Spent Fuel Project Office
Draft Interim Staff Guidance-22
Potential Rod Splitting Due to Exposure to an Oxidizing Atmosphere During Short-term Cask Loading Operations in LWR or Other Uranium Oxide Based Fuel

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Issue:

Under the current guidance in ISG-1, Revision 1, "Damaged Fuel," the definition of intact fuel includes fuel rods containing no cladding defects greater than pinhole leaks or hairline cracks. During the cask water removal process (also known as blow-down), parts of, or all of, the fuel rods will be exposed to a gaseous atmosphere. If the gaseous atmosphere is oxidizing, oxidation of fuel pellets or fuel fragments can occur if a cladding breach exists (such as a pinhole). Oxidation may occur rapidly and cause significant swelling of fuel pellets and fragments, which could result in gross fuel cladding breaches.

Regulatory Basis:

The regulations for storage in 10 CFR Part 72, and those for transportation in 10 CFR Part 71, have the following common safety objectives: (1) ensure that the radiation doses do not exceed the limits prescribed in the regulations, (2) maintain subcriticality, and (3) ensure there is adequate confinement or containment of the spent fuel. Additionally, 10 CFR Part 72 regulations require that the spent fuel be readily retrievable from the storage systems. In particular, the following regulations are applicable to this ISG:

10 CFR 72.120(d) states in part – "no significant chemical, galvanic or other reactions between or among the storage system components, spent fuel...The behavior of materials under irradiation and thermal conditions must be taken into account."

10 CFR 72.122(h)(1) states in part – "The spent fuel cladding must be protected during storage against degradation that leads to gross ruptures in the fuel or the fuel must be otherwise confined such that the degradation of the fuel during storage will not pose operational safety problems with respect to its removal from storage."

10 CFR 72.122(l) states in part – "Retrievability…allow ready retrieval of spent nuclear fuel... for further processing or disposal."

10 CFR 72.236(m) states in part – "To the extent practicable...consideration should be given to compatibility with removal of the stored spent fuel from reactor sites, ...transportation, and ultimate disposal by the DOE."

The requirements of 10 CFR 72.122 (h)(1) ensure safe fuel storage and handling and minimize post-operational safety problems with respect to the removal of the fuel from storage. As required by this regulation, the spent fuel cladding must be protected during storage against degradation that leads to gross rupture of the fuel and must be otherwise confined such that degradation of the fuel during storage will not pose operational problems with respect to its removal from storage. Additionally, 10 CFR 72.122(l) and 72.236(m) require that the storage system be designed to allow ready retrieval of the spent fuel from the storage system for further transportation, processing or disposal.

10 CFR 71.33(b) states that applications for NRC approval must include a description of the proposed package in sufficient detail to identify the package accurately and provide a sufficient basis for evaluation of the package; including, with respect to the contents of the package -- the chemical and physical form of the contents. Thus, any significant oxidation of the UO_2 fuel pellets to U_3O_8 would change the chemical form from that which was approved in the certificate of compliance.

Applicability:

This guidance applies to reviews of spent fuel dry cask storage systems and spent fuel transportation packages conducted in accordance with NUREG-1536, "Standard Review Plan for Dry Cask Storage Systems" (January 1997); NUREG-1567, "Standard Review Plan for Spent Fuel Dry Storage Facilities" (March 2000); and NUREG-1617, "Standard Review Plan for Transportation Packages for Spent Nuclear Fuel" (March 2000).

Technical Review Guidance:

This ISG is only applicable to applications for storage or transportation of irradiated LWR fuel.

Once the fuel rods are placed inside of the storage cask and water is removed to a level that exposes any part of the rods to a gaseous atmosphere, reasonable assurance that the spent fuel cladding will be protected against splitting due to fuel oxidation that might occur is encouraged. If oxidation occurred, it may lead to loss of retrievability, or to a configuration not adequately analyzed for radiation dose rates or criticality. Further, the release of fuel fines or grain-sized powder into the inner cask environment from ruptured fuel may be a condition outside the licensing basis for the cask system. Three possible approaches to address the potential for and consequences of fuel oxidation are:

1) Maintain the fuel rods in an appropriate environment such as Ar, N_2, or He to prevent oxidation.

2) Assure that there are not any cladding breaches (including hairline cracks and pinhole leaks) in the fuel pin sections that will be exposed to an oxidizing atmosphere. This can be done by a review of records (for example, sipping records) or 100% eddy current inspection of assemblies.

3) Determine the time-at-temperature profile of the rods while they are exposed to an air atmosphere and calculate the expected oxidation to determine if a gross breach would occur. The analysis should indicate that the time required to incubate the splitting process will not be exceeded. Any analysis would have to address expected differences in characteristics between the fuel to be loaded and the fuel that was tested to determine the basis for the analysis. Conversely, the maximum allowable temperature of the rods could be limited to the temperature that calculations show cladding splitting will not be expected to occur. Such evaluations must incorporate the effects of uncertainty in the data base.

