NE 591: Advanced Reactor Materials

Fall 2021

Dr. Benjamin Beeler

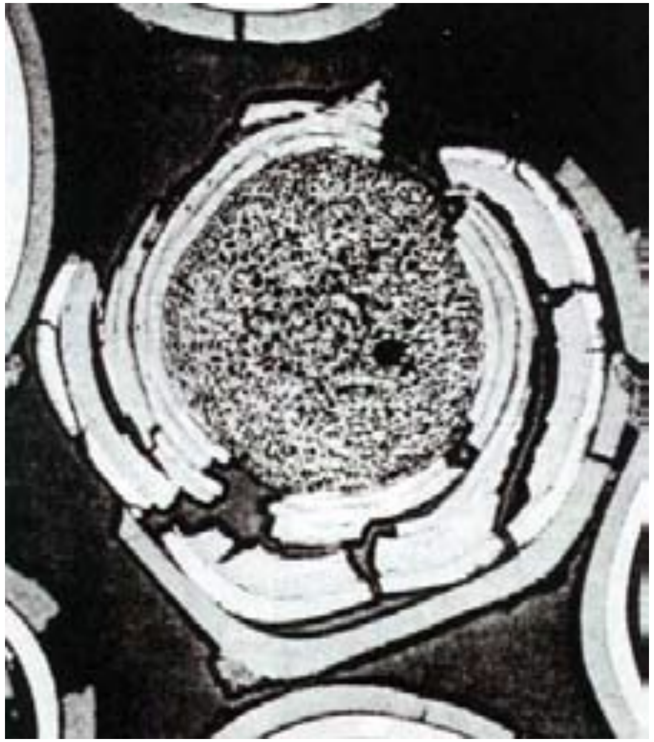
Last Time

- Graphite has a lot of texture and porosity, dependent upon fabrication
- Graphite shrinks, then undergoes turnaround
- This behavior is governed by point defects and Mrozowski cracks
- Turnaround happens quicker at higher temperatures, and is dependent upon applied stress
- Irradiation creep is critically important in graphite

TRISO FUEL FAILURES

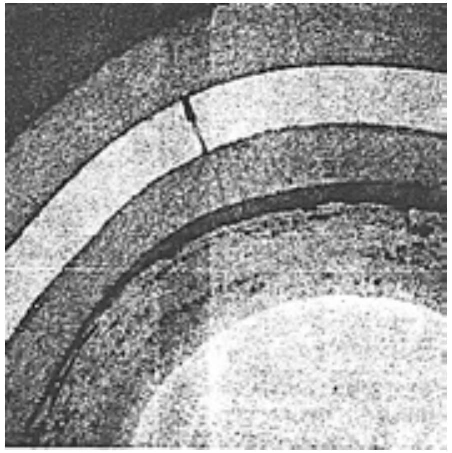
Fuel Failure Mechanisms

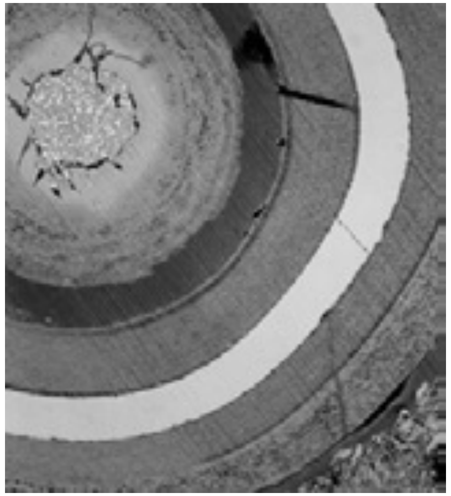
- Overpressure
- During irradiation, fission gases are released from the kernel into the porous buffer layer. The pressure that is generated exerts tensile forces on the SiC layer of the particle
- For UO2 kernels, there is also excess oxygen released during fission which will react with carbon from the buffer to form CO and CO2 gas
- Particles are generally sized with a large enough buffer void volume to ensure that they do not fail by overpressure during irradiation
- Particle failure is postulated to occur as a result of an insufficient or missing buffer layer that occurs during the coating process



Fuel Failure Mechanisms

- Irradiation-induced IPyC cracking
- Under irradiation, pyrocarbon (PyC) shrinks in both the radial and tangential directions
- At modest fluences it begins to swell in the radial direction
- This behavior puts the PyC layers into tension in the tangential direction
- However, irradiation-induced creep works to relieve the tensile stress in the PyC layer
- If the PyC is strongly attached to the SiC layer, the PyC shrinkage provides a strong compressive stress that offsets the tensile stresses generated by gas production in the kernel





Fuel Failure Mechanisms

- Debonding between IPyC and SiC
- The debonding is related to the strength of the IPyC/SiC interface
- Weakly bonded coating layers can partially detach because of the tensile stresses generated by the IPyC shrinkage under irradiation
- A particle for which partial debonding of the IPyC from the SiC has occurred can develop relatively large tensile stresses in the SiC
- Irradiation induced creep relieves the stress at longer times

Fuel Failure Mechanisms

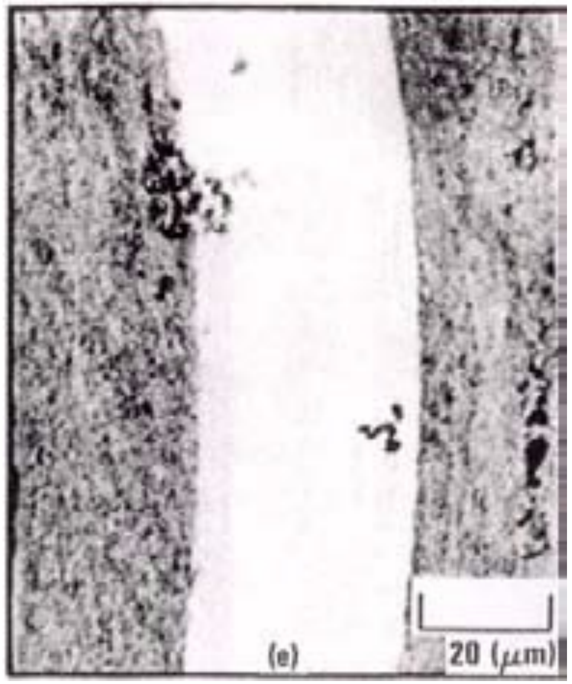
- Kernel migration

- Kernel migration is defined simply as movement of the kernel toward the TRISO-coating, which can lead to the kernel penetrating the TRISO-coating
- Kernel migration is associated with carbon transport in the particle in the presence of a temperature gradient
- A thermal gradient generates different equilibrium conditions for C, UO2, CO and CO2 at either side of the particle
- Carbon diffuses up the temperature gradient, the kernel diffuses down

