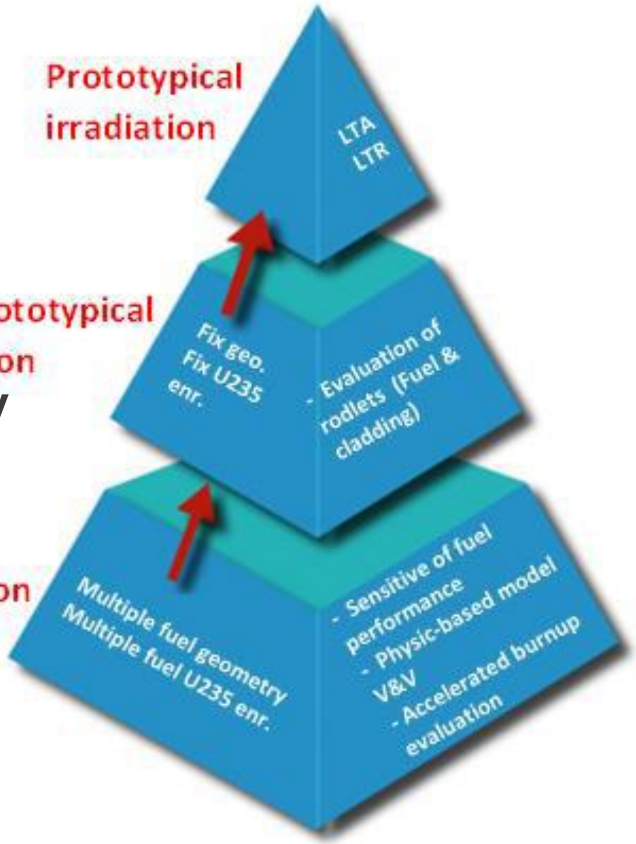
Reaction Product

Addressing variability in performance as a function of sample quality

- **Oxygen and carbon are common fabrication impurities from the CTR-N process.**
- **Further, sintered density variability can cause enhanced oxidation and potentially irradiation performance degradation.**
- **Two new projects led by UTSA aim to assess the impact that impurities which arise at fabrication have on performance.**
  – International NEUP with UTSA, BSU, LANL and University of Manchester to assess impurities which arise at fabrication.
  – These samples along with others fabricated at LANL are leveraged here.
- **The present NSUF irradiation in HFIR will expose samples with varied impurity concentration and varied density to a range of irradiation temperatures and levels of burn up**

High density average grain size: 15.4 μm
Low density average grain size: 13.0 μm
High carbon content average grain size: 19.4 μm





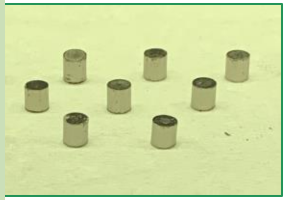
Irradiation Goals

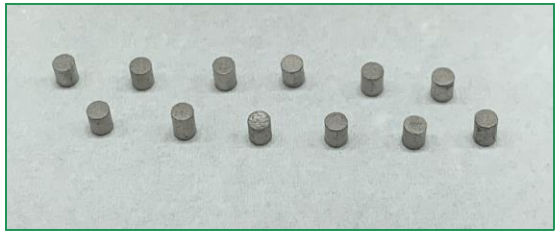
- **There is a lack of Uranium Nitride data in the literature.**
  - Most data is from mixed U,Pu(C,N) irradiations from the 70s that have spotty documentation.
  - Many of these irradiations occurred in EBR-II, but the PIE data from these experiments is hard to find.
  - The exact impurity levels of this material is also not well documented
  - Other more recent tests focused on minor actinide additions to a base mixed nitride (AFC-1, FUTURIX-FTA)
- **MiniFuel provide an opportunity to produce baseline UN irradiation performance without Pu and with well defined C and O content at several different temperatures and densities**
- **Goals for PIE**
  - Fission gas release
    - No data exists at lower temperatures typical of LWR's with the exception of other MiniFuel experiments which is on UN kernels
  - Swelling
    - The basic swelling behavior of phase pure UN is not well understood. EBR-II and AFC irradiations used low smear density pellets bonded with Na to the cladding. It appears that Russian fuel (BN-800) uses a He bond, but open data is sparse from these irradiations
    - Swelling will be measured by geometric changes in the discs before and after irradiation, but it is difficult to measure swelling below 1-5%.
  - Microstructure
    - Porosity and fission gas bubble evolution during irradiation should provide direct feedback to fuel performance modeling of this material
    - The existence or absence of metallic fission products will be established at higher burnups and provide valuable feedback to thermodynamic modeling of the U(C,N) + F.P. systems
    - Phase stability of the UN can also be established, lattice parameters can be compared to pre-irradiation data
    - This work will look for evidence of high burnup structure formation or grain restructuring as seen in UO_2 fuel. However the conditions of this experiment may not be favorable for the formation of those structures even if those structures form in UN.
  - Thermal properties
    - Depending on dose, it may be possible to evaluate the thermal properties (thermal conductivity) of the irradiated discs

