Quantification of Uncertainty in Materials Science
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# Certainty and Uncertainty at Multiple Scales


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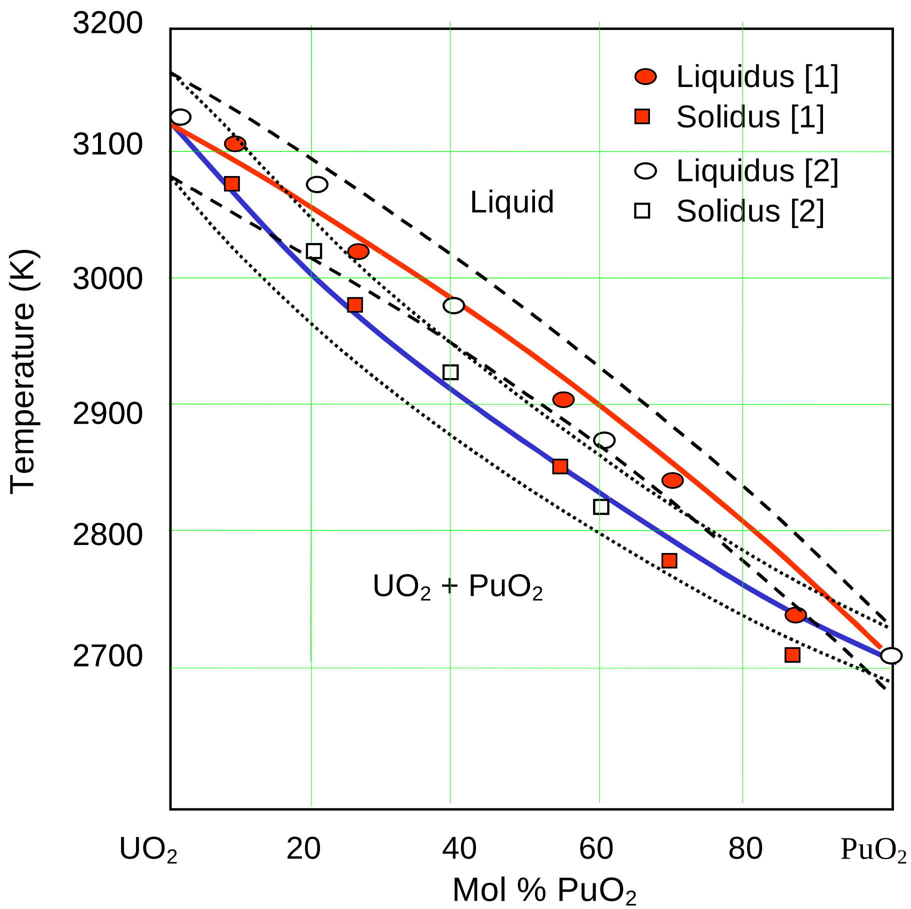
# Uncertainty of Nuclear Fuels Data

- Uncertainty in fuel thermo-mechanical properties is often >10%
- Uncertainty of chemical properties (free energy) can be 10-15 %

Example:

- Uncertainty quantification the $UO_2$-$PuO_2$ phase diagram*. ΔT = 50K, Δc = 3%
- Bayesian analysis of 15 data sets (melting temperatures , transformation enthalpies, …).
- Optimization via a genetic algorithm.





* M. Stan and B. J. Reardon, *CALPHAD*, **27** (2003) 319-323.

[1] M. G. Adamson, E. A. Aitken, and R. W. Caputi, J. Nucl. Mater., **130** (1985) 349-365.

[2] T. D. Chikalla, J. Am. Ceram. Soc., **47** (1964) 309-309.

# Pu-Ga phase diagram from DFT and MD

Electronic Structure

Molecular Dynamics

Free energy of all phases

Chemical potentials

$$\mu^{\delta}_{Pu}(T,P,x) = G^{\delta}(T,P,x) - x\left[\frac{\partial G^{\delta}(T,P,x)}{\partial x}\right]_{T,P}$$

$$\mu^{\delta}_{Ga}(T,P,x) = G^{\delta}(T,P,x) + (1-x)\left[\frac{\partial G^{\delta}(T,P,x)}{\partial x}\right]_{T,P}$$

