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September 9, 2004  
Contract No. NRC-02-02-012  
Account No. 20.06002.01.081

U.S. Nuclear Regulatory Commission  
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Subject: Submittal of Intermediate Milestone—A Review Report on High Burnup Spent Nuclear Fuel—Disposal Issues, IM 06002.01.081.410

Reference: Letter dated July 16, 2004 from D. Galvin to V. Jain—Acceptance of Container Life and Source Term Intermediate Milestone 06002.01.081.410: A Review Report on High Burnup Spent Nuclear Fuel—Disposal Issues

Dear Mr. Galvin:

Enclosed are the subject intermediate milestone and our responses to the NRC staff comments on the draft report. The report has been revised to incorporate NRC comments. This report provides a review of disposal issues related to high burnup spent nuclear fuel. Based on 1998 projections, high burnup spent nuclear fuels will constitute more than 70 percent of the total inventory of the spent nuclear fuel generated between 1998 and 2015. Spent nuclear fuel exceeding 45 GWd/MTU is classified as high burnup spent nuclear fuel. Projections show that the potential Yucca Mountain repository could receive 30 percent of the spent nuclear fuel that could be classified as high burnup. Currently burnup in nuclear reactors in the United States is limited to an average burnup of 62-GWd/MTU for the fuel rod with a maximum burnup. This report summarizes pertinent information and presents current understanding of the issues related to the disposal of the high burnup spent nuclear fuel. Topics discussed in this report include radionuclide inventory, instant release fraction, spent nuclear fuel dissolution, and cladding.

If you have any questions regarding this report, please feel free to contact me (210) 522-5439.

Sincerely yours,

Vijay Jain  
Element Manager  
Corrosion Science & Process Engineering

VJ:jg

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Response to NRC Staff Comments on "A Review Report on High Burnup Spent Nuclear Fuel—Disposal Issues": CNWRA Intermediate Milestone 06002.01.081.410

#	NRC Comment	CNWRA Response
1	At some place, possibility of drift degradation needs to be addressed. This will affect (i) cladding behavior in hydride reorientation and creep, (ii) dry oxidation of the matrix, and (iii) diffusion of dose contributing radionuclides (e.g., to grain boundaries), all at expected higher temperatures.	A 3 <sup>rd</sup> paragraph was added at the beginning of the Introduction referring to the possibility of drift degradation and the associated temperature increase. The paragraph contains all the points mentioned in the comment.
2	In the cladding performance, many concerns (e.g., creep, hydride embrittlement, or localized corrosion) are raised. At least it should state that quantitative analyses are needed to assess the risk significance of these concerns with respect to performance and inventory. For example, in assessing the significance of higher stress expected, some available information on the distribution of incipient crack sizes can be considered.	A paragraph was added at the end of Chapter 7, indicating the need of quantitative analyses of the various cladding failure processes to assess the risk significance to waste disposal of each of these processes. It is also indicated that performance confirmation activities will allow an evaluation of the relative significance of the various failure processes.
3	The bases of the justification in p. 4-8 are not well supported. Give references and examples in 4 <sup>th</sup> and 7 <sup>th</sup> bullets.	Text and reference was added for 4 <sup>th</sup> and 7 <sup>th</sup> bullet.
4	In presenting dissolution rates in Chapter 5, some details used in determining the surface area for each dissolution rate need to be included. Also, the solution chemistry needs to be stated in details (e.g., figure 5-1, 5-2, table 5-1).	Surface area and solution chemistry details were added as needed.

