Establishing Risk-Informed Non-Fixed Surface Contamination Limits for Spent Fuel Transportation Casks

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ABSTRACT

Current limits for non-fixed surface contamination on spent fuel casks are the same as for other transportation package types: 4 Bq cm^{-2} [110 pCi cm^{-2}] for beta and gamma emitters and low toxicity alpha emitters, and 0.4 Bq cm^{-2} [11 pCi cm^{-2}] for all other alpha emitters. These limits have been called into question in recent years, particularly with regard to spent fuel casks, because they are based on conditions, practices, and radiation dosimetry of the 1950s; they are not optimized with regard to the total doses received from spent fuel casks by workers and members of the public; and the radionuclides considered in their derivation do not include the generally dominant sources of activity on cask surfaces. In 2001 the International Atomic Energy Agency (IAEA) initiated a Coordinated Research Project (CRP) to reexamine limits for non-fixed contamination on all types of packages. This paper provides an update on the results of the CRP as they apply to limits for contamination on spent fuel casks. Because the CRP's proposed limits are radionuclide-specific, much of the discussion is concerned with identification of radionuclides likely to be present on cask surfaces.

I.0. INTRODUCTION

Current limits for non-fixed surface contamination on spent fuel casks are the same as for other transportation package types: 4 Bq cm^{-2} [110 pCi cm^{-2}] for beta and gamma emitters and low toxicity alpha emitters, and 0.4 Bq cm^{-2} [11 pCi cm^{-2}] for all other alpha emitters. These limits have been called into question in recent years, particularly with regard to spent fuel casks, for several reasons. For example, they are based on conditions, practices, and radiation dosimetry of the 1950s; they are not optimized with regard to the total doses received from spent fuel casks by workers and members of the public; and the radionuclides considered in their derivation (Pu-239 and Sr-90) do not include the generally dominant sources of activity on cask surfaces.

In 2001 the International Atomic Energy Agency (IAEA) initiated a Coordinated Research Project (CRP) to reexamine limits for non-fixed contamination on all types of packages. The CRP reviewed the scientific basis for current surface contamination limits and developed a model to estimate doses to workers and members of the public due to package surface contamination. The "CRP Basic Model" is described in IAEA TECDOC-1449, "Radiological aspects of non-fixed contamination of packages and conveyances" [1].

This paper provides an update on the results of the CRP and implications of the CRP Basic Model with regard to limits for contamination on spent fuel casks. The CRP Basic Model and some general implications of this model are summarized in Section 2.0. Because the CRP's proposed limits are radionuclide-specific, much of the discussion in subsequent sections is concerned with characterization of radionuclides typically found on cask surfaces. This characterization is based on typical sources of the contamination (Section 3.0) as well as reported measurements of radionuclides in spent fuel pool water and on cask surfaces (Section 4.0). The characterization of dominant radionuclides on cask surfaces is used, together with radionuclide-specific dose predictions of the CRP Basic Model and proposed dose limits (Reference Doses, Section 5.0), to derive proposed surface contamination limits for spent fuel cask surfaces (Section 6.0).

2.0. SUMMARY OF THE CRP BASIC MODEL

The CRP Basic Model and its predictions of maximal doses to workers and members of the public are summarized in IAEA-TECDOC-1449 [1]. Four categories of packages are considered: small manually handled packages (e.g., medical isotopes); small remotely handled packages (e.g., waste or UO2 drums moved with fork lifts); large remotely handled packages (e.g., standard ISO freight containers); and fuel flasks. Persons are assumed to be exposed to removable surface contamination on packages during the following transport steps: final inspection (workers); loading onto conveyance (workers); movement on vehicles (workers and public); transfers during transport (workers and public); and receiving and unloading (workers and public). Each transfer step is further divided by subtask, package type, worker type, and (if applicable) sites of public exposure. The workers addressed include those involved in package preparation, fork-lift drivers, conveyance drivers, transfer site workers, loading operators, health physics workers, and unloading workers.

For each radionuclide and each occupational or public exposure situation considered, the CRP estimated doses to workers and members of the public for each plausible mode of intake or external exposure, e.g., deposition on the skin of the hands and face, ingestion from the hands after touching a package, inhalation of surface contaminants that become airborne, external exposure from contamination on the package surface, or external exposure from contamination transferred from the package surface to the ground. Cautious assumptions were made concerning exposure times or other model parameters in cases where there was inadequate information to determine realistic values.