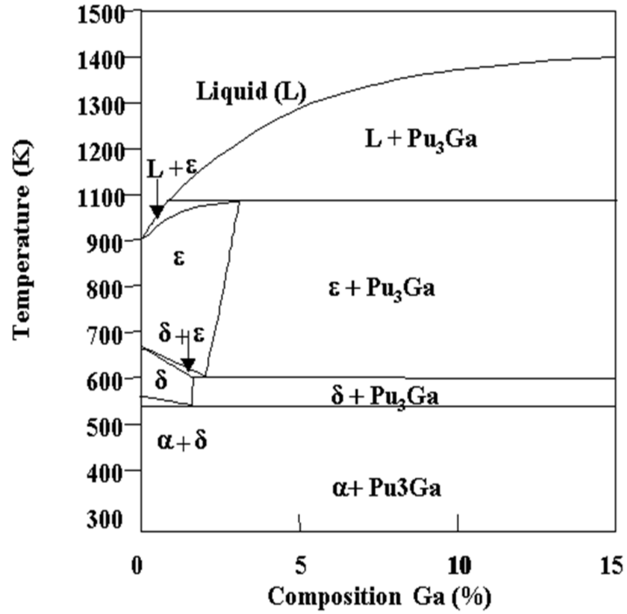
Thermodynamic equilibrium

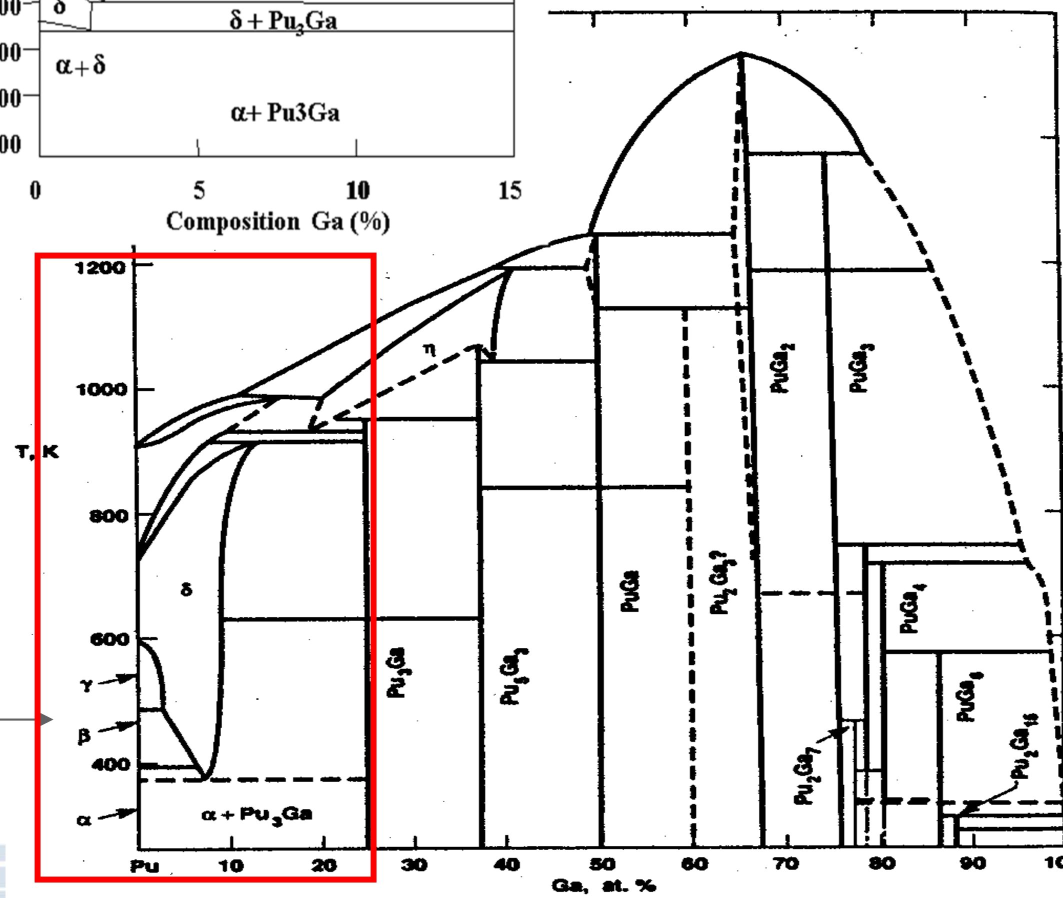
$$\mu^{\delta}_{Pu}(T,P,x^{\delta}) = \mu^{\alpha}_{Pu}(T,P,x^{\alpha})$$

$$\mu^{\delta}_{Ga}(T,P,x^{\delta}) = \mu^{\alpha}_{Ga}(T,P,x^{\alpha})$$

New phase diagram

Minimal input from the binary[1]




[1]M. I. Baskes, K. Muralidharan, M. Stan, S. M. Valone, and F. J. Cherne, JOM, **55** (2003) 41-50.

# Major sources of uncertainty - Nuclear Energy[1]

- Models of material properties are oversimplified. Often ranges of model validity are not specified.
- Extensive  use of empirical correlations. These are needed ‘to close’ the balance equations and are also reported as ‘constitutive equations’ or ‘closure relationships’.
- Imperfect knowledge of boundary conditions and initial conditions.
- Approximate equations are solved by approximate numerical methods.
- Software errors.
- Computer/compiler errors.
- The 2nd principle of thermodynamics is not necessarily fulfilled.
- Different groups of users having the same code and the same information for modeling a Nuclear Power Plant do not achieve the same results.
- …

[1]IAEA Report (authors: Allison C., Balabanov E., D’Auria F., Jankowski M., Misak J., Salvatores S., Snell V.) “Accident Analysis for Nuclear Power Plants” IAEA Safety Reports Series No 23, pp 1-121, ISSN 1020-6450; ISBN 92-0-115602-2, Vienna (A), 2002.

