Proceedings of the 15th International Symposium on the
Packaging and Transportation of Radioactive Materials
PATRAM 2007
October 21-26, 2007, Miami, Florida, USA

Source Terms for Spent Fuel Transportation and Storage Cask Evaluation

Robert E. Einziger Ph.D.
Division of Spent Fuel Storage and Transportation
United States Nuclear Regulatory Commission

ABSTRACT

NUREG/CR-6487 and Interim Staff Guidance (ISG)–5 provide guidance on source terms to use when evaluating the ability of spent fuel storage and transportation casks to meet containment regulations in 10 CFR Part 71 and 72. These source terms, while intended to be bounding are based only on the properties of lower burnup (<45 GWd/MTU) fuel. The guidance is based on the same number of failed fuel rods, and breach sites per rod for both storage and transportation casks, while in fact the normal and accident conditions are quite different. Currently fuel is being driven to much higher burnup (current limit is 62.5 GWd/MTU peak rod average) that has changed the properties of the fuel. The change in the mechanical properties of the cladding will increase the potential for multiple fracture sites in a rod. The formation of a fuel rim at higher burnup may change the propensity of the fuel to fracture under impact and the resulting particulate size distribution. The analysis in this paper accounts for fuel fracture that occurs during an accident event and removal of particulate from the cask cover gas prior to the gases release from the cask. This paper reevaluates the particulate release fractions using high burnup fuel properties. Uncertainties in the release fractions due to uncertainties in the fuel properties data base are also evaluated.

INTRODUCTION

10 CFR Parts 71 and 72 have requirements for release limits of radioactive material that relate to the leak tightness of the cask or its containment capability [1, 2]. ISG-5 has recommended source terms for applicants to use when determining if their system meets the requirements [3]. These source terms are based on NUREG/CR-6487 [4] which used the properties of lower burnup fuel (<45 GWd/MTU) and does not account for depletion of radioisotopes after they are released from the fuel rod but before they are swept out of the cask through the leak site. In addition, these source terms neglect additional potential sources of release such as fracturing of the fuel pellet into fines and release of additional fission gases and volatiles to the gap during an accident. The same source terms were developed for both normal and accident conditions but did not account for the differences in conditions between storage and transportation.

There are four types of releasable radionuclides contained within a fuel rod: 1) CRUD (the only radionuclide source that does not require breach of the fuel rod cladding) on the outside surface of the fuel rod, 2) fission gases released from the fuel pellet to the rod void space during irradiation, 3) volatiles such as Cs that usually are released concurrently to the void space with the fission gases, and 4) fuel particulate that has either formed in the reactor due to abrasion of the pellets and thermal gradients, or during an impact accident such as a dropped cask. Each of these sources contains its own mix of radionuclides and must be individually treated.

In recent years, utilities have been driving fuel to higher and higher burnup for economic purposes. Burnup ≤ 62 GWd/MTU has been approved for LWR reactors by the NRC. To have satisfactory in-reactor performance the fuel vendors have been changing the properties of the fuel pellet including the addition of burnable poisons, and the use of newer cladding alloys that have a greater corrosion resistance. Coolant water chemistry has been adjusted to minimize CRUD. These changes to the fuel rod design and higher exposures have led to changes in the post-irradiation properties of the fuel rod that may affect the magnitude of the potential release source term.

Even though the rate of corrosion has been reduced in the newer cladding alloys, the higher exposure in some of these alloys has resulted in significantly more hydrogen pickup, and the potential for the cladding to become significantly more brittle if hydride reorientation occurs during the drying process. At high burnup the fuel pellet has swelled so there is virtually no fuel/cladding gap and the fission gas release (FGR) has accelerated so that instead of an FGR of 1-3% like in lower burnup fuel there is now 8-12% [5]. The fuel pellet has formed a rim of submicron size grains on its external surface. This rim can be as large as 100 μm in PWR fuel and 150 μm in BWR fuel. This rim region retains its fission gas in large voids and has an actinide inventory approximately double that of the center of the fuel [5]. While this rim is only 4-6% of the fuel volume, its ultra small grain size is already in the respirable range and may have different fracture distributions of particle sizes than the larger grains comprising the bulk of the fuel. Therefore in any analysis of the source term for high burnup fuel, the rim and body of the fuel pellet must be treated as separate entities.

