

# Research and Development Needs for Used Fuel Storage

## Fuel Cycle Research & Development

*Prepared for*  
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*Used Fuel Disposition Campaign*  
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## SUMMARY

The primary objective of the Used Fuel Long Term Storage Research and Development Program is to formulate a technical basis for continued safe, long-term storage of used nuclear fuel that will enhance final disposition options. The program will focus on dry storage at reactor and centralized locations with storage times exceeding the current regulatory basis of 40 to 60 yrs up to 200 yrs. This is a contribution report to a joint effort on identifying research and development needs for long term storage of spent fuel. This report focuses on fuel clad failure mechanisms associated with long term dry storage. The research and development needs identified in this report will be assimilated into a Research and Development Program Plan due in September, 2010. Research and development needs were identified as a result of exhaustive literature searches for each known and hypothesized failure mechanism. The results are documented in summary tables provided in this report.

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## ACRONYMS

ANL – Argonne National Laboratory

DCSS – Dry Cask Storage System

DHC – Delayed Hydride Cracking

EPRI – Electric Power Research Institute

MOX – Mixed Oxide

NRC – Nuclear Regulatory Commission

PNNL – Pacific Northwest National Laboratory

R&D – Research and Development

# USED FUEL DISPOSITION CAMPAIGN RESEARCH AND DEVELOPMENT NEEDS FOR USED FUEL STORAGE

## 1. INTRODUCTION

The primary objective of the Used Fuel Long Term Storage Research and Development (R&D) Program is to formulate a technical basis for continued safe, long-term storage of used nuclear fuel that will enhance final disposition options. The program will focus on dry storage at reactor and centralized locations with storage times exceeding the current regulatory basis of 40 to 60 yrs up to 200 yrs. Although the initial emphasis of the program will be on light water reactor, uranium oxide fuel, alternative and advanced fuel concepts will be addressed in this program. Because very little information is available on long term storage of high burnup fuel (up to and exceeding 45 GWd/MTU), and because much of the fuel in today's reactors are moving to higher burnups, a particular emphasis of this program will be focused on high burnup fuels.

The first step in formulating a R&D Program Plan is to identify the research needs associated with establishing the technical basis for long term storage. To that end, an exhaustive literature search was conducted to identify known and hypothetical failure mechanisms, what is known about each of those mechanisms, and the consequential R&D data needs associated understanding the behavior of these mechanisms. The results of this study are provided in tabular format. This report focuses on fuel clad interactions.

## 2. R&D NEEDS

### 2.1 Functional Requirements

Each of the failure mechanisms identified were associated with one or more of five functional requirements for the system:

- Thermal performance: the ability of the system to withstand thermal conditions and stresses
- Radiological protection: the ability of the system to provide radiological dose protection
- Confinement: the ability of the system to maintain confinement of dispersible material
- Sub-criticality: the ability of the system to avoid exceeding criticality safety conditions
- Retrievability and transportability: the ability of the system to withstand forces and conditions associated with handling and transporting the used fuel after prolonged storage.

These functional requirements are the principal drivers for setting the performance conditions of the fuel, and to which each failure mechanism must be evaluated.

### 2.2 Summary Tables of Technical Issues and R&D Needs

The following tables summarize the results of the literature studies to define failure mechanisms, the functional requirements, results of the literature search, resultant data needs, and the importance of the mechanism to licensing for extended dry storage.

