Nuclear Fuel Performance

NE-533

Spring 2025

Exam 2

- Grading catchup:
  - Exam 2 avg: 86.4; Curve of 5 pts.
- MOOSE grading:
  - sent out today
- Paper grading:
  - sent out today
- Let me know of any comments or questions on any of this

MOOSE Project Notes

- Two ways of doing the meshes, both work
  - Single mesh, divide into subdomains
  - Three separate meshes, stitch meshes together, define subdomains
- First way, need to be careful in subdividing, as you can specify multiple blocks at same point
- Second way can give you more mesh flexibility and defines points on the block transitions; may help with convergence
  - can try, but not needed
- Can try adaptive timestepping; can check for steady-state with transient
- Overall, everyone did very good on the MOOSE project

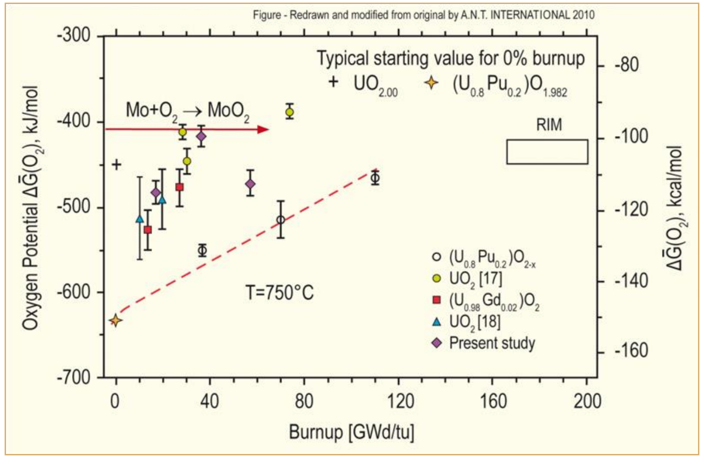
Last Time

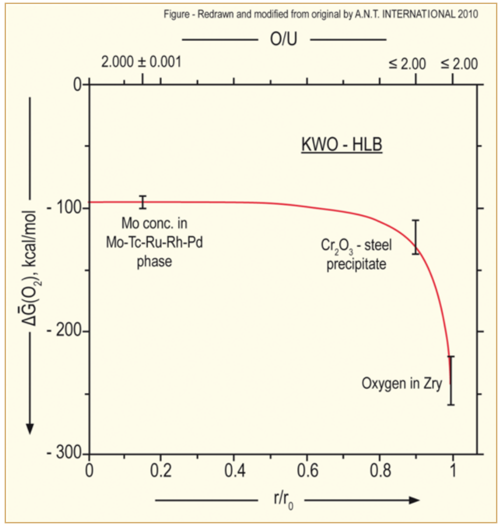
- Grain growth, grain boundaries, densification
- Started on fuel chemistry

FUEL CHEMISTRY CONT.

Oxygen potential

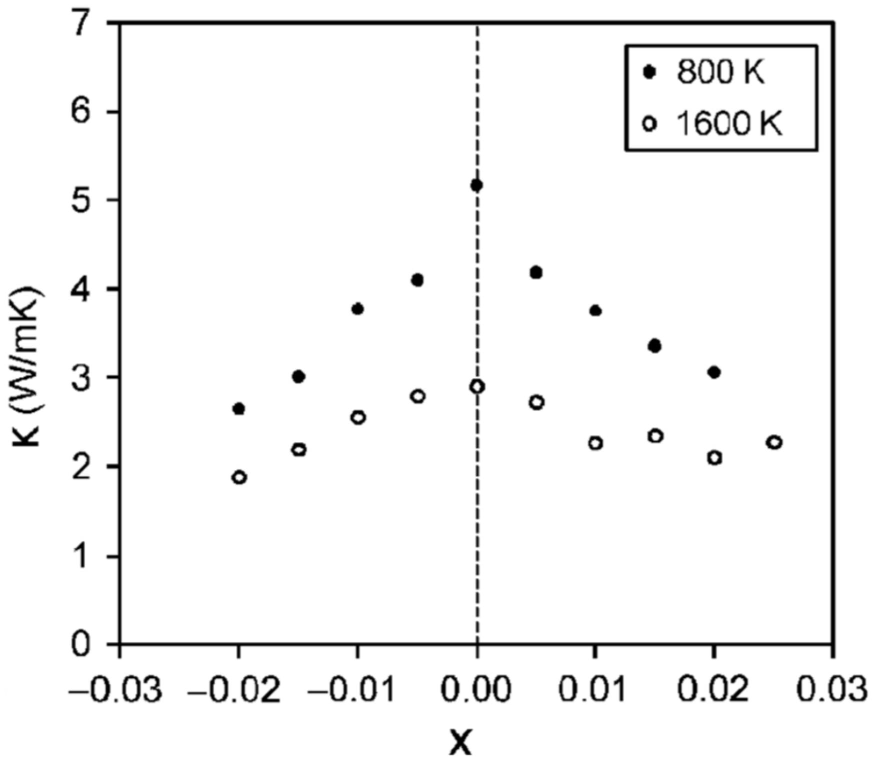
- The oxygen potential changes during irradiation, indicating change in the O/M ratio
- Oxygen potential changes during irradiation due to
  - Liberation of oxygen by fission; Generation of fission products; Conversion of uranium to plutonium; Reaction of oxygen with U, Pu, fission products, and cladding
- Oxygen potential across pellet radius observed to be constant at the approximate value of Mo/MoO$_2$ reaction (from calculations)
- Mo serves as a buffer to the O potential, or a means of inferring what the oxygen potential may have been in the fuel from PIE
- Oxygen potential is low near the cladding, because the oxygen enters the cladding





