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Neutron based evaluation in support of NEAMS

Fuel Cycle Research & Development

Prepared for U.S. Department of Energy Campaign or Program

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SUMMARY

The primary objective of the Advanced Non-Destructive fuel Examination (ANDE) work package is to develop capability that has the potential to accelerate insight and development of ceramic and metallic fuels. Establishing unique validation opportunities for new models is a key component of this effort. To explore opportunities a series of interactions were held with NEAMS modelers at LANL. The focus was to identify experiments that draw on the unique capabilities of neutron scattering and imaging for studies of nuclear fuel particularly in areas where experimental data can be valuable for of models validation.

The neutron characterization techniques applied in the ANDE program span length scales from millimeter to micrometer to angstroms. Spatial heterogeneities of interest include cracks, pores and inclusions, crystal structure, phase composition, stoichiometry texture, chemistry and atomic thermal motion. Neutrons offer characterization opportunities that are distinct from other probes such as X-rays, electrons or protons. This report describes a variety of opportunities whereby neutron data can be related to models and lists some opportunities.

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ACRONYMS

AECL	Atomic Energy of Canada Limited
ANDE	Advanced Non-Destructive fuel Examination
APS	Advanced Photon Source
AFC	Advanced Fuel Cycle
ATF	Accident Tolerant Fuel
ATR	Advanced Test Reactor
BWR	Boiling Water Reactor
CCD	Charge-Coupled Device
CEA	French Alternative Energies and Atomic Energy Commission
DOE	Department of Energy
DOT	Department of Transportation
EBS	Ethylene bis-stearate
EPMA	Electron Probe Microanalysis
FCCI	Fuel Cladding Chemical Interaction
FCMI	Fuel Cladding Mechanical Interaction
FCRD	Fuel Cycle Research and Development
FOV	Field of View
FR	Fast Reactor
HEU	Highly Enriched Uranium
HFEF	Hot Fuels Examination Facility
HI	High Intensity
HIPPO	High Pressure/Preferred Orientation neutron diffractometer
IAEA	International Atomic Energy Agency
IMCL	Irradiated Materials Characterization Laboratory
INL	Idaho National Laboratory
JAEA	Japan Atomic Energy Agency
LANL	Los Alamos National Laboratory
LANSCE	Los Alamos Neutron Science Center
LEU	Low enriched uranium
LWR	Light Water Reactor
MA	Minor Actinides
MAMOX	Minor Actinide bearing Mixed Oxide Fuels
MSD	Mean Square Displacement

MOX	Mixed Oxide Fuels
NDE	Non-Destructive Evaluation
ND-PIE	Non-Destructive Post Irradiation Examination
NEAMS	Nuclear Energy Advanced Modeling and Simulation Program
NPDF	Neutron Powder Diffractometer/Neutron Pair Distribution Function
NRC	Nuclear Regulatory Commission
NRC	National Research Council Canada
NSLS	National Synchrotron Light Source
ORNL	Oak Ridge National Laboratory
O/M	Oxygen-to-Metal ratio
PCMI	Pellet Cladding Mechanical Interaction
PIE	Post Irradiation Examination
pRAD	Proton Radiography
PSI	Paul Scherrer Institute
PWR	Pressurized Water Reactor
RE	Rare Earth element
ROI	Region of Interest
SEM	Scanning Electron Microscopy
SME	Subject Matter Expert
SMARTS	Spectrometer for Materials Research at Temperature and Stress
TFRE	Transmutation Fuel with Rare earth inclusions
TMRS	Target-Moderator-Reflector System
TEM	Transmission Electron Microscopy
TOF	Time-of-flight
UN	Uranium nitride
U-Si	Uranium silicide (unspecified stoichiometry)
US	United States

1. Introduction

1.1 Science-based Approach and Modelling

Precedent suggests that it takes at least 20 years to qualify a new nuclear fuel. This long maturation time and the associated cost of testing and certification impede innovation. Over the last decade advances in atomistic and mesoscale modelling, coupled with increasing computational power offer a possible route accelerate innovation at reduced cost. However realizing the opportunity is predicated on finding a path whereby the regulators, informed by the scientific community, recognize that insights from new models are trustworthy for certifying the implementation of new materials, technology or operations.

The AFC execution plan notes that. *"The Office of Nuclear Energy FCRD has the responsibility for developing advanced fuel technologies for the Department of Energy (DOE) using a "science-based approach". This is focused on developing a fundamental understanding of nuclear fuels and materials. The goal is to combines theory, experiments, and multi-scale modeling and simulation to achieve predictive understanding of the fuel fabrication processes and fuel and clad performance under irradiation.*

and

"…advances made in fundamental understanding of materials, instrumentation and measurement techniques, and development and growth of high performance computing provide a means to overcome these barriers and implement a new approach to research and development. Termed the "science-based" approach, this process involves small-scale experiments, coupled with theory development and advanced modeling and simulation to reduce the number and cost of engineering-scale tests"

The value of models derive from their potential to rationalize observations and predict observable quantities. If their trustworthiness is established by a rigorous and efficient validation program then it is reasonable to ask what experiments on which materials under which conditions (e.g. temperature, atmosphere) over what length (and time) scales are critical. For fuels and cladding the challenges are inherently hierarchical with behaviors and phenomena that range from atomistic to macroscale, from nanometer porosity to millimeter cracks. It is this multiscale aspect that motivates a range of experimental approaches to assess performance over a range of length scales

"As opposed to large-scale, integrated experiments typical of demonstration-based programs, the focus on experiments for a science-based approach shifts to smaller-scale".

The Nuclear Energy Advanced Modeling & Simulation (NEAMS) initiative is a key stakeholder and potential customer for the results of mesoscale experiments that can underpin modelling initiatives is. Within NEAMS two codes that can benefit from experiments from mesoscale microstructural data are Marmot (microstructural evolution) and Bison (fuel performance) (Fig 1). In some cases the current suite of models have validation that need differ from the insights that can be achieved with conventional PIE approaches. It is in this context that novel experimental validation approaches are considered using atypical approaches. It is that context that this report explores some anecdotal opportunities for using neutrons in support of modelling initiatives.

One feature of a "science based approach" to nuclear fuel development is an understanding of material behavior at the microstructural and atomistic level. For new fuel formulations there is often a compelling need for experimental data to inform and validate predictive models. Since models predict material parameters on length scales from millimeters (e.g. cracks) to micrometers (e.g. phase

composition and texture evolution) to nanometers (e.g. crystal structure, interstitial sites for fission gases, phonons), experimental capabilities to complement all of these parameters are required.