**Table 2.2.1 Summary of Technical Issues – Fuel/Clad System**

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Cladding Creep	<p>Confinement</p> <p>Sub-criticality</p> <p>Retrievability &amp; Transportability</p>	<ol style="list-style-type: none"> <li>1. The main driving force is the hoop stress caused by internal rod pressure, which will decrease over time as the decay heat decreases.</li> <li>2. Similarly, creep is considered self-limiting. As the internal volume of the rod increases, the pressure decreases and reduces the hoop stress. Once a rod is breached, with either pinhole or hairline cracks the most likely defect, the internal pressure decreases and reduces the hoop stress.</li> <li>3. Examination of used fuel with burnups of up to ~35 GWd/MtU that had been stored for approximately 15 years in a Castor V/21 dry storage cask showed that the maximum creep was no more than 0.1%.<sup>a</sup> However, no rod profilometry data was available for the rods prior to storage, so this conclusion is based on comparison to as-fabricated data and not comparison of actual experimental data.</li> <li>4. In creep tests at temperatures between 250-400°C of Zircaloy</li> </ol>	<ol style="list-style-type: none"> <li>1. Creep/strain models have not considered strain associated with the fuel pellets, which at high burnup, tend to have closed the gap and are in direct contact with the cladding with fuel-clad bonding during in-reactor operation such that cladding stresses are introduced on cooling. This strain would be present even at low temperatures and after the rod has been depressurized due to a breach. Even though this residual stress would not likely drive much additional creep as additional creep strain would remove the cladding stress, confirmatory data is advised.</li> <li>2. While the main driving force decreases over time, data only exists for up to 15 years of storage for Zry-2 and Zry-4 fuels. <ol style="list-style-type: none"> <li>a. Extrapolation of data up to 120 or more years is necessary.</li> <li>b. There is only limited data on new cladding types (e.g., ZIRLO™ or M5™).</li> <li>c. There is only limited data</li> </ol> </li> </ol>	<p>Intact fuel cladding is important in containing and confining radionuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.</p> <p>Current NRC regulations and guidance<sup>c</sup> are directed at preventing “gross rupture” of the cladding. While creep is not expected to result in gross rupture, it can lead to breach that allows other degradation mechanisms to occur.</p> <p>Even though clad breach may not occur, it is important to determine if creep over</p>



		<p>cladding irradiated up to a burnup of 64 GWd/MtU, no failures have been observed below 2% strain.<sup>b</sup> However, all test durations were between a few days to at most 83 days (2000 hours).</p> <ol style="list-style-type: none"> <li>5. At temperatures below 300°C, creep may be considered to be immeasurably slow and is not a factor in extended storage under normal operation.<sup>b</sup></li> <li>6. For all fuel burnups, the maximum calculated fuel cladding temperature should not exceed 400°C for normal conditions of storage and short-term loading operations (e.g., drying, backfilling with inert gas, and transfer of the cask to the storage pad).<sup>c</sup></li> <li>7. For low burnup fuel, a higher short-term temperature limit may be used if it can be shown that the cladding hoop stress is equal to or less than 90 MPa for the temperature limit proposed.<sup>c</sup></li> <li>8. During loading operations, repeated thermal cycling should be limited to less than 10 cycles, with cladding temperature variations less than 65°C each.<sup>c</sup></li> <li>9. For off-normal and accident conditions, the maximum cladding temperature should not exceed 570°C.<sup>c</sup></li> </ol>	<p>on creep in high burnup fuels, and even then for very limited times.<sup>d,e</sup></p> <ol style="list-style-type: none"> <li>d. Temperatures may be higher for longer periods of time not only for higher burnup fuels or MOX, but for the newer, larger DSS designs.</li> <li>e. Fission gas release and He release may increase internal pressure over time, especially for MOX fuels.</li> </ol> <ol style="list-style-type: none"> <li>3. Only one documented case of crack initiation (from an intentional defect for oxidation response testing) at a strain rate of at most 1% exists.<sup>f</sup> Still, the influence of material parameters like alloy composition, fabrication steps, hydride content, fast neutron fluence, and annealing effects must be considered when applying creep models.<sup>b</sup> A better understanding of the relationship between data on unirradiated cladding, data on irradiated cladding, and data on annealed cladding is necessary.<sup>g</sup></li> <li>4. There are multiple mechanisms for cladding creep.<sup>h</sup> These mechanisms come into play under different temperature and stress regimes. Murty warns that “blind extrapolations of the</li> </ol>	<p>extended periods would limit the future transportability of the fuel.</p>
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			<p>short-term, high stress data to low stresses and temperatures could lead to nonconservative predictions of the creep rates, creep strains, and lifetimes due to the dominance of viscous creep mechanisms, such as Nabarro-Herring, Coble, and Harper-Dorn creep at low stresses.”<sup>h</sup></p> <p>5. No data or analyses on the effects of long-term storage on transportability of the fuel.</p>	
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- a) *Dry Cask Storage Characterization Project*, EPRI, Palo Alto, CA, U.S. Nuclear Regulatory Commission - Office of Nuclear Regulatory Research, Washington D.C., U.S. Department of Energy – Office of Civilian Radioactive Waste Management, North Las Vegas, NV, U.S. Department of Energy – Idaho Operations Office, Idaho Falls, ID: 2002. 1002882.
- b) *Technical Bases for Extended Dry Storage of Spent Nuclear Fuel*, EPRI, Palo Alto, CA: 2002. 1003416.
- c) USNRC, 2003. Interim Staff Guidance, 11, ISG-11 Rev-3, U.S. Nuclear Regulatory Commission, Washington DC, November 2003.
- d) Goll, W., H. Spilker, and EH Toscano, “Short-time creep and rupture tests on high burnup fuel rod cladding”, *Journal of Nuclear Materials* **289**(2001)247-253.
- e) Quecedo, M., M. Lloret, JM Conde, C. Alejano, JA Gago, and FJ Fernandez, “Results of Thermal Creep Test on Highly Irradiated ZIRLO”, *Nuclear Engineering and Technology* **41**, 2 (2009) 179.
- f) Novak, J., IJ Hastings, E Mizzan, and RJ Chenier, “Postirradiation Behavior of UO<sub>2</sub> Fuel I: Elements at 220 to 250°C in Air”, *Nuclear Technology* **63** 254 (1983).
- g) Ito, K., K. Kamimura, and Y. Tsukuda, “Evaluation of Irradiation Effect on Spent Fuel Cladding Creep Properties”, *Proceedings of the 2004 International Meeting on LWR Fuel Performance*, Orlando, FL, September 19-22, 2004. p. 440. American Nuclear Society.
- h) Murty, KL, “The Internal Pressurization Creep of Zr Alloys for Spent-Fuel Dry Storage Feasibility”, *Journal of the Minerals, Metals and Materials Society (JOM)*, **52** 9(2000)34.

