INL/CON-23-73191-Revision-1

Effective Lanthanide Diffusivity through U-Zr Metallic Fuel

October 2023

Larry Kenneth Aagesen Jr, Chao Jiang, Jacob Aaron Hirschhorn

Changing the World's Energy Future

INL is a U.S. Department of Energy National Laboratory operated by Battelle Energy Alliance, LLC

INL/CON-23-73191-Revision-1

Effective Lanthanide Diffusivity through U-Zr Metallic Fuel

Larry Kenneth Aagesen Jr, Chao Jiang, Jacob Aaron Hirschhorn

October 2023

Idaho National Laboratory
Idaho Falls, Idaho 83415

http://www.inl.gov

Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517

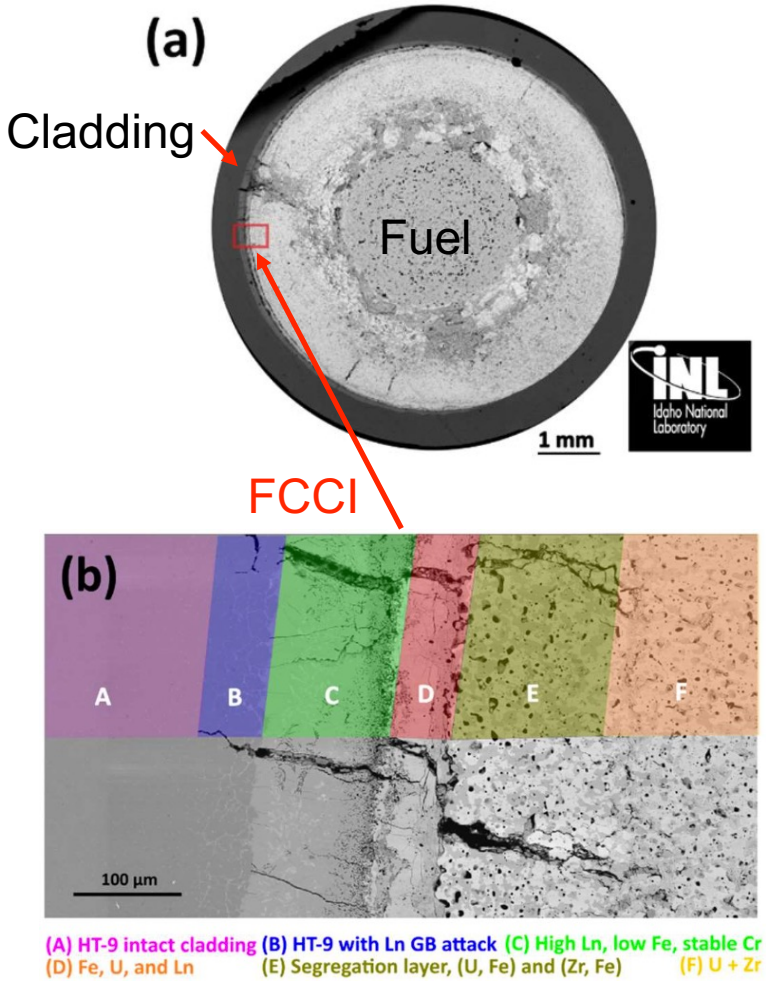
Effective Lanthanide Diffusivity through U-Zr Metallic Fuel

Larry Aagesen, Jacob Hirschhorn, Chao Jiang

Idaho National Laboratory

Idaho National Laboratory

Fuel-Cladding Chemical Interaction (FCCI) in U-(Pu)-Zr metallic fuels

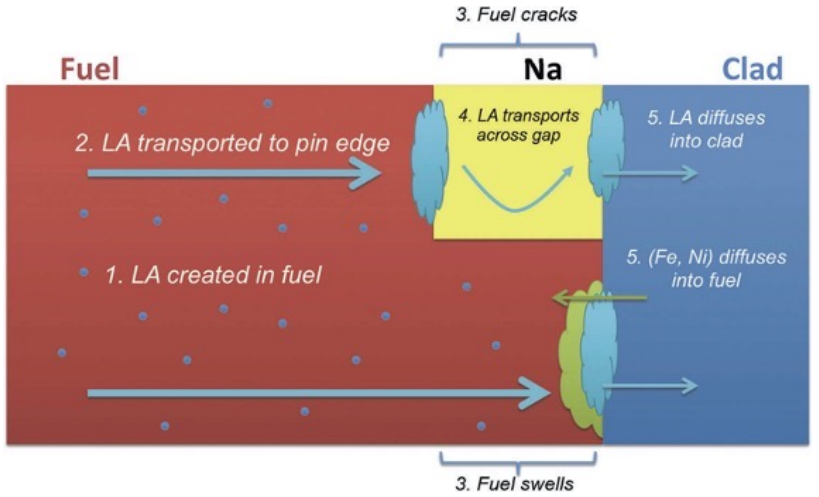


- FCCI is a major performance limitation for U-Zr fuels
- Complex interactions resulting from interdiffusion at fuel-cladding interface
  - Cladding: various stainless steels have been used (HT9, D9)
- Fuel
  - Diffusion of Fe into U can result in low-melting temperature eutectic
- Cladding
  - Diffusion of lanthanide fission products from fuel to cladding leads to formation of brittle intermetallic phases, referred to as “wastage”
  - Most prevalent intermetallic: $(Fe,Cr)_{17}(Nd,Ce)_2$
  - Degrades cladding mechanical properties

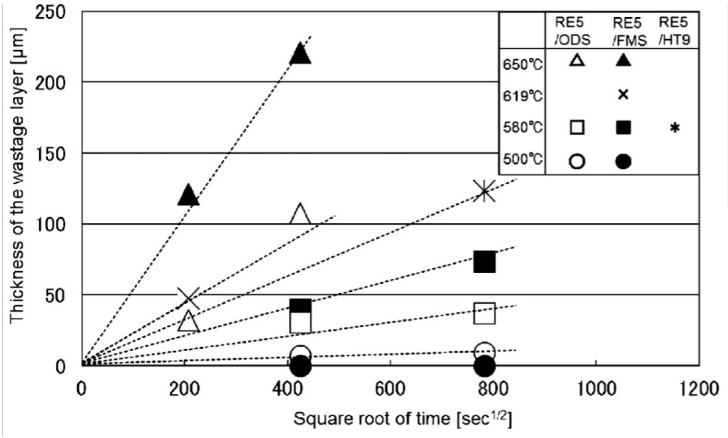
Wang et al., J. Nucl. Mater., 572, 153990, 2022

