Nuclear Fuel Performance

NE-533

Spring 2023

Last Time

- Dr. Larry Aagesen spoke about phase field simulations for nuclear fuel performance modeling
- Exploring fuel fragmentation of the HBS
- Developing new models for fission gas swelling and release of metallic fuels
- Utilizing mechanistic, lower length scale, modeling tools to inform engineering scale fuel performance simulations

- Thanks for turning in MOOSE project
- Problem sessions and short lecture today; Exam on Thursday

UO_2 RADIATION EFFECTS

Thermal Conductivity Degradation

- The physical effects caused by atomic displacements are rather complex and depend on the relative sink strengths of a given material for interstitials and vacancies and on the temperature
- The defects that survive migrate through the crystal lattice where they cluster to form extended defects like dislocation loops, lines and networks, which can potentially be absorbed in grain boundaries, gas bubbles or precipitates, which act as sinks
- Damage by fission also leads to changes in chemistry due to production of fission products
- One of the major effects of defect formation is the degradation of thermal diffusivity, due to phonon scattering
- Results obtained on irradiated UO2 show that the thermal conductivity is decreasing with increasing burn-up, and that at equal burn-up, samples with higher irradiation temperatures have higher thermal conductivity

Thermal Conductivity Degradation

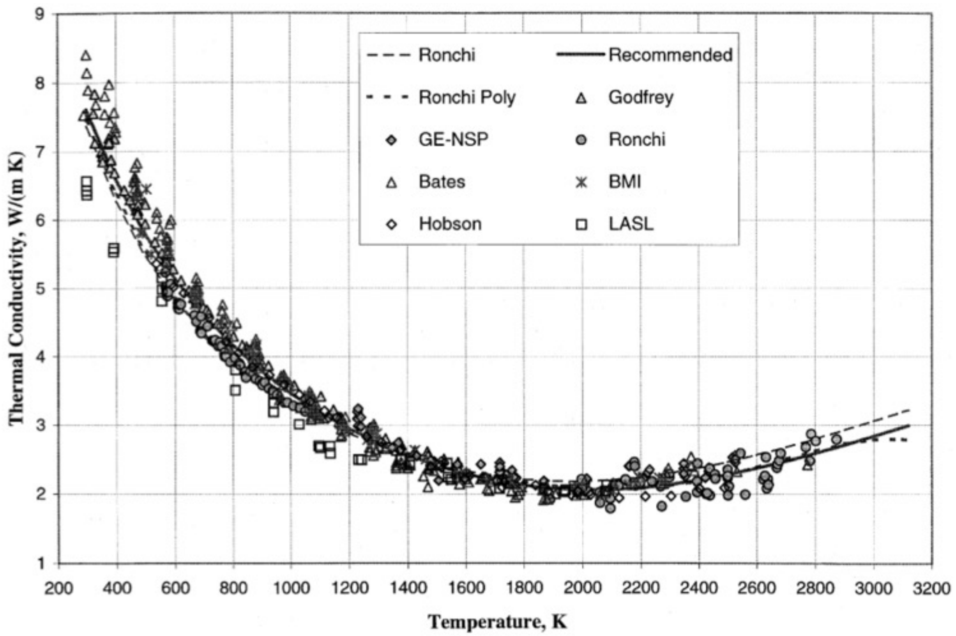
- There are mesoscopic and atomistic features that affect the thermal conductivity
- The porosity existing in fresh fuels evolves during irradiation and has a strong impact on heat transport, because voids/bubbles have significantly lower kth than the lattice
- Precipitates are formed by fission products that are insoluble in the UO2 lattice, forming oxide or metallic inclusions: can use composite theory to get effective thermal conductivities; typically, these precipitates INCREASE the thermal conductivity of the fuel
- Actinides, rare earths, and transition metals can form mixed oxides with UO2 (soluble fission products): these atoms act as phonon scattering centers as a result of the differences in bonding, radii, or mass, and thus decrease kth
- Volatile fission products/gases are partially in solution and partially in bubbles, where they act as phonon scatterers or as low kth composite fractions
- Point defects and extended defects contribute to the degradation of the thermal conductivity by scattering or limiting the mean free path of the phonons

