Nuclear Materials Modeling Lab.

KYUNG HEE UNIVERSITY

Effective Thermal Conductivity of UO_2

A Computational approach

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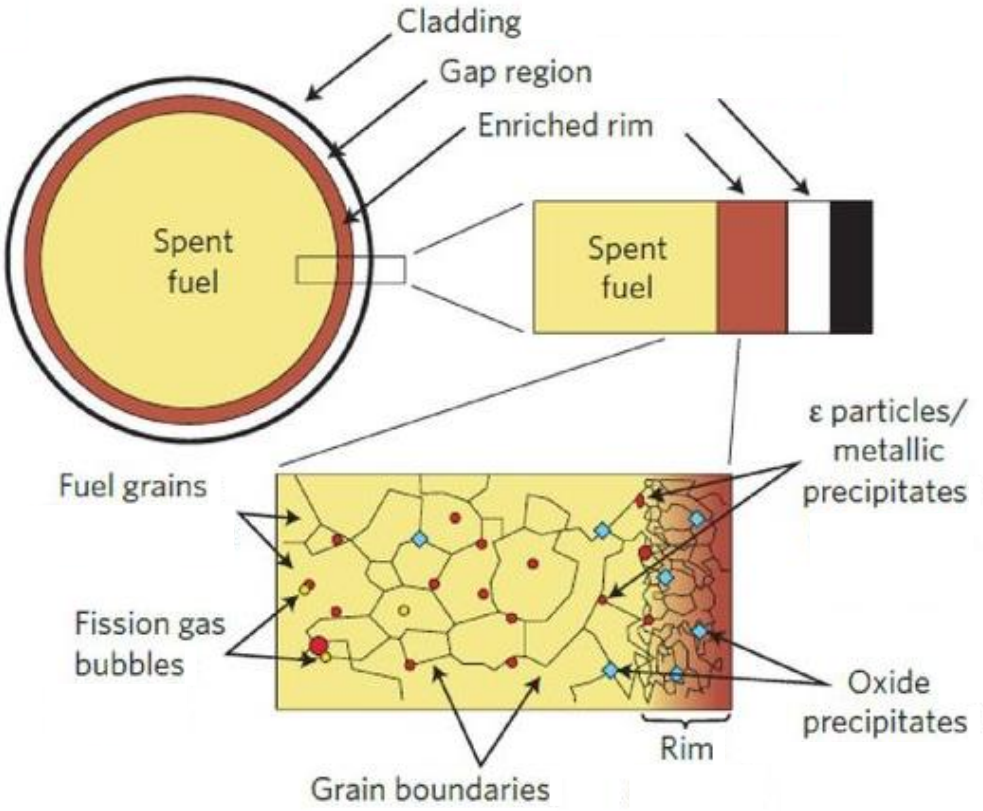
Research Motivation

- Spent fuel microstructure
  - Grain subdivision
  - Defects accumulations
  - Fission product precipitations
  - Fission gas bubbles growth



[1] J. Bruno and R. C. Ewing (2006)

- Thermal conductivity degradation of UO_2 fuel with increasing burn-up, caused by the accumulation of defects in the crystalline lattice.

