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***Updated FY12
Ceramic Fuels
Irradiation Test Plan***

LA-UR 12-X

***Prepared for
U.S. Department of Energy
FC R&D Program***

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EXECUTIVE SUMMARY

The Fuel Cycle Research and Development program is currently devoting resources to study of numerous fuel types with the aim of furthering understanding applicable to a range of reactors and fuel cycles. In FY11, effort within the ceramic fuels campaign focused on planning and preparation for a series of rabbit irradiations to be conducted at the High Flux Isotope Reactor located at Oak Ridge National Laboratory. The emphasis of these planned tests was to study the evolution of thermal conductivity in uranium dioxide and derivative compositions as a function of damage induced by neutron damage. Current fiscal realities have resulted in a scenario where completion of the planned rabbit irradiations is unlikely.

Possibilities for execution of irradiation testing within the ceramic fuels campaign in the next several years will thus likely be restricted to avenues where strong synergies exist both within and outside the Fuel Cycle Research and Development program. Opportunities to augment the interests and needs of modeling, advanced characterization, and other campaigns present the most likely avenues for further work. These possibilities will be pursued with the hope of securing future funding.

Utilization of synthetic microstructures prepared to better understand the most relevant actors encountered during irradiation of ceramic fuels thus represents the ceramic fuel campaign's most efficient means to enhance understanding of fuel response to burnup. This approach offers many of the favorable attributes embraced by the Separate Effects Testing paradigm, namely production of samples suitable to study specific, isolated phenomena. The recent success of xenon-imbedded thick films is representative of this approach. In the coming years, this strategy will be expanded to address a wider range of problems in conjunction with use of national user facilities' novel characterization techniques to best utilize programmatic resources to support a science-based research program.

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ACRONYMS

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|--------|---------------------------------------------|
| APS | Advanced Photon Source |
| FCRD | Fuel Cycle Research and Development Program |
| HFIR | High Flux Isotope Reactor |
| INL | Idaho National Laboratory |
| LANL | Los Alamos National Laboratory |
| LANSCE | Los Alamos Neutron Science Center |
| LFA | Laser Flash Analysis |
| O/M | Oxygen to Metal Ratio |
| ORNL | Oak Ridge National Laboratory |
| PIE | Post Irradiation Examination |
| SET | Separate Effects Testing |

1. Current Status of Ceramic Fuel Irradiation Campaigns within the Department of Energy Complex

The Fuel Cycle Research and Development (FCRD) program is currently devoting resources to study of numerous fuel types with the aim of furthering understanding applicable to a range of reactors and fuel cycles. Current fiscal realities have combined with the events at Fukushima Daiichi in March of 2011 to create a situation where the bulk of national resources provided to nuclear materials research will be devoted to programs focused on enhanced accident tolerance. Such endeavors, regardless of the system or techniques advocated, are largely dependent first on establishing superior performance from a corrosion standpoint. A fuel/cladding system demonstrated to be principally inert when exposed to water vapor at temperatures to 1500°C would represent a supreme advantage in light water reactor loss of coolant accidents. Any new, unproven fuel/clad system would undoubtedly require test irradiations to establish in-pile performance, but the preliminary focus of experimental efforts will be the execution of corrosion testing to vet proposed concepts and demonstrate performance advantages.

It is against this backdrop that traditional ceramic fuels, principally uranium dioxide, must successfully justify the dedication of considerable resources for the many facets required in planning, design, fabrication, execution, and examination of materials included in even the most basic irradiation campaign. In contrast to metallic fuels or various composite options, uranium based ceramic fuels occupy the position of needing no development for deployment, as they have been used nearly exclusively for many decades by the nuclear industry. However, this stature also dictates that these fuel forms are the most relevant to both the existing industry and commercial builds initiating in the coming decades. Uranium dioxide's prevalence has also resulted in significant attention from modelers and theorists. The broad goal of this community is development of an integrated performance code that unites neutronics and thermal hydraulics with material performance across length scales in order to predict the response of core materials under all envisioned reactor states. To be truly revolutionary, such a code would replace existing empirical relations that provide extrapolated property data as a function of engineering parameters with science based theory that instead supplies a given fuel property (e.g. fracture toughness, fission gas release rates, thermal conductivity) based on intrinsic local material parameters (e.g. anion and cation chemistry, defect chemistry, stress state, microstructure).

The primary aim in development of any irradiation plan focused on uranium dioxide is thus intimate integration with modeling, simulation, and theory to both inform and validate their efforts. Researchers working in these areas during the past decade have increasingly expressed concern regarding the nature of experimental data historically produced by the nuclear materials community. Archival data sets were often acquired solely with the qualification of a given fuel chemistry and reactor in mind with little note to properties or factors deemed nonessential. Even in cases where property studies of interest were executed, the omission of details now known to be critical to understanding material performance often limits applicability of such results within a modern framework.