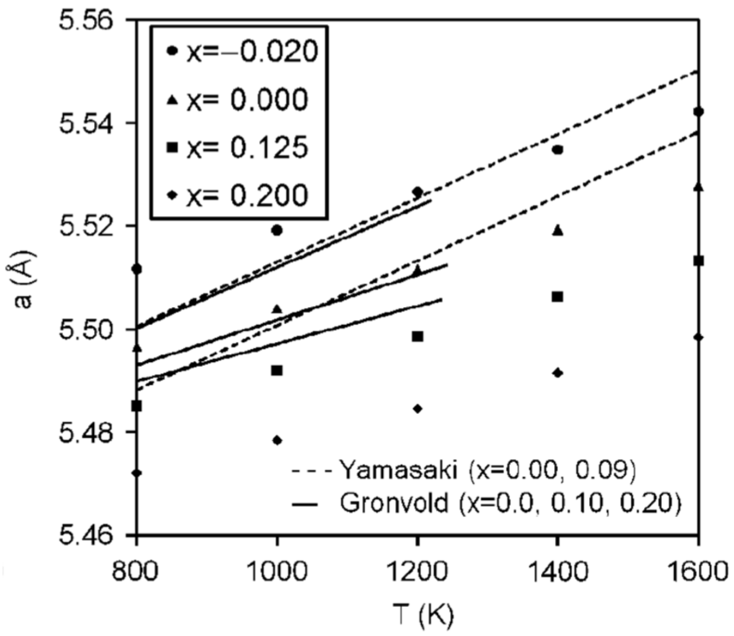
Fuel Stoichiometry/Properties

- The stoichiometry of the fuel directly impacts the fuel performance
- Stoichiometry impacts
  - Melting temperature
  - Thermal conductivity
  - Processes dependent on diffusion
    - Grain growth
    - Fission gas release
    - Creep
  - Chemical state and behavior of fission products
  - Chemical reactions at inner cladding surface
- Thermal conductivity is highest for stoichiometric UO2