Cladding wastage formation

- Lanthanide fission products (LA) generated in fuel
  - Most prevalent: Nd, Ce, La, Pr, Sm
- Diffuse to pin edge
- EBR-II design: liquid Na in fuel-cladding gap
  - Prior to contact by swelling: LA diffusion through liquid sodium
  - Pre and post contact: fast transport from pin edge to cladding
- Several intermetallic phases are observed, most prevalent: $(Fe,Cr)_{17}LA_2$
- Diffusion couple data suggests wastage thickness $\delta = \sqrt{Kt}$



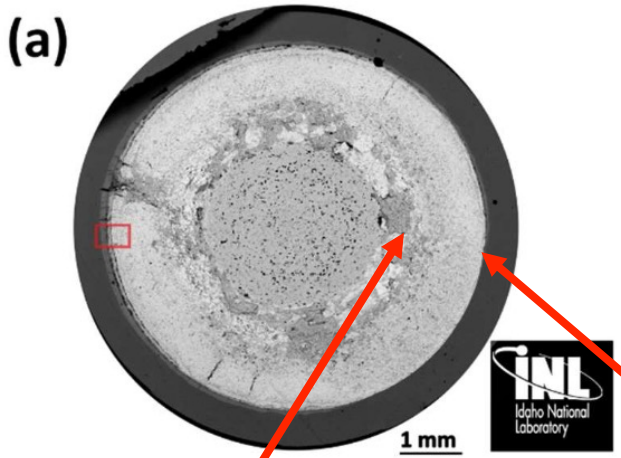
Matthews et al., Nucl. Tech. 198 (2017) 231

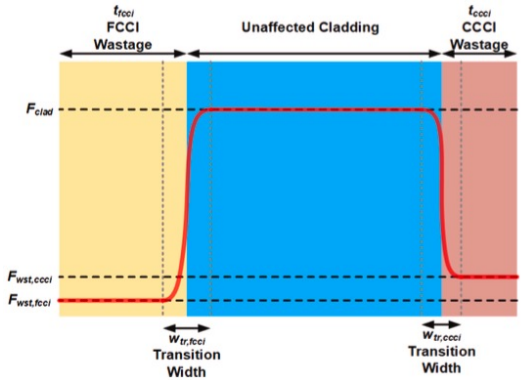


Diffusion couple data from
Inagaki et al., Trans. AES Japan, 12, p.149 (2013)

FCCI Mechanistic Modeling approach

- Empirical FCCI model available in BISON based on EBR-II data
  - May not be readily extensible to new reactor designs
- Mechanistic model under development, initial focus on Nd





Cladding degradation

Miao et al., NED, 385, 111531, 2021

Nd diffusion in fuel:

$$\frac{\partial c}{\partial t} = \nabla \cdot D_{eff} \nabla c + y\dot{F} - \lambda c$$

Growth rate of $Fe_{17}Nd_2$

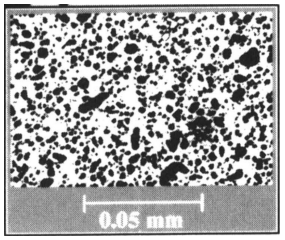
$$\delta = (Kt)^{\alpha}$$

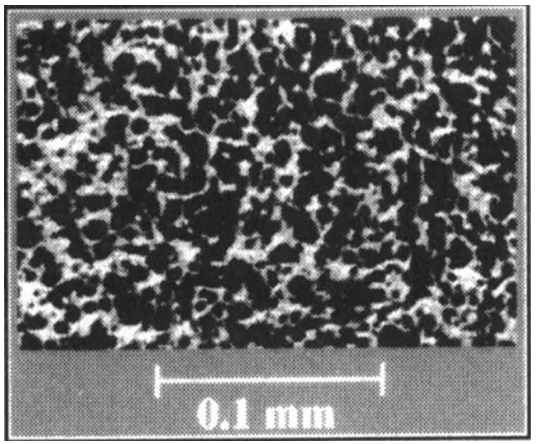
$$K = K_0 c e^{-E_a/kT}$$

δ

Determining effective diffusivity of Nd

- Need to account for fuel microstructure
  - Isolated porosity
  - Interconnected porosity
    - Interconnectivity function f_V
  - Interconnected, sodium-logged porosity due to infiltration from bond sodium
    - BISON model: $p_{logged} = f_V p_{tot} f_{ref}$ (based on data from Bauer)
- Need diffusivity of Nd through bulk, surface, and sodium

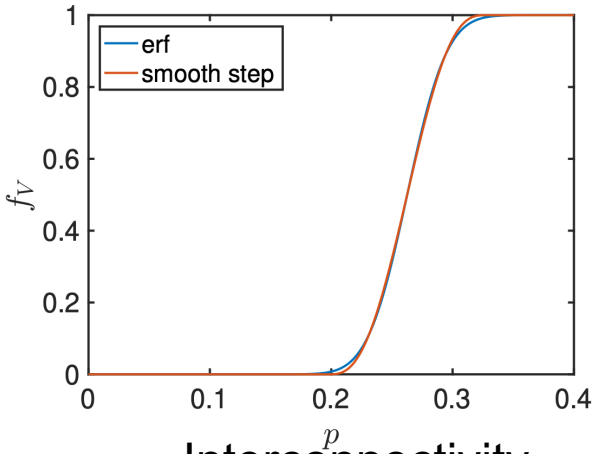




1.3% burnup
Isolated

2.1% burnup
Interconnected

Bauer & Holland, Nucl. Tech, 110, p. 407 (1995)

