An Estimate of the Contribution of "Crud" to the Radioactivity Source Term of a Spent Fuel Transport Cask*

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INTRODUCTION

There are three sources of releasable radioactive material in a loaded, spent fuel transport cask: (1) the residual contamination activity on the cask surfaces as a result of loading operations and previous shipments, (2) fission and activation-product activity associated with deposited material on the fuel assembly surfaces ("crud"), and (3) the radionuclides contained within the individual fuel rods comprising the fuel assemblies.

The U.S. regulations for the containment requirements of a spent fuel transport cask limit radionuclide release rates to 10^{-6} A_2 per hour and A_2 per week for normal and accident conditions of transport, respectively (10 CFR 71). Compliance with these release limits, R_i, may be demonstrated by directly measurable leak rates, L_i, through the mathematical expression

$$L_i = R_i/C_i \ , \ (i = N \text{ for normal or } A \text{ for accident conditions})\ , \qquad (1)$$

where C_i represents the concentration of airborne radioactive material in the cask that could escape from the containment system for the appropriate conditions of transport. This paper describes a method for estimating the contribution to C_N and C_A due to crud and residual contaminant activity.

METHODOLOGY FOR DETERMINING CRUD CONTRIBUTIONS TO C_N AND C_A

A conservative upper bound on L_i of Eq. (1) can be obtained by determining possible maximum values of the airborne activity concentration C_i for each radioisotope associated with the crud and contaminant inventories. To obtain a more realistic estimate of L_i, however, it is necessary to develop a more mechanistic description of radioactive particle behavior in the cask, which includes the distribution of particle sizes.

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Crud can contribute to the radiological source term during transport only if it spalls from surfaces and becomes airborne. No attempts appear to have been made to determine undisturbed particle size distributions of crud contained on or released from fuel rod surfaces, although a number of observations have been made of crud particle sizes following abrasive removal (Anstine 1982; Strasser et al., 1985). We examined the only two existing scanning electron micrographs of crud deposits in an effort to obtain a particle size distribution; the fuel rod was from Quad Cities BWR fuel. A value of the diameter of each particle in the photomicrograph was determined from the average of the major and minor chords. The resulting distribution, which we have provisionally assumed to be representative of all LWR crud, is given in Figure 1. The distribution exhibits a lognormal character with a geometric mean diameter of 3.0 μm and the geometric standard deviation of 1.87μm. About 99% of the particles have diameters below 10 μm. The distribution is readily converted to a mass distribution by assuming all particles have the same density; the resulting mass median diameter is 9.7μm.

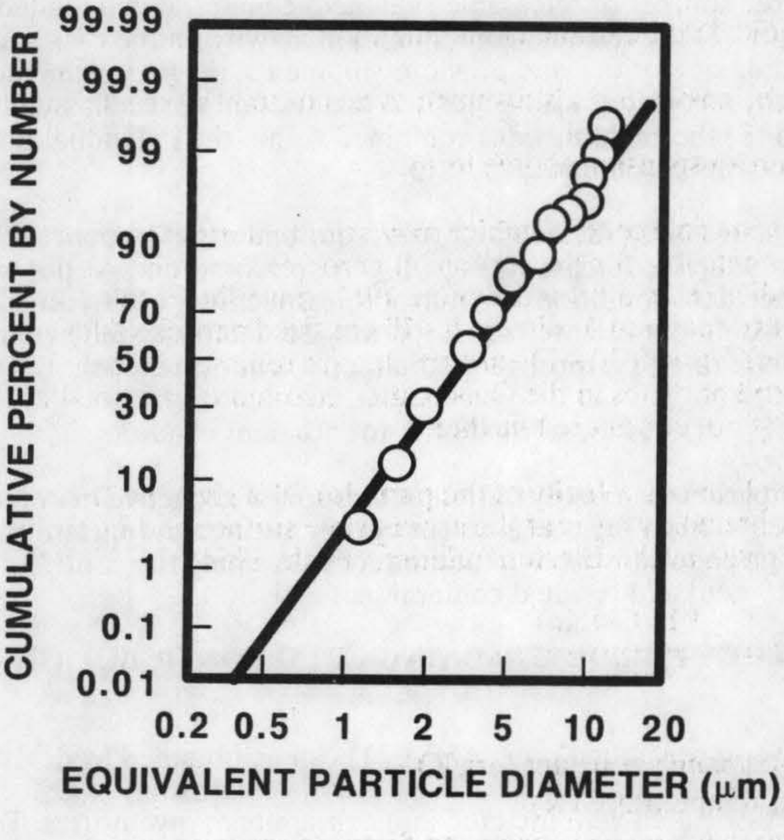

Figure 1. Crud Particle Size Distribution for Quad Cities BWR Fuel

A mass balance equation for C_i, for particles of size d_p, which includes sinks from plating on surfaces, gravitational settling and particle out-leakage, can be written:

$$dC_i/dt = -\beta_i C_i + B \tag{2}$$

where

$C_i = C_i(t)$ = time dependent particulate activity suspended in the cask cavity gas volume (in Ci/cm^3), i = N for normal, A for accident conditions,

