Nuclear Fuel Concepts

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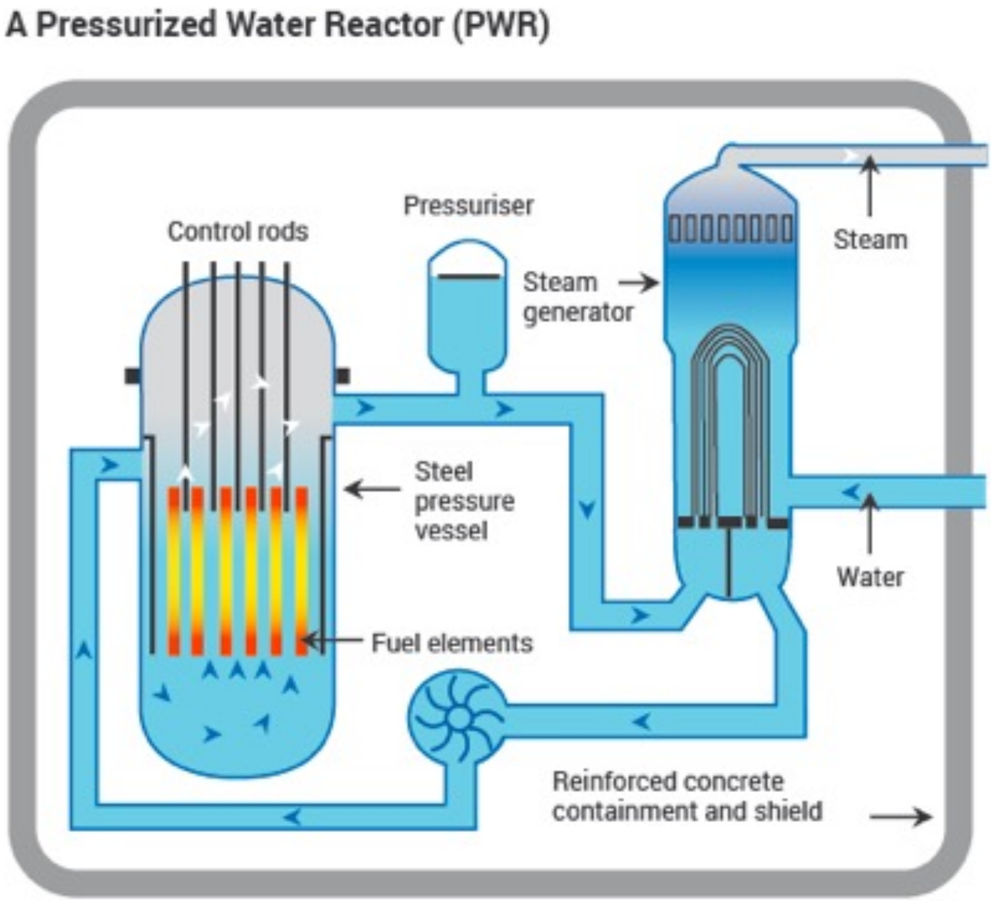
Who am I?

- From rural western Kentucky
- PhD in Nuclear Engineering from Georgia Tech
- Worked at Idaho National Laboratory before joining NCSU 4 years ago

- I am a computational nuclear materials scientist
  – Computational tools
  – Geared for material science research
  – Applied to nuclear materials (especially advanced fuels)
  – Spans physics, chemistry, and engineering

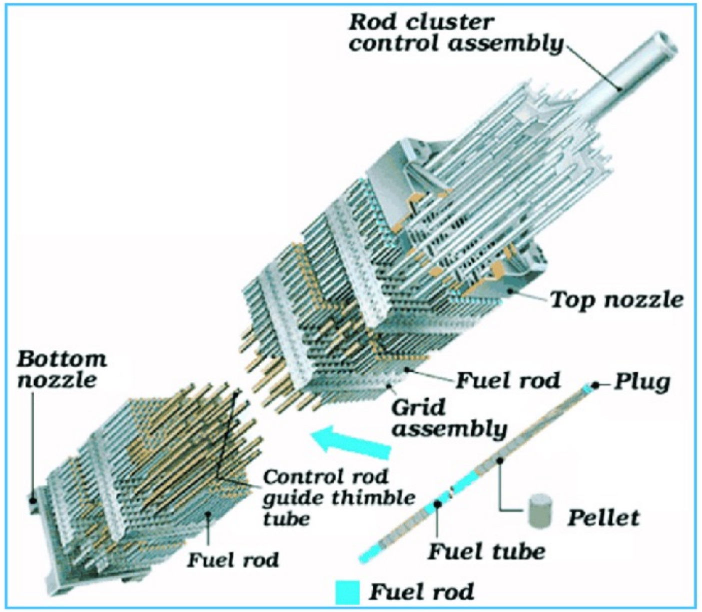
Fuel is the Heat Source

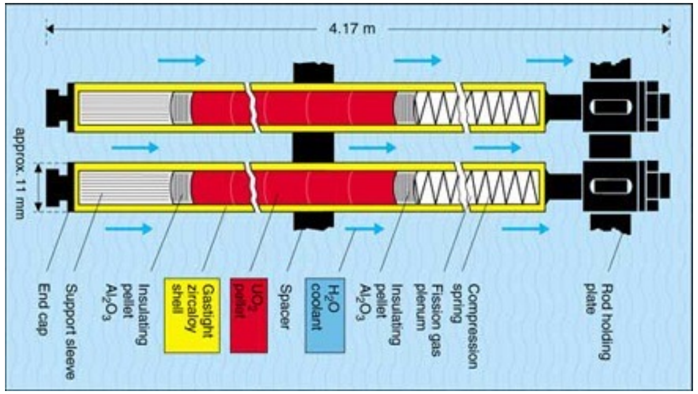
- Fuel functions as a heat source, generating heat that is transferred to a coolant, which transfers heat through external loops, heat exchangers, etc., to run a generator
- While the fuel must meet neutronic requirements, limiting possible options of materials choice, the lifetime of the fuel is governed by material constraints
- What I care about is how the fuel material evolves/behaves as a function of time



Fuel System

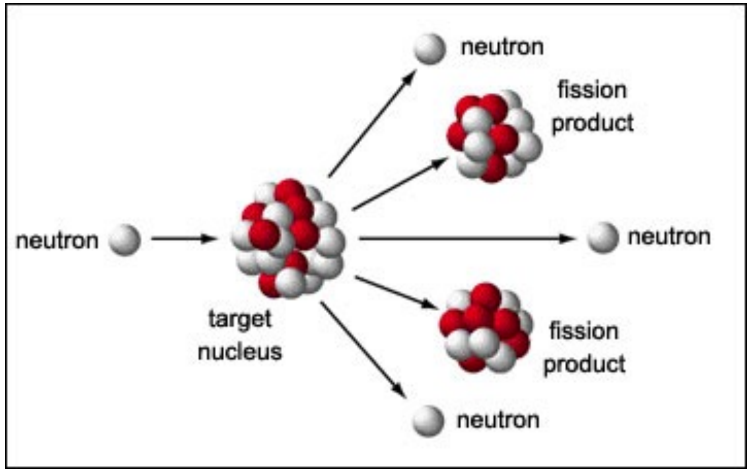
- Fissile material compound (fuel), surrounded by a cladding, surrounded by coolant
- Fuel generates your neutrons and heat, while cladding keeps the nasty stuff in the fuel from getting to the coolant
- Coolant transports the heat away to secondary systems
- Coolant can also reduce energy of (moderate) neutrons
- I focus on the fuel here, but this concept of a fuel system is more complete





Fuel is limited to select elements

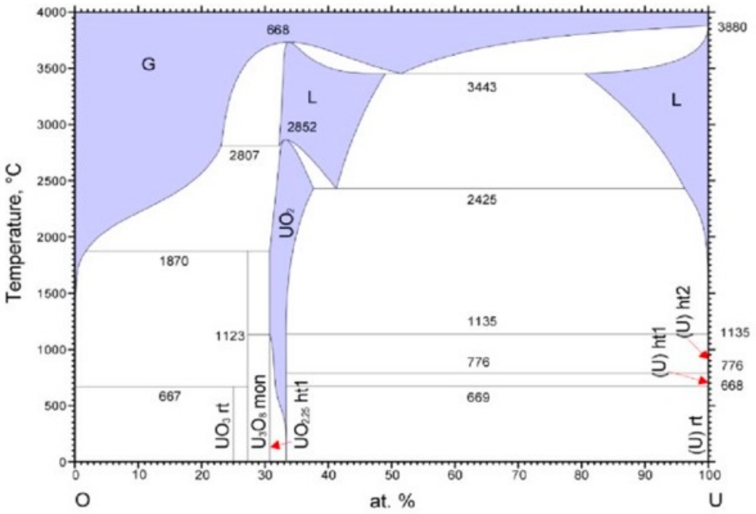
- A **fissionable nuclide** is capable of undergoing fission (even with a low probability) after capturing a high energy neutron
- A **fissile nuclide** is capable of sustaining a nuclear fission chain reaction with neutrons of any energy
- A **fertile material** is a material that, although not itself fissionable, can be converted into a fissile nuclide by neutron absorption and subsequent nuclei conversions.
- U-235 is a fissile isotope that naturally occurs in uranium in small amounts (0.7%). Can be enriched.

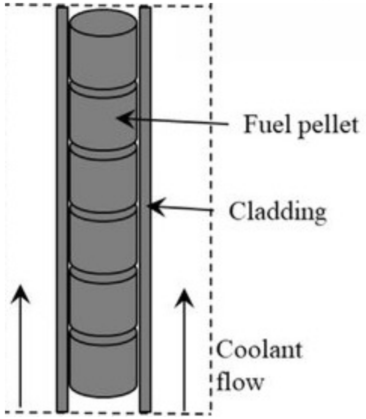


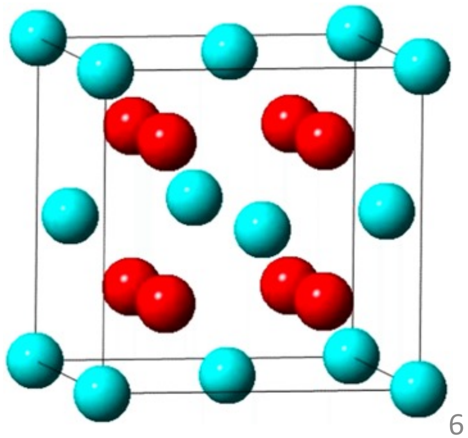
Neutron + Target Nucleus -> Two fission products, 2-3 neutrons; Fission releases around 210 MeV of energy

Uranium Dioxide (UO2)

- Reference fuel for nuclear power industry
- Single phase, fluorite structure
- Fabrication via sintering UO2 powder into pellets
- Water coolant
- Pellets are stacked inside Zircaloy cladding tubes (zirconium alloy)
- Good behaviors
  - Very high melting point, about 2800 C
  - Maintains a stable fluorite phase up to melting
  - Very compatible with Zircaloy clad Relative stability in water
  - Reasonably radiation resistant







Uranium Dioxide (UO2)

- Bad behaviors
  - Brittle (thermal stress fractures, fragmentation)
  - Poor thermal conductivity
  - Properties very sensitive to stoichiometry
  - Limited linear heating rates
  - Non-negligible thermal expansion/swelling
  - Higher stored thermal energy than other fuel material

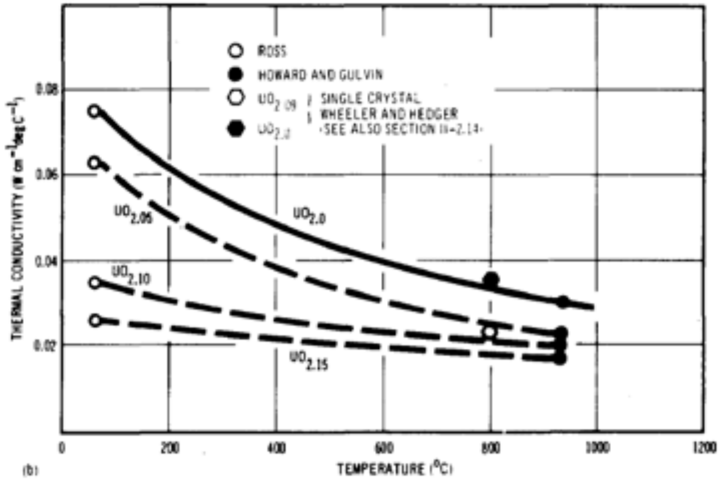
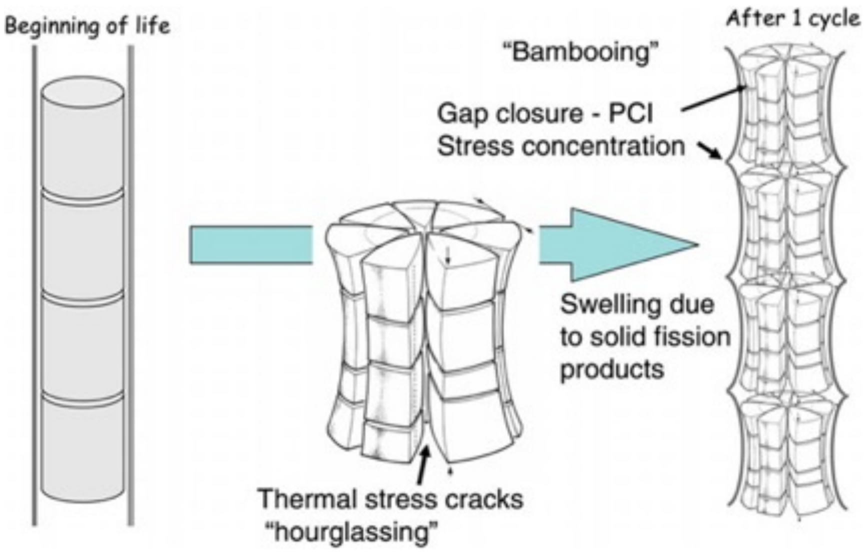
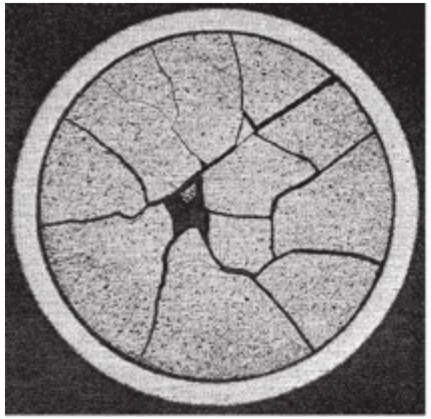


Fig. 9. Effect of hyperstoichiometry on conductivity.

