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SAND2017-3463C

Preparation and Characterization of UO_2 Epitaxial Thin Films

AML
Advanced Materials Laboratory

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U.S. DEPARTMENT OF ENERGY NNSA

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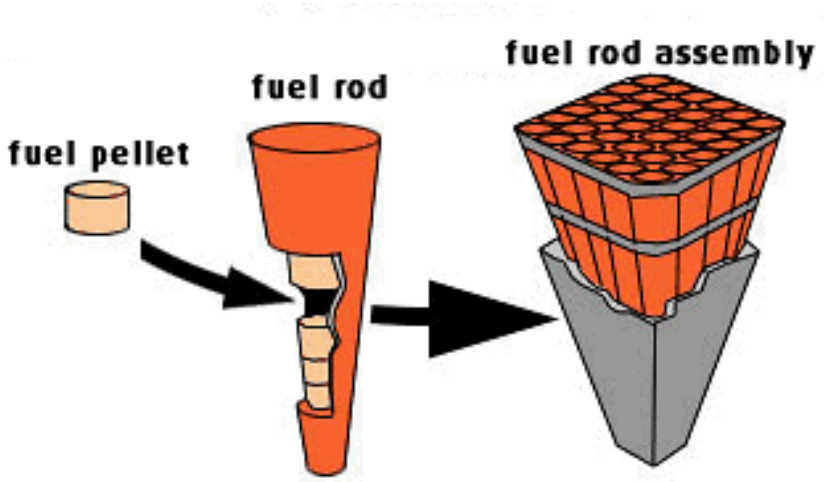
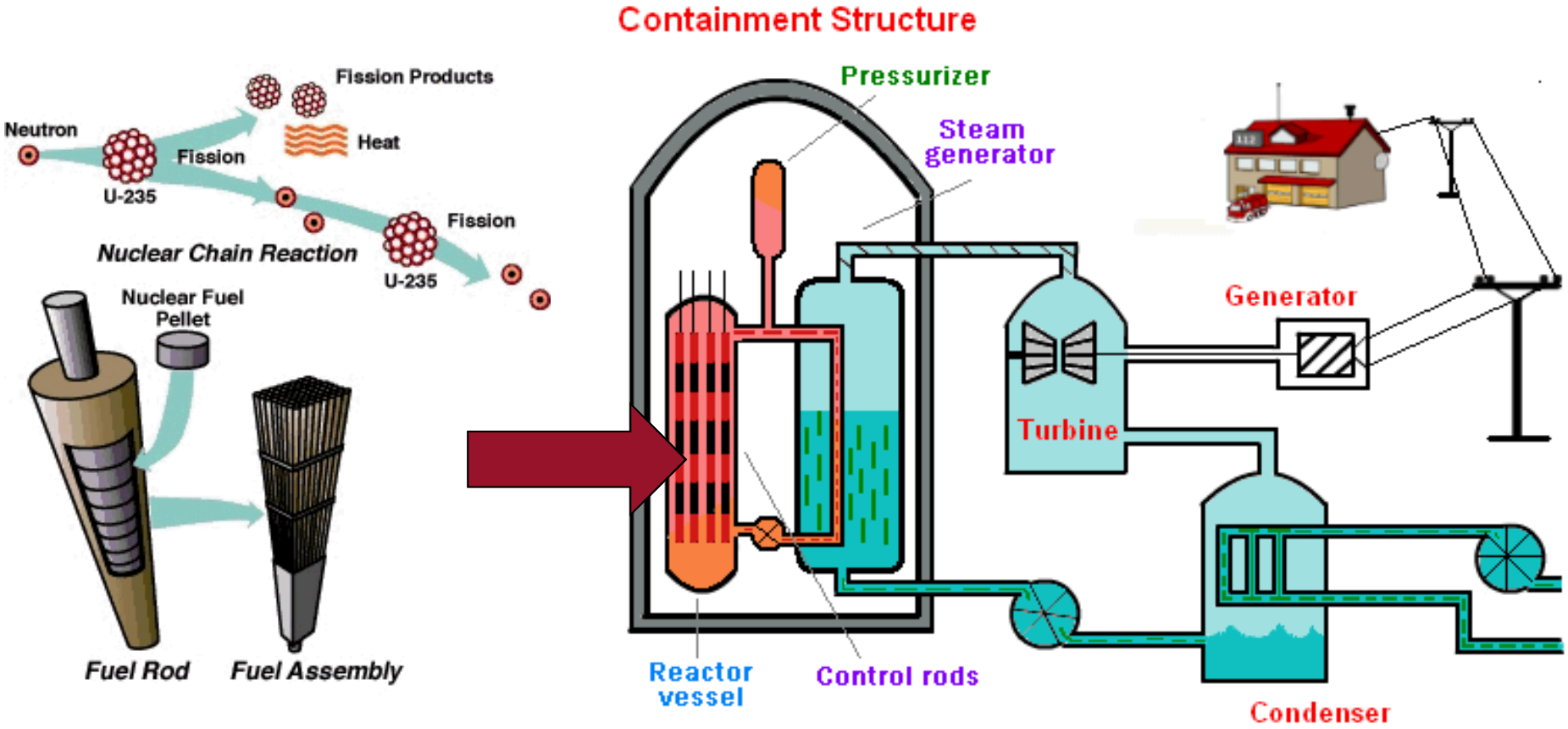
Outline