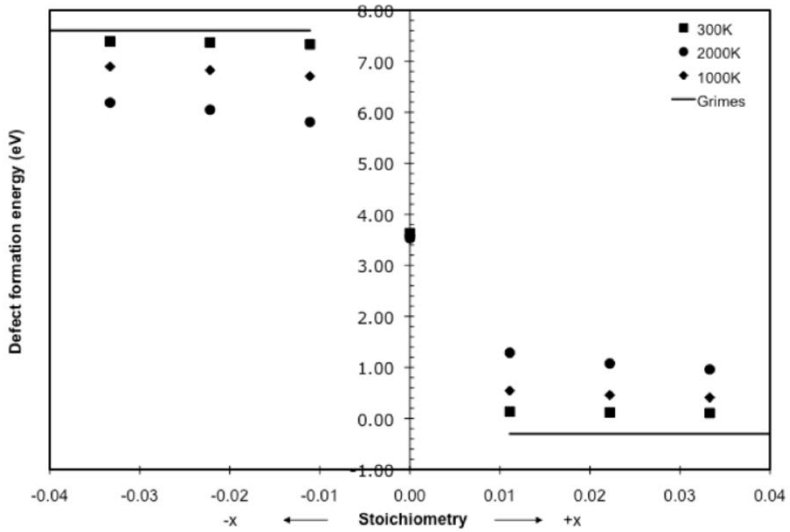


Fuel Stoichiometry/Properties

- The lattice constant of the material decreases with increasing stoichiometry



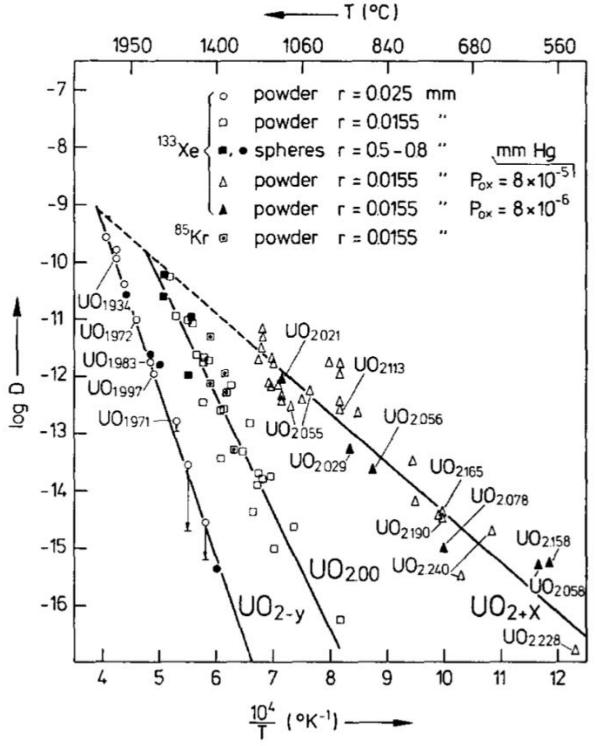
- The vacancy formation energy also changes with stoichiometry



Fuel Stoichiometry/Properties

- The solution energy of Xe, Cs, and Sr in UO_2 depends on stoichiometry as well
- The coefficient defining Xe diffusion also changes with stoichiometry
- **Though stoichiometry matters, most fuel performance codes ignore it**

Fission product	$UO_{1.97}$	UO_2	$UO_{2.03}$
Xe	3.88 eV	3.88 ev	2.61 eV
Cs	1.7 eV	-0.04 eV	-3.29 eV
Sr	-3.71 eV	-6.03 eV	-9.55 eV



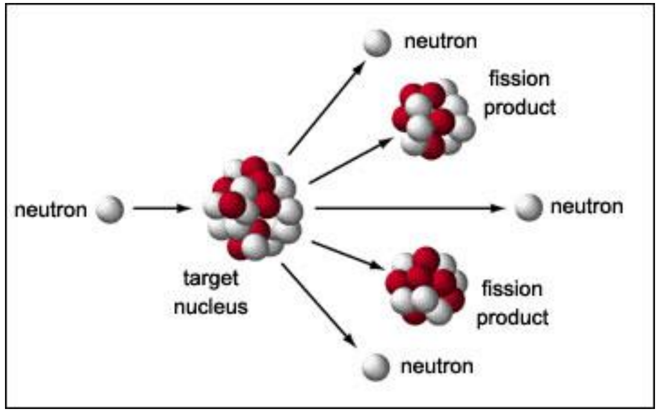
Fuel Chemistry Summary

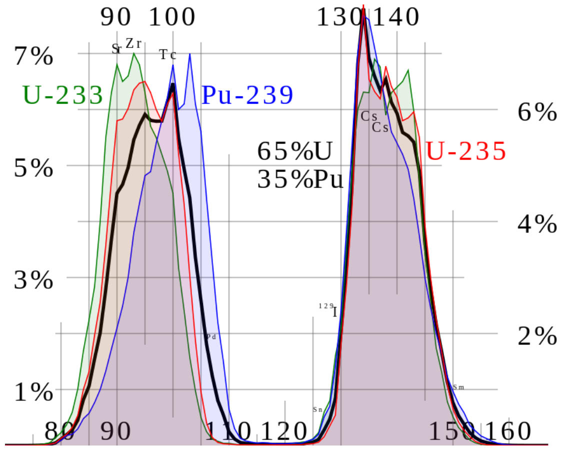
- UO_2 has a cubic fluorite structure that is very stable
- The charges are balanced with a U^{4+} valence state
- However, the ratio of oxygen to uranium can change. We call this the stoichiometry and abbreviate it as O/M ratio
- The O/M ratio changes during reactor operation, but it is complicated
- The O/M ratio impacts many properties of the fuel

