SOURCE TERM ANALYSIS IN HANDLING CANISTER-BASED SPENT NUCLEAR FUEL: PRELIMINARY DOSE ESTIMATE

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ABSTRACT

This paper evaluates the source term of radionuclide release from Commercial Spent Nuclear Fuel (CSNF) and its potential dose consequences during the handling. CSNF may be primarily packaged in a canister. Some uncanistered CSNF may be handled under water in a CSNF pool inside a wet handling building. This scoping calculation discusses radionuclide release from the drop or collision of CSNF assemblies for the assumed radionuclide release scenarios in the pool and the pool area, and outside the wet handling building. From the analog studies, the canister appears to be robust against drop or equivalent impact accidents. When the expected robust canister is assumed to be breached, radionuclide release may be limited through small surface opening area. The source term from oxidation and high burn up of CSNF is also discussed.

I. INTRODUCTION

There will be a pre-closure period for operations to receive, and handle and emplace CSNF and high-level waste into a repository prior to permanent closure (pre-closure period). Most of the CSNF assemblies may be packaged in canisters prior to transportation to minimize radionuclide release from any accident associated with handling bare CSNF assemblies. Some of the CSNF that may be in other canisters may be repackaged in canisters in a CSNF pool in a wet handling building. With the canister, the geological operations area (GROA) no longer requires a dry transfer building (i.e., a hot cell) to handle bare assemblies. In this paper, we present analysis results of radionuclide release and dose assessments associated with CSNF handling in the GROA. This paper will first present the results associated with the dry transfer building for bare assemblies. Then, the results will be interpreted with respect to the design with the canister and the wet handling building. The scope of this work emphasizes source term in the dose (i.e., consequence) assessment.

Several event sequences may lead to radionuclide release from the waste handling building. This paper considered event sequences from these three event sequence categories in which radionuclides may be released into the atmosphere, as presented by Kamas, et al. (2006). An emphasis was placed on the source term. These analyses and assessments are scoping in nature and are not intended to represent staff positions on methods and assumptions for consequence assessments. They represent an analytical testing methodology.

Some CSNF rods shipped in canisters may have breached cladding. If any accident (e.g., drop) leads to the canister breach, CSNF pellets in rods with breached cladding may oxidize in air, as the temperature inside the breached canister could initially rise upon the air inflow. Higher oxidation, which converts UO_2 to U_3O_8, may lead to pellet pulverization and cladding unzipping [Ahn, 1996], potentially increasing the radionuclide release. High burnup CSNF is also included in this assessment because the rim structure can increase the radionuclide release too.

The analyses and assessments made in this paper for the canister include: (i) integrity of the canister under accident conditions; (ii) release in air from the breach of the canister; (iii) release in the pool; (iv) release from transfer cask/canister in the pool area under accident conditions; and (v) release in normally uncontaminated adjacent areas. For the analyses described in this paper, it is assumed that canisters may fail during a drop/collision event and the overpacks do not provide any confinement of radionuclides.

II. DOSE ASSESSMEMT METHOD

The dose assessment utilized a method developed by the Center for Nuclear Waste Regulatory Analyses [Dasgupta, et al., 2002]. The method is based on the MELCOR code [Gauntt, et al., 2000] and the RSAC code [Shonka Research Associates, Inc., 1993]. The MELCOR code allows the user to calculate the building discharge fractions of radionuclides and the RSAC code performs the consequence analysis of an atmospheric radionuclide release. The MELCOR code models the breach of CSNF assemblies as an instantaneous release of radionuclides (i.e., release fraction) into the room air of the waste handling building. The code also estimates the fractional release of radionuclides transported through the waste handling building and its ventilation system, and to the atmosphere. The RSAC code models the radionuclide dispersion in the atmosphere to calculate radiological consequences to an off-site member or worker from an atmospheric release of radionuclides.