Table 2.2.2 Summary of Technical Issues – Fuel/Clad System

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Cladding Annealing	Confinement  Sub-criticality  Retrievalability & Transportability	<ol style="list-style-type: none"> <li>1. Some properties, such as strength, ductility, and fracture toughness, of the cladding may change because of irradiation (see Table 2.2.5). These changes are brought about because of radiation damage effects in the cladding. These effects can be annealed (removed or lessened) over time at temperature. The extent of annealing depends on the irradiation fluence, temperature, and time.<sup>a</sup></li> <li>2. It is typically believed that if annealing occurs, it will be in the early stages of storage when the temperatures are high and that annealing is not an issue for extended storage because the cladding temperature will be too low for additional annealing to occur.<sup>a</sup></li> <li>3. The temperature for 50% recovery of irradiation damage in Zry-2 was determined to be 380°C. Extrapolation of data indicates 40 to 50% recovery may be expected in six months at temperatures between 325 and 350°C. Below 200°C, there is virtually no annealing reported.<sup>b</sup></li> <li>4. FRAPCON currently has an</li> </ol>	<ol style="list-style-type: none"> <li>1. Most data on annealing is for Zry-2 and Zry-4 fuels.                         <ol style="list-style-type: none"> <li>a. Extrapolation of data up to 120 or more years is necessary.</li> <li>b. There is only limited data on new cladding types (e.g., ZIRLO™ or M5™).</li> <li>c. There is only limited data on effects in high burnup fuels.</li> <li>d. Temperatures may be higher for longer periods of time not only for higher burnup fuels or MOX, but for the newer, larger DCSS designs.</li> </ol> </li> <li>2. The link between unirradiated cladding behavior with that of fully- or partially-annealed cladding must be established.</li> </ol>	<p>Intact fuel cladding is important in containing and confining radio-nuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.</p> <p>The mechanical properties of Zircaloy (e.g., yield strength, elastic modulus, fracture toughness, ductility) are known to change because of irradiation effects (i.e., fast neutron flux). Much of the irradiation hardening is expected to anneal out early in dry storage because of the relatively high temperatures. Some additional irradiation effects will occur during storage, but these will be minimal compared to the in-reactor damage, with the exception that this damage will occur at relatively low</p>

