UNITED STATES
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139

MEMORANDUM FOR: John G. Davis, Director
Office of Nuclear Materials Safety and Safeguards

FROM: Robert B. Minogue, Director
Office of Nuclear Regulatory Research

SUBJECT: RESEARCH INFORMATION LETTER NO. 139, POTENTIAL OXIDATION OF UO_2 IN IRRADIATED FUEL AND ITS REGULATORY IMPLICATIONS

References:

1. Letter to C. E. MacDonald from M. Resnikoff, Sierra Club, November 7, 1983.
2. Memorandum to J. G. Davis from R. B. Minogue, "Plans for Technical Response to Address Concerns Regarding Oxidation of UO_2 in Irradiated Fuel," dated November 25, 1983.

Introduction

The purpose of this memorandum is to provide an appraisal of UO_2 fuel oxidation phenomena and its potential impacts on the transportation of irradiated power reactor fuel assemblies. Concerns regarding the potential regulatory implications of this oxidation process were raised in reference 1. The basis for these concerns is an incident which took place at the Battelle Columbus Laboratory (BCL) in May 1980. The incident involved the release of radioactivity to the BCL fuel storage pool and the enclosing building interior during unloading operations of a fuel assembly known to have fuel pins with large cladding splits. The assembly had been transported in an air and temperature environment in which the irradiated UO_2 fuel material could have been oxidized to higher oxidation states. One of these oxides, U_3O_8, can result in spalling and powdering of the irradiated fuel segments. The significance of this potential oxidation process on past consequence and risk assessments is assessed.

Background

The appraisal of the status and regulatory implications of the potential oxidation of irradiated UO_2 was carried out by members of both the RES and NMSS staffs. The outline proposed for this appraisal was transmitted in reference 2 and indicated that three major topics would be addressed. The first topic was to include a review and reevaluation of the incident at the Battelle Columbus Hot Cell Laboratory. The second topic was to include documentation of available information on the potential for UO_2 oxidation, specifically related to transportation and unloading/loading operations of irradiated fuel assemblies. This information would be used to assess the potential extent of UO_2 oxidation during these activities. The third topic was to include definition of the regulatory implications of the potential oxidation process.

Results

The detailed results of this appraisal are presented in the appendix of this memorandum. The BCL incident is evaluated in section I. The conclusions reached are that during the unloading of a fuel assembly, known to contain fuel pins with large cladding splits, an estimated 120-240 Ci of fuel material and solid and volatile fission products were released to the unloading pool and the surrounding high bay building atmosphere. Two pathways can be identified as potential major contributors to this release. First, some fraction of potentially oxidized fuel could have been released from the failed fuel rods to the cask interior during transport (i.e., normal shocks and vibrations). Second, the flooding of the cask interior during cooldown involved water interactions with thermally hot fuel rods and this interaction could have caused washout of additional material contained in the fuel rods. The exact mechanism of release of fuel from the cask cavity into the pool and building atmosphere and the contribution of any fuel oxidation on the magnitude of release is not known.

The important parameters and uncertainties associated with the oxidation process are discussed in the appendix, section II. The maximum extent of irradiated fuel oxidation is evaluated for normal and abnormal circumstances potentially associated with transportation. In order to evaluate the change in past risk estimates, a bounding likelihood for failed fuel shipments is proposed.

The possible impacts of the oxidation process on the risk and consequences associated with normal transport activities, severe accidents and sabotage events are discussed in section III of the appendix. The conclusion reached is that the impact on past risk estimates is small and the maximum release estimates would not be increased by more than a factor of 4.*

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* Based on tables 5-3, 5-12 and 5.13 in NUREG-0170, the 0.000422 expected latent cancer fatalities from spent fuel shipments assuming the 1985 shipment's model would not increase by more than 15%. The calculated consequences of extremely severe accidents in very high population density urban areas would increase from predictions of 1 latent cancer fatality to 4 latent cancer fatalities.

Findings

The major impact of irradiated fuel oxidation is the potential for occupational exposures during unloading operations. Three corrective actions are possible. First, proper unloading procedures could be devised to preclude a reoccurrence of the exposures associated with the BCL incident. Second, a non-oxidizing gas could be provided for dry spent fuel shipments which have temperatures sufficiently high to result in fuel oxidation ($>150^{\circ}C$). This provision could result in a reduction in both the likelihood of occupational exposures and the potential for contamination incidents affecting the cask or unloading facilities and equipment. Finally, canning of known failed fuel assemblies would provide an additional barrier against the release of solid fuel particles and would minimize the contamination potential inherent to such fuel assemblies.

