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# OAK RIDGE NATIONAL LABORATORY

MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

P.O. Box 2008 Oak Ridge, TN 37831-6170 Tel: (865) 241-3570 Fax: (865) 574-3527 Email: wagnerjc@ornl.gov

Reference: ORNL/LTR-2011/281

August 30, 2011

Dr. Brady D. Hanson Radiochemical Science & Engineering Group Pacific Northwest National Laboratory 902 Battelle Boulevard P.O. Box 999, MSIN P7-27 Richland, Washington 99352

Dear Brady:

#### **End-of-Year Status Report on Storage R&D Opportunities Support – ORNL Fuel Cycle Research and Development (FCR&D) Milestone FCRD-USED-2011-000270 – Due 8/31//2011**

This letter documents the completion of the End-of-Year Status Report on Storage R&D Opportunities Support **–** ORNL Fuel Cycle Research and Development (FCR&D) Milestone FCRD-USED-2011- 000270, due September 2, 2011. This report covers work performed during FY11 in support of the Storage R&D Opportunities work package. The Oak Ridge National Laboratory (ORNL) FY11 scope of work focused on three areas:

- Providing general support for meetings, travel, and document development and review of Storage R&D Opportunities milestone reports on what is needed to form the technical basis for very longterm storage of used nuclear fuel
- Developing an ORNL Used Fuel Inventory Report
- Initiating the development of plans for experimental clad testing work to support UFD

If you have any questions, please contact Rob Howard at (865) 241-5750 or me at (865) 274-1184.

Sincerely,

John C. Wagner

John C. Wagner, Ph.D. Reactor and Nuclear Systems Division

JCW:dw

Enclosure

c: G. L. Bell R. L. Howard J. M. Scaglione K. Sorenson, SNL Y. Yan

#### **LETTER REPORT**

Reactor and Nuclear Systems Division



Prepared for the Used Fuel Disposition Campaign Fuel Cycle Research and Development (FCR&D) Program Office of Nuclear Energy U.S. Department of Energy

> Prepared by Oak Ridge National Laboratory P.O. Box 2008 Oak Ridge, Tennessee 37831-6170 managed by UT-BATTELLE, LLC for the U.S. Department of Energy under contract DE-AC05-00OR22725

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## **1. PURPOSE AND BACKGROUND**

This status letter report documents work performed by Oak Ridge National Laboratory (ORNL) staff for the Department of Energy (DOE) Office of Nuclear Energy, Office of Fuel Cycle Technology Used Fuel Disposition Campaign (UFDC). Within the UFDC, the Storage and Transportation task has been created to address issues of extended or long-term storage and transportation. This report covers work performed during FY11 in support of the Storage R&D Opportunities work package. A mid-year status report that covered the period from October 1, 2010 through March 31, 2011 was provided on April 21, 2011.

When fuel is no longer capable of efficiently sustaining a chain reaction, it is removed from the reactor and is termed used nuclear fuel (UNF) or spent nuclear fuel (SNF). Because of the high heat load and radioactivity, UNF is initially stored in water-filled pools to provide both cooling and shielding. Reactors were not designed or built to store all of the UNF produced over their lifetime of operation. This is especially true for reactors applying for license extensions of up to 20 years, bringing their total operating lifetime to 60 years. Most reactors initially addressed this storage shortfall by reracking their pools to increase the in-pool storage capability by decreasing the spacing between assemblies. Typically this also requires the use of additional fixed neutron poisons and burnup credit to provide the required reactivity margin to demonstrate subcriticality. As the pools reach capacity, it is necessary to remove assemblies that have been sufficiently cooled so the utility can maintain the desired full-core offload capability and to prevent premature shutdown of the reactor. Without an operating repository, centralized storage facility, or reprocessing facility, the only option is to build additional onsite storage, either wet or dry. Because dry storage systems are designed to allow passive cooling, their overall cost and maintenance are expected to be less than the cost and maintenance for an additional pool. The commercial nuclear industry has been actively pursuing dry storage to meet its fuel storage needs.

Until a disposition pathway, either recycling or geologic disposal, is chosen and implemented the storage periods for UNF will likely be longer than were originally intended. The ability of the important-to-safety structures, systems, and components (SSCs) to continue to meet safety functions over extended times must be determined. In addition, it needs to be determined if these SSCs can also meet applicable safety functions when the used nuclear fuel is transported to its final location. To facilitate all options for disposition and to maintain retrievability and normal back-end operations, the likelihood that the used nuclear fuel remains undamaged after extended storage needs to be evaluated. This does not preclude consideration of other options, such as canning of all UNF, from a total systems perspective to determine overall benefit to nuclear waste management.

The US Department of Energy Office of Nuclear Energy (DOE-NE), Office of Fuel Cycle Technology has established the UFDC to conduct the research and development (R&D) activities related to storage, transportation, and disposal of UNF and high-level radioactive waste (HLW). The mission of the UFDC is:

*To identify alternatives and conduct scientific research and technology development to enable storage, transportation and disposal of used nuclear fuel and wastes generated by existing and future nuclear fuel cycles.*

The near-term objectives of the Storage and Transportation task within the UFDC are to use a sciencebased approach to

- Develop the technical bases to support the continued safe and secure storage of UNF for extended periods
- Develop the technical bases for retrieval of UNF after extended storage

 Develop the technical bases for transport of high burnup fuel; as well as low and high burnup fuel after dry storage

Together, these objectives will help formulate the technical bases to support licensing for extended storage of UNF that will accommodate all disposition options.

