High Temperature Thermochemical and Isotopic Simulations of High Burnup UO_2 Fuel

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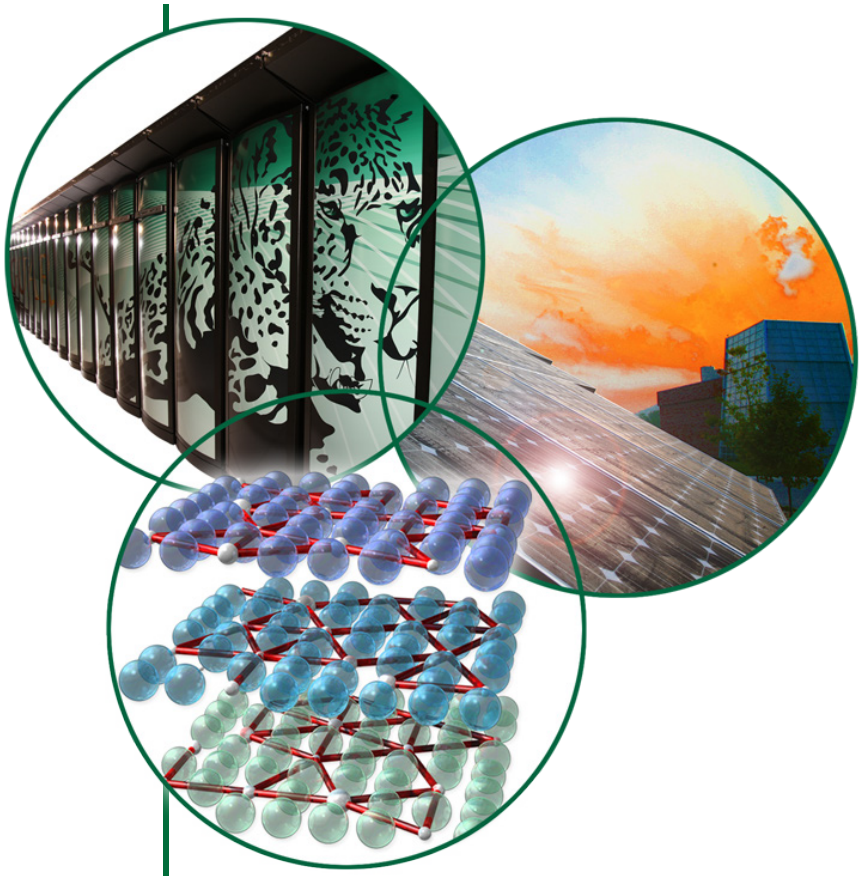
[c] Computer Science & Mathematics Division, ORNL

[*] Currently at Chalk River Laboratory and Royal Military College of Canada



Materials Modelling and Simulation of Nuclear Fuels (MMSNF)

Chicago, IL

October 14-16, 2013

U.S. DEPARTMENT OF ENERGY

NEAMS
NUCLEAR ENERGY ADVANCED MODELING & SIMULATION PROGRAM

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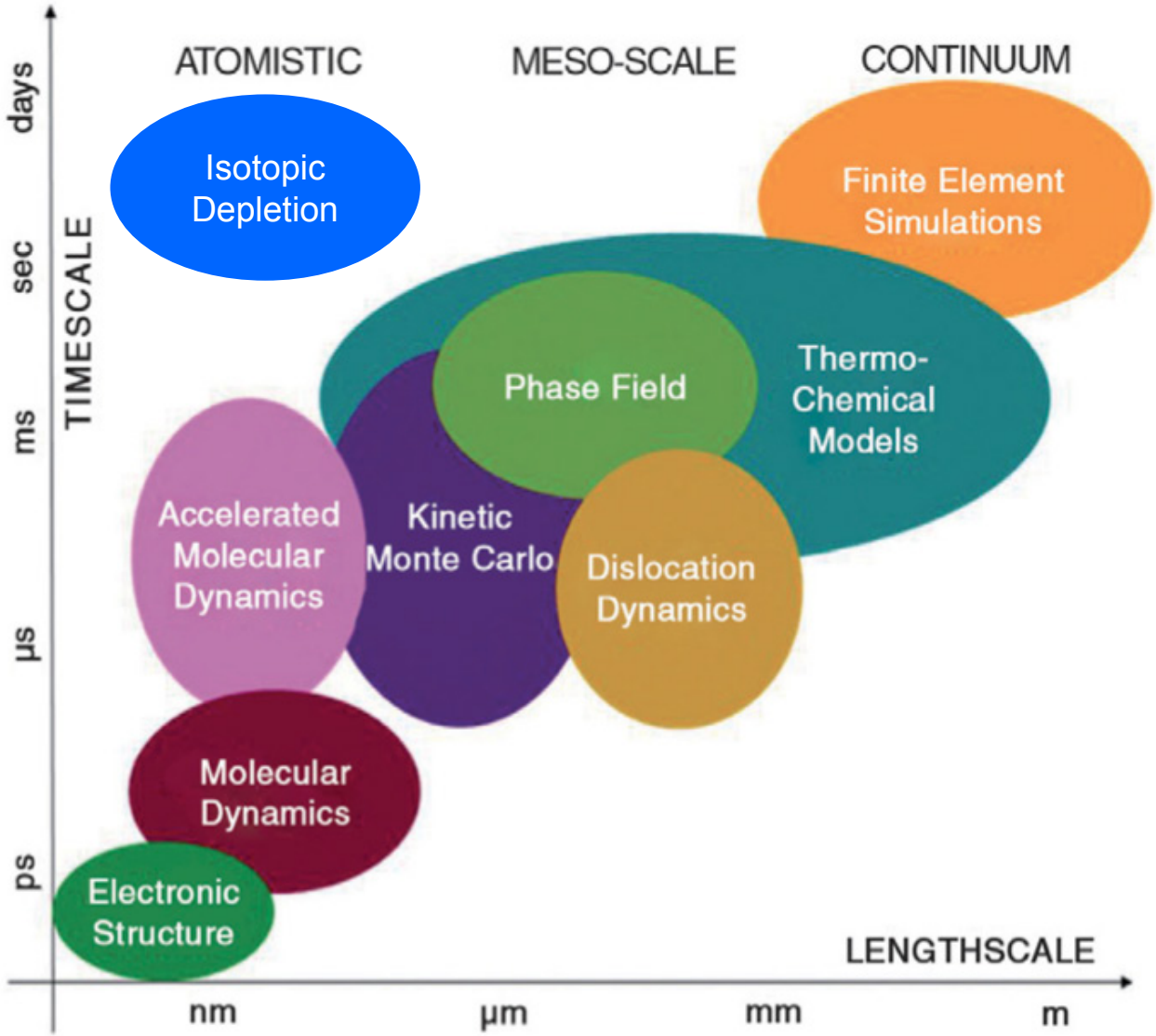
Introduction



Adapted from M. Stan, Materials Today, 12, 11 (2009) 20-28.

