Advanced Fuels Campaign

Fundamentals of Nuclear Fuel

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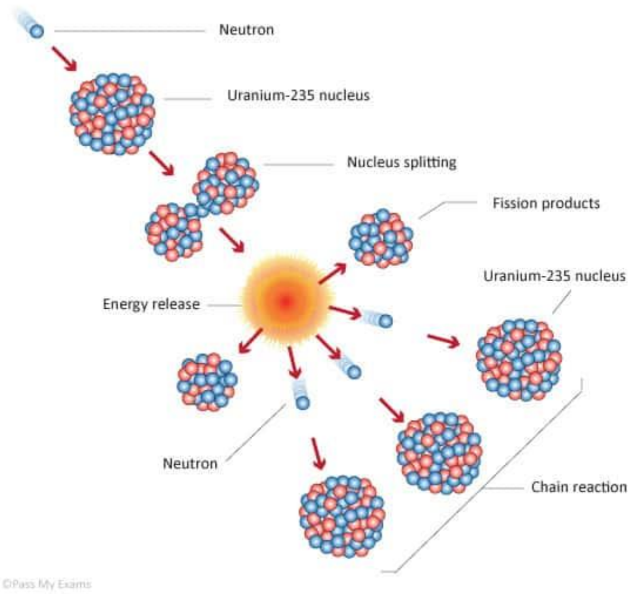
Nuclear Physics 101

- Free neutrons are eventually either absorbed or scattered by atomic nuclei
  - The probability of these reactions depends on both the target nuclide and the neutron energy
  - Atoms which strongly absorb neutrons, fission, and release more neutron are called fissile isotopes or **“fuel”** (U, Pu)
  - Atoms which strongly absorb neutrons and transmute without releasing more neutrons are called poisons (B, Cd, Gd)
  - Neutrons born of fission are considered “fast” neutron (~2MeV)
  - Fast neutrons are less likely to be absorbed, but more likely to knock atoms out of their crystal lattice (atom displacement)
  - Fast neutrons can be scattered or “moderated”, usually by small nuclei (H, C, Be) to become lower energy “thermal” neutrons (~0.025 eV)
  - Thermal neutrons are more likely to absorbed and cause transmutation
- A self-sustaining fission chain reaction is the whole point
  - Releases heat, more neutrons, and leaves behind radioactive fission products

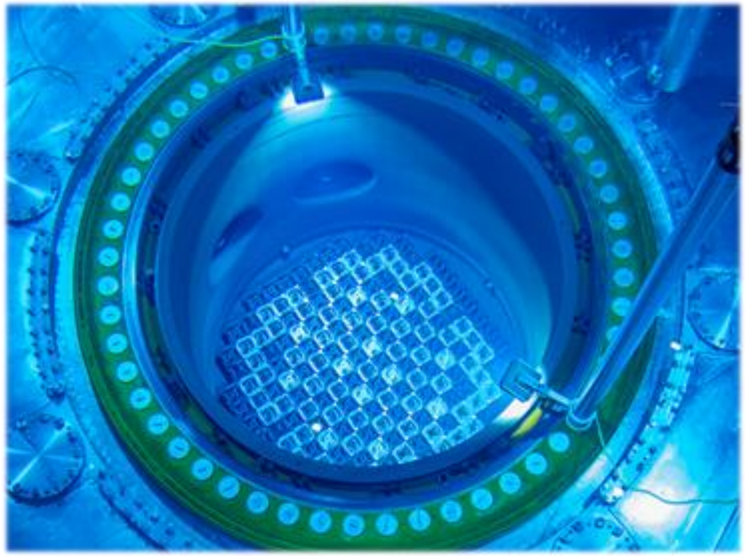


The Role of Nuclear Fuel

- There are two useful products of nuclear fission/reactors:
  1. Release Energy: For electrical energy production, process heat, etc.
  2. Release Neutrons: To test nuclear materials, transmute isotopes, create neutron beamlines, etc.
- Nuclear fuel must do three things to support nuclear reactors
  1. Sustain Chain Reaction and Fuel Cycle
  2. Transfer Heat
  3. Retain Radioactive Fission Products

Operators, customers, & stakeholders care most about efficiency of these functions on an expected normal day

Regulators care most about failure probability and severity of these functions on a postulated bad day



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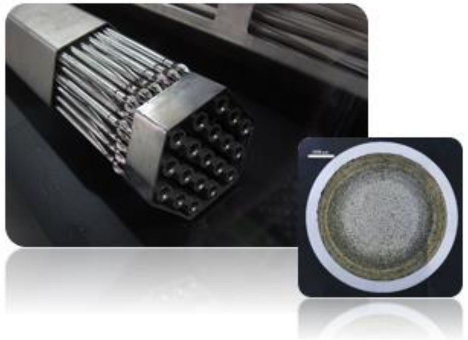
Some Vocabulary

Most fuel systems are a two-part system

1. The material bearing fissile, fissionable, and fertile actinides:
   - Often categorized by material type: Ceramic (UO_2), Metallic (U-Zr), Intermetallic (U_3Si_2), Coated Particle (TRISO), or liquid (molten salt)
   - Referred to as fuel material, fissile phase, and simply as "fuel"
2. The hermetic thing that contains radionuclides:
   - Various designs/dialects includes "cladding" (MTR, SFR, LWR), "coatings" (TRISO), "canisters", "sheaths" (Canadian word for cladding), or reactor vessel/piping (MSR)
   - Most often metallic materials, but sometimes ceramics/composites