		<p>annealing model for Zircaloy that is recrystallized (RXA) and cold-work stress relief annealed (CWSRA). The model assumes that the annealing of cold work is separate from annealing of irradiation damage. The lowest temperature data for RXA cladding is at 773 K for 8 mins<sup>c</sup> and for CWSRA cladding is at 755 K for 1 hr.<sup>d</sup></p>		<p>temperatures.</p> <p>Since much of the data on cladding behavior comes from unirradiated or “fresh” fuel that has not undergone annealing, it is important to determine the mechanical properties of the cladding as it will be in dry storage and the effects on future retrievability and transportation.</p>
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- a) *Technical Bases for Extended Dry Storage of Spent Nuclear Fuel*, EPRI, Palo Alto, CA: 2002. 1003416.
- b) Einziger, RE, MA McKinnon, and AJ Machiels, “Extending Dry Storage of Spent LWR Fuel For Up To 100 Years”, International Symposium on Storage of Spent Fuel from Power Reactors, Vienna, Austria, November 9-13, 1998.
- c) Torimaru, T., T. Yasuda, and M. Nakatsuka, “Changes in mechanical properties of irradiated Zircaloy-2 fuel cladding due to short term annealing,” *Journal of Nuclear Materials* **238**(1996) 169-174.
- d) Lowry, L. M., A. J. Markworth, J. S. Perrin, and M. P. Landow, *Evaluating Strength and Ductility of Irradiated Zircaloy: Task 5, Experimental Data: Final Report*, NUREG/CR-1729 (BMI-2066) Vol. 1, prepared by Battelle Columbus Laboratories for the U.S. Nuclear Regulatory Commission, Washington, D.C.

Table 2.2.3 Summary of Technical Issues – Fuel/Clad System

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
<p>Cladding Phase Transformation</p> <p>Cladding Eutectic Formation</p> <p>Cladding Melting</p> <p>Cladding Breach Due to Rapid Heating</p>	<p>Confinement</p> <p>Sub-criticality</p> <p>Retrievability &amp; Transportability</p>	<p>1. A change in phase (e.g., from <math>\alpha</math> to <math>\beta</math>) can affect the mechanical properties of the cladding. Such phase transitions are dependent on the type of alloy (e.g., Zry-2 or Zry-4) and the microstructure of the cladding. However, the <math>\alpha</math> to <math>\beta</math> transition occurs at a temperature (~810°C) much higher than for normal dry storage and even most postulated short-term off-normal situations. This mechanism is considered important during accident scenarios such as exposing the cask to fire.</p> <p>2. Zry-4 melting point is 1850°C and thus melting is applicable only under severe accident conditions.</p>	<p>1. These mechanisms are not relevant to normal operations, even for extended storage times, different materials or designs, or high burnup fuels. They are, however, important under accident (esp. fire) scenarios. Analysis of the potential effects of such an accident on future retrievability or transportability should be performed, but should not be different than for currently licensed facilities.</p>	<p>Intact fuel cladding is important in containing and confining radio-nuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.</p> <p>All of these mechanisms are applicable under accident scenarios and apply to all fuels and cladding types, but less data exists for newer cladding types and for impacts of high burnup (if any).</p>

Table 2.2.4 Summary of Technical Issues – Fuel/Clad System

<b>Mechanism</b>	<b>Functional Requirement</b>	<b>Results of Literature Search</b>	<b>Data Needs (gaps)</b>	<b>Importance to Licensing</b>
Cladding Null Ductility Transition	Confinement Sub-criticality Retrievalability & Transportability	1. Cladding ductility is significantly reduced due to irradiation effects (see Table 2.2.5) and hydrogen effects (see Table 2.2.9).	1. Refer to Tables 2.2.5 and 2.2.9 for data needs.	Intact fuel cladding is important in containing and confining radio-nuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievalability of the fuel for repackaging or other processes.

Table 2.2.5 Summary of Technical Issues – Fuel/Clad System

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Cladding Embrittlement (radiation effects)	Confinement Sub-criticality Retrievability & Transportability	<ol style="list-style-type: none"> <li>1. Radiation causes a decrease in ductility from ~10% to about 1-2%.<sup>a,b</sup></li> <li>2. Hydrogen pickup and the distribution of hydrogen in circumferential hydrides along the outer rim of the cladding can cause cladding embrittlement when the hydride rim gets thick enough at high burnup.</li> </ol>	<ol style="list-style-type: none"> <li>1. Newer cladding designs (e.g., ZIRLO™ and M5™) were designed to undergo less corrosion during reactor operation, and therefore have less hydrogen pickup. However, more data on the effects of higher burnups and radiation damage to these newer materials are necessary, especially as related to potential annealing (see Table 2.2.2).</li> </ol>	Intact fuel cladding is important in containing and confining radionuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.

