Nuclear Fuel Performance

NE-533

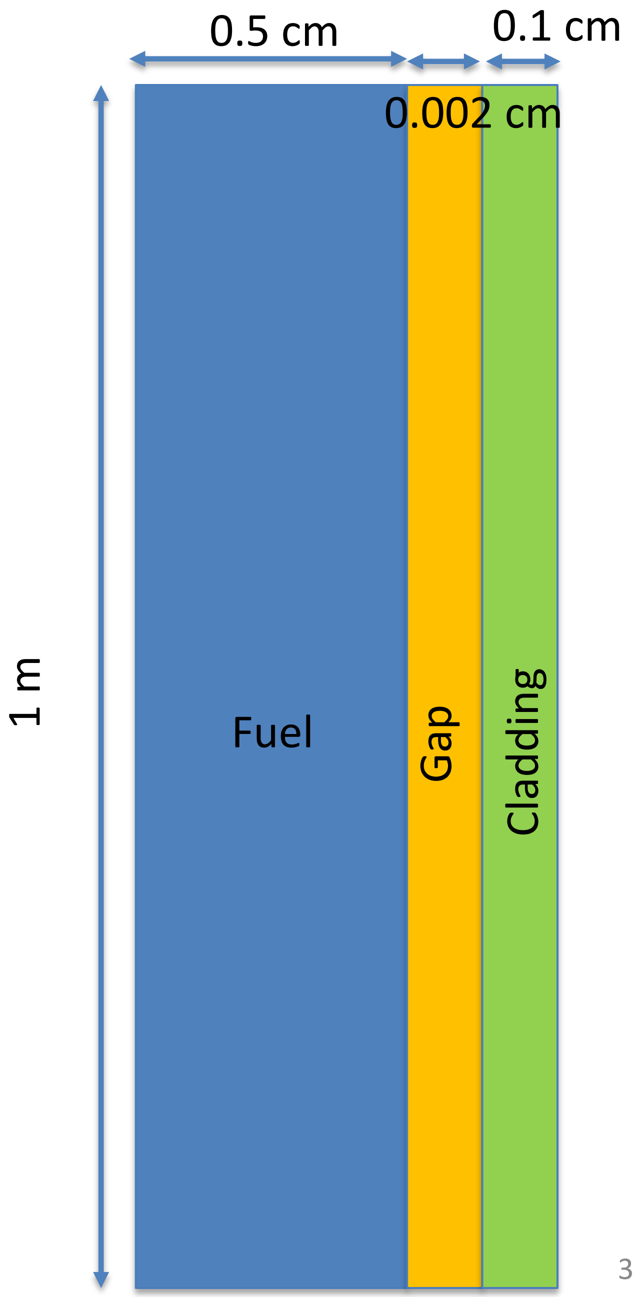
Spring 2022

Last time

- If you want to opt into the paper project 2, please do that asap
- Reminder, MOOSE project due April 26

- Covered RIA and LOCA type accidents
- RIA is often caused in PWR/BWR by control rod ejection/drop
- Shorter pulses have greater impact than longer pulses (given same energy deposition)
- Effects of RIA depend on burnup includefission gas, FCMI, oxide layer, hydrides, cladding pressure, etc
- In loss of coolant accidents (LOCA), the fuel and cladding experience increases temperature and decrease in coolant pressure – occurs more slowly than a RIA
- Accident tolerant fuel includes different fuel and cladding materials to increase time before catastrophic behavior during an accident

MOOSE Project Part 2



- Fuel pin dimensions listed – 2D RZ
- Assume reasonable values for thermal conductivities, constant
- Utilize axial T_{cool}, with T_{cool}^{in}= 400 K, reasonable flow rate, heat capacity, etc.
- Utilize axial LHR, with LHR^0=150 W/cm

$$LHR\left(\frac{z}{Z_o}\right) = LHR^o \cos\left[\frac{\pi}{2\gamma}\left(\frac{z}{Z_o}-1\right)\right] = LHR^o F\left(\frac{z}{Z_o}\right) \qquad T_{cool} - T_{cool}^{in} = \frac{1}{1.2}\frac{Z_o \times LHR^o}{\dot{m}C_{PW}}\left\{\sin(1.2) + \sin\left[1.2\left(\frac{z}{Z_o}-1\right)\right]\right\}$$

- Solve temperature profile for:
  - @ z=0.25, z=0.5, z=1
- Solve for centerline temperature vs time
  - Transient: Volumetric/Areal heating rate
    $LHR^0(t) = LHR^0*(1\text{-}EXP(-0.1*time))+50$ for up to t=100
  - @ z=0.25, z=0.5, z=1
- Find location of peak centerline temperature at steady-state and at t=100 in transient

LIMITING PHENOMENA

Limiting Phenomena

- Concerning limiting phenomena, some criteria have been established for the UO2 and MOX fuel designs
- Engineering must demonstrate that all relevant parameters fulfill those criteria at any time from the loading of the fuel to reprocessing, or during long-term storage
- The key performance limiting phenomena are Pellet-clad mechanical interaction; Cladding elongation and assembly bow; Cladding oxidation and hydrogen pickup; Cladding wear; Power to melt; Fuel rod internal pressure; Departure from nucleate boiling; Normal operation limits