# Goal: Understand, predict, and control thermal conductivity of uranium dioxide ($UO_2$)

Thermal conductivity of UO2 decreases with

- temperature
- burnup

Empirical model [1]

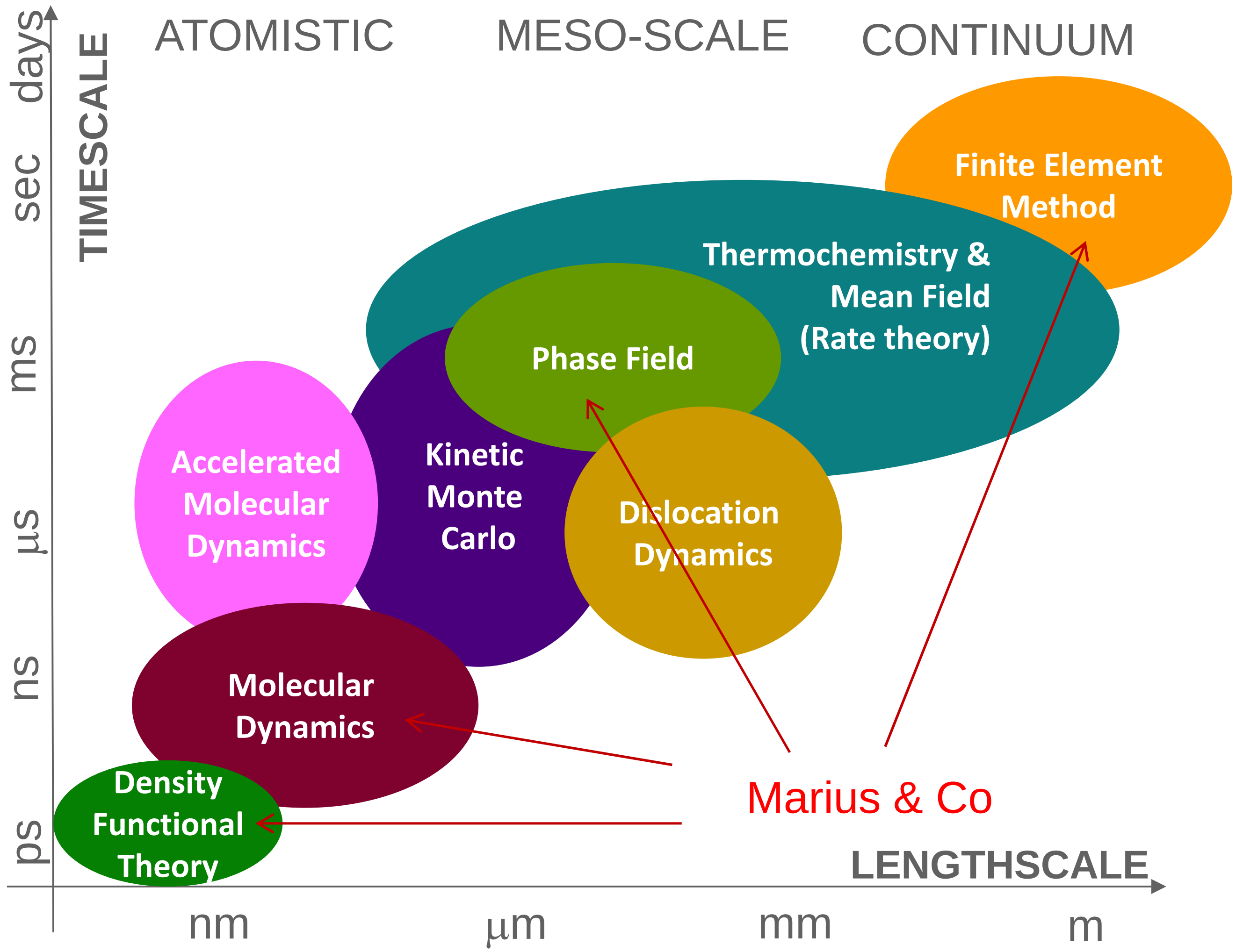
$$k(b) = \frac{1}{1+e^{\frac{20-b}{6}}} - 0.015267$$

Target model:

$$k(T, x, p, b, microstructure, time)$$

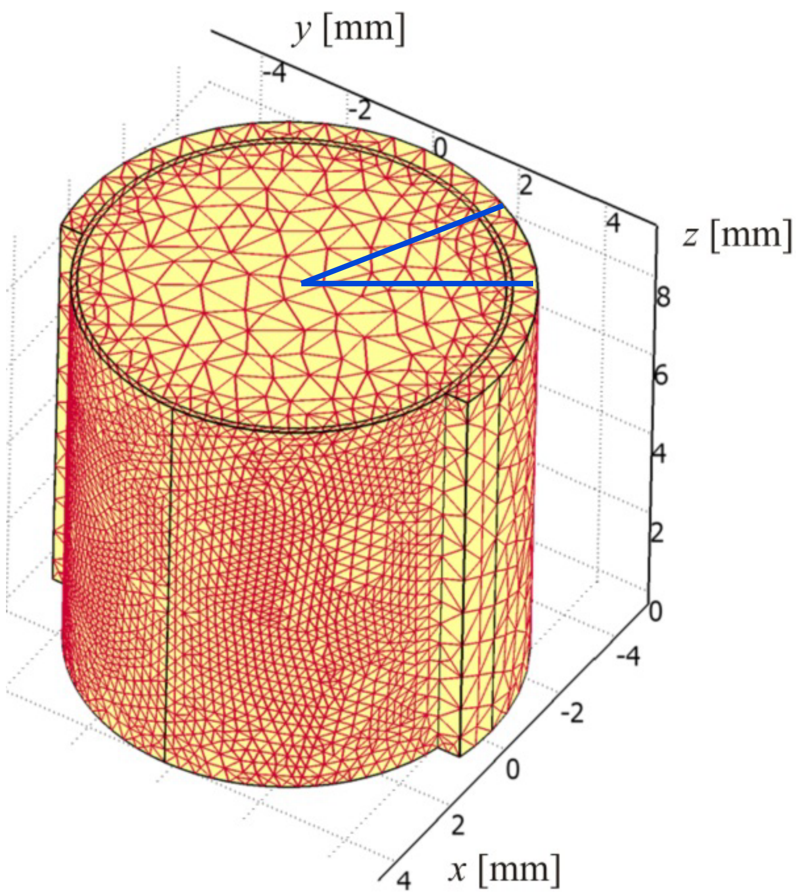
[1] C. Ronchi, et al., J. Nucl. Mater. 327 (2004) 58.