- a) Garde, A. M. 1989. "Effects of Irradiation and Hydriding on the Mechanical Properties of Zircaloy-4 at High Fluence." In Zirconium in the Nuclear Industry: Eighth International Symposium, ASTM STP 1023, pp. 548-569, Eds. L.F.P. VanSwam and C. M. Eucken. American Society for Testing and Materials, Philadelphia, Pennsylvania.
- b) Lowry, L. M., A. J. Markworth, J. S. Perrin, and M. P. Landow, *Evaluating Strength and Ductility of Irradiated Zircaloy: Task 5, Experimental Data: Final Report*, NUREG/CR-1729 (BMI-2066) Vol. 1, prepared by Battelle Columbus Laboratories for the U.S. Nuclear Regulatory Commission, Washington, D.C.

Table 2.2.6 Summary of Technical Issues – Fuel/Clad System

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Cladding Oxidation	<p>Thermal performance</p> <p>Confinement</p> <p>Sub-criticality</p> <p>Retrievability &amp; Transportability</p>	<ol style="list-style-type: none"> <li>Oxidation of Zircaloy is a thermally induced process and requires an oxidant. During normal cask operations where a DCSS is filled with inert gas (e.g., He), oxidation cannot occur. Oxidation may occur due to reaction with oxygen if the DCSS was mistakenly backfilled with air, if a leak allows oxygen into the DCSS, or due to oxygen production from radiolysis of residual water (including waterlogged rods),.</li> <li>Rothman<sup>a</sup> examined multiple cladding oxidation conditions and predicted cladding thinning of 4 to 53 <math>\mu\text{m}</math> (up to 9% of cladding thickness) after 10,000 years at 180°C. Based on this, oxidation, whether from steam, water, or air is considered non-consequential for extended dry storage.</li> <li>However, tests conducted on spent fuel samples at ANL showed extensive fuel-side cladding oxidation (estimated at 18% of the through-wall thickness), enough to cause the rod segment to unzip, within 1.5 years in an air-limited, 100% relative humidity test at 175°C.<sup>b</sup></li> </ol>	<ol style="list-style-type: none"> <li>While the conditions of 100% relative humidity and a temperature of 175°C are not expected to occur during dry storage, and the corrosion observed was fuel-side (requiring a through-wall defect in the cladding), it still remains that the oxidation rate observed in the ANL tests is faster than any existing model predicts. It is necessary to determine the cause of this rapid oxidation and then determine if it could ever occur in dry storage.</li> </ol>	<p>Intact fuel cladding is important in containing and confining radionuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.</p> <p>Cladding oxidation reduces the thickness of the cladding, thereby reducing its strength. In addition, an increasing oxide layer could change the emissivity and thus increase the fuel/clad temperature.</p>



- a) Rothman, AJ, "Potential Corrosion and Degradation Mechanisms of Zircaloy™ Cladding on Spent Nuclear Fuel in a Tuff Repository," Report Attachment 10 to MRB-0418, JUCID-20172, Lawrence Livermore National Laboratory, Livermore, CA, 1984.
- b) Bechtel SAIC Co. LLC, *CSNF Waste Form Degradation: Summary Abstraction*. ANL-EBS-MD-000015 REV 02. Las Vegas, NV. 2004.

Table 2.2.7 Summary of Technical Issues – Fuel/Clad System

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Waterlogged Rods	<p>Thermal performance</p> <p>Confinement</p> <p>Sub-criticality</p> <p>Retrievability &amp; Transportability</p>	<ol style="list-style-type: none"> <li data-bbox="661 344 1121 711">1. Kohli et al.<sup>a</sup> showed that the bulk of water in a water-logged rod was released not just during vacuum drying, but vacuum drying with the temperature at 100°C. Because of decay heat, the fuel in most DCSS drying can approach the NRC limit of 400°C.<sup>b</sup> It would be expected that most water would be removed during drying.</li> <li data-bbox="661 711 1121 1214">2. However, the reactor-induced breaches were relatively large and could be seen with visual examination and in addition “Holes (~3.0 mm in diameter) were drilled in the plenum region of each rod to release any water that may have been trapped in the plenum.” Multiple, large defects allow water to be released and have less chance of icing up (sealing) during vacuum drying. Even then, the rods continued to outgas for about 1000 hours at 325°C.</li> <li data-bbox="661 1214 1121 1398">3. Water-logged rods would be capable of producing the high relative humidity at temperatures higher than 175°C and could result in clad unzipping as was observed in the ANL tests (See</li> </ol>	<ol style="list-style-type: none"> <li data-bbox="1138 344 1579 841">1. More realistic calculations of how much water may be in casks after vacuum drying are needed. Most utilities are not putting known failed fuel into dry storage at present. If they did, even under current canning requirements- with a screen at either end of the can-, it is still possible that more water than has been calculated (7.7 g in a waste package<sup>d</sup> or 65 g in a cask<sup>e</sup>) could remain in a cask after vacuum drying, even if the proper procedures are followed.</li> </ol>	<p>Intact fuel cladding is important in containing and confining radionuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.</p> <p>Rods that fail during in-core operation or in the spent fuel pool could have water fill the void space in the rod. If properly dried, there should be minimal water in a cask. However, this drying is a function of time, temperature, and most importantly, the size of the defect through which water must be released.</p> <p>Residual water can lead to corrosion and further rod degradation and could pose a potential criticality risk.</p>

