HTGR Technology Course for the Nuclear Regulatory Commission

May 24 – 27, 2010

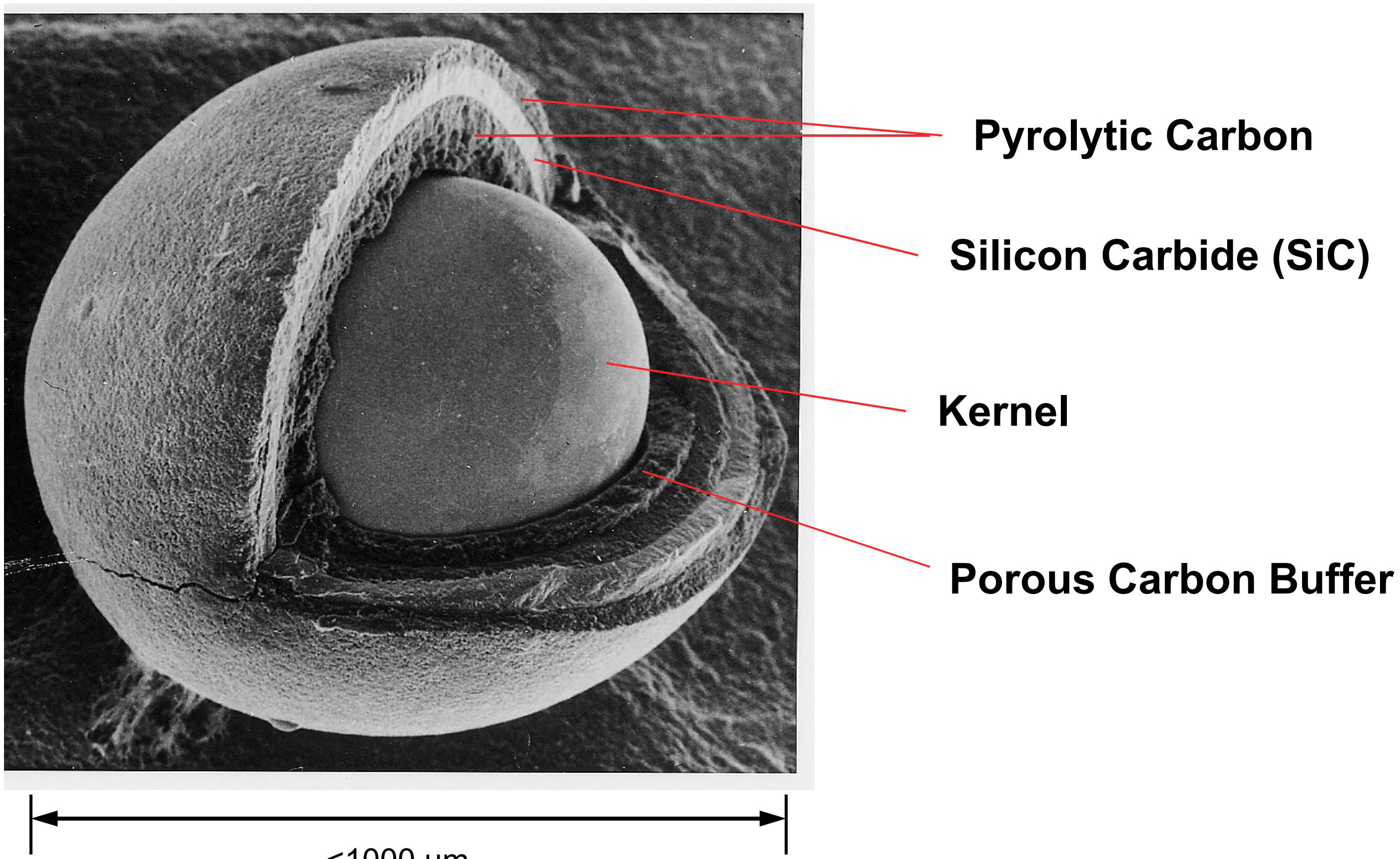
Module 7a

TRISO Fuel Design, Properties, and Requirements

Outline

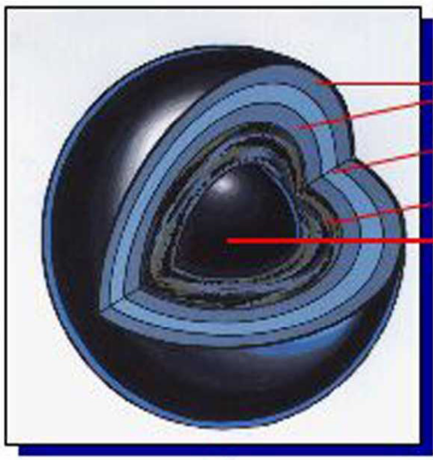
- TRISO coated fuel design and functions
- Coating structures and properties
- Fuel failure mechanisms
- Fuel performance requirements and specifications

The TRISO-coated Fuel Particle is Fundamental to HTGRs



But different HTGRs have different fuel element designs

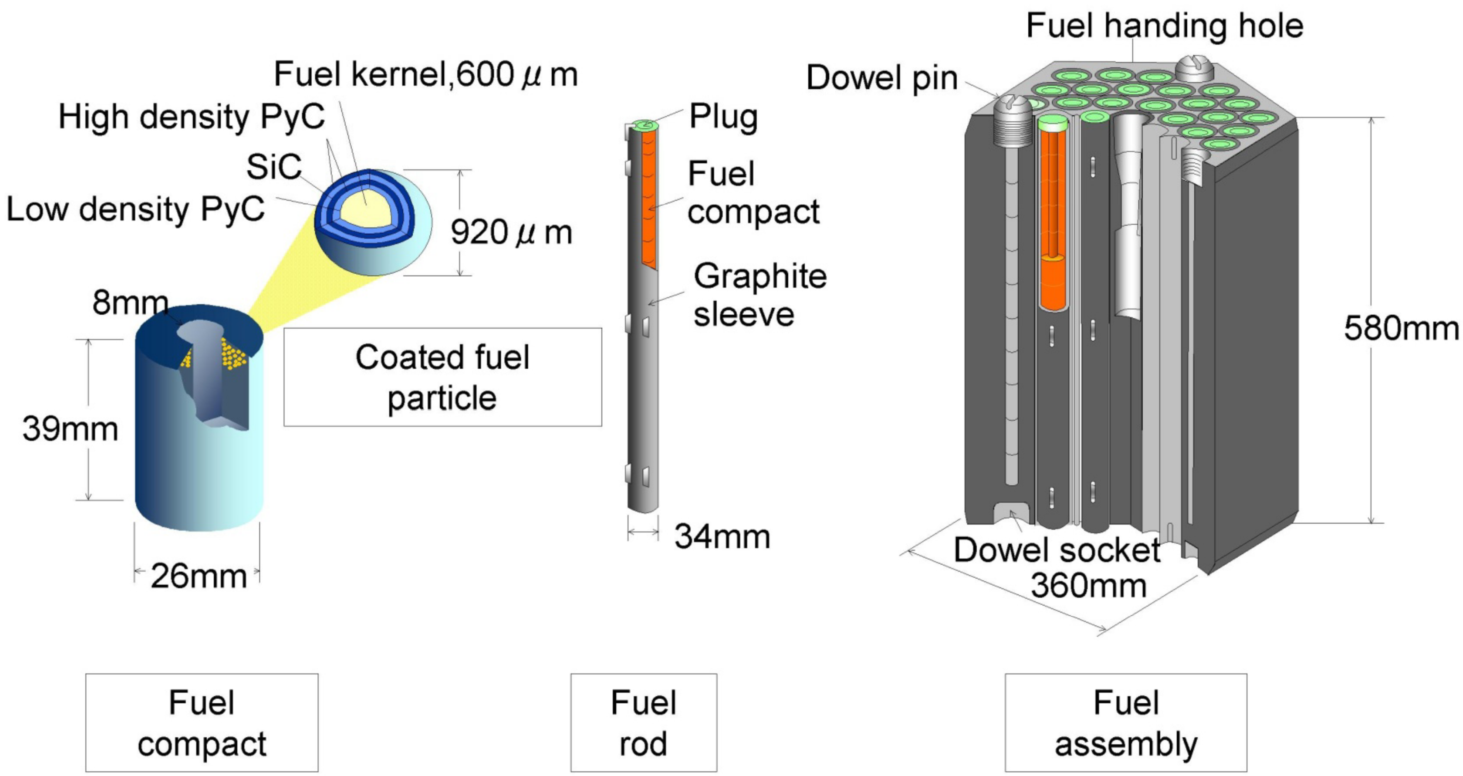
Prismatic HTGR Fuel Element - MHTGR



TRISO coated fuel particles (left) are formed into fuel rods (center) and inserted into graphite fuel elements (right).