5	In cladding performance, many potential failure mechanisms are presented. Examples are crud-induced localized corrosion; oxides formed in reactors regardless of burn up. It may need to state that risk insight will be studied for these concerns. Further examples include locally enhanced corrosion, radiolysis-induced hydrogen absorption (p. 6-1) and localized corrosion (p.6-14), nodular corrosion (old design will be disposed too), hydrogen concentration on the outer cladding surface, iron oxide effects on localized corrosion (p. 6-16), hydride embrittlement (p. 6-23), hydride content and distribution on the creep behavior (p. 6-26), and full range of environmental conditions (4 <sup>th</sup> line from the end in p. 6-26).	See response to comment # 2.
6	The inventory of newly developed cladding is of very minor amount. Therefore, it should be cautious to present any concern associated with the new cladding (the last sentence of p.v).	Sentence was modified to include all cladding materials subject to high burnups.
7	In the 3 <sup>rd</sup> paragraph of p. xviii, the criterion of 250 C is valid for the SNF matrix to avoid higher oxidation. However, this criterion for cladding is not performance-based.	Text was modified, eliminating the mention to cladding, to indicate that the temperature value is chosen only for the irradiated fuel pellets to limit oxidation to a higher oxidation state.
8	In the 2 <sup>nd</sup> paragraph of p. 1-1, indicate what fraction of cladding protection was used in getting the dose.	It is now indicated that cladding protection allowed radionuclide release from only one percent of the spent fuel.
9	In the 2 <sup>nd</sup> paragraph of p. 3-1, present the actual temperatures of the centerline.	Information on actual centerline temperature added.
10	In the 2 <sup>nd</sup> paragraph of p. 3-7, "cladding performance" is not connected to the following discussion on the matrix behavior.	Information on cladding was removed from the text.
11	In the last sentence of "4. INSTANT RELEASE FRACTION," add "in the total system performance assessment." Specify all radionuclides with atomic numbers.	Information on radionuclides was added.
12	In the 5 <sup>th</sup> sentence, change $10^{-3.5}$ , to a rational number $3.2 \times 10^{-4}$ .	Text changed as suggested.

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13	In p. 4-12, Cl-36 volatility is compared with that of I-129. It doesn't seem to be right because Cl-36 is gas and I-129 is solid.	Boiling point of iodine is 184 C. Under reactor operating conditions both chlorine and iodine are expected in gaseous phase. Text revised in the report.
14	In the 9 <sup>th</sup> line of p. 5-5, specify how the dissolution rate was determined after 4.5 years (e.g., Sr release or U release).	Release was measured by Sr-90. Information added in the report.
15	In the 1 <sup>st</sup> sentence of p. 5.2, specify the solution chemistry.	Information already exists on Page 5-1. No change made to the text.
16	In Eq. (5-3), quote TPA4 and MRS presentation by T. Ahn and S. Mohanty in ADAMS. Delete two "extremely" in the 1 <sup>st</sup> sentence after Eq. (5-3), because it is consistent with literature such as Oversby. Also it doesn't seem to be right that lower end of the pre-exponential term is sampled more frequently (see S. Mohanty). Rather higher end contributions are significant. That's why model 1 and model 2 of TPA show only a factor of 2.5 difference in dose.	Text revised as suggested.
17	In p. 6-1, add (Total System Performance) Assessment.	Text added as suggested.
18	What is Pilling-Bedworth ratio in p. 6-8?	The Pilling-Bedworth (PB) ratio is the ratio between the molar volume of the oxide in the film and that of the metal underneath. The PB criterion establishes that oxides with a ratio higher than 1 are protective. See comment # 49.
19	In the 2 <sup>nd</sup> line of p. 6-2, several forms [such as?]	It was clarified that internal cladding oxidation and pressurization can result from fission gases and helium.
20	The 6 <sup>th</sup> line from the end of 6.4.3, what is the basis for sufficient hoop stress?	Text revised to indicate that hoop stresses close to 150 MPa can be reached at high burnups as a result of increasing pressure caused by fission gases and helium.