Limiting Phenomena

- PCMI
- PCMI is a complex process with a maximum risk for failure when the fuel pellet to cladding gap closes firmly and the reactivity of the fuel is still high
- The risk is enhanced by pellet fragments inducing a local shear strain on the cladding, and by the chemical interaction kinetic at the interface
- In order to prevent SCC, the cladding hoop stress calculated for normal operation and transients is limited
- The extent of the total permanent hoop strain is limited during the whole lifetime of the fuel rods, typically to 1%

Limiting Phenomena

- Cladding elongation and assembly bow
- During irradiation, the anisotropic character of the cladding material and the preferential migration of vacancies and interstitials in specific lattice planes drive an overall cladding axial growth, activated by the fast neutron flux
- When contact is established between the pellet and the cladding, pellet axial elongation causes an additional axial cladding strain
- This can lead to fuel rod bow with pitch reduction between the rods, reducing thermal margins
- Differential elongation of guide tubes in a PWR assembly can lead to an overall assembly bow

Limiting Phenomena

- Cladding oxidation and hydrogen pickup
- For the ZrO_2 formation at the cladding waterside surface, a typical criterion is related to the ASTM criterion of a maximum cladding wall thickness reduction of 10%, which corresponds to an oxide thickness of the order of 100 microns
- When the hydrogen concentration in the cladding exceeds the solubility limit, 70–100 ppm by weight at operating temperatures, zirconium hydrides will form
- The impact of hydrides on key mechanical properties depends strongly on hydride distribution and orientation
- Oxidation and hydrogen pickup are increasingly important at higher exposures, as the dependence on burnup is nonlinear

Limiting Phenomena

- Cladding wear
- The criterion for cladding wear at the contact points between grid spring/dimples and the fuel rod is often also related to the ASTM criterion of a maximum cladding wall thickness reduction of 10%
- More wear is technically acceptable, as evidenced from operational experience

Limiting Phenomena

- Power to melt
- The use of uranium dioxide or MOX provides a comfortable power to melt margin
- The melting temperature decreases slightly with burnup, but remains above 2750C
- At high burnup, above 50 MWd/kgM, considering the fuel thermal conductivity decrease, the power to melt was estimated to be around 600 W/cm, which is an unrealistic high LHR

Limiting Phenomena

- Fuel rod internal pressure
- Significant reopening of the radial gap between the fuel stack and the cladding must be avoided to ensure at the heat transfer to the coolant
- If a gap opens, fuel overheating and excessive fission gas release can occur, ultimately leading to fuel failure
- The original criterion required that the rod inner pressure must never exceed the outer coolant pressure
- This criterion was over-conservative and has been replaced by a ‘nonlift-off’ criterion, where the radial creep-out of the cladding (driven by gas pressure in excess of the system pressure) must never exceed the expansion rate of the pellet
- Experiments have shown that a large overpressure of the gas (considerably more than 5.0 MPa) is needed to initiate the reopening

Limiting Phenomena

- Departure from nucleate boiling
- With increasing heat flux there comes a point at which the heat transfer from a fuel rod rapidly decreases due to the insulating effect of a steam blanket that forms on the rod surface, resulting in a severe increase of cladding temperature and possibly cladding failure
- The ratio of the heat flux needed to cause departure from nucleate boiling (DNB) at given local coolant properties (pressure, enthalpy, mass flow rate) to the actual local heat flux of a fuel rod is defined as the DNBR
- This phenomenon may limit the maximum allowed thermal power of a given PWR

Limiting Phenomena

- Normal operation limits
- Constraints on the axial LHR distribution are typically applied at the core design level and during normal operation to guarantee that the conditions are never worse than those assumed in scenarios considered in the accident analyses
- The maximum allowed LHR may depend on the axial position and on burnup, and can be reactor- and even cycle-specific
- The fulfilment of the constraint is verified during the reload safety evaluation process as well as during plant operation

Summary

- There are a variety of limiting phenomena in LWR fuel systems that provide the boundaries of operation and lifetime
- These limits include phenomena in the fuel, gap, cladding, corrosion, and assembly levels
- The performance of uranium dioxide and MOX fuels in LWR nuclear reactors is well established
- These fuels have demonstrated a very good behavior during irradiation, favored by their high melting temperature, providing large operating temperature margins