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		Table 2.2.6). <sup>c</sup>		
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- a) Kohli, R., D Stahl, V Paupathi, AB Johnson, Jr., and ER Gilbert, “The Behavior of Breached Boiling Water Reactor Fuel Rods On Long-Term Exposure to Air and Argon at 598 K,” *Nuclear Technology* **69**(1985)186.
- b) USNRC, 2003. Interim Staff Guidance, 11, ISG-11 Rev-3, U.S. Nuclear Regulatory Commission, Washington DC, November 2003.
- c) Bechtel SAIC Co. LLC, *CSNF Waste Form Degradation: Summary Abstraction*. ANL-EBS-MD-000015 REV 02. Las Vegas, NV. 2004.
- d) Bechtel SAIC Co. LLC, *Clad Degradation – FEPs Screening Arguments*. ANL-WIS-MD-000008 REV 02. Las Vegas, NV. 2004.
- e) *Data Needs for Long-Term Dry Storage of LWR Fuel*, EPRI, Palo Alto, CA: 1998. 108757.

Table 2.2.8 Summary of Technical Issues – Fuel/Clad System

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Radiolysis	Confinement Sub-criticality Retrievability & Transportability	<ol style="list-style-type: none"> <li>1. Radiolysis of water can result in production of oxygen or highly oxidizing species (e.g., OH or H<sub>2</sub>O<sub>2</sub>) that can then corrode fuel, cladding, or cask internals. However, water is limited to what may be left in a cask after vacuum drying (as free-, chemisorbed- or physisorbed-water) or in water-logged rods.</li> <li>2. Radiolysis of nitrogen (either from air ingress or mistaken backfill) can result in the production of very aggressive oxidants such as nitric acid, even though concentrations may be low.<sup>a, b</sup></li> <li>3. Water reactions could produce hydrogen that could react with the cladding (see Table 2.2.9 for hydrogen effects).</li> </ol>	<ol style="list-style-type: none"> <li>1. More realistic calculations of how much water may be in casks after vacuum drying are needed. Most utilities are not putting known failed fuel into dry storage at present. If they did, even under current canning requirements- with a screen at either end of the can-, it is still possible that more water than has been calculated (7.7 g in a waste package<sup>c</sup> or 65 g in a cask<sup>d</sup>) could remain in a cask after vacuum drying, even if the proper procedures are followed.</li> </ol>	<p>Intact fuel cladding is important in containing and confining radionuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.</p> <p>Rods that fail during in-core operation or in the spent fuel pool could have water fill the void space in the rod. If properly dried, there should be minimal water in a cask. However, this drying is a function of time, temperature, and most importantly, the size of the defect through which water must be released.</p>

a) Sunder, S. and NH Miller, "Oxidation of CANDU uranium oxide fuel by air in gamma radiation at 150°C", *Journal of Nuclear Materials* **231**(1996)121-131.

b) Delegard, CH, MR Elmore, KJ Geelhood, MA Lilga, WG Luscher, GT MacLean, JK Magnuson, RT Pagh, SG Pitman, and RS Wittman, *Final Report-Evaluation of Chemical Effects Phenomena in Post-LOCA Coolant*, NUREG/CR-6988, US Nuclear Regulatory Commission, March 2009.

c) Bechtel SAIC Co. LLC, *Clad Degradation – FEPs Screening Arguments*. ANL-WIS-MD-000008 REV 02. Las Vegas, NV. 2004.

d) *Data Needs for Long-Term Dry Storage of LWR Fuel*, EPRI, Palo Alto, CA: 1998. 108757.

