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ABSTRACT

Representative accident source terms patterned after the NUREG-1465 Source Term have been developed for high burnup fuel in BWRs and PWRs and for MOX fuel in a PWR with an ice-condenser containment. These source terms have been derived using nonparametric order statistics to develop distributions for the timing of radionuclide release during four accident phases and for release fractions of nine chemical classes of radionuclides as calculated with the MELCOR 1.8.5 accident analysis computer code. The accident phases are those defined in the NUREG-1465 Source Term – gap release, in-vessel release, ex-vessel release, and late in-vessel release. Important differences among the accident source terms derived here and the NUREG-1465 Source Term are not attributable to either fuel burnup or use of MOX fuel. Rather, differences among the source terms are due predominantly to improved understanding of the physics of core meltdown accidents. Heat losses from the degrading reactor core prolong the process of in-vessel release of radionuclides. Improved understanding of the chemistries of tellurium and cesium under reactor accidents changes the predicted behavior characteristics of these radioactive elements relative to what was assumed in the derivation of the NUREG-1465 Source

Term. An additional radionuclide chemical class has been defined to account for release of cesium as cesium molybdate which enhances molybdenum release relative to other metallic fission products.

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NOMENCLATURE

ATWS	anticipated transient without scram
BWR	boiling water reactor
CDF	cumulative distribution function
DC	direct current
ECCS	emergency core cooling system
GWd/t	gigaWatt-days per ton of heavy metal
HBU	high burnup reactor fuel
LBU	low burnup reactor fuel
LEU	low enrichment uranium dioxide fuel
LOCA	loss of coolant accident
MIMAS	micronized master blend method for MOX fuel fabrication
MOX	mixed oxide fuel
NRC	Nuclear Regulatory Commission
PWR	pressurized water reactor
RCIC	reactor core isolation cooling

1. INTRODUCTION AND BACKGROUND

1.1 Regulatory Use of Source Terms

The use of postulated, accidental releases of radionuclides is an important feature of the regulatory practices and policies adopted by the U.S. Nuclear Regulatory Commission (NRC) in pursuit of a defense-in-depth safety philosophy. The reactor site criteria (10 CFR Part 100) require for the purposes of licensing nuclear power plants that radionuclide releases to reactor containments associated with a "substantial meltdown" of the reactor core be postulated. The consequences of these radionuclide releases are evaluated assuming that the containment remains intact and leaks at the design-basis leak rate. Radionuclides that leak from the containment are termed the "radiological release to the environment." The magnitude of the radiological release to the environment can be estimated from the containment leak rate and the radionuclide inventory suspended in the containment atmosphere as a function of time. The radionuclide inventory suspended in the containment atmosphere depends on the amount released to the containment as well as the effectiveness of natural and engineered processes that lead to radionuclide deposition within containment. The postulated radionuclide release to the containment is termed the "in-containment source term." It is this in-containment source term that is the topic of this report.

The nuclear power plants currently operating in the country were licensed originally based on "in-containment source terms" specified in Regulatory Guides 1.3 [NRC, 1974a] and 1.4 [NRC, 1974b]. These specifications were derived from the descriptions of accidental radionuclide releases found in the 1962 report TID-14844 [DiNunno *et al.*, 1962]. Following the reactor accident at Three Mile Island, which did involve a "substantial meltdown" of the reactor core, the U.S. Nuclear Regulatory Commission launched a major research initiative to better understand the likely releases of radionuclides to the containment in the event of accidents that progressed well beyond the design bases of nuclear power plants. An important result of the research was establishing the relationships among radionuclide releases and the details of accident progression. The research effort culminated in evaluations of accident risks at five selected types of nuclear power plants [NRC, 1990]. Results obtained in this assessment of accident risks as well as the results of an enormous body of source term research done both in the USA and elsewhere in the world were used to formulate an alternative to the postulated source terms used in the past [Soffer *et al.*, 1995]. The alternative has come to be known as the "NUREG-1465 Source Term". This alternative was adapted into regulatory practices of NRC through Regulatory Guide 1.183 [NRC, 2000].

