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Title: Ceramic Fuels: Separate Effects Test Plan & Thermophysical Properties Update

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Paris, France
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Los Alamos NATIONAL LABORATORY
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Ceramic Fuels: Separate Effects Test Plan & Thermophysical Properties Update

A.T. Nelson, K.J. McClellan, S.L. Voit

The emphasis of U.S. work in ceramic nuclear fuels under the Fuel Cycle Research and Development (FCRD) Advanced Fuels Campaign (AFC) has shifted to predominantly fundamental research under the Separate Effects paradigm. A summary of proposed FY12 ceramics fuel work in this area will be presented. Potential leveraging of a Separate Effects approach to address Light Water Reactor accident tolerance will be discussed in context of FY12 planning and existing work packages. Finally, an update on work in the area of basic thermophysical property studies will be presented.

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Fuel Cycle Research and Development

Ceramic Fuels: Separate Effects Test Plan & Thermophysical Properties Update

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Global Actinide Cycle International Demonstration Technical Meeting
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Outline

- **Ceramic fuels SET plan update**
  - *Fabrication of samples for first HFIR rabbit irradiation*
  - *FY12: Update and prioritization of ceramic fuels SET plan*
  - *SET approach to performance/design of accident tolerant fuels*
  - *FY12: Kinetics of UO_2 oxidation in steam*
- **Thermophysical properties of ceramic fuels update**
  - *Thermal conductivity of solid solution oxides*
  - *FY12: Thermal conductivity of UO_{2+X}*
  - *FY12: Thermal conductivity of Xe-implanted UO_2 thick films*

Ceramic Fuels SET Plan

- **FY11 work provided initial ceramic fuel irradiation test plan and funding fabrication of initial sample set**
- **Importance of thermal conductivity to ceramic fuel performance prompted focus on irradiation defects' role in degradation of thermal conductivity as a function of processing & chemistry**

Traditional Compositions	MDD Compositions
Single Crystal UO_2	MDD UO_2
AREVA UO_2	95/5 MDD $(U,Ce)O_2$
95/5 SS $(U,Ce)O_2$	80/20 MDD $(U,Ce)O_2$
80/20 SS $(U,Ce)O_2$	95/5 MDD UO_2 / Nd_2O_3
95/5 CP $(U,Ce)O_2$	80/20 MDD UO_2 / Nd_2O_3
80/20 CP $(U,Ce)O_2$	95/5 MDD $(Th,U)O_2$
	80/20 MDD $(Th,U)O_2$

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Ceramic Fuels SET Plan

- Selected compositions and processing routes will be fabricated to 'traceable' O/M
- Goal is three doses ~(0.01, 0.1, 1.0 dpa) and three temperatures (minimum possible, 600-1000 K, < 1300 K)
- Preference is use of LFA at LANL for measurement of diffusivity change following irradiation… but not required
- Integrated TEM to characterize generated defect structures greatly strengthens applicability to fundamental science

Successful execution of this study will provide unprecedented detail on the types and production rates of neutron-generated damage in oxide fuels and the associated degradation of thermal conductivity

Ceramic Fuels SET Plan: FY11 Accomplishments

- Received authorization for dUO_2 / ThO_2 operations in LANL Fuels Research Lab (455)
- Baseline irradiation plan was developed with an emphasis on initiation of rabbit testing in FY12
- Initial fabrication of sample compositions and characterization of resultant microstructures completed
- Quality assurance document completed and all criteria / controls approved by FCRD QA program
- Baseline characterization of thermal transport in unirradiated samples intended for initial HFIR irradiation completed

Ceramic Fuels SET Plan: FY12 Planned Activities

- **L2 Milestone for fabrication pushed to March**
  - *FY11 carryover funding supports fabrication and analysis*
  - *Allows for further optimization of sample microstructures and verification of chemistry, unirradiated properties*
- **No funding for engineering, design support or further sample fabrication**
- **Funding for further refinement of ceramic fuels SET plan**

Ceramic Fuels SET Plan: Fundamental 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

All modules describe actors of significant and interrelated importance to understanding the response of a fuel form (UO_2, ThO_2, UN, etc) across all components of the fuel cycle.

BUT... construction / execution of test plan must be balanced against other factors

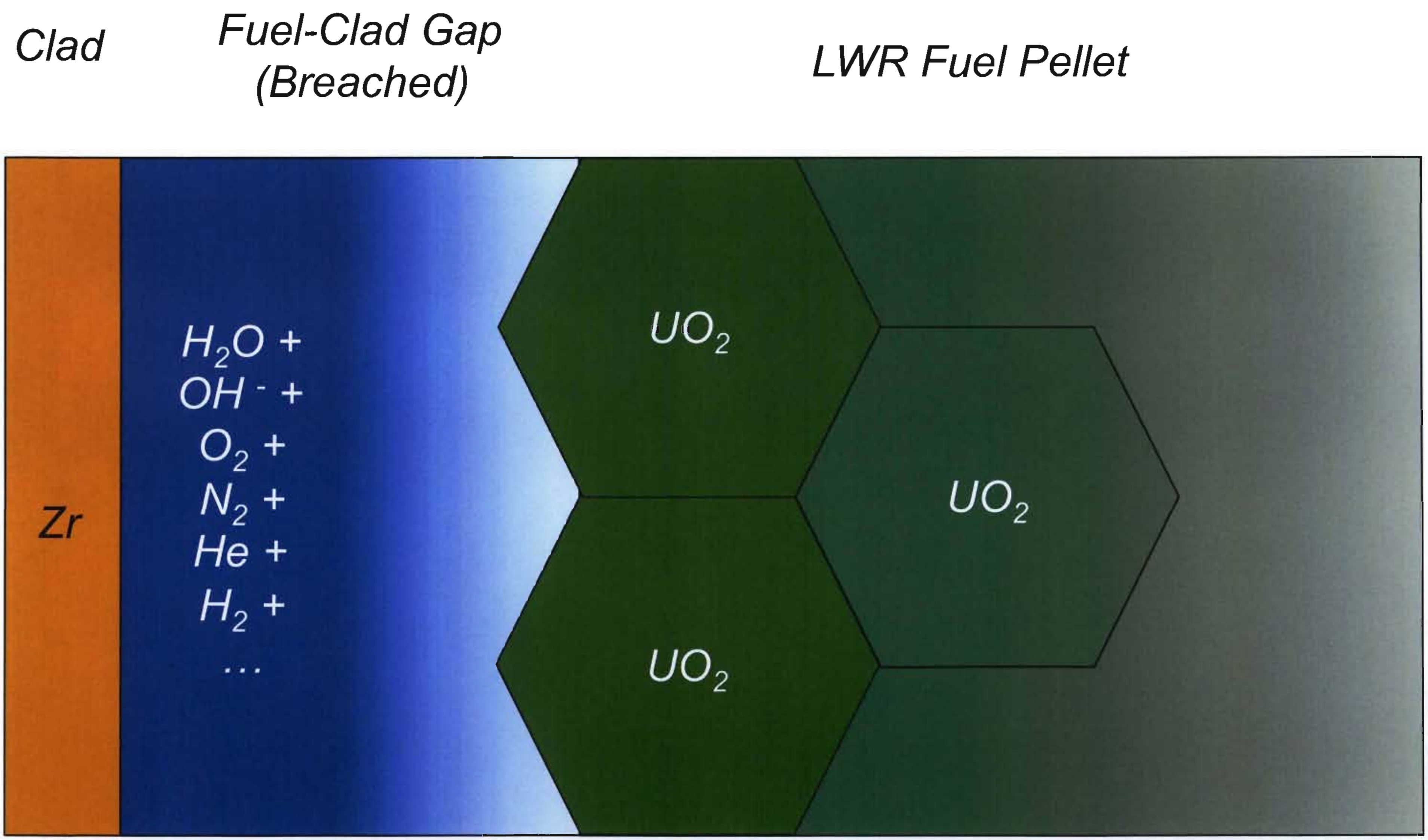
Ceramic Fuels SET Plan: Constructing FY12/FY13 Goals

