Fabrication of Planar PyC/SiC Diffusion Couples to Study Radiation Enhanced Diffusion in Representative TRISO-Fuel SiC

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ADVANCED GAS REACTOR TRISO FUELS PROGRAM REVIEW

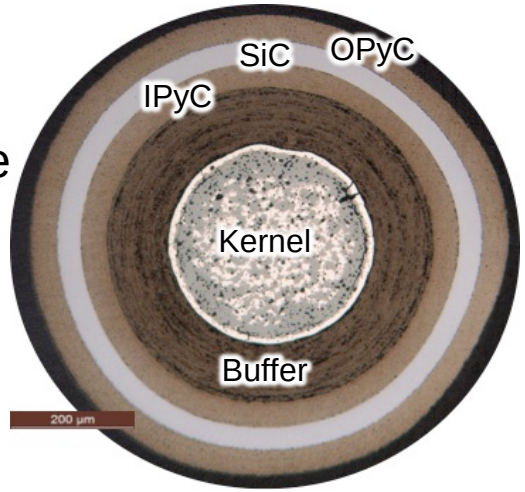
JULY 18-19, 2017

CENTER FOR ADVANCED ENERGY STUDIES (CAES)

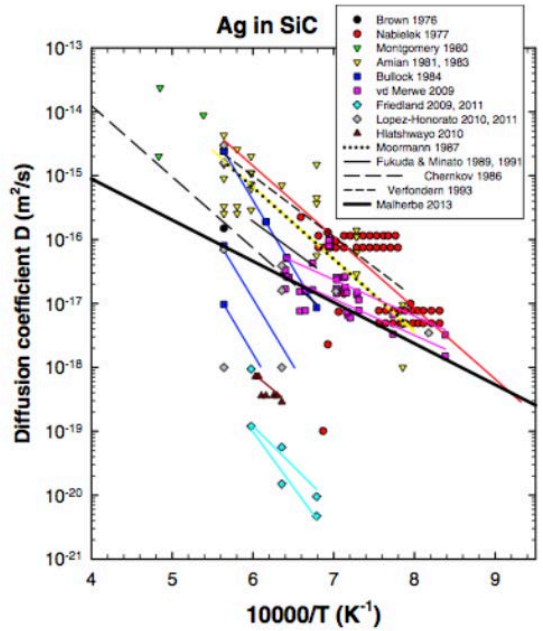
Idaho Falls, ID

Project Motivation

As-fabricated TRISO particle



- Determine **accurate** diffusion kinetics for fission products in **SiC representative of the tristructural-isotropic (TRISO) coated particle fuel system**.
  - Gain a more complete understanding of release of fission products (FP) from intact TRISO fuel
    - Influence of SiC microstructure, PyC/SiC interface (characteristics that can be tailored) and radiation enhanced diffusion
  - Provide data to performance models for safe and efficient high temperature gas-cooled reactor operation

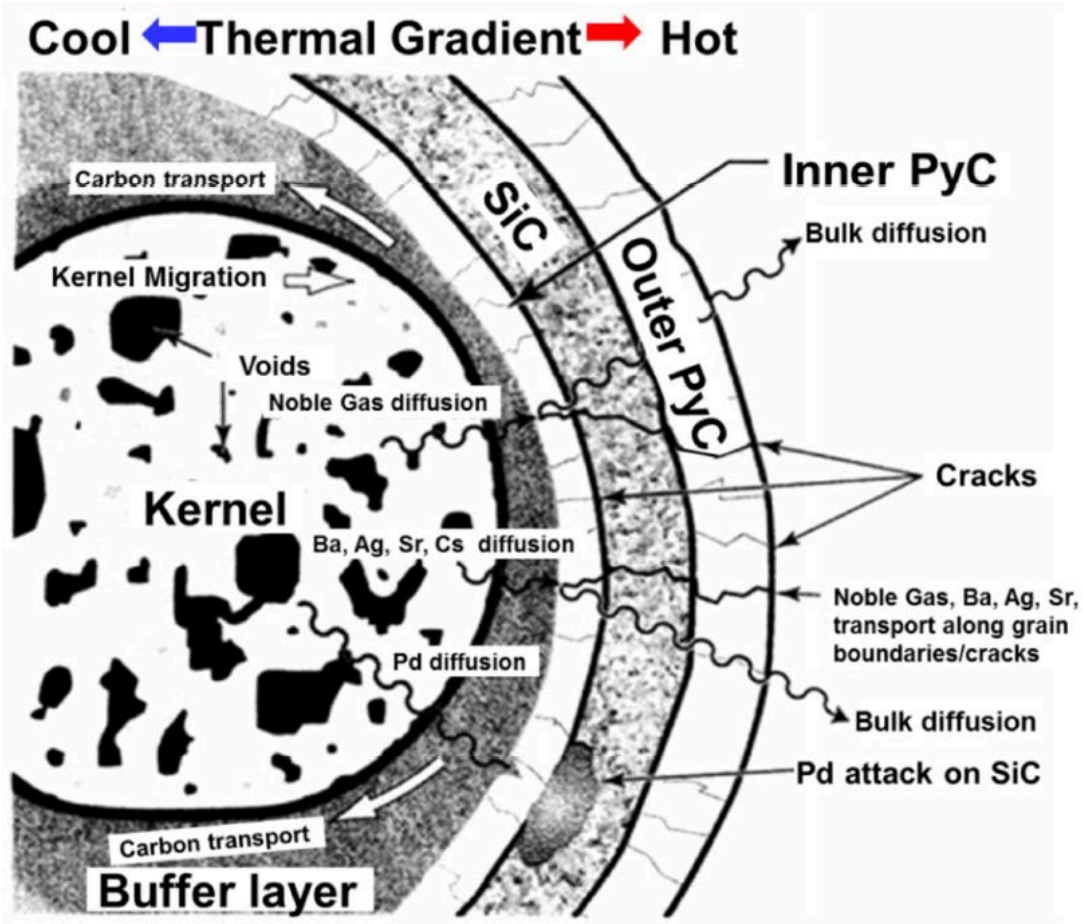


Reported diffusion coefficients for Ag/SiC reproduced from [1]

[1] J.B. Malherbe, J. Phys. D. Appl. Phys. 46 (2013) 473001.