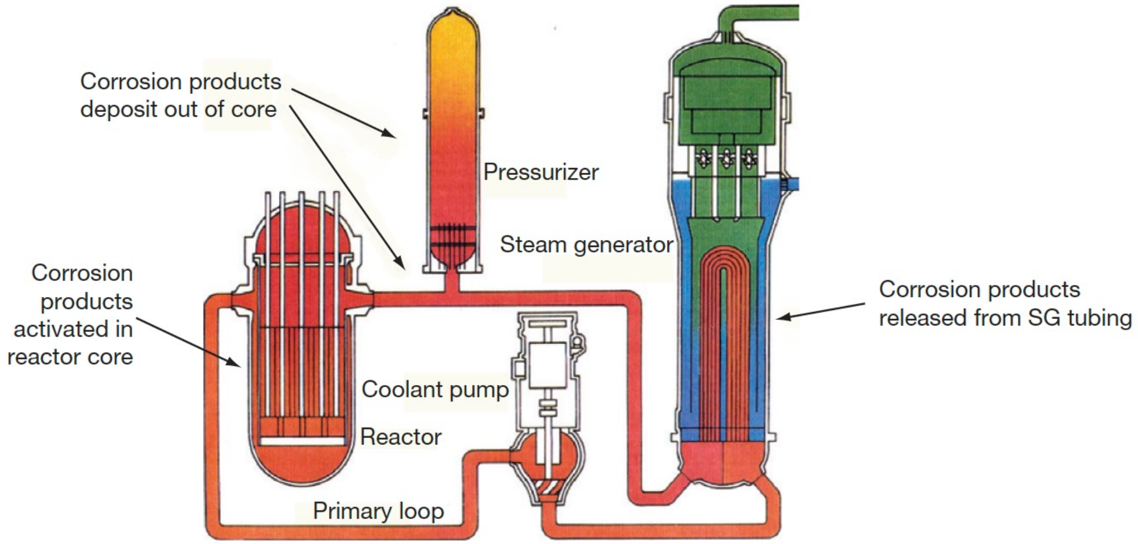
WATER CHEMISTRY

Water Chemistry

- Excellent water quality is essential if material degradation is to be controlled
- Primary system water chemistry affects fuel performance through the deposition of corrosion products on fuel pin surfaces
- Chlorides and sulfates are particularly aggressive in increasing intergranular stress corrosion cracking (IGSCC) and other corrosion processes
- Initial efforts to improve water quality brought about a slow but steady reduction in impurities through improved design and operation of purification systems
- Excellent water chemistry alone is not sufficient to control corrosion, thus programs to modify water chemistry, including minimizing oxygen to reduce the electrochemical corrosion potential (ECP) in BWRs, and oxygen and pH control in PWRs, have been implemented
- Water chemistry advances are now an important part of the overall operating strategy to control material degradation

PWR Water Chemistry

- In the very early days of PWR operation, heavy crud buildup on fuel cladding surfaces was caused by the transport of corrosion products from the steam generators into the reactor core
- Activated corrosion products caused high-radiation fields on out-of-core surfaces fuel performance was compromised, and even coolant flow issues were observed



CRUD

- A corrosion product called Chalk River unidentified deposit (CRUD) accumulates on the Ni alloy and stainless-steel surfaces
- CRUD is an accumulation of materials and corrosion products that is composed of either dissolved ions or solid particles such as Ni, Fe, and Co on fuel rod cladding surfaces in NPPs
- CRUD degrades heat production by nuclear fuel because it is slowly eroded by the circulation of the hot pressurized water and later deposited on the cladding or outer housing of fuel rods
- The chemical composition of CRUD varies depending on the types of refueling cycles and the constituents of the basic metal material.
- Irradiation can produce radionuclides in the CRUD, such as 60Co and 63Ni