Uranium Dioxide (UO2)

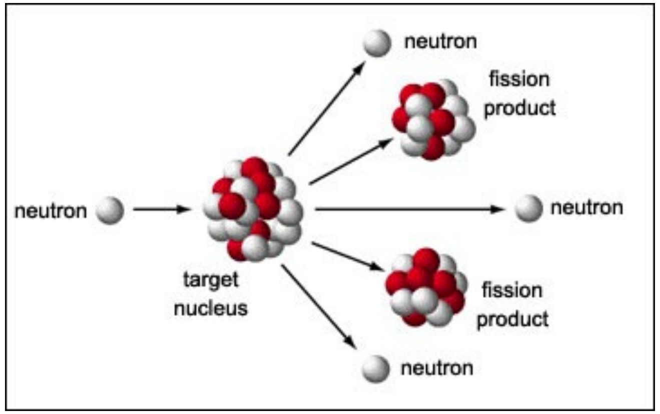
- Critical Phenomena
  - Thermal conductivity (and degradation)
  - Fission gas release, leading to pressure increase inside the cladding
  - Fuel fragmentation and relocation under transients
  - Bambooing creating stress concentrations
- All 98 operational reactors in US utilize UO2
  - we sufficiently understand its limitations, and don't push it beyond

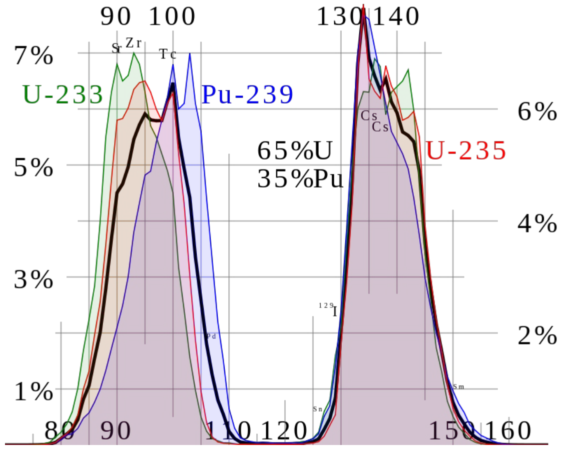




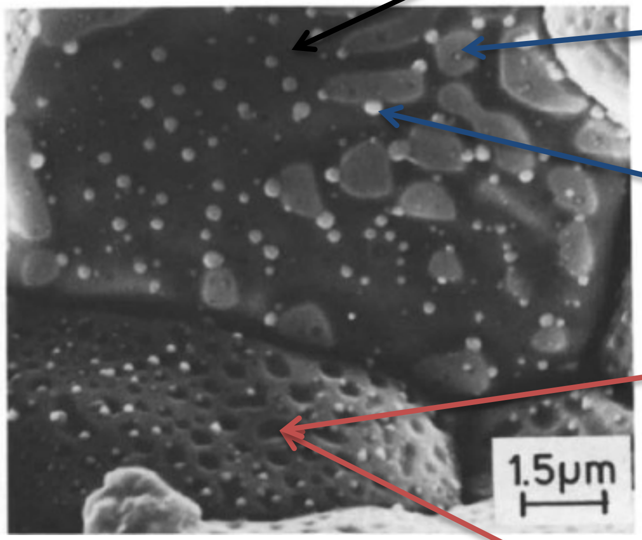
Fission Product Generation

- 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 microstructure





Various types of fission products in the fuel



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

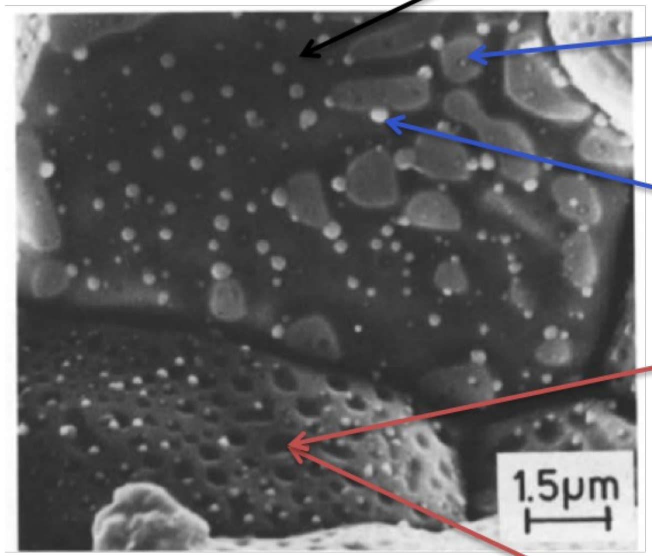
Insoluble oxides (Zr, Ba and Sr)
- Form insoluble oxides in the fluorite lattice

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

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

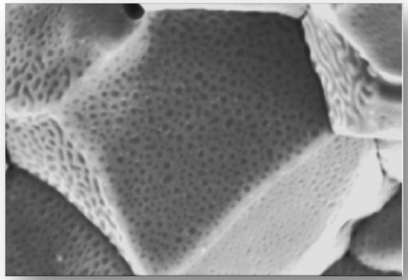
Metals (Mo, Ru, Pd, and Tc)
- Slightly raise thermal conductivity,

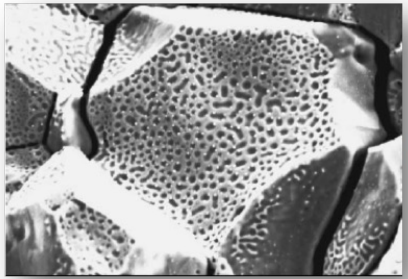
Volatiles (Br, Rb, Te, I and Cs)
- Cause swelling, decrease thermal conductivity
- Escape from fuel, corrode the cladding

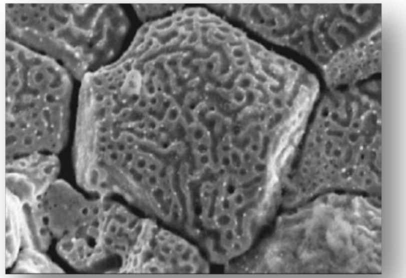
Noble gases (Xe, Kr)
- Cause swelling
- Decrease thermal conductivity
- After release, raise gap pressure and lower thermal conductivity

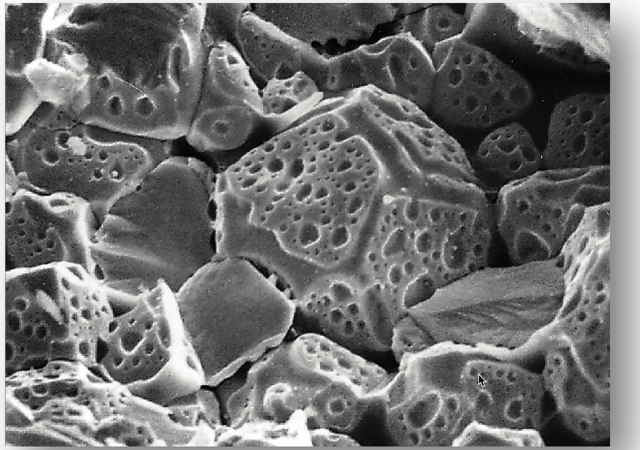
Fission Gas Release

- 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
- Stage 2: Gas bubbles nucleate on grain boundaries, growing and interconnecting
- Stage 3: Gas travels through interconnected bubbles to a free surface


Nucleation


Growth


Percolation

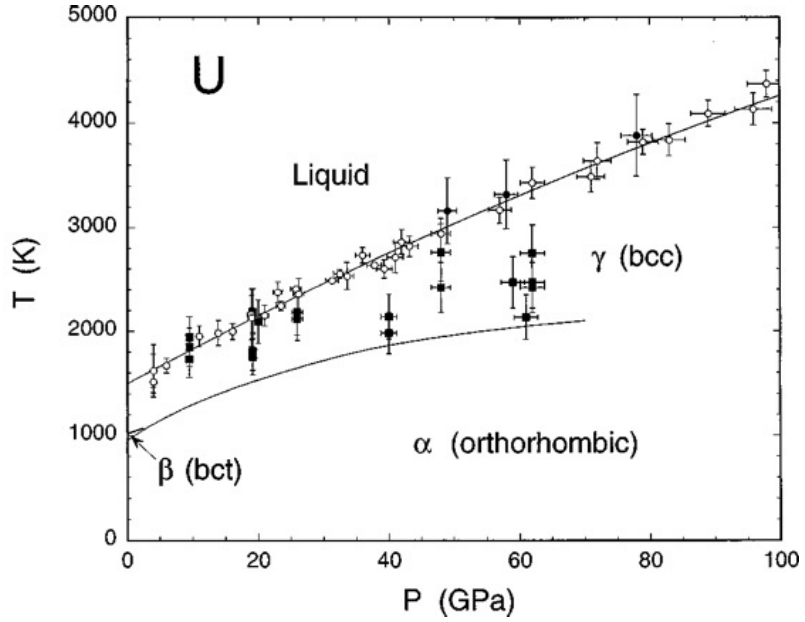


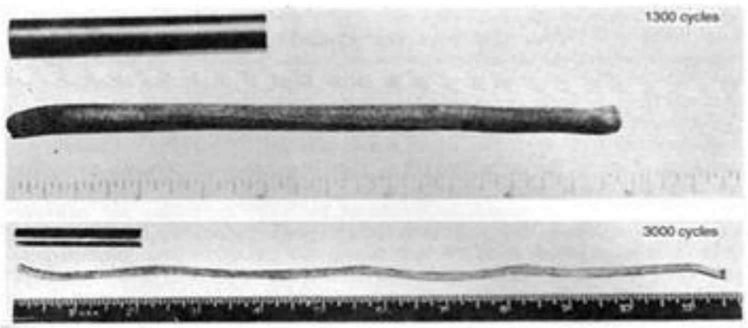
Advanced Fuel Types and Associated Reactor Types

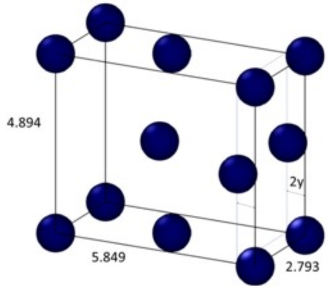
- Mixed Oxide (MOX) and Accident tolerant fuel (ATF) – Light Water Reactors
- UZr – Sodium Cooled Fast Reactors
- UMo – Research Reactors
- UC/UCO – High Temperature Gas Reactors
- UF4/UCl3 – Molten Salt Reactors

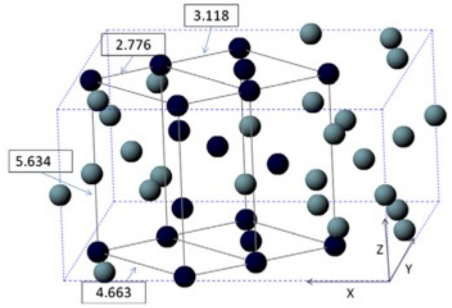
Why not use pure uranium metal?

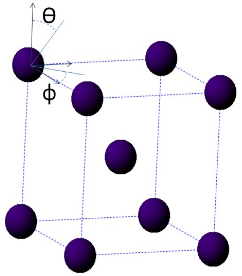
- Pure uranium has three solid phases
  - α-phase is orthorhombic
  - β-phase is tetragonal
  - γ-phase is body-centered cubic
  - During thermal cycling, pure uranium dramatically swells
  - α-U has both anisotropic thermal expansion and anisotropic irradiation growth





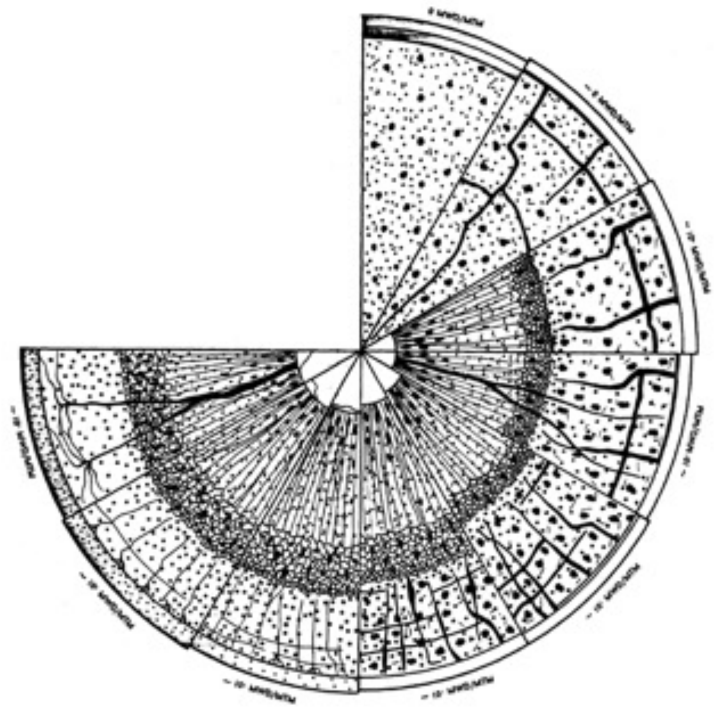






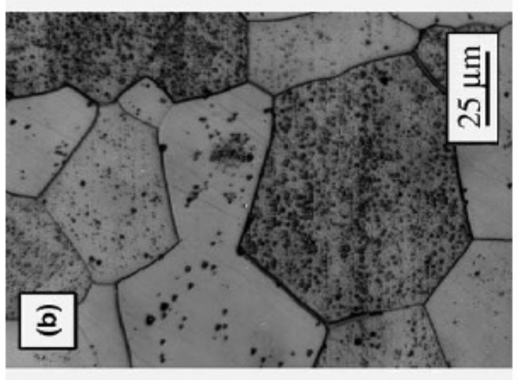
Mixed Oxides

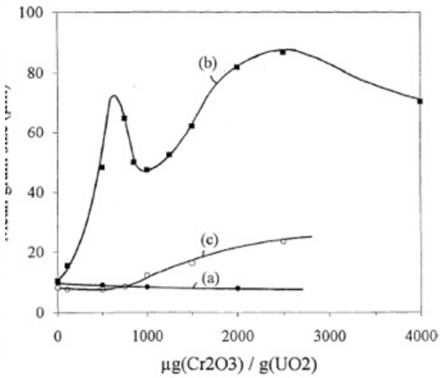
- UO2 can be combined with PuO2 for a mixed oxide (MOX) fuel for use in fast reactors
  - fast reactor uses higher energy neutrons
- Allows to burn excess weapons grade plutonium, or breed additional fuel isotopes
- Less common is inclusion of minor actinides in MOX to burn waste
- ~30 reactors in Europe currently utilize a partial MOX core
- Similar behavior to UO2, but different neutronics, fission gas release, thermal conductivity, etc.

