Integrated analysis of microstructure and thermal conductivity in irradiated materials

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Acknowledgements

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Collaborators:

Thermal conductivity measurements:

- David Hurley, Zilong Hua, Amey Khanolkar, Tsveti Pavlov (INL),

Samples, Irradiation and characterization:

- Lingfeng He (NSCU), Kaustubh Bawane (INL), Matt Mann (AFRL), Lin Shao (TAMU), and Fabiola Cappia (INL)

Modeling:

- Linu Malakkal, Shuxiang Zhou, Chao Jiang (INL), Anter el Azab (Purdue), Chris Mariannetti (Columbia), Miaomiao Jin (PSU),

Temperature dependent microstructure

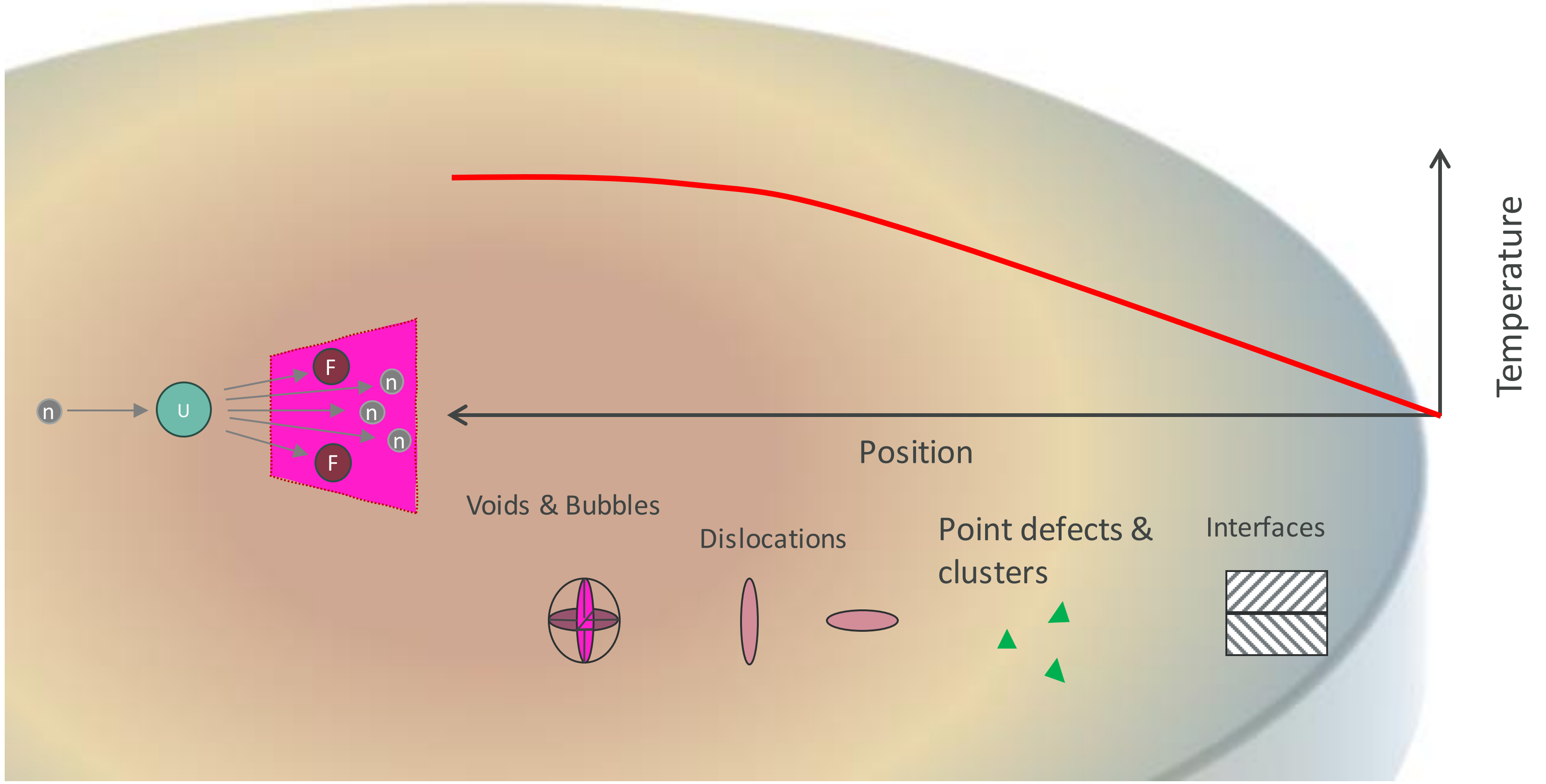
$$D = D_0 \exp(-Q_a/k_B T)$$

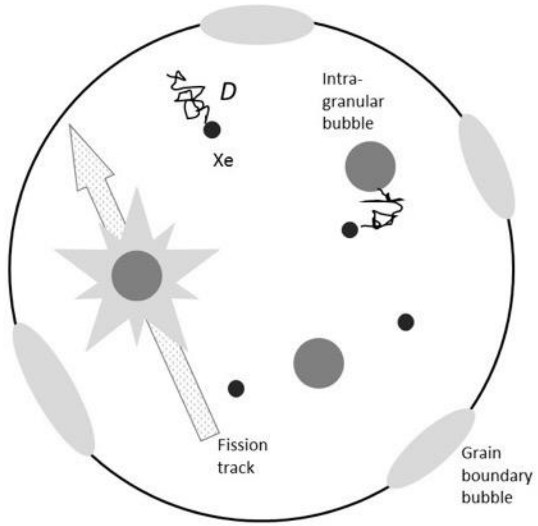
Grain growth: $d^2 - d_0^2 = \boldsymbol{D}t$

Fission gas release: $f_c = 4\left(\frac{\boldsymbol{D}t}{\pi a^2}\right)^{\frac{1}{2}} - \frac{3}{2}\frac{\boldsymbol{D}t}{a^2}$

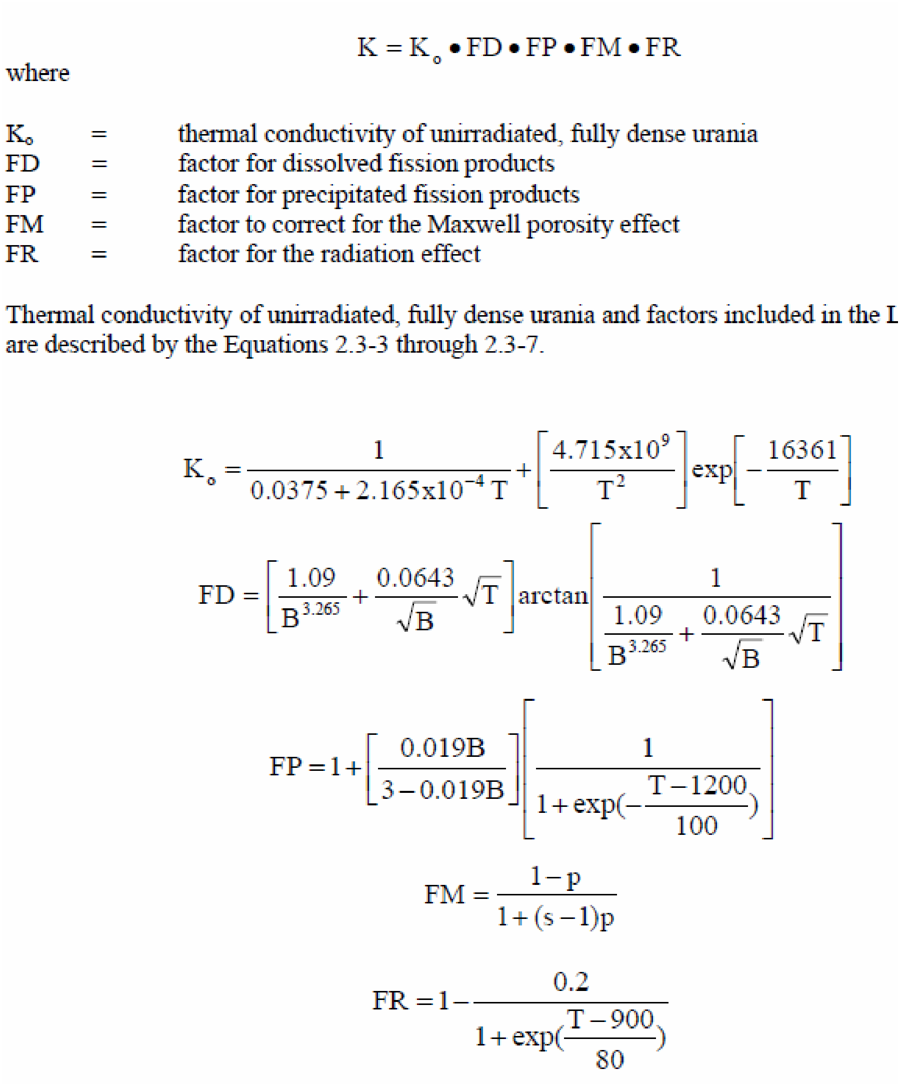
Nuclear fuel

- Strong temperature gradient
- Fission rate is also not uniform
- Microstructure evolution is governed by atomic diffusion with Arrhenius-type temperature dependence





UO_2 conductivity correlations in FPCs



$$K = K_o \bullet FD \bullet FP \bullet FM \bullet FR$$

where

K_o = thermal conductivity of unirradiated, fully dense urania
FD = factor for dissolved fission products
FP = factor for precipitated fission products
FM = factor to correct for the Maxwell porosity effect
FR = factor for the radiation effect

Thermal conductivity of unirradiated, fully dense urania and factors included in the I are described by the Equations 2.3-3 through 2.3-7.

$$K_o = \frac{1}{0.0375 + 2.165\text{x}10^{-4}\,T} + \left[\frac{4.715\text{x}10^9}{T^2}\right]\exp\left[-\frac{16361}{T}\right]$$