- Update of ceramic fuels SET plan in FY11 included timeline and effort prioritization based on unified approach to providing fundamental data for all areas of modeling and simulation
- Focus of FY12 and FY13 work will be refinement and recommendations based upon most urgent needs of modeling and simulation coupled with near term infrastructure, budgetary, and political realities

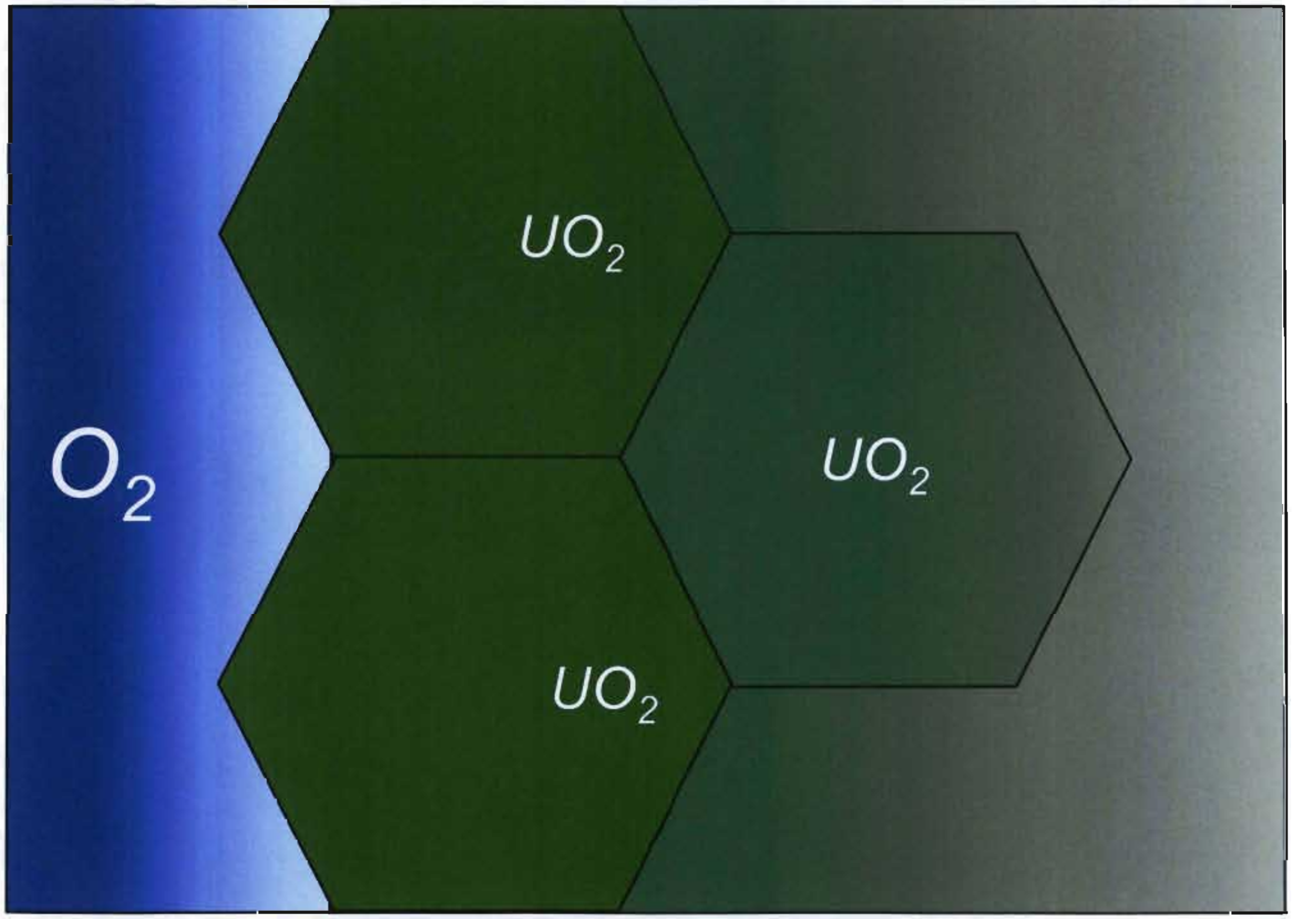
Ceramic Fuels SET Plan: Constructing FY12/FY13 Goals

- **Presently area of immediate attention is performance of UO_2 under accident scenarios**
- **All modules discussed are involved in accident performance, but some can be prioritized based on criteria of previous slide**
  - *Test reactor irradiations & extensive PIE unlikely in short term*
  - *Certain areas of modeling, application to length scales more mature than others*
- **Investigating UO_2 response to clad failure can be considered in terms of SET modules**
  - *Thermochemistry (O-U binary phase stability & kinetics)*
  - *Microstructural evolution (species transport: oxygen)*

