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High Density LWR Fuels

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Accident Tolerant Fuel (ATF) Goal: Enhanced "Grace Time"

*Fuels with **enhanced accident tolerance** are those that, in comparison with the standard UO_2 – Zr system, can **tolerate loss of active cooling** in the core for a **considerably longer time period** (depending on the LWR system and accident scenario) while maintaining or improving the fuel performance during normal operations.*

Enhanced Tolerance to Loss of Active Core Cooling

Improved Reaction Kinetics with Steam
- Decreased heat of oxidation
- Lower oxidation rate
- Reduced hydrogen production (or other combustible gases)
- Reduced hydrogen embrittlement of cladding

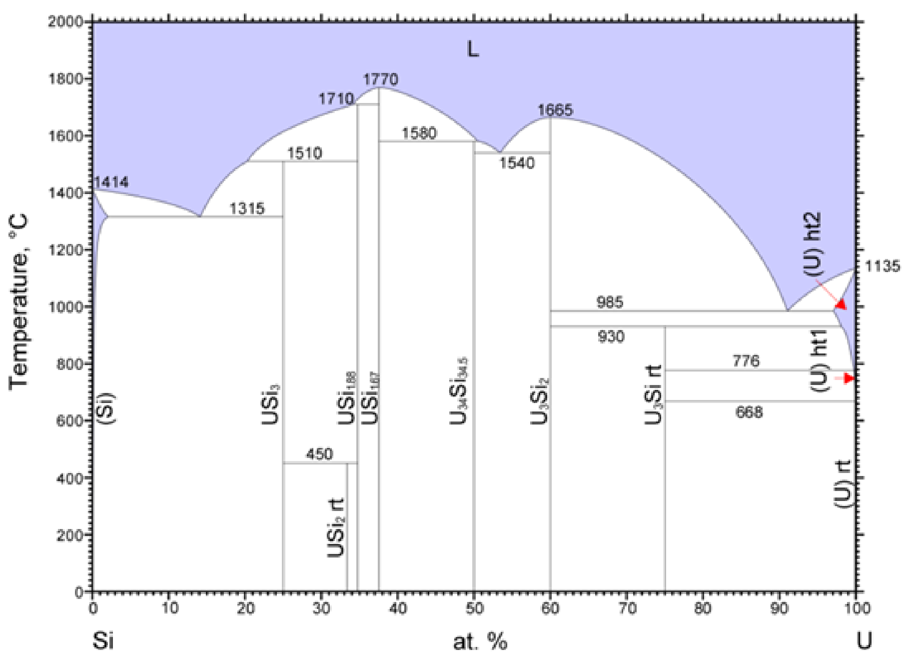
Improved Fuel Properties
- Lower fuel operating temperatures
- Minimized cladding internal oxidation
- Minimized fuel relocation/dispersion
- Higher fuel melt temperature

Improved Cladding Properties
- Resilience to clad fracture
- Robust geometric stability
- Thermal shock resistance
- Higher cladding melt temperature
- Minimized fuel - cladding interactions

Enhanced Retention of Fission Products
- Gaseous fission products
- Solid/liquid fission products

High Density Fuels with oxidation resistance may aid the implementation of ATF technologies

- **Uranium silicide could provide both economic and safety benefits as a replacement for uranium oxide**
  - Greater thermal conductivity
  - Appropriate melting temperature
  - Better uranium loading
    - *U_3Si_2 has 17% more uranium atoms than equivalent density UO_2*



Okamoto, H., Si-U Phase Diagram (1990), ASM Alloy Phase Diagrams Center, P. Villars, editor-in-chief; H. Okamoto and K. Cenzual, section editors; http://www1.asminternational.org/AsmEnterprise/APD, ASM International, Materials Park, OH, 2006.

Property	UO_2	U_3Si_2
Density (g/cm^3)	10.96	12.2
Uranium Number Density ($atom/cm^3$)	2.45×10^{22}	2.86×10^{22}
Thermal Conductivity (W/m·K 400-1200° C)	6 to 2.5	38 to 21 [1] 13.0 to 22.3 [2] 14.5 to 29.1 [3]
Melting Point	2847° C	1665° C
LWR Operating Centerline Temperature Range	1200-1700° C	400-800° C

[1] K.M. Taylor, and C.H. McMurtry, "Synthesis and Fabrication of Refractory Uranium Compounds," TID-6591, No. ORO-340. Carborundum Co., Niagara Falls, NY, 1960
[2] H. Shimizu, "The Properties and irradiation behavior of U3Si2," NAA-SR-10621, Atomics International, 1965
[3] J.T. White, A.T. Nelson, "Report on Performance of Advanced Ceramic LWR Fuel Candidates," LA-UR 13-23673, 2013

Matrix of candidate high fissile density ceramic fuel systems

Matrix/2nd Phase (U g/cm^3)	UO_2 (9.7)	U_3O_8 (7.1)	UN (13.5)	U_3Si_2 (11.3)	U_3Si_5 (7.5)	UB_2 (11.7)	UB_4 (7.9)
UO_2 (9.7)	Reference Case	Neutronic	Rxn	Rxn	Rxn	Thermo	Thermo
UN (13.5)	Rxn	Rxn	Rxn	WEC FOA	Thermo / Neutronic	Thermo	Thermo
U_3Si_2 (11.3)	Rxn	Rxn	WEC FOA	WEC FOA	Rxn	Rxn (Thermo)	Rxn (Thermo)
UB_2 (11.7)	Thermo				Rxn (Thermo)		

Red	Discarded due to reaction with 2nd phase or coolant, and/or neutronics
Yellow	Under initial screening
Green	Viable after initial screening; continuing for further assessment

- **Initial screening by neutronics, thermodynamic calculations, constituent and composite property measurements**
- **LANL-1 test targets UN-U_3Si_5 composite system due to relative maturity of properties and processing**
- **Complements UN-U_3Si_2 development under the Westinghouse-led FOA**

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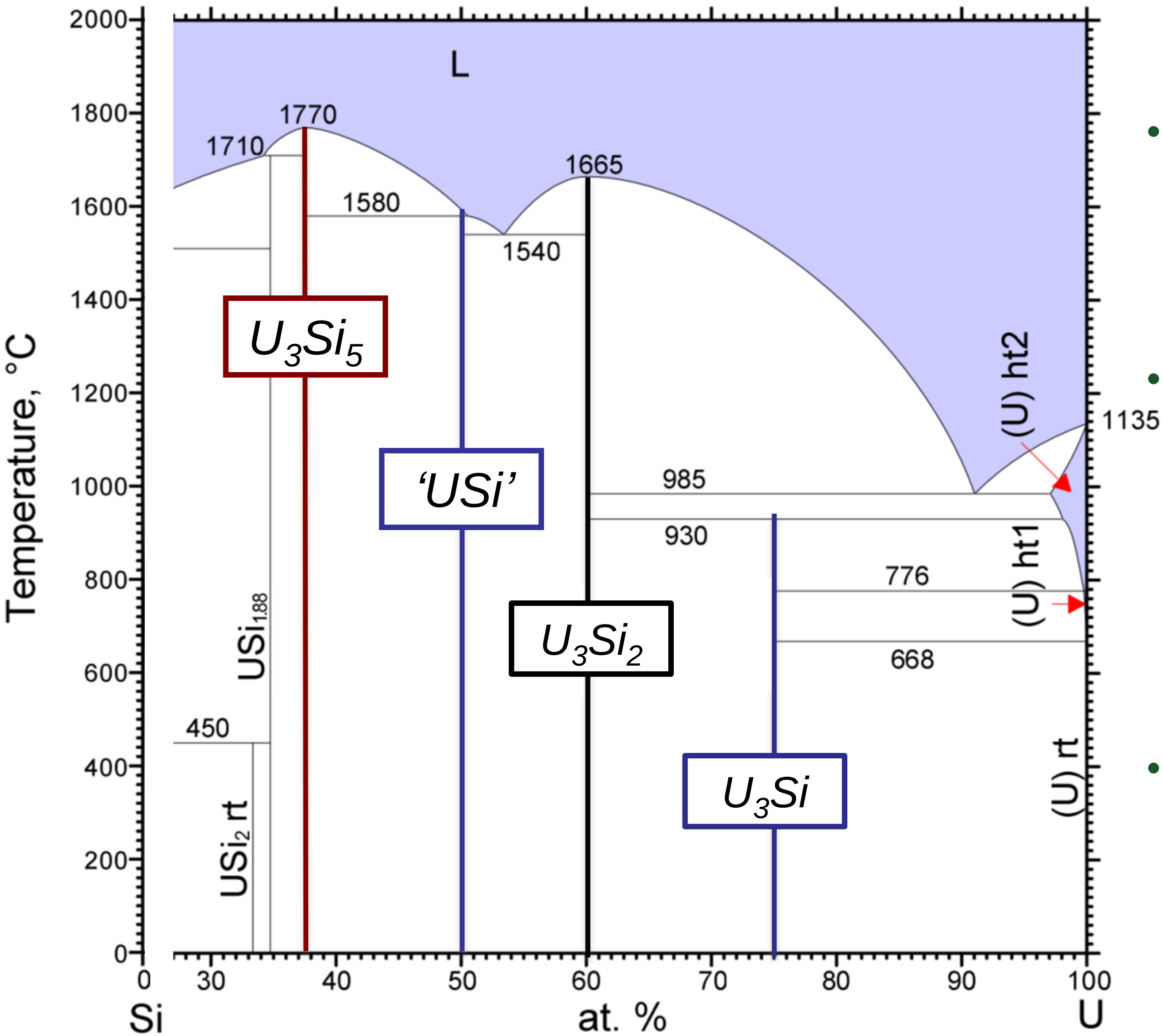
Uranium-Silicon Binary System (U_3Si_2 and U_3Si_5)

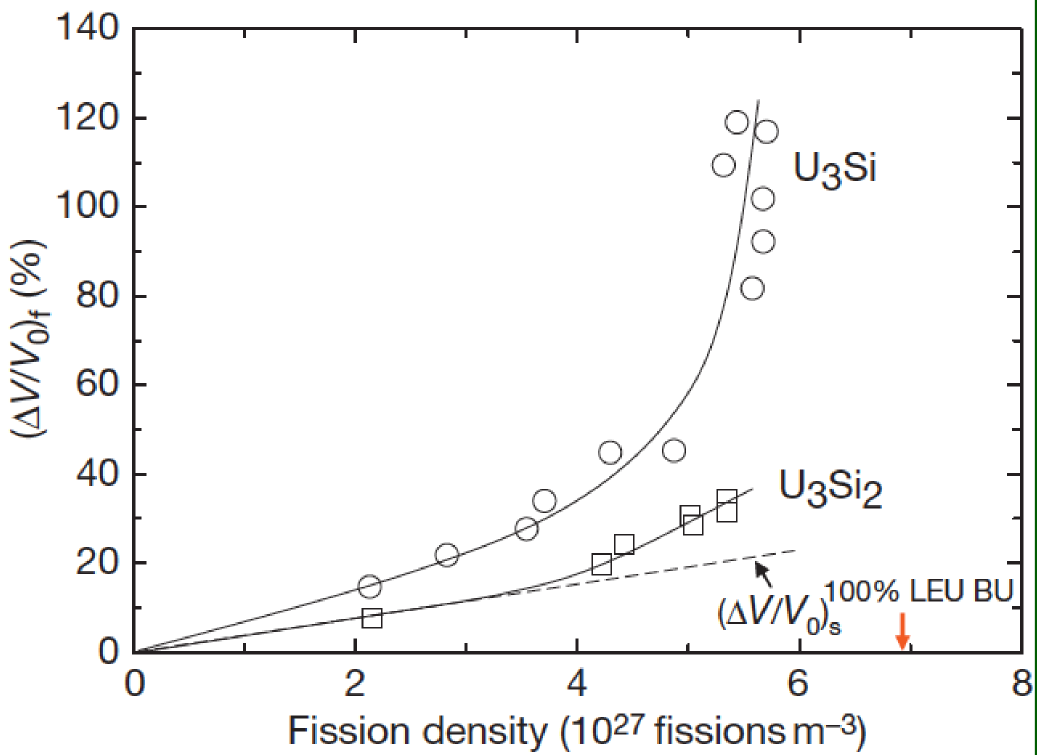


- Restrict discussion to U-Si phases with comparatively high U density
- Moving from U-rich to Si-rich portion of phase diagrams greatly decreases knowledge of U-Si compounds (both fundamental and as fuels)
- *Experimental investigations have suggested uncertainty in 30-40 at. % Si range*