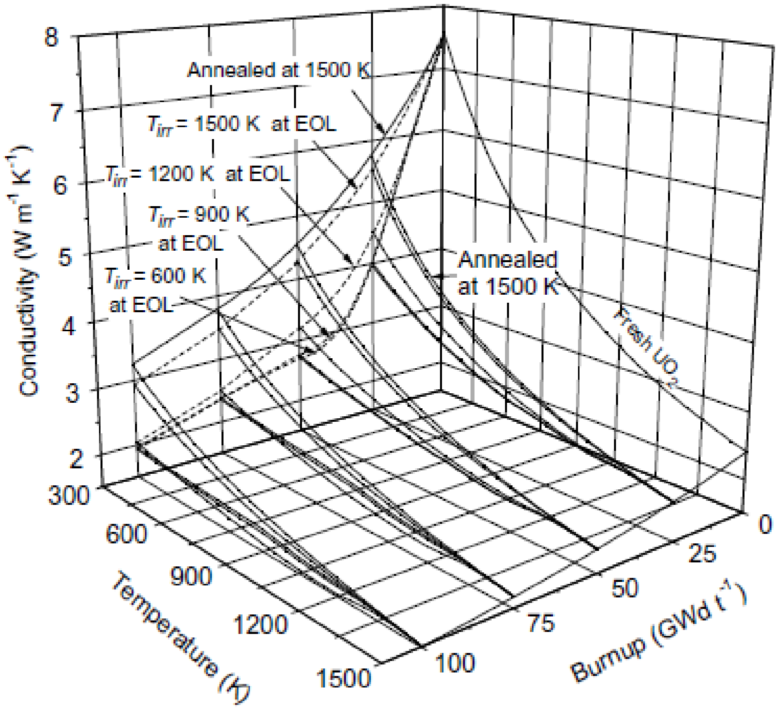
$$FD = \left[\frac{1.09}{B^{3.265}} + \frac{0.0643}{\sqrt{B}}\sqrt{T}\right]\arctan\left[\frac{1}{\frac{1.09}{B^{3.265}} + \frac{0.0643}{\sqrt{B}}\sqrt{T}}\right]$$

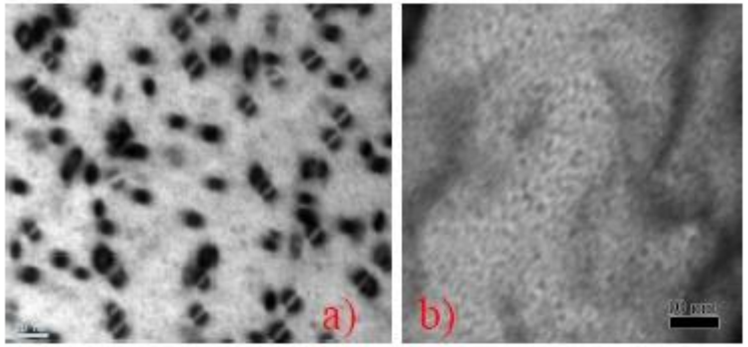
$$FP = 1 + \left[\frac{0.019B}{3 - 0.019B}\right]\left[\frac{1}{1 + \exp(-\frac{T-1200}{100})}\right]$$

$$FM = \frac{1-p}{1+(s-1)p}$$

$$FR = 1 - \frac{0.2}{1 + \exp(\frac{T-900}{80})}$$

- Common correlation are by Lucuta and Ronchi ˙
  - G. Lucuta, *et al.*,. JNM 232, 166-180 (1996)
  - Ronchi et al, JNM 327 (2004) 58
- Limitation of current fuel performance codes
  - No spatial resolution
  - No detailed microstructure information
- NE's NEAMS and BES's EFRC address these limitations

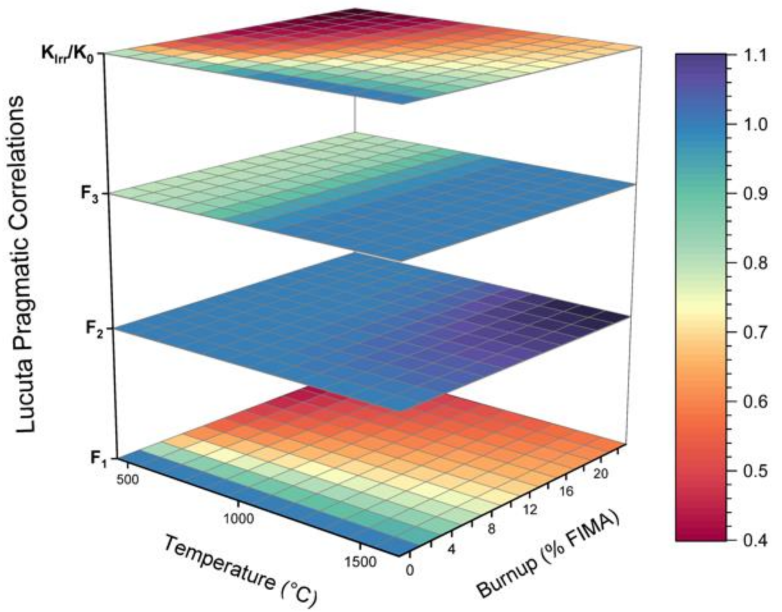




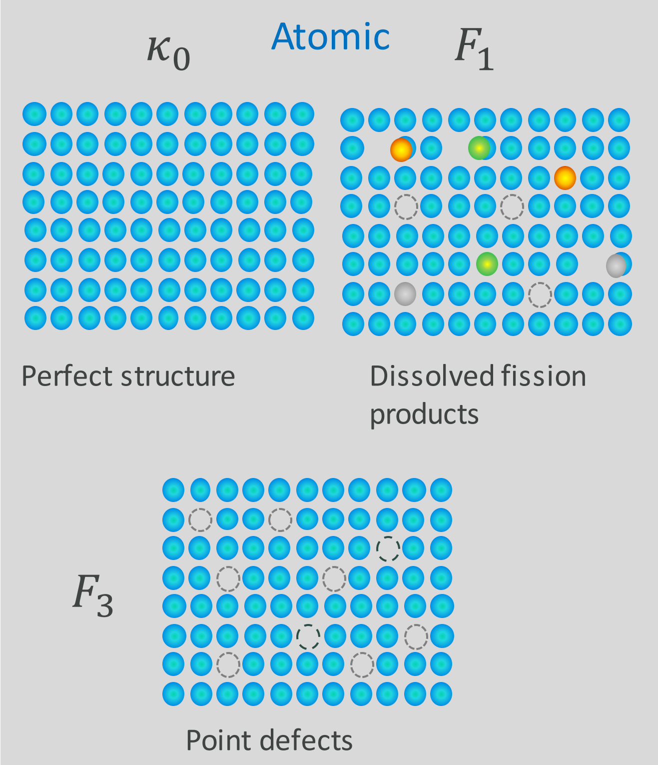
TEM images of Krypton implantation in UO2 showing a) dislocation loops, b) bubbles over focus, and c) bubbles under focus.

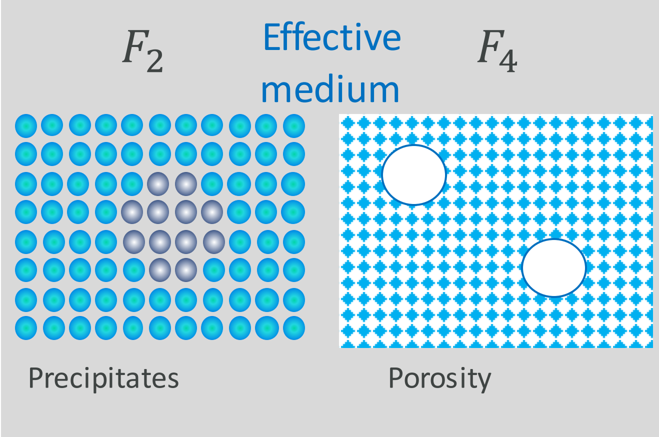
Thermal conductivity in engineering fuel performance codes

$$\kappa_{\mathrm{irr}} = \kappa_0 F_1 F_2 F_3 F_4$$

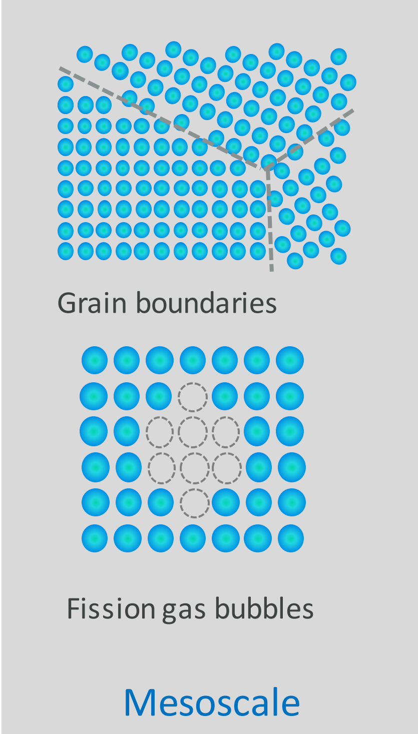


- Conductivity reduction by dissolved fission product is governed by burnup
- Impact of radiation defects was deemed to complex to model and relied in empirical model