These two codes were used to calculate doses for a range of event sequences based upon the source term. The outdoor worker dose is estimated by the maximum onsite dose. This maximum dose was assumed to occur in the cavity zone. The cavity zone is an area adjacent to a building from which radionuclide release may occur, in which the recirculation of contaminated air may lead to increased dose (relative to the Gaussian plume model). The worker dose consequence computes ground surface, submersion, and inhalation dose components. The indoor worker dose is estimated based on the outdoor worker dose, considering event sequences associated with a hypothetical case of no building.

The downwind dose to the public is estimated by the RSAC code. The shortest distance from the CSNF transfer facility to a site boundary is approximately 11 kilometers (km, 6.84 miles). The code computes ground surface, submersion, inhalation, and ingestion doses. Ingestion dose is calculated for 1 year, assuming that the receptor eats some locally produced food that is grown at the site boundary of 11 km from release. Most farming activity in the area takes place about 30 km (18.65 miles) away. This paper presents the total effective dose equivalent.

III. CANISTER INTEGRITY, AND RETENTION OF RADIONUCLIDE IN A BREACHED CANISTER

Canisters will be weld-closed for radionuclide containment [DOE, 2008]. Therefore, it is important to assess the structural integrity of the canister under various normal operation and accident conditions. To obtain insights on the structural integrity of the canister, staffs of NRC and CNWRA have conducted a literature review of the structural integrity of analog canisters. This review compiles literature evaluating the structural behavior of the DOE standard canister of DOE SNF [Snow, et al., 2000], Idaho Spent Fuel Project canisters [Snow and Morton, 2003], multi-canister overpacks [Snow, 2004], and HLW Canisters [Peterson, 1985]. Analog studies suggest that the canister is robust to keep radionuclides contained against drop or equivalent impact accidents during operations. Details of the analog studies will be presented elsewhere. Even if the canister is breached, radionuclide release may be limited through small surface opening areas.

Finite element studies of canister designs for HLW disposal were evaluated. The 610- and 1,676-mm (24- and 66-in) dished bottom canisters have an energy-absorbing skirt attached to the bottom, and one 1,676-mm (66-in) canister had a flat bottom. The canisters with the skirt had maximum surface strains of 65 to 48 percent for the 610-mm (24-in) and 1,676-mm (66-in) dished bottom, respectively. These strains are below the 60- to 80-percent maximum strain. Specifications for the canister are a flat bottom; a diameter of approximately 1,676 mm (66 in); and a length of 5.5 m (18 ft) as specified in the Revision 1 [DOE, 2008]. The 1,676-mm (66-in) flat bottom canister (with no skirt) had a maximum surface strain of 72 percent, which is in between the 60 to 80 percent maximum strain. Mid-surface strains for all canisters averaged 30 percent, which is well below failure; therefore, no surface cracks would propagate through the thickness.

The finite element method simulations for these canisters have demonstrated that variations in the types and designs of the canisters can withstand dynamic loads due to drop events of 9 m (30 ft) or below. These canisters were evaluated in terms of changes in geometry, maximum equivalent plastic strains, and their ability to pass a leak test. The standardized canister, for example, utilized longitudinally welded 316L stainless steel pipe as opposed to seamless pipe, and there are welds where the skirt joins the canister body. Using this type of pipe provided very limited experimental testing of the welds. The mechanical properties of the welds and the factors that may affect the weld were discussed. The fracture toughness of welds was discussed, including a method used to measure toughness - namely the Charpy V-Notch impact energy, which may provide estimates of the weld open surface area. Generally, the tests at 9 m (30 ft) are conservative with respect to the surface operational conditions (about 0.3 m, 1 ft) at the GROA. The canister is expected to be handled in an overpack which provides some protection against impact.

Radionuclide Release from Failed Canister

The retention factor of radionuclide release (or mitigation factor) of a failed canister can be determined primarily by what fraction of the total geometric surface area of canister would be open to air upon the canister failure. The surface area opening will in turn depend on the impact energy from drop or collision of canisters. For example, Sprung, et al. [2000] indicates a retention factor of radionuclide particulates, 0.02, for an impact speed of 27 m/s (60 mph) pressurized to 5 atmospheres (506.6 kPa). For a drop case, this impact speed is determined primarily by the drop height. Alternatively, the drop height or the impact speed gives the impact energy onto the canister.