FISSION PRODUCTS

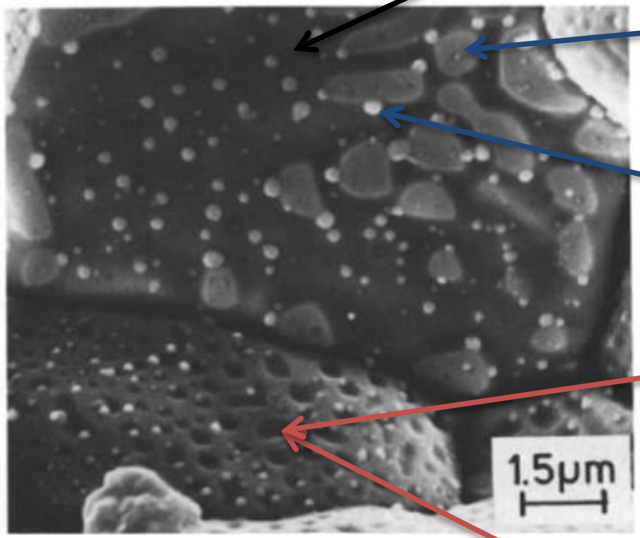
Fission Product Generation

- Fission releases around 200 MeV
  - The fission fragments have 169 MeV of kinetic energy
  - 2 to 3 neutrons with an average energy of 2 MeV
  - 7 MeV of prompt gamma ray photons
  - The remaining energy is released by beta decay
- Every fission product that is produced is now in the crystal lattice of the fuel, changing the chemistry/microstructure





There are various types of fission products that form



Soluble oxides (Y, La and the rare earths)
- Dissolved in the cation sublattice

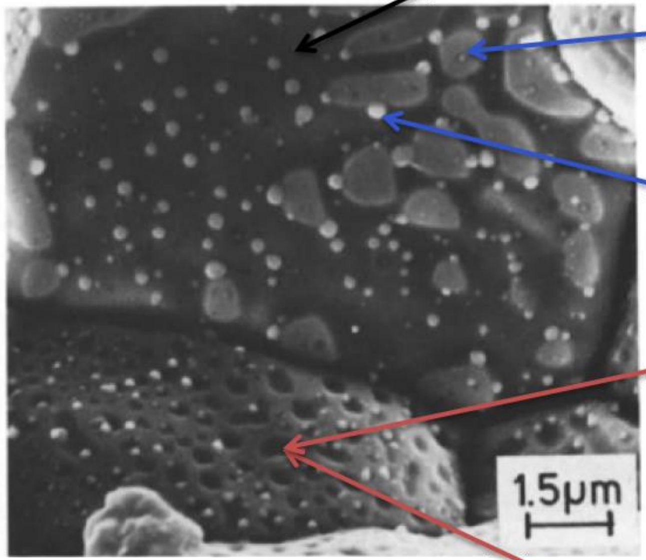
Insoluble oxides (Zr, Ba and Sr)
- Form insoluble oxides in the fluorite lattice

Metals (Mo, Ru, Pd, and Tc)
- Form metallic precipitates

Volatiles (Br, Rb, Te, I and Cs)
- Exist as gases at high temperatures of the pellet interior
- Also exist as solids at the cooler pellet exterior

Noble gases (Xe, Kr)
- Essentially insoluble in the fuel matrix
- Form either intragranular (within grain) voids or bubbles or intergranular (grain boundary) bubbles

All fission products impact the behavior of the fuel



Soluble oxides (Y, La and the rare earths)
- Cause swelling, decrease thermal conductivity