Fuel Failure Mechanisms

- Fission product attack

- Fission products can be transported from the kernel to the inner surface of the SiC where they interact and can damage and potentially fail the SiC layer
- This is more of an issue in UC kernels
- In UCO kernels, the oxycarbide form of the kernel generally ties up all but the noble fission products (e.g., Pd) as either carbides or oxides, which tend to limit their mobility in the UCO system
- However, Pd and Ag transport have still been observed in UCO coated particle fuel



Fuel Failure Mechanisms

- Matrix-OPyC interactions and OPyC irradiation-induced cracking
- During manufacture, infiltration of the liquid graphitic matrix into the porosity of OPyC caused mechanical weakening
- During subsequent dimensional change under irradiation this caused OPyC layer to fail by cracking and debonding from the SiC layer
- This has largely been addressed through specifications on the matrix material and on the microstructure of the OPyC

- Non-retentive SiC
- There are situations where the SiC layer becomes functionally failed or degraded in some way and is no longer retentive of fission products
- Diffusive release through intact layers
  - If fuel temperatures during normal operation approach 1300C, then some of the fission products that are usually retained by the TRISO coating (e.g., cesium) will be able to diffuse out of the particle during normal operation
- SiC degradation resulting in permeability
  - Cesium attack of the SiC and/or CO corrosion of the SiC can allow fission products to be released at high burnup

Fuel Failure Mechanisms

- Creep failure of PyC

- Under stress, thermal creep of the PyC will occur
- In some post-irradiation heating tests, photomicrographs reveal a thinning and failure of the PyC
- Such failure has not led to failure of the SiC layer

- SiC thermal decomposition

- At very high temperatures (> 2000C) the SiC layer undergoes thermal decomposition
- The phenomenon is primarily a function of temperature and time and has not played a major role in fuel failure at lower accident temperatures

Fuel Failure Mechanisms

- Kernel-coating mechanical interaction (KCMI)

- At sufficiently high burn-up values, it is inevitable that all gas gaps between the kernel and coatings will close, thereby resulting in a mechanical interaction between the two
- Modelling studies predict that the SiC layer will fail shortly after the onset of KCMI
- This failure mechanism could be of increasing importance as attempts are made to achieve higher burn-up values

Fuel Failure Mechanisms

Failure mechanism	Reactor service conditions	Particle design and performance parameters that affect the failure mechanism	Comments
Pressure vessel failure	Temperature Burnup Fast fluence	Strength of SiC Buffer density (void volume) Fission gas release CO production Layer thicknesses Kernel type (UO_2, UCO)	
Irradiation-induced PyC failure	Fast fluence Temperature	Dimensional change of PyC Irradiation-induced creep of PyC Anisotropy of PyC Strength of PyC PyC thickness PyC density	Can be ameliorated by proper coating conditions
IPyC partial debonding	Temperature Fast fluence	Nature of the interface Interfacial strength Dimensional change of PyC Irradiation-induced creep of PyC	Can be ameliorated by proper coating conditions
Kernel migration	Temperature Burnup Temperature gradient	Layer thicknesses Kernel type	UO_2 only. Not important for UCO. Reasonably well understood
Fission product attack	Temperature Burnup Temperature gradient Time at temperature	Fission product transport behavior Diffusion Buffer densification and cracking Chemical state/transport behavior of fission products Microstructure of PyC and SiC	Could be more important at high burnup in LEU fuels because of greater yields of palladium from plutonium fissions
Non-retentive SiC Layer: Diffusive release through intact layers	Temperature Burnup Temperature gradient Time at temperature	Chemical state/transport behavior of fission products Microstructure of SiC SiC thickness	More important at higher temperatures (> 1200*C) where existing data suggest diffusion will contribute to the source term.
Non-retentive SiC layer:	Burnup Temperature Fluence	Kernel type (UO_2, UCO)	CO is generated in particles with UO_2 kernels.
SiC Corrosion by CO	Time at temperature	IPyC performance	At elevated temperatures, CO can attack the SiC layer if the IPyC layer is porous or has failed.
SiC degradation by cesium		Microstructure of SiC Thickness of SiC	Exact mechanism is unclear but limited data suggest cesium may degrade SiC layer
PyC thermal creep	Time at temperature	Thickness of PyC and stress state of PyC	Not important in traditional accident envelope (peak temperature < 1600°C)
SiC thermal decomposition	Temperature Time at temperature	SiC thickness Microstructure of SiC	Not important in traditional accident envelope (peak temperature < 1600°C)
Kernel Coating Mechanical Interaction (KCMI)	Burnup Fast Fluence Temperature	Initial Kernel – Coating Gas Gap Buffer properties IPyC Properties Kernel Swelling Rate	Failure of SiC Layer shortly after Gas Gap closed at sufficiently high Burn-ups

Fuel Failure Summary

- Overpressure
- IPyC cracking
- IPyC and SiC debonding
- Kernel migration
- Fission product attack
- Creep failure of PyC
- SiC decomposition
- KCMI

- These failure mechanisms have been observed to some extent in TRISO-coated fuel testing activities conducted around the world
- They are in general functions of temperature, burnup, fluence and temperature gradient in the particle and details of the particle design
- TRISO-coated fuel is usually designed such that none of the fuel failure mechanisms are expected to be significant