Validated empirical correlations



Empirically treated needs more validation

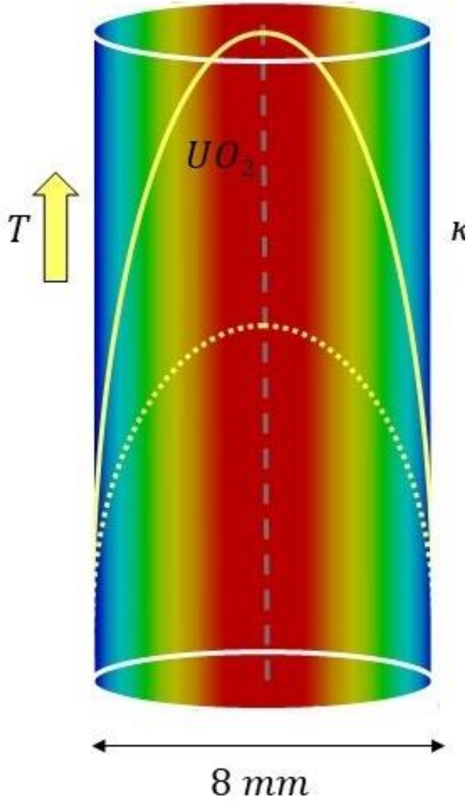
Not considered

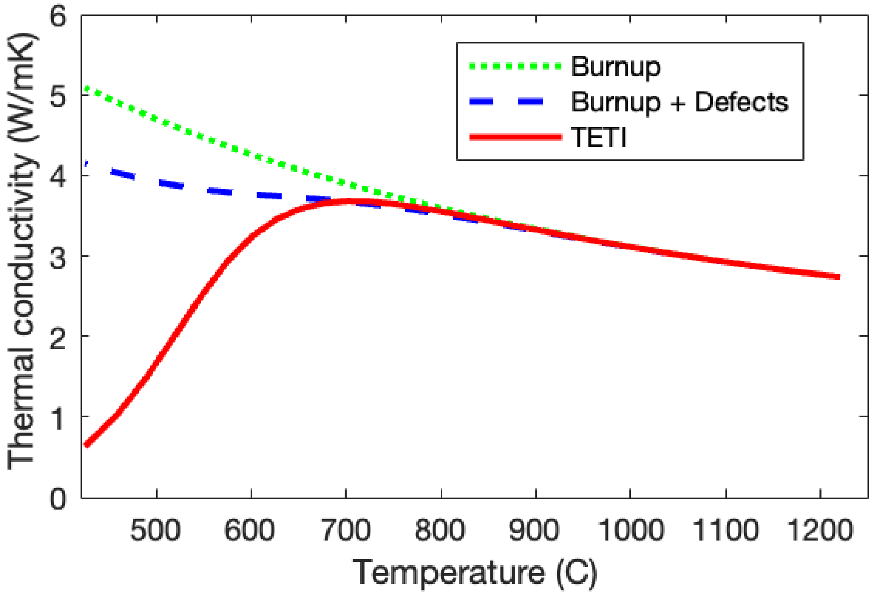
Hurley *et al.*, Chem. Rev. 122, 3711 (2022)
Lucuta, *et al.*,. JNM 232, 166-180 (1996)
Ronchi et al., JNM 327, 58 (2004)

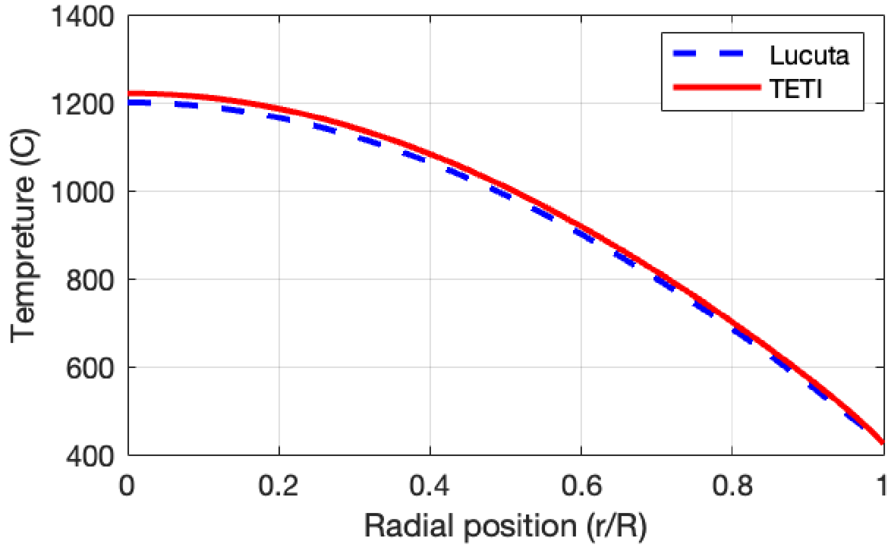
Lucuta Model model refinement

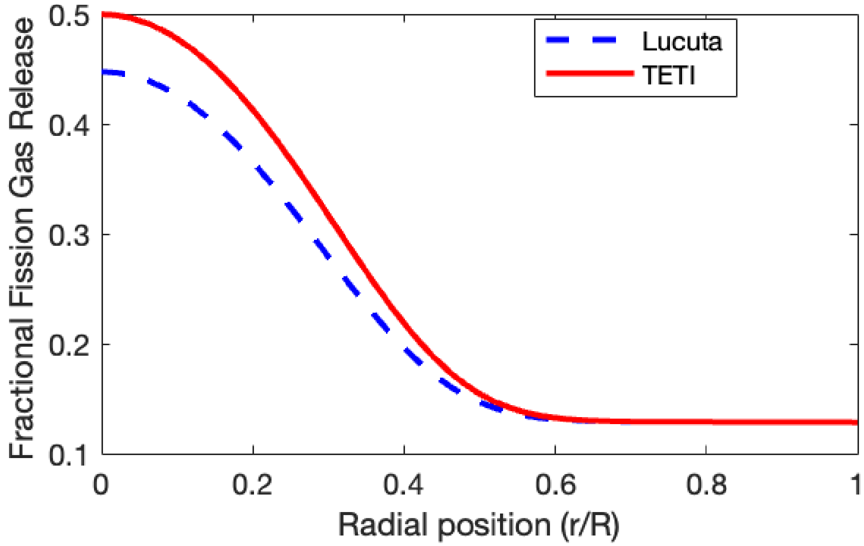
$$\kappa_0 F_1 = \frac{1}{A + A_{BU} + BT}$$

$$\boxed{F_3 = 1 - \frac{0.2}{1 + \exp\left(\frac{T - 900}{80}\right)}}$$









P. G. Lucuta, *et al.*,. JNM 232, 166-180 (1996)
Ferrigno et. al J. Nucl. Mater. 573, 154108 (2023)

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Measurement of thermal conductivity/diffusivity

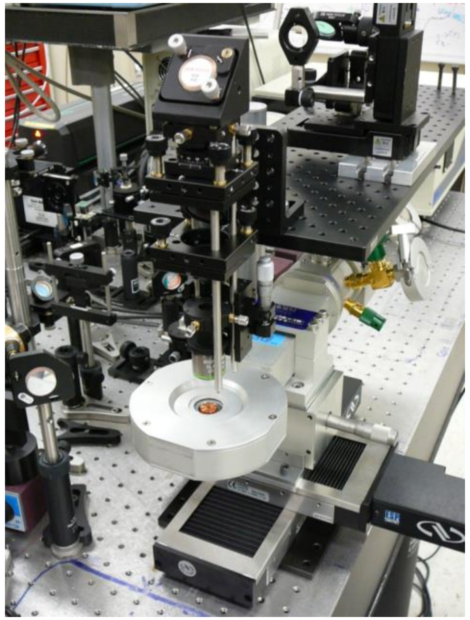
$$Q'' = -\kappa \vec{\nabla} T$$

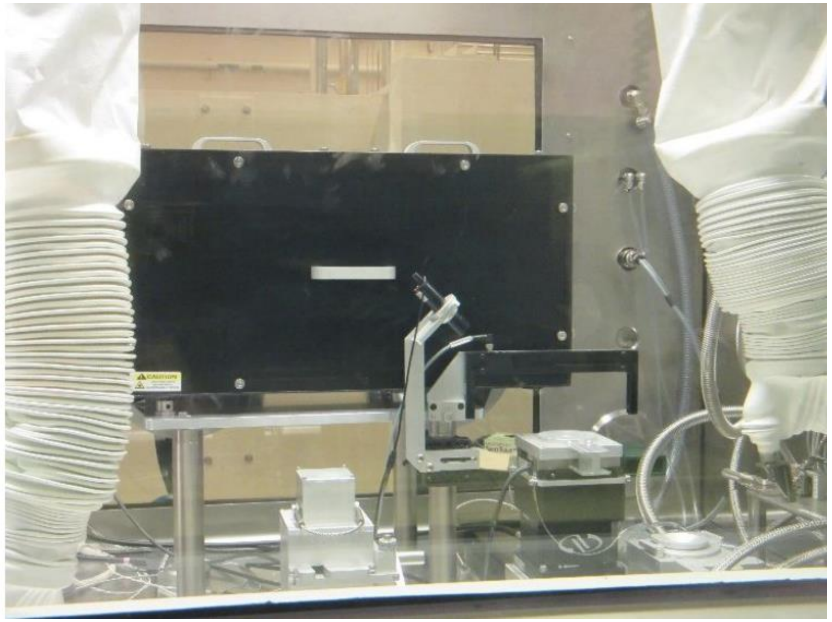
$$\rho C \frac{\partial T}{\partial t} = \vec{\nabla} \cdot \left(\kappa \vec{\nabla} T\right) + q$$

