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Title: Ceramic Fuels Technologies

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Intended for: Presentation at the Fuel Cycle Technology Annual Review Meeting-- Advanced Fuels Campaign session

Argonne National Laboratory
Argonne, IL
November 8-10, 2011

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Presentation Title: Ceramic Fuels Technologies

Meeting: Fuel Cycle Technologies Annual Review Meeting, Argonne, IL, November 8-10, 2011

Summary:

This presentation is effectively a "highlights" presentation for the FY11 progress within the ceramic fuels development technical area of the advanced fuels campaign. The main audience targeted in this talk is DOE-NE headquarters staff (e.g. NE-5) so there is substantial emphasis on program approach, and goals along with the recent highlights. This is one of four 30 minute talks that along with a broader fuels overview by the campaign director that will make up the 2 hour fuels review session. The work included in this presentation will not be covered in detail during the talk. Highlights from all areas are included for the hard copy version but only a subset will be described during the presentation.

The talk consists of both basic and applied R&D on oxide fuel systems to support the fundamental research approach that is being implemented in the FCRD program. The work has the main goal of increasing our fundamental understanding of ceramic fuel systems to 1) enable improved fuel fabrication processes to be developed, and 2) to support improvements in performance of existing fuels and development of advanced fuels. The work (and talk) is structured along the lines of employing a basic materials R&D approach of identifying and understanding the relationships between feedstock, processing, microstructure, properties and performance. The highlights that are presented are derived from work performed at LANL, ORNL, INL, BNL and Arizona State University.

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Fuel Cycle Technologies

Ceramic Fuels Technologies

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Annual Review Meeting
Argonne National Laboratory
November 8-10, 2011

LA-UR-11-XXXXX

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Key: Ceramic Fuels Irradiation Testing Post Irradiation Examination Analytical Support NEAMS

Outline

- Approach and structure for Ceramic-based Fuel Development Technical Area
- Mission, goals and objectives (Fuel Fabrication and Performance)
- FY11 structure and work scope
- Examples of progress and highlights
- FY12 scope and objectives

DOE-NE R&D Roadmap and Ceramic Fuels

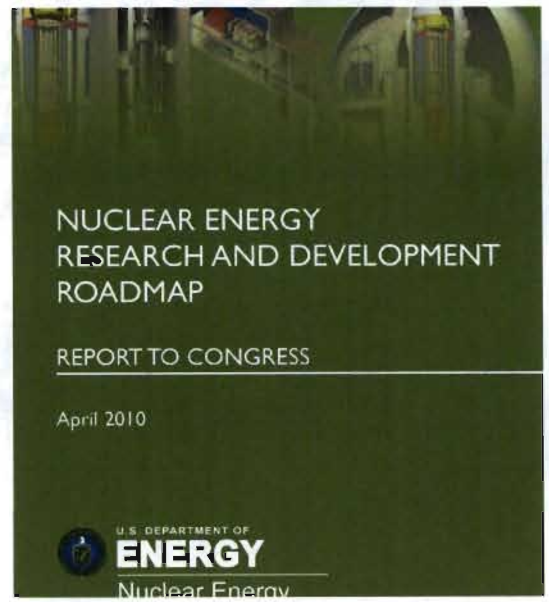
Nuclear Energy R&D Objectives

R&D OBJECTIVE 1: Develop technologies and other solutions that can improve the reliability, sustain the safety, and extend the life of current reactors

R&D OBJECTIVE 2: Develop improvements in the affordability of new reactors to enable nuclear energy to help meet the Administration's energy security and climate change goals

R&D OBJECTIVE 3: Develop Sustainable Nuclear Fuel Cycles

R&D OBJECTIVE 4: Understand and minimize the risks of nuclear proliferation and terrorism



http://www.ne.doe.gov/pdfFiles/NuclearEnergy_Roadmap_Final.pdf

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Ceramic Fuels Technical Area Structure and Approach

- Advanced Fuels Campaign
  - Ceramic Fuels
    - UO2-based Fuels
    - ThO2-based Fuels
    - Alternative Ceramic Fuel Systems
  - Metallic Fuels
  - Coated particle Fuels
  - Cladding & Structural Materials
  - Analytical Support

- **Each fuel type has fabrication and performance areas**
- **Focus on increasing fundamental understanding of phenomena governing behavior to enable revolutionary advances but structure for data sets that support concurrent evolutionary advances**
- **Experimental efforts emphasize Separate Effects approach but Integral Effects tests (ongoing and new) conducted in parallel**
- **Separate effects testing and characterization targeted to specific, near-term data needs for current model development and validation**

Separate Effects Test Focus Areas (Modules)

Module	Primary Variables
Thermal Transport	T, O/M, μ-structure*, impurity, burnup, phase content
Fission Gas Behavior	T, O/M, Conc., μ-structure, burnup
Thermo-chemical	O/M, T gradient, starting composition
Microstructural Evolution	O/M, μ-structure, T gradient, burnup, composition
Thermo-mechanical	O/M, μ-structure, T gradient, burnup, composition
Fission Product Behavior	O/M, μ-structure, T gradient, burnup, phase content

These focus areas roughly map to "modules" in an Integrated Performance and Safety Code

These "modules" address key fuel development (optimization or design) issues of FCCI and FCMI

Even these "modules" are interdependent; there is another level of complexity before a full up integrated performance code

**Note: Microstructure = fcn (grain size, porosity, texture, etc)*

Key 5-Year Ceramic Fuel Goals

- **Fuel Fabrication**

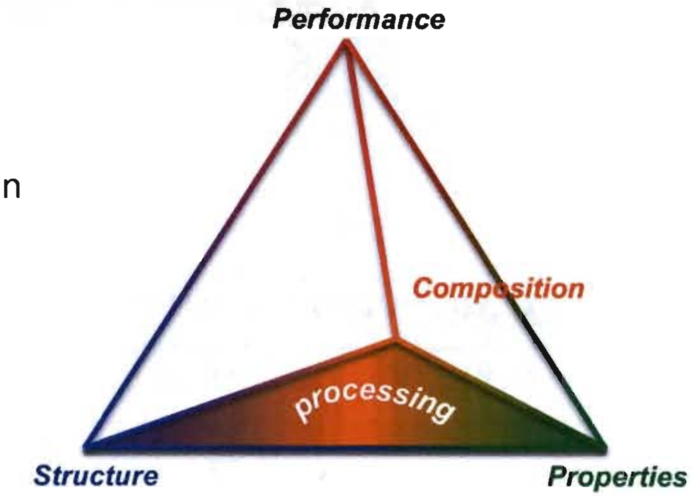
  Goal: *"Turning the (black) art of ceramic fuel fabrication to science."*

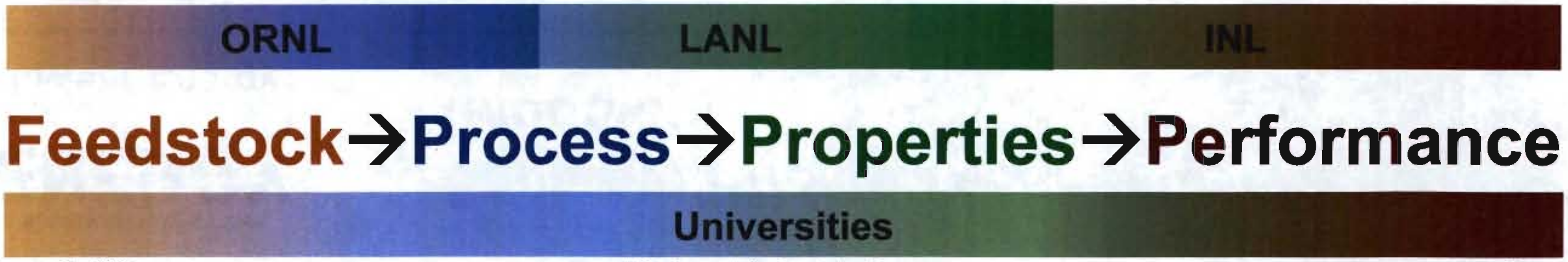
  Demonstrate advanced fabrication techniques that enable production of ceramic fuel pellets with substantial reduction in process losses, increased efficiency and greater reproducibility.

- **Fuel Performance and Safety**

  Goal: *"Improved operation efficiency and safety through mechanistic thermal conduction model."*

  Establish the ability to predict with high fidelity the thermal profile in ceramic fuel pellets (conventional UO_2 and advanced designs) through improved fundamental understanding of the contributions of chemistry, microstructure and in-service damage that govern thermal conductivity.





Nominal SET schedule* showing basic prioritization by focus area

Focus Area	Year 1	Year 2	Year 3
Thermal Transport	Benchtop → Rabbit 1 →	Rabbit 2 →	Instrumented 1 →
Fission Gas Behavior	Benchtop →	Rabbit 1 →	
Thermo-chemical	Benchtop →	→	Rabbit 1 →
Microstructural Evolution	Benchtop →	Rabbit 1 →	Instrumented 1 →
Thermo-mechanical	Benchtop →	→	Rabbit 1 →
Fission Product Behavior	Benchtop →	→	→

- *Schedule limited by capabilities for advanced characterization on actinides but primarily by advanced PIE capabilities for SET samples after even short irradiations*
- *Prioritization includes experimental considerations and ability to support model development and validation*

**Note: Integral effects testing happens in parallel*

Thermal Transport: UO_2 Thermal Conductivity

$$k_{uo_{2\pm x}} = \left[\frac{1}{A(Bu,x,Gd,T\ldots) + B(Bu,x,Gd,T\ldots)\cdot T} + C(x,Gd,Bu)\exp^{D(x,Gd,Bu)\cdot T} \right] \cdot F(\rho)$$

Phonon-Defect Scattering — Phonon-Phonon Scattering — Polaron-Defect Generation

Semi-empirical models consider the effects of temperature, porosity, burnup, and burnable absorber
→ Extend/generalize models

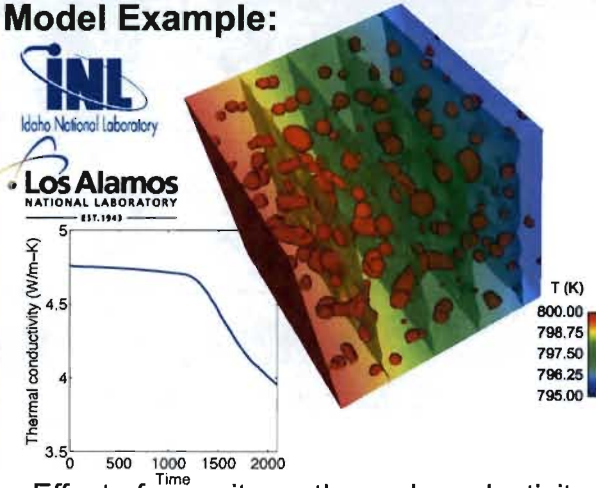
$$\kappa_{UO_{2\pm x}} = \kappa_0(T)\cdot\kappa_1(\beta)\cdot\kappa_2(p)\cdot\kappa_3(x)\cdot\kappa_4(r)$$

Baseline — Burnup "chemistry" — Porosity — Stoichiometry — Burnup "damage"

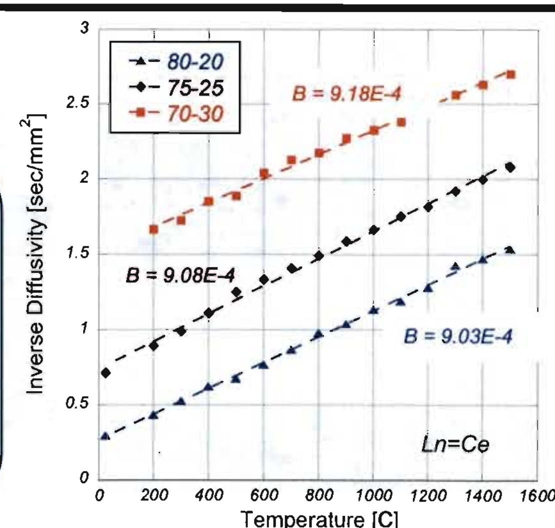
***Separate Effects Tests** (starting with dUO_2)*

- *Benchtop: Controlled microstructures, stoichiometry, additives/ impurities, fission gas(see FG module)*
- *Rabbit: in-pile thermal effects and low damage*
- *Emphasis:*
  - *Data sets for lower length scale models → get the physics right*
  - *Reduce uncertainty in existing models*

Model Example:



Effect of porosity on thermal conductivity



Fission Gas Behavior: Xenon segregation and transport

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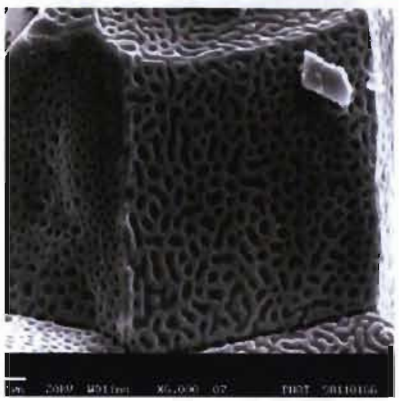

Model Example:

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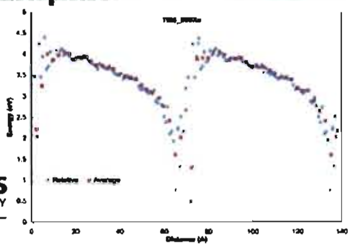
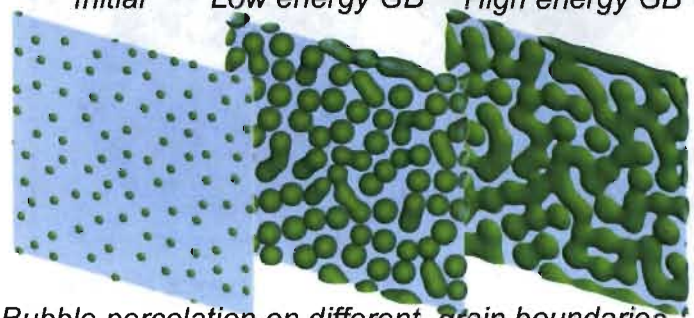
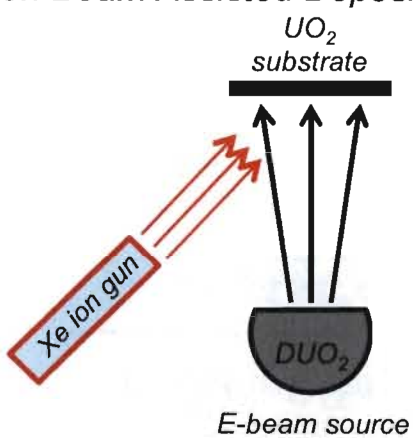