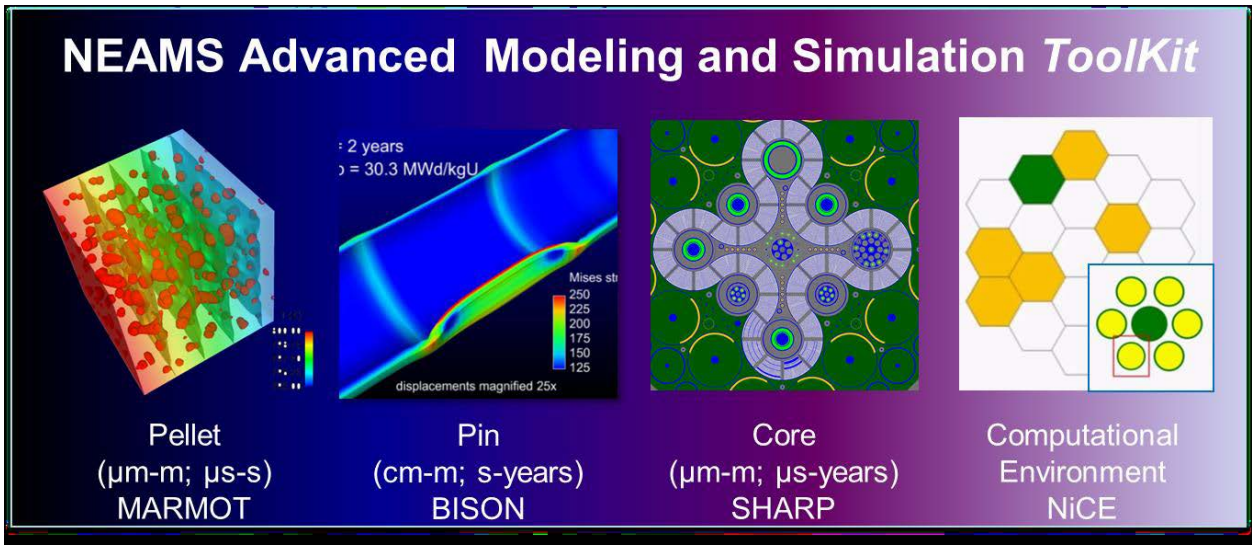

Figure 1 NEAMS *ToolKit* components and representative length and time scales.

1.2 Neutrons as a characterization tool

Many neutron experiments are accompanied by modeling studies. At LANL alone there have been hundreds of scientific publications over the past 15 years with modeling/experiment complementarity. A mature arena of study is understanding polycrystalline deformation in which in situ neutron deformation studies (combined with pre- and post-deformation neutron texture characterization) are compared with self-consistent deformation models to contribute to the understanding of deformation of materials with cubic crystal structure (e.g. steel [55]), hexagonal crystal structures (e.g. magnesium [54], zirconium [56], beryllium [57] and their alloys), orthorhombic crystal structures (e.g. uranium [58]), to monoclinic crystal structures (e.g. U-6Nb alloys [59]) and even triclinic crystal structures (e.g. TATB [60]). While the experimentalists and modelers can excel in their respective fields, the close interactions between the communities have established a long standing collaboration that accelerates advance and provides a precedent for how advance could be achieved in FCRD/NEAMS relevant problems.

A key objective for the development of neutron based PIE techniques is the routine characterization of the chemistry and microstructure of fuels via 3D nondestructive examination techniques and measurement of the properties at micron to submicron scale to support the science based fuel development program. The key attributes of neutrons for examination of nuclear fuels include non destructive determination of isotopic concentrations, tomography of cracks and engineering scale features and microstructure and crystal structure determination. On fresh fuels they can complement X-ray characterization by examining bulk pellets. However since thermal neutron are not prohibitively attenuated by lead, measurements in shielded containers are possible despite shielding and significant gamma ray activity.

This initiative focuses on examination of fuels; this requires other characterization techniques than the more frequently utilized destructive TEM or microscopic examination, which are more robust when it comes to highly radioactive samples and allow bulk characterization through fuel cladding or shielding. Those techniques are X-ray, neutron and proton radiography/tomography as well as neutron resonance spectroscopy and neutron diffraction. These techniques are usually only negatively impacted by

radioactive materials if the detector is exposed to radioactive particle emission from the material being analyzed and are used routinely in materials science to evaluate microstructure and chemistry of materials down to the submicron level. Furthermore, the Los Alamos Neutron Science Center (LANSCE) at Los Alamos National Laboratory (LANL) has the unique advantage of an existing infrastructure which allows receiving and handling of radioactive materials.

Optimal synergy between modelers and experimentalists has the potential to significantly accelerate fuel development and ultimately licensing of new fuel forms. This report is intended to document some of our efforts to facilitate such interaction.

Techniques developed at LANSCE using the pulsed neutron source at the Lujan center may, within a decade, have potential for utilization at the irradiation facilities (e.g. ATR at INL) as well as commercial sites for fuel production or power plants. Recent advances in small-scale accelerators for neutron generation (by D-D fusion) or laser-drive pulsed neutron sources [1] while not as yet capable of sufficient neutron production for measurements of the type described here, are nevertheless promising. If such a "portable" source with sufficient neutron performance were to become available then the synergy between modeling and experimental techniques described in this report would be immediately available to be deployed to the field and become routinely available, leveraging this initiative to develop and apply techniques not routinely used for fuel characterization and providing new characterization capability relevant to benchmarking models used in fuel development and related areas.

Characterization of nuclear materials at non-ambient conditions is of importance for predicting performance under operation and accident conditions. Similarly characterization under synthesis conditions is of economic interest. However, in either case the characterization tools that can be applied to nuclear materials are limited. Neutrons are one probe that can be used to penetrate sample containment in high temperatures, pressures, or chemical environments. Moreover neutron facilities, due to the activation of some materials that can occur when exposed to neutron beams, are administratively prepared to handle radioactive materials. Since neutrons can sample volumes ranging from a cubic millimeter to several cubic centimeters, sample preparation is usually minimal and sample can be characterized without ever taking them out of their containment.

1.3 Scope and Goals of this activity

This activity, part of the ANDE work package, is focused on the application of neutrons to optimally support modeling and simulation efforts in the field of fuel cycle R&D. Neutron imaging and scattering techniques can examine volumes of up to several cubic centimeters, consistent with typical rod assemblies of ceramic fuel pellets or metallic fuel forms. In contrast with X-ray scattering, neutron scattering lengths for high and low atomic number elements are often comparable, allowing the crystallographic study of systems consisting of heavy and light elements such as the uranium silicide and uranium nitride systems actively researched as accident tolerant fuel forms. We illustrate below with an example of a uranium carbide how this ability is clearly superior to X-ray characterization and directly relevant to various modeling efforts.

In FY 2012, at a LANL/INL workshop a plan was conceived to develop, assess and use the advanced non-destructive evaluation techniques including neutron and proton radiography available at LANL and INL for analysis of irradiated fuel rodlets from the AFC-2C irradiation at the ATR in Idaho [5]. A plan was developed to create a set of mock-up rodlets containing pellets of depleted uranium dioxide (dUO_2) with defects similar to that seen in irradiated fuel rodlets to assess the capabilities of the NDE techniques at LANL. The goal was to determine the spatial resolution and the ability to resolve microstructure and chemistry variations in light water reactor (LWR) and fast reactor (FR) size rodlets.