1. **Recreate surface chemistry**
   - UO_2 -Zircaloy interface (Pellet Cladding Interface)
     - Profilometry – film thickness
     - Transmission Electron Microscopy – microstructure information
     - Powder X-ray Diffraction – phase identification
2. Simulate radiation damage
   - In-situ ion bombardment
   - Compare to INL spent fuel
3. Characterize damage

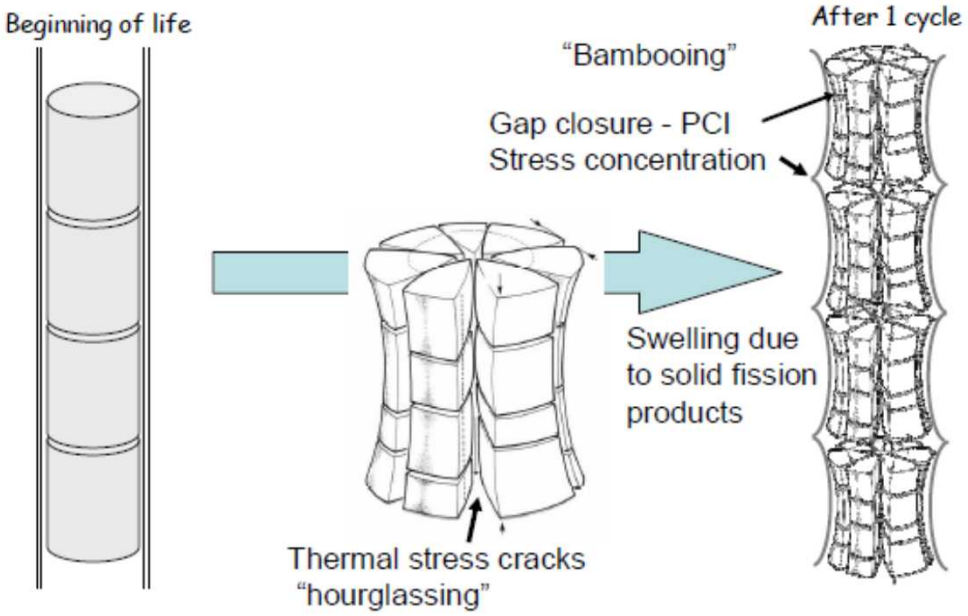


Nuclear Reactor Overview

