A Source Term Methodology for Spent Fuel Cask Containment Evaluations*

T.L. Sanders[1], R.E. Einziger[2], H. Jordan[3], A.P. Malinauskas[4]

[1]*Sandia National Laboratories**, Albuquerque, New Mexico*
[2]*Battelle Pacific Northwest Laboratory, Richland, Washington*
[3]*Battelle Memorial Institute, Columbus, Ohio*
[4]*Oak Ridge National Laboratory, Oak Ridge, Tennessee, United States of America*

INTRODUCTION

A cask containment system can generally be designed from two perspectives; either the cask and its associated hardware alone are assumed to provide containment, or the contents of the cask, in this case the spent fuel, are also considered part of the containment system. In the latter approach, known as a source-term methodology, some credit is derived for those contents based on their material, physical, or chemical properties that tend to limit or inhibit radionuclide "release." The former approach in which no such credit is taken is generally called a "leak tight" design basis.

While previous containment analyses for transport casks in the United States have used both leak tight and source term approaches, recent practice on the part of both designers and regulators has been to rely totally on the cask for containment. This evolution to a "leak tight" criterion primarily resulted from the lack of a standardized source term characterization methodology that might have addressed specific properties of the cask contents.

The containment requirements for transport casks in the United States are defined by both International Atomic Energy Agency (IAEA) and U.S. Nuclear Regulatory Commission (NRC) regulations (IAEA 1985;US Code 1987). Procedures generally acceptable to the NRC for assessing compliance with these provisions have been identified in Regulatory Guide 7.4 (US Code 1975). This guide, in turn, endorses the containment and leak test procedures that are specified in the American National Standards Institute Standard ANSI N14.5 (ANSI 1987). The ANSI standard permits time-integrated values for the activity releases R_N and R_A under normal or hypothetical accident conditions, respectively. The requirements become

$$\int_o^{\tau} r_N(t)dt \leq 10^{-6}A_2 = R_N \quad , \text{ N => normal conditions} \quad , \qquad (1)$$

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** A United States Department of Energy Facility where τ is one hour, and

$$\int_0^{\tau} r_A(t)dt \leq A_2 = R_A \text{ , } A \rightarrow \text{ accident conditions, } \tag{2}$$

where τ is one week.

Compliance with these release limits may be demonstrated by direct measurement. In the event this is not practical, the standard provides an alternative method for demonstrating containment adequacy that evolves from the definitions of R_N and R_A. The alternate approach relates measurable gas leak rates, L_i (cm^3/s), to the appropriate activity release rates, R_i, through the expression

$$L_i = R_i/C_i \text{ , } (i = N \text{ or } A) \text{ , } \tag{3}$$

where C_i represents suspended radioactivity (in curies/cm^3 in the cask medium) that could escape from the containment system during transport.

Although ANSI N14.5 is quite prescriptive for measuring and correlating leak rates, little guidance is given regarding the determination of the activity concentrations C_N and C_A. When C_N and C_A cannot be definitively established, the "leak tight" design criterion is required.

Our source term objective is to develop methodologies for defining C_N and C_A for all releasable activity associated with spent fuel. It is important to note that our intent is to develop effective methods to assure compliance with the regulations pertaining to containment, and to have a sound, comprehensive technical basis for those methods. Our goals are to provide a consistent and well documented approach to demonstrating containment compliance, and to assure that the result of the approach minimizes exposure to the general public and the radiation worker population to the extent reasonably possible.

FACTORS AFFECTING THE RELEASABLE ACTIVITY IN A SPENT FUEL SHIPPING CASK

The activity concentration or source term, C_i, available in a loaded spent fuel transport cask consists of radionuclides which can be released from three sources: (1) any residual activity on internal cask cavity surfaces resulting from loading and/or previous transport operations; (2) fission and activation-product activity associated with radioactive material deposited on the surfaces of fuel assemblies (CRUD); and (3) the fission and activation product radionuclides contained within the individual rods comprising the spent fuel assemblies. These individual sources are additive, as illustrated by Equation (4):

$$C_i = C_{RC_i} + C_{CR_i} + C_{F_i} \text{ , } \tag{4}$$

where

C_{RC} = residual contaminants contribution
C_{CR} = CRUD contribution
C_F = fuel contribution
i = N for normal conditions of transport
i = A for accident conditions of transport.

Each of these three distinct sources has differing concentrations, physical and chemical forms, characteristics impeding release, and release mechanisms. Estimates of the contributions of each are treated in the following two papers.