TRISO FUEL PERFORMANCE

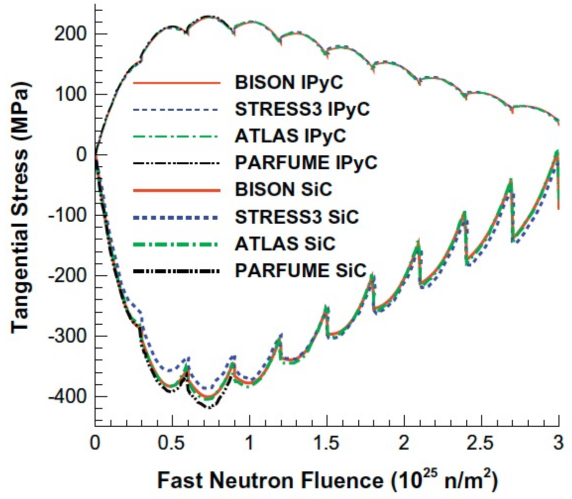
TRISO Fuel Performance

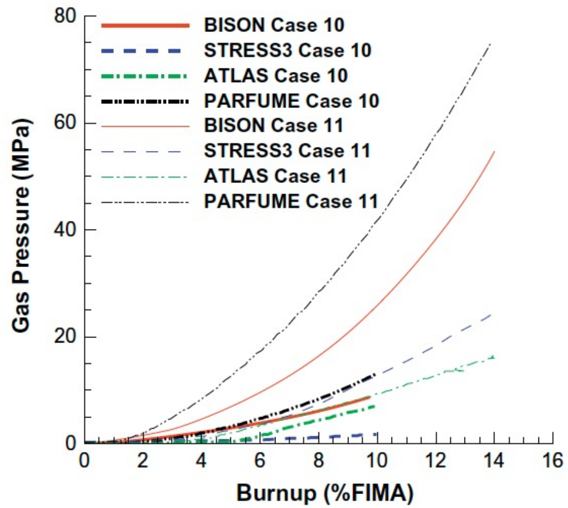
- Fuel performance models need to be able to account for numerous inter-related phenomena that affect temperatures, stresses, and failure of the material systems
- Must be multiphysics, incorporating thermodynamics, chemistry, kinetics, microstructure, mechanics, etc.

- Solid fission product swelling
- Gaseous fission product swelling
- Densification
- Thermal conductivity
- Fission gas release
- CO production
- Oxygen transport
- PyC layer dimensional change
- Kernel migration
- SiC failure
- Fission product attack

Fuel Performance Codes

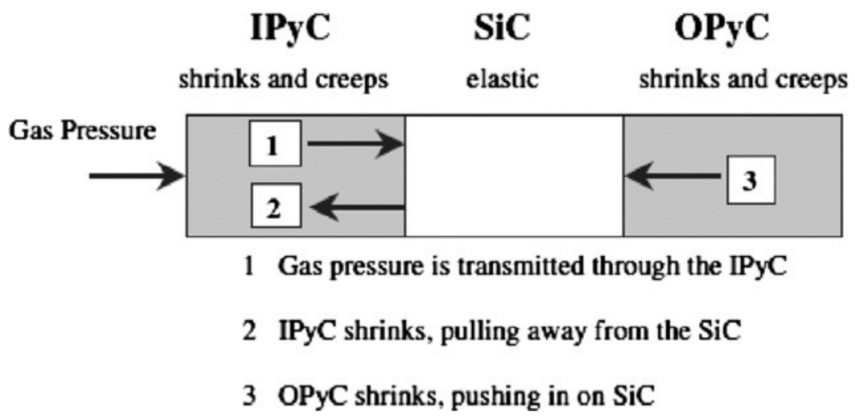
Code	PARFUME	PASTA	ATLAS	STRESS3	TIMCOAT	GA/KFA	JAERI
Developer	INL (US)	TU Delft (NL)	CEA (FR)	BNFL/NS (UK)	MIT (US)	GA/KFA (US)/(DE)	JAERI (JP)
References	[9,18,45–47]	[44,48,49]	[9,17]	[9,17,50]	[16]	[17,52]	[17,51]
Mission	NPR/AGR, NGNP	PUMA (EU)	FBR MOX	None specified	HTRs	Multiple	HTTR
Assumed geometry	Pebble bed, prismatic	None	None?	None	Pebble bed, prismatic	None	None
Pressure calculation	R-K EOS	R-K EOS	R-K EOS	Unknown	IGL	R-K EOS	IGL
CO production method	HSC-based yield	Custom (Nabielek?)	unknown	Martin	Karsten (KFA)	None, LEU, HEU	Proksch
Heat transfer calculation	1D finite difference with buffer/IPyC gap	THERMIX calculation with buffer/IPyC gap	Finite element	Unknown	Full-core then particle	Single irr. temp. used	Single irr. temp. used?
Phenomena modeled	Pressure, PyC IIDC, PyC irr. creep, thermal expansion, SFP swelling, FP diffusion	Pressure, PyC IIDC, PyC irr. creep, Thermal expansion	Pressure, PyC IIDC, PyC irr. creep, SFP swelling, GFP swelling	Pressure, PyC IIDC, PyC irr. creep, SFP swelling, SiC elasticity	Pressure, PyC IIDC, PyC irr. creep	Pressure, PyC IIDC, PyC irr. creep, SFP swelling	Pressure, PyC IIDC
Failure mechanisms modeled	PV, IPyC cracking, debonding, asphericity, SiC thinning, SiC thermal decomposition, kernel migration	PV, IPyC cracking	PV, IPyC cracking, debonding, asphericity	PV, IPyC cracking, debonding	PV, IPyC cracking via fracture mechanics	PV	PV
PyC shrinkage correlation	Custom	FZJ	Unknown	Custom	Unknown	Unclear	Unknown
PyC irr. creep coefficient $(\mathrm{MPa\ n\ m^{-2}})^{-1}$	$c = 5 \times 10^{-29}/$ $\nu = 0.5$ or $c = 4 \times 10^{-29}/$ $\nu = 0.4$	3.0×10^{-29}	Unknown	4.9×10^{-29}	CEGA function	2.0×10^{-29}	Unknown
Fission gas release model (s)	Recoil + booth	Modified booth (cyclic situation)	Unknown	Unknown	UT/KFA (booth-based?)	Booth	Booth (single species)
Displacement calculations?	Yes	Yes	Yes	Yes	Yes	No	No