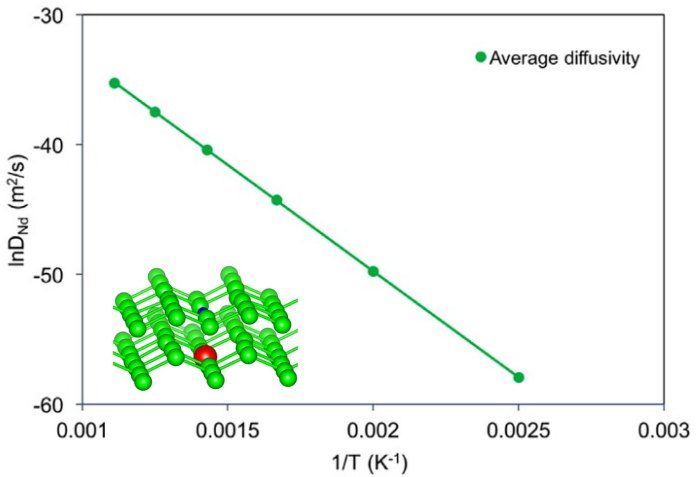


Interconnectivity

Aagesen et al., Mat. Theory, 6:8, 2022.

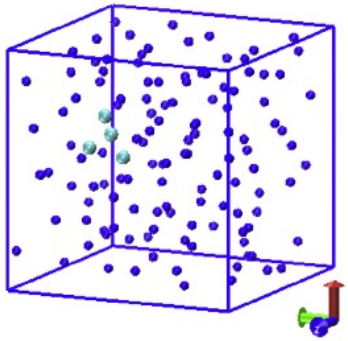
Nd diffusivity: atomistic calculations through bulk, surface, Na

- Bulk solid diffusivity
  - Assume α phase dominates U microstructure
  - DFT calculation showed Nd interstitial in α-U is unstable, so assume vacancy mechanism
  - Use Nudged Elastic Band to calculate energy barriers
  - Kinetic Monte Carlo to calculate diffusivity in a,b,c directions, polycrystalline average
- Surface diffusivity
  - Same methodology, calculate on (001) α-U surface
- Diffusivity through liquid sodium
  - Previous ab-initio molecular dynamics (AIMD) simulations from literature



Diffusivity of Nd in polycrystal α-U

Jiang et al, JNM, 557, 153307 (2021)

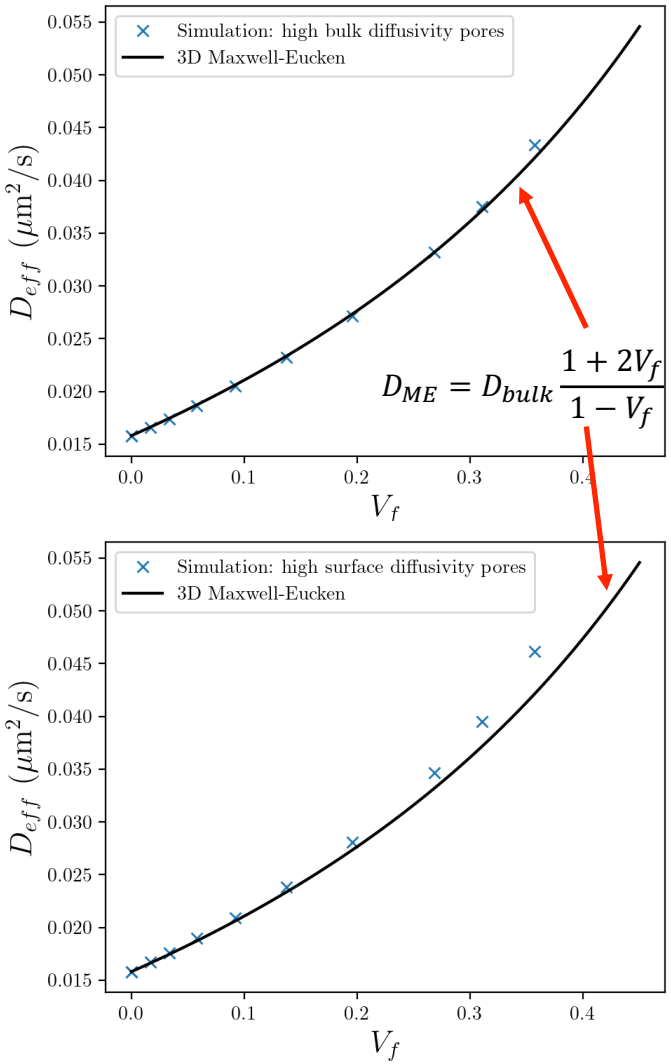


Nd diffusivity in Na: AIMD

Li et al., JNM, 484, p. 98-102 (2017)

Effective diffusivity of Nd through isolated porosity

- Higher diffusivity on pore surfaces may increase effective diffusivity somewhat
- Maxwell-Eucken model of effective transport
  - When diffusivity in spherical inclusions is much higher than matrix,
  - $D_{ME} = D_{bulk} \frac{1+2V_f}{1-V_f}$
- Hypothesis: this expression still valid for high surface diffusivity
  - Test by calculating D_{eff} using asymptotic expansion homogenization (AEH) in MOOSE*, for case of isolated spherical pores
- **Use Maxwell-Eucken model for isolated porosity**



* Hales et al., Comp. Mat. Sci., 99, p. 290 (2015)

Effective diffusivity of Nd through interconnected porosity, no sodium infiltration

- No analytical theory available, so need to calculate and fit a function
  - Need to account for interconnected microstructure
- Large 3D simulations required to get statistics, so start with the simplest model possible
  - Cahn-Hilliard model, single defect species with source term for production in the solid
  - Free energy with minima at normalized defect concentrations $c = 0$ and $c = 1$

$$F = \int_v (f_b + f_{grad}) dV \qquad f_b = W c^2 (1-c)^2 \qquad f_{grad} = \frac{\kappa}{2} |\nabla c|^2$$