Challenges: Difficulty in real fuel analysis

- Release from intact fuel is known but the kinetics and influence of the layer properties on release is not well understood.
- Irradiated TRISO fuel systems are not ideal for determining kinetics properties.
  - FP evolution is complex: production, stabilization and transport
  - Fuel experiences variable thermal histories
  - Non-ideal geometries for analysis
- These challenges yield a wide range of reported diffusion kinetics.

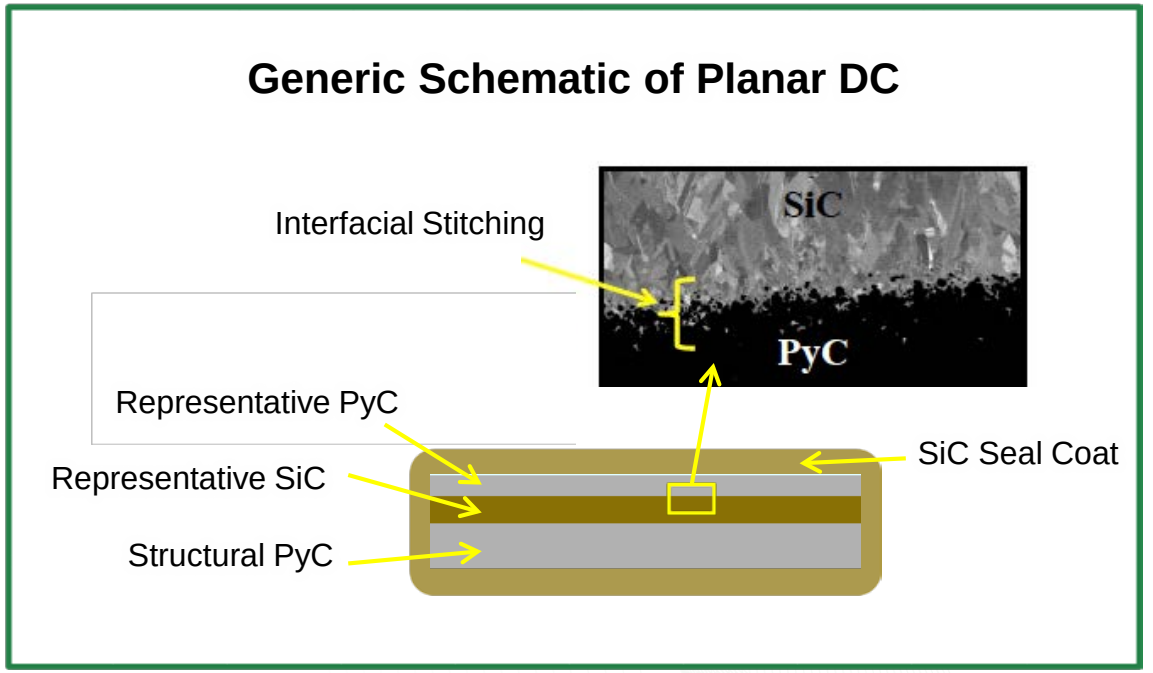


Processes in high burnup TRISO fuel influencing lifetime, reproduced from [2]

[2] B.G. Kim, *et. al.,* Nucl. Eng. Technol. 47 (2015) 608-616.

Focus of planar diffusion couple experiment: explore layer variables and radiation enhanced diffusion

- Leverage Advanced Gas reactor (AGR) Program TRISO fluidized-bed coating capabilities to build planar TRISO-layer diffusion couples (DC) with controlled ”FP” inventory and known thermal history.
  - Baseline: Layers identical to AGR-2 TRISO
  - IPyC Variant: Low density IPyC (relative to AGR-2)
  - SiC Variant: Large grained SiC (relative to AGR-2)
    - Design is influenced by previous work by Dwaraknath and Was 2016 [3].
- Utilize HFIR to conduct comparative study between neutron irradiated DCs and thermally exposed DCs – *Radiation Enhanced Diffusion?*
- Investigate transport in SiC via depth profiling (Rutherford Backscattering Spectrometry) and cross-sectional electron microscopy.



First step in the process is to develop planar diffusion couple

[3] S.S. Dwaraknath and G.S. Was, J. Nucl. Mater. 474 (2016) 76-87.

Diffusion couple test matrix

Description	Condition	Samples*	Exposure Times
Neutron Irradiation	0.5 dpa, 1100±50°C	Baseline: *Ag, Ag+Pd, Eu*, Sr SiC Variant: *Ag*, Eu, Sr RH-SiC: Ag	~168 hours
	1.0 dpa, 1100±50°C		~336 hours
Thermal Equivalent	Temp. and time equivalent to 0.5 dpa capsule		~168 hours
	Temp. and time equivalent to 1.0 dpa capsule		~336 hours
High Temperature Thermal Diffusion	1500°C	Baseline: Ag, Ag+Pd, Eu, Sr IPyC Variant: Ag, Ag+Pd SiC Variant: Ag, Ag+Pd, Eu, Sr	300, 600 hours
	1600°C		
	1700°C		

*IPyC variant will be included if space is available, ^ italics denote priority neutron irradiated samples

- Isolate influence of radiation enhanced fission product diffusion
- Determine diffusion kinetics at elevated temperatures to compare to safety testing analysis
- Understand the influence of layer properties on diffusional transport and Ag+Pd cooperative effects
  - Aggressive test matrix, not all comparisons will be made for irradiated samples with non-analyzed samples going to NEUP sample archive