Neutron imaging and diffraction offer opportunities for the characterization of nuclear materials [6]. At LANL, these opportunities are explored with the goal of advancing pre- and post-irradiation examination to provide data sets of irradiation-induced changes with unprecedented level of detail. For this purpose, various demonstration and mock-up samples of the major nuclear fuel types (Urania-based, SIMFUEL, metallic fuel) were characterized. Furthermore, samples nominally identical to specimens planned to be irradiated were characterized to provide first-of-its-kind pre- and post-irradiation bulk characterization. In this document we discuss how these or similar measurements can inform and complement modeling efforts.the progress in the past months is reported. Activity to date is summarized in Table 1 and Table 2.

Over the last few years, a series of measurements have demonstrated the applicability of neutrons, protons and synchrotron X-rays to a range of ceramic and metallic nuclear fuel systems. Anecdotal results were provided in previous reports. Meanwhile recent initiatives have focused on developing fuels with enhanced accident tolerance, increased burn-up and reduced waste. One method to enhance accident tolerance of the fuels is to improve the thermal conductivity to reduce the fuel centerline temperature. This increases the time to reach critical temperature in an accident. At LANL, U_3Si_5 [3,4] and a composite of UN with 15-vol% U_3Si_5 are under consideration as potential ATF forms. They have been fabricated to determine material compatibility and measure the associated thermal properties. Thermal conductivity and expansion values were then supplied to modeling and simulation to further refine the design of the fuel pellets and the associated fuel rodlet and cladding design.

Completion	***Activity***
2013 February	Evaluation of neutron NDE techniques and capabilities
2013 May	UO_2 mock-ups fabricated
2013 December	Neutron and Proton examination of UO_2 mock-ups
2014 September	Purchase of neutron resonance imaging detector
2014 October	Graduate student hire (UC Berkeley)
2014 December	dU-10Zr (metallic fuel) mock-ups fabricated (INL)
2015 January	Neutron examination of dU-10Zr metallic fuel mock-ups
2015 January	SIMFUEL mixed oxide fuel mock-ups fabricated (LANL)
2015 February	Neutron examination of SIMFUEL mixed oxide fuel mock-ups
2015 June	UO_2 mock-ups shipped to UCBerkeley) for destructive examination
2015 August	Data analysis for metallic and mixed oxide fuel mock-ups – Ongoing
2015 September	Undergraduate student hire (LANL)
2015 October	Postdoc conversion (UC Berkeley)
2015 November	UN U3Si5 UN U3Si2 fabricated (LANL)
2015 December	Neutron examination of LANL ATF-1 fuels (HEU)
2016 February	Assessment of ability to measure fission gas partial pressures
2016 May	Data analysis for LANL ATF-1 fuels
2016 May	U Pu Zr (Transmutation fuel) fabricated (INL)

2016 August	Data analysis for fission gas measurements

Table 1: Activity starting 2013.

Sample	Composition	Purpose	Measured
dUO_2 pellets	W inclusions & diffusion Cracks	Establish sensitivity for density, visualization of voids/cracks and inclusions of different materials	FY12-13
Metallic foils	Au, Ag, ^{238}U, Cd,	Calibrate thickness & concentrations measurement Assess instrument performance	FY12-15
Gases	Kr, Xe	Demonstrate fission gas detection	FY14
UN U_3Si_5	UN 15 vol.% U_3Si_5, Depleted Uranium	Assessment fabrication homogeneity	FY15
Metallic Fuel	U-10Zr w/ La,Pr,Ce,Nd.	Demonstrate characterization of rare earths inclusions	FY15
SIMFUEL mock rodlets	UO_2 , La_2O_3, Rh_2O_3, ZrO_2	Demonstrate identification of phase and fission products	FY15
UN U_3Si_5, UN U_3Si_2, U_3Si_5	235-U enriched	Enrichment level measurements Pre-irradiation – batch characterization	FY16

Table 2: Samples characterized to date.

2. Neutron characterization and modeling; a case study for the U-C system

2.1 Neutron and X-ray Diffraction for U-C system

For neutrons the scattering power of different elements (and therefore the contribution of each elemental species in a given crystal structure to the diffraction signal) is *not* dependent on the weight or Z-number of the element. Instead, it is isotope specific and varies irregularly over the periodic table of the elements. This means that neutrons are well suited for characterizing sub-lattices of light elements, such as C, N, O etc. in the presence of heavy elements, such as U. This contrasts with X-rays or electrons which interact with the electronic shell rather than the nucleus and thus the diffraction signal is biased towards the heavier element, making it harder to determine changes in the sub-lattices of the lighter element.

Since phase transitions, thermodynamic properties etc. of uranium-based compounds (uranium oxides, nitrides, carbides, silicides etc.) manifest frequently in the anion sublattice, neutrons are an excellent probe to study these. One key example is the Bredig transition in UO_2 or the example of UC_2. For uranium-based compounds, the fraction of studies using neutrons rather than X-ray as reported for instance in the Inorganic Crystal Structure Database, is with 55% much higher than for all systems reported in the database (16% of all studies reported use neutrons) [6]. Figure 1 illustrates the advantages of neutrons for crystallographic studies of uranium-based systems with the crystal structure of δ-UC_2 as an example.

