Nuclear Fuel Concepts

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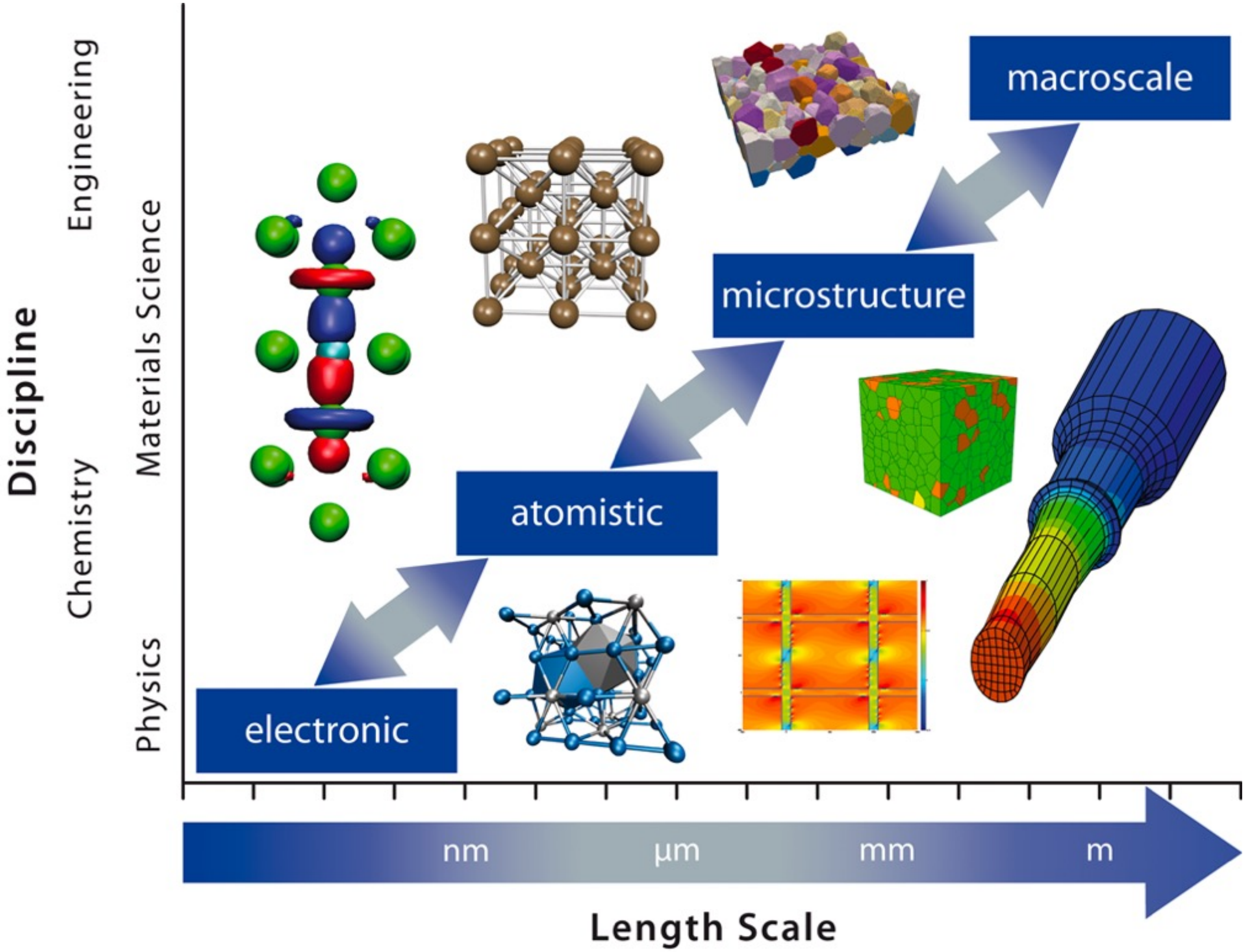
My viewpoint/approach

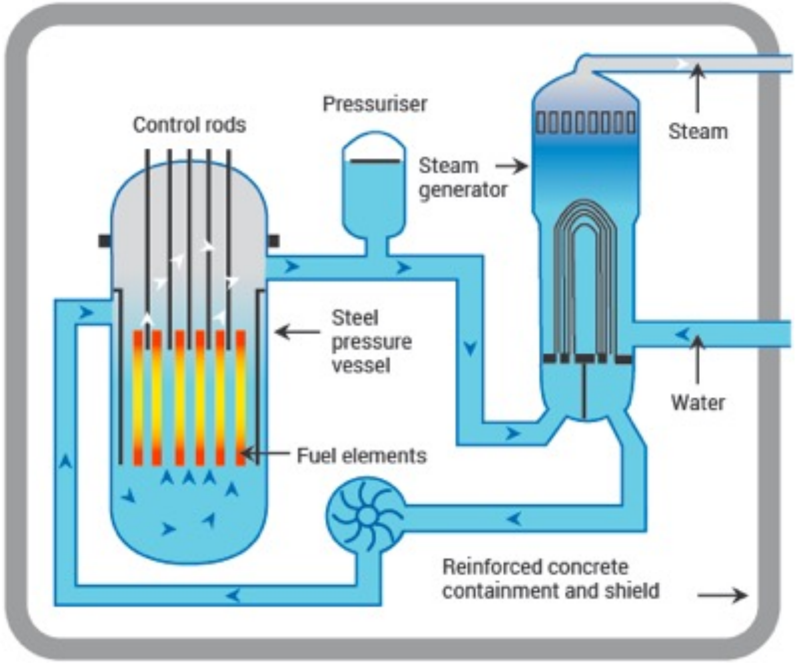
- Scientifically informed engineering is a multiscale problem
- Critical phenomena need to be identified from the macroscale, and informed from the lower length scales
- Perform good science, sometimes for science's sake, but often to address a key engineering problem or need



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

A Pressurized Water Reactor (PWR)



Fuel Considerations

- Safety:
  - Stable and predictable behavior
  - No melting, no phase changes under transients that lead to deleterious behavior
- Uranium density
- Mechanical integrity
- Cladding interactions
- Swelling and fission gas release
- Operating temperatures

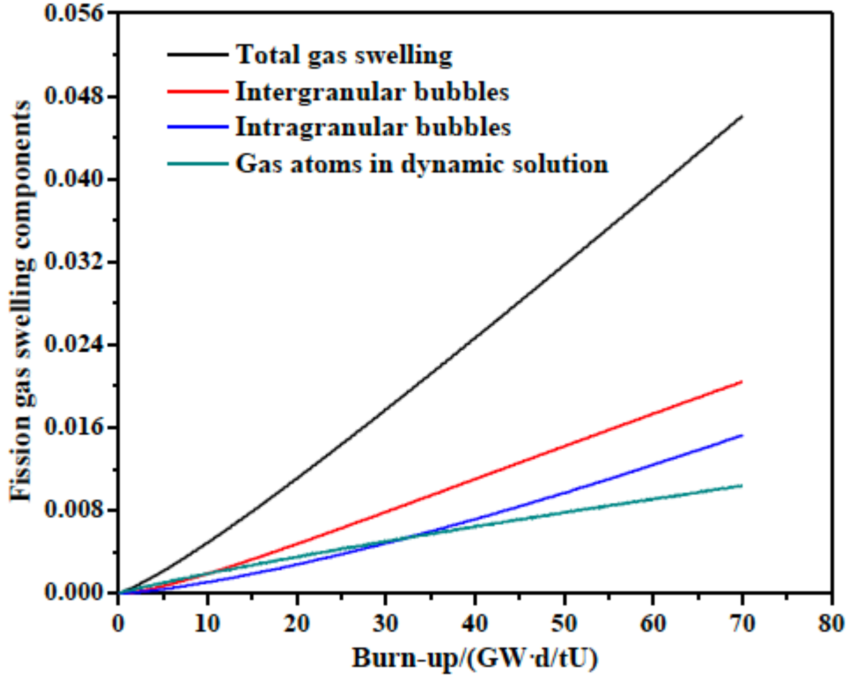


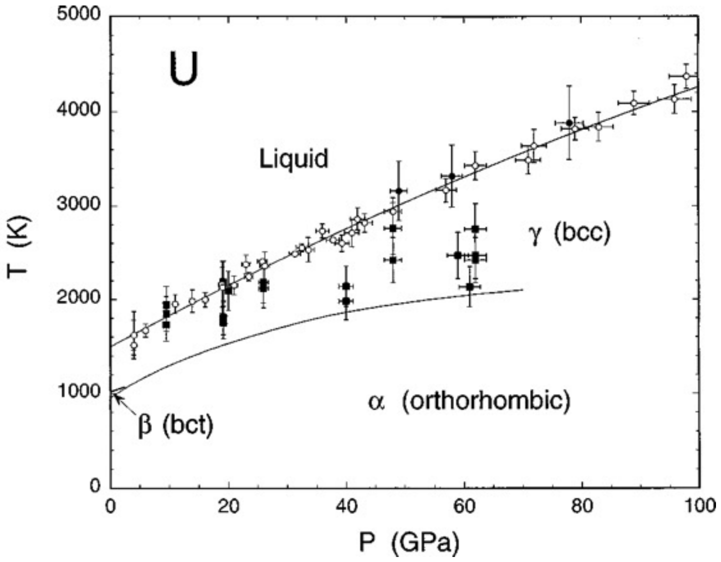
Fig. 4. Variation of swelling rate components of fission gas in UO_2 fuel with the burn-up.

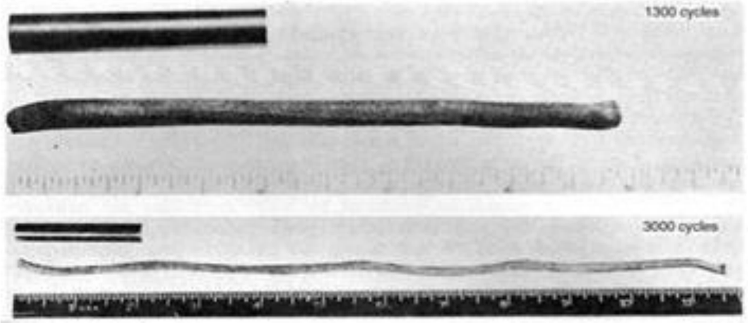
Fuel Types and Associated Reactor Types

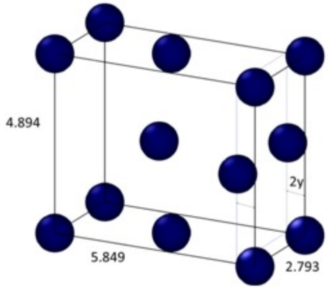
- UO_2 – Light Water Reactors
  - Mixed oxide (MOX)
  - Accident tolerant fuel (ATF)
- UZr – Sodium Cooled Fast Reactors
- UMo – Research Reactors
- UC/UCO – High Temperature Gas Reactors
- UN – Lead Cooled Fast Reactors
- NaCl-UCl_3 – Molten Salt Reactors

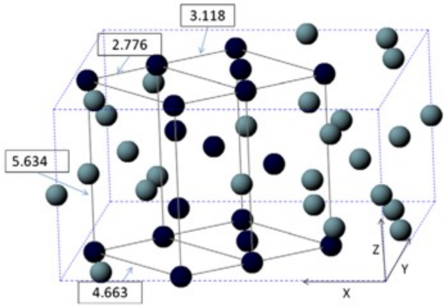
Why not use pure uranium metal?