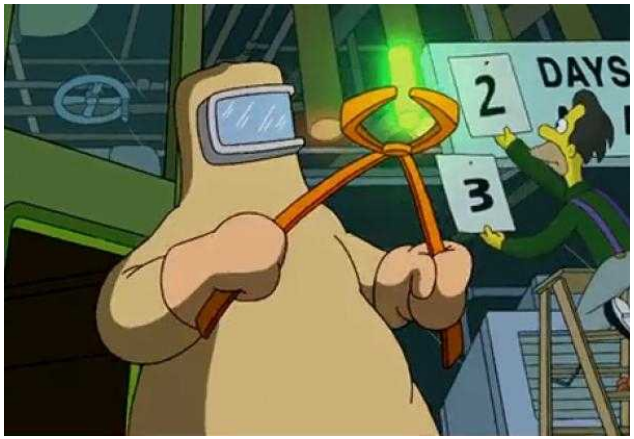


Pressurized Water Reactor

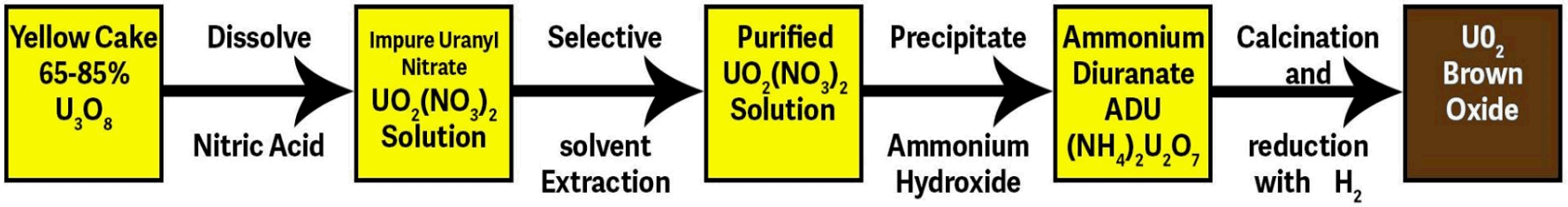
Reactor Chamber

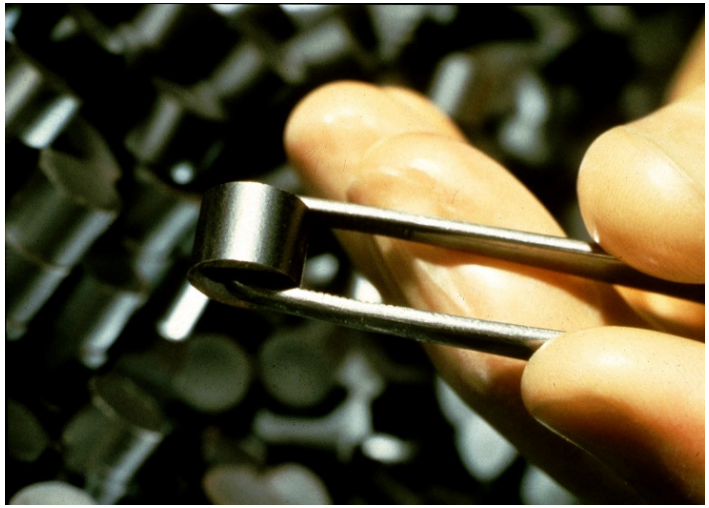


What happens at the microscale?



UO_2 Fabrication





$$UF_6 + 2H_2O \longrightarrow UO_2F_2 + 4HF$$

(gas)(excess steam) (solid)(gas)

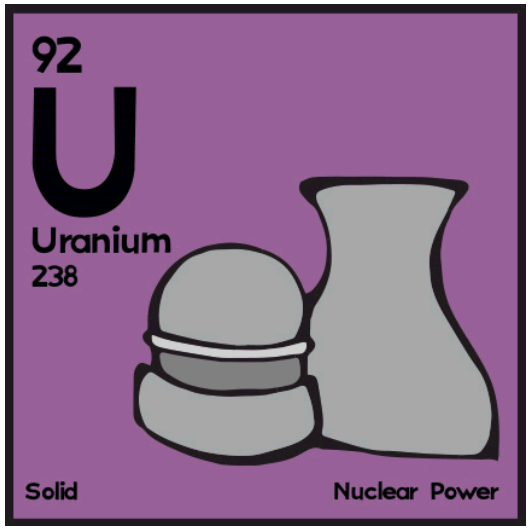
$$UO_2F_2 + \text{(heat)} \longrightarrow UO_2 + 2HF$$

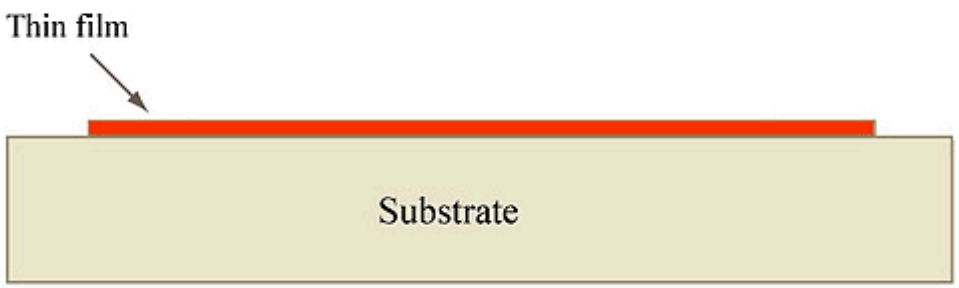
(solid) (powder)(gas)