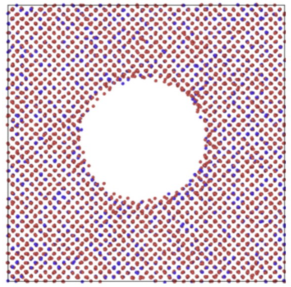
$$\mu = \frac{\delta F}{\delta c} = \frac{\partial f_b}{\partial c} - \kappa \nabla^2 c$$

$$\frac{\partial c}{\partial t} = \nabla \cdot (M \nabla \mu) + S$$

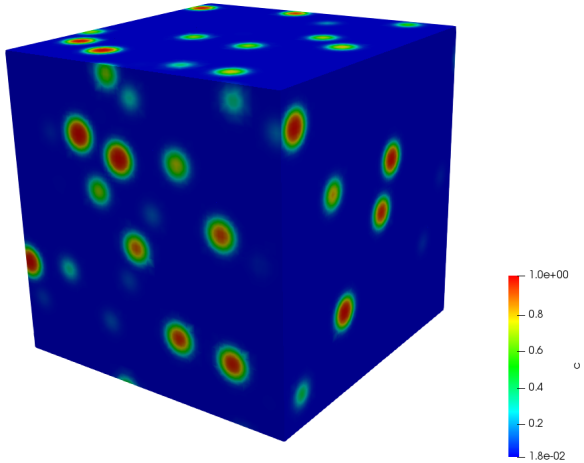
$$S = s_0 [1 - h(c)]$$

Effective diffusivity of Nd through interconnected porosity: phase-field simulations

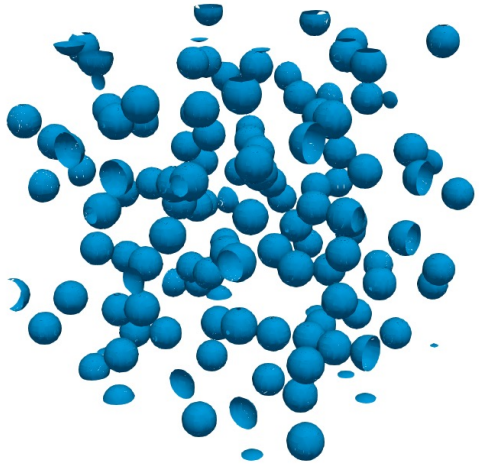
- Initial conditions: Randomly placed isolated bubbles at $N = 3 \times 10^{14}/m^3$, as determined from experiment
- Interfacial energy 1.8 J/m^2 from atomistic calculations
- Diffusivity not well known, so vary parametrically with fixed source strength to see effect



MD calculations to determine interfacial energy (Beeler et al., J. Nucl. Mat., 540, 152271, 2020)



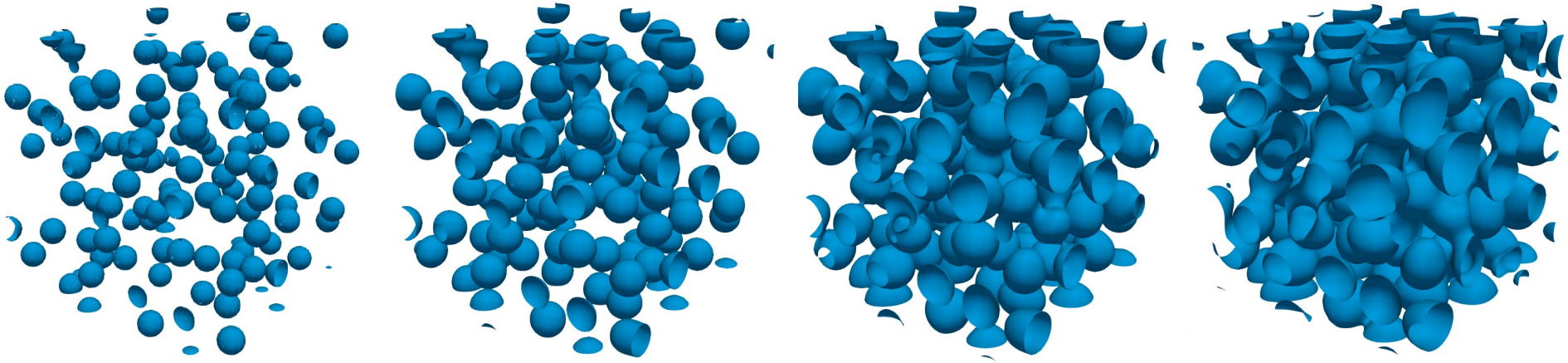
Initial conditions, simulation volume $(72\ \mu m)^3$



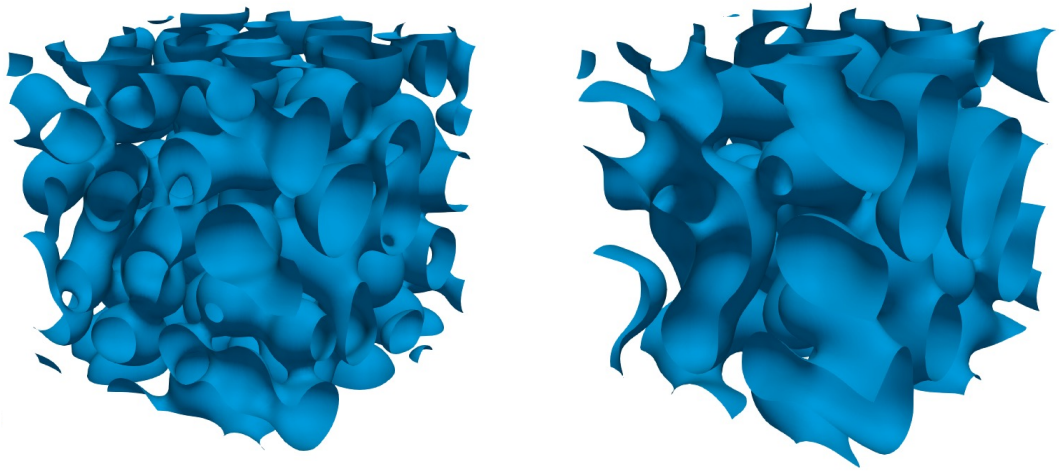
c = 0.5 contour

Phase-field simulation results

- $(72\ \mu m)^3$ domain, 112 bubbles for $N = 3 \times 10^{14}/m^3$
- Time evolution for $D = 2\ nm^2/s$:



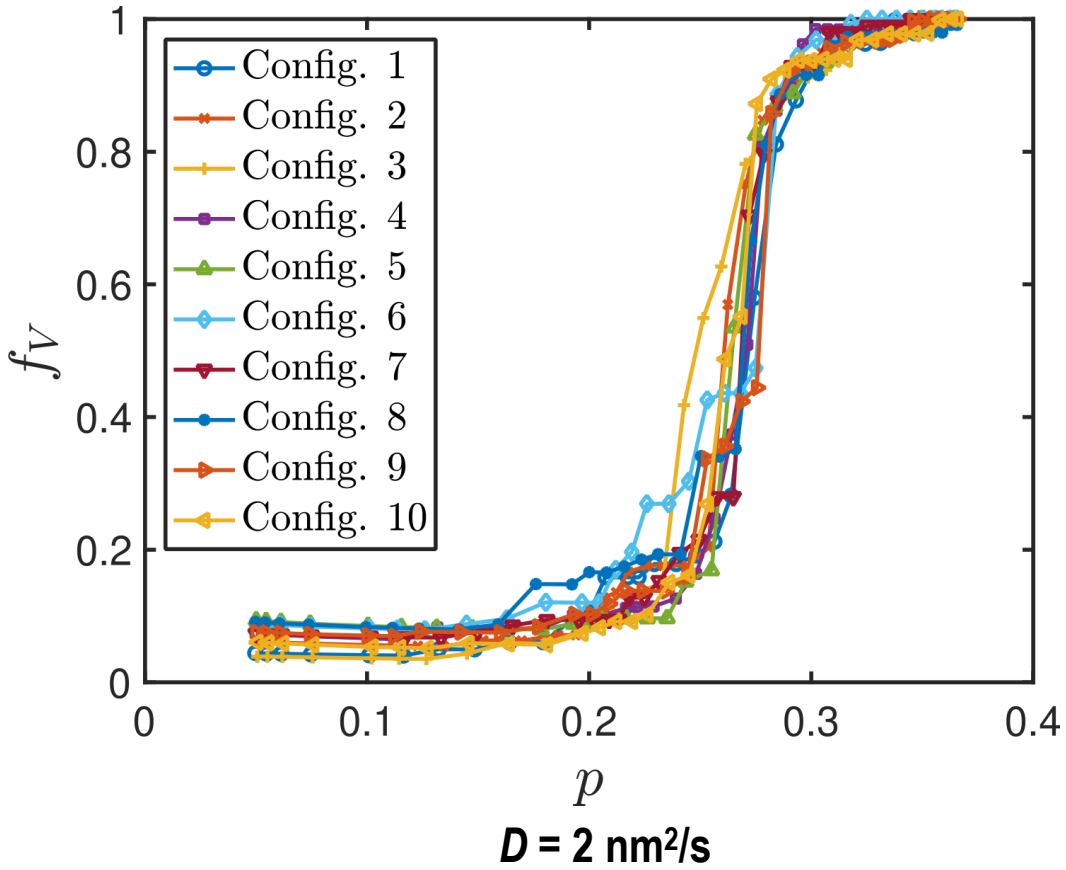
- Effect of diffusivity on morphology:



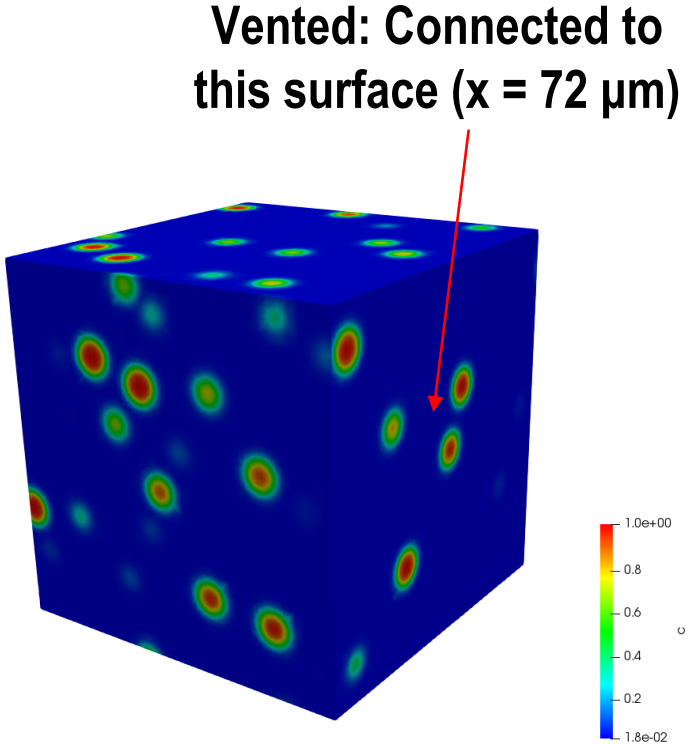
$D = 2\ nm^2/s$, $t = 1.14 \times 10^8$ s $D = 10\ nm^2/s$, $t = 1.14 \times 10^8$ s

Effective diffusivity of Nd through interconnected porosity: interconnectivity function