## **2. WORK ACTIVITIES SUPPORTING STORAGE R&D OPPORTUNITIES**

The Oak Ridge National Laboratory (ORNL) FY11 scope of work supporting the Storage R&D Opportunities Work Package focused on three areas:

- Providing general support for meetings, travel, and document development and review of Storage R&D Opportunities milestone reports on what is needed to form the technical basis for very long term storage of used nuclear fuel
- Developing an ORNL Used Fuel Inventory Report
- Initiating the development of plans for experimental clad testing work to support UFD

## **3. YEAR END STATUS OF WORK**

## **3.1 General Support for Meetings, Travel, and Document Development and Review of Storage R&D Opportunities Milestone Reports**

FY11 work for this area began in October 2010. ORNL staff initiated this effort by performing an in-depth technical review of the FY10 *Used Fuel Disposition Campaign Storage Research and Development Opportunities Work Package Year End Report.* Comments on the report were provided to the document authors on November 23, 2010. As a follow-on effort, ORNL staff provided technical review and comment on several drafts of *Gap Analysis to Support Extended Storage of Used Nuclear Fuel FCRD-USED-2011-000136* in order to support timely completion of that deliverable.

John Scaglione, Rob Howard, and Yong Yan participated in the UFD Storage Research and Development Opportunities Workshop, which was held in Las Vegas, Nevada, February 1-3, 2011. The objectives of the meeting were to (1) describe the required R&D approach to close gaps, including preliminary assessment of cost, timeline and essential materials or conditions for success; and (2) outline a reasonable approach/roadmap for the first (and subsequent, if possible) phase(s) of a research program. The results of the workshop were used as input to the Storage R&D Research Opportunities Level 1 Milestone M11UF041401 *Gap Analysis to Support Extended Storage of Used Nuclear Fuel FCRD-USED-2011- 000136.* Input provided by ORNLwas included as Appendix A to the mid-year status report.

## **3.2 Development of an ORNL Used Fuel Inventory Report**

A significant issue related to developing the experimental program necessary to support the technical bases for continued safe, long-term storage of used nuclear fuel that will accommodate all final used fuel management options is the identification and acquisition of representative and appropriate used fuel for testing. All DOE sites are limited to varying degrees on the receipt, and extended storage of additional used fuel due to state restrictions and the questions involving the transfer of fuel ownership (a final disposition path does not exist). Presently, it is a challenge to move (and ultimately dispose of) even small research quantities of UNF within the DOE complex. Since this issue will likely not be resolved in the immediate future, ORNL has developed a Used Fuel Inventory Report to identify a list of existing used fuel rod segments at ORNL and their characteristics (sample types, sample lengths, estimated burnup, initial enrichment) for potential use in the UFD campaign. Cladding materials from some high burnup rods are also available and have been characterized to some extent (the corrosion layer, hydrogen content, hydride morphology). Samples include used fuel clad with traditional zirconium alloys Zry-2 and Zry-4, and advanced alloy M5. The Used Fuel Inventory Report is included as Appendix A.

## **3.3 Initiating the Development Plans for Experimental Clad Testing Work to Support UFD**

Hanson et al. 2011 noted that significant data is needed to determine the effects of high burnup and different clad alloys on hydrogen embrittlement and reorientation and their subsequent effect on the ability of cladding to remain in the same condition it was in when emplaced in dry storage. Very little, if any, data is publicly available on the newer cladding alloys and on high-burnup cladding. Data that is available is often on unirradiated cladding.

Experimental work must be performed to obtain this data and to obtain additional information to address the numerous disagreements regarding cladding behavior, such as whether low-temperature mechanisms (e.g., for DHC, annealing, and creep) are important over extended storage.

Irradiation is known to have a significant impact on the properties and performance of Zircaloy cladding and structural materials. High-energy neutrons (>1 MeV) are known to produce two different dislocation loops, the  $\langle a \rangle$  and  $\langle c \rangle$  loops. The size and density of the dislocation loops alter the mechanical properties, specifically the strength (e.g., hardness, tensile strength, burst strength) and ductility (e.g., uniform and total elongation strains). Irradiation increases the cladding strength and decreases the cladding ductility by creating these dislocation loops and by changing the configuration (amorphization) of the second-phase precipitates (SPPs) such as Zr(Nb,Fe)2. The processes that lead to dislocation formation or SSP amorphization depend on the material temperature; as a result, the irradiation temperature has an important effect on the cladding microstructure and consequently the mechanical properties. Higher irradiation temperatures result in larger <a> loop dislocations, whereas <c> loops do not form at 77°C (EPRI 2006). Thermal annealing, such as can occur at the higher clad temperatures during drying or initial dry storage, can result in a dramatic decrease in hardness and corresponding increase in ductility.

