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Oxidation of Spent Fuel During Fuel Handling Accidents Test Plan

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1. INTRODUCTION

The transfer of commercial spent nuclear fuel (CSNF) from transport or dry storage cask into final repository cask represents a scenario where fuel elements are potentially at risk to exposure to air and elevated temperatures. In the event of an accident during handling operations, the facility cooling may cease to function, and the temperature of the fuel would begin to rise due to decay heat generation. CSNF is generally stored in an inert environment, but handling operations could be performed in an air environment. If any damaged fuel elements are present during this postulated accident the fuel could begin to oxidize from UO_2 to U_3O_8. If the oxidation event is severe, volume expansion from oxidation could result in large-scale cladding failure and large releases of CSNF debris into the transfer facility. Depending on the design of the fuel handling facility, any SNF release could have an impact on off-site dose.

To build a facility that can reasonably process the inventory of CSNF that is ready for a repository at a reasonable cost, handling in air may be necessary. To evaluate accidents at this facility the oxidation behavior of damaged CSNF needs to be well understood and characterized. It may also be necessary to understand the behavior of intact cladding in a fuel handling accident as well. Fortunately, some relevant information already exists since the oxidation of CSNF has been previously studied [1–4].

The progression of UO_2 oxidation is well known. When exposed to an oxidizing atmosphere at elevated temperatures, UO_2 will begin to oxidize along its grain boundaries to U_4O_9 which is denser than UO_2. This densification of the fuel continues to expose fresh UO_2 surfaces and pulverizes the UO_2. As the reaction continues, U_4O_9 oxidizes further to U_3O_7 (at some temperatures and this step may not occur in irradiated fuel) then it oxidizes to U_3O_8, which is less dense than UO_2. The U_3O_8 phase volume expansion can become particularly problematic in a spent fuel handling accident, and will begin to stress the cladding, potentially causing further cladding failure by widening the original gap in cladding that initially allowed air ingress and the fuel to begin oxidizing. Because the fuel is pulverized during oxidation, the expanded cladding breach can begin releasing a fine particulate of spent nuclear fuel (SNF) which could lead to off-site dose and will certainly require a significant decontamination effort in the CSNF handling facility. The oxidation of fuel and subsequent increased cladding failure also leads to the loss of containment of fission gases and other volatile fission products in the SNF, particularly Cs.

There are still some gaps that exist in the understanding of CSNF behavior. Specifically the following phenomena warrant further study to determine how they affect UO_2 oxidation kinetics:

- The effect of final UO_2 grain size
- The effect of final fuel chemistry (burnup)
- The effect of the High Burnup Structure or high burnup rim structure that can form under certain conditions
- Commercial reactor type: pressurized water reactor (PWR) vs. boiling water reactor (BWR), which changes cladding behavior (zircaloy-4 based cladding vs. zircaloy-2 based cladding), pellet size where BWR pellets are larger than PWR pellets and other differences.

The progression of fuel failure is also dependent on the properties of the cladding after irradiation and possible long term wet/dry storage. Other factors that impact SNF behavior in air include:

- High-temperature zirconium alloy creep
- Zirconium alloy fracture toughness as it relates to crack propagation

Another factor to consider is the size and shape of the initial defect in the fuel. The progression of oxidation is also impacted by the amount of moisture in the air.

This report reviews the existing literature on CSNF oxidation and identifies gaps in the current knowledge of the oxidation of CNSF during a fuel handling accident. A proposed test plan to begin closing the knowledge gaps in CNSF oxidation follows the gap analysis.