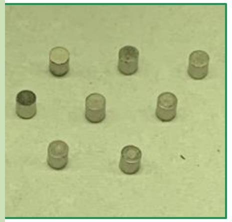
Sample Testing Matrix

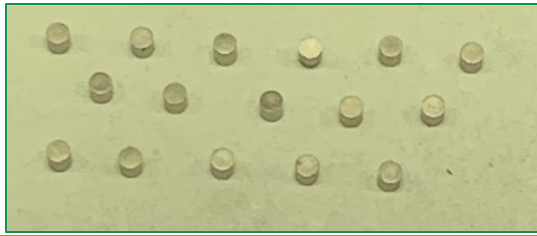
- **First and foremost- we need a name!**
  - **<u>ROADRUNNER</u>** minifuel - Research On ADvancing the peRformance of UraNium Nitrides in Extreme enviRonments
- Next, we need to identify samples and irradiation conditions.

Target #	SubCapsule (labeled by the testing objective)	Sample Geometry	Sample Origin	Targeted Burnup (MWd/KgU)	Temperature (°C)	Pellet Number	Furnace Program	Density	Purity O (%)	O (ppm)	C (%)	C (ppm)
1	High-level Burnup: Control	disc	LANL	75	900	35-P-24-068	24-005-MF		0.0507	507	0.0961	961
1	High-level Burnup: Control	disc	LANL	75	900	35-P-24-067	24-005-MF		0.0507	507	0.0961	961
1	High-level Burnup: Control	disc	LANL	75	900	35-P-24-066	24-005-MF		0.0507	507	0.0961	961
1	High-level Burnup: Control	disc	LANL	75	900	35-P-24-065	24-005-MF	95.167	0.0507	507	0.0961	961
1	High-level Burnup: Control	disc	LANL	75	900	35-P-24-064	24-005-MF	95.422	0.0507	507	0.0961	961
1	High-level Burnup: Control	disc	LANL	75	900	35-P-24-063	24-005-MF	95.253	0.0507	507	0.0961	961
2	High-level Burnup: Varied Temp	disc	LANL	75	1200	35-P-24-087	24-006-MF		0.0725	725	0.134	1340
2	High-level Burnup: Varied Temp	disc	LANL	75	600	35-P-24-082	24-006-MF		0.0725	725	0.134	1340
2	High-level Burnup: Varied Temp	disc	LANL	75	900	35-P-24-081	24-006-MF	96.474	0.0725	725	0.134	1340
2	High-level Burnup: Varied Temp	disc	LANL	75	900	35-P-24-071	24-005-MF		0.0507	507	0.0961	961
2	High-level Burnup: Varied Temp	disc	LANL	75	1200	35-P-24-070	24-005-MF		0.0507	507	0.0961	961
2	High-level Burnup: Varied Temp	disc	LANL	75	600	35-P-24-069	24-005-MF		0.0507	507	0.0961	961
3	Density Variation	disc	LANL	37.5	900	35-P-24-158	24-014-MF	91.6	0.186	1860	0.409	4090
3	Density Variation	disc	LANL	37.5	900	35-P-24-147	24-013-MF	88.4	0.189	1890	0.249	2490
3	Density Variation	disc	LANL	37.5	900	35-P-24-145	24-013-MF	88.4	0.189	1890	0.249	2490