Flow chart for fabricating planar diffusion couples

Determine Substrate Geometry

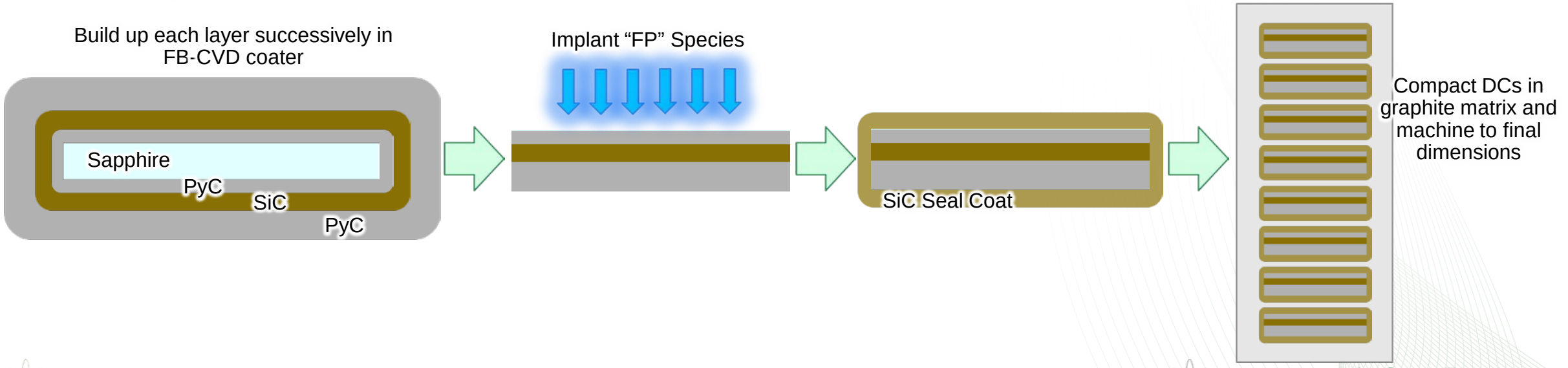
Determine Layer Parameters and Properties