Robert B. Minogue, Director
Office of Nuclear Regulatory Research

Enclosure: As stated

APPENDIX

I. Circumstances of May 1980 Contamination Incident at the Battelle Columbus Laboratory (BCL) Hot Cell

In May 1980, BCL received a fuel assembly containing known large cladding splits from the Connecticut Yankee (Haddam Neck) Power Plant in a NFS-4 spent fuel shipping cask. The assembly identified by BCL* as Batch 3 Assembly H07 was a (15 x 15) PWR assembly with stainless steel clad fuel rods. The assembly was to be used in an investigation of fuel rod failures being conducted by BCL for the Electric Power Research Institute (EPRI). Visual and photographic examination at the reactor facility, of the outer rods (outer two rows) in this assembly had identified several rod cladding failures including a 4-5 ft. long split approximately 1/8 inch wide. In several of these rods with split cladding an absence of some of the fuel pellets was noted.** During the subsequent examination program at BCL, a number of rods were removed for examination while others were identified to be in a failed condition. Figure 1, taken from reference 2, illustrates the location of removed rods and failed rods following the incident. Since the removed rods were from the highest burnup regions of the assembly (~37Gwd/MTU), it was believed that all failed rods were identified, although it is conceivable that one or two others may exist.*** The visual appearance of the failures at the reactor pool prior to shipment was stated as being similar to their appearance in the BCL pool after the incident.***

* Personal communication between V. Pasupathi and R. Klingensmith of BCL and NRC Review Group (W. Lake, W. Lahs, D. Reisenweaver and S. Turel).

** Figure 1 of reference 1 shows a view of one such rod.

*** Personal communication between V. Pasupathi and W. Lahs.

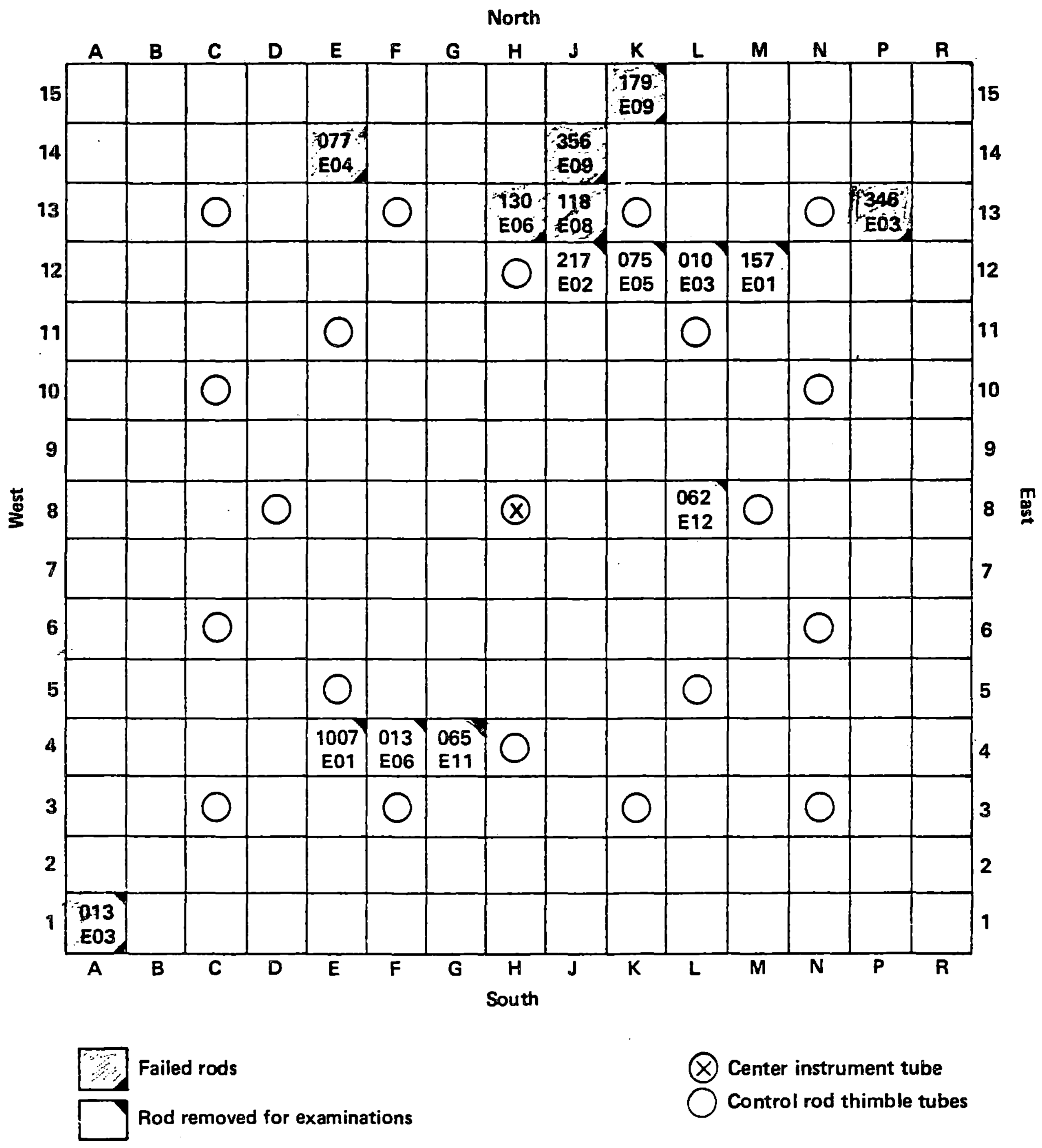

FIGURE 1 Locations Of Rods Removed From Batch 8 Assembly (1)

The assembly with decay heat between 3-3.5Kw (mistakenly believed to be about 2Kw) was transported to BCL in the NFS-4 (NAC-IE) shipping cask. The loaded cask cavity environment was normal atmospheric air. The loaded cask had remained on the reactor site for about 5 days and was in transit ∿1 day.