$$D = \frac{\kappa}{\rho C}$$

Laser flash

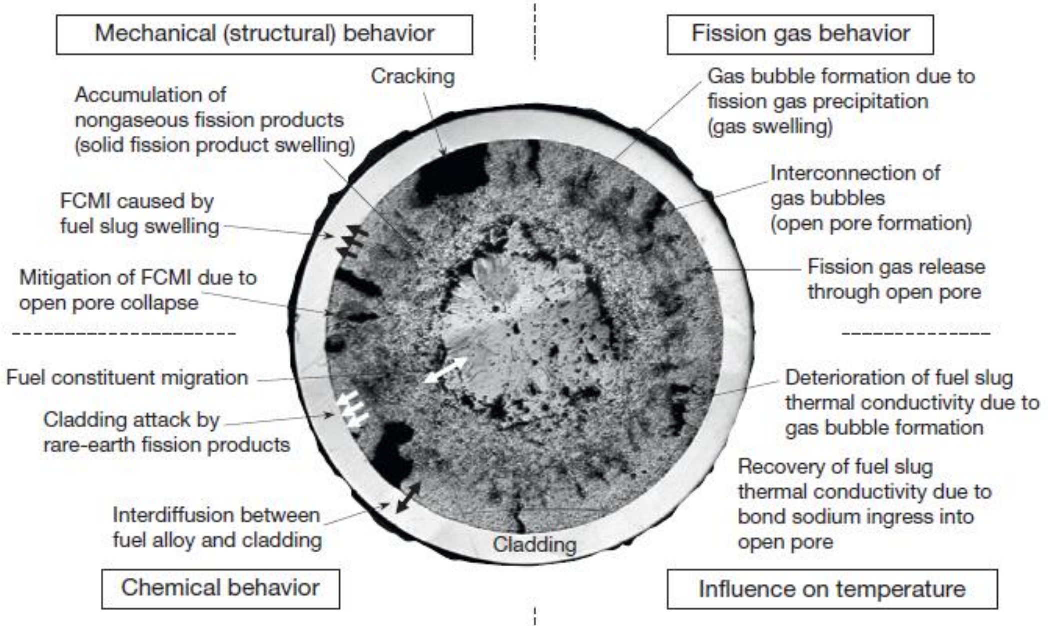
Modulated thermoreflectance / thermal conductivity microscope



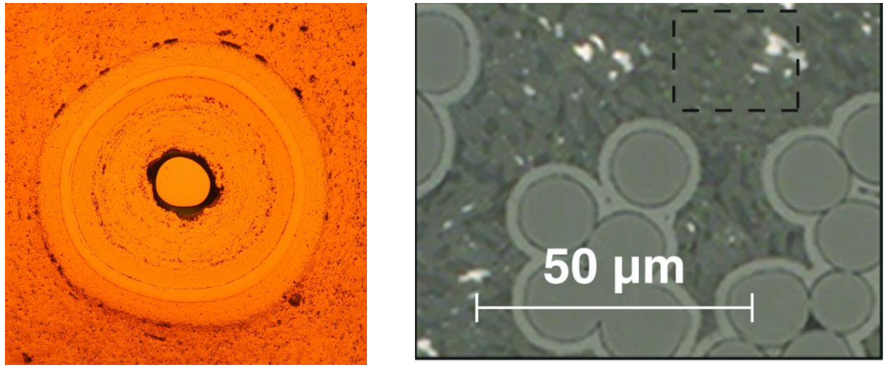


- Methods applied to fuels are typically transient and thus measure thermal diffusivity
- Laser flash analysis is typically used for bulk measurements
- Modulated thermoreflectance is commonly used for studying thermal transport in thin films for thermal management of electronic devices and thermoelectrics

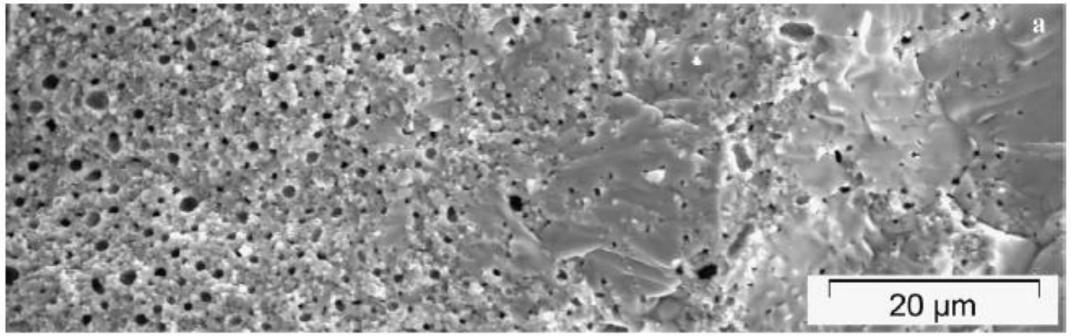
Need for spatial resolution techniques



T. Ogata, Compreh. Nuc. Mat. Vol 3, pp 1-40 (2012)



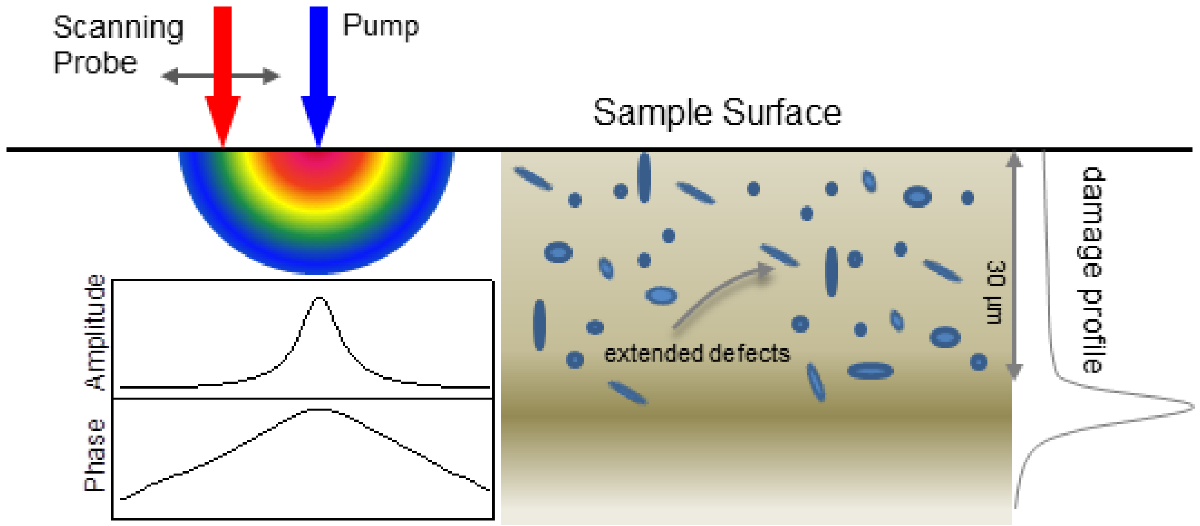
- Irradiated fuels have nonhomogeneous damage profile
  - Constituent redistribution in U-metal fuels, UO_2 high burnup structure, TRISO fuel
- Advanced composite materials
  - SiC/SiC fiber composites
  - Corrosion resistant coatings for cladding
- Fundamental studies that utilize ion beam irradiation
- Samples with small dimensions and irregular shapes

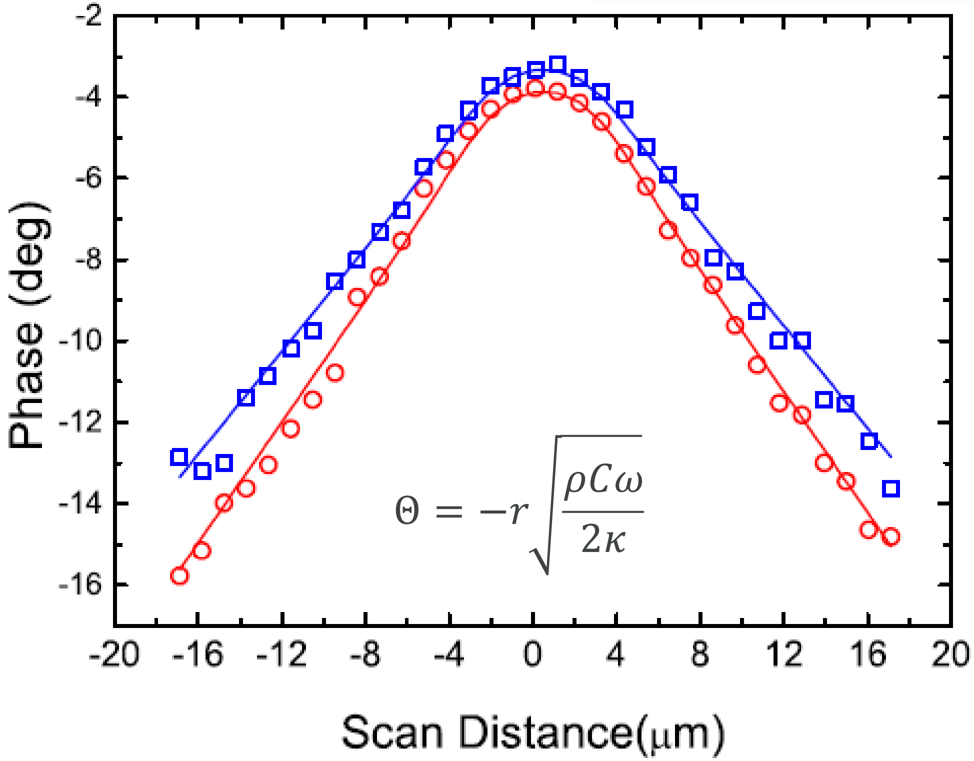


Sonoda et al,

Modulated thermoreflectance method

κ_{ref} = 146 ± 5 W/mK
κ_{irr} = 137 ± 4 W/mK





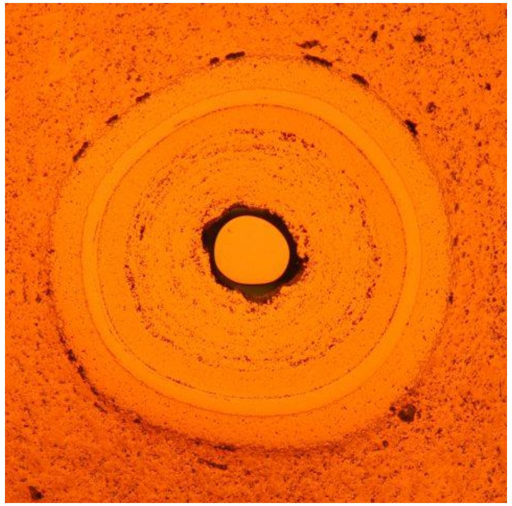
$$\rho C \frac{\partial T}{\partial t} = \vec{\nabla} \cdot \left(\kappa \vec{\nabla} T\right) + q e^{-i\omega t}$$