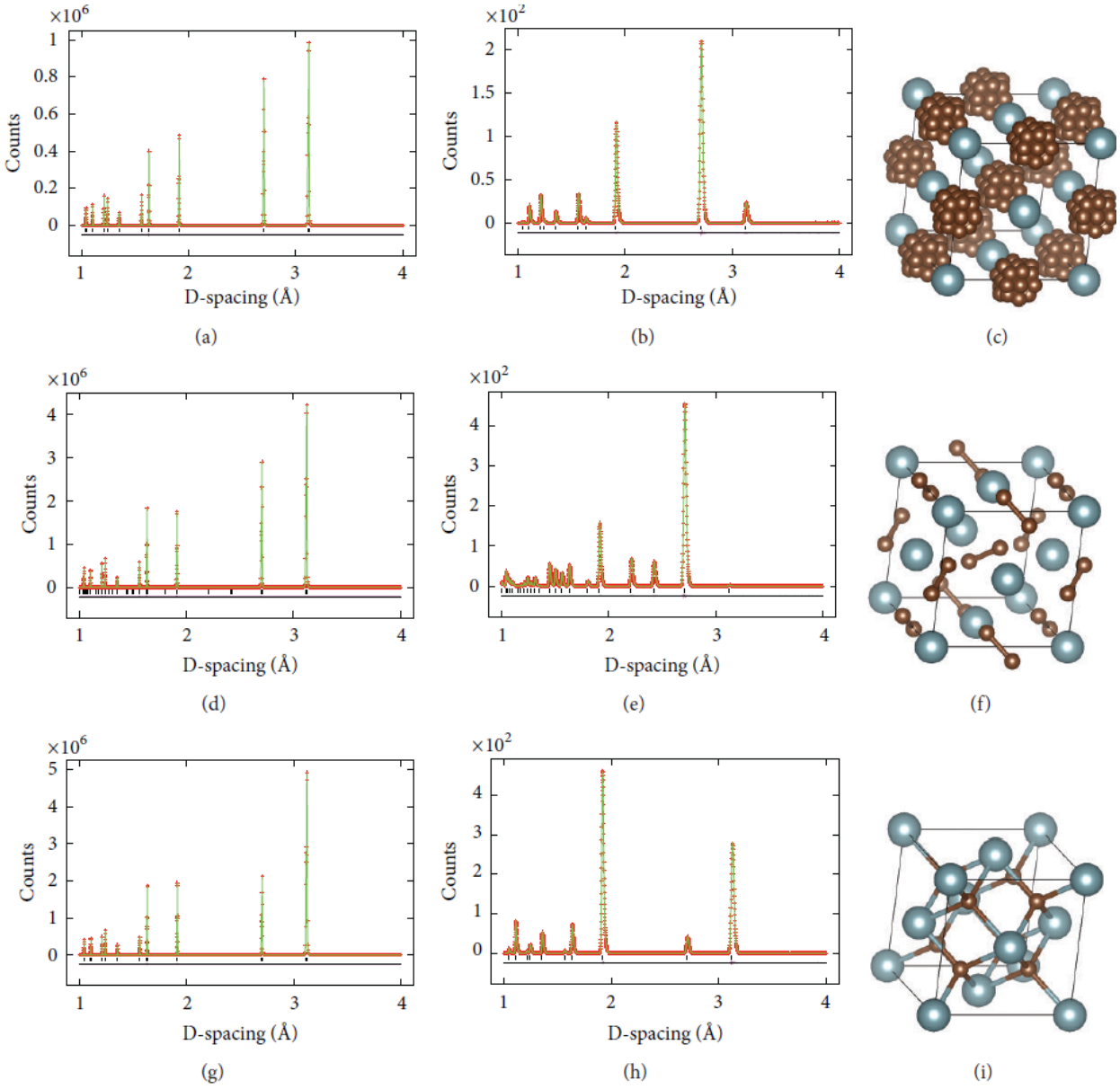

Figure 2: Simulated X-ray (left) and Neutron (right) diffraction patterns for three published crystal structures of cubic UC_2. First row – disordered carbon structure proposed by Bowman et al [23], second row pyrite-structure proposed by Bredig [24], and third row fluorite structure proposed by Wilson [25].

As shown in Figure 2, the three crystal structures reported in the literature for the cubic high temperature phase of UC_2 are substantially different. However, the difference occurs in the carbon sub-lattice and the uranium sub-lattice is a face-centered cubic lattice in all cases. Consequently, the diffraction patterns for X-ray diffraction are dominated by the uranium sub-lattice and are similar for all three structures. For neutrons, however, the simulated patterns are clearly different. See for example the two peaks between 2.0 and 2.5 Å in d-spacing only occurring for the pyrite-type structure, or inverted peak intensity ratios for the two highest d-spacing diffraction peaks, all of which make crystal structure solution for systems such as UC_2 much more reliable with neutrons. Simulations of the diffraction patterns were conducted with the gsaslanguage [26] and GSAS [27] packages. The crystal structures are visualized using VESTA [28] [6].

2.2 In situ high temperature Neutron Diffraction Urania

Taking advantage of the neutron diffraction capabilities at the Lujan center, the reaction of urania with graphite to form the cubic UC_2 phase was studied using the High Pressure-Preferred Orientation (HIPPO) [9,30] diffractometer at the Los Alamos Neutron Science Center (LANSCE) [31,32]. Figure 3 shows a schematic of the instrument. HIPPO views a high-flux/medium resolution moderator emitting neutron pulses at a frequency of 20 Hz. The comparably short flight path between moderator and sample (~8.89 m) combined with five rings of a total of 1200 ^{3}He detector tubes arranged around the incident beam allows for short integration times, enabling the kinetic studies reported here. The detectors are mounted on 45 panels which are arranged on rings with nominal diffraction angles ranging from 40° to 145° diffraction angle. For data analysis, time-of-flight histograms of each detector tube are integrated either per panel, resulting in 45 histograms suitable for texture analysis, or per ring, suitable for diffraction data analysis assuming no preferred orientation. The sample chamber has a volume of ~2 m^3 and accommodates various sample environments.

Samples for this experiment were mounted in a high temperature furnace using a graphite resistive tube heater to achieve temperatures in excess of 2200°C [33]. The sample was a pressed pellet of fine grained UO_2 (grain size ~100 nm) and graphite powders of appropriate weight fractions. Isothermal neutron diffraction data were collected for 30 min every 250˚C between 250˚C and 2000˚C. The 30 min were collected as six data sets of five minutes each to enable the study of kinetics. During heating (at 10˚C/min) data were collected in 2 min intervals. The experiment (excluding setup and alignment) was completed in ~11 h. Details about the experimental setup, data analysis, and sample characterization can be found elsewhere [34].

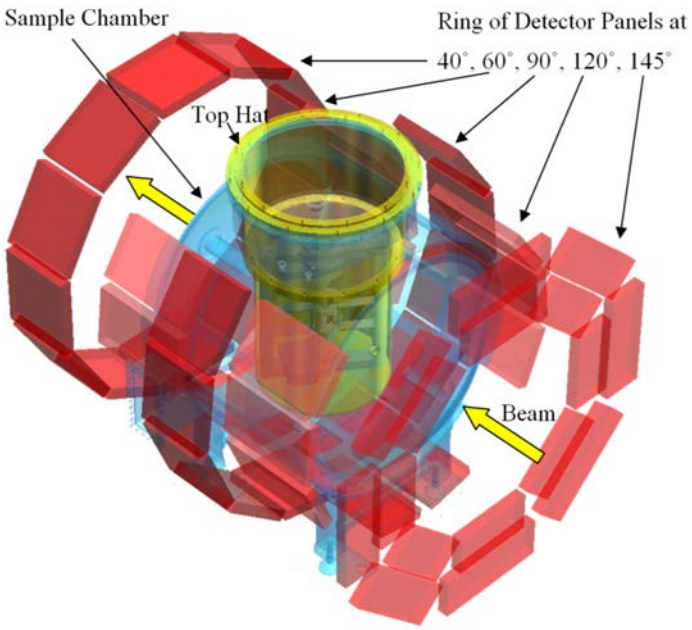

Figure 3: Schematic of the HIPPO neutron time-of-flight diffractometer at LANSCE. Red areas are detector panels, the blue volume is the sample chamber, and the yellow structure is the furnace enclosure.

An overview of the diffraction data is given in Figure 4. The contour plot shows the diffracted intensity as a function of lattice spacing (d-spacing) and time/temperature. The temperature is indicated on the left part of the plot. At the bottom of the plot, diffraction lines for UO_2 and graphite (in the 2H modification) are visible. During heating from 250°C the thermal expansion is visible as traces for each diffraction peaks move to the right (higher d-spacing). At 750°C a change in the diffraction patterns is apparent when the UO_2 diffraction peaks change in width and intensity. This corresponds to the temperature in the U-O phase diagram when the two crystallographically similar phases UO_2 and U_4O_{9-x} become a single oxygen super-saturated phase UO_{2+y}. Then just below 1500°C, the reaction $UO_2+C \rightarrow UC_x+CO_y$ occurs, with the carbon-oxide leaving the system as a gas. This leads to an almost complete disappearance of the UO_2 and weakening of graphite diffraction signals as well as the formation of a new set of diffraction peaks associated with the formation of uranium carbide, which we could index with the cubic UC mono-carbide phase.

