Nuclear Fuel Performance

NE-533

Spring 2022

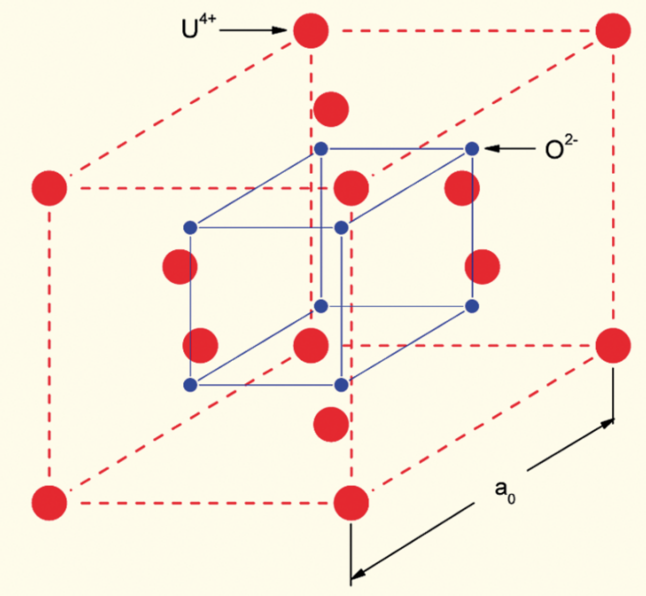
Last Time

- Fuel and pellet conditions change with time due to microstructure evolution
- All materials have defects, radiation damage causes many more defects
- Microstructure can be tailored through processing
- Thermal conductivity degrades with time
- High Burnup Structure can form along the rim
- Started on some fuel chemistry

FUEL CHEMISTRY

Incorporation of Fission Products

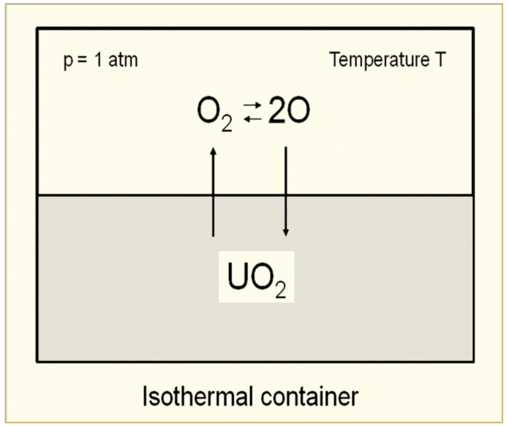
- As fission products form, the valence state of the uranium can change
- Typical valence of soluble fission products is M^{3+}
- The uranium valence state changes to compensate
  - Oxygen liberated by fission
  - Fission products produced with M^{3+} valance state incorporated in fuel lattice
  - Uranium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