3	Impurity Variation	disc	LANL	37.5	900	35-P-24-172	24-015-MF		0.0436	436	0.524	5240
3	Impurity Variation	disc	LANL	37.5	900	35-P-24-041	24-004-MF	95.148	0.0255	255	0.215	2150
3	Impurity Variation	disc	LANL	37.5	900	35-P-24-166	24-015-MF	95.115	0.0436	436	0.524	5240
4	Mid-level Burnup: Varied Temp	disc	LANL	60	900	35-P-24-083	24-006-MF		0.0725	725	0.134	1340
4	Mid-level Burnup: Varied Temp	disc	LANL	60	600	35-P-24-080	24-006-MF	95.093	0.0725	725	0.134	1340
4	Mid-level Burnup: Varied Temp	disc	LANL	60	1200	35-P-24-079	24-006-MF	96.931	0.0725	725	0.134	1340
4	Mid-level Burnup: Varied Temp	disc	LANL	60	900	35-P-24-077	24-005-MF		0.0507	507	0.0961	961
4	Mid-level Burnup: Varied Temp	disc	LANL	60	1200	35-P-24-076	24-005-MF		0.0507	507	0.0961	961
4	Mid-level Burnup: Varied Temp	disc	LANL	60	600	35-P-24-075	24-005-MF		0.0507	507	0.0961	961
5	Density Variation	disc	LANL	60	900	35-P-24-159	24-014-MF	91.6	0.186	1860	0.409	4090
5	Density Variation	disc	LANL	60	900	35-P-24-148	24-013-MF	88.4	0.189	1890	0.249	2490
5	Density Variation	disc	LANL	60	900	35-P-24-146	24-013-MF	88.9	0.189	1890	0.249	2490
5	Impurity Variation	disc	LANL	60	900	35-P-24-170	24-015-MF		0.0436	436	0.524	5240
5	Impurity Variation	disc	LANL	60	900	35-P-24-042	24-004-MF	95.148	0.0255	255	0.215	2150
5	Impurity Variation	disc	LANL	60	900	35-P-24-167	24-015-MF	94.282	0.0436	436	0.524	5240
6	Low-level Burnup: Varied Temp	disc	LANL	37.5	600	35-P-24-086	24-006-MF		0.0725	725	0.134	1340
6	Low-level Burnup: Varied Temp	disc	LANL	37.5	1200	35-P-24-085	24-006-MF		0.0725	725	0.134	1340
6	Low-level Burnup: Varied Temp	disc	LANL	37.5	900	35-P-24-084	24-006-MF		0.0725	725	0.134	1340
6	Low-level Burnup: Varied Temp	disc	LANL	37.5	900	35-P-24-074	24-005-MF		0.0507	507	0.0961	961
6	Low-level Burnup: Varied Temp	disc	LANL	37.5	1200	35-P-24-073	24-005-MF		0.0507	507	0.0961	961
6	Low-level Burnup: Varied Temp	disc	LANL	37.5	600	35-P-24-072	24-005-MF		0.0507	507	0.0961	961