Insoluble oxides (Zr, Ba and Sr)
- Can cause swelling

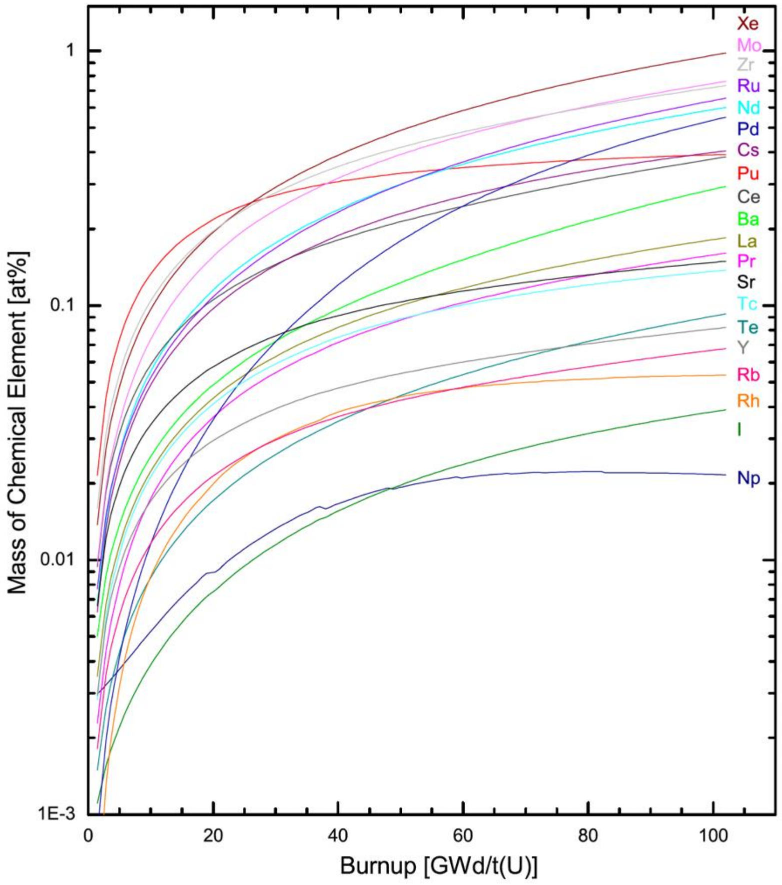
Metals (Mo, Ru, Pd, and Tc)
- Slightly raise thermal conductivity,

Volatiles (Br, Rb, Te, I and Cs)
- Cause swelling, decrease thermal conductivity
- Escape from fuel, corrode the cladding

Noble gases (Xe, Kr)
- Cause swelling
- Decrease thermal conductivity
- After release, raise gap pressure and lower thermal conductivity

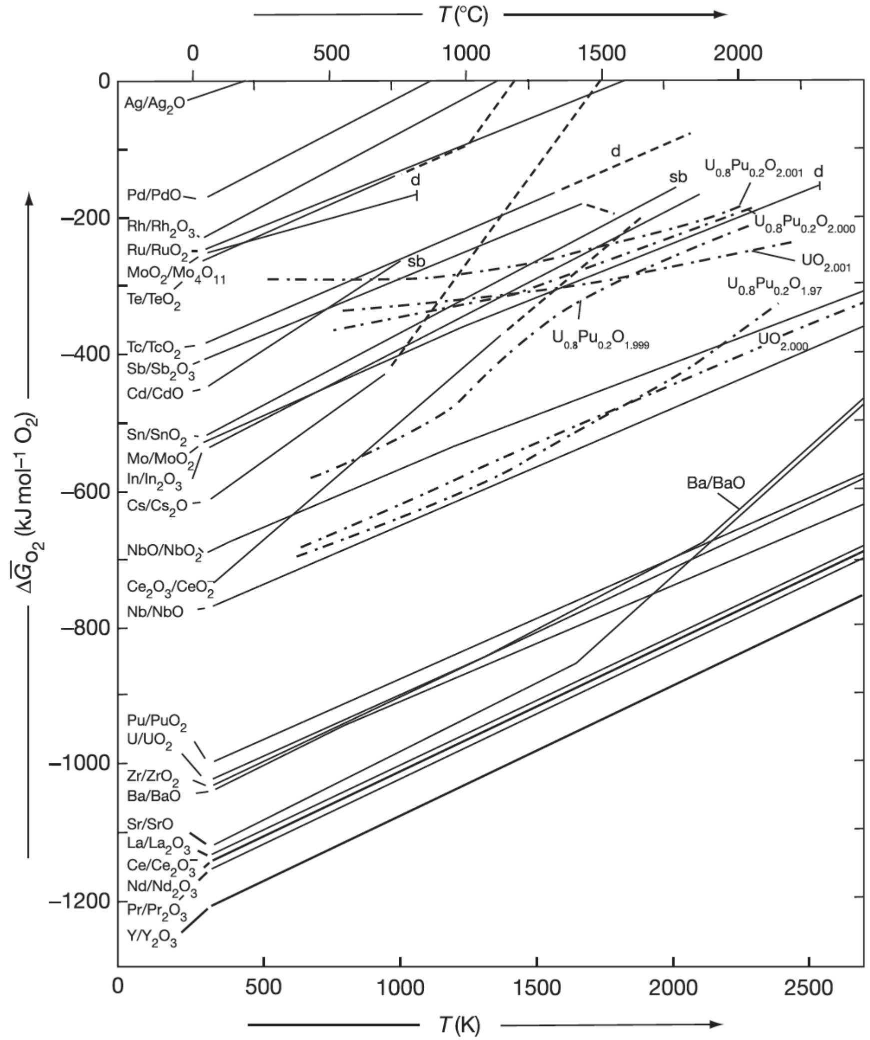
Fission Product Evolution

- Not only are the absolute proportion of fission products accumulating with time, but the relative proportions are changing with respect to one another
- There are continuous transitions between the groups as the critical concentration conditions for new phase development are surpassed because of increased burnup
- Since fission releases two oxygen but only one metal atom, and not all metal atoms will react to bond the oxygen, the O/M ratio of the fuel slowly increases during burnup