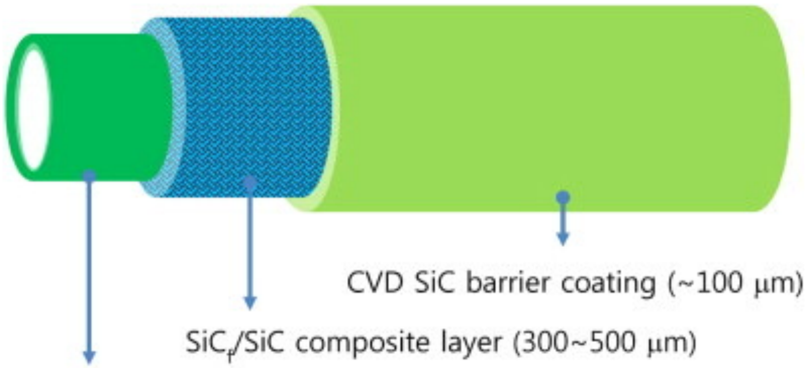


Accident Tolerant Fuel/Advanced Technology Fuel

- Cr-doped UO2
  – Larger grain size, theoretically reduced fission gas release
  – Cr changes the O potential present within the fuel, changing defect concentrations and mobilities
- Coatings or alternate claddings – FeCrAl or SiC
  – Improved radiation resistance, corrosion resistance, etc.

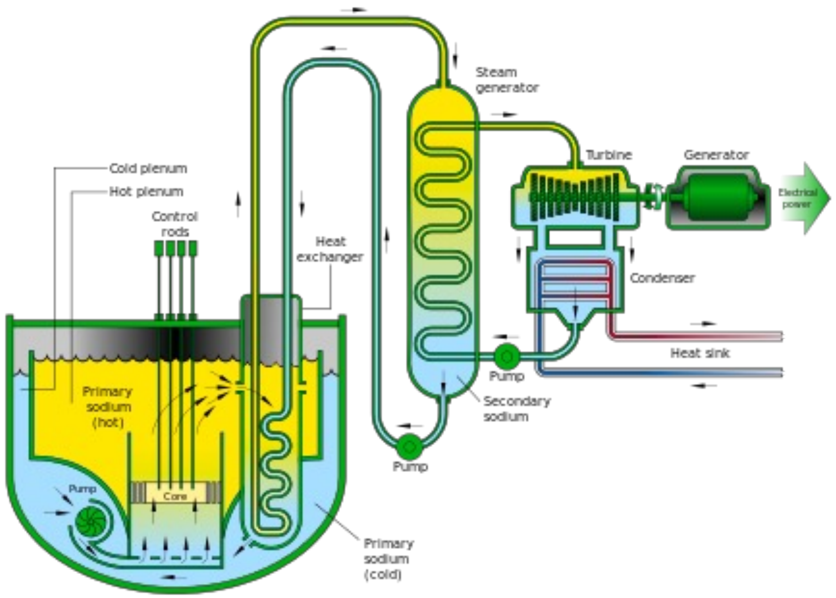


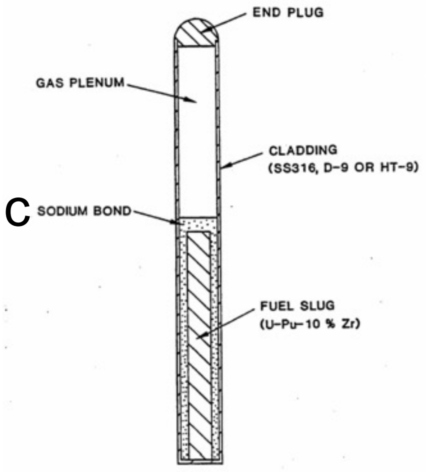


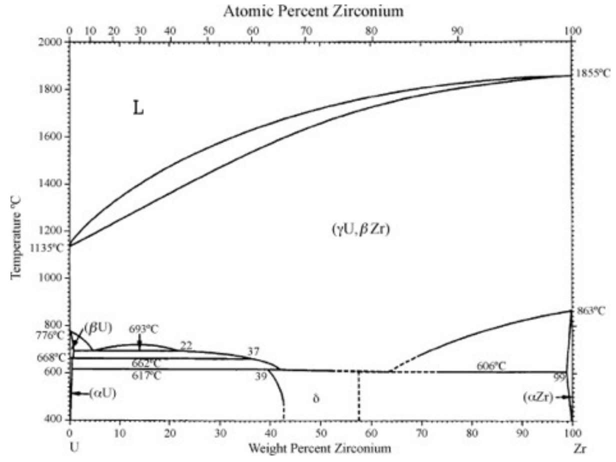


Uranium-Zirconium (UZr)

- Utilized in sodium cooled fast reactors (SFRs) - EBR I, EBR II
- Varied crystal structure and compositional environment
- Easily alloyed with Pu, minor actinides (MA)
- Can function as a breeder/burner fuel
- Sodium coolant
- Fe-based cladding
- Alloy with Zr to increase the melting point and to stabilize the high temperature body-centered cubic phase

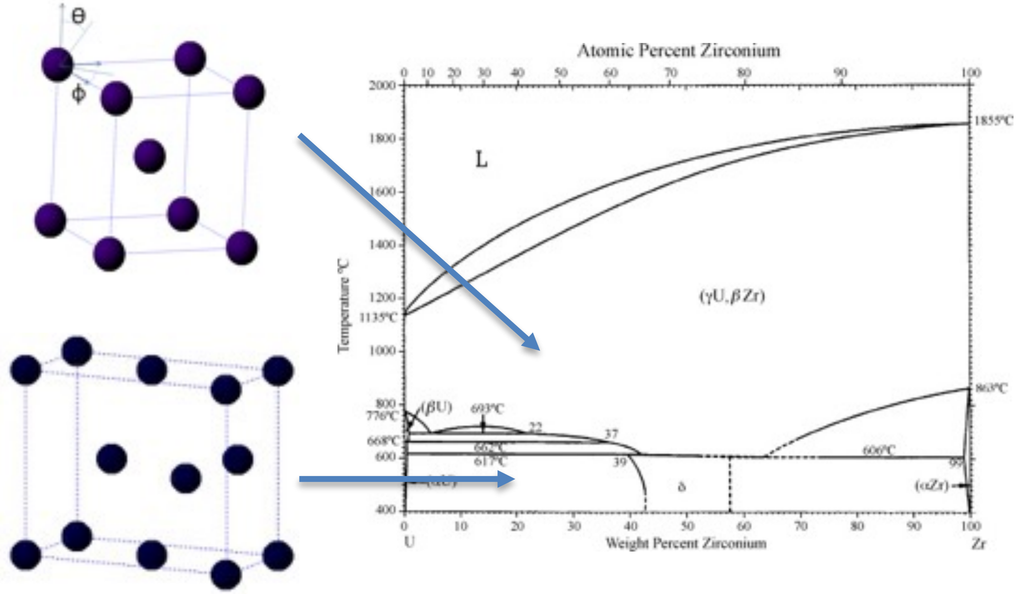


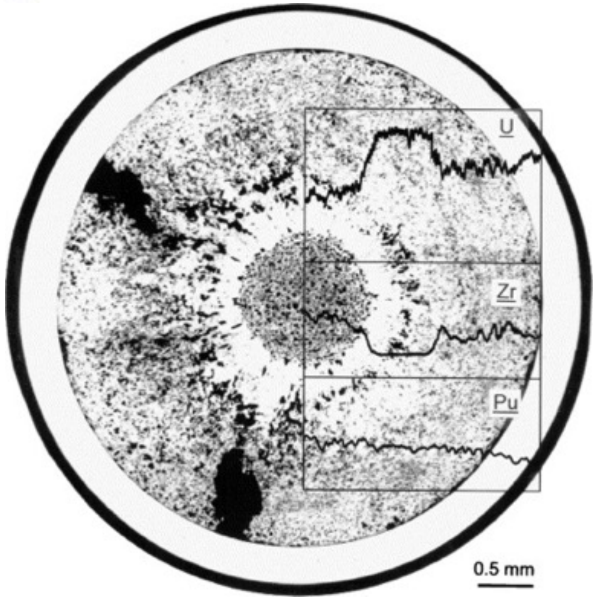




Uranium-Zirconium (UZr)

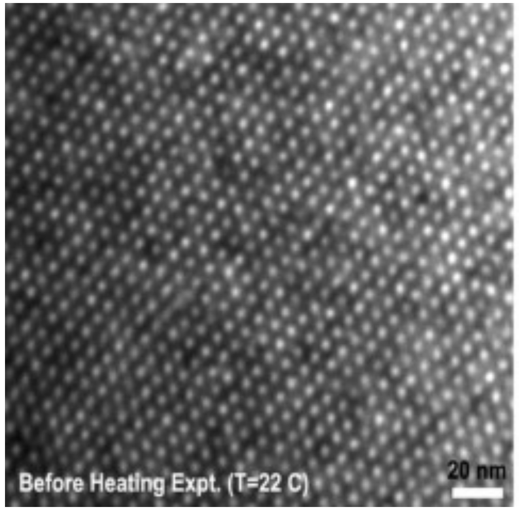
- Phenomena of interest
  - 30-50% swelling
  - Constituent redistribution
  - Alpha tearing
  - FCCI
- Good: High thermal conductivity; Stability to high burnups (> 20%); Flexible composition; Inherent safety- negative reactivity feedback, etc.
- Bad: Low melting point; Dramatic fuel swelling that must be accounted for; Incredibly complex microstructures; Fuel-Clad Chemical Interaction, Easily oxidized, etc.

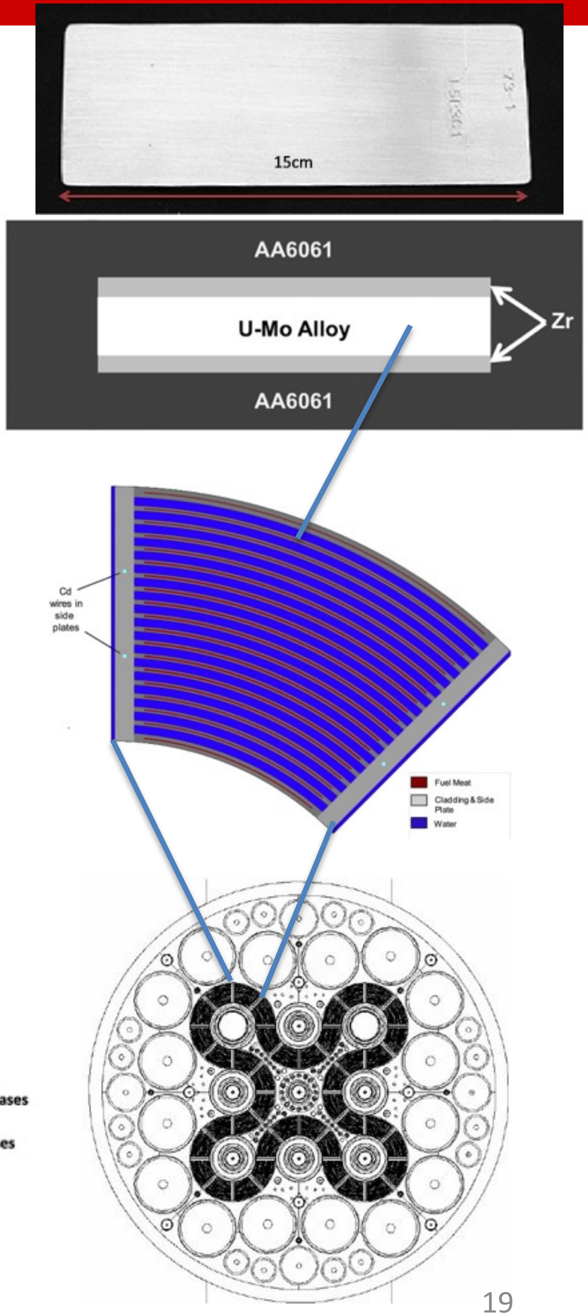


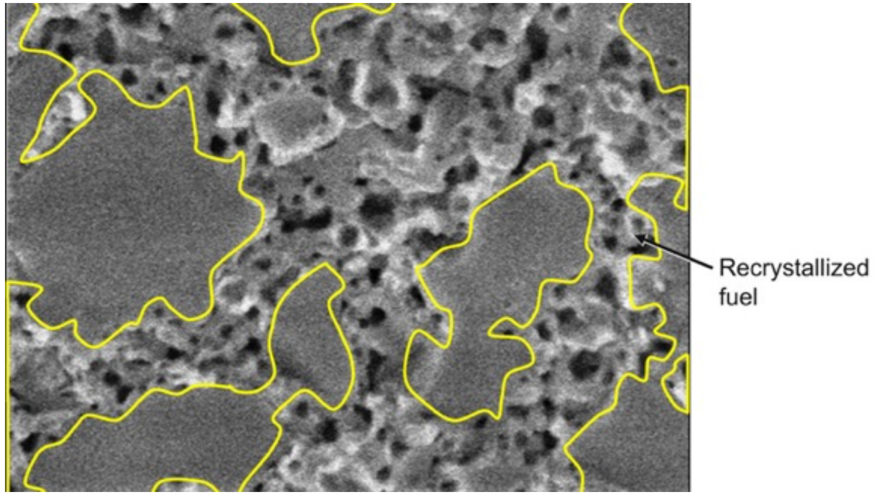


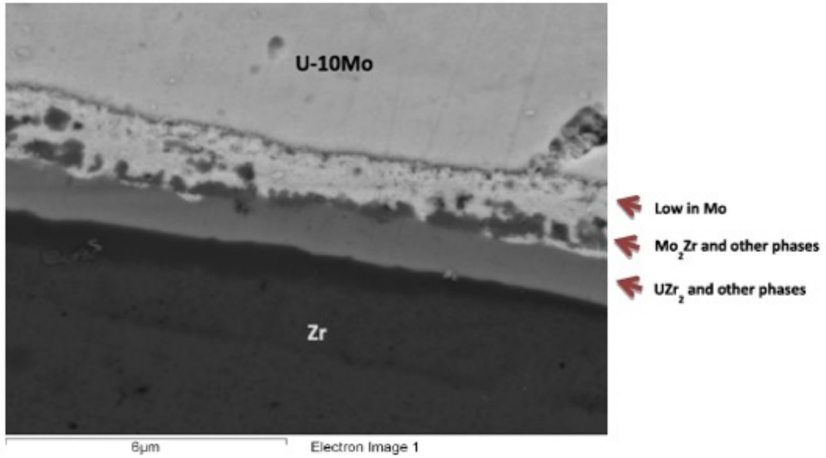
Uranium-Molybdenum (UMo)

- New fuel being qualified for research reactors
- Fuel foil, with Zr diffusion barrier, Al cladding
- Will be utilized in ATR, NBSR, MITR, MURR
- Phenomena: Decomposition; Fission Gas Superlattice; Recrystallization; Interdiffusion region; Carbides