2. PREVIOUS STUDIES

The consequences of a fuel handling accident resulting in the oxidation of damaged fuel have been studied when determining plans for permanent CSNF geologic repositories. Einziger and Strain summarize much of the work performed in the 1980s in their report and in a paper on the oxidation of PWR fuel [1,2]. From the mid-1980s, the basic problem was clear. If UO_2 is exposed to air at elevated temperatures, it oxidizes, pulverizes, expands, and then produces a stress on the cladding that can continue down the length of cladding, resulting in a massive release of irradiated fuel. With the available data, Einziger and Strain established conservative time and temperature limits for in-air handling of fuel with pre-existing defects. Data needs were also identified that specified the need to understand some specific radionuclide release pathways, specifically for H-3 and C-14. Only PWR fuel was evaluated in their study. While no significant difference between PWR and BWR fuel behavior was expected, there was a desire to confirm BWR behavior. The specific behavior of cladding splitting was not fully understood at all temperatures of interest, as data and testing of SNF were sparse. This is an area where targeted experimentation and parametric studies with modeling may prove very beneficial in new studies in this area.

Gaps in the understanding of UO_2 oxidation are further defined in a report by the Civilian Radioactive Waste Management Service (CRWMS) [4]. This report summarizes much of the research conducted between the Einziger and Strain summaries [1,2] in 1986 and the writing of the Civilian Radioactive Waste Management report in 2000. One of the most significant derivations from Hanson's CRWMS report [4] is the derivation of incubation time to U_3O_8 as a function of burnup based on the data in Hanson's earlier work [5]. For dry air, the following model from the CRWMS report [4] is conservative and relatively easy to apply to a variety of fuel handling situations.

$$t_{inc} = k_{2.4} e^{\frac{Q_{2.4}}{RT}} + \lambda_{inc} k_{75} e^{\frac{Q_{75} + \alpha \times BU}{RT}} \tag{1}$$

where
t_{inc} = incubation time in hours
$k_{2.4}$ = is the pre-exponential factor for the UO_2 to $UO_{2.4}$ transition (h) Nominal Case: 1.40×10^{-8}, Bounding Case 2.93×10^{-9}
$Q_{2.4}$ = in the activation energy (105 kJ mol^{-1})
R = the universal gas constant
T = the temperature in Kelvin
k_{75} = the pre-exponential factor for the $UO_{2.4}$ to U_3O_8 transition (h) Nominal Case: 4.84×10^{-14}, Bounding Case: 1.48×10^{-14}
λ_{inc} = This is a geometric term related to the outer radius of the fuel and the inner radius of the cladding. For the no gap case it is 6.30×10^{-3}.
Q_{75} = Arrhenius activation energy for $UO_{2.4}$ to U_3O_8 150 kJ mol^{-1}
α = 1.0 kJ mol^{-1} per MWd/kgU
BU = local burnup of the sample (MWd/kgU)

The nominal and bounding case curves from these values are shown in Figure 1. Operational scenarios would have fuel exposed to for about 100 hours and accident scenarios could have the fuel exposed to air for 1000 hours. For 100 hours of incubation time, the key temperature to stay below is

250°C for the bounding case and 280°C for the nominal case. At 1000 hours of incubation time, the key temperature to stay below is 200°C for the bounding case and 230°C for the nominal case.