Normal operation in reactors can not only result in irradiation damage of cladding, but also introduce hydrogen into the metal due to formation of a waterside corrosion layer. During reactor operations, the cladding undergoes outer surface corrosion as the high-temperature water reacts with the cladding, producing a zirconium oxide layer. Hydrogen is released during this chemical reaction, and a fraction of this hydrogen is absorbed by the Zircaloy (hydrogen pickup). The solubility of hydrogen in zirconium is highly temperature-dependent, with increased solubility at higher temperatures. When the concentration of hydrogen exceeds the solubility limit, zirconium hydrides form. Depending on the size, distribution, and orientation, these hydrides can embrittle the cladding and reduce ductility. Furthermore, the presence of hydrides can facilitate cracking if the hydrides are aligned radially, perpendicular to the tensile stress field. Cladding hydrides are typically observed to be oriented in the circumferential direction but can reorient to the radial direction, depending on the stress level of the cladding when it is cooled from a higher temperature, such as will occur following the drying process. Hydrides have also been shown to diffuse to colder regions of the cladding under a relatively small temperature gradient. The reorientation and diffusion of hydrides can result in cracking of the cladding.

One of the primary needs identified in the *Gap Analysis to Support Extended Storage of Used Nuclear Fuel FCRD-USED-2011-000136* is to establish the link between the behavior and performance of unirradiated cladding and actual irradiated cladding. The UFDC is planning to address this need through both testing and modeling and in collaboration with university partners under NE Universities Program (NEUP). ORNL has begun to address part of the testing component of this linkage with and experimental

concept to simulate high-burnup used cladding by irradiating cladding material in the High Flux Isotope Reactor.

Concept development in this area began during the second half of FY11. Laboratory testing of used nuclear fuel can require a long lead time for preparation, be of long duration, difficult to repeat, relatively expensive to perform, and can potentially be used either directly or indirectly in future licensing activities. Because of these factors, it is important that appropriate planning and controls for this work be developed in advance. The general concept is as follows:

**Title:** Neutron Irradiation of Hydrided Zirconium-alloy Cladding in the HFIR: a Simulation Approach for High Burnup Used Fuel Cladding

Fast neutron irradiation of pre-hydrided zirconium-alloy cladding in the High Flux Isotope Reactor (HFIR) at elevated temperatures is proposed to simulate the effects of high burnup on used fuel cladding for use in understanding the materials properties relevant to very long-term storage (VLTS). The irradiated pre-hydrided metallic materials will generate baseline data to benchmark hot-cell testing of high-burnup used fuel cladding at relatively low cost, and more importantly, samples free of alpha contamination can be provided to the researchers/students in universities that do not have hot cell facilities to handle highly contaminated high-burnup used fuel cladding to support their research projects for the UFDC.

This simulation approach should provide well-controlled neutron irradiation of pre-hydrided materials in the desired temperature range  $(200-350^{\circ}C)$ , similar to the service temperatures of the BWR and PWR. The pre-hydrided specimens will be fabricated by using the existing pre-hydrogen charging system at the ORNL, which has already been developed by an ongoing program for the NRC. The hydrogen content will be in the range of 100-800 wppm, the typical values for high burnup used fuel cladding. The hydrogen content will be analyzed, and the hydride morphology will be characterized and compared to the high burnup used fuel cladding to optimize experimental conditions. The pre-hydrided specimens will be irradiated up to a fast fluence of  $2.3 \times 10^{21}$  neutrons/cm<sup>2</sup> (>1 MeV), corresponding to burnup level of 70 GWd/MT. In addition, the project can be expanded by providing the service to universities or other customers to hydride and irradiate their customer-designed/fabricated specimens in the future. Furthermore, samples could be provided to support long-term annealing tests to investigate the evolution of clad strength in the dry storage conditions.

Additionally, initial collaboration with Sandia National Laboratory has begun to identify possible sources of cladding material that may be considered excess or scrap from previous clad testing efforts. Appendix B contains the preliminary scope, ROM cost, and schedule summary of activities that will likely be initiated in FY12 related to producing simulated high burn up clad samples.

Irradiation	$200 - 250$ °C	Service temperature of the cladding for BWR			
temperature	$300 - 350$ °C	Service temperature of the cladding for PWR			
Typical sample size	Cladding OD: 9.5-10 mm	Varies with cladding type			
	Wall-thickness: $0.60-0.76$ mm	Varies with cladding type			
	Length: $25-75$ mm	75 mm for as-received, 25 mm for hydrided samples			
<b>Neutron</b> fluence $(>1$ MeV)	$1.0 \times 10^{21}$ n/cm <sup>2</sup>	Corresponding to burnup level of 30 GWd/MT			
	$1.7 \times 10^{21}$ n/cm <sup>3</sup>	Corresponding to burnup level of 50 GWd/MT			
	$2.3 \times 10^{21}$ n/cm <sup>4</sup>	Corresponding to burnup level of 70 GWd/MT			
Materials	Zry-2, Zry-4, M5, and ZIRLO	M5 and ZIRLO: advanced zirconium alloy for PWR			
	As-received and hydrided Zr alloys	Pre-hydrided materials will be fabricated at ORNL			
Total sample length	$36-72$ inch	May need be irradiated in separated core locations			

**Table 1. Proposed neutron irradiation at the HFIR**

## **3.4 References**

B. Hanson, H. Alseaed, C. Stockman, D. Enos, R. Meyer, K. Sorenson, *Gap Analysis to Support Extended Storage of Used Nuclear Fuel FCRD-USED-2011-000136,* PNNL-20509, Pacific Northwest Laboratory, Richland, WA (2011)*.*

*Recovery of Irradiation Damage by Post-Irradiation Thermal Annealing:Relevance to Hydrogen Solubility and Dry Storage Issues,* TR-1013446, Electric Power Research Institute, Palo Alto, CA (2006).