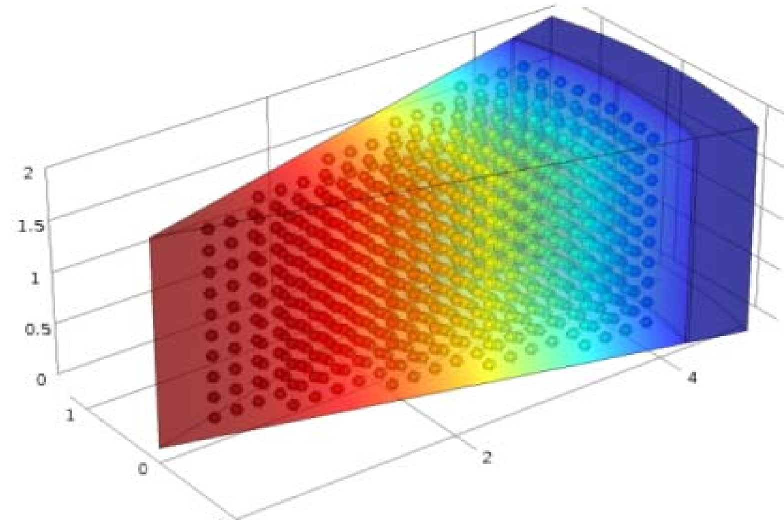
# Multi-scale theoretical and computational methods