Oxide Formation

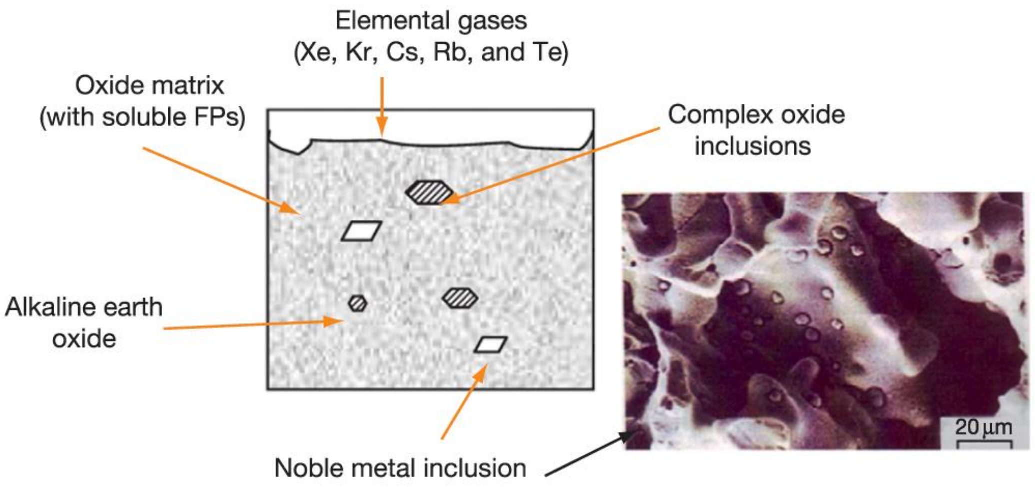
- With burnup, the liberated oxygen will associate with U, Pu, and La but may not be in sufficient supply to combine with Pd and other less oxidizing elements
- If the Gibbs energy of formation of the given fission product lies below the fuel oxygen potential, the element will be capable of forming an oxide (Ce, Sr, Ba)
- If the reaction line is above the oxygen potential in the fuel, the fission product will exist as an element in the fuel (in a separate metallic phase) (Ru, Pd, Tc)



Soluble/Insoluble Fission Products

- When a 4+ valence fission product (e.g.,Zr4+) enters the lattice, there is no change in the electrical neutrality
- If the charge of the fission-product cation is lower than U+4, the site occupancy of the lattice is altered to achieve electrical neutrality
- The alkaline earth cations Ba+ and Sr2+ have large ionic radii and form a separate oxide phase
- Fission products that have limited solubility in UO2 will segregate to the grain boundaries and voids

Chemical group	*Physical state*	*Probable valence*
Zr and Nb	Oxide in fuel matrix; some Zr in alkaline earth oxide phase	4+
Rare earths	Oxide in fuel matrix	3+
Ba and Sr	Alkaline earth oxide phase	2+
Mo	Oxide in fuel matrix or element in metallic inclusion	4+ or 0
Ru, Tc, Rh, and Pd	Elements in metallic inclusion	0+
Cs and Rb	Elemental vapor or separate oxide phase in cool regions of fuel	1+ or 0
I and Te	Elemental vapor; I may be combined with Cs and CsI	0 or 1 −
Xe and Kr	Elemental gas	0