BCL had received failed fuel previously with no significant incidents; however, these shipments involved individual fuel rods or fuel assemblies with lower thermal power levels. When the cask was received at BCL, cavity pressure was checked (no pressurization indicated) and a cavity gas sample was taken prior to cooling and removal of fuel. The cavity gas sample was analyzed subsequent to the incident and the results indicated that the internal cavity gas was essentially depleted of oxygen (reference 1, pg. 649).

To accomplish cooldown prior to cask unloading, the cask cavity was slowly filled with water while the cask was vented to the hot cell through a connected hose. Steam was initially discharged from the hose indicating that the assembly was thermally hotter than fuel previously handled. A high radiation level alarm was also actuated within the hot cell. A sludge discharge from the hose was also noted.*

Following cavity flooding, the cask was lowered into the unloading pool where the head was removed. It was at this point that a dark cloud of material emanated from the cask resulting in contamination of pool water and airborne contamination of the cask handling area. Fuel particle release was confirmed by a β/γ activity ratio of 4 - this is much higher than ratios typical from normally observed contamination or "crud" which is dominated by the strong γ-emitter Co^{60}.

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* Personal communication between V. Pasupathi and NRC Review Group.

Battelle estimated that the peak fuel rod temperatures had reached 550°F. NRC predictions have also been made and these results are in good agreement given the uncertainties in evaluating radiative heat transfer between fuel rods and the inner cask structure.* Oxidation of UO_2 to U_3O_7, U_3O_8 of higher oxides can take place at these temperatures but the degree of oxidation can not be well defined. U_3O_8 is of particular interest since formation of this oxide can cause flaking of UO_2 pellets. Theoretically, about three moles of O_2 were available to react with the UO_2 in the cask cavity. Fuel samples were sent to Pacific Northwest Laboratories for analysis to clarify the composition of any existing higher fuel oxidation states; however, this analysis has not been performed due to higher priority work.

No precise estimates of the total activity released in the incident are available.** However, based on oxygen availability and the limited data on the physical condition of the fuel assembly, several estimates can be made which may be of use in assessing the significance of the incident. Considering the limited availability of oxygen and depending on the specific higher oxidation state, a maximum of 2.4Kg of original UO_2 conceivably could have been involved in the oxidation process <u>if U_3O_8 was the only oxidation product</u>. This 2.4Kg is approximately equivalent to the weight of UO_2 contained in one rod of an approximately 200 rod PWR fuel assembly. Post-incident examination of the failed fuel rods and estimates of the amount of radioactivity released from the fuel assembly indicate that the release of the disrupted or oxidized UO_2 from the fuel rods was much less than this theoretically available 2.4Kg (more on the order of a few hundred grams).

* Personal communication between W. Lake and W. Lahs.

** Personal conversation between V. Pasupathi and NRC Review Group.

Figure 2 taken from reference 1 shows a photograph of a section taken through the failed region of a rod. If this void extends through a length of 4-5 ft., its volume would be about 2% of the fuel. This material could have been lost prior to shipment; however, as a point for comparison, this amount of missing material in each of 3 rods would contain a fission product and transuranic inventory (excluding Kr^{85}) of about 180 Ci. This 180 Ci value compares with the rough estimate of the radioactivity which apparently was lost from the fuel assembly in the incident.

From the increase in the activity of the 125,000 gallon BCL pool after the incident, up to 70 Ci of radioactivity can be accounted for in the pool water. Based on airborne samples taken after the incident, the release to the building atmosphere was negligible compared to this value.(17) Isotopes identified in both water and air samples included Ru^{106}, Cs^{134}, Cs^{137}, and Ce^{144}. A highly radioactive dark liquid residue in the bottom of the cask cavity and a dark coating of the cask interior and internal supporting spacer were sampled and analyzed. The contaminating material was identified as fuel and fission products. Decontamination efforts performed both at BCL and subsequently, Rockwell International are believed to have removed a total of 40-50 Ci. A few tens of grams of fuel material may remain in the cask.

In summary, the BCL incident involved the following: (1) some irradiated fuel material in the form of UO_2 and higher oxides could have been released from failed fuel rods into the confines of the shipping cask during transportation; (2) during the cask cooldown and internal cavity flooding processes, fractions of material in the cask and material within the failed rods were released to the BCL fuel pool;

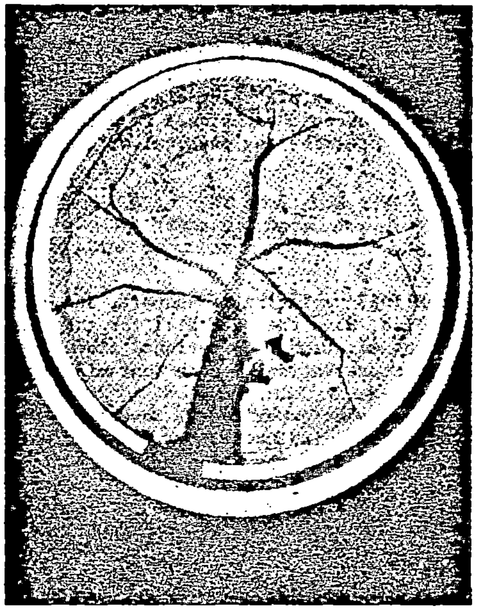

Fig. 2. Section Through Failed Region of Rod (1)

(3) the total release of solid and volatile fission products (excluding noble gases) from the fuel rods during this process is estimated to be in the range of 120 - 240 Ci; and (4) a few tens of grams of this released fuel material may still reside in the cask. The estimated release would represent from 3 - 7% of activity within a single rod. Oxidation of the UO_2 to higher oxidation states is believed to be the major contributor to the radioactivity release; however, it is conceivable that the cask cavity cooldown procedures could have played a significant role.