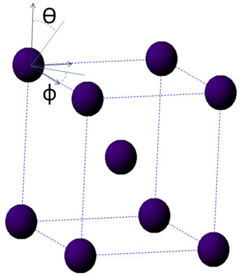
- Pure uranium has three phases
  - α-phase is orthorhombic
  - β-phase is tetragonal
  - γ-phase is body-centered cubic
  - During thermal cycling, pure uranium dramatically swells
  - Alpha U has both anisotropic thermal expansion and anisotropic irradiation growth





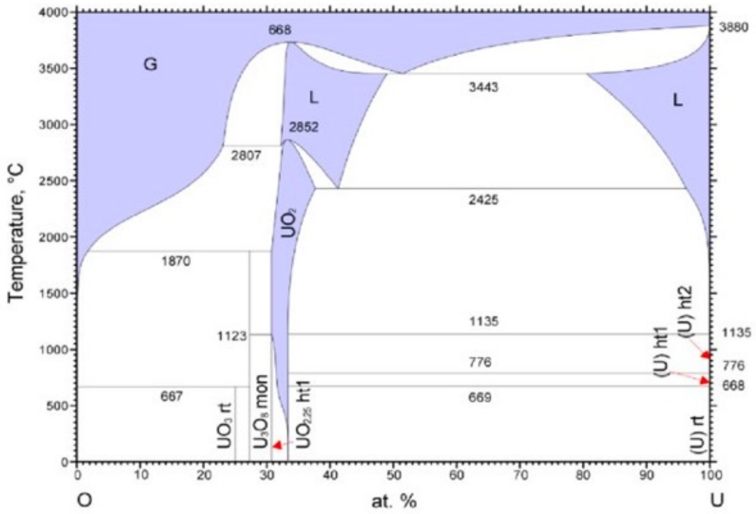


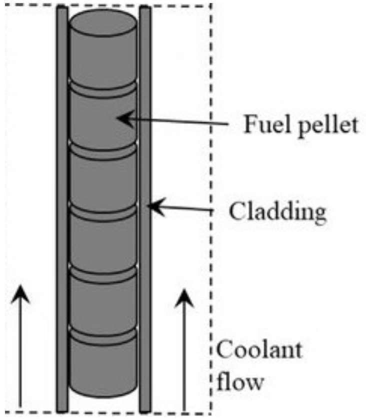


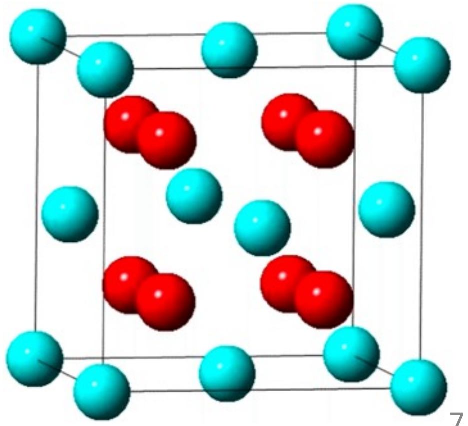


Uranium Dioxide (UO2)

- 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
- Good behaviors
  - Very high melting point, about 2800 C
  - Maintains a stable fluorite phase up to melting
  - Very compatible with Zircaloy clad (no interaction zones forming/no FCCI)
  - Relative stability in water
  - Reasonably radiation resistant (no amorphization can incorporate a large number of fission products as substitutional defects)







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

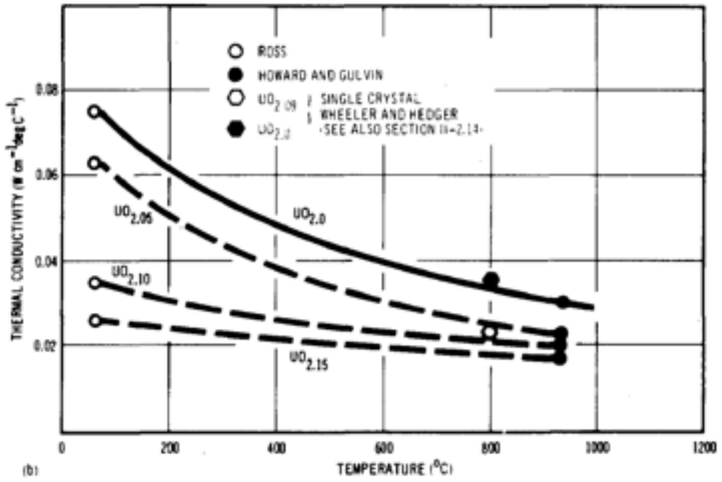
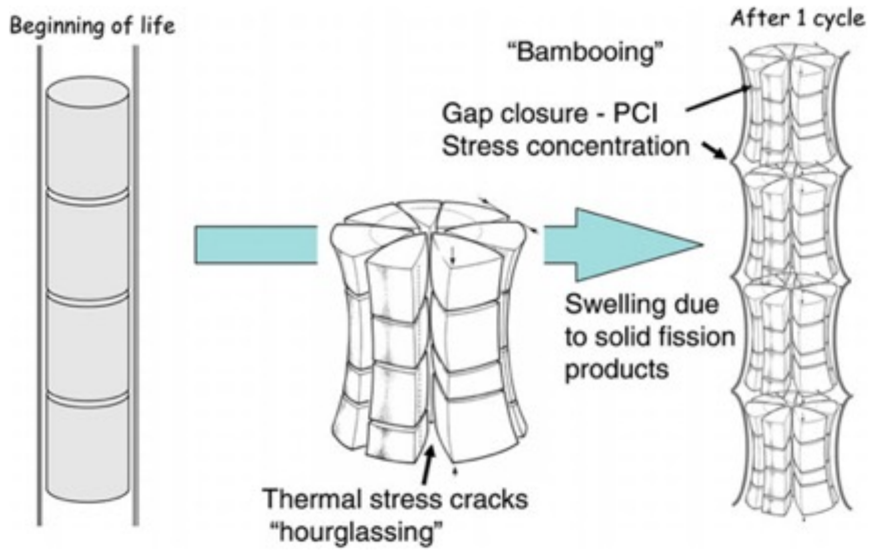
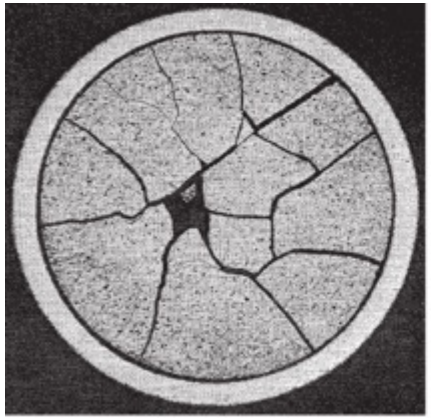


Fig. 9. Effect of hyperstoichiometry on conductivity.

Uranium Dioxide (UO2)

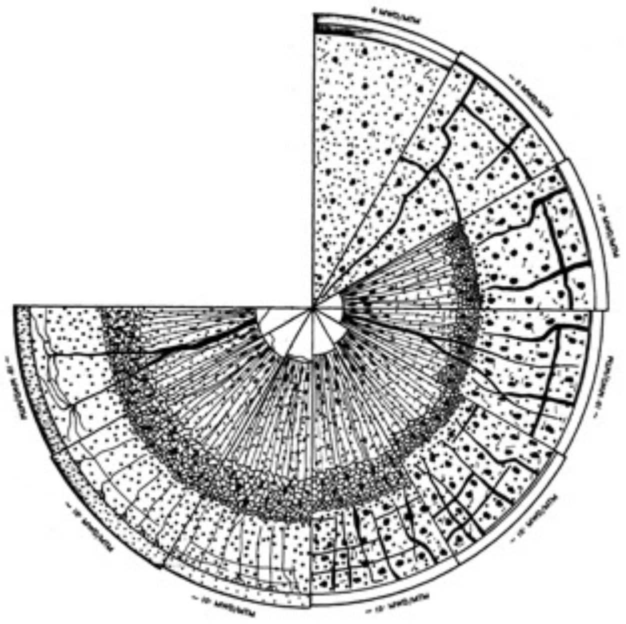
- Critical Phenomena
  - Thermal conductivity (and degradation)
  - Fission gas release, leading to pressure increase inside the cladding
  - Fuel fragmentation and relocation under transients
  - Bambooing creating stress concentrations
- All 98 operational reactors in US utilize UO2





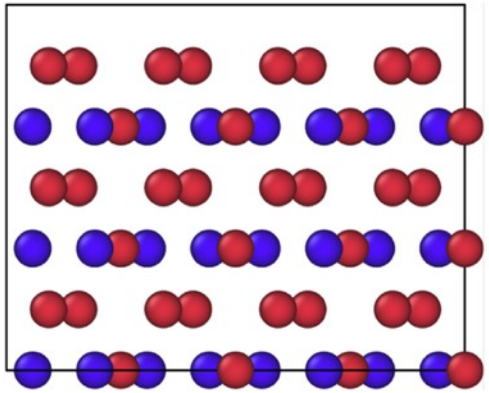
Mixed Oxides

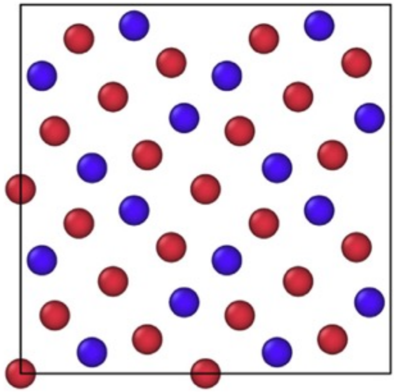
- UO2 can be combined with PuO2 for a mixed oxide (MOX) fuel for use in fast reactors
- Allows to burn excess weapons grade plutonium
- About 30 reactors in Europe currently utilize a partial MOX core
- Similar behavior to UO2, but different neutronics, fission gas release, thermal conductivity, etc.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

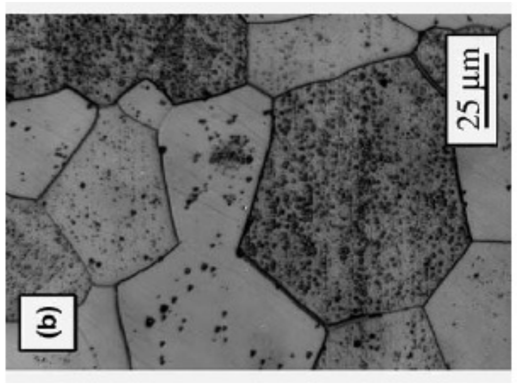


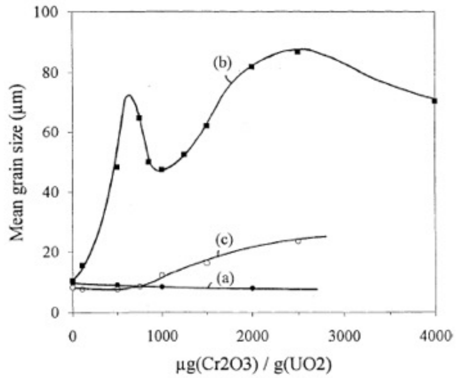
Accident Tolerant Fuel/Advanced Technology Fuel

- USi fuels (U_3Si_2)
  - A metal-ish compound, with higher thermal conductivity, higher uranium density
- Cr-doped UO_2
  - 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.