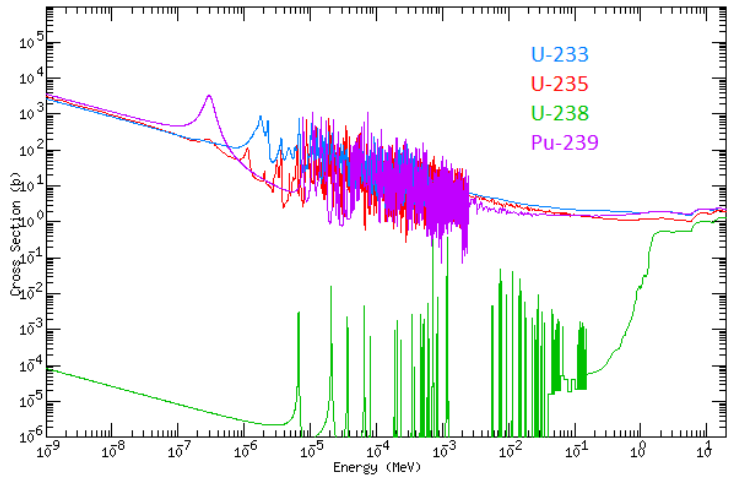
- The fuel and cladding together create the "fuel system"
  - Confusingly is also called "fuel"
  - Less confusingly also referred to as description of assembly type:
    - Fuel rods, fuel pins, fuel plates, fuel bundles, fuel assemblies, fuel elements, fuel pebbles, fuel compacts

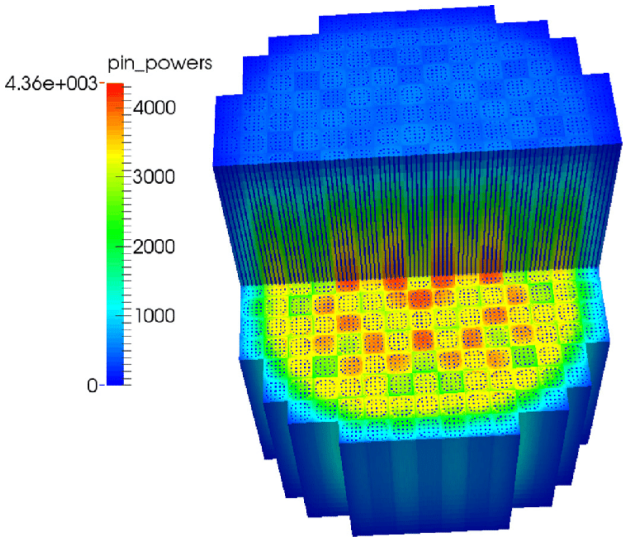





Fuel Performance for the Nuclear Physicist

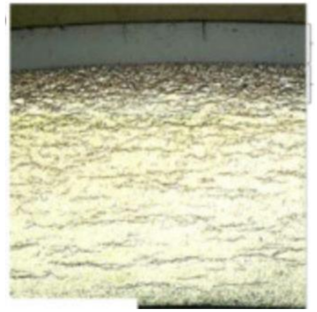
- Most neutrons are released immediately by fission as “prompt neutrons”
  - If there were only prompt neutrons, then the only thing reactors could do would be an amusing but useless power pulse
- Some neutrons (<1%) are released slightly later as “delayed neutrons”
  - The rates at which neutrons are born (from fission) to those lost (by absorption and leakage) must be controlled within the delayed neutron fraction for a reactor to have stable power
  - The composition and physical arrangement of fuel, moderator, and poison are crucial in getting it right
  - Nuclear data cross sections describe probabilities of fission, capture, and scattering depending on isotope and neutron energy, and they are not simple
  - Neutron transport codes rely on nuclear data
  - Neutronic models of nuclear reactor must be validated against system scale physics experiments
- Nuclear physicists’ favorite test reactors are “zero-power reactor” for well characterized criticality experiments

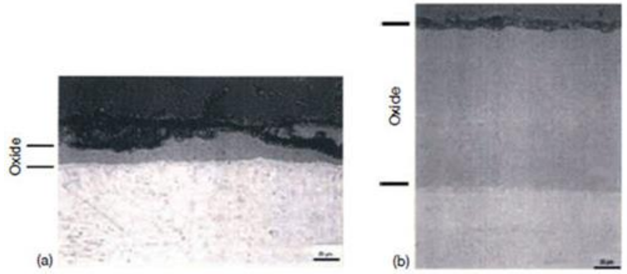




Fuel Performance for the Corrosion Scientist

- Reactors usually have a fluid convection system to remove heat from core
  - Cladding coolant interactions crucial considerations
  - Corrosion consumes cladding thickness thus weakening mechanical function
  - Build up of surface oxide and other deposits (e.g., CRUD) can impede heat transfer and cause fuel temperature to increase
  - Diffusion of reaction products can embrittle cladding (e.g., hydrides)
- A reactor dependent problem
  - Light water reactor fuel development is a multidecadal story about getting zirconium alloys and pressurized hot water chemistry to cooperate
  - With proper control of coolant contaminants, helium and sodium cooled reactors can have very benign corrosion behaviors
  - Some other proposed reactor coolants (molten salt, lead-alloys, supercritical water) are expected to put corrosion centerstage for fuel performance
  - Non-nuclear corrosion testing useful, but in-reactor phenomena (e.g., radiolysis) can dramatically affect behavior
- Corrosion scientists' favorite test reactors are material test reactors with independently controllable flowing coolant loops