Cask Contaminants and Crud

Consideration of the potential for escape of crud or contaminant deposits on fuel assemblies or cask surfaces begins with estimates of the total amount of material present and its physiochemical form. This total inventory defines the maximum amount of non-fuel activity that could be released into the biosphere in the event of a breach in the integrity of a cask. If crud in the cask were the only source of activity, a conservative upper bound on L_i of Equation 3 could be established by determining possible maximum values of the gas-borne activity concentration, C_i, for each isotope associated with the crud inventory. Conservative limits on C_i, together with the upper limits imposed on R_i by 10 CFR Part 71, result in an acceptable upper bound on L_i, the volumetric gas leak rate, under normal and accident conditions. To make a more realistic estimate of L_i, one must develop a more mechanistic picture of particle behavior in the cask, and the time history of C_i. To do this, one must consider C_i as a function of the following:

(1) The total crud activity inventory. The activity inventory on the fuel surfaces is a function of the fuel type (PWR or BWR), fuel age, isotope composition, and reactor system chemistry. The residual cask surface activity is a function of previous fuel characteristics and cask operational history.

(2) The cask design. Cask cavity dimensions and configuration, number and type of fuel assemblies that the cask may transport, surface area of cask cavity, basket, and fuel rods, etc.

(3) Crud spallation fractions and initial particle size distribution of dispersed crud. These are in turn functions of the normal and hypothetical accident conditions of transport, the fuel type, and the reactor history.

(4) Diffusive deposition and gravitational settling on cask cavity surfaces, fuel rod surfaces, fuel basket surfaces, and other hardware surfaces in the cask cavity. These are in turn dependent on aerosol residence time, crud particle size distribution, and cask design.

(5) The transport conditions causing the release for containment evaluations. These are the normal and accident conditions as specified in 10 CFR Part 71.

Spent Fuel

The spent fuel/cask configuration may be thought of as a multibarrier confinement and containment system. For a release to occur, radionuclides contained in the spent fuel pellets must first be in a dispersable condition, a forcing function is necessary to initiate and maintain radionuclide transport, and some form of barrier release pathway must exist or be initiated.

Thus, a radiological consequence in the environment external to a cask from activation and fission products contained in spent fuel can only occur if radionuclides are first somehow released from the spent fuel to the cask cavity and subsequently escape from the cask cavity to the external environment. The first phase is governed by the multibarrier characteristics of the spent fuel and the second phase is governed by the characteristics of both the material released to the cask cavity and the pathway through the cask to the external environment. For the purpose of this analysis, the pathway through the cask is that required to restrict the potential release within the limits, R_N and R_A, for normal and accident conditions, respectively. This corresponds to definitive containment limits as defined in terms of limiting leak rates L_i given by Equation (3)

Figure 1 is a logic network for tracing the potential consequences of an initiating event, commonly called a fault or event tree. The upper most "event," which is simply "Radioactive Material Released to Cask Cavity from Spent Fuel," can

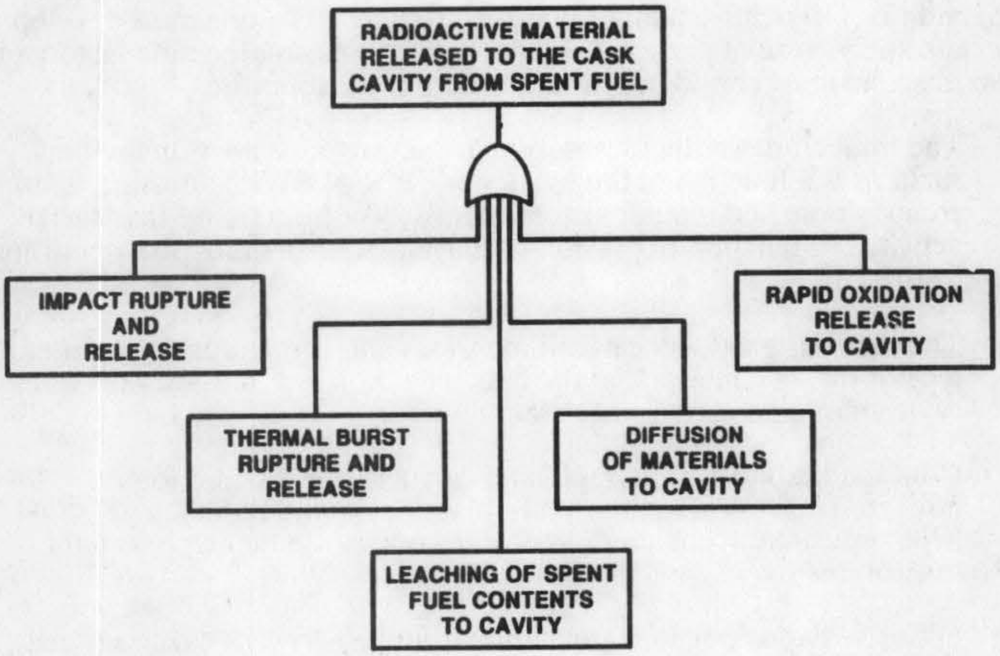

Figure 1. Release Scenarios Affecting the Fuel Contained Source Term in a Spent Fuel Transport Cask