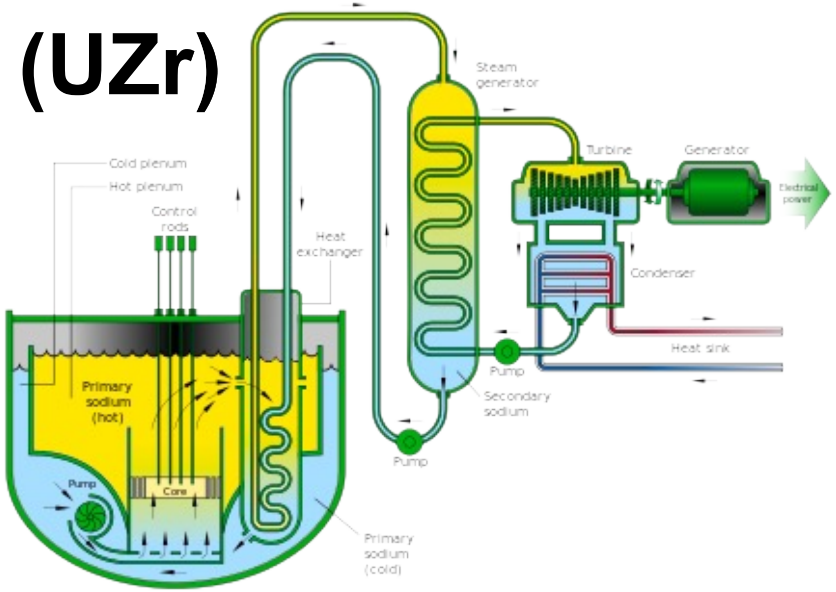




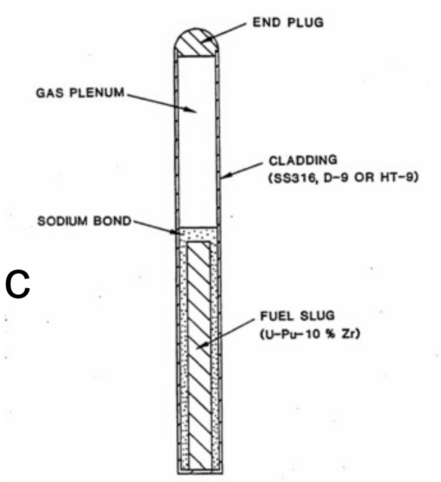


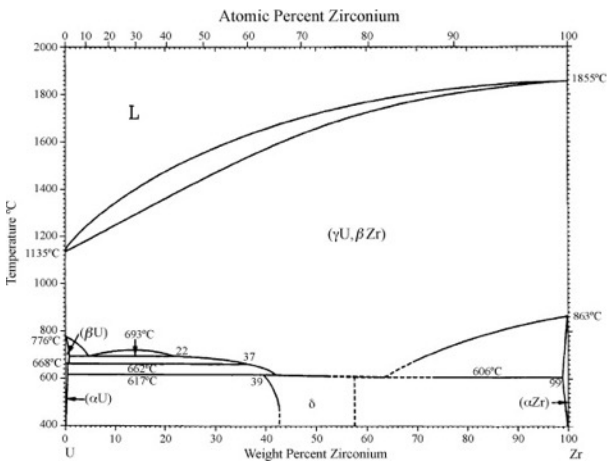


Uranium-Zirconium (UZr)



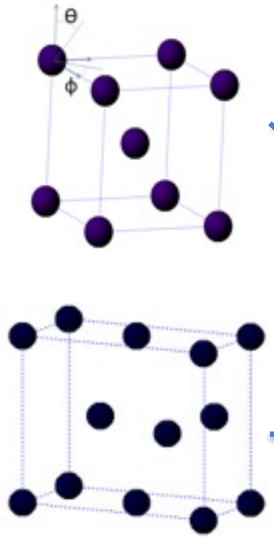
- Utilized in sodium cooled fast reactors (SFRs) - EBR I, EBR II
- Varied crystal structure and compositional environment
- Easily alloyed with Pu, minor actinides (MA)
- Can function as a breeder/burner fuel
- Sodium coolant
- Fe-based cladding
- Alloy with Zr to increase the melting point and to stabilize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

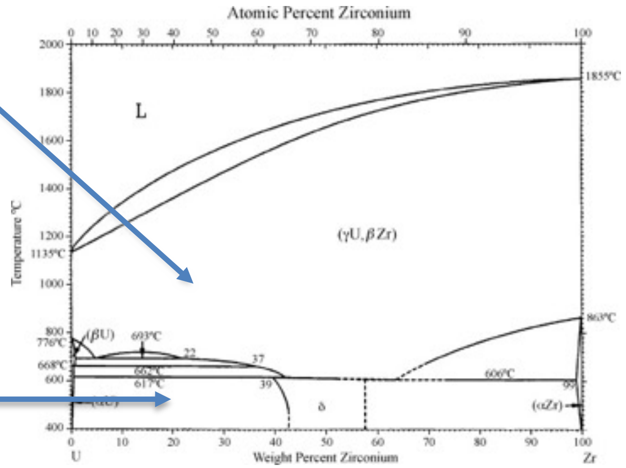


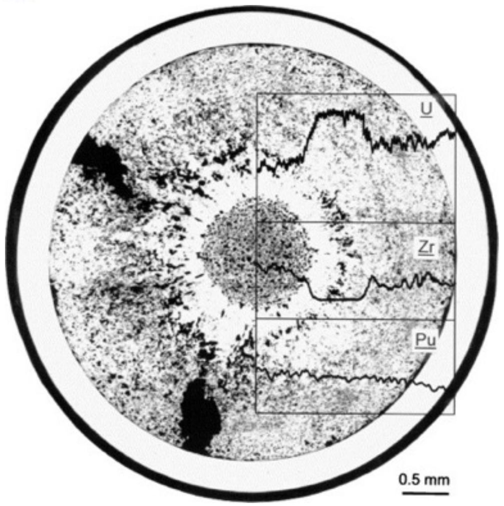


Uranium-Zirconium (UZr)

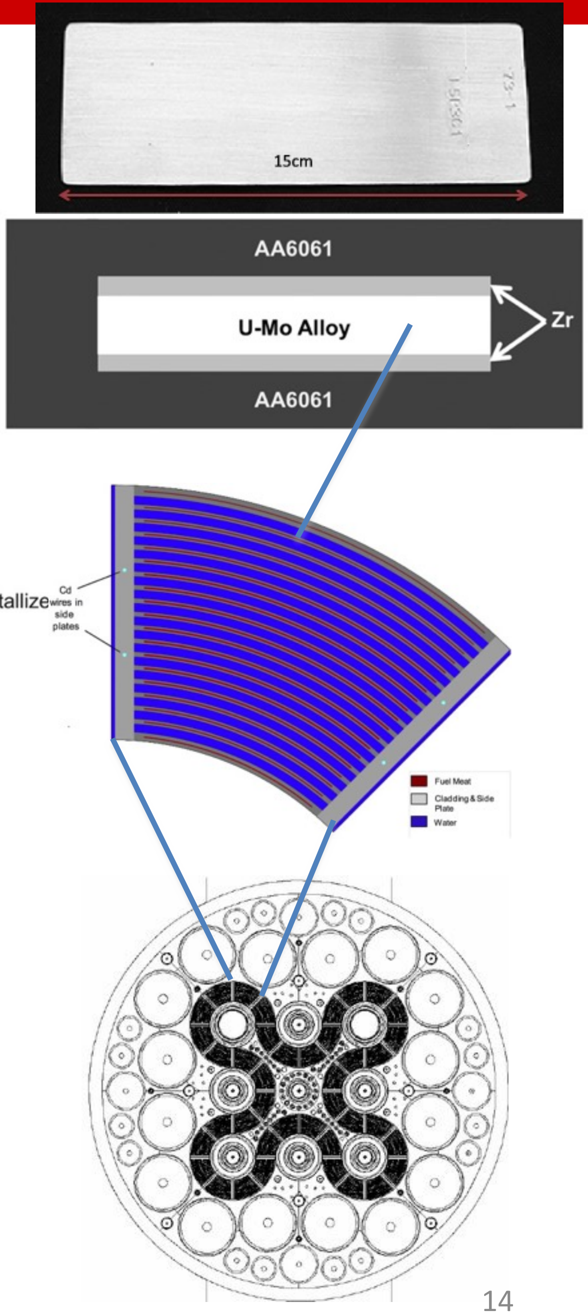
- Phenomena of interest
  - 30-50% swelling
  - Constituent redistribution
  - Alpha tearing
  - FCCI
- Good: High thermal conductivity; Stability to high burnups (> 20%); Flexible composition; Inherent safety- negative reactivity feedback, etc.
- Bad: Low melting point; Dramatic fuel swelling that must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



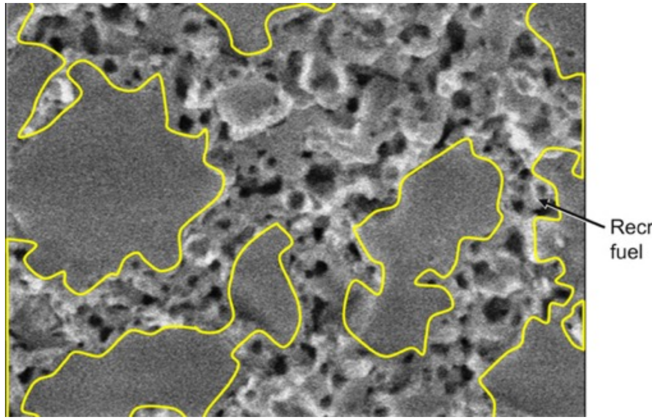


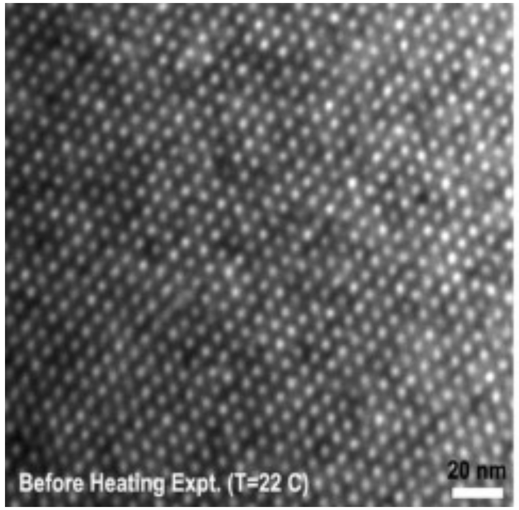


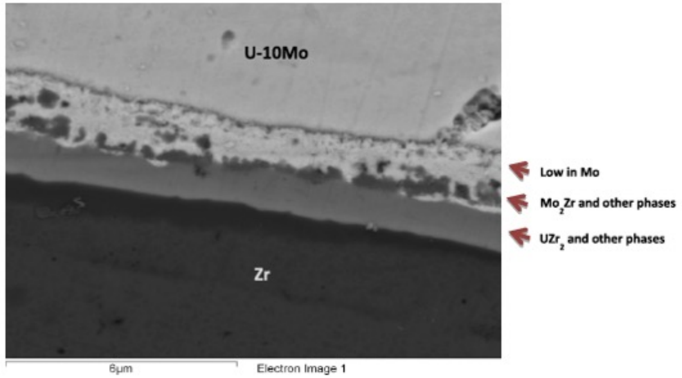
Uranium-Molybdenum (UMo)