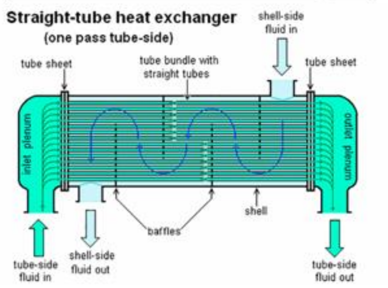




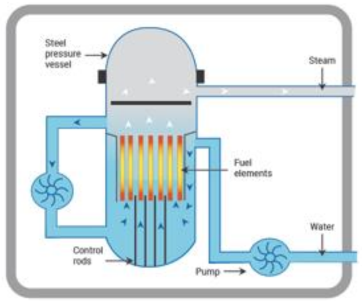
Fuel Performance for the Mechanical Engineer

- Not surprisingly, mechanical engineers also get rather excited about nuclear fuel
  - Fuel manufacturing methods deeply intertwined with its behavior
  - Cladding has key roles in retaining fission product swelling/pressure & maintaining fuel assembly structure
  - Fuel assembly materials/geometry defines thermal hydraulics of core
  - Complex multiphase thermal hydraulics behaviors in some coolant (especially water)
- Mechanical engineers' favorite test reactors are material test reactors and transient test reactor with all kinds of adaptable, versatile, and extreme condition test capabilities

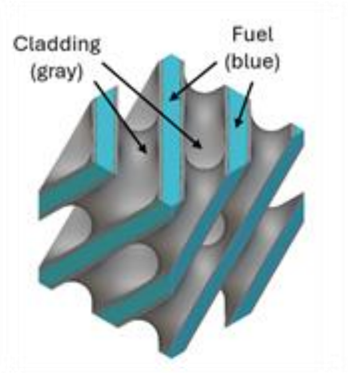
1950's heat exchanger



1950's reactor

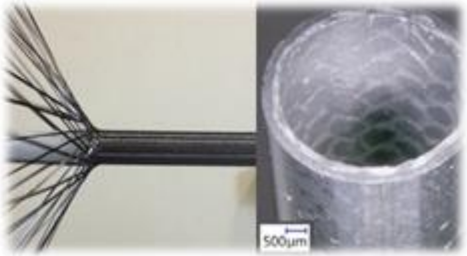


Advanced AM heat exchangers



Concept being researched using AM for nature-inspired nuclear fuel

Advanced LWR Bundle Nozzle Made by AM



Ceramic fiber composite cladding being researched

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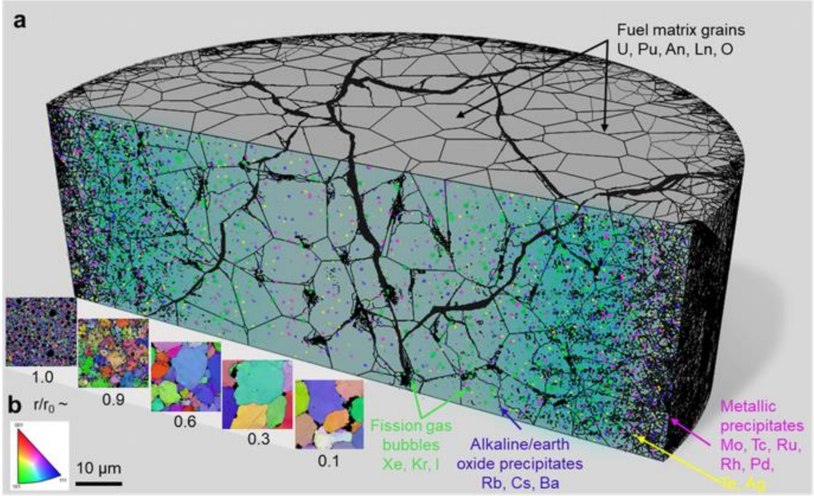
Nuclear Fuel Performance for the Material Scientist

- Material science is probably the most represented discipline in nuclear fuel performance
- Nuclear fuel is interesting stuff
  - Not the atoms you started with: Fission products and transmutation nearly yield “one of everything” from periodic table
  - High temperatures and thermal gradients to drive diffusion alongside dissimilar material interfaces to drive interactions
  - Fast neutron damage and fission gas production constantly battering microstructures, causing volumetric swelling, and messing with creep rates
- Material Scientists’ favorite test reactors are material test reactors
  - They love material test reactors almost as much as microscopes!



The Basic of Fuel Material Performance

- Nuclear fuel material performance usually falls into two battlegrounds*
  1. Fuel-cladding interaction
  2. Fission gas behavior