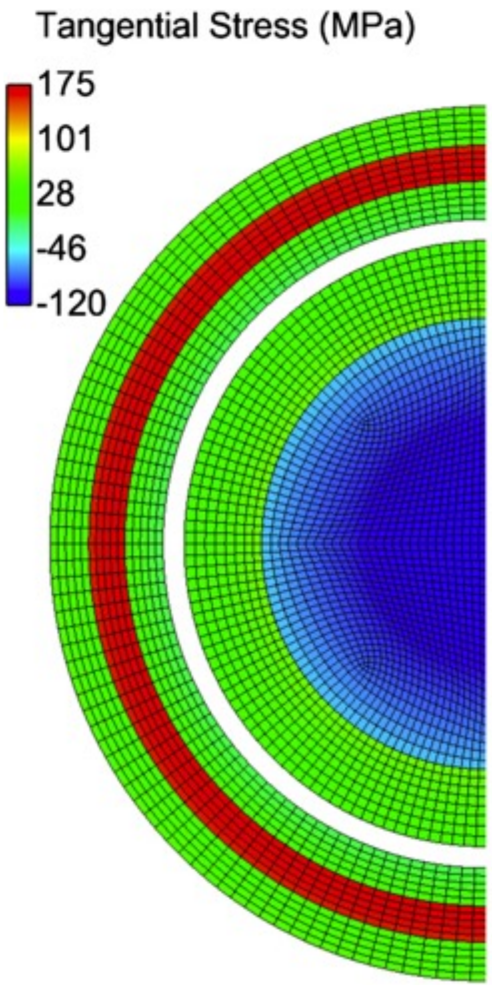
SiC Failure

- The dominant 1D failure mechanism for TRISO fuel particles involves pressure vessel failure, where the SiC layer develops a through-thickness crack resulting from a tensile stress that exceeds the fracture strength of the material
- Modern approaches to pressure vessel failure explicitly calculate separate stresses and through-layer crack probabilities for the SiC layer and each PyC layer



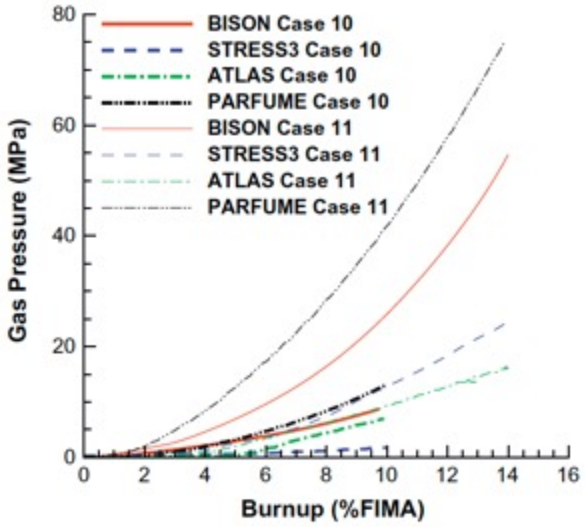
TRISO Stresses

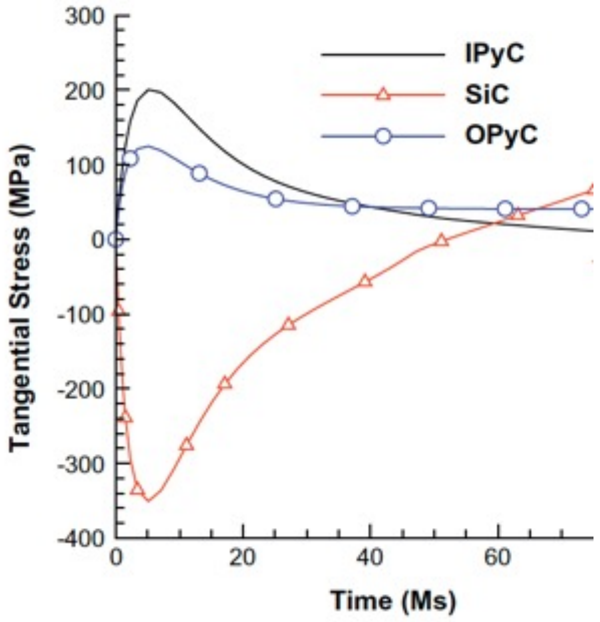
- The irradiation causes the kernel to swell and the buffer layer to shrink, thereby modifying the internal void volume available to the fission and reaction gases
- The porosity of the buffer results in the only force impinging on the three load bearing layers being the gas pressure on this surface
- If the IPyC and OPyC were absent, the gases produced in the fission process would simply stress the SiC layer and, if the pressure were high enough, cause it to fail
- However, these tangential stresses are reduced by the presence of the two PyC layers because; under unrestrained conditions they would shrink during irradiation



TRISO Stresses

- PyC shrinkage will be largely restrained by the SiC layer, and tangential stresses will be generated in both PyC layers and radial, inward acting forces imposed on the two surfaces of the SiC layer
- In practice, these tangential stresses in the two PyC layers will attain a quasi-equilibrium stress state because their shrinkage is counter-balanced by irradiation creep
- The SiC elastic modulus is at least an order of magnitude greater than that of PyC and thus, the dimensional changes and creep strains of SiC are comparatively much smaller than for PyC





Material Evolution

- Ideally, would be able to model and describe 3D modes of failure
  - Shrinkage cracks within the IPyC layer
  - IPyC/SiC debonding
  - Particle asphericity
  - Kernel migration
  - SiC coating thinning
  - Fission product attack

$$k_{\mathrm{fuel}} = K_{1d} K_{1p} K_{2p} K_{4r} k_{0,\mathrm{fuel}}$$

$$K_{1d} = \left(\frac{1.09}{\tau^{3.265}} + \frac{0.0643}{\sqrt{\tau}}\sqrt{T_{\mathrm{kern}}}\right) \arctan\left(\frac{1}{\frac{1.09}{\tau^{3.265}} + \frac{0.0643}{\sqrt{\tau}}\sqrt{T_{\mathrm{kern}}}}\right)$$

$$K_{1p} = 1 + \frac{0.019}{3 - 0.019\tau} \frac{1}{\left(1 + \exp\left(-\frac{T_{\mathrm{kern}} - 1200}{100}\right)\right)}$$

$$K_{2p} = \frac{1 - P}{1 + 2P}$$

$$K_{4r} = 1 - \frac{0.2}{1 + \exp\left(\frac{T_{\mathrm{kern}} - 900}{80}\right)}(1 - \exp(-\tau))$$

$$k_{PyC} = 10.98222\left(\frac{1 - P}{1 + 2P}\right) + 0.00444$$

$$k_{\mathrm{SiC}} = k_{0,\mathrm{SiC}}(3.91112 \times 10^{-2} \cdot \exp(2.24732 \times 10^{-3} \cdot T_{\mathrm{SiC}}))(1 - P)$$

$$k_{0,\mathrm{SiC}} = 42.58 + \frac{-1.5564 \times 10^4}{T_{\mathrm{SiC}}} + \frac{1.2977 \times 10^7}{(T_{\mathrm{SiC}})^2} + \frac{-1.8458 \times 10^9}{(T_{\mathrm{SiC}})^3}$$