#### **APPENDIX A**

#### **A.1. Spent Fuel Rod Segments at ORNL and their Characteristics**

This report provides an inventory of existing used fuel rod segments at Oak Ridge National Laboratory (ORNL) and their characteristics (sample types, sample lengths, estimated burnup, initial enrichment…) for potential use in the Used Fuel Disposition Campaign (UFDC). Cladding materials from some high-burnup rods have been characterized (the corrosion layer, hydrogen content, hydride morphology...) to meet the needs of previous research programs. Besides the traditional zirconium alloys Zry-2 and Zry-4, the high burnup advanced alloy M5 cladding is also available at ORNL. In addition, the post-irradiation examination (PIE) on the mixed uranium and plutonium oxide fuel (MOX) rods has been underway at ORNL since 2008. The leftover material from the MOX fuel rods and other fuel rods are potential candidates for use in the UFD campaign.

The irradiated materials were supplied to ORNL for several ongoing PIE programs taking place in laboratory hot cells. This material includes TMI-1 PWR fueled rods at 48-50 GWd/MTU, H. B. Robinson PWR fueled rods at 64-67 GWd/MTU, Limerick BWR fueled rods at 54-57 GWd/MTU, North Anna M5 fueled rods at 63-70 GWd/MTU, Surry-2 PWR fueled rods at 36 GWd/MTU, Calvert Cliffs PWR fueled rod at 38-46 GWd/MTU, Cooper BWR fueled rod at 24 GWd/MTU, and Catawba MOX fueled rod at 45 GWd/MTU. Spent fuel rod segments stored in Building 3525 are summarized in Table A.1, and are detailed in the following sections.

<b>Reactor name</b>	H. B. Robinson	Limerick	Surry	$TMI-1$	Calvert Cliffs	Copper	North Anna	Catawba (MOX)
<b>Reactor type</b>	<b>PWR</b>	<b>BWR</b>	<b>PWR</b>	<b>PWR</b>	<b>PWR</b>	<b>BWR</b>	<b>PWR</b>	<b>PWR</b>
Enrichment, $wt$ %	2.90	3.40-3.95	3.1%	4.00	$2.45$ to 3.04%	1.33-2.93	4.20	2.4 to 5%
Burnup, <b>GWd/MTU</b>	63-67	54-57	36	$48 - 50$	43	28	63-70	40-47
<b>Discharge</b> date	1995	1998	1981	1997	1982	1982	2004	2008
<b>Cladding</b>	$Zry-4$	Zr-lined $Zry-2$	$Zry-4$	Low-Sn $Zry-4$	$Zry-4$	$Zry-2$	M <sub>5</sub>	M <sub>5</sub>
Nominal OD, mm	10.76	11.18	10.72	10.92	11.18	14.3	9.50	9.50
<b>Initial wall</b> thickness, mm	0.76	0.71	0.62	0.69	0.66	0.94	0.57	0.57
OD oxide, $\mu$ m	$\leq 100$	$\approx 10$	<40	$\leq 50$	<b>Not</b> provided	<b>Not</b> provided	$<$ 20	${<}10**$
Hydrogen pickup, wppm	$\leq 800$	70	$<$ 300	$\leq 300$	<b>Not</b> provided	<b>Not</b> provided	< 120	$<$ 55
<b>Fueled</b>	Yes	Yes	Yes	Yes	Yes	Yes	Yes	Yes
Total length, inch	248*	216	151	146	22	8	163	472
<b>Availability</b> for ORNL <b>Tests</b>	$EPT,***$ tensile	EPT,*** tensile. bend, fatigue	EPT,*** tensile. bend, fatigue	$EPT,***$ tensile, bend, fatigue	N <sub>o</sub>	<b>No</b>	EPT***	<b>TBD</b>
<b>Availability</b> for other labs	<b>No</b>	<b>TBD</b>	<b>TBD</b>	<b>TBD</b>	N <sub>o</sub>	<b>No</b>	<b>No</b>	<b>TBD</b>

**Table A.1. Characteristics of high-burnup fuel rod segments in ORNL Building 3525 (burnup values are rod averaged)**

\*Including 39" defueled cladding.

\*\*Estimated.

\*\*\*Expanding plug test.

#### **A.1.1 High-Burnup H. B. Robinson PWR Fuel Rods**

The high-burnup PWR rods received by ORNL in 2008 were from a  $15\times15$  assembly of the H.B. Robinson plant Unit 2 [1]. They operated for seven cycles and reached a rod-average burnup of 67 GWd/MTU (73 GWd/MTU peak pellet). The fuel enrichment is 2.90%. The nominal fuel pellet dimensions are 9.06 mm dia.  $\times$  9.93 mm height and the active fuel height is 3.66 m. The cladding is cold-worked/stress-relieved Zircaloy-4, 10.77 mm OD  $\times$  9.25 mm ID, with a nominal tin content of 1.42%. The rods were pressurized with helium to 2.0 MPa during fabrication. A detailed description of the as-fabricated cladding, the irradiation history, the nondestructive testing results (eddy current, profilometry, fission-gas release, etc.) is given by the plant operator and fuel vender [1].