Introduce “FPs” to DC

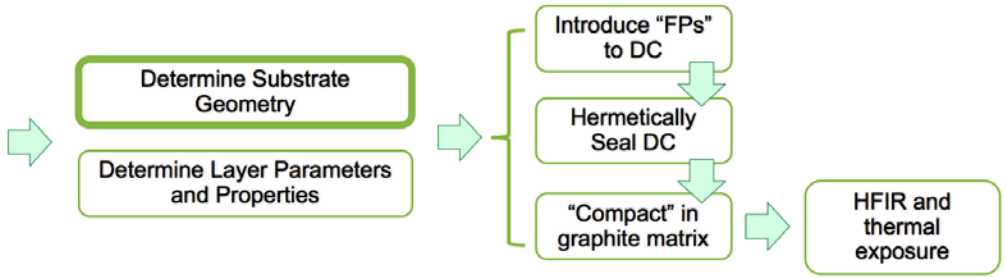
Hermetically Seal DC

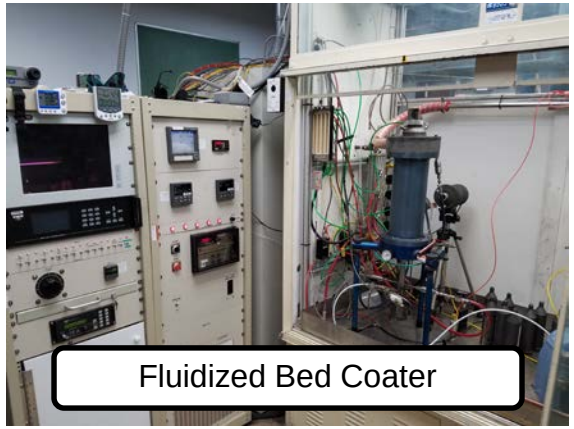
“Compact” in graphite matrix

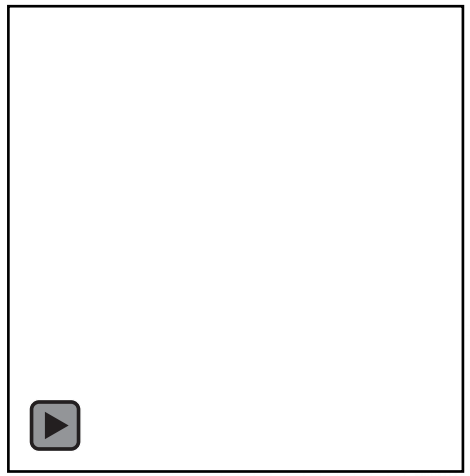
HFIR and thermal exposure

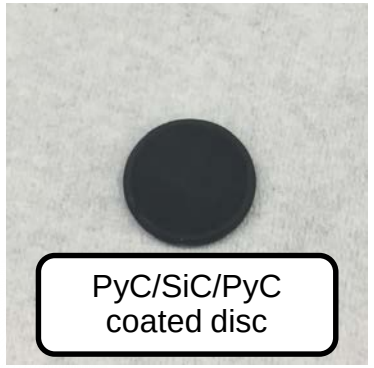


Determining substrate geometry



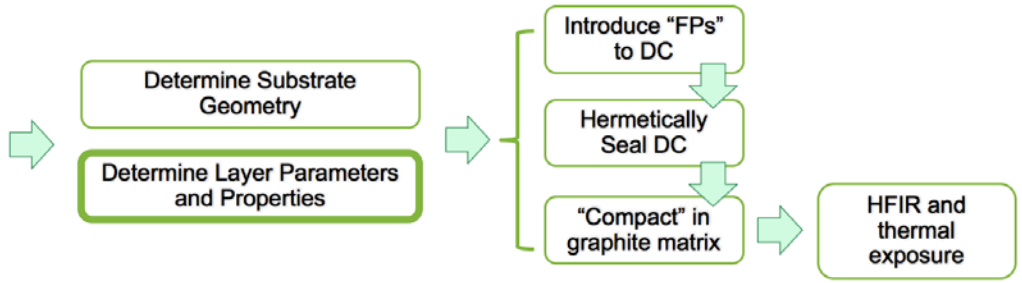


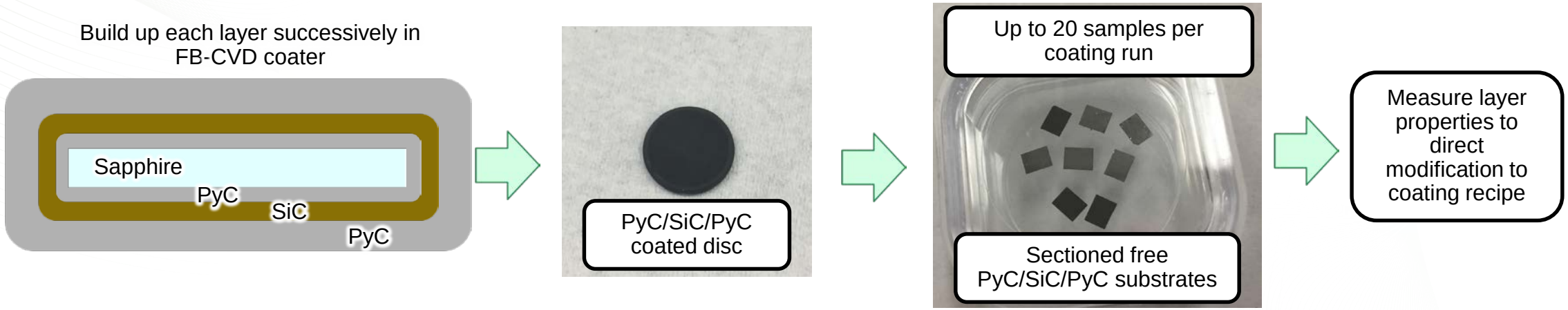




- Fluidized bed coater is identical to the system used to produce AGR-1 fuel (dedicated to surrogate fuel)
- Select sapphire disc diameter (~3-9 mm diameter) and thickness (0.5-1 mm) to build up PyC/SiC/PyC layers
  - Deposition and fluidization behavior were unknown
  - Used ~102 g of 400-µm-diameter ZrO_2 kernels with 12-µm-thick IPyC layer as a fluidization medium
- All diameters of sapphire discs fluidized appropriately, selected ~9 mm diameter disc due to ability to obtain up to 4 samples from each disc

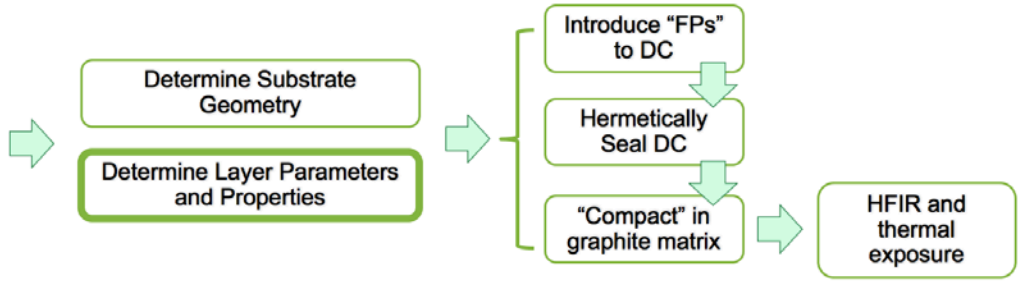
Determining Layer Parameters and Properties



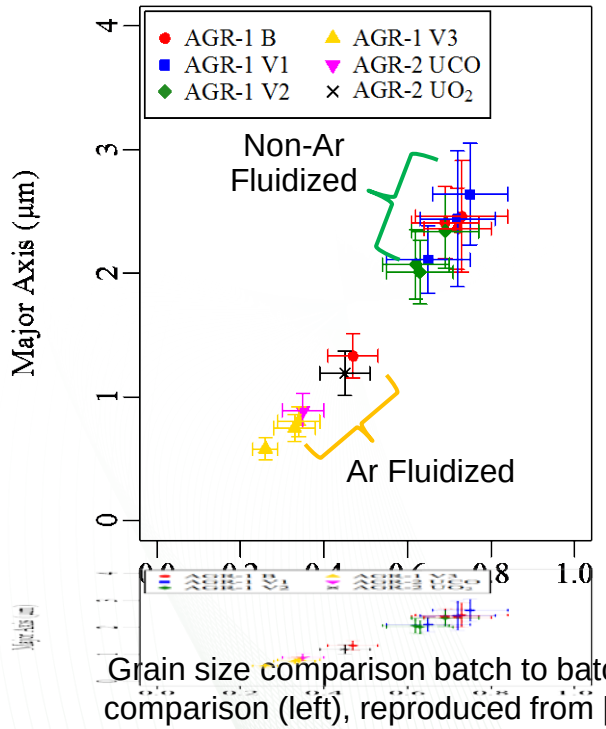


- Identify optimal conditions for achieving AGR-2 layer properties on planar geometry
  - Follow modified AGR-Surrogate-Coat-SOP-01
- Add "thick" secondary PyC support layer to improve handling and add robustness
- Check layer properties to compare to AGR-2: SiC microstructure, density, anisotropy
  - PyC density measured by interrupted coating runs

Targeted properties for each DC variant



		IPyC Density [1] (g/cm³)	IPyC Anisotropy [1] (BAF)	SiC Density [1] (g/cm³)	SiC Grain Size [2] (major axis, μm)	SiC Grain Size [2] (minor axis, μm)
	AGR-2 TRISO	1.890±0.011	1.0349±0.0012	3.197±0.004	0.89±0.14	0.35±0.05
Targets	Baseline	1.90±0.05	≤1.045	≥3.19	0.89±0.14	0.35±0.05
	PyC Variant	<1.85	≤1.045	≥3.19	0.89±0.14	0.35±0.05
	SiC Variant	1.90±0.05	≤1.045	≥3.19	2.39±0.24	0.71±0.05