U-Si Compounds for LWR Accident Tolerant Fuel Applications

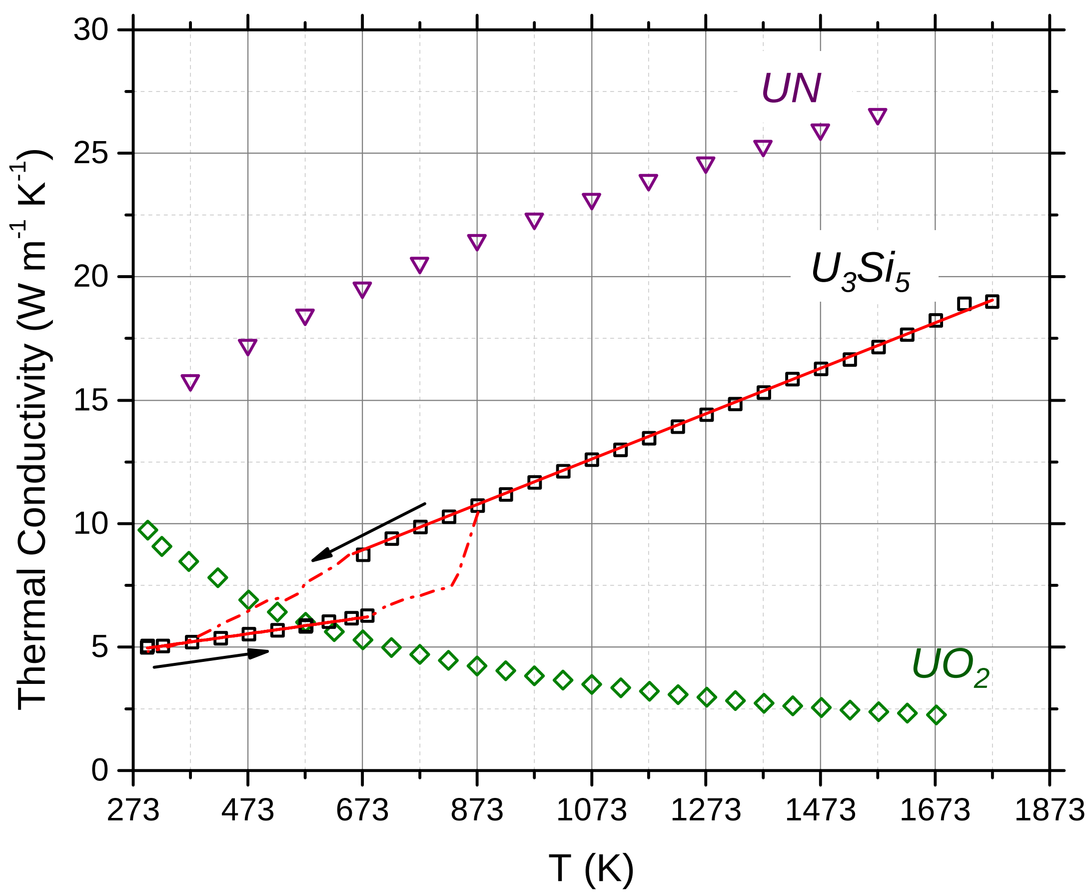
- **Industry (WEC), NEUP-IRP (Ga Tech), and core AFC program have identified U-Si compounds as potential monolithic or constituent phase for advanced LWR fuels**
  - Higher U-density (improve reactor performance or break-even to enable non-Zr cladding)
  - Improved thermal conductivity (initially hypothesized, quantified by AFC in FY13-14)
  - Hypothesized oxidation resistance
- **Historic use as research reactor fuel (vastly different requirements than LWR)**
- **Numerous uncertainties remain**
  - Irradiation performance must be demonstrated
  - Silicide fission product thermochem?
  - Industrial fabrication must be developed

Swelling of U_3Si and U_3Si_2

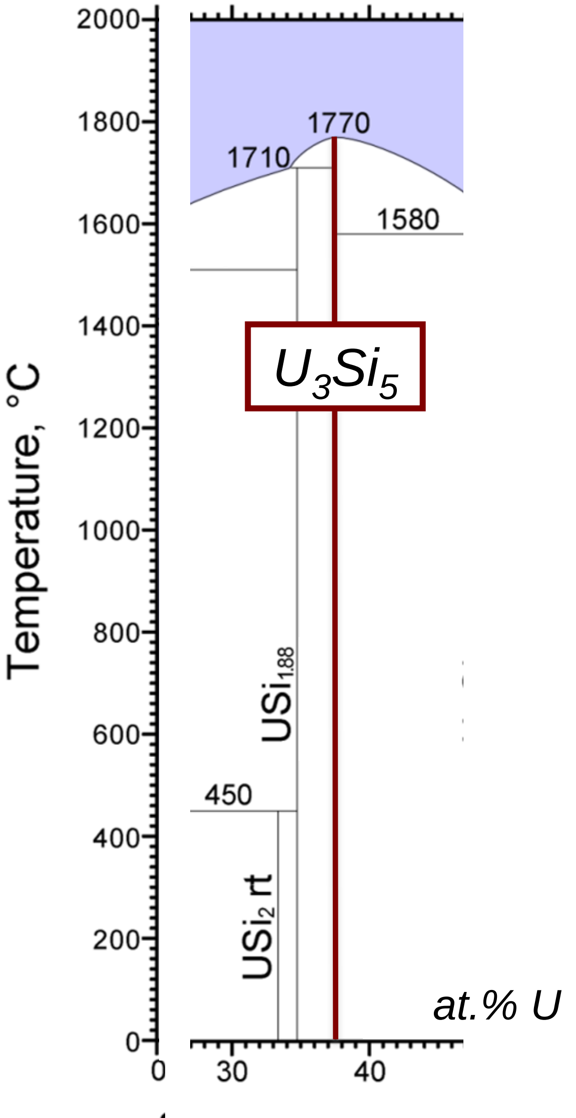


Kim, Comprehensive Nuclear Materials (3.14), 2012

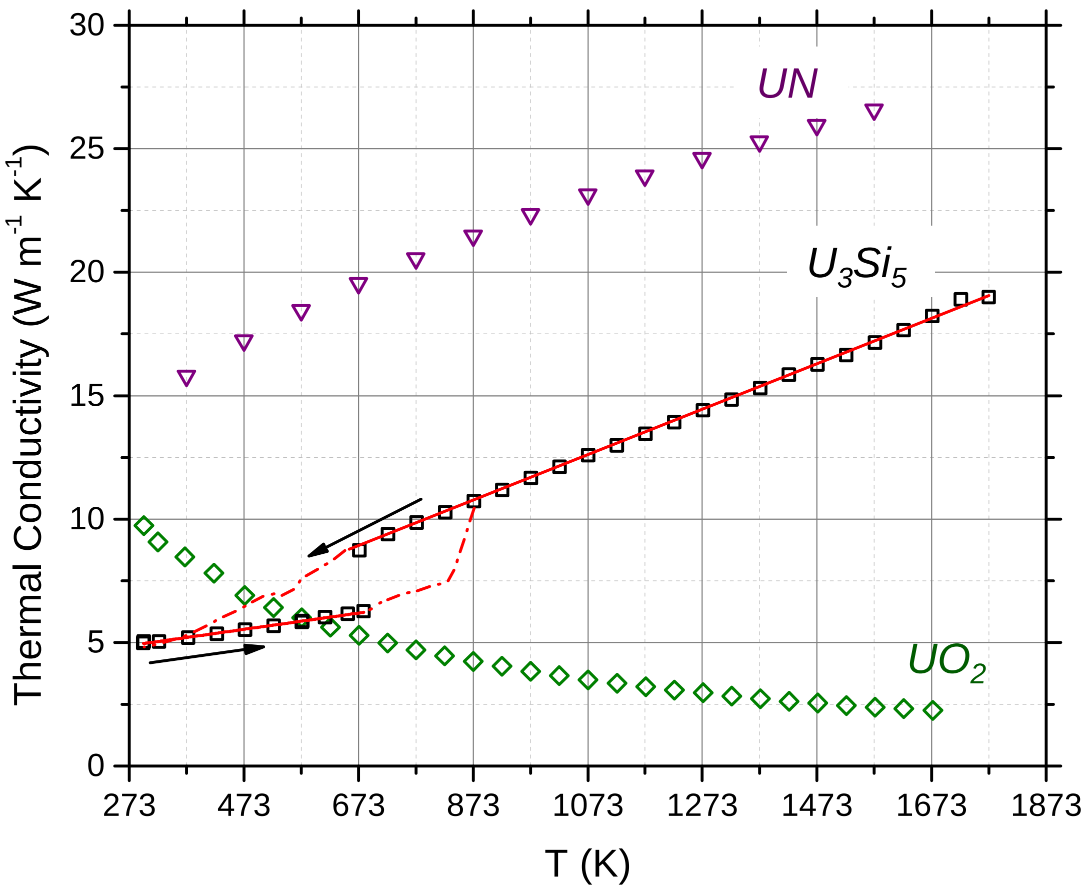
Thermal Conductivity of U_3Si_5: Phase Transformation and Effects



White et al., Journal of Nuclear Materials, Accepted (in press)