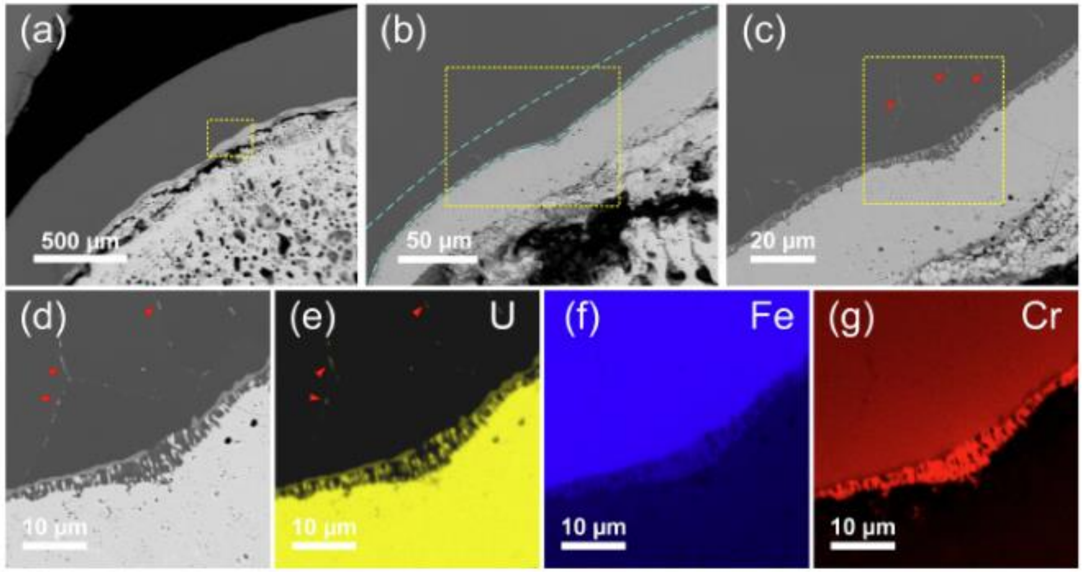
  * Setting aside radiation damage cladding behavior (assuming well covered by preceding talks)

- Fuel-cladding interaction, dissimilar material interfaces
  - Chemically aggressive fission product species corroding the cladding from the inside
  - Complex thermo-mechanical stress and fuel swelling creep interactions, especially ceramic fuels in contact with metallic claddings

- Noble gases are boring (Xe, Kr), so why are they so interesting as a fission product?
  - Cannot “tied up” in compounds, always a free non-condensable monoatomic gas
  - If they stay in the fuel material, they cause it swell and push on the cladding
  - If they get released from the fuel material, they apply internal pressure on cladding
  - Hence, fission gas behavior is at the heart of fuel performance

Fuel Cladding Chemical Interaction in Metallic Fuel

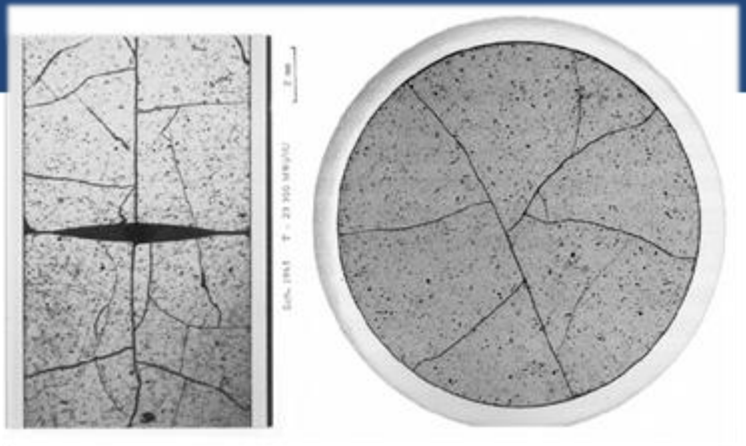
- Uranium alloy metallic fuel preferred in sodium fast reactors for its high fissile density and thermal conductivity
- Iron-based claddings needed for strength at temperature and resistance to fast neutron damage
- Lanthanide fission products migrate through toward cladding and react, essentially corroding the cladding from the inside out
- Understanding behavior crucial to determining temperature and burnup limits to ensure fuel performs reliably



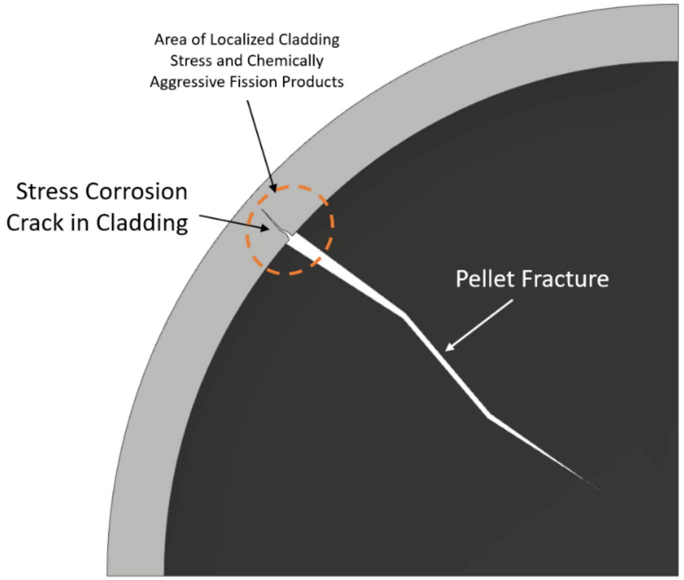
FCCI Microscopy in U-10Zr metallic fuel
https://doi.org/10.1016/j.jnucmat.2020.152588

Summary of Pellet Cladding Interaction in LWRs

- Thermal gradients causes radial fractures in UO_2 on first rise to power
- External coolant pressure on Zr-alloy cladding “creep down” closes pellet-cladding gap by mid-life burnup
- From this state, power ramps (planned power maneuvers) create a stress state susceptible to stress corrosion cracking
  - Chemically aggressive fission products (e.g., iodine) concentrates phenomena near pellet fracture-cladding interfaces
  - Can penetrate through cladding to create pin hole “leaker rod”
- PCI ramp testing in material test reactors used to investigate behavior
  - Irradiation at low power long enough for cladding creep down
  - Followed by power ramp and hold, repeat until leak is detected



Fractured UO_2 LWR Fuel
Michel et al. Eng. Frac. Mech., 75, 3581 (2008)



Pellet Cross Section Illustrating PCI Behaviors (Quarter Section Shown)