SET Approach to LWR Accident Tolerance

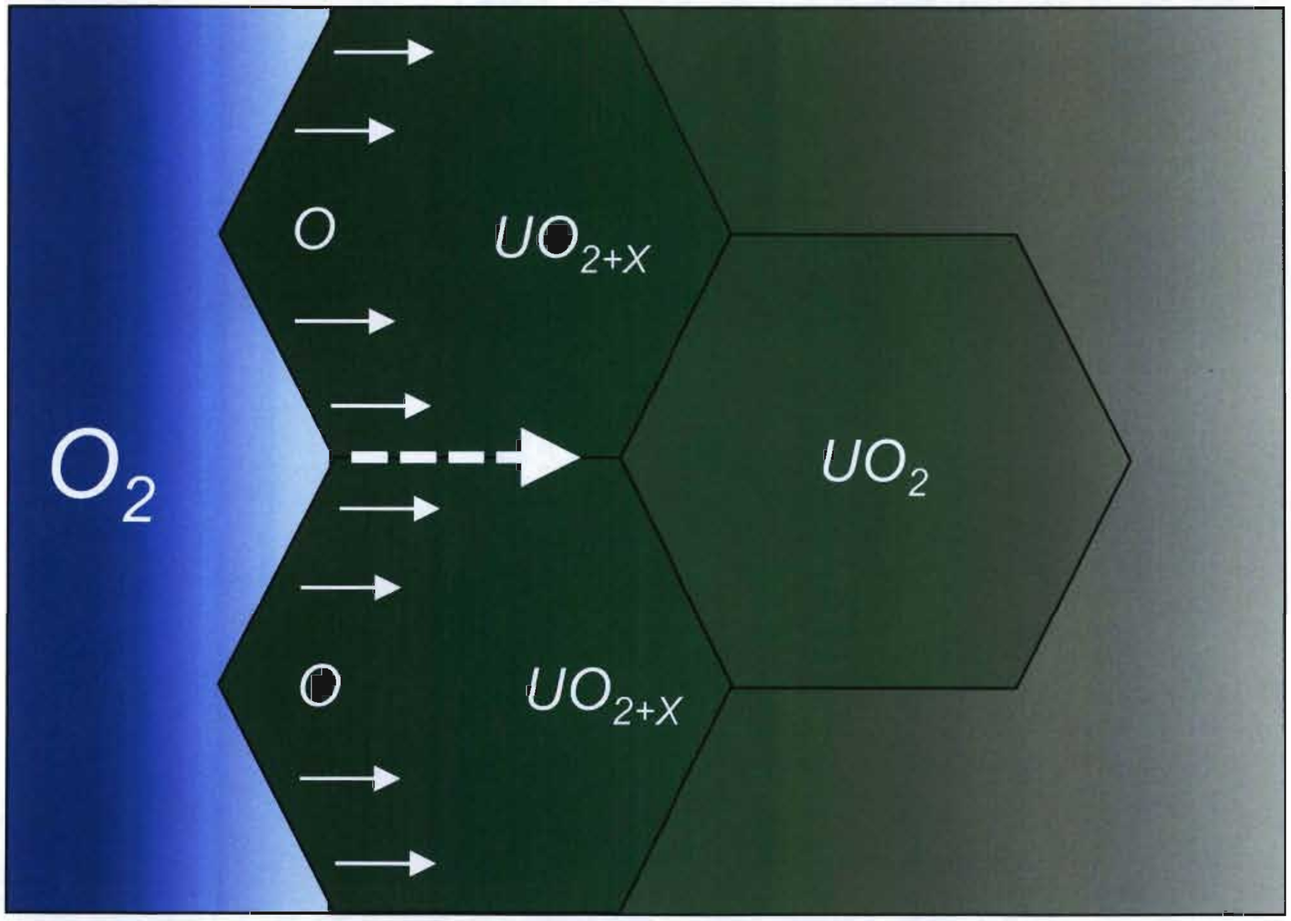


LWR Fuel Response to Cladding Failure



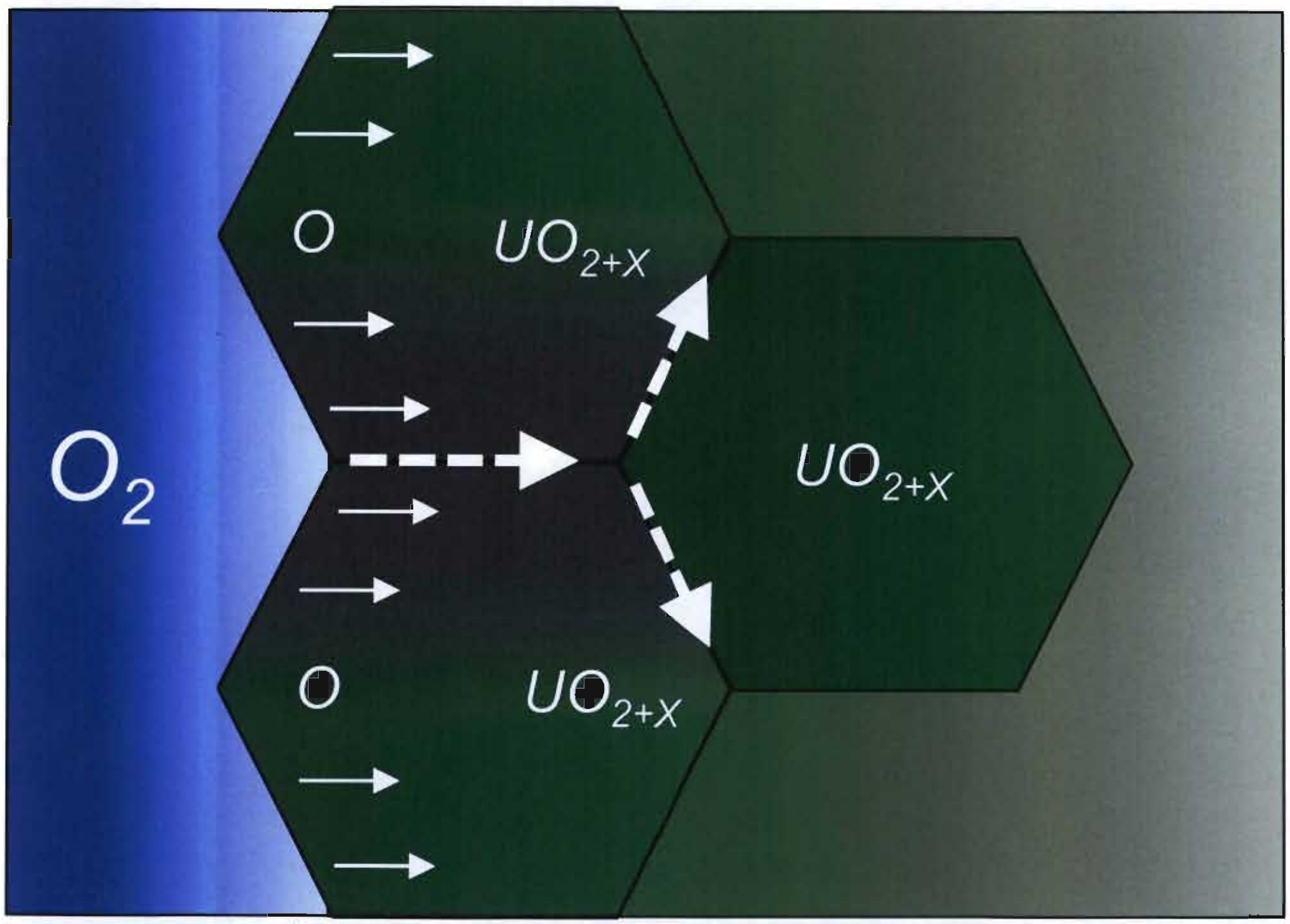
- To first order, system is oxygen source and urania
- Initial state of fuel is dominantly UO_2 grains