In contrast to the TID-14844 source term, the NUREG-1465 Source Term recognizes that the radionuclide releases during a severe accident would take place over substantial periods. Four phases of radionuclide release are addressed – the "gap release phase", the "in-vessel release phase", the "ex-vessel release phase", and the "late in-vessel release phase." Furthermore, the NUREG-1465 source term takes into account a greater diversity of radionuclide behavior than was done in the TID-14844 source term.

There are, of course many different radioactive elements in reactor fuel as well as activation products produced in reactor core structures. These many elements make varying contributions to the radiological consequences of reactor accidents. Comparisons of the relative contributions made by different radioactive elements to short-term and long-term consequences are shown in Table 1 [Alpert, *et al*., 1986]. The comparisons are made for equal release fractions of various elements and the results are normalized to those of iodine for short term effects and to cesium for long-term effects. Bolded entries in the table indicate elements that can produce consequences commensurate with those of either iodine or cesium.

To account for elements that have the potential of producing significant consequences, the NUREG-1465 source term defines eight chemical classes of radionuclides rather than the three physical classes considered in the TID-14844 source term. These chemical classes are shown in Table 2. Different fractional releases of each chemical class of radionuclide are specified for each of the sequential phases of radionuclide release to the reactor containment. Timing of the phases and release fractions are taken to be different for pressurized water reactors (PWRs) and boiling water reactors (BWRs). The NUREG-1465 source term highlights the importance of radionuclides released to the containment as aerosol particles. At the same time, the NUREG-1465 source term recognizes the complexity expected of iodine chemistry within containment including the formation of gaseous molecular iodine and volatile organic iodides as well as particulate metal iodides.

The NUREG-1465 source term, based as it is on substantial experimental and analytic research, is a more realistic representation of radionuclide release to containment associated with a "substantial meltdown" of a reactor core as envisaged in the reactor site criteria (10 CFR 100). Though the alternative source term was developed for "future" light water reactors, it has been endorsed by the NRC for use in regulatory processes involving currently operating nuclear power plants. The source term has found a variety of applications beyond evaluation of site suitability [Lee, 2006]. Applications include definition of the post-accident environment for equipment qualification, post-accident habitability of the reactor control room and leakage specifications for main steam isolation valves. Timing features of the

Table 1. Relative importances of various radioactive elements in reactor fuel [Alpert, *et al*., 1986].

	Early Exposure* (normalized to iodine)			**Long-term Exposure** (Normalized to cesium
Element	**4 hr. bone marrow dose**	**24 hr. bone marrow dose**	**Lung dose**	**Latent cancers**
Co	0.007	0.008	0.01	0.07
Kr	0.2	0.1	0.04	0.001
Rb	0.0002	0.0002	0.0002	0.00001
Sr	**1.0**	**0.7**	**0.9**	**0.7**
Y	0.07	0.07	**3.5**	0.4
Zr	**1.0**	**1.0**	**2.0**	**0.7**
Nb	0.3	0.3	0.4	0.2
Mo	0.1	0.1	**0.7**	0.06
Tc	0.02	0.03	0.03	0.06
Ru	0.3	0.3	**3.0**	**1.0**
Rh	0.01	0.01	0.08	0.004
Sb	0.06	0.06	0.1	0.004
Te	**0.8**	**0.8**	**0.6**	0.1
I	**1.0**	**1.0**	**1.0**	0.1
Xe	0.02	0.01	0.005	0.0001
Cs	0.15	0.14	0.09	**1.0**
Ba	**0.6**	**0.5**	**0.6**	0.2
La	**1.1**	**1.2**	**1.6**	0.08
Ce	0.1	0.2	**8.0**	**2.0**
Pr	0.004	0.003	0.8	0.08
Nd	0.03	0.03	0.3	0.03
Np	**1.6**	**1.4**	**5.0**	0.04
Pu	0.004	0.003	**1.4**	**3.0**
Am	0.002	0.001	0.01	0.03
Cm	**0.6**	0.4	**5.0**	**1.1**

* early exposure via cloud, inhalation, and either 4 or 24 hours of groundshine

Table 2. Chemical classes of radionuclides adopted in the NUREG-1465 source term.