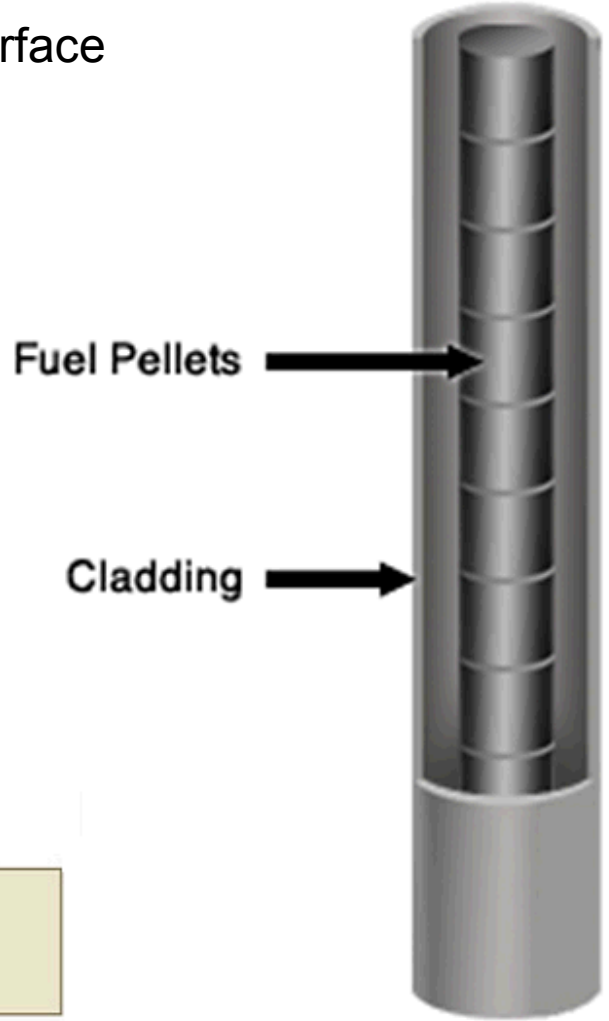
Epitaxial Thin Films

Strategy for studying the pellet cladding interface

$UO_2(NO_3)_2 \cdot 6H_2O$



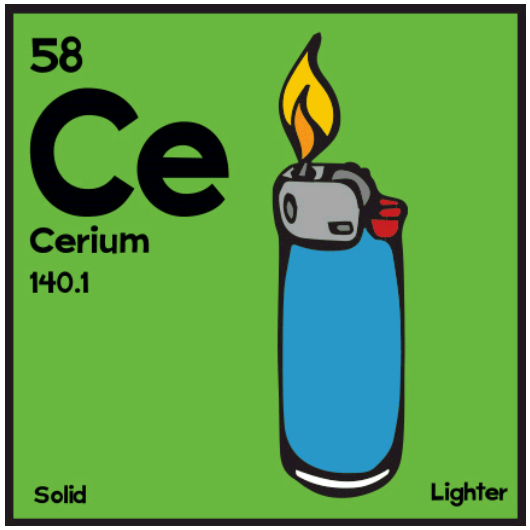


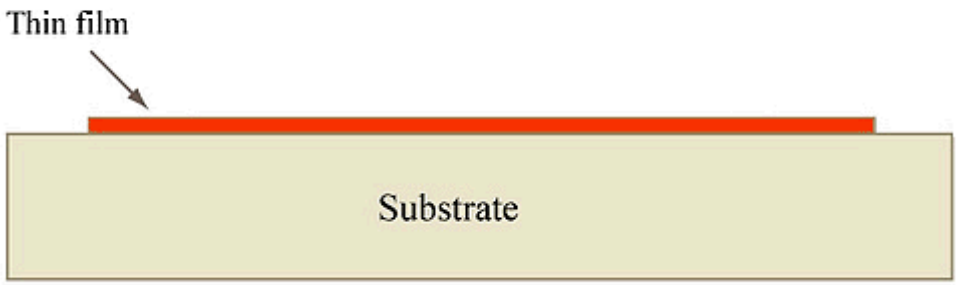


Epitaxial Thin Films

Strategy for studying the pellet cladding interface

$Ce(NO_3)_3 \cdot 6H_2O$





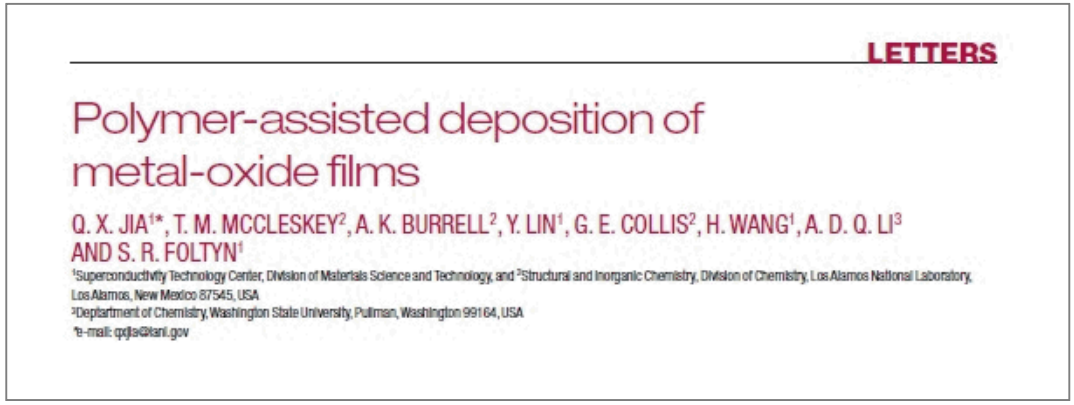
"Surrogate for a Surrogate"