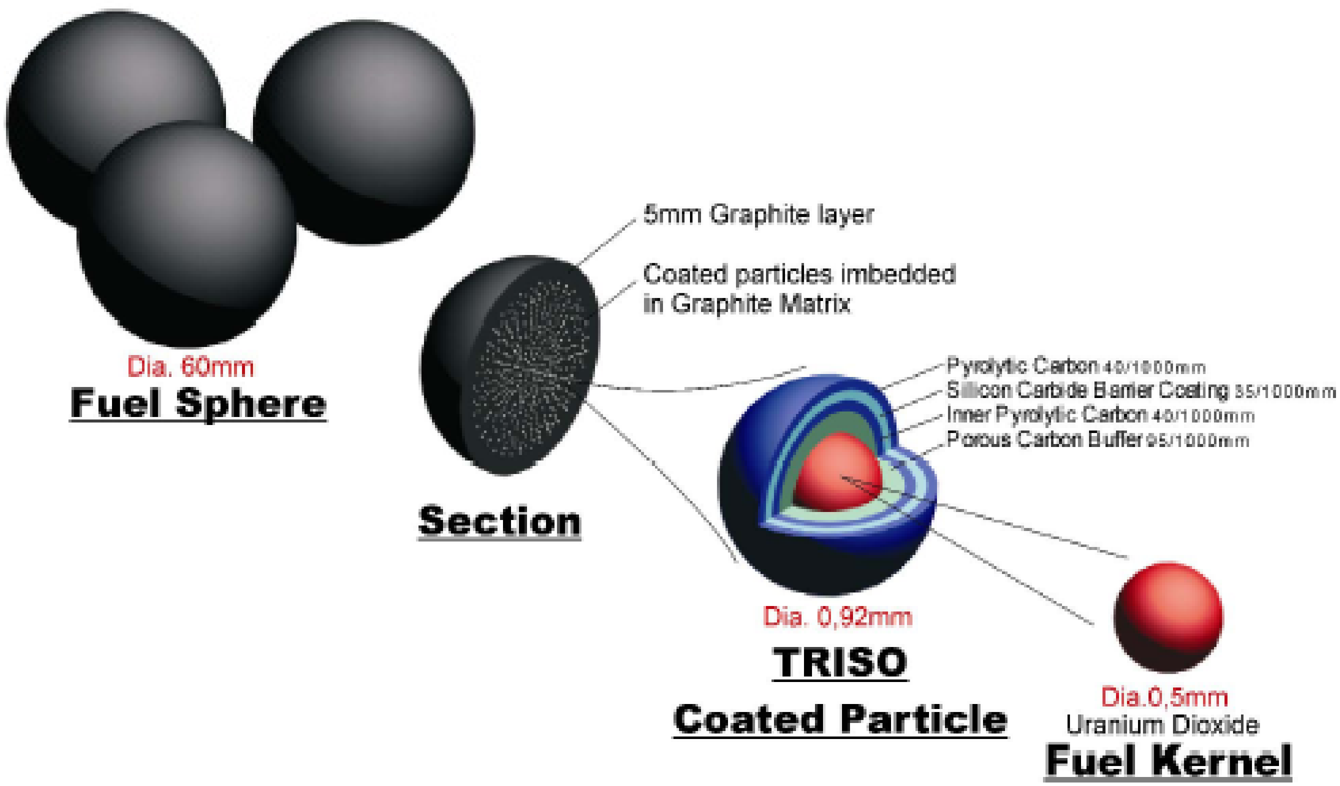
PARTICLES COMPACTS FUEL ELEMENTS

Prismatic HTGR Fuel Element – HTTR

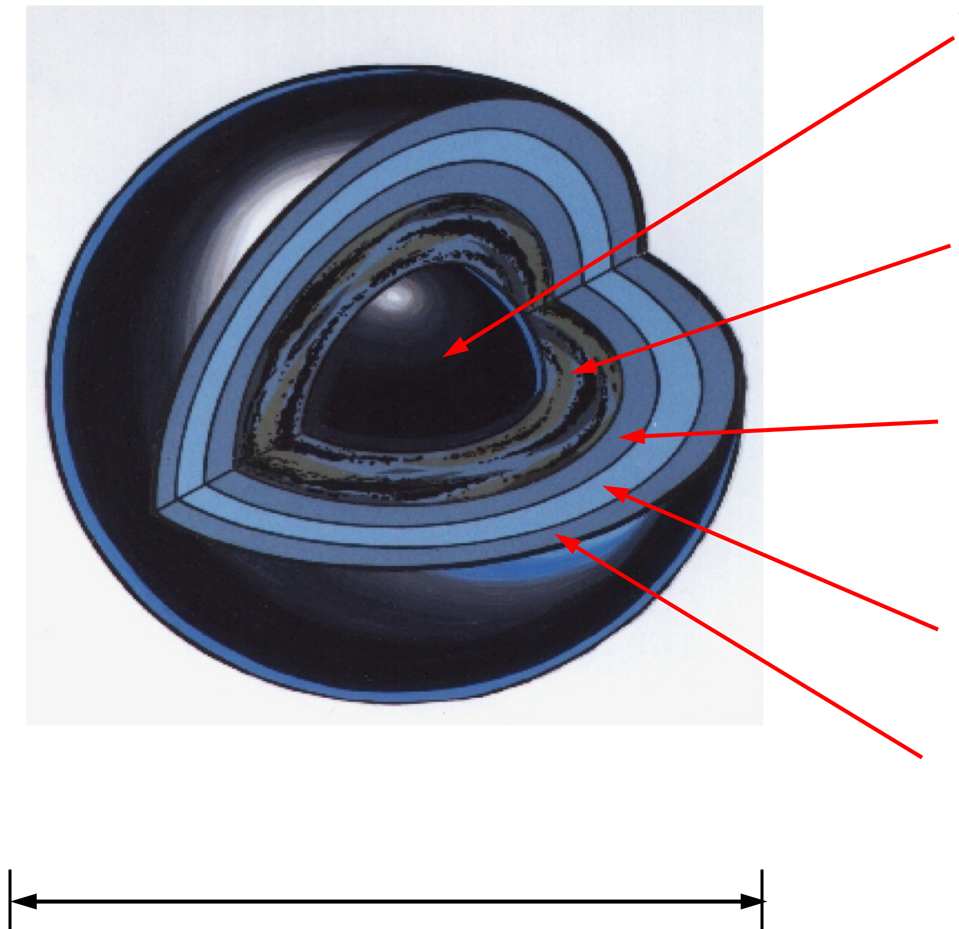


Pebble Bed HTGR Fuel Element - PBMR

Fuel Sphere



The TRISO Fuel Particle is a Mini Pressure Vessel



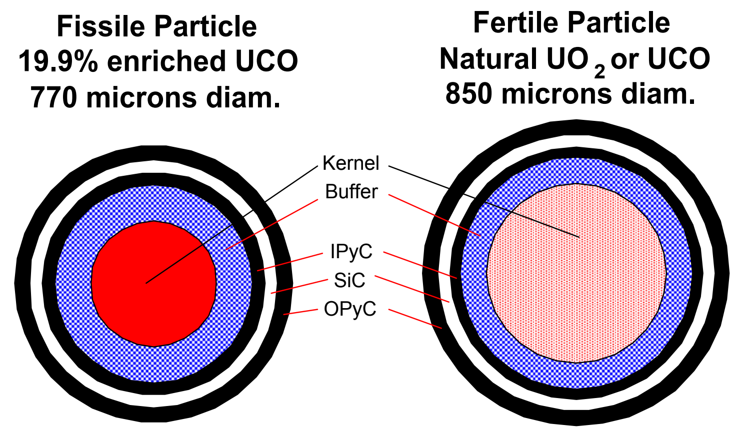
COMPONENT/PURPOSE

- **Fuel Kernel (UO_2 , UCO, UC_2 PuO_x, $(Th,U)C_2$)**
  - Provides fission energy and neutrons
  - Retains fission products
  - Controls particle oxygen potential
- **Buffer layer (porous carbon layer)**
  - Attenuate fission recoils
  - Void volume for fission gases
  - Accommodates kernel swelling
- **Inner Pyrocarbon (IPyC)**
  - Prevent Cl attack of kernel during manufacture
  - Provides structural support for SiC
  - Retains gaseous fission products
- **Silicon Carbide (SiC)**
  - Primary load bearing member
  - Retain gas and metal fission products
- **Outer Pyrocarbon (OPyC)**
  - Provides structural support for SiC
  - Provide fission product barrier in particles with defective SiC

Fissile and Fertile Fuel Particles

- **Past prismatic core designs (FSV, MHTGR, GT-MHR) included fissile and fertile fuel particles**
  - **Fertile particles breed U-233 or Pu**
  - **Enhances capability to minimize power peaking and maximize fuel cycle length**
- **NGNP/AGR Fuel Program is qualifying a single UCO fissile fuel particle**
  - **425-µm diameter kernel**
  - **14% U-235 enrichment**
- **Preliminary NGNP core design studies suggest feasibility of using single fuel particle design with one or more U-235 enrichments**