This gap between the data needs of modelers employing modern computational techniques to better understand the behavior of nuclear fuels and what the experimental community has conventionally produced prompted the FCRD program to hold a series of workshops in 2010 and 2011. The aim of these meetings was to convene modelers and experimentalists within the nuclear materials community in order to produce a unified approach to addressing this challenge.

FCRD Ceramic Fuels Irradiation Test Plan

The conclusions of the meeting and recommendations grown out of imbedded discussions became the FCRD Advanced Fuels Separate Effects Test (SET) Research and Development Plan. Initial thoughts were put forth describing the methodology upon which experimental plans would be designed and executed in order to best integrate with both existing and planned modeling efforts. The term ‘Separate Effects’ is used to denote experiments that focus on understanding and cataloging the role of a single variable on a single property in order to maximize the utility of the resultant data set to the modeling community.

This document presents the current status and priorities of the FCRD ceramic fuels irradiation campaign as integrated into a larger Separate Effects philosophy. A pragmatic view of the funding outlook for test irradiations is taken looking toward FY13 and beyond. While it is possible that world or national events may instigate yet another sea change in priorities with respect to nuclear energy and nuclear materials research and development, discussion here centers on the assumption that funding levels and distribution within the FCRD campaign will remain essentially flat.

2. Update on Ongoing and Foreseeable Irradiation Tests

In FY11, effort within the Ceramic Fuels campaign focused on planning and preparation for a series of rabbit irradiations to be conducted at the High Flux Isotope Reactor (HFIR) located at Oak Ridge National Laboratory (ORNL). The emphasis of these planned tests was to study the evolution of thermal conductivity in uranium dioxide and derivative compositions as a function of damage induced by neutron damage. Thermal conductivity degradation in ceramic fuels brought about by disturbances to lattice order through structural damage is one of the least understood aspects contributing to the evolution of thermal conductivity in uranium dioxide fuel.

Although this mechanism can be significant, to date it has yet to be studied. This is due to the fact that the only means to produce samples where the effects of lattice damage on thermal transport could be studied would be very short test irradiations performed on materials with negligible fissile isotope contents. Integral tests performed in the traditional manner will undergo many other drastic structural and chemical evolutions that may either degrade or enhance thermal transport. Decoupling of these many effects to isolate the contribution of a single actor proves challenging if not impossible.

The FCRD Ceramic Fuels campaign thus proposed a series of irradiations utilizing the rabbit irradiation capability at HFIR in order to attack this challenging problem. This facility allows for irradiation of material for comparatively short periods of time (hours to hundreds of hours) as is necessary to view the formation and influence of defects in ceramic fuels before they become obscured by the numerous and complex other evolutions within a fuel. Depleted uranium would be used to eliminate fissions from clouding the defect generation mechanisms. Irradiation of carefully controlled microstructures and chemistries of uranium dioxide and derivative compositions chosen for maximum relevance to the modeling community would provide insight into the role of specific defect structures on phonon scattering. Samples were characterized before irradiation through both laser flash analysis, and transmission electron microscopy efforts were planned. Following irradiation to different doses at different temperatures, the same analyses were to be repeated in order to gain a clear understanding of what specific defect types were formed as a function of the irradiation variables (dose and temperature) and sample chemistries. Thermal diffusivity following irradiation coupled with graduated thermal anneals will provide the final piece of information necessary to provide modelers with the first major piece of experimental data needed to simulate the basic evolutions of oxide fuels under neutron irradiation. Details on the planning and execution of the irradiation can be found in previous FCRD reports [1-3].

Work in FY11 and continuing into FY12 at Los Alamos National Laboratory (LANL) centered on development and refinement of fabrication techniques to produce samples of known and highly controlled chemistries and microstructures for the first set of irradiations. A milestone encompassing fabrication and characterization of the initial sample set was completed in March of 2012. These samples were then intended to be shipped to ORNL for insertion in the rabbit irradiation vehicle to meet an early-FY13 insertion schedule.

Unfortunately, further programmatic support for the irradiation was not available in FY12. Appreciable work by ORNL staff is still needed in the areas of irradiation capsule design and irradiation planning; administrative work is also required at LANL to develop approvals and controls for analysis of irradiated fuels. Should executing this characterization at the Fuels Research Laboratory at LANL prove untenable, collaboration with Idaho National Laboratory (INL) staff for sample analysis is viewed as the fallback option.