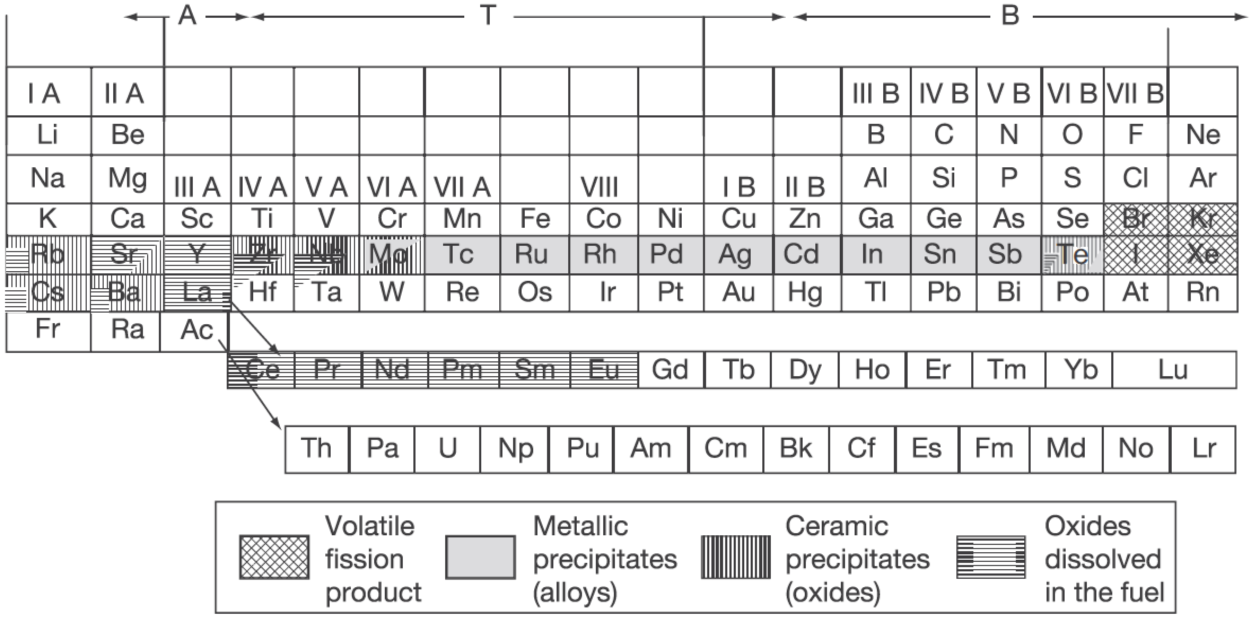


Fission Product Segregation/Precipitation

- Fission products can be segregated because of their migration to specific locations such as intragranular segregations, grain boundaries, or pellet surfaces
- Metallic inclusions are commonly observed in ceramographs from irradiated samples
- They are formed by isotopes of Mo, Tc, Ru, Rh, Pd, Ag, Cd, In, Sn, Sb, and Te
- These inclusions are found at the surface of the grain boundaries and are associated, in general, with grain boundaries and intragranular bubbles
- Some of the components of these precipitates can be oxidized or reduced forming other compounds that can be released from the fuel matrix
- Cs, Ru, Te, and Ba have been consistently found at the cracks in the pellet and on the clad inner surface
- The white inclusions are metallic precipitates, which are composed of Mo, Tc, Rh, Ru, and Pd, form a quinary alloy in an hcp structure
- A “gray oxide phase” perovskite structure containing different combinations of Ba, Cs, Zr, Mo, and U can also form

Volatile/Gaseous Fission Products

- Xe release occurs at the central region of the pellet where the highest temperatures were achieved during irradiation
- Volatile FPs are released from the fuel matrix similar to that of the noble gases
- Volatile fission products in the gap can react among themselves, resulting in a changing chemical speciation
- The kinetics of formation/decomposition of CsI and Zr iodides are possible factors in the mechanism of SCC



FP Interaction with Zr Cladding

- Fission fragments will deposit on the inner surface of the cladding
- Some of these species can diffuse into the cladding, while others can attack the cladding thereby initiating cracks that can later progress with the formation of through-wall cracks
- Oxygen will diffuse into the cladding and contribute to its oxidation state
- Measurements have shown deposits of Sr, Cs, Pu, and Am
- The corrosion behavior of zirconium by Te has been reported
- The chemical reaction between some corrosive fission products and the cladding can lead to PCI
- Fission products such as Cs, Cd, or I can attack the cladding inducing crack initiation, which then progress through the cladding by intragranular and transgranular cracking modes

FISSION GAS RELEASE

Fission Gas Release

- Fission gases (Xe, Kr) are released in a process composed of three stages in UO2
- Stage 1: Gas atoms are produced throughout the fuel due to fission and diffuse towards grain boundaries
- Small intragranular bubbles form within the grains, but never get larger than a few nm radius due to resolution from energized particles
- Gas atoms that don’t get trapped within the intragranular bubbles migrate to grain boundaries