II. Potential for UO_2 Fuel Oxidation During Transport

The purpose of this section is twofold: first, to summarize the state-of-knowledge regarding UO_2 oxidation to higher oxidation states in transportation environments, and second, to present projections on the likelihood and extent of oxidation which could occur during normal and abnormal transportation situations.

Subsection II.a. will briefly describe the oxidation process - discussing the parameters affecting oxidation rates, the large uncertainties in predicting rates of oxidation of irradiated fuel, and the preliminary insights gained from on-going research projects.

Subsections II.b. through II.e, will provide both qualitiative and quantitative engineering judgements regarding the extent and bounding likelihood of irradiated fuel oxidation during normal and specified abnormal transportation situations. These judgements include uncertainties in understanding the process itself and rely on predictions based on the single BCL operational occurrence.

a. UO_2 Oxidation - Important Parameters and Uncertainties

UO_2 Oxidation

Until about one year ago, there was general agreement among investigators that the oxidation of UO_2 was a two-stage process. It was believed that the UO_2 was first converted to U_3O_7 and if further oxygen was available, U_3O_8 and eventually UO_3 was formed. Within the past year, this process has been questioned.

It is now believed by many investigators that U_3O_7 is a transition phase between UO_2 and U_3O_8. There is evidence that some U_3O_8 is formed before all of the UO_2 is converted to U_3O_7. The rate of oxidation is based on various factors which will be discussed later. Data presented by K. Simpson(3) indicates that after 6 weeks in air at 225°C (437°F), loose powder was formed from unirradiated UO_2 pellets. This powder was analyzed by x-ray diffractometry and was found to consist of ∿30% U_3O_7 and ∿70% U_3O_8.

During dry transportation of spent fuel, existing casks may be filled with air. If failed or breached rods are present in air, the UO_2 will be exposed to the oxygen and oxidation may occur. The density of UO_2 is 10.96 g/cc. There is a small density change when U_3O_7 is formed (11.4 g/cc). There is no visible evidence when this change occurs. However, the density of U_3O_8 is 8.35 g/cc and when this form of the oxide is formed, spalling or powder formation occurs due to the volume increase associated with the U_3O_8 formation. It is the U_3O_8 powder spalling which is of primary concern during handling operations and accident situations.

Parameters Affecting Oxidation Rate

It has been found that the rate of formation of U_3O_8 is dependent upon various pellet properties and transportation environment factors. The factors, besides oxygen availability, which have the greatest influence are temperature, time, surface area, and irradiation. Each of these factors is discussed below.

1. Temperature-Time:

There appears to be a period of time in which the formation of U_3O_8 is not visibly apparent. This time period is called the "incubation period." Once the incubation period is completed, it appears that the formation of U_3O_8 is accelerated and the powder spalls from the pellet. The incubation period is related to the temperature. Table 1 provides the incubation periods for unirradiated UO_2 at various temperatures. It can be seen that as the temperature of the fuel is increased, the incubation period is shortened.

2. Surface Areas:

The rate of oxidation is directly proportional to the exposed surface area. As the surface area of the fuel is increased, more grains of UO_2 are exposed to the available oxygen. The surface area can be increased in many ways. As pellets are irradiated, they tend to crack or fracture. This cracking appears to be proportional to the burnup. Another factor affecting surface area is the porosity of the pellet after manufacturing - the greater the porosity, the greater the surface area.

INCUBATION PERIODS FOR UNIRRADIATED FUEL PELLETS

TABLE 1

Temperature (°C)	Incubation Period	Weight Gain (mg cm^{-2} hr^{-1})
200	4000 hr (167 da)	6×10^{-5}
225	1000 hr (42 da)	1×10^{-3}
230	250 hr (10.4 da)	8×10^{-3}
250	200 hr (8.3 da)	2×10^{-2}
300	20 hr	4×10^{-2}
350	2 hr	1.5
400	0.5 hr	33

NOTE: This material is a best estimate based on the information presented in references 3, 5, 6, and 7.

The incubation periods in this table are based on unirradiated UO_2 pellets or fragments without cladding.

The size of the breach in the cladding will be a large contributing factor in the oxidation rate. Very small defects, such as stress corrosion cracks, tend to limit the amount of oxygen which can enter the rod. Defects 30 mils and larger allow the ingress of oxygen into the rod and the oxidation process occurs. The limiting size of defects which allow the oxidation process to occur and continue has not yet been determined. The small defect sizes necessary to allow oxidation raises related concerns regarding defect detectability.

During visual examinations conducted underwater with remote video equipment at the reactor site, it is very difficult to observe the fuel rods beyond the outer few rows of an assembly. Defects contained in these inner rods may not be detected. If the rods are more than a few years old, dry sipping of the assemblies to detect defects may also prove ineffective. After this amount of time, most of the volatile isotopes will have decayed away and the gaseous longer-lived isotopes will have escaped from the defects and dissipated. A wet sipping technique for determining the presence of fission products may be appropriate if the detection of failed fuel rods is desired.