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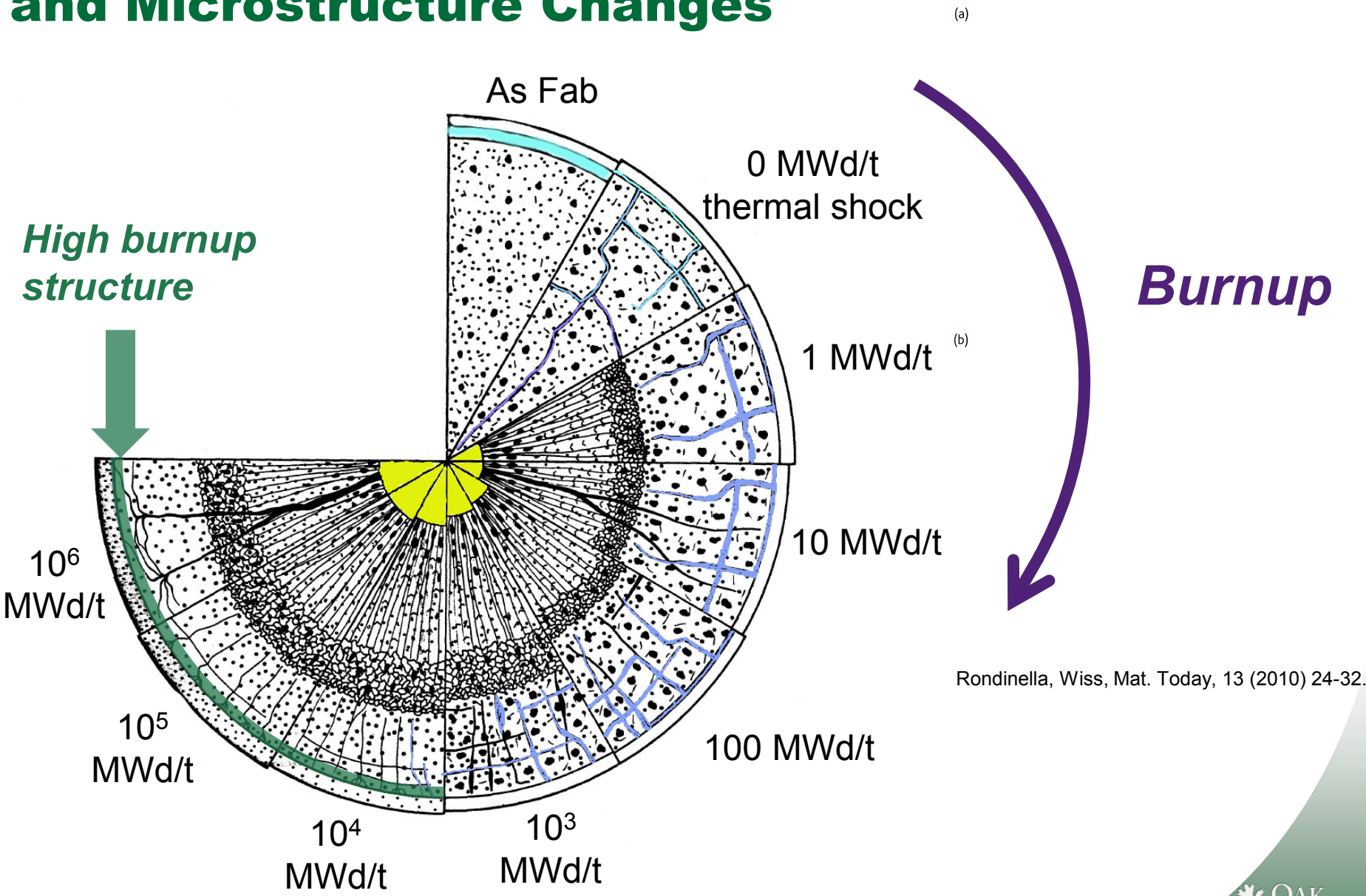
Evolution of MOX Fuel Shows Extensive Phase and Microstructure Changes



Rondinella, Wiss, Mat. Today, 13 (2010) 24-32.

Adapted from Christensen, J.A. Trans. Am. Nucl. Soc., 15 (1972) 214.

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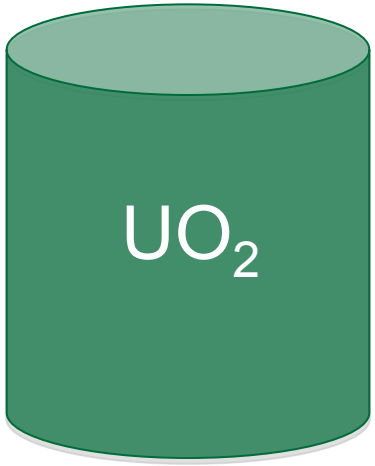
Isotopic Modelling of Nuclear Fuel

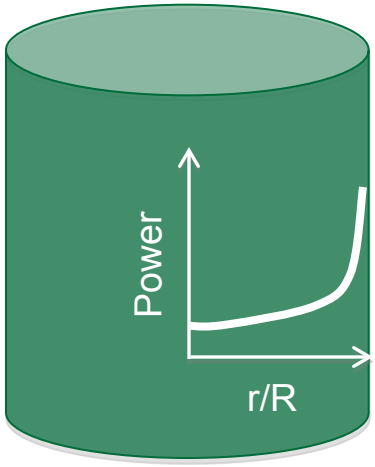
Fresh Fuel



Isotopes

^{235}U, ^{238}U and ^{16}O

Irradiated Fuel



Isotopes

^{235}U, ^{238}U and ^{16}O
+ >2000 isotopes

ORIGEN-S:

- ***Depletion***
- ***Decay***
- ***Transmutation***

- ***Neutron flux depression***
- ***Rim effect***

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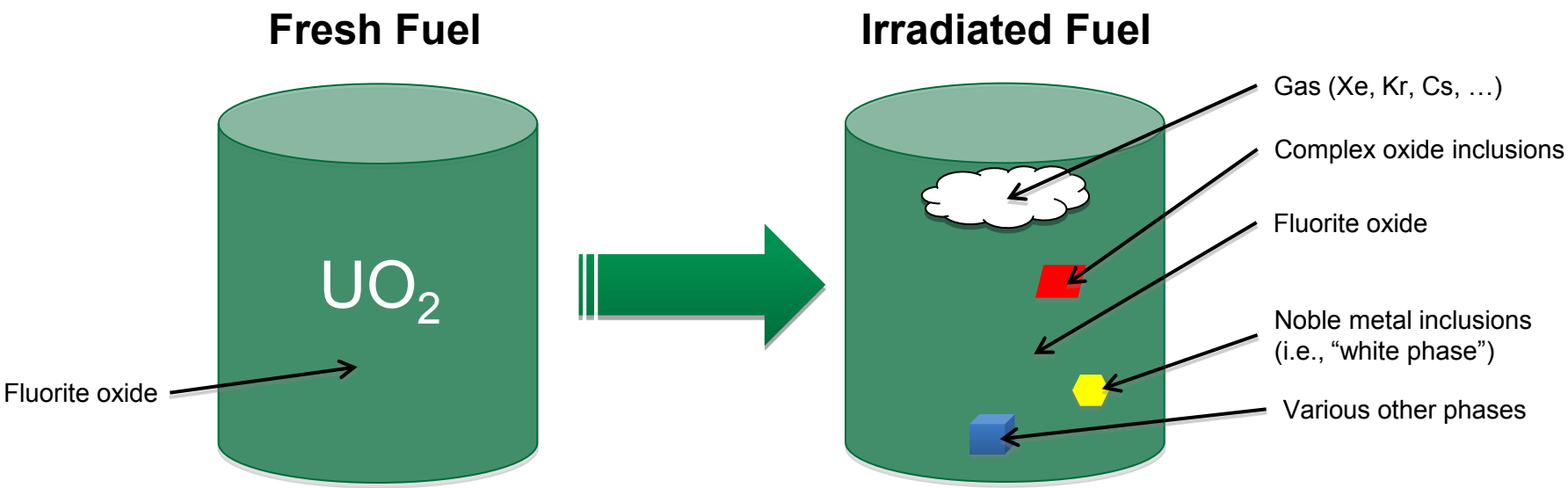
Thermodynamic Modelling of Nuclear Fuel



Elements

U and O

Elements

U, O, Pu, Xe, Zr, Mo, Ru, Nd, Ce, Cs, Sr, Ba, Pd, La, Tc, Pr, Y, Rh, Kr, Sm, Rb, Te, Np, I, Pm, Nb, Ag, Se, Eu, Cd, Sn, Gd, Br, Am, Sb, Hg, In, Cu, Cm, Tb, Ge, Dy, As, Ho, Er, Th, Zn, Pa, Ga, Pb, Ra, Ac, Bk, Cf, Bi, Po, Rn, Tl, Es, Fr, At

Adapted from D.R. Olander, "Fundamental Aspects of Nuclear Reactor Fuel Elements," U.S. Dept. of Commerce (1976).

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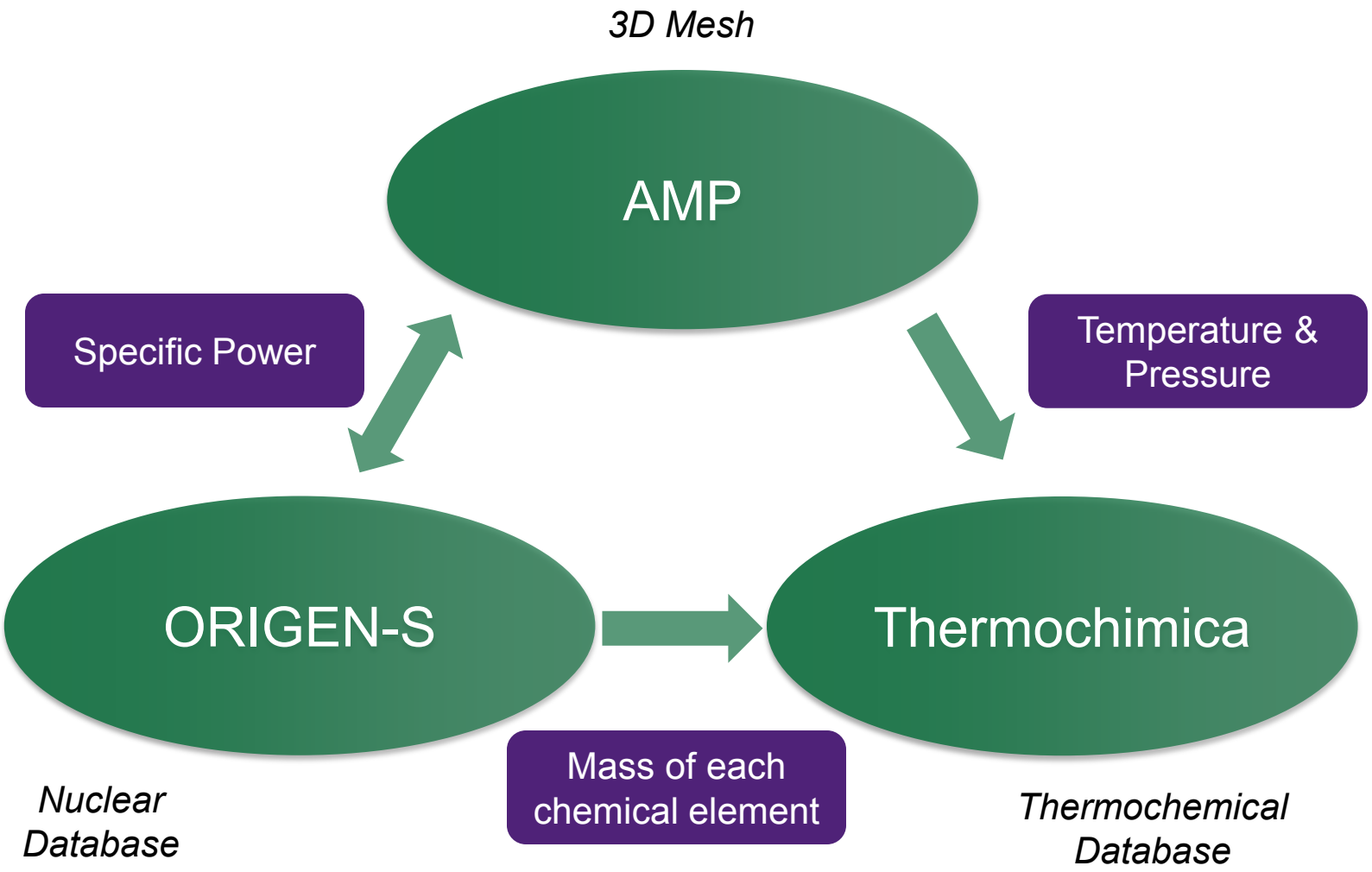
Simulation Hierarchy



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Thermochimica Overview

UO_2 Fluorite Crystal Structure

Å)

Anion sublattice

Cation sublattice

M.H.A. Piro, PhD Thesis, Royal Military College, 2011.
M.H.A. Piro, S. Simunovic, T.M. Besmann, B.J. Lewis, W.T. Thompson, Comp. Mat. Sci., 67 (2013) 266-272.