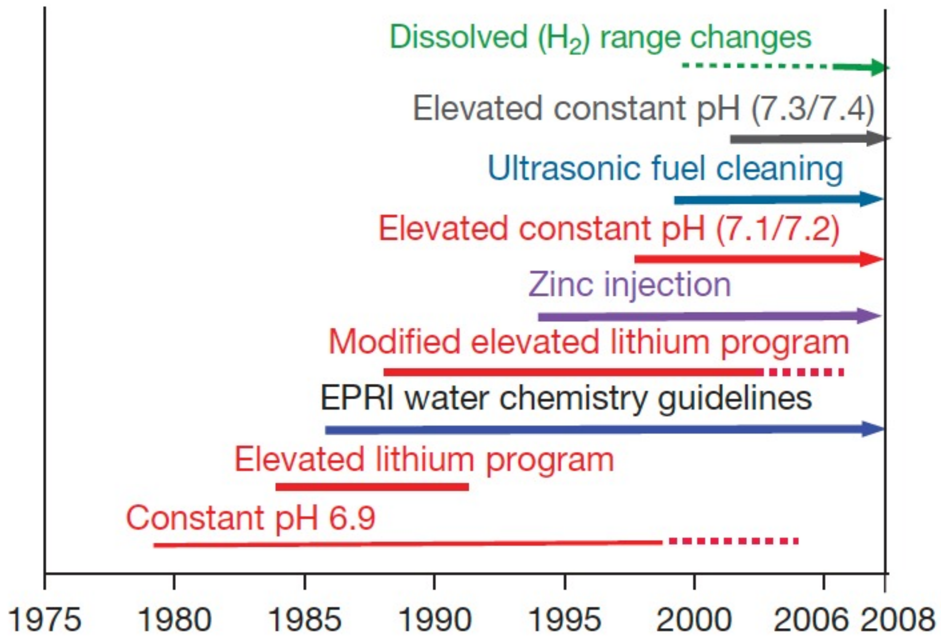
PWR Water Chemistry

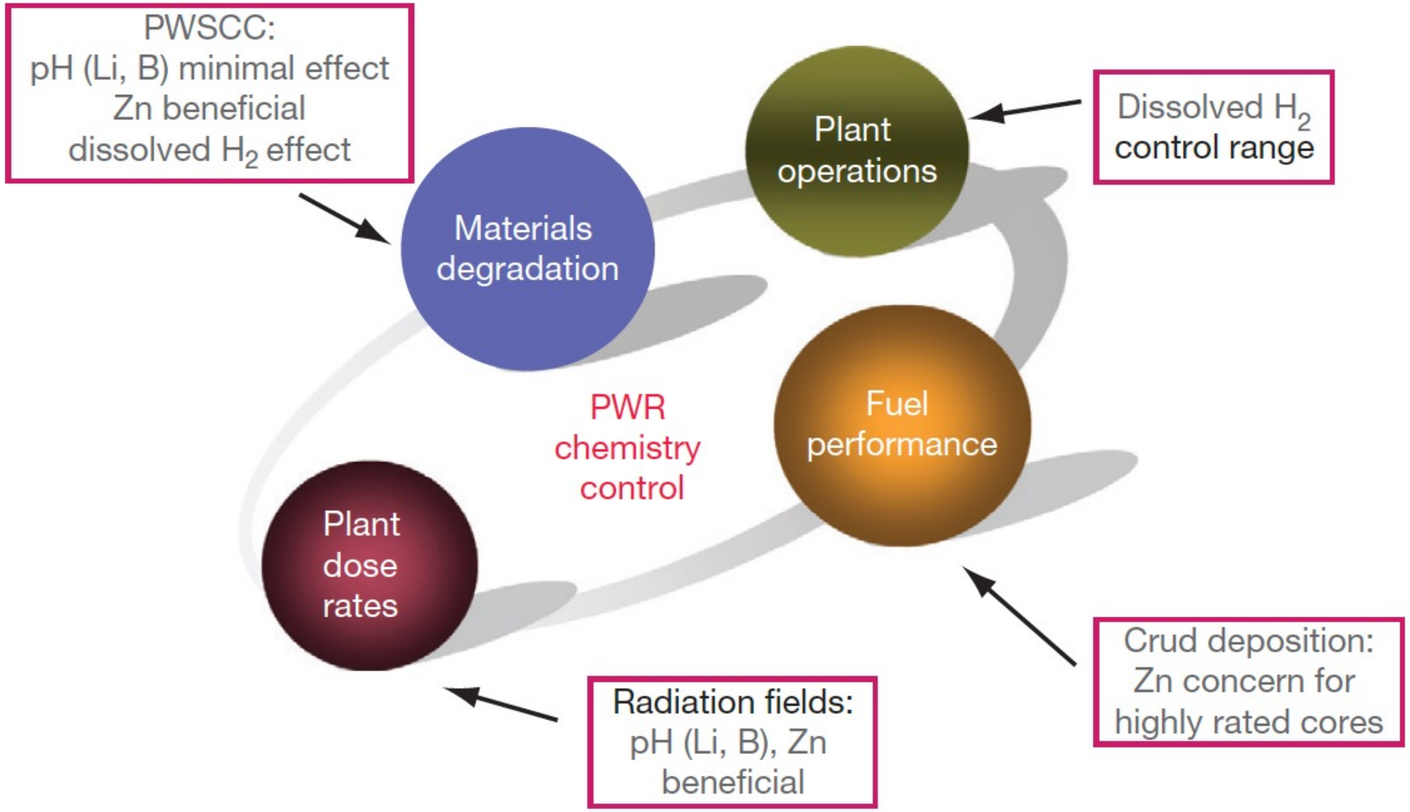
- PWR problems were initially mitigated by imposing a hydrogen overpressure on the primary system, reducing the corrosion potential, and raising the primary chemistry pH
- Commercial PWR power plants use a steadily decreasing concentration of boric acid as a chemical shim (for reactor control) throughout the fuel cycle, which results in the use of lithium hydroxide to control pH
- The concept of ‘coordinated boron and lithium’ was developed, whereby the concentration of LiOH was gradually reduced in line with the boric acid reduction to maintain a constant pH
- It was determined that heavy fuel crud buildup was avoided if a constant pH of at least 6.9 was maintained
- Zinc injection is utilized to reduce radiation fields, and also inhibits SCC

Radiation Control

- Corrosion products deposited on the fuel become activated, are released back into the coolant, and may be deposited on out-of-core surfaces
- During shutdowns, the major radiation source for personnel exposure is activated corrosion products, deposited on primary system surfaces
- The mechanism of the zinc ion effect is complex, as release of ^{60}Co from fuel crud is reduced, and deposition out-core is also reduced
- Aqueous zinc ion promotes the formation of a more protective spinel-structured corrosion film on stainless steel, especially when reducing conditions are present
- Both cobalt and zinc favor tetrahedral sites in the spinel structure, but the site preference energy favors zinc incorporation
- The ^{60}Co remains longer in the water and is eventually removed by the cleanup system