UO2 Thermal Conductivity

- We have established correlations for thermal conductivity of fresh UO2 fuel as a function of temperature

$$k_0 = \frac{100}{7.5408 + 17.629t + 3.6142t^2} + \frac{6400}{t^{5/2}} \exp\left(\frac{-16.35}{t}\right)$$

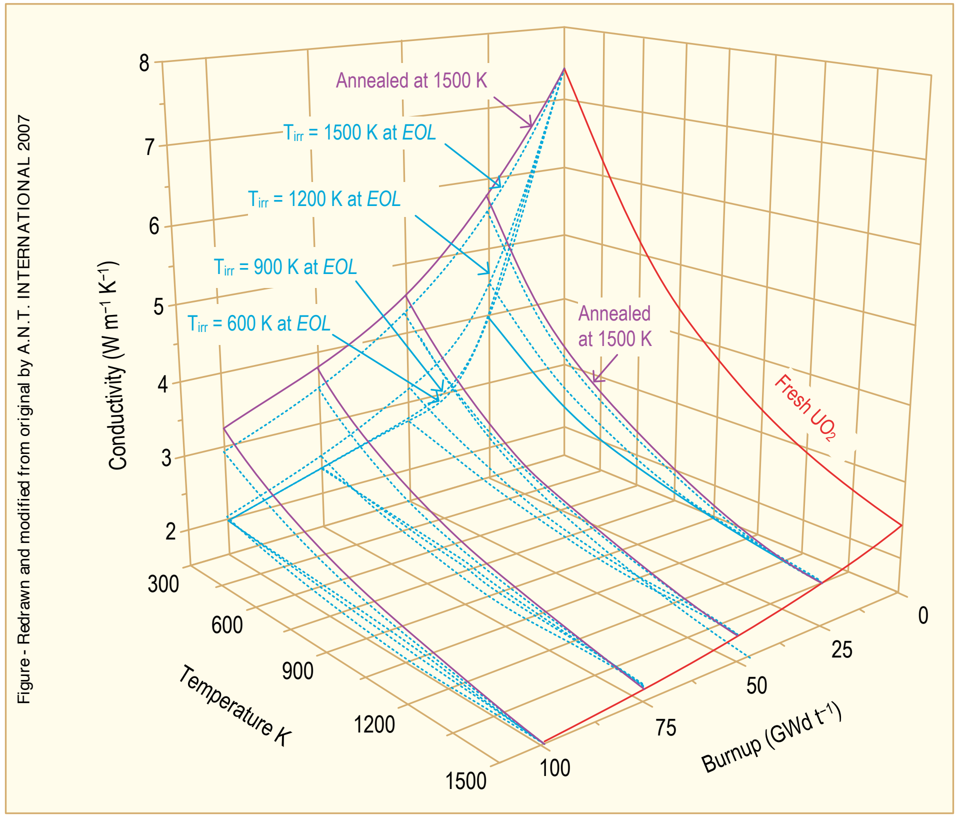
  - Where t = T/1000
  - The first part of the equation describes the phonon interactions
  - The second part describes electronic transport which becomes significant at high temperature



UO2 Thermal Conductivity

- UO_2 thermal conductivity is low and decreases more during reactor operation
- The thermal conductivity has been collected after various amounts of burnup to make empirical fits

$$\lambda_{95} = \frac{1}{A(x) + aG + B(x)T + f_1(Bu) + f_2(Bu)g(Bu)h(T)} + \frac{C}{T^2}\exp\left(\frac{-D}{T}\right)$$



UO2 Thermal Conductivity

- The primary empirical model used in BISON is the NFIR model
- The model is a function of the temperature T (in °C) and the burnup β (in MWD/kgU