GT-MHR fissile and fertile fuel particles



	Fissile	Fertile
Kernel Diameter, µm	350	500
Buffer thickness, µm	100	65
IPyC thickness, µm	35	35
SiC thickness, µm	35	35
OPyC thickness, µm	40	40

HTGR Fuel Types

- **HTGRs can use many fuel types**
  - Fissile: UC_2, PuO_x, $(Th,U)C_2$, $(Th,U)O_2$, UO_2, UCO
  - Fertile: ThC_2, ThO_2, UO_2, UCO
- **UO_2 is most widely used fuel type**
  - Used in AVR (Germany), HTTR (Japan), HTR-10 (China)
  - Extensive irradiation and heating test data base from German HTGR Program
  - Reference fuel type for PBMR
- **UCO offers potential for improved fuel performance at higher fuel burnup**

UCO Fuel Being Qualified as Improved Fuel

- **UCO (UC_xO_y) is UO_2 with UC and UC_2 added**
- **UCO designed to provide superior fuel performance at high burnup**
  - Kernel migration suppressed (most important for prismatic designs because of larger thermal gradients)
  - Eliminates CO formation; internal gas pressure reduced
  - Fission products still immobilized as oxides
  - Allows longer, more economical fuel cycle
- **Reference fuel for NGNP prismatic reactor designs**
- **Potential higher burnup alternative for pebble bed HTGRs**
- **AGR-1 irradiation test provided a highly successful demonstration of UCO irradiation performance under normal reactor operating conditions**

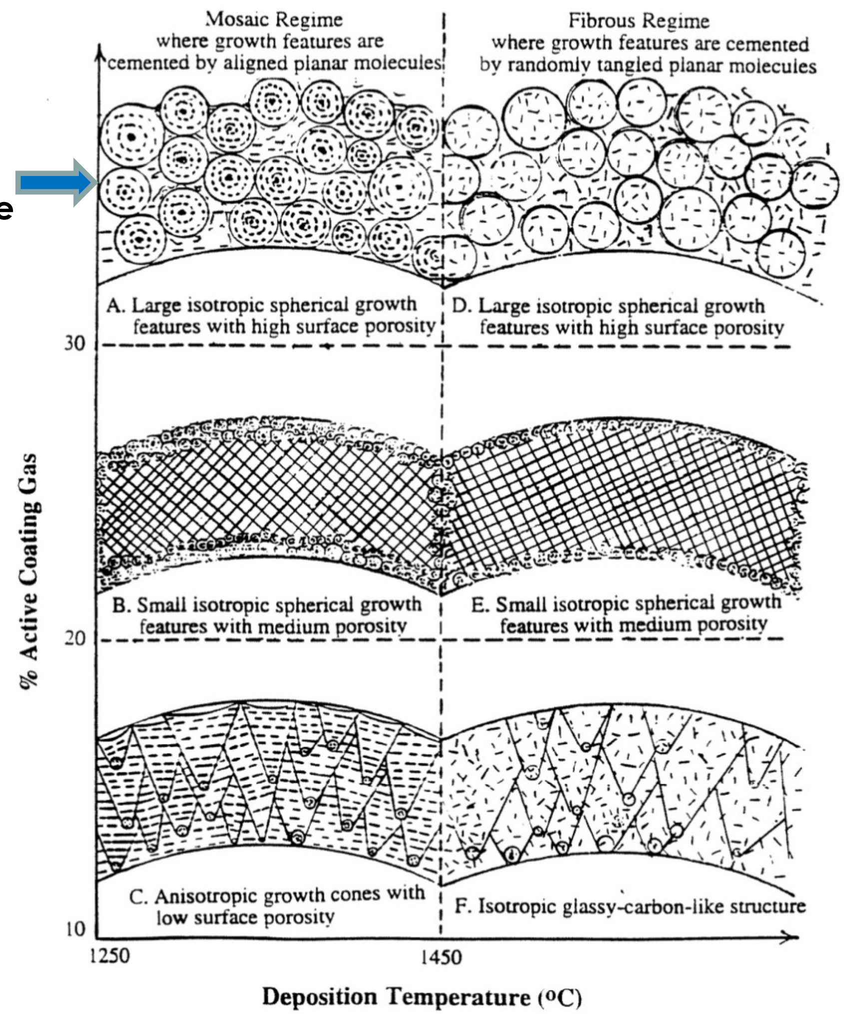
Outline

- TRISO coated fuel design and functions
- Coating structures and properties
- Fuel failure mechanisms
- Fuel performance requirements and specifications

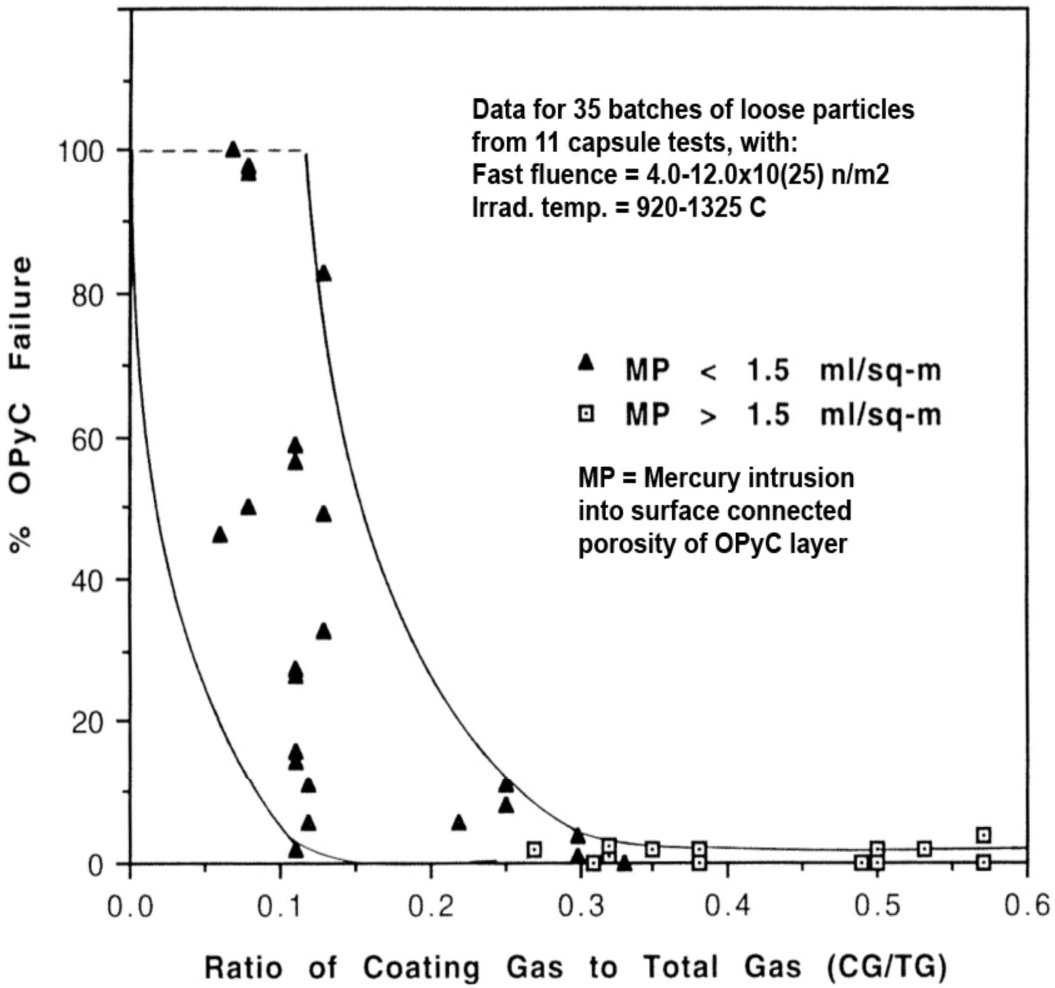
PyC Irradiation Performance is Highly Dependent on Microstructure

- **PyC microstructure varies greatly with key coating conditions**
  - Coating gas to total gas ratio
  - Deposition temperature
  - Coating rate
- **Low PyC anisotropy is essential, but not sufficient, to ensure good irradiation performance**
- **High density (~1.90 g/cc) and low permeability is also important.**