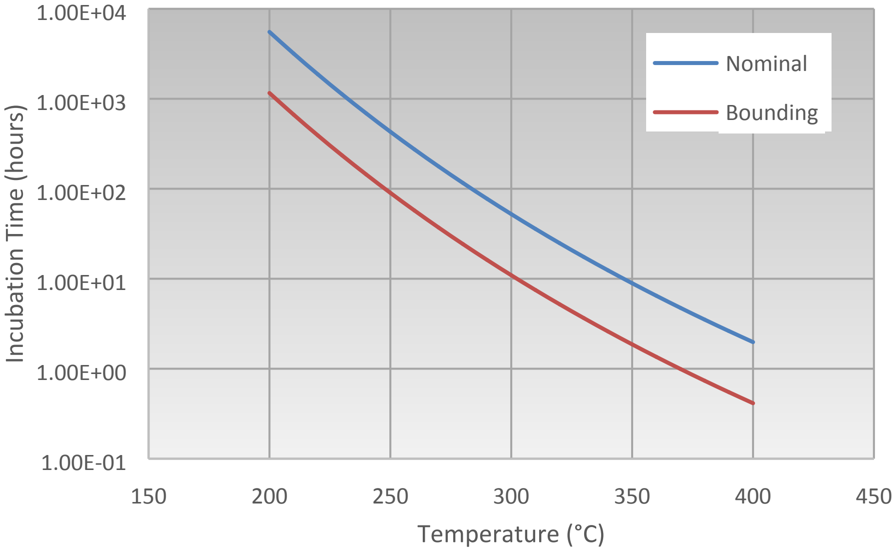

Figure 1. Incubation time to the initiation of a defect due to dry oxidation of the fuel

Grain size at end of life is expected to have some effect on the progression of oxidation in CSNF and some effect on the final respirable fraction released from an accident. This is due to the progression of oxidation along the grain boundaries [3]. Work to specify the effect of grain size on oxidation was also performed [6], but there was not a clear relationship established for typical UO_2 grains, and the effect of high burnup structure (HBS) UO_2 [7,8] was not established. This research on grain size notes that oxidation data for high burnup UO_2 above 45 MWd/kgU needed to be established. The final conclusion from Hanson [3] was that assuming an as-fabricated grain structure for most fuels was still a conservative assumption for most fuels. Grains tend to grow in UO_2 under irradiation, with the exception of HBS formation.

Work on BWR fuel [9] and CANDU fuel [10] is also presented in the CRWMS report [4], but no readily significant change in the behavior of these fuels vs. PWR fuel is reported. Typical BWR discharge burnups are also lower than PWR burnups, making BWR assemblies less likely to be the bounding case for a fuel handling facility. The effect of fission products has also been considered in some research [11,12] and is currently incorporated into incubation time by the burnup term, as indicated in Eq. (1). However, with higher burnup fuel that has experienced varied burnup histories, it may be important to consider rare earth content directly and not just as an empirical burnup factor.

Extensive work on irradiated higher burnup CSNF is summarized in Hanson's 2008 report [3]. One of the major conclusions from this work was that high burnup structure had little impact on the particle size distribution of oxidized fuel. Although in the summary it is acknowledged that there is some conflict in the literature. This is also in contradiction to some more recent observations of HBS behavior under certain accident scenarios [7,13–16]. The behavior of HBS at temperatures used in fuel handling scenarios may be significantly different from its behavior under in-pile conditions. It will be important to resolve this inconsistency in any future proposed work. Unfortunately, the method used to evaluate mass gain in previous work had high uncertainties. Irradiated fuel was oxidized in a test apparatus, and then the mass was measured again after oxidation. It is not clear why mass gain was not monitored continuously. The proposed work will seek to conduct similar measurements in a thermogravimetric analyzer to better capture the kinetics of oxidation.

The effect of water is less well understood than the effects of air, but has also been studied previously [17–19]. When considering water, two distinct scenarios must be considered. In one case, failed fuel may be exposed to a water environment either in a fuel handling pool or from a pre-existing defect that causes a pin to become filled with water. In another case, the fuel may be exposed to an environment with a mixture of air and water vapor. Either scenario changes the kinetics of oxidation and introduces hydrogen, which further complicates the progression of UO_2 oxidation, or may accelerate cladding failure by hydriding the cladding.

Research has also investigated the particle sized distribution of aerosol powders released from CSNF from oxidation and mechanical agitation [3]. This study was limited to only 4 irradiated fuel samples. Clearly there is need for additional data in this area. The overall study of fuel particle dispersal from various different defects and oxidation scenarios is not yet fully defined in the literature.

3. GAPS IN THE UNDERSTANDING OF CSNF OXIDATION

The previous section briefly covers several decades of extensive research into the oxidation of UO_2. To the first order, the oxidation of CSNF with preexisting flaws is understood. However, the limitations that arise from working with irradiated fuel and the evolution of light-water reactor (LWR) fuel over the past few decades have still left some gaps in the full understanding of CSNF in an air environment.

contains several variables that may impact CNSF oxidation and the dispersal of particulates following an accident. The state of knowledge on each variable is summarized, and proposed testing to improve understanding of each effect is defined, with some justification provided.