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Thermochimica Overview

UO_2 Fluorite Crystal Structure

UO_2

l Site

$UO_{2\pm x}$

U^{3+}, U^{4+}, U^{5+}, O^{2-}, Va

Defect complex of UO_2

M.H.A. Piro, PhD Thesis, Royal Military College, 2011.
M.H.A. Piro, S. Simunovic, T.M. Besmann, B.J. Lewis, W.T. Thompson, Comp. Mat. Sci., 67 (2013) 266-272.

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Thermochimica Overview

UO_2 Fluorite Crystal Structure

UO_2 + FPs

l Site

Defect complex of UO_2

Phases:

- *Fluorite oxide*
- *Noble metals (HCP, BCC, …)*
- *Oxides*
- *Uranates*
- *Molybdates*
- *Zirconates*
- *Gas*

M.H.A. Piro, PhD Thesis, Royal Military College, 2011.
M.H.A. Piro, S. Simunovic, T.M. Besmann, B.J. Lewis, W.T. Thompson, Comp. Mat. Sci., 67 (2013) 266-272.

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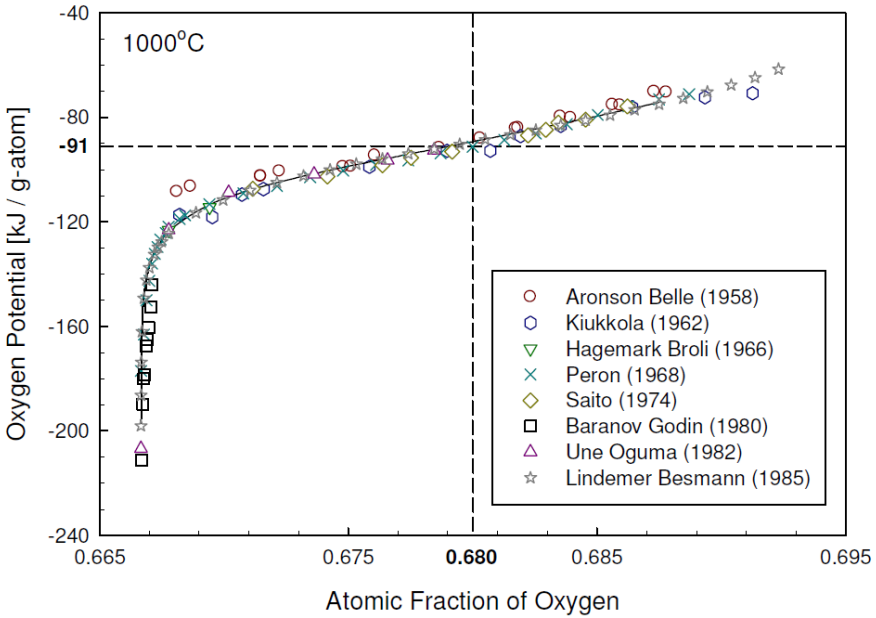
Comparison with Experiments – Isolated Validation Studies of Models

UO_{2+x} Thermodynamic Model



$$\chi_O = \left(\frac{b_O}{b_U + b_O} \right)$$

M.H.A. Piro, PhD Thesis, Royal Military College, 2011.

Gamma Spectra Model after ^{235}U Fission

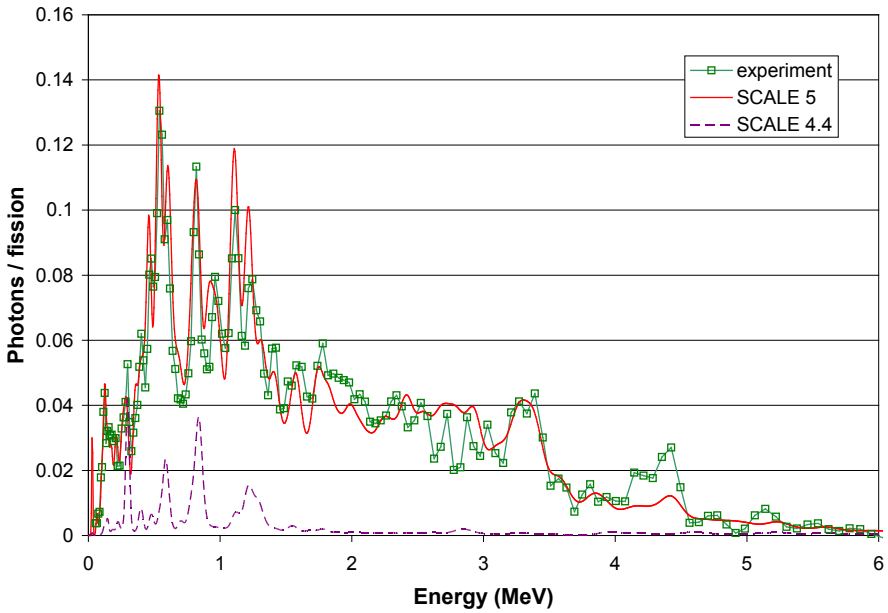


Figure kindly provided by I. Gauld, SCALE group, ORNL

Note: many other isolated validation studies have been incorporated.
The above are intended to provide simple examples.

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Comparison with Experiments – Integrated Validation Studies of Simulations

- 3.5% enriched UO_2 fuel irradiated in a European PWR to ~102 GWd/t(U).

- Sample discs (~1 mm thick) were taken from upper end of fuel stack for various measurements at ITU:
  - Integral fuel composition
  - Local fuel composition
  - Oxygen partial pressure

C.T. Walker, V.V. Rondinella, D. Papaioannou, S. Van Winckel, W. Goll, R. Manzel, J. Nucl. Mater., 345 (2005) 192-205.