3. Unirradiated vs. Irradiated Fuel:

The incubation times listed in Table 1 were derived from experiments using unirradiated UO_2 pellets and fragments. Various researchers have tried to corrolate the effects of irradiation on unirradiated UO_2 and determine how the irradiation affects the incubation period. All researchers agree that the incubation period is less for irradiated UO_2 than for unirradiated UO_2. However, they do not agree on the degree of difference. Factors of 2 to 50 have been proposed for this difference. No conclusions can be drawn on the applicability of the incubation period for irradiated fuel based on the unirradiated UO_2 data.

4. Environment:

G. White(5) has conducted an experiment as to the effect of the presence of quantities of nitrous oxide on the UO_2 oxidation process. Small quantities of NO_2 are generated by radiolysis of moist air. When NO_2 is formed, numerous reaction products are formed. One of these products may be nitric acid.

The experiment using air with 1% NO_2 indicated that the rate of oxidation of UO_2 was much higher than for a total air environment. The final product was UO_3 rather than U_3O_8.

There is some contradictory evidence from other investigators indicating that the oxidation rate is not affected by the introduction of moist air. No conclusions can be drawn from the presently available data.

5. Particle Size:

A limited amount of data has been collected concerning particle size of the formed U_3O_8 powder. The research sponsored by the NRC(4) has shown that most of the material formed by the oxidation process tends to move very little if not disturbed. The powder falls out of the rod via the breach, but due to its density does not appear to become airborne. The fuel material which fell from the breach in the NRC experiment laid in the bottom of the containers. However, some fission products did become airborne. Although this research is not directed at determination of fission product particle size, indications are that this fission product material included sizes between 2 and 15 microns.

6. Additional Research:

Additional research in the area of UO_2 oxidation is being conducted in the U.S. by the NRC, DOE, TVA, and EPRI. Research is also being conducted in the United Kingdom, West Germany, and Canada. As additional information becomes available, this research information letter will be updated.

b. Potential for Irradiated UO_2 Fuel Oxidation to Higher Oxidation States During Normal Transport

Two major conditions must be met before any significant oxidation of irradiated UO_2 to higher oxidation states can take place during normal transportation activities: (1) the UO_2 must be exposed to oxygen and (2) the exposure must take place at sufficiently elevated temperatures. Since UO_2 fuel pellets are normally clad in inerted rods (typically Zirc-2 or Zirc-4, although some fuel from older reactors is stainless steel clad), the first condition can be met only if the integrity of the cladding is violated and the external environment includes oxygen.

The temperatures required by the second condition can be generated in the transportation process by the confinement in a spent fuel shipping cask of an irradiated fuel assembly with sufficient decay heat.

Five currently licensed shipping casks have been identified in which irradiated fuel could reach potential oxidizing temperatures and which have an air filled cavity. Two other designs (NLI-1/2 and NLI-10/24) were considered which have sufficiently high fuel pin temperatures but have helium filled cavities. The five casks of concern and the predicted maximum fuel rod temperatures based on maximum allowable cask heat loads are shown in Table 2.

Table 2: Predicted Maximum Fuel Pin Temperatures Assuming Maximum Authorized Heat Loads and Normal Transport Conditions

CASK	No. Assy PWR	BWR	Q max* (KW)	Predicted max. temp. Fuel Pin, °F (°C)
NFS-4	1	2	2.5	415-447°F (213-231°C)
TN-8/TN-8L	3	-	35.5/23.7	837°F (447°C)
TN-9	-	7	24.4	712°F (378°C)
IF-300	7		11.72	650°F (343°C)
	-	18	11.72	510°F (266°C)

* Maximum authorized heat load for respective shipping casks.

All the predicted temperatures are sufficiently high to promote the potential oxidation of UO_2 to U_3O_8 over time periods typical to irradiated fuel shipments if the UO_2 is exposed to the cavity air. The free volume of air within the identified shipping casks ranges from about 12 ft^3 in truck casks to 96 ft^3 in rail casks. The oxidizing potential of this volume of air could convert UO_2 to a wide range of oxides (e.g., U_3O_7, U_3O_8, UO_3). The smaller volume, representative of a single PWR assembly truck cask, has been calculated to convert ∿2.4Kg of UO_2 to U_3O_8.** This value is conservative since available evidence indicates that other oxides would also be formed. The 2.4Kg value is approximately equivalent to the weight of the UO_2 contained in one rod of an ∿200 rod PWR fuel assembly which for 150 day cooled power reactor fuel would be expected to contain ∿ 9,000 Ci of solid and volatile fission products and transuranics.

** Values of 20 Kg presented in reference 1 are apparently in error. This error was carried over into the summary of the BCL incident described in the memorandum to files from W. Lahs, dated December 5, 1983.

However, based on the actual oxidation occurrence discussed in section I, it appears that the oxidation process contributed to the release of 120 to 240 Ci of fission product and fuel material from the fuel rods. This level of radioactivity is equal in magnitude to that contained in 70 - 130 grams of irradiated UO_2 fuel.