occur as a result of one of five different "mechanisms." Each is associated with a particular initiating event that is brought about by some environmental condition. The physical mechanisms which can cause release of radionuclides from the spent fuel are: (1) impact failure of intact fuel cladding; (2) burst rupture of intact fuel cladding; (3) diffusion of vaporized fission products from rods with defected cladding; (4) leaching of soluble fission products from rods with defected cladding in contact with water or from water-logged rods; or (5) rapid fuel oxidation in severely failed fuel rods (IAEA 1987).

Each of the above mechanisms can be investigated further to identify subevents and parameters that may influence how each is initiated and maintained. For example, impact rupture of a sound (i.e., pressurized) spent fuel rod is relatively easy to understand: the release of radioactive material occurs as a result of mechanical disruption of the cladding and subsequent depressurization of the fuel rod. As a result of the impact, small particles of the fuel (fuel fines) may exist or be generated with a size and mass distribution such that some particles are transported to the cavity by the venting gases. The fuel rod venting will also release fission product gases and, depending on temperature, volatile species contained in the void regions of the fuel rod. Environmental conditions are defined by the regulatory tests for normal and accident conditions of transport. As noted above, these conditions represent prescriptive performance requirements. One task involves determining the fuel response to these conditions, which in turn depends largely on the cask response.

The physical, chemical, and radioactive characteristics of the radionuclide products contained in the spent fuel are important because they affect not only the releasability of particular species, but also the radiological implications of specific products as characterized by the A_2 values assigned to each. Releasability is a qualitative measure of the ease with which a particular species can migrate across pellet and clad barriers to a release point under varying pressure and/or temperature forcing functions or during impact. Physical and chemical characteristics can affect both the initiation of species transport and the transport process. For example, while many fission products are gaseous or volatile, bonding within the fuel matrix is such that in the event of a cladding breach only a fraction of the total gaseous or vaporized inventory may be transported across the cladding even under pressurization.

Both the failure mode and the failure threshold for the cladding depend on initial conditions of cladding, the loading condition experienced by the fuel assembly, and internal pressure. Depending on these initial conditions, the cladding may exhibit ductile or brittle behavior and rupture may occur as a result of crack initiation followed by through-the-wall propagation or by propagation of a pre-existing crack. The response behavior depends on a number of initial condition variables such as temperature, irradiation history, the presence of pre-existing partial through-the-wall flaws such as cracks resulting from pellet-clad interactions, or embrittlement caused by hydriding.

The loading conditions experienced by fuel depend on the cask design, i.e., how the cask responds to the impact event and how the forces involved are translated through the cask to the fuel. The loads experienced will range from severe impact conditions over short time durations to the long duration, time-varying accelerations experienced during normal transport.

Some percentage of the rods will have "failed" during in-reactor service, venting internal pressures to equilibrium with reactor or storage system pressures. Interestingly, from a source term perspective, there is a benefit gained because (1) much of the highly mobile gaseous and volatile fission products are gone, and (2) the potential high-pressure source for transporting products to the cask cavity is also no longer available. Thus, in an impact condition, dispersion can only occur as a result of local mechanical responses.

In some cases, previously "failed" fuel may become water-logged during in-reactor service, i.e., the differential pressure between the rod and reactor system pressure is sufficient to force some amount of coolant into the pellet-clad gap regions. Hydriding of the cladding can occur when hydrogen gas, liberated from the moisture in the fuel, reacts with cladding to form zirconium hydrides, resulting in localized embrittlement. While such embrittlement can significantly reduce the impact failure criteria for the rod, the failure consequences are offset by the absence of the high-pressure mobilizing force present in rods with intact cladding.

Burst rupture of a fuel rod is initiated by high internal rod pressures generated by a severe thermal environment. As a rod is heated by either excessive internal heat generation or as a result of an external heat source such as a fire, the internal pressure of the rod will increase until a local failure threshold is exceeded. The cladding then yields, and burst rupture occurs at the location of a stress concentration or a pre-existing flaw.