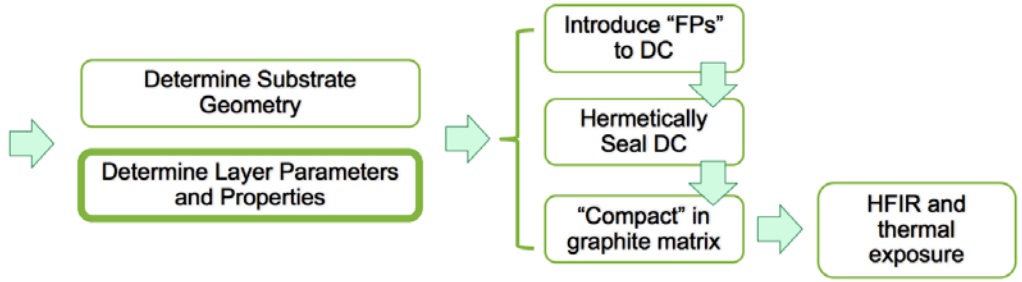


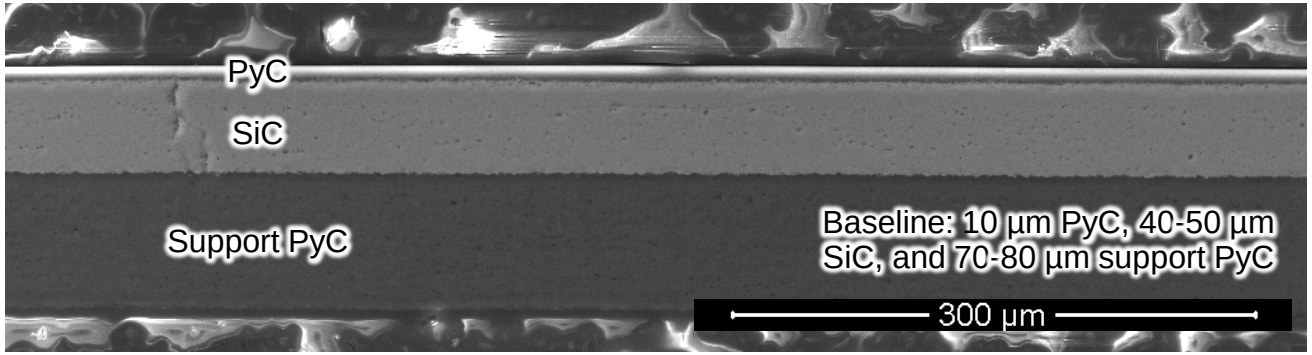
Grain size comparison batch to batch comparison (left), reproduced from [5]

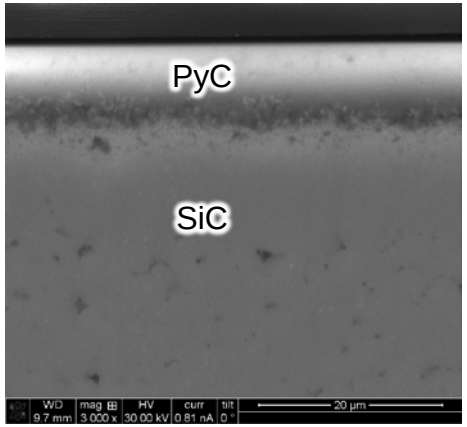
- Targets are based on specified values from AGR-2 UCO fuel [4]
- A lower density PyC variant is sought to explore the effects of a different PyC/SiC interface on FP interfacial reactions (introduction of FPs to SiC)
- The SiC variant is aimed to target the large-grained SiC microstructure from AGR-1 (B, V1, V2) [5]

[4] B.P. Collin, INL/EXT-14-32277 R1 (2014)
[5] T.J. Gerczak, *et al.*, J. Nucl. Mater. 480 (2016) 257-270.

Use AGR experience to target wanted properties







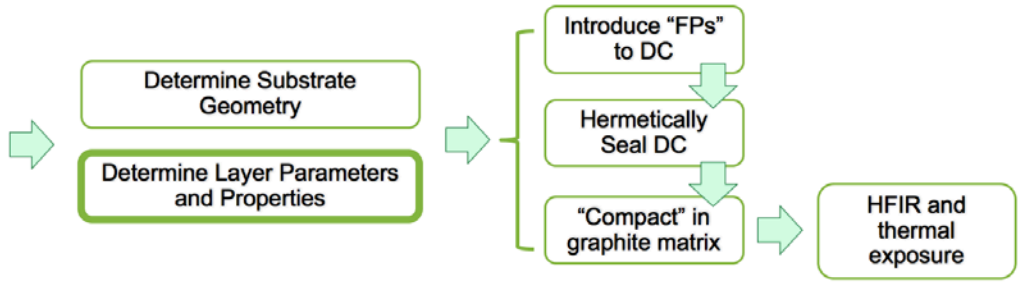
Example of run conditions from run DCCD-06

	Run Time	Temperature	Ar Fluid. Gas	MTS used	Addi. Gas	Acetyl.	Propyl.	H_2 carrier
PyC	3 min	1335°C	6300	-	-	1460	1240	-
SiC	180 min	1425°C	3250	180.1 g	2600 H_2	-	-	650
Support PyC	45 min	1320°C	6300	-	-	1460	1240	-

- Layer thickness and porosity is checked by optical and scanning electron microscopy
  - Observed porosity beyond ~20 µm in SiC, initiated EBSD analysis to confirm microstructure following past experiences [5]
- Baseline: 10 µm PyC, 40-50 µm SiC, and 70-80 µm support PyC