This paper reevaluates the particulate release fractions using high burnup fuel properties for 0.3 m and 9 meter side drops of a cask undergoing no thermal transient. Uncertainties in the release fractions due to uncertainties in the fuel properties data base are also evaluated.

RELEASE FRACTION MODEL

The original classified model was developed by Sprung for analyzing security events. An unclassified model was developed and discussed in detail as an appendix to the NRC Storage cask PRA [6]. In this paper the model will be used to determine the release rate from a cask containing high burnup fuel that is dropped on its side. A vertical drop, where fuel can settle on the grid spacers, will have a larger depletion rate. The release rate is per rod with the assumption that the leak in the cask is small enough so that the rod depressurizes rapidly upon cladding breach compared to the depressurization of the cask. This decouples the release from the rod and the release from the cask. In addition it is assumed that all the energy from the fall is transmitted to the rods and none is absorbed by impact limiters if they are present. This set of assumptions should give the largest source terms. The model is in a form that it can be exercised with the removal of these assumptions if so desired.

The thickness of the rim layer (t_{rim}) in high-burnup BWR spent fuel is about 150 µm [5]. PWR rods have a somewhat thinner rim at an equivalent burnup. Because the body and the rim of high-burnup spent fuel pellets have quite different morphologies [5], release of fuel fines from these two regions of a high-burnup pellet may be quite different. Like fresh UO_2 pellets, the body regions of high-burnup spent fuel pellets consist of sintered 10 µm UO_2 particles. In the rim layer, 0.1 to 0.3 µm subgrains are generated by the recrystallization of UO_2.

The rim layer has 1.3 to 2 times the concentration of actinides compared to their concentration in the pellet body [6]. For a typical BWR rod about 11.3 percent of the total radionuclide inventory in the pellets resides in the rim layer of these pellets and the remaining 88.7 percent of the total inventory is contained in the body of the pellets. Accordingly, for BWR rods:

$$F_{RC,k} = (0.113)(\text{rim release fraction}) + (0.887)(\text{body release fraction}) \qquad (1)$$

where $F_{RC,k}$ is the release fraction from the rod to the cask for isotope k. A similar equation can be written for PWR fuel. The release fraction for particles is calculated as shown in Equation 2 below, using the fractional releases from three locations within he rod:

(1) the rim of the pellet located under the rod failure site and ejected during the event

(2) the rims of other pellets (pellets not located right next to the rod failure)

(3) on the surfaces of the network of internal pellet cracks caused by abrasion, vibration, and impact fracturing

$$F_{RC,particles} = \{0.113(F_{init,ri}+F_{imp,r})(n\,F_{tear} + F_{ent}\,F_{bed}) + 0.887\,n\,(F_{init,b} + F_{imp,b})(F_{bed})\}(1-f) \qquad (2)$$

where:

F_{init} = the fraction of the mass of the UO_2 fuel pellets (body or rim) in a rod that has been converted to respirable fuel fines by abrasion and vibration before the impact event takes place.

F_{imp} = the fraction of the mass of the UO_2 in the body or rim of a spent fuel pellet that is converted to respirable fuel fines by brittle fracture as a result of the impacts.

F_{tear} = the fraction of the mass of the UO_2 in the rim layer that is blown out of the rod from one rod tear during rod depressurization without filtering by passage through a particle bed.

F_{ent} = the fraction of the mass of the UO_2 in the rim layer that is entrained in the depressurization gas flow through the rim layer-cladding gap and transported to the rod tear and out into the cask.