A bounding likelihood of a spent fuel shipment including a failed fuel assembly can be inferred from data in fuel performance reports.(8) For the calender year 1981, 0.6% of the fuel assemblies (both discharged and in core) were estimated to contain failed rods. If the number of failed assemblies is compared to the number of assemblies discharged, a bounding 3% occurrence rate can be calculated. These failed fuel assemblies typically average 3 failed rods/assembly with widely varied conditions of failure.(8)

c. Potential for Irradiated UO_2 Fuel Oxidation to Higher Oxidation States During Loading/Unloading Operations

The potential for oxidation of irradiated UO_2 to U_3O_8 or other higher oxidation states during loading or unloading operations requires conditions similar to those identified during normal transport activities. Since the typical loading operation involves transfer of a fuel assembly from a water-filled pool to the shipping cask, no significant oxidation would be expected until the water coolant is removed from the cask and the assembly temperature increases from its initial value of 70-100°F. (Temperatures sufficient to promote oxidation could be achieved.) Any potential for oxidation during loading is limited however, by the time of fuel exposure to oxygen. That potential should be clearly far less than the potential for oxidation during transport. For loading processes involving purging or vacuum drying and backfilling of the shipping cask with non-oxidizing gas, the times of exposure to air would be extremely limited. Unloading operations are also performed under water. Therefore, no significant fuel oxidation is expected.

d. Potential for Irradiated UO_2 Fuel Oxidation to Higher Oxidation States During Extremely Severe Transport Accidents

Analysis and testing have been performed to assess the response of spent fuel shipping casks to severe accident forces. These assessments indicate that no significant violations to containment integrity would occur for the severe conditions evaluated. However, since all severe accident conditions could not be evaluated, both references 9 and 10 assumed some cask containment violations sufficient to release small fractions of volatile fission products or activation products dissolved in cask cooling water. Some fuel rod cladding failures were also assumed to result from accident forces. Extending these reference assessments, it can be argued that under extremely severe conditions, the inert or oxygen depleted cavity gas could be replaced in time with atmospheric air.* For minor cracks or small penetrations of containments, the time for cavity gas replacement could involve several hours. For accidents which result in small cask breaches not affecting normal cask heat transfer capabilities, fuel temperatures could remain at values indicated in table 2 or table 3. Table 4 gives similar temperature maxima calculated for the severe accident conditions specified in 10 CFR 71. Therefore, the conditions for fuel oxidation conceivably could be attained sometime following the accident event. Following the incubation period

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* For failed fuel shipped in closed failed fuel connisters, an additional barrier to both oxygen ingress and fuel particle release would be provided.

Table 3: Predicted Maximum Fuel Pin Temperatures for He Cooled Casks Assuming Maximum Authorized Heat Load and Normal Transport Conditions

	No. Ass'y		Qmax*	Predicted max. temp.
Cask	PWR	BWR	(Kw)	Fuel Pin °F (°C)
NLI 1/2	1	2	10.6	1013°F (545°C)
NLI 10/24	10	-	70	900°F (482°C)
	-	24	40	724°F (384°C)

Table 4: Predicted Maximum Fuel Pin Temperatures Assuming Maximum Authorized Heat Load and Severe Accident Conditions

	No. Ass'y		Qmax*	Predicted max. temp.
Cask	PWR	BWR	(Kw)	Fuel Pin °F (°C)
NFS-4	1	2	2.5	438-472°F (226-244°C)
NLI-1/2	1	2	10.6	1102°F (594°C)
TN-8/TN-8L	3	-	35.5/23.7	998°F (537°C)
TN-9	-	7	24.4	825°F (441°C)
IF 300	7	-	11.72	754°F (401°C)
	-	18	11.72	614°F (323°C)
NLI-10/24	10	-	70	991°F (533°C)
	-	24	70	854°F (457°C)

* Maximum authorized heat load for respective shipping casks.

(referrred to in the generalized oxidation discussion), fuel within failed rods could oxidize to U_3O_8 at a rate which is a strong function of fuel rod temperatures and oxygen availability, until the process reached completion. Available data indicates that this process could take hundreds of hours at 450°F (232°C) but would occur rapidly at 750°F (399°C).(5)(11) Much of the oxidized material would tend to remain in place in the fuel rod if not disturbed, but any that fell from a fuel cladding breach could fall to the bottom of the fuel basket or shipping cask. The postulated circumstances that allow replacement of the preaccident cover gas with atmospheric air through some small containment violation are not conducive to allowing a significant release of this material from the cask.

Undefined accident forces even greater than those discussed above could be hypothesized to cause gross cask containment violations. Even under these conditions, immediate exposure of the fuel to atmospheric air can not be presumed. The geometric location of the violation would not only be critical to the rapid replacement of the cask cavity gas but also to the subsequent release of oxidized material. And, as noted before, if the fuel had cladding damage before shipment, making it more vulnerable to these most severe conditions, it would be further contained in an internal failed fuel cannister. Based on this subjective evaluation, a substantial release pathway for fuel material oxidized subsequent to the accident event is not considered a significant possibility.

e. Potential for Irradiated UO_2 Fuel Oxidation to Higher Oxidation States Following a Transportation Sabotage Incident