FCRD Ceramic Fuels Irradiation Test Plan

Without the campaign's support in these areas, it is not possible to execute any of the multiple planned test irradiations. Capsule design is of particular importance given its role in driving the temperature of the samples during irradiation. Integration of sample fabrication efforts at LANL with capsule design at ORNL is vital, as not only must samples meet the geometric restrictions imposed by the capsule, but must also be sized in conjunction with calculated heating rates and gas pressures to maintain the desired irradiation temperature. At present it appears that funding for irradiation design efforts at ORNL is unlikely, suggesting that advancement of the planned irradiations is unlikely in the FY13/FY14 timeframe.

Despite this, the ceramics campaign continues to pursue other opportunities to execute test irradiations. The most likely mechanism by which this will be achievable is integration with other areas of interest both within the FCRD campaign as well as other organizations within DOE-NE. An example of the latter is the problem presented by missing pellet surfaces. Quality assurance of fuel pellet geometry, while extensively studied by fuel vendors, cannot ever hope to ensure reactor operators that all pellets are delivered in perfect geometries. Some small fraction will satisfy all checks yet possess small chips, thus deviating from the prescribed geometries. In pile, these anomalies will perturb the thermal and mechanical response of the fuel/clad system.

This problem was one of the first attacked by the Nuclear Energy Advanced Modeling and Simulation (NEAMS) program. Computational efforts have provided detailed spatial and temporal predictions of the effect that missing pellet surfaces will have on a fuel rod during irradiation. Their predictions suggest that the effect may be extreme in some circumstances. However, at present it is not possible to benchmark these models.

The opportunity to design a test irradiation to focus on study of the effects of missing pellet surfaces could prove an interesting opportunity. Uranium dioxide pellets could be fabricated to industrial norms, with the important exception of the intentional inclusion of various types of defects designed in collaboration with NEAMS predictions. A test irradiation executed to study the effects of missing pellet surfaces or other defects could be planned to not only inform and validate current models, but also serve to demonstrate various proposed advanced post irradiation examination (PIE) and non-destructive evaluation (NDE) techniques. One of the most significant beginning-of-life effects of a missing pellet surface will be substantially increased temperatures generated by the larger fuel-clad gap in the location of the missing surface. The focus of a wide range of advanced PIE proposals has been in situ temperature diagnostics; this type of test may prove to be a significant opportunity for the demonstration and evaluation of these proposals.

The ceramic fuels campaign may also be able to leverage the test irradiation needs of other areas within the FCRD program. As discussed, the near term focus on accident tolerance will likely result in the development of new or modified cladding materials. Revolutionary cladding materials found to possess significant advantages in terms of corrosion performance will clearly require irradiation testing in order to demonstrate to industry that larger scale rod and assembly testing is reasonably assured of success. The requirement to produce ceramic fuels to drive these test irradiations may allow the ceramics campaign the chance to include evolutionary modifications to either reference oxide or nitride fuel. For example, if techniques are found to improve the corrosion performance of uranium nitride through minor impurity additions, it may be possible to include samples of these materials in a test irradiation primarily intended to demonstrate the performance of a new cladding variant. These opportunities will be pursued in collaboration with the Structural Materials campaign as they arise.

3. Opportunities for Advancement Beyond Reactor Irradiations

It is clear that while ceramic fuels may be able to leverage the needs of other programs in the near term to obtain support for reactor irradiations, this is by no means a certainty. Fortunately, the progress made in design and execution of the Separate Effects program has provided means by which many actors encountered during irradiation of ceramic fuels can be studied without requiring the resources of reactor irradiations and associated PIE. The fundamental tenants of an SET approach center around understanding the role that specific phenomenon have in governing system response. In an irradiated nuclear fuel, it is readily possible to list the numerous evolutions encountered during burnup and consider how they will drive system response, whether the response is thermal transport, mechanical properties, or other areas of interest. A summary of the many actors and how they can be considered with respect to different system responses of relevance to ceramic fuels can be found in Reference 4.

Many of the avenues required to understand the structure-property relations of unirradiated ceramic fuel remain equally important to modeling the performance of fuel during irradiation. For example, work currently ongoing within the program to understand oxygen-to-metal ratio (O/M) effects on thermal transport remain equally important to performance in-pile. Similarly, examination and understanding of grain texture and porosity's role on various other properties will remain valid throughout the fuel lifetime. This work will continue within the program core in the foreseeable future.