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21	Do we know burnup of the creep results in p. 6-18? Likewise, please state the burnup if known in delayed hydride embrittlement in p.6-22.	A mean value of 44 GWd/MTU is included for the data used by Siegman and Macheret (2002). This value is considered to be representative for the reported creep results. For the case of hydride reorientation, the burnup of 31 GWd/MTU, quoted by Eizinger and Kohli (1983) in their paper, was included as reference.
22	The report attempts to quantify the fraction of high burnup fuel, "Assuming 63,000 MTU capacity for commercial spent nuclear fuel of a total capacity of 70,000 MTU at the potential Yucca Mountain repository, 30 percent of the 63,000 MTU commercial spent nuclear fuel could be classified as high burnup fuel". The final repository will need to contain significantly more than 63,000 MTU of commercial spent nuclear fuel. With the growing trend to increase assembly average discharge burnup, the percentage of high burnup fuel will increase.	Review is limited to the current YM repository capacity. Text added to clarify the objective of the report.

23	The report assumes current operating strategies and an upper peak rod burnup of 62 Mwd/kgU.	These issues at this time are outside the scope of YM Repository work.
	a The nuclear industry seeks to increase peak rod burnup from 62 to 75 Mwd/kgU. An increase in burnup will impact pellet morphology, fission gas distribution, and clad performance and must be accounted for in any repository study.	
	b Trends in the nuclear industry, including longer cycle lengths, higher assembly average discharge burnups, core power uprates, new burnable absorbers, and changes in plant chemistry, may negate some of the assumptions in this study. These items have a detrimental effect on fission gas release and clad performance. All of these items need to be addressed in any repository study.	
	i For example. Fission gas release and isotope distribution is dependent on power history more than burnup. With lower U235 enriched and/or annular pellet axial blankets, power uprates, and pressure on utilities to lower fuel costs, local power density has been increasing. This trend will promote higher fission gas release.	
24	The report mentions irradiation hardening and its beneficial impact on creep resistance. It is important that the potential for partial annealing during vacuum drying and initial dry storage be further addressed. This phenomena may impact plant procedures for processing spent fuel into dry cask storage.	Indeed, this is an area that requires additional research. In the report it is noted that results from Tsai (2003) indicated that a substantial fraction of irradiation hardening can be annealed out above 420 °C in a matter of hours. A sentence is added to clarify the point on the basis of a recent paper by Tsai and Billone (2004).

25	RES is sponsoring ongoing research at ANL on high burnup creep performance. While the report appears to capture some of the early results from the Surry fuel rods (at ~ 36 MWd/kgU), it does not capture the Robinson fuel rods (at ~ 67 Mwd/kgU). Note that some of the ANL tests were run out to cladding breach. In addition to the long term creep behavior, these failed rods may provide insight into repository criteria.	Information from this ongoing research is now added on the basis of the paper by Tsai and Billone (2004) mentioned in comment #24.
26	RES is sponsoring ongoing research at SNL on high burnup fission gas release and isotopic distribution. This research should be reflected in this report.	No public documents were available for review on this program. Therefore, no changes were made to the report.
27	The report identifies new corrosion resistant clad alloys (e.g. ZIRLO and M5) as decreasing the likelihood of hydride embrittlement due to reorientation. It should be realized that even these clad alloys will absorb 100 ppm of hydrogen at their expected burnups and fuel duty. Since the solubility limit is ~100 ppm (at 300 degC) for most zirconium alloys, these new alloys will exhibit the same degree of precipitation upon cooling.	A sentence was added in order to qualify the statement in the report.
28	The report claims that circumferentially oriented hydrides have little impact on cladding performance. While newer, low corrosion alloys will absorb less hydrogen, current fuels may reach above 800 ppm. Due to the migration of hydrides, local concentrations near the outer diameter may exceed 2000 ppm. I am not sure that such a blanket statement can be made.	A sentence was added to in order to qualify the statement in the report.