Several reference basis explosive threats to spent fuel shipping casks have been evaluated in past studies.(12)(13) The most damaging was a large shaped charge which caused a cask wall penetration represented by approximately a 6 inch (avg. dia.) hole. The corresponding hole through the fuel bundle was approximately 3 inches in diameter. Therefore, it can be postulated that following such a hypothetical explosive attack, atmospheric air could replace the cask cavity gas. Full scale experiments indicate that fuel rod damage could be as high as one half the fuel rods in a fuel assembly. Fuel disrupted by the attack could be deposited within the cask or in the environment external to the cask. This disrupted material would be expected to cool quickly to temperatures below which oxidation would not be a significant problem. Depending on heat transfer conditions, fuel material remaining within the damaged fuel rods could remain at temperatures close to the previously tabulated values. Following an incubation period, UO_2 fuel within these damaged rods could oxidize to U_3O_8 at a rate governed by rod temperature. This oxidation process could continue until all damaged fuel is fully oxidized. Most of the oxidized material would tend to remain in place in the damaged rods, although material near points of gross cladding failure could fall to the bottom of the shipping cask. This material, formed sometime subsequent to the hypothesized explosive attack, could only be released from the cask through the entrance hole created by the shaped charge. A significant release of this material would require proper geometric orientation of the entrance hole and a driving force, subsequent to the explosive attack, to move the oxidized material from the cask and fuel rods to the external environment. Based on this subjective evaluation, a substantial release pathway for fuel material oxidized subsequent to the sabotage event is not considered a significant possibility.

III. Regulatory Implications of UO_2 Oxidation to Higher Oxidation States

The regulatory implications of UO_2 oxidation, discussed in this section, are all based on the assumption that the oxidation process contributed to or could have contributed significantly to the radioactive material release which occurred in the BCL incident. The descriptions of the incident and limited experimental evidence supports this assumption. The information and experimental evidence, however, do not preclude the possibility that the procedures used to flood the cask cavity and cool the cask could have been a major cause of fuel and fission product release to the pool and cask handling area.

a) Normal Conditions of Transport*

Potential oxidation of UO_2 has limited significance during normal transportation. Because of the containment system, oxidation products would not be released from the cask. Any public health and safety impacts would be related to the potential effects of fission product transfer from fuel rods to the interior surfaces of

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* For accident conditions that fall within those described by 10 CFR 71, the containment of any oxidized fuel would be assured by the cask design requirements. Therefore, the arguments presented in this paragraph apply to hypothetical accident conditions of 10 CFR 71.

the shipping cask. By moving the radioactivity closer to the shipping cask walls the radiation level external to the cask could increase slightly. Fuel oxidation during transport is limited by available oxygen and the maximum amount of fuel oxidized to U_3O_8 has been calculated to be approximately one rod in a typical truck shipment or 8 rods in rail cask shipments. This amount of fuel material and any additional radioactive volatiles released from the failed rods would not significantly change the radiation levels typical to routine shipments of unfailed irradiated fuel.

b) Unloading and Loading Operations

The regulatory implications of UO_2 oxidation appear most pertinent to unloading operations involving grossly failed fuel assemblies. A conservative bounding estimate would indicate that failed fuel assemblies could account for 0.6% to 3% of the fuel assembly population. For significant oxidation to occur, the extent of cladding failure must be beyond that associated with minor failure modes such as stress corrosion cracking. Moreover, the failed assembly must be exposed to air and the fuel must reach temperatures high enough to promote oxidation. If significant oxidation does occur, the maximum potential health and safety impacts would be reasonably bounded by the occupational exposures and facility/equipment contamination experienced in the BCL incident and its related recovery operations.(14)(15) The rationale for representing the impacts of this BCL incident as upper bound values is as follows: (1) the fuel assembly was an atypical design containing grossly failed stainless steel clad fuel rods, (2) the extent of fuel rod damage was far greater than the average, (3) the thermal power of the assembly was greater than most past or expected near term spent fuel shipments,

(4) the assembly was shipped in an air environment without any added containment provisions, and (5) the receiving parties did not fully appreciate the potential hazards of the unloading operation.

c) Extremely Severe Transport Accidents

Past assessments(9)(10) have estimated both the risks associated with spent fuel transportation and the radiological consequences of extremely severe accidents involving spent fuel shipments in highly urbanized areas. This section will evaluate the impacts, if any, that UO_2 oxidation could have on these past assessments.

As discussed in section II.d., no significant cask containment violations are expected following a spent fuel shipping cask involvement in a severe transport accident. Nevertheless, some cask containment violations were assumed and the resulting fission product releases from both truck and rail shipping casks were evaluated.(9) The release from the truck cask included 1700 Ci of Kr^{85}, 0.022 Ci of I^{131}, and 200 Ci of volatile fission products. The rail cask release included 10,900 Ci of Kr^{85}, 0.138 Ci of I^{131} and 1280 Ci of volatile fission products. The consequence calculation in reference 10, assumed that the radioactivity release was 154 Ci of Co^{60}. This release represented a bounding estimate for the amount of activated corrosion products which could be present on the cask inner surfaces and the exterior surfaces of the fuel elements (known as reactor "crud").

A reassessment of these past release estimates, accounting for potential UO_2 fuel oxidation, must include two major considerations: (1) the effect of potential UO_2 oxidation prior to the accident and (2) the potential for UO_2 oxidation subsequent to the accident.