F_{bed} = the fraction of the respirable particles that are not captured during flow through a particle bed

n = the number of tears in an average failed rod in the cask.

f = fraction of particulate that settles out of the gas in the cask prior to release from the cask

EVALUATION OF TERMS

In-reactor pellet fracture (F_{init})

Abrasion during insertion of the pellets into the rod and vibration during reactor operation causes fuel fines to form on pellet external surfaces and also on pellet crack surfaces, especially at the pellet-pellet interfaces. Lorenz [7] heated lengths of low-burnup PWR fuel to burst and measured the amount and size of the particulate emanating from the burst site. The mass fraction of respirable fines that escaped the rod is estimated by Lorenz, from the mass of fuel that stayed in the collection tube and the amount of fuel that escape the tube in an airborne fashion, to be 4×10^{-6} Due to the size of the burst opening and the 99% filtering efficiency of a particle bed for respirable particles, Sprung calculated [8] that the fraction of the fuel that was fines is ($F_{int,b}$) was 2.4×10^{-5}. A similar value is not available for the rim region since no tests similar to Lorenz's using high-burnup fuel have been conducted to our knowledge. Since the rim occurs after pellet abrasion has stopped $F_{int,rim} = 0$.

Impact fracture of pellets (F_{imp})

The DOE Handbook [9] gives a formula for F_{imp}, the fraction of a brittle material that is converted to respirable fines by impact fracturing. Comparing this equation to experimental data for the impact fracturing of a variety of brittle materials including depleted UO_2 and average burnup spent fuel pellets, but no high-burnup spent fuel pellets, shows that the handbook equation lies about a factor of 10 higher than the experimental data. Accordingly, in order to be somewhat conservative, for average burnup spent fuel pellets and for the body of high-burnup spent fuel pellets; the leading coefficient in the DOE Handbook equation is reduced by a factor of five, which gives $F_{imp,body} = 4\times10^{-12}$ E/V where E/V is the energy, imparted to the fuel by the drop, per unit volume of the brittle material that is subject to fracturing. The units of the leading coefficient are cm^3/erg.

None of the material used to develop this relationship was similar to the rim structure in high-burnup fuel (i.e., very fine grained material, already in the respirable range, having large pores containing high-pressure gas). Making assumptions about the pore energy, Sprung calculated that the rim fracture and the fracture of the body should be approximately equal. This is supported by German microhardness measurements [10]. By contrast, anecdotal evidence from attempts to prepare metallurgical samples of the rim indicated that this region is much more friable than the body of the fuel. At best, it can be concluded that there is a value of high uncertainty that can range over several orders of magnitude. This uncertainty must be accounted for in any release determination. In the current study, the fraction of the rim is being allowed vary from a low equal to the same fraction as the body of the fuel to a high of complete fracture.

Rim tear (F_{tear})

If rod failure causes a plug of fuel fines to be blown out of the rod tear from the possibly friable rim layer located next to the tear, F_{tear} can be estimated from the equation: $F_{tear} = w \ell d / \pi d_p t_{rim} L$ where w, ℓ, and d are the final width, length, and depth of the ejected rim materials and d_p, t_{rim} and L are the diameter of the fuel pellets, the thickness of the pellets friable rim layer, and the active length of the spent fuel rod (the length of the pellet stack in the rod). After the ejection of a rim layer plug, the depressurization flow of rod gases over the sides of the hole left by plug ejection should entrain more rim material into that flow, which will increase the final width of the total mass of rim material lost by plug ejection plus entrainment. Since defect sizes are unknown, an estimate must be made. For an Atrium 10 ×10 BWR rod having a half circular defect with a 10/1 aspect ratio, $F_{tear} = 2.8\times10^{-4}$.

