STACE: Source Term Analyses for Containment Evaluations of Transport Casks*

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INTRODUCTION

The containment requirements for the transportation of radioactive material are defined by both International Atomic Energy Agency (IAEA) and U.S. Nuclear Regulatory Commission (NRC) regulations (IAEA 1990; 10 CFR 71). Procedures generally acceptable to the NRC for assessing compliance with these provisions have been identified in Regulatory Guide 7.4 (US NRC 1975). This guide endorses the containment and leak test procedures that are specified in American National Standards Institute (ANSI) Standard N14.5 (ANSI 1987). ANSI N14.5 states that "compliance with package containment requirements shall be demonstrated either by determination of the radioactive contents release rate or by measurement of a tracer material leakage rate." The maximum permissible leakage rates from the transport cask L_i (cm^3/s), where i represents either normal (N) or accident (A) conditions of transport, can be determined from the maximum permissible release rates R_i (Ci/s) and the time-averaged volumetric concentrations of suspended radioactivity within the cask C_i (Ci/cm^3) by:

$$L_i = R_i / C_i \tag{1}$$

The maximum permissible release rates are specified in ANSI N14.5 to be $A_2 \times 10^{-6}$ per hour for i = N, and A_2 per week for i = A (the exception is ^{85}Kr in which 10,000 Curies are permitted to be released in one week). The quantity A_2 is an activity limit which, under specific release scenarios, would prevent radiological effects from exceeding a specified level consistent with radiological protection standards of the International Commission on Radiological Protection (ICRP). Values of A_2 (e.g., A_2 = 7 Ci for ^{60}Co; A_2 = 10 Ci for ^{137}Cs) are tabulated in Appendix A of 10 CFR 71.

ANSI N14.5 further states that "C_N and C_A shall be determined by the performance of tests on prototypes or models, reference to previous demonstrations, calculations, or reasoned argument," with "consideration given to the chemical and physical forms of the materials within the containment system, the possible release modes, and the maximum temperature, pressure, vibration, and the like, to which the contained material would be subjected for normal and accident conditions of transport." The development of the Source Term Analyses for Containment Evaluations (STACE) methodology provides a unique means for estimating the probability of cladding breach within transport casks, quantifying the amount of radioactive material released into the cask interior, and calculating the releasable radionuclide concentrations and corresponding maximum permissible leakage rates. STACE, which is being developed at Sandia National Laboratories (SNL), is a task of the Cask Systems Development Program (CSDP) sponsored by the United States Department of Energy's Office of Civilian Radioactive Waste Management (OCRWM). The STACE methodology follows the procedures of ANSI N14.5 by estimating the releasable radionuclide concentrations for specific cask designs, fuel assemblies, and initial conditions. These calculations are based on defensible analysis techniques that credit multiple release barriers, including the internal fuel structure, the cladding, and the internal cask walls.

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An alternative to computing C_N and C_A is to limit the maximum permissible leakage rate to 10^{-7} std cm^3/s, which is the practical limit of "leak tightness" as defined by ANSI N14.5. This approach generally leads to increased cask maintenance costs, personnel exposure, and limited lifetime usage of the casks in the certification and recertification process. By directly computing C_N and C_A the source term methodology is expected to significantly improve cask economics and safety by relaxing the maximum permissible leakage rates.

SOURCE TERM METHODOLOGY

The development of a source term methodology considers the individual contributions of the three distinct media from which the radionuclides in a spent fuel transport cask originate:

- radionuclides that can be released through breaches in the spent fuel cladding;
- activated corrosion and free fission products, referred to as CRUD, adhering to the surface of spent-fuel rods; and
- residual contamination that may build up in the cavity of a cask over time.

Containment of cask contents by a transport cask is a function of the cask body, one or more closure lids, and various bolting, hardware, and seals associated with the cavity closure and other containment penetrations. In addition, characteristics of cask contents that impede the ability of radionuclides to move from an origin to the external environment also provide containment. In essence, multiple release barriers exist in series in transport casks, and the magnitude of the releasable activity available in the cask is considerably lower than the total activity of its contents. A source term approach accounts for the magnitude of the releasable activity available in the cask by assessing the degree of barrier resistance to release provided by material characteristics and inherent barriers that impede the release of radioactive contents.