The Charpy V-Notch impact test is used to determine the energy input required to fracture the V-Notched rectangular metal specimen by applying known impact energy by force. The V-Notch gives an identifiable fracture path. The Charpy V-Notch impact energy suggests how the open surface area of the canister could be estimated upon impact. The American Standard Testing and Materials accepted Charpy V-Notch specimen has a normal size of $1 \times 1 \times 5.5$ centimeter (cm, $0.4 \times 0.4 \times 2.2$ inch) [ASTM, 2007] with a 0.2-cm (0.08-in) notch. Assuming the total impact energy is used to create the surface area of the notched ligament (i.e., the specimen thickness minus the notch depth), a correlation of the impact energy input and the open surface area can be estimated. For example, 100 J (23.90 cal) of Charpy V-Notch impact energy may create a fraction of ~0.03 of the surface area. The specific impact energy in this case would be higher than normally expected from the canister drop test at a 7-m (23-ft) height. This type of estimate can be normally used for ductile materials such as 316L stainless steel. The background information for the estimate of the specific impact energy for a canister could be obtained from the references [Peterson, et al., 1985; Kamas, et al., 2006] which also presented some exercise results of pre-closure safety analyses. The impact energy is the canister weight times gravity times drop height, given the surface conditions of the impact. Energy dissipated by heat is normally negligible.

III. RADIONUCLIDE RELEASE FROM DRY TRANSFER

The atmospheric release of radionuclides is assessed from a hypothetical accident during the dry transfer of bare assemblies. The assessment made can be reinterpreted with respect to the canister design in Sections IV to VIII. One assembly from a pressurized water reactor (PWR) is used, unless otherwise specified in the release assessment. The inventory of radionuclides in the PWR assembly was determined with a loading of 0.43 metric tons (0.47 tons) of uranium, a burnup of 49 GWd/MTU (4.4×10^{12} Btu/ton), an enrichment of 4.0 percent, and a decay time of 25 years. It is assumed that this PWR assembly bounds other types of CSNF in the dose assessment. To assess the actual risk, this dose per assembly needs to be multiplied by the assemblies at risk from the event sequence assessment [Kamas, et al., 2006].

Bare assemblies would be handled in a hot cell. The hot cell uses negative air-pressure and HEPA filters. The connected room is a normally uncontaminated adjacent area where any leakage from the work area will release radionuclides into the connected room and result in a dose to the indoor worker. Information from the assessment of hot cell operations is used. Under normal and category 1 accident conditions, HEPA filters will be operable. The outdoor worker dose at the site within 11 km and the public dose beyond 11 km were calculated. Under category 2 accident conditions, the HEPA filter was assumed to be inoperable and only the public dose beyond 11 km was calculated.

The source term for a given event sequence is the product of the following:

- release fraction
- material at risk
- damage ratio
- leak path factors
- canister retention factor (sections III and V on canister integrity)

The release fraction is the fraction of the total radionuclide inventory released under normal and accident conditions. Table 1 shows the analysis results of release fraction from an impact on assemblies [Kamas, et al., 2006]. This impact simulates the drop/collision of assemblies (i.e., drop) and the impact from a seismic event. The release fraction for assembly drop/collision assumes an impact speed of 13 m/s (30 mph, equivalent to 9 m (29.5 ft) drop) and the seismic event assumes an impact speed of 27 m/s (60 mph, equivalent to 36 m (118.1 ft) drop). The impact speed or the drop height is in turn correlated to the impact input energy. The 36 m height is to simulate seismic impacts that may cause building degradation and its following impacts on the assemblies. The release fraction on oxidation increased by (i) dividing the UO_2 matrix of a diameter of ~0.1 cm into grains/subgrains of a diameter of ~10^{-3} cm, and (ii) more vaporization at higher temperatures. In the high burnup SNF, the rim structure may further divide the UO_2 matrix into subgrains on oxidation. The release fraction of cruds was from references [Sprung, et al., 2000; NRC, 1997; Ahn, 1996].