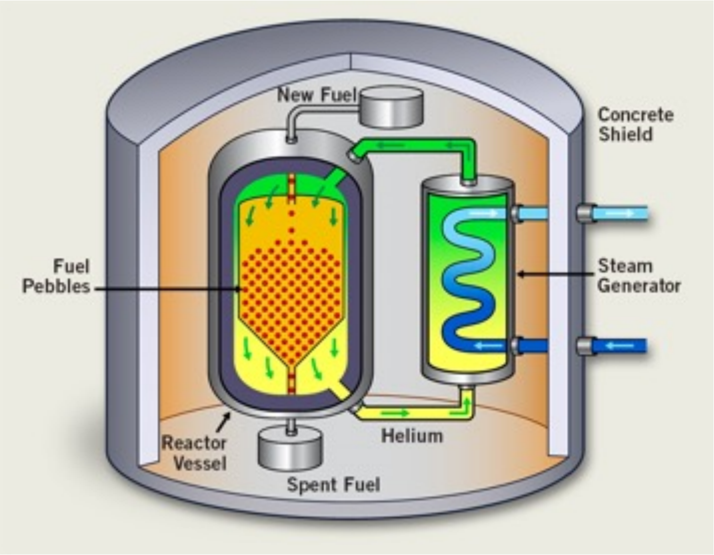


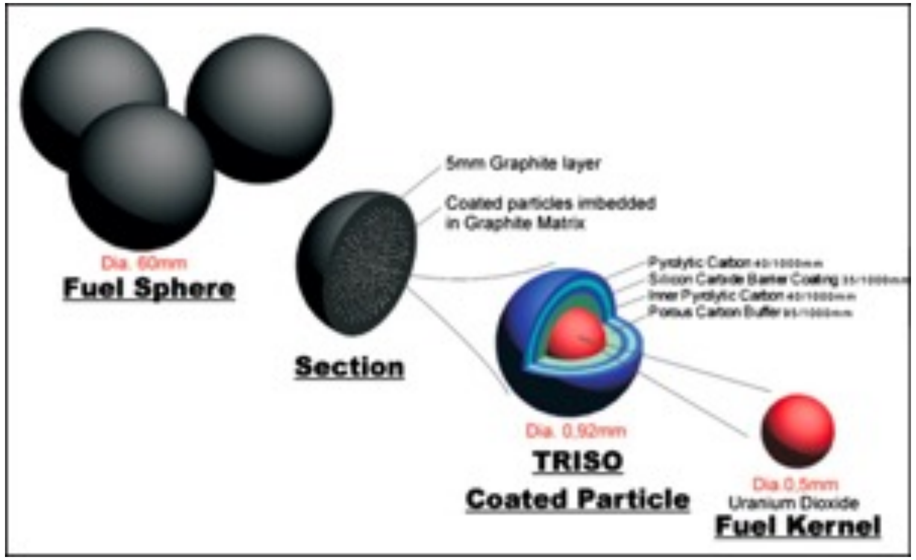




TRISO Particle Reactors

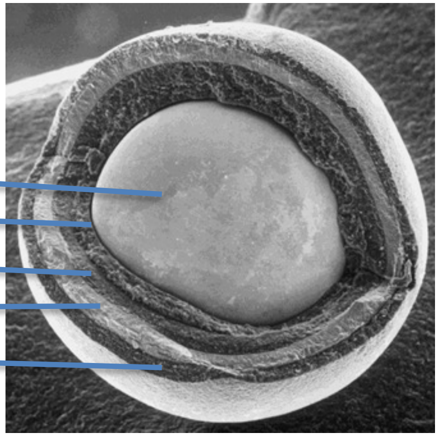
- High temperature gas reactors (HTGRs) or molten salt reactors (MSRs) can utilize TRISO particles as the fuel source
- Pebble bed and prismatic types of HTGRs
- Particles are agglomerated with graphite into a larger pebble, or into a cylindrical block
- Current designs utilize UCO (in the US), which is a heterogeneous mixture of UO2 and UC fuel
- Helium cooled gas or molten salt cooled

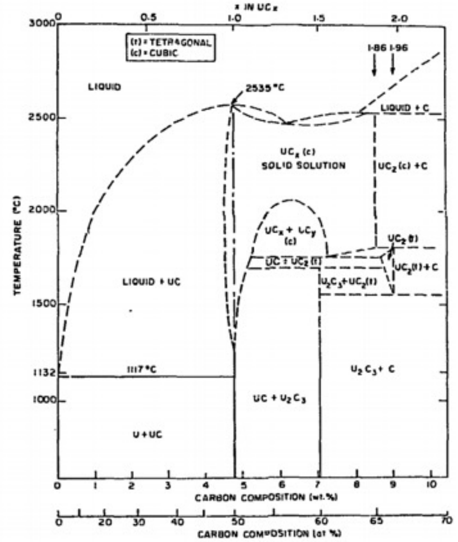


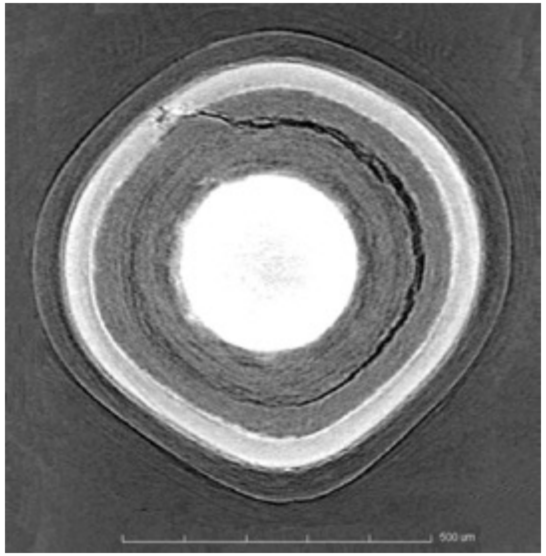


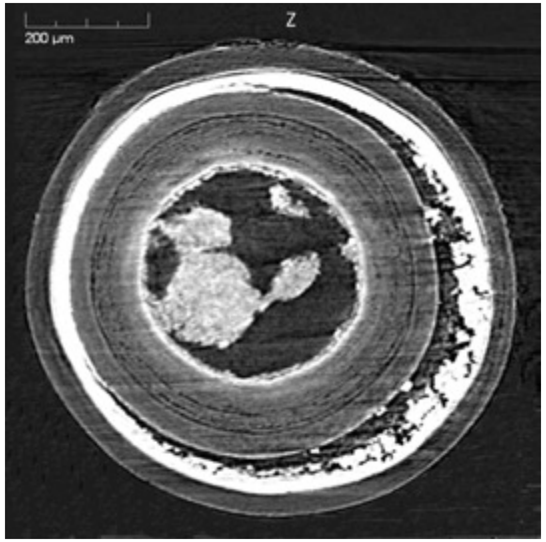
UC/UCO TRISO Fuels

- TRISO: TRistructural ISOtropic particle fuel
- Layered fuel in mm-sized particles
- Layers:
  - Fuel Kernel
  - Buffer
  - Inner Pyrolitic Carbon (IPyC)
  - SiC
  - Outer Pyrolitic Carbon (OPyC)
- Can appear as UC, U_2C_3, or UC_2
- Advantages
  - High thermal conductivity
  - High fuel density
  - Thermally stable
  - High melting temperature
- Disadvantages
  - Rapidly corrodes in water
  - Reacts with some cladding



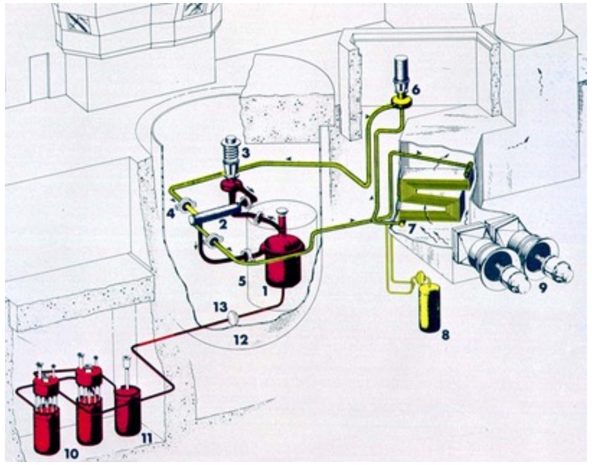


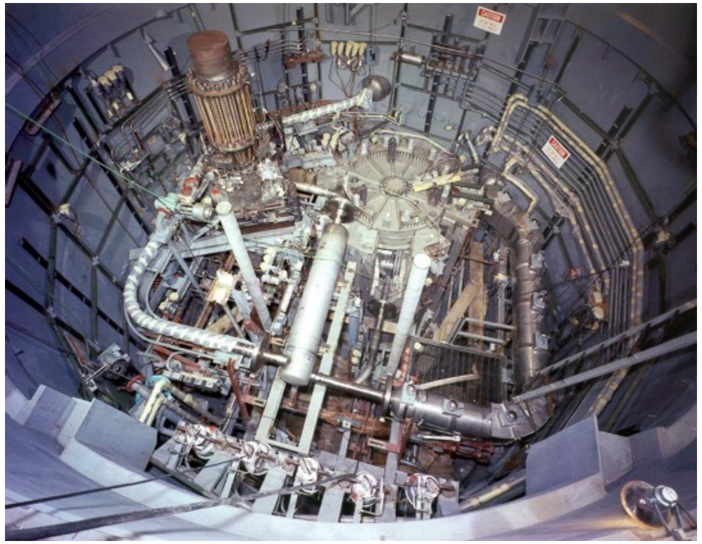




Molten Salt Reactors

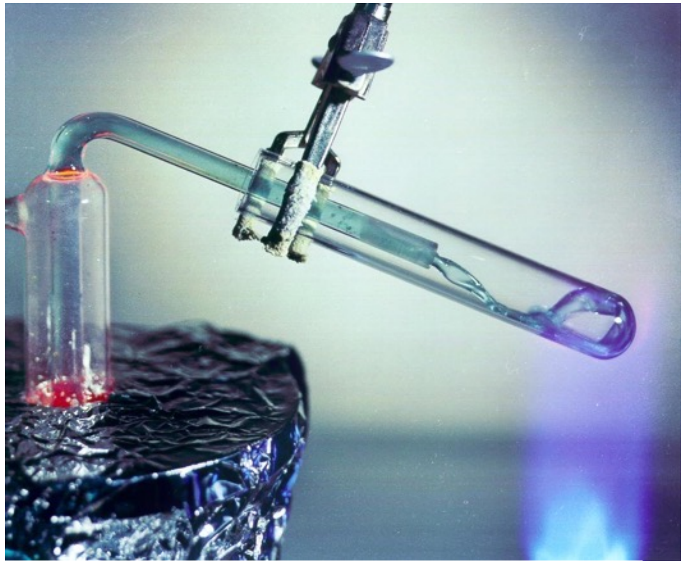
- Utilize molten salt (alkali/alkaline-halides), e.g., NaCl, as a heat transfer medium, functioning as coolant, fuel, or both
- The molten salt reactor experiment (MSRE) operated at Oak Ridge in the 1960s and utilized flowing fuel molten salt: LiF-BeF2-ZrF4-UF4
- MSRs can operate at ambient pressures and high temperatures
- MSRs have a very large margin to boiling, and combined with a negative reactivity feedback coefficient, have inherent safety

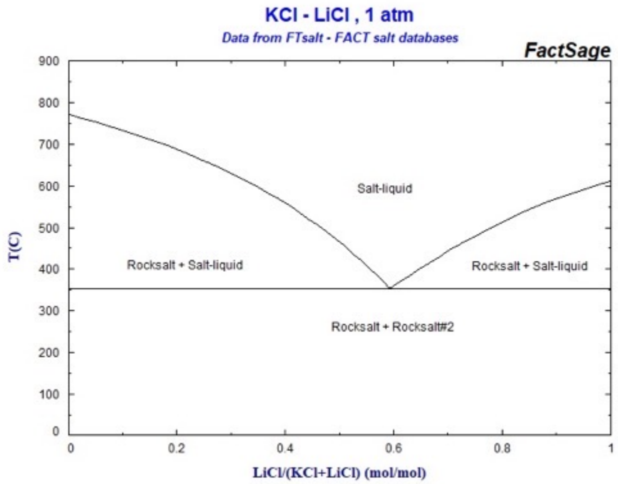




Molten Salt Reactors

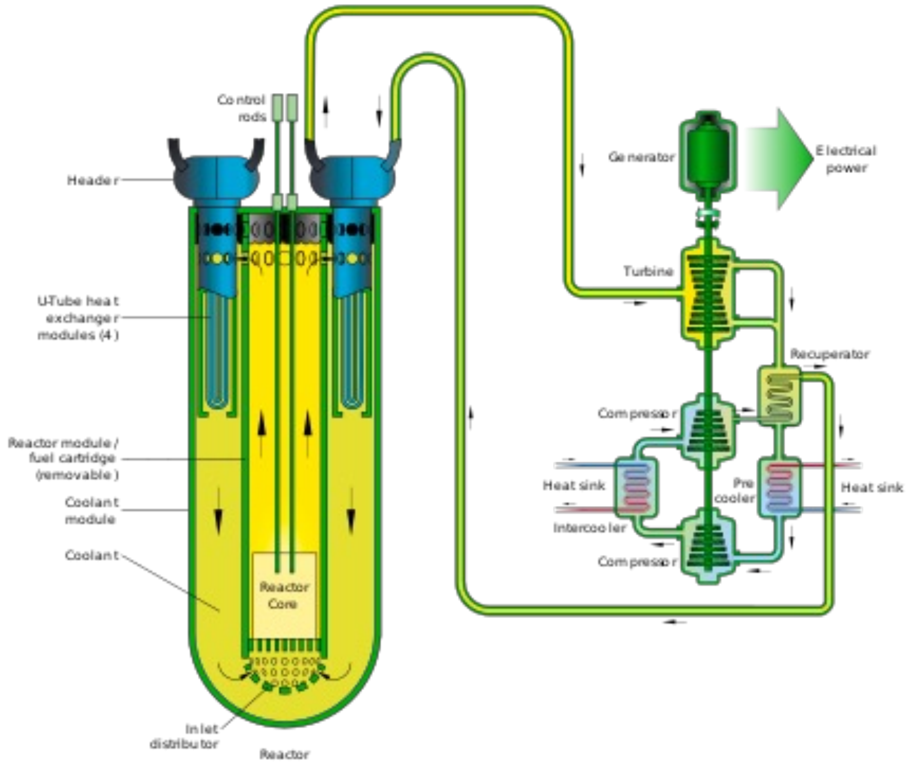
- Molten salts have high heat capacity, high boiling point, and high thermal conductivity, and can easily absorb fission products
- The changing chemical composition of the salt as it is transmuted by reactor radiation leads to changing properties
- The corrosivity of hot salts can break down cladding components and leech alloying elements
- LWR type reactor structural materials cannot be used, but some Ni alloys and graphite are sufficiently compatible with molten salts