Effective Thermal Conductivity of Porous Media

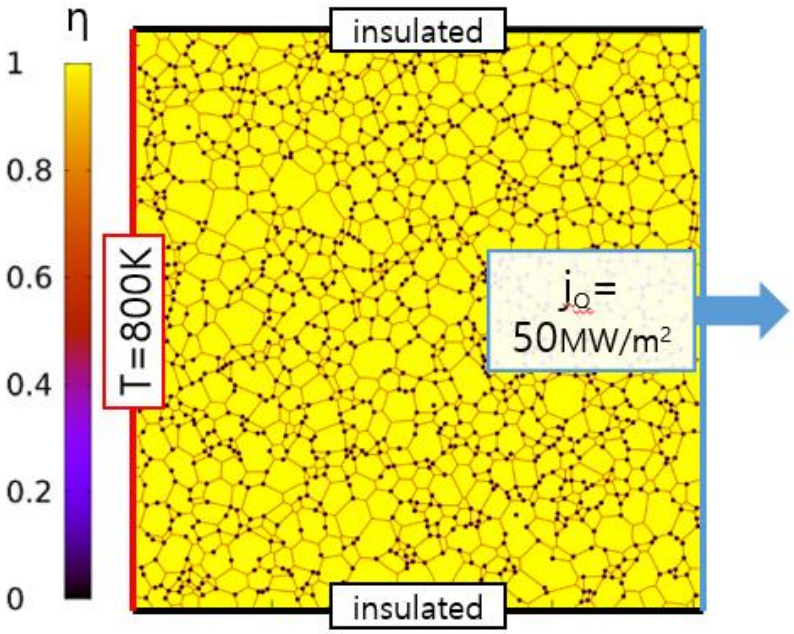
- Thermal conductivity models for UO_2 in FRAPCON
  - Based on extensive databases that have been accumulated over decades.
  - Cannot provide property values for the systems outside its range of experience.
  - Needs for developing more generally applicable models based on physical understanding of the thermophysical behavior of system microstructures.

- Models and simulation techniques
  - **Atomistic/particulate models** : The acoustic mismatch model, diffuse mismatch model, and molecular dynamics (MD) simulations preserve the underlying particulate nature of heat carriers.
  - **Continuum models** : The continuum models and finite-element simulations utilize specific constitutive laws and interfacial boundary conditions that coarse the grain atomistic details.

Simulation Geometry

- UO_2 fuel containing defects
  - Helium bubbles (in pores)
  - Grain boundaries
  - Microstructure is defined by structural order parameter, η.

$$\begin{cases} UO_2\ \text{Crystal}: \eta = 1 \\ \text{Helium Bubble}: \eta = 0 \\ \text{Grain Boundary}: 0 < \eta < 1 \end{cases}$$



- Local conductivity depends on the parameter, η.

$$\begin{cases} k_{crystal} = \dfrac{1}{0.0375 + 0.0002165T} + \dfrac{4.715 \times 10^9}{T^2} \exp\left(-\dfrac{16361}{T}\right) \\ k_{local} = (k_{crystal} - 0.152) \times \eta^{7.7} + 0.152 \quad \text{(units: W/K/m)} \end{cases}$$

[2] J. Harding and D. G. Martin (1989)

[3] P. C. Millett et al. (2008)

Kapitza conductance ~10MW/m_2
(GB의 thermal resistance값 결정)

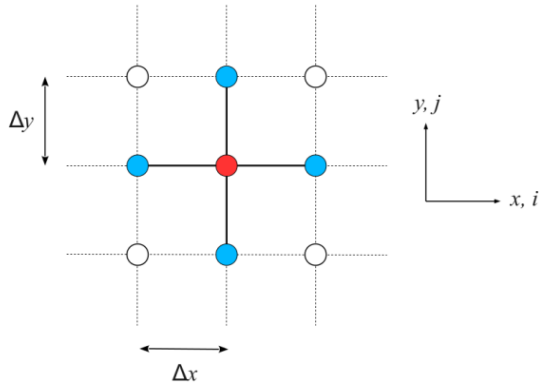
Effective Thermal Conductivity Simulation