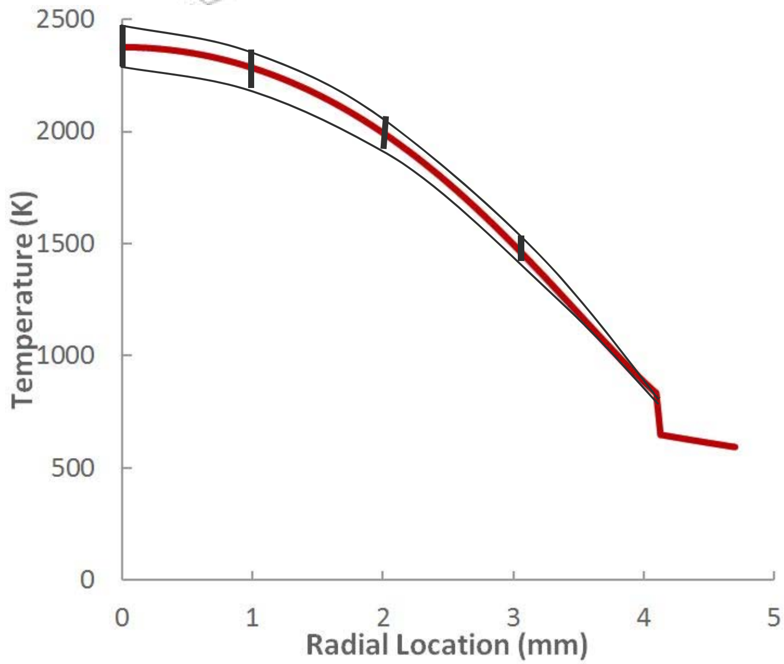



M. Stan, Materials Today, 12 (2009) 20.

# FEM simulations of porosity effects on thermal transport in $UO_2$ fuels



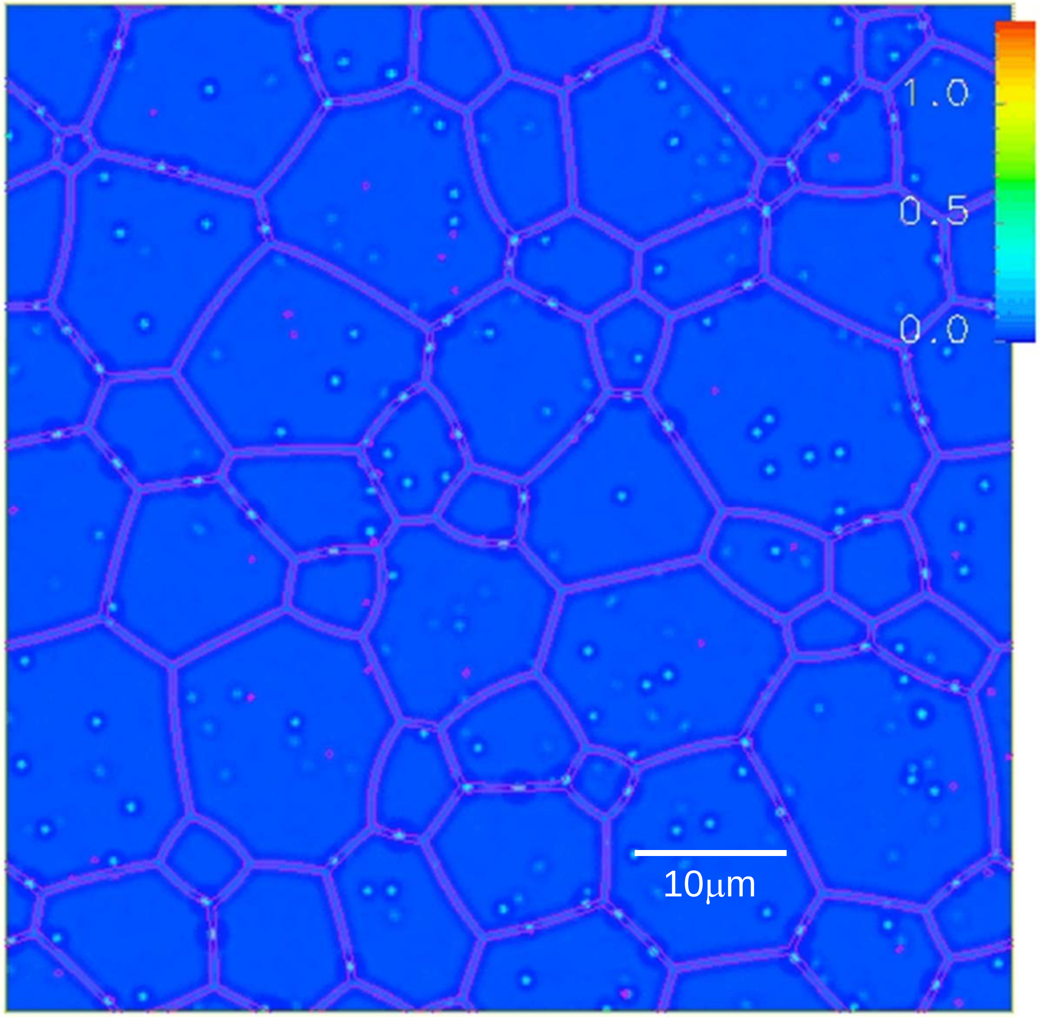



Coupled heat and chemical transport with thermal expansion [1]

Effective thermal conductivity [2] $k_{porous}^{eff}(T, x, p)$

[1] B. Mihaila et al., J. Nucl. Mater. 430 (2012) 221.

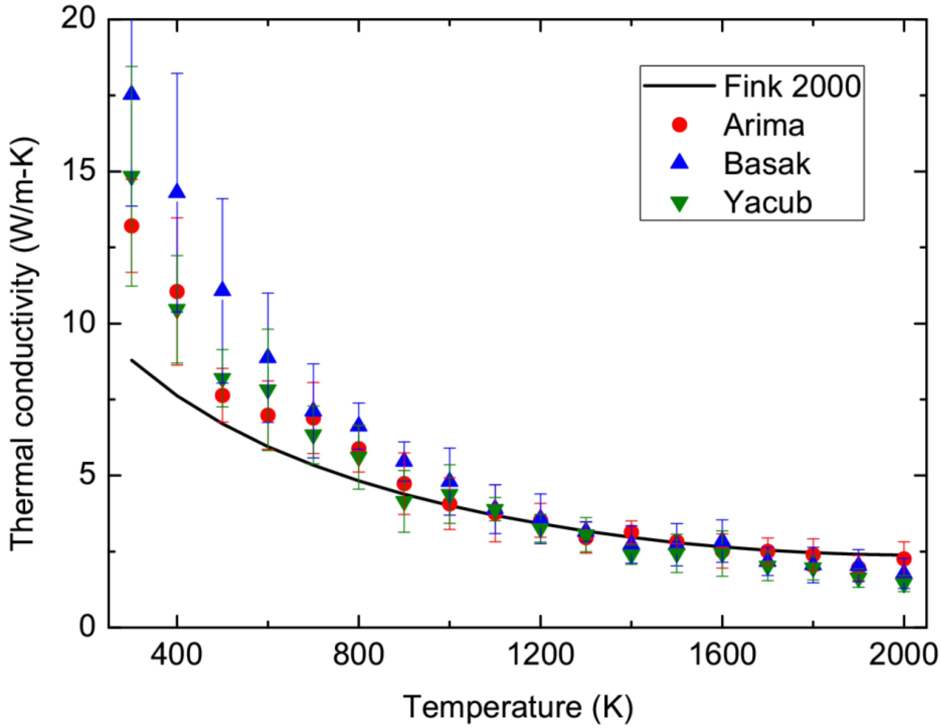
# Microstructure of $UO_2$ - Phase Field vs Experiment