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Table 1. Variables That May Impact CNSF Oxidation and the Dispersal of Particulates Following an Accident

Variable	**State of knowledge**	**Proposed testing**
	Tests on UO_2	
Basic oxidation kinetics of UO_2	This variable is well known and has been tested at several temperatures for standard UO_2 powders [1–4].	Some basic testing of UO_2 powder oxidation is proposed to baseline other measurements proposed in this test plan. Also, some oxidation tests may be performed inside an XRD to better characterize the progression of UO_2 through the different oxidation states.
Grain size effect	No effect due to grain size has been observed in the literature to date. This includes some integral work on irradiated fuel that contained HBS [3].	No testing is proposed at this time; future capabilities in synthesis of nano-grain UO_2 may enable characterization of grain size effects in unirradiated UO_2.
Dissolved fission products and plutonium content (burnup) effect	A factor accounting for burnup has been proposed for the working model derived in the CRWMS report [4], but the data used to determine this factor were quite scattered. The oxidation behavior of UO_2 will be impacted by the oxygen potential in the fuel, which is impacted by the fuel's fission product and Pu content. To a first order, this content can be captured by a simple burnup term, but a more mechanistic model should be developed, especially to capture the behavior of higher burnup LWR fuels that have become the norm in the fleet.	Oxidation kinetics of unirradiated UO_2 doped with known amounts of dissolved surrogate fission products and surrogate Pu, primarily Ce, will be fabricated to mechanistically understand the oxidation kinetics of this simulated burnup fuel (SIMFUEL). The results can be applied more consistently to all fuel destined for the repository.
Additive impact	Vendors have begun to introduce doped UO_2 fuels into light LWRs [20] to enlarge grain size, which is intended to slow fission gas release during normal operating conditions and to limit pellet-cladding mechanical interaction by modifying the fuel's mechanical properties. This fuel has not been evaluated by prior studies.	The impact of additives such as aluminum oxide, chromium oxide and gadolinium oxide will be evaluated by oxidation kinetics studies over the temperature range applicable to fuel accident scenarios. This will be very complementary to studies on dissolved fission products and Pu surrogates.
Integral irradiated UO_2 testing	The current data set on high burnup LWR UO_2 is limited to the few samples evaluated in the Hanson et al. Fuel-in-Air FY07 Summary Report [3].	The scatter in the available data and increasing discharge burnups from the commercial fleet indicate that the need remains to further test the oxidation kinetics of irradiated UO_2.

Table 2. Variables That May Impact CNSF Oxidation and the Dispersal of Particulates Following an Accident (continued)