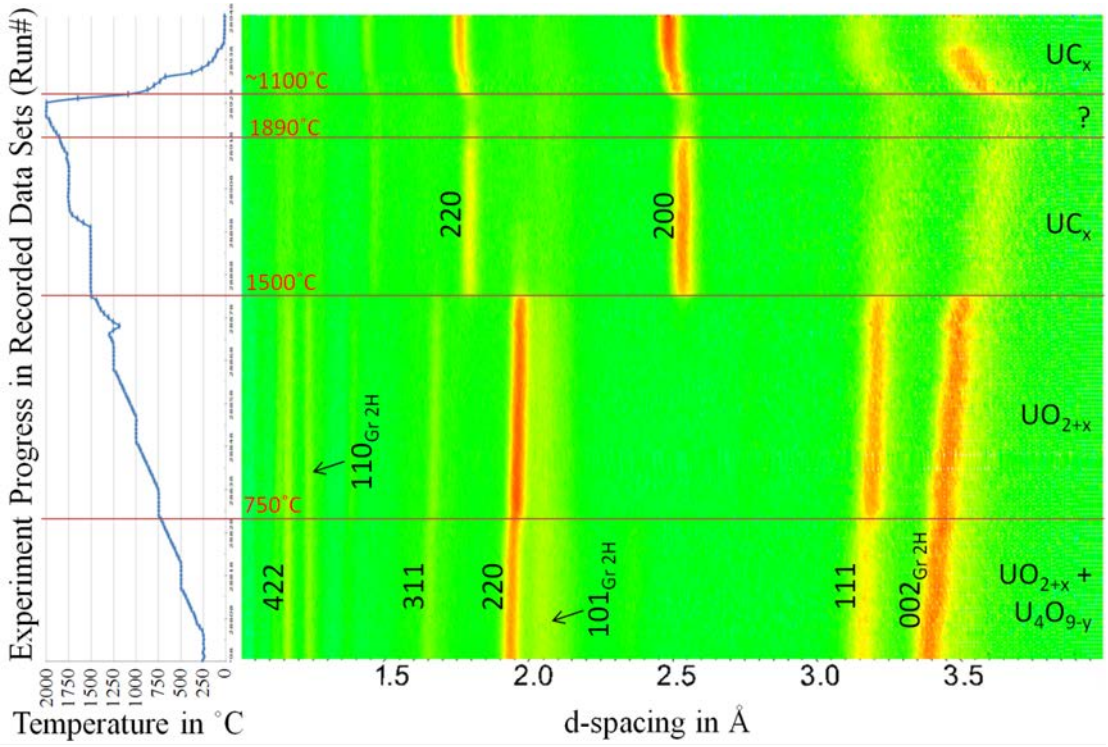

Figure 4: Overview of the experiment as contour plot of diffraction intensity as a function of d-spacing (horizontal axis) and time/temperature (vertical axis). Major diffraction lines are labeled with their (hkl) indices with those of graphite having subscripts. The phases present in each horizontal section are listed on the right. The sample temperature is shown on the left.

The onset of the reaction occurred at 1440°C with a completion at 1500°C. With the heating rate set to 10°C/minute, the reaction was therefore completed within 6 minutes. Further increase of the sample temperature allowed us to follow the thermal expansion of the UC_x phase. However, even during the dwell times at constant temperatures we observed changes in lattice parameters [34]. We attribute this to the continuation of the migration of carbon atoms into the uranium carbide. At 1890°C an unexpected drop in the intensity of the uranium carbide peaks was noted and the furnace was cooled as shown in Figure 4 upon which the diffraction signal intensity was recovered.

The ability to follow the reaction $UO_2+C \rightarrow UC_x+CO_y$ in situ with neutron diffraction allowed us to conclude that the reaction occurs within minutes over a temperature range of ~60°C. The initiation of the reaction is far below the reaction temperature of 2000°C reported for this reaction in standard text books on actinide chemistry [38], and even below the 1500 to 2000°C range reported previously [39]. This may be due to the unusually small ~100 nm grain size of our UO_2 initial powder. Furthermore, the holding time reported e.g. by Carraz et al. [40] of several hours would be unnecessary. The neutron diffraction data collected for the cubic high temperature UC_2 phase unambiguously confirmed the disordered C-C dumbbells reported by Bowman et al. [23] and excluded the structures proposed by Bredig [24] and Wilson [25], both determined by X-ray diffraction.

Since calculations of properties of this phase are reported in the literature, e.g. [41,42], accurate knowledge of the crystal structure is of great practical relevance. However, since the phase in question is not quenchable (since the dynamic disorder of the C-C dumbbells is only stable at high temperatures), in

situ studies in the stability field of the phase are unavoidable. These experimental results exemplify the advantages of neutron diffraction for such investigations.

2.3 Modeling relevance of neutron diffraction data

2.3.1 High temperature structure

To explain the diminished diffraction intensity at 1890°C [34] and its subsequent recovery, molecular dynamics simulations of the cubic high temperature phase of the U-C system [35] suggest an explanation. With increasing temperature, the uranium carbide system absorbs increasing amounts of carbon without changing its crystal structure (except for an increase in lattice parameter). (We did not observe a change to the tetragonal phase of uranium-carbide). However once the carbon up-take reaches two carbon atoms per uranium atom, the carbon atoms form rotating C-C dumbbells. This rotation leads to a dynamic disorder, which disturbs the periodicity of the crystal lattice with the potential to exhibit the observed decrease in diffracted intensity.

The effect is different from the Bredig transition in UO_2, where the oxygen atoms start to move freely in the uranium sublattice, forming a superionic conductor [36]. The phenomenon is better described by the terms ‘sub-lattice melting’ or ‘premelting’ [37], where one species in a binary, ternary or higher system is highly disordered, resulting in an atomic motion similar to a melt. Rotation of the C-C dumbbells was already described by Bowman et al. [23] in their determination of the crystal structure using neutron diffraction. Wen et al. [35] found, however, that the oscillation is not spherical, as described by Bowman, but rather an oscillation approximately around the static location of the C-C dumbbells in the pyrite structure. *Analysis of our diffraction data [34] confirm the structure reported by Bowman et al., (also determined by neutron diffraction).*

2.3.2 Interstitial Sites

Modelling of the U-C system, as reported by Freyss [42], requires the assumption of a crystal structure. Freyss assumed a CaF_2 structure as proposed by Wilson (section E.1 in ref [42]) to predict the lattice parameter (Table III in ref [42]). In this work, modelling for crystal structures of several uranium carbide phases (α-UC_2, β-UC_2 and U_2C_3) was pursued using a standard first-principles projector augmented wave method as implemented in the VASP code, using exchange correlation interaction in the generalized gradient approximation as parametrized by Perdew-Burke-Ernzerhof. While agreement of simulated and experimental lattice parameters is excellent, the lattice parameter value is not sensitive to the atomic position of the carbon atoms and thus, by itself, is not a rigorous test of the model. *To discuss interstitials, their formation energies in various lattice positions etc., the correct atomic positions of the carbon atoms are required.* The conclusions for locations of fission gas atoms in the crystal lattice in the UC_2 phase are therefore questionable.