Transitioning to the many actors that are created by irradiation leads to a different challenge, as historically the only way to generate the chemistry, structures, and effects encountered by a fuel during irradiation has been through execution of integral tests. As mentioned in discussions above, the challenge with such an approach is the complications that arise from separating the contributions of many different and often interlinked changes to the fuel. Within the ceramics campaign approach to SET, the proposed solution to this challenge is the generation of synthetic samples to understand various actors encountered by nuclear fuels during burnup. This approach thus avoids the time and expense of reactor irradiations with the additional advantages of providing samples that both isolate specific phenomena and additionally can be characterized without the controls needed for irradiated nuclear fuel.

This approach will become even more prevalent to advance ceramic fuels research in the current climate. The fabrication of synthetic samples in order to study the various actors encountered during burnup provides an economic and extremely relevant means by which to advance our understanding of ceramic fuels to support ongoing modeling and simulation efforts as well as increase our fundamental understanding of how nuclear fuels evolve during irradiation. The most recent success achieved by LANL has been the development of a technique to grow thick films of uranium dioxide containing xenon gas in controlled morphologies [5]. This approach allows the preparation of samples containing up to several percent xenon in a controlled, characterized manner. Existing modeling work at INL has predicted the effect of fission gas bubbles on thermophysical properties; extension of this work at both INL and LANL to understand other transport phenomena is ongoing. Prior to development of the thick film technique, the only way to produce uranium dioxide samples containing two atomic percent xenon would be execution of an extensive test irradiation performed to roughly ten percent burnup. This would require several years' irradiation time. Furthermore, the samples produced by this technique would possess not only significant radioactivity, but also all the other evolutions of irradiated fuel.

Xenon-embedded uranium dioxide thick films possess none of these disadvantages. Work is ongoing to evaluate the capability of multilayer laser flash analysis (LFA) to measure the degradation of thermal conductivity brought about by xenon bubbles both in solution and collected along grain boundaries. If successfully demonstrated, this approach would represent a revolutionary means by which to evaluate the effect of different xenon, krypton, or other relevant gasses on thermal transport in nuclear fuels.

This philosophy is being extended to other areas relevant to understanding the performance of ceramic fuels during irradiation. The basic question posed within the SET framework is: what changes does a ceramic fuel undergo during irradiation, and is there a means to meaningfully replicate it using modern synthesis techniques? The initial pursuit of thick films was chosen due to its relevance to fuel performance and attention from the modeling and simulation field. Similar criteria can be used to prioritize future efforts. Production of some synthetic structures may not pose a great experimental challenge, but would also be of minimal relevance to either fuel performance or ongoing modeling work. For example, it would be possible to synthesize the ternary oxides formed by fission products during burnup and fully characterize their properties, but this would not be strongly relevant.

One challenge that the campaign is currently considering is the evolutions brought about by the steep radial thermal gradients formed within ceramic fuels. This gradient is responsible for many of the complex evolutions observed within a fuel during burnup, namely the formation of columnar microstructures and the migration of many species. Traditionally, study of the effects of this gradient was achieved through test irradiations. However, it may be possible to build furnaces that could impose a steep gradient (1000°C over 5 mm) on unirradiated depleted uranium dioxide. This capability may provide a means to generate structures and chemical gradients out of pile to produce samples that are much more amenable to analysis using the wide array of modern characterization techniques in comparison to spent fuel. Samples produced in this manner would provide unprecedented detail on thermomigration as well as provide material that could be used to investigate the effect of widely varied structures on thermal or mechanical properties.

Finally, use of techniques to fabricate synthetic microstructures opens the door for collaboration with the many national user facilities to execute scientific programs. Handling and transport of spent fuel requires substantial effort. Depleted uranium, while still requiring some controls, is much easier to ship and characterize at different facilities around the country. Current collaborations with both the Los Alamos Neutron Science Center (LANSCE) and the Advanced Photon Source (APS) at Argonne National Laboratory are set to explore the resolution and capabilities of their techniques to monitor evolutions relevant to understanding the evolution of ceramic fuels during irradiation. Use of neutron or photon tomography may provide a means to map the evolution of grains, porosity, chemical distributions, and crack networks as a function of temperature, stress, or other stimulus.

This union of modern NDE with ceramic fuels research could offer a very powerful tool. For example, if neutron or photon analysis could be combined with a capability to impose a steep temperature gradient on uranium dioxide, it may be possible to temporally and spatially map the changes of microstructure and migration of porosity during reactor startup. Furthermore, imposing even more extreme temperatures, atmospheres, or pressures could mimic accident scenarios. Interactions with cladding, the degradation of the fuel, or expansive fracture and failure of uranium dioxide could be monitored in order to better understand the response of existing or alternative fuel/cladding systems to various scenarios.

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