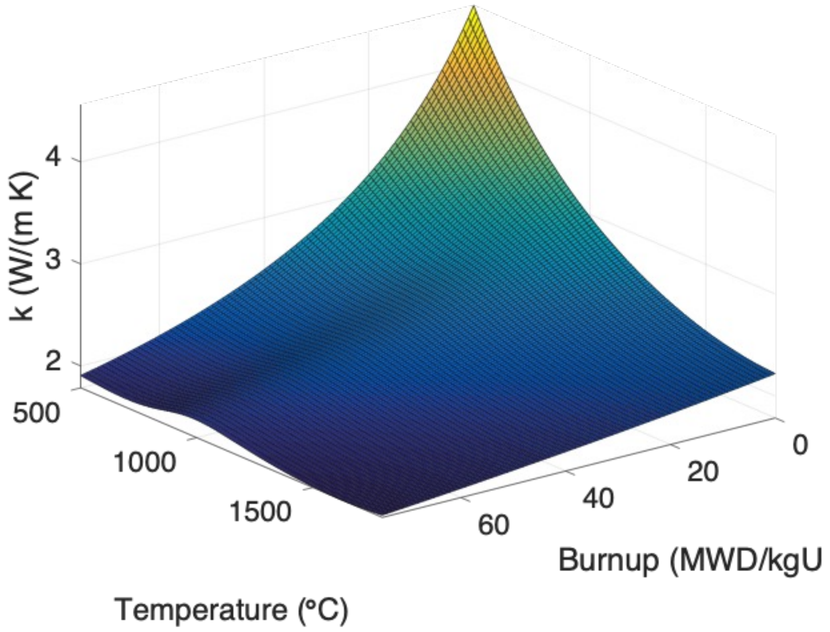
$$k = (1 - R_f(T))k_{ph1}(T,\beta) + R_f(T)k_{ph2}(T,\beta) + k_{el}(T)$$

$$R_f(T) = \frac{1}{2}\left(1 + \tanh\left(\frac{T - 900}{150}\right)\right)$$

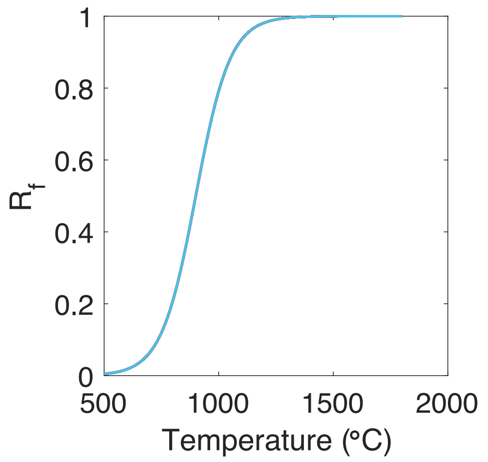
$$k_{ph1} = \frac{1}{(9.592 \times 10^{-2} + 6.14 \times 10^{-3}\beta - 1.4 \times 10^{-5}\beta^2 + (2.5 \times 10^{-4} - 1.81 \times 10^{-6}\beta)T}$$

$$k_{ph2} = \frac{1}{(9.592 \times 10^{-2} + 2.6 \times 10^{-3} \cdot \beta + (2.5 \times 10^{-4} - 2.7 \times 10^{-7}\beta)T}$$