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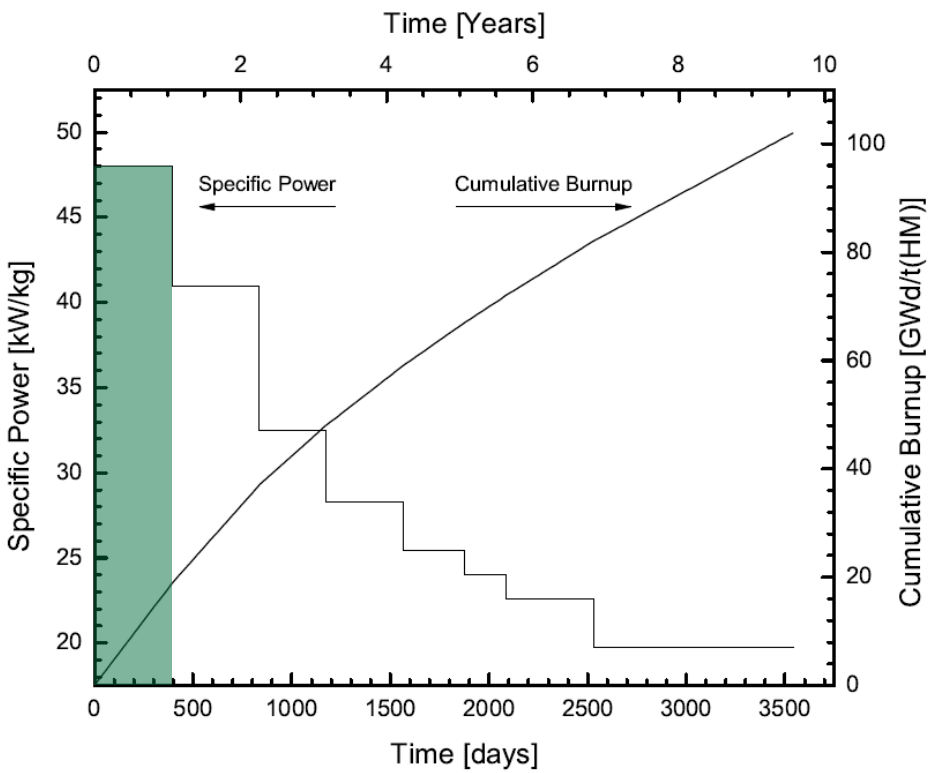
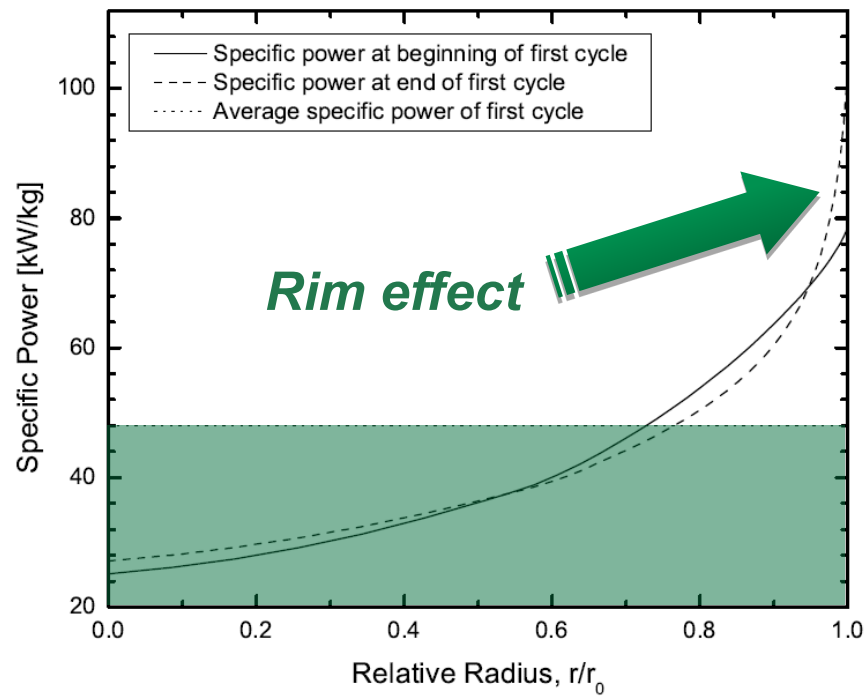
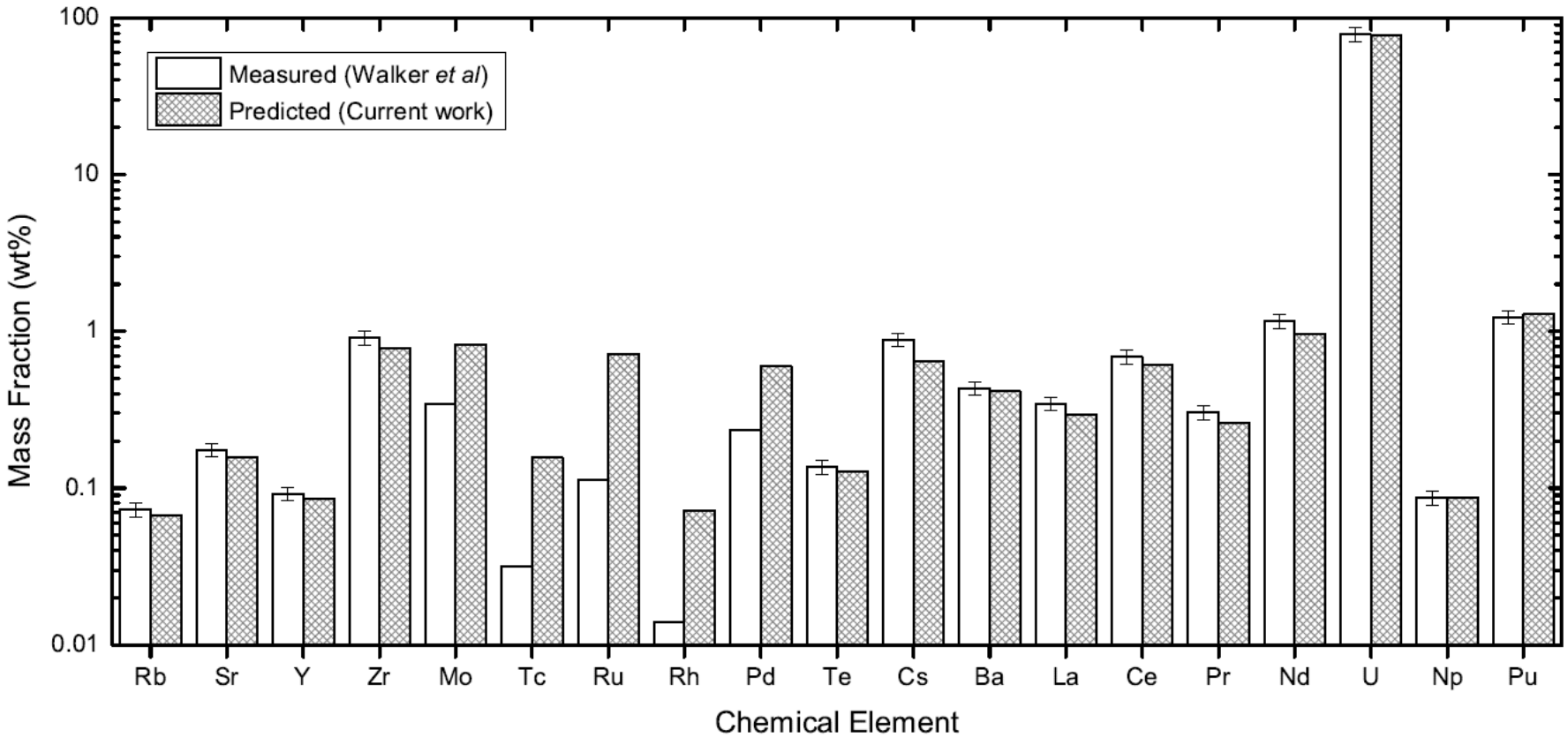
Irradiation History





M.H.A. Piro, J. Banfield, K.T. Clarno, S. Simunovic, T.M. Besmann, B.J. Lewis and W.T. Thompson, J. Nucl. Mater., 441 (2013) 240-251.