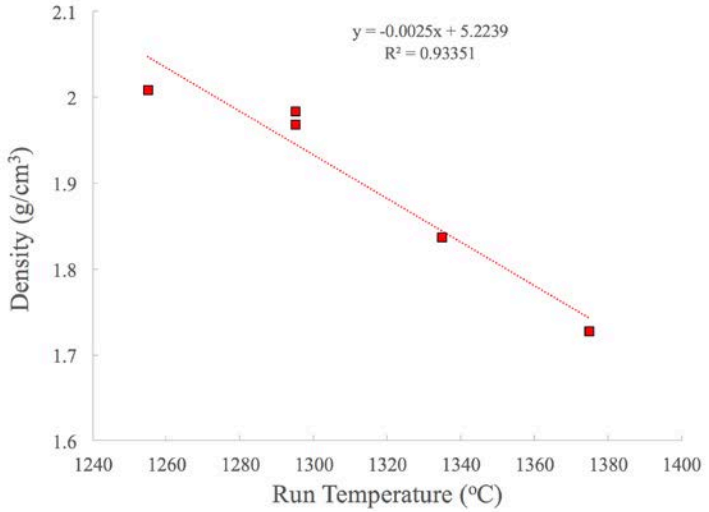
[5] T.J. Gerczak, *et al.*, J. Nucl. Mater. 480 (2016) 257-270.

Systematic determination of PyC properties



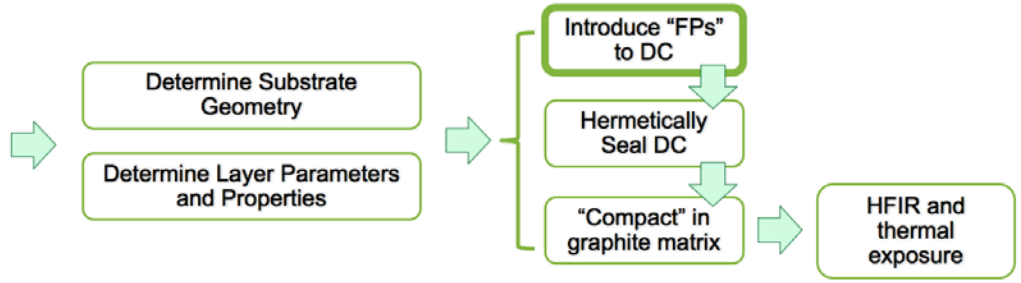
Run details for interrupted PyC only samples for density analysis (all gas flows in sccm)

	Run Time	Temperature	Ar Fluid. Gas	Acetyl.	Propyl.	Press. (torr)	PyC density (g/cm^3)
PyC-05I	4.6 min	1295°C	6300	1460	1240	790	1.984±0.002
PyC-07I	4.5 min	1335°C	4200	973	827	746	1.837±0.018
PyC-08I	3.9 min	1295°C	4200	973	827	747	1.968±0.020
PyC-09I	3.9 min	1375°C	4200	973	827	749	1.727±0.004
PyC-10I	5.8 min	1255°C	4200	973	827	748	2.008±0.008
PyC-11I	5.8 min	1295°C	4200	973	827	750	In progress

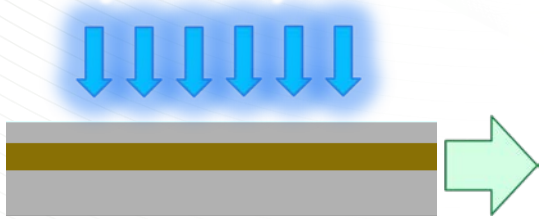


- Identified optimal PyC deposition condition based on density column measurements

Implantation condition chosen to reflect end of life fission product inventory from AGR-2

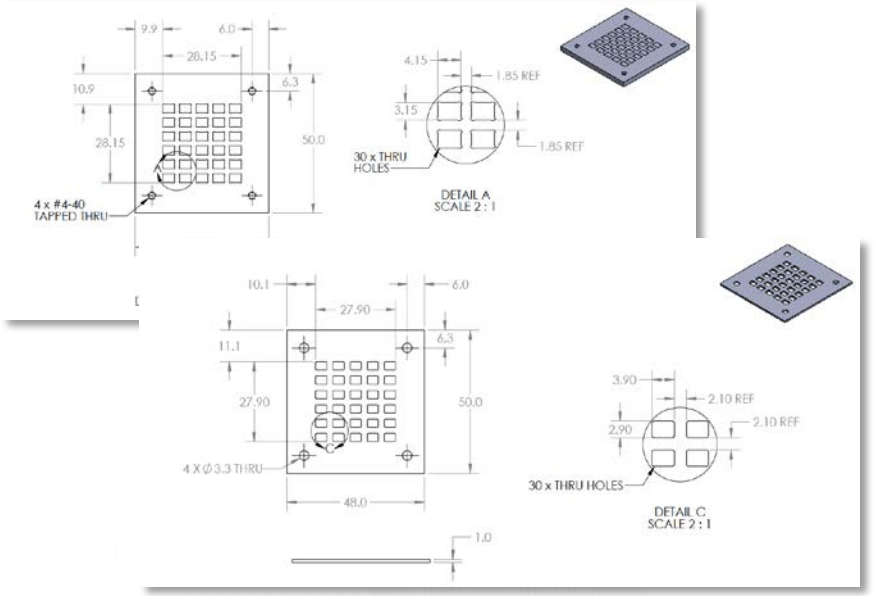


Implant "FP" Species



Implantation conditions

Diffusion Couple Type	Energy (kV)	Fluence (ions/cm^2)
Ag	400	4.20×10^{16}
Ag+Pd	400	(Ag) 4.20×10^{16}, (Pd) 7.76×10^{17}
Eu	400	7.98×10^{16}
Sr	400	1.36×10^{18}