$$k_{el} = 1.32 \times 10^{-2}\, e^{1.88 \times 10^{-3} T}$$

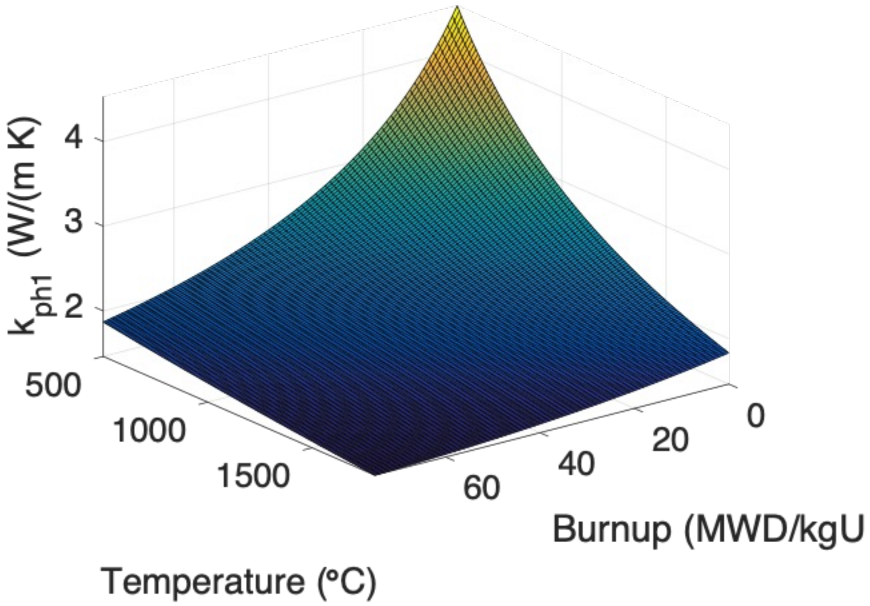


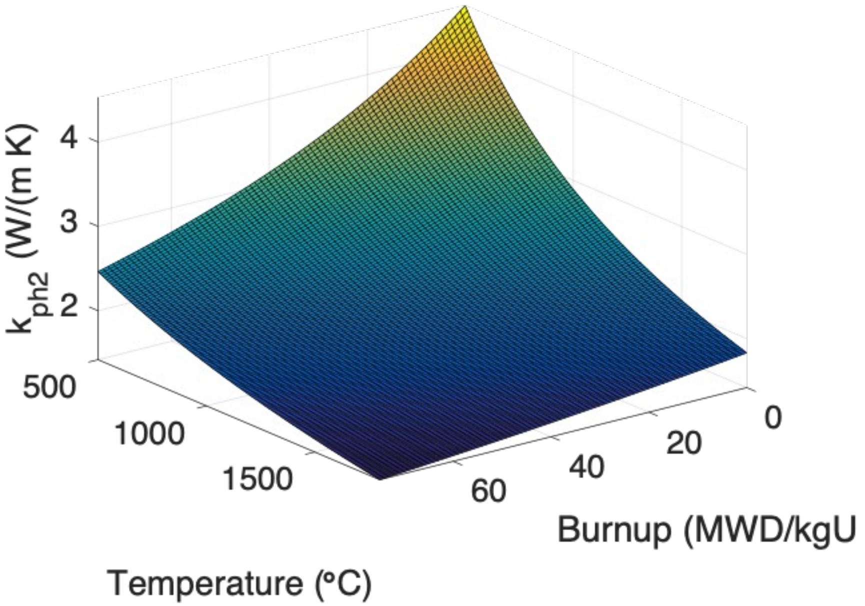
UO2 Thermal Conductivity



- The R_f function switches between two k_{ph} functions

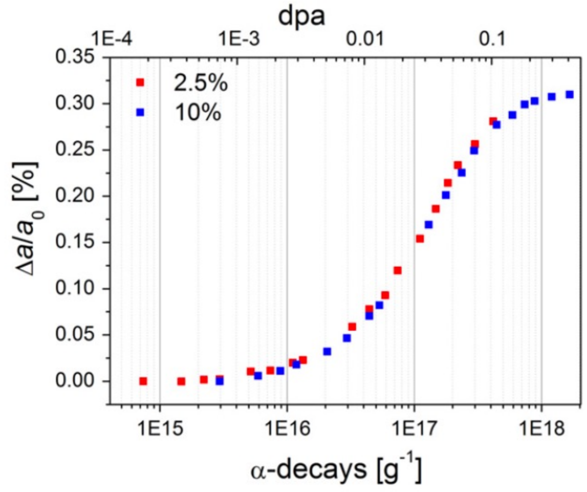
$$R_f(T) = \frac{1}{2}\left(1 + \tanh\left(\frac{T-900}{150}\right)\right)$$