Atomistic simulations of Xe GB segregation inform mesoscale percolation simulations



Bubble percolation on different grain boundaries

*Ion Beam Assisted Deposition**



**IBAD method enables FG in solution with little damage and microstructural control*

Thermal treatments of thick films
Compare and contrast in-situ data from micro-SIMS/TEM for reference and rabbit samples

Separate Effects Tests *(starting with dUO_2)*

Benchtop: Controlled microstructures (single crystal, bi-crystal, HBS), O/M
Rabbit: in-pile thermal effects and low damage
Emphasis: Data sets for atomistic and meso-scale models

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Thermochemistry

Literature and targeted experiments provide data for thermochemical values and models

U-Pu-O System (relatively complete)

5-metal phases Mo-Pd-Rh-Ru-Tc (relatively complete)

Single Component FP oxides Ba,Sr,Cs,Rb,Zr,Mo (available)

Iodides (Rb,Cs)-I (relatively complete)

Single component MA oxides Am, Np, Cm

Rare earth oxides – A_2O_3- Y,La,Ce,Nd,Dy, Gd,Eu,Pr

Perovskite – ABO_3 Ba,Sr,Cs,Zr,Mo

Rock salt – AO Ba,Sr

Secondary metal phases

Oxide fuel phase fluorite structure U-Pu-Ma

Oxide fuel phase fluorite structure with rare earths

Perovskite – ABO_3 addtion of rare earths

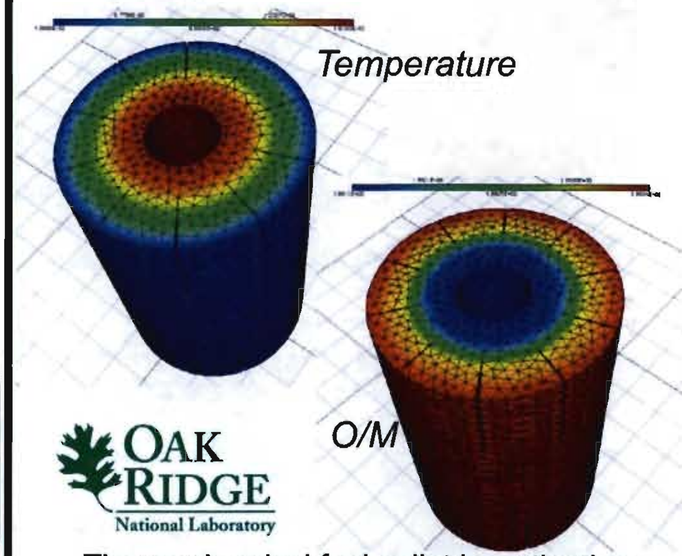
(Rb,Cs) and (Ba,Sr) uranates, zirconates, molybdates

Indicates solution model will be built up from binary and ternary oxides subsystems

Increasingly accurate database/simulation

- *Good models exist for UO_2 and MOX*
- *Employ CEF approach*
- *Focus on U-Ln-O systems coordinated with atomistic models*
- *Using co-converted feedstock for solid solutions*
- *In-situ test to validate thermochemical models as fcn of T*

Model Example:



Thermochemical fuel pellet investigation

Separate Effects Tests *(UO_2-based pseudo binary systems)*

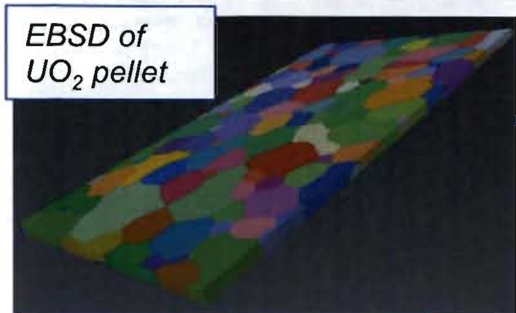
Benchtop: thermodynamics and kinetics

Rabbit: lower priority, in-pile thermal effects and low damage

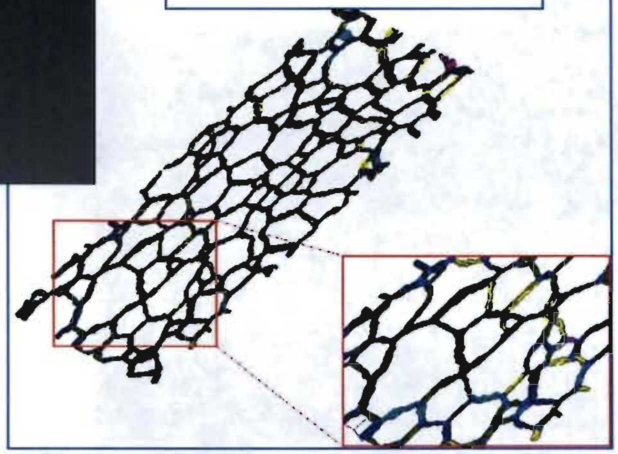
Emphasis: data sets for processing and atomistic models, and continuum model validation

Microstructural Evolution: 3D and statistical data sets

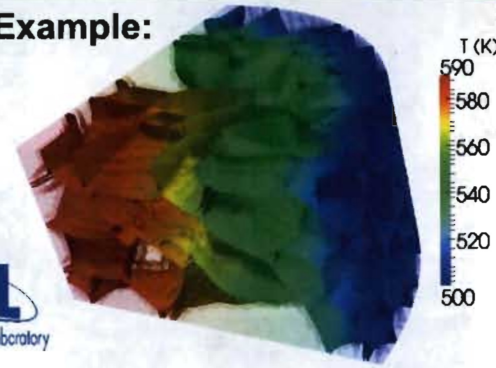
EBSD of UO_2 pellet



Map of boundary types from EBSD data

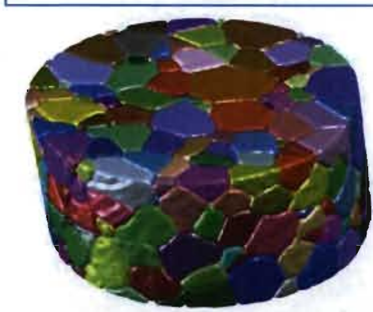


Model Example:

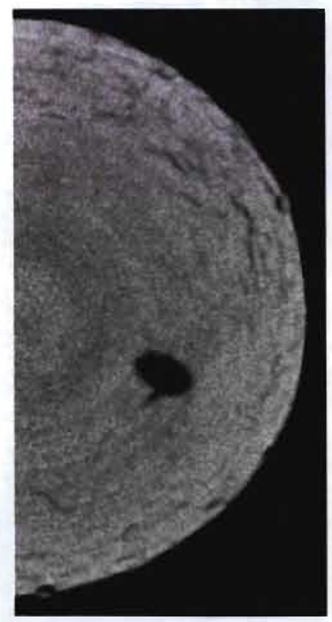


Grain growth in a temperature gradient

HEDM of metal compact



Advanced in–situ measurements, tomography or HEDM, can be used for at least urania samples in the near term



High energy x-ray tomography image of void in SS clad UO_2

APS, 200-250keV, "pink" beam, 6μm resolution 5mm pellet

***Separate Effects Tests** (starting with dUO_2)*
- *Benchtop: Post mortem and in-situ*
- *Rabbit: in-pile thermal effects and low damage*
- *Emphasis: 3D and statistical data sets*

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Thermomechanics: creep and basic properties

- *Improve creep data sets for single crystals and specific boundaries*
- *Obtain basic thermophysical properties via RUS, dilatometry*
- *In-situ testing at neutron scattering facilities*

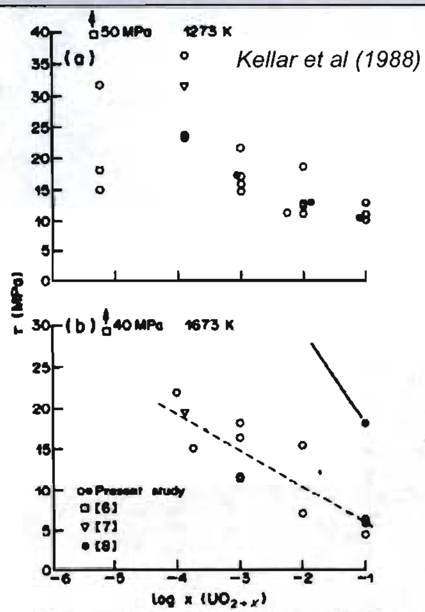
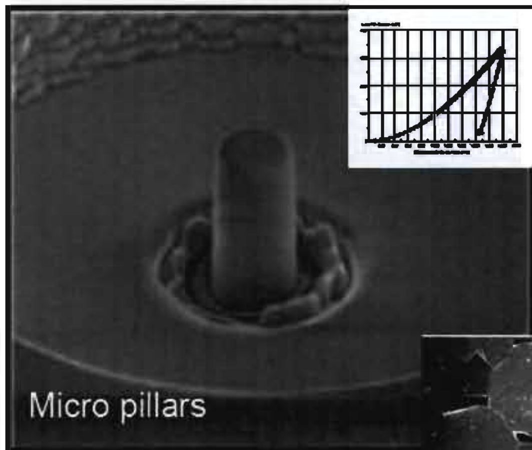
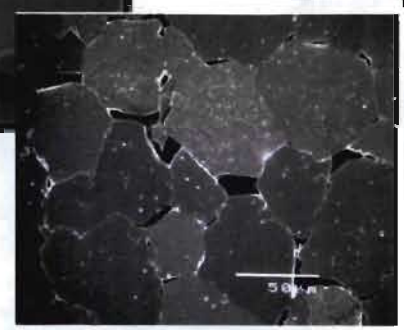
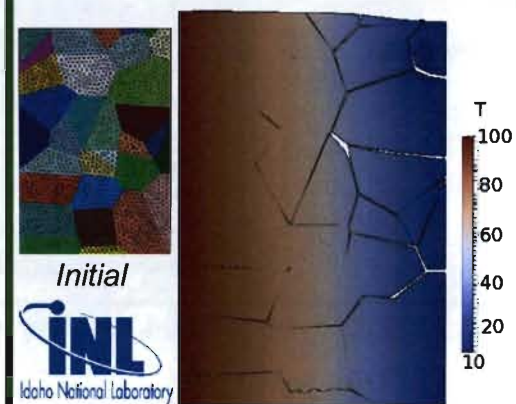


Fig. 6. Yield stresses at 0.4% shear strain vs O/U ratio for specimens deformed at (a) 1273 K and (b) 1673 K which deformed either by {111} glide or noncrystallographic {111}-{110} glide.





Model Example:



Intergranular fracture investigation

Incorporate RUS/NEWS

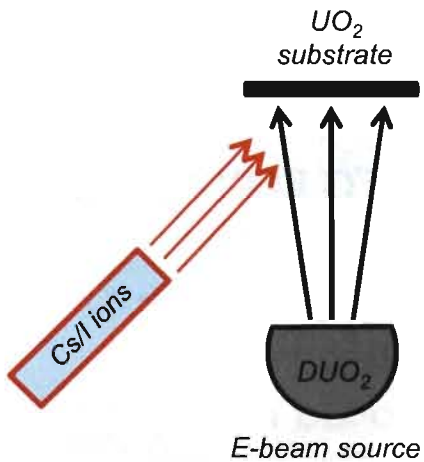
Pellet reconstruction

Separate Effects Tests *(starting with dUO_2)*

Benchtop: microstructure and O/M (bulk and micro/nano testing)
Rabbit: thermal & impurity effects and low damage
Emphasis: reduce uncertainty in baseline properties

Fission Product/Actinide Behavior: Cs/I segregation and transport

*Ion Beam Assisted Deposition**

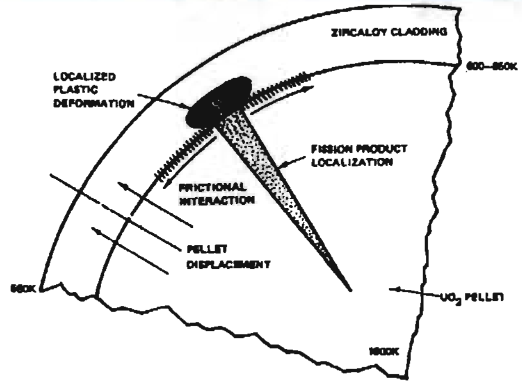


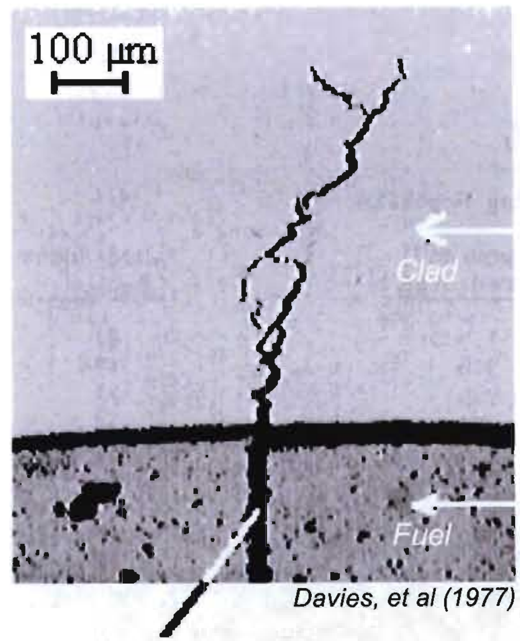
**IBAD method enables FPs in solution with little damage and microstructural control*