2.3.3 Phase Stability

The paper by Chevalier and Fischer [41] reports on thermodynamic modelling of the U-C system “*performed in the framework of the development of a thermodynamic database for nuclear materials, for increasing the basic knowledge of key phenomena which may occur in the incident of a severe accident in a nuclear power plant*”. The primary goal was modelling the interactions of UO_2 with B_4C control rods during accidents. It surveys the experimental and calculated properties of the U-C system. In Table 1 of that work, UC_2 is listed as “fcc_B1 type, isotypic with CaF_2”, citing again Wilson’s structure

determination (reference [40] in their paper). The monocarbide UC is described as NaCl structure type, consistent with our data. The authors note that the CaF_2 structure of UC_2 as reported by Wilson is incompatible with a complete miscibility of the solid solution (UC, UC_2) at high temperature. They do not, however, offer an alternate crystal structure or refer to Bowman's structure report (despite their otherwise comprehensive literature review). The authors report experimental methods used by others to investigate these phases as diffusion of couples, quenching experiments, metallography, optical pyrometry, and high temperature X-ray diffraction. This serves to emphasize that those methods are inadequate to resolve the crystal structure of the non-quenchable cubic UC_2 phase. Indeed when the models only use interatomic interaction potentials *without specific atomic positions in a unit cell*, they are less sensitive to accurate description of the crystal structure.

2.3.4 Predicting High Temperature Behavior using Pair Potentials

Basak used a semi-ionic pair potentials to model the lattice parameters, thermal expansion and isothermal bulk modulus of UC [43]. In this case, experimental lattice parameter data was utilized to determine appropriate pair potential parameters for the monocarbe UC by fitting predicted lattice parameters to experimental ones. This procedure assumes however that no phase transformation or variation in carbon content, leading to chemical strains, occurs, which in case of residual graphite from sample synthesis cannot be excluded. As seen in the neutron diffraction data, the phase transformation from UC to UC_2 does *not* change the lattice parameters significantly nor does it result in additional diffraction peaks. Only a weakening of the diffraction signal is observed. In the case of X-ray diffraction the diffraction signal for X-rays is dominated by the unaffected uranium sub-lattice and the weakening of the diffracted intensity is harder to observe.

Thus, while lattice parameters may be determined equally well with X-rays or neutrons, the neutron data are more sensitive to phase transformation observed in U-C, U-O, U-N, U-Si etc. The data are thus a valuable complement to benchmark, constrain, or calibrate simulation data. From the derived potential, Basak predicts the mean square displacement (MSD) of the carbon ions in UC. The time-average mean square displacement can be compared to the experimental thermal motion or atomic displacement parameters derived from the diffraction analysis. *Neutrons have a twofold benefit here: a) the neutron diffraction signal for the carbon sub-lattice that is comparable to that of the uranium sub-lattice and b) the absence of a form factor, as in X-ray diffraction, allows a larger number of diffraction peaks to be used for determining thermal motion parameters.* This was also utilized in a series of papers on MAX phases (ternary carbides), where simulations of the thermal motion, derived from simulated phonon spectra, were compared to anisotropic thermal motion parameters as a function of temperature determined by neutron diffraction with HIPPO.

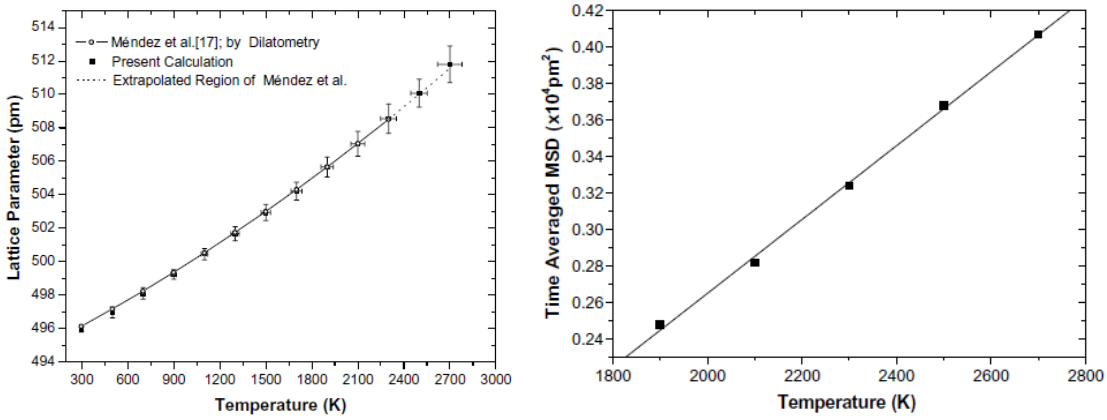

Figure 5: Lattice parameters for UC as derived from dilatometry data and used to fit atomic potential parameters for the U-C system (left). Predicted time-averaged mean square displacement parameters of the carbon ion in UC (right). Neutron diffraction could provide experimental benchmarks for the thermal expansion that would be also sensitive to possible re-distribution of carbon atoms (dilatometry is insensitive to length change due to thermal expansion or redistribution of ions). It could also experimentally verify the predicted thermal motion parameters.

2.3.5 DFT Calculations on the Role of Interstitials for Phase Stability

Changes of stability fields due to intercalation of interstitial species, such as carbon into uranium in this example, are hard to predict with structural simulations. *Experimental data such as neutron diffraction measurements that address the kinetics, intermediate phases, or changes to transition temperatures of phase transformations are of paramount importance*. For example in work done by Winkler et al. [44] on formation of titanium carbide from the elements, a stabilization of the hexagonal α phase was observed concurrently with an anomalous increase of the c lattice parameter of this phase. Pure Ti transforms from the hexagonal α to cubic β phase. In the presence of carbon, as is the case here, α-Ti is still present at temperatures >1200K. DFT calculations indicated that this is due to stabilization by carbon atoms which manifests itself in the anomalous increase of the c-lattice parameter. Density functional theory based calculations supported the interpretation that the interstitial carbon atoms in the α-Ti lattice stabilize this phase.

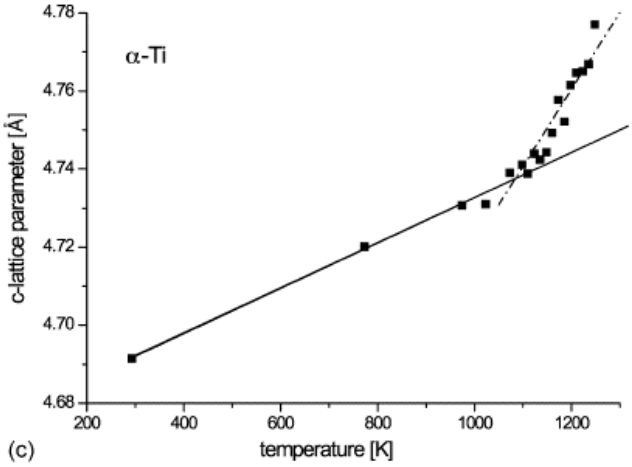

Figure 6: Evolution of the c-lattice parameter of pure Ti in the presence of graphite as a function of temperature measured by neutron diffraction during the in situ observation of the reaction Ti+C→TiC [44].