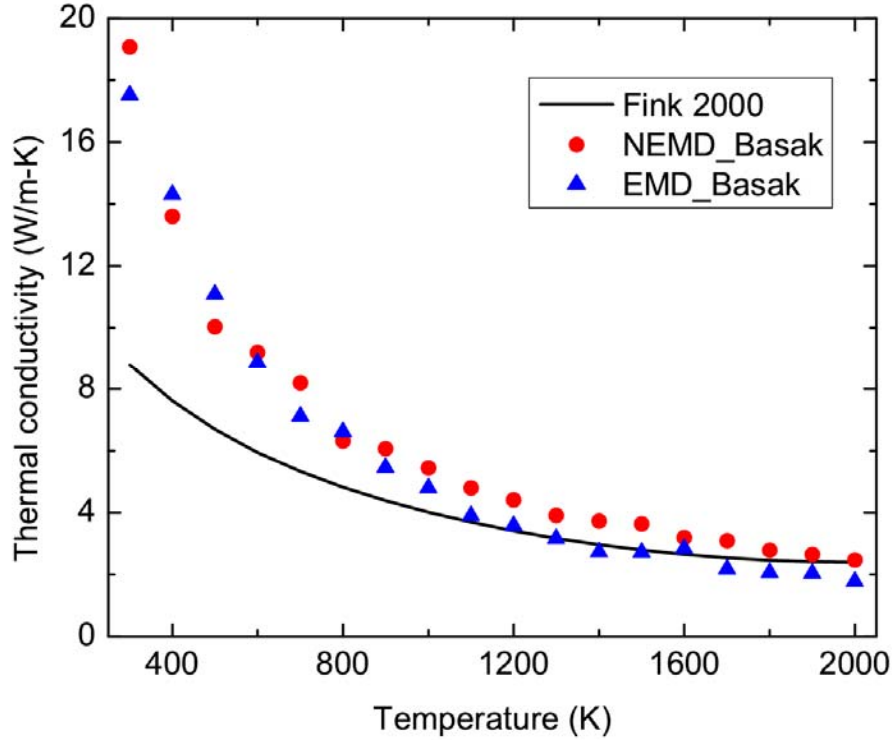

Simulation of gas bubbles evolution in polycrystalline $UO_2$ fuel[1-3].
Color scheme of FP concentration: red = high, blue = low.

[1]M. Stan, J. Nucl. Eng. Technology, **41** (2009) 39-52.
[2]S.Y. Hu et al., J. Nucl. Mater. **392** (2009) 292–300.
[3]I. Zacharie *et. al.,* J. Nucl. Mater. **255** (1998), 92-104.

# Thermal Conductivity of $UO_2$ by Molecular Dynamics




Thermal conductivity of $UO_2$ calculated by EMD with various potentials. Good agreement with experiment above 1000K.