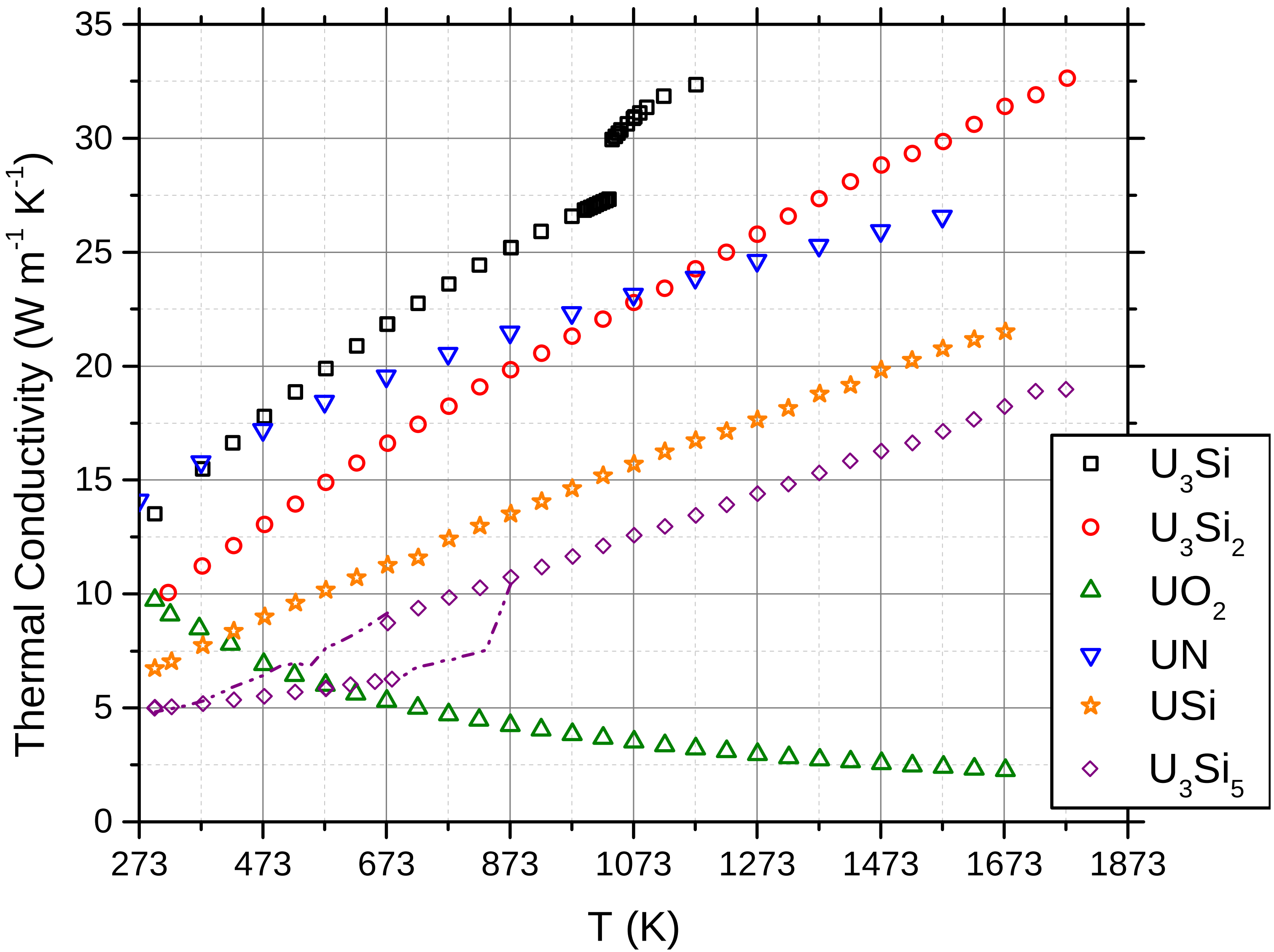
Thermal Conductivity of U_3Si_5: Phase Transformation and Effects



White et al., Journal of Nuclear Materials, Accepted (in press)

- **Kinetics and impact of low temperature (~450° C) phase transformation in U_3Si_5 must be better understood**
- **FY15 work in two areas will focus on this phenomena**
  - Fundamental high temperature XRD and neutron diffraction work to characterize structure and transformation
  - Microstructural characterization following varied thermal profiles

Compilation of Thermal Conductivity of Candidate Uranium Silicides

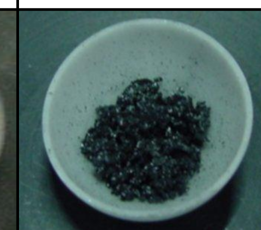
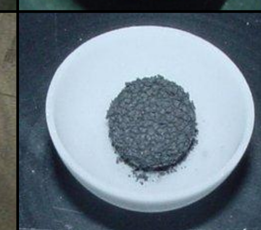


Consequences of AFC Thermophysical Property Study of U-Si System for ATF Development

- **Heat capacity follows theory**
  - Confirmed δ-δ' in U_3Si
  - Revealed inconsistency in U-Si phase diagram (USi_{2-x} region)
- **Thermal conductivity trends as expected (values f(T) critical for design work)**
- **CTE data is again important for design work as well as for composite fuel development**
  - UN, 1073 K: 8.6E-6 /K (Hayes *et al. J Nuc Mat* 171 1990)
  - U_3Si_2, 1073 K: 17.0E-6 /K
  - U_3Si_5, 1073 K: 11.2E-6 /K

Oxidation Performance of U-Si Compounds under Steam

- **Fundamental question of LWR ATF development: what happens when fuel is exposed to H_2O?**
- **Thermogravimetric analysis is being performed under steam-dominated atmospheres with two aims**
  - Understand pulverization threshold (when does monolithic pellet turn into a powder)
  - Characterize oxidation kinetics
- **Success in the second aim may provide pathways to development of even more robust ATF systems**

	4 hr STEAM TESTING		
	400° C	800° C	1000° C
U_3Si_2			
U_3Si_5			
UO_2			

Ø5 x 1 mm test pellets

Extension of Thermophysical Property Studies to Composite Systems

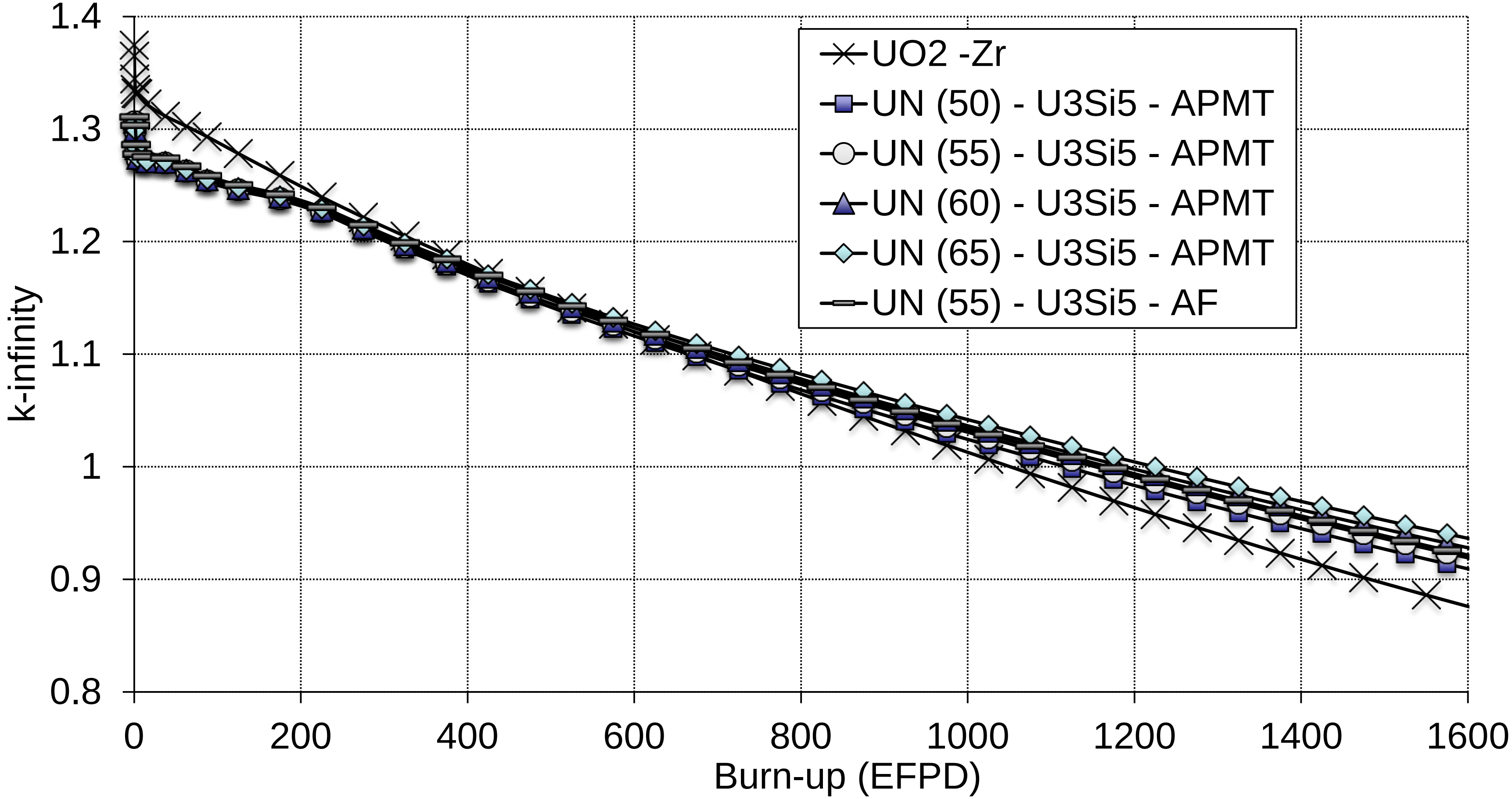
- **UN/U_XSi_Y composite fuels identified as potential ATF fuels (or enabling component)**
  - Improved U density at fixed enrichment (enable ATF cladding and/or augment reactor performance)
  - Improved thermal conductivity
  - UN/U_XSi_Y composite may ‘enable’ UN in LWR
  - BNL analyses have found UN-U_3Si_5 composites feasible ‘drop in’ fuels (N. Brown, BNL)
- **Development of UN-U_3Si_5 fabrication techniques in FY14 has facilitated thermophysical property measurements on high density composite materials**
- **Steam oxidation studies will follow in coming months**

Combination of UN and U-Si system may offer improvements

- **UN/U_3Si_5 and $UN/U_3Si_5/UB_2$ (4.9%-enriched) fuel clad with: Zircaloy, SS304, SS316, Kanthal AF, and Kanthal APMT**
- **Of the 50+ cases studied, these cases exhibited the “most similar” performance to UO_2-Zr in terms of 3-batch fuel management and reactivity coefficients:**