- 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

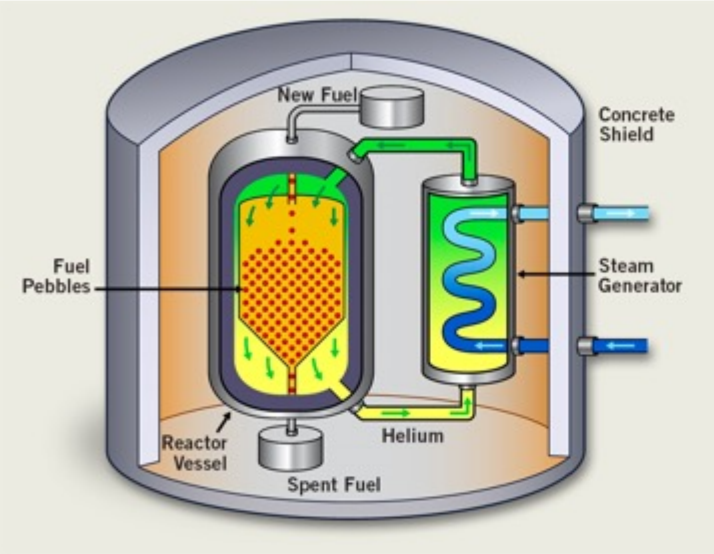


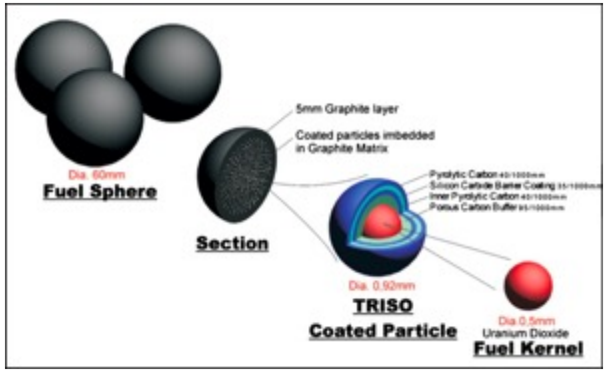




TRISO Particle Reactors

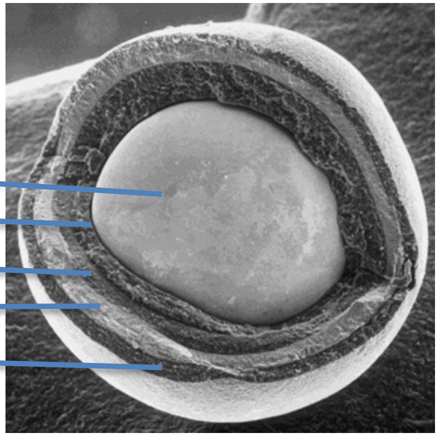
- High temperature gas reactors (HTGRs) or molten salt reactors (MSRs) can utilize TRISO particles as the fuel source
- Pebble bed and prismatic types of HTGRs
- Particles are agglomerated with graphite into a larger pebble, or into a cylindrical block
- Current designs utilize UCO (in the US), which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

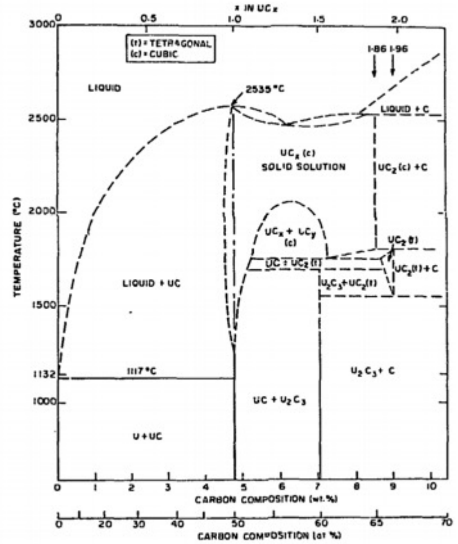


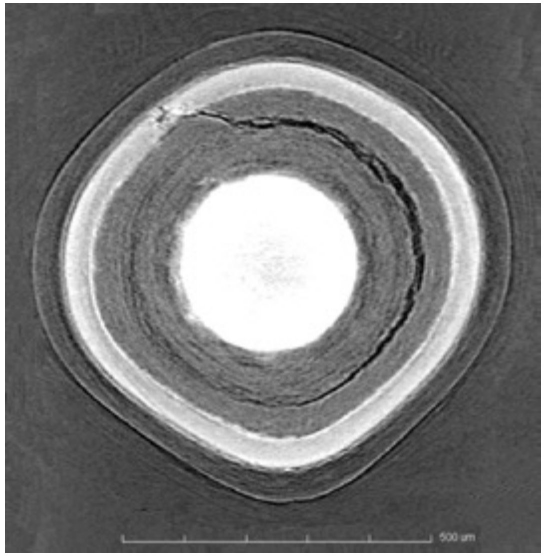


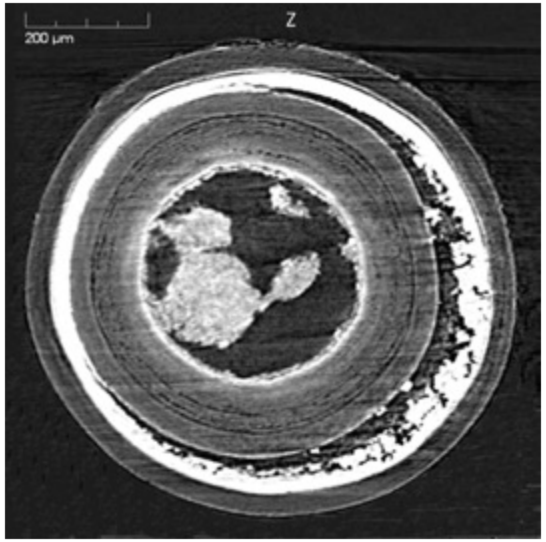
UC/UCO TRISO Fuels

- 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



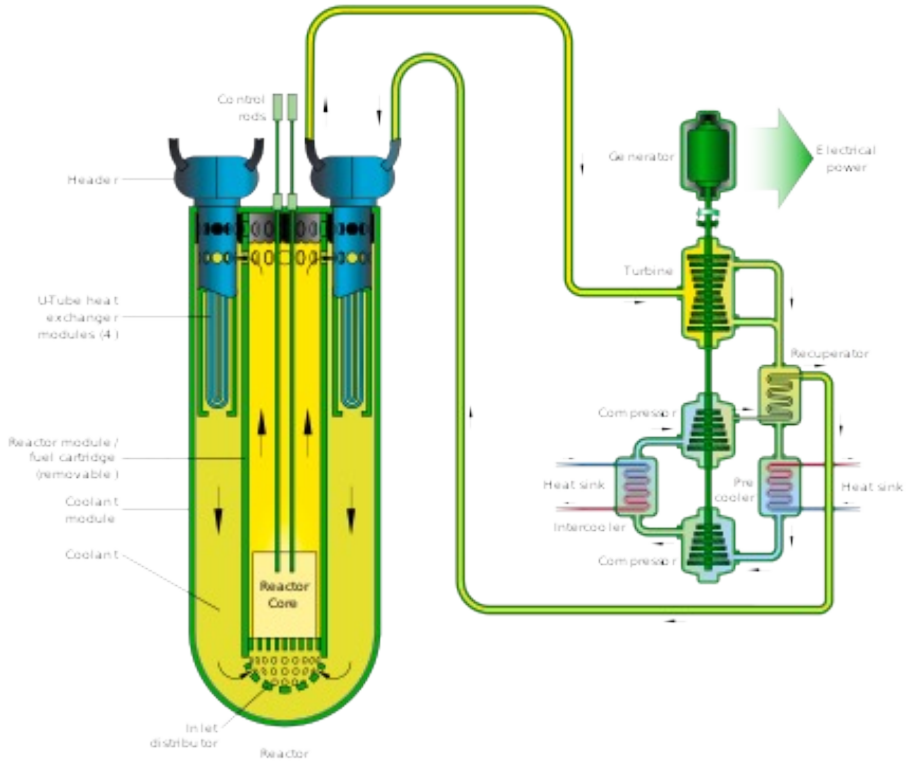






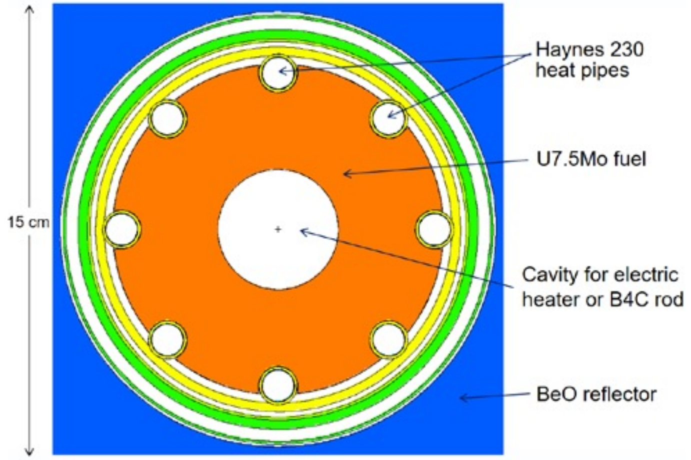
Uranium Nitride

- Potential fuel for gas cooled reactors, 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

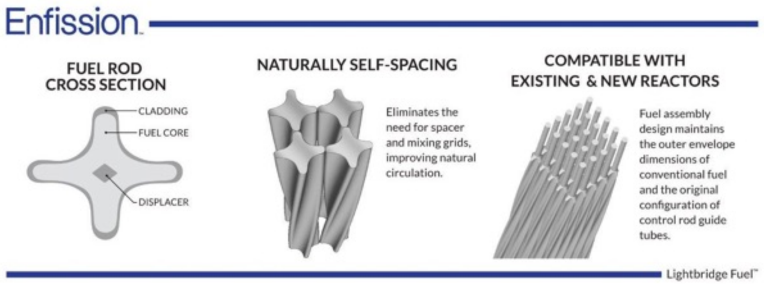


Unique Fuel Designs

KRUSTY



LightBridge



SSR

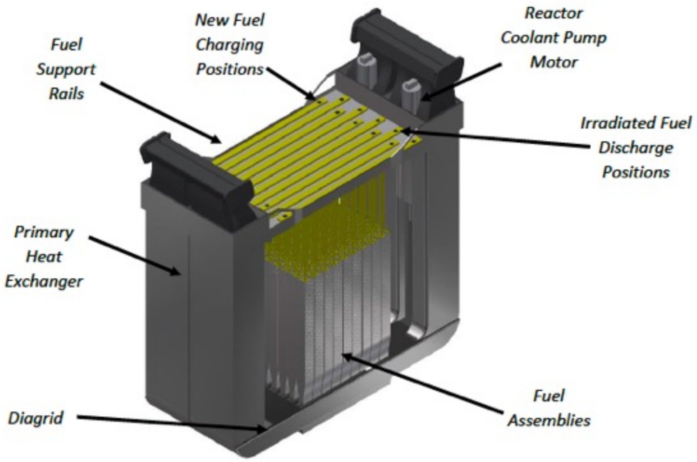
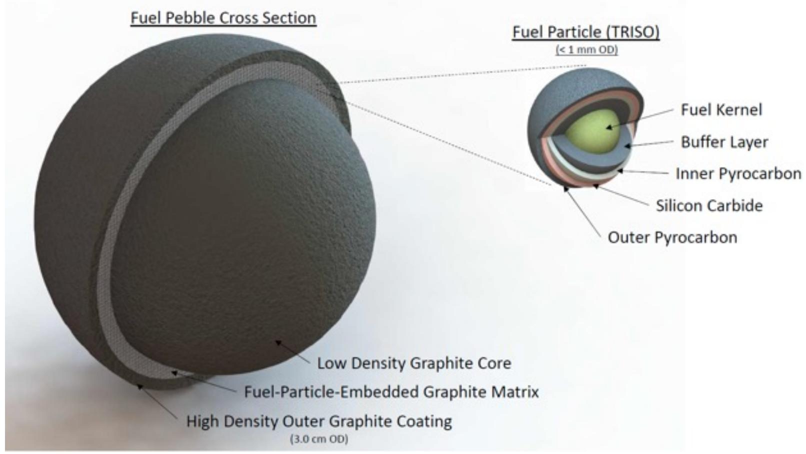


Figure 6: Overview of Stable Salt Reactor Core Module

Kairos



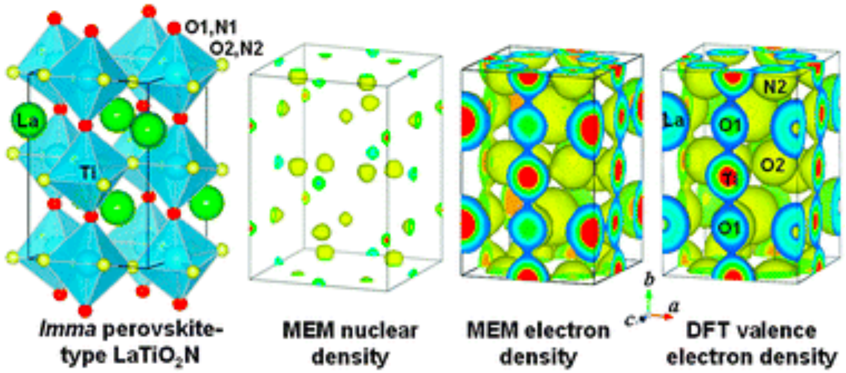
Summary

- There exist a number of nuclear fuels in different stages of utilization and development
- The more established ones are the less interesting ones…
- 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.