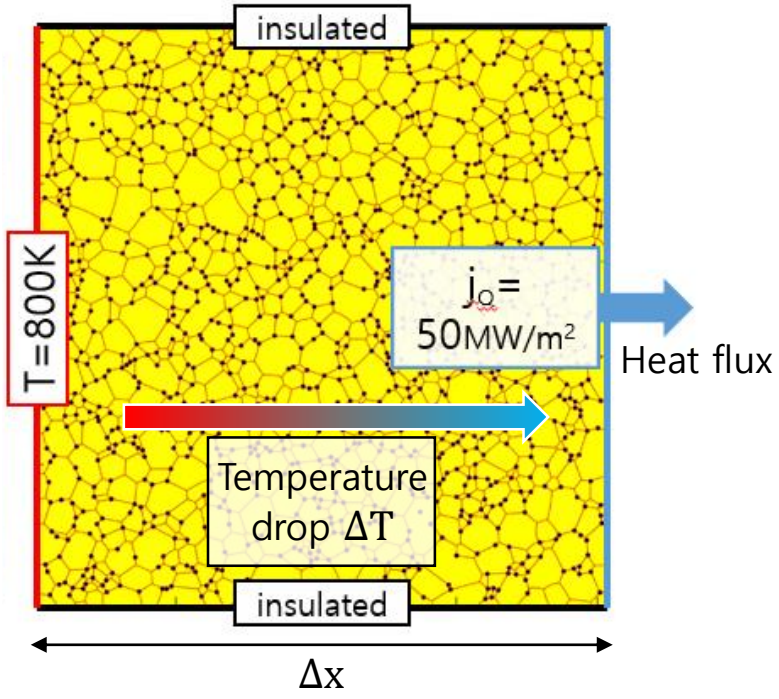
- Steady-state heat equation

$$\nabla \cdot (k(r)\nabla T) = 0$$

→ Steady-state temperature distribution

- Finite difference approximations





- Effective conductivity relation (rearranged Fourier's law)

$$k_{eff} = \frac{j_Q \times \Delta x}{\Delta T}$$

Numerical Method

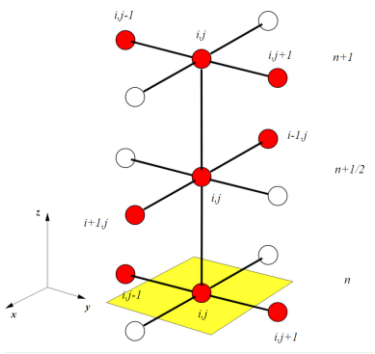
- Crank-Nicolson Scheme (CN)

$$\frac{T_{i,j}^{n+1} - T_{i,j}^{n}}{\Delta t} = \frac{1}{2(\Delta x)^2}(\delta_x^2 + \delta_y^2)(T_{i,j}^{n+1} - T_{i,j}^{n})$$

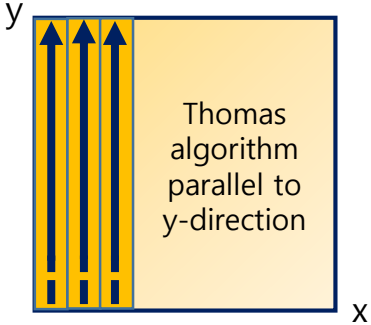
  - Finite difference method
  - Second-order accurate in both time and space
  - Numerically stable comparing to simple Euler method

- Alternating Direction Implicit Method (ADI)

$$\begin{cases} \dfrac{T_{i,j}^{n+1/2} - T_{i,j}^{n}}{\Delta t/2} = \dfrac{(\delta_x^2 T_{i,j}^{n+1/2} - \delta_y^2 T_{i,j}^{n})}{\Delta x^2} \\ \dfrac{T_{i,j}^{n+1} - T_{i,j}^{n+1/2}}{\Delta t/2} = \dfrac{(\delta_x^2 T_{i,j}^{n+1} - \delta_y^2 T_{i,j}^{n+1/2})}{\Delta y^2} \end{cases}$$