Chemical Group Name	Elements in the Group
Noble Gases	Kr, Xe
Halogens	Br, I
Alkali Metals	Rb, Cs
Tellurium Group	Se, Sb, Te
Barium, Strontium Group (Alkaline Earths)	Sr, Ba
Noble Metals	Ru, Rh, Pd, Mo, Tc, Co
Lanthanides	La, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am
Cerium Group	Ce, Pu, Np, Zr

alternate source term have been especially useful in the regulatory review process.

1.2 Insights Since Publication of the Alternative Source Term

Since publication of the NUREG-1465 alternative source term, research into the behavior of radionuclides under reactor accident conditions has continued. Notable undertakings include the PHÉBUS-FP project [Simondi-Teisseire and March, 2008] to investigate radionuclide release from degrading, irradiated, reactor fuel, transport of the released radionuclides through a simulated reactor coolant system and behavior of radioactive particles and vapors in a simulated PWR containment. There have also been studies of aerosol transport through steam generators with either wet or dry secondary sides [Güntay *et al*., 2008], iodine chemistry under accident conditions, and mitigation of aerosol production during the "ex-vessel release phase" of severe accidents.

Phébus-FP experiments have shown substantial radionuclide release from degrading reactor fuel in the form of aerosols. But, these tests have shown also that some fraction of the radioactive iodine is released to the containment in gaseous as well as particulate form much as has been anticipated by the NUREG-1465 source term. Furthermore, the tests have shown that much of the cesium released from degrading fuel is not present as CsOH. Rather, it is present as some other, less volatile, species – evidence suggests cesium molybdate.

There have been substantial improvements in the computational resources available for prediction of radionuclide release and behavior during reactor accidents. The MELCOR computer code [Gauntt, *et al*., 2000] has been developed by the NRC to consolidate the numerous specialized codes

[Gieseke *et al*., 1986] that were used to produce the risk assessments of five representative nuclear power plants (NUREG-1150) and the technical bases for the NUREG-1465 source term. The consolidated computational vehicle is more reliable and there have been many opportunities to further validate and refine the predictions of the progression of reactor accidents including the releases of radionuclides to the containment. Notable developments have been refinements in the prediction of core debris relocation within the reactor core during fuel degradation and a better quantification of heat transfer from degrading fuel to reactor coolant system structures.

At the same time, there have been evolutions in the operations of nuclear power plants in the country. It is becoming increasingly common for reactor fuel to be used to burnups in excess of 40 GWd/t. (Note that fuel burnup values cited in this document are assembly averages and not peak pin values.) As burnup increases, the fuel undergoes changes including the development of the so-called “rim effect” [Lee and Jung, 2000]. High burnup fuel is known to affect the behavior of fuel during reactivity transients [Meyer, *et al*., 1996]. The high porosity and fission product concentration in the “rim” region could be expected to affect the radionuclide release during core damaging accidents. For these reasons, the authors of the NUREG-1465 Source Term cautioned against using the source term for high burnup fuel.

There are also proposals in the USA to use mixed oxide fuel (MOX) composed of mixtures of uranium dioxide and plutonium dioxide as fuel for pressurized water reactors with ice condenser containments under the US-Russia Excess Weapon-grade Plutonium Disposition Program. Mixed oxide fuel has been used extensively in Europe, but there is much less experience with this fuel in the US nuclear industry and the US regulatory process. There is evidence to suggest that fission product releases from mixed oxide fuel will not be the same as release of radionuclides from low-enrichment uranium dioxide fuel more commonly used in nuclear power plants in the USA. [Ashbaugh, *et al*., 2005]. There may be changes in the nature of high temperature core degradation when MOX interacts with cladding during reactor accidents that progress to the point of substantial meltdown [ERI, 2002]. Again, the authors of NUREG-1465 cautioned against using the source term for analysis of accidents at plants fueled with MOX.

2. OBJECTIVES AND SCOPE

2.1 General

The primary objective of this report is to define alternative accident source terms for regulatory applications that can be applied to reactors that use fuel to burnups in excess of 40 GWd/t or reactors that use MOX fuel. At the same time, development of these source terms affords an opportunity to ascertain how improved understanding of severe reactor accidents and improved abilities to predict the progression of such accidents including prediction of radionuclide release to the containment could affect derivation of regulatory source terms appropriate for accidents involving substantial meltdown of the reactor cores.