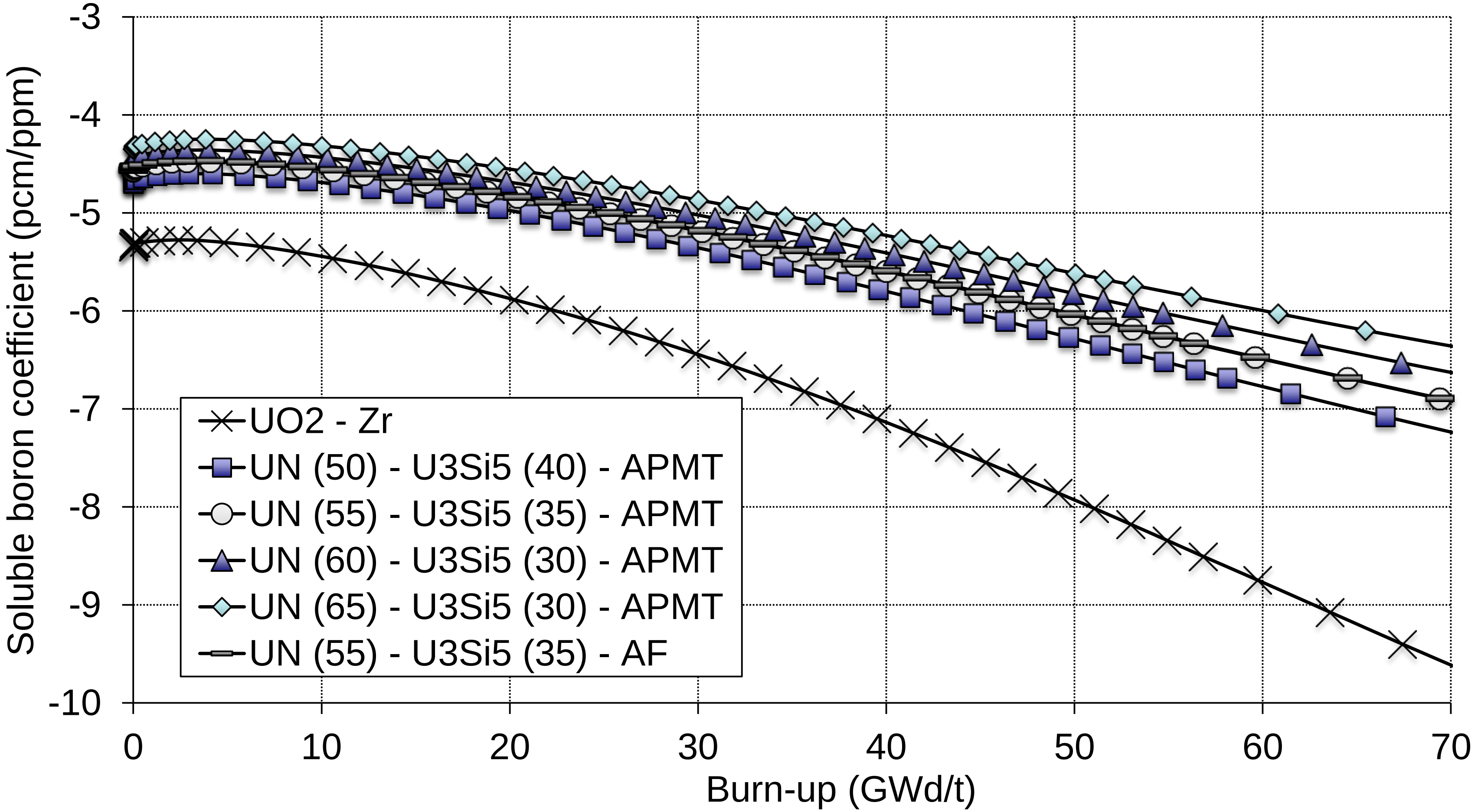
Fuel/Clad	Primary phase content	Secondary phase content	Tertiary phase content	Cycle length (EFPD)	Batch burn-up (GWd/t)	Discharge burn-up (GWd/t)
UO2 - Zr	**95.0%**	**0.0%**	**0%**	**532.9**	**20.5**	**61.6**
UN - U_3Si_5 - Zr	**50.0%**	**40.0%**	**0%**	**566.5**	**20.5**	**61.6**
UN - U_3Si_5 - UB_2 - Zr	60.0%	29.5%	0.5%	546.6	18.6	55.8
UN - U_3Si_5 - UB_2 - Zr	**55.0%**	**34.5%**	**0.5%**	**524.4**	**18.4**	**55.2**
UN - U_3Si_5 - AF	**55.0%**	**35.0%**	**0%**	**533.1**	**17.4**	**52.2**
UN - U_3Si_5 - APMT	65.0%	25.0%	0%	558.3	17.2	51.6
UN - U_3Si_5 - APMT	60.0%	30.0%	0%	542.4	17.2	51.6
UN - U_3Si_5 - APMT	**55.0%**	**35.0%**	**0%**	**526.0**	**17.2**	**51.5**
UN - U_3Si_5 - APMT	50.0%	40.0%	0%	509.2	17.2	51.5
UN - U_3Si_5 - UB_2 - APMT	75.0%	14.5%	0.5%	510.5	14.8	44.5

Example: UN/U_3Si_5 with Kanthal APMT or AF, Cycle Length



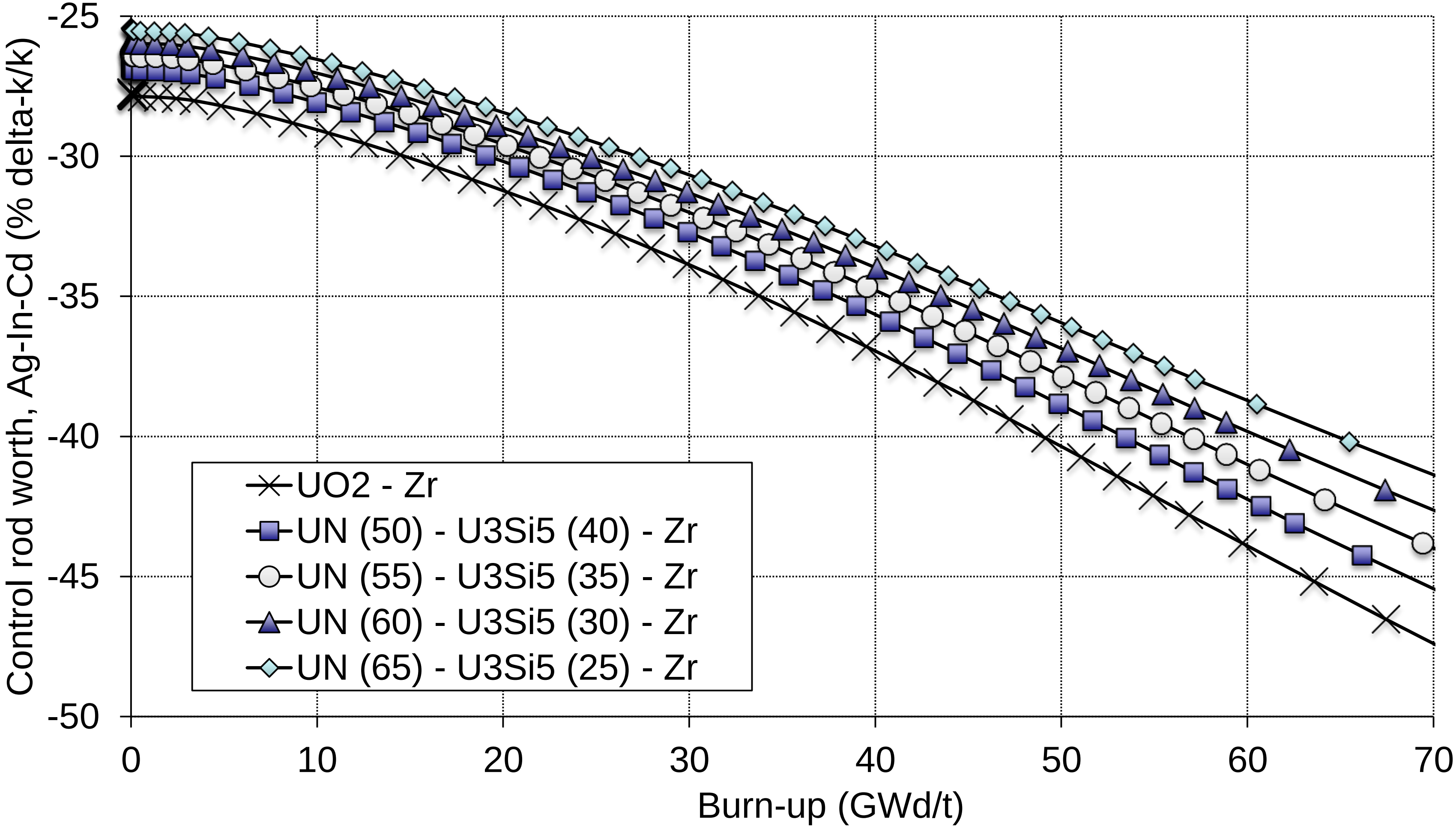
UN (55%)-U3Si5 (35%) – AF (533.1 EFPD) is most similar to UO_2 –Zr (95%) (532.9 EFPD)

Example: UN/U_3Si_5 with Kanthal APMT or AF, Soluble Boron Worth



Increased U_3Si_5 fraction softens spectrum, increases magnitude of soluble boron worth

Example: UN/U_3Si_5 with Zircaloy, Control Rod Worth



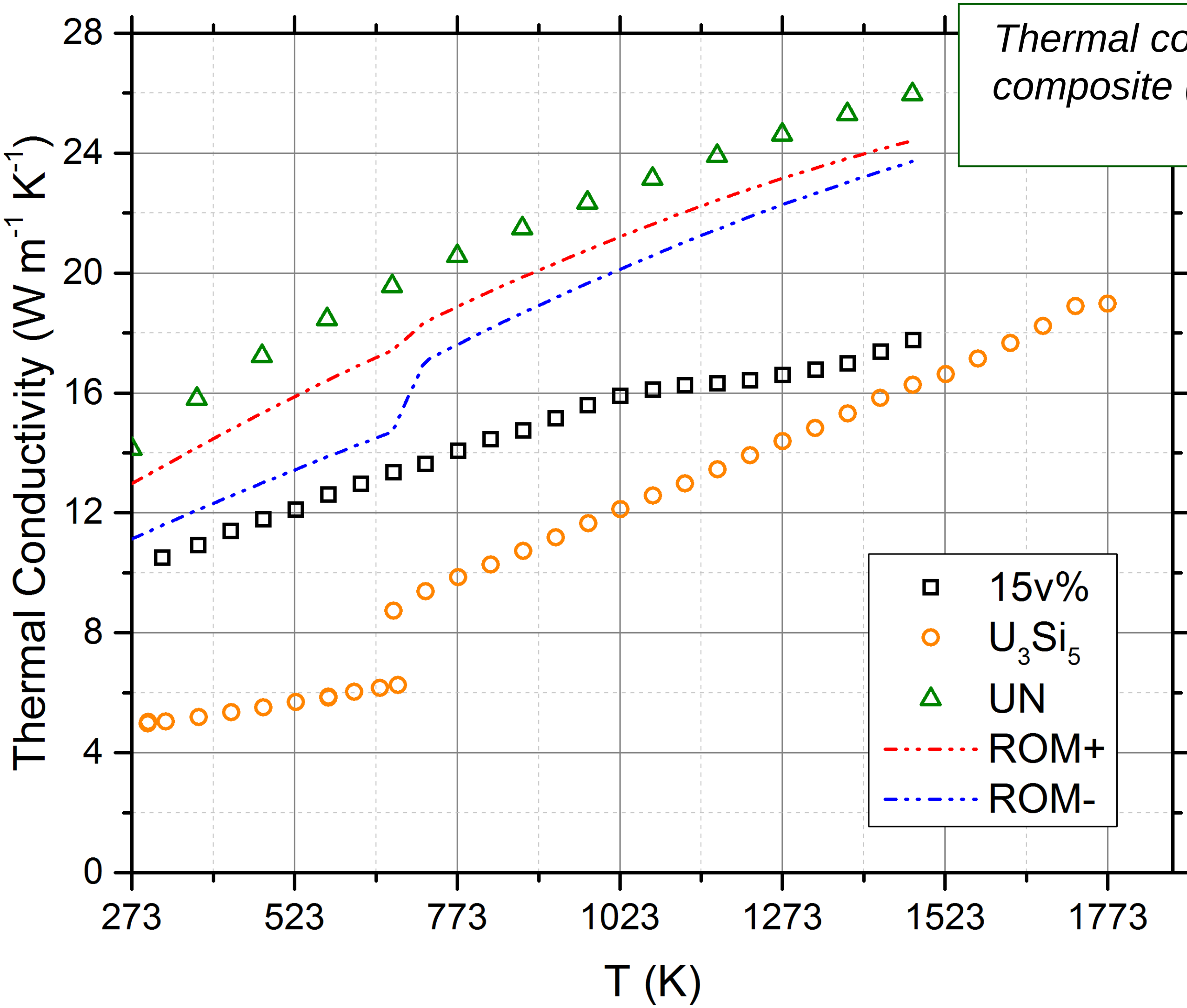
Increased U_3Si_5 fraction softens spectrum, control rod worth converges toward UO_2-Zr