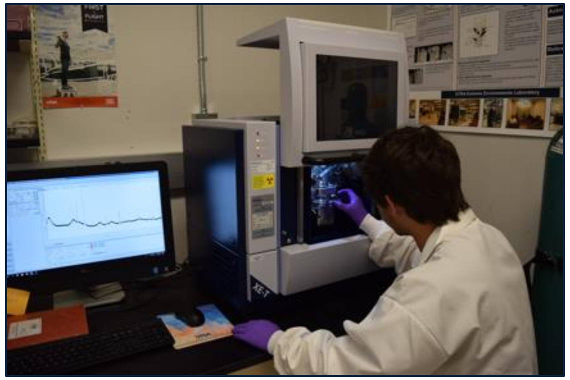






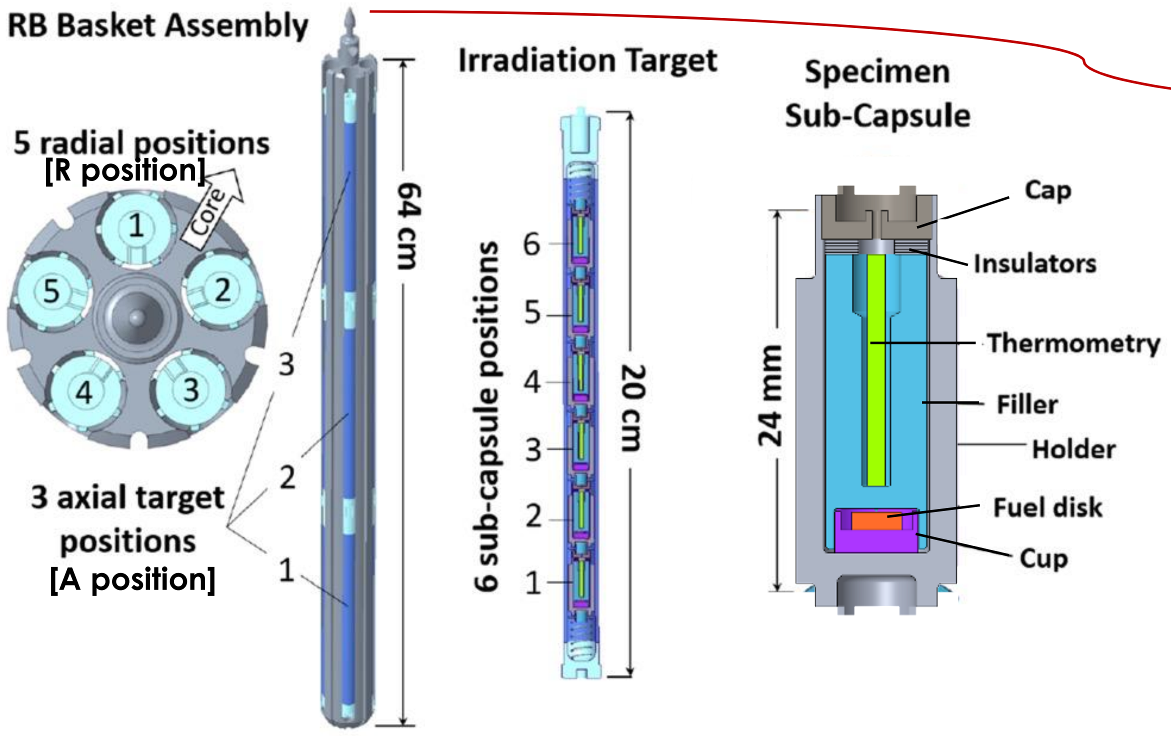
Sample Characterization in anticipation of PIE

- **Geometric Density Measurements (LANL to be repeated at ORNL)**
  - done
- **Light Element contamination Measurements (LANL)**
  - done
- **Scanning Electron Microscopy on samples post thinning (UTSA)**
  - Projected April-May 2024
- **XRD (UTSA)**
  - Projected April 2024
- **Laser Flash Analysis/Thermal Diffusivity (LANL)**
  - To be performed on spare samples from fabrication runs
- **Advanced Microscopy (TEM, EBSD, etc) (ORNL**
  - To be performed on spare samples from fabrication runs
- **XCT (ORNL)**
  - Upon arrival at ORNL
- **Shipment from LANL to UTSA**
  - April 2024
- **Shipment from UTSA to ORNL**
  - May-June 2024

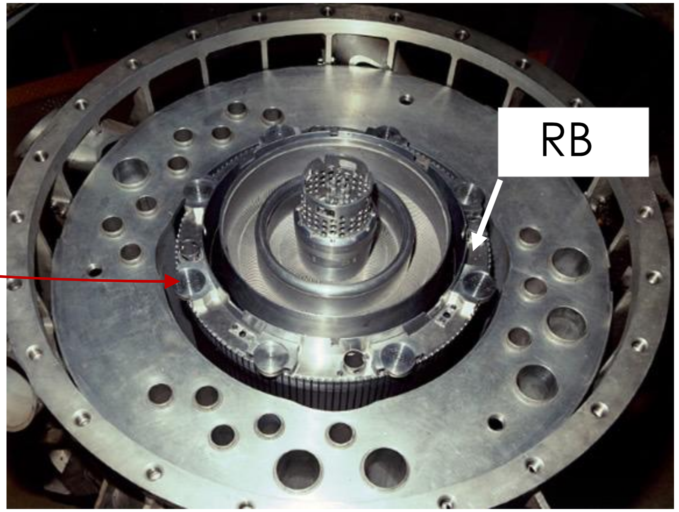


MiniFuel Experiment Design

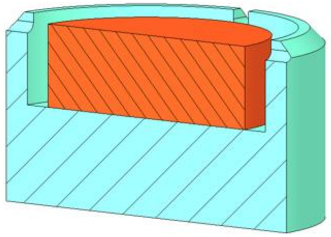
- **Modification of the existing MiniFuel experiment design for insertion in HFIR Removable Beryllium (RB) positions to accommodate 1-mm thick disk specimens**
  - 1mm thickness supports PIE efforts
- **New HFIR drawing approved**