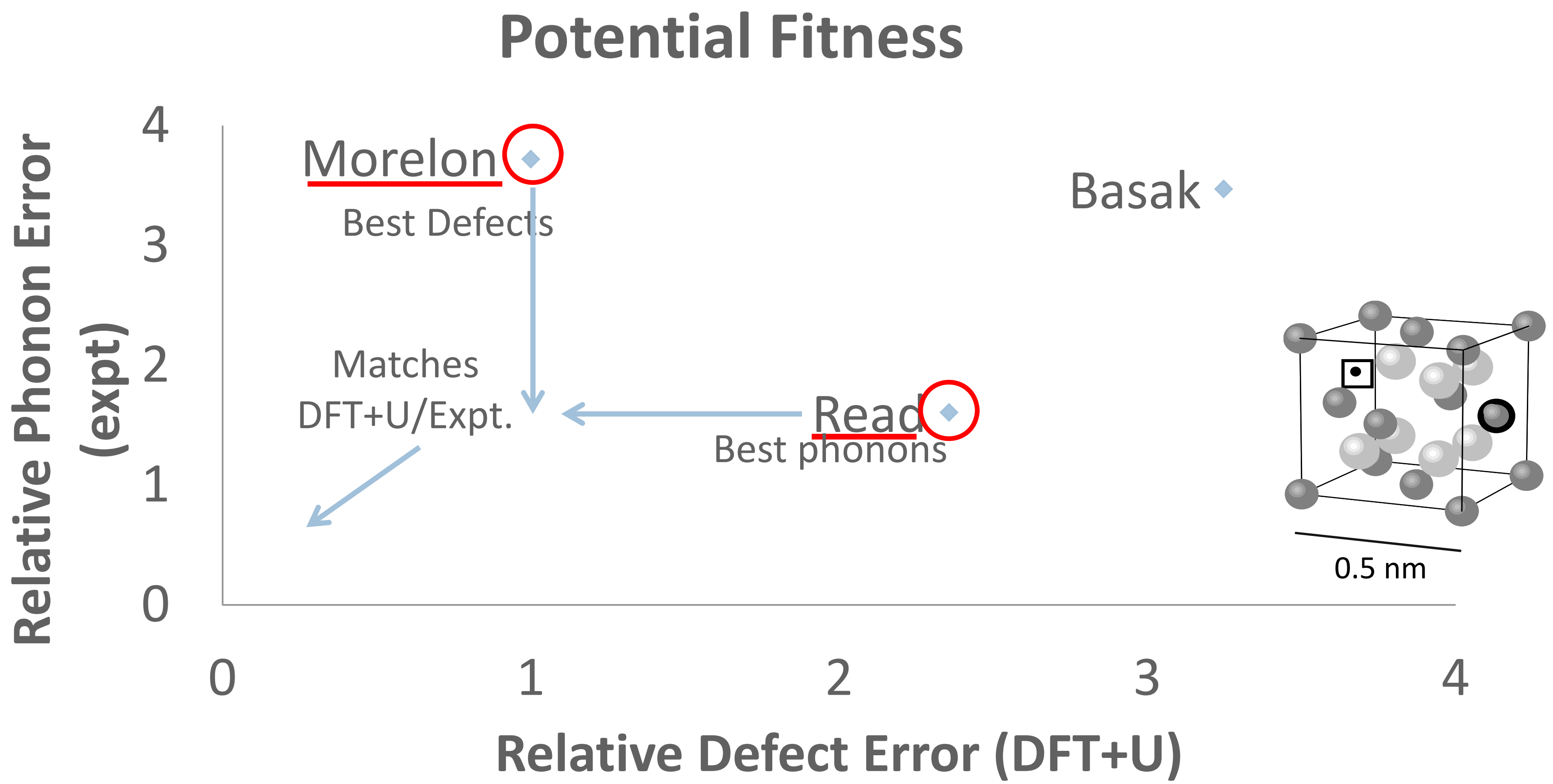



Comparison of thermal conductivity calculated by EMD and NEMD methods using the Basak potential.



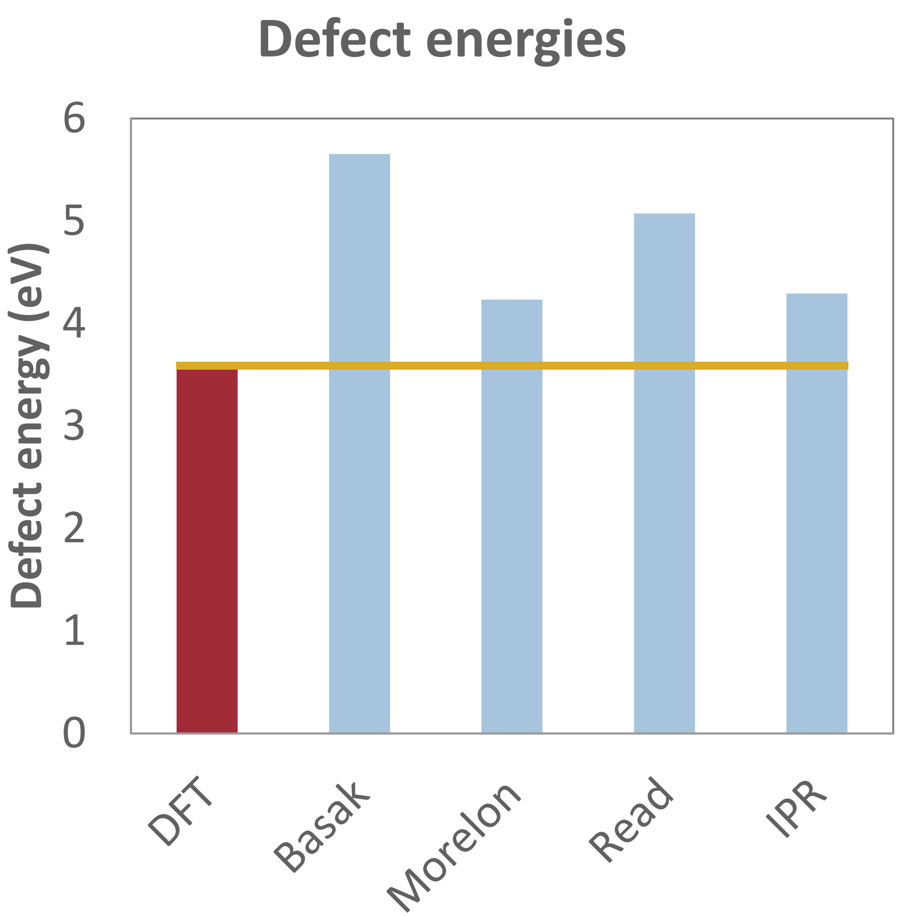
Z. G. Mei, M. Stan, and J. Yang, J. Alloys Comp. 603, (2014) 282

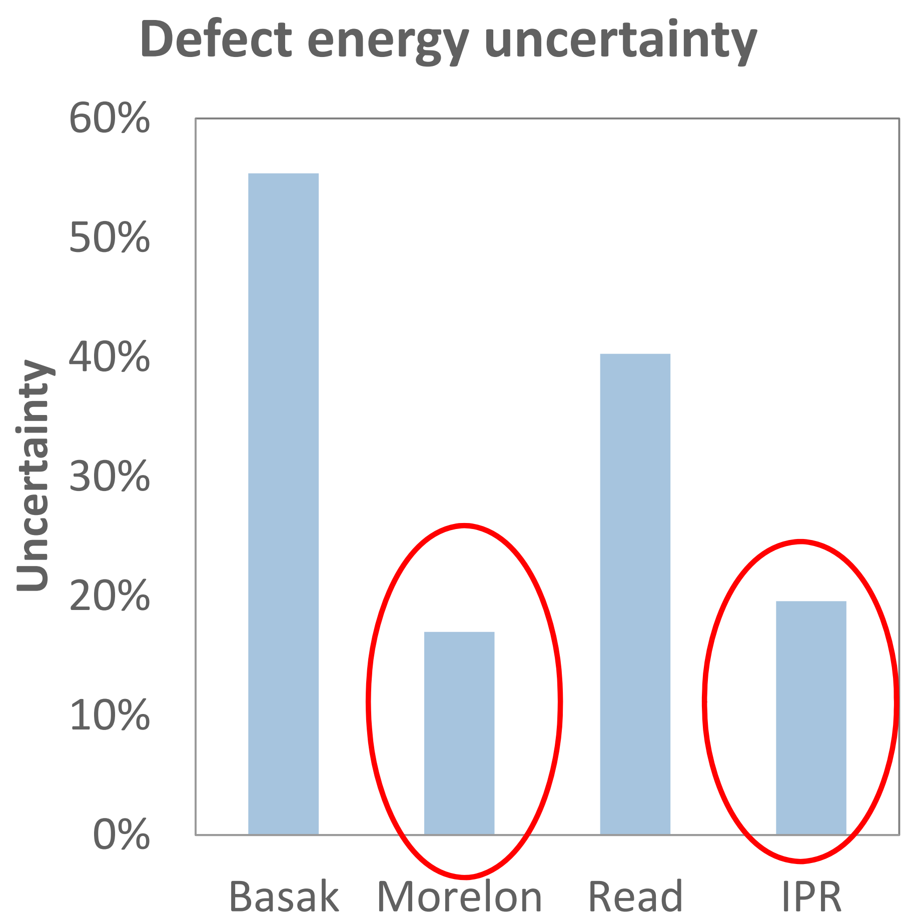
# Accuracy of Interatomic Potentials – Ab Initio MD