The source terms developed here are patterned after the source terms developed in NUREG-1465. They are expressed in terms of the timing and rates of appearance of various chemical classes of radionuclides in the containment. That is, the source terms account for the retention of radionuclides in the reactor coolant system. They do not account for the effects of natural and engineering processes that will lead to radionuclide deposition in containment and removal from the inventory of airborne radionuclides available to leak from the containment to the environment.

The source terms developed here are not intended to be bounding or otherwise deliberately conservative. They are intended to be representative of the source terms to the containment for the class of accidents that involve substantial meltdown of the core.

The source terms developed here are not expected to be definitive for all future light water reactors. Technically justifiable changes may be made in source term parameters (timing, release fractions and chemical form) if warranted by specific features of a reactor.

Finally, it is important to note that in this work, no effort was made to account for the state-of-knowledge or epistemic uncertainty in the source term. The intention of the work is to develop source terms that reflect the current state of knowledge much as the NUREG-1465 source term reflected the state of knowledge when it was developed. Aleatory uncertainty regarding the nature of reactor accidents that could lead to significant core melting and release of radionuclides to the containment is considered.

2.2 Approach

The approach adopted here for the development of representative source terms follows to a significant extent the strategy adopted in the development of the NUREG-1465 Source Term. It is assumed that source terms for high burnup fuel in BWRs and PWRs should be distinguished. At this time, a MOX source term is needed only for ice-condenser containment PWRs. It is now planned in the USA to use MOX fuel only in PWRs with ice condenser containments as part of the program to dispose of excess weapons grade plutonium dioxide. Radionuclide releases to the containments of a selection of plants during a variety of accidents of regulatory interest are calculated. The computational vehicle for these calculations is the MELCOR 1.8.5 systems-level accident analysis computer code [Gauntt, *et al*.,2000]. Releases to the containment are segmented into the four periods adopted in the development of the NUREG-1465 source term:

- **Gap Release Phase**: The release, following clad rupture, of fission gases and radionuclide vapors that accumulate during operation in the fuel-cladding gap and fuel rod plenums.
- **In-vessel Release Phase**: The release of radionuclides during core degradation and prior to core debris penetration of the reactor vessel.
- **Ex-vessel Release Phase**: The release of radionuclides from core debris expelled from the reactor vessel into the reactor cavity following failure of the bottom head of the reactor pressure vessel. This release is predominantly due to releases during core debris interactions with concrete but could include releases caused by high pressure melt expulsion phenomena and even ex-vessel steam explosions.
- **Late In-vessel Release Phase**: This phase of release starts once core debris has penetrated the reactor vessel. Radionuclide releases come from fuel that has not been expelled from the reactor vessel and revaporization of radionuclides that have been deposited on surfaces within the reactor coolant system.

Before this sequence of accident phases involving the intense release of radionuclides can begin, the water in the reactor coolant system must be expelled. In the case of accidents initiated by breaks in large coolant lines, the water inventory is depleted quickly. For accidents initiated by small breaks in the reactor coolant system or by transient events such as station blackout, depletion takes place mainly by boiling driven by the decay heat in the core. For these

accident sequences, there can be a very substantial delay between accident initiation and the intense release of radionuclides addressed here and in the NUREG-1465 Source Term.

Coolant expelled following accident initiation will contain some amount of radioactive material. The concentrations of radionuclides in the coolant are limited by plant Technical Specifications or other regulatory measures. Consequently, any release of radioactivity to the containment brought on just by the expulsion or boiling of the coolant will be minuscule relative to the releases that begin with the Gap Release Phase of an accident. The coolant releases are typically ignored in source term analyses as they are here.

The delay between the initiation of an accident and the onset of radionuclide releases during the Gap Release Phase and the In-vessel Release Phase of an accident is a regulatory issue. This issue is explored thoroughly in NUREG-1465. A prescription that meets the needs of the regulatory process is described in NUREG-1465. The prescription is not dependent on the burnup of the fuel and need not be altered should MOX fuel be used in a reactor core. Consequently, the treatment of delay between accident initiation and the onset of release described in NUREG-1465 is adopted here and the issue is not further explored.