Average Fuel Composition



Measurements made with Inductively Coupled Plasma Mass Spectrometry (ICP-MS)

C.T. Walker, V.V. Rondinella, D. Papaioannou, S. Van Winckel, W. Goll, R. Manzel, J. Nucl. Mater., 345 (2005) 192-205.

M.H.A. Piro, J. Banfield, K.T. Clarno, S. Simunovic, T.M. Besmann, B.J. Lewis and W.T. Thompson, J. Nucl. Mater., 441 (2013) 240-251.

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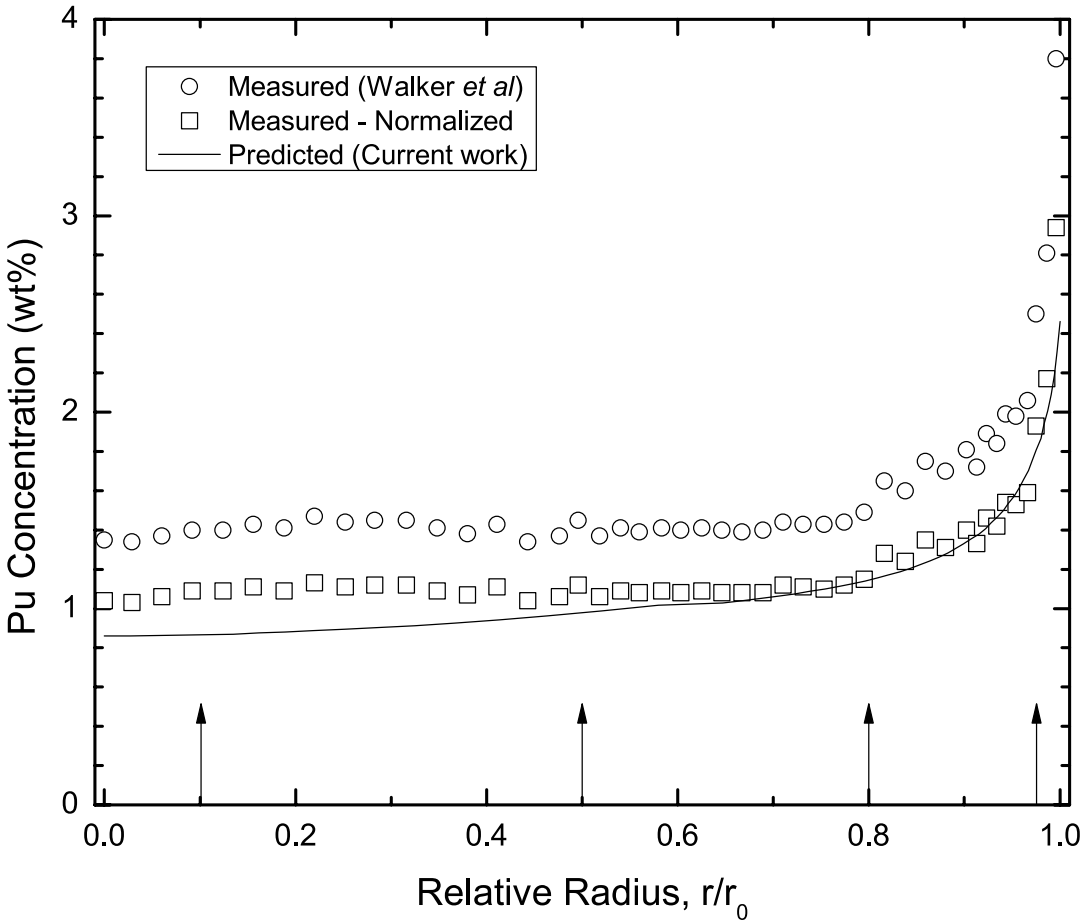
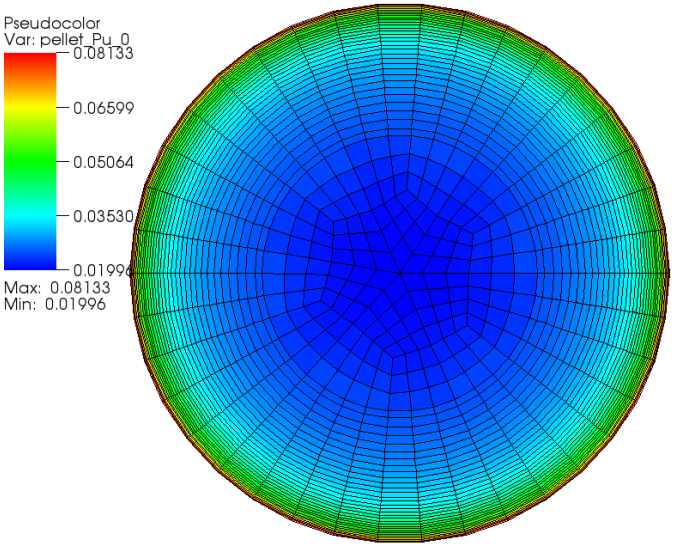
Predicted Pu Distribution



Pu Concentration
[g-at/g-at]



Measurements made with Electron Probe Micro Analysis (EPMA)

C.T. Walker, V.V. Rondinella, D. Papaioannou, S. Van Winckel, W. Goll, R. Manzel, J. Nucl. Mater., 345 (2005) 192-205.

M.H.A. Piro, J. Banfield, K.T. Clarno, S. Simunovic, T.M. Besmann, B.J. Lewis and W.T. Thompson, J. Nucl. Mater., 441 (2013) 240-251.