LWR Fuel Response to Cladding Failure



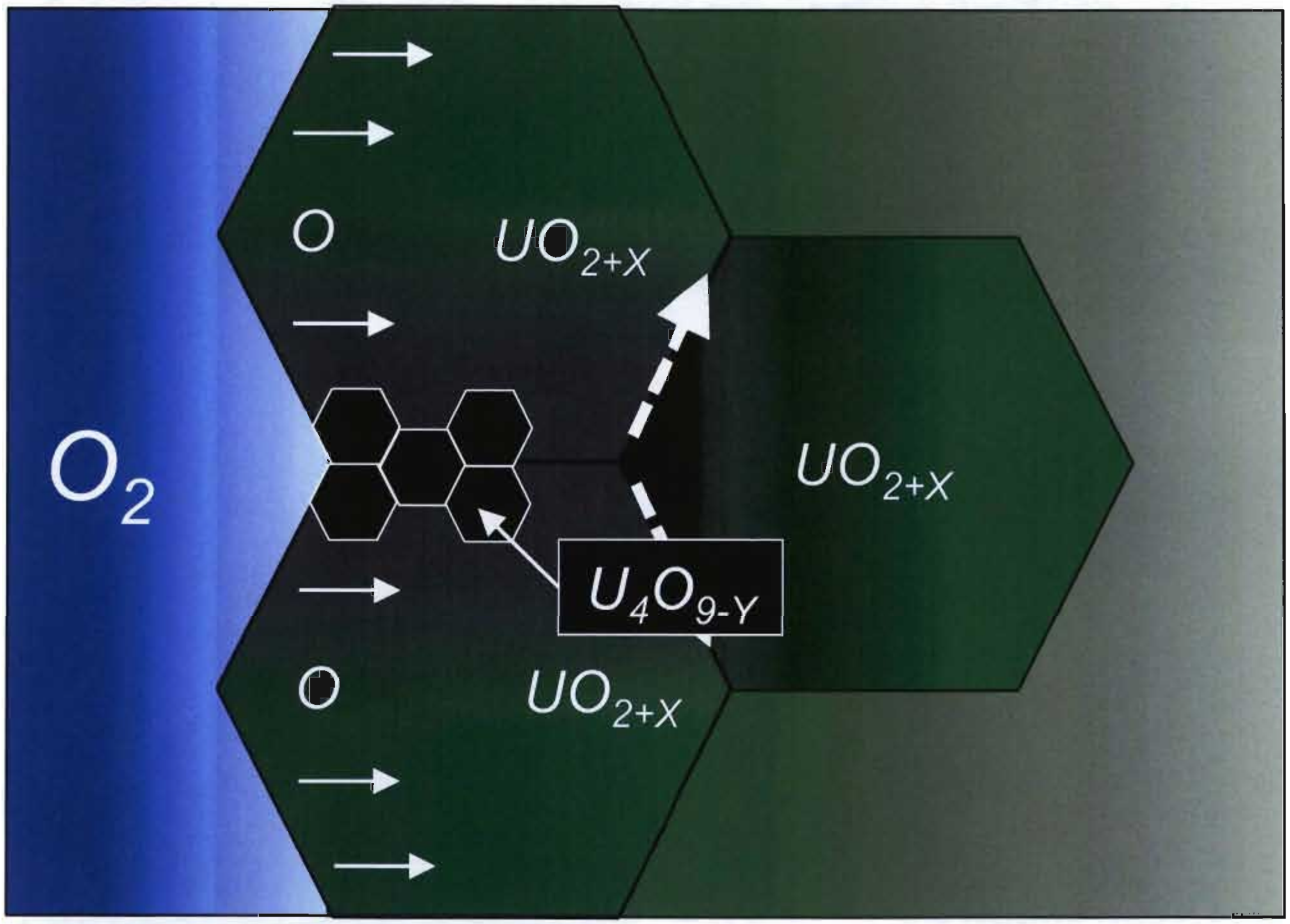
- Oxygen diffusion into matrix begins at surface as available
- Grain boundary diffusion transports oxygen more rapidly into pellet interior

LWR Fuel Response to Cladding Failure



- When source of oxygen is extreme, difference in mobility greatly impacts resulting distribution
- Previous studies performed in air reveal significant segregation along boundaries

LWR Fuel Response to Cladding Failure



- When solubility limit of fluorite phase is exceeded, higher oxide phase begins to precipitate
- U_4O_{9-Y} precipitation along grain boundaries generates strain within lattice