- *Analogous to/leverage FG studies*
- *Thermal treatments of thick films*
- *Compare and contrast in-situ data from micro-SIMS/TEM for reference and rabbit samples*
- *Source term for PCI-SCC*

Separate Effects Tests *(starting with dUO_2)*

Benchtop: Controlled microstructures, O/M
Rabbit: in-pile thermal effects and low damage
Emphasis: data sets for atomistic and meso-scale models





Davies, et al (1977)

UO_2 pellet cracking

FY11 Ceramic Fuels Effort Summary

- **U-based fuel design, development & testing (Int' l)**
  - Feedstock, process/structure/property, rabbit test
- **Ceramic Process Modeling: Sintering**
  - MSC and MC
- **MOX fuel processing and properties (Int' l)**
  - AFC-2c/d compositions
- **Th-based fuel design, development & testing**
  - Planning and initial operations
- **Technique Development and Reference Materials (Int' l)**
  - Melt point, crystal growth, thick films, advanced processing
- **Vented Fuel/Getter Concept for High Burnup**
  - Focus on FG getter and "diode plate"

13 Ceramic WPs

2 Analytical Support WPs

49 Milestones
11 L2
38 L3

- **Substantial effort on planning**
  - Integration within ceramic fuels technical area
  - Integration with other technical areas (e.g. irradiation testing & Mod/Sim)
  - Integration with/roll up to overall AFC plans

Reference Materials, Techniques, & Thermochemistry

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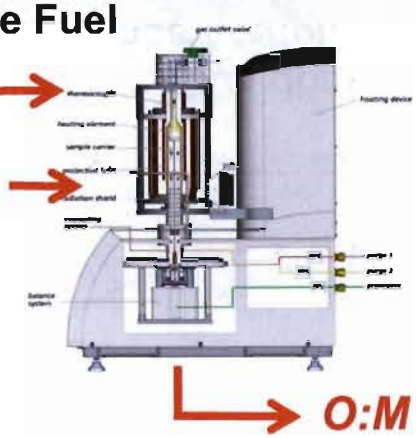
- **Thermochemical Understanding / Models Key to Efficient Oxide Fuel Development**
  - **Fuel Processing**
    - Control of O/M ratio for optimizing sintering
    - Homogeneity in multicomponent oxide systems
  - **In-Reactor Behavior of LWR and Transmutation Fuels**
    - *Behavior of O/M ratio during burnup and secondary phase formation*
    - *Chemical interactions with clad (e.g., oxidation, stress corrosion cracking)*



- **Work Scope / Task Summary**
  - Develop a thermochemical models of uranium-rare earth-oxygen systems
  - Prepare mixed oxide solid solutions for use as reference materials

FY11 Highlights / Progress

- ***Optimized a (U,Ce)O model in the hypostoichiometric region***
- **Developed an initial (U,La)O model**
- **Synthesized *$U_{80}Nd_{20}O$ and $U_{95}Nd_{05}O$ for use as reference materials***
- ***Collected oxygen potential data on (U,Ce)O in support of modeling effort***

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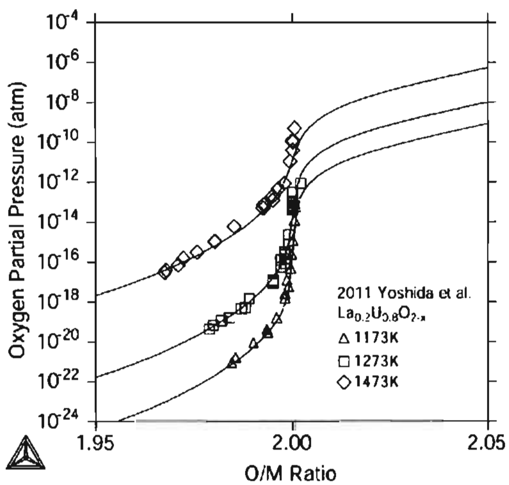
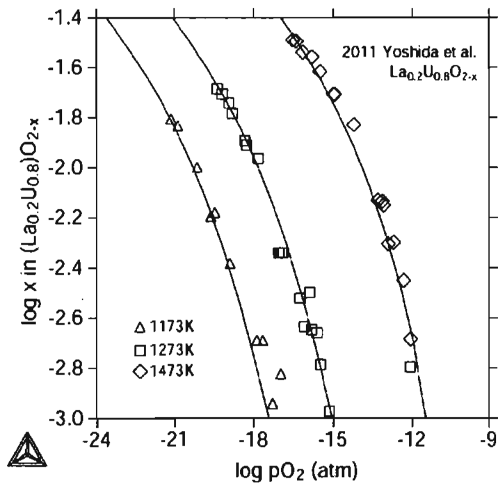
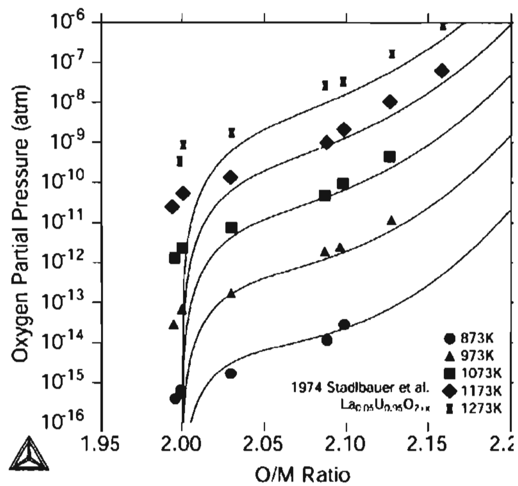
Thermochemistry of Oxide Materials

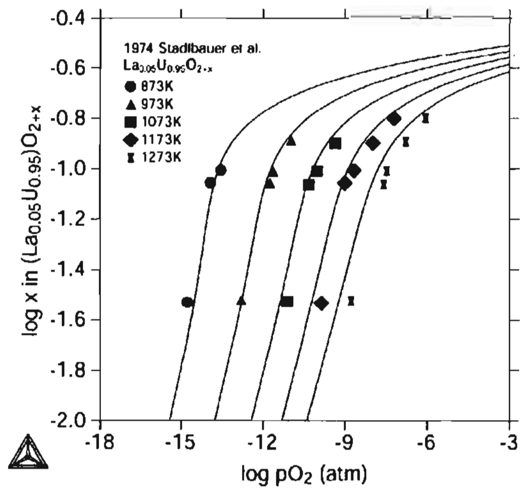
$U_{80}La_{20}O_{2-x}$



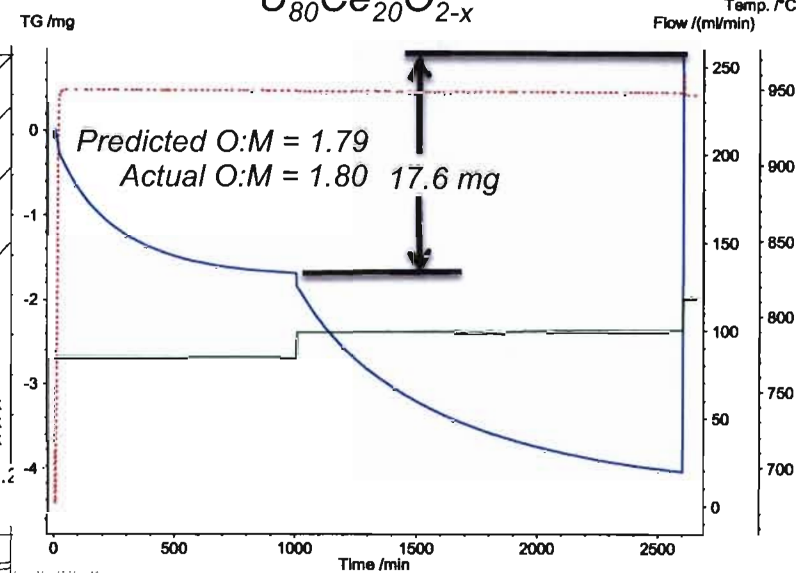


$U_{95}La_{05}O_{2+x}$





$U_{80}Ce_{20}O_{2-x}$

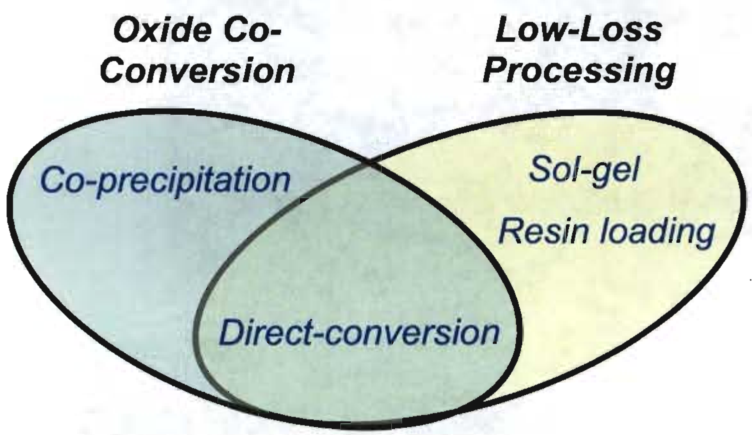


- **Phase equilibrium of La-doped UO_2 at any La concentration, T, O/M can be predicted from thermochemical model**
- **Measurement of oxygen potential versus temperature and O:M with uranium-cerium-oxide**

Oxide Feedstock Synthesis, Characterization and Conditioning

- **Development of Advanced Fuel Feedstock Synthesis Methods**
  - Tailor feedstock properties for advanced fuel systems
  - Develop methods to minimize processing losses

- **Work Scope / Task Summary**
  - Prepare and characterize oxide fuel feedstock material for use at LANL with fuel development activities and for fabrication of articles for irradiation testing
  - Evaluate porous spherical particles for use as a near-dustless feedstock



FY11 Highlights / Progress

- ***Synthesized six batches of oxide feedstock powders for use in fuel fabrication development. Four additional batches for feedstock synthesis process development.***
- ***Generated a Ceramic Fuel Feedstock Development Plan.***
- ***Performed an evaluation of co-precipitation processes for the synthesis of mixed-oxide fuel feedstock materials.***
- ***Prepared and characterized porous spherical particles for low-loss processing study.***

Oxide Feedstock Synthesis, Characterization and Conditioning

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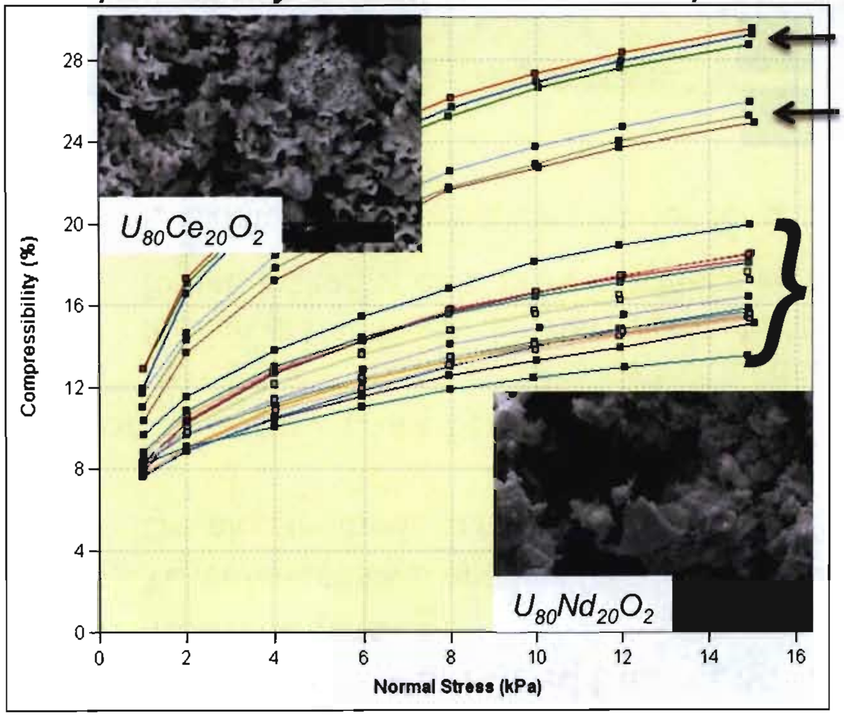
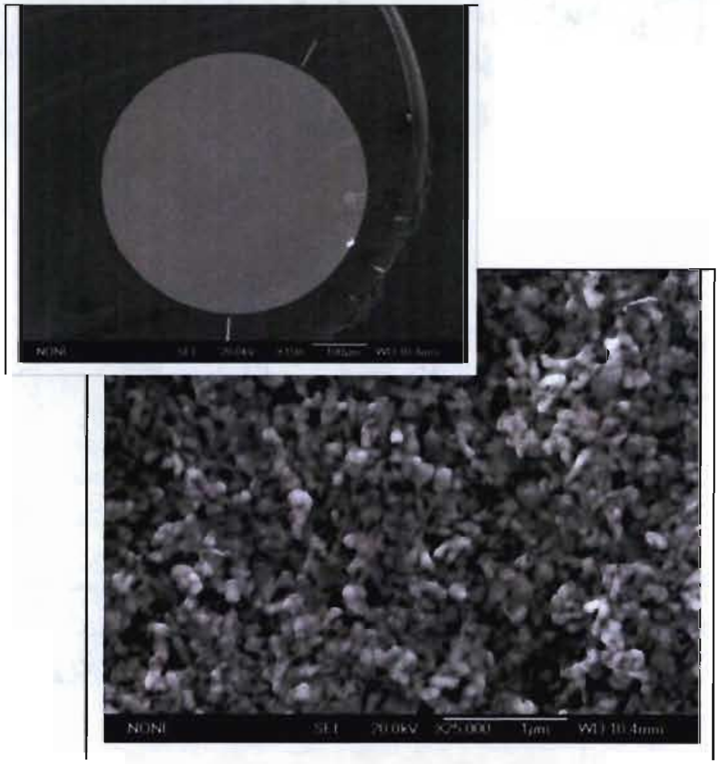
Compressibility of Various MDD Compositions