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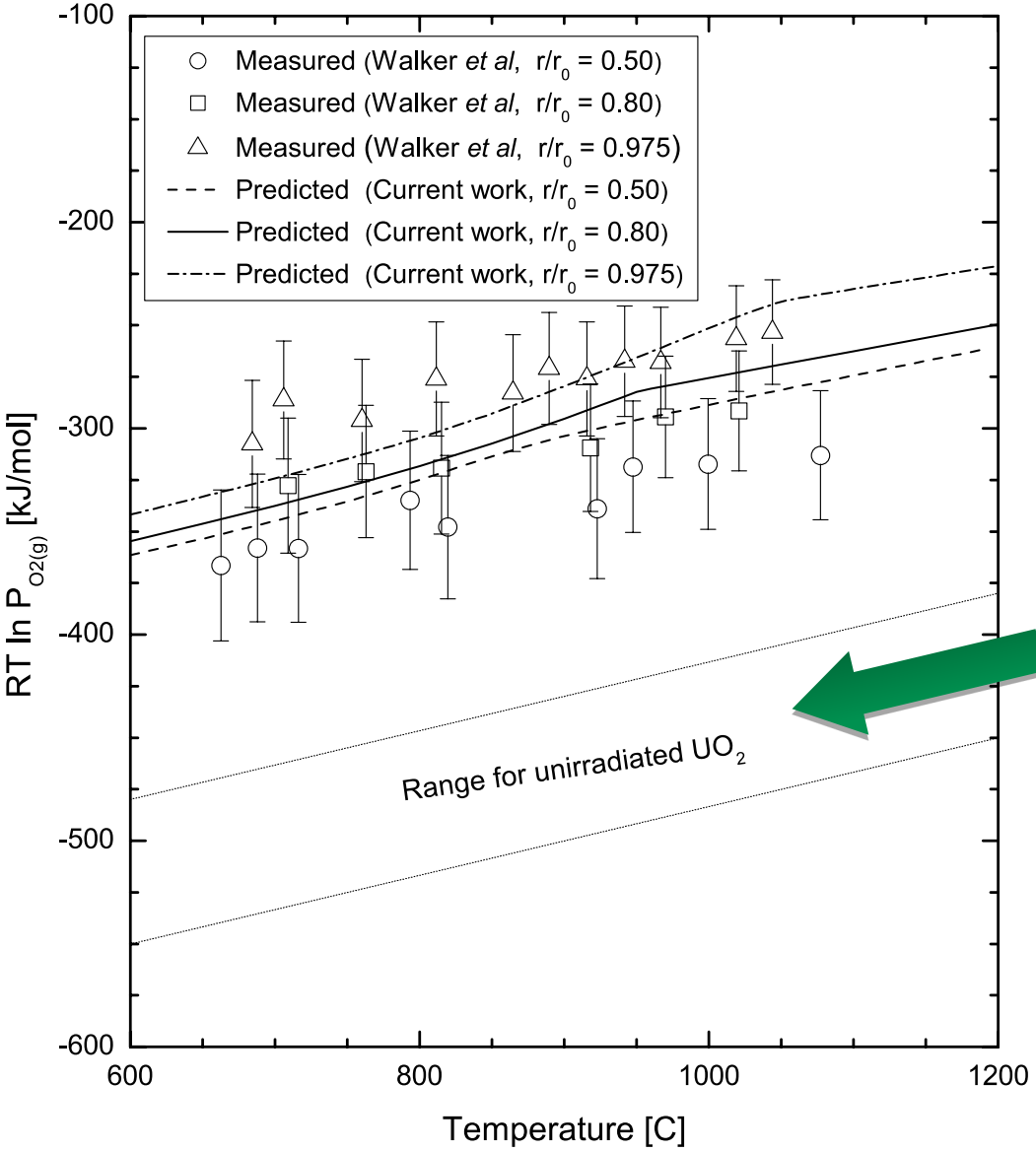
Oxygen Partial Pressure ($RT \ln p_{O2(g)}$)



Most codes that account for fuel chemistry assume "fresh fuel"

Measurements made with a miniature solid state galvanic cell

C.T. Walker, V.V. Rondinella, D. Papaioannou, S. Van Winckel, W. Goll, R. Manzel, J. Nucl. Mater., 345 (2005) 192-205.

M.H.A. Piro, J. Banfield, K.T. Clarno, S. Simunovic, T.M. Besmann, B.J. Lewis and W.T. Thompson, J. Nucl. Mater., 441 (2013) 240-251.

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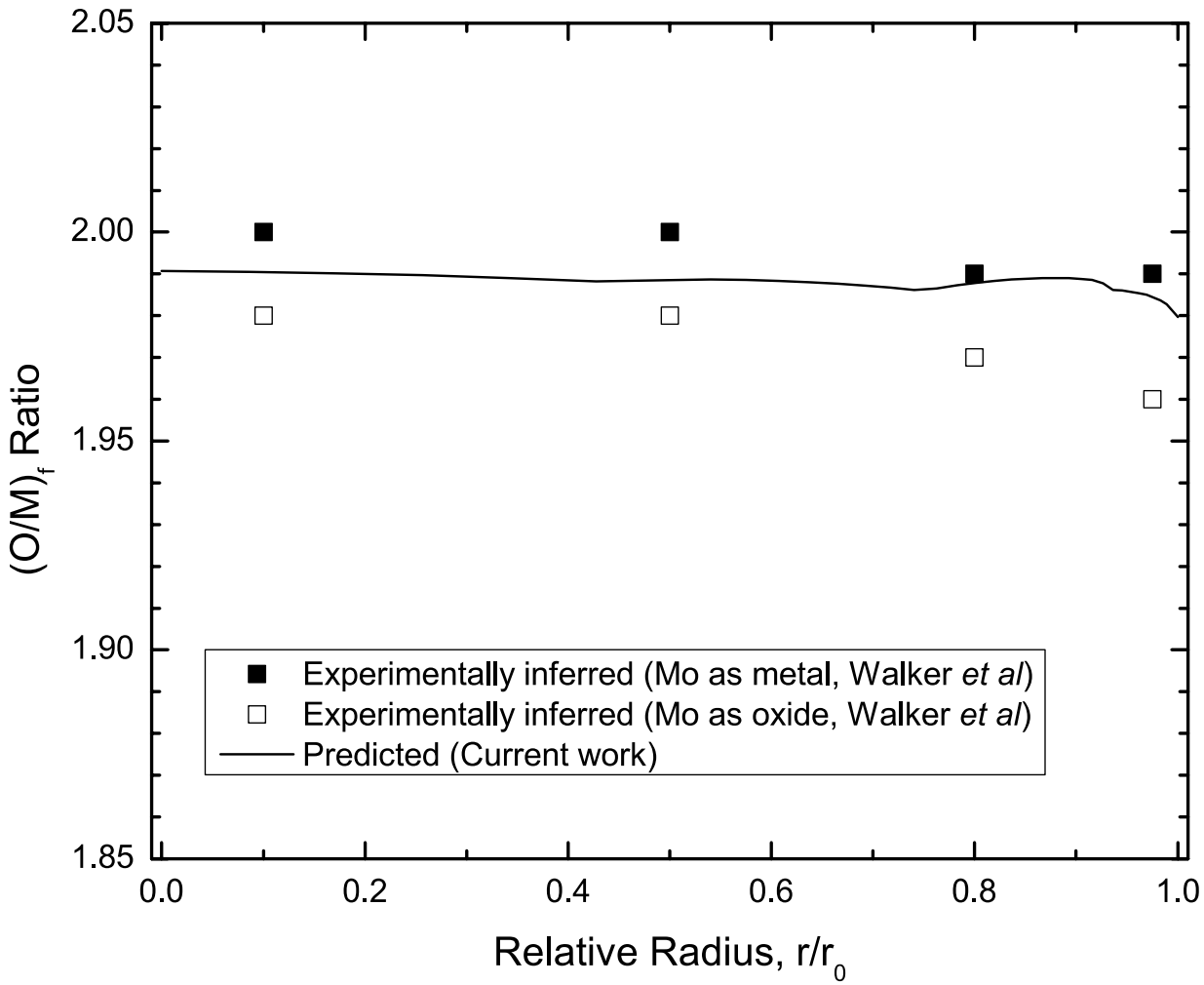
Oxygen-to-Metal Ratio



Experimentally inferred values for O/M were derived by ICP-MS, EPMA and assumptions regarding phase equilibria.