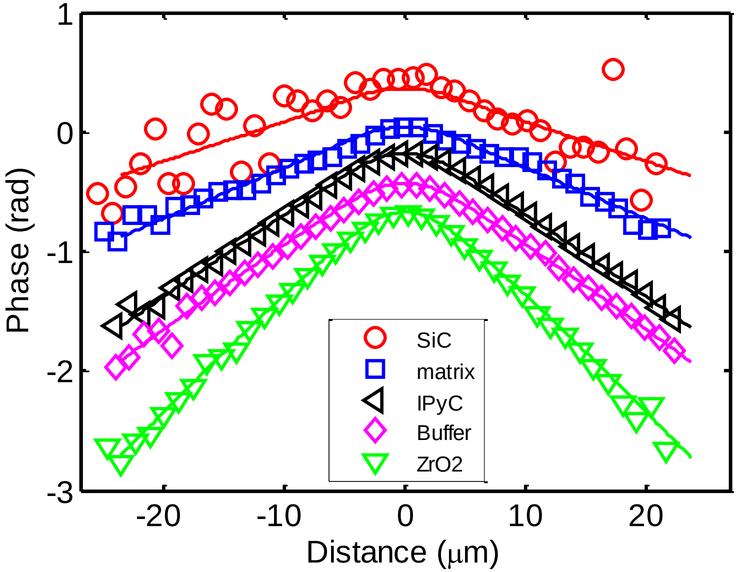
$$i\omega T_{\omega} = \frac{k}{\rho C} \nabla^{2} T_{\omega} + \frac{q'''}{\rho C}$$

Khafizov *et al.*, Nucl. Instrum. Meth. B 325, 11 (2014)
Hurley *et al.*, Rev. Sci. Instrum. 86, 123901 (2015)
Khafizov et al., J. Mater. Res. 32, 204 (2017)

The slope of phase profile is larger in irradiated sample (red) than in reference (blue), an indication of thermal conductivity reduction in irradiated sample

Conductivity of individual layers in TRISO





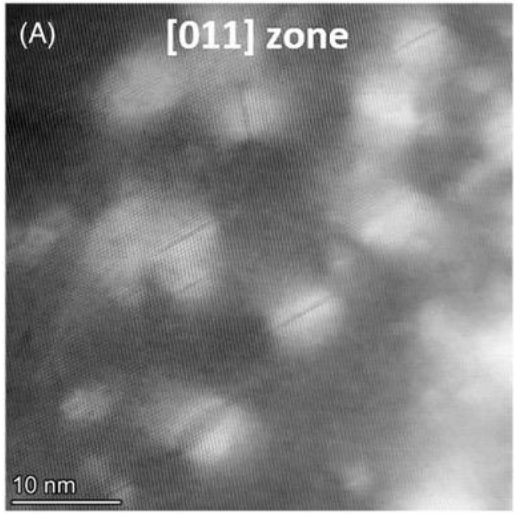
	Experiment (W/m K)	PARFUME (W/m K)	Rochais *et al.*
Matrix	26.2 ± 0.9	39.7	
OPyC	6.7 ± 0.3	4	4.8
SiC	62.1 ± 1.7	61	NA
IPyC	8.4 ± 0.1	4	10.3
Buffer*	6.5 ± 0.1	0.5	7.0
ZrO_2	3.1 ± 0.5		2.3

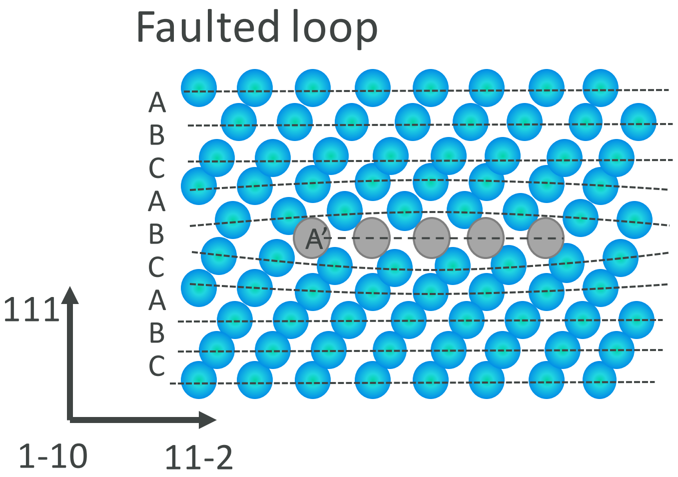
*Bulk, nonporous region
Rochais *et al*. Nucl Engin. Des. 238 (2008) 3047

- TRISO particle is a primary candidate fuel for advanced high temperature reactors and fully ceramic encapsulated ATF
- Conductivity of individual layers important for predicting transient behavior was measured

Khafizov et al., J. Mater. Res. 32, 204 (2017)
Moorehead et al., Materials Today Advances 21, 100455 (2024)

Ion irradiation and PIE (Transmission electron microscopy)



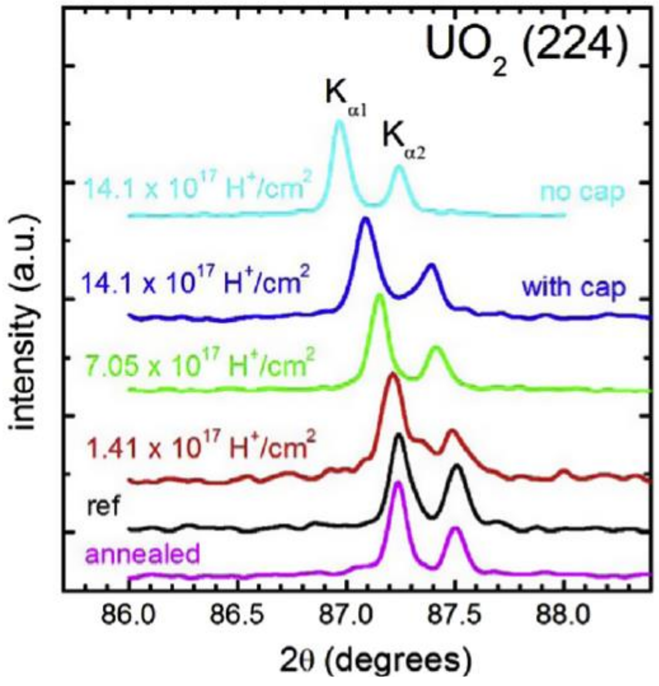


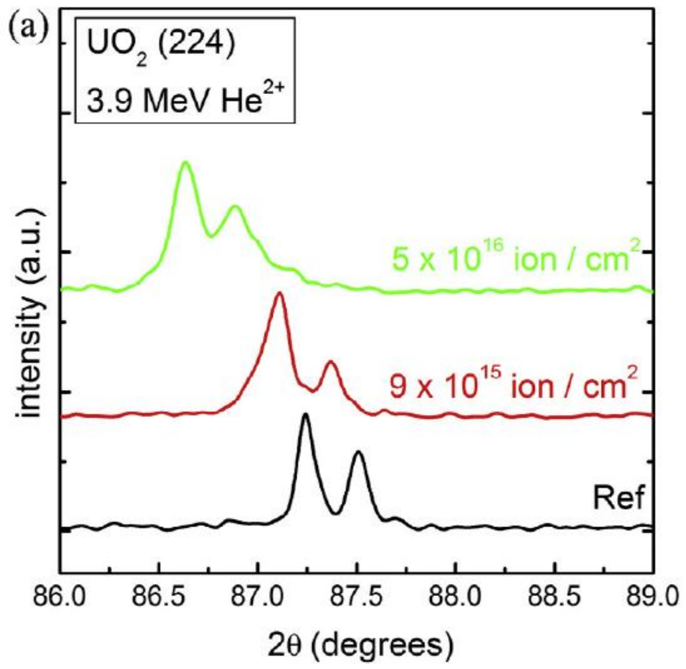
- Ion irradiations for ex-situ characterization are performed using ion beams at Texas A&M and U Wisconsin (2 MeV protons)
- In-situ ion irradiation and TEM were performed at ANL- IVEM (1 MeV Kr ions)
- Access through US DOE Nuclear Science User Facility (NSUF)

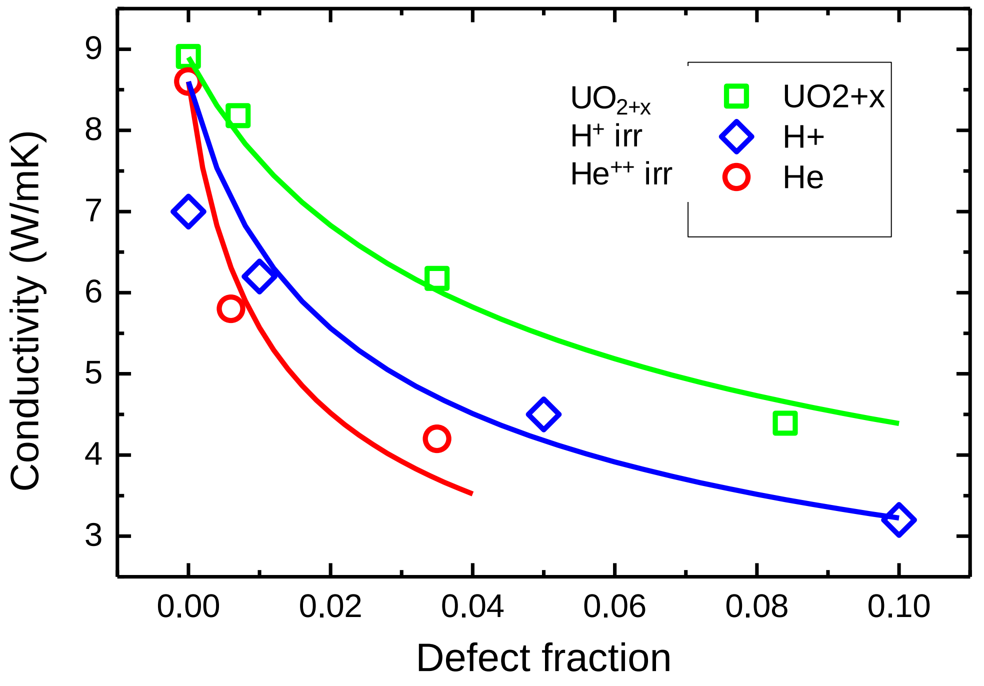
Pakarinen et al., J. Nucl. Mater. 454 ,283 (2014)
Chauhan *et al.*, Materialia 15, 101019 (2021)