Inspection of the rods by either eddy current or visual inspection, to the extent needed to assure there are no pinhole cracks is difficult, time consuming, and subject to error. Calculation of the possibility of cladding splitting is fraught with all the uncertainties discussed above. Lowering the maximum allowable temperature may impose an economic penalty by limiting the heat load

in the cask. The selection of the methodology used to address this issue is up to the applicant. The use of an inert atmosphere to prevent an oxidizing atmosphere is one method accepted by the staff, to address the issue

The materials reviewer should coordinate with the thermal reviewer to determine that the operating procedures, technical specification, and associated licensing documentation, as submitted by the applicants, provide a supportable analysis of the potential for cladding splitting, should fuel rods be exposed to a oxidizing gaseous atmosphere.

Appendix A provides detailed technical discussion for ISG-22 on oxidation of LWR spent fuel or other uranium oxide based fuel in an oxidizing environment.

Recommendation:

The staff proposes that NUREG-1536, NUREG-1567, and NUREG-1617 be modified to add the technical review guidance and technical discussion contained in this ISG. This ISG will result in modifications to the thermal chapter and operating procedures of these SRPs.

Approved:_____

E. William Brach, Director — Date
Spent Fuel Project Office

APPENDIX A
Technical Discussion for ISG-22

Fuel Oxidation and Cladding Splitting

Irradiated uranium dioxide exposed to an oxidizing atmosphere will eventually oxidize to U_3O_8. The time it takes to oxidize is a function of temperature that follows an Arrhenius function. However, at temperatures that may be expected for some spent fuel, this reaction can occur within a matter of hours.

The grain boundaries of irradiated fuel are highly populated with voids and gas bubbles. Initially the grain boundaries are oxidized to U_4O_9 resulting in a slight matrix shrinkage and further opening of the pellet structure. Oxidation then proceeds into the grain until there is complete transformation of the grains to U_4O_9 [EIN92]. The grains remain in this phase for a temperature dependent duration until the fuel resumes oxidizing to the U_3O_8 state. The transformation to U_3O_8 occurs with ~33 % lattice expansion that breaks the ceramic fragment structure into grain sized particles. At higher temperatures, the two transformations occur so rapidly that they are difficult to distinguish. The mechanism of oxidation in irradiated fuel appears to be different than in unirradiated fuel where U_3O_7 is formed and oxidation proceeds from the fragment surface and not down the grain boundaries. This mechanistic change occurs at or below ~10 GWd/MTU.

When the UO_2 is in the form of a fuel rod, the expansion of the fuel, when it transforms to U_3O_8, induces a circumferential stress in the cladding. Due to the swelling of the fuel, the process is usually initially localized to the original cladding crack site. The cladding strains due to this stress range from 2-6% before the initial crack starts to propagate along the rod. The incubation time to initiate the propagation and the rate of propagation have an Arrhenius temperature dependence. Axial propagation, spiral propagation and a combination of the modes that result in splitting have been observed in PWR rods [EIN86].

Data Base

The data base for oxidation was developed mostly in the 1980s in the US, Canada, England, and Germany. The data can usually appear in four forms: 1) O/M ratio [ratio of oxygen to metal content of the oxide] vs. time, 2) time to the $UO_{2.4}$ plateau vs. time, 3) cladding splitting incubation vs. time, and 4) cladding splitting rate vs. time. Some later work was done by the Japanese, and most recently work is on-going by the French primarily on MOX fuel. Much of the work was done on unirradiated fuel. All the work on cladding splitting was done in the early 1980's by the US [EIN86, EIN84, JOH84] and Canadians [NOV84, BOA77] and is limited. Recently DOE [BEC05] has issued an analysis of the oxidation issue in relationship to handling of potentially breached fuel in their proposed handling facility at the repository. This analysis depends on variables such as the gap between the fuel and the cladding, and burnup in a manner that is currently under technical review. In total, this research has shown that there are a number of variables that can affect the rates at which the fuel oxidizes and the cladding splits: burnup, moisture content of the air, cladding material, and type of initial defect.

The DOE study [BEC05] for Yucca Mountain uses a model for the cladding splitting that tries to account for the fuel-to-cladding gap and burnup of the fuel. The gap is the as-measured cold

gap and does not account for the closing of the gap due to differential thermal expansion of the cladding and fuel material, which could be calculated. There is very limited data on burnup effects. Therefore, it cannot be determined if the DOE model is correct. Plots in the Einziger document [EIN86] present actual data and comparisons with the data taken by other researchers at 30 GWd/MTU. The gap closure is implicitly accounted for in the measurements of splitting. However, no burnup effects can be inferred from this data.