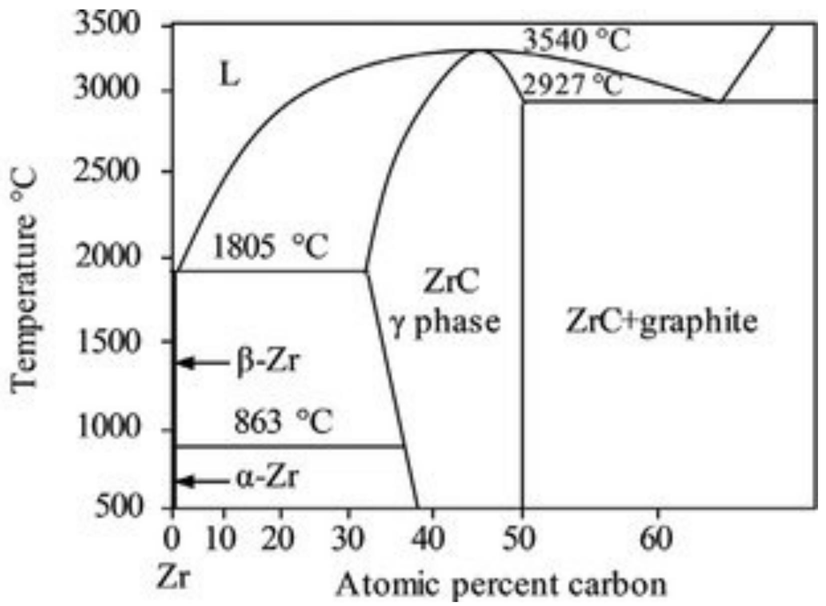
Data needs

- There are a number of areas where we lack requisite data, either behavioral or fundamental properties, in order to accurately describe TRISO particle evolution
- High-quality high burnup data on TRISO particles is generally lacking, which would enable benchmarking of fuel performance models
- Uncertainties and unknowns in fundamental material properties are also abundant, especially with regard to how properties vary as functions of temperature, fast fluence, and burnup
- There is significant uncertainty in the irradiation creep constant, the value of Poisson’s ratio in irradiation creep models, and the thermal conductivity of PyC
- Predictions for the chemical attack of SiC require effective diffusion coefficients of key fission products in various fuel materials, which are unknown
- Fuel properties are the least well understood, especially as the fuel evolves during operation

ADVANCED CONCEPTS IN TRISO

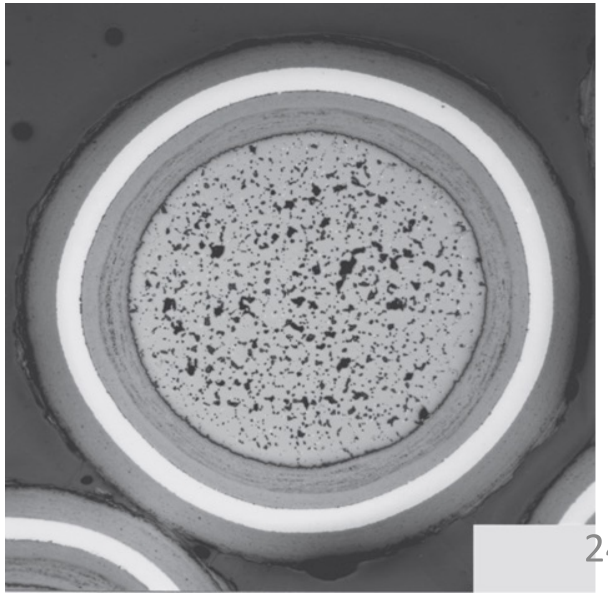
ZrC replacing SiC

- Zirconium carbide (ZrC) is a refractory and chemically stable compound, which melts eutectically with carbon at 3123 K
- ZrC can potentially improve the high-temperature stability, the resistance to chemical attack by fission products, and the retention of fission products
- However, ZrC is significantly less oxidation resistant than SiC



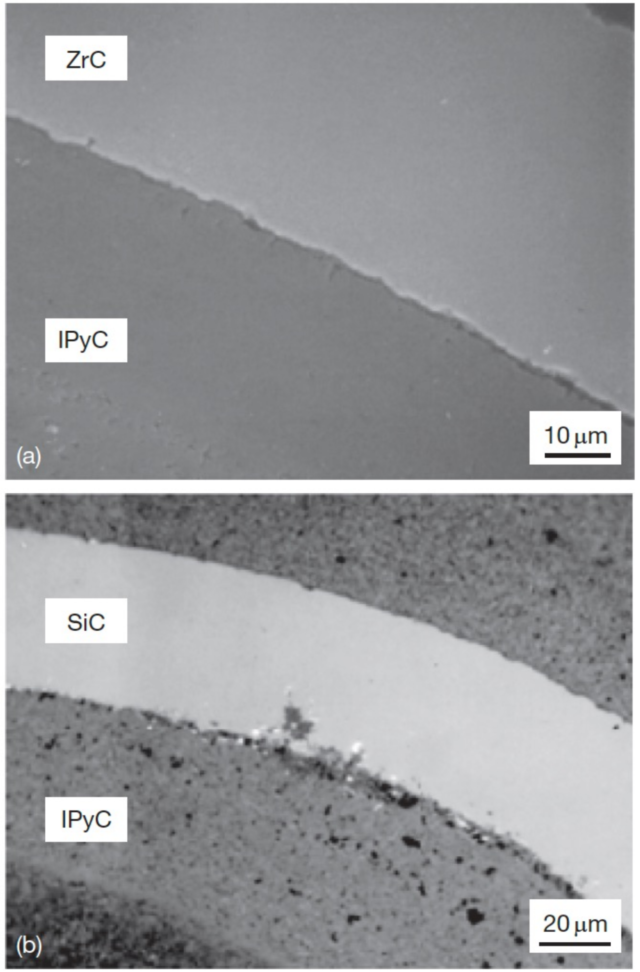
ZrC replacing SiC

- Four types of ZrC inclusions have been studied (1) ZrC-TRISO coated particles, (2) ZrC-TRISO type coated particles without OPyC layer, (3) ZrC-coated particles with ZrC-doped OPyC layer, and (4) ZrC coated particles with graded C–ZrC layer(s)
- All four types have been fabricated and irradiated
- Primary focus has been on option 1, however usage of ZrC started from option 2 in the 1970s
- ZrC-TRISO-coated UO2 particle after irradiation at 1673–1923K to 4.5% FIMA