The objective definition of a representative source term requires some appreciation of the distribution of source terms that are possible. To develop such distributions, it is assumed that releases calculated for each accident phase constitute an independent, random sample of the distribution of releases that could occur during the particular accident phase for accidents involving substantial melting of the reactor core and release of radionuclides to the containment. Manifestly, the accidents analyzed here are not random samples. As discussed in Section 3 of this document, the accidents were selected deliberately because they were of regulatory interest and figure in the risk profiles of the plants considered in the analysis. The assumption is, then, a first approximation that allows progress in the development of distributions.

Cumulative distribution functions (CDFs) were developed from the accident analysis results using nonparametric order statistics [Hogg and Craig, 1978]. The distributions of interest here are, of course, unknown and may not be simple. The underlying probability density functions could be bimodal or even multimodal in character. That is, the density functions for release fractions and timing could have more than one local maximum if dominating processes vary among the accident sequences analyzed. Imposition of arbitrary functional forms on the distributions derived from the accident analyses is avoided by the use of nonparametric statistical analyses. Nonparametric methods place equal weight on each of the sampled values of timing or release fractions derived from accident analysis. The nonparametric methods as applied here yield quantiles associated with probabilities of 0.05 to 0.95 in steps of 0.05. The distributions were refined using a bootstrap resampling of the accident analysis results and

reanalysis using the nonparametric methods. This was done to better characterize the uncertainties in the quantiles based on the mean and standard deviation of their respective order statistics.

Example distributions obtained in the analysis of accidents involving substantial core melting in ice-condenser PWRs using MOX fuel are shown in Figure 1 and 2. Figure 1 shows the cumulative distribution function (CDF) estimated for the duration of the In-vessel Release Phase of the accidents. Figure 2 shows the estimated cumulative distribution function (CDF) for the fraction of the initial core inventory of halogens released to containment during the In-vessel Release Phase of the accidents. In both figures, the cumulative distribution functions are shown as probability plotted against quantiles of the uncertain output from accident analyses. Because small numbers of accidents were analyzed (See Section 3.), the cumulative distributions can only be determined to some arbitrary level of confidence. Instead, uncertainty bounds on the distributions were determined using a "bootstrap" resampling with replacement [Efron, 1981] and are one standard deviation on either side of the estimated location of the quantile at each level of probability. About 10,000 iterations of the bootstrap refinement were used. Random numbers for the resampling were obtained using a Mersenne twister algorithm that has a very long period and performs quite well for most of the tests in the DIEHARD test program [Matsumoto, *et al.*, 1988]. In general, bootstrap uncertainty bounds found this way will not be symmetrical. The symmetrical uncertainty bounds adopted here are first approximations that are fairly accurate for the medians where skew in the resampling results is small. Symmetrical uncertainty bounds become less accurate at the probability extremes (approaching probability 0 and 1) where both skew and kurtosis of the resampling results become large.

The median is taken to be the representative value of the source term distribution. Selection of the median avoids over-emphasis of source terms from large break loss of coolant accidents that would have occurred inevitably if the mean of the distribution had been selected as representative. Similarly, selection of the median rather than the mean avoids having the timing results being dominated by the slower developing accidents such as station blackout events. The nonparametric methods adopted here do, in fact, weight equally each of the accident analysis results used to formulate a distribution for timing or for release fraction. This would not be the case for the mean. A percentile other than the median as the representative magnitude would require justification from some other source. By adopting the median, half the accidents have larger release fractions and longer release times than the representative accident and half have smaller release fractions and shorter release times.