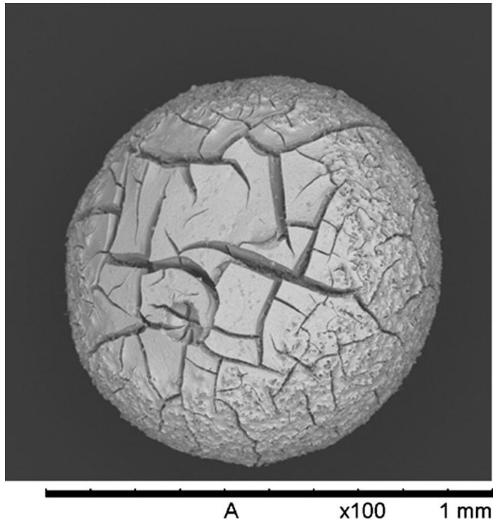


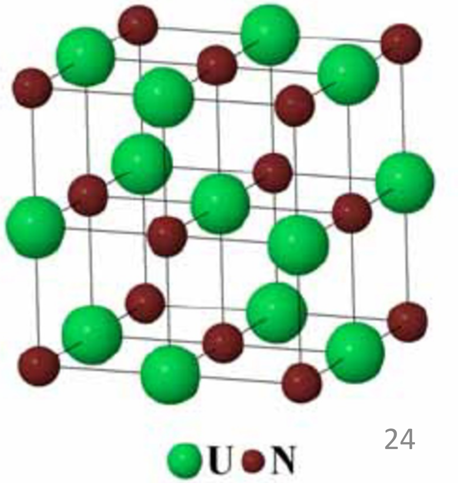


Uranium Nitride

- Potential fuel for gas cooled reactors as compacts or TRISO kernels, as well as lead cooled reactors Pb-cooled reactors are similar in concept to SFRs, pool-type
- High fissile density, high thermal conductivity, and chemical stability with cladding
- Very difficult to manufacture, poor oxidation resistance, requires N-enrichment
- Also being explored as a potential accident tolerant fuel for LWRs

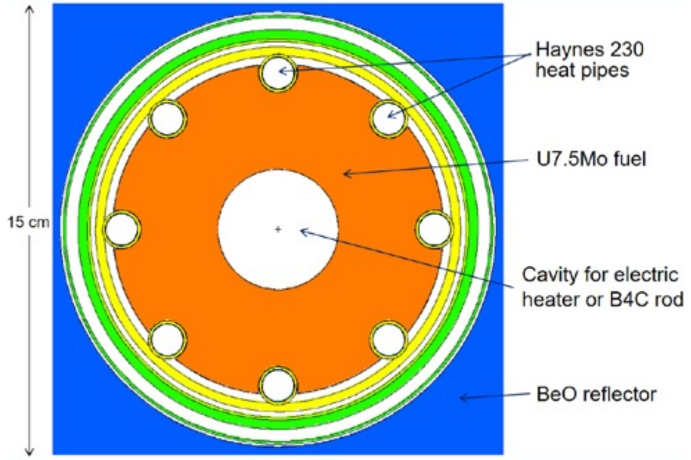




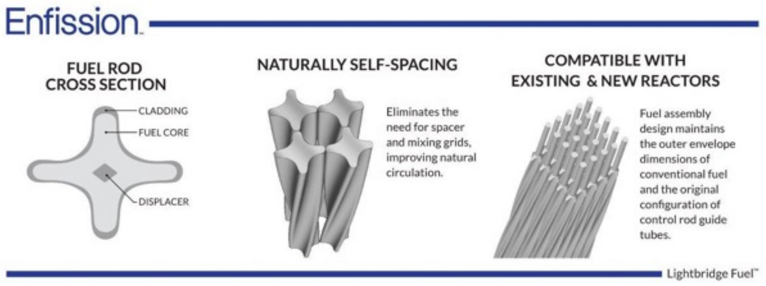


Unique Fuel Designs

KRUSTY



LightBridge



SSR

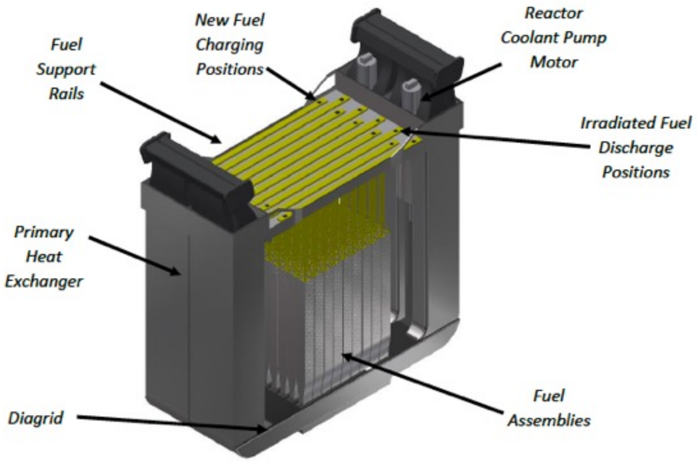
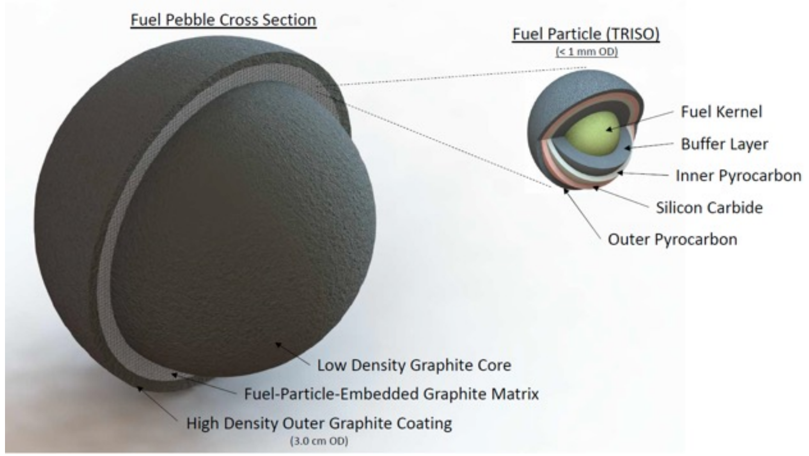


Figure 6: Overview of Stable Salt Reactor Core Module

Kairos



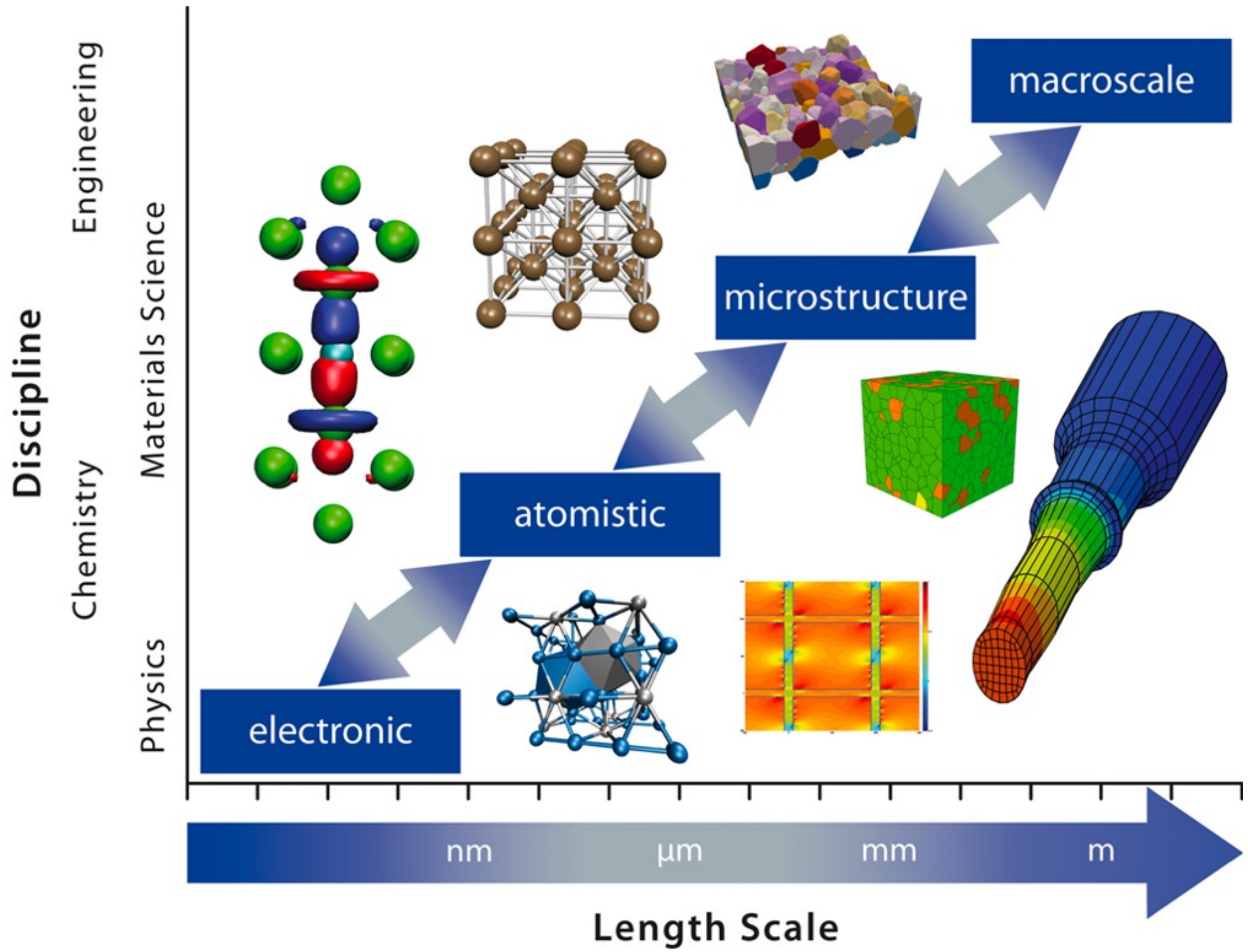
Summary

- There exist a number of nuclear fuels in different stages of utilization and development
- The more established ones are the less interesting ones… but still have a lot to understand
- Active research on existing, underutilized, and developmental fuel designs
- Each reactor design or application has individual needs, and no one fuel is one size fits all
- Need to balance safety, performance (normal, off-normal, extended), manufacture, processing, waste, etc.

How do we model advanced fuels?

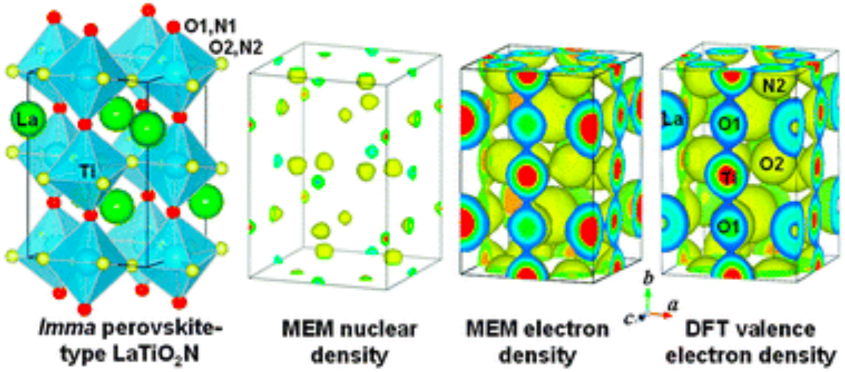
My viewpoint/approach

- Scientifically informed engineering is a multiscale problem
- Critical phenomena need to be identified from the macroscale, and informed from the lower length scales
- Perform good science, sometimes for science's sake, but often to address a key engineering problem or need

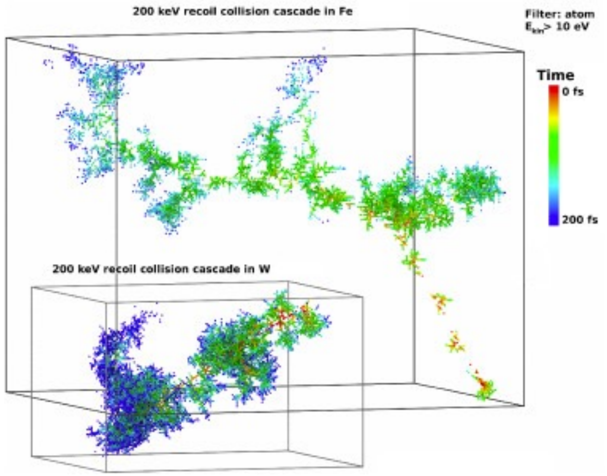


Atomistic Modeler's Toolkit

- 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
- 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



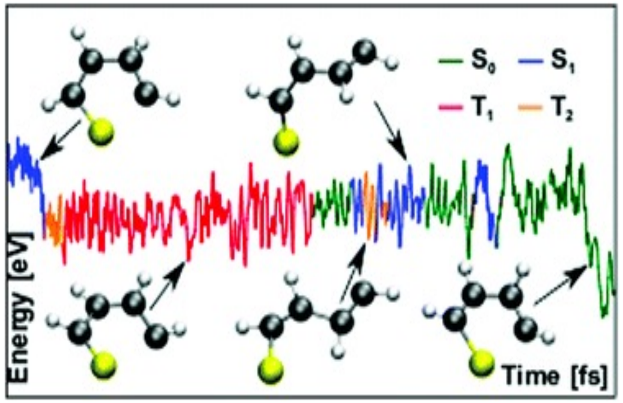
Yashima, https://doi.org/10.1039/C0CC00573H



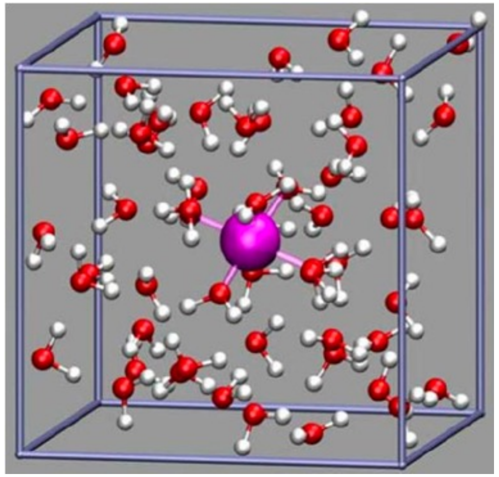
Sand, https://doi.org/10.1016/j.jnucmat.2018.08.049

Atomistic Modeler's Toolkit

- *ab initio* Molecular Dynamics (AIMD)
  - is a computer simulation method in which finite temperature molecular dynamics (MD) trajectories are generated with forces obtained from accurate 'on the fly' electronic structure calculations
  - hybrid mixing between the accuracy of electronic structure calculations, while allowing for real temperature effects to be considered