Key Findings for Phase Fraction Study

- **Adjusting fractions of a secondary phase informs fuel designers about envelope for engineering a "drop-in" replacement for UO_2**
- **Higher heavy metal loading of UN-U_3Si_5 fuels can compensate for the significant reactivity penalty associated with Fe-based cladding options**
- **The reactor physics calculations in this study assume 100% ^{15}N, but natural nitrogen would yield less desirable performance results due to the (n,p) reaction in ^{14}N and associated reactivity penalty**
- **These are preliminary results from an ongoing study**
- **These scoping calculations assume a constant enrichment and assembly power**
- **"Most similar" to UO_2 is not necessarily always best**
  - E.g. higher density fuels with Zr-based cladding could enable four-batch operation with the same cycle length

Thermal Conductivity of UN-U_3Si_5 Composites

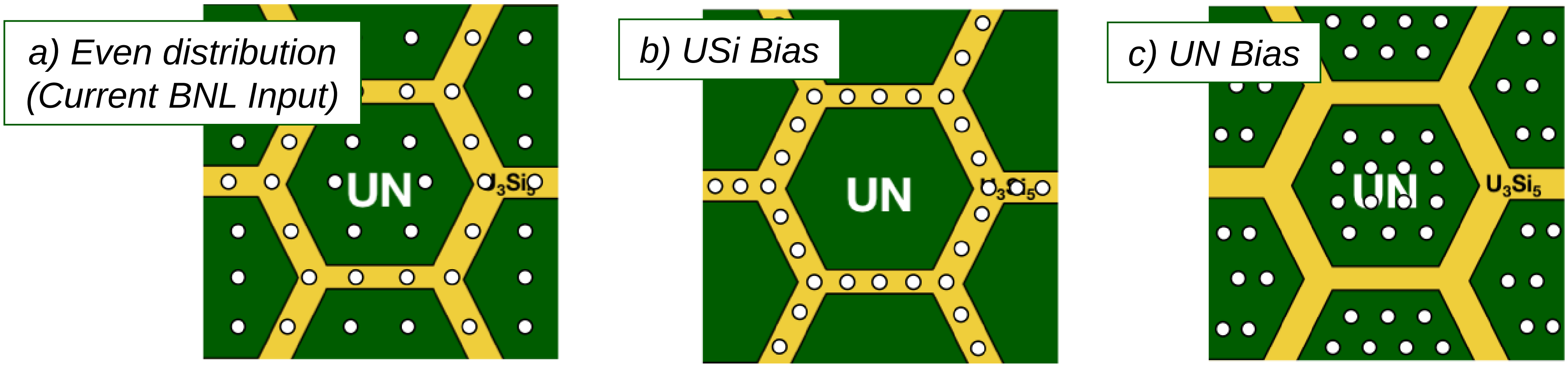
Thermal conductivity of UN-15%U_3Si_5 composite (94% TD) in comparison to UN and U_3Si_5



- Performance measured to date does not indicate significant flaws
- Microstructural characterization is ongoing to elucidate factors dictating structure-property relations

Considerations in Structure-Property Relations for Composites

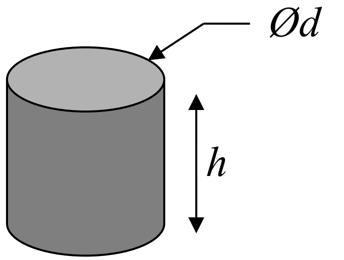
Representations of UN/U_3Si_5 Composite (15 vol% U_3Si_5)



Each graphic above contains ~5% porosity… but this can be distributed in multiple ways

- **Porosity is distributed uniformly in both phases (or displaces both phases without bias)**
  - Current assumption in BNL neutronics calculations
  - Unlikely in a real system
- **Porosity is localized in either U_3Si_5 (b) or UN (c)**
  - May introduce uncertainty in U loading
  - How much biasing is possible for a given bulk geometric density?

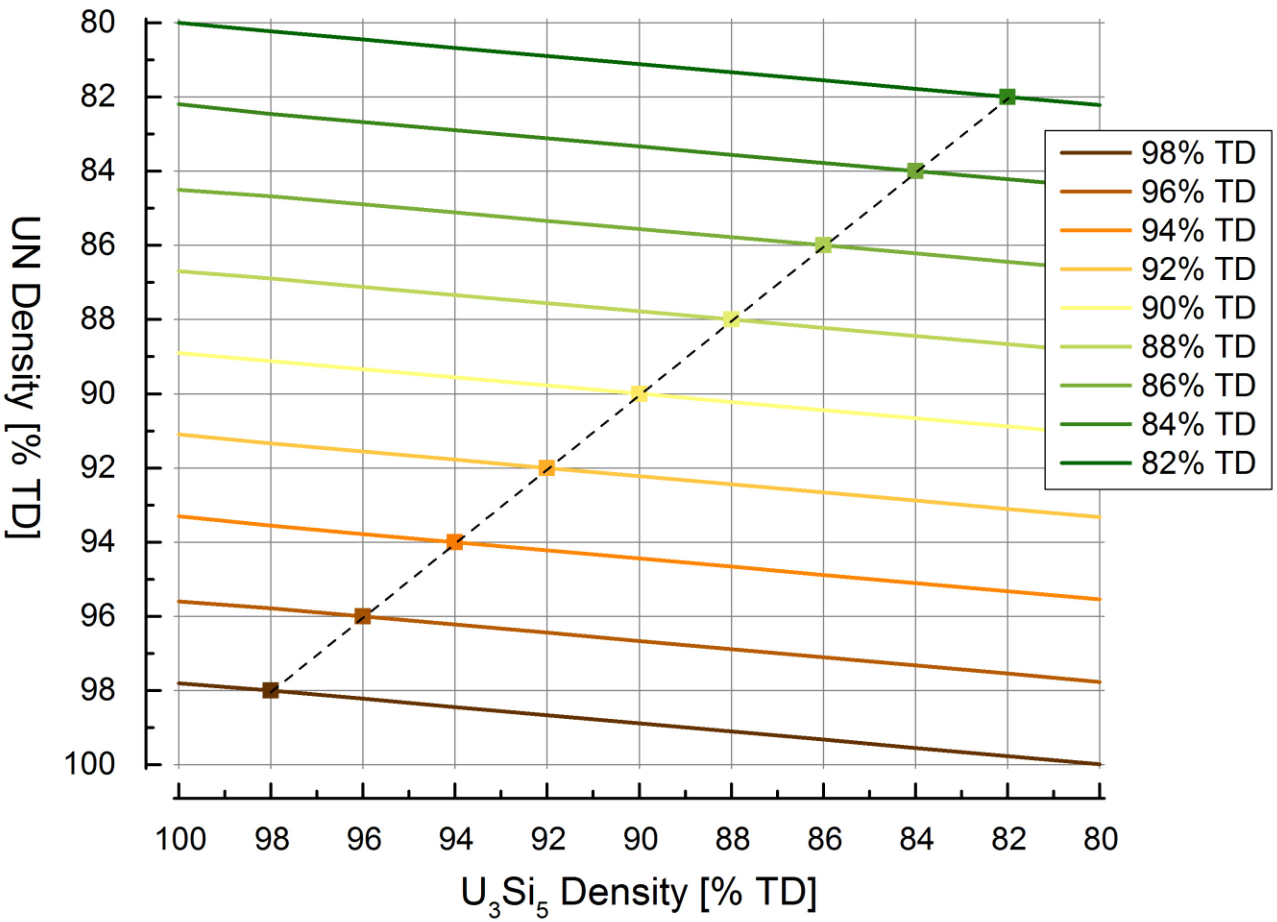
Considerations in Structure-Property Relations for Composites



ρ_{UN} = 14.33 g/cm^3 (13.54 U)
ρ_{U3Si5} = 8.96 g/cm^3 (7.48 U)