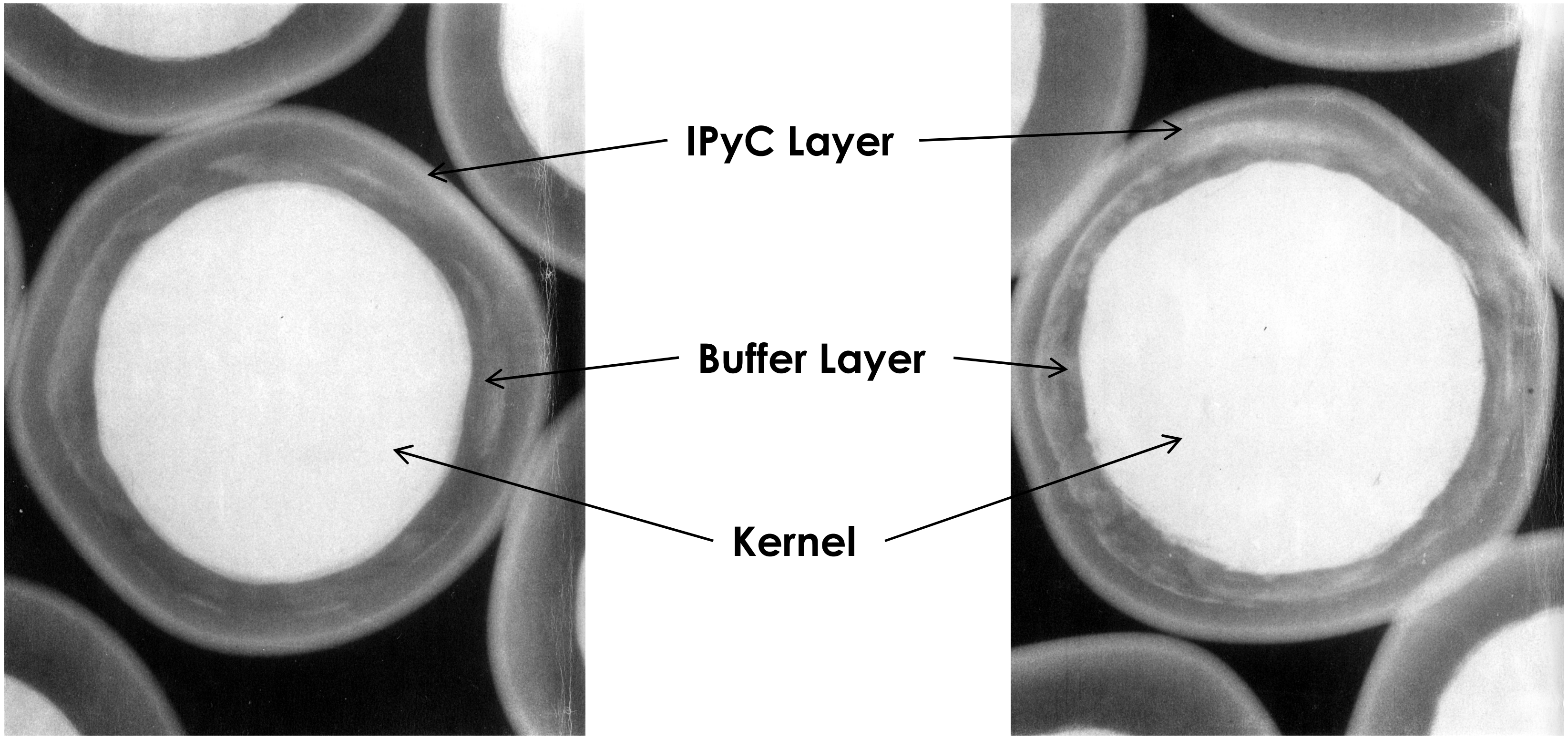
Desired structure



PyC Irradiation Performance is Well Correlated with Coating Gas Fraction



Uranium Dispersion May Result From Permeable or Cracked IPyC Coatings

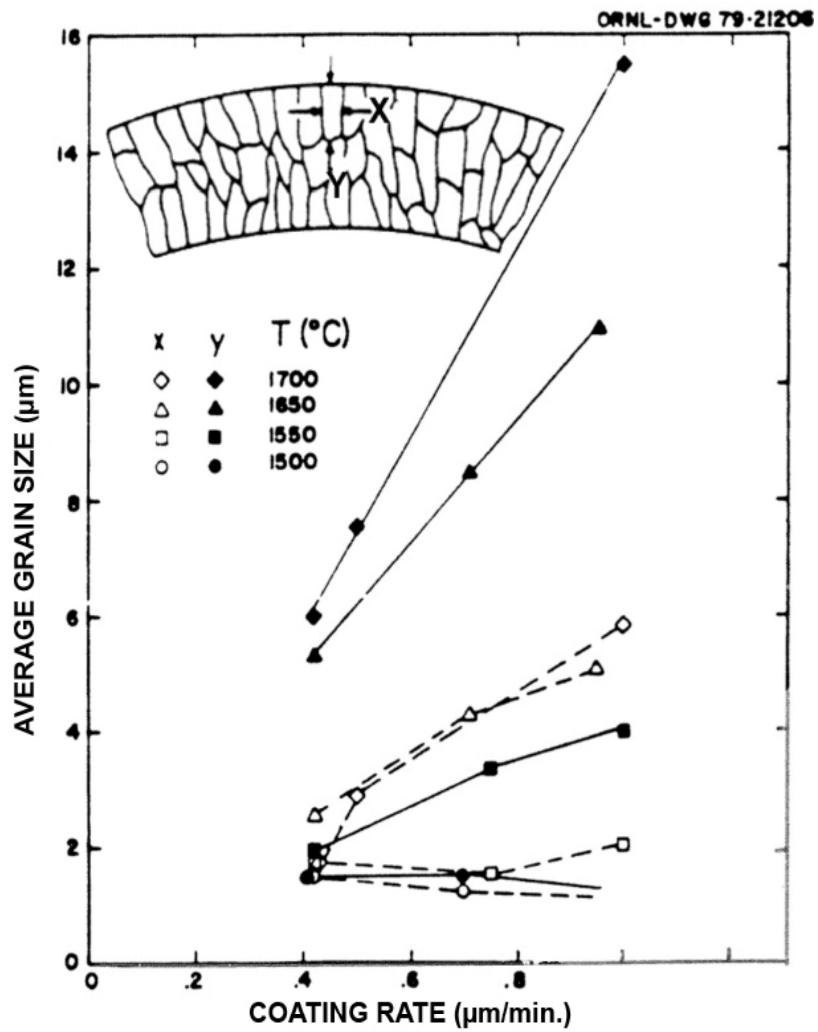


X-radiographs of IPyC-coated fuel particles showing varying degrees of uranium dispersion into the buffer coating layer

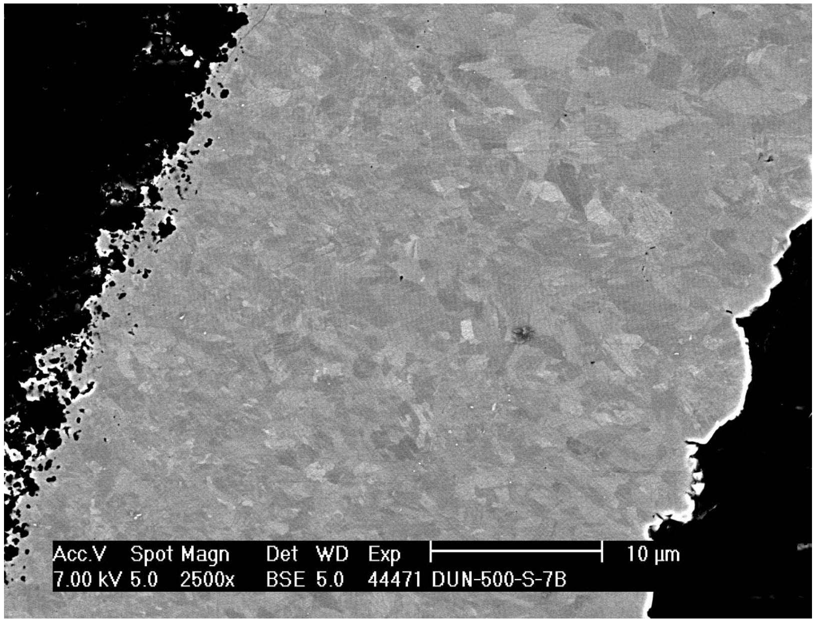
Uranium dispersion caused by reaction of chlorine with uranium kernel to form volatile uranium chlorides during SiC coating layer deposition

Density Measurements and Process Specifications Control SiC Quality

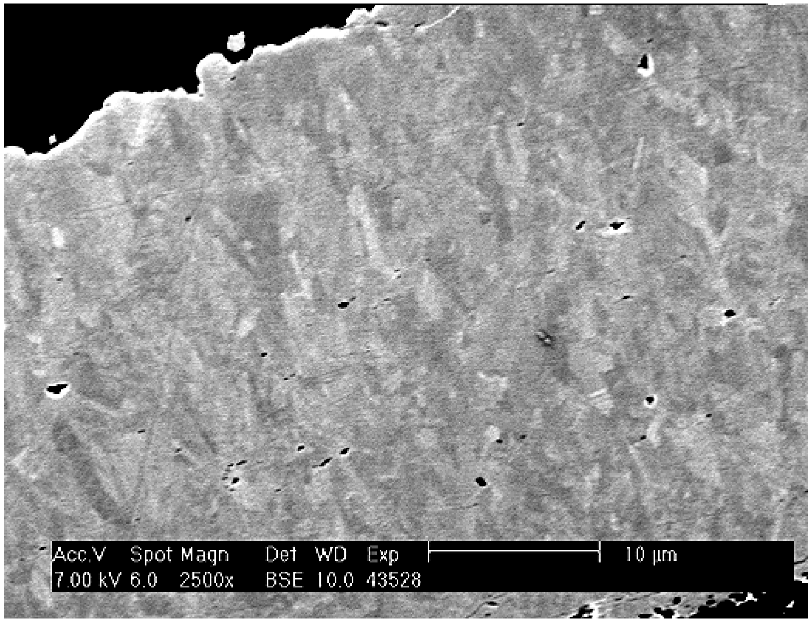
- **SiC microstructure varies with coating conditions**
  - Methyltrichlorosilane (MTS)/H_2
  - Deposition temperature
  - Coating rate
- **Properties important to SiC strength and fission product retentiveness**
  - Morphology (cubic β-SiC)
  - Small grains (1 – 2 µm)
  - No free Si or C
  - Defects
- **Desired SiC properties obtained at temperature of ~1500°C (coating bed) and coating rates < ~1 µm/min**