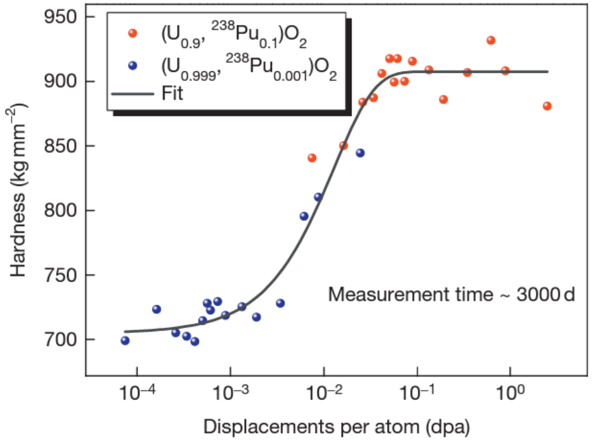


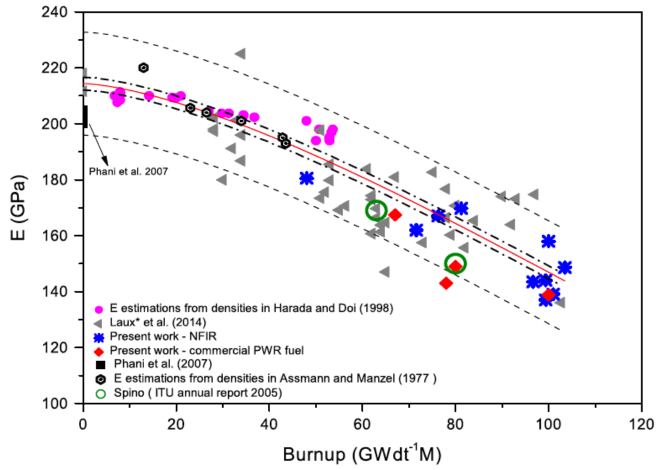


Physical Changes

- An important effect of radiation damage is the volume increase of the crystal lattice, leading to macroscopic swelling of the fuel material
- Alpha particle damage alone can cause approximately 0.4% swelling
- Mechanical properties also vary with damage, increasing with dpa
- UO2 does not become amorphous, but damage accumulation can eventually cause polygonization: transformed into a material consisting of very small grains





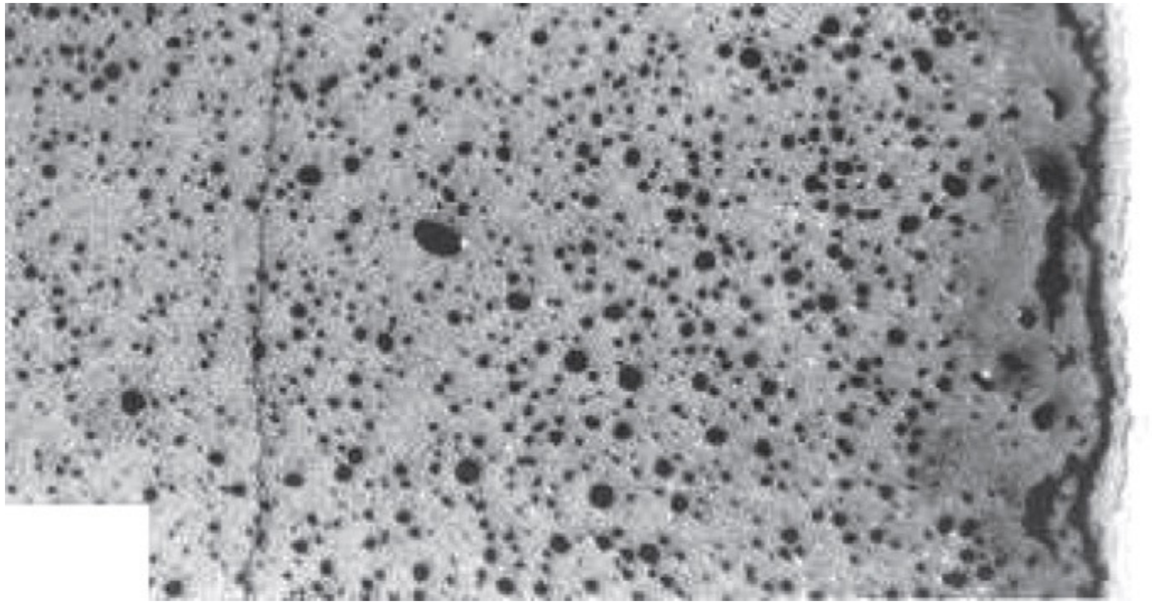


High Burnup Structure (HBS)