The condition of the fuel column and axial fission product migration were evaluated with scanning gamma spectroscopy for selected rods as part of the NRC spent-nuclear-fuel program at ANL [2, 3]. The results are shown in Fig. A.1 for one of the rods, A02. The gross distribution shows no unusual features

such as fuel column disruption or excessive fission product migration. Slight dips were observed at the grid spacer locations due to flux depression. Fuel ceramography, cladding metallography, and hydrogen concentration measurements were performed for rod F7, which was neither near assembly edges nor next to guide tubes. The expectation was that cladding sectioned from this rod would have more uniform hydrogen concentration and hydride morphology.



**Fig. A.1. Cross gamma scan profile for H. B. Robinson Rod A2. The profile is the composite of five scan as the rod was pre-sectioned into five segments.**



**Fig. A.2. High-burnup HBR fuel morphology for the fuel midplane cross section of rod F07 showing indications of asymmetric power and temperature distributions relative to the center of the pellet.**

Detailed characterization was performed for the midplane region of the F07 rod to determine the fuel, fuel-cladding bond, corrosion layer, and hydride morphology. Figure A.2 shows a low magnification image of the fuel morphology. The central darkened region is not symmetric with respect to the center of the pellet. This result indicates asymmetric power distribution and fuel temperature, as well as circumferential variation in cladding temperature. The cross-section of the fuel in Fig. A.2 also reveals the typical start-up and shut-down cracks. The fuel-cladding bond appears to be well developed at the F07 rod midplane. Figure A.3 shows the fuel-cladding bond within one circumferential sector of the fuel shown in Fig. A.2. The bond thickness is  $11\pm4$  μm. According to Une et al. [4], the bond layer is primarily  $ZrO<sub>2</sub>$  with some  $UO<sub>2</sub>$  in solid solution.



(a)



(b)

**Fig. A.3. Fuel (dark), cladding (light), and fuel-cladding bond layer (gray) at the midplane of HBR rod F07. The image of Fig. A.3(b) is a magnification of a small region from the fuel cross section shown in Fig A.3(a).**

The corrosion layer thickness was measured to be  $71\pm5$  μm at the midplane and  $95\pm4$  μm at 650 mm above the midplane. These values are in good agreement with the poolside NDE data. Within the layer, an occasional radial crack can be seen, but oxide spallation is not prevalent. Metallographic images of the corrosion layer are shown in Fig. A.4 at two relevant axial locations along the rod.







Hydrogen content in the cladding was analyzed using a fusion/thermal conductivity technique. The results show hydrogen contents of  $\approx 550$  wppm at axial midplane and  $\approx 740$  wppm at 650 mm above the midplane. The concentrations are consistent with the observed oxide layer thickness, corresponding to a hydrogen uptake ratio of  $\approx$ 23%. With the cladding in etched condition, a dense hydride band adjacent to the cladding outer surface oxide layer can be seen (see Fig. A.5). The densities of hydrides decreases towards to the cladding inner surface. No unusual hydride morphology was observed in the area examined. Orientation of the precipitates is mostly circumferential. Since the terminal solubility of hydrogen [5] in the cladding is 200 wppm at the reactor operation temperature, some of the precipitates existed in the cladding during operation.



**Fig. A.5. Hydride distribution and morphology in HBR rod F07 cladding at locations: (a) fuel midplane (550-wppm H); (b) 650 mm above midplane (740-wppm H).**

## **A.1.2 High-Burnup Limerick BWR Fuel Rods**

The high burnup Limerick BWR rods were from a 9×9 assembly. They operated for three cycles and reached a rod-average burnup of 56 GWd/MTU (64 GWd/MTU peak pellet). The fuel enrichment is 3.40-3.95%. The nominal fuel pellet height is 3.71. The cladding was Zr-lined Zircaloy-2, recrystallizedannealed with an OD of 11.18 mm and an ID of 9.75 mm. The rods were pressurized with helium during the fabrication.

The condition of the fuel column and axial fission product migration were evaluated with scanning gamma spectroscopy for selected rods as part of the NRC LOCA program at ANL [6, 7]. The results are shown in Fig. A.6 for Rod F9. The gross distribution shows no unusual features such as fuel column disruption or other unusual behavior. Slight dips were observed at the grid spacer locations due to flux depression.



**Fig. A.6. Cross gamma scan profile for Limerick Rod F9. The profile is the composite of five scans as the rod was pre-sectioned into five segments.**

Figure A.7a shows the fuel structure of the as-received Limerick fuel at an axial elevation of ≈700 mm above the midplane. The off-center restructuring might be partially due to the edge location of the F9 rod in the assembly. The fuel-cladding gap is closed and a fission product phase can be seen in the F9 rod at high magnification images. Figure A.7b shows a high-magnification of a small area of the mid-radius of the Limerick pre-LOCA fuel with the high concentration of fission gas within the grains, as well as some fission gas bubbles on grain boundaries.