Argon Dilution Allows High-Density β-SiC to be Deposited at Lower Deposition Temperatures

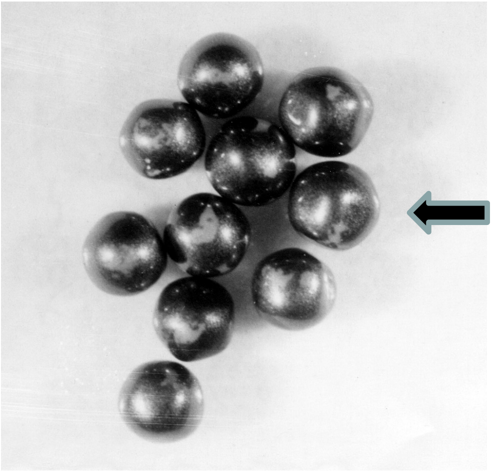


ORNL SiC, deposited at ~1400ºC with MTS/(H_2 & Ar); grain size ~ 1 to 2 um

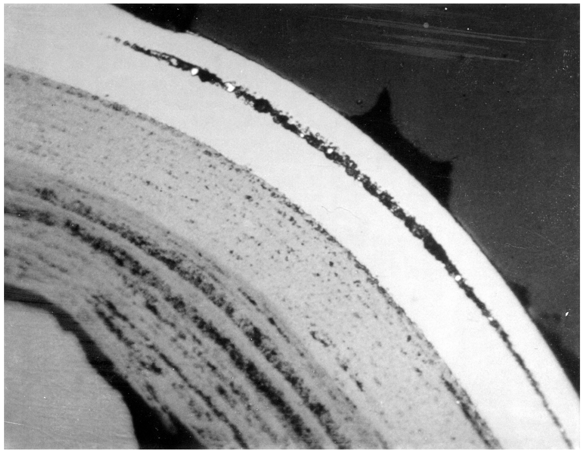


German SiC, deposited at ~1500ºC with MTS/H_2 (from HTR-Modul proof test composite EUO 2358 – 2365); grain size ~ 2 to 5 µm

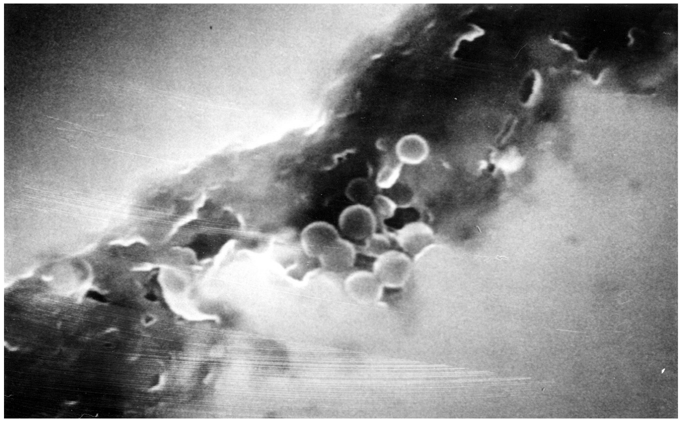
“Gold Spots” are an Optical Manifestation of Structural Defects in the SiC Coating



Gold spots on surface of SiC coating layer



Photomicrograph showing crescent-shaped defect in the SiC coating layer



Scanning electron microscope photo showing SiC soot in the defect

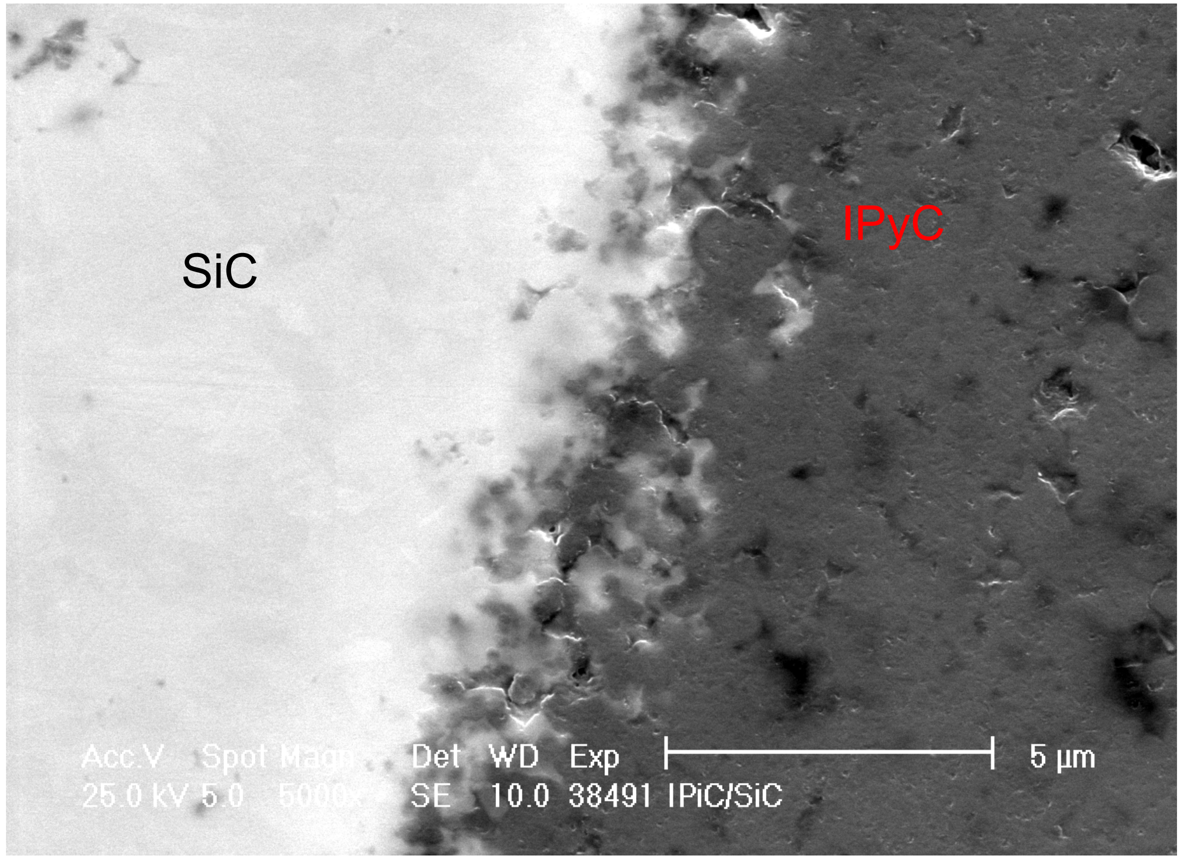
Defects caused by poor fluidization of the particle bed during chemical vapor deposition of the SiC coating layer

No clear correlation established between gold spots and irradiation performance

Strong Bonding Between the IPyC and SiC Layers is Beneficial to Good Irradiation Performance

Achieved using proper deposition conditions for the IPyC coating layer

Strong bonding results in compressive stress on SiC as IPyC shrinks with irradiation and prevents tensile stresses associated with IPyC pealing away from the SiC



Photomicrograph of German fuel particle (from HTR-Modul proof test composite EUO 2358 – 2365) exhibiting extensive penetration of SiC into the IPyC layer

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TRISO Fuel Failure Mechanisms