- In the late 1950s, it was observed that a strong capture of neutrons by U-238 in the resonance range occurs at the periphery of the fuel
- This leads to the production of Pu-239, and leads to a higher fissile density and thus an increase in the local burnup
- This high local burnup area is on the outer ring of the fuel and is about 200 microns thick, which represents about 8% of the fuel
- This leads to microstructure changes…

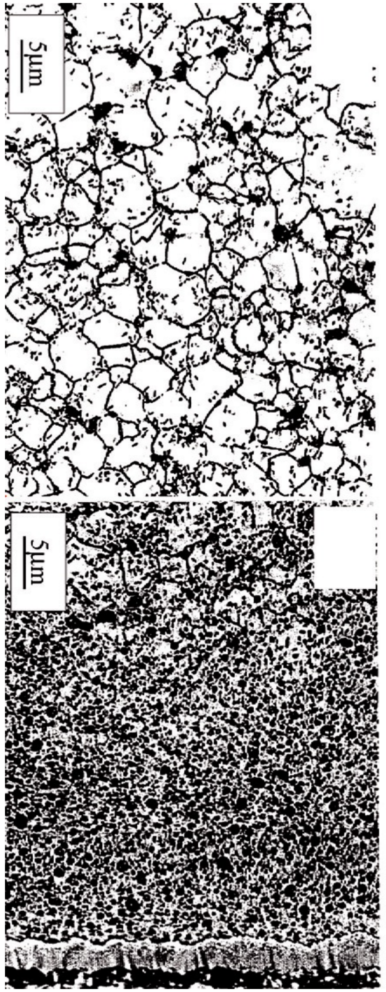
High Burnup Structure (HBS)

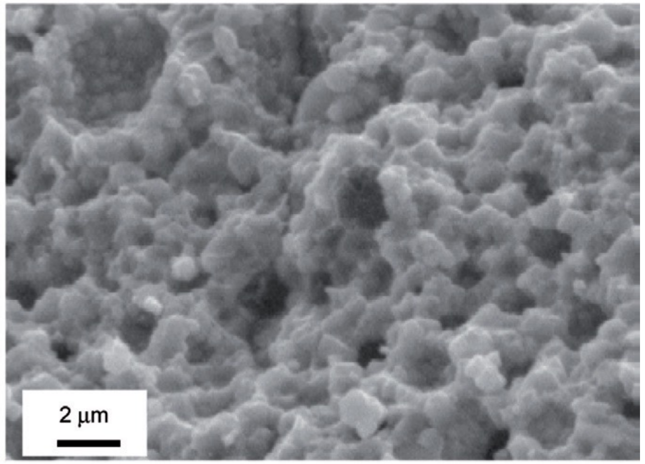
- When thermal recovery is not sufficiently efficient (at lower temperatures), the accumulation of defects at high burnup can lead to the instability of the crystalline structure, initiating a restructuring driven by the energy stored in the material
- In UO_2, grains subdivide from 10microns in size to 100-200 nm size, and a densely porous structure is formed (~20% porosity)



High Burnup Structure (HBS)

- The increase of the relative porosity volume degrades the material conductivity and reduces the mean grain size
- On the other hand, the intragranular irradiation defect-cleaning improves the fuel intrinsic thermal conductivity
- Fission gas in HBS bubble is retained, not released
- Exact mechanism of HBS formation is not understood