Lingfeng He *et al.*, Acta Materialia 208, 116778 (2021)
Dennett *et. al.*, Acta Mater. 213, 116934 (2021)

Impact of point defects on conductivity of UO_2





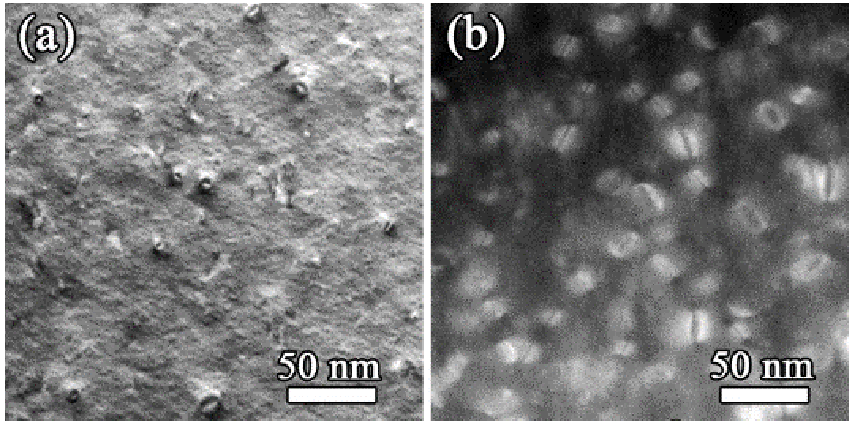


- UO_2 sample have been irradiated at Wisconsin IBL using light ions
- There is correlation between conductivity reduction and lattice constant expansion

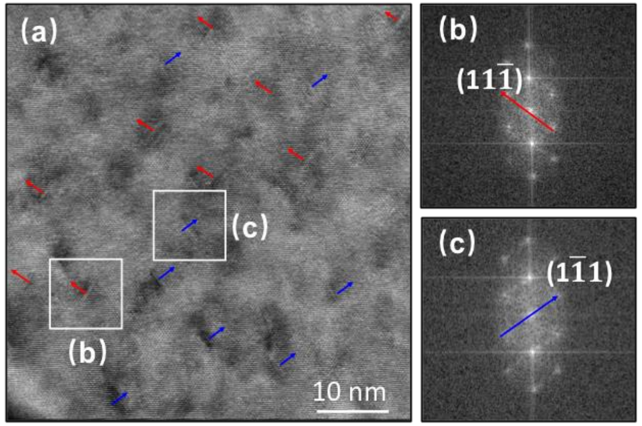
Pakarinen et al., J. Nucl. Mater. 454 (2014) 283
Khafizov *et al.*, Acta Materialia 193, 61 (2020)

Impact of dislocation loops in H^+ irradiated CeO_2

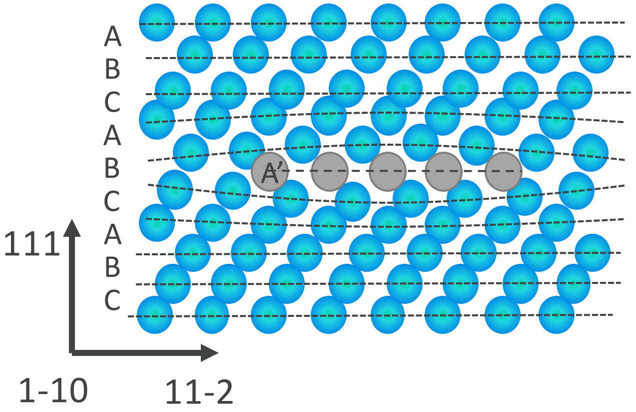
Dislocation loops in bright field TEM



Nature of loops by HRTEM



Faulted loop

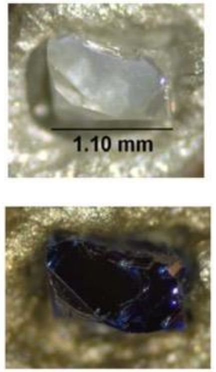


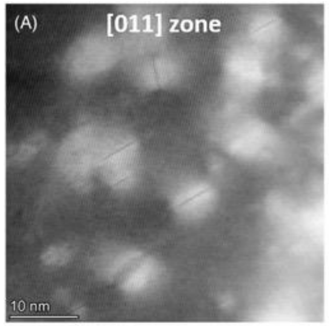
ID	T	Dose (dpa)	Dislocation loops (TEM)		XRD
	(°C)	(dpa)	diameter (nm)	density (10^{22} m^{-3})	(10^4 $\Delta a/a_0$)
LF	600	0.14	3.6 ± 1.0	0.65	7.0
HF	600	0.14	4.0 ± 1.0	1.18	32
HF	700	0.20	7.4 ± 2.2	0.39	3.0

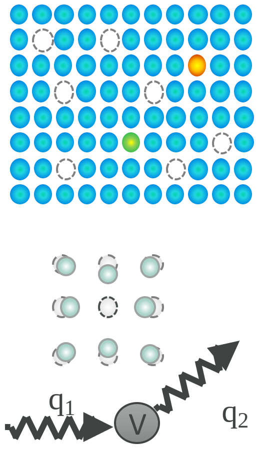
Khafizov *et. al.*, J. Amer. Ceram. Soc . 102, 7533 (2019)
Chauhan *et. al.*, Materialia 15, 101019 (2021)

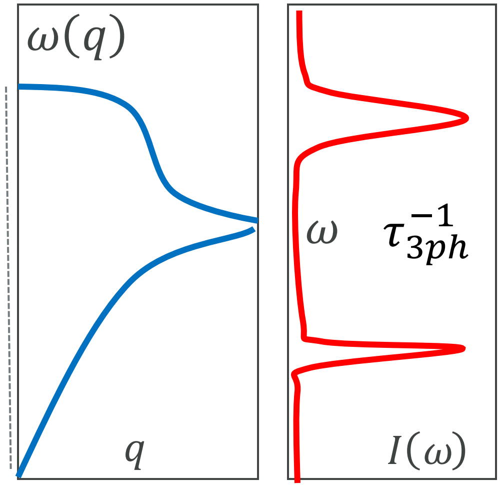
Integrated analysis in proton irradiated ThO_2

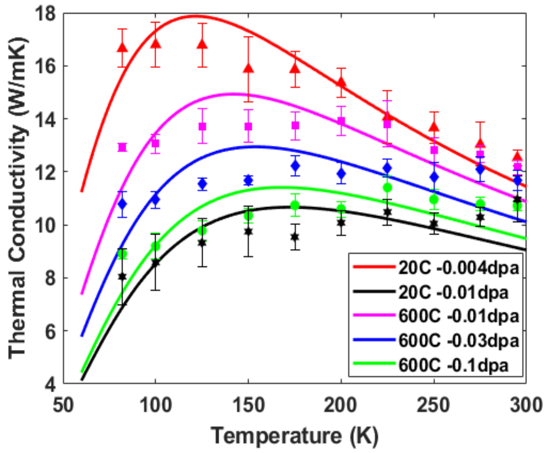
Irradiations at Texas A&M, In-situ Argonne IVEM, PIE INL CAES, INL MFC











Microstructure evolution model

$$\frac{\partial C_i}{\partial t} = p_i + \sum_j R_{ij} C_i C_j$$

$$R_{ij} \propto D_0 e^{-E_m/k_B T}$$

$$\tau_{pd}^{-1}(C_i, S_i^2)$$

$$\tau_{loop}^{-1}(N_L, R_L)$$

$$\tau_k^{-1} = \tau_{3ph}^{-1} + \tau_{pd}^{-1} + \tau_{loop}^{-1}$$

$$\kappa = \frac{1}{V} q \sum C_q v_q^2 \tau_q$$

Experimental characterization

(N_L, R_L) - TEM
$\Delta a/a = C_i(\delta a/a)_i$ - XRD Lattice expansion
$PS = C_i PS_i$ - Positron spectroscopy
$A = C_i A_i$ - Absorption spectra
$P = C_i P_i$ - Photoluminescence spectra

Chauhan *et. al.*, Materialia 15, 101019 (2021)
Dennett *et. al.*, Acta Materialia 213, 116934 (2021)
Deskins *et. al.*, Acta Materialia 241, 118379 (2022)
Lingfeng He et al., J Amer. Ceram. Soc. 105, 5419 (20220