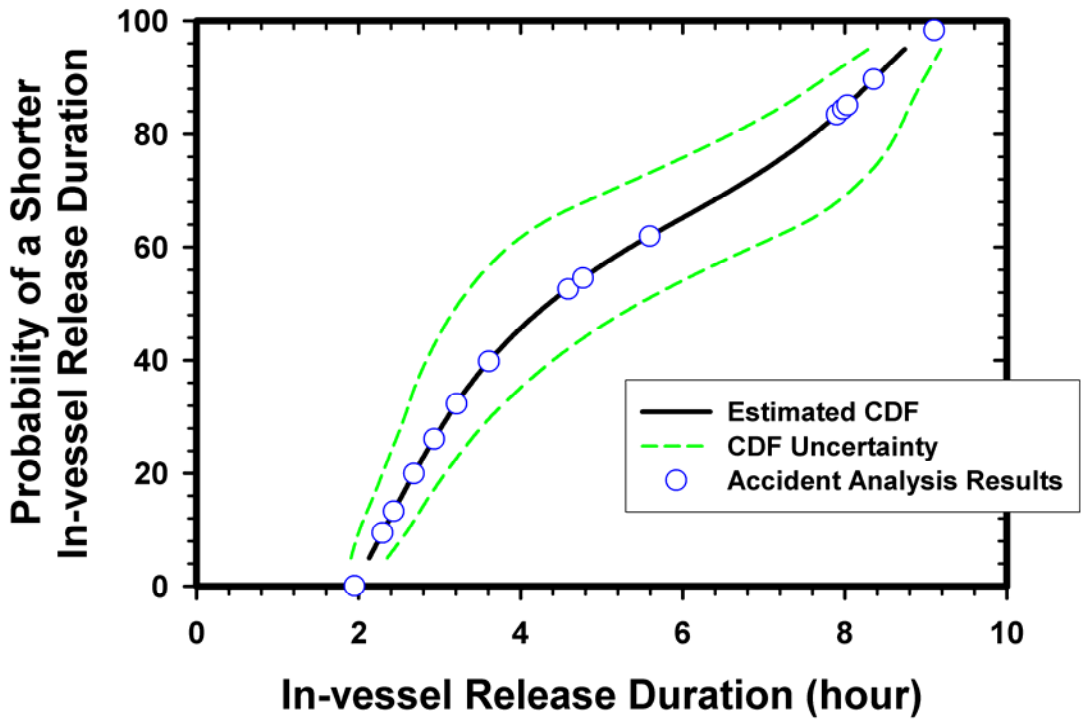

Figure 1. Example distribution of the duration of in-vessel release during accidents in an ice-condenser containment using MOX fuel. Results of accident analyses are shown as circular points. The estimated cumulative distribution function (CDF) obtained by nonparametric analysis and bootstrap resampling is shown as a solid line. Uncertainties in the quantile locations are indicated by dashed lines located one standard deviation on either side of the estimated cumulative distribution function.