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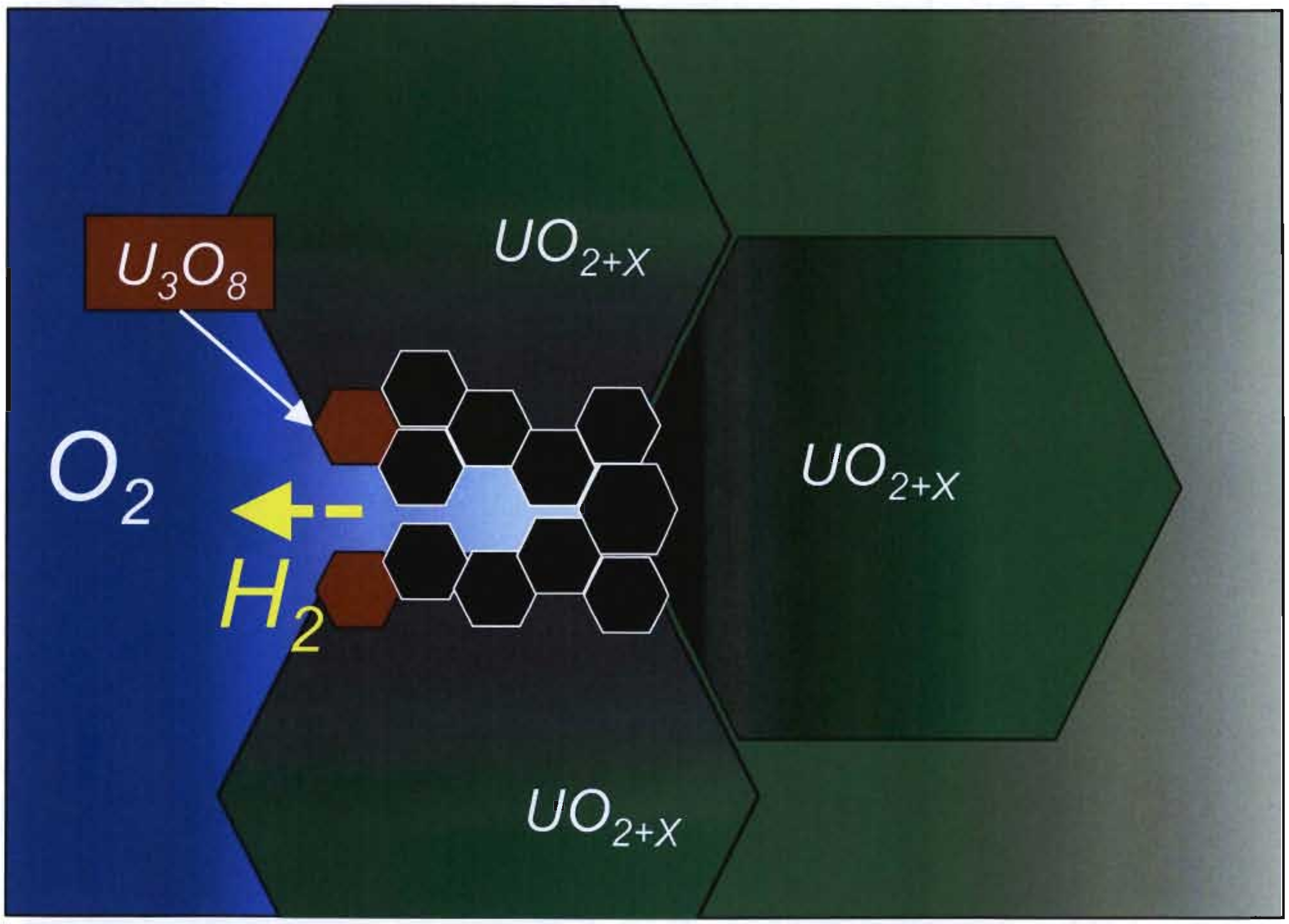
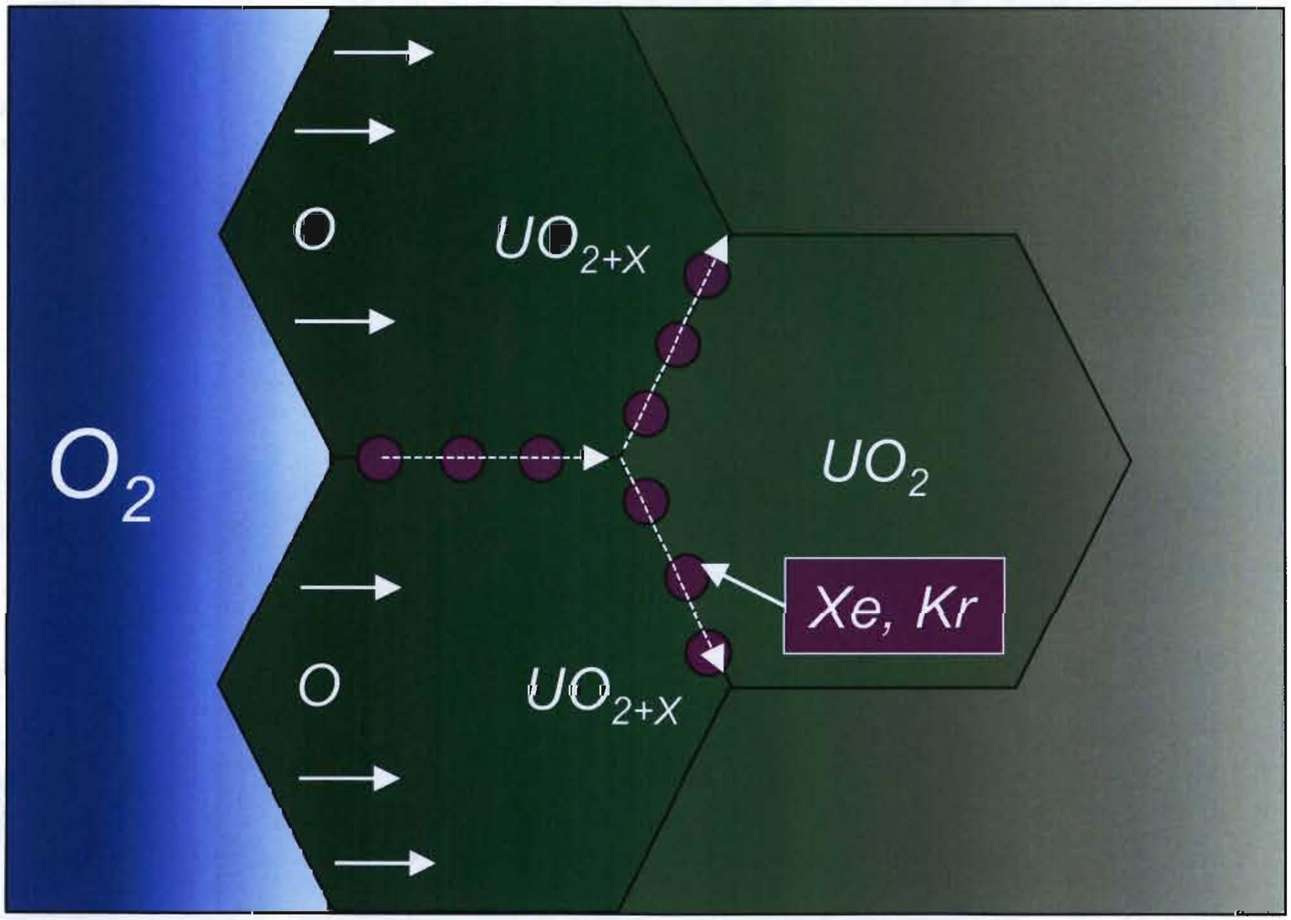
LWR Fuel Response to Cladding Failure



- Pellet cracking increases surface area of available for oxidation
- More rapid oxidation results in additional H_2 production
- Loss of pellet mechanical integrity intensifies

LWR Fuel Response to Cladding Failure



- Similar fast diffusion profiles may be opened up by fission gas accumulation on grain boundaries

Kinetics of UO_2 Oxidation in Representative LWR Accident Atmospheres

- Driven by renewed interest in LWR performance under accident scenarios, ceramics campaign has integrated characterization of UO_2 response to water vapor
- Acquisition and installation of water vapor furnace for STA supported by LANL LDRD funds in late FY11
- *Note STA (±1 ng/cm^2) and traditional test loops have complimentary application ranges*
- System is currently being used to investigate conventional cladding alloys (Maloy talk)
- Extension to investigate oxidation of UO_2 as a function of environmental and sample variables is a major LANL/ORNL FY12 focus within campaign

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Kinetics of UO_2 Oxidation in Representative LWR Accident Atmospheres

- Baseline studies for FY12: effect of water vapor content (5-95%) and temperature (RT-1250°C) on oxidation of UO_2 and development of kinetic modeling framework
- Straightforward extension: incorporation of secondary cation effects e.g. $(U,Ce)O_2$ or $(Th,U)O_2$
- Mid term adaptation: examination of coupled clad/fuel effects
- Longer term adaptation:
  - *Characterization of candidate accident-tolerant systems*
  - *Evaluation of irradiation effects on kinetics*
  - *Fission gas effects on oxidation (potential use of Xe-implanted films)*
- Pressure effects must be considered