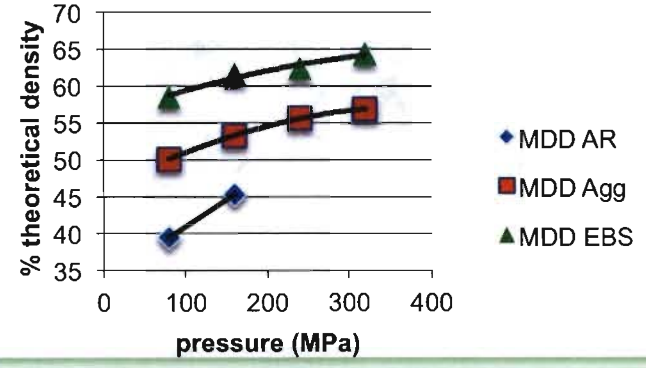


- **Particle morphology has a direct impact on bulk powder properties such as flowability and compressibility.**
- **Powder cohesion appears to increase with increasing ceria concentration.**
- ***Sol-gel derived UO_2 μ-spheres have been reduced under various conditions to produce porous, crushable feedstock particles.***
- ***SEM images confirm a porous microstructure.***

Oxide Feedstock Synthesis, Characterization and Conditioning

- **Support refinement and improvement of urania-based fuels for existing LWRs and technologies for transmutation applications to improve safety and performance**
  - In accident scenario, pellets will tend to fail at defect locations – weakest link
  - Defects in a powder create defects in a pellet both physical and chemical
  - Consistent, low defect (fewer missing surfaces subsurface cracks, pores)
  - Allows minor actinide, burnable poison and property modifier additions

- **Work Scope / Task Summary**
  - Synthesis and characterization of urania-based powders by conventional methods
  - Acquisition and characterization of urania-based powders from ORNL made by direct conversion
  - Conditioning and fabrication of pellets from feedstocks

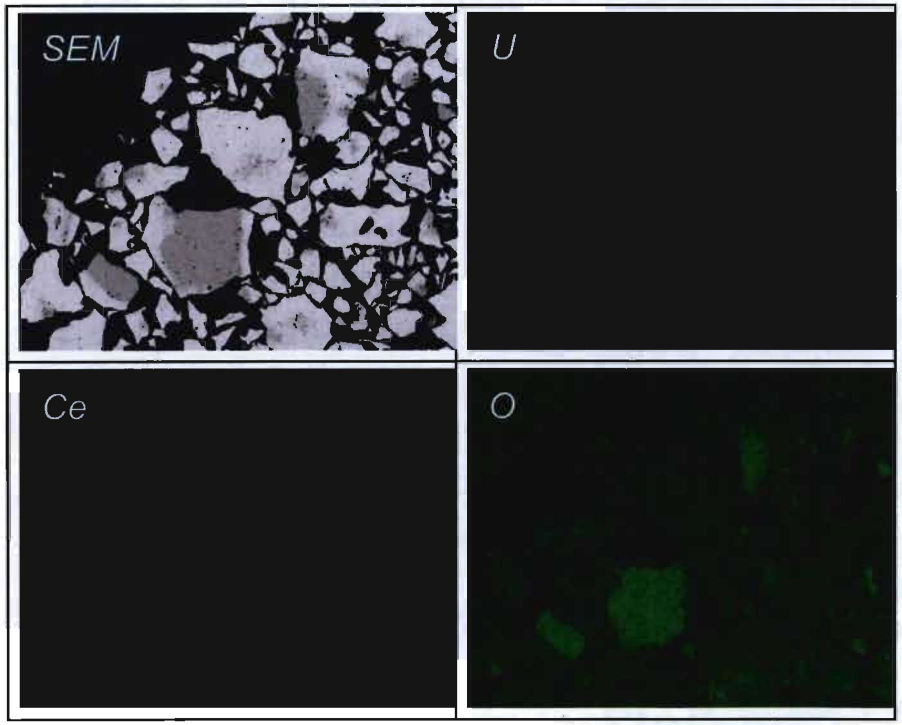


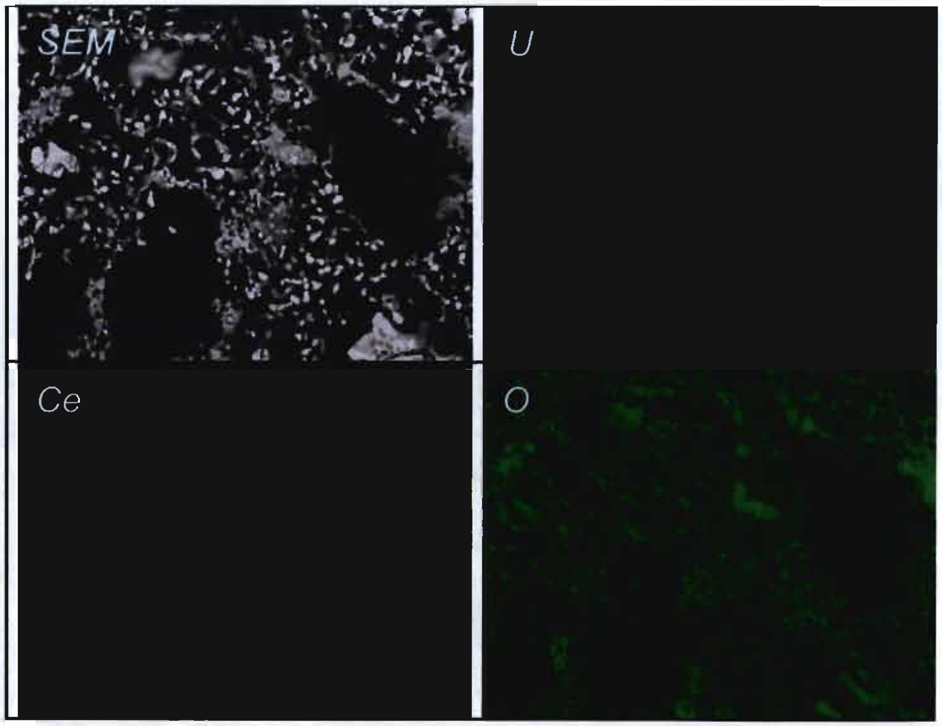
FY11 Highlights / Progress

- ***Acquired, characterized and conditioned three urania powders made by direct conversion***
- ***Acquired, characterized and conditioned six mixed oxide powders (U with Ce, Nd and Th additions) made by direct conversion***
- ***Synthesized and characterized uranium/cerium powders by conventional methods***

Oxide Feedstock Synthesis, Characterization and Conditioning

- Elemental - Energy Dispersive Spectroscopy Mapping (EDS)
- Conventional and direct denitration synthesis of 80% UO_2 20% CeO_2
- Conventional – heterogeneous, coarse
- Direct denitration – largely homogeneous, fine





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Advanced Processing

- **Improve fabrication methods to produce pellets from low loss feedstocks under controlled conditions for better performance**
  - Pellet performance *i.e. higher power, deeper burn* governed by defects fail at weakest link
  - Low loss fabrication supports advanced fuel cycle – *worker and environmental safety*
  - Alter sintering rate and O/M ratio as desired

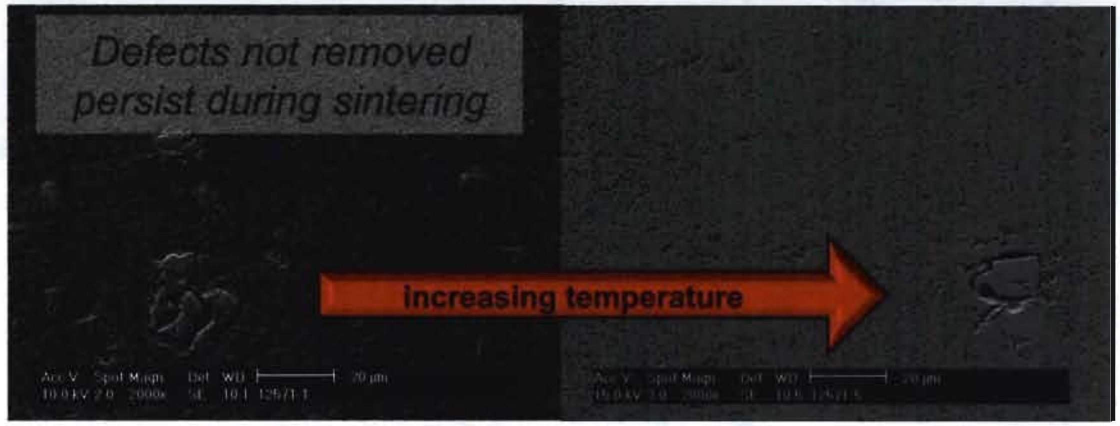
- **Work Scope / Task Summary**
  - Develop process methods (powder conditioning/ pressing/sintering) for low loss powders
  - Build equipment to control sintering atmosphere to allow dynamic O/M control throughout sintering process
  - Demonstrate microwave sintering of uranium oxide

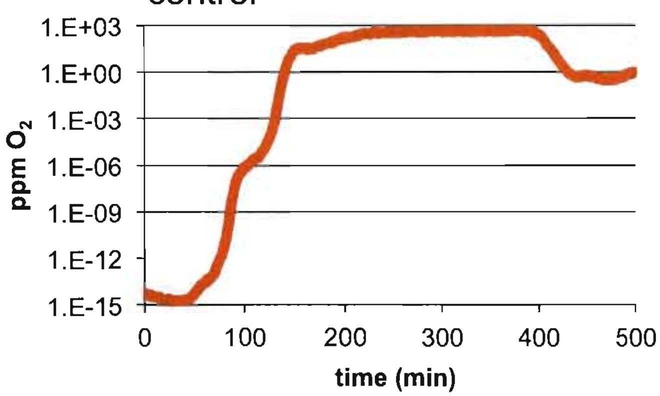
FY11 Highlights / Progress

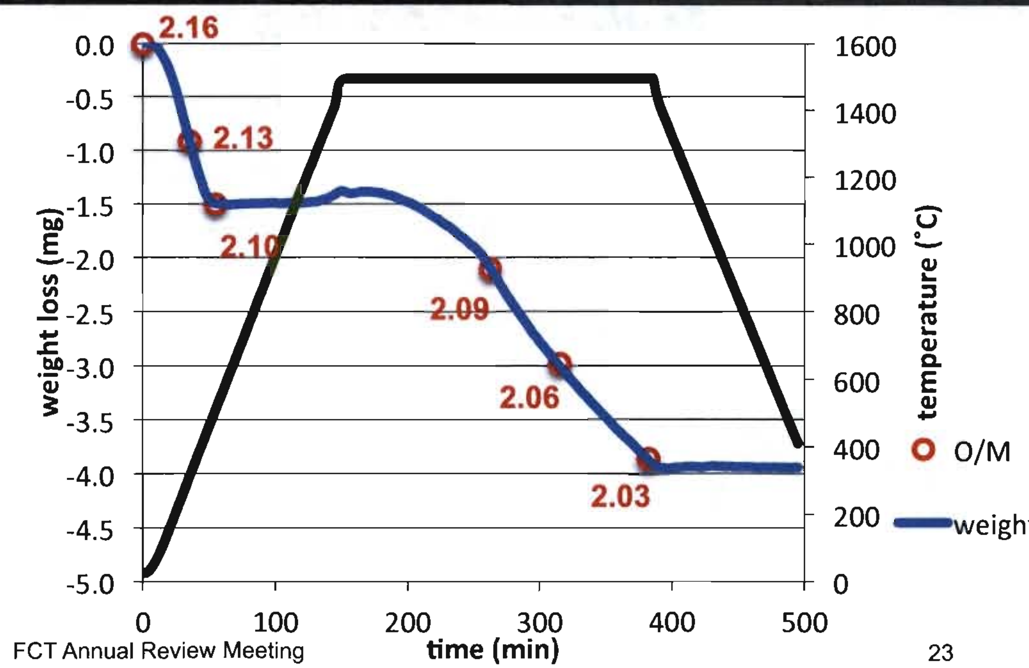
- ***Fabricated high density pellets from low loss powders***
- ***Developed sophisticated atmosphere control during sintering/ characterization for equipment***
- ***Produced microwave sintered pellets***

Advanced Processing

- **Sintering atmosphere controlled dynamically to adjust O/M**
  - Guided by thermodynamic modeling at ORNL
  - Achieved with sophisticated gas control system
  - Argon carrier with sub ppm hydrogen and oxygen control
  - Oxygen concentration controlled over 18 orders of magnitude
  - Allows precise sintering control







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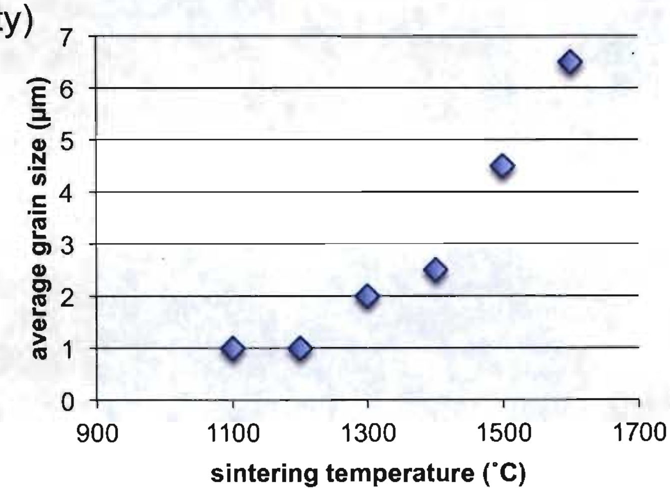
Fabrication Process Development