- Fraction vented to surface as a function of porosity



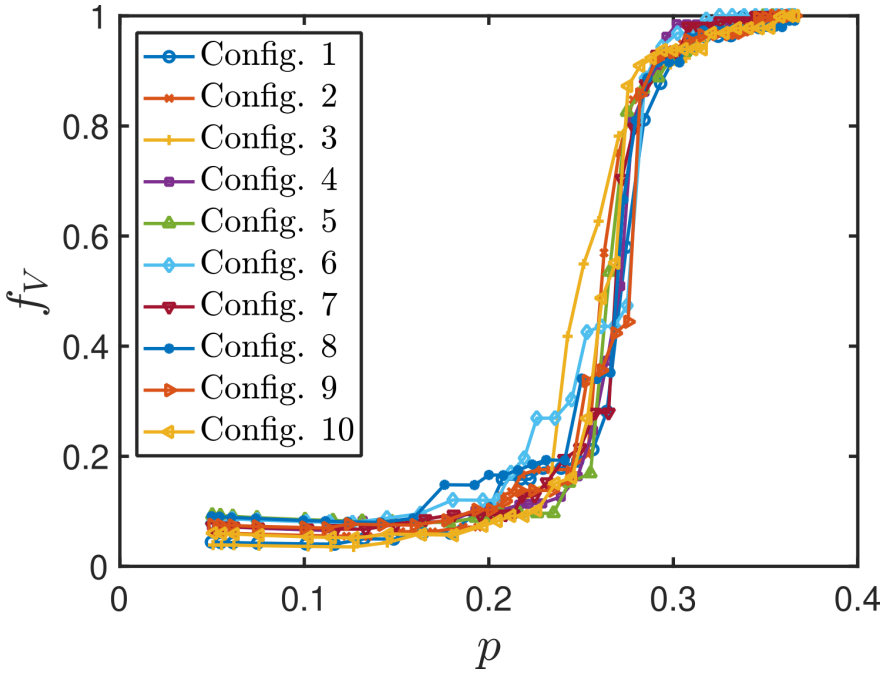
D = 2 nm²/s

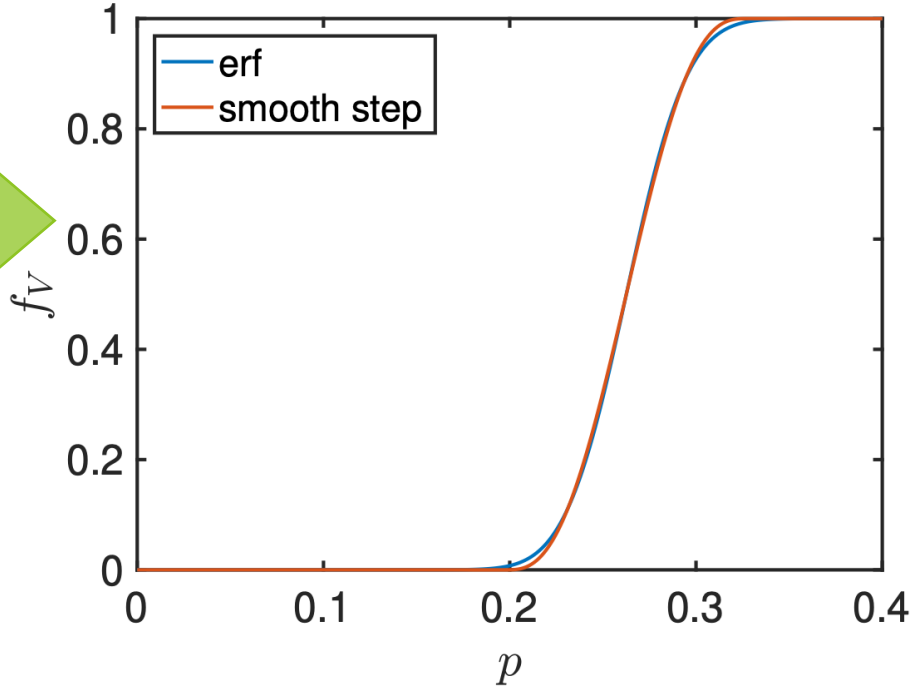


Aagesen et al., Mat. Theory, 6:8, 2022.

Effective diffusivity of Nd through interconnected porosity: interconnectivity function

- Fit interconnectivity function based on simulation results

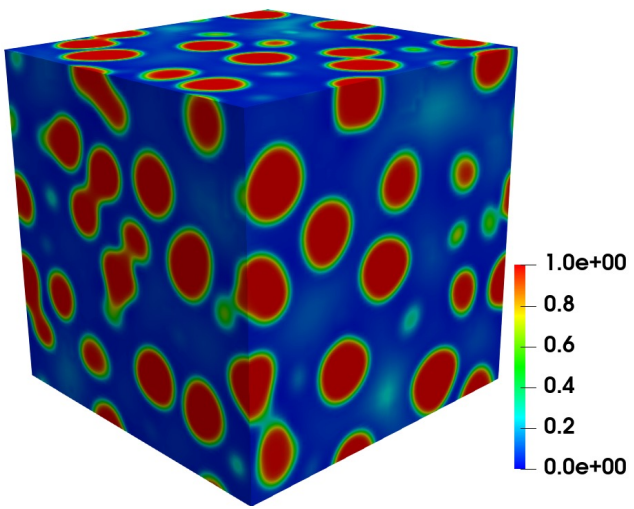


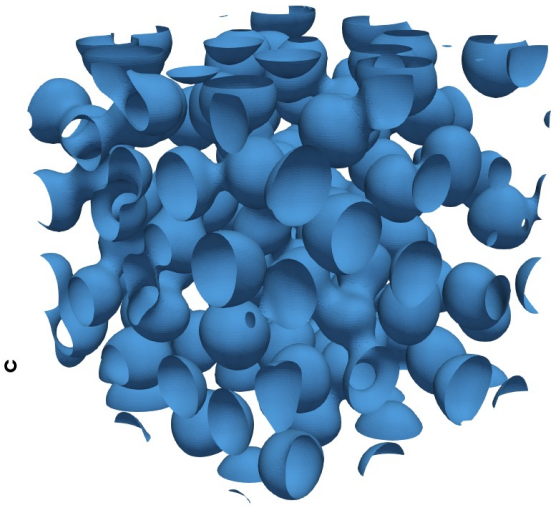


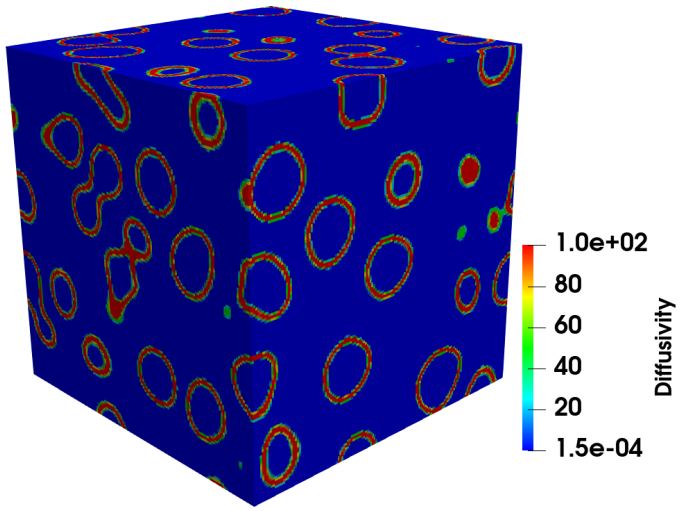
Aagesen et al., Mat. Theory, 6:8, 2022.