Table 1 Release Fractions for Spent Nuclear Fuel [Kamas, et al, 2006]

	Release Fraction		
Material	Dropped Assembly	Fuel Oxi-dation	Seismic Event
Tritium	0.3	0.3	0.3
Noble Gases	0.3	0.3	0.3
Iodine	0.3	0.3	0.3
Crud	0.15	0.15	0.15
Ruthenium	0.0002	0.002	0.0002
Cesium	0.0002	0.002	0.0002
Strontium	2.0×10^{-6}	1.2×10^{-3}	8.5×10^{-6}
Fuel Fines	2.0×10^{-6}	1.2×10^{-3}	8.5×10^{-6}

The material at risk depends on the event sequence. The damage ratio is the fraction of the material at risk that is damaged during the event sequence; and leak path factor is the product of the building discharge fraction and mitigation factor of the HEPA filter. The building discharge fraction accounts for deposition and agglomeration as radionuclides travel through the building ventilation system. The mitigation factor of the HEPA filter accounts for the presence of high efficiency particulate air filters that are capable of removing over 99 percent of particulate materials from the air prior to atmosphere release.

Figure 1 is the dose as a function of distance from a dry transfer hot cell area, under the inoperable HEPA condition, and with and without building (as a sensitivity study without building protection). The release fraction was for the dropped assembly in Table 1. The leak path factor of the building discharge fraction is still operable in the calculation. The public doses are similarly obtained at 11 km boundary. Figure 2 shows example calculations results of these doses by varying leak path factor values from 10^{-7} to 10^{-3}. When the assembly is not oxidized, the dose does not change with varying leak path factor. However, if CSNF oxidation (and/or high burnup) occurs upon the canister breach, the dose could go up to a factor of 35 [Kamas, et al., 2006]. The oxidation (and/or high burnup) could pulverize the assembly matrix into a respirable size by volume expansion or rim structure, effectively increasing release fraction.

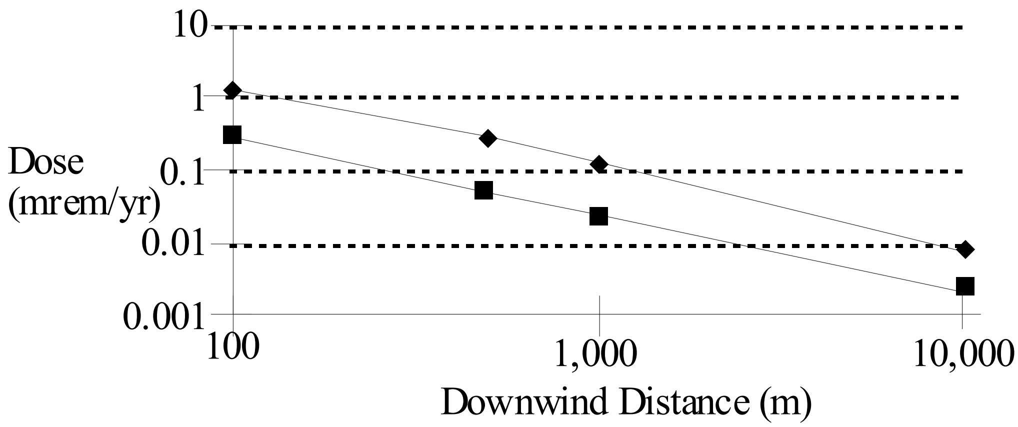

Figure 1 Building size sensitivity in dose from CSNF drop. Building size from Dasgupta, et al [2002]. Dose within the site boundary, 11 km from the release point. Legend: ◆: no building. ■: small building (mrem is milirem)