Differences in oxidation states:

Ce(III) → Ce(IV) oxidation

U(IV) → U(IV) reduction

Polymer-Assisted Deposition



LETTERS

Polymer-assisted deposition of metal-oxide films

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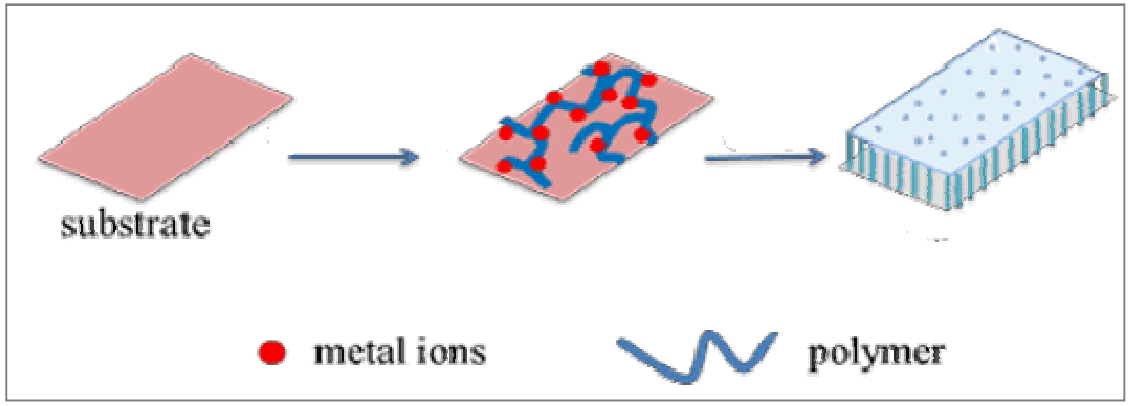
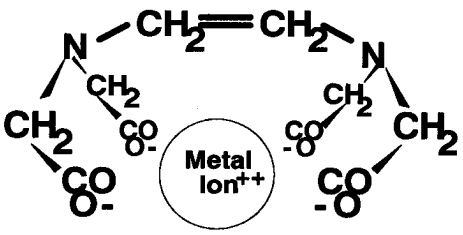
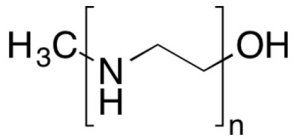


- Viscosity
- Stabilization
- Homogeneity
- Uniform distribution



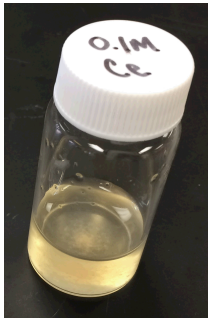
Ethylenediaminetetraacetic acid (EDTA) chelates a metal ion



Polyethylenimine (PEI)