Application of SET Approach to Accident Tolerance

- **Motivation – if blocking (limiting) short circuit oxygen diffusion within UO_2 is possible, fuel performance during transient/accident scenarios will be enhanced**
  - *Rapid loss of pellet mechanical integrity will be delayed*
  - *Release of solid fission products*
- **Potential solution is grain boundary engineering**
  - *Provide impurity atoms that segregate to grain boundaries and block oxygen diffusion*
  - *Synthesize small quantities of second phases that decorate grain boundaries and block, getter oxygen*
- **Must verify that any such approaches do not meaningfully impact other areas critical to fuel performance**
  - *Fabrication*
  - *Thermal transport*
  - *Evolution of in-pile fuel properties/performance*

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Motivation: AFC Core Ceramics Experimental Efforts

AFC CORE EXPERIMENTAL PROGRAMS
E.G. THERMAL TRANSPORT IN UO_2 AS A FUNCTION OF X

INFORM / BENCHMARK MODELING AND SIMULATION

DRIVE ENHANCED FUNDAMENTAL NUCLEAR MATERIALS THEORY

FACILITATE NEXT GENERATION FUEL PERFORMANCE CODES

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FACILITATE NEXT GENERATION FUEL PERFORMANCE CODES

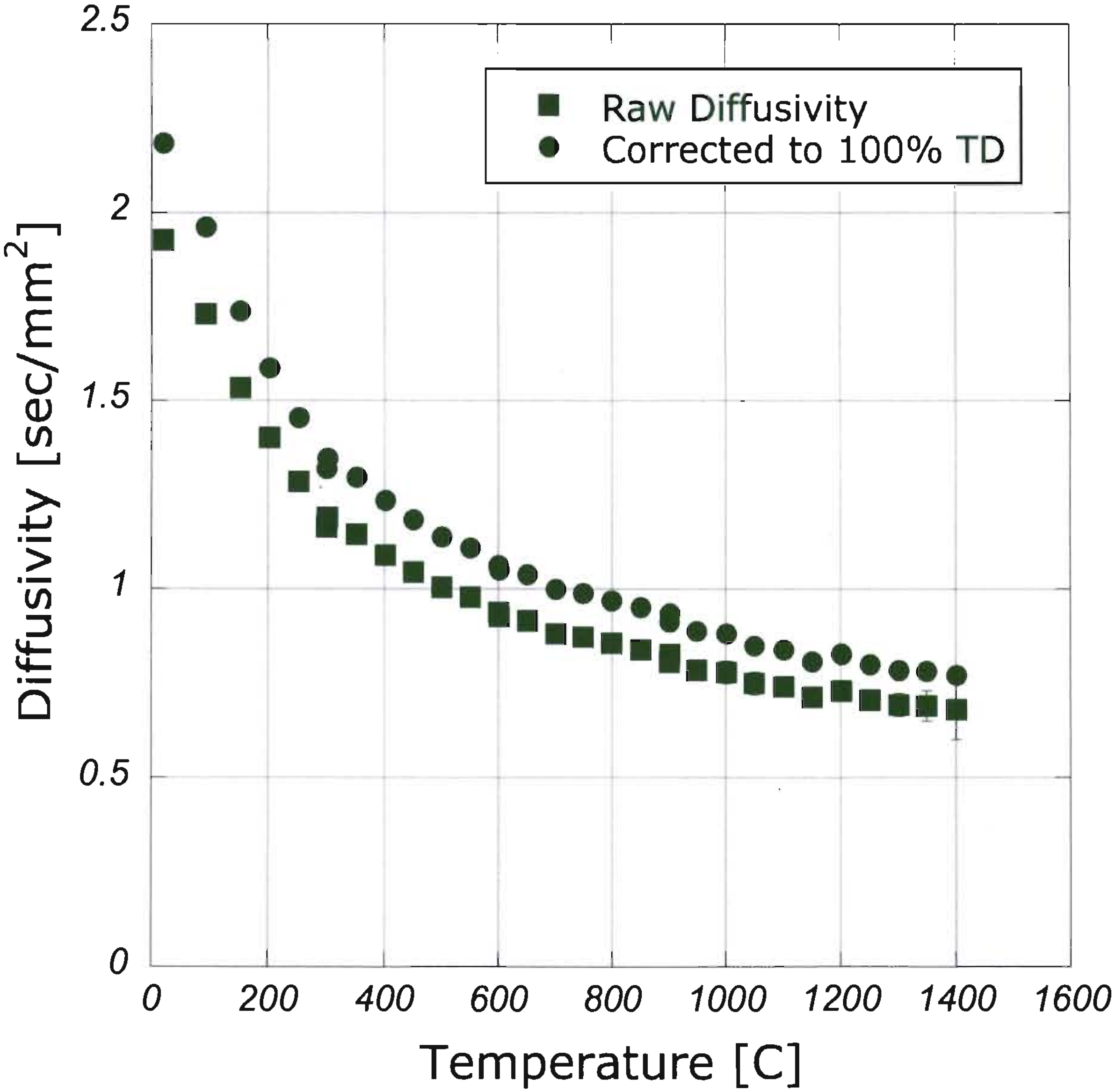
IMPROVE ABILITY TO MODEL / EVALUATE EXISTING LICENSURE AND REGULATORY REQUIREMENTS