Entrainment in gas flow ($F_{ent,}$)

If during storage the rod cladding separates slightly from the pellet rim, a small rim layer-cladding gap might open along the length of the rod. If flow through this rim layer-cladding gap during rod depressurization is significant, some particles formed from rim materials might become entrained in this flow and, if not captured by particle bed filtering, be carried out of the rod to the cask interior. In actuality, entrainment may be insignificant, since any reopened pellet-cladding gap will be clogged with large particles formed by impact fracturing (mechanical or shock loading), i.e., the large particles will interfere with the boundary layer flow needed to entrain particles off of surfaces, and high burnup fuel usually doesn't have a pellet-cladding gap remaining [5]. Using entrainment values for similar density materials, Sprung found that the uncertainty in the calculations led to the physically impossible entrainment fraction greater than 1 [6]. Because $F_{ent,}$ could not be larger than 1 but could be significantly less with no gap, F_{ent} will be varied from 0 to 1 in this evaluation. $F_{ent,}$ should be set to 1 to obtain a maximum release fraction.

Particle filtering by rubble bed (F_{bed})

F_{bed} is the fraction of the respirable particles that are not captured during flow through a particle bed. Aerosol physics [11] was used to estimate that a 0.3-cm long bed (l) of 200 µm particles [14] would have a filtering efficiency 0.99 for the maximum respirable size particles. Assuming 100% release from $2l$ is the length of the rod from which respirable particles escape with only partial (0 to 0.99) filtering (i.e., the particle bed length is too short to provide significant filtering) and 1% from 2L is the length of the rod from which respirable particles escape but are filtered as a result of passage through particle beds, it can be shown that: $((0.99)l/L +0.01) = F_{bed}$. Depending on the estimate of L, which should depend on "n", F_{bed} can range from 0.01 to 0.02 for n=1 to 5. Assuming $L = 10l$ or ~ 3 cm filtering bed, then $F_{bed} =0.1$. The ramifications of this uncertainty are discussed later.

Cladding fracture sites (n)

Due to Zircaloy cladding being pushed to higher burnup and to the use of new cladding alloys, such as Zirlo, there is higher cladding hydrogen content. The fracture strains of these higher burnup claddings are not well known therefore the number of cladding fracture sites during an impact is uncertain. Up to 5 fracture sites have been estimated for lower burnup fuel [12]. Therefore, "n" will be varied from 1 to 5 for the current evaluation.

In cask settling of particulate (f)

Because gas flow rates out of the failed rods are high, removal of particles by inertial impaction onto spacer plates or rod surfaces located next to cladding failures may occur, provided particle sticking probabilities are significant at the particle impact speed. Those particles not sticking as a result of impaction may settle before reaching the cask opening. Since gas flow velocities through the failed cask will be slow, particle removal during transport through the cask to the location of the cask=s closure failure will occur by Brownian and turbulent diffusion to cask interior surfaces and by gravitational settling onto upward facing interior surfaces. Removal of the larger UO_2 particles (UO_2 particles with geometric diameters ≥ 3.16 μm) will occur primarily by gravitational settling, while small particles may be removed primarily by Brownian and turbulent diffusion.

Sprung [6] calculated the typical impingement distance from a rod breach to the next nearest surface for a variety of pre-2004 cask designs and concluded that that the largest size respirable particulate released from a rod would impinge on the neighboring surface. However, the impact velocity would be so great that the sticking coefficient for UO_2 particles is most likely # 1H10$^{!5}$ Few particles that impact the spacer plates will stick to the plates or an adjacent rod surface, therefore, only gravitational settling will be considered further for depletion.