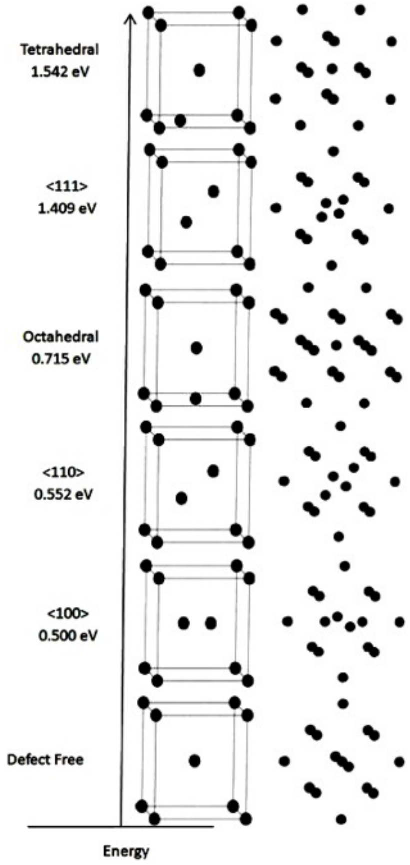
Schnappinger, https://doi.org/10.1039/C7CP05061E

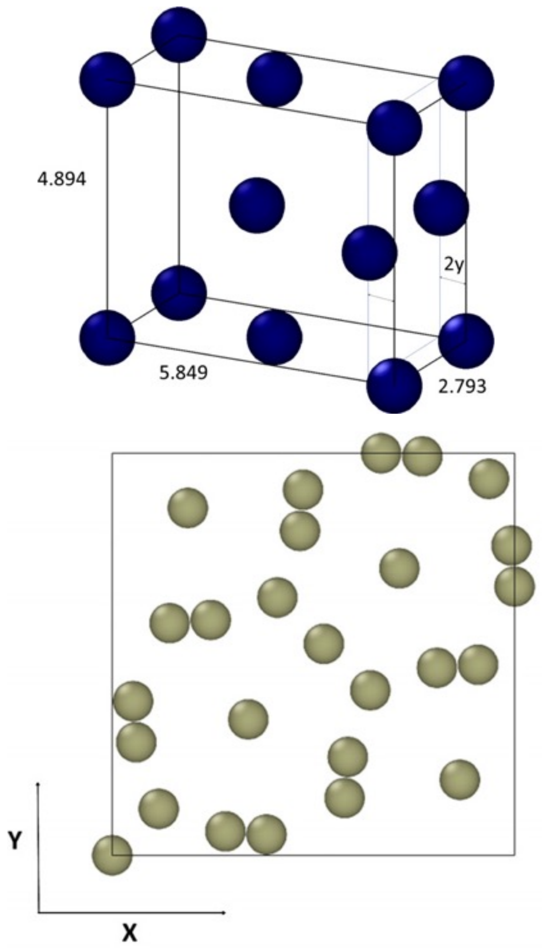


Yazyev, https://doi.org/10.1007/s00214-005-0052-6

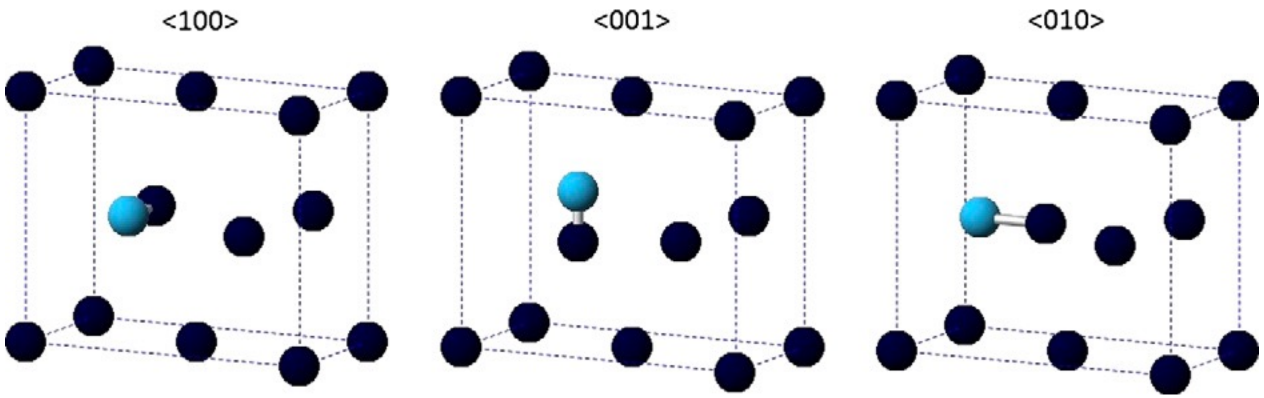
DFT applied to Nuclear Fuels

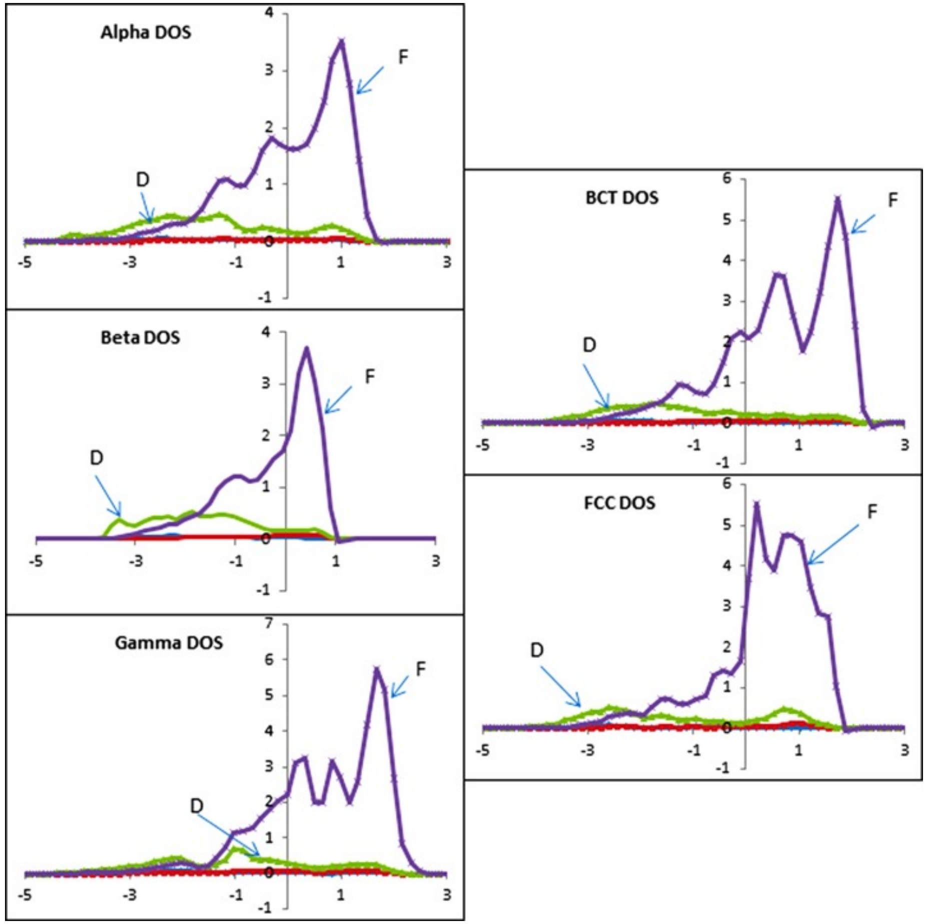
- Since we are dealing with the electronic structure, these calculations are very computationally expensive and limited to a few hundred atoms
- Typically employed to calculate lattice constants, elastic constants, point defects, electronic densities, phonon DOS, etc.
- DFT on advanced nuclear fuels has really only existed for about 15 years

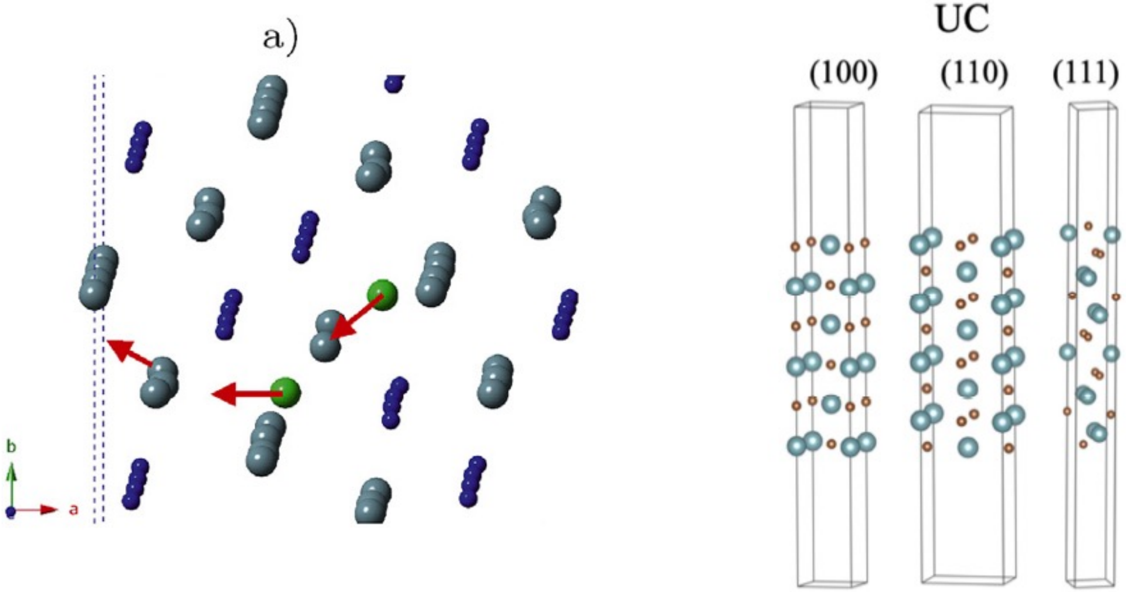




DFT applied to Nuclear Fuels

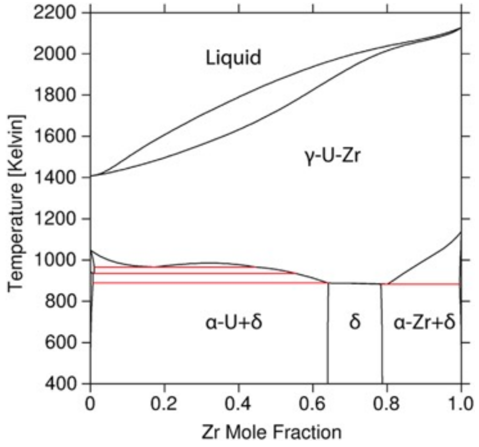


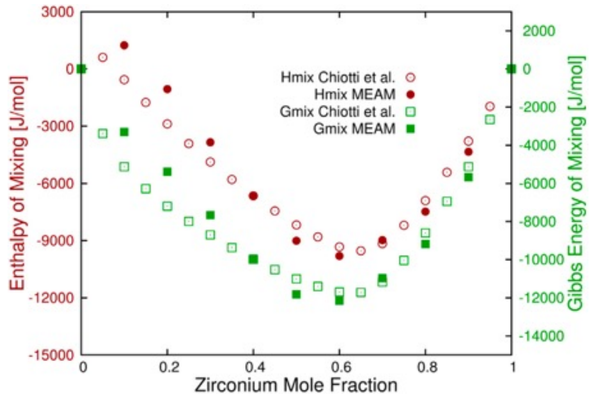


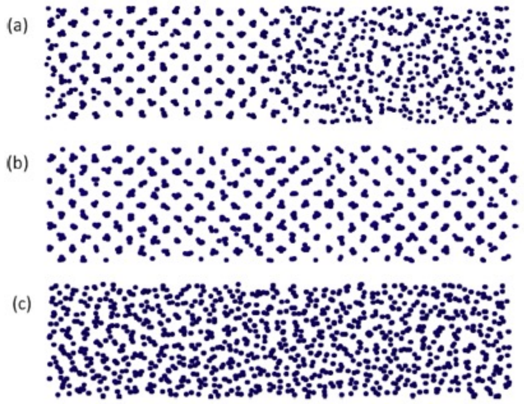


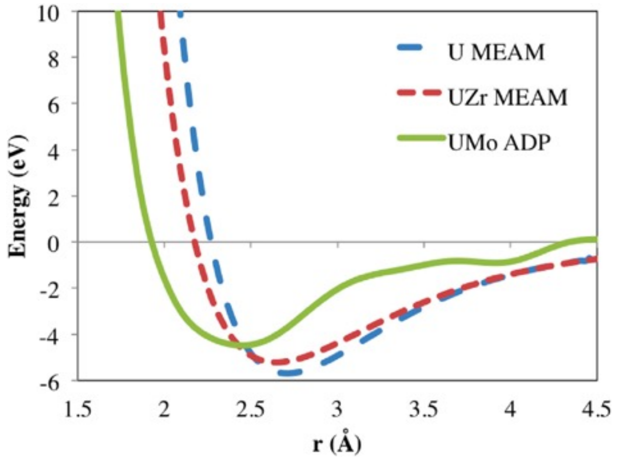
MD applied to Nuclear Fuels

- MD can span longer time scales and larger length scales, up to millions of atoms and nanoseconds
- These simulations require interatomic potentials capable of accurately predicting a number of properties
- No interatomic potentials for advanced nuclear fuels existed prior to about 15 years ago
- Potential development is still a major area for research