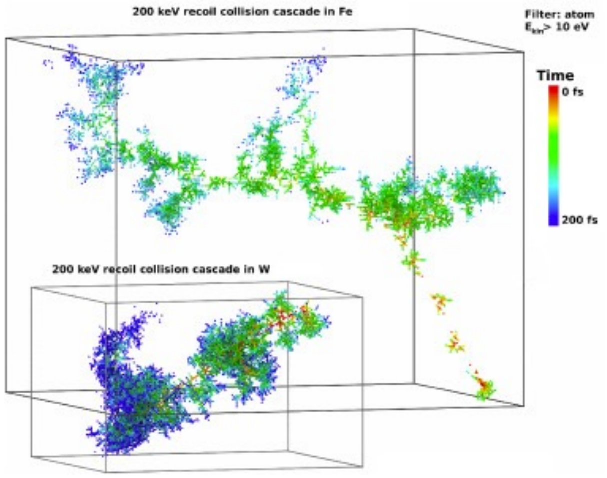
How do we model advanced fuels?

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



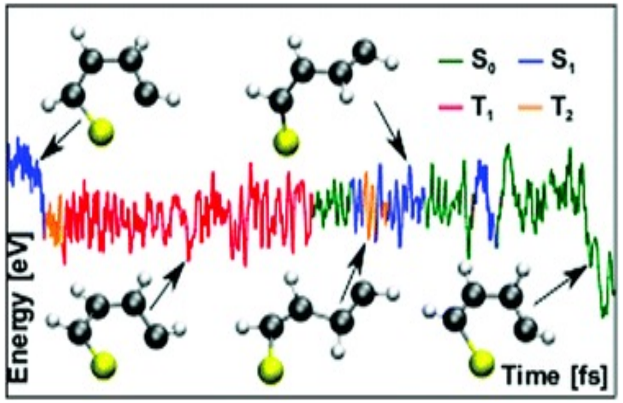
Yashima, https://doi.org/10.1039/C0CC00573H



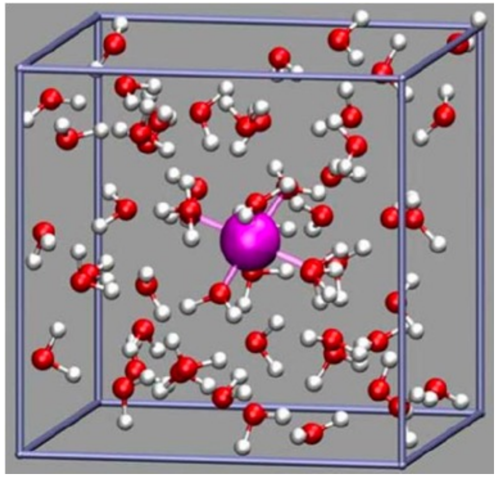
Sand, https://doi.org/10.1016/j.jnucmat.2018.08.049

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



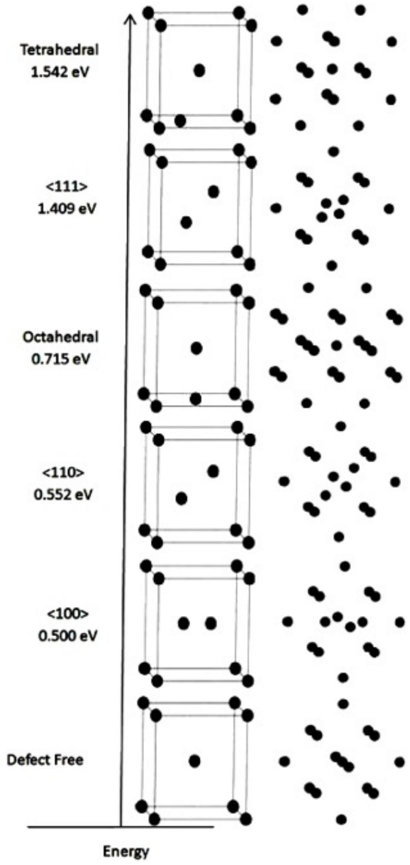
Schnappinger, https://doi.org/10.1039/C7CP05061E

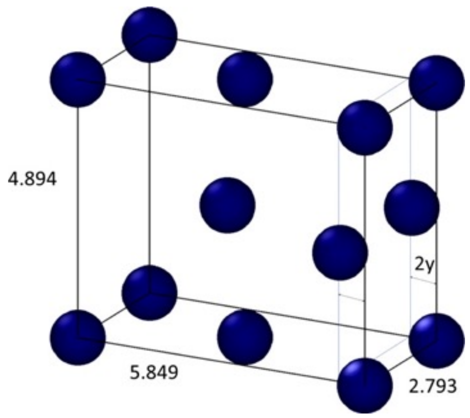


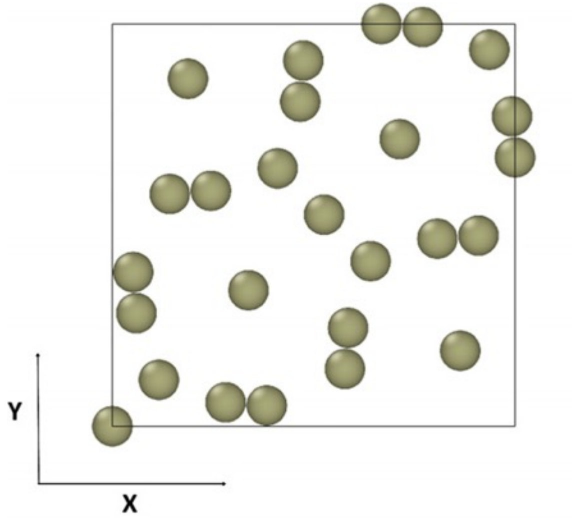
Yazyev, https://doi.org/10.1007/s00214-005-0052-6

DFT applied to Nuclear Fuels

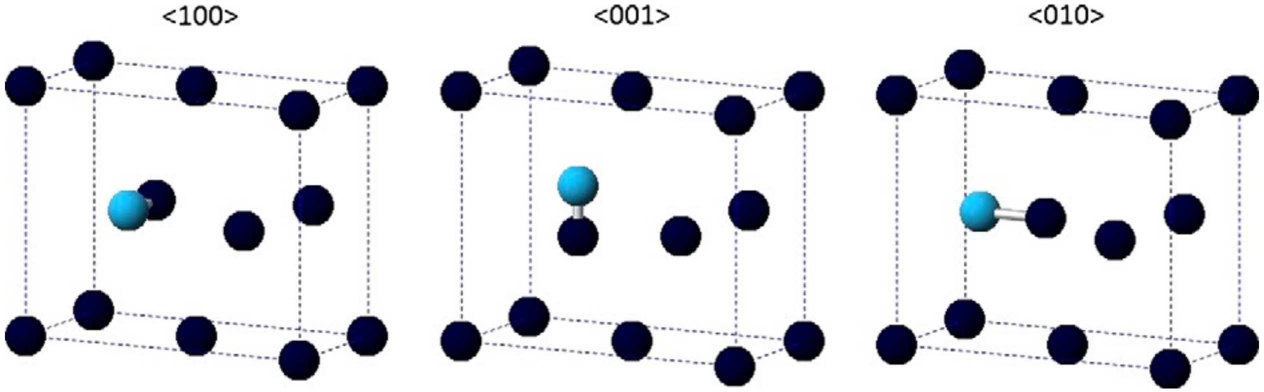
- Since we are dealing with the electronic structure, these calculations are very computationally expensive and limited to a few hundred atoms
- Typically employed to calculate lattice constants, elastic constants, point defects, electronic densities, phonon DOS, etc.
- DFT on advanced nuclear fuels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