3. Potential for Neutron Characterization to inform NEAMS Models

Section 2 described in detail one example of how neutron diffraction data can be relevant to NEAMS relevant models. There are many areas where connections can be made. Besides the pre-irradiation characterization of accident tolerant fuels described in detail in previous reports, fundamental work on e.g. anisotropic thermal expansion (e.g. a- and c-lattice expansion in hexagonal U_3Si_5), phase stability (heating and cooling of U-Si systems in situ to determine phase transition temperatures), simulation of accident conditions, or synthesis of e.g. silicides to assess hold times and temperatures used in processes and their influence on the resulting microstructure (similar to the uranium carbide formation experiment described above). All of these experiments would immediately affect development of accident tolerant fuels. Besides the provision of experimental data to inform and validate simulations, such experimental

work would also play a substantial role in training the next generation of FCRD/NEAMS researchers in the use of state-of-the-art characterization techniques and their influence on predictive tools.

3.1 Established Techniques

At the Lujan center a series of beam lines offer different characterization opportunities. They include flight path 5 for imaging, flight path 4 (HIPPO) for rapid throughput and extreme environment studies measurements, FP2 (SMARTS) for spatially resolved measurements and FP11 (Asterix) for surface corrosion studies. Each of these beamlines offer unique capabilities to perform experiments that can inform and constrain NEAMS models. Some of the possibilities that have arisen from discussions with NEAMS experts are listed in Table 3. For these experiments precedence exists as previous work applied this techniques to NE relevant or similar systems. Examples are the tomographic characterization of ATF on FP5, deformation and heat treatment for Zr-Nb alloys on SMARTS and HIPPO, and oxidation studies on thorium and uranium on Asterix.

Instrument	Technique	Measurement
FP5	Imaging	Characterization of Fresh ATF to (a) understand the initial conditions prior to irradiation, (b) correlate performance parameters with initial conditions, and (c) ensure uniformity of the fabrication processes.
FP5	Imaging	Characterization of Fresh Metallic fuel to (a) understand the initial conditions prior to irradiation, (b) correlate performance parameters with initial conditions, and (c) ensure uniformity of the fabrication processes.
FP5	Imaging	Species redistribution in metallic fuels in thermal gradient
FP5	Imaging	Fission gas modeling (trapped Xe) , macroscopic diffusional flow
FP5	Imaging	Fuel Properties – melt, relocation. fuel clad chemical interactions and fuel clad mechanical interactions.
FP5	Imaging	Observation of cracking and swelling during simulated LOCA events
FP5	Imaging	Tomography of cracks at engineering scales
FP5	Imaging	Temp and response missing pellet surface
FP5	Imaging	Steam kinetics studies - oxidation of the cladding
HIPPO/SMARTS	Diffraction	UPuZr f(T,t) stoichiometry
HIPPO/SMARTS	Diffraction	Cladding behavior in environment of missing pellet surface
HIPPO/SMARTS	Diffraction	Microstructure uniformity for different synthesis conditions
HIPPO/SMARTS	Diffraction	Microstructure evolution during heating
HIPPO/SMARTS	Diffraction	UN/U-Si f(T,t) stoichiometry
HIPPO/SMARTS	Diffraction	Reaction kinetics for synthesis reactions
HIPPO/SMARTS	Diffraction	Reaction kinetics for accident conditions (e.g. oxidation, control rod material etc.)
HIPPO/SMARTS	Diffraction	Deformation studies e.g. to investigate changes in deformation behavior due to irradiation

HIPPO/SMARTS	Diffraction	Dislocation studies, e.g. to investigate changes in dislocation networks and their annealing behavior due to annealing
HIPPO/SMARTS	Diffraction	FCCI interaction
HIPPO/SMARTS	Diffraction	Creep Models
HIPPO/SMARTS	Diffraction	In situ melt behavior
Asterix	Reflectometry	Chemical speciation in Uranium Oxide Films
Asterix	Reflectometry	Evolution of uranium oxide in an oxidizing environment
Asterix	Reflectometry	Hydride formation

Table 3: **Neutron experiments with NEAMS modelling relevance**

3.2 Potential Applications relevant to Data Needs for Validation

The Fiscal Year 2016 Consolidated Innovative Nuclear Research Funding Opportunity Announcement (DE-FOA-0001281) spells out several data needs for validation (Appendix D therein). In this section we address several opportunities for potential provision of such data by neutron methods which require some development and collaboration with modelers to establish requirements for e.g. sensitivity, preparation of samples, design of sample environments etc. Such efforts are somewhat more exploratory than the aforementioned proven approaches and may also require refining the existent capabilities.

3.2.1 Fission Gas Diffusion

The Fuels Product Line of the NEAMS program is in need of “improved temperature-dependent diffusion coefficient measurements of Xe in UO2”. If samples with appropriate amounts of Xe in a urania matrix can be prepare, e.g. by ion implantation, neutron diffraction and energy-resolved neutron imaging could be used to characterize the microstructure of the initial state of the specimen. Using the ability of energy-resolved neutron imaging to map and measure the density of isotopes, the sample could be heated in the neutron beam and the changes in xenon concentration could be measured, thus providing the desired diffusion coefficient. Figure 7 illustrates the ability of energy-resolved neutron imaging to visualize and measure densities specific for different isotopes.