As indicated in section I. and II.b. the radioactivity release from the fuel assembly in the BCL incident has been estimated as 120 to 240 Ci excluding noble gases. This release occurred primarily during the flooding of the cask cavity. The washout caused by water and steam generated in this process should conservatively bound the expected release from the fuel rods to the cask cavity under most extremely severe accident conditions. Adjusting the activity release to account for the 150 day-cooled fuel evaluated in the past assessments would increase these values by a factor of about 2. In the BCL incident, however, some of this radioactivity remained in the cask following the release to the pool and high bay of the cask handling area. This circumstance suggests that a release from the cask under most extremely severe accident conditions would be bounded by an undefined fraction of the adjusted release estimate of 240 to 480 Ci. Depending on the accident scenario postulated, this release estimate could be considered as an upper bound replacement value for the 200 Ci of volatiles or 154 Ci of "crud" assumed in the past evaluations. However, if the 240 to 480 Ci release is added to past estimates, the calculated releases would change by less than a factor of 4. Because an accident involving a failed fuel assembly is less likely by at least a factor of 30 (3% incidence of failed fuel), the calculated change in overall risk would be extremely small.

Following an extremely severe accident event, air could conceivably replace the pre-accident cask environment, i.e., either an inert gas or air depleted of oxygen. The fuel oxidation process could then proceed as dictated by time and fuel temperature considerations. This post-accident oxidation could produce long term radioactive material releases which would be limited to volatile fission products that could diffuse from either irradiated unoxidized UO_2 or UO_3 which had undergone oxidation to U_3O_8 or higher oxides. Reference 16 indicates that at 500°C (932°F) diffusion release rates from oxidized fuel may be greater than from unoxidized fuel. Even if the fuel pin should be at this temperature level, the rate of release is not expected to be significant relative to the prompt releases already evaluated.

In conclusion, indications are that the oxidation of UO_2 to higher oxidation states during or following an extremely severe transportation accident does not contribute significantly to either the radiological consequences or risks calculated in previous evaluations. Based on tables 5-9, 5-12, and 5-13 in reference 9, the 0.000422 expected latent cancer fatalities from spent fuel shipments assuming the 1985 shipments model would not increase by more than 15%. The calculated consequences of extremely severe accidents in very high population density urban areas could increase from predictions of 1 latent cancer fatality to 4 latent cancer fatalities.

d) Sabotage

Recently completed research programs by both NRC and DOE have evaluated the chemical and physical form of radioactive material release resulting from specified explosive attacks against spent fuel shipments. Of the explosive threats evaluated, the shaped charge was determined to be the bounding explosive threat. The DOE sponsored program included a full scale test involving an obsolete, but structurally typical shipping cask containing a section of an unirradiated fuel assembly. The shaped charge penetrated the cask wall, damaged 50% of the fuel rods in the assembly, and removed 2.5Kg of fuel from the cask. The amount of potentially respirable irradiated fuel material was estimated by the DOE and NRC programs as 17 grams and 9 grams, respectively. The evaluations of the effects that UO_2 fuel oxidation can have on these results must consider the possibility of oxidation before and after the hypothesized sabotage event and the dispersion of oxidized fuel in the event.

From the discussion in section II.b., it has been calculated that shipment of a grossly failed fuel assembly in an air filled cask at temperatures sufficient to cause oxidation, could result in the equivalent of a single fuel rod of UO_2 being oxidized to U_3O_8.

The disruption of material caused by a shaped charge attack on a fuel rod containing powdered U_3O_8 is uncertain. Since the high pressure shock wave would be expected to dissipate more rapidly in U_3O_8 powder than in large solid fuel fragments, the formation of respirable particles through shock compression should be significantly less in oxidized vs. unoxidized fuel. If it is assumed that only the failed fuel rod is in the path of the shaped charge jet, so that the shaped charge jet penetrates this rod, approximately 40 grams of fuel material could be initially disrupted by the jet passage. The fraction of respirable sized material is unknown but a maximum value of one half the total should be conservative. The disruption of any further U_3O_8 caused by the detonation of the initiating explosive is also unknown, but would be limited to the amount of powdered oxide available at the time of the incident. The estimated likelihood of attack on a failed vs. unfailed fuel assembly would reflect the failed fuel occurrence rate of 0.6 to 3% estimated in section II.b.

Following a sabotage event, air could replace the pre-event cask atmosphere. Again the post-event oxidation process and potential for radioactive material release would be controlled by diffusion processes discussed in section III.c.

In conclusion, the estimated total impact of UO_2 fuel oxidation on the sabotage source term is, at most, a doubling of the larger solid material release from the cask to 5Kg and a respirable release bounded by the extremely severe accident releases.

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Findings

The major impact of irradiated fuel oxidation is the potential for occupational exposures during unloading operations. Three corrective actions are possible. First, proper unloading procedures could be devised to preclude a reoccurrence of the exposures associated with the BCL incident. Second, a non-oxidizing gas could be provided for dry spent fuel shipments which have temperatures sufficiently high to result in fuel oxidation (>150°C). This provision could result in a reduction in both the likelihood of occupational exposures and the potential for contamination incidents affecting the cask or unloading facilities and equipment. Finally, canning of known failed fuel assemblies would provide an additional barrier against the release of solid fuel particles and would minimize the contamination potential inherent to such fuel assemblies.

D.F. Ross
for Robert B. Minogue, Director
Office of Nuclear Regulatory Research

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