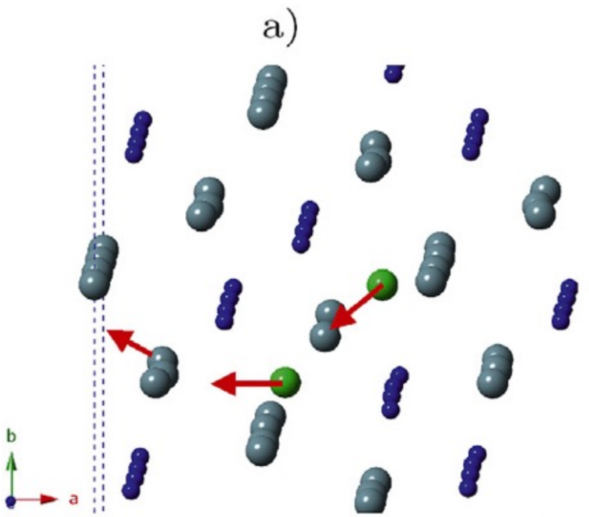


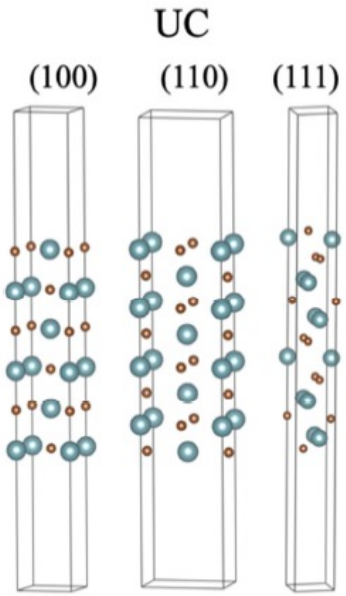


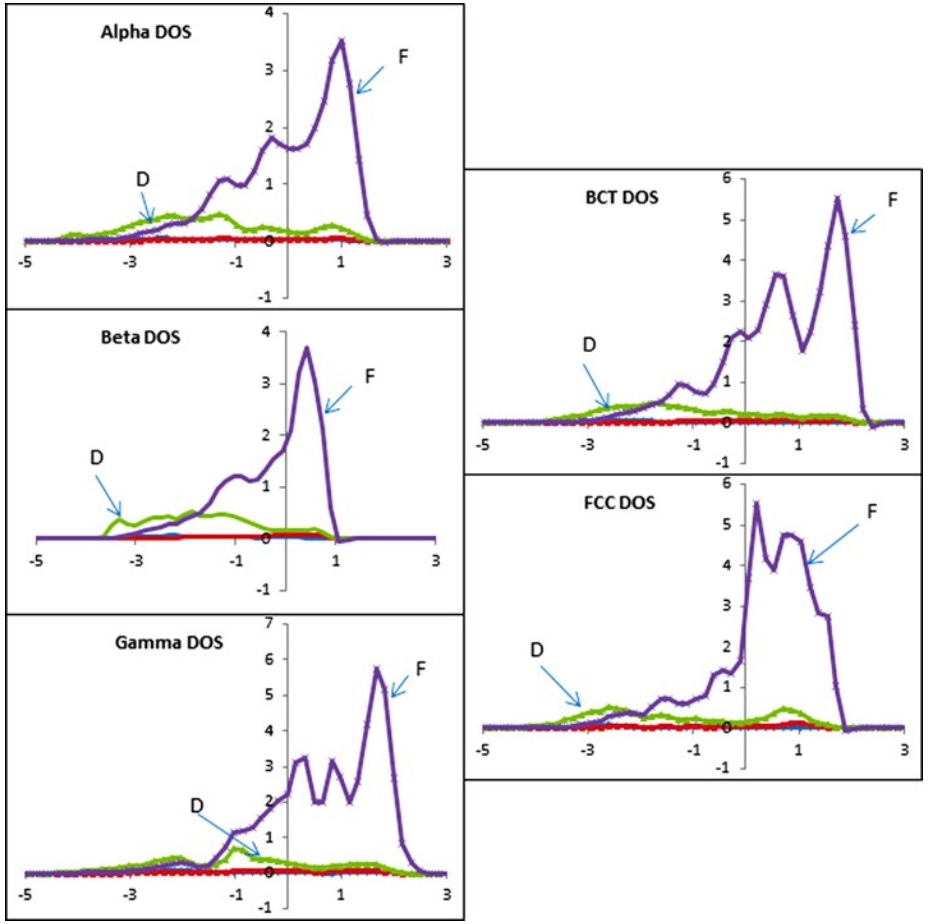


DFT applied to Nuclear Fuels



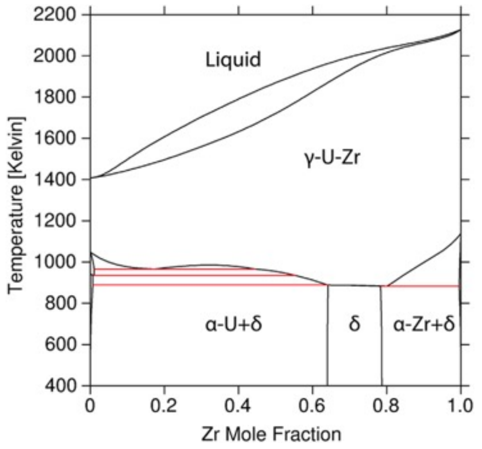


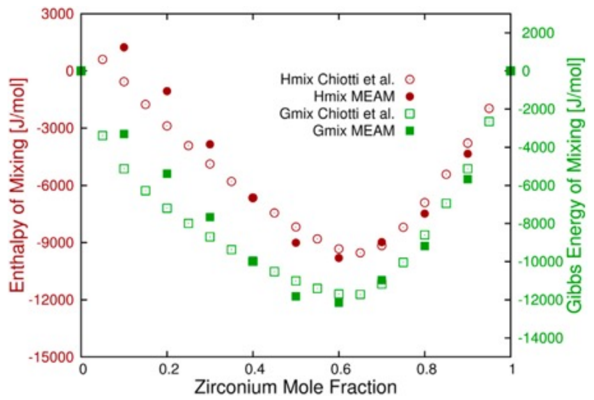


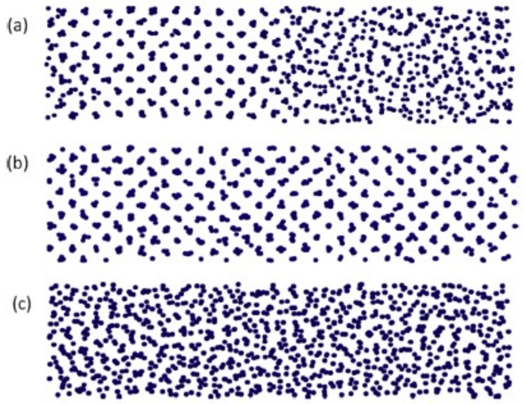


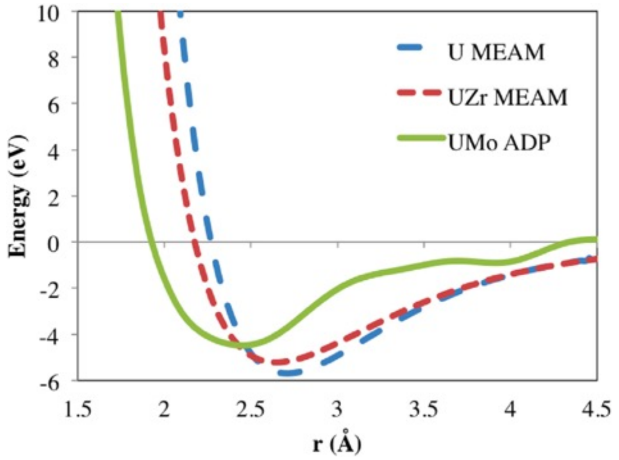
MD applied to Nuclear Fuels

- 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

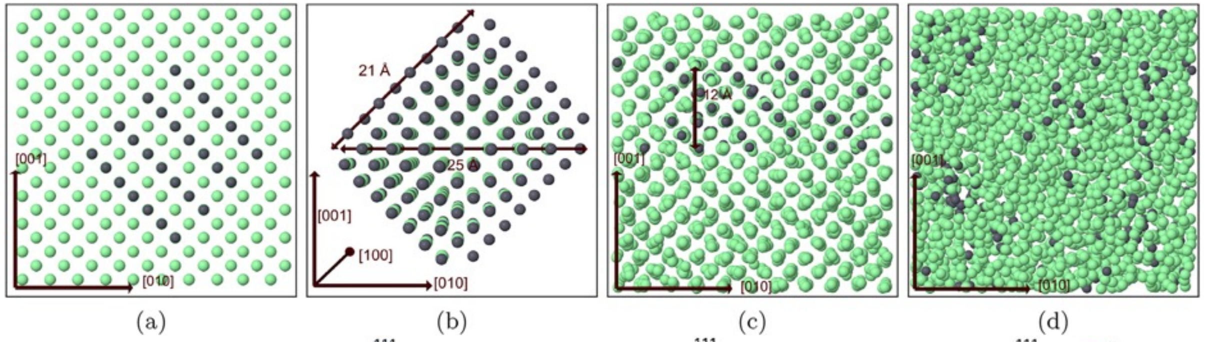


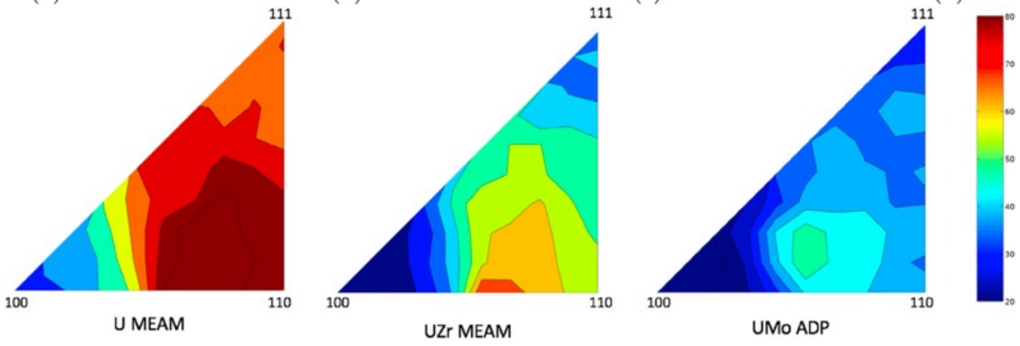


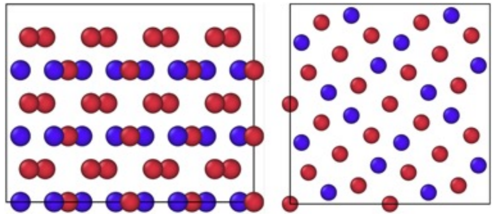


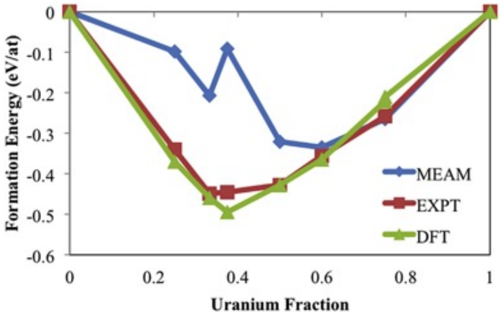


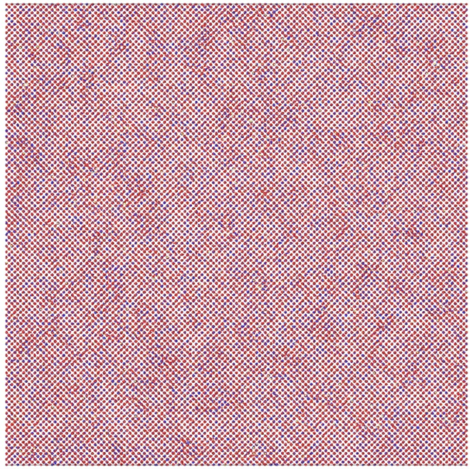
MD applied to Nuclear Fuels