Effective diffusivity of Nd through interconnected porosity, no sodium infiltration

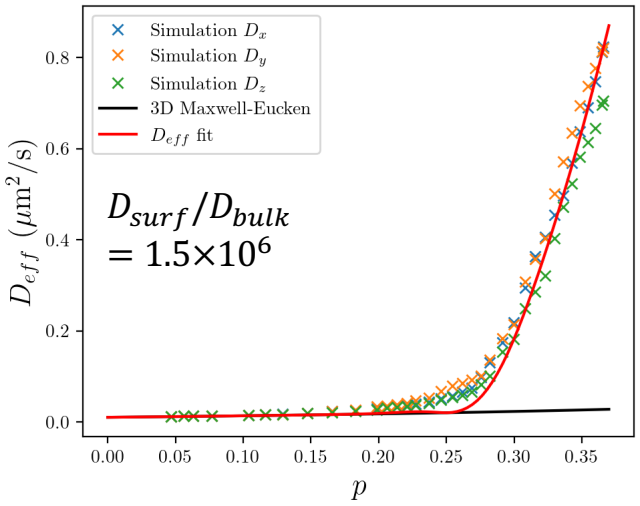
- Use simulations of bubble growth and interconnection
- Assign diffusivity at each position including surfaces
  - Account for interface thickness in phase-field model
- **Calculate porosity, effective diffusivity at each simulation time step using AEH method**

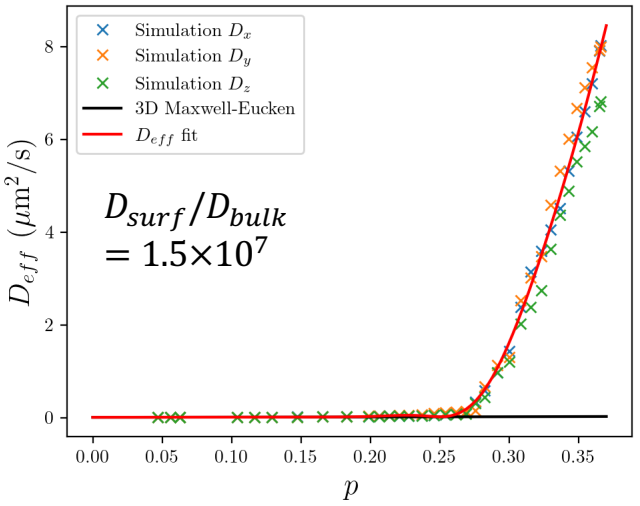


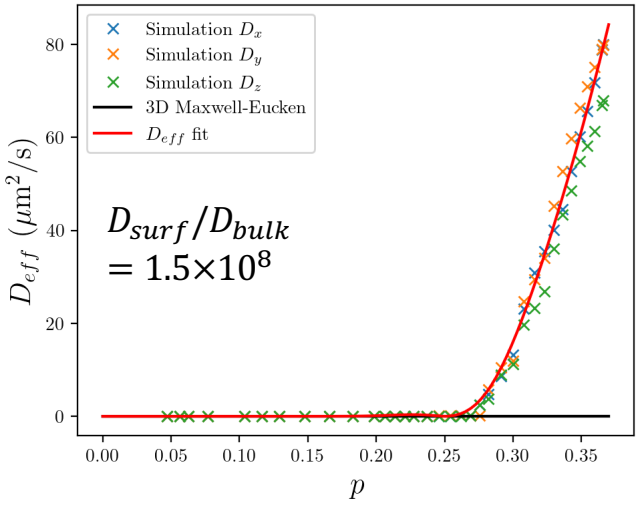


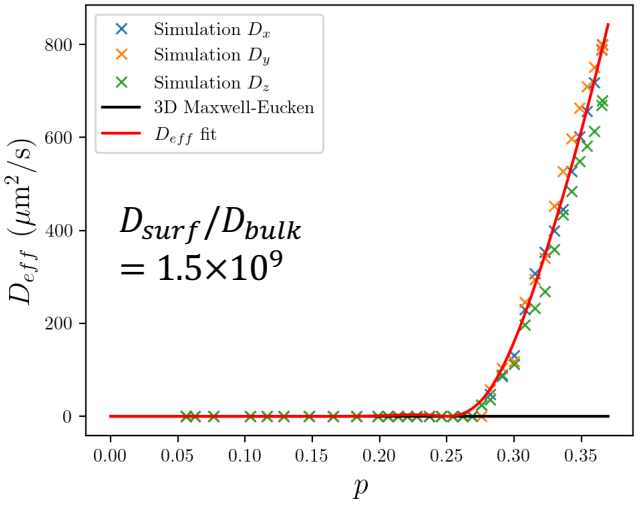


Effective diffusivity of Nd through interconnected porosity, no sodium infiltration: results



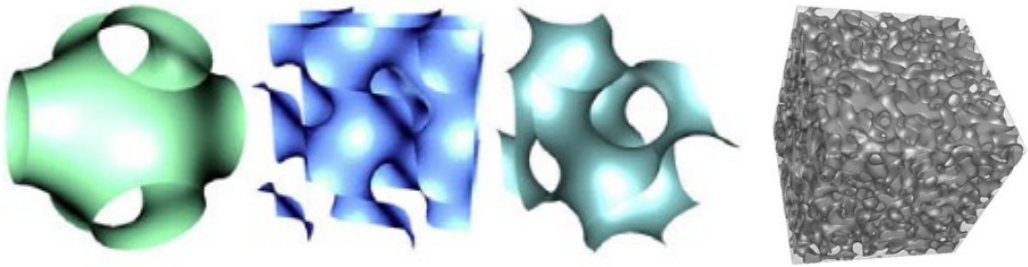






Effective diffusivity of Nd through sodium-logged interconnected porosity

- Interconnectivity occurs when $p > 0.26$
- Sodium-filled porosity: $D_{Na} \gg D_{bulk}$
  - When filled (logged) with sodium, transport through interconnected pores is dominated by diffusivity through sodium
- Fuel with interconnected porosity: bicontinuous structure
  - Past work has shown that when diffusivity through one phase dominates, can use
  - $D_{bi} = \frac{D_{high} p}{\tau}$
  - τ: tortuosity, $\tau \approx 1.5$ for many bicontinuous structures