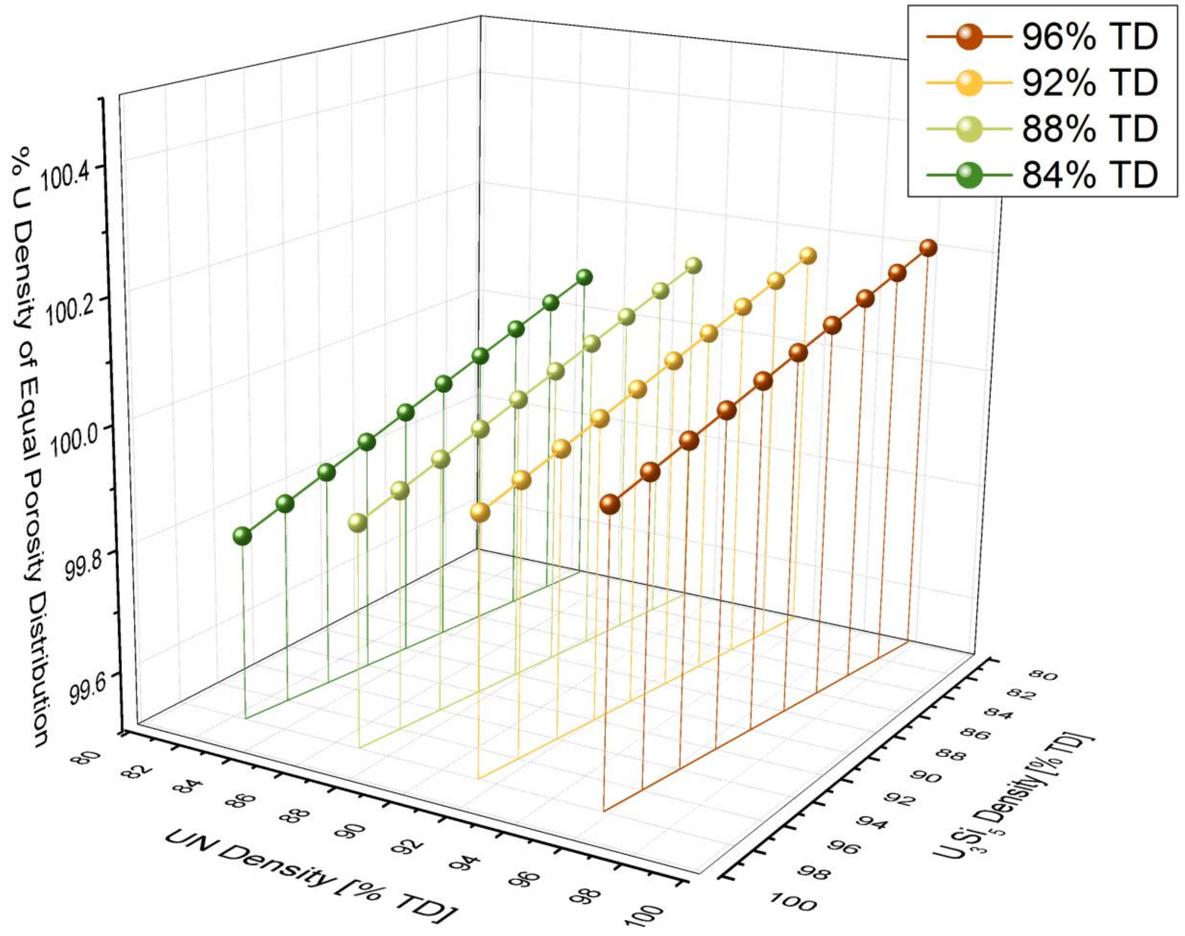
$\rho_{15\%USiCOMP}$= 13.53 g/cm^3

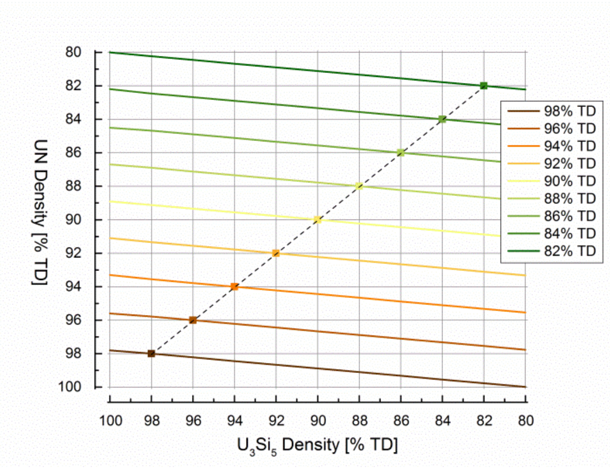
- **Measured bulk density does not explicitly define composition of system (two degrees of freedom)**
- **Possible range of compositions for a given bulk density are shown at right**



Effect of Porosity Bias on U Density of Composites

- Biasing will increase or decrease U smear density of composite
- Magnitude shown at right will not change qualitative BNL conclusions…
- … Effect on transport or oxidation properties could be significant





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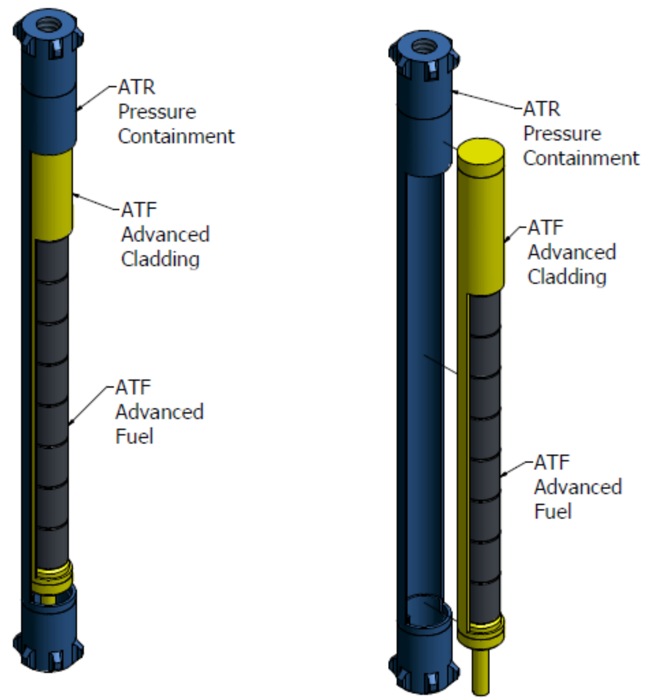
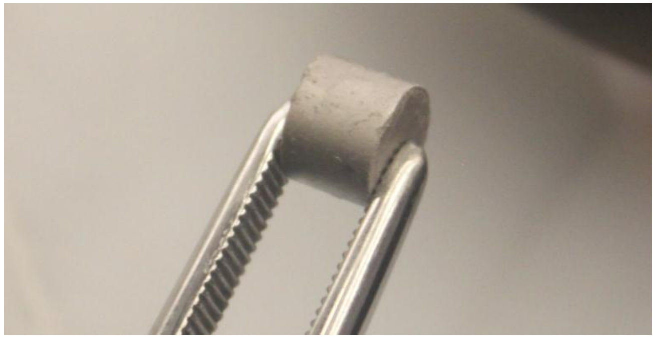
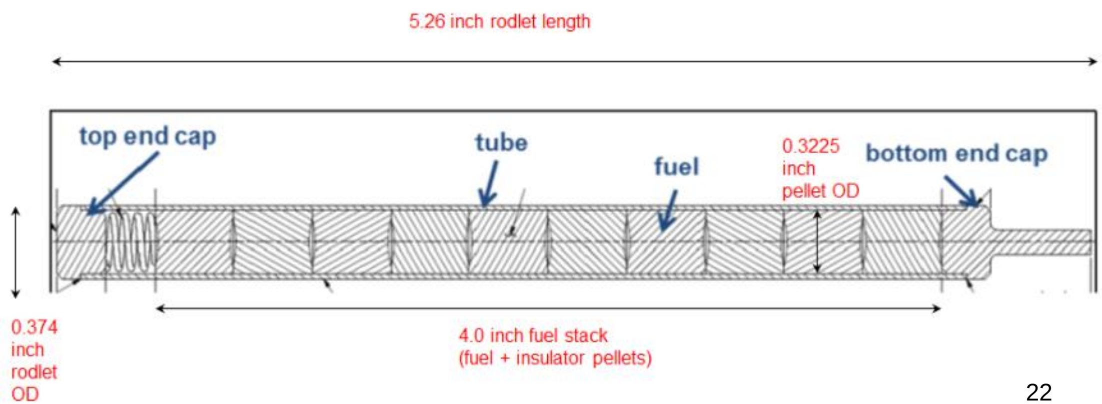
ATF-1W Irradiation

- **U_3Si_2 pellets have been fabricated at INL for the ATF-1 Irradiation as part of the collaboration with Westinghouse**
- **6 rodlets with 5.5% enriched U_3Si_2 fuel pellets with Zirlo cladding at 3 different fuel centerline temperatures:**
  - ~480° C
  - ~800° C
  - ~1200° C
- **Under typical PWR conditions**
  - 180-500 W/cm LHGR
  - 300-400° C cladding temperature
  - 20, 40, 70 GWd/mTU burnup



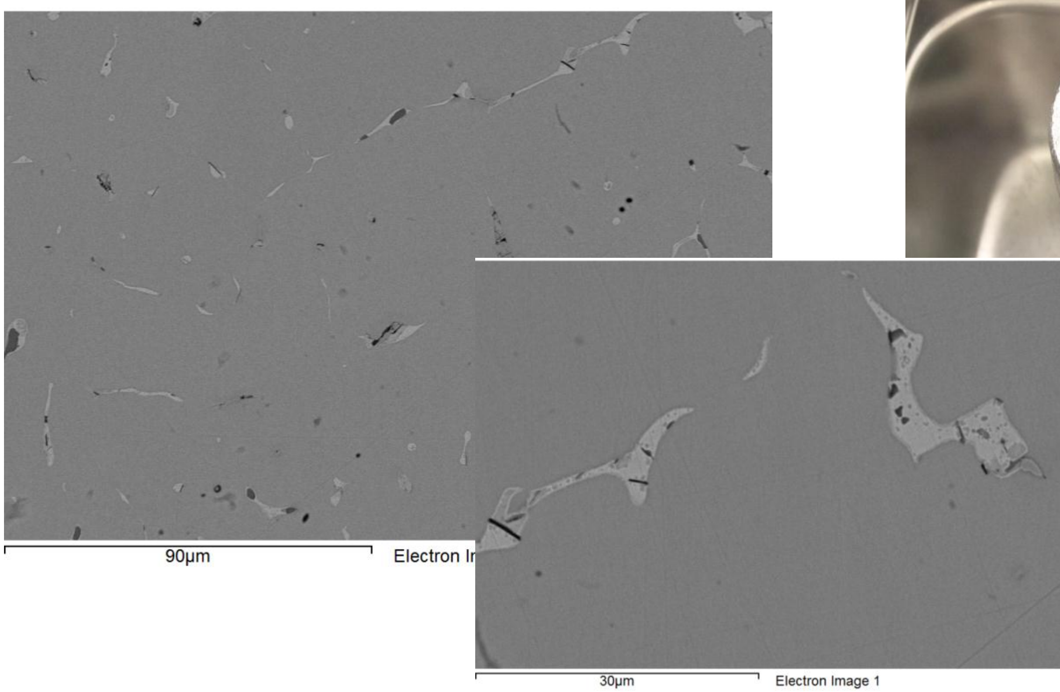


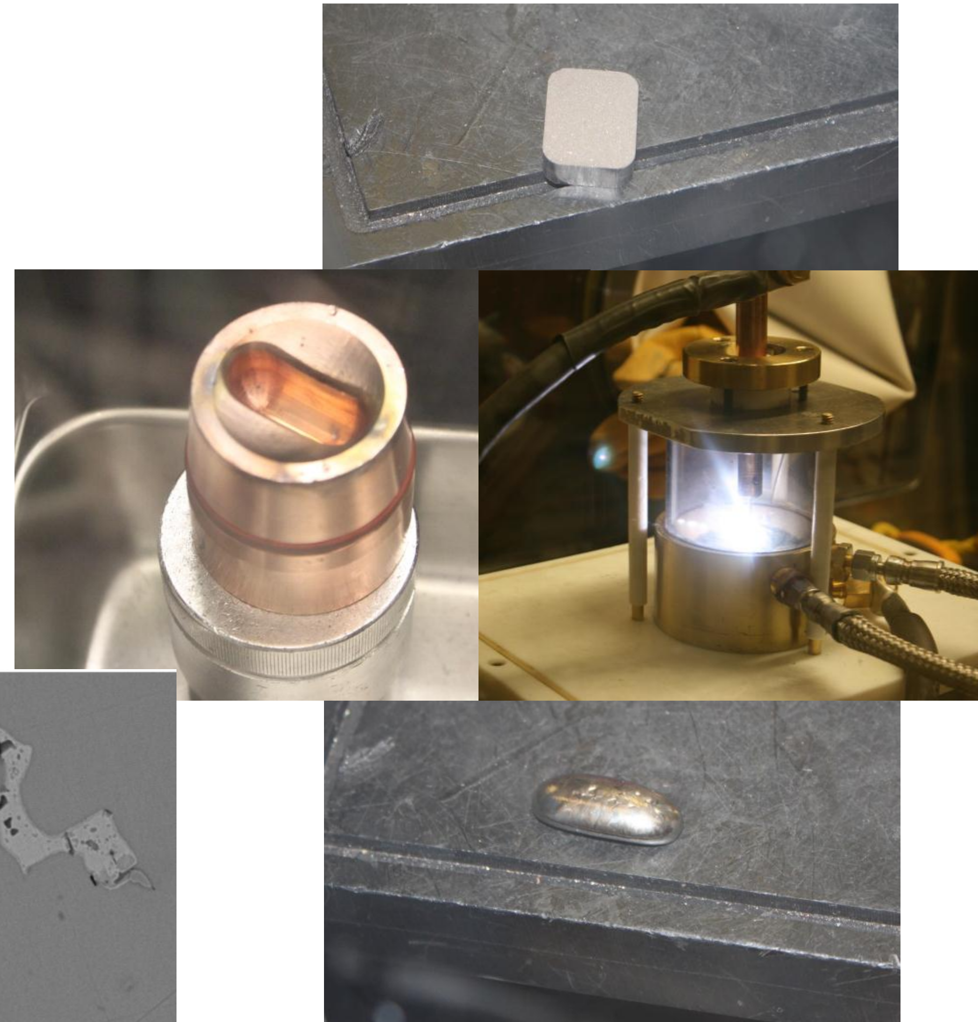
ATF Rodlet Sketch



U_3Si_2 Fabrication – Arc Melting for Westinghouse ATF-1 test

- **Compacting the U and Si**
  - 92.5 wt. % U 7.5 wt. % Si
- **Arc melting**
- **Confirmation of phase by XRD and SEM analysis (~0.5 to 3% U_3Si)**