**Fig. A.7. Low-magnification cross-sectional photo-composite of Limerick Rod F9 at an elevation 700 mm above the fuel axial midplane (a) and high-magnification of a small area of the mid-radius of the Limerick fuel with the high concentration of fission gas within the grains, as well as some fission gas bubbles on grain boundaries (b).**

The oxide layer on the Limerick cladding surface is thin,  $\approx 10$  µm average. No oxide spallation was observed. At some locations, a thin crud  $(10 \mu m)$  can be found over the oxide (see Fig. 8a). The crud contains zinc, probably related to the zinc injection procedure used in the Limerick plant for dose-buildup control. Measured hydrogen content in the cladding is  $\approx$ 70 wppm. Figure A.7b shows the hydrides precipitated in the cladding. The precipitates in the Zry-2 are small  $(100 \mu m)$  and uniformly distributed across the wall thickness.





**Fig. A.8. (a) Micrographs showing cruds over the oxide layer on the OD surface of Limerick Rod F9, (b) distribution of hydrides precipitated in the Limerick cladding.**

## **A.1.3 Dry-Cask Stored Surry PWR Rods**

The Surry fuel rods in ORNL were from one of the  $15\times15$  Westinghouse assemblies loaded in a castor-V/21 dry cask in the mid-1980s for benchmarking the thermal and radiological codes for dry-cask storage [8]. After the benchmark test, the fuel rod was stored in a dry inert atmosphere Castor V/21 cask at the Idaho National Environmental and Engineering Laboratory (INEEL) for 15 years at peak cladding temperatures decreasing from about 350 to 150°C. The cask was opened to examine the fuel for degradation and to determine if it was suitable for extended storage. No rod breaches had occurred and no visible degradation or crud/oxide spallation was observed for the rod retrieved.

The assembly was irradiated for three cycles to achieve a burnup of 36 GWd/MTU (40 GWd/MTU peak pellet) and attained near the highest cladding temperature among the rods in the cask during the benchmark tests ( $\approx$ 415°C peak for several days). The assembly-average fast (E>1 MeV) neutron fluence is calculated to be  $6.38 \times 10^{25}$  n/m<sup>2</sup>. It was discharged in November 1981 and was in water storage until transported to INEEL (now INL) and loaded into the Castor cask in July 1985. The fuel enrichment is 3.1% and the nominal fuel pellet dimensions are 9.29 mm dia.  $\times$  15.2 mm height, with an active fuel height 3.66 m. The cladding is Zircaloy-4, cold-worked and partially annealed, with dimensions of 10.72 mm  $OD \times 9.5$  mm ID.



**Fig. A.9. Average outer diameter profile for Surry Rod H9 after 15 years of dry cask storage in He.** 

Profilometer of 12 of the post-storage rods shows the cladding creep-down was 0.6% [9]. The results are shown in Fig. A.9 for Rod H9. As this value is typical of PWR rods at this burnup, it suggests no significant outward creep of the cladding during the benchmark tests or the extended cask storage. The condition of the post-storage fuel is shown in Fig. A.10 for Rod H9. All features appear to be normal with no evidence if degradation from the extended storage, perhaps due to the lower temperature of the fuel during the storage compared to in-reactor temperatures. The fuel/cladding gap is open and fuel/cladding chemical interaction and fission product deposit in the gap are both insignificant. Cross-sectional photo-composite (see Fig. A.10) shows a pellet cracked into 10-25 pieces, which is prototypical of this fuel at this burnup.



**Fig. A.10. Low-magnification cross-sectional photo-composite of Surry Rod H9 at an elevation 510 mm above the fuel axial midplane.**

The cladding oxide thickness over the outer surface in the post-storage Surry rods ranged from 25 µm at the midplane (see Fig. A.11) to 40  $\mu$ m at 1.0 m above the midplane. These values are within the normal range for Zry-4 PWR rods of this burnup and suggest that no additional oxidation during cask storage. Measured hydrogen contents in the cladding are 250±40 wppm at the axial midplane, consistent with the observed oxide thickness. The concentrations are consistent with the observed oxide layer thickness, corresponding to a hydrogen uptake ratio of  $\approx$  24% at the midplane by using a Pillings-Bedworth Ratio of 1.75 for the corrosion layer.

The cladding was etched to reveal the hydrides. The radial location and the density of hydrides are shown in Fig. A.12. In spite of the internal pressure during the benchmark tests and after 15 years storage in a dry inert atmosphere Castor V/21 cask, the hydrides are aligned in the circumferential direction with no hydride re-orientation in the radial direction. Probably because of a lack of strong radial temperature gradient while in the cask, the hydride is fairly uniform across the cladding wall.



**Fig. A.11. Cladding outer surface corrosion layer for Surry-2 rod H9 at axial midplane.**



**Fig. A.12. Hydride structure in post-storage Surry Rod H9. The axial elevation is 0.50 m above the midplane.**

## **A.1.4 High Burnup TMI PWR Rods**

The high-burnup TMI PWR rods were from a  $15\times15$  assembly. They reached a rod-average burnup of 48-50 GWd/MTU before they were discharged in 1995. The fuel enrichment is 4.00%. The nominal fuel pellet diameter is 9.36 mm and the active fuel column length is 3.60 m. The cladding is Zircaloy-4, 9.36 mm  $OD \times 7.98$  mm ID.