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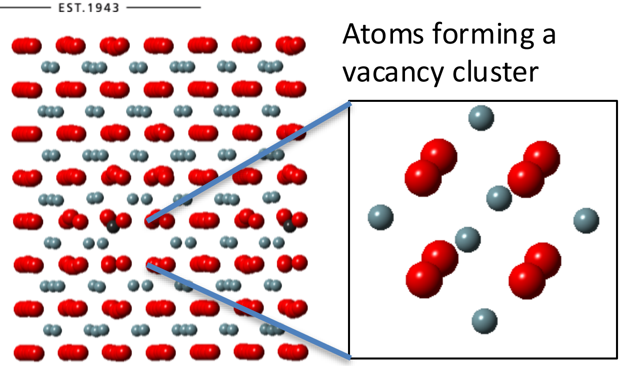
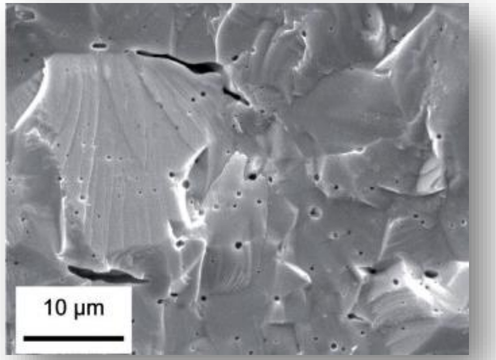
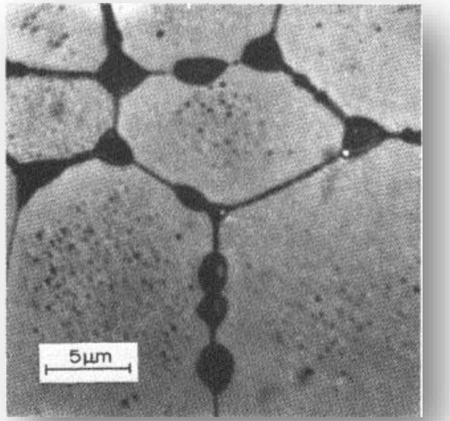
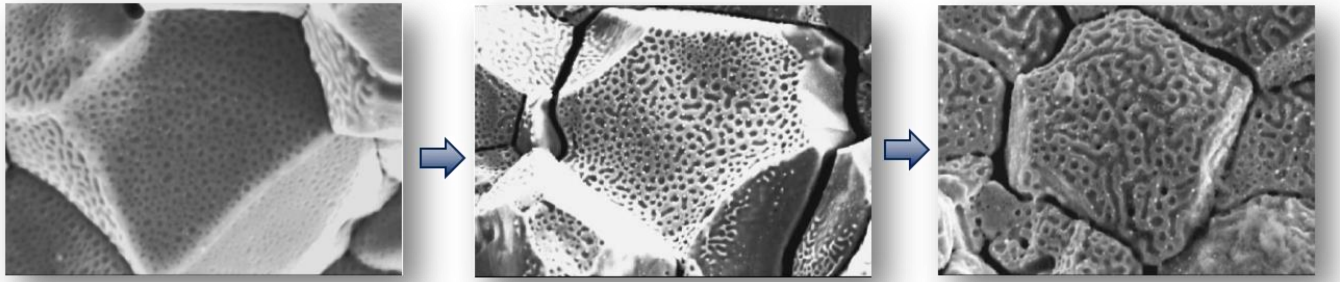






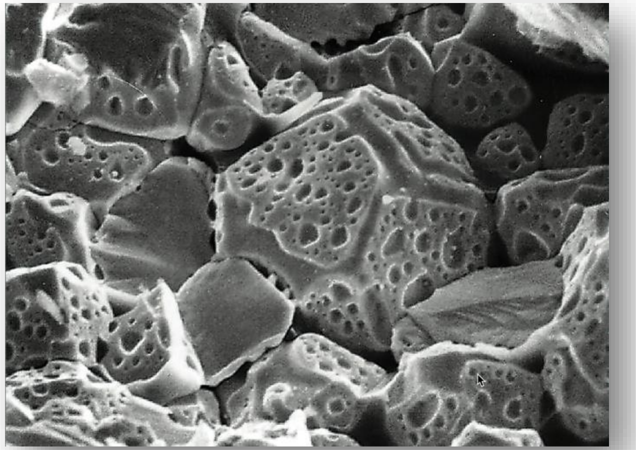
Fission Gas Release

- Stage 2: Gas bubbles nucleate on grain boundaries, growing and interconnecting



Nucleation Growth Percolation

- Stage 3: Gas travels through interconnected bubbles to a free surface



Fission Gas Release

- Fission gas release also occurs due to mechanisms that don't depend on diffusion
- Release can occur to particle recoil and knockout at low temperature
- It can occur due to fracture during rapid transients