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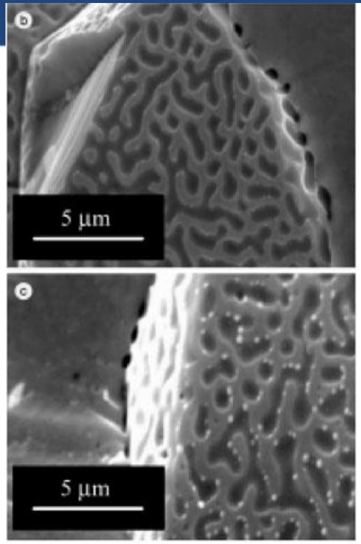
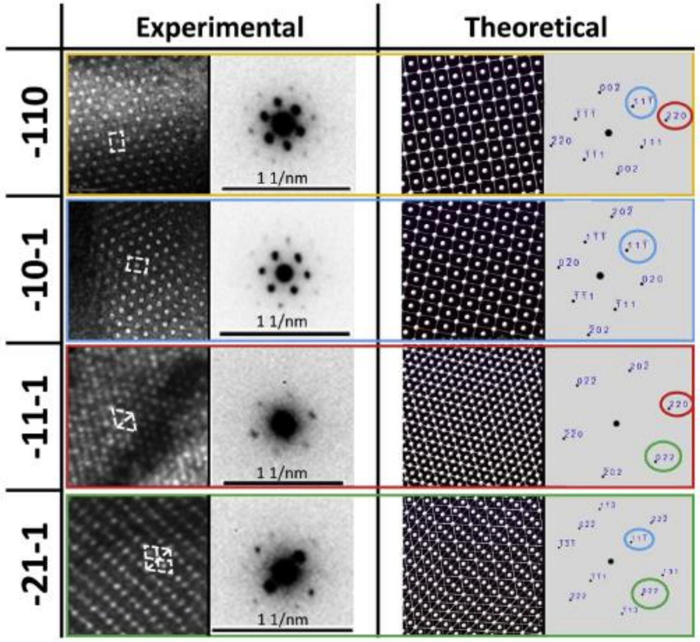
Let's talk about fission gas

- Early life: Fission gas atoms recoil microns in fuel material and get "stuck"
- As gas inventory grows, more gas atoms collect in nano bubbles, finding stable homes in the microstructure
  - Ordering into beautiful patterns in fuels with ordered crystal structures (U-Mo at MTR temperatures, UO_2 at LWR temperatures)
  - Or just chaotically "bubbling around" in glassy/amorphous microstructures (U_3Si_2 at MTR temperatures, U-Zr at SFR temperatures)
  - Geometric swelling starts to manifest
- As gas bubble pressure grows, so does the driving force for bubble growth and migration
  - Bubbles often find each other and coalesce and drive toward a free surface to get released into the cladding
  - Gas diffusion through the microstructure tends to follow thermal gradients, larger bubbles tend to collect at grain boundaries first
  - Fractures in some fuel systems (e.g., pelletized ceramics) reduce gas diffusion length to a free surface
  - In ductile fuel systems (e.g., metallic) bubbles interconnect and form "wormholes" to the surface



Intragranular fission gas bubbles in UO2
https://doi.org/10.3389/fenrg.2022.766865



Comparison of measurement and simulated fission gas lattice in U-Mo,
https://doi.org/10.1016/j.jnucmat.2019.151947

Fission Gas Management Strategy

Resist, 0% release to cladding

Comply, 100% release to cladding

MTR	VHTR	LWR	SFR	MSR

- Some things that increase FGR in solid fuels
  - Burnup (or fission density) → more fission gas inventory
  - Isotope (Pu-239, U-233 have different higher fission gas yields than U-235)
  - Increasing temperature (gas micro-bubble pressure goes up, and diffusion rates increase as atoms in the fuel material “wiggle” more)
  - Amorphous (gas atoms diffuse more easily through irregular “messy” non-crystalline microstructures)
  - Time under irradiation (neutron atom displacement, fission, etc. are ”mixing” the microstructure, helping gas atoms move around)
- In liquid fuels virtually all fission gas comes out and the piping/vessel system must deal with it

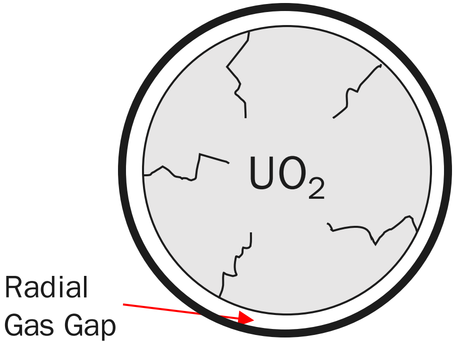
Fission Gas in Rod Type Fuel

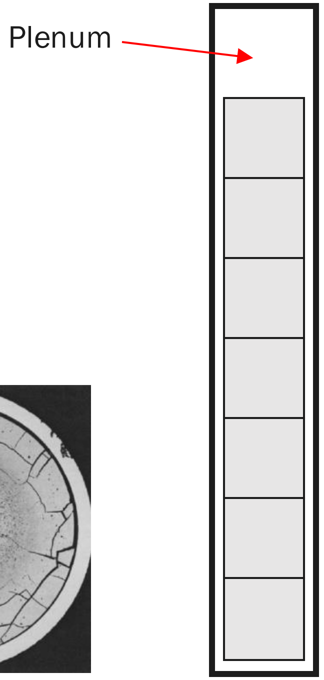
LWR: Fresh state gap, plenum, pellet porosity (1-3%), initial plenum pressure, cladding properties/geometry all define the capacity for accommodating FGR