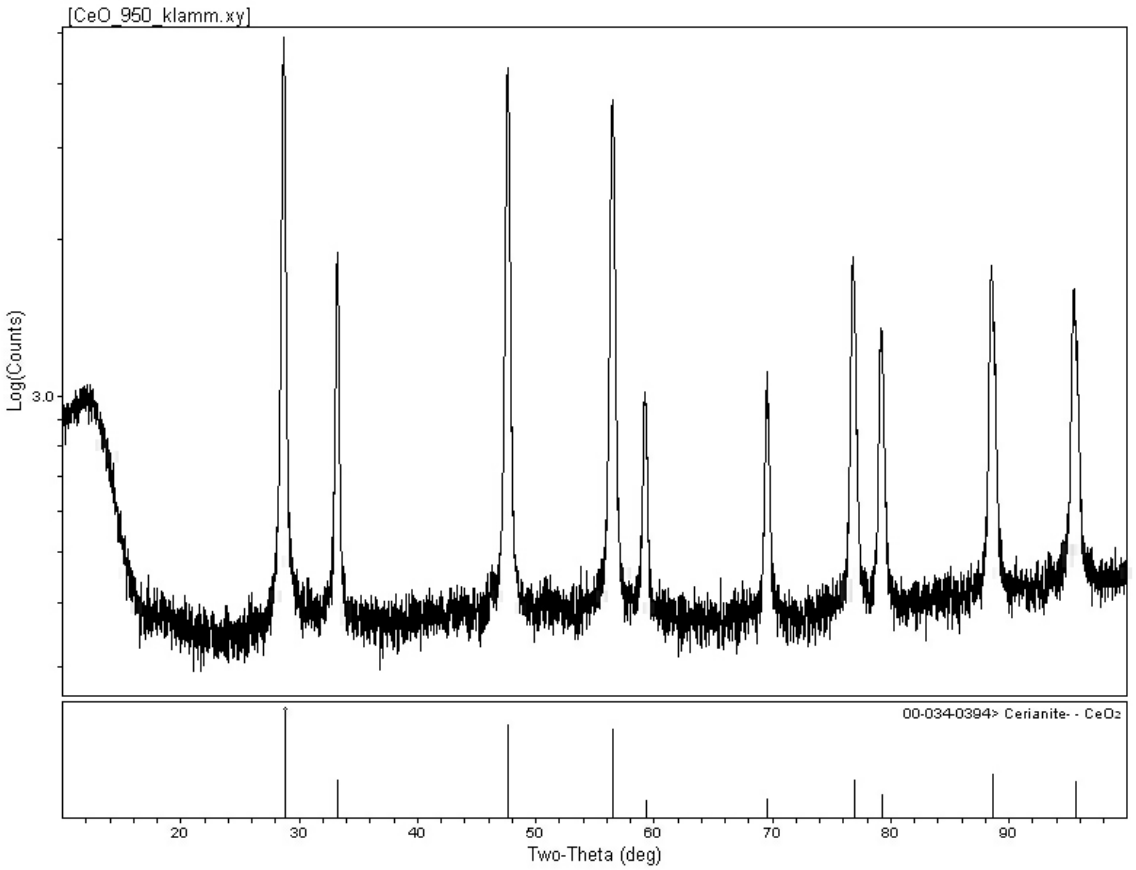
A. Synthesis of bulk $Ce(IV)O_2$

$0.1M\ Ce(NO_3)_3 \cdot 6H_2O$
$0.125M$ EDTA
$0.125M$ PEI



1hr
350C

1hr
1000C



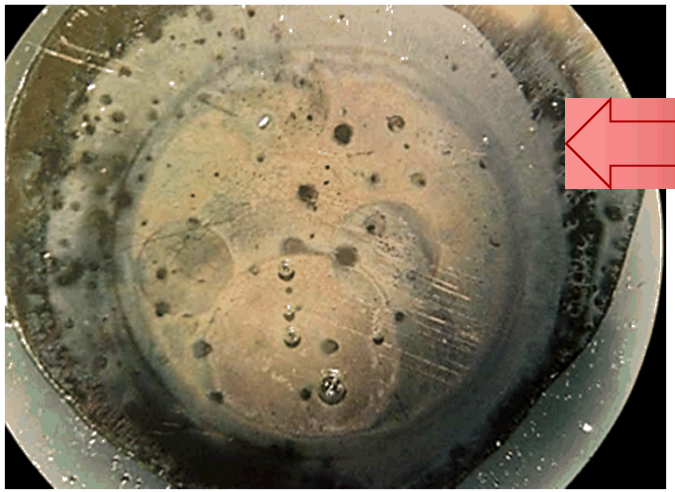
Powder X-ray Diffraction Pattern
$Ce(IV)O_2$

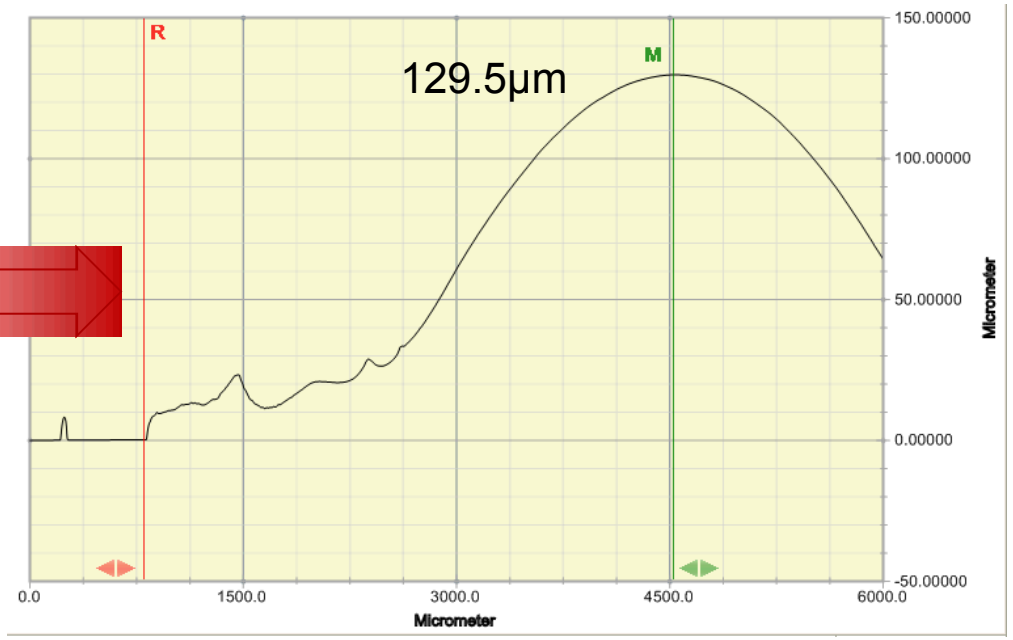
B. CeO_2 Thin Films: Drop Casting

Applying same cerium nitrate solution onto a substrate

- Polymer-Metal ratio 1.25 : 1
- 5-10 layers (drops)
- 200C on hotplate (in air)