High-magnification images were taken from three axial locations of a TMI rod to determine corrosion layer thickness, cladding metal thickness, and hydride distribution and morphology. The corrosion layer thickness over the cladding outer surface was measured to be  $30 \mu m$  at  $0.8 \text{ m}$  above the midplane, 19  $\mu m$ at 0.2 m above the midplane, and 13 um at 0.6 m below the midplane, as shown in Fig. A.13. These values are within the normal range for Zry-4 PWR rods of this burnup. Hydrogen content determination was made using LECO fusion extraction analysis. Measured hydrogen contents in the cladding are ≤300 wppm, consistent with the observed oxide thickness.



**Fig. A.13. Outer-surface corrosion layer from a TMI rod: (a) 0.8 m above the midplane (30 μm); (b) 0.2 m above the midplane (19 μm), and (c) 0.6 m below the midplane (13 μm).**

The cladding was etched to reveal the hydrides. The radial location and the density of hydrides are shown in Fig. A.14. In most cases, the hydrides were circumferentially oriented. However the hydride is not uniformly distributed across the cladding wall; high density of hydride rims are observed near the corrosion layer. The fuel-cladding gap is closed and a fuel-cladding bond appears to be well developed for the TMI rod examined. According to Une et al. [4], the bond layer is primarily  $ZrO<sub>2</sub>$  with some UO<sub>2</sub> in solid solution.



**Fig. A.14. Hydride distribution and morphology in a TMI cladding sample at low magnification (a) and at high magnification (b).**



**Fig. A.15. Fuel-cladding bond layer for a TMI rod: (a) 30 μm at 0.8 m above the midplane; (b) 19 μm at 0.2 m above the midplane; and (c) 13 μm at 0.6 m below the midplane.**

#### **A.1.5 Calvert Cliffs PWR Rods**

The Calvert Cliffs PWR fuel rods in ORNL include two groups: Approved Testing Materials-104  $(ATM-104)$  and  $ATM-106$  [9].  $ATM-104$  has  $UO<sub>2</sub>$ -fueled rods with a medium burnup of about 43 GWd/MTU. The fuel enrichment is 3.04%. The nominal fuel pellet diameter is 9.56 mm and the fuel pellet is 11.43 mm long. The active fuel column length is 3.47 m. These segments were from a  $14\times14$  fuel assembly (BT03) fabricated in the mid-1970s and irradiated for four cycles from March 1977 to April 1982. The assembly operated at reduced power for a period of five months during the last reactor cycle.

All ATM-104 rods are clad with Zry-4 fabricated by Sandvik Special Metal with 9.36 mm OD  $\times$  7.98 mm ID. The condition of the fuel column was evaluated with scanning gamma spectroscopy by Guenther et al. [9]. The results are shown in Fig. A.16 for Rod MKP070. The gross distribution shows no unusual features such as fuel column disruption or other unusual behavior. Slight dips were observed at the grid spacer locations due to flux depression.



**Fig. A.16. Cross gamma scan profile for Calvert Cliffs Rod MKP070 of ATM-104. The profile is the composite of eight scans as the rod was pre-sectioned into 8 segments.**

The Calvert Cliffs PWR ATM-106 fuel rods also have  $UO_2$ -fueled rods with a medium burnup of about 43 GWd/MTU. Like ATM-104, the ATM-106 rods were from a standard 14×14 fuel assembly (BT03), but the fuel pellets are approximately 1.5 times longer than the ATM-104 fuel pellets. The fuel enrichment is 2.45%. The nominal fuel pellet diameter is 9.64 mm and the active fuel column length is 3.47 m. The ATM-106 fuel rods were irradiated for four cycles from October 1974 to October 1980. The ATM-106 fuel rods were also operated at reduced power for a period of 5 months during Cycle 4.

All ATM-106 rods are clad with Zry-4 fabricated by Sandvik Special Metal with 9.36 mm OD  $\times$  7.98 mm ID. The condition of the fuel column was evaluated with scanning gamma spectroscopy by Guenther et al. [9]. The gamma scan results are shown in Fig. A.17 for ATM-106 Rod NBD095. The gross distribution shows no unusual features such as fuel column disruption or other unusual behavior. Slight dips were observed at the grid spacer locations due to flux depression.



**Fig. A.17. Gamma scan of the ATM-6 rod NBD905.**

## **A.1.6 Cooper BWR Rods**

The Copper BWR fuel rods ARM-108 in ORNL were from a GE 7×7 fuel assembly fabricated in 1972. ATM-108 has  $UO_2-Gd_2O_3$ -fueled rods with a medium burnup of about 28 GWd/MTU. The fuel enrichment is 1.33-2.93%. The rod AND-2006 has 3 wt % gadolinia. The nominal fuel pellet diameter is 12.1 and the active fuel column length is 2.74 m. These rods were irradiated in the Cooper BWR for five cycles from March 1974 to May 1982. Because the gadolinia eventually burns out, the power in the rods would typically start at a low value and increase with time depending on the rate of burnup, the gadolinia content, and overall power.