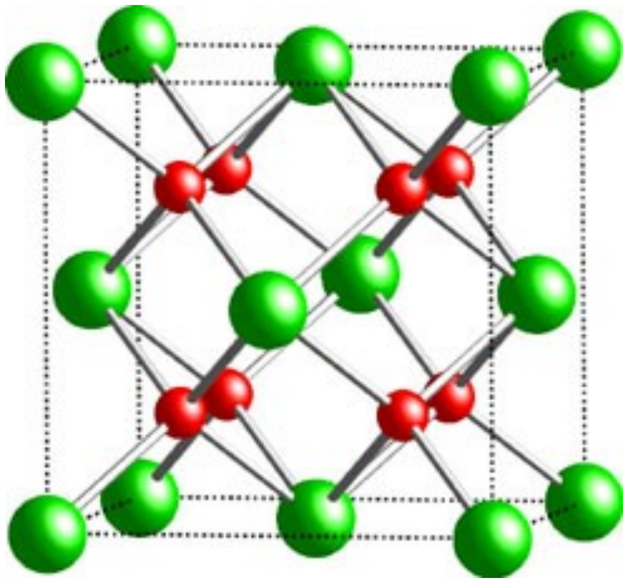




FUEL CHEMISTRY

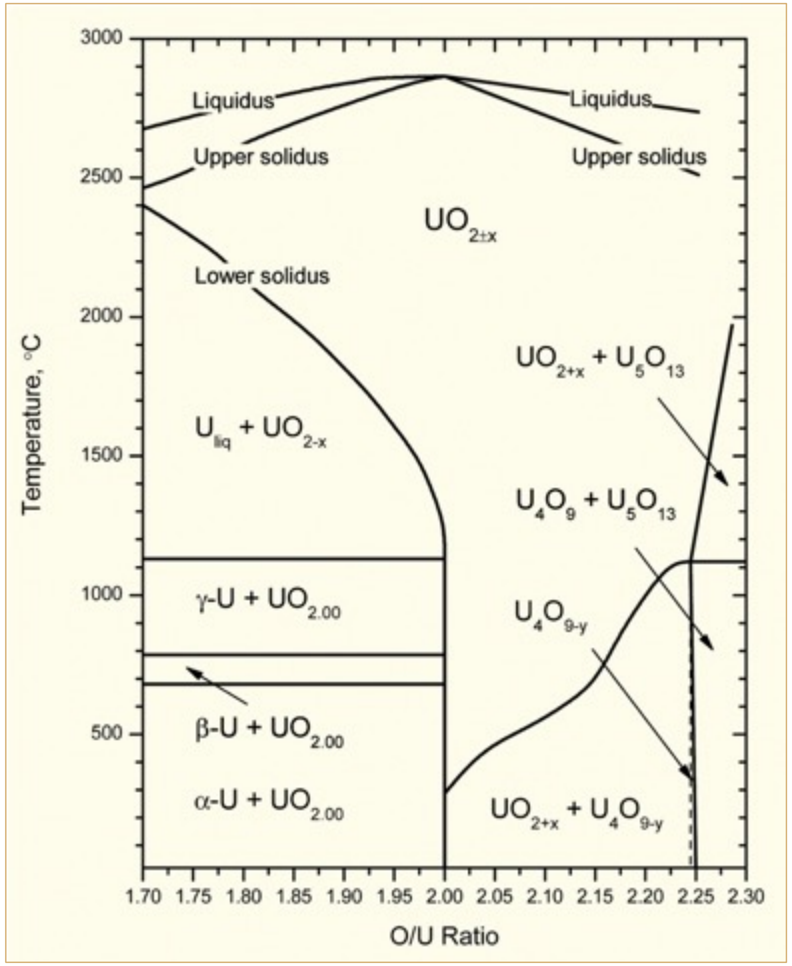
Fuel Chemistry

- UO_2 is an ionic compound that must have balanced charges
- What is the charge of a typical oxygen ion?
  - O^{2-}
- Uranium valance states
  - Possible: U^{3+}, U^{4+}, U^{5+}, U^{6+}
  - Most stable: U^{4+}, U^{5+}, U^{6+}
  - Beyond UO2, can have U4O9, U3O8, UO3
- The structure is very stable all the way up to the melting temperature and down to extremely low temperatures, even with irradiation damage
- There is space in the uranium lattice that can accommodate fission products