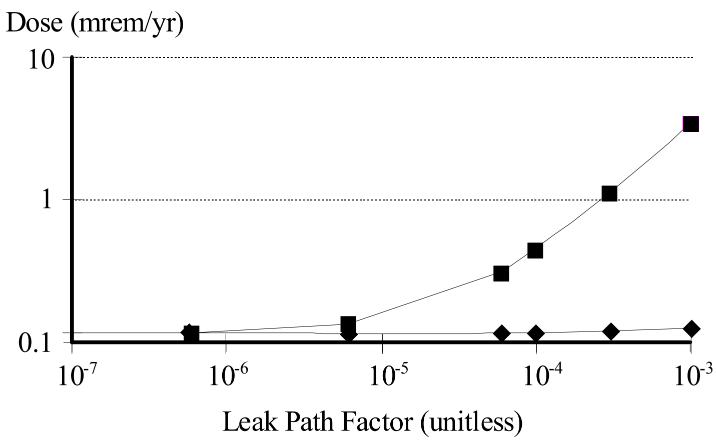

Figure 2 Dose at the site boundary, 11 km from release: lower curve for assembly drop/collision; and upper curve for fuel oxidation/pulverization

Using the release fraction associated with additional impact events, an example calculation was made for the public dose consequence for release fractions associated with 13 m/s (30 mph) and 27 m/s (60 mph) impact on assemblies. Equivalent speeds would be reached at drop heights of 9 m (29.5 ft) and 36 m (118.1 ft) respectively. Figure 3 shows the increase of respirable mass (i.e., equivalent to release fraction) from impact by increasing various impact energy. This drop height varied to simulate the impact of building degradation under category 2 seismic conditions. There was about a factor of 3 increases in the maximum off-site dose with 100 assemblies damaged in ground release without leak path factor.

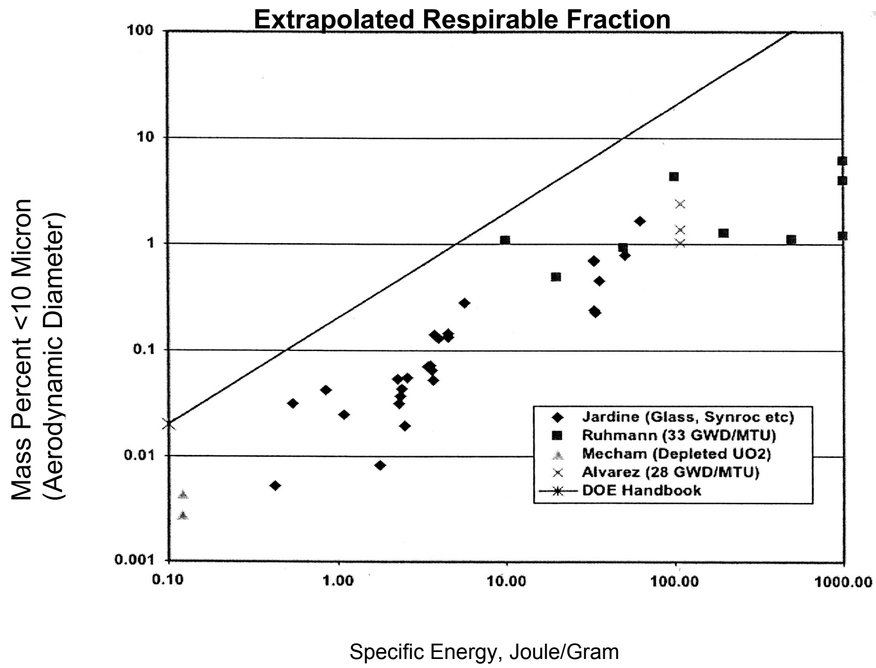

Figure 3. Comparison of the DOE handbook Respirable Fraction Equation to Experimental Values of the Specific Energy Input into the Brittle Material. Legend: ◆: Jardine (Glass, Synroc, etc). ■: Ruhmann (33 GWD/MTU). ▲: Mecham (depleted UO_2). ✕: Alvarez (28 GWD/MTU). —: DOE Handbook. Source: NRC, 2007