PWR Water Chemistry





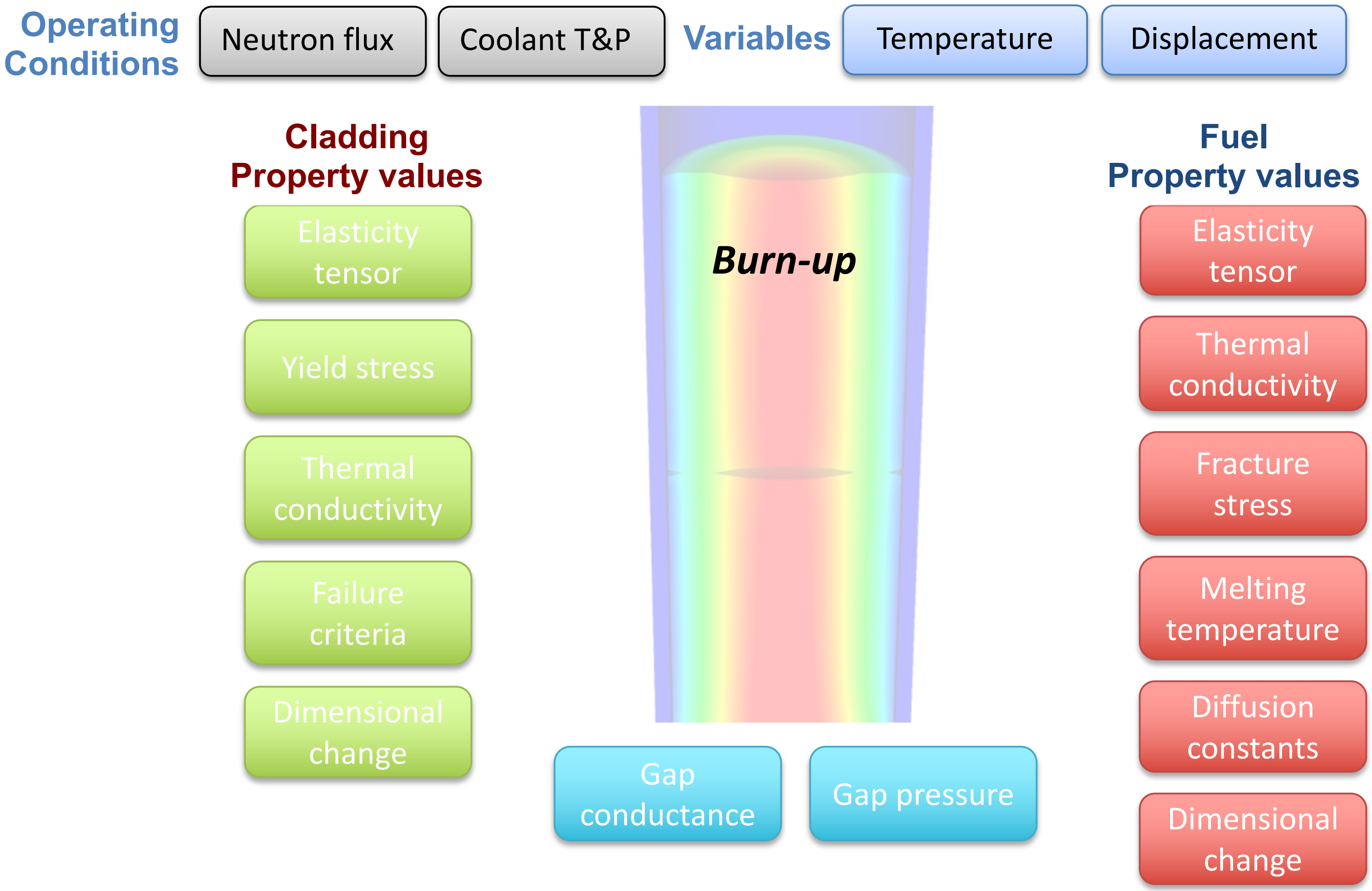
Summary

- Brief overview of water chemistry concerns
- Primary system water chemistry affects fuel performance through the deposition of corrosion products on fuel pin surfaces
- Control measures such as dissolved H2 and balancing LiOH to boron content are utilized to control the pH
- Zinc injection is utilized to control radiation fields

MECHANISTIC FUEL PERFORMANCE MODELING

Going beyond burnup…

- Fuel performance codes historically rely on materials models correlated to temperature and burn-up
- Development has begun on models based on microstructure rather than burn-up