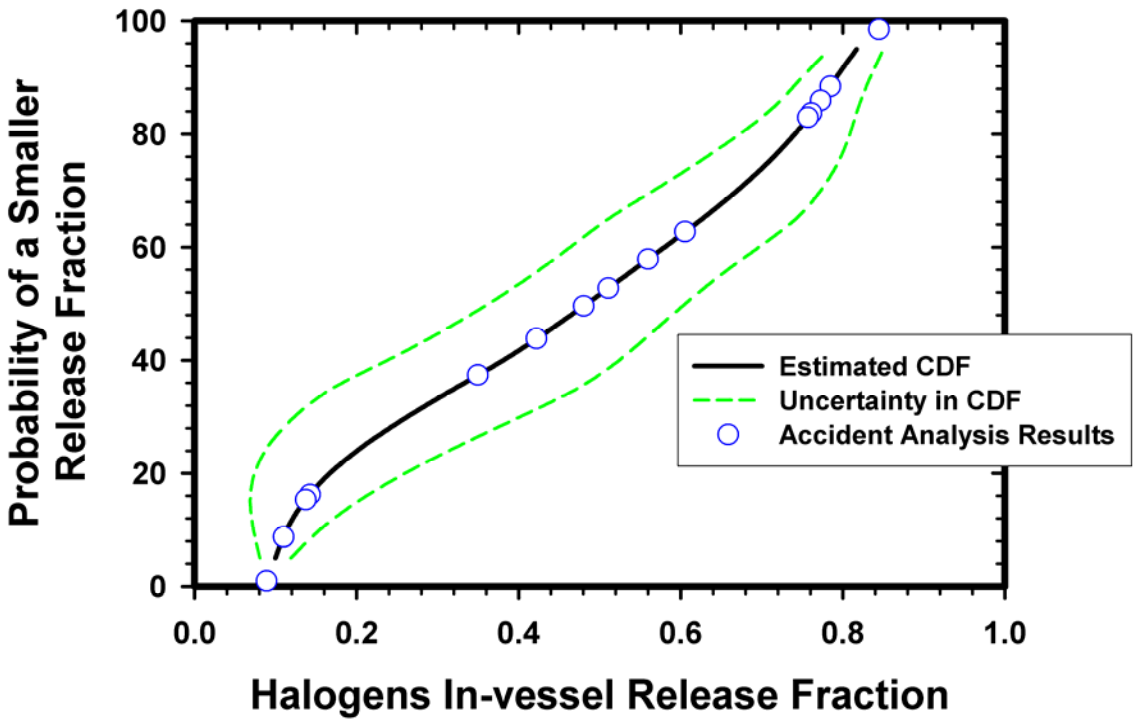

Figure 2. Example distribution of the halogen fraction released to containment during the in-vessel release phase of accidents in an ice-condenser containment using MOX fuel. Results of accident analyses are shown as circular points. The estimated cumulative distribution function (CDF) obtained by nonparametric analysis and bootstrap resampling is shown as a solid line. Uncertainties in the quantile locations are indicated by dashed lines located one standard deviation on either side of the estimated cumulative distribution function.

2.3 Limitations

The results provided here have been obtained based on accident analyses done with the MELCOR 1.8.5 computer code (Gauntt, *et al*., 2000). These results reflect the current understanding of severe accident progression and source term processes as this understanding is embodied in the computer code. No attempt has been made to systematically quantify the state of knowledge uncertainty (epistemic uncertainty) in the results. As noted in more detail in the discussion of the results, the models in MELCOR for radionuclide escape from fuel were modified to reproduce data for high burnup fuel and for MOX fuel. Data were not available to modify gap inventories. Also, data are not available to suggest that degradation of high burnup fuel or MOX fuel differ from predictions of MELCOR models of fuel degradation based on experiments with moderate burnup (<40 GWd/t), low-enrichment, uranium dioxide fuel. Consequently, core degradation was modeled in the analyses in the same way for low and high burnup fuel and for low-enrichment uranium dioxide fuel and MOX fuel.

Source terms are, of course, products of the physics and chemistry of core degradation accidents which, in turn, depend very much on the nature of the reactor. The results presented here have been obtained for light-water reactors using zirconium-alloy-clad, oxide fuel. These results should not be used in connection with source term analyses for non-light-water reactors such as graphite-moderated, gas-cooled reactors using TRISO fuel or sodium-cooled fast reactors with metal or oxide fuel. Also, results are not applicable to small modular reactors that could have accident processes that differ substantially from those of the large power plants considered here.

3. ACCIDENT SEQUENCES

3.1 Accidents Used for the High Burnup Source Term

To develop the source term for high burnup fuel, accidents were analyzed for both BWRs and PWRs. The BWRs considered were the Grand Gulf reactor with a Mark III pressure suppression containment and the Peach Bottom reactor with a Mark I containment. Summary descriptions of the BWR accidents are provided in Table 3. Station blackout accidents, transients involving loss of injection or loss of decay heat removal and breaks in the reactor coolant system are included. Together the accidents consider 92% of the core damage frequency expected for the Peach Bottom reactor and 98% of the core damage frequency expected for the Grand Gulf reactor [Office of Nuclear Regulatory Research, 1997]. Interfacing systems loss of coolant accidents are not considered because such accidents would not release large amounts of radionuclides to the containment.

Details of the accident analyses are provided by Leonard *et al.* [2007]. Data for the fuel assembly geometry, fuel mass, cladding mass, control poison mass, nose pieces and the upper and lower tie plate masses and materials were derived from vendor design information. These specifications differed for low and high burnup operations. At both Peach Bottom and Grand Gulf, GE 8x8 fuel assemblies were used for low burnup operations. For high burnup operations, Peach Bottom uses GE14C (10x10) fuel assemblies and Grand Gulf uses Atrium-10 (10x10) fuel assemblies.

Data concerning core axial and radial power, end-of-cycle fission product inventories and decay heat generation were obtained using plant operating data. Axial and radial profiles for low burnup and high burnup designs were developed from licensee data for three consecutive early cycles and three recent cycles. The core-average discharge burnup for the low burnup cycles ranged from 26 GWd/t to 38 GWd/t. The maximum assembly-average discharge burnup from the high burnup cycles was 45-48 GWd/t. ORIGEN [Croff *et al*., 1978] calculations of the fission product inventories extrapolated the actual level of discharge burnup from the high burnup core to the regulatory limit of 62 GWd/t for the highest exposure pin, which was reduced to 59 GWd/t on an assembly-average basis.