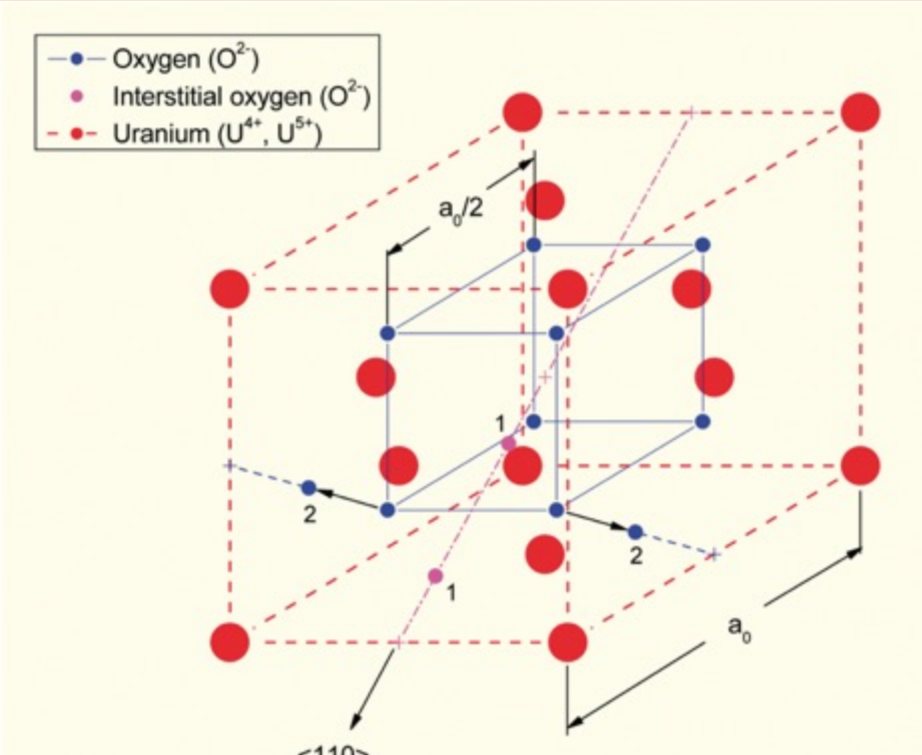
O/M ratio

- The ratio of oxygen to uranium metal (O/M ratio) can vary
- This is the stoichiometry
- The homogeneity range of uranium dioxide extends to both hypo- and hyperstoichiometric compositions in oxygen
- The minimum and maximum oxygen contents in the dioxide correspond to the compounds with the formula of respectively $UO_{1.67}$ at 2720 K and $UO_{2.25}$ at approximately 2030 K
- Will the O/M ratio go up or down during reactor operation?
  - It is complicated, because of the formation of fission products that also react with the oxygen, and O interaction with Zr cladding



Excess O

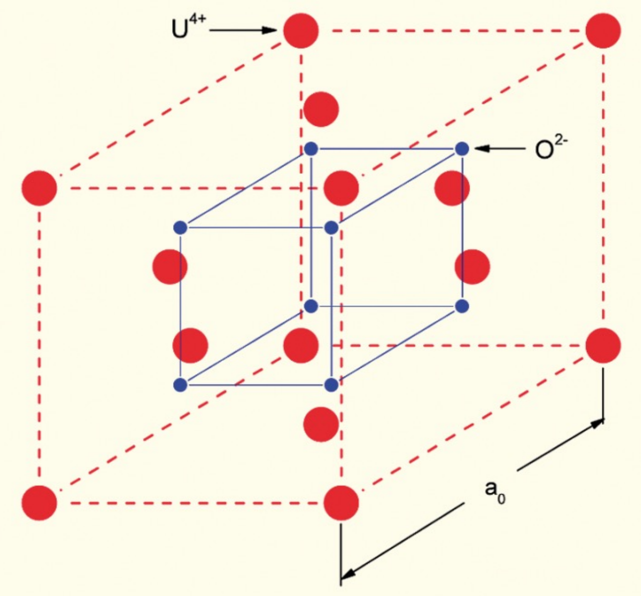
- The crystal structure of UO_2 can accommodate extra oxygen
- Excess oxygen resides at interstitial locations
- Oxygen in neighboring sites is displaced
- Cation valence increases to maintain electrical neutrality
- Fuel fabricated to be nearly stoichiometric; i.e., UO2.00± because:
  - It is the most stable
  - It has the highest melting temperature



[after Olander, 1976]

Incorporation of Fission Products

- As fission products form, the valence state of the uranium can change
- Typical valence of soluble fission products is M^{3+}
- The uranium valence state changes to compensate
  - Oxygen liberated by fission
  - Fission products produced with M^{3+} valance state incorporated in fuel lattice
  - Uranium oxidizes from U^{4+} to U^{5+} or U^{6+} to maintain local electrical neutrality



b) Cation sublattice