MiniFuel Experiment Design



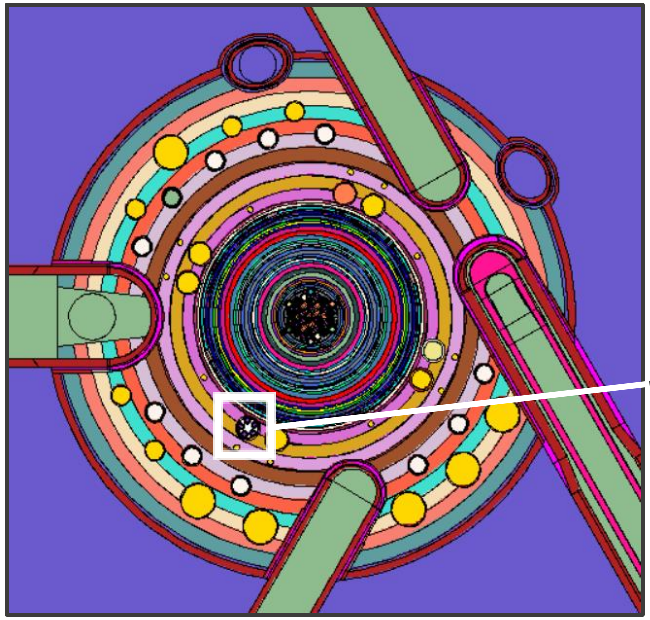
HFIR



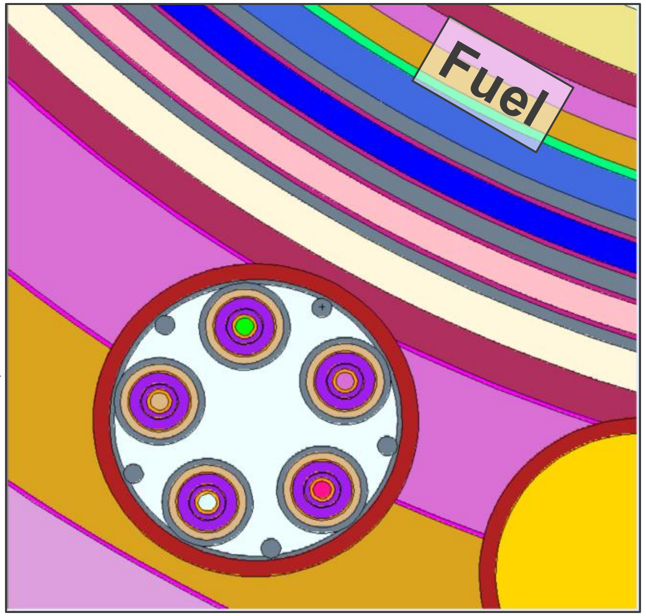
Cross-sectional view of the cup containing the fuel disk (modification of existing design)

Neutronics work

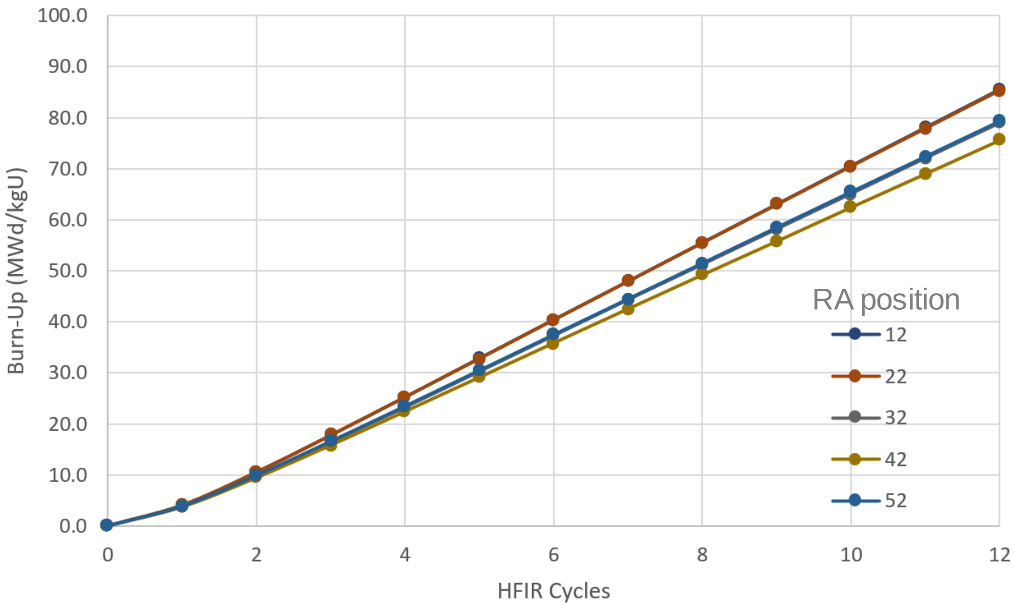
- **Neutronics simulations completed, using HFIRCON (MCNP coupled with ORIGEN)**
- **Results:**
  - Fuel specimen burnup as a function of the number of HFIR cycles
  - Heat generation rates imported into the ANSYS thermal model to calculate experiment temperatures
- **Safety calculations performed and under HFIR review for approval of the experiment**