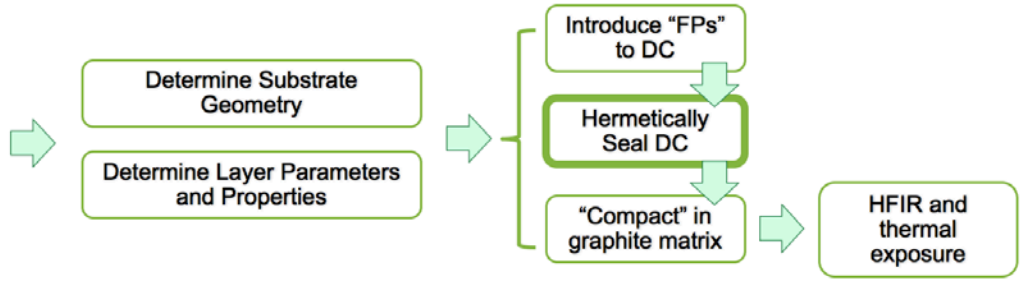


Drawing of sample holder

- Implantation conditions aim to match fission product inventory at end-of-life [6]: areal density relative to PyC/SiC surface areas
  - Aim for representative fission product concentrations interacting with SiC layer, approach does not account for the kernels ability to retain FPs
- Implantation will be conducted by the University of Michigan on the Michigan Ion Beam Laboratory (MIBL) 400-kV inline implanter
  - Subsequent analysis will also be completed by MIBL

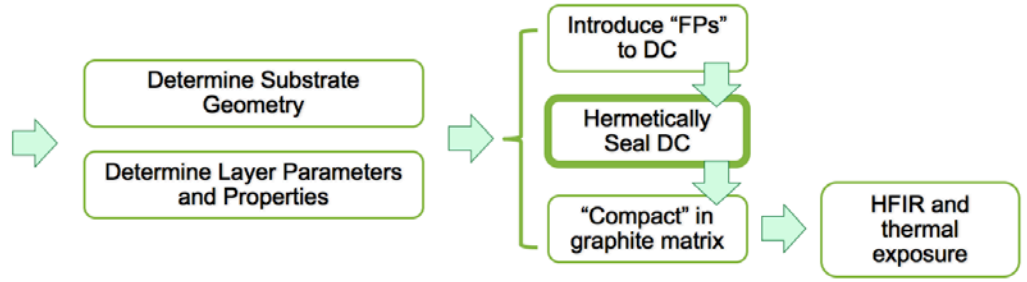
[6] J.W. Sterbentz, "JMOCUP as-run daily depletion calculation for the AGR-1 experiment in ATR B-10 position," ECAR-958 Rev. 2, Idaho National Laboratory (2013)

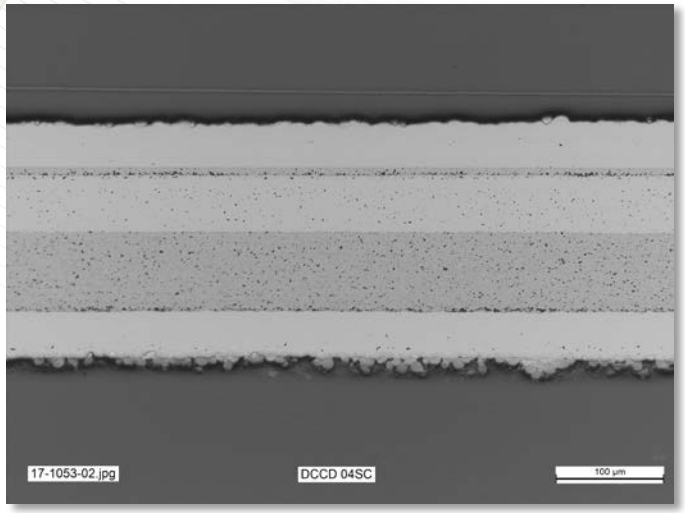
Maintaining a hermetic seal for thermal exposure

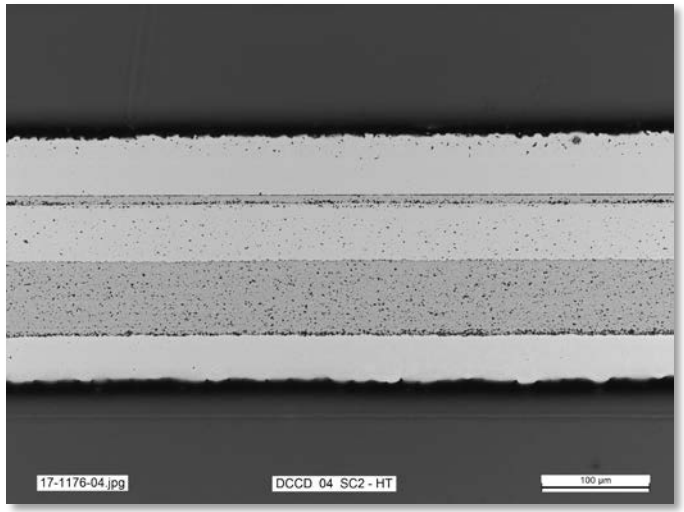


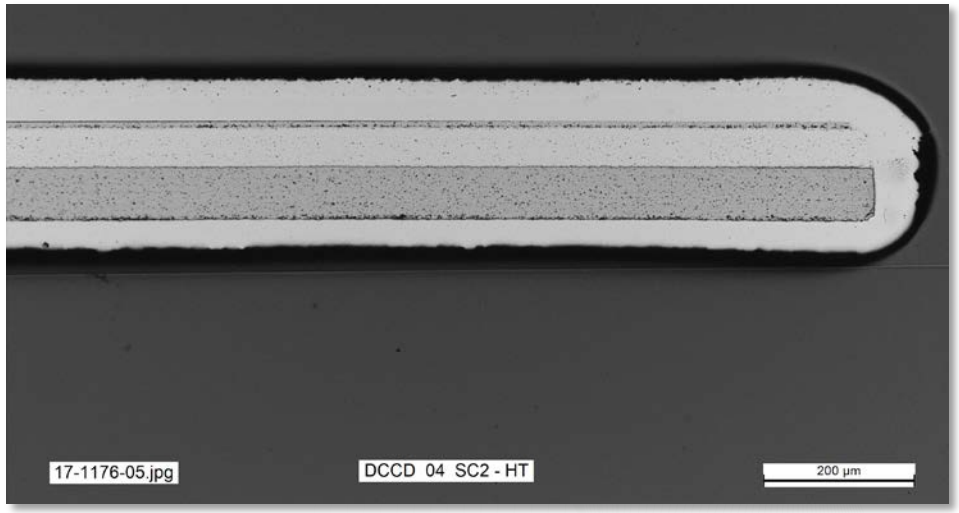
- Attempt to add SiC seal coating to retain fission products during thermal exposure
  - Using "rapid" initial coating rate to create a hermetic seal to retain implanted FPs during coating
  - Samples are introduced to a "hot" fluidized bed but experience ~3 hours at 1425°C
  - Fluidized along with "soft" buffer coated ZrO_2 kernels to minimize fracture
- Hermetic coating is tested by burning coated samples in air at 900°C for 8 hours