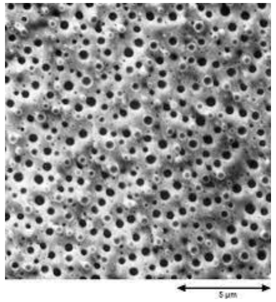
High temperature ceramic, well-order crystal structure, relatively low ^{235}U density (5% enrichment) ⟶ Relatively low FGR (~10-20%)

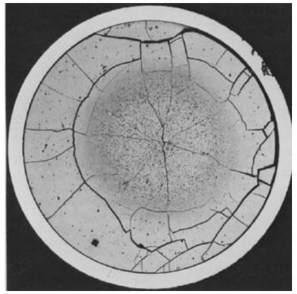
Gas diffusion through microstructure to surfaces/fractures permits a slow-and-steady release to modestly-sized plenum

General idea is to maintain $P_{outer} > P_{inner}$







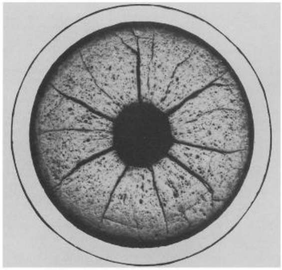


SFR: Higher fission rates and coolant temperatures

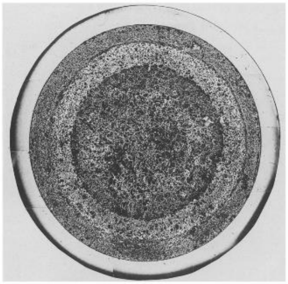
Extreme thermal gradients drive porosity migration to centerline in oxide systems (e.g., SFR-MOX), pathway for FGR to plenum

Fresh state 25% free volume designed into metallic systems, swells like bread until pores fully interconnect, then swelling ceases and FGR releases to plenum through "open-cell foam" microstructure

Larger FGR fractions (~80% in metallic fuel) require longer plenums (thankfully the cores are much shorter and fast spectrum neutronics allow cladding alloys to be stronger (Fe-based)



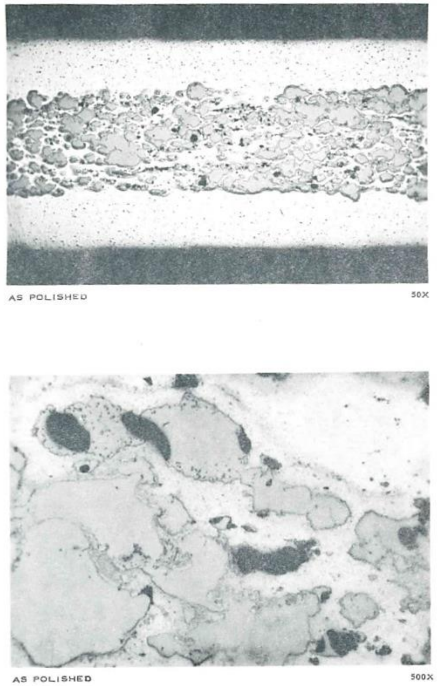
SFR-MOX



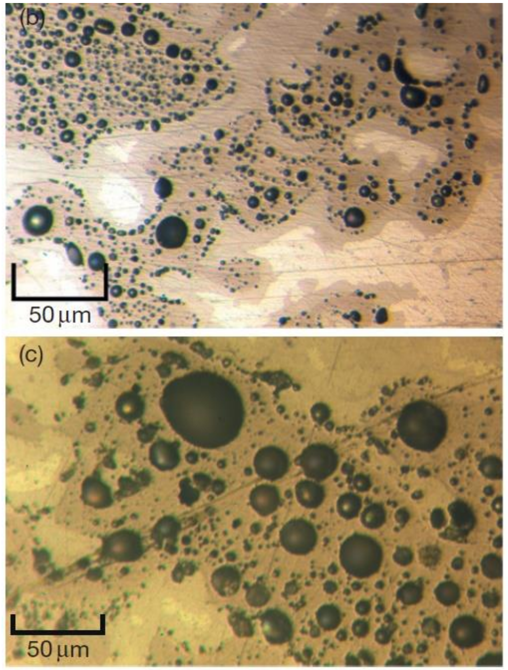
SFR-Metallic Fuel

Fission Gas in MTR Plate-Type Fuel

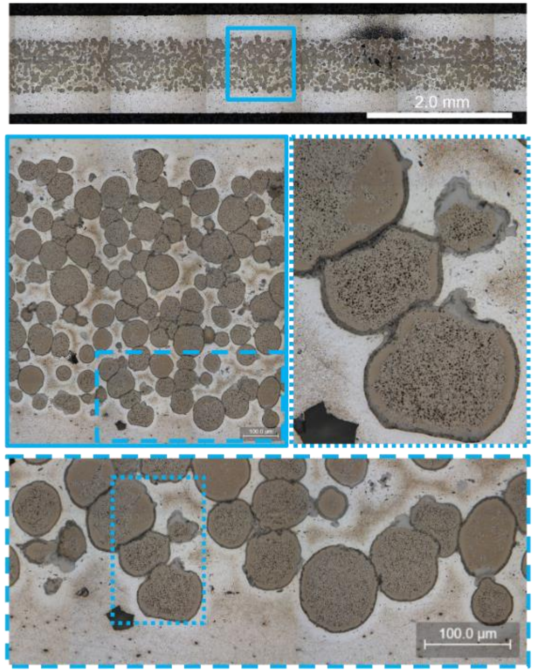
- 0% FGR very difficult to achieve in useful burnup/temperature regimes
  - Most fuel systems must have some free volume to accommodate gas
- Rectangles make terrible pressure vessels, must retain nearly all fission gas in the microstructure (never grow up, stay an adolescent fuel forever)
  - Composite architecture ("dispersion" fuel) helps provide some initial porosity (~10%) and holds composite together in a 3D web of metal
  - But introduces new problem of managing particle matrix chemical interaction