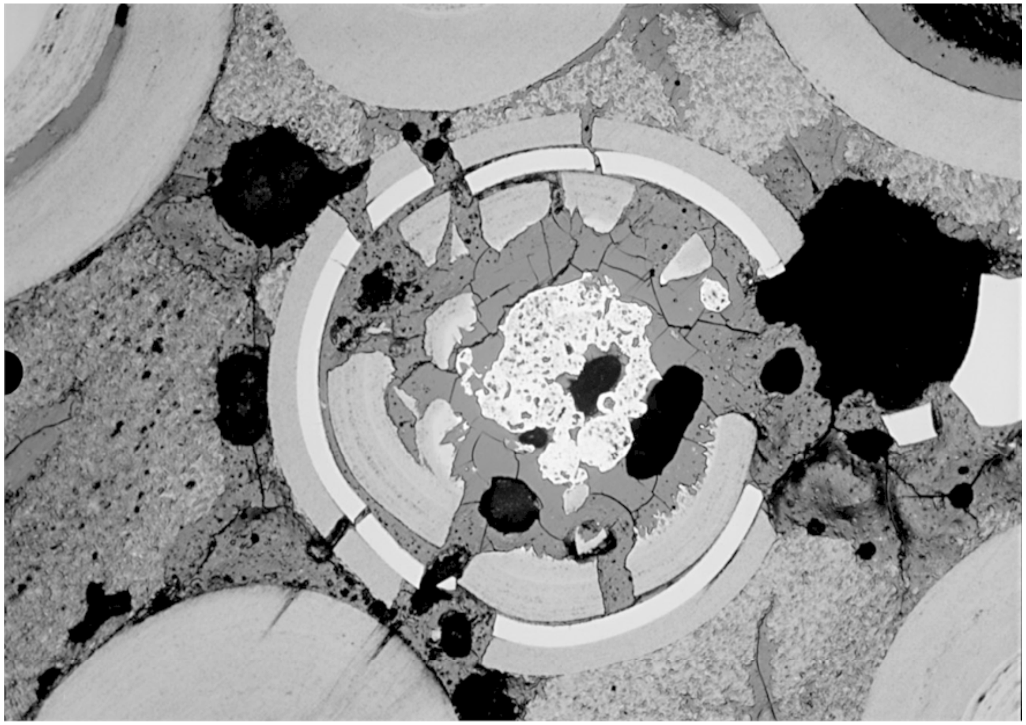
- **Mechanical**
  - Pressure vessel failure
  - PyC and SiC failure due to stress from excessive anisotropic shrinkage of the IPyC
  - OPyC failure due to OPyC - compact matrix bonding
- **Thermochemical**
  - Kernel migration
  - Fission product and impurity reactions with SiC
  - Uranium dispersion
  - CO reaction with SiC
  - SiC decomposition (at higher temperatures)

Pressure Vessel Failure

- **Internal gas pressure breaks particle**

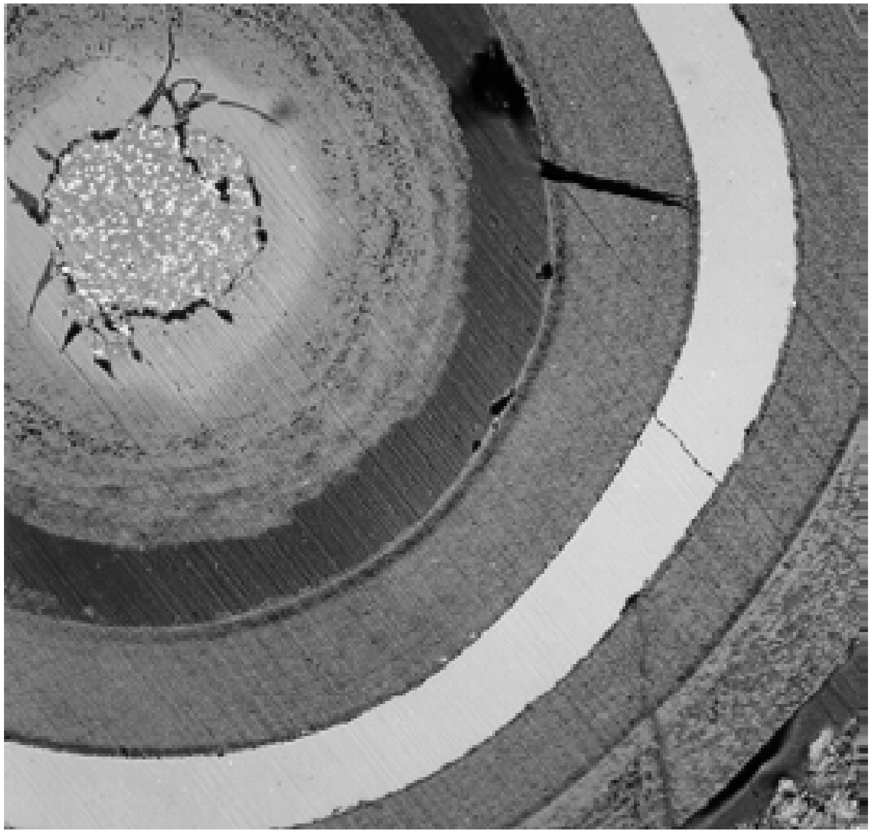
  $$P = P_{FP} + P_{CO}$$

- **Probability of pressure vessel failure in standard (in spec.) fuel particles is very low**



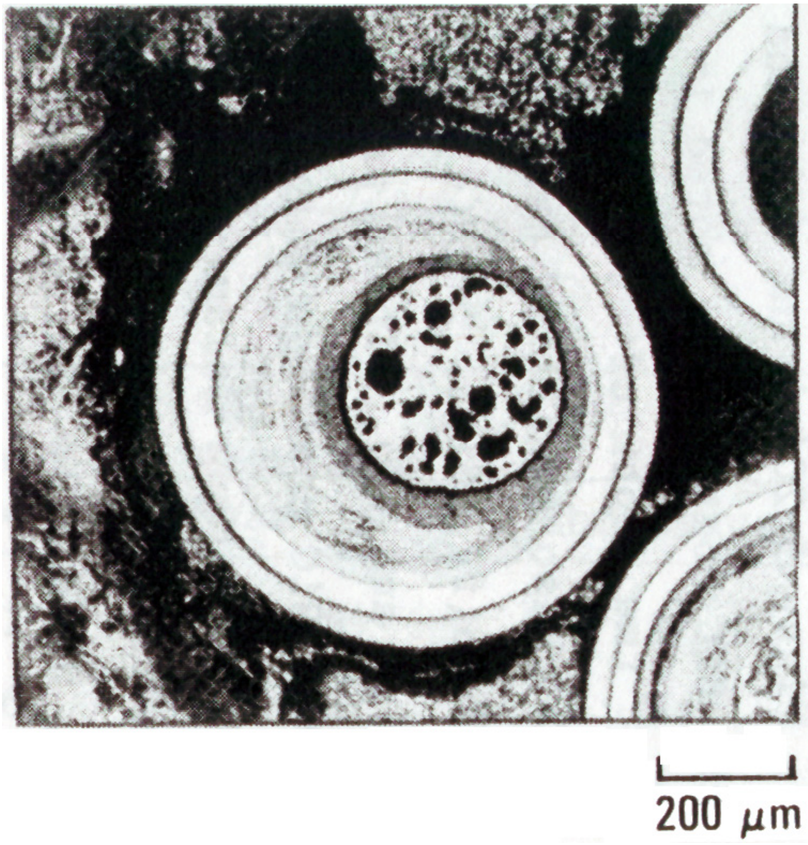
SiC Failure Due to Stress from Excessive Shrinkage of IPyC

Large shrinkage cracks in the IPyC layer led to stress concentrations in the SiC layer and to subsequent failure of the SiC layer in UCO fuel particles in HRB-21 and NP-MHTGR irradiation test fuel



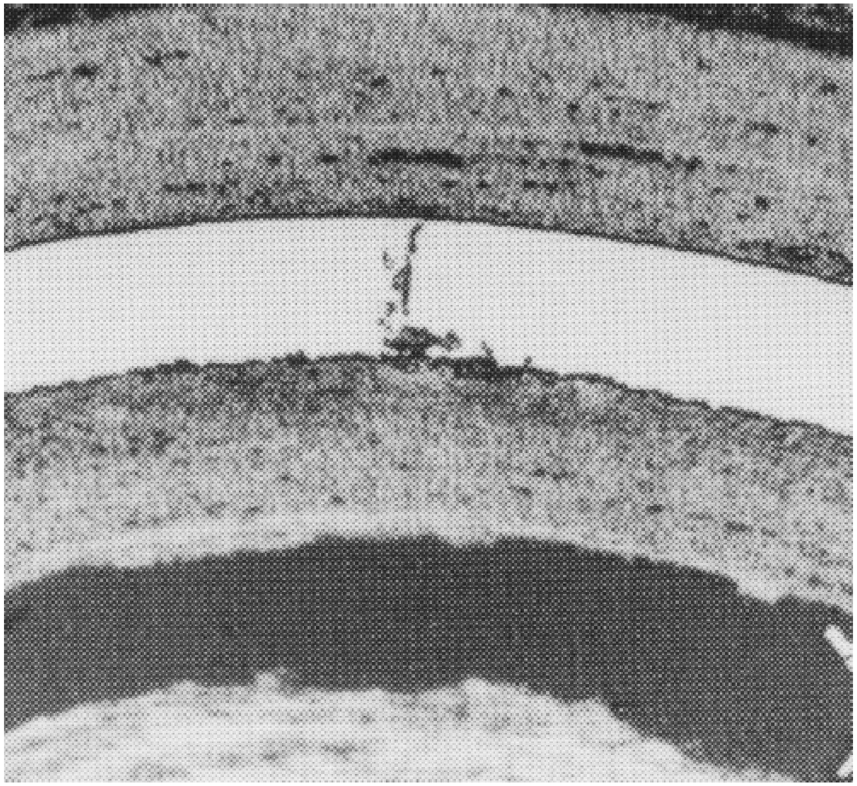
Kernel Migration

- Kernel migration in UO_2 fuel particles driven by thermal gradient across the particle
- Occurs due to carbon transport by reactions with oxygen
- Kernel migrates to hotter side of particle
- Migration rate increases with temperature, burnup, and thermal gradient
- Excessive migration results in SiC failure due to chemical reaction of SiC with uranium