29	<p>Current high duty fuel, especially when fuel pellets are coated with ZrB<sub>2</sub> burnable absorber, are expected to reach rod internal pressures above 3,000 psia at operating temperatures (clad temperature ~ 340 degC).</p> <p>a     The report states that SCC is not a concern because "both the critical stress and the stress intensity levels required for iodine stress corrosion cracking, as well as the limiting temperatures of 280-290 degC, are not reached".</p> <p>i     Does this conclusion consider ISG-11 limit on clad temperatures of 400 degC (which would exceed 280-290 degC)?</p> <p>ii    Does this conclusion consider rod internal pressure above 3000 psia at normal operating temperature (340 degC) and its corresponding hoop stress during vacuum drying at 400 degC?</p> <p>b     For long-term storage, rod internal pressure plays a major role in SCC, long-term creep, and hydride reorientation. An evaluation needs to be performed to determine whether fuel design limits related to rod internal pressure will be governed by in-reactor steady state (no clad lift-off), in-reactor transient performance (clad balloon and rupture), initial dry cask storage (drying and storage), or long-term storage concerns.</p>	<p>The conclusion regarding SCC is valid under disposal conditions once the cladding temperature has decreased below the temperature quoted in the report. It does not include considerations for temporary dry storage. The authors fully agree that a more detailed evaluation needs to be performed regarding rod internal pressure and its effect on hoop stresses at temperatures typical of the initial stages of dry storage. A sentence was added to that effect in the report.</p>
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30	The report (Page 6-10) includes a discussion on helium partial pressure within the fuel rod. In addition to fill gas and alpha emission, the report needs to consider ZrB2 burnable absorber (B10 coating on the fuel pellets) which is a significant source of helium ( $B10 + n = He + Li$ ).	A paragraph was added according to the comment, quoting the report of Lanning and Beyer (2004)..
31	The report identifies hydride reorientation as a limiting concern with dry cask storage. Due to current uncertainties concerning reorientation, maybe RES's long term creep program at ANL should be expanded to include a study of hydride reorientation (variables including burnup, composition, texture, hydrides and oxides, time at temperature, and hoop stress). The results of such a comprehensive study could then be used to better define regulatory limits on hoop stress and clad temperatures.	This aspect is out of the scope of the report. We are pleased to know that the interest to conduct research on hydride reorientation has been stimulated by our report.
32	The report did not consider MOX fuel which will be coming into use and is planned per DOE to be stored in the repository.	A brief discussion was added in Chapter 3. It should be noted that there is no data on instant release fraction for MOX fuel and that the total amount of MOX fuel anticipated to be disposed at the potential Yucca Mountain repository is negligible compared to $UO_2$ fuel.
33	The report did not consider the effects of the different burnable poisons - ZrB2, Gd, and Erbia.	The study of burnable poisons is outside the scope of this report.
34	The report did not consider the long range goals of the industry include going to burnups on the level of 100 GWD/MT peak average rod and Westinghouse has proposed going even higher with the new IRIS design they are still working on.	These issues at this time are outside the scope of potential Yucca Mountain repository work.
35	On page 7-3 of conclusions, a sentence reads "Creep failure, on the other hand, seems to be less possible for high burnup fuel than for medium burnup fuel." I was unable to find supporting evidence for this statement in section 6.4.4.	Sentence was deleted. It came from a general reference but the recent results of Tsai and Billone (2004) do not support such statement. It is indicated that creep failure for high burnup fuel requires further evaluation through experimental work.