Maintaining a hermetic seal for thermal exposure







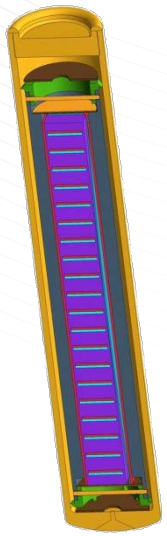


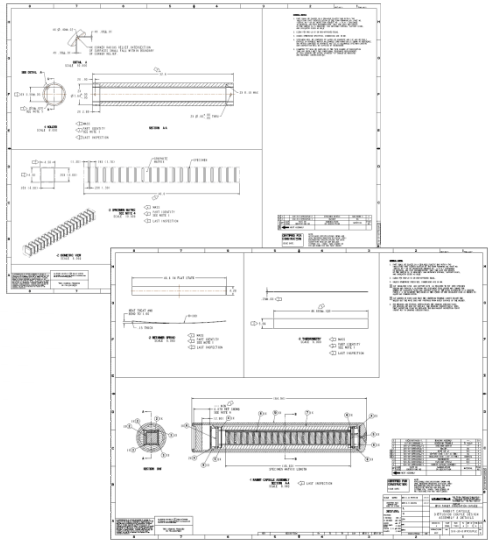
Cross-section of seal coated sample

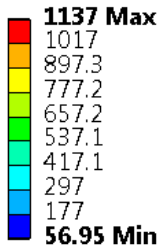
Cross-sections of seal coated sample after 900°C 8 hour burn in air

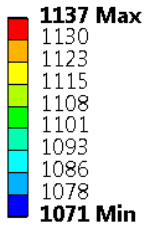
- Explored seal coating thicknesses from 25 to 50 µm (~3 hours at 1425°C)
- All PyC layers remained intact after air burn for 50-µm-thick seal coating layers
  - Need to determine retained fraction of implanted species after seal coat, utilize burn-leach to confirm retained concentration in PyC

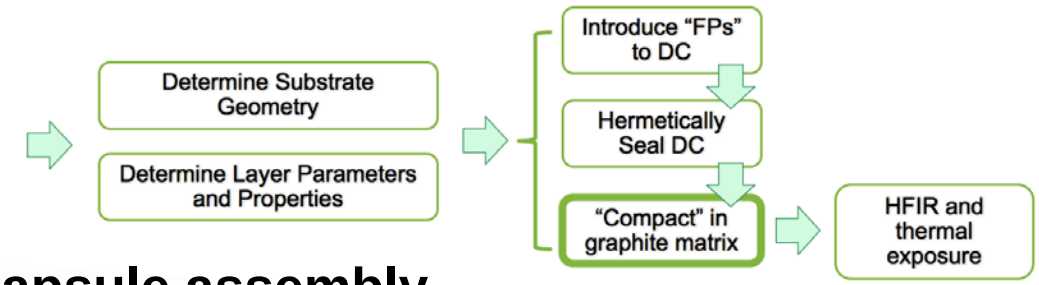
Design of DC “compact” and thermal analysis of capsule design



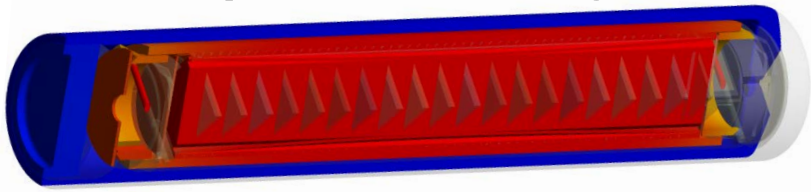




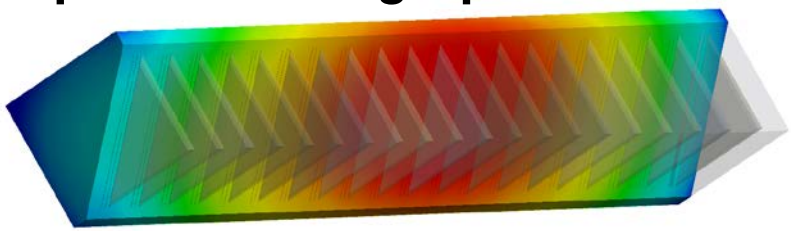




Capsule assembly



Specimens in graphite matrix



- Embed samples in a graphite monolith and machine to dimensional tolerances
  - Compact design has been validated to achieve targeted conditions
- Overcoating and compacting will be performed in the Coated Particle Fuel Development Laboratory at ORNL

Summary

- Demonstrated ability to fabricate hermetic, planar diffusion couple substrates using fluidized bed coater identical to those used to fabricate AGR TRISO fuel
- Performed study of coating conditions to obtain ideal layer properties
- Defined sample goals – understand the target variables, how to produce them, and how to measure them
- R&D ongoing for FP retention after seal coat and compacting.

General Discussion and ????

- This work supported by the US DOE, Office of Nuclear Energy Nuclear Energy University Program (NEUP), CFA-16-10764 and by the US DOE, Office of Nuclear Energy under DOE Idaho Operations Office Contract DE-FOA-00001281.

Thank you for your attention:

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