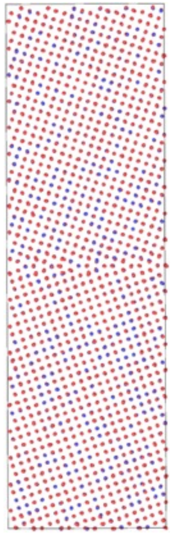


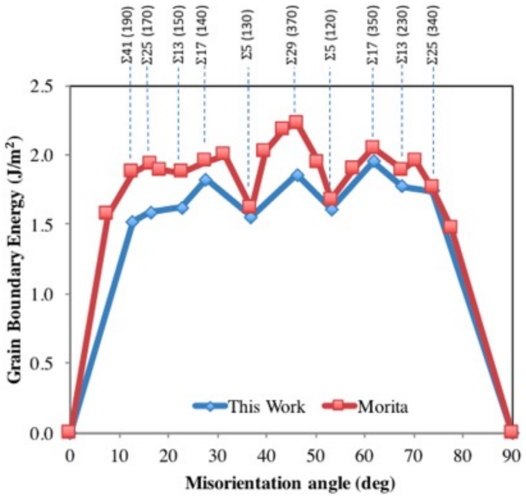


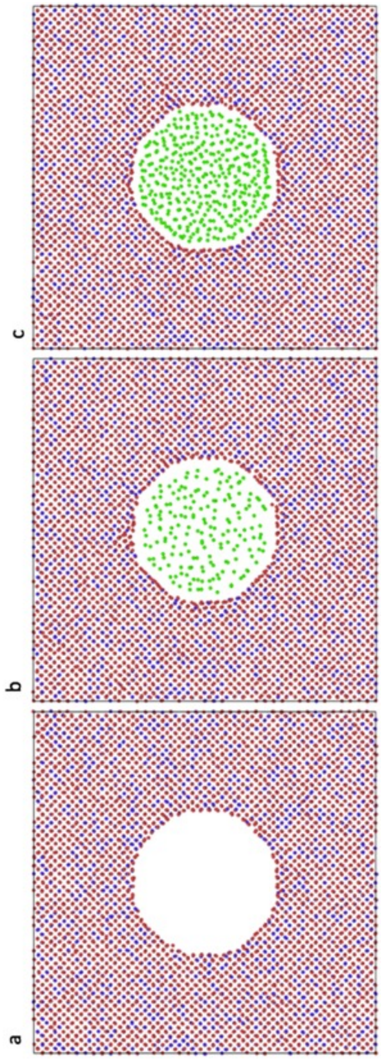






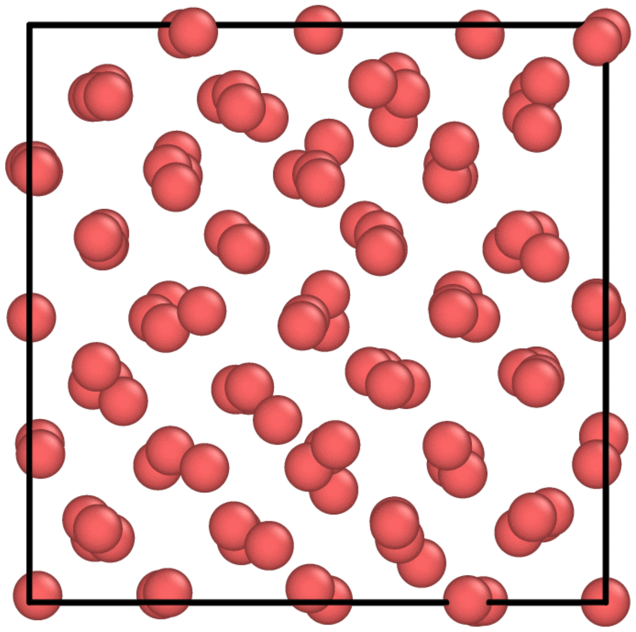




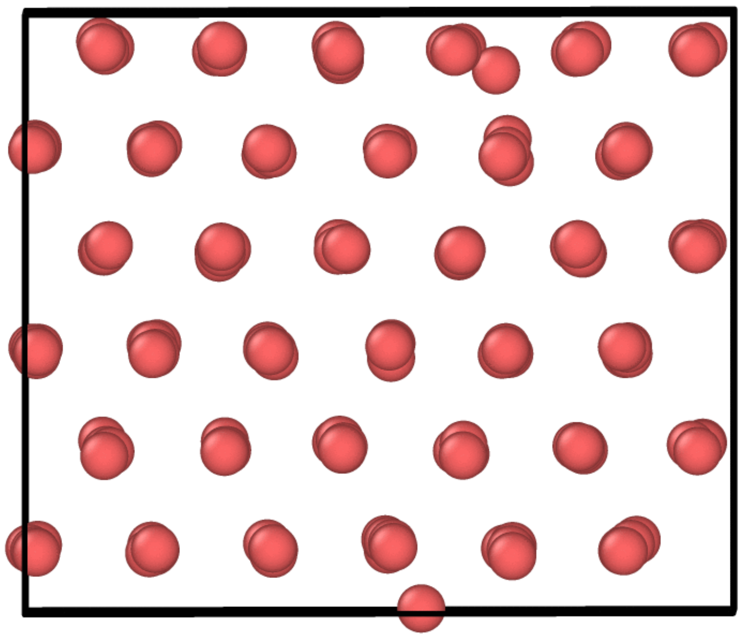


AIMD applied to Nuclear Fuels

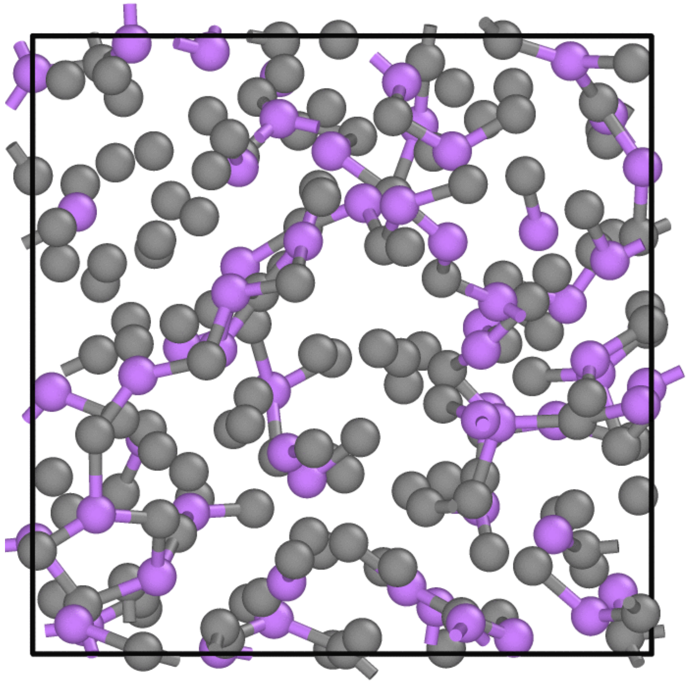
BCC U



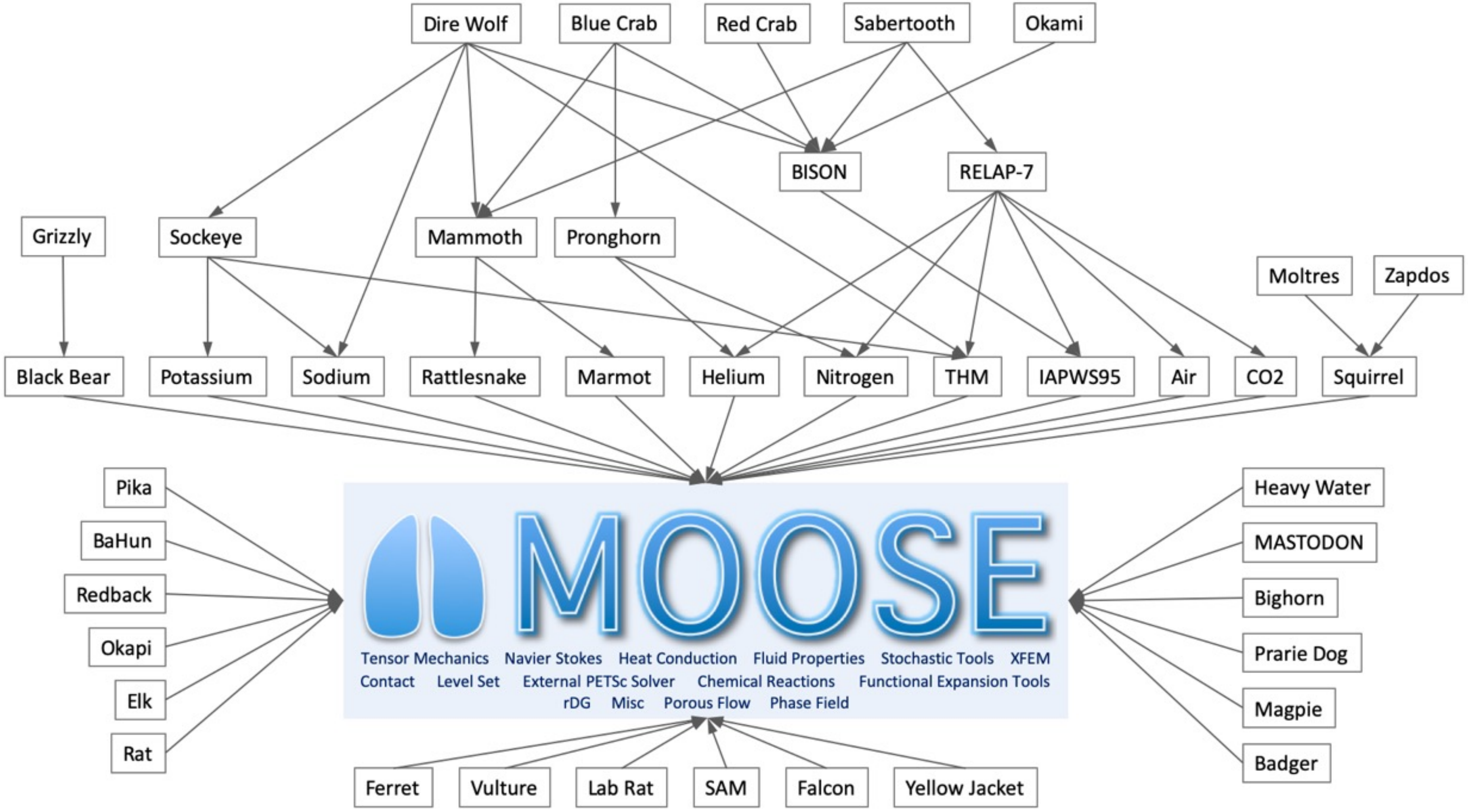
Alpha U with int



Eutectic LiCl-KCl

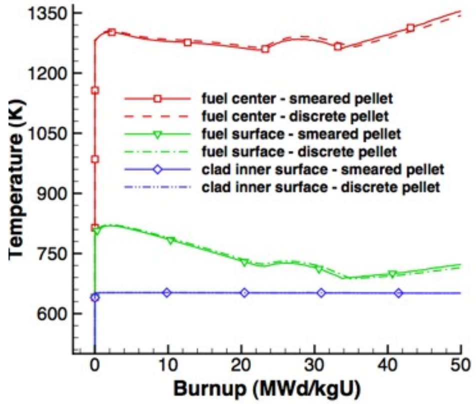


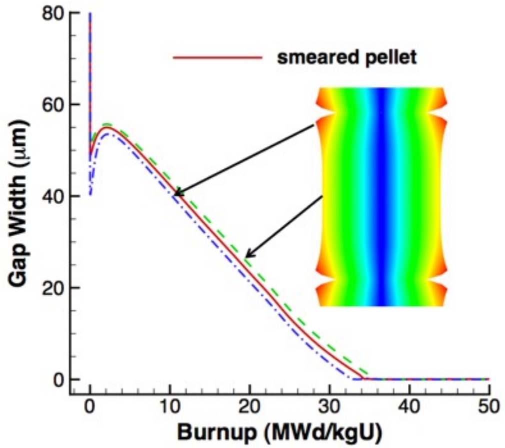
Integration into Fuel Performance Models

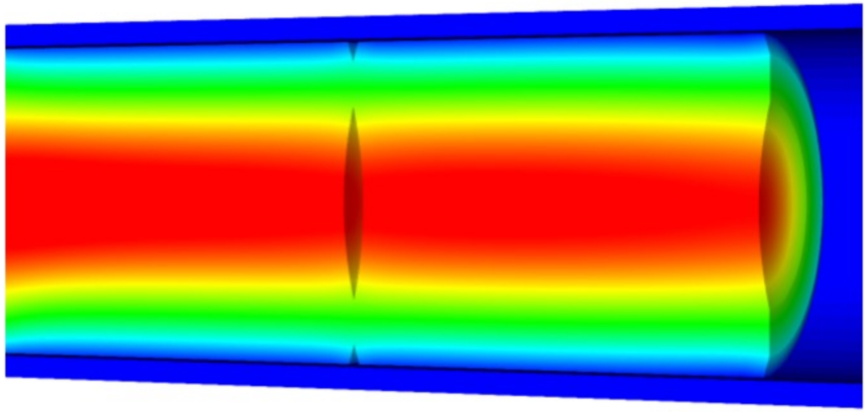


Integration into Fuel Performance Models

- 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!