$\beta_i = -(A_T D'/V + A_s v_s/V + L_i/V)$,

A_T = total surface area of the cask cavity, including the fuel and the basket (cm^2),

A_s = settling area normal to settling direction inside the cask cavity, including the fuel and the basket (cm^2),

v_s = particle settling velocity,

V = void volume of the cask (cm^3),

L_i = gas leak rate (L_i is assumed to be constant) (cm^3/s), and

B = a re-suspension source term.

The resuspension source term, which may arise in normal transport from road vibrations, is negligible for the very small particles shown in our distribution. Adhesion studies (Deryagin and Zimon, 1961) have shown vibration forces of up to 24,000 g's are required to disrupt 5 - 10 μm sized particles (depending on residence time), though 30 - 40 μm particles are removed by only 1.3 g's. Since 99% of the crud particles in the Quad Cities distribution are smaller than 10 μm, resuspension is not considered further.

The diffusive plate-out velocity of the particles D' is given by D/δ, where δ is the mass transfer boundary layer at the appropriate surface and D, the particle diffusivity, is given by the Einstein relation (Fuchs 1964):

$$D = \frac{kT_g\, Cu(Kn)}{3\pi\mu\kappa d_p} , \tag{3}$$

in which

k = Boltzmann's constant (erg/K),
T_g = gas temperature (K),
Cu = Cunningham slip correction factor (dimensionless),
Kn = Knudsen number (dimensionless),
κ = dynamic shape factor (dimensionless),
μ = bulk gas viscosity (g/cm•s), and
d_p = diameter of a spherical particle of equal volume (cm).

The particle settling velocity is given by (Fuchs 1964):

$$v_s = \rho_p g \, Cu(Kn) \, d_p^2 / (18 \, \mu \, \kappa) \, , \tag{4}$$

where ρ_p is the particle material density, and g is the gravitational acceleration. The initial condition for C_i is: $C_i(t=0) = f_i \, M_T/V$, where M_T is the total activity inventory, and f_i is the spallation fraction. Since resuspension is negligible, the solution to Eq. 2 is,

$$C_i = (f_i \, M_T/V) \exp[-\beta_i t] \, . \tag{5}$$

The time-averaged activity concentration, C during a leak period τ, assuming a constant leak rate L_i is given by

$$\bar{C}_i = \frac{1}{\tau} \int_0^\tau C_i(t) \, dt \, . \tag{6}$$

Inserting Eq. (5) into Eq. (6), and generalizing to a polydisperse particle mass distribution, $C(d_{p,o})$, yields

$$\bar{C}_i = (f_i M_T/V) \frac{\int_0^\infty \left(\frac{1 - e^{-\beta_i \tau}}{\beta_i \tau} \right) C(d_p, o) \, d(d_p)}{\int_0^\infty e^{-\beta_i \tau} \, C(d_p, o) \, d(d_p)} \tag{7}$$

in which

$$\beta_i = A_s v_s/V + A_T D'/V + L_i/V \tag{8}$$

CRUD AND CONTAMINANT CHARACTERISTICS

Crud (and most residual contamination) is a mixture of reactor primary-cooling-system corrosion products that have deposited on fuel assembly or cask surfaces. These deposits contain neutron-activated nuclides, and may also contain particulates of fissile material and fission products. There are two types of crud: (1) a fluffy, easily removed crud, usually found on BWR rods, and composed mainly of hematite (Fe_2O_3); and (2) a tenacious, tightly bound crud, usually found on PWR rods, and composed mainly of a nickel substituted spinal (Einziger and Cook 1984a).

Many variables are required to characterize spent fuel and, unfortunately, insufficient data exist to completely specify the crud characteristics for each fuel variable. Also, there are, in fact, only a dozen or so comprehensive measurements of crud characteristics on rods that show discernible amounts of crud, and hundreds of qualitative visual observations of rods that indicate little or no crud. A compilation of measured Cobalt-60 (^{60}Co) maximum "spot" activity densities and visual observations versus the number of rods, assemblies, or samples was developed. Figure 2 and Figure 3 illustrate histograms that were constructed for PWR and BWR fuel, respectively, to display the percentage of rods in selected