HFIRCON model of HFIR



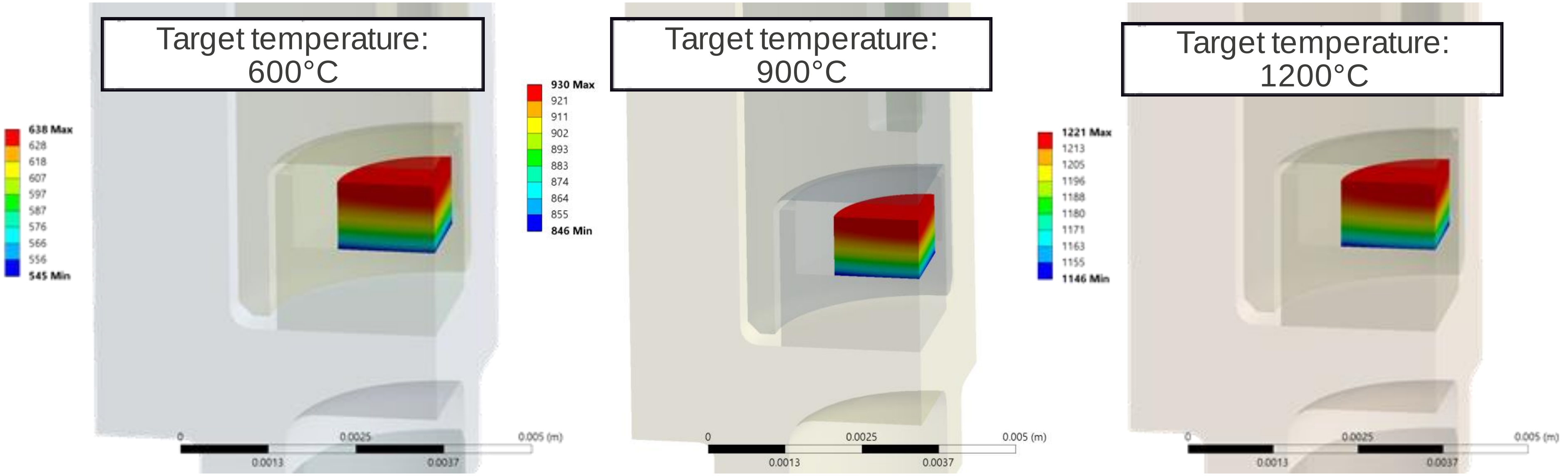
View of the MiniFuel basket in the RB position in the HFIRCON model



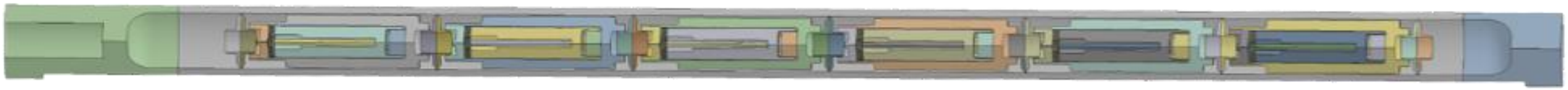
Average fuel specimen burnup for 5 targets positions as a function of the number of HFIR cycles

Thermal analysis

- Axial position 2 chosen for all the targets to prevent large burnup variations per sub-capsules within the same target (5 positions per MiniFuel basket)
- Experiment temperatures calculated with ANSYS
- Holder outside diameters determined to reach target temperatures