UAlx in Al "aluminide" fuel (ATR fuel), a fuel system that succeeds by being mostly aluminum



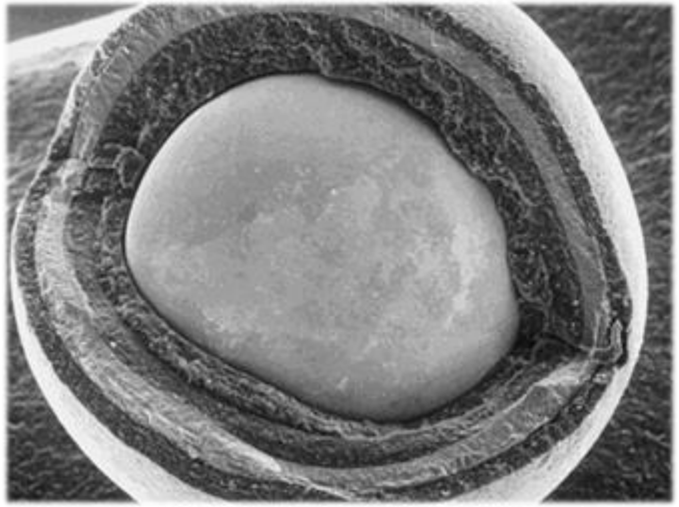
U_3Si_2 in Al "silicide" fuel, an amorphous MTR fuel system that succeeds [barely] by managing fission rate and temperature

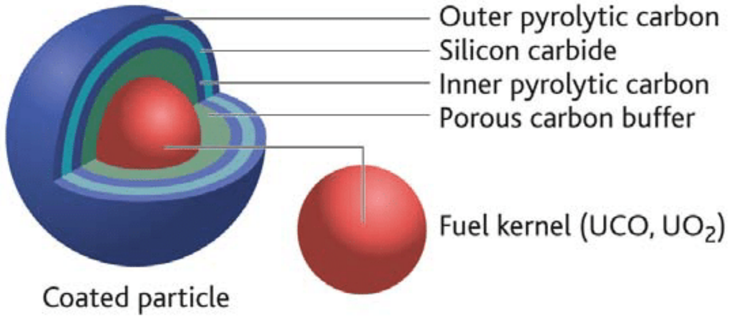


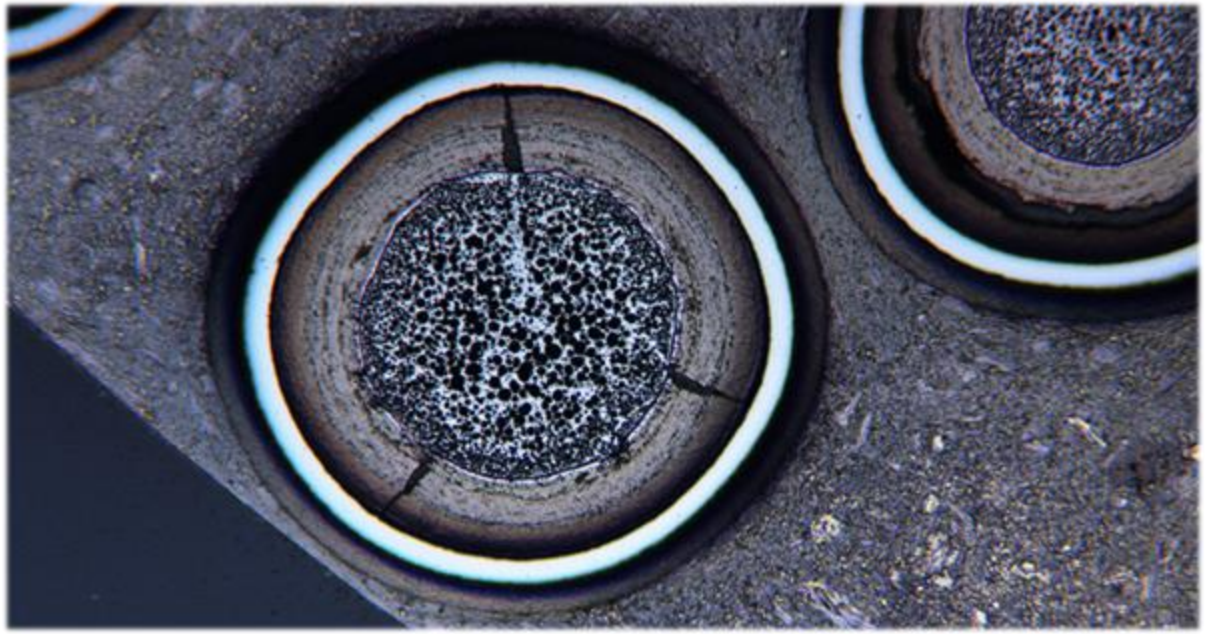
U-Mo in Al dispersion fuel, a high-density fuel system that succeeds by managing matrix chemical interaction