# The Iterative Potential Refinement (IPR) potential of $UO_2$ makes excellent predictions of both phonons and defect energetics

Schottky defect formation energies and uncertainty



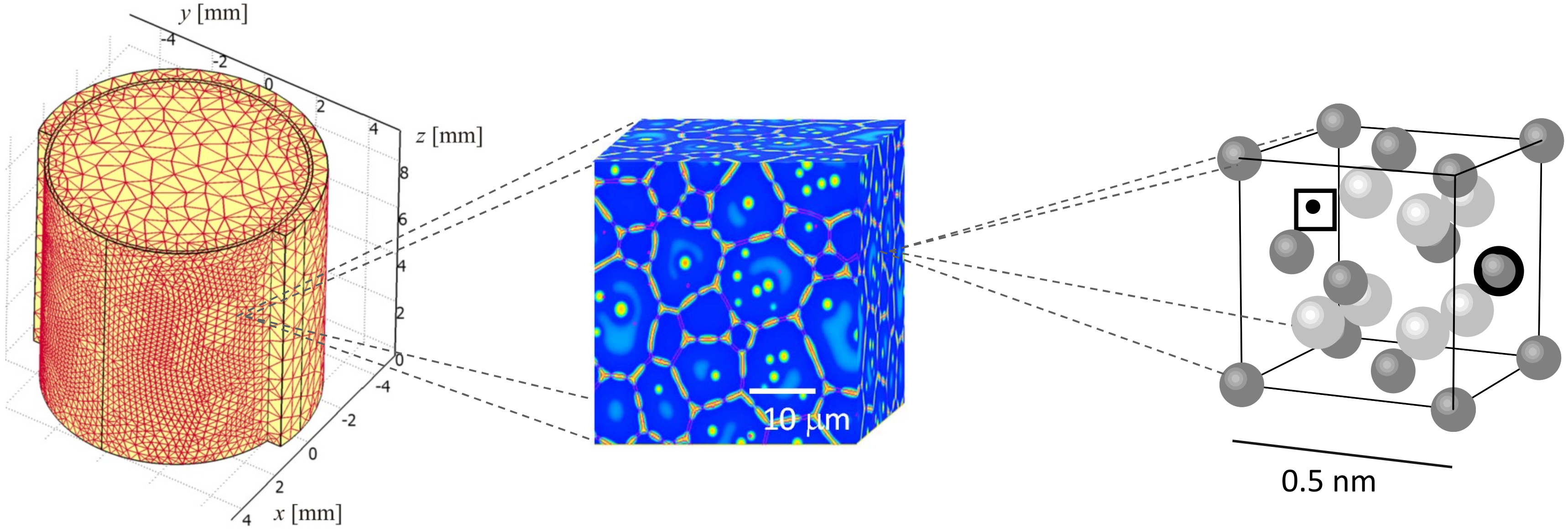

A. E. Thompson, B. Meredig, M. Stan, and C. Wolverton, J. Nucl. Mater., 446 (2014) 155

# Computational Microscopy: zoom in and out

Transport and deformation (Finite Element Method)

Microstructure evolution (Phase Field Method)

Defect formation/phase nucleation (Ab Initio Molecular Dynamics)




Bridging scales expands the investigation time and space domains.
- Lower scales help improve the understanding of underlying mechanisms.
- Higher scales help improve the prediction of global properties.

**ZOOM - a multi-scale computational microscope (ANL-Univ. of Chicago)**

Contact mstan@uchicago.edu

M. Stan, in Characterization of Materials, John Wiley & Sons, 2012.

# Summary

Evaluating uncertainty improves

- Understanding – identifying the key physics
- Prediction – qualitative is important!
- Control – optimizing properties, materials design

Uncertainty is not only a calculation output; it provides feedback to establish the necessary accuracy of measurements and simulations

Ideas for collaboration

- Quantify uncertainty of 2-D and 3-D exp/comp images
- Evaluate uncertainty propagation across time and length scales, e.g. phase stability, phase transformations
- Use machine learning for UQ, big data
- Write position paper titled “Sometimes UQ Matters”