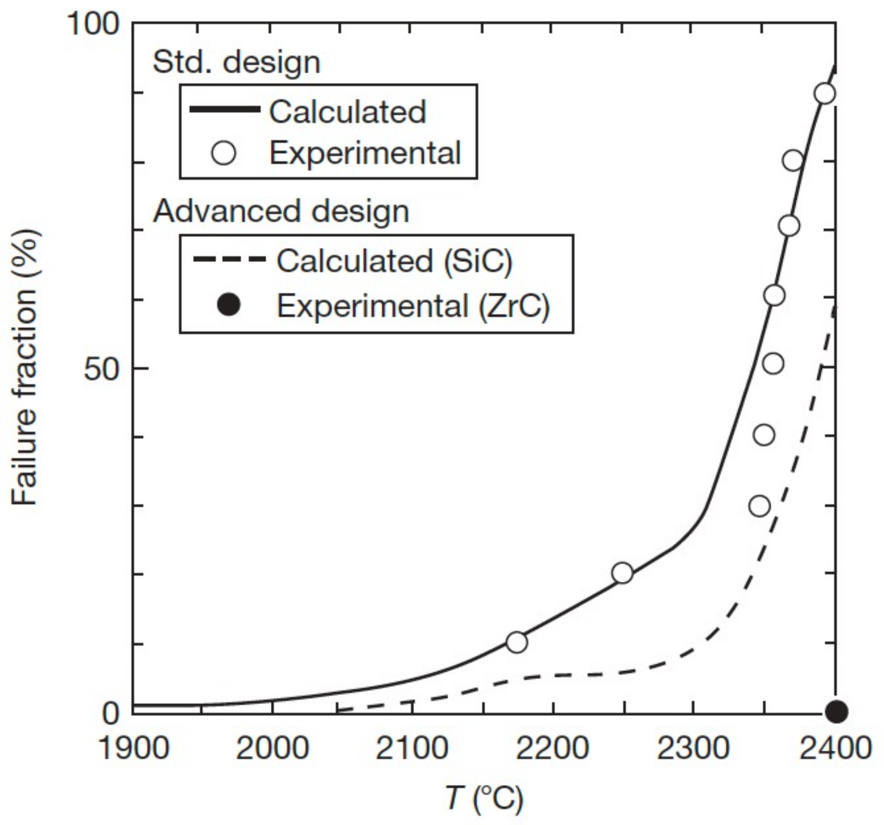
ZrC replacing SiC

- The improved performance of the ZrC coating layer against chemical attack by Pd has been demonstrated in both out-of-reactor experiments and irradiation tests
- $ZrPd_3$ can form in lab conditions, but insufficient Pd exists in fuel particles to cause formation and thus degradation
- ZrC does not seem to solve the silver problem
- Ruthenium release might be an issue, which it is not for SiC



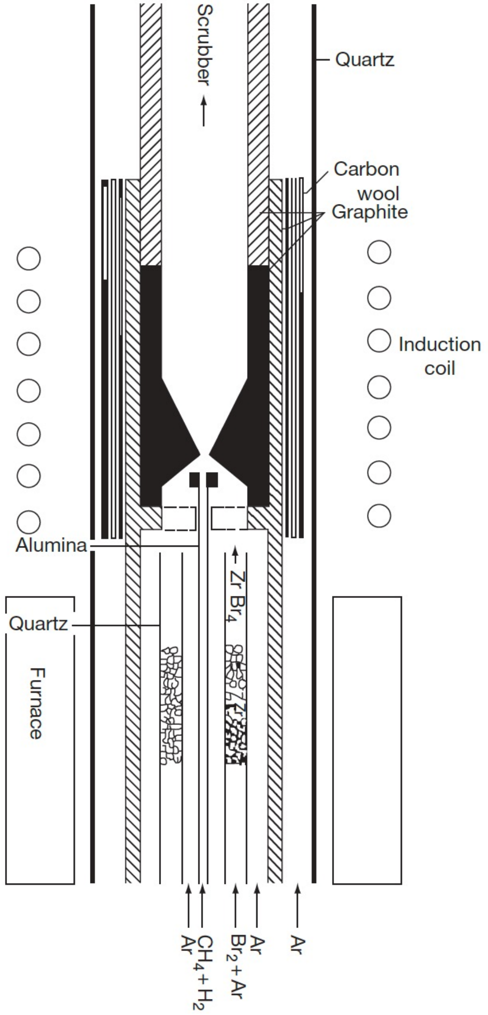
ZrC replacing SiC

- High-temperature behavior of ZrC is superior to SiC
- Right is a heating experiment on irradiated TRISO particles
- No ZrC failures up to 2400C, where an extended hold produced a failure
- Thus, better stability during fabrication, ability to operate at higher temperatures, better stability in off-normal temperature spikes, better fission gas retention at high temperature