IV. HANDLING OF ASSEMBLIES IN A WET HANDLING BUILDING

Pool

Under normal conditions, radionuclides will be released in the pool due to initially failed cladding (maximum about 1 percent failure from older SNF). An assessment was made to determine the amount of radionuclides that could be released in the pool under normal conditions. Radionuclide release data in the SNF pool were reviewed [Johnson, Jr., 1997]. An example analysis from the literature data is presented in Table 2, showing release fractions of radionuclides at various times in the pool. The release fractions shown here are more than three orders of magnitude smaller than those that were estimated under drop conditions (Table 1). The release from any accident in the pool can be bounded by the release in the pool area (as discussed below).

Table 2 CSNF Pool Source Term.

Time in a Spent Fuel Pool	Release Fraction *
120 days	1.82×10^{-9}
1 year	4.32×10^{-9}
10 years	2.52×10^{-8}

* Current data at the Gen Basins site with high integrity fuel

Pool Area in a Wet Handling Building

An exercise was performed to assess the worker dose under accident conditions in the pool area. The scenario that was assessed assumed that assemblies were handled in a negative air-pressured work area. The heating, ventilation, and air-conditioning (HVAC) system has a HEPA filter. Section III assessed the dose for a hot cell with HEPA filters inoperable in the previous design. This hot cell dose assessment is the same for the operations of the wet handling building where there is no confinement of assembly handling. In this assessment, the release fraction is from the drop of assemblies and other leak path factors such as building discharge factor were from default values in the Tool, as used in Section III. Radionuclide releases are insensitive to these leak path factors if there is no deposit or agglomeration of particulates from the oxidation. The dose is mainly from gaseous release because the canister during the transfer is filled with water. The oxidation is not expected to occur in the canister filled with water. The particulates from the rim will be in the water too. The gaseous radionuclide releases will not be affected by the building discharge factor (i.e., leak path factor). Therefore, the dose calculated from the outdoor work area without filtering in the hot cell of the previous design simulates the dose to the worker in the wet handling building. Figure 4 shows a simulated dose curve from 1 meter (m) from the hot cell under an accident of dropping one assembly inside the hot cell. The dose at 1 m is regarded as the worker dose in the wet handling building in the new design because there was no dispersion within 10 m as shown in the figure. As expected, the calculated dose is similar to that in Figure 1 of no building case in the release from the hot cell (e.g., dose at 100 m).

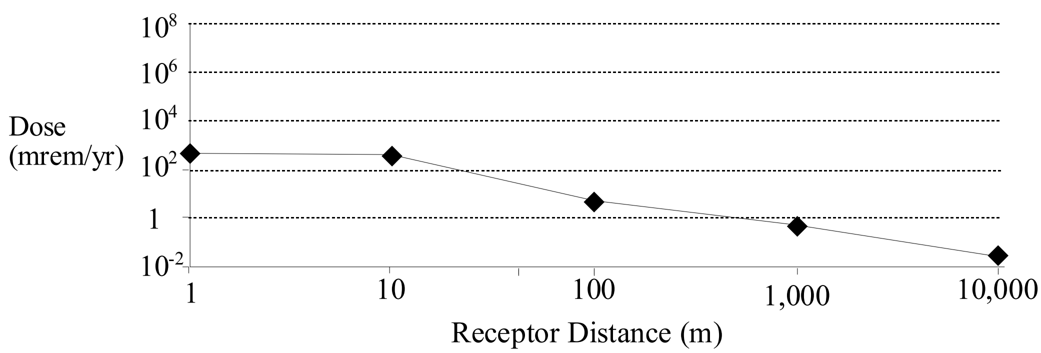

Figure 4. Dose from 1 m from the hot cell under drop condition; simulate dose to the worker in the pool room