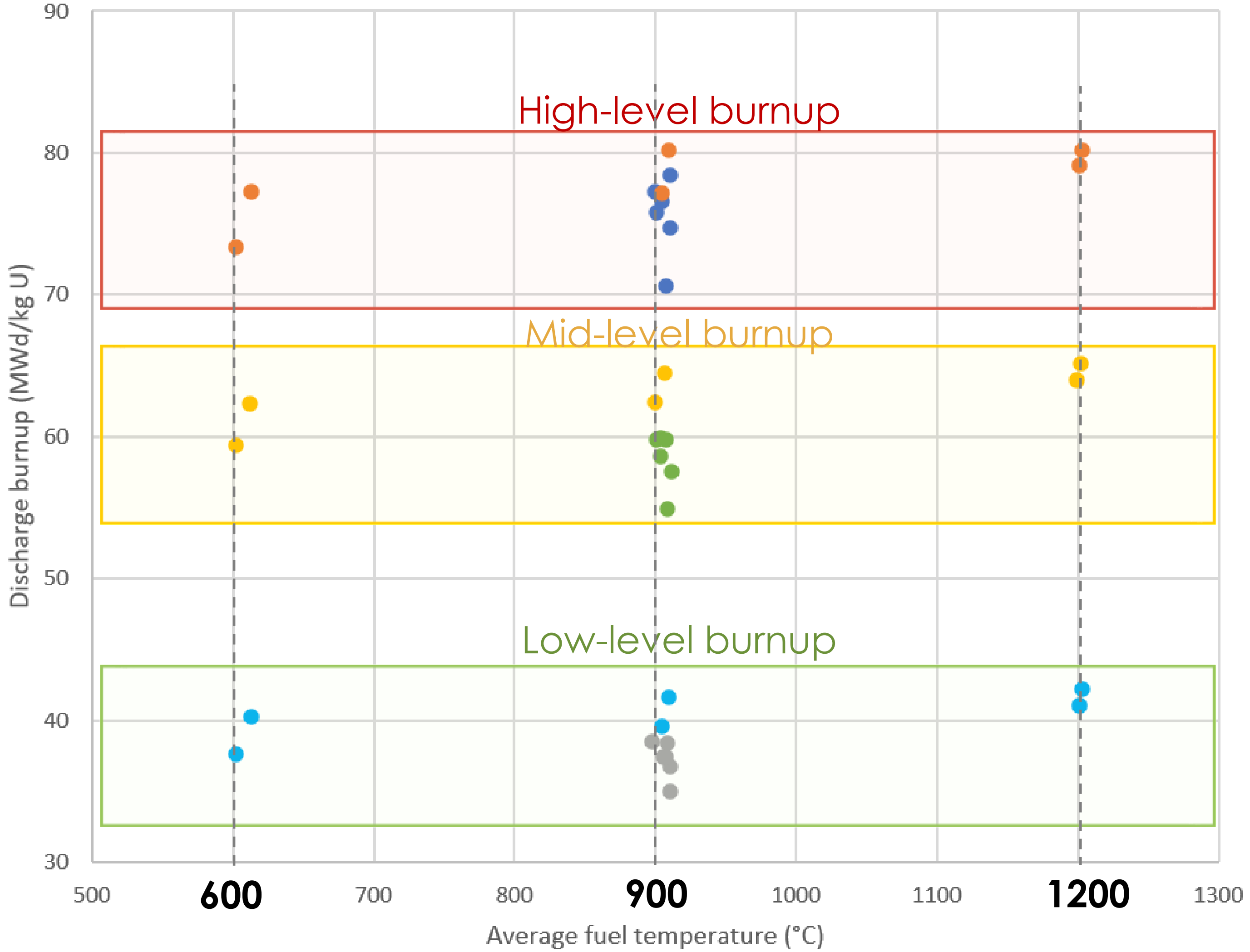
Temperature contours of the fuel specimens for the 3 target temperatures