36	Medium burnup fuel should be defined in the executive summary.	Medium or moderate burnup fuel are the fuels with burnup below 45 GWd/MTU. Text added in the executive summary.
37	Page 2-1 The input decks for ORIGEN-ARP used to calculate the inventories should be placed in an appendix.	Input summary for 65 GWd/MTU case added as Appendix A.
38	Page 2-1 TPA 5.0 has not been released to the public and is still being worked on. This should be clarified in a foot-note. It may also be useful to compare the information with TPA 4.1j, the last public version of the code.	Footnote added to clarify on code used for this analysis. TPA 4.1j used the same inventory in the nuclides.dat file as the base case TPA 5.0 nuclides.dat file.
39	Page 2-5 to 2-6 Figure 2-3 and Section 2-3 do not discuss the extent that high burnup fuel would lengthen the thermal pulse or the extent of fuel experiencing the thermal pulse. Since the models in TPA 5.0 are being actively worked in this area, TPA 5.0 may not give an accurate assessment of risk significance and the conclusion in the summary may be inappropriate. It would be useful to discuss the effect of high burnup fuel on the thermal pulse.	The TPA 5.0 models are being actively worked in the areas of drift degradation effects on waste package life, temperature, amount of water contacting the waste form, and changes in water chemistry. This includes analysis in the Risk Analysis for Risk Insights Report investigating the effects of natural backfill on waste package temperature. The analysis and conclusion in the summary of this report was intended to show the small change that high burnup fuel would have on the thermal profile that is used in the burnup.dat file of TPA 5.0 code, and that it is a representative profile which can be used for these future analyses. This is particularly true in light of loading of waste packages and other controls that will have to be used to maintain limits on the linear thermal loading in the potential repository. The length of the thermal pulse is dependent on radionuclide decay rates, which as shown in Figure 2-3, are not dependent on burnup.
40	Chapter 4 The CANDU fuel has burnups ranging from 5.6 to 15.5 GWd/MTU and yet this data is being used to support conclusions about burnups 4 times higher. While the CANDU data provides information concerning the effects of higher power densities at low burnups, the applicability of this data to high burnup fuel is not clearly shown.	Discussion added to the text justifying the use of CANDU fuel data.

41	Tables 4-3 and 4-4 provides estimates of fission product release for high burnups based upon linear extrapolation from lower burnups. However, Figures 3.3, 3.5, and 3.6 indicate that the effects of burnup above 60 GWd/MTU are not linear. Better justification is needed for the linear models or non-linear models should be considered.	Table limited to 75 GwdMTU. Fission gas release can be linearly estimated upto 75 GWd/MTU. Figure added from the paper of Koo et al. (2001). Beyond 75 GWd/MTU non-linear models may be required.
42	Section 5.1 Remove DOE from the last line of the first paragraph. Comment 38 also applies.	Accepted. Text corrected.
43	In table 6-1, M5 has no tin so put in a dash. Zirlo has oxygen so a value is needed.	The mistake in the table regarding tin was corrected. No oxygen content was provided in the referenced publication.
44	Section 8 References: Provide the ADAMs number for the Brach reference.	Accepted. ADAMS number added.
45	Section 8 References: For the Bremier reference, the organization is the Nuclear Energy Agency, not Institute.	Accepted. Text changed to Agency.
46	Section 6.3.2 Proprietary sources have identified lower value for the Pilling-Bedworth ratio, which would result in a higher hydrogen pickup fraction.  Section 6.3.2 calculates a hydrogen burnup fraction for PWR cladding. This section needs equivalent hydrogen pickup information for BWR cladding.	A sentence is added to clarify this point. A theoretical value of 1.56 can be calculated if no correction is introduced to account for lower oxide density or compositional changes. The hydrogen pickup for BWR cladding was not discussed because performance assessments are usually confined to PWR fuel as the predominate fuel in the repository.

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47	<p>The information contained in ISG-11, rev3. UNITED STATES NUCLEAR REGULATORY COMMISSION, Interim Staff Guidance No. 11, Revision 3, "Cladding Considerations for the Transportation and Storage of Spent Fuel" (2003), <a href="http://www.nrc.gov/reading-rm/doc-collections/isg/ISG-11R3.pdf">http://www.nrc.gov/reading-rm/doc-collections/isg/ISG-11R3.pdf</a>. and GRUSS, K., BROWN, C., "U.S. Nuclear Regulatory Commission Acceptance Criteria and Cladding Considerations for the Dry Storage of Spent Fuel," (TOPFUEL Conf., Wurtzburg, Germany, 2003) can serve as background for the report. The ISG and the paper provides background information as to why the NRC attempts to minimize the formation of radial hydrides in the cladding and how cladding stress are kept below the 90 MPa.</p>	<p>This information has been taken into account in the report. Reference is made to the paper by Gruss et al. (2003) that was inadvertently omitted.</p>
48	<p>SFPO recently issued a PNNL white paper on cladding stresses in PWR fuel, ML040290474. This paper analyzes the hoop stress in a Westinghouse design burnable poisons with ZrB2 used at Diablo Canyon. The burnable poisons with ZrB2 were found to have a higher hoop stress than the standard Westinghouse designs. A question was raised by HLW that release of helium in ZrB2 should be addressed. The author of the report can refer to the above white paper for a technical basis for a response to the question and what effect helium has on the pressure in the cladding.</p>	<p>The comment was considered. See response to comment #30.</p>