CUDA-accelerated parallel computing

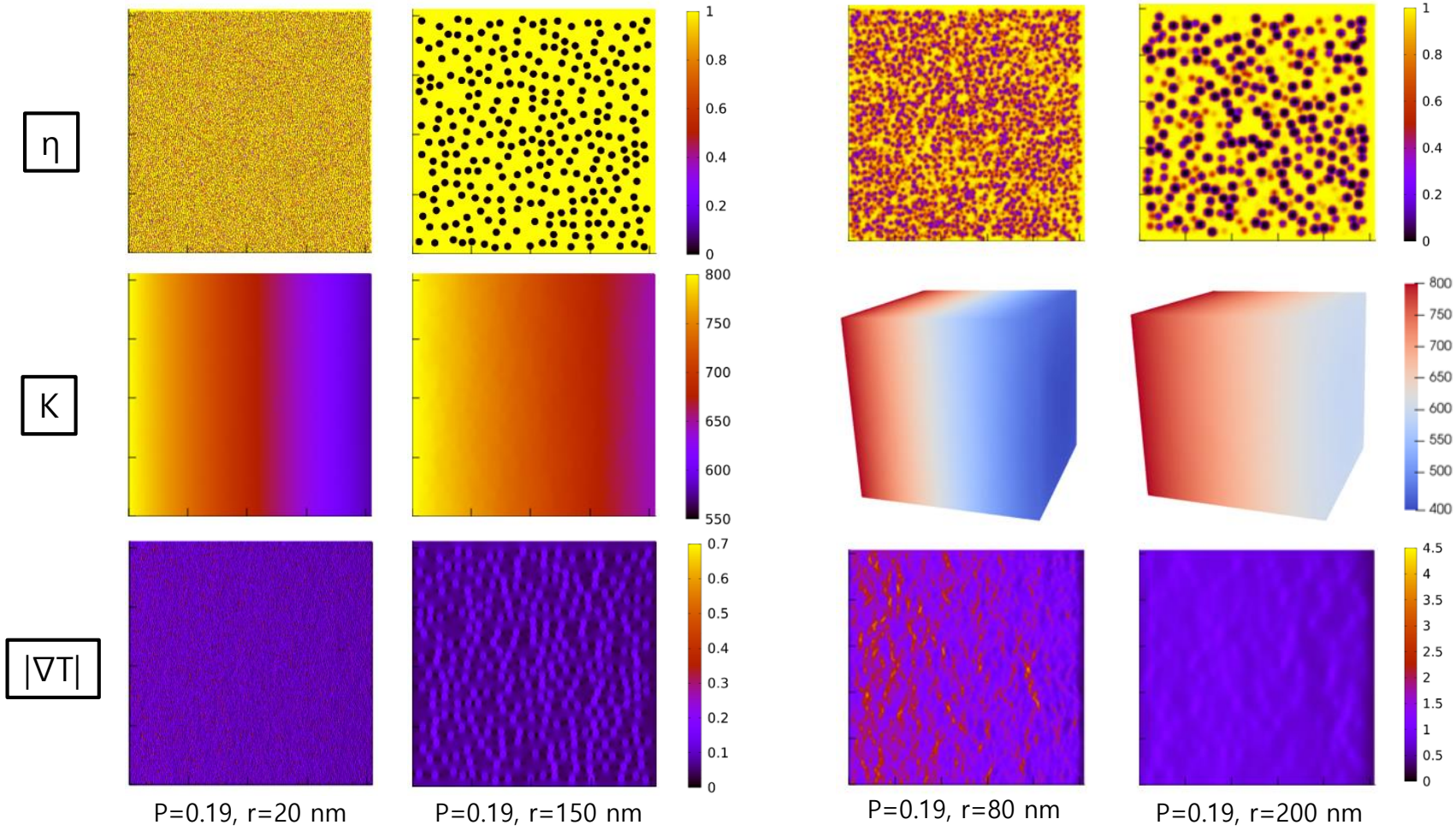


10 times faster than serial code

Simulational Result (without grain boundary)

[4] B. Yoon and K. Chang (2020)