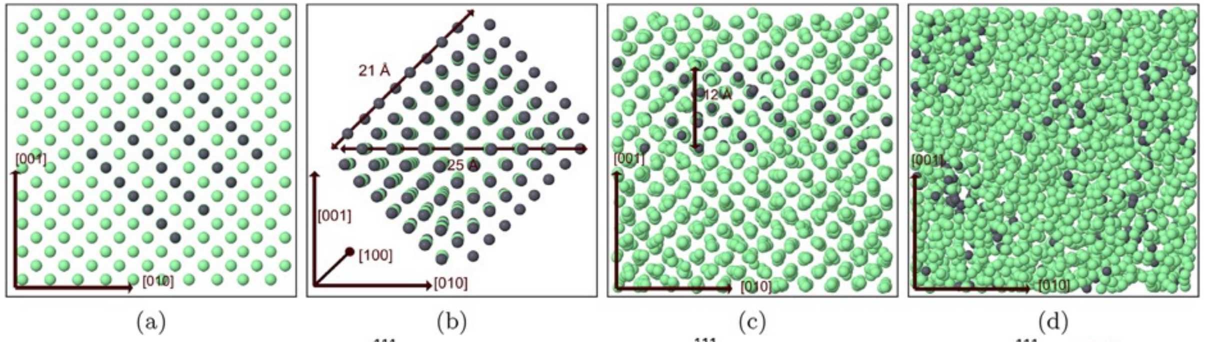


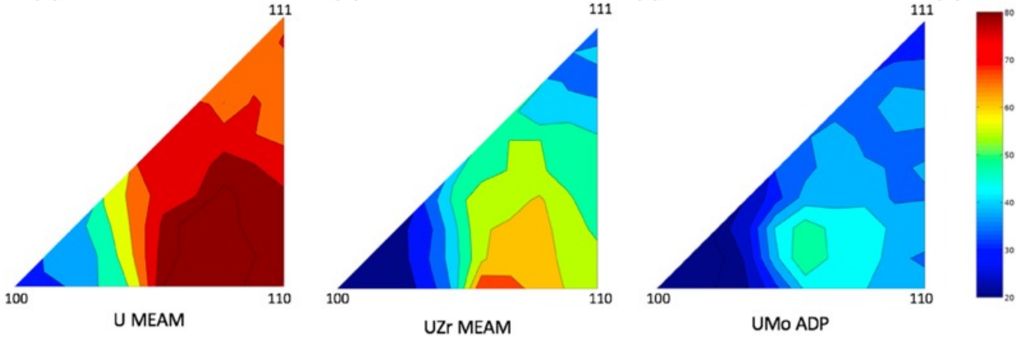


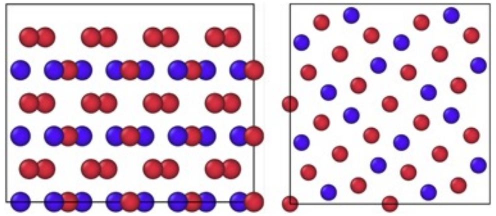


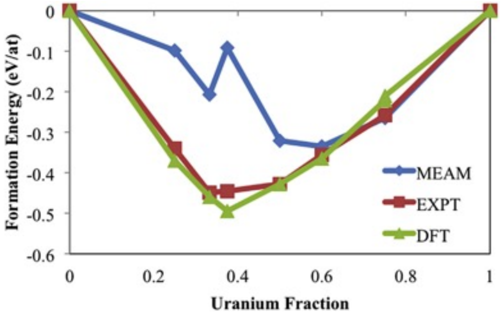


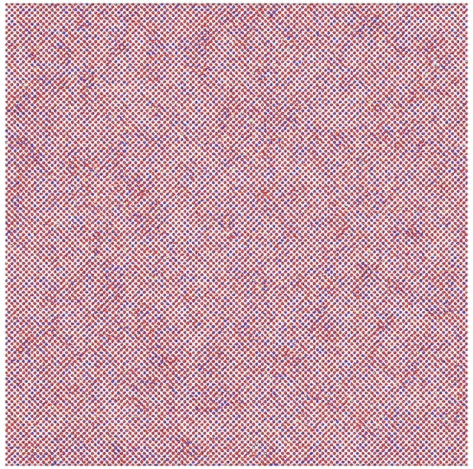
MD applied to Nuclear Fuels

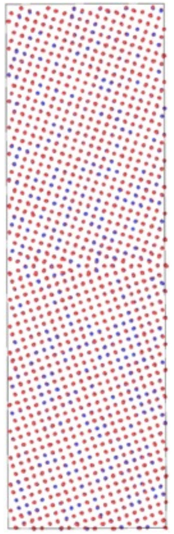


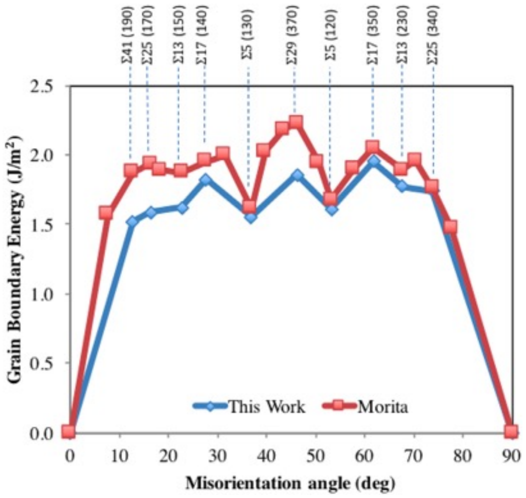


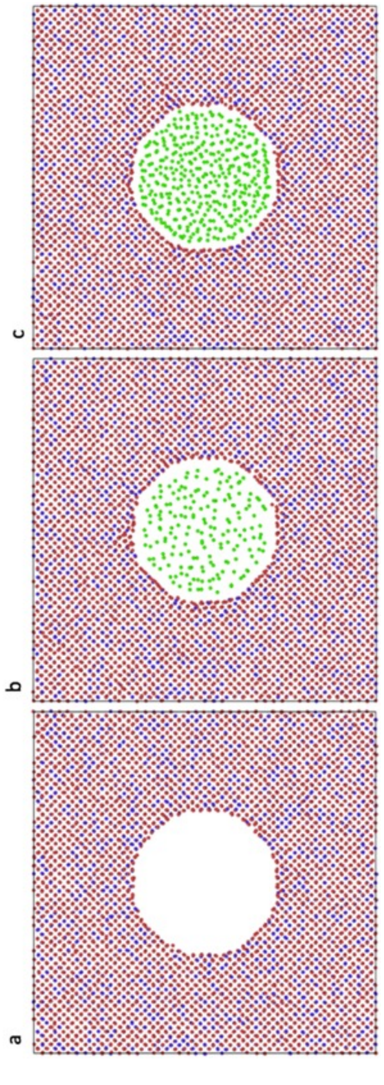






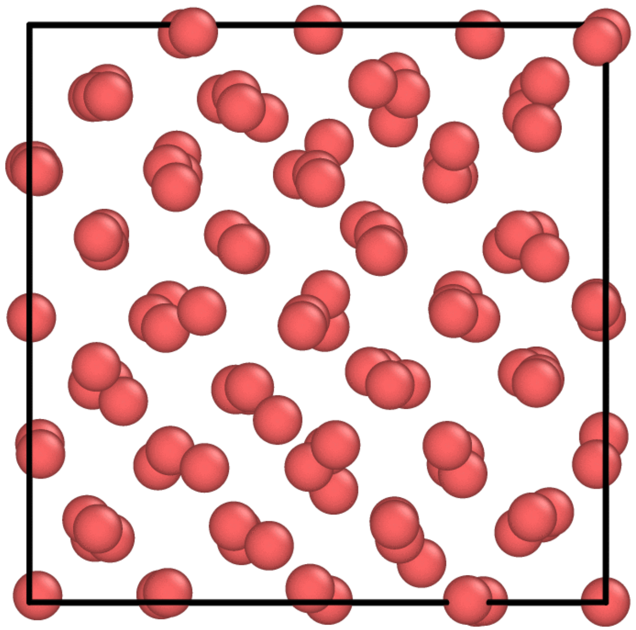




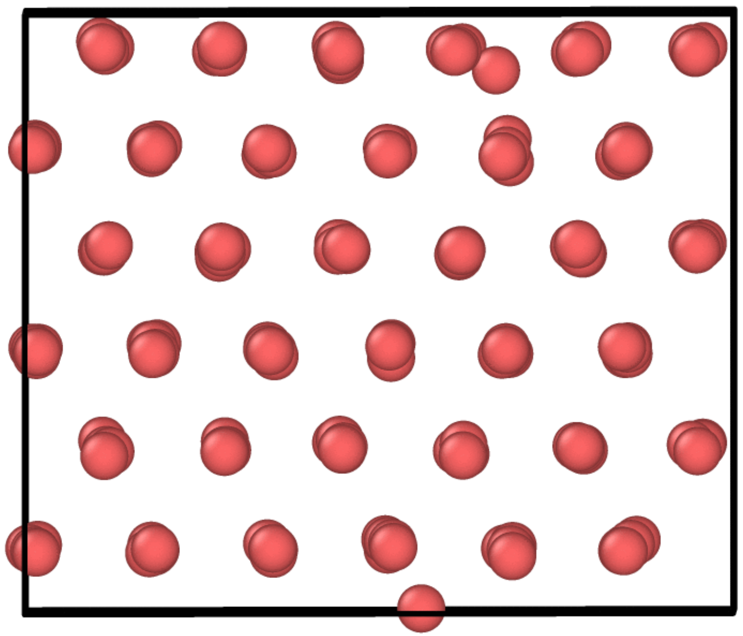


AIMD applied to Nuclear Fuels

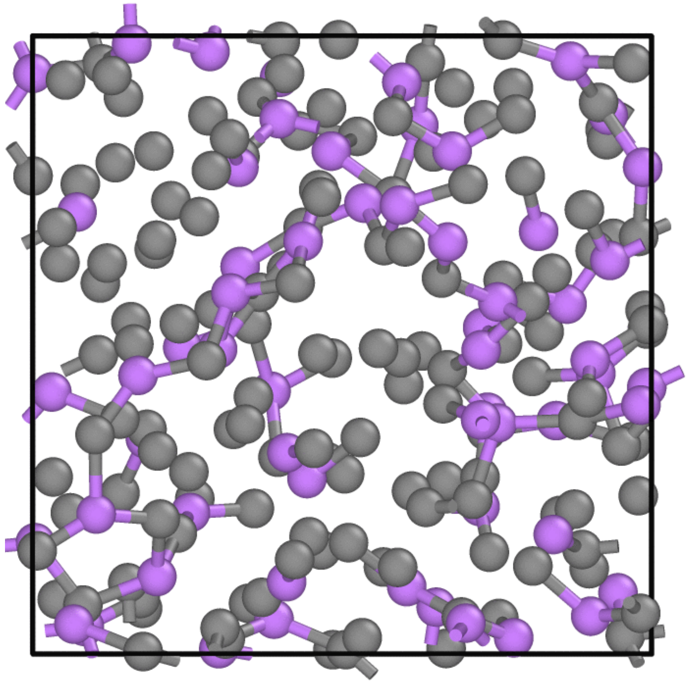
BCC U



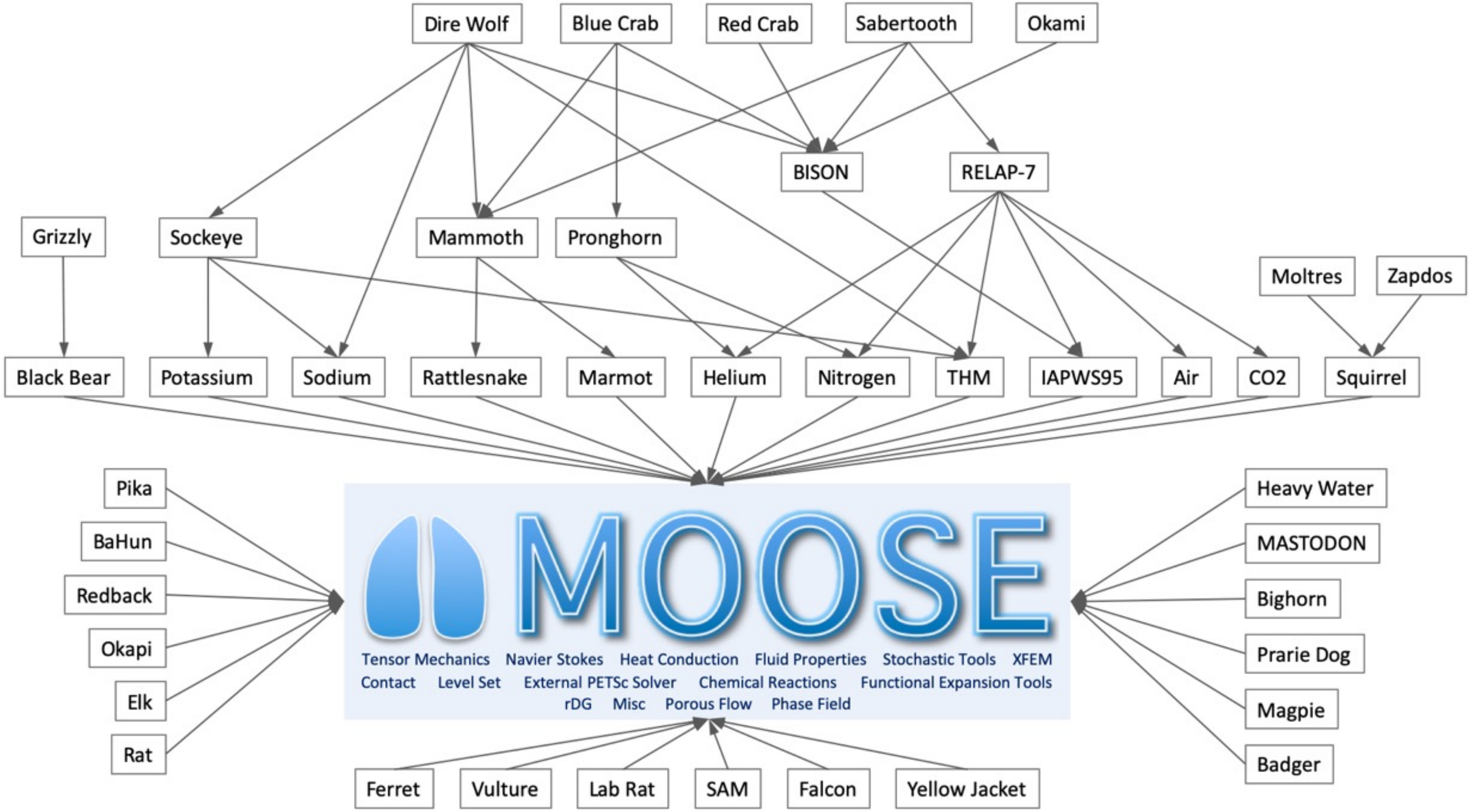
Alpha U with int



Eutectic LiCl-KCl

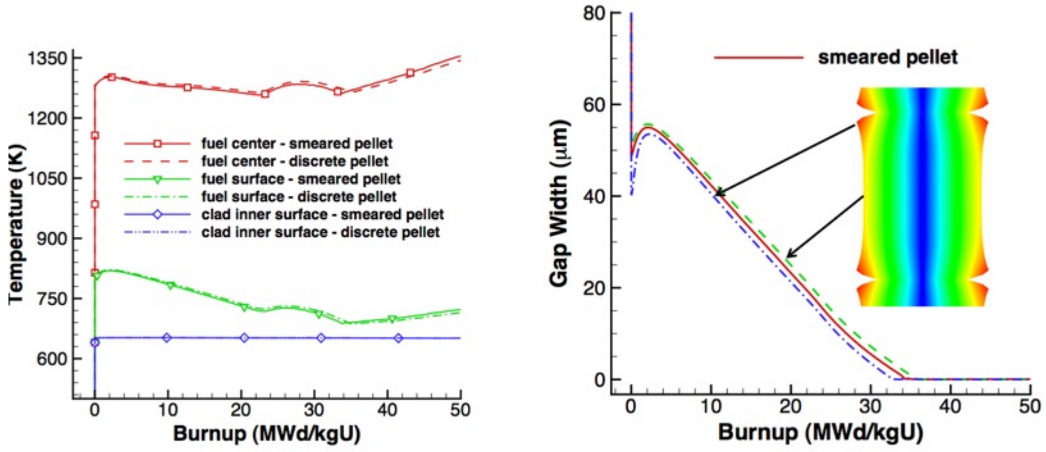


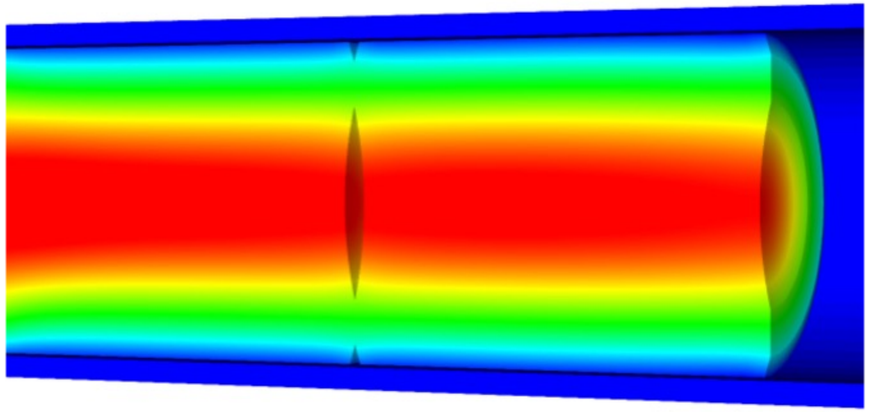
Integration into Fuel Performance Models