49	A question was raised concerning the following: What is Pilling-Bedworth ratio in p. 6-8? It should be noted that it is the theoretical oxide-to-metal ration (O/M) is referred to as Pilling-Bedworth ratio. When water reacts with zirconium-based alloys, the surface of the metal is converted to an oxide. Due to difference in the densities of the oxide and the base metal, there is a volumetric change from the metal consumed to the oxide generated. This volumetric difference results in a thicker oxide than the metal that was consumed. The value cited of the ratio cited in the report is not the same as the value cited in a <b>Westinghouse Proprietary report</b> . The value in WCAP-15168, "Dry Storage of High Burnup Spent Nuclear Fuel," 1.56. This value is proprietary. Note: In response to the comment, a NRC staff member believes this value is available in a nonproprietary document but could not immediately identify the document. If the CNWRA identifies this nonproprietary source, the hydrogen pickup fraction should be recalculated.	This was clarified in detail in a paragraph added to the report.
50	Page 6-23, paragraph two is not correct. Neither Argonne East nor SFPO believe the failure of the rod was due to hydride reorientation, but perhaps the tests set-up. See vol 29, July 5, 2004 issue of Nuclear Fuel.	The text in the paragraph was corrected according to new information available, as provided by SFPO staff and the presentation by Tsai and Billone (2004) was quoted.
51	For background, the author should review SFPO's User Need document (ML040650621) for work that is being conducted on the Robinson Fuel at ANL. Test include post-test characterization of the creep samples, effects of hydride reorientation, impact testing, and fracture toughness testing.	Information is included on the basis of a paper by Tsai and Billone (2004).

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52	Note: On page 6-18, paragraph 3, it should be noted that NRC does not endorse the Methodology used to derive the maximum cladding temperature limit by Livermoore. NUREG-1536 endorsement of the diffusion-controlled cavity growth (DCCG) method to calculate the maximum cladding temperature limit during dry storage was overly restrictive and relatively inflexible. Literature did not support the use of this model for zirconium-based materials. This is documented in ISG-11, rev0.	A sentence was added in the following paragraph in page 6-19 to clarify this point according to the comment.
53	The report should consider some of the corrosion data contained in Westinghouse and EPRI reports (Hot cell data from Calvert Cliffs, Fort Calhoun, etc). It should be noted that most of the reports are proprietary, but can be found in ADAMs or in the NRC technical library. These reports have a substantial amount of data and information that can help substantiate this report.	To review of such information is out of the scope of the present report.
54	The author should consider using the data in the following three journals: Vizcaino, P., A.D. Banchik and J.P. Abriata. 2002. "Solubility of Hydrogen in Zircaloy-4: Irradiation-Induced Increase and Thermal Recovery," Journal of Nuclear Materials, Vol. 304 (2002), pp. 96-106 and Pyecha, T.D., et al. 1985. "Waterside Corrosion of PWR Fuel Rods Through Burnups of 50,000 MWd/MTU", Published in ANS Topical Meeting on Light Water Reactor Fuel Performance, Orlando, Florida April 21-24, 1985 and, Kilp, G.R., et al. 1991. "Corrosion Experience with Zircaloy and Zirlo In Operating PWR's," Published in International Topical Meeting on LWR Fuel Performance, Avignon, France April 21-24, 1991.	In the report more recent information than that provided in Pyecha et al. (1985) and Kilp et al. (1991) is used. Nevertheless the information provided in these papers was evaluated and it was concluded that does not change the main conclusions of the report regarding oxide thickness and hydrogen content. The information contained in Vizcaino et al, (2002) deserves an detailed analysis that cannot be accomplished in the context of the present report.