Variable	State of knowledge	Proposed testing
Zirconium testing		
High-temperature creep and burst behavior of irradiated zircaloy	The thermal creep properties of irradiated zircaloy are well known up to 400°C, but they are not well known above that temperature.	If an applicable accident can be identified, then creep behavior of fresh and pre-hydrided zircaloy will be investigated above 400°C. The orientation of zircaloy hydrides may also be analyzed. This could include investigating zircaloy burst behavior in the absence of a through-cladding defect. At some temperatures and internal pressures, it may be possible to fail zircaloy with only partial damage to wall thickness.
Defect size and shape effect	This variable has not been fully studied.	Some work with nonradioactive surrogate ceramic pellets that oxidize in a manner similar to that of UO_2 will be performed. Additional work with UO_2 and SIMFUEL UO_2 will benchmark the behaviors of different defect sizes and shapes.
Integral system testing		
Cladding unzipping from oxidation	Cladding unzipping during fuel handling has been a known issue since the early 1980s [21]. It is known that UO_2 will strain cladding as it oxidizes to U_3O_8, but most work has been focused on understanding the time to U_3O_8 formation because the unzipping process is very complex.	Small lengths of refabricated, irradiated LWR fuel with different defect sizes will be tested in existing furnaces to allow for the study of the progression of cladding unzipping.
Defect size and shape effect	Not fully studied	This will be covered by the testing campaign described in the item above; surrogates will be used to gain a better understanding of the problem before irradiated material is used.
Aerosol behavior	Aerosol behavior has been studied [3], but more work should be done to expand this data set. The distribution of particles is known, but the composition of different particle sizes is not known.	Additional aerosol samples will be collected in parallel with other tests planned on irradiated fuel. The compositions of the different particle sizes will be investigated by electron microscopy to determine if they vary. If particles form from different regions in the fuel, then certain high-dose consequence fission products (Cs, I) and actinides (Pu, Am) may reside in the aerosol particles of a specific side.
Water vapor		
Effect of water vapor on oxidation kinetics	The effect of water is less well understood but has been studied previously [17–19]. It is known that water influences the sintering kinetics of UO_2 at much higher temperatures.	An initial experiment to study the oxidation kinetics of UO_2 at fuel handling accident relevant temperatures 300-600°C is proposed for cases in which air plus significant humidity are used in testing. If the result differs significantly from the dry air case, then many of the other experiments proposed will need to be repeated with water vapor.

4. PROPOSED WORK SCOPE

After considering the gaps discussed in the previous section, a work scope was developed to begin closing the gaps:

1. Compile a complete data set of oxidation kinetics tests on UO_2 powders, UO_2 powders with known concentrations of dopants, and irradiated materials at temperatures that span dry storage and fuel handling accident conditions. Dopants include simulated fission products, simulated Pu surrogates, and fuel performance additives used in the most recent LWR fuel.
2. Evaluate the behavior of fresh and irradiated zirconium alloys at elevated temperatures relevant to fuel handling accidents.
3. Execute a novel experimental approach to assess the impact of rapid depressurization following cladding failure at low temperature storage conditions on fuel integrity and particulate release. Perform experiments to facilitate improved understanding of cladding failure propagation (unzipping) as a function of fuel state.
4. Provide first-of-a-kind mechanistic data on the impact of moisture in storage and fuel handling accident oxidation behavior and radioactivity release.

Oxidation kinetics studies on UO_2 are proposed to better mechanistically quantify the incubation time behavior of irradiated UO_2. Two primary methods are proposed. The kinetics of the UO_2 to U_3O_8 reaction will be monitored using thermogravimetric analysis (TGA) from approximately 250°C up to 500 or 600°C, depending on input to be obtained from thermal analysis of fuel handling accidents. In addition to the mass change information that can be attained from TGA, crystallographic information can be obtained from in-situ oxidation testing in an x-ray diffractometer (XRD). Both of these techniques will be baselined on fresh UO_2 powder and compared to the standard incubation time equation. UO_2 with dopants (SIMFUEL) and additives will then be tested in a similar manner. The SIMFUEL measurements will allow for the modification of the incubation time equation to more accurately reflect the final state of the fuel prior to procedurally complex irradiated fuel measurements are taken. Finally, TGA measurements on irradiated UO_2 will be performed to confirm the SIMFUEL measurements and to obtain improved data on irradiated fuel oxidation testing.

The severity of a fuel handling accident is exacerbated by the crack behavior of zircaloy alloys that continue to expose fresh UO_2 to air at elevated temperatures. The burst and unzipping behavior of irradiated zircaloy is driven by the thermal creep and fracture toughness of irradiated zircaloy, which is not well known above LWR operating temperatures (>~400°C). These mechanical properties are also impacted by the prior thermal history of the cladding. Particularly, if hydride reorientation has occurred in the cladding, the creep and fracture toughness may cause the zirconium to fail particularly severely. Thermal creep tests can be performed on fresh and pre-hydrided cladding to simulate high burnup and dry storage conditions. Fracture toughness testing can also be performed on fresh and pre-hydrided cladding. These basic mechanical properties are imperative to mechanistically understand fuel unzipping, and they could simulate the progression of fuel handling accidents beyond the experimental basis.