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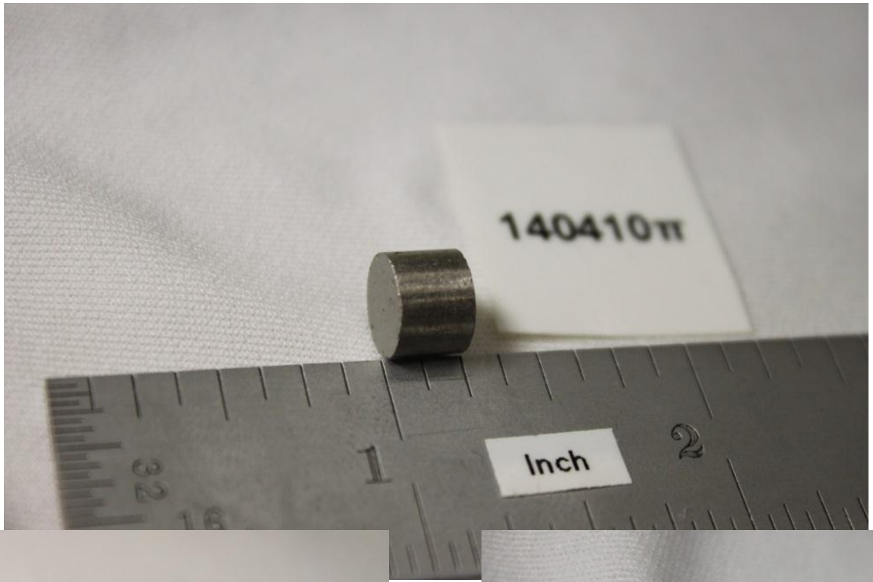
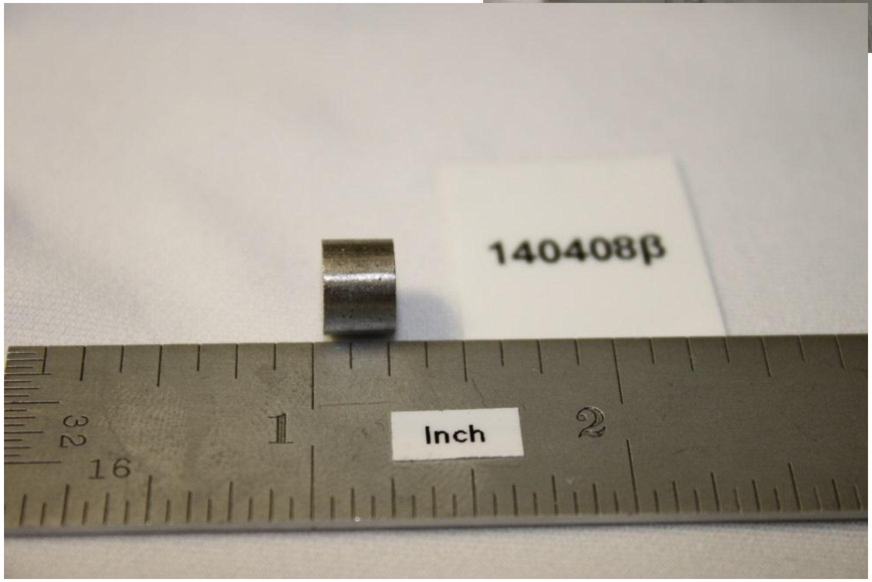
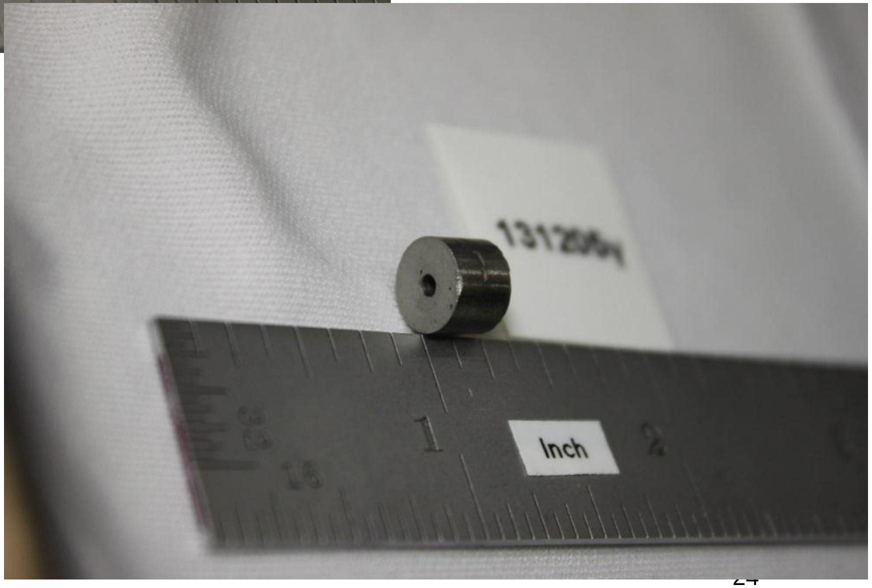
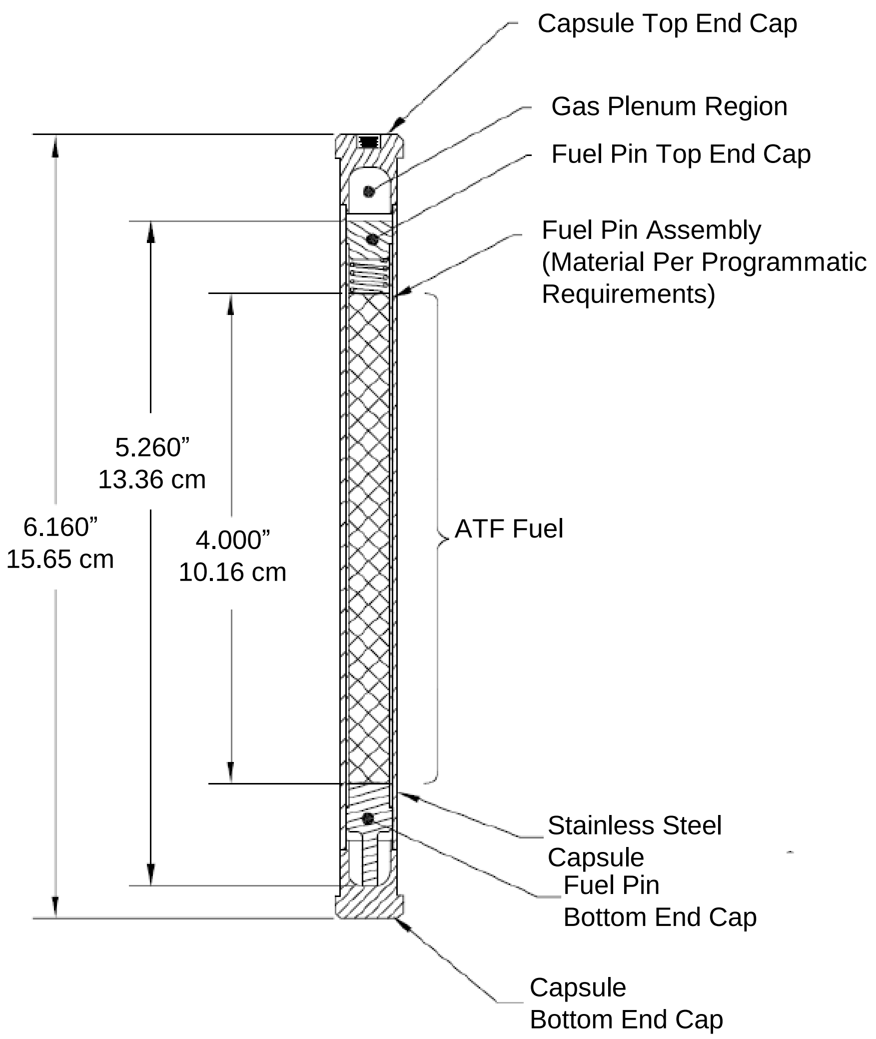
Machined Pellets for ATF-1W



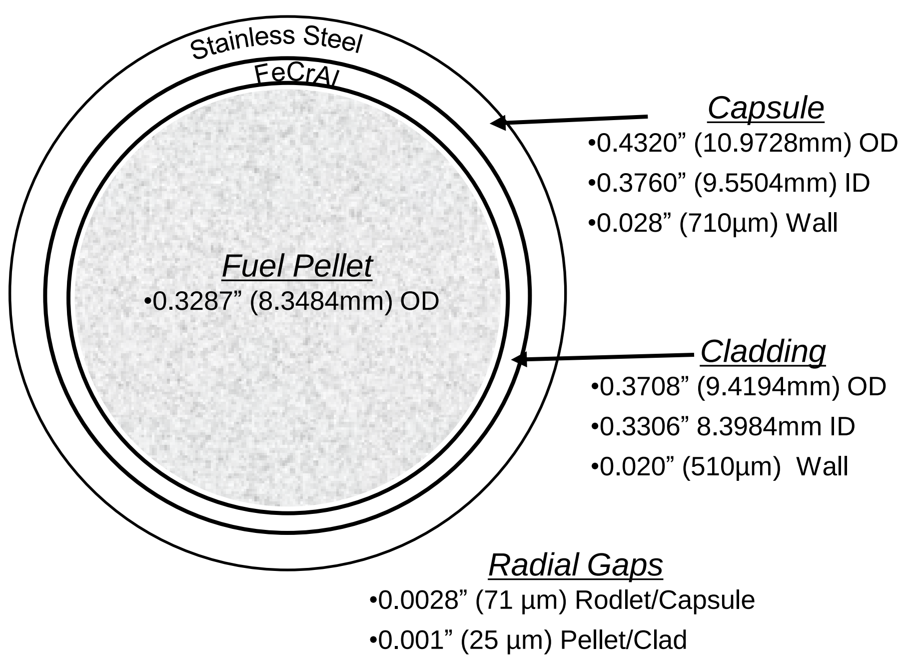




Rodlet and assembly configuration



ATF-1 Test Capsule (LANL-1)



Conclusion

- **High density fuels may help enable ATF technologies (cladding) by:**
  - Increasing fuel density offsetting neutron penalty
  - Improving thermal properties
- **Technology still at the research and development level**
- **Oxidation and performance must be proven**