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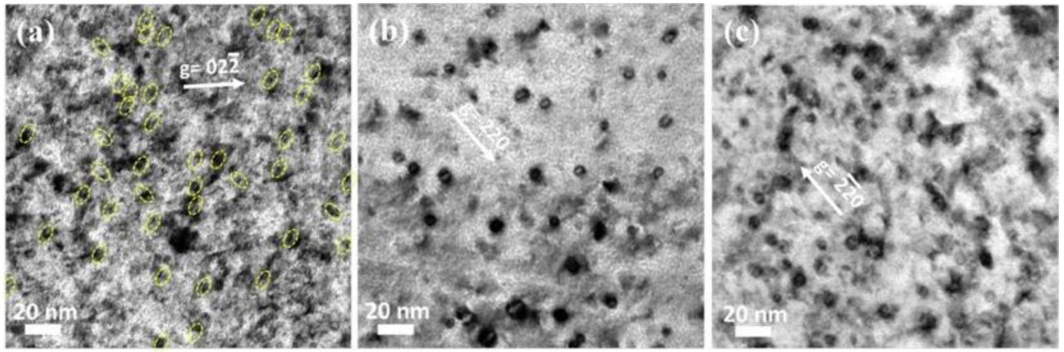
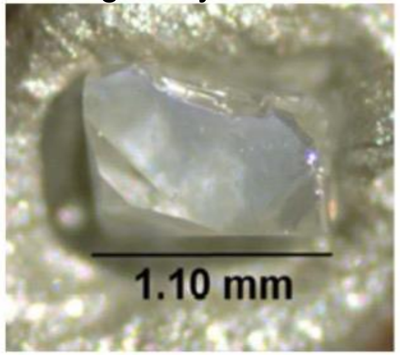
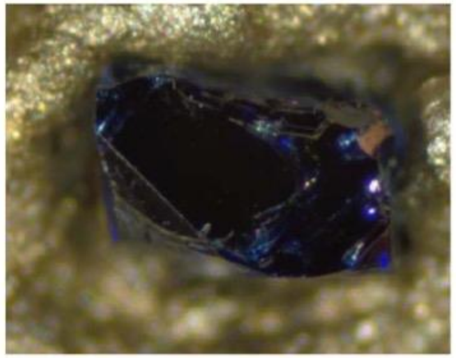
Integrated analysis in proton irradiated ThO_2



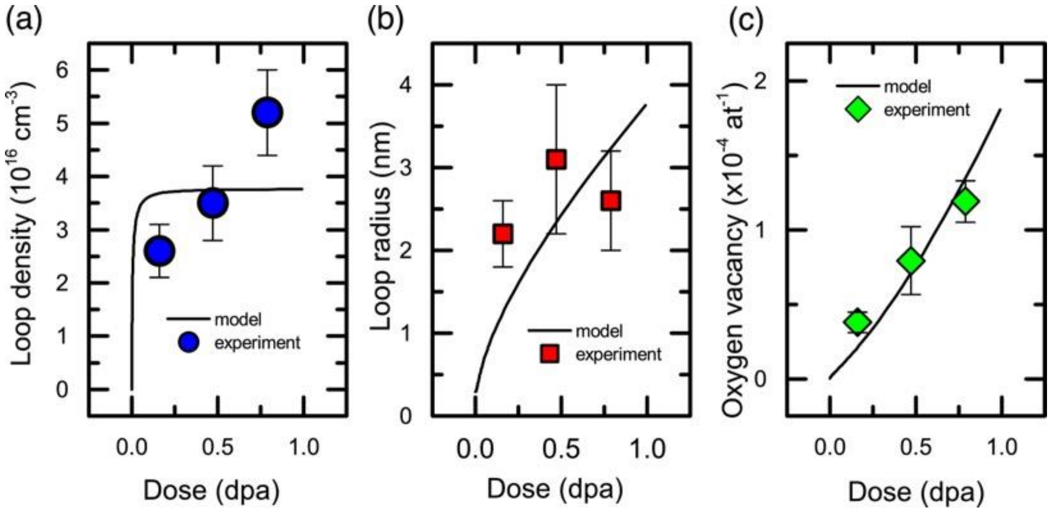
Pristine ThO_2 single crystal

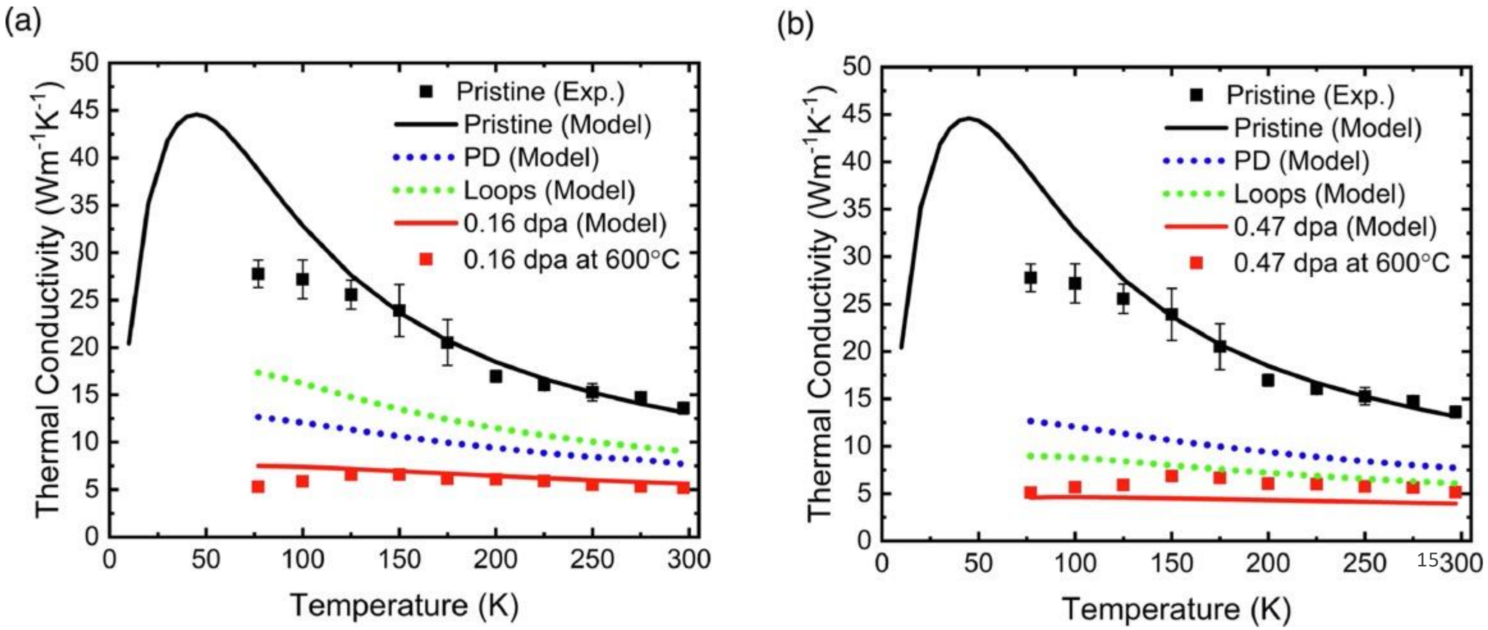


Post-Irradiation with 2 MeV H^+ ions



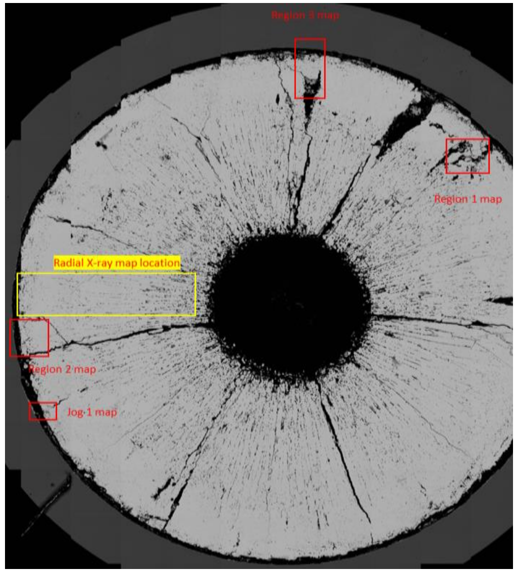
- Hydrothermally grown single crystal ThO_2 irradiated at Texas A&M IBL





Dennett *et. al.*, Acta Materialia 213, 116934 (2021)

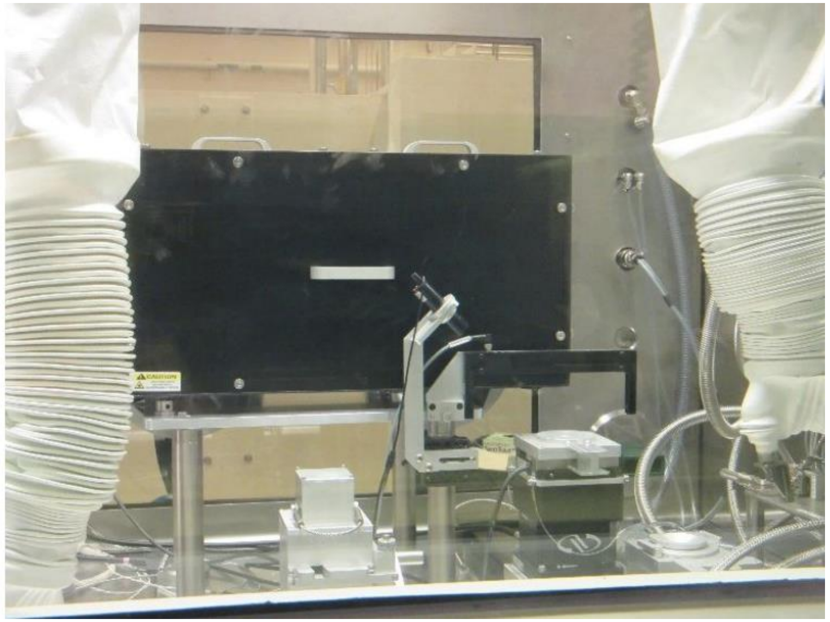
Radial characterization of thermal conductivity for the validation of fuel performance models



6.3% FIMA SFR MOX sample

F. Cappia, et al., INL/EXT-21-61757, 2021

- Characterization of MOX fuel along the radial direction provides a validation of material properties of nuclear fuel performance models.
- Access through DOE NE NSUF RTE led by BYU and Tsveti Pavlov