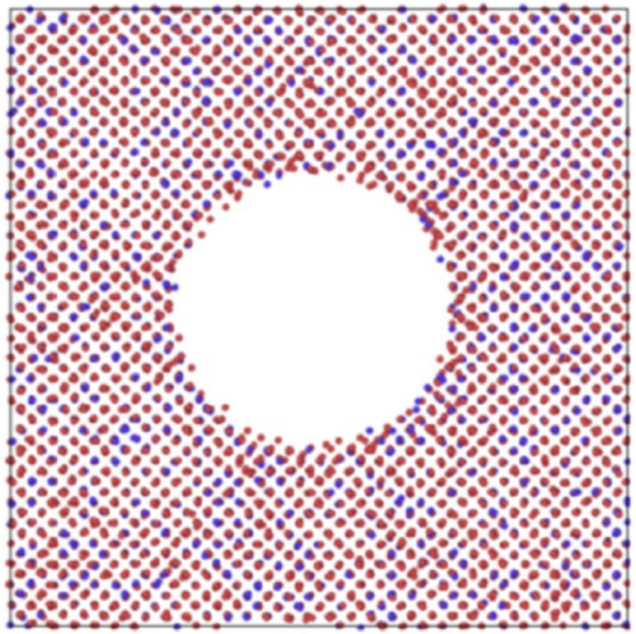


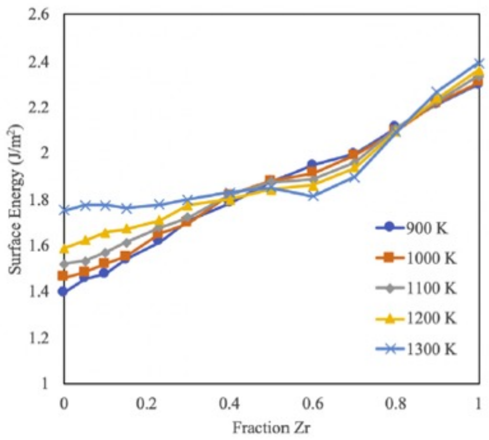


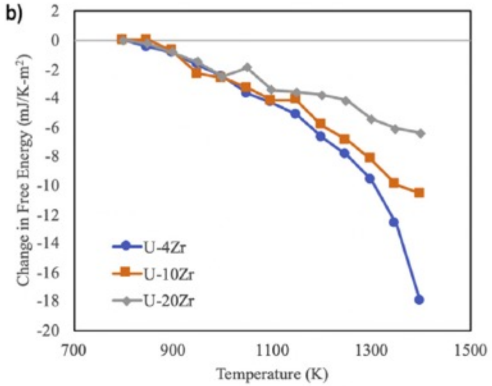
Integration into Fuel Performance Models

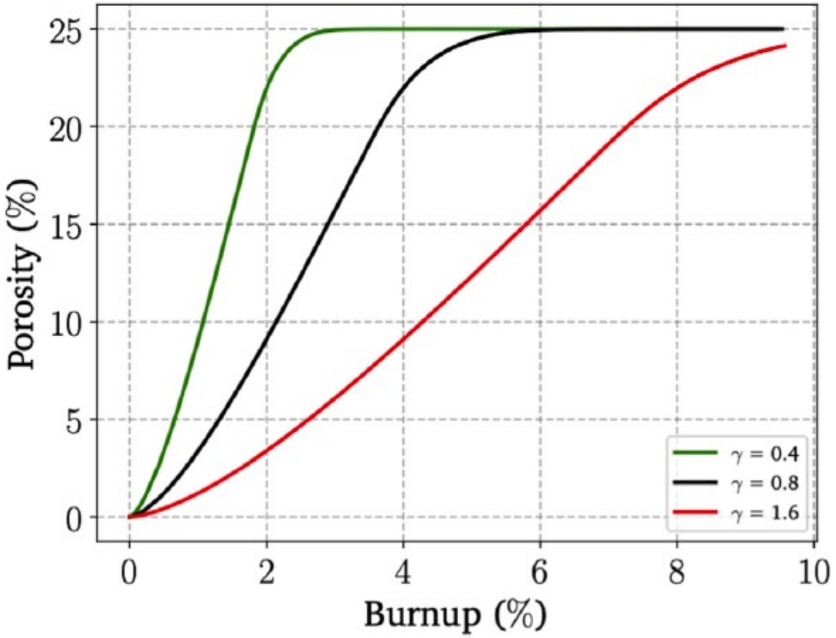
- Example of U-Zr fission gas swelling

$$\left(\frac{\Delta V}{V_0}\right)_g = \frac{4\pi}{3}R^3 N \qquad R = \sqrt{\frac{3k_B T Y_{gas}\dot{F}t}{4\pi 2\gamma N}},$$



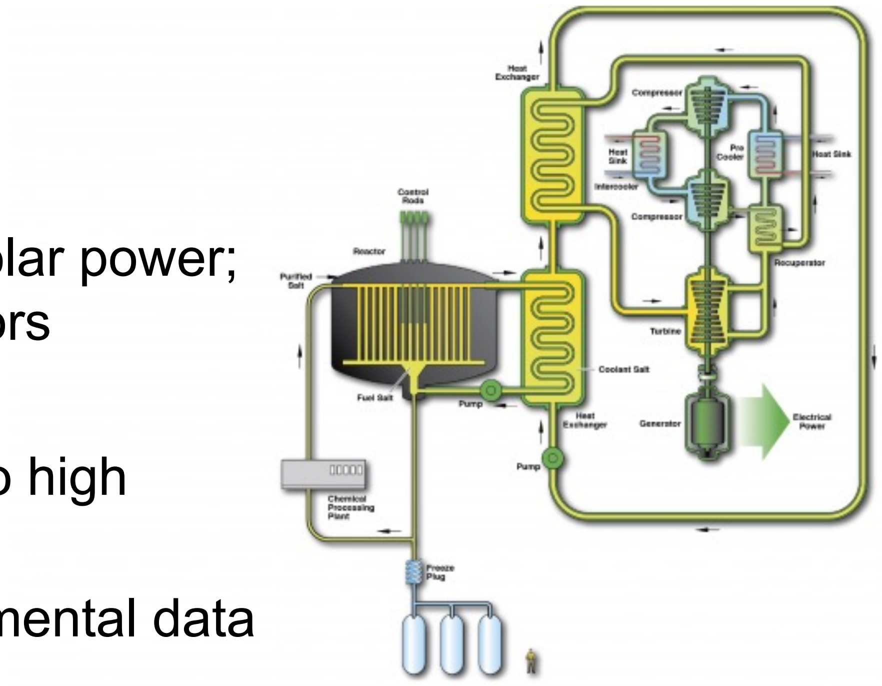


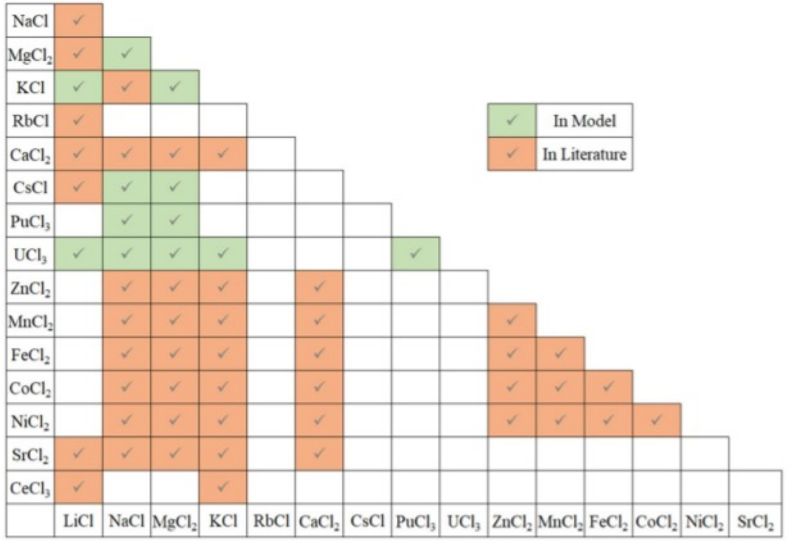


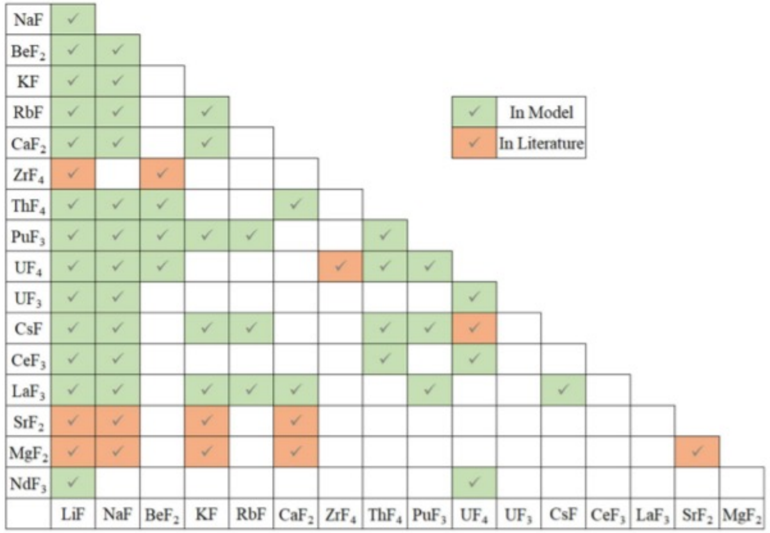


Molten Salts

- 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





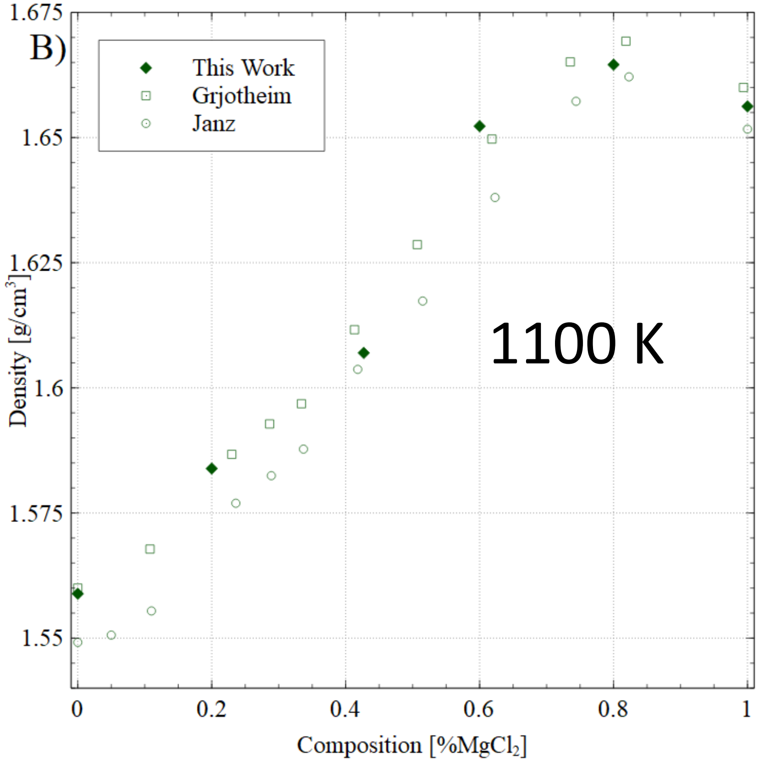


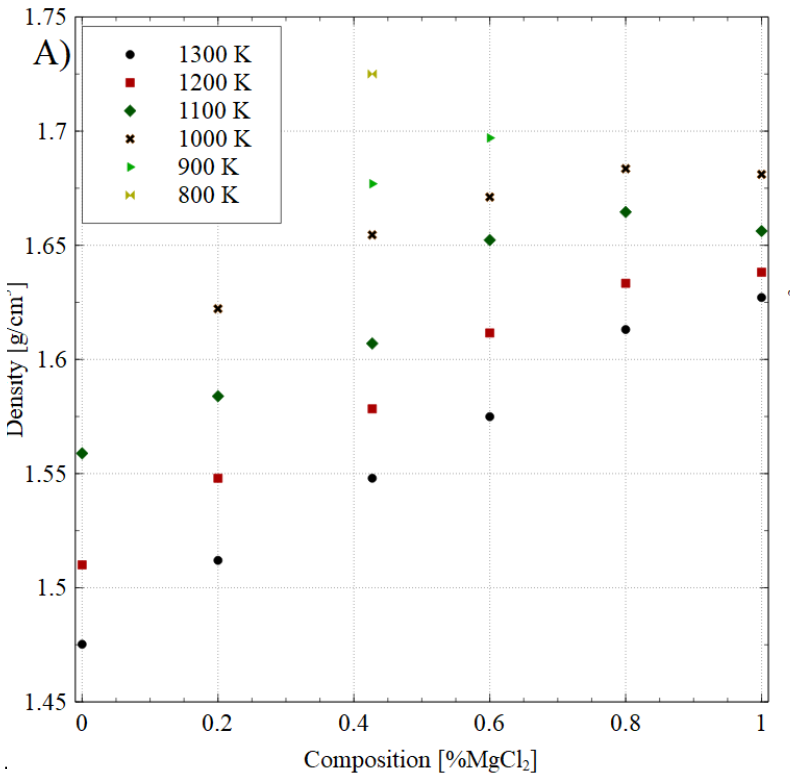
What do we do?

- 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