Overview of Powder Metallurgy Fabrication Route used for U-Si

SYNTHESIS OF PHASE OF INTEREST VIA ARC MELTING (SMALL EXCESS OF Si USED)

HEAT TREATMENT AT ~80% T_{MELT} FOR 24-72 HOURS

MECHANICAL MILLING TO PRODUCE FEEDSTOCK

COLD PRESSING WITHOUT BINDER OR LUBRICANTS

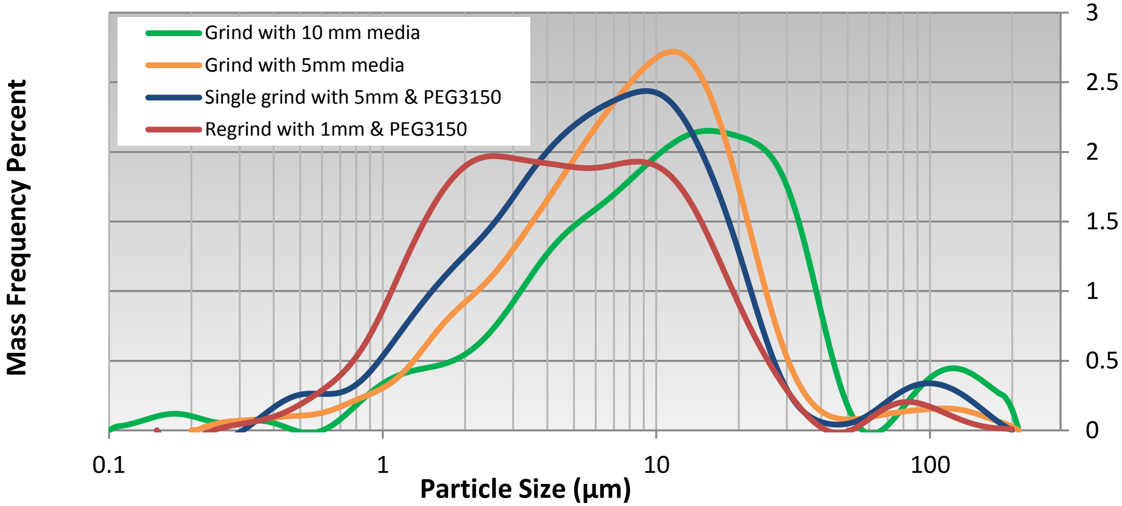
SINTERING AT >80% T_{MELT} for 24-48 HOURS

Material	Fixturing	Temp [C]	Time [h]	Density [%TD]
U_3Si_2	Y_2O_{3-X}	1385	12	96+
U_3Si_5	W	1450	12	97+

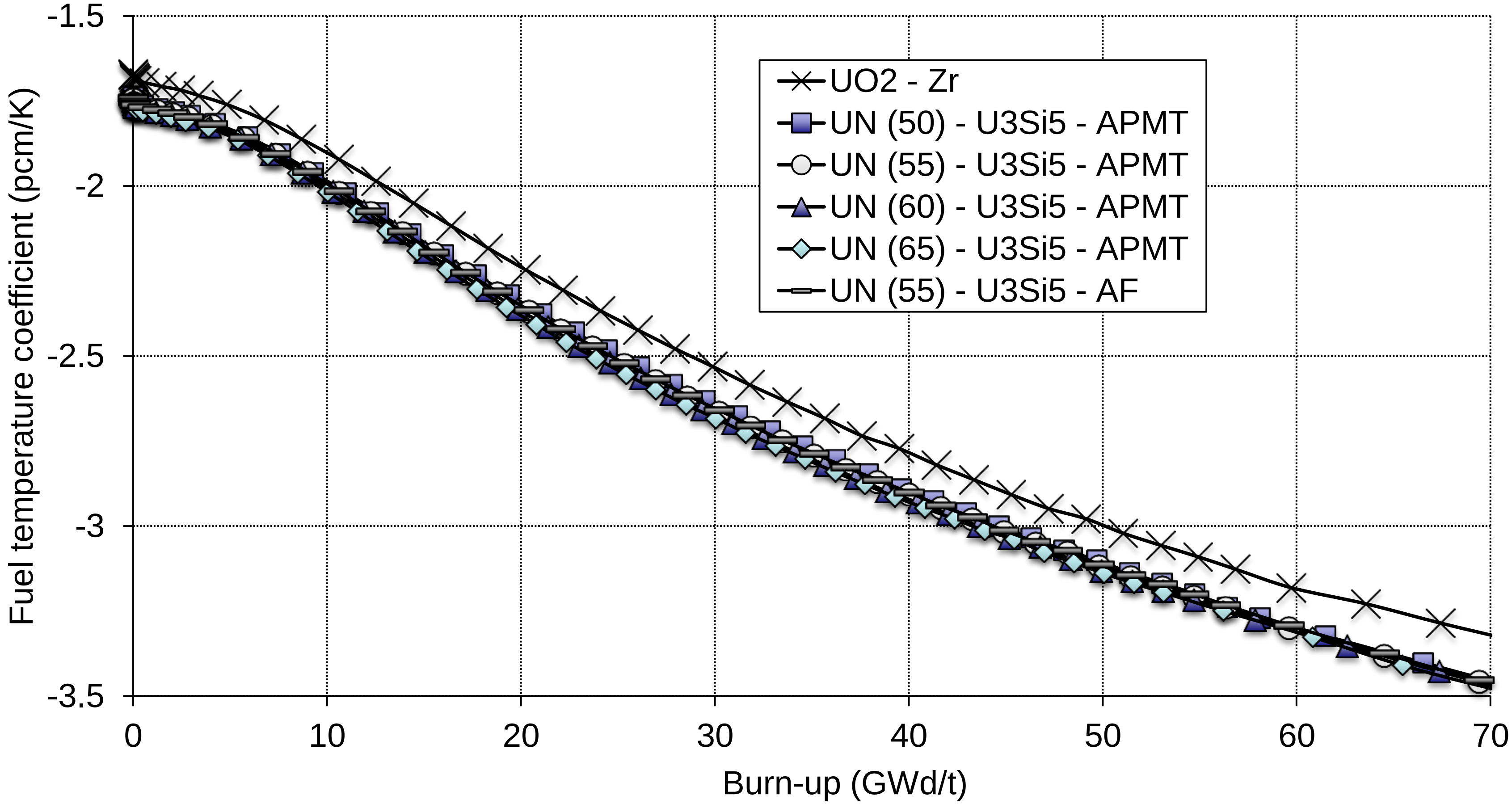
Samples of U_3Si_5 prepared via powder metallurgy route for property analysis

Fabrication – Comminution

- Planetary Ball Milling quickly comminutes arc melted ingots to a fine powder
- The final sintered pellet densities that can be reliably achieved are highly dependent on the particle size distribution in the final powder
- Significant iteration between milling, pressing and sintering was needed to find the final powder product
- Particle size analysis has been performed by sedimentation theory sub-sieve particle analysis
- The best results were achieved by adding a milling lubricant (PEG 3150) and running a two step milling process

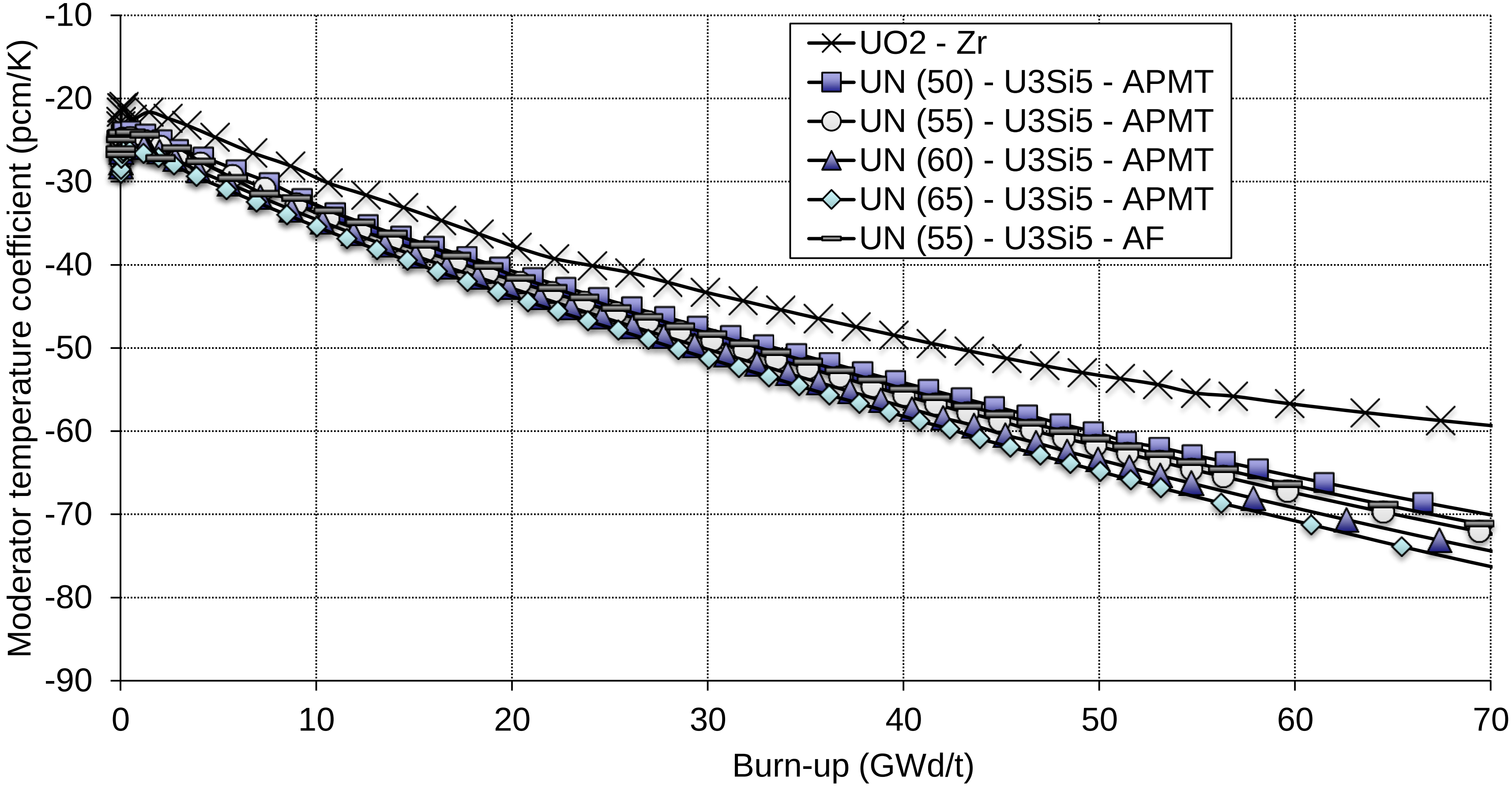


Example: UN/U_3Si_5 with Kanthal APMT or AF, Fuel Temperature Coefficient (Doppler)



FTC (Doppler) is similar for all options and is within design limit of reference LWR

Example: UN/U_3Si_5 with Kanthal APMT or AF, Moderator Temperature Coefficient



Hardened spectrum increases magnitude of MTC, but all options are within design limit of reference LWR