The first two parts of this test plan will lead to the confirmatory integral testing . Integral tests will begin with nonradioactive surrogate testing. Pellets that approximate the oxidation behavior of UO_2, perhaps CeO_2 or ZrO_2, will be placed in zirconium cladding with preexisting flaws under an oxidizing atmosphere at elevated temperatures. This testing on fresh and hydrided cladding should inform the severity of oxidation accidents prior to testing of UO_2–bearing fuel or irradiated fuel. This will also inform the sample lengths necessary for irradiated fuel testing. The confirmatory integral testing will leverage prior Advanced Fuels Campaign work on burst testing in the prototype Severe Accident Test

Station (SATS), and it may also be conducted in the prototype SATS furnace. Similar tests on unirradiated UO_2 and unirradiated doped UO_2 should follow in equivalent furnaces that are approved for uranium work. The surrogate and unirradiated UO_2 tests will also identify if there is a defect size and shape effect on cladding unzipping. Testing of round hole, longitudinal cracks, and circumferential cracks can all be tested in these configurations before tests are conducting using irradiated fuel testing. These tests will inform irradiated fuel testing in the hot cell SATS. During the testing, particles produced by cladding failure will be collected to determine the distribution of aerosol particles that may be produced during an accident. This distribution ultimately informs the potential dose to workers or the public. This approach to irradiated fuel testing should produce a more mechanistic data set that is well understood and that minimizes expensive hot cell testing.

At temperatures relevant to sintering UO_2 (~1600°C), small additions of water can greatly affect the progression of oxygen diffusion and sintering kinetics. It is less clear how H_2O in different concentrations will impact fuel handling accidents. All prior parts of this test plan will be performed under dry air conditions. Many of the tests proposed previously will be repeated with controlled H_2O additions to the atmosphere to capture the impact of humidity on oxidation. If it is shown that humidity does not have a significant impact on oxidation at relevant temperatures, then humidity testing will not progress all the way to full-scale integral irradiated fuel testing.

The proposed schedule is subject to funding levels as follows. In the first year, testing on UO_2 and UO_2 with additives will be performed using existing equipment. Some development of UO_2 with additives will be necessary, but existing sol-gel technology will allow for at least Ce incorporation into prepared UO_2 SIMFUEL powders. An XRD for in-situ testing will be identified, and an environmental chamber for oxidation testing in the XRD will be procured. A TGA for irradiated fuel testing will be procured and installed in the Irradiated Fuels Examination Laboratory (IFEL) at Oak Ridge National Laboratory (ORNL) for future irradiated fuel TGA testing in the next year. Further review of existing zirconium alloy creep and fracture toughness data will be performed. Efforts to fill gaps in the data up to hydriding levels expected in high burnup LWR fuels will be initiated in the first year of the project. Integral testing in prototype SATS with surrogate pellets will also be initiated in the first year. Integral UO_2 and irradiated fuel testing will be delayed to the second year and out years. Only limited H_2O work if any will be performed in the first year of the project subject to initial dry air testing.

5. SUMMARY

Fully understanding the progression of irradiated fuel handling accidents requires additional data. Gaps exist primarily (1) at the highest burnup levels, (2) in new fuel forms that incorporated in-pile fuel performance–enhancing additives, and (3) concerning the impact of water on the progression of oxidation. This test plan proposes to close these gaps through a tiered approach that maximizes the mechanistic understanding of this problem while minimizing the necessary irradiated fuel testing. These tests will include the study of oxidation kinetics in UO_2, SIMFUEL UO_2, and irradiated LWR fuel, as well as the mechanical properties of zirconium alloys. This new understanding will be leveraged to support integral testing, and where applicable, it will be applied to tests regarding the impact of water vapor on oxidation.

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