Example leakage rate calculations in ANSI N14.5 conservatively assume that 3% of the fuel rods fail during normal conditions of transport and 100% fail under hypothetical accident conditions of transport. This paper presents a defensible methodology to be used in conjunction with ANSI N14.5 to estimate fuel rod failure rates and the corresponding releasable fission products. The critical normal and hypothetical accident conditions are, respectively, a 0.3-meter drop and 9-meter drop of the cask containing the fuel rods onto an unyielding target. STACE models the response of the fuel rods to these impacts and evaluates the release of radioactive materials in the event of fuel cladding failure. The fuel assemblies and cask internal hardware are modeled in detail, allowing for interactions between assemblies and the cask basket, and between spacer grids and fuel rods.

Three reports have been prepared which together present a methodology for determining the concentration of freely suspended radioactive materials within a spent-fuel transport cask. Each report treats one of the three sources of radioactivity: (1) the loaded spent fuel (Sanders et al. 1992), (2) the radioactive material, CRUD, attached to the external surface of the cladding (Sandoval et al. 1991), and (3) the residual contamination adhering to the interior surfaces of the cask cavity (Sanders et al. 1991). Since the concentrations of the individual sources are additive, the maximum permissible leakage rate for the combined source is written:

$$L_{total} = \frac{R}{C_{SF} + C_{CRUD} + C_{RC}} . \tag{2}$$

The individual concentrations C_{SF}, C_{CRUD}, and C_{RC} determine individual leakage rates L_{SF}, L_{CRUD}, and L_{RC}, respectively, when considered as sole sources of radioactivity. Expressing the individual concentrations in terms of the individual leakage rates through Equation (1), Equation (2) can be rewritten in terms of the individually determined maximum permissible leakage rates:

$$L_{total} = \frac{L_{SF} \times L_{CRUD} \times L_{RC}}{L_{CRUD}\, L_{RC} + L_{SF}\, L_{RC} + L_{SF}\, L_{CRUD}} . \tag{3}$$

This method of combining individually determined containment requirements should only be done after all terms are converted to the same temperature and pressure conditions.

Spent Fuel Contribution

Spent fuel contains the largest potential source of releasable radioactivity (Sanders et al. 1992). The contribution of spent fuel to the overall maximum permissible leakage rate largely depends upon its initial pre-transport condition and on subsequent fuel rod response to transportation conditions. The type and amount of radioactive materials that may be released from the fuel rod to the cask cavity are governed by fuel cladding failure which is a function of cask and assembly designs, transport loading conditions, fuel irradiation histories, and other initial conditions. Since cladding failures are highly statistical events, criteria for evaluating the spent fuel source term is probabilistic, although it may depend upon deterministically derived response characteristics. Therefore, the source term methodology combines a detailed deterministic mechanical response of fuel rods and assemblies with probabilistic failure evaluations and release estimates.

Four steps are used to apply the source term methodology to spent fuel for normal and hypothetical accident conditions:

1. Characterization of the dynamic environment experienced by the cask and its contents.

2. Deterministic modeling of the stresses induced in spent fuel cladding by the dynamic environment.

3. Evaluation of these stresses against probabilistic failure criteria for ductile tearing and material fracture at generated or pre-existing cracks partially extending through the cladding wall's thickness.

4. Prediction of the activity concentration in a cask cavity using knowledge of the cask void volume, the inventory of radionuclides residing in fuel-cladding gaps, and estimates of the fraction of gases, volatile species, and fuel fines released.

The dynamic environments in the first step are defined in 10 CFR 71 and are divided into normal and hypothetical accident conditions. The most severe normal and hypothetical accident conditions are the 0.3-m and 9-m free drop impacts onto unyielding targets, respectively (Sanders et al. 1992). Other regulatory events such as shock and vibration, a fully engulfing fire, and puncture events, have been evaluated and shown to have minimal impact on the assemblies' response (Sanders et al. 1992). A rigid-body kinematics model is used to analyze the impact event by characterizing the crushing behavior of the impact limiters for all possible drop orientations. This analysis defines the center of gravity deceleration load history applied to the fuel assemblies. This is then input to the assembly computational models.