- 2D Simulation
- 3D Simulation



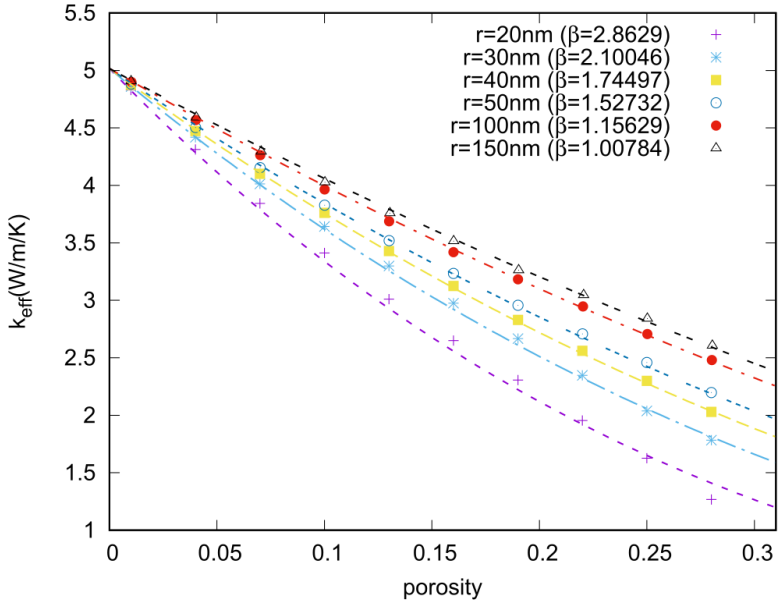
Pore Size Effect

- Schulz Model [5] B. Schulz (1981)

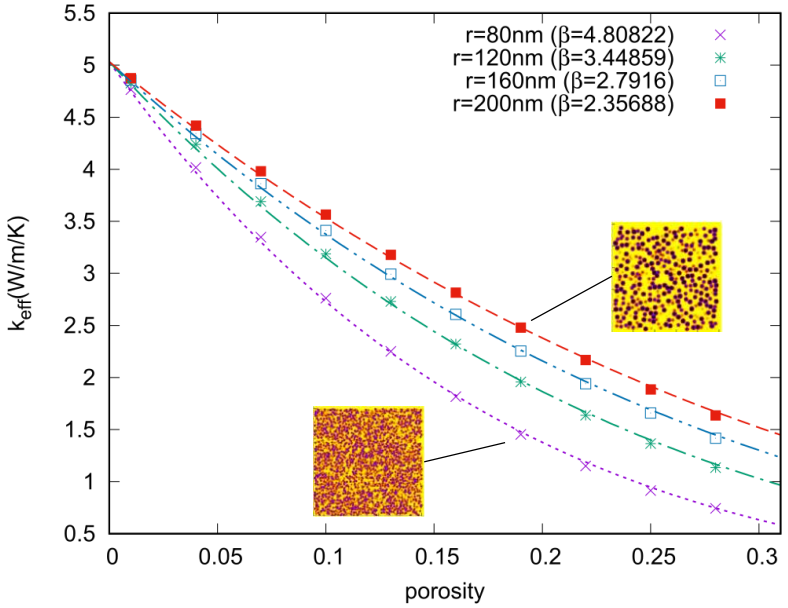
$$k_{eff} = k_0(1-P)^{\beta}$$

- P : porosity
- k_0 : conductivity of the corresponding nonporous material

[4] B. Yoon and K. Chang (2020)