IMPROVE ABILITY TO MODEL / EVALUATE UNANTICIPATED REACTOR CONDITIONS

IMPROVE ABILITY TO MODEL / EVALUATE EVOLUTIONARY IMPROVEMENTS TO STEADY STATE REACTOR OPERATION

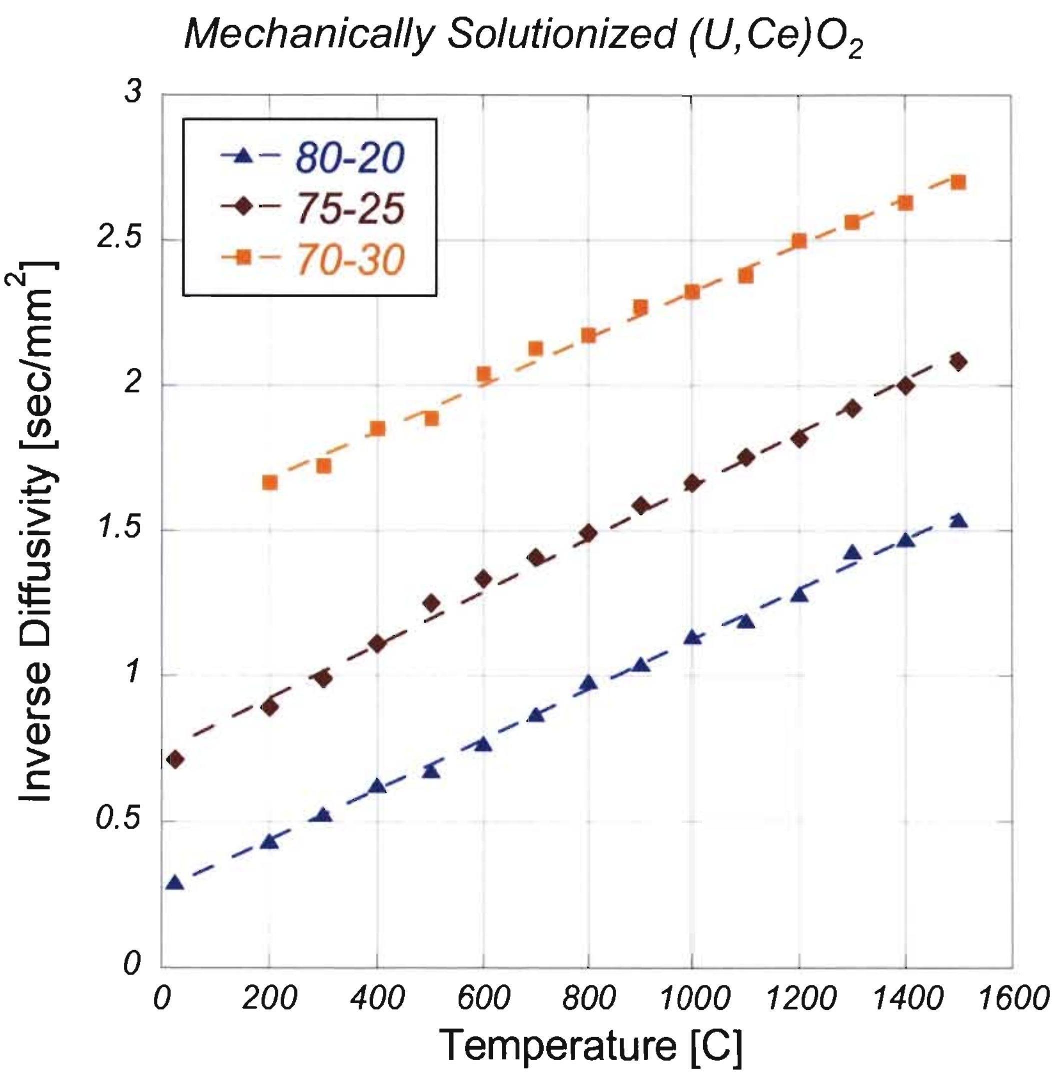
ULTIMATE MOTIVATION: **DESIGN 'BETTER' FUELS AND REACTOR SYSTEMS**

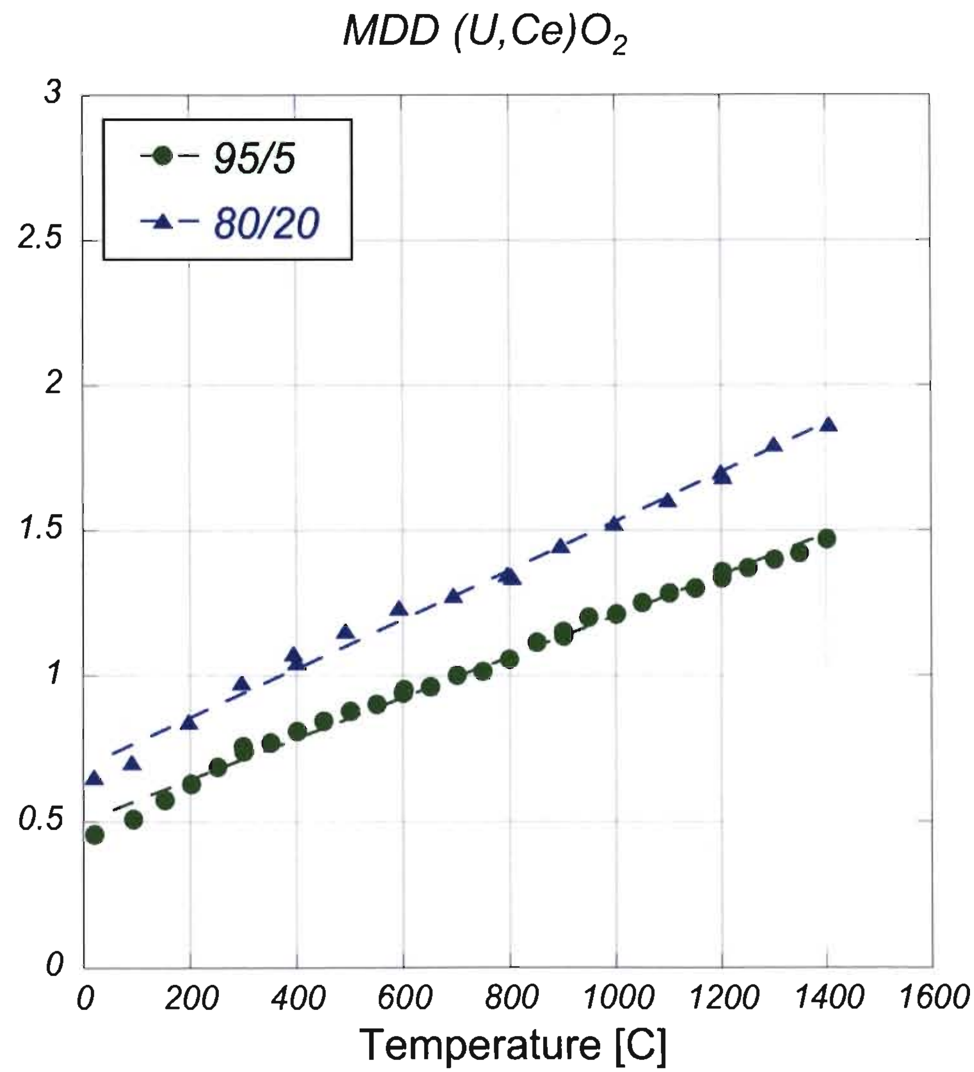
Thermal Diffusivity of MDD Oxide Solutions



- Investigating cation charge/mass effects on thermal transport in $(U,X)O_2$
- MDD powders synthesized by ORNL pressed and sintered for SET development
- 95/5 $(U,Ce)O_2$ shown at left typical of LFA curves

Thermal Diffusivity of MDD Oxide Solutions





Thermal Diffusivity of MDD Oxide Solutions

- MDD $(U,Ce)O_2$ as measured thus far contains far inferior thermal transport behavior compared to mechanically synthesized material
- Pending technique refinement may reduce uncertainty at high temperatures (> 1200°C) but discrepancy exists at room temperature
- Synthesis and analysis of additional samples is ongoing and may provide further insight into discrepancy