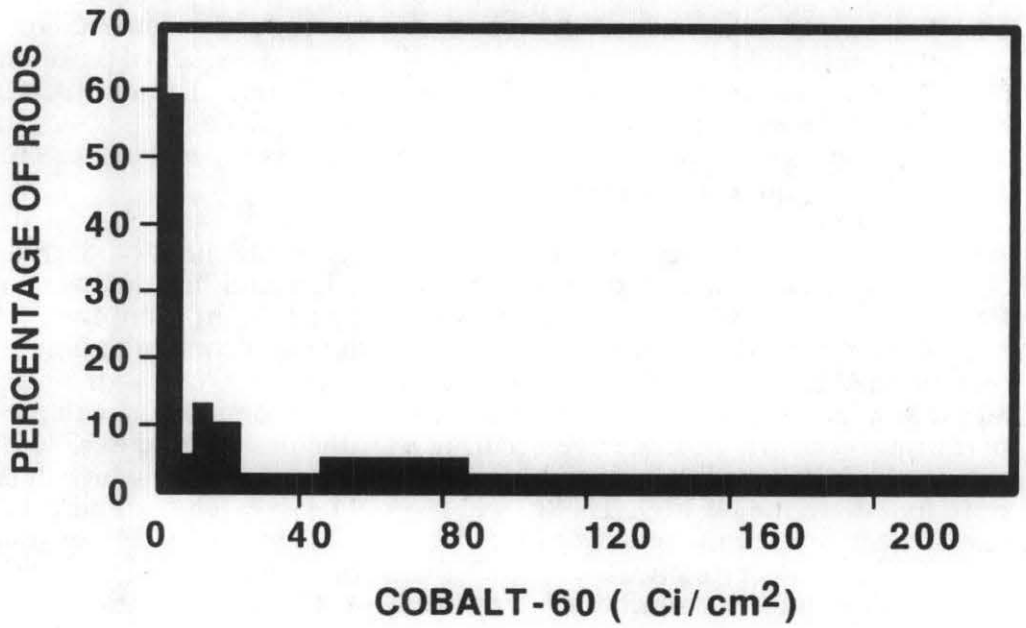

Figure 2. Distribution of PWR Spent Fuel Rods as a Function of Maximum "Spot" Crud Activity

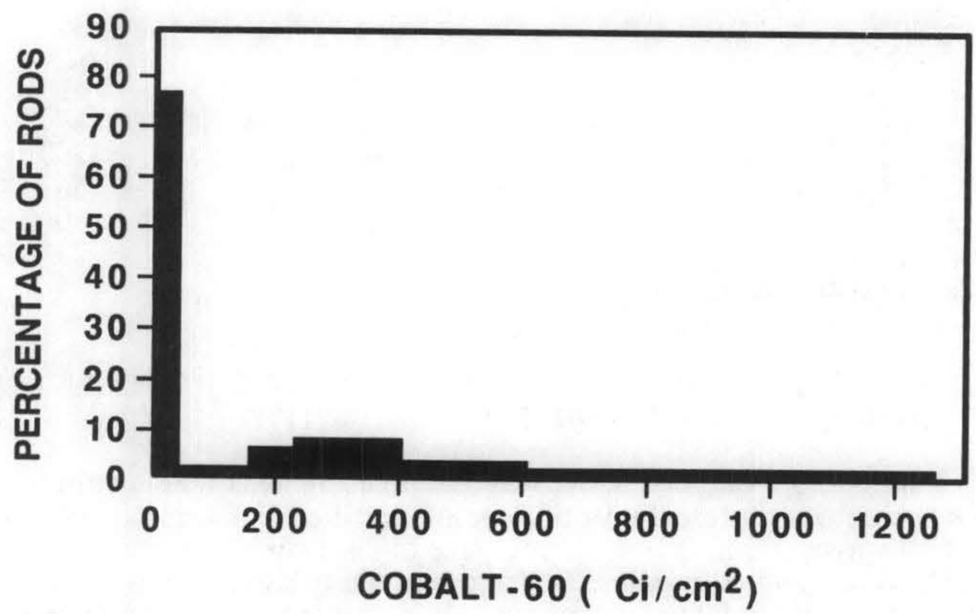

Figure 3. Distribution of BWR Spent Fuel Rods as a Function of Maximum "Spot" Crud Activity

activity density intervals. Both types of rods exhibit bimodal distributions that are dominated by an essentially crud-free mode. However, for purposes of the present analysis maximal values of radioactivity density, as reported from examinations of crud, were selected to represent "worst case" activity densities of pertinent radioisotopes contained in BWR and PWR fuel rod crud. These values are presented in Table 1. In addition, these maximum "spot" activities were assumed to be distributed over the entire surface of the rod. ^{60}Co accounts for most of the activity 5 years after fuel discharge.