Fission Product Corrosion of SiC

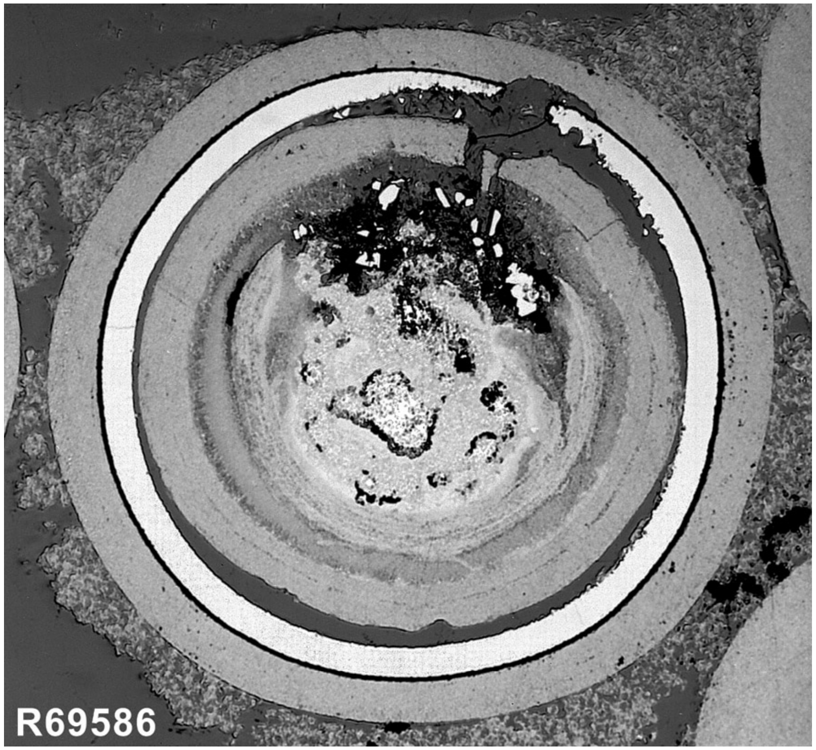
- Greatest effect from lanthanides and palladium above ~1300°C operating temperature
- Important to keep lanthanides as oxides in kernel (UO_2 and UCO)
- Lanthanides are mobile (as La_2C_3) in UC_2 fuel
- Pd not retained in kernel (UO_2 and UCO)



Localized Pd Attack of the SiC Layer

Corrosion of SiC - CO Attack

- In UO_2 fuel particles, cracked IPyC can allow internal CO attack of SiC under normal reactor operating conditions
- Use of oxygen getter (C, ZrC, etc.) in fuel particles prevents CO formation and reaction with SiC
- Under accident conditions, exposed SiC can be externally attacked by CO
  - Air or water ingress can result in reactions with graphite to produce CO
  - Failed or reacted OPyC exposes SiC layer



Parameters that Strongly Influence Mechanical Failure Mechanisms

Failure Mechanism	Service Conditions	Fuel Properties and Performance
Pressure vessel failure	Temperature, burnup, fast neutron fluence	SiC strength, buffer layer void volume, kernel type, fission gas release from kernel, IPyC and OPyC performance
SiC failure due to stress from excessive anisotropic shrinkage of the IPyC	Temperature, fast neutron fluence	SiC strength; IPyC thickness, density, anisotropy, and strength; dimensional change of IPyC; Irradiation-induced creep of IPyC
OPyC failure due to OPyC - compact matrix bonding	Temperature, fast neutron fluence	OPyC strength, OPyC surface connected porosity, intrusion of matrix into OPyC, irradiation-induced dimensional change of matrix

Parameters that Strongly Influence Thermochemical Failure Mechanisms

Failure Mechanism	Service Conditions	Fuel Properties and Performance
Kernel migration	Temperature, burnup, temperature gradient	Kernel type, buffer and IPyC thickness
Fission product and impurity reactions with SiC	Temperature, time at temperature, burnup	Kernel type, SiC thickness, SiC microstructure, transport behavior of fission products
Uranium dispersion	Temperature, burnup	IPyC permeability and defects, IPyC thickness, SiC thickness
CO reaction with SiC	Temperature, time at temperature, burnup	Kernel type, SiC thickness, IPyC integrity and permeability
SiC thermal decomposition	Temperature, time at temperature	SiC thickness and microstructure

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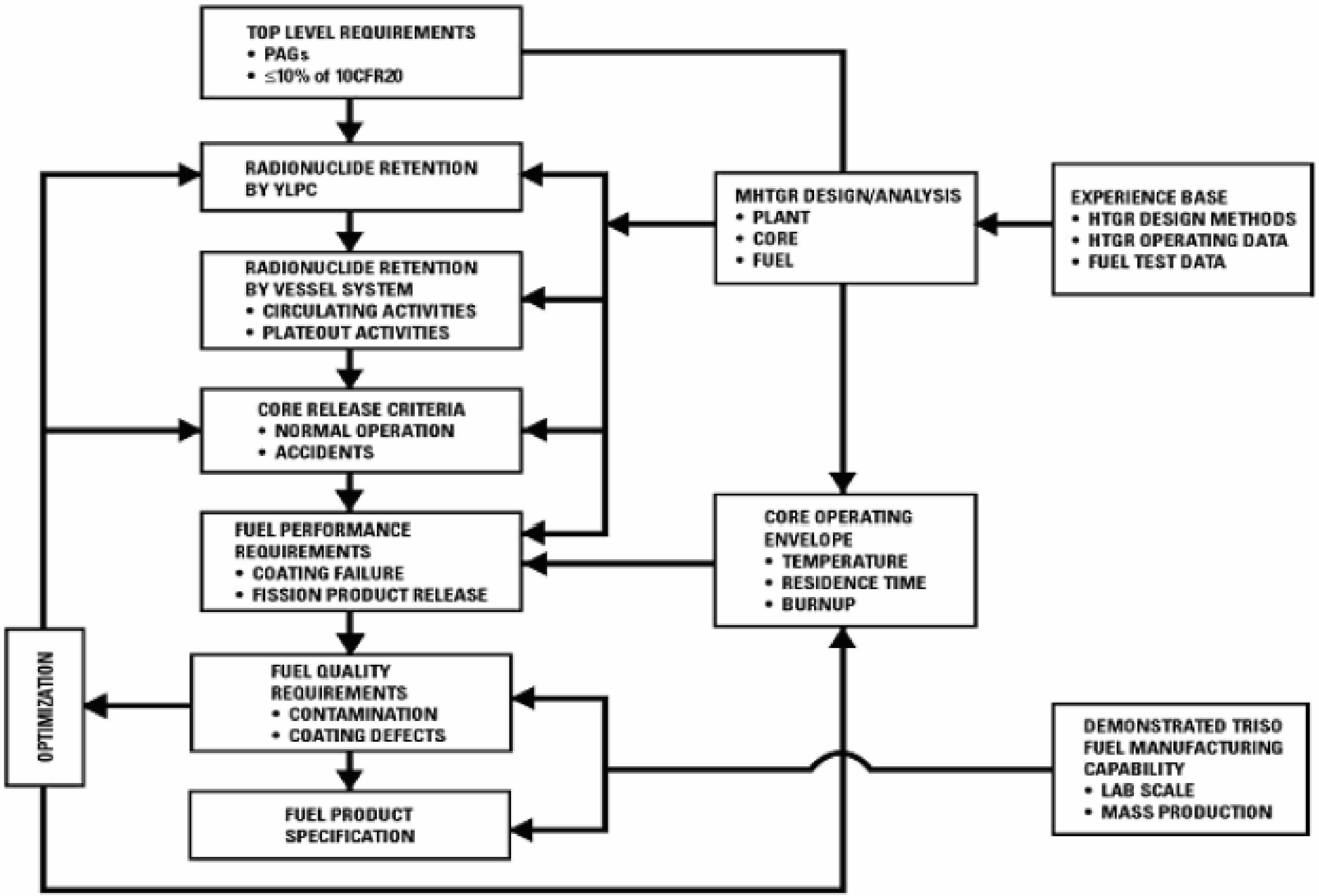
Product Specifications and Key Process Specifications Control Fuel Quality

- **Product specifications define property requirements for as-manufactured fuel**
- **Process specifications define how fuel is made**
  - Raw material specifications
  - Equipment specifications
  - Process conditions
- **Reactor designer uses both product and key process specifications to control fuel quality and fuel performance**
  - Kernels and compacts - product specifications
  - Coated particles - product and process specifications