ATM-108 rods are Zry-2 clad, 14.3 mm OD  $\times$  12.42 mm ID. The condition of the fuel column was evaluated with scanning gamma spectroscopy by Guenther et al. [9]. The results are shown in Fig. A.18 for ATM-108 Rod AND-0206. The gross distribution shows no unusual features such as fuel column disruption or other unusual behavior. Gamma scan for portion of bottom is not available.



**Fig. A.18. Gamma scan of ATM-8 rod ADN0206 (gamma scan for portion of bottom is not available).**

## **A.1.7 High Burnup North Anna PWR Rods**

The high-burnup North Anna PWR rods received by ORNL were from a 15×15 assembly. They reached a rod-average burnup of 63-70 GWd/MTU. The fuel enrichment is 4.20%. The nominal fuel pellet diameter is 8.19 mm. The active fuel height is 3.66 m and the fuel rod length is 3.86 m. The cladding is M5, 9.50 mm OD and 8.36 mm ID, with a nominal Nb content of 1.02%. The nondestructive testing results (eddy current, profilometry, fission-gas release, etc.) were not provided to ORNL by the plant operator and fuel vender. The oxide layer on the North Ann M5 cladding surface is thin,  $\langle 20 \mu m \rangle$  average. No oxide spallation was observed. The hydrogen content in the cladding is low, <120 wppm.

## **A.1.8 Catawba MOX Fuel Rods**

The MOX fuel rods received by ORNL for post-irradiation examination (PIE) work were contained in the MOX Lead Test Assemblies (LTA) irradiated at the Catawba Nuclear Power Plant. The full length rods were pulled from reactor LTAs. The rods have a length of approximately 153" (3.89 m) and a diameter of approximately 0.4" (19.16 mm) before they were discharged in 2008. Exposure time is 984 equivalent full-power days and they have received burn-ups of approximately 45 GWd/MT.

Each rod contains approximately 330 pellets of MOX fuel in an M5 alloy cladding with an end plug welded on each end. The fuel enrichment is 2.4-5.0%. The cladding is 9.50 mm OD and 8.36 mm ID, with a nominal Nb content 1.02%. The oxide layer on the Catawba MOX fuel cladding surface is thin, estimated to be  $\lt 10$  µm average. No oxide spallation was observed. The hydrogen content in the cladding is low, approximately 30-55 wppm.

The goal of the MOX PIE work is to experimentally verify the presence or absence of materials interactions in the MOX test fuel, to examine the fuel and cladding for unusual behavior, and to provide information for comparison with prediction. This PIE includes profilometry, gamma scanning, metallography, radiochemistry, and cladding mechanical testing have been underway since 2009. The PIE data are proprietary and are not publically available. However, the results can be provided with approval of the vender and sponsor.

## **A.1.9 References**

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## **Appendix B**

## **Neutron Irradiation of Hydrided Zirconium-alloy Cladding in the HFIR: a Simulation Approach for High Burnup Used Fuel Cladding—Initial Scope, Schedule and Cost Summary**



**Activity:** Perform Post-charging/Pre-irradiation Non-Destructive Examination (NDE) and Destructive Examination (DE) Materials Characterization

- **Scope**: Characterization should include hydrogen analyses and optic metallographic examinations on samples sectioned from pieces that were charged. Additionally SEM and TEM on selected samples (minimum of two SEM and two TEM examinations). TEM study will provide a baseline for the post irradiation examination (irradiation induced hydride evolution, defects, etc.). Results documented in a letter report and provided to all participants.
- **Duration:** 4 weeks (in parallel with hydrogen charging activity above)
- **ROM Cost**: \$55K

**Activity**: Prepare HFIR Experiment Design and Safety Analysis Documentation

**Scope:** 1) Create design drawings for test equipment that will be used within HFIR. These must be reviewed and approved by the designer, technical reviewer, lead QA, HFIR Safety,

- and HFIR QA. 2) Create a design document that provides supporting calculations for key design parameters, reviewed and approved by the designer, technical reviewer, and Thermal Hydraulics and Irradiation Safety Group Leader
- 3) Create one or more safety calculations that consider the following scenarios:
	- (a) steady-state operation at 130% reactor power
	- (b) small-break loss of coolant accident
	- (c) loss of offsite power
	- (d) 50% flow blockage.
- 4) HFIR Safety organization develops an Unreviewed Safety Question Document (USQD) and creates an Experiment Authorization Basis Document (EABD) based on 1), 2), and 3) above
- 5) Fabricate necessary test equipment.

**Duration**: 6 Months

- **ROM Cost**: \$250-300K
- **Activity**: Irradiate Samples in HFIR
- **Scope:** Load samples in appropriate HFIR target position(s). Irradiate for required number of operating cycles
- **Duration:** 7-14 operating cycles, depending on total neutron fluence target and experiment design.
- **Basis:** Operating cycle lengths vary between 22 and 27 days. A 25-day duration operating cycle is used for planning purposes. Operating cycles are followed by outages that typically vary between 17 and 45 days. Nominally, seven operating cycles occur within a calendar year.
- **ROM Cost**: No dollar cost, however, researchers are obligated after completing experiment to (1) publish results, with suitable acknowledgement of the ORNL facility and (2) notify ORNL of such publications. Assume journal publication costs will be included in the costs for performing detailed material performance and characterization testing on irradiated samples to be performed by the individual national lab or university participants described below.