Maximal annual dose per unit concentration on surfaces predicted by the CRP Basic Model depend strongly on the radionuclide, with values spanning about seven orders of magnitude for workers and eight orders of magnitude for members of the public. The most restrictive values are for long-lived alpha emitters, and the least restrictive values are for weak beta emitters. The maximal doses per unit surface contamination do not vary greatly with package type. The model predicts that the maximum worker dose is at least two orders of magnitude greater than maximum public dose for any package type or surface contaminant.

For spent fuel casks, the CRP concluded that the optimal surface contamination limits, meaning the limits that result in the lowest total dose to workers and members of the public, are higher than the current limits. This is because the doses to workers increase with decreasing surface contamination limits due to increased cleaning time.

3.0. SOURCES OF RADIOACTIVITY ON SPENT FUEL CASK SURFACES

The radionuclide composition of cask surface contamination depends on the mixture of radionuclides present in the spent fuel pool water, although different radionuclides in the pool water may not bind to cask surfaces to the same extent. Radionuclides can enter the pool water by various pathways including: (1) mixing of reactor coolant and pool water during fuel discharges (in light water reactors), (2) desorption of soluble forms of radionuclides and spallation of particulates from fuel assembly surfaces, and (3) storage of fuel assemblies with reactor-induced defects [3]. Radionuclides that reach the pool water by these pathways are primarily activation products and fission products.

Activation products that reach the pool originate mainly inside the reactor coolant system [3]. Corrosion products formed inside the coolant system and circulating with the coolant are adsorbed on the fuel rod surfaces as crud deposits or oxide films and become radioactive when they are irradiated with neutrons [3,4]. They are transferred to the spent fuel pool water along with the fuel. Activated corrosion products in the reactor coolant may include Co-58, Fe-55, Fe-59, Mn-54, Cr-51, Sb-124, Sb-125, and Zr–95.

Cobalt-60, produced by neutron irradiation of Co-59 in metals, generally represents a significant portion of the activity in reactor coolant systems. This is illustrated in Table 1 for light water reactors but is also true of other types of reactors, including heavy water reactors (HWRs) and Magnox Gas Cooled Reactors (GCRs) [4].

Fission products in the fuel can come into contact with the reactor coolant if there is a cladding failure [3]. Fission products sometimes found in relatively high concentrations in reactor coolant systems include Cs-137, Cs-134, and I-131 (Table 1). Most gaseous fission products are released from the fuel inside the reactor and removed in the gas collection system. Soluble fission products that circulate in the coolant system may be transported to the spent fuel pool by adsorption on the external surfaces of the fuel assemblies. The storage pool water continues to leach fission products from the stored spent fuel. The leaching rate varies from one radionuclide to another, with cesium isotopes having a particularly high leaching rate.

Table 1. Radionuclides typically representing most of the activity in reactor cooling systems of light water reactors.

		% of total activity		
Nuclide	Half-life	Primary coolant, Reference BWR[a]	Primary coolant, Reference PWR[a]	Surfaces of PWRs and BWRs[b]
Fe-55	2.74 y	37	2.2	43
Co-60	5.27 y	29	7.5	32
Cs-137	30.2 y	18	75	--
Cr-51	27.7 d	5.3	0.07	--
Cs-134	2.06 y	0.88	12	--
Co-58	70.9 d	0.56	0.75	--
Ni-63	100 y	0.34	--	25
I-131	8 d	1.5	1.4	--
Transuranics[c]	c	--	--	3×10^{-6}

[a]Based on tabulations in [5].
[b]Based on samples from 11 pressurized water reactors (PWRs) and >8 boiling water reactors (BWRs). Surface samples from inside stainless steel piping, a main coolant system check valve, and fuel element hardware [6].
[c]Pu-238 ($T_{1/2}$ = 87.7 y), Pu-239 (24,000 y), Pu-240 (6564 y), Cm-244 (18.1 y).

According to NUREG-1714 [7], contamination on the exterior surface of a spent fuel canister is most likely to come from the radionuclides in particulate material suspended in the spent fuel pool water. The authors contended that most of the activity in suspended particles in the pool at the time of loading would be the long half-life corrosion products from spent nuclear fuel surfaces that might dislodge during movement of the spent fuel. The most prominent radionuclides in particulate form were projected to be the activation products Co-60, Co-58, Fe-55, Fe-59, Mn-54, Cr-51, and Zn-65.