Bicontinuous structures: Chen et al., Scr. Mater., 61, p. 52-55 (2009)

Effective diffusivity function to capture behavior of three regimes

$$D_{eff} = D_{bulk}\frac{1+2p}{1-p} + D^{p}_{surf} f_V a|p - p_{start}|^{\alpha} + \frac{D^{Na}_{Nd} p}{\tau} f_V$$

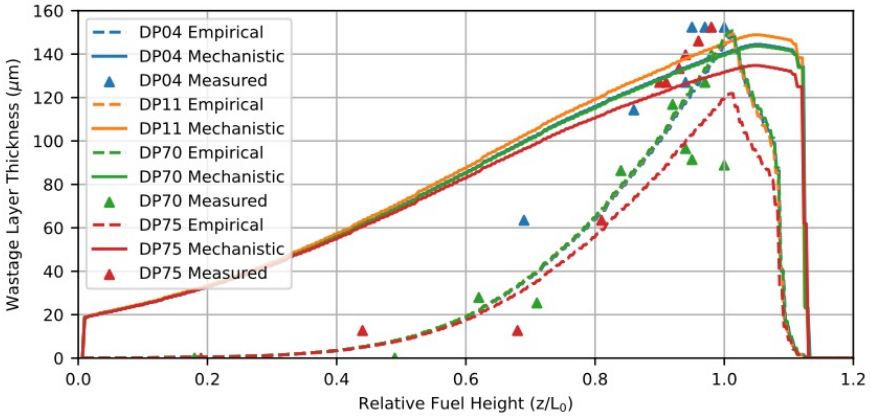
Isolated porosity

Interconnected porosity, no sodium

Interconnected porosity, with sodium

```
[Nd_diffusivity]
    type = DerivativeParsedMaterial
    property_name = D_Nd
    coupled_variables = 'T'
    material_property_names = 'porosity sodium_logged_porosity' # interconnectivity'
    constant_names = '      D0_bulk  Ea_bulk  D0_surf  Ea_surf  D0_Na    Ea_Na  kB
a           p_start alpha   p_cen delta   tortuosity'
    constant_expressions = '4.007e-8 1.4076   5.916e-8 0.079234 7.86e-9  0.0421 8.6173324e-5
2.17695e-3  0.2508  1.71912 0.269 0.0392  1.5'
    expression = 'D_bulk:=D0_bulk * exp(-Ea_bulk / (kB*T));
                  D_surf:=D0_surf * exp(-Ea_surf / (kB*T));
                  D_Na:=D0_Na * exp(-Ea_Na / (kB*T));
                  interconnectivity:=0.5 * (1 + erf((porosity - p_cen) / delta));
                  D_bulk * (1 + 2*porosity) / (1-porosity)
                  + D_surf * a * interconnectivity * (abs(porosity - p_start))^alpha
                  + D_Na * sodium_logged_porosity * interconnectivity * porosity / tortuosity'
```

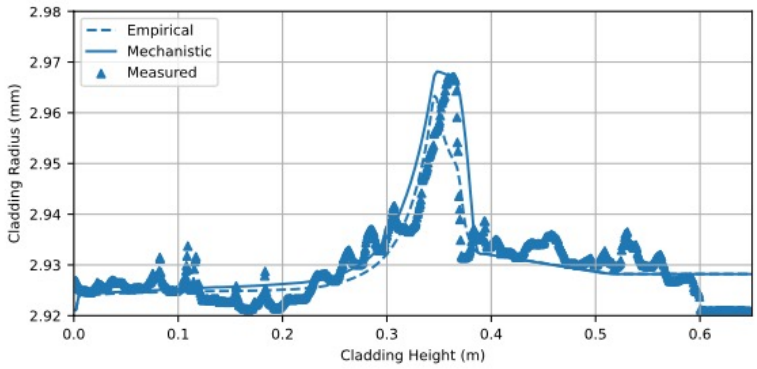
BISON model results: EBR-II X447 Assessment Case



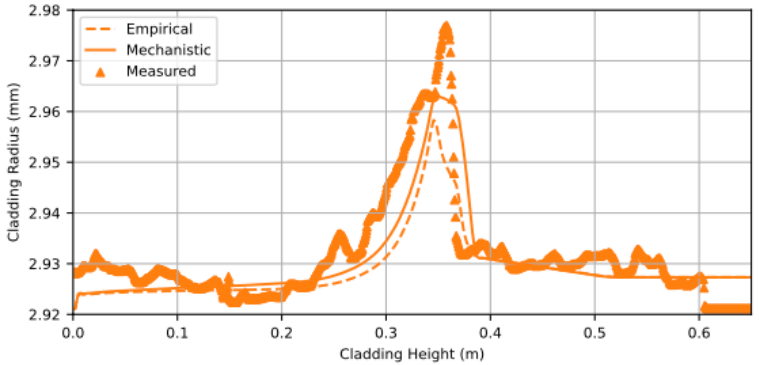
$$\delta = (Kt)^{1/2}$$

$$K = K_0 c e^{-E_a/kT}$$

- Used effective diffusivity, growth rate of $Fe_{17}Nd_2$ expression assuming $\alpha = 1/2$
- Calibrated K_0 using data from pin DP04 at maximum wastage thickness
- Apply to other pins, good agreement for maximum wastage thickness, cladding strain
  - Maximum thickness: fuel design criterion
  - Improvement at lower height: growth exponent



DP04



DP11

Conclusions

- Used atomistic and mesoscale methods together to answer questions for engineering-scale fuel performance modeling
  - Developed mechanistic model of cladding wastage layer growth
- Provides insight into mechanisms that is not available from empirical models
- Model improvements extend capability of BISON to consider broader range of metallic fuel reactor designs, e.g.
  - Annular fuel, without bond sodium
  - Cladding inner liners

Thanks for your attention!

Funding Support: DOE-NE NEAMS Program

Questions?

Idaho National Laboratory