The sequence and governing conditions for burst rupture failure are qualitatively similar to those for impact failure. As in the impact case, for failure to occur, rod failure criteria that are a function of initial fuel conditions and the translation of the cask response to input conditions on the fuel must reach some threshold values. The heat load experienced by the cask will range from the extreme accident fire environment to the normal heat load experienced during shipment, which depend on ambient conditions, internal heat generation rates, and the heat transfer design of the cask. Burst rupture typically results in a localized hole that has a diameter approaching a few millimeters. The release mechanism (pressure driven) is the same as the impact release from an intact rod. The release products, however, will likely include more volatile products.

Leaching of fission products by water or steam is another mechanism for release of fission products from spent fuel. Its importance for future transport in the United States is much less than for previous scenarios because future shipments are expected to require a dry inert gas atmosphere in cask cavities. Water may be present as a result of operational error prior to shipment, in water-logged fuel, or by gross containment failure. In the event of high heat input, sufficiently water-logged fuel could generate and release steam, possibly carrying fission products in the exiting steam. Direct contact between water and spent fuel pellets can result in contact leaching of fuel products which can later be released via steam or, in the event of gross water in-leakage, be carried directly throughout the cask cavity.

Previously failed or ruptured fuel rods can also release fission products by a diffusion process. Once a spent fuel rod is ruptured, and after the initial release transient remaining vaporized fission products can diffuse through the fuel to the gap region and out of a breach point. The likelihood of diffusion occurring increases with temperature.

Fuel oxidation as a release mechanism is included here primarily for completeness. The events and sequences required for fuel oxidation to be feasible fall in the realm of low probability ("extra regulatory") extra severe events which lie beyond the range of the regulatory conditions and thus, beyond the scope of this analysis. Oxidation can only be initiated on a large scale after severe disruption of fuel cladding followed by a severe thermal environment with the fuel immersed in flowing air or steam. Thus, for cask cavities filled with inert gases, rapid oxidation is credible only if (1) gross operational error occurs prior to shipment, or gross containment failure results in an exchange of the inert cavity gas for air, and (2) gross mechanical failure of the fuel cladding occurs and, (3) a high-temperature environment exists.

Following a release from the spent fuel, the mobility or mass transport characteristics of the released material will govern how the material will disperse throughout the cask cavity and to a containment release point. The internal movement of the material occurs under the influence of gradients in temperature, pressure or concentration.

Particles released will have a size and mass distribution; gases will have some density distribution. To reach the external environment through a containment leak path, radioactive gases must diffuse through the cavity gas and particles must remain gas-borne long enough to cross the shortest cavity void space and be small enough to be able to pass through the leak path, all before the containment release transient subsides and equilibrium conditions are reached. For particles, diffusive deposition and gravitational settling processes in transit will significantly reduce the particle source term exiting the containment. The attenuation capabilities of these processes are dependent on the aerosol resident time, particle size and mass distribution, and the fuel and cask design. Influencing design parameters include the total surface area in the cask cavity, the cask orientation and the gravitational settling area described by that orientation, and the gas void volume of the cask and fuel.

CONCLUSION

The development of an integrated source term methodology for the containment analysis of spent fuel transport casks is not a trivial task. A realistic estimate of an acceptable containment limit requires a mechanistic description of release mechanisms for both crud and other contaminants that are exterior to the fuel and activation and fission products contained within the fuel. The concentration of radionuclides in a cask available for release through a pathway to the external environment is a function of the following:

> Cask design parameters such as cavity dimensions, and configuration, number and type of fuel assemblies that the cask may transport, surface area of the cask cavity and fuel rods, etc.
>
> The total radionuclide inventory within the cask. The available crud inventory depends on cask operating history, fuel type, fuel age, isotope composition, and reactor system chemistry. The spent fuel inventory depends on the fuel type and its burnup, initial enrichment, and age or decay time since discharge.

The physical, chemical, and radioactive characteristics of the radioactive materials contained within the cask.

The environmental condition initiating a release. These conditions are the regulatory performance requirements for normal and accident conditions of transport.

The response of a cask to the regulatory environmental conditions and how that response translates to forcing functions on the cask contents.

The fuel response to initiating events. This is determined primarily by fuel design, type of initiating event, material characteristics and constitutive relationships, and fuel history and pre-transport condition.

Spallation fractions and particle size distributions of dispersed crud and other surface contaminants. These, in turn, depend on initiating conditions, fuel type, and history.

The mechanisms and pathways for release of radionuclides from fuel pellets to gap regions and the cask cavity. These depend on fuel history and structure, release mechanism, material properties, and many other variables.

The mobility and mass transport characteristics of the radionuclides contained within the cask. This depends on numerous parameters including release mechanism, inventory, particle size distribution, diffusive deposition and settling characteristics, aerosol residence time, and cask design.

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