**Table 2.2.9 Summary of Technical Issues – Fuel/Clad System**

Mechanism	Functional Requirement	Results of Literature Search	Data Needs (gaps)	Importance to Licensing
Delayed Hydride Cracking (DHC)	Confinement  Sub-criticality  Retrievability & Transportability	<ol style="list-style-type: none"> <li>1. DHC is a time-dependent mechanism traditionally thought of as diffusion of hydrogen to a flaw tip, followed by nucleation, growth, and fracture of the hydride at the flaw tip. The process continues to repeat as long as a sufficient stress to promote the hydrogen diffusion occurs.<sup>a,b</sup></li> <li>2. DHC has traditionally been ruled out as a possible mechanism for cladding degradation during extended storage because as the temperatures decrease, the stress decreases and is insufficient to sponsor crack propagation.<sup>c,d,e</sup></li> <li>3. However, Rothman noted that additional data is necessary for larger crack depths (~50% of wall thickness).<sup>e</sup></li> <li>4. The earlier models did not account for the hysteresis in the solvus, which has important effects on the temperature dependence of the DHC velocity.<sup>f</sup> This hysteresis shows that the hydrogen concentration can be substantially higher on a cooling solvus line than for a heating solvus line.<sup>g</sup></li> <li>5. DHC is a known failure</li> </ol>	<ol style="list-style-type: none"> <li>1. While there is much disagreement<sup>b,m</sup> with Kim’s model<sup>l</sup>, if Kim’s hypotheses are correct, then spent fuel will be more likely to fail by DHC upon cooling below 180°C if there are stress raisers inside the rod such as the end cap weld region or incipient cracks due to an interaction of fuel and cladding during reactor operation. This may explain why DHC failure has not been observed in the 15-year demonstration project where the fuel temperature in an open cask (so lower than in the sealed cask) was measured as 154°C.</li> <li>2. Higher burnup fuels with higher hydrogen loading and more extensive cladding oxidation may be more susceptible to DHC.</li> </ol>	Intact fuel cladding is important in containing and confining radionuclides, in keeping the fuel in a sub-critical configuration, and for allowing retrievability of the fuel for repackaging or other processes.

		<p>mechanism in pressure tubes of Zr-2.5% Nb alloy as used in CANDU and RBMK reactors<sup>h</sup> as well as in Zry-2 tubing used in the Hanford N-reactor.<sup>i</sup></p> <p>6. Simpson and Ells reported DHC failure of unirradiated Zr-2.5% Nb specimens at room temperature over periods of four to five weeks to up to 24 months.<sup>j</sup></p> <p>7. Huang and Mills reported that cracking only occurred above a critical temperature (180°C) when specimens were subjected to an overtemperature cycle. Below that temperature, DHC occurred regardless of whether specimens were heated or cooled to the test temperature.<sup>i</sup></p> <p>8. Chao et al.<sup>k</sup> report that the location of the crack is important in determining whether or not the cladding will break, with the greatest chance being when the crack is located on the outer side of the cladding.</p> <p>9. Kim has proposed a new model for DHC. In this model, creep deformation, prior creep strain, higher burnup, the solvus hysteresis, and the <math>\gamma</math> to <math>\delta</math> hydride phase transition all play important roles in DHC.<sup>l</sup></p>		
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a) Puls, Manfred P., "Review of the thermodynamic basis for models of delayed hydride cracking rate in zirconium alloys", *Journal of Nuclear Materials* **393**(2009) 350-367.

- b) McRae, GA, CE Coleman, and BW Leitch, “The first step for delayed hydride cracking in zirconium alloys”, *Journal of Nuclear Materials* **396**(2010) 130-143.
- c) Bechtel SAIC Co. LLC, *Clad Degradation – FEPS Screening Arguments*. ANL-WIS-MD-000008 REV 02. Las Vegas, NV. 2004.

### **3. CONCLUSIONS**

Several failure mechanisms were identified in the literature searches associated with fuel clad interactions. These mechanisms include cladding creep, annealing, phase transformation, melting, eutectic formation, embrittlement, ductility transition, oxidation, waterlogged fuel rods, radiolysis, and delayed hydride cracking. Data needs have been identified from this literature search. However, not all of these data needs and mechanisms will have the same priority in a comprehensive R&D program. Future efforts will focus on identifying additional mechanisms, especially for systems other than the fuel/clad interactions, and prioritizing the data needs that the R&D program must address.