Contamination at an aging power plant generally is widespread on surfaces, particularly near the fuel discharging equipment, the processing and storage facilities for radioactive effluents and wastes, and the storage pools [4]. It is conceivable that radioactive contamination could reach the spent fuel pools in significant quantities by pathways other than the three scenarios summarized earlier (mixing of reactor coolant and pool water, release from fuel assembly surfaces into the pool water, and leakage through defects in fuel assemblies). Based on reported inventories of contaminant radionuclides at nuclear power plants that have operated for at least a few years [4-6], any of the following radionuclides conceivably could also be important contributors to total activity on some spent fuel casks, in addition to activation and fission products already mentioned: H-3, P-32, Ni-59, Ni-63, Sr-89, Sr-90, Zr-95, Nb-93m, Nb-95, Ag-108m, Cs-136, I-131, Ba-140, and Eu-152. Each of these radionuclides represented at least 0.1% of total activity estimated either for a total facility at shutdown or for a reactor coolant system during operation, for at least one reactor site. Data for different types of reactors in different countries were

considered, e.g., light water reactors in the U.S. and Italy, heavy water reactors in Canada, and Magnox GCRs in the U.K.

Alpha-emitting actinides formed in the fuel can contaminate the pool water to some extent through failed cladding, but the rate of escape of these radionuclides from the fuel is low compared with fission products such as cesium. Thus, the contribution of actinides to the total activity in the coolant system or to contamination at other parts of a power plant generally is small, except possibly in cases where they have been dispersed due to a reactor accident. The radionuclides expected to be the most important alpha emitters on surfaces at power plants are ^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{241}Am, ^{242}Cm, ^{244}Cm, ^{232}U, ^{233}U, ^{234}U, ^{235}U, ^{236}U, and ^{238}U [4, 6, 8].

4.0. REPORTED RADIONUCLIDES IN SPENT FUEL POOLS OR ON FUEL CASKS

Radionuclides reported to be present in moderate to high concentrations in at least some spent fuel pools include Cr-51, Mn-54, Co-58, Co-60, Zn-65, Ag-110m, Sb-124, Sb-125, Cs-134, and Cs-137 [3, 9-15]. Their relative activities depend on a variety of factors including the reactor type, the storage times of the spent fuel assemblies, the efficiency of the pool's purification system, and the frequency and severity of cladding failures. Relative activities of these radionuclides determined in one spent fuel pool are given in Table 2 [9].

Table 2. Radionuclides in spent fuel pool water at the Forsmark 2 facility in Sweden [9].

Radionuclide	% of total reported activity
Co-60	37.1
Sb-125	34.8
Sb-124	9.7
Cs-137	5.7
Ag-110m	4.2
Zn-65	4.1
Cs-134	2.2
Cr-51	1.7
Mn-54	0.3
Co-58	0.2

Based on reported measurements of spent fuel pool water, Co-60 appears to represent a major portion of the activity in the pool in most cases [10, 13, 14, 16]. Cesium-137 or Cs-134 may represent a significant portion of the activity in the pool water in the event of cladding failure [3, 14], but the concentration of these two radionuclides is highly variable and often below detection limits. For example, Cs-137 was detectable in 15% of samples taken over a three-year period from spent fuel pool water at a PWR [16]. Over that period the average Co-60 concentration was 27 times greater than the average Cs-137 concentration [16].

Rawl et al. [15] identified four common contaminants in LWR spent fuel pool water based on a literature search and qualitative information from power plant operators: ^{134}Cs, ^{137}Cs, ^{58}Co and ^{60}Co. A reference pool water composition was developed on the basis of typical proportional activities of these radionuclides in spent fuel pool water (Table 3) [15].

Table 3. Reference spent fuel pool water radionuclide composition [15].

Radionuclide	% of activity present
^{60}Co	72.2
^{58}Co	21.7
^{137}Cs	3.6
^{134}Cs	2.5

As is the case for spent fuel pool water, reported measurements of radionuclides on cask surfaces are variable. Douce et al. [10] concluded from a review of reported measurements of contamination on cask surfaces that Co-60 typically is responsible for about 80% of activity on the cask surfaces and that Mn-54, Co-58, and Ag-110m can also be present but on a smaller scale. On the other hand, Jung et al. [11] identified Cs-137 and Cs-134 as well as Co-60 as the dominant sources of activity on cask surfaces.