Thermal conductivity microscope

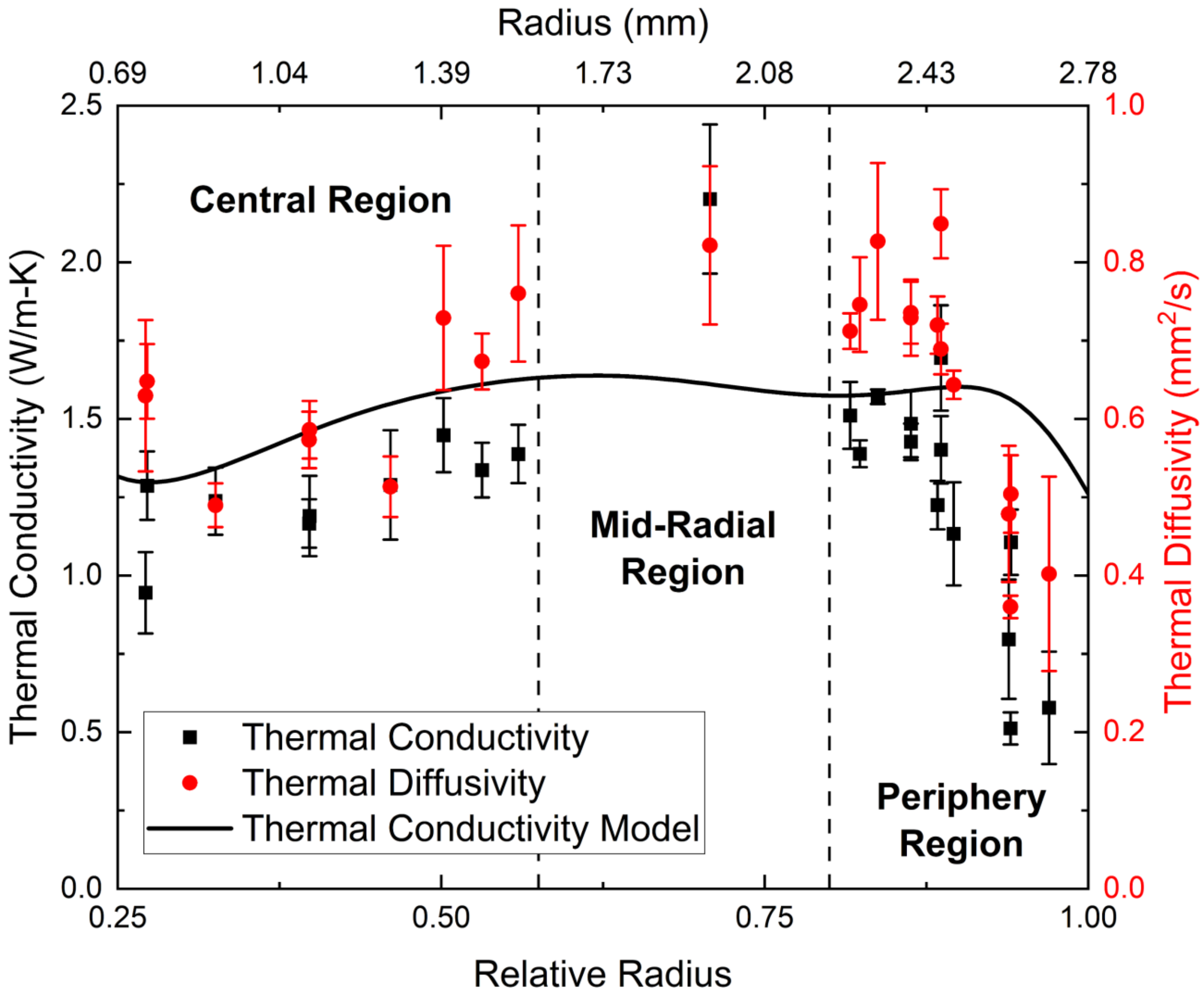
Adkins, Cynthia A, et al.,. INL/EXT-19-55902 (2019)

- Thermoreflectance-based conduction measurement.
- Capable of measuring irradiated nuclear fuels with very high burnup
- Discretized measurements down to 15 μm.
- Allows for validation of radial thermal conductivity models and integrals

Estimating radial thermal conductivity

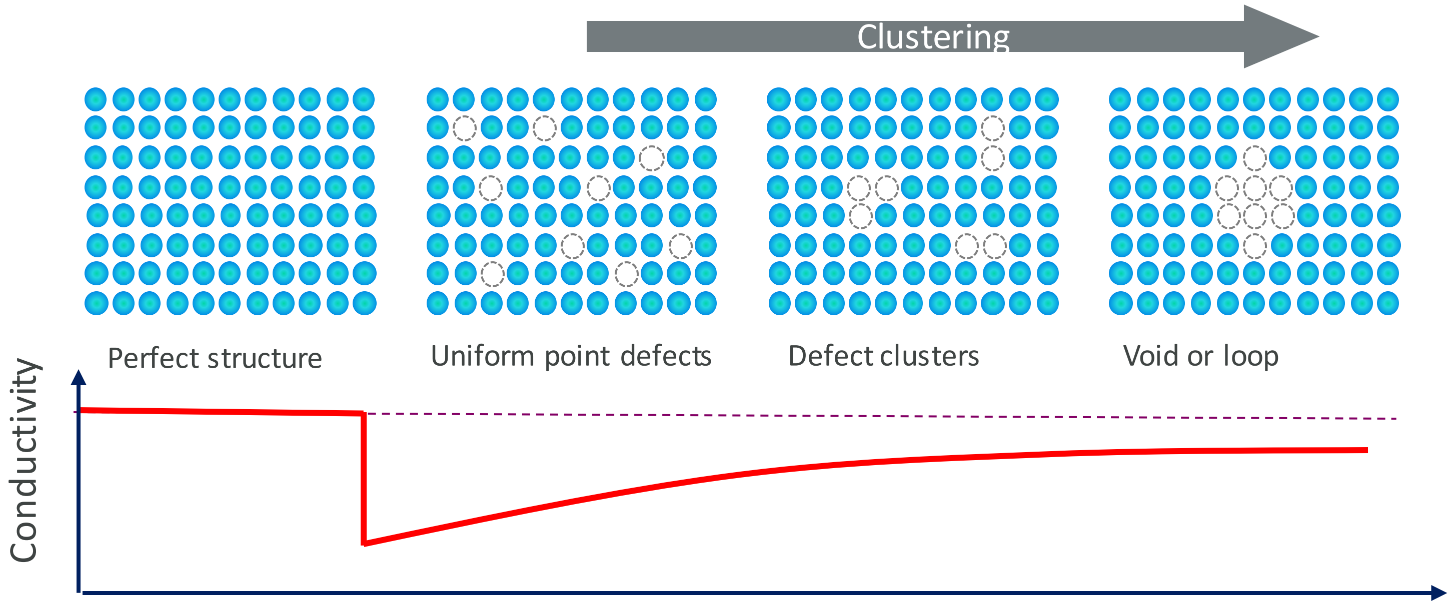
- Comparison to Lucuta-Inoue correlation
- Good agreement in the central region of fuel, but unable to capture the complexity of the high burnup structure.
- Further data needed about the mid-radial regime to adequately describe porosity migration, and lack of Pu enrichment.

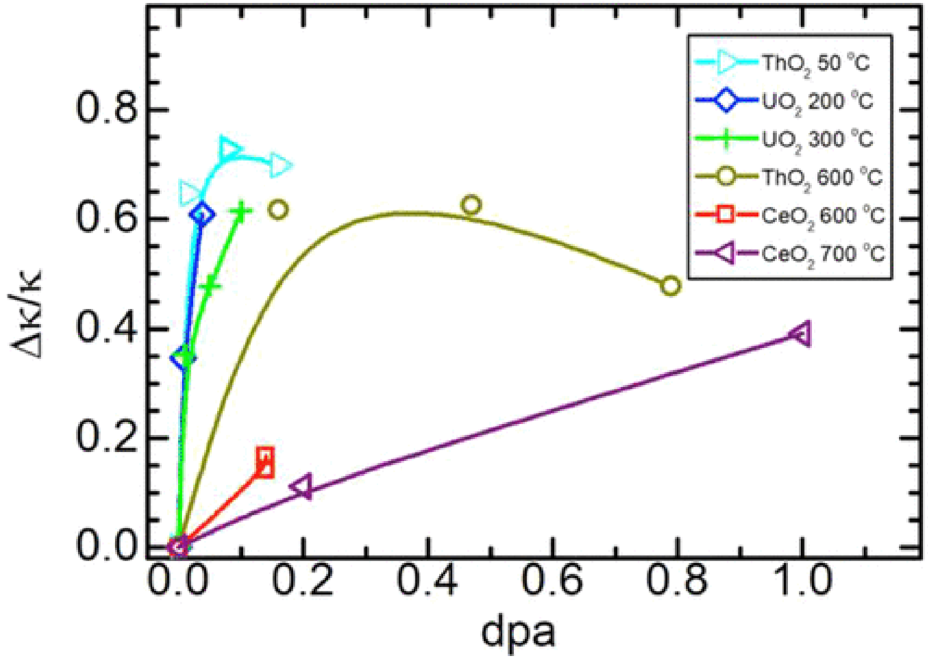
J. Ferrigno, *et al.*, submitted to JNM, 2023



19% FIMA SFR MOX sample

Impact of clustering on thermal conductivity





- In insulating materials, defects lead to reduction in thermal conductivity
- Clustering of point defect leads to recovery of conductivity
- NEMD simulations using empirical potentials support this picture

Hurley *et al.*, Chemical Reviews 122, 3711 (2022)
Miaomiao Jin *et. al.*, J. Nucl. Mater. 566, 153758 (2022);
Dennett *et. al.*, APL Materials 8, 111103 (2020)