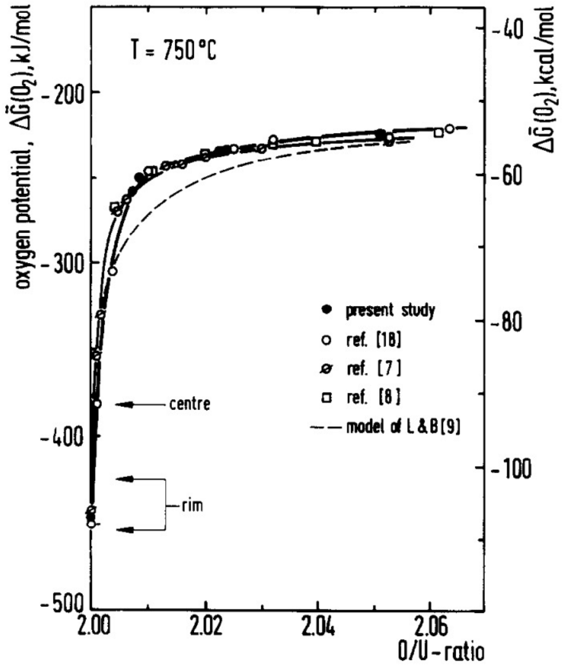


b) Cation sublattice

Oxygen potential

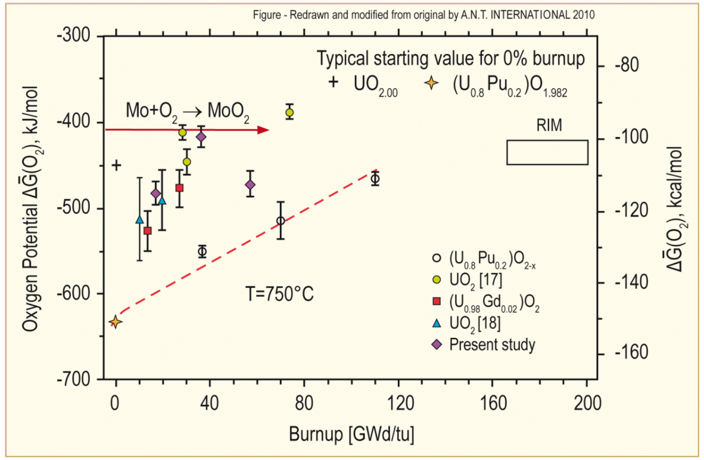
- The oxygen potential is a measure of how free the oxygen is to escape/move around
- Oxygen potential is defined as: $\Delta \bar{G}_{O_2} \equiv RT \ln\left(p_{O_2}\right)$
- or, equivalently: $\Delta \bar{G}_{O_2} = 2\mu_{O_{solution}} - G^{\circ}_{O_2}$
  - With $\mu_{O_{solution}}$ = Chemical potential of oxygen in solution
  - $G^{\circ}_{O_2}$ = Gibbs free energy of gaseous oxygen at temperature T and a standard pressure (1 atm)
- It is possible to determine the stoichiometry from the oxygen potential

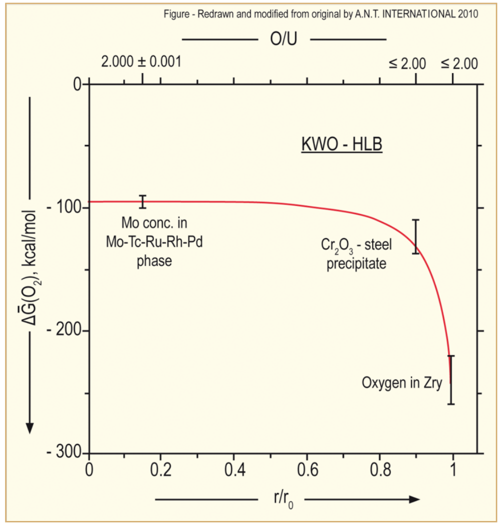




Oxygen potential

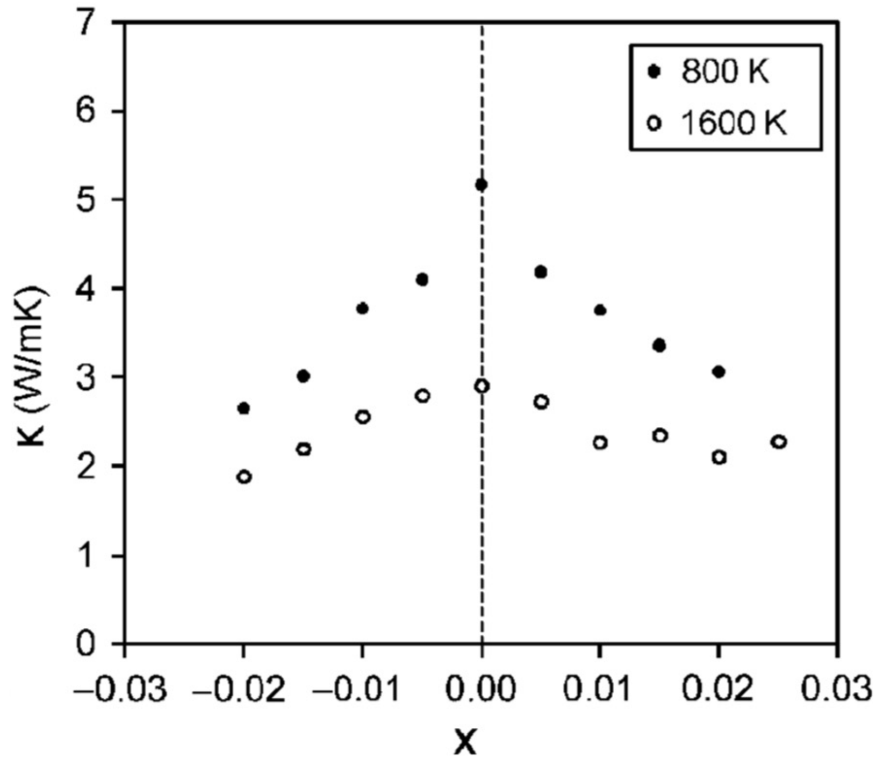
- The oxygen potential changes during irradiation, indicating change in the O/M ratio
- Oxygen potential changes during irradiation due to
  - Liberation of oxygen by fission; Generation of fission products; Conversion of uranium to plutonium; Reaction of oxygen with U, Pu, fission products, and cladding
- 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