An attempt was also made to characterize residual contamination of transport casks. Very little published data were available. Unpublished data were obtained from U.S. nuclear utilities and other facilities that transport spent fuel. The data ranged from cursory estimates to detailed radiation and contamination measurements. Nonetheless, the data survey identified 20 cases for which detailed measurements were made. This is a small sample in comparison with the hundreds of transport operations that have occurred over the past several decades; however, the sample is believed to be representative. The cask types for which detailed data were found are the NLI 1/2, TN-8L, and NAC-ID casks. The estimated total residual activities in these casks ranged from 0.01 Ci to 7.7 Ci. These values are well within the uncertainty of those in Table 1 for fuel deposited crud, thus the residual contaminant contribution is probably of secondary significance.

TABLE 1. RADIOISOTOPIC COMPOSITION OF "WORST CASE" CRUD

	Crud Activity Density[a] ($\mu Ci/cm^2$)	
Nuclide	PWR Fuel	BWR Fuel
^{51}Cr	391	35
^{54}Mn	380	625
^{58}Co	1400	63
^{59}Fe	300	87
^{60}Co	140	1250
^{65}Zn	-	56
^{95}Zr	36	30

[a]Corrected to reactor shutdown

EXAMPLE CALCULATIONS OF CONTAINMENT REQUIREMENTS RELATIVE TO THE RELEASE OF CRUD-BORN RADIOACTIVITY

The methodology outlined above was used to estimate containment requirements for cask geometries representative of currently certified U.S. spent fuel transport casks under the assumption that the only source of dispersible radioactivity is that associated with crud on the fuel assemblies. The age of the fuel assemblies was assumed to be 5 years, so that about 90 percent of the radioactivity associated with the crud results from ^{60}Co. In addition, the entire spectrum of known crud burdens was used. Histograms relating containment requirements in terms of leak rate with the total fuel rod population were developed. Those histograms, in principle, can be employed in a probabilistic risk assessment of crud release. In this paper, however, we shall present only the requirements that embrace the entire population, i.e., those representing the bounding cases for spot activity. This worst case spot measurement was assumed to exist on the entire surface of the fuel rods.

The observed crud distributions were used to determine average values of C_i from Eq. (7), and corresponding histograms of L_i by Eq. (1) for each of the transport casks examined. In all cases, 100 percent spallation was assumed and helium was assumed for the cavity fill gas. These histograms, in turn, were translated into distribution plots which relate L_i to the fraction of the total fuel rod population being transported. From such cumulative distributions, it was possible to obtain values of L_N and L_A that envelope the entire population of fuel rods. The results for normal transport yielded acceptable leak rates in the range of 10^{-4} to 10^{-3} cm^3/sec. Acceptable leak rates for accident conditions are orders of magnitude higher. The particle size distribution, the settling velocity, and the settling area are the most sensitive elements in these calculations.

RESULTS AND CONCLUSIONS

A methodology has been developed to determine the concentrations C_N and C_A and the associated transport cask containment criteria L_N and L_A resulting from radioactive debris (crud) contained on spent fuel and cask surfaces. The methodology accounts for crud particle characteristics (such as particle size distribution) and for attenuation of the suspended particulate concentration. Calculations were performed for typical spent fuel transport cask geometries for the normal and accident conditions prescribed in 10 CFR Part 71. The most current published data on crud composition and structure, specific activity, spallation mechanisms and fractions, and crud particle size were used. For parameters where no useful quantitative data could be found, conservative upper-bound values were used; e.g., 100% spallation for regulatory impact and shock loading transport conditions and uniform worst case activity densities. The containment criteria leak rates were calculated assuming 5-year-cooled spent fuel. The results of the calculations lead to the following conclusions:

(1) Cask surfaces do not appear to become contaminated to the extent that this contamination competes with the crud on fuel rods as a dominant source term. This indicates very little crud spalls during normal transport conditions.

(2) The maximum allowable leak rates for the transport of BWR fuel are 10 to 20 times smaller than those for PWR fuel, depending on cask type. This occurs primarily because much higher crud activity densities have been observed on BWR fuel.

(3) The calculated leak rates are most sensitive to the crud particle size distribution and settling area. The effect on leak rate could be a factor of 100 to 1000 in each case, depending on cask design, type of fuel, and particle size.

(4) The leak rate values presented in this report are believed to be quite conservative for crud releases because:

(a) the crud activity inventory used for this analysis conservatively assumed that the measured maximum crud activity is distributed uniformly over the entire surface area of the fuel assembly, even though data indicate that the average crud activity is significantly less than the maximum values used. The effect of decay beyond five years was ignored.

(b) All crud (100% spallation) was assumed to be instantaneously released to the cask cavity.

(c) the likelihood of particle plugging of the containment leak path was not considered.

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