Limitations of the Data Base

No oxidation or cladding splitting studies have been conducted on fuel with burnup greater that 45 GWd/MTU. Data between 30 and 45 GWd/MTU, shows a decrease in the oxidation rate due to the presence of certain actinides and fission products that are burned into the fuel. There is no reason that this should not continue at higher burnups, but the strength of the effect may change with burnup. Higher burnup fuel (>55 GWd/MTU) forms an external rim on the pellets that consists of very fine grains (1 micron vs 10 micron). As indicated earlier, the oxidation process is a grain boundary effect. The fuel pellet must be divided into two regions for the purpose of oxidation analysis; the center of the pellet where the grains have grown slightly, and the rim. While the rate of the oxidation may decrease with burnup, the total amount of fuel that is oxidized may increase due to a much greater intergranular surface area in the rim region. The DOE model [BEC05] uses a linear decrease in oxidation with burnup but this has, as yet, not been substantiated. A burnup effect is supported by Hanson's analysis [HAN98] of Einziger and Cook's data from the NRC whole-rod tests in which defect propagation was observed to occur earlier at the defects at the lower end of the rod where the burnup was lower.

Studies using a low partial pressure of water vapor in air have not shown any dependence of the oxidation rate on the moisture content of the air [FER05]. On the other hand, there are some studies that have shown a large increase in the oxidation rate when the moisture content is above 50% of the dew point [CUN03]. Oxidation in a 100% steam atmosphere is a different process. There are also studies that indicate that the oxidation rate will decrease if the oxygen content in the atmosphere drops into the range of a few torr or less. It does not appear that there is an effect of oxygen content at higher oxygen levels but the data is sparse.

All oxidation studies on fuel, with few exceptions, have been conducted on PWR fuel [EIN86, JOH84]. However, the UO_2 matrix is essentially the same in both PWR and BWR fuel. At the higher burnups, oxidation behavior may vary slightly as the actinide and fission product burn-in varies. The effect of the process on the splitting of the cladding may vary considerably due to the difference in gap size between the cladding types, and the thicker cladding in BWR rods.

The limited cladding splitting studies have been conducted on Zircaloy clad PWR [EIN86, EIN84, JOH84] and CANDU fuel. Defects were put in the fuel either by an SCC (stress corrosion cracking) process producing small sharp holes more typical of those found in reactor initiated SCC and by drilling that produced a larger duller hole. Most of the defects used in the studies were of the latter type. No measurements were made in cladding above 30 GWd/MTU. Very few data points were measured to determine the splitting rate and the time to start splitting has to be determined by interpolation. As a result, there is large uncertainty in both measurements. No measurements have been made on other alloy types (e.g., M5 and Zirlo) or at higher burnups where the cladding may be more brittle.
In light of the uncertainties that oxidation would introduce for fuel performance during accidents and fuel retrievability, ISG-22 provides technical review guidance to minimize the potential for

oxidation.

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References

BEC05 - Commercial Spent Fuel Handling in Air Study, 000-30R-MGR0-00700-000-000, (March 2005).

BOA77 - DG Boase, and TT Vandergraaf, "The Canadian Spent Fuel Storage Canister: Some Materials Aspects, "*Nucl Techol*., 32, 60, (1977).

EIN84 - RE Einziger and JA Cook, "LWR Spent Fuel Dry Storage Behavior at 229 C," HEDL-TME 84-17, NUREG/CR-3708, Hanford Engineering Development Laboratory (Aug 1984).

EIN86 - RE Einziger and RV Strain, "Oxidation of Spent Fuel at Between 250 and 400ºC," EPRI-NP-4524, Electric Power Research Institute (April 1986).

EIN92 - RE Einziger, LE Thomas, HV Buchanan, and RB Stout. 1992 "Oxidation of Spent Fuel in Air at 175 to 195 C", *J Nucl. Mater*., 190, p53.

FER05- C Ferry et.al. - Synthesis on the Spent Fuel Long Term Evolution, Rapport CEA-R-6084, (2005).

HAN98 - BD Hanson 1998, *The Burnup Dependence of Light Water Reactor Spent Fuel Oxidation*, PNNL-11929, Richland, Washington,: Pacific Northwest National Laboratory. TIC: 238459.

JOH84 - AB Johnson et.al., "Exposure of Breached BWR Fuel Rods at 325C to Air and Argon," *Proc. NRC Workshop on Spent Fuel/Cladding Reaction During Dry Storage*, Gaithersburg, Maryland, Aug 1983, NUREG/CR-0049, D. REISENWEAVER, Ed., .S. Nuclear Regulatory Commission (1984).

NOV84 - J Novak, and IJ Hastings, "Post-Irradiation Behavior of Defected UO2 in Air at 220-250 C," *Proc. NRC Workshop on Spent Fuel/Cladding Reaction During Dry Storage*, Gaithersburg, Maryland, Aug 1983, NUREG/CR-0049, D. REISENWEAVER, Ed., .S. Nuclear Regulatory Commission (1984).