- **Develop fabrication methods to produce pellets with controlled microstructures for better performance**
  - Pellet performance *i.e. higher power, deeper burn* governed by material properties dictated by microstructural features
  - Achieve microstructural control (grain size and porosity)
- **Work Scope / Task Summary**
  - Improve understanding of interrelationships of composition, processing and microstructure
  - Develop methods for microstructure control
  - Study properties as a function of composition and microstructure
  - Detailed microstructural characterization



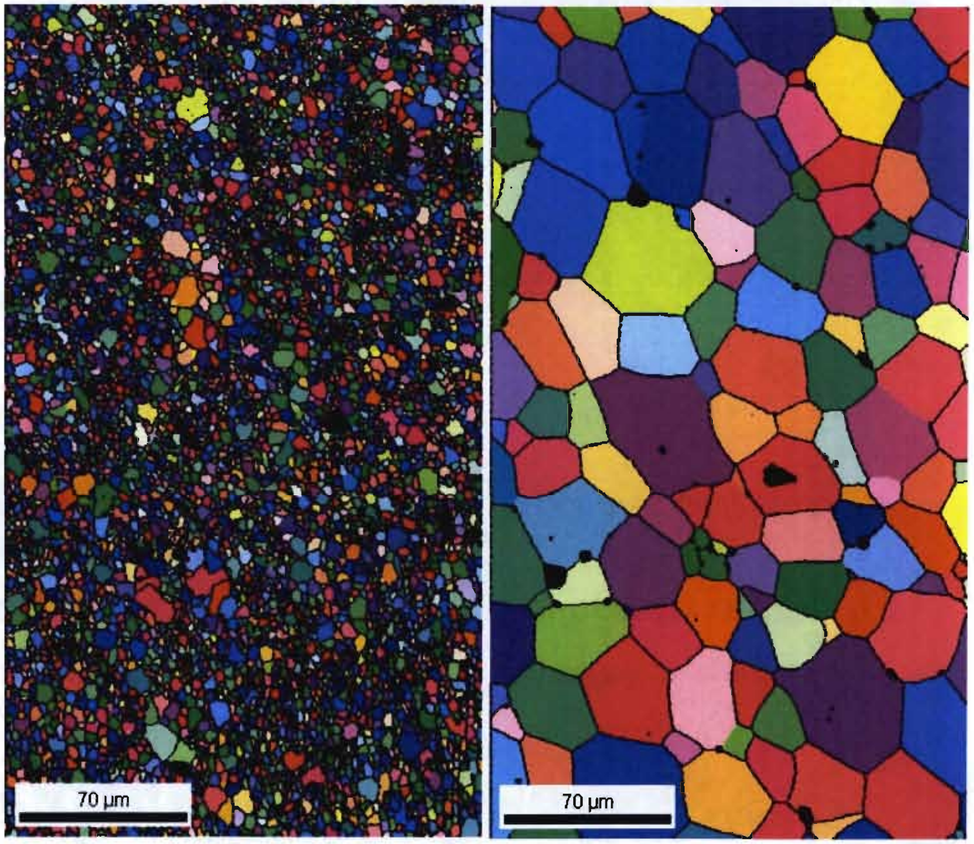
FY11 Highlights / Progress

- ***Produced pellets with controlled grain sizes***
- ***Studied thermal diffusivity as function of processing and composition***
- ***Fabricated apparatus for melt point determination***
- ***Detailed study of grain boundary orientation***

Fabrication Process Development

- **Controlled grain size in nominally identical pellets**
  - Grains grown from 3 microns to 21 microns
  - Density relatively constant 92% and 93% respectively
  - EBSD images of pellets sintered under gettered argon shows grain orientation, morphology and size
  - No texture (preferred orientation) is induced by processing methods

Grain size controlled by processing parameters



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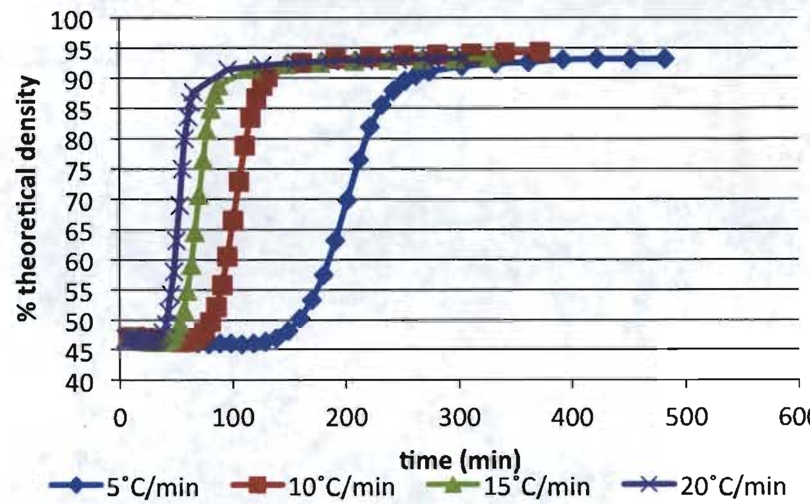
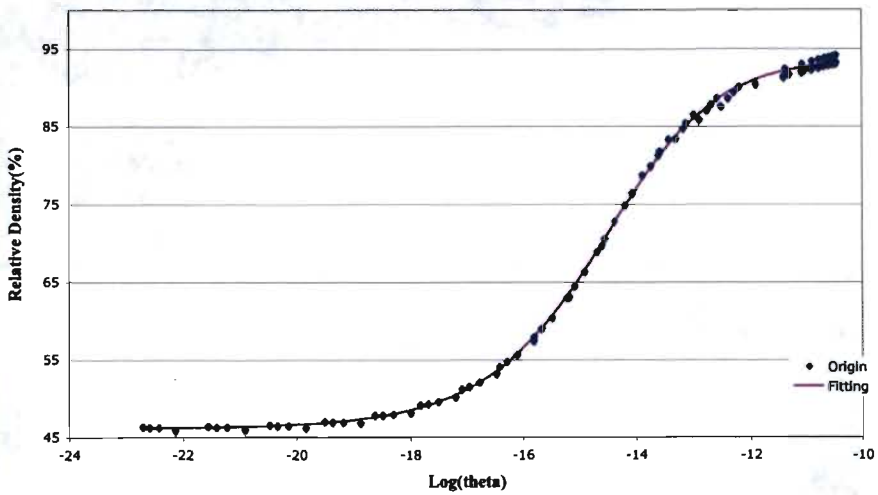
Analytical Support: Sintering Modeling

- **Predict and optimize sintering of ceramic pellets to produce consistent, robust, minimally flawed fuel**
  - Improve control over ceramic microstructure
  - Minimize costly process steps and reduce waste
  - Improve pellet reproducibility and reliability
- **Work Scope / Task Summary**
  - Collect data to construct Master Sintering Curve for uranium oxide
  - Extend MSC approach to aspects of special importance to sintering behavior
  - Optimize sintering

FY11 Highlights / Progress

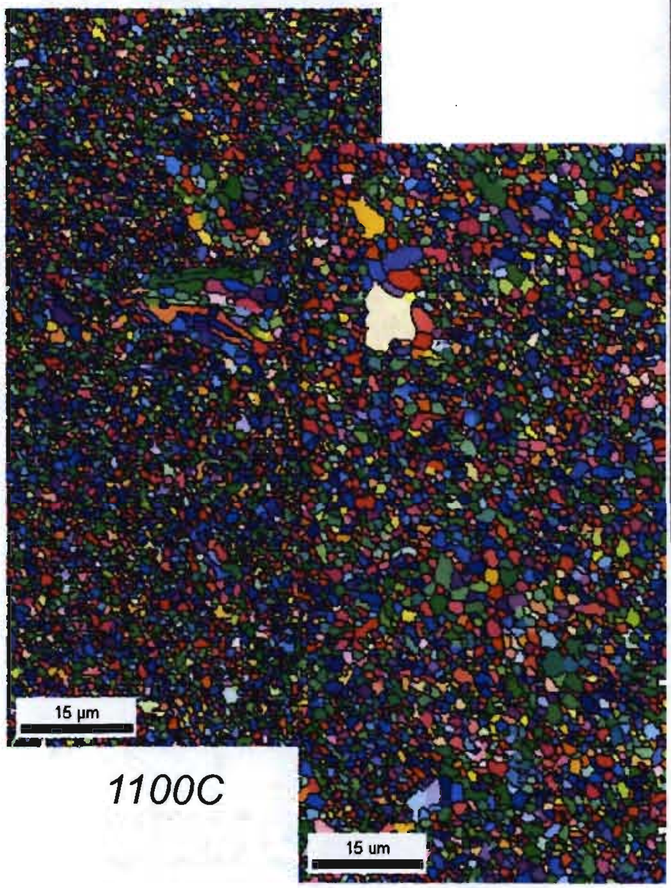
- ***Generated Master Sintering Curve***
- ***Collected sintering data for three different O/M ratios***
- ***Collected grain size evolution data***





Analytical Support: Sintering Modeling

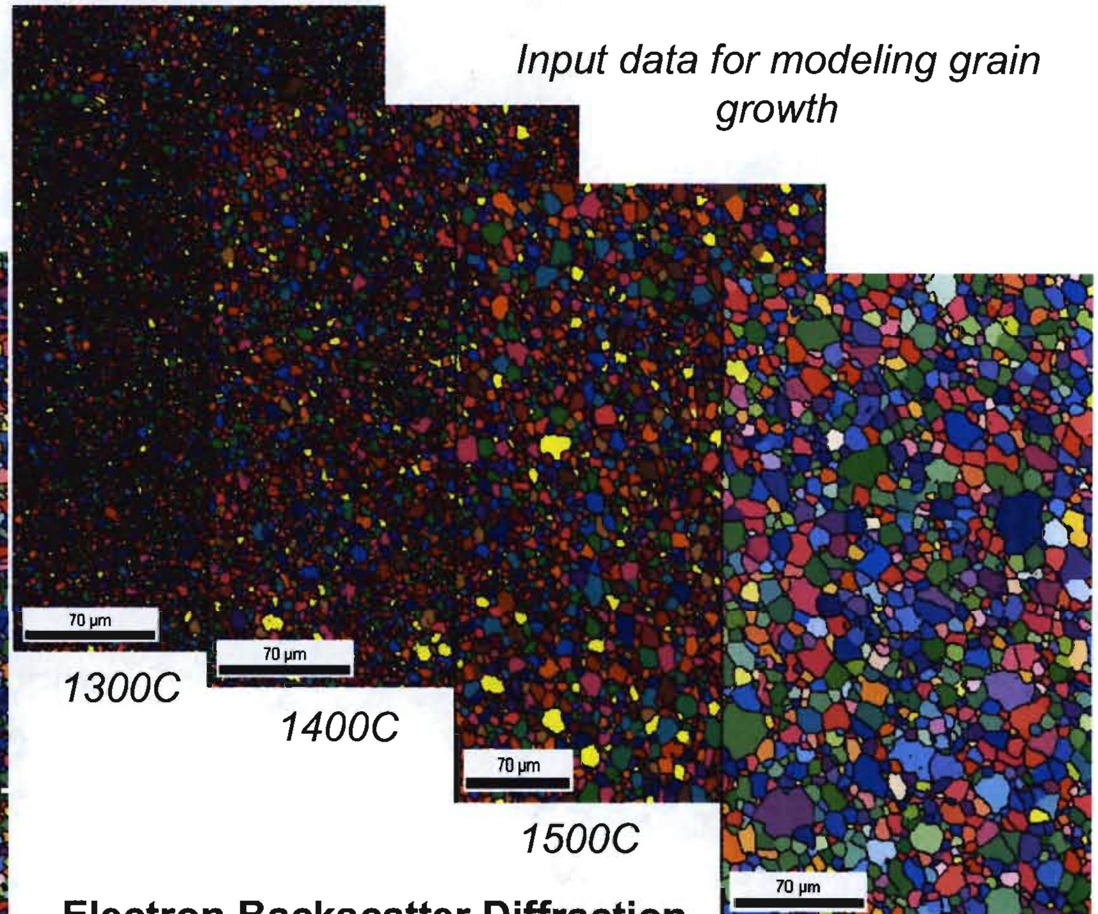
Grain Evolution During Sintering



1100C

1200C

Input data for modeling grain growth



1300C

1400C

1500C

1600C

Electron Backscatter Diffraction (EBSD)

Thermophysical Properties of Ceramic Fuels

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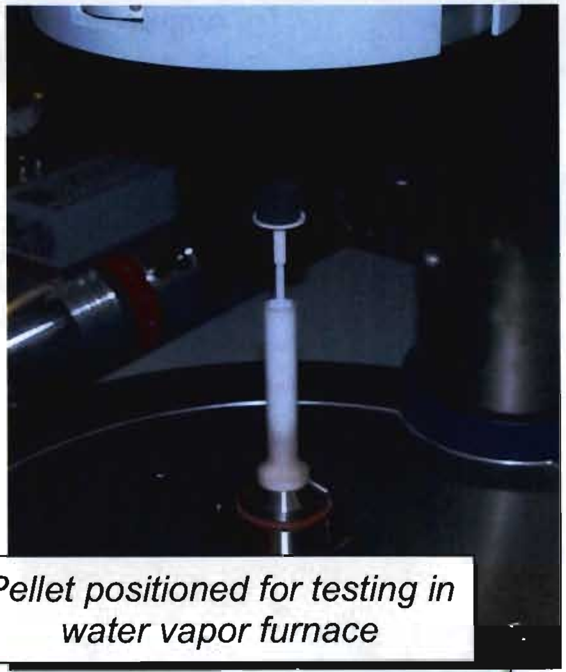
- **Thermal transport dominates ceramic fuel performance during both normal operation and transients**
- **Modern property measurement techniques feed numerous other areas**
  - Strong ties to ongoing modeling and simulation work
  - Improvement of existing fuel performance codes
  - Enhanced theoretical understanding to facilitate design of evolutionary and revolutionary fuel forms
- **Work Scope / Task Summary**
  - Characterize fundamental aspects of thermophysical performance as a function of chemistry and structure
  - Provide datasets to facilitate improved models describing oxidation of LWR fuel during accidents