The second step develops detailed geometric and computational models which are analyzed using discrete finite element methods to obtain the deterministic mechanical response of the fuel rods and assemblies. A deterministic response analysis of a loaded transport cask and its contents is performed by isolating smaller substructures from the total system and analyzing them separately. This isolation takes place at naturally identifiable interfaces so that force or displacement boundary conditions can be properly defined. The smallest structural element that is isolated is a single fuel rod. The force transfer interfaces for the rod are tie plates, spacer grids, and to a lesser extent, adjacent rods. Under certain conditions (e.g., end impact) the single rod model response adequately represents the response of the whole assembly, assuming that all rods in the assembly have similar deformation patterns. This assumption is conservative because the predominant end-drop failure mode is caused by high tensile bending strains produced by lateral displacement. However, a single rod model is inapplicable to other impact orientations (e.g., side drop and slapdown), and the more complex structure of a single assembly is required.

The single assembly models individual rods, spacer grids, and end-tie plates. The force transfer interface for the single assembly is the basket structure. Depending on the structural design of the basket, force transfer between assemblies may be replaced by displacement boundary conditions that isolate the object assembly from surrounding assemblies. The basket/assembly interface is replaced by a line support for a typical continuous-plate basket design. The basket structure itself is not part of the structural model. The detailed geometric model of the single assembly consists of several hundred beam-column elements that represent individual rods, and special nonlinear hysteretic truss elements that represent spacer grids at each interface (Barrett et al. 1992). To capture the detailed deformation characteristics of the fuel rods, each rod is represented by no fewer than thirty beam-column elements with elastic-plastic and large displacement and strain capabilities. Rod-to-rod interaction is simulated by nonlinear contact spring elements with contract/release capabilities. Deterministic response parameters, cladding stresses and strains, and rod interaction forces are obtained at critical points along each length of the rod. These quantities are then used in the third step.

The third step involves the application of probabilistic methods to determine the likelihood of cladding breach in the spent fuel (Foadian et al. 1992). Two properties specifically used in this evaluation are the material ductility, which is related to ductile tearing from excessive strain, and fracture toughness, which is used to determine the extension of generated or pre-existing partial (partially through the wall) cracks. Three cladding failure modes which can occur are transverse tearing, rod breakage, and longitudinal tearing. Experimental data are used to define the various failure modes, and these data have been translated into specific failure criteria (Bauer et al. 1977, Miyamoto et al. 1976, Barsell 1987). Transverse tearing requires that the strain exceed the material ductility limits. It is assumed that once a crack is initiated, it will immediately extend through the wall, thus forming a pinhole or narrow transverse crack. Rod breakage is the extension of an existing transverse crack, and it requires a bending stress intensity that exceeds the fracture toughness of the intact material. Depending on the amount of available energy, a narrow transverse tear could extend through a large portion of the cladding cross section, or even result in a guillotine break. Longitudinal tearing, the opening of a part-wall longitudinal crack on the inside of the cladding, requires a hoop stress intensity that exceeds the fracture toughness. The driving force for the hoop stress intensity is a pinch load arising from rod-to-rod interaction. The source term methodology determines probabilities for the three different types of cladding breach. This is an essential prerequisite for defining release mechanisms, because the physical composition of fuel rod contents that could be released through a cladding breach is strongly dependent on the geometry of the cladding breach. A pinhole failure, for example, could result in the release of fission gases, volatile species, and finely dispersed fuel, whereas a guillotine break could further permit the release of fuel fragments.