2D simulation

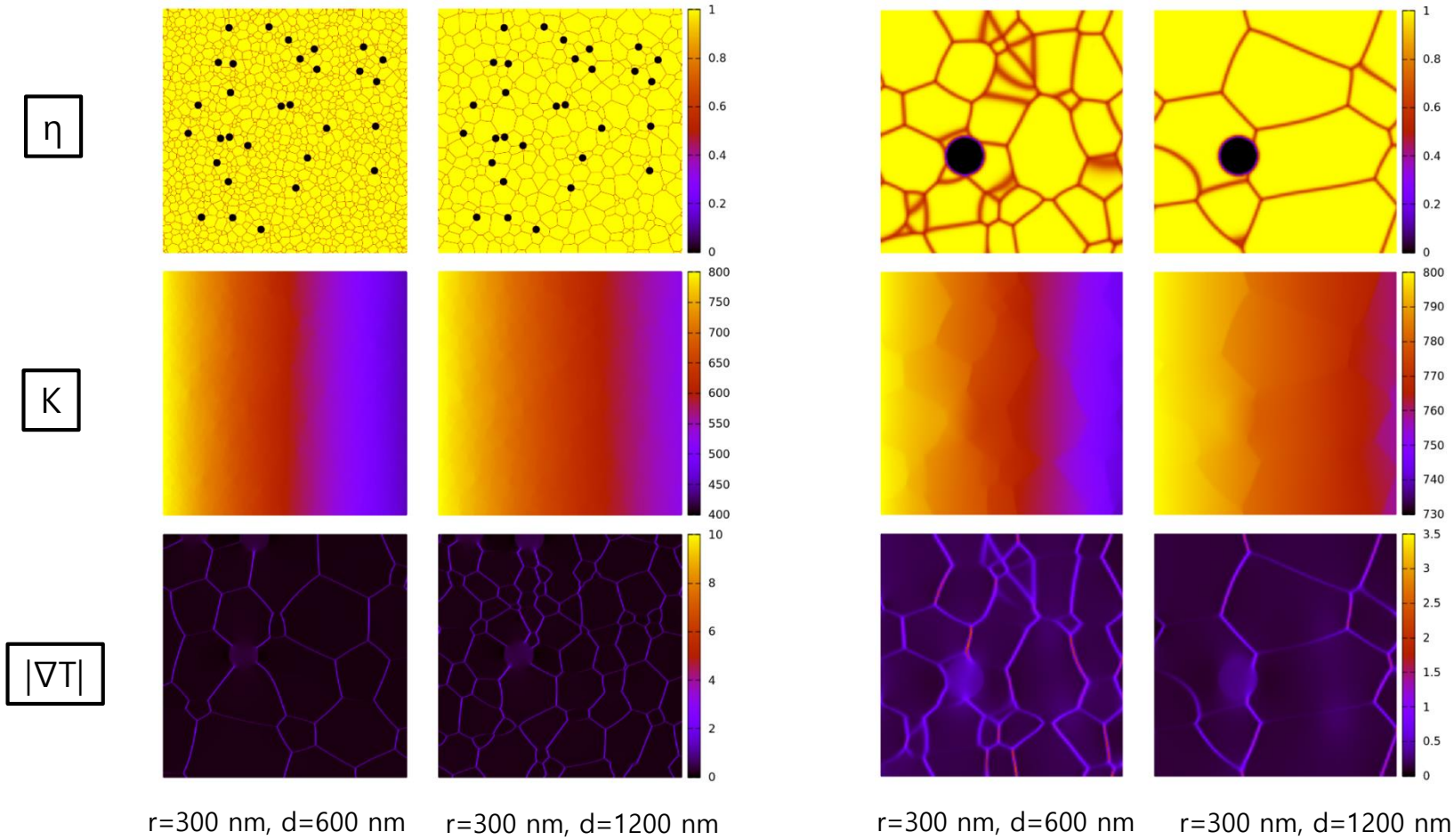


3D simulation

Simulational Result (with grain boundaries)

[6] B. Yoon and K. Chang (2021)

- 2D Simulation (20.48^2 um^2)
- 3D Simulation (2.56^3 um^3)

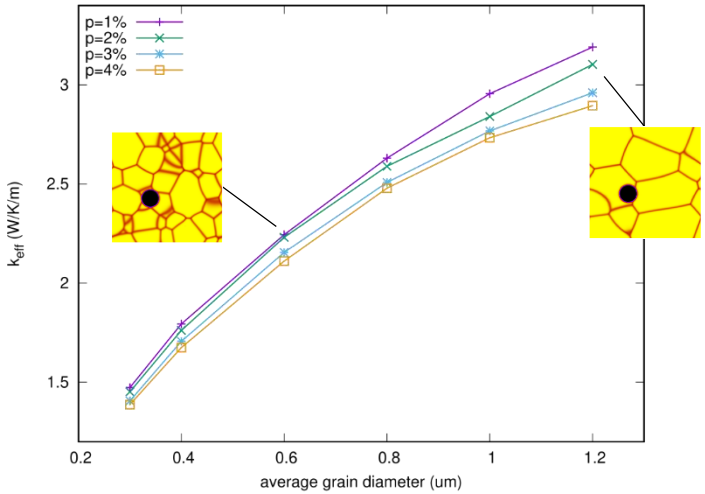


r=300 nm, d=600 nm r=300 nm, d=1200 nm r=300 nm, d=600 nm r=300 nm, d=1200 nm

Effective Thermal Conductivity of Porous Polycrystalline UO_2

[6] B. Yoon and K. Chang (2021)