Fission Gas in Coated Particle TRISO Fuel

- Uranium Oxide/Carbide ceramic kernels (UCO)
  - Small kernel size (~500 µm) dispersed in high thermal conductivity matrix (graphite) reduces thermal gradients/cracking in fuel
  - Despite LWR-like FGR microstructural behavior, increased enrichment (20%) and high burnups → Particles end up virtually decimated by fission gas in late life due to high fission density
  - Innermost porous carbon buffer coating provides free volume
  - SiC and pyrolytic carbon coatings function like cladding (tiny little spherical pressure vessels) to retain FGR







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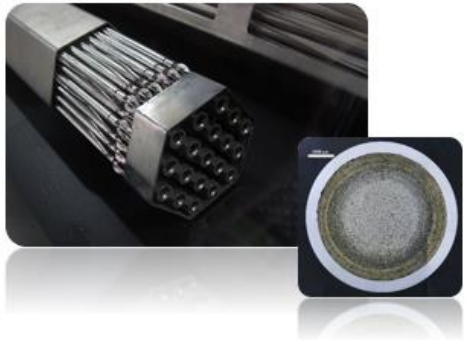
Conclusions

- Nuclear fuels are the heart of a reactor, alongside coolant and spectra, constitute the most important decisions a reactor designer can make
- Several disciplines and phenomena significant in fuel performance, test reactors make crucial contributions to developing nuclear fuels
  - Oldest and most well-known fuel system (UO_2 in Zr-alloy) still being actively researched to unleash new gains
  - Near future deployment of TRISO and U-Zr metallic fuels only possible because of DOE's investment in understanding fuel behavior, now starting the journey toward their maximum potential
  - Relatively little data for other fuel systems and innovative concepts, data will be key to unlocking their performance potential
- Irradiation testing crucial to understand nuclear fuel performance, better optimize behavior, and maximize reactor performance



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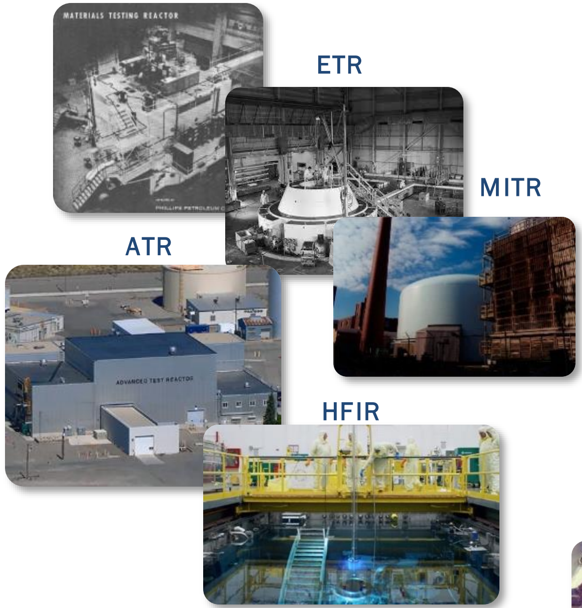
A Foreshadow to Tomorrow's Session: Test Reactors

The Past

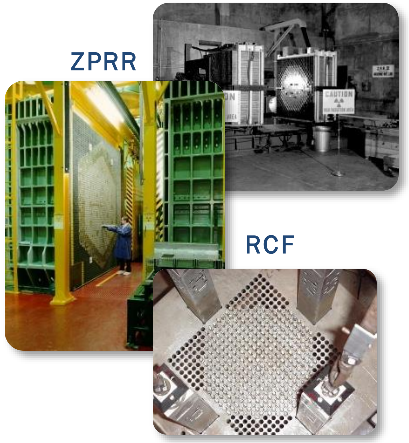
Thermal Spectrum

MTR
ETR
MITR
ATR
HFIR



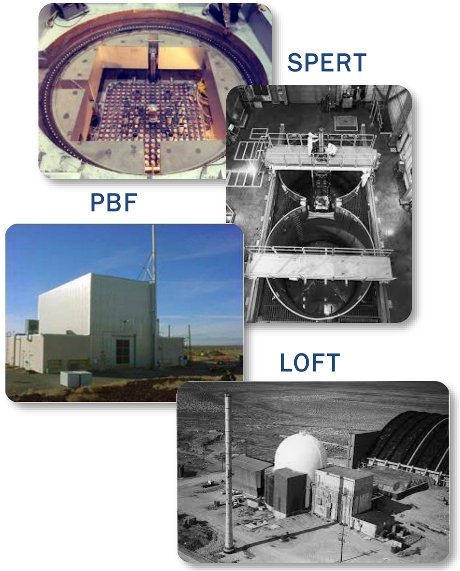
Critical Facilities

ZPR-3
ZPRR
RCF



Transient

TREAT
SPERT
PBF
LOFT



Fast Spectrum

EBR-II
FFTF

The Future

ATR

-High flux, large volume
-Water loops for fuels testing
-Unique dynamic testing

MITR

-LWR-like flux
-Cladding corrosion testing in water loops

HFIR

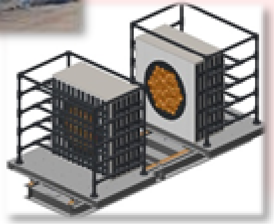
-Very high flux on subsize tests
-Unique capabilities for accelerated testing

TREAT

-Extreme power maneuvers
-Unique fuel safety testing abilities

SPARC

-Reactor scale critical tests
-Project recently initiated with near future timeline



Fast Spectrum Test Reactor

-High fast flux
-Crucially needed, timeline TBD