The fourth step concerns the prediction of the activity concentration in the cask cavity. Many radionuclides are produced within fuel rods during reactor operation. The specific nuclide composition depends on the initial enrichment, irradiation history, and length of time since reactor discharge. Estimates of the radionuclide composition are obtained from the Oak Ridge LWR Spent Fuel Database (OCRWM 1987). ORIGEN2 calculations (Croff 1983) were carried out for several burnup levels to represent current and projected (extended) burnup limits for BWR and PWR fuel assemblies. These results are compiled in a radionuclide inventory database within STACE and input data for specific analyses are obtained by interpolation for the specified fuel type, burnup level, and time of transport after reactor discharge.

Having quantified the radionuclide inventory of fuel assemblies prepared for transport, the amount of this source released to the cask cavity during a cladding breach is determined. The spent fuel source term includes radionuclides released from the fuel matrix to the fuel-cladding gap in gaseous and vapor form, as well as gas-borne particulate fines. A model for the gap inventory has been developed to account for the buildup of xenon and krypton isotopes in the fuel-cladding gap. To determine the buildup of moderately volatile species (iodine, cesium, and tellurium) in the gap, it is assumed that they have the same mobility and diffusion characteristics as the noble gases, thereby establishing relationships for magnitude and distribution between the long-lived isotopes of volatile species and fission gases. The entire gap inventory is conservatively assumed to be readily available for release in the event of cladding breach, irrespective of breach location or size. The mass of fuel fines released through the cladding breach is taken to be 0.003 percent of the fuel mass in the rod, based on observation of the quantity of material released from rod segments in burst rupture experiments at Oak Ridge National Laboratory and Battelle Columbus Laboratory (Burian et al. 1985, Lorenz et al. 1980, Lorenz et al. 1981). This does not account for new fines, if any, produced due to crushing of the fuel pellets. Ninety percent of the fuel fines that reach the cask cavity are assumed to settle or plate-out, and thus be unavailable for release (Sanders et al. 1992).

CRUD Contribution

The methodology for modeling the CRUD source term differs from that for spent fuel due to the wider range and better quality of available data (Sandoval et al. 1991). There are two types of CRUD: a fluffy, easily removed CRUD composed mostly of hematite that is usually found on BWR rods; and a tenacious type composed of nickel-substituted spinel occurring on PWR rods. In a few BWR reactors, copper is also an important constituent. Along individual rod cladding, the average to peak observed density of CRUD radioactivity is approximately two, independent of the radionuclide. The nuclides which are important in the CRUD total activity depend on the time since discharge from the reactor; for shipments of five-year or older fuel, ^{60}Co accounts for over 92% of the activity in PWR fuel and 98% of the activity in BWR fuel.

The concentration of CRUD suspended in the cavity of a loaded spent fuel transport cask depends on the amount of CRUD initially adhering to the transported assemblies, on the fraction spalled in normal and hypothetical accident transport conditions, and on depletion and resuspension mechanisms acting on the suspended particles. The amount of CRUD present on spent fuel rods has been characterized in prior work (Sandoval et al. 1991). Most recently discharged fuel has no discernible or only slight CRUD deposits. CRUD aerosols have a time-dependent concentration after a spallation-inducing event. An expected particle size distribution for CRUD has been developed based on one sample of fuel that is believed to be representative of BWR fuel. The distribution has a precise log-normal shape with a mean number diameter equal to 3 μm and a standard deviation of 1.87 μm. Since a detailed distribution is available, it is possible to account for the behavior of aerosols inside the cask cavity. In the absence of resuspension, the rate of decrease in aerosol concentration is proportional to the concentration itself. The average CRUD concentrations in a cask cavity can be expressed as the concentration immediately after spallation and initial mixing, multiplied by a Release Reduction Factor that incorporates all geometrical information on the cask volume, settling and collection areas, and the aerosols' time-varying size distribution. C_{CRUD} can then be calculated directly, based on the specified cask cavity.

Residual Contamination Contribution

After casks have been used to transport spent fuel, their interior surfaces (especially the bottom) accumulate a residual contamination from CRUD spalled off the transported assemblies, or from immersion in storage pool water during loading and unloading of the assemblies. The residual contamination report (Sanders et al. 1991) discusses the mechanisms leading to spallation but does not quantify the adhesion forces themselves, and it presents previously unpublished data that clarify the amount of residual contamination present.