Fuel Quality Specifications are Derived from Regulatory Dose Limits

- **SiC defect fraction allocated from allowable core Cs release derived from occupational exposure limit**
- **Heavy metal contamination fraction allocated from EPA Protective Actions Guide (PAG) thyroid dose limit during rapid depressurization**
- **Missing buffer fraction allocated from exposed kernel limit derived from PAG thyroid dose limit during rapid depressurization**

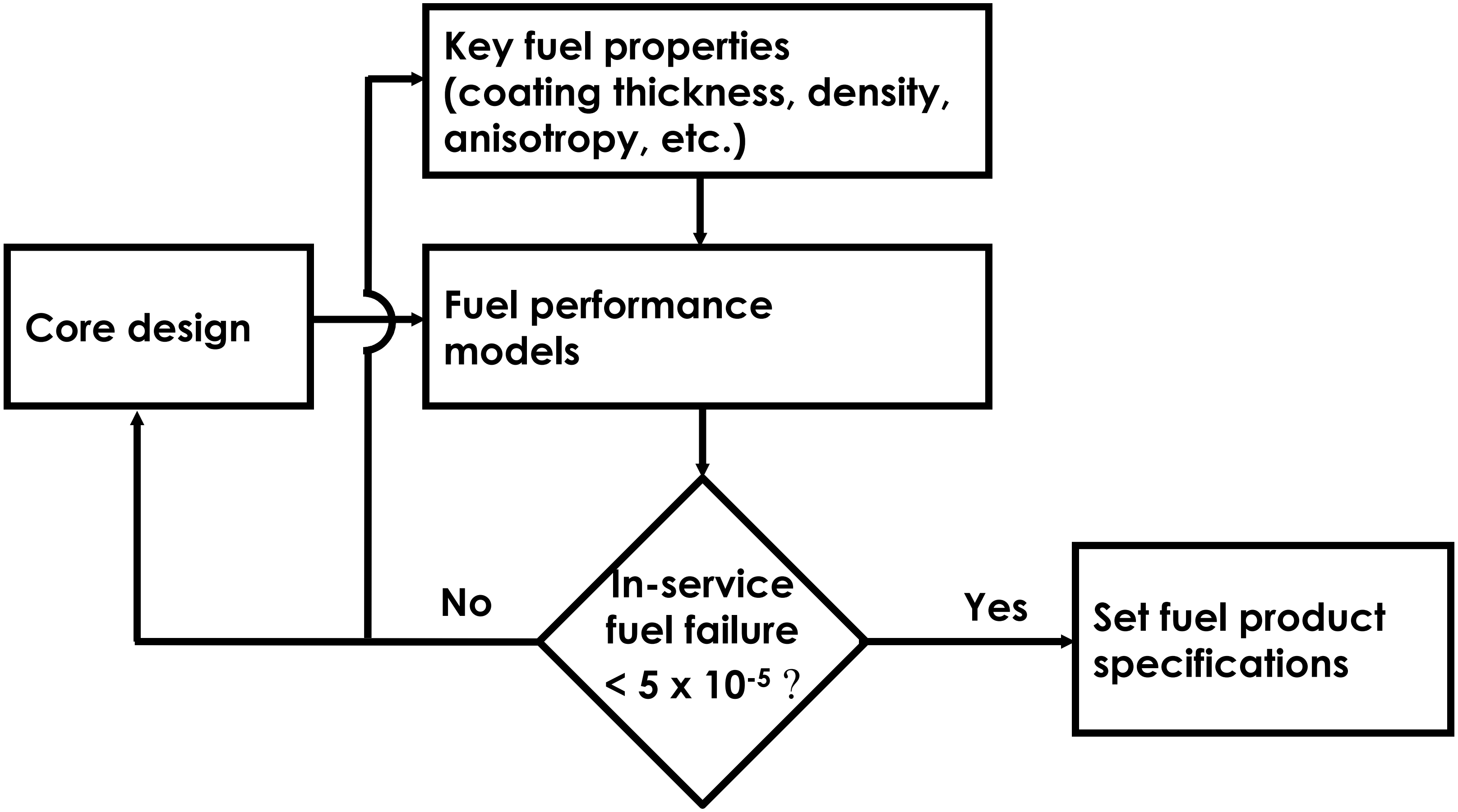
Logic for Deriving Fuel Performance Requirements



Preliminary Prismatic NGNP Fuel Requirements

Parameter	NGNP – 750°C Core Outlet Temperature	
	“Maximum Expected”	“Design”
As-Manufactured Fuel Quality		
HM contamination	≤ 1.0×10^{-5}	≤ 2.0×10^{-5}
Missing or defective buffer	≤ 1.0×10^{-5}	≤ 2.0×10^{-5}
Missing or defective IPyC	≤ 4.0×10^{-5}	≤ 1.0×10^{-4}
Defective SiC	≤ 5.0×10^{-5}	≤ 1.0×10^{-4}
Missing or defective OPyC	≤ 0.01	≤ 0.02
In-Service Fuel Failure		
Normal operation	≤5.0×10^{-5}	≤2.0×10^{-4}
Core heatup accidents	≤1.5×10^{-4}	≤6.0×10^{-4}

Fuel Properties are Also Specified to Prevent In-Service Fuel Failure



Fuel performance models establish link between in-service fuel failure and measurable fuel properties

NGNP/AGR Fuel Program - Reference UCO Fuel Product Specifications

Property	Property Type	Mean Value	Critical Region	Defect Fraction
Kernels				
C/U atomic ratio	Variable	0.40 ± 0.10	NA	NA
O/U atomic ratio	Variable	1.50 ± 0.20	NA	NA
Kernel diameter (μm)	Variable	425 ± 10	≤0.01 ≥375 ≤0.01 ≥475	NA
Kernel density (Mg/m^3)	Variable	≥ 10.4	NA	NA
Sphericity	Attribute	NA	≥1.05	≤0.10
Coated Particles				
Buffer thickness (μm)	Variable	100 ± 15	≤0.01 ≤58	NA
Buffer bulk density (Mg/m^3)	Variable	1.05 ± 0.10	NA	NA
IPyC thickness (μm)	Variable	40 ± 4	≤0.01 ≤30 ≤0.01 ≥52	NA
IPyC density (Mg/m^3)	Variable	1.90 ± 0.05	≤0.01 ≤1.80 ≤0.01 ≥ 2.00	NA
IPyC anisotropy (BAF_o)	Variable	≤1.045	≤0.01 ≥1.06	NA
SiC thickness (μm)	Variable	35 ± 3	≤0.01 ≤23	NA
SiC density (Mg/m^3)	Variable	≥ 3.19	≤0.01 ≤3.17	NA
OPyC thickness (μm)	Variable	40 ± 4	≤0.01 ≤20	NA
OPyC density (Mg/m^3)	Variable	1.90 ± 0.05	≤0.01 ≤1.80 ≤0.01 ≥ 2.00	NA
OPyC anisotropy	Variable	≤1.035	≤0.01 ≥1.06	NA
Missing OPyC (≤20 μm)	Discrete	NA	NA	≤3.0 x 10^{-4}

NGNP/AGR Fuel Program - Reference UCO Fuel Product Specifications (2/2)

Property	Property Type	Mean Value	Critical Region	Defect Fraction
Fuel Compacts				
Defective SiC coating fraction	Discrete	NA	NA	≤5.0 x 10^{-5}
Defective IPyC coating fraction (heavy metal dispersion)	Discrete	NA	NA	≤4.0 x 10^{-5}
Defective OPyC coating fraction	Discrete	NA	NA	≤1.0 x 10^{-2}
Heavy metal contamination (fraction exposed uranium)	Discrete	NA	NA	≤1.0 x 10^{-5}
Iron content outside SiC, ppm	Variable	≤50	≤01 ≥ 100	NA

Summary

- TRISO fuel is fundamental to modern HTGR designs
- The TRISO coating system is the primary barrier to release of fission products in HTGRs
- PyC and SiC irradiation performance is highly dependent on microstructure
- TRISO particles are subject to several mechanical and thermochemical failure mechanisms, which are avoided by careful control of coating properties and fuel service conditions
- Fuel specifications are derived to ensure compliance with regulatory dose limits
- Both product and process specifications are used to control as-manufactured fuel quality and in-service fuel performance

Suggested Reading

- "Fuel Performance and Fission Product Behavior in Gas Cooled Reactors," TECDOC-978, International Atomic Energy Agency, November 1997
- Hanson, D. L., "Technical Basis for NGNP Fuel Performance and Quality Requirements," Report 911168, Rev. 0, General Atomics, September 2009
- K. Minato et al., "Carbon Monoxide – Silicon Carbide Interaction in HTGR Fuel Particles, *Journal of Materials Science, 26, 2379 (1972)*
- "Effects of Deposition Conditions on the Properties of Pyrolytic Carbon Deposited in a Fluidized Bed," ORNL/TM-2005/533, Oak Ridge National Laboratory, September 2005