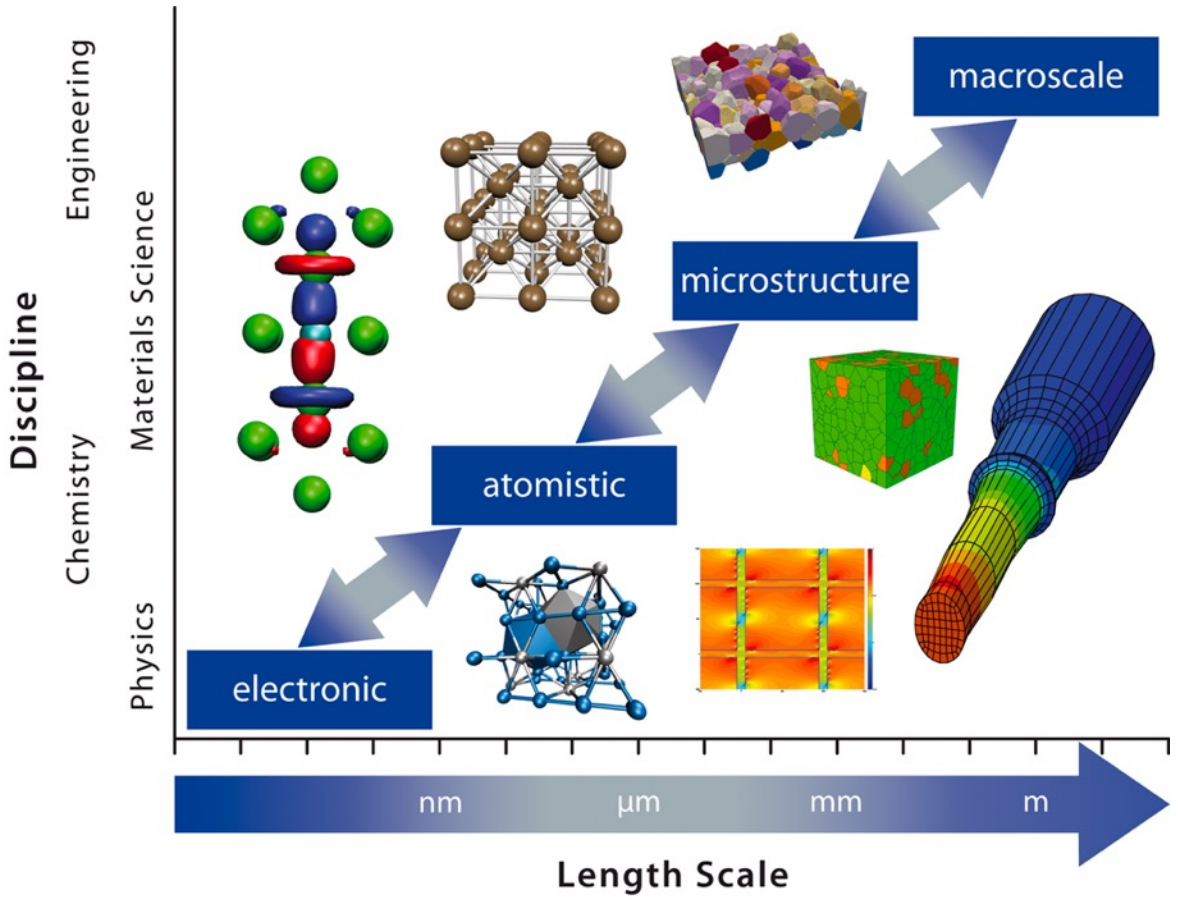


Microstructure/Mechanistic/Multiscale

- Mechanistic: relating to theories which explain phenomena in purely physical or deterministic terms
- There is a drive for fuel performance codes to employ mechanistic materials models that are based on the current state of the evolving microstructure rather than burn-up
- A series of state variables define the current state of the microstructure, and the evolution of these state variables is defined by mechanistic models that are functions of fuel conditions and other state variables
- The material properties of the fuel and cladding are determined from microstructure/property relationships that are functions of the state variables and the current fuel conditions
- Multiscale modeling and simulation is being used in conjunction with experimental data to inform the development of these models
- This mechanistic, microstructure-based approach has the potential to provide a more predictive fuel performance capability

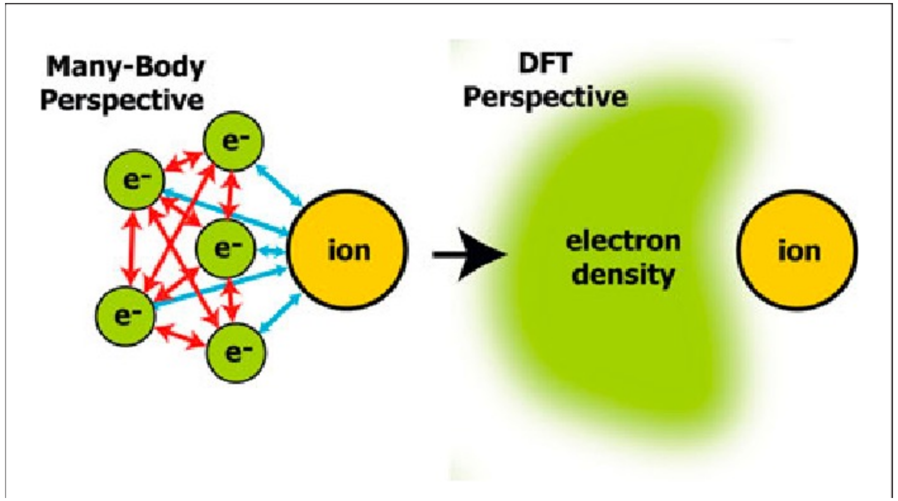
Multiscale Modeling

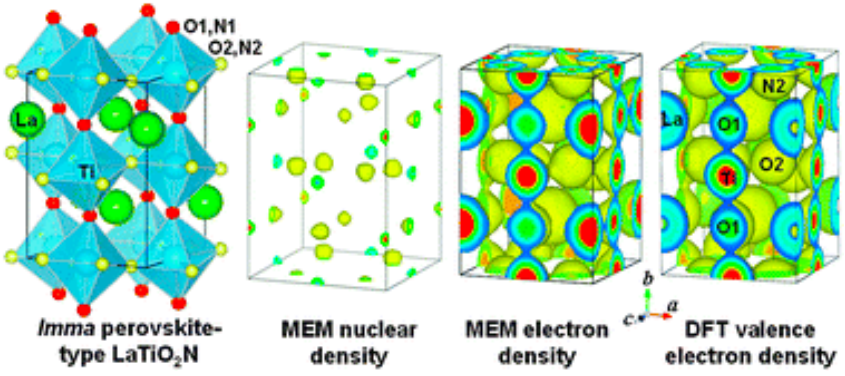
- Modeling and simulation is the essential bridge from good science to good engineering, spanning from fundamental understanding of materials behavior to deliberate design of new materials technologies leveraging new properties and processes
- Lower length scale modeling can provide insight, properties, mechanisms, behaviors, etc., that can be input into higher length scale modeling tools to describe, mechanistically, macroscale behavior