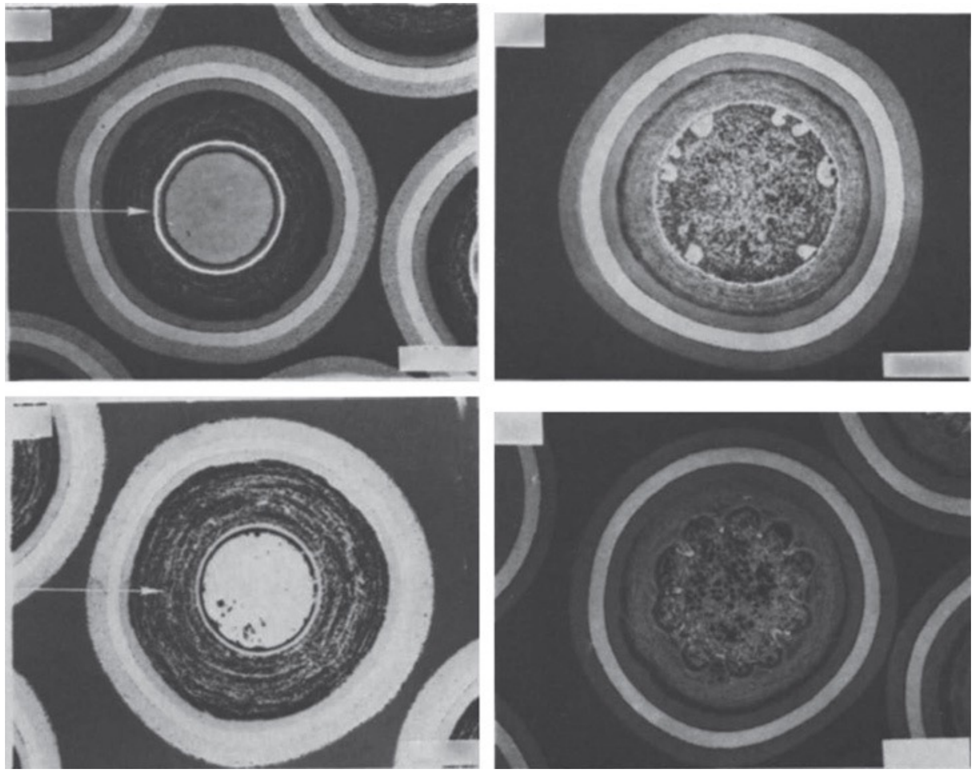
ZrC Fabrication

- Fabrication of ZrC coatings has been established in a lab setting
- Not yet an industrial process, thus not feasible for large scale production



Non-fissile Carbides in Fuel

- To inhibit the buildup of CO gas in UO_2 fuel kernels, UC_2 is often added to getter oxygen and reduce the oxygen potential
- Similar results can be achieved by the addition of nonfissionable carbides, such as SiC or ZrC around fuel kernel
- Additional benefits might be gained by placing getters of this type at the outer boundary of the UO_2 kernels

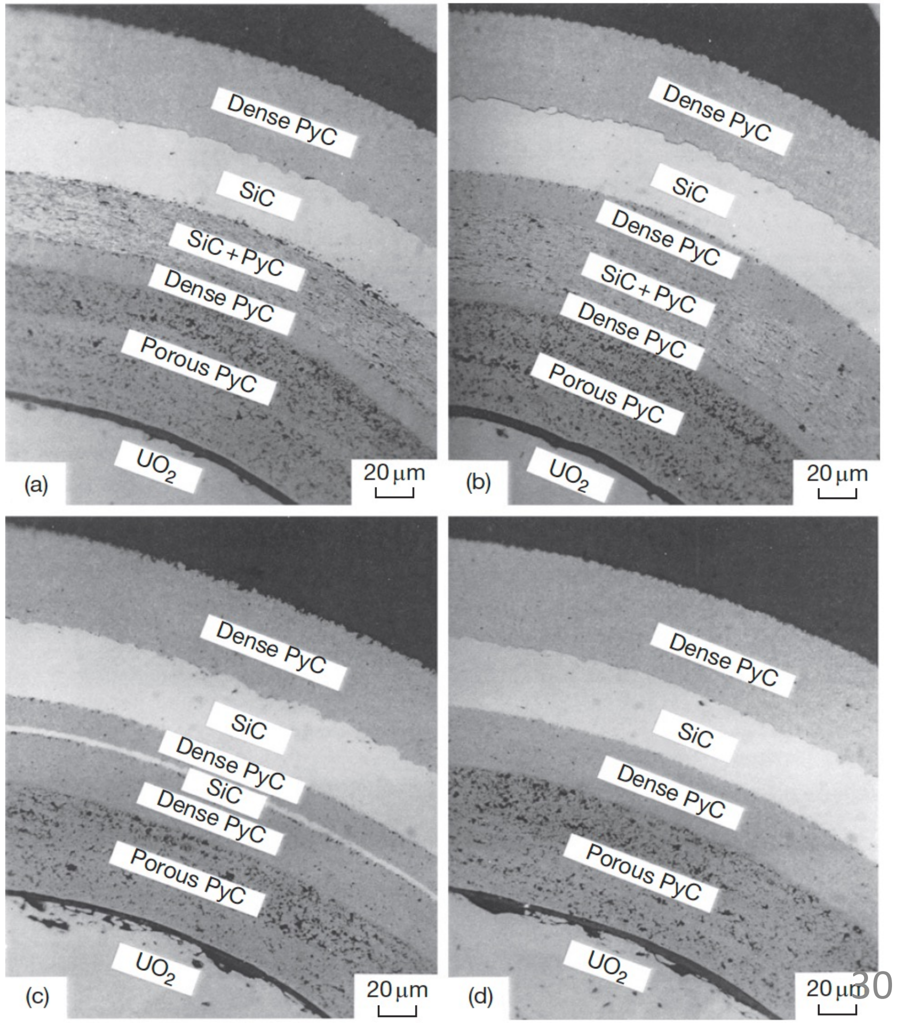


Non-fissile Carbides in Fuel

- Introduction of ZrC around fuel kernel getters released oxygen
- This seems to inhibit fuel kernel migration
- If the layered ZrC is intact, can inhibit fuel swelling as well, potentially reduced stresses on outer layers
- Have performed limited irradiation experiments with no failures
- If the layered ZrC is intact, no release occurs for Ag and Eu
- Release occurs for both species under same conditions for nominal TRISO and for ZrC dispersed in buffer
- An intact ZrC layer seems to be necessary for explaining the outstanding retention of these particles, in addition to the benefits of O gettering