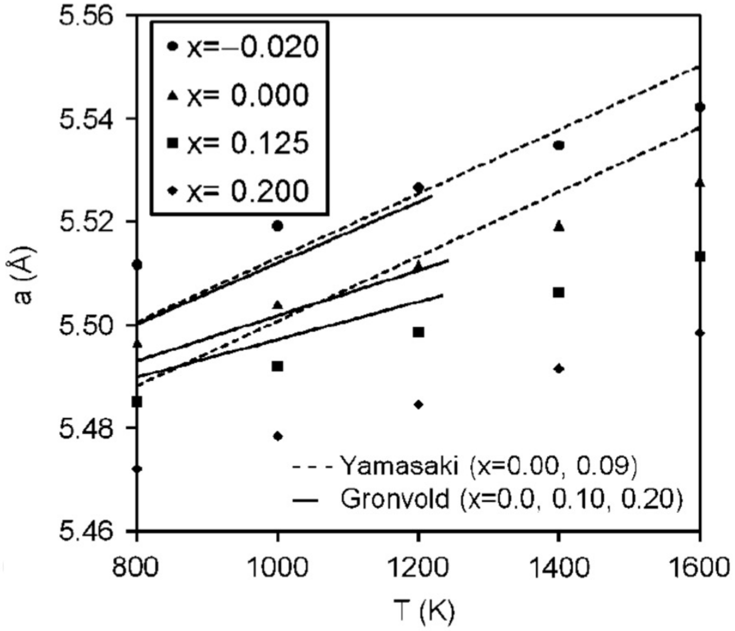
Fuel Stoichiometry/Properties

- 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

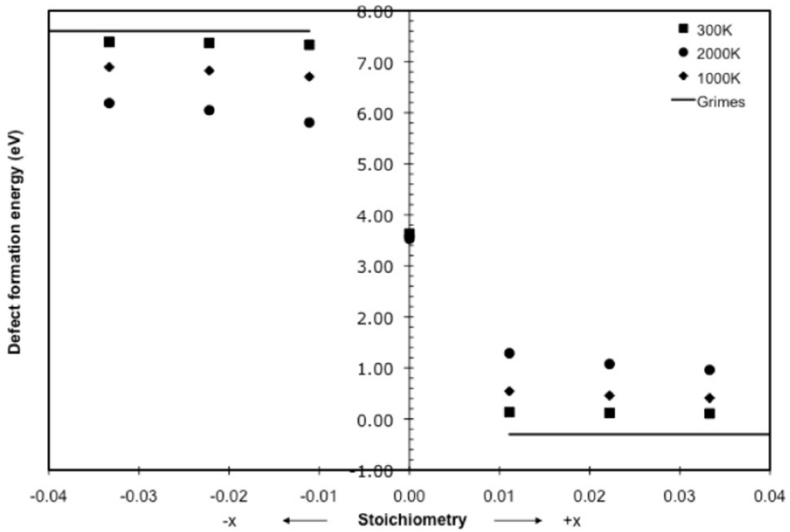


Fuel Stoichiometry/Properties

- The lattice constant of the material decreases with increasing stoichiometry



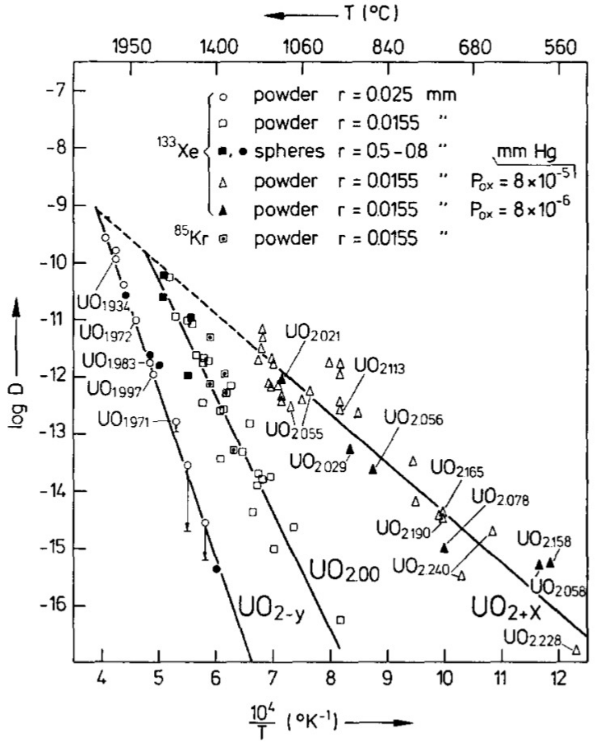
- The vacancy formation energy also changes with stoichiometry



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