C.T. Walker, V.V. Rondinella, D. Papaioannou, S. Van Winckel, W. Goll, R. Manzel, J. Nucl. Mater., 345 (2005) 192-205.

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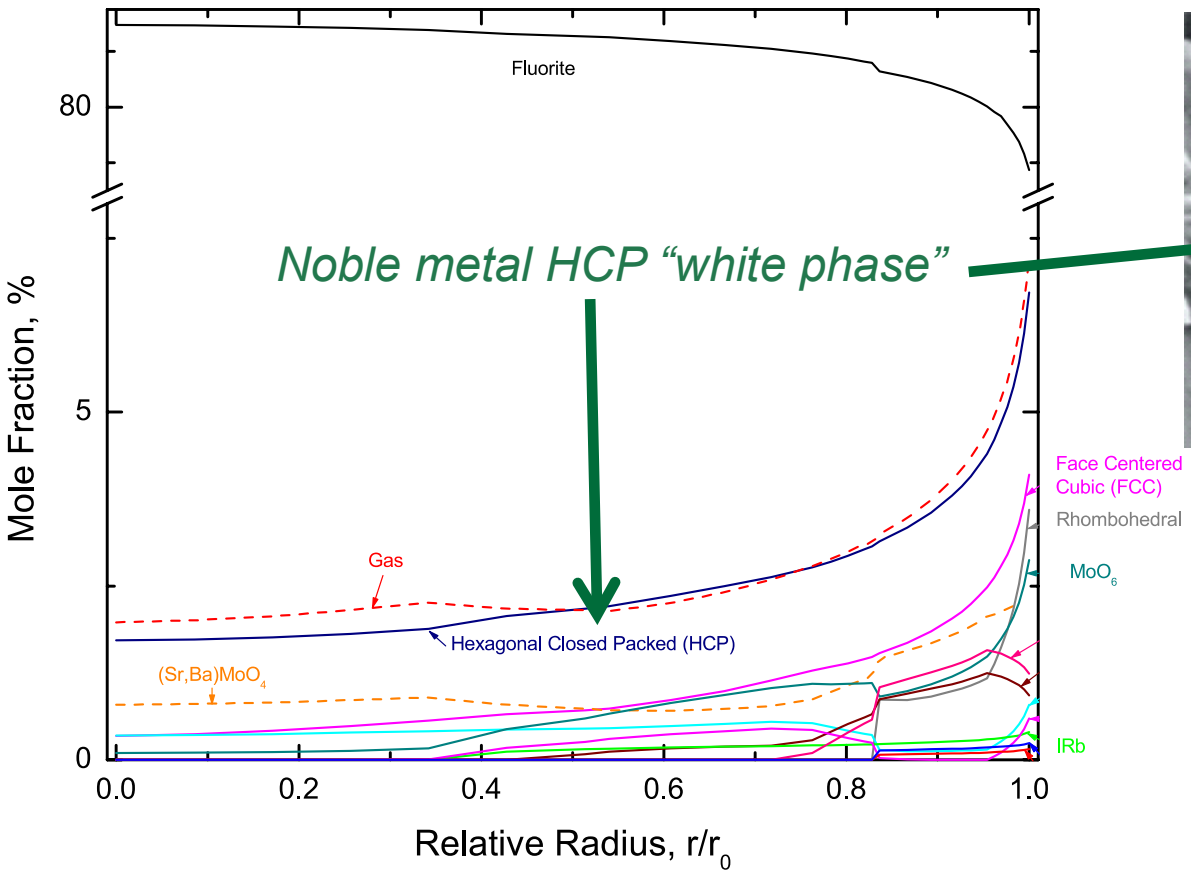
Phase Composition



SEM in high burnup structure [~75 GWd/t(U)]

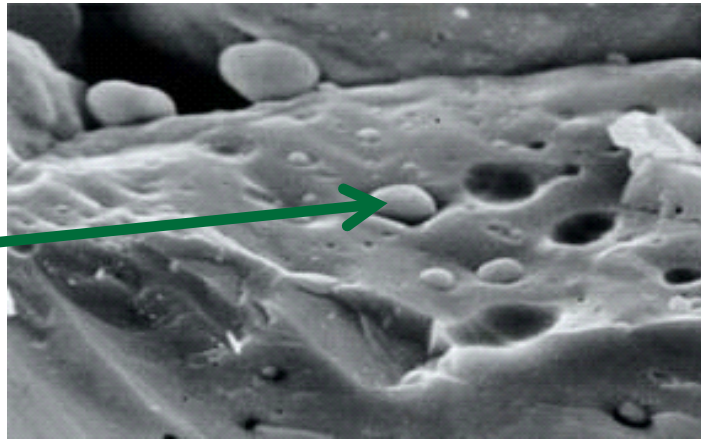


Figure kindly provided by T. Wiss and V.V. Rondinella (ITU)

T. Sonoda, M. Kinoshita, I.L.F. Ray, T. Wiss, H. Thiele, D. Pellottiero, V.V. Rondinella, Hj. Matzke, Nucl. Inst. Meth. (2002).

M.H.A. Piro, J. Banfield, K.T. Clarno, S. Simunovic, T.M. Besmann, B.J. Lewis and W.T. Thompson, J. Nucl. Mater., 441 (2013) 240-251.

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Summary

- Advanced simulation capability has been developed that enhances fuel behaviour predictions by integrating thermochemistry and isotopic evolution.

- Simulation predictions are in relatively good agreement with experiments…

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Final thought…

"In theory, there is no difference between theory and practice. But in practice, there is."

- Yogi Berra

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Acknowledgements

- V.V. Rondinella, C.T. Walker, D. Papaioannou,T. Wiss (ITU), I. Gauld and L. Ott (ORNL) for helpful technical discussions.
- Colleagues on the AMP and SCALE teams (ORNL).
- The development of the Advanced Multi-Physics (AMP) code was funded by the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program of the U.S. Department of Energy Office of Nuclear Energy, Advanced Modeling and Simulation Office.

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