Density Functional Theory

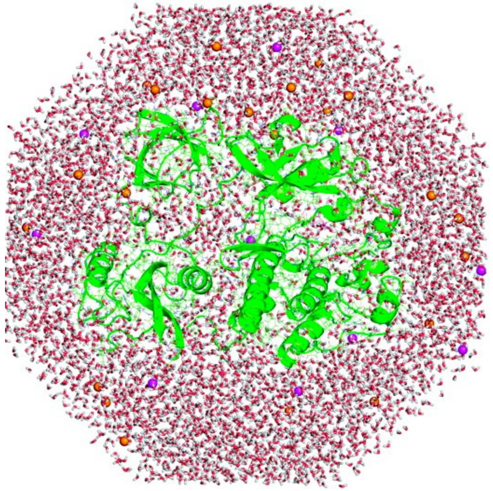
- DFT is a computational quantum mechanical modelling method used to investigate the electronic structure of many-body systems, in particular atoms, molecules, and the condensed phases
- DFT is primarily utilized to investigate electronic structure, cohesive energy, elastic constants, phonons, entropies, etc.

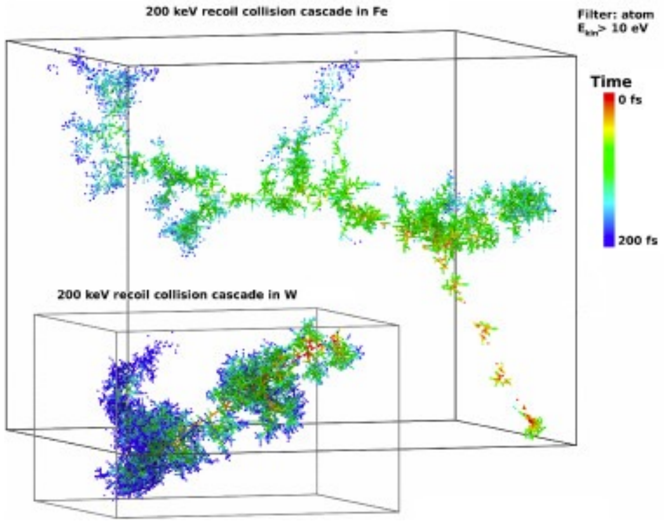




Molecular Dynamics

- MD is a computer simulation method for analyzing the physical movements of atoms and molecules, determined by numerically solving Newton's equations of motion for a system of interacting particles
- MD is still atomistic, and is often utilized to calculate transport properties, melting temperature, clustering, etc., that are properties just beyond DFT capabilities





Rate Theory/Cluster Dynamics

- Provides recipes for calculating reaction rates between individual species of the types which are ubiquitous in chemistry and physics
- Is typically a mean-field approach which uses transition state theory as a tool for describing reactions involving radiation-produced defects
- Can be utilized to describe clustering, absorption, emission, growth, resolution, etc. to describe microstructural phenomena on an intermediate time scale

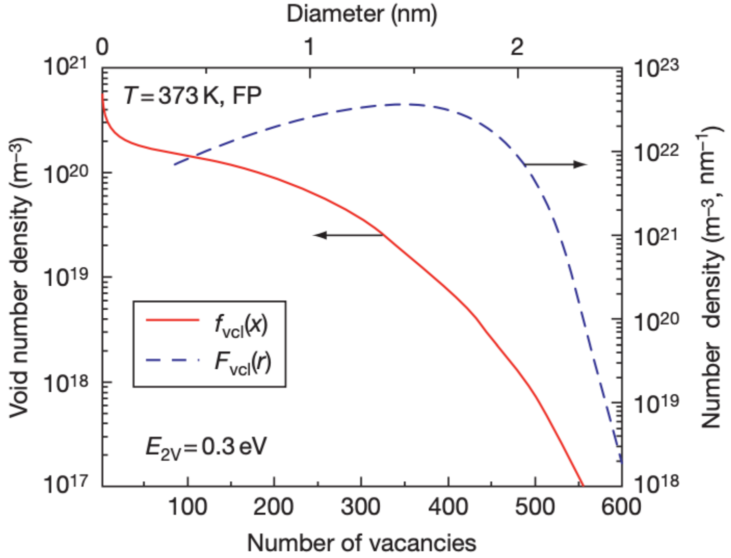
$$\nu(\tilde{E}) = 0.8\frac{E^{\mathrm{PKA}}(\tilde{E})}{2E_{\mathrm{d}}}$$

$$G_{\mathrm{v}} = G^{\mathrm{NRT}}(1-\varepsilon_{\mathrm{r}})(1-\varepsilon_{\mathrm{v}})$$

$$G_{\mathrm{i}} = G^{\mathrm{NRT}}(1-\varepsilon_{\mathrm{r}})(1-\varepsilon_{\mathrm{i}})$$