ANSYS model of a MiniFuel target

Test Matrix – 3 distinct temperatures and burnup levels

- Use of axial positions 2 of the 2 MiniFuel baskets to load the 6 targets

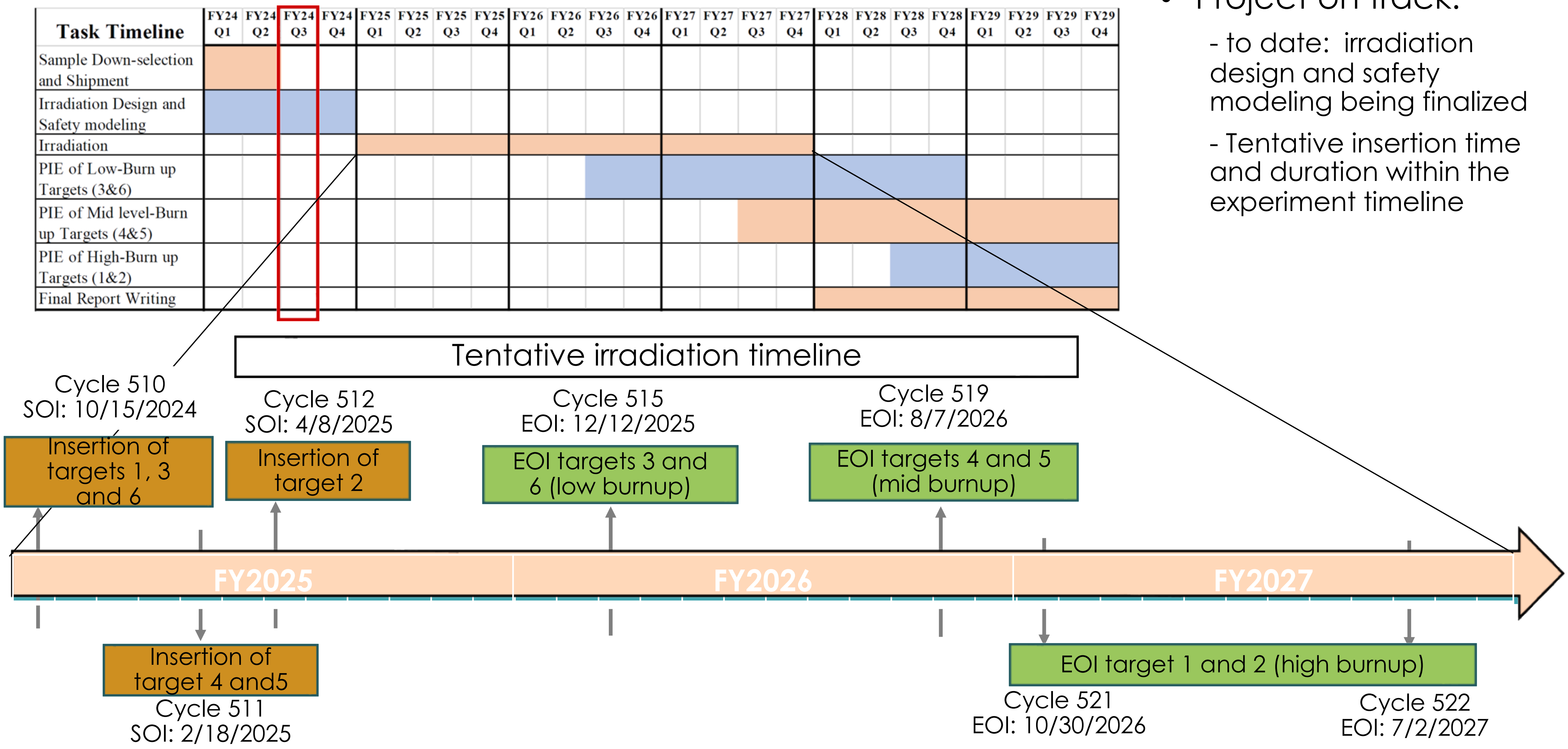


Target	RA position	Burnup	Temp. (°C)	No. of HFIR cycles
Target 1	42	High	900	12
Target 2	12	High	600-900-1200	11
Target 3	32	Low	900	6
Target 4	22	Mid	600-900-1200	9
Target 5	52	Mid	900	9
Target 6	12	Low	600-900-1200	6

- Preliminary results considering up to 7 cycles irradiation – additional cycles not expecting to change the average fuel temperature

EEML Extreme Environment Materials Laboratory
Los Alamos NATIONAL LABORATORY
OAK RIDGE National Laboratory
Westinghouse

Experimental timeline



- Project on track:
  - to date: irradiation design and safety modeling being finalized
  - Tentative insertion time and duration within the experiment timeline

SOI: start of irradiation
EOI: end of irradiation

Acknowledgments

- **Sample fabrication for this experiment was supported by a FY2023 NEUP, award DE-NE0009298, "International Collaboration to Advance the Technical Readiness of High Uranium Density Fuels and Composites for Small Modular Reactors." The fabrication work was also supported by the DOE-NE and Westinghouse Electric Company under contract DE-NE0008824.**
- **Support for the irradiation experiment is funded by NSUF: "UN Multi-design Irradiation Campaign: A Critical Assessment of Accelerated Burnup and Main Correlations for Mechanistic Fuel Performance Modeling"**
  - Proposing team:
    - Elizabeth Sooby (PI) - University of Texas at San Antonio;
    - Denise Adorno Lopes (co-PI) – Oak Ridge National Laboratory
    - Antoine Claisse– Westinghouse Electric Company; (Denise Adorno Lopes and Luke Olson at WEC during proposal development)
    - Kory Linton, Nathan Capps, and Jason Harp – Oak Ridge National Laboratory;
    - Joshua White and Michael Cooper – Los Alamos National Laboratory

- **Questions? Elizabeth.Sooby@utsa.edu**

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