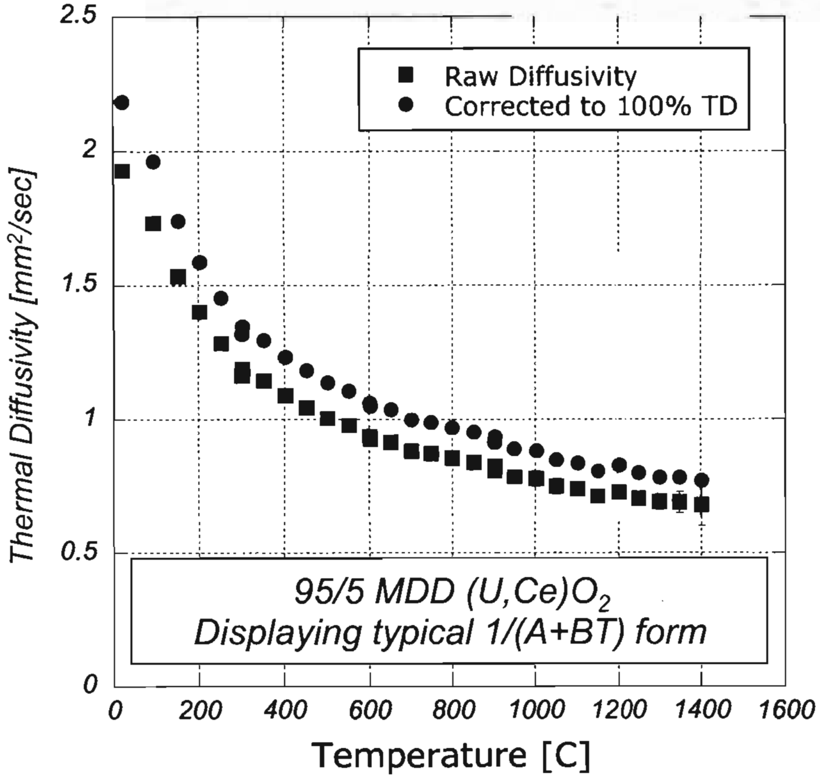


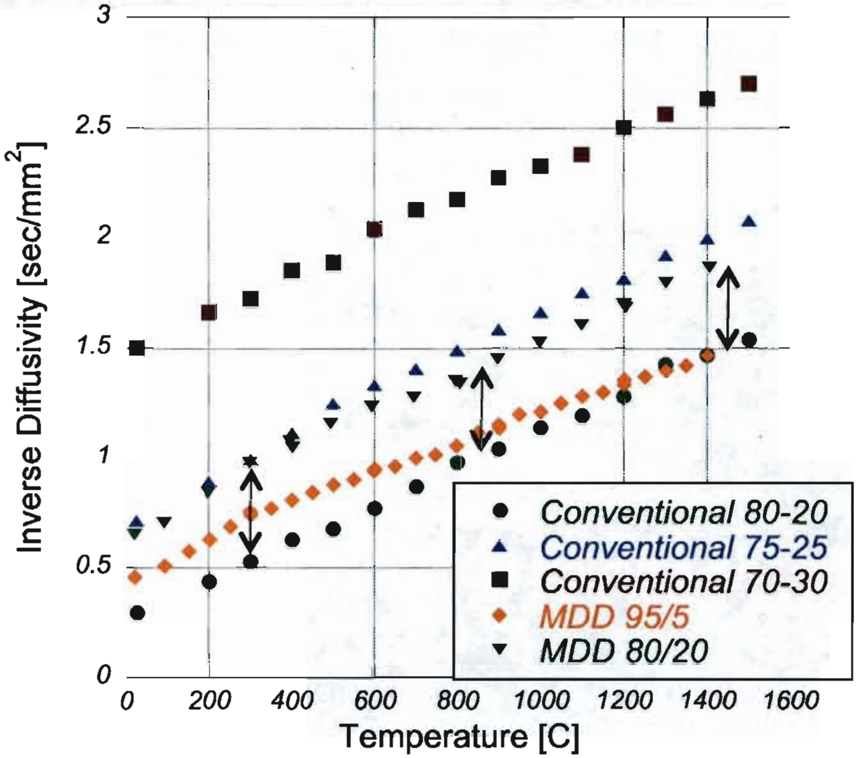
Pellet positioned for testing in water vapor furnace

FY11 Highlight list/Progress

- ***Thermal conductivity of MDD solid solution oxides has been measured to understand the role of cation charge state on thermal transport***
- ***Design and initial sample fabrication was completed for the first HFIR rabbit irradiation campaign for the separate effects studies on thermal conductivity in oxide fuels***

Thermophysical Properties of Ceramic Fuels: Diffusivity of Advanced MDD Oxide Solutions

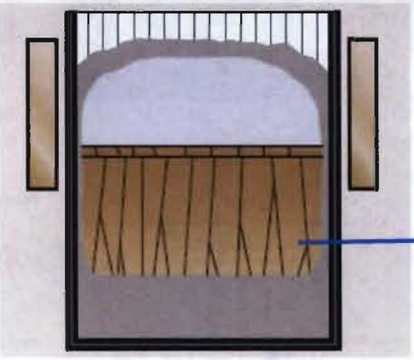




- FY11/12 emphasis is investigating cation charge/mass effects on thermal transport in $(U,X)O_2$
- MDD powders synthesized by ORNL pressed and sintered for SET development
- Disparity observed between MDD and conventionally produced material

Reference Materials and Technique Development – Crystal Growth

- **Fundamental studies of fuel material properties necessary to support Mod/Sim through SET**
  - Improves available data sets and fills in data gaps
  - Improves fundamental understanding of materials characteristics
  - Requires use of single, bi-and Multi-crystals to separate individual effects
- **Work Scope / Task Summary**
  - Growth and characterization of single and multi-crystals of UO_2
  - Produce single and engineered substrates for UO_2 Thick Film Deposition



Skull Melting Technique

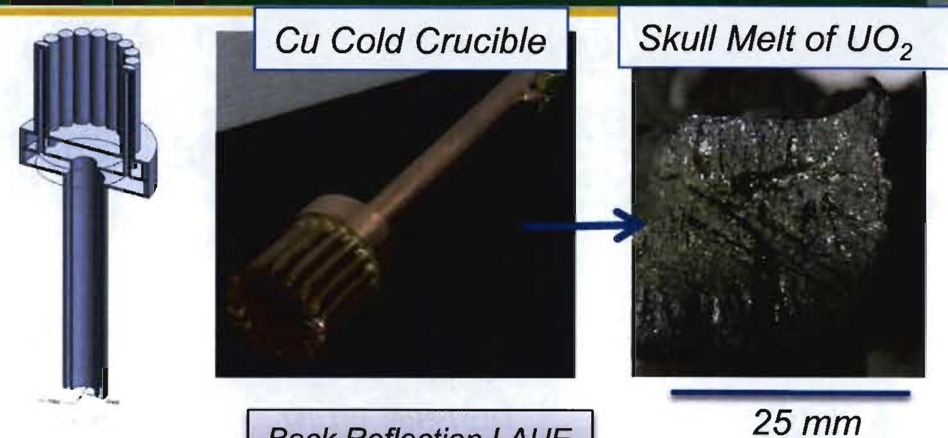
UO_2

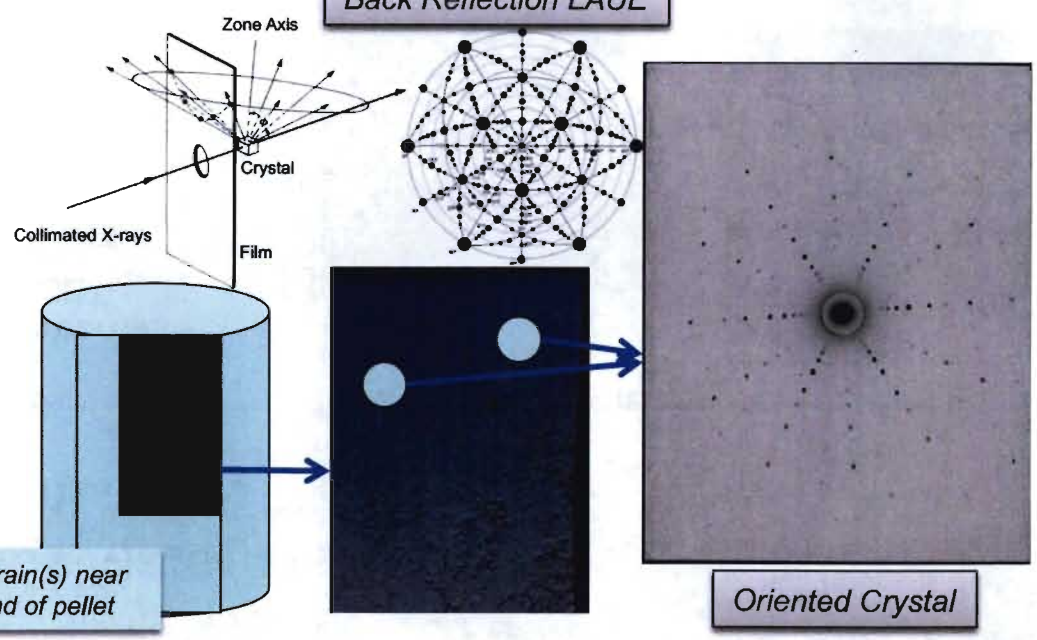
FY11 Highlight list/Progress

- ***Successfully melted large quantity of UO_2 using "Skull" method and W-susceptor***
- ***Melted UO_2 in Tri-arc indicating path to single crystal growth by modified-CZ***
- ***Generated single crystals with embedded grains using exaggerated grain growth technique.***

Reference Materials and Technique Development – Crystal Growth

- **Skull Melting Technique**
  - *350g UO_2 melted with tungsten susceptor*
  - Some columnar crystal growth with *fine grained regions under rapid cool*
- **Tri-arc Melting Technique**
  - 10g *of UO_2 melted and sustained arc under G-Ar*
  - High UO_2 vapor pressure caused quartz window to be coated and difficult to see through.
  - Provides path for single crystal growth *by CZ*
- **Exaggerated Grain Growth Method**
  - Utilizes controlled atmosphere to cause grains to grow preferentially
  - Normal 5 mm L x 5 mm dia. pellet was treated
  - Pellet has upper and lower regions of large grains with embedded smaller grains.
  - Center region 1.5 x 3 mm has small grains with higher porosity





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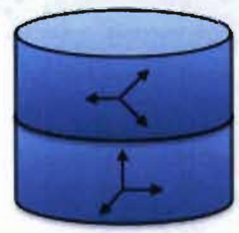
Reference Materials and Technique Development – Thick Film Growth for FG studies

- "Key" to understanding the effects of fission gas release during irradiation is separate effects testing on single and multicrystalline sub
  - FG bubble growth
  - FG behavior at grain boundaries
  - FG effects on microstructure and properties
- **Work Scope / Task Summary**
  - Deposition of UO_2 thick films with embedded xenon on various substrates
  - Deposition and characterization of UO_2 thick films with controlled levels of Xe



Engineered Bicrystal

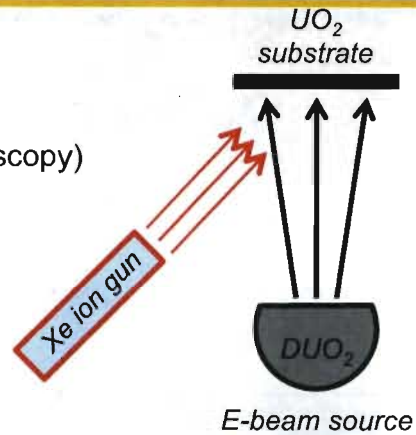
UO_2 Substrate with TF of UO_2

FY11 Highlight list/Progress

- ***Evaluated analytical techniques for quantitative Xe analysis***
- ***Demonstrated embedding of low Xe content in thin films***
- ***Generated nano-grained thin films***
- ***Grew thick film of UO_2 with Xe on UO_2 polycrystalline substrate***

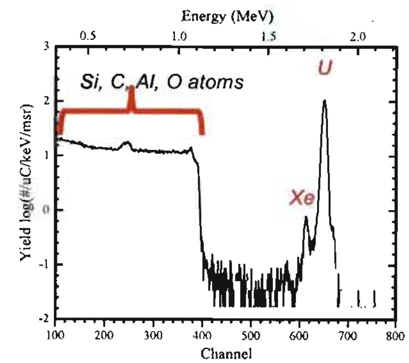
Reference Materials and Technique Development – Thick Film Growth

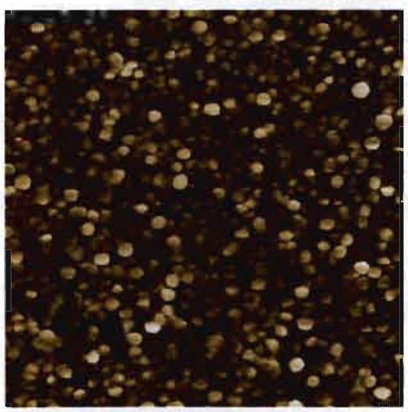
- **Screened several Xe analysis techniques**
  - Thick films (MXRF- micro X-ray fluorescence)
  - Thin films (RBS - Rutherford backscattering spectroscopy)
- **Demonstrated embedded low Xe content**
  - 0.6 at.% Xe
  - Supports mod/sim efforts
- **Nano-grained microstructure**
  - Support FG behavior studies at Rim structure
- **Assessed of xenon gas with temperature**
  - Annealing experiments up to 1300°C
  - No Xe redistribution after annealing to 1300°C
  - Can calculate release fraction *f*(Temp.) – Diffusion coefficient and activation energies