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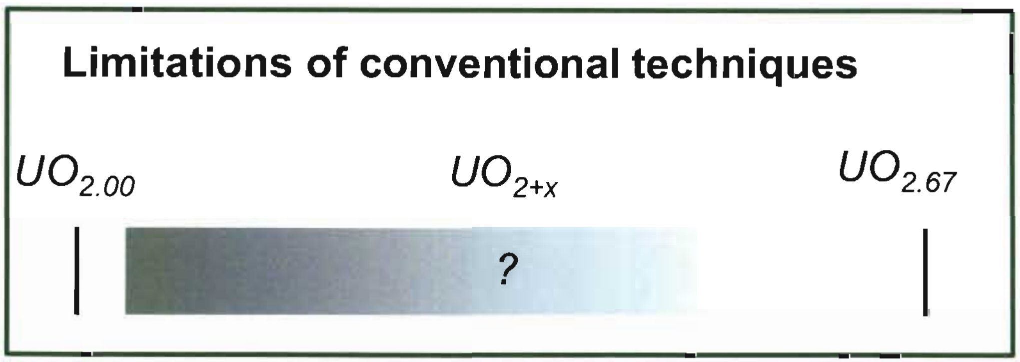
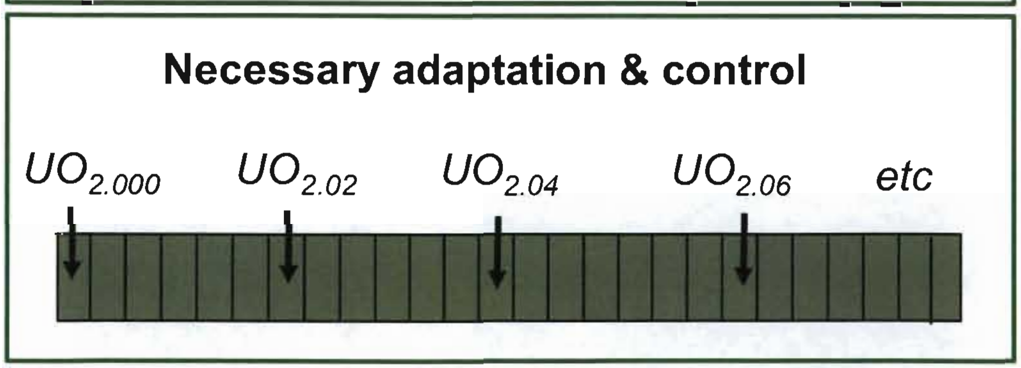
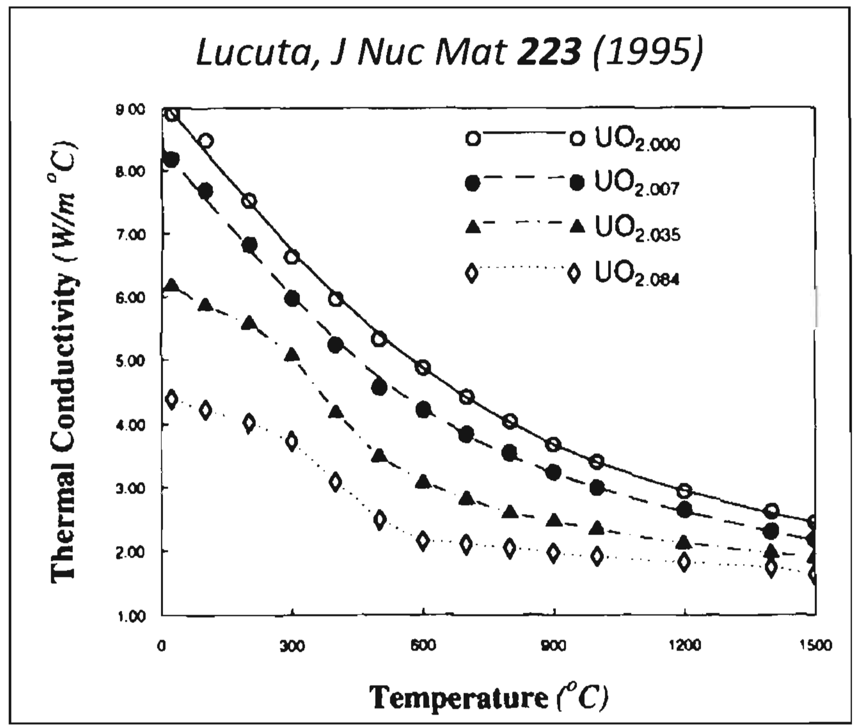
Thermal Conductivity of Novel Systems: UO_{2+X}

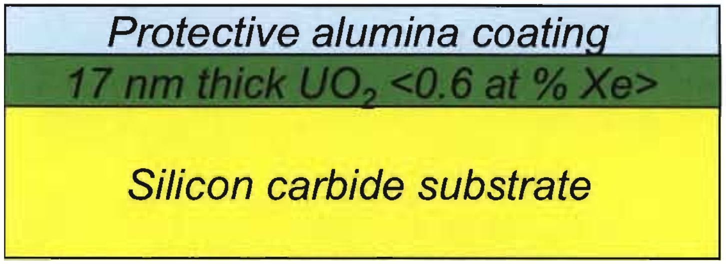
- Installation of dynamic atmosphere controls is currently underway on high temperature LFA (2000°C)
- Demonstration and verification of technique to measure UO_{2+X} with fixed stoichiometry early FY12, deliverable for measurements August



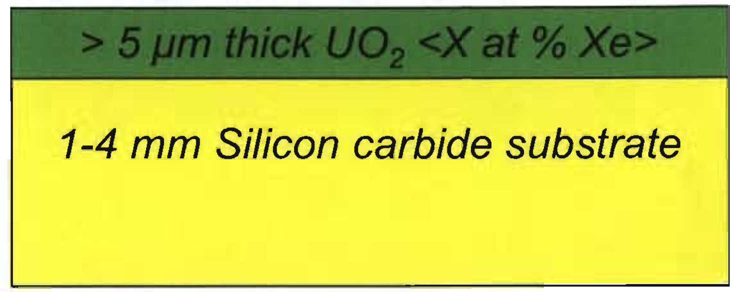




Thermal Conductivity of Novel Systems: UO_2+Xe



Current Xe-implanted thick film geometry



Geometry required for LFA analysis of Xe-implanted thick films

- **Multilayer LFA technique allows for in situ measurement of thermal conductivity of UO_2 films containing Xe in various morphologies**
- **When matched with structural and chemical analysis of films, significant insight into the role of Xe on thermal conductivity of oxide fuels**
- **Significant FY12 emphasis on synthesis and measurement**

Near Term Milestones / Activities

- November: Two L3 (melt point and thermal transport) reports due
- Dec-Jan: Focus will be initial exploration of oxidation kinetics of UO_2, and installation of dynamic atmosphere controls on LFA, and optimization of microstructures for HFIR rabbit irradiations
- Feb-Mar: Finalization of HFIR sample fabrication, execution of initial O/M studies and oxidation kinetics work in UO_2 system