- As expected, the effective conductivity decreases with increasing porosity and decreasing grain diameter.



- We found that under the given porosity and grain size, thermal conductivity decreases as the pore radius decreases.

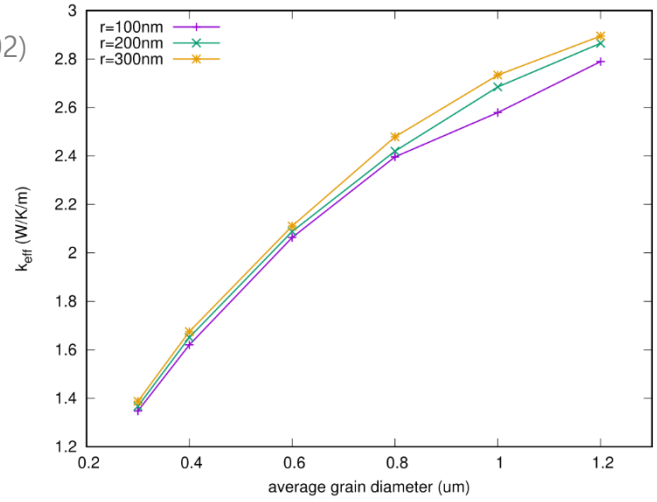
[7] H.-S. Yang et al. (2002)

- Schulz Model + Yang's polycrystal model

(Multiplier model)

$$k_{eff} = k_0 \frac{(1-P)^{\beta}}{1 + k_0/(G_k d)}$$

  - P : porosity
  - d : average grain diameter
  - k_0 : conductivity of corresponding nonporous material
  - G_k : kapitza conductance



Effective Thermal Conductivity Model of Porous Polycrystal

- Multiplier Model

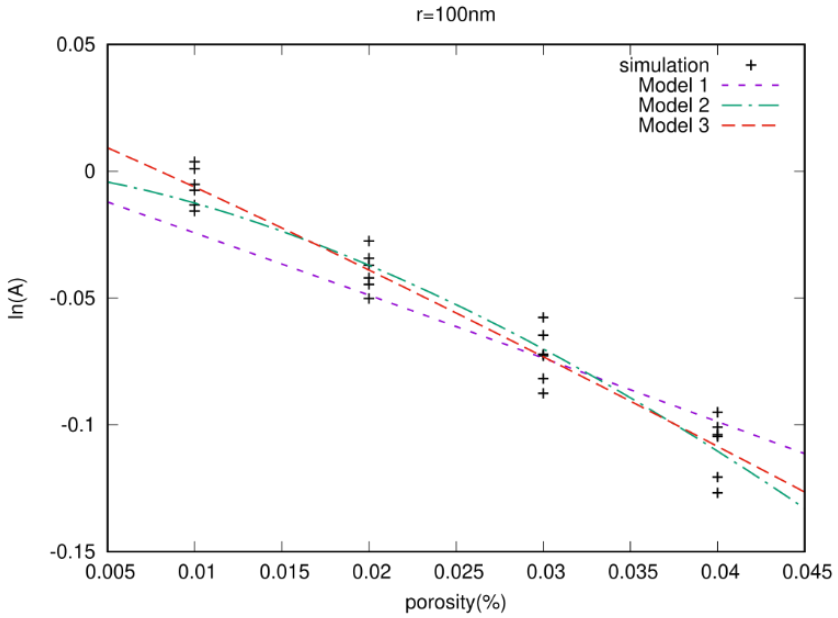
$$k_{eff} = k_0 \frac{(1-P)^{\beta}}{1 + k_0/(G_k d)}$$

The value of exponent β in multiplier model		
	2D	3D
r=100nm	2.46884	2.41845
r=200nm	1.20273	1.88383
r=300nm	0.77723	1.66590

- The exponent β is 1.5 in Nikolopoulos and Ondracek's analysis of experimental data for spherical pores. [8] P. Nikolopoulos and G. Ondracek (1983)
- We found that the pore size affects the exponent β in both single crystal and polycrystal models.