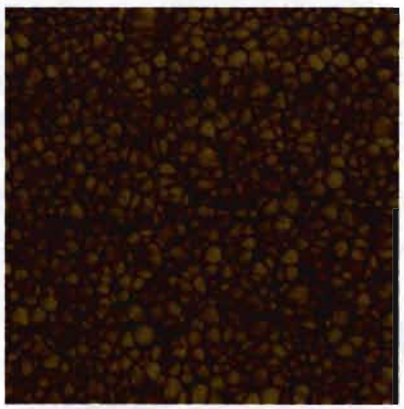
UO_2 Substrate with TF of UO_2

RBS of UO_2 Thin Film

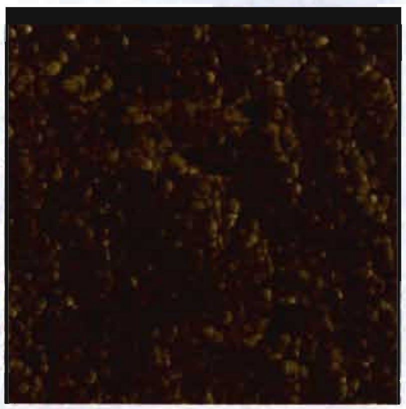




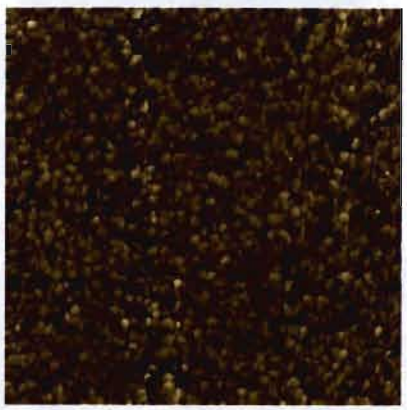
Carbon substrate



as-deposited UO_2 film (27 nm)



T_{ann} =600° C (35 nm)

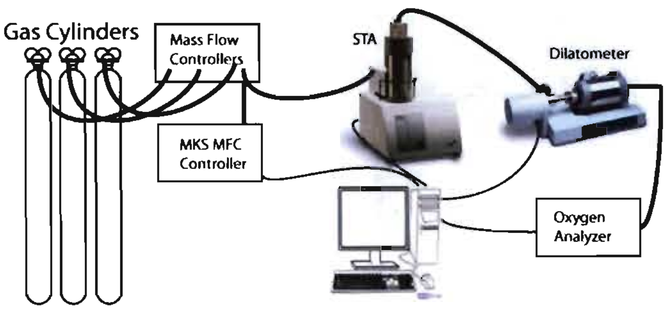


T_{ann} =800° C (15 nm)

Reference Materials and Technique Development – Dynamic O/M control

- **O/M (Oxygen-to-Metal ratio) is responsible for numerous effects in fuel properties, processing and performance.**
  - O/M linked to FCCI, FCMI, fuel swelling, heat capacity, thermal conductivity, melting point, etc.
  - Required for modeling and simulation input
  - Influences sintering efficiency -- low-loss processing
- **Work Scope / Task Summary**
  - Dynamically control O/M to set oxygen activity with changing temperatures
  - Investigate effects of O/M on sintering kinetics
  - Implement control on analytical instruments

Schematic of O/M Gas Control System



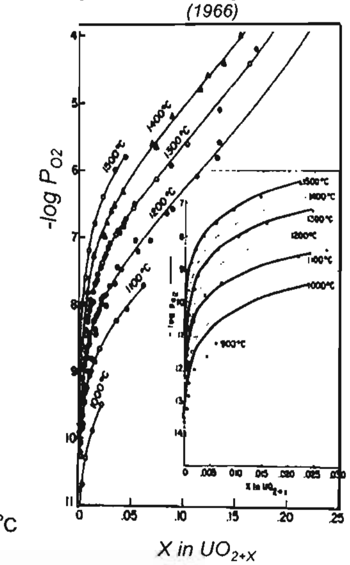
FY11 Highlight list/Progress

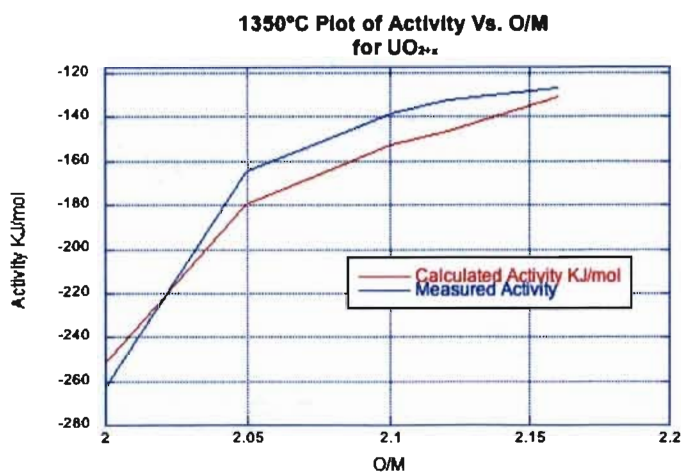
- ***O/M has been effectively controlled using mixtures of $Ar/H_2/H_2O/O_2$ to dynamically set the oxygen activity above UO_2 pellets***
- ***Preliminary comparisons of measured O/M as a function of oxygen activity have been made against those determined by ORNL and agree fairly well.***

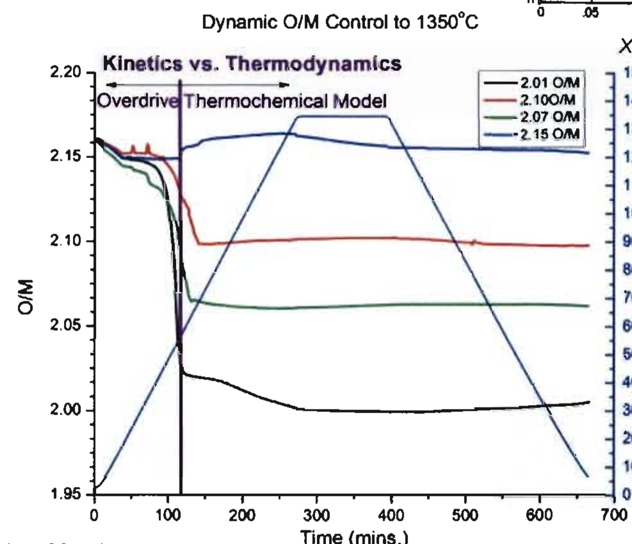
Reference Materials and Technique Development – Dynamic O/M control

- **Dynamic O/M control via precise gas mixtures**
  - Controlled O/M by dynamically changing pO_2 f(temp.)
    - *Large range of pO_2 values necessary to control over wide temperature range*
  - Based on UO_2 data set from ORNL – good match
    - *Data values available from 600°C*
    - *Used reduction from starting O/M of 2.16*
    - *Experimentally determined pO_2 values below 600°C*
  - Calculated ΔG from predicted and measured pO_2
    - *Some discrepancy between measured and calculated values –miscellaneous source terms*
    - *Reproduced experiments with O/M variance of ±0.005*

Hagemar et al., J Inorg. Nucl. Chem ***28*** *(1966)*







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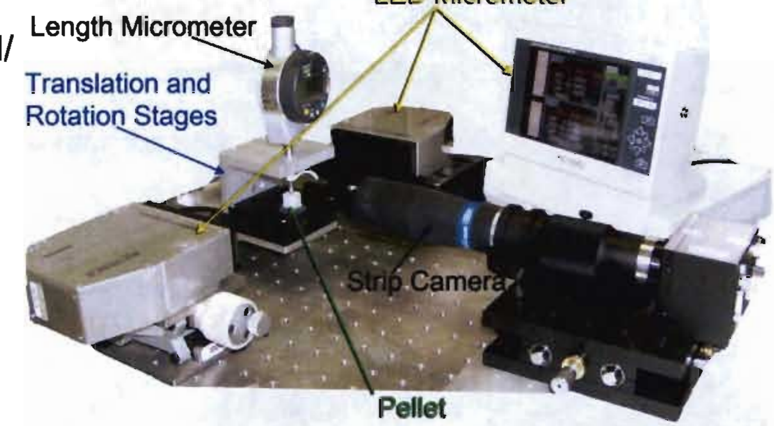
Technique Development – Pellet Measurement System for Advanced Characterization

- **Capability to measure fresh and irradiated fuels**
  - Reduces radiological exposure
  - Provides accurate representation of pellets for mod/sim data
  - Net shape processing – modeling and experiment
  - Has high speed industrial applications
- **Work Scope / Task Summary**
  - Create a prototype pellet measuring system
  - Enable control and measurement via a LabView interface
  - Evaluate potential for glovebox application

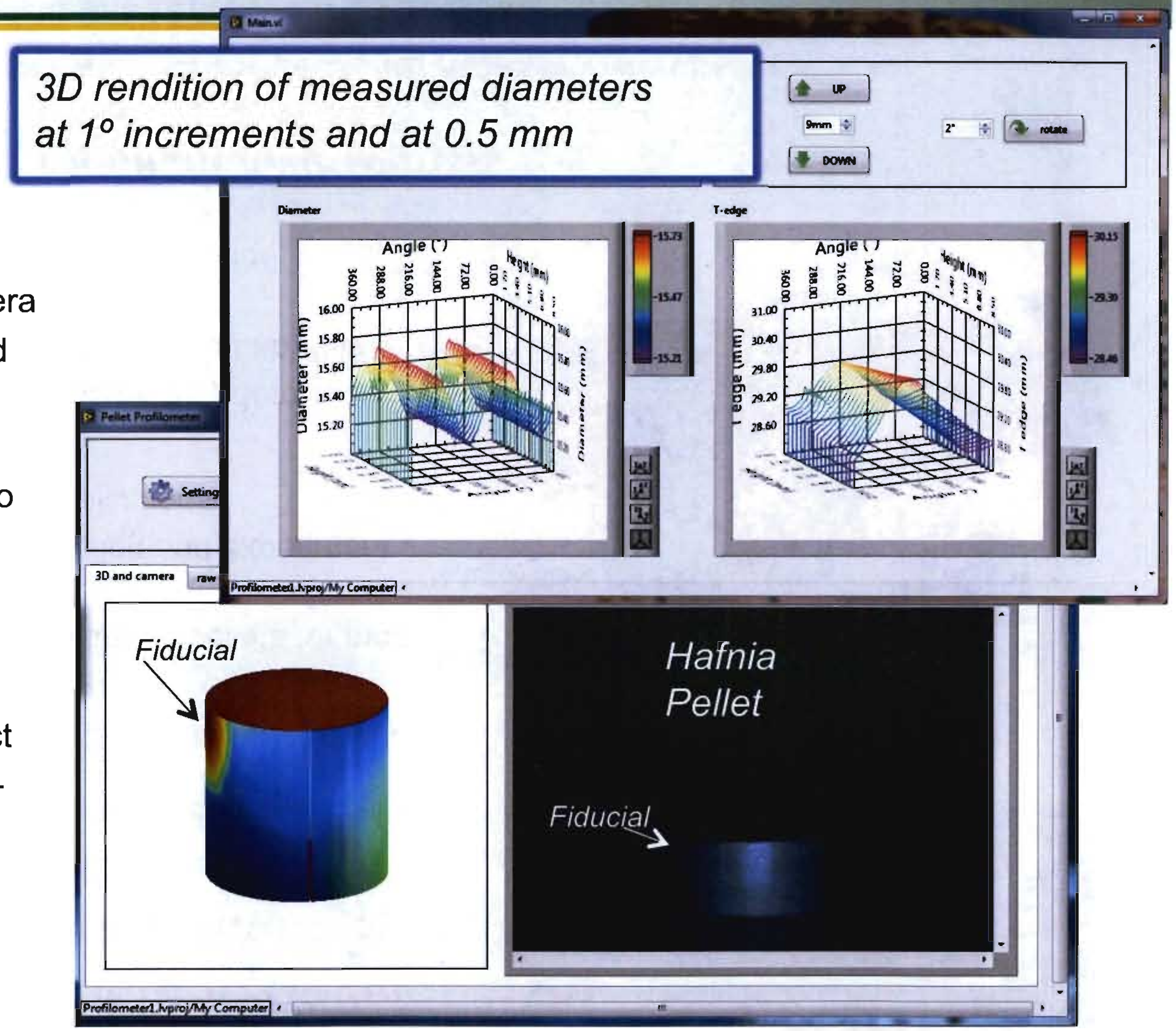


FY11 Highlight list/Progress

- ***Procured components for a complete pellet measuring system***
- ***Established a prototype pellet measuring system***
- ***Made initial measurements of hafnia pellet***

Technique Development – Pellet Measurement System for Advanced Characterization

- **Pellet measurement system created**
  - LED measurement of pellet diameter
  - Optical height measurement by camera
  - Fully synchronized and recorded to data file
- **Advancements**
  - Measurements down to 10 nm on Z-axis
  - Fine rotational control
- **Potential Add-ons – quality assessment**
  - RUS – physical contact
  - Non-linear RUS – non-contact method



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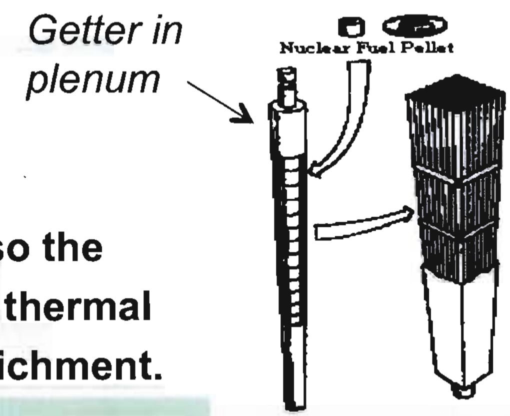
Vented Fuel Pellet/ Getter Concept For High Burnup