In this calculation, 24,000 m^3 is assumed to be the mixing air volume in the pool room. If this volume is further confined within a portion of the work area height, the dose will increase accordingly. On the other hand, the smaller confined volume will be directed to the ventilation fan. In this case, the likelihood of indoor worker standing in that direction will be inversely proportional to the smaller confined volume. Therefore, 24,000 m^3 is a reasonable approximate value in the dose calculation. Lastly, an additional conservative assumption was that all CSNF rods will be subject to cladding failure under accident conditions. In the analysis of cladding integrity under drop conditions, less than 1 percent failure rate was reported in the presence of impact limiters [Sanders, et al., 1992]. This will be especially the case at lower drop heights.

V. BREACHED CANISTER RETENTION FACTOR OF RADIONUCLIDE RELEASE

The retention factor in the release from the failed canister was defined in section III. The canister retention factor needs to be considered. The current version of NRC and CNWRA's PCSA Tool does not include the canister retention factor. However, the results of the exercise of the MELCOR and RSAC codes are discussed qualitatively with respect to the canister retention factor. For example, the building discharge factor is ineffective inside the building, i.e., effectively LPF = 1. If any residual building discharge factor was credited in the dose calculation inside the building, it can be compensated by the canister retention factor for no building discharge factor. If the dose did not consider the canister retention factor based on the old design, the canister retention factor will further correct the dose in the new design.

VI. RELEASE FROM ACCIDENTS OUTSIDE THE WET HANDLING BUILDING

Assuming that the canister has failed and overpack does not provide any confinement, the outdoor worker dose will be the dose within 11 km (Figure 4) and the public dose will be the dose at 11 km (Figure 1). The case of no building and HEPA filters inoperable is the same as that of canister drop/collision outside the building. The canister retention factor will further lower the dose. If the initial temperature upon the canister failure rises, the CSNF matrix may oxidize and will result in the in the increase of the outdoor worker and public doses more than by a factor of 100 [Kamas, et al., 2006].

VII. NORMALLY UNCONTAMINATED ADJACENT AREAS

Under the same conditions of the assessment made for the dose in the SNF pool area, the dose in adjacent areas, that are normally uncontaminated, was assessed. Figure 5 shows various doses calculated at the leak rate from 0.05 to 1.9 m^3/second.

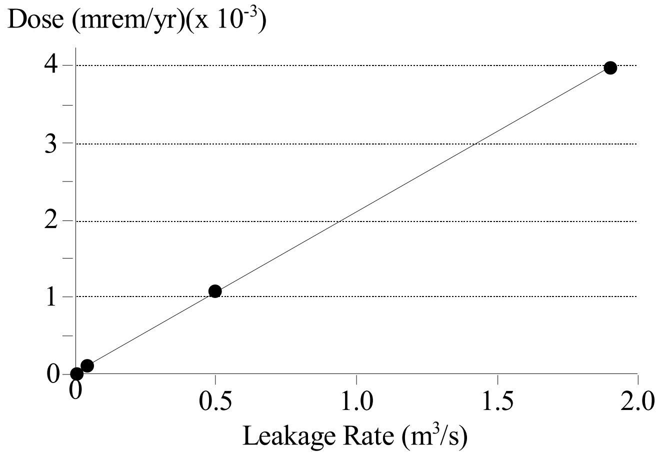

Figure 5. Dose in the normally uncontaminated area adjacent to the building

VIII. HIGH BURNUP FUEL

High burnup CSNF is defined as CSNF with burnup values exceeding 45 GWd/MTU (4.0×10^{12} Btu/ton). When the burnup exceeds 60–65 GWd/MTU (5.4×10^{12} – 5.8×10^{12} Btu/ton) in high burnup CSNF with Zircaloy cladding, the properties of the irradiated UO_2 matrix (i.e., the matrix) and cladding could be significantly altered [Ahn and Jain, 2005]. The matrix will form a fine grained rim structure on the surface. Typically, the grain size is 0.1 to 0.3 microm in diameter compared to the matrix grain size of about 10 micrometers. Also, the rim structure is generally known to be enriched with plutonium [Johnson, et al., 2005]. Cladding might become embrittled by potential hydride reorientation and delayed-hydride cracking [Ahn, et al., 2007].