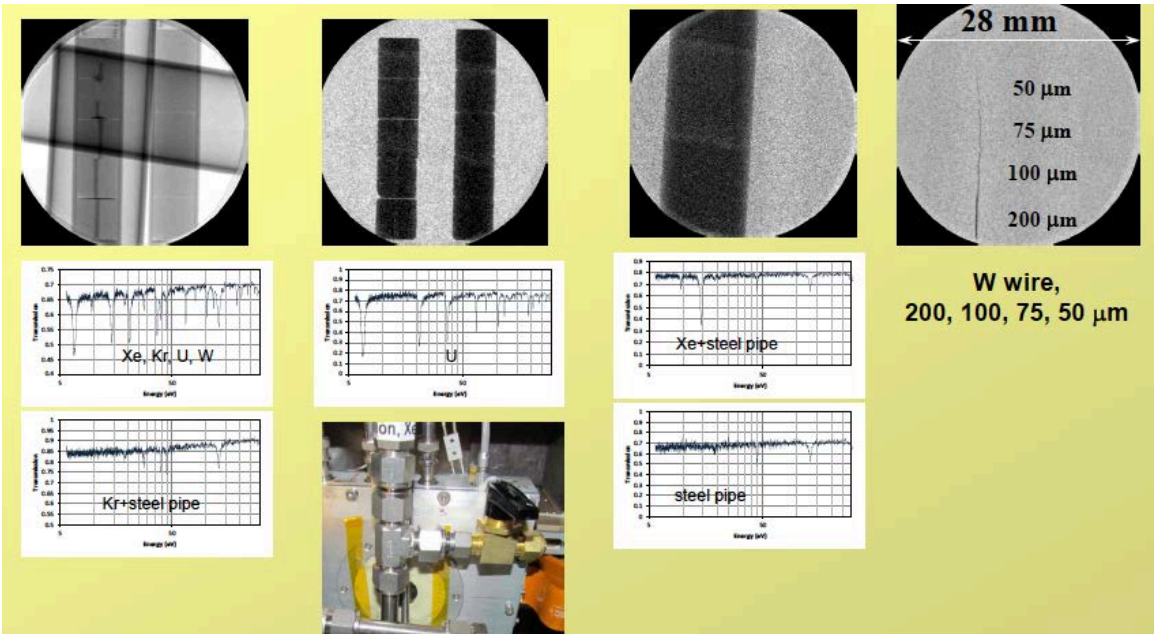

Figure 7: Example of two fuel rodlets with different thickness of tungsten wires embedded together with steel pipes filled with Xe and Kr (photograph) and viewed by neutron radiography over the whole energy range (left), resonance energies of uranium, xenon, and tungsten. The ability to map and measure xenon density could be applied to measuring diffusion of xenon in Urania.

3.2.2 Fission Gas Release

Similarly, data for the validation of fission gas release models could be provided by characterizing fuel pellets containing xenon for their xenon distribution as well as microstructure, cracks, voids etc. with neutron diffraction and imaging. These specimen could then be exposed to thermal transients followed by the same characterization as before. Differences in xenon concentration can then be correlated with the local microstructure and the availability of the xenon concentrations as well as micro-structure pre- and post-transient provides unique data for modeling. Figure 8 explains this concept with CT reconstruction of several Urania fuel pellets with intentional flaws. Two of them were exposed to tungsten receptors during RF heating with temperatures high enough to let some of the tungsten diffuse into the Urania matrix.

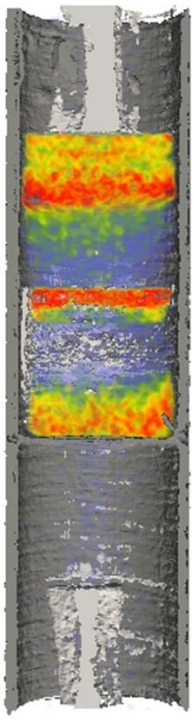

Figure 8: Overlaid CT reconstruction of five urania pellets (8mm diameter) in steel cladding. Displayed are CT reconstructions of strong thermal neutron attenuation (dark grey, e.g. steel cladding), weak neutron attenuation (bright grey, voids and cracks) together with tungsten density measurements (color scheme, maximum intensity 1%).

3.2.3 Mechanical Behavior of Irradiated Zircaloy Cladding

The Fuel Product Line of NEAMS also requires data on "mechanical behavior (yield stress, creep behavior, failure data) for zircaloy cladding that has been irradiated and exposed to chemical environments conducive to stress corrosion cracking." The unique capability of LANSCE to handle radioactive materials would allow to characterize a series of zircaloy specimen with different irradiation and corrosion histories and systematically characterize their texture and volume fractions of α/β phase on HIPPO, then measure the flow curves in situ on SMARTS. The in situ neutron diffraction data would allow to address differences in deformation behavior on a microscopic level while at the same time providing macroscopic flow curves to establish e.g. yield stress or ultimate tensile strength. Such studies on irradiated Zr-2.5Nb were already demonstrated [61].

3.2.4 Pellet Cladding Interaction

The need to "evaluate pellet cracking and fragment movement during normal operation" where the "UO_2 fracture behavior and frictional interaction between pieces would be studied under representative thermal and stress conditions" offers another potential where neutron radiography could be utilized to study electrically heated pellets to obtain fracture characteristics. Neutrons would provide the additional benefit of being able to characterize also the microstructure of pellets prior to the experiment, including spatially resolved scanning of stresses.

4. Discussion

One element of the FCR&D program is *"Capability Development integrates the infrastructure, tool development, and resource application to enable the science required for all nuclear fuels development within DOE"*. Progress in last decade has placed a scientific focus on mesoscale science potentially enabled by future exascale computational performance. Realizing the promise of such computational power will place demands on validation and a range of tools will be needed.

To date materials development has heavily relied on an Edisonian approach to innovation with a tendency for materials development to focus on utility rather than novelty. Conversely, there are many examples of materials innovation such as metal matrix composites, bulk metallic glasses and ultra fine grain material where their commercial impact has been impeded, in part, because of challenges in certification. It is hoped that the GAIN initiative will accelerate innovation but it is likely that trust in new modelling approaches will be a necessary element of its success.

It is a truism to suggest that model validation can always use more experimental data but the crucial question is how much and what data is needed to satisfy regulators that conclusions from a model or code contribute to a certification decision. The hierarchical nature of fuels modelling in which multiscale models embed lower length scale models suggest that a range of tools capable of examining fuel clad systems over a range of length scales will be needed.

In the specific case of actinides in general, and plutonium in particular, LANL has a unique institutional interest. The LANL materials strategy explicitly calls out plutonium production science as an area of focus. This includes developing characterization tools as well as developing models capable of predicting performance under a variety of conditions. Thus there is strong synergy with FCR&D initiatives in studies of transmutation fuels. Many of the production challenges in the NNSA stockpile have analogues in the development and certification of new transuranic transmutation fuels and relies on modelling.

Key elements of the LANL materials strategy are

- Processing-aware actinide materials science: Controlled synthesis of materials meeting performance requirements with predictive process-structure-properties-performance relationships from ambient to extreme conditions. Computation coupling across length and time scales with 3D microstructure modeling capability.
- Age-aware actinide materials science: Forward predict aging effects on performance with a quantified uncertainty with the identification of fundamental control variables (dose, dose rate, temperature) and computation coupling across length and time scales supporting the development of a robust QMU framework.

The Lujan neutron scattering center offers access to multiple beam lines and staff that are stakeholders in developing neutron (and synchrotron X-ray characterization tools). LANL has an ongoing need to develop models and experimental tools that can support FC R&D goals. The range of experimental characterization opportunities include but are not limited to the neutron scattering facility. LANL is exploring membership in the NSUF program. Consequently, LANL is an excellent location to initiate relevant experimental characterization to optimize the insight gained by modelling efforts.

This report has focused on the NDE opportunities pre irradiation but the potential examination post irradiation is of equal import. Indeed the ability of neutrons to characterize complete pellets of research

rodlets has the potential to generate large data sets relevant to both Bison and Marmot. Ultimately the same techniques could applied to studies of used fuel disposition.

5. Conclusions

Neutron scattering and imaging techniques can provide data that is uniquely relevant to NEAMS multiscale models. Unique aspects of the imaging include bulk characterization of complete pellets for microstructural features flaws and isotopic distributions. Unique aspects of the diffraction techniques derive from the ability to make crystallographically significant measurements over a wide range of temperature conditions. There are many possibilities for unique separate effects tests. The LANL institutional focus on plutonium makes the study of metallic fuels of particular interest.

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