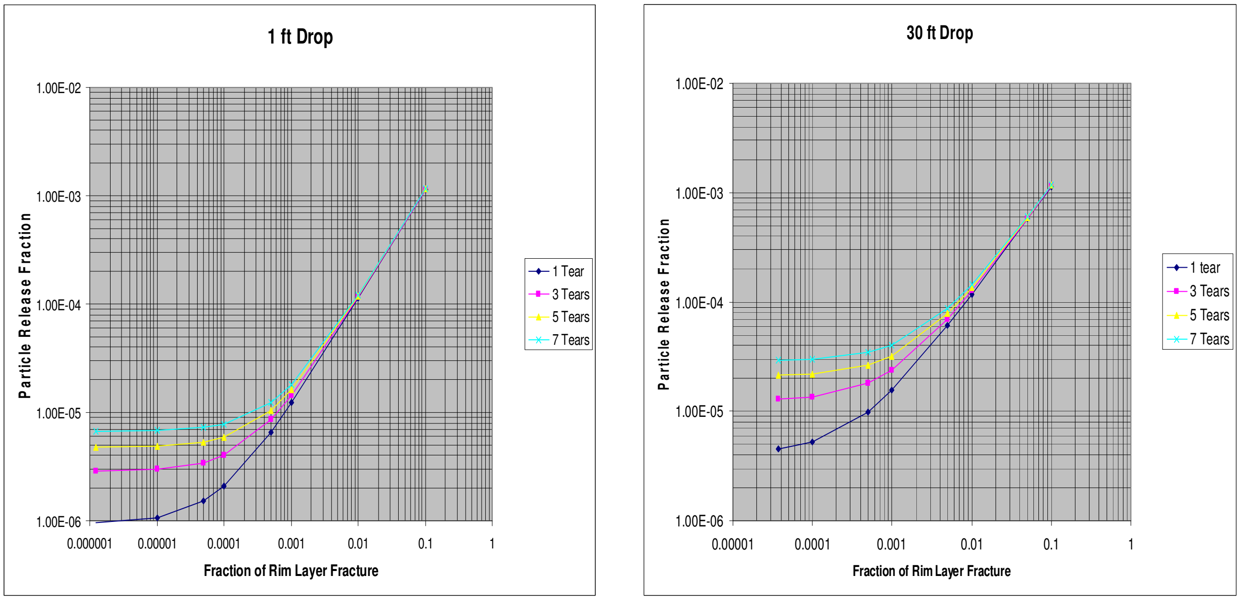
During horizontal cask depressurization, one side of each of the spent fuel assembly basket tubes and half of the outer surface area of the spent fuel rods in each tube will be facing upward and will be surfaces onto which particles can deposit by gravitational settling. The force on the particulate in the gas stream attributable to the gas flow is in the horizontal direction, while the gravitational force is in the vertical direction. If the time to traverse the distance to the adjacent rod or basket surface is less than the time for the particle to be swept to the breach site, the particle can be considered to be gravitationally settled. Under the relatively quiescent environment that will prevail during the passage of rod gases and fission products from rod failures to the cask closure failure, any respirable UO_2 particles not removed by inertial impaction should encounter a horizontal surface before they exit the cask. For 3.16 μm UO_2 particles at their gravitational settling impact speed the sticking coefficient is ∃0.9 [13], $F_{\text{gravitational settling}} = F_{\text{surface}}F_{\text{stick}} = (1.0)(0.9) = 0.9$.

ENVELOPE OF SOURCE TERMS

The large uncertainty in the release fraction for a given impact is from the uncertainty in the number of fracture sites in the cladding due to the unknown fracture response of the hydrogen-containing high burnup cladding (1-5), extent of the fracture of the rim region under impact (1×10^{-6} to 1), amount of rim material that can be entrained in the depressurizing rod gas stream (0 – 1), and the filtering efficiency of the particle bed (0.01 – 0.1). Plots of release fractions, as a function of the fraction of the rim that is fractured on impact, is shown in Figure 1 for 1 ft and 30 ft drops. The term that dominates will depend on the relative magnitude of the rim fracture. In this case, once 1% of the rim fractures the release rate is independent of the extent of rim fracture due to the domination of the rim fines not at the fracture site that are entrained in the gas flow. At lower rim fracture this term becomes less dominant. Lower entrainment will lower the release fraction nominally (factor of 5) until the entrainment is reduced to below 10% at which point it reduces the release fraction significantly.