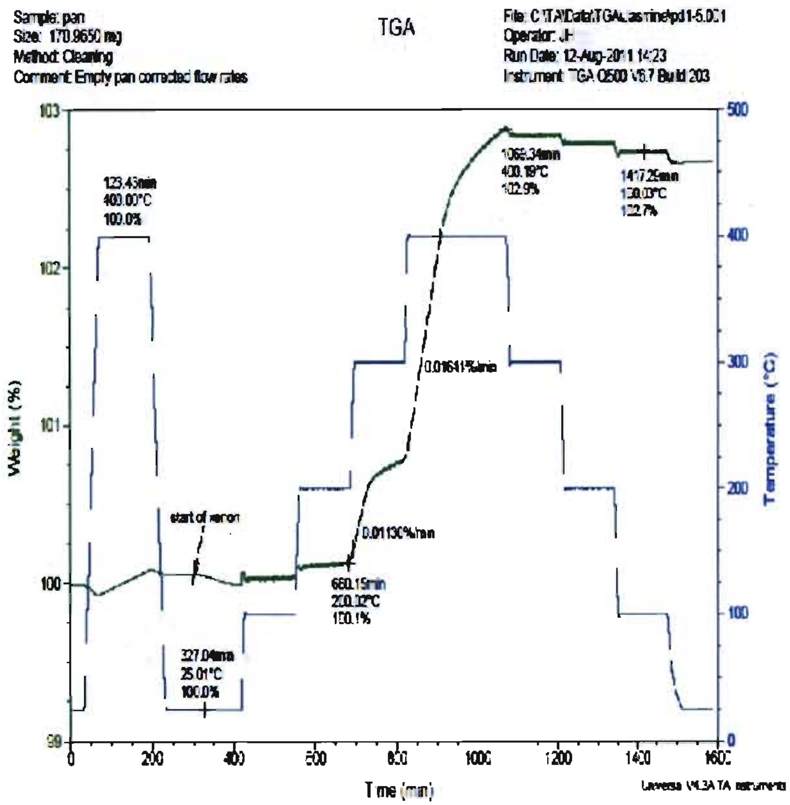
- The Project's goal is to extend the burnup capacity of nuclear fuel. Emphasis is on the present US nuclear reactor fleet, but this can be expanded to include advanced reactors.
- The Concept lends itself to aiding in the utilization of Used Fuel and development of Accident Tolerant Fuel.
- Examine getter materials that can be located in the fuel pin plenum and which will preferentially adsorb Xe and Kr, the major fission product gases. By doing so the concept would reduce internal clad pressure, improve thermal conduction, and enhance reactivity without higher enrichment.



FY11 Highlight list/Progress

- *Thermogravimetric Analysis (TGA)was carried out on a number of candidate getter materials at BNL; additional measurements were performed on a Quartz Crystal Microbalance at LANL.*
- *Pd micron particle powder was shown to exhibit very positive adsorption characteristics for Xe.*

Vented Fuel Pellet/ Getter Concept For High Burnup

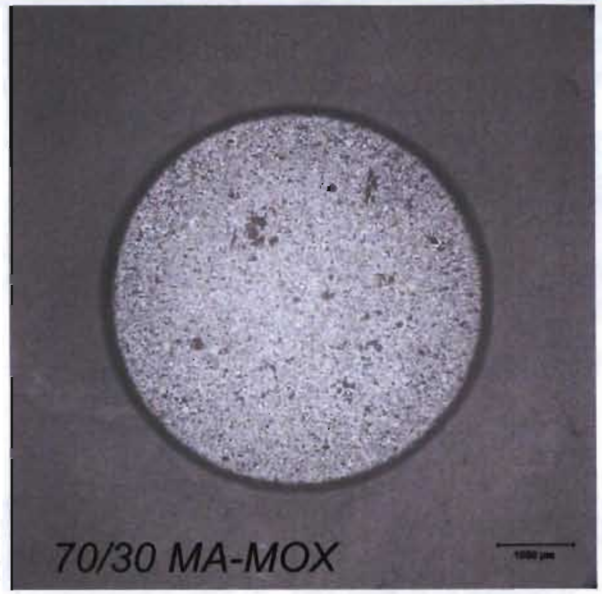


Adsorbent	Excess Adsorption	Pressure - temperature Eq	Surface area (sq. m/g)	Suspected phenomena
MOF	0.317	Yes	1500-2100	physisorption
Micro pore Carbon	0.500	Yes	400	physisorption
Micron Pd	104	?	1	? chemisorption /phase change
Milli Pd	7.8	?	<< 1	?
Activated Carbon 10% Pd	0.65	Yes	750 - 1000	physisorption
Activated Carbon 30% Pd	1.45	Yes	950	physisorption
SWNT	0.65	Yes	2200	physisorption

The TGA plot to the left shows how Xe is adsorbed on Pd powder exhibiting characteristics of chemisorption. From the table to the right one can observe that the excess adsorption for Pd shows that Pd can adsorb about 100 times the Xe in the same plenum volume then if the Xe remained as a free gas. In doing so the getter (Pd) improves the thermal-mechanical properties of the fuel pin, and removes parasitic neutron adsorbing Xe from the active fuel region to the plenum.

MA-MOX Sintering Kinetics

- **Establish understanding and control of sintering kinetics for MA-MOX fuel pellet fabrication**
  - Process optimization to improve efficiency, quality and reliability
  - Decrease process loss via improved process control
  - Improve microstructural and O/M control of product



- **Work Scope / Task Summary**
  - Kinetics of sintering as a function of time, temperature and atmosphere via dilatometry
  - Interrupted sintering in metal furnace
  - Relate sintering conditions to chemistry and microstructure

FY11 Highlight list/Progress

- ***Completed dilatometry runs on 70/30 and 80/20 MOX and MA-MOX analogue compositions (MA= 2% Np and 3% Am substituted for U)***
- ***Extended interrupted metal furnace sintering data sets to higher temperatures (1800 °C max)***
- ***Initiated chemical and microstructural characterization***

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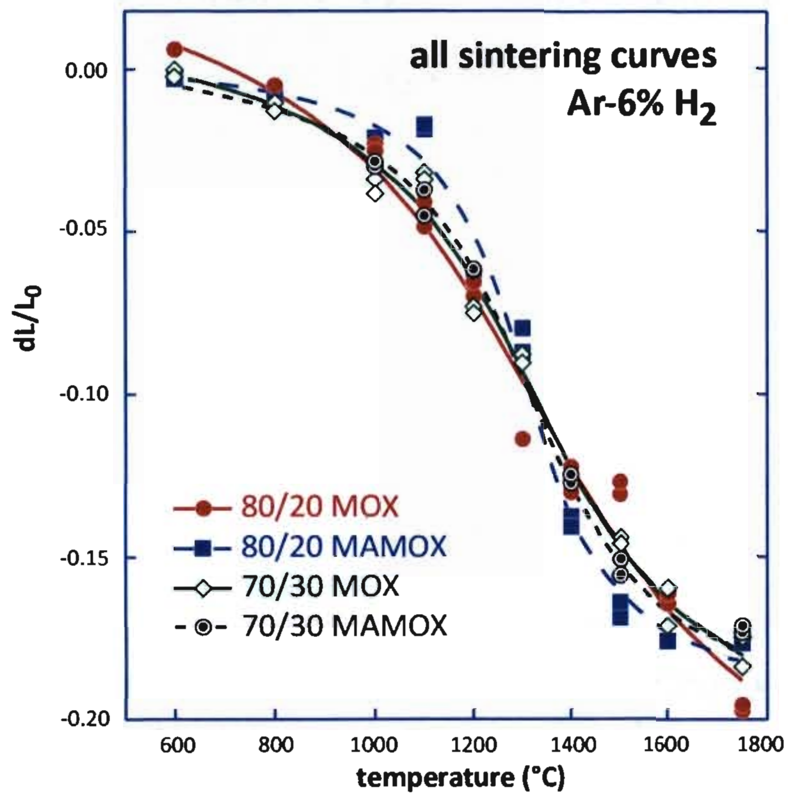
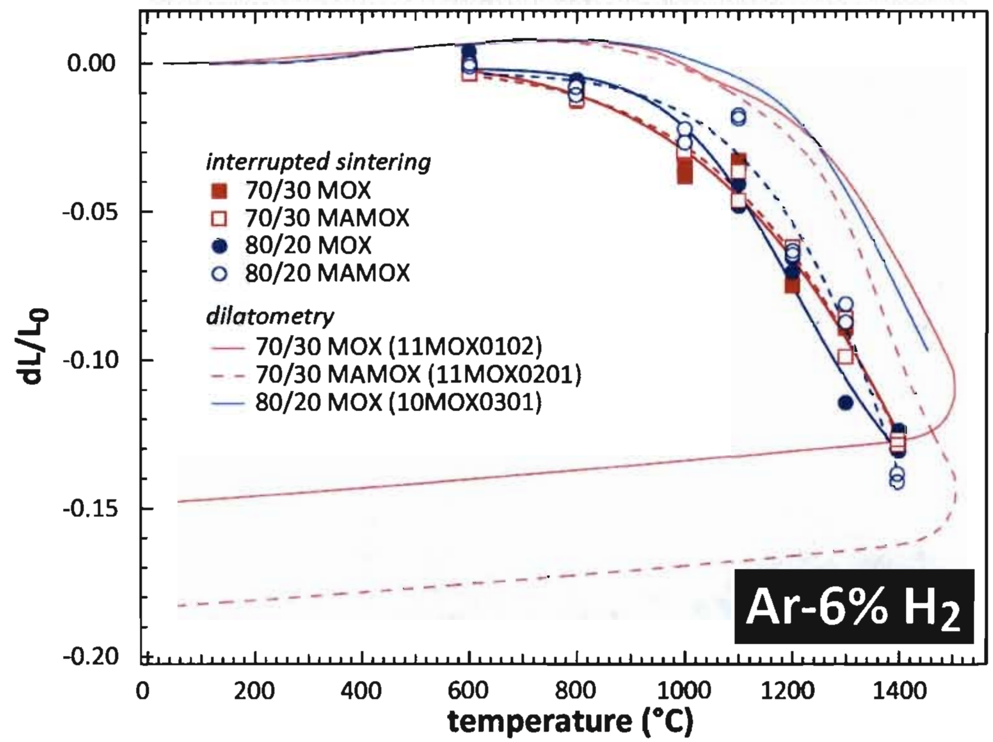
MA-MOX Sintering Kinetics





- **Gross sintering kinetics are similar for different compositions**
- **Atmosphere has a strong influence on kinetics**
  - Process parameters are furnace specific

Increased emphasis in FY12 on LWR fuels (R&D Objective 1)

Ceramic fuels development technical area provides core expertise, mod/sim datasets, and technical database for improving existing fuels and developing new fuels

- R&D OBJECTIVE 1: Develop technologies and other solutions that can improve the reliability, sustain the safety, and extend the life of current reactors

- Transitioned over the last ~2 years to an oxide-based fuels R&D program supporting the NE R&D roadmap which also provides the basis (data and expertise) to respond to evolving priorities, e.g. accident-tolerant fuel development

- Oxide fuel R&D transitions in FY12 from a focus on NE Roadmap R&D Objective 3 (Fuel Cycle) to NE Roadmap R&D Objective 1 (LWRs)
  - Change involves prioritization on UO_2 performance and safety for knowledge base, data sets for model development and validation, and reference basis for advanced LWR development

FY12 Work Scope and Task List

- **FY12 again focused on oxide systems (increased LWR focus)**
  - Fuel Pellet Fabrication: "Turning art to science"
  - Fuel Performance: "Control through fundamental understanding"

- **Advanced Process Development**
- **Properties and Advanced Characterization**
- **Performance and Separate Effects Testing**
- **Reference Materials and Technique Development**
- **Innovative Fuels Concept: Fission Gas Getter**

7 Ceramic WPs

1 Analytical Support WP

1 Innovative Concept WP

29 Milestones
4 L2
25 L3

FY12 Work Scope and Tasks (1/3)

- **Advanced Process Development**
  - Improve direct converted oxide feedstock characteristics to support fabrication process simplification
  - Demonstrate ability to predict and improve dimensional control of pellets during sintering (*e.g. reduce hourglassing*) in coordination with sintering modeling to improve safety and reliability
  - Demonstrate fabrication high quality pellets from low loss feedstocks to support reduced defects and potential for enhanced safety
  - Demonstrate fabrication of pellets with controlled microstructure for performance by design - support modeling with data from well characterized microstructures
  - Complete MA-MOX sintering kinetics write-up (FY-11)

FY12 Work Scope and Tasks (2/3)

- **Properties and Advanced Characterization**
  - Provide a framework for modeling the thermokinetics of UO_2 oxidation, including a definition of relevant temperature and atmosphere conditions as well as reaction mechanisms
  - Generate initial experimental datasets describing the reaction of UO_2 with water vapor at high temperature to drive thermokinetic modeling efforts
  - Construct thermophysical property datasets linking quantitative degradation of UO_2 thermal conductivity to hyperstoichiometry, cation species, and Xe content/ morphology
- **Performance and Separate Effects Testing**
  - Continue refinement of ceramic fuels Separate Effects Test plan to leverage available experimental capabilities to inform and benchmark ongoing modeling and simulation work
  - Fabricate initial samples for first HFIR rabbit test to provide insight into the effect of neutron damage on thermal conductivity of oxide ceramic fuels

FY12 Work Scope and Tasks (3/3)

- **Reference Materials and Technique Development**
  - Thermochemical models of the U-Ce-O and U-Nd-O systems will be optimized using new experimental data
  - Growth of single, bi and multi-crystals to support fundamental studies and provide reference materials for separate effects testing
  - Thick film growth of urania and urania compounds to evaluate FG evolution, migration and effects on fuel properties
- **Innovative Fuels Concept: Fission Gas Getter**
  - Select leading cadidate getter material and establish Environmental Qualification Envelope (pressure, temperature, radiation and chemical interaction)

ALTERNATIVE HIGHLIGHTS

BACKUP MATERIAL