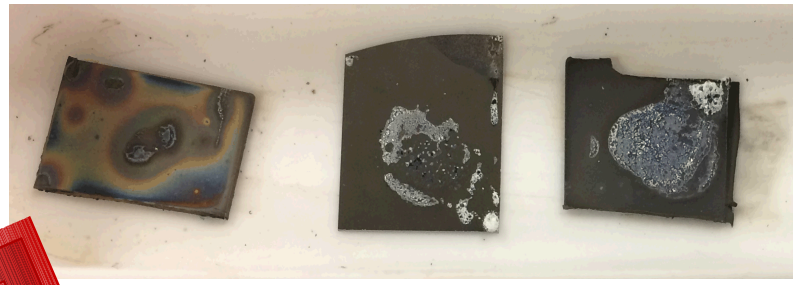
Profile of cerium nitrate on platinized Si



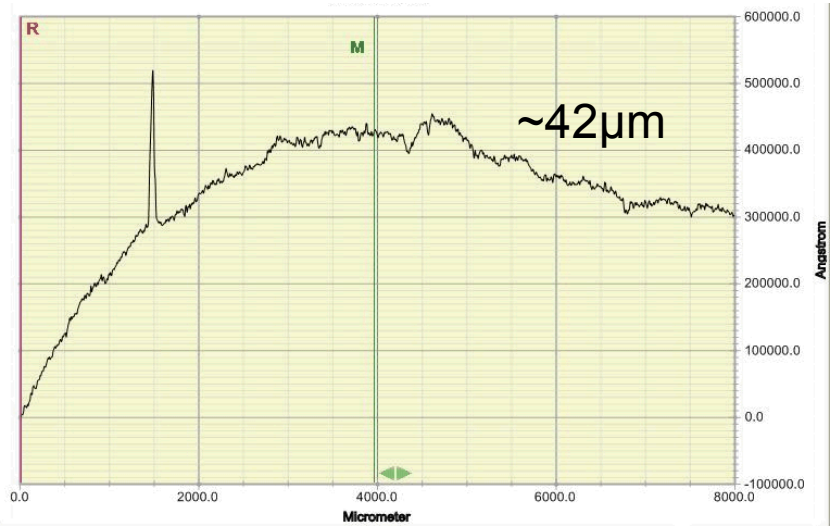


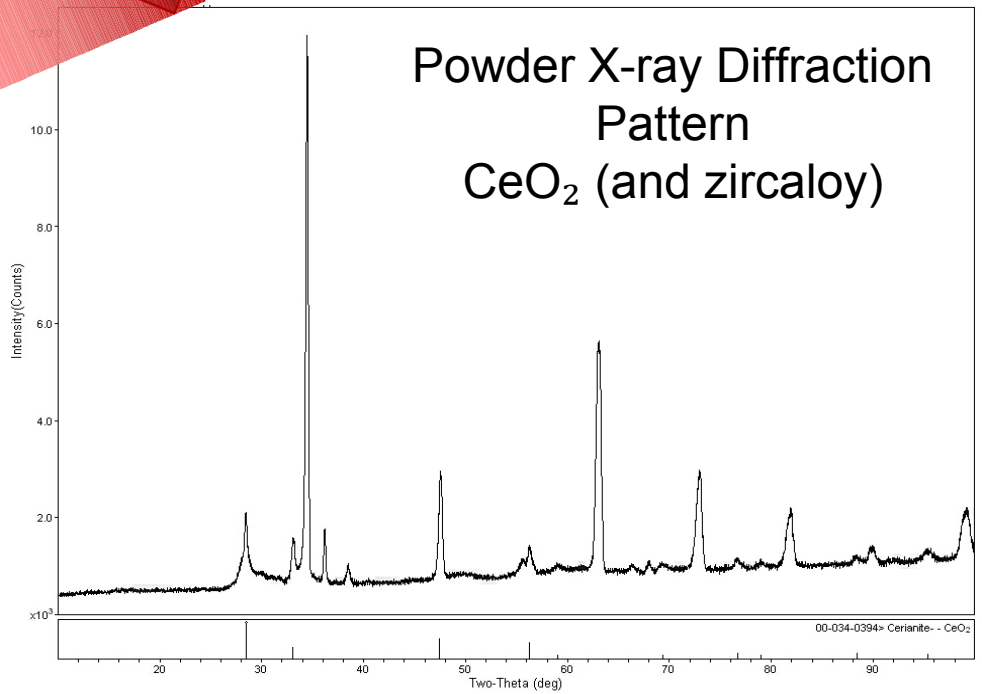
C. CeO_2 Thin Films: Spin Coating

- Polymer-Metal ratio 0.625M:1M
- 4-6 layers (drops)
- Annealed 1000C in Ar



Profile of cerium nitrate on zircaloy





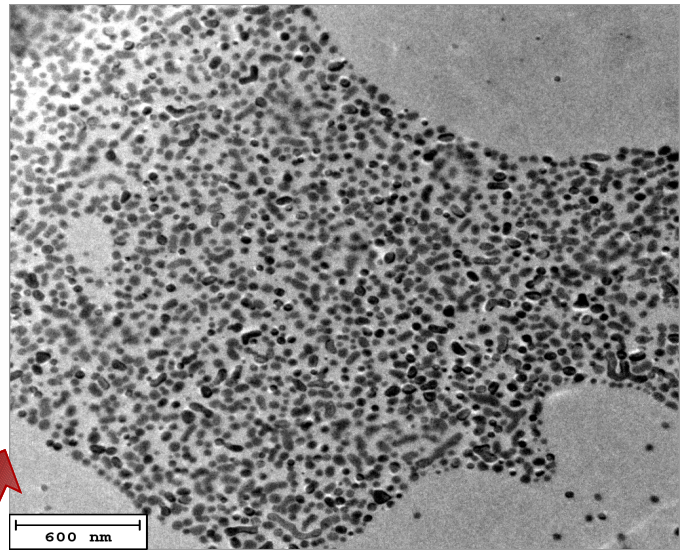
D. Synthesis of $U(IV)O_2$

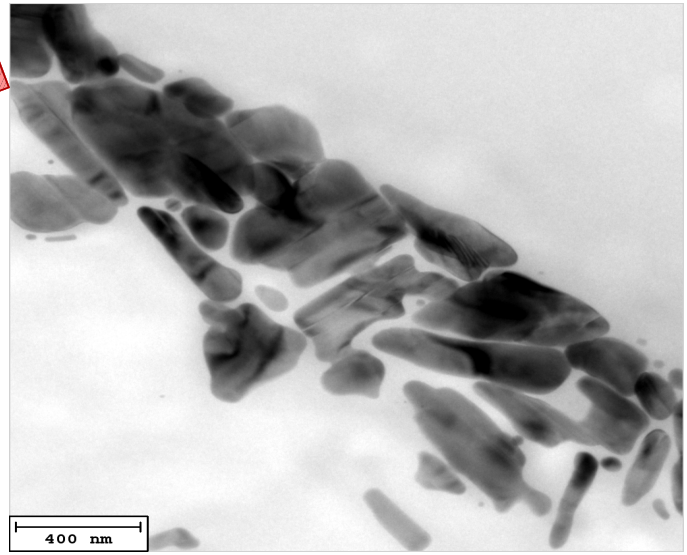
Uranyl nitrate

- Polymer-Metal ratio 0.625M : 1M
- 1000C
- Annealed in Ar and air

TEM on Silicon Nitride Grids

0.1M uranium nitrate annealed in Ar (top) and in air (bottom)





Summary

1. **Recreate Surface Chemistry**
   - Ce(IV) oxidation state as a UO_2 surrogate
   - Deposition & annealing process
   - Characterize

2. Future Steps
   - Further characterization of films
   - Mechanical testing
   - In-situ TEM

Provide a surrogate thin film that will enable us to asses the damage and integrity of nuclear systems at the microscale

Acknowledgments

Samuel Briggs –TEM
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Remi Dingreville –Project PI

Sandia Laboratory Directed Research and Development program for funding

Backup Slides

Substrate

- Platinized Silica
- Zircaloy 4

	Element	Weight%
Zircaloy-2	Sn	1.2-1.7
	Fe	0.07-0.20
	Cr	0.05-0.15
	Ni	0.03-0.08
	Zr	balance
		+ impurities
Zircaloy-4	Sn	1.2-1.7
	Fe	0.18-0.24
	Cr	0.07-0.13
	Zr	balance
		+ impurities

Sources

- Steve Skutnik. A background on spent fuel. Neutron Economy. May 6. https://neutroneconomy.blogspot.com/2012_05_01_archive.html
- Q. X. J, et al. *Polymer-assisted deposition of metal-oxide films*. Nature Materials. Vol 3. August 2004.
- Olander, Donald R. *Nuclear Fuels*. Engineering Journal. Vol 13. Jan. 2009