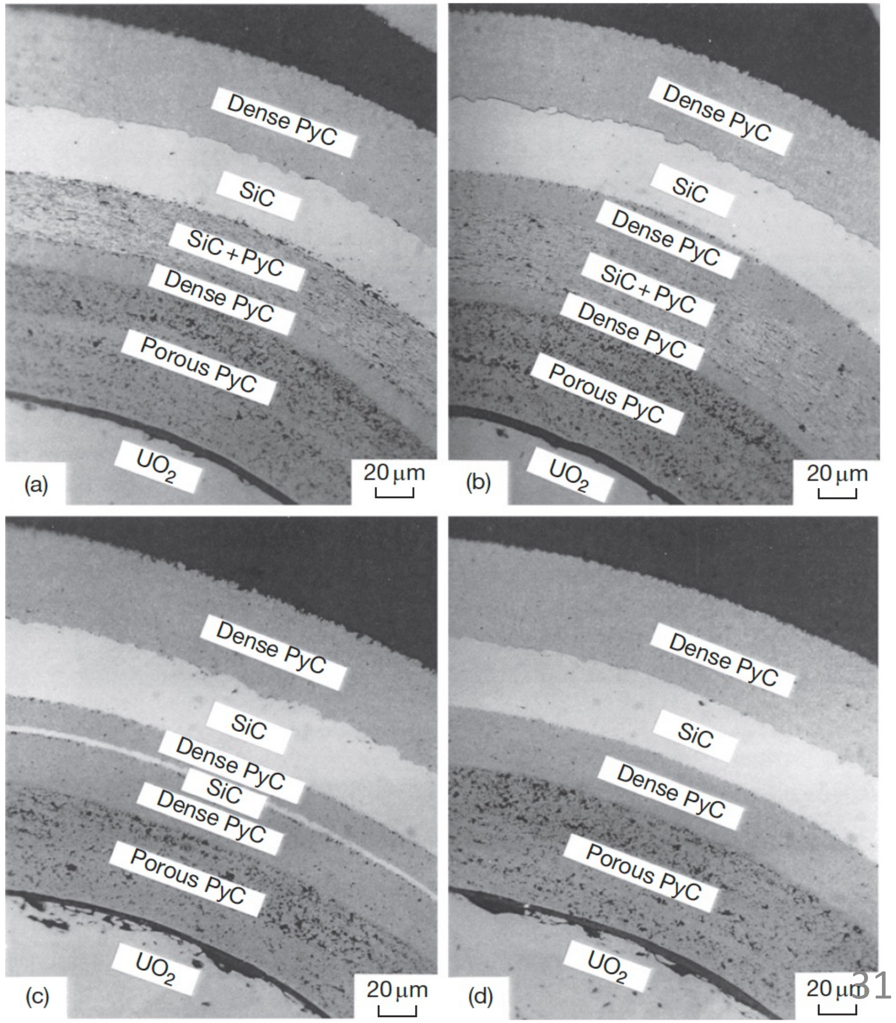
Alternate Layering

- Two methods are available to prevent the corrosion of the SiC layer: (1) to keep the fission products within the fuel kernel and (2) to make a barrier to the diffusion of the fission products to the SiC layer
- Can potentially add a layer that traps palladium by chemical reaction inside the SiC layer of the TRISO coating
- Can add either an additional SiC layer, or a SiC/PyC composite layer



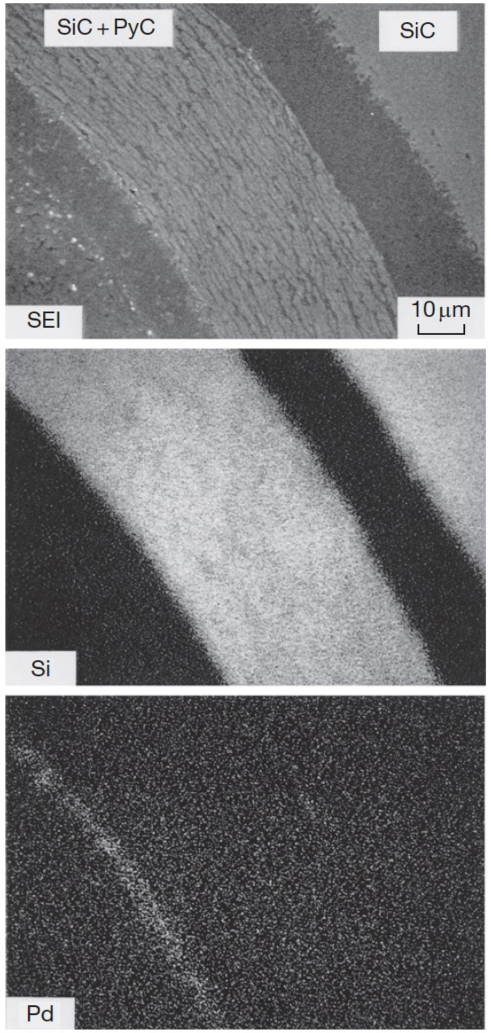
Alternate Layering

- The type-A coating has an additional layer of SiC+PyC adjacent to the inside of the SiC layer; the thickness of the IPyC layer can be reduced
- The type-B coating has the dense PyC layer between the SiC+PyC and SiC layers; the intermediate dense PyC layer will interrupt extension of the corrosion zone from the SiC+PyC layer to the SiC layer
- In the type-C coating, SiC is used for an additional layer; SiC layer will react with fission products and PyC will interrupt corrosion of outer SiC



Alternate Layering

- Irradiation of these advanced coatings was performed in the JRR-2 reactor
- Ceramography revealed no crack in the advanced coating layers or in the TRISO coating layers
- All three types trapped Pd at the inner SiC-based barrier, preventing corrosion of the outer SiC
- At high heating, a PyC layer between the outer SiC and the inner SiC-based layer proved more effective at fission product retention
- Also, the SiC/PyC layer proved more effective than pure SiC
- No new technology needed to fabricate these particles



Type B Coating

Summary

- Fuel Failure mechanisms
  - design and proper fabrication can limit most failure mechanisms
  - as we drive toward higher burnup, additional problems may arise
- TRISO Fuel performance
  - there are a number of fuel performance codes
  - data needs, and the evolutionary complexity of particles hold back more sophisticated modelling
- Advanced concepts in TRISO particles
  - ZrC instead of SiC
  - ZrC/SiC as a layer around kernel
  - alternate layering of SiC/PyC

QUESTIONS?

Quiz Upcoming!

- Will cover TRISO particle-based fuels
- Quiz will be on Sept 7th
  - Will keep it here unless there is a strong will to move it to the 2nd
- Open ended questions covering the concepts presented in class