The largest amount of residual contamination reported is approximately 1 Ci. This amount is conservatively assumed to be present in the transport cask, and all of it is assumed to spall in both normal and hypothetical accident conditions of transport. An extensive set of example calculations for normal and hypothetical accident conditions is presented in the residual contamination report.

STACE

The methodology developed in the previous sections is implemented through the integrated STACE software package (Seager et al. 1992). STACE is a system of software modules operating under a graphics controller that performs source term analyses of spent fuel transport casks. STACE extracts relevant data from its built-in database module to perform thermal, mechanical, cladding breach, and release analyses. Figure 1 summarizes the STACE design elements. The output of STACE includes steady-state thermal contours for normal transport, temperature versus time in hypothetical accidents, and the structural response of the fuel rods and spacer grids. The probabilities of cladding breach are given for three different failure modes, and an isotopic breakdown is given of the initial and time-averaged activity released to the cask cavity for normal and hypothetical accident conditions. Finally, the maximum permissible leakage rates are given for normal and hypothetical accident conditions.

EXAMPLE SOURCE TERM ANALYSES

Table 1 presents the time-averaged volumetric concentrations of suspended radioactivity, C_i, and the maximum permissible leakage rates, L_i, for the spent fuel, CRUD, and residual contamination contributions to the releasable source term for the example case in which one Westinghouse 17x17 PWR assembly is transported in a representative lead-shielded truck cask. The assembly is assumed to have an average burnup of 30 GWd/MTU, and to be transported 10 years following reactor discharge. Calculations are performed for the normal condition of a 0.3-m end drop and the hypothetical accident condition of a 9-m side drop of the truck cask with impact limiters onto an unyielding target. The cladding temperature during the normal and hypothetical accident conditions is assumed to be 27°C.

The mechanical and cladding breach analyses predict a single rod failure probability of 5×10^{-5} for the 0.3-m normal transport event. Since the assembly contains 264 fuel-bearing rods, 0.013 fuel rods are expected to fail. However, these analyses conservatively assume that one rod fails due to normal conditions of transport. The peak cask accelerations are conservatively assumed to be 100 g during the 9-m side drop, and the analyses predict a single rod failure probability of 3.7×10^{-3}. Therefore, slightly less than one fuel rod is expected to fail in the 17x17 assembly due to the hypothetical accident condition. The CRUD and residual contamination are assumed to be composed entirely of ^{60}Co and to completely spall during both normal and hypothetical accident conditions. The maximum permissible leakage rates due to the spent fuel, CRUD, and residual contamination contributions to the source term are combined using Equation (3) to give total maximum permissible leakage rates for both normal and hypothetical accident conditions of transport. These results are also given in Table 1.