Based on Otani [11], the model used in this paper assumes 99% filtering for any fines arising more than 0.3 cm from the breach site, and no filtering closer to the breach site. The farthest that a fine can travel is half the distance between breach sites (180 cm for n=1 and 30 cm for n=5). If a large travel path from particulate origin to breach site is available then there is more efficient filtering and the release fraction is reduced. A bed of 3 cm was chosen for this analysis resulting in $F_{bed} = 0.1$ or 90% efficiency. The total release fraction $F_{RC,\ particles}$ can drop as much as an order of magnitude as F_{bed} decreases from 0.1 to 0.01.

Figure 1- Release Fraction as a Function of the Amount of Rim Fractured by Impact. 100% of the fuel is entrained and $F_{bed} = 0.1$. $F_{imp,b}$ is 4×10^{-5} for 30 ft and 1×10^{-6} for 1 ft.



The release terms from the rods (no including deposition in the cask) are tabulated in Tables 1 and 2. The source term includes deposition in the cask. As long as there is not complete rim fracture and complete entrainment, then the respirable source term is at or below the currently recommended value of 3×10^{-5}. For 1 to 5 breaches, complete entrainment and equal rim and body impact fracture, a source term of 1×10^{-7} to 5×10^{-7} should be typical of lower burnup fuel that has no rim and still maintains a fuel/cladding gap. The source term for accidents should be a factor of 4-5 higher. The currently recommended value of 3×10^{-5} is higher because it; 1) does not account for depletion in the cask (90%), and 2) uses the full release fraction not the respirable release fraction calculated by Lorenz. [1]

Table 1. Release fractions from rods from 1 foot drop – Normal transport

Breaches	1	1	1	1	5	5	5	5
Entrainment	0	0	1	1	0	0	1	1
Rim fracture	B	C	B	C	B	C	B	C
Rod release fraction	1E-06	3E-05	1E-06	1E-02	5E-06	2E-04	5E-06	1E-02
Source term/rod	1E-07	1E-06	1E-07	1E-03	5E-07	2E-05	5E-07	1E-03

Entrainment = none (0) or complete (1), Rim fracture = complete (C), or same as the body (B)

Table 2. Release fractions from rods from 30 ft drop – HAC Condition

Breaches	1	1	1	1	5	5	5	5
Entrainment	0	0	1	1	0	0	1	1
Rim fracture	B	C	B	C	B	C	B	C
Rod release fraction	4E-06	4E-05	5E-06	1E-02	2E-04	2E-04	2E-05	1E-02
Source term/rod	4E-07	4E-06	5E-07	1E-03	2E-05	2E-05	2E-06	1E-03

Entrainment = none (0) or complete (1), Rim fracture = complete (C), or same as the body (B)

Since high burnup fuel has almost no gap and will continue to have a smaller gap with increased burnup, the tendency will be to have a lower and lower entrainment of the fuel in the gas stream as the rate of depressurization through the smaller gap decreases. An entrainment closer to zero should be used. On the other hand there may be a propensity towards more cracks per rod as the cladding becomes more brittle with additional in-rector oxidation infusing more hydrogen into the cladding. This is especially the case if drying for storage results in extensive radial hydride reorientation. Probably one should consider only 5 breach sites for both normal and accident drops.

The major uncertainty in the source term for the high burnup fuel particulate is the degree of fracture of the rim. If there is both complete fracture of the rim into respirable particles and complete entrainment of these particles, then the source term is 2 orders of magnitude higher than currently recommended. Fortunately the entrainment is probably much lower in high burnup fuel due to the narrowing gap. Recent work [14] using microindentation and porosity theory points towards the rim fracture being very similar and possibly even less than the fracture of the larger grained body of the fuel under similar impacts. The impact used to calculate the fracture of the fuel assumes all the energy is transmitted to the rods. Impact limiters on the cask should reduce the energy transmitted to the rods and lower the fracture propensity. If in fact the rim fracture is not excessive, then values for the source term for normal transport of high burnup fuel are probably in the range of 10^{-7} to 10^{-6}. Similarly for accidents the source term is probably in the range of 2×10^{-5}.