5.0. REFERENCES DOSES USED TO DERIVE PROPOSED CONTAMINATION LIMITS

Reference Doses are required to convert estimated maximum doses per unit surface contamination on spent fuel flasks to surface contamination limits. Exemption values in the Basic Safety Standards [17] and IAEA Transport Regulations [2] were based on the primary dose criterion that the effective dose should not exceed 10 μSv y^{-1} [1 mrem y^{-1}] or the most exposed individual. This may be a reasonable limiting dose to apply to members of the public due to surface contamination of spent fuel casks, but a separate value is needed for workers because an annual dose as low as 10 μSv y^{-1} [1 mrem y^{-1}] is not reasonably achievable for persons who regularly work with spent fuel casks.

A value of 1 mSv y^{-1} [100 mrem y^{-1}] has been proposed as a Reference Dose for workers for determination of package surface contamination limits [18]. This value is used in some current IAEA documents as an acceptable dose level for worst-case or low probability situations, or situations in which it is not practical to limit doses to values on the order of 10 μSv y^{-1} [1 mrem y^{-1}] [19-21]. Moreover, the CRP Basic Model and its parameters values are recognized as being conservative, and the expected doses from exposure to packages contaminated at limits based on a given dose constraint are considerably lower than that dose constraint [22]. Another important consideration in selection of a Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] for workers is that the Basic Model, as well as a similar but generally less conservative model developed by the World Nuclear Transport Institute (WNTI) [22], predicts that the maximum dose to workers from contamination on packages including spent fuel casks is more than 100 times greater than the maximum dose to members of the public, regardless of the radionuclides present. This means that a Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] for workers should serve to keep doses to members of the public below 10 μSv y^{-1} [1 mrem y^{-1}]. Finally, a Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] for workers seems to be a reasonable dose constraint as this term is defined by the International Commission on Radiological Protection (ICRP), because it is only 5% of the ICRP dose limit for workers [23].

6.0. CONCLUSIONS: PROPOSED CONTAMINATION LIMITS FOR SPENT FUEL CASKS

Beta-gamma emitters judged, on the basis of information discussed earlier, to have the greatest potential to reach spent fuel cask surfaces are listed in Table 4. Cobalt-60 appears to be a major source of beta-gamma emissions from spent fuel cask surfaces in most cases. Cesium-137 and Cs-134 are important contributors to the total activity in some situations, but their contribution to total activity in spent fuel pools or on fuel casks is highly variable. Reported mixtures of radionuclides in pool water or on casks indicate that other potentially important contributors are Cr-51, Mn-54, Co-58, Zn-65, Ag-110m, Sb-124, and Sb-125. Radionuclides listed in the last row of Table 4 are considered as potentially important sources of beta-gamma activity because they are important contaminants at some facilities and conceivably could reach spent fuel pools.

For each of the radionuclides listed in Table 4, maximum annual doses to workers and members of the public from a surface activity of 1 Bq cm^{-2} [27 pCi cm^{-2}] were assessed in IAEA-TECDOC-1449 [1]. Highest estimated annual doses per unit concentration on spent fuel flask surfaces are associated with Co-60, Ag-110m, and Ba-140. These radionuclides yield similar estimates of dose per unit activity on surfaces of fuel flasks or other package types. For example, Table 25 of TECDOC-1449, which lists maximal doses to the most exposed workers from any of the package types, gives values in the range 0.024-0.027 mSv y^{-1} / Bq cm^{-2} [~0.1 mrem y^{-1} / pCi cm^{-2}] for these three radionuclides.

Table 4. Potential beta-gamma emitters on spent fuel casks	
Radionuclides	Expected contribution to total activity on cask surfaces
Co-60	Typically dominant contributor
Cs-137, Cs-134	Major contributors in some cases
Cr-51, Mn-54, Co-58, Zn-65, Ag-110m, Sb-124, Sb-125	Other non-trivial contributors identified in some cases
H-3, P-32, Fe-55, Fe-59, Ni-59, Ni-63, Sr-89, Sr-90, Zr-95, Nb-93m, Nb-95, Ag-108m, Cs-136, I-131, Ba-140, Eu-152	Theoretically non-trivial contributors in limited situations