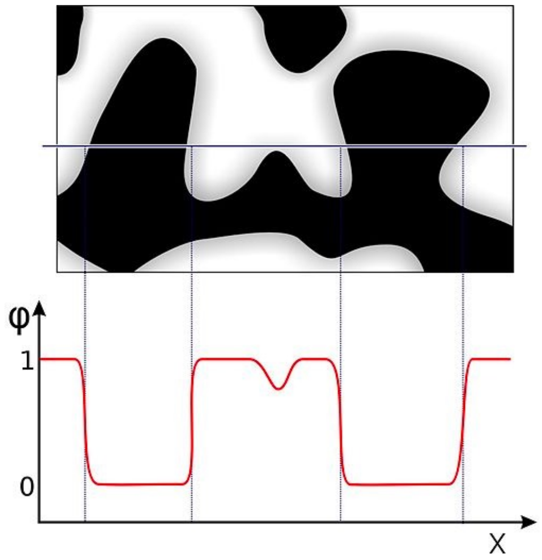
$$\frac{\mathrm{d}C_{\mathrm{v}}}{\mathrm{d}t} = G^{\mathrm{NRT}}(1-\varepsilon_{\mathrm{r}})(1-\varepsilon_{\mathrm{v}}) + G_{\mathrm{v}}^{\mathrm{th}} - k_{\mathrm{v}}^2 D_{\mathrm{v}} C_{\mathrm{v}} - \mu_{\mathrm{R}} D_{\mathrm{i}} C_{\mathrm{i}} C_{\mathrm{v}}$$

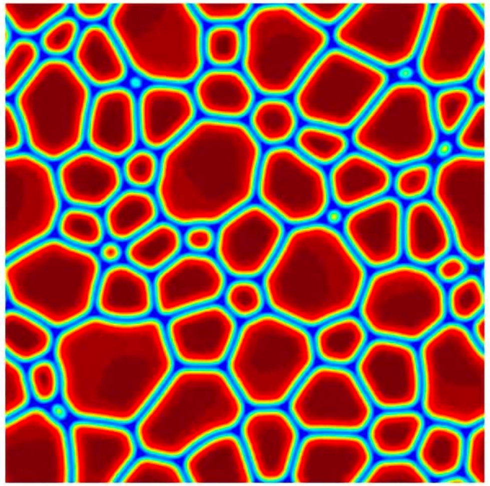
$$\frac{\mathrm{d}C_{\mathrm{i}}}{\mathrm{d}t} = G^{\mathrm{NRT}}(1-\varepsilon_{\mathrm{r}})(1-\varepsilon_{\mathrm{i}}) - k_{\mathrm{i}}^2 D_{\mathrm{i}} C_{\mathrm{i}} - \mu_{\mathrm{R}} D_{\mathrm{i}} C_{\mathrm{i}} C_{\mathrm{v}}$$



Phase-Field

- PF is mathematical model for solving interfacial problems that substitutes boundary conditions at the interface by a partial differential equation for the evolution of an auxiliary field (the phase field) that takes the role of an order parameter
- The order parameter takes two distinct values (for instance 1 and 0) in each of the phases, with a smooth change between both values in the zone around the interface, which is then diffuse with a finite width
- PF is commonly used for grain growth, phase separation, bubble coalescence, recrystallization, etc.





Microstructure-based fuel performance modeling

Structure/property relationships connect the microstructure variables to the property values

Operating Conditions: Neutron flux | Coolant T&P

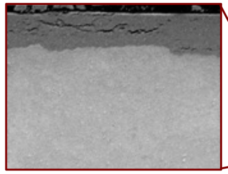
Variables: Temperature | Displacement | Stoichiometry

Cladding Property values

- Elasticity tensor
- Yield stress
- Thermal conductivity
- Failure criteria
- Dimensional change

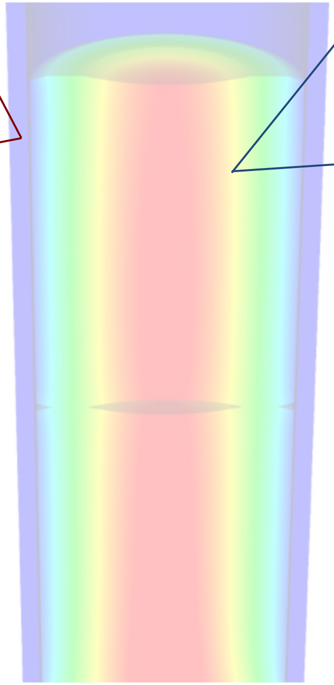
Microstructure evolution models

- Grain growth
- Plasticity
- Defect prod. & transport
- Hydrogen transport
- Loop evolution



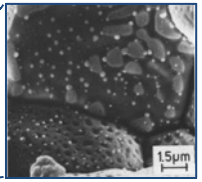
Cladding Microstructure variables

- Average grain size
- Dislocation density
- H concentration
- Hydride fraction
- Point defect concentrations
- Loop density
- 2nd phase particle fraction



Gap variable

- Gap fission gas concentration



Fuel Microstructure variables

- Average grain size
- Dislocation density
- U defect concentrations
- Pu concentration
- Fission product precipitate fractions
- Dissolved fission products
- Fission gas bubble fraction
- Grain boundary fission gas bubble coverage
- Crack parameter

Microstructure evolution models

- Grain growth
- Plasticity
- Defect prod. & transport
- Fission gas behavior
- Densification
- Fracture

Fuel Property values

- Elasticity tensor
- Thermal conductivity
- Fracture stress
- Melting temperature
- Diffusion constants
- Dimensional change

Summary

- Researchers are working to develop materials models for the fuel and cladding that are mechanistic rather than empirical and that are based on the evolution of the microstructure rather than the burnup.