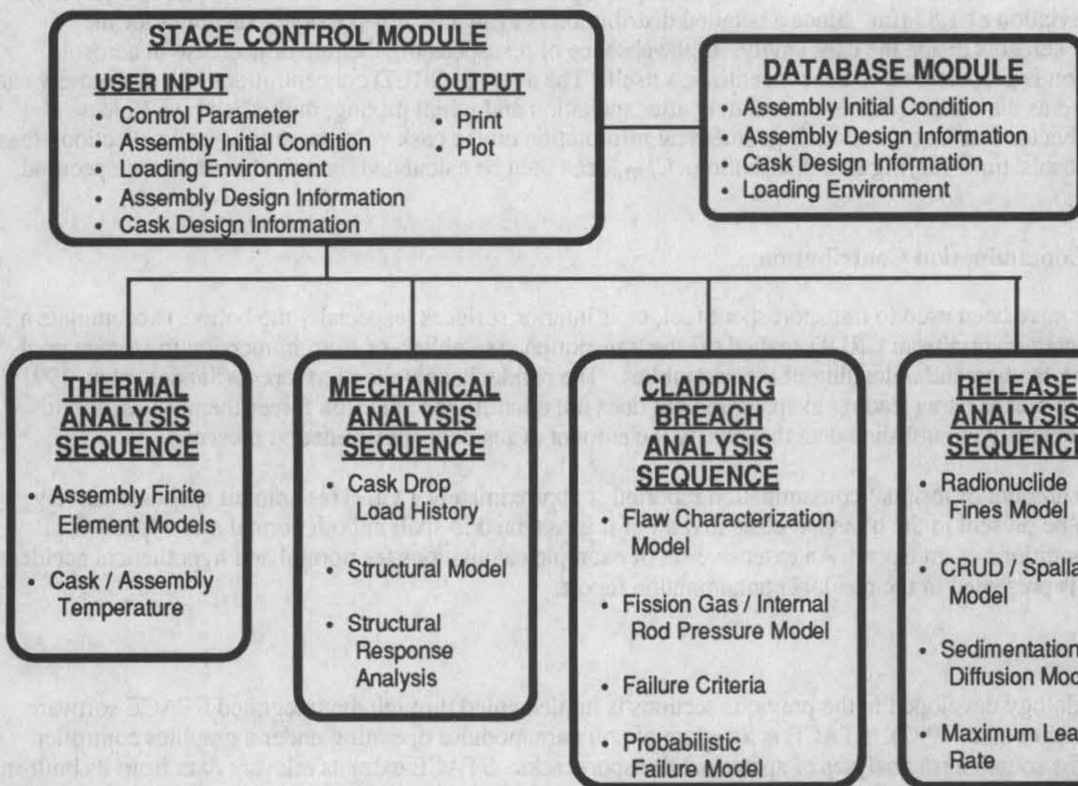

Figure 1. Design elements of STACE software system.

Table 1. Example Source Term Analyses for a Westinghouse 17x17 PWR Assembly.

	Spent Fuel		CRUD		Residual Contamination		Total
Transport Condition	C_{SF} (Ci/cm^3)	L_{SF} (cm^3/s)	C_{CRUD} (Ci/cm^3)	L_{CRUD} (cm^3/s)	C_{RC} (Ci/cm^3)	L_{RC} (cm^3/s)	L_T (cm^3/s)
Normal (0.3-m drop)	2.1×10^{-6}	5.6×10^{-3}	3.4×10^{-7}	5.6×10^{-3}	4.7×10^{-9}	4.1×10^{-1}	2.8×10^{-3}
Accident (9-m drop)	2.1×10^{-6}	33	2.1×10^{-9}	5.6×10^{3}	2.8×10^{-11}	4.1×10^{5}	33

CONCLUSIONS

Following the guidance of ANSI N14.5, the STACE methodology provides a technically defensible means for estimating maximum permissible leakage rates. These containment criteria attempt to reflect the true radiological hazard by performing a detailed examination of the spent fuel, CRUD, and residual contamination contributions to the releasable source term.

The evaluation of the spent fuel contribution to the source term has been modeled fairly accurately using the STACE methodology. The structural model predicts the cask drop load history, the mechanical response of the fuel assembly, and the probability of cladding breach. These data are then used to predict the amount of fission gas, volatile species, and fuel fines that are releasable from the cask. There are some areas where data are sparse or lacking (e.g., the quantity and size distribution of fines released from fuel rod breaches) in which experimental validation is planned. The CRUD spallation fraction is the major area where no quantitative data has been found; therefore, this also requires experimental validation. In the interim, STACE conservatively assumes a 100% spallation fraction for computing the releasable activity. The source term methodology also conservatively assumes that there is 1 Ci of residual contamination available for release in the transport cask. However, residual contamination is still by far the smallest contributor to the source term activity.

Finally, the ANSI N14.5 recommendation that 3% and 100% of the fuel rods fail during normal and hypothetical accident conditions of transport, respectively, has been shown to be overly conservative by several orders of magnitude for these example analyses. Furthermore, the maximum permissible leakage rates for this example assembly under normal and hypothetical accident conditions, estimated to be 2.8×10^{-3} cm^3/s and 33 cm^3/s, respectively, are significantly higher than the leaktight requirement of 10^{-7} std cm^3/s. By relaxing the maximum permissible leakage rates, the source term methodology is expected to significantly improve cask economics and safety.

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