For the radionuclides listed in Table 4, contamination limits for packages based on a Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] and maximal estimated doses to workers per unit activity on packages are shown in Table 5. The maximal estimated doses to workers per unit activity on package surfaces were taken from Table 25 of TECDOC-1449 [1] and represent the maximum for all package types considered in the CRP model, including spent nuclear fuel flasks. The lowest limit among likely contaminants on spent fuel flasks, shared by Co-60, Ag-110m, and Ba-140, is 40 Bq cm^{-2} [1100 pCi cm^{-2}]. Because derived limits for other potentially important beta-gamma emitters on fuel cask surfaces are higher than 40 Bq cm^{-2} [1100 pCi cm^{-2}], a contamination limit of 40 Bq cm^{-2} [1100 pCi cm^{-2}] should be adequately cautious for beta-gamma emitters on spent fuel casks. Because Co-60 is expected to represent a major portion of the surface activity fuel on casks in most cases, this is not an unreasonably conservative approach.

To address the possibility that spent fuel casks may occasionally be contaminated with non-trivial quantities of alpha emitters, it is useful to specify a separate limiting concentration for alpha emitters. Due to lack of direct information concerning alpha emitters on spent fuel casks, it is assumed that some combination of the typically most important alpha-emitters on surfaces at nuclear power plants is the source of alpha emissions on the casks. As indicated earlier, these are U-232, U-233, U-234, U-235, U-236, U-238, Pu-238, Pu-239, Pu-240, Pu-241, Am-241, Cm-242, and Cm-244.

Table 5. Derived surface contamination limits for spent fuel casks for potentially important β-γ emitters. Based on Reference Doses of 1 mSv y^{-1} (worker) and 10 μS y^{-1} (public)	
Radionuclide	Limit in Bq cm^{-2} [pCi cm^{-2}]
Co-60, Ag-110m, Ba-140	40[a] [1100]
Cs-134, Cs-136	50[a] [1350]
Ag-108m, Sb-124	60[a] [1600]
Fe-59, Eu-152	80[a] [2200]
Mn-54, Co-58, Sr-90, Nb-95, Zr-95, I-131, Cs-137	100[a] [2700]
H-3, P-32, Zn-65, Sr-89, Sb-125	200-1000[a,b] [5400-27,000]
Cr-51, Fe-55, Ni-59, Ni-63, Nb-93m	3000-40,000[a,b] [81,000-1,100,000]

[a]Rounded to one significant digit.
[b]A maximum limit of 100 Bq cm^{-2} was proposed at a recent IAEA Consultants Meeting [18].

Contamination limits for these 13 alpha emitters were derived on the basis of the Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] for workers, together with maximal dose estimates to workers derived using the CRP Basic Model. The lowest derived limits are for Pu-238, Pu-239, Pu-240, Am-241, and U-232 and are in the range 0.3-0.4 Bq cm^{-2} [8-11 pCi cm^{-2}]. As in the earlier calculations for beta-gamma emitters, these limits are based on maximal doses to workers given in Table 25 of TECDOC-1449 [1] and represent the maximum over all package types considered in the CRP Basic Model. Maximal doses to workers based on spent nuclear fuel casks are somewhat lower

for each of the alpha emitters considered here. As is the case for beta-gamma emitters, derivation of limits based on dose estimates and a Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] for workers yields at least as restrictive a contamination limit as would be derived from the maximal dose estimates for members of the public together with a Reference Dose of 10 μSv y^{-1} [1 mrem y^{-1}]. Thus, the predictions of CRP Basic Model together with a Reference Dose of 1 mSv y^{-1} [100 mrem y^{-1}] for workers provide support for the existing contamination limit of 0.4 Bq cm^{-2} [11 pCi cm^{-2}] for high-toxicity alpha emitters.

The concentration of alpha emitters on a spent fuel cask is expected to be orders of magnitude lower than that of beta-gamma emitters. Thus, a contamination limit of 40 Bq cm^{-2} [110 pCi cm^{-2}] for beta-gamma emitters should ensure that the concentration of alpha emitters is well below 0.4 Bq cm^{-2} [11 pCi cm^{-2}]. Nevertheless, a separate contamination limit for alpha emitters is warranted to prevent any unexpectedly high alpha activity on cask surfaces. If beta-gamma and alpha emitting radionuclides are present on the cask surface, proportioning the activity present with the activity limit for each type of radiation should be done to stay within the reference dose.

7.0. REFERENCES

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