CONCLUSIONS

The properties of high burnup fuel have been applied to the source term analysis method developed by Sprung to calculate source terms for a horizontal transport cask carrying high burnup fuel. In general the currently recommended value (3×10^{-5}) for the source term is an upper bound. There is reason to believe it could be lowered by 1-1.5 orders of magnitude for lower burnup fuel (≤45 GWd/MTU). Except in the unlikely case that there is complete fracture of the pellet rim into submicron size grains, the current recommended source term is suitable for high burnup fuel during accidents and an order of magnitude high for normal transport conditions. More information on the fracture of the rim region is needed before new source term values can be recommended.

REFERENCES

1. 10CFR71 Code of Federal Regulations 10 CFR Part 71, Packaging and Transportation of Radioactive Material, p 278, Jan 2006
2. 10CFR72 Code of Federal Regulations 10 CFR Part 72, Licensing Requirements for the Independent Storage of Spent Nuclear Fuel, High- Level Radioactive Waste, and Reactor-Related Greater than Class C Waste, p 335, Jan 2006
3. NRC Spent Fuel Project Office Interim Staff Guidance -5 Rev1, Confinement Evaluation, http://www.nrc.gov/reading-rm/doc-collections/isg/spent-fuel.html
4. BL Anderson, RE Carlson, and LE Fischer, Containment Analysis for Type B Packages Used to transport Various Contents, NUREG/CR-6487, UCRL-ID-124822. LLNL, Nov 1996
5. RE Einziger and CE Beyer, Characteristics and Behavior of High Burnup Fuel that May Affect the Source Terms for Cask Accidents, *Nuclear Tech*, Aug 2007
6. NUREG-1864, A pilot Probabilistic Risk Assessment of a Dry Storage Cask Storage System at a Nuclear Plant, March 2007, USNRC. Washington, DC
7. Lorenz, R.A., et al., “Fission Product Release from Highly Irradiated LWR Fuel,” NUREG/CR-0722, Oak Ridge National Laboratory, Oak Ridge, Tennessee, pp. 48–80, February 1980.
8. JL Sprung et.al., Reexamination of Spent Fuel Shipment Risk Estimates NUREG/CR- 6672, March 2000
9. U.S. Department of Energy, “DOE Handbook: Airborne Release Fractions/Rates and Respirable Fractions for Non-Reactor Nuclear Facilities,” DOE-HDBK-3010-94, Section 4.3.3, “Free-Fall Spill and Impaction Stress,” Washington, DC, December 1994, p. 4-52.
10. W Goll, et.al., UO_2 Fuel Behavior at Rod Burnups to 105 MWd/gHM, Atw52 Jg. (2007) HEFT 2-Febrruar, p 95
11. Otani, Y., *Aerosol Science & Technology,* 10:463, American Association for Aerosol Research, Mt. Laurel, New Jersey, 1989.
12. TL Sander, KD Seager, YR Rashid, PR Barrett, AP Malinauskas, RE Einziger, H Jordan, TA Duffy, SH Sutherland, and PC Reardon, A method for Determining the Spent-Fuel Contribution to Transport Cask Containment Requirements, Sandia Report, SAND-2406, November 1992.Sandia report
13. Beal, S.K., “Correlations for the Sticking Probability and Erosion of Particles,” *Journal of Aerosol Science*, 9:455, Elsevier, Inc., Burlington, Massachusetts, 1978.
14. J Spino, J.Cobos-Sabate, and F Rousseau, Room temperature microindication behavior of LWR-fuels, Part 1: fuel microhardness, J. Nucl. Mater. **322,** (2003), 204-216

---

[1] It should be noted that the release fractions in ISG-5 are intended to be the complete fraction of fuel released from the rod irrespective of particle size. Since only airborne particles can escape the cask and, as shown [6] any particle larger than respirable will settle in the cask due to the relatively small leak rate, the respirable release rate and the total release rate should be the same.