Integration into Fuel Performance Models

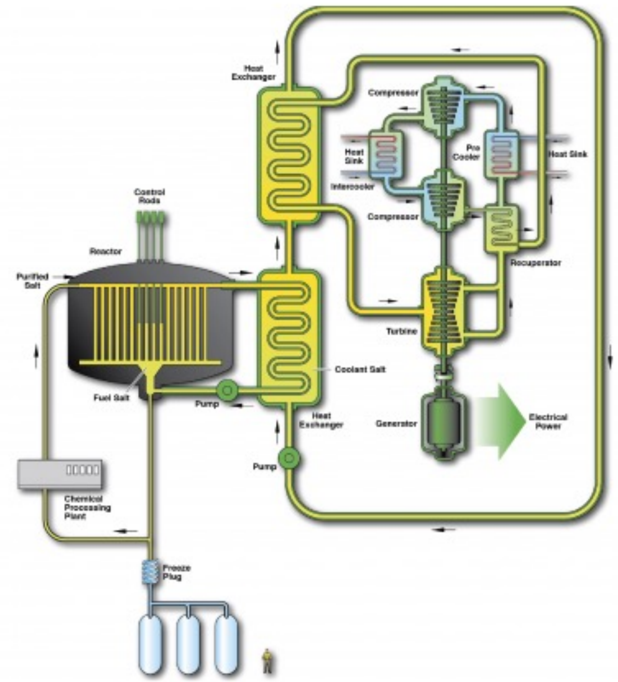
- BISON is the multiphysics MOOSE-based fuel performance code developed under NEAMS and includes thermal transport, stress, and underlying material evolution, etc.
- BISON requires models for material properties and how they evolve during operation
- A lot of these fundamental properties are unknown, and too difficult to obtain experimentally
- Lower Length Scale Modeling!





Molten Salts

- Applications of molten salts: Catalysis; Metal production; Solar power; Thermal energy storage; Pyroprocessing; Molten salt reactors
- There is a gap in the fundamental properties of molten salts
- These properties are difficult to collect experimentally due to high temperatures, corrosivity, toxicity, etc.
- Computational methods are used to supplement the experimental data



✓ M	In Model
✓ L	In Literature

NaCl	✓ L														
$MgCl_2$	✓ L	✓ M													
KCl	✓ M	✓ L	✓ M												
RbCl	✓ L														
$CaCl_2$	✓ L	✓ L	✓ L	✓ L											
CsCl	✓ L	✓ M	✓ M												
$PuCl_3$		✓ M	✓ M												
UCl_3	✓ M	✓ M	✓ M	✓ M				✓ M							
$ZnCl_2$		✓ L	✓ L	✓ L		✓ L									
$MnCl_2$		✓ L	✓ L	✓ L		✓ L				✓ L					
$FeCl_2$		✓ L	✓ L	✓ L		✓ L				✓ L	✓ L				
$CoCl_2$		✓ L	✓ L	✓ L		✓ L				✓ L	✓ L	✓ L			
$NiCl_2$		✓ L	✓ L	✓ L		✓ L				✓ L	✓ L	✓ L	✓ L		
$SrCl_2$	✓ L	✓ L	✓ L	✓ L		✓ L									
$CeCl_3$	✓ L			✓ L											
	LiCl	NaCl	$MgCl_2$	KCl	RbCl	$CaCl_2$	CsCl	$PuCl_3$	UCl_3	$ZnCl_2$	$MnCl_2$	$FeCl_2$	$CoCl_2$	$NiCl_2$	$SrCl_2$

✓ M	In Model
✓ L	In Literature

NaF	✓ M															
BeF_2	✓ M	✓ M														
KF	✓ M	✓ M														
RbF	✓ M	✓ M		✓ M												
CaF_2	✓ M	✓ M		✓ M												
ZrF_4	✓ L		✓ L													
ThF_4	✓ M	✓ M	✓ M			✓ M										
PuF_3	✓ M	✓ M	✓ M	✓ M	✓ M			✓ M								
UF_4	✓ M	✓ M	✓ M				✓ L	✓ M	✓ M							
UF_3	✓ M	✓ M								✓ M						
CsF	✓ M	✓ M		✓ M	✓ M			✓ M	✓ M	✓ L						
CeF_3	✓ M	✓ M						✓ M		✓ M						
LaF_3	✓ M	✓ M		✓ M	✓ M	✓ M			✓ M			✓ M				
SrF_2	✓ L	✓ L		✓ L		✓ L										
MgF_2	✓ L	✓ L		✓ L		✓ L									✓ L	
NdF_3	✓ M									✓ M						
	LiF	NaF	BeF_2	KF	RbF	CaF_2	ZrF_4	ThF_4	PuF_3	UF_4	UF_3	CsF	CeF_3	LaF_3	SrF_2	MgF_2

What do we do?

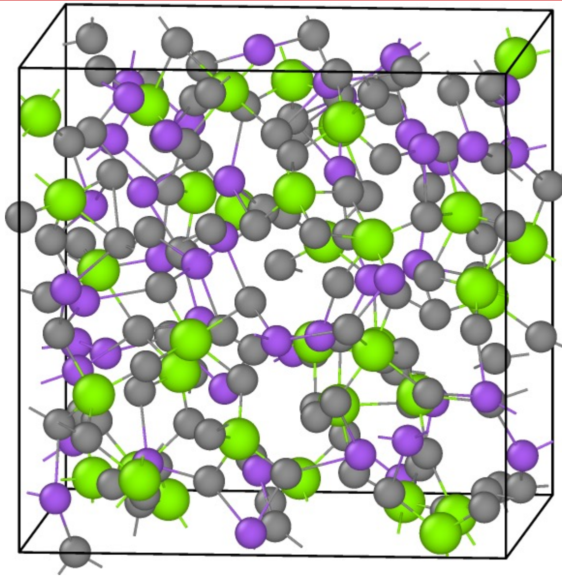
- Work with experimentalists to target specific salt pseudo-binaries and ternaries and validate computationally determined properties, then expand beyond the experimental data
- Apply AIMD to determine density, compressibility, thermal expansion, heat capacity, enthalpy of mixing, etc.
- Utilize these calculations to determine transport properties: diffusivity, viscosity, thermal conductivity, etc.

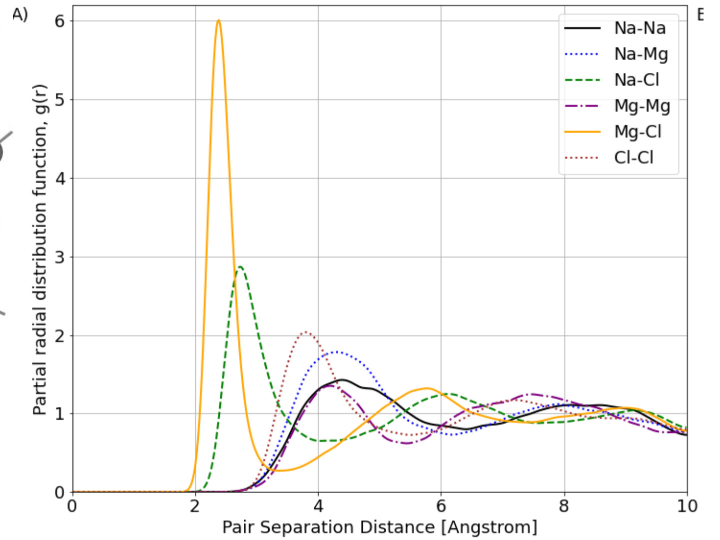
NaCl-MgCl2 Density

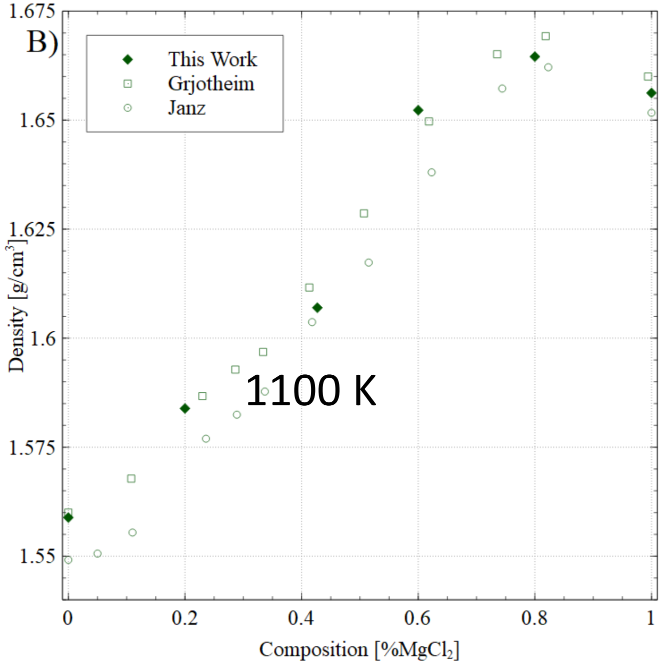
- Great agreement with the literature
- Identified a temperature transition
- Fit with first order Redlich-Kister expansion (n=2)
- $\rho_{RK} = \rho_{id} + \rho_{ex}$

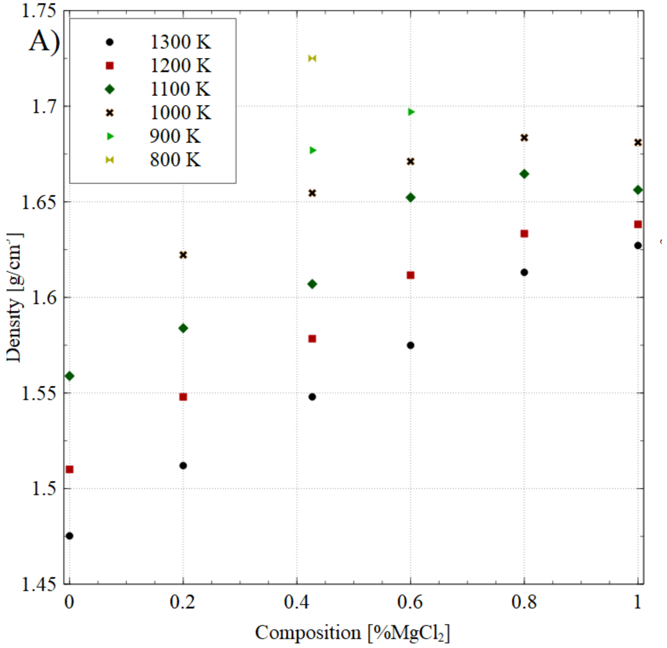
$$\rho_{ex} = x(1-x)\sum_{n-1}^{N}(A_n + B_n T)(2x-1)^{n-1}$$

$$\rho_{id} = \frac{\sum_i x_i M_i}{\sum_i x_i V_i}$$

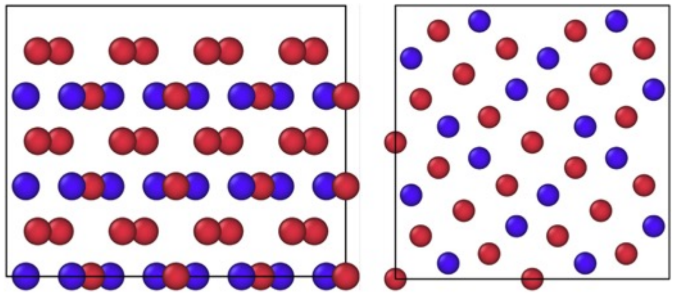


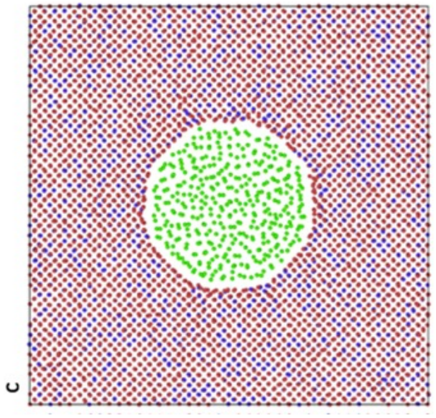


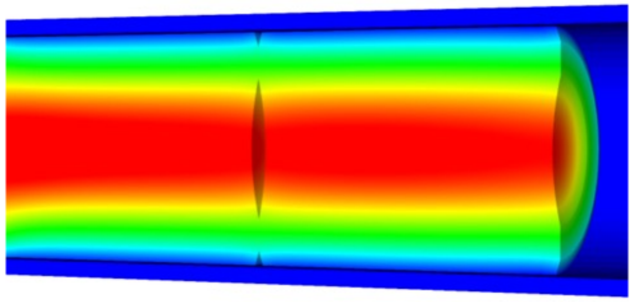




Summary







- Scientifically informed engineering is a multiscale problem
- There exist a number of scientific knowledge gaps for advanced nuclear fuels
- Atomistic methods (DFT, MD, AIMD) are a rapidly growing area that can fill in a number of these gaps
- Atomistic data can be implemented into higher length scale methodologies to increase fidelity of fuel performance simulations