The release fraction of actinide particulates might increase within the rim structure, because the rim is enriched with plutonium and is fine grained. Intergranular fracture of the rim upon accidental impact might increase the fraction of respirable particulates (less than ~10 micrometer in diameter), along with the contribution of the higher concentration of plutonium. However, it is likely that the release fraction values used in this report (e.g., Table 1) are conservative. Also, the rim might be more ductile than expected because a new microstructure might form. Therefore, the current release fraction might bound the release fraction of high burnup CSNF with the rim structure. More detailed data on the impact properties of the rim is needed to reduce uncertainties associated with the release fraction of particulates from the rim under impact accident conditions.

In this paper, a detailed analysis is conducted to determine the potential increase in the release fraction when the cladding becomes embrittled due to high burnup. Under impact accident conditions (e.g., drop or collision), an assembly could become rubbles; composed of small pieces of clad segments. In each segment, cladding will partially protect the release of particulates (i.e., filtering effects), while the gap between the cladding and the matrix keeps some residual gas pressure. When the cladding becomes embrittled, the partial cladding protection will disappear and there will be no residual gas pressure in the gap. Mathematically, the filtering efficiency of particulates is described by Reynolds number in the equations below.

$$\eta_R = 16R^{\left\{2-\frac{\mathrm{Re}}{\left(\mathrm{Re}^{1/3}+1\right)^3}\right\}}$$

Where η_R = intercept removal efficiency
Re = Reynolds number

$$R = \frac{d_p}{d_g}$$

Where d_p = particle diameter
d_g = granular diameter

As Re decreases, the air flow becomes less turbulent decreasing the filtering efficiency. In order to maintain a 99 percent filtering efficiency, the length of the cladding segment would need to increase. Table 3 below shows the calculated filtering efficiency for (1) previously used Re with partial cladding protection [Sanders, et al., 1992], Re = 77 and 311, and (2) conservative values for no cladding protection, Re = 10, 20. The conservative values were calculated in the definition of Re, using ambient pressure (that was converted to energy and velocity) without partial cladding protection. As seen in the Table 3, the changes in the filtering efficiency were minor even without cladding. To confirm the analysis results here, some further experiments with high burnup CSNF might be needed.

Table 3 Release Fraction with High Burnup Cladding.

Impact Speed (MPH)	Accepted Release Fraction	Calculated (conservative) Release Fraction
No impact	3.9×10^{-7}	3.9×10^{-7}
30	2.2×10^{-6}	2.5×10^{-6}
60	8.5×10^{-6}	9.7×10^{-6}
90	1.9×10^{-5}	2.2×10^{-5}
120	3.4×10^{-5}	3.9×10^{-5}

VIII. SUMMARY

This assessment indicates that radionuclide release from the drop or collision of a CSNF assembly appears to be small for the assumed release scenarios in the pool and pool area, and the outside wet handling building. In the pool, the release fraction is very low, $10^{-9} – 10^{-8}$, compared with that from drop or collision of bare CSNF. In the pool area, the worker dose from the drop of cask/canister containing bare assemblies in the wet handling building is as low as that from the drop of bare assemblies in the open system. From the analog studies, the canister appears to be robust against drop or equivalent accident. The analog canisters are robust against impact from drop at the height as high as 9 m. It is recognized that the radionuclide release could be restricted through small surface opening areas, even if the expected robust canister is assumed to be breached. There is potential for increased dose from the oxidation of the CSNF matrix. However, this could be mitigated by the robust canister. For high burnup CSNF greater than 45 MWd/MTU, the release fraction may not change significantly compared with lower burnup CSNF than 45 MWd/MTU, because the rim structure may not be embrittled and the cladding embrittlment may not change the release flow pattern.

DISCLAIMER

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