Modified Effective Thermal Conductivity Models

The effective thermal conductivity models of UO_2	
Model 1	$\ln\left[\frac{k_{eff}\times(1+k_0/(G_k d))}{k_0}\right]=(\beta+1)\times\ln(1-p)$
Model 2	$\ln\left[\frac{k_{eff}\times(1+k_0/(G_k d))}{k_0}\right]=(\beta+1)\times\ln(1-p)^n$
Model 3	$\ln\left[\frac{k_{eff}\times(1+k_0/(G_k d))}{k_0}\right]=(\beta+1)\times\ln(1-p)^n+\alpha(1-p)$



Root Mean Square Deviations of Three Models

- Root mean square deviation

$$RMSD = \sqrt{\frac{1}{N}\sum^{N}|k_{simulation} - k_{model}|^2}$$

  - $k_{simulation}$: effective conductivity value obtained from simulation
  - k_{model} : effective conductivity value predicted by model 1, 2 and 3
  - N : number of effective conductivity values being compared

Root mean square deviations of 2D system

Pore radius	Model 1	Model 2	Model 3
100nm	0.0560	0.0381	0.0339
200nm	0.0471	0.0349	0.0331
300nm	0.0598	0.0509	0.0507

Root mean square deviations of 3D system

Pore radius	Model 1	Model 2	Model 3
100nm	0.0341	0.0235	0.0217
200nm	0.0355	0.0179	0.0157
300nm	0.0334	0.0249	0.0209

Conclusion

- In the mesoscale regime, we have computationally investigated the effect of porosity, pore size, and grain boundaries on the effective conductivity of UO_2 by means of Crank-Nicolson and ADI method with implementation of CUDA GPU parallelization in 2D and 3D systems.

- We found that under the given porosity and grain size, thermal conductivity decreases as the pore radius decreases.

- We proposed empirical models for effective conductivity of porous polycrystal UO_2 based on microstructure generated by phase-field method in the presence of pores and grain boundaries.

References

[1] J. Bruno and R. C. Ewing, "Spent Nuclear Fuel," *Elements*, vol. 2, pp. 343-349, 2006.

[2] J. Harding and D. G. Martin, "A recommendation for the thermal conductivity of UO2," *Journal of Nuclear Materials*, vol. 166, no. 3, pp. 223-226, 1989.

[3] P. C. Millett et al., "Phase-field simulation of thermal conductivity in porous polycrystalline microstructures," *Journal of Applied Physics*, vol. 104, no. 3, p. 033512, 2008.

[4] B. Yoon and K. Chang, "Effect of the pore radius on the effective conductivity of UO2 in 2D and 3D: A computational approach," *Results in Physics*, vol. 19, no. 2-3, p. 103440, 2020.

[5] B. Schulz, "Thermal conductivity of porous and highly porous materials," *High Temperatures – High Pressures*, vol. 13, no. 6, pp. 649-660, 1981.

[6] B. Yoon and K. Chang, "Effective Thermal Conductivity Model of Porous Polycrystalline UO2: A computational Approach," *In Press*, 2021.

[7] H.-S. Yang et al. "Interfacial Thermal Resistance in Nanocrystalline Yttria-Stabilized Zirconia," *Acta Materialia*, vol. 50, no. 9, pp. 2309-2317, 2002.

[8] P. Nikolopoulos and G. Ondracek, *Journal of Nuclear Materials*, vol. 114, no. 2-3, p. 231-233, 1983.

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