The core power data along with data regarding fuel enrichment, fresh fuel loading and partially burned assembly movement, and core power histories were used to perform ORIGEN calculations of fuel depletion and fission product generation. The ORIGEN calculations were performed on an assembly-specific basis for different axial locations along each assembly based on the axial power distribution. This provided spatially dependent fission product inventories and decay heat histories that were based on plant operational history.

Core-wide inventories of radionuclides are compared in Table 4.

Accidents in PWRs at the Surry and the Sequoyah reactors are summarily described in Table 5. Station blackout accidents, breaks in the reactor coolant system, transients, and reactor coolant pump seal failure accidents are included. Together, accident analyses were done covering 85% of the core damage frequency of Surry and 88% of the core damage frequency of the Sequoyah plant [Office of Nuclear Regulatory Research, 1997]. Interfacing systems loss of coolant accidents and steam generator tube rupture accidents were not included because they do not release large amounts of radioactive material to the containment.

Details of the accident analysis with the MELCOR computer code are provided by Ashbaugh *et al.* [2008]. Core design and performance information was obtained for several recent cycles in the PWR plants to reflect current assembly geometry and mass. Fuel loading patterns and assembly power and burnup histories were obtained in order to calculate representative spatial distributions of fission product inventories and the associated decay heat. Inputs for fuel assembly geometry, fuel mass, cladding mass, control poison mass, grid support material and mass and other important physical characteristics were developed based on the specific fuel design used in each PWR plant during early periods of reactor operation for low burnup calculation and current operations for high burnup operations. Sequoyah used Westinghouse 17x17 fuel assemblies for low burnup operations and Mark-BW 17x17 fuel assemblies for high burnup operations. Surry used Westinghouse 15x15 assemblies for low burnup operations and SIF 15x15 assemblies for high burnup operations.

End-of-cycle fission product mass inventories and decay heat information were based on plant-specific, cycle-specific nuclear design reports obtained from the licensees. For each PWR plant, three recent cycles were examined to ensure that significant cycle-to-cycle variations did not occur.

Radial power profiles for the low burnup cores were not provided by the licensees. Power profiles were obtained from LBU SCDAP/RELAP5 models developed at the Idaho National Laboratory for similar plants.

Axial power distribution data were not available from the licensees, but it was indicated that the axial profiles are reasonably consistent from plant to plant and cycle to cycle. Axial profiles were obtained from SCDAP/RELAP5 models developed at the Idaho National Laboratory for similar plants and these were compared to a generic PWR axial power profile [O'Donnell, *et al.*, 2001]. Licensees indicated that the axial profiles for PWRs did not vary significantly between low burnup and high burnup cores.

Decay heat and fission product inventories were calculated with the ORIGEN code [Croff, *et al*., 1978] assuming an assembly average discharge burnup of 59 GWd/t for high burnup operations and 28 GWd/t for low burnup operations.

Core-wide inventories of fission products are compared in Table 6.

For modeling the radionuclide releases from high burnup fuel, the MELCOR description of radionuclide releases from fuel were re-parameterized to better match results of the VERCORS RT-6 test of radionuclide release from high burnup fuel [Gauntt, 2010a, b]. This test showed more rapid release of volatile radionuclides such as cesium from high burnup fuel than had been observed in earlier tests with lower burnup fuel. As fuel temperatures increase, the release rates of volatile radionuclides from low burnup and high burnup fuel tend to converge.

Table 3. Accidents considered in development of source term for high burnup fuel in BWRs.

#	Containment	Accident Initiator	Other Failures	Containment Failure
1	Mark I	Short-term station blackout	No coolant injection; no DC power; low vessel pressure	Early liner melt
2	Mark I	Short-term station blackout	No coolant injection; no DC power; low vessel pressure	Early liner melt; high vessel pressure
3	Mark I	Short-term station blackout	No coolant injection; no DC power; low vessel pressure	Late head flange leakage
4	Mark I	Short-term station blackout	No coolant injection; no DC power; high vessel pressure	Early liner melt
5	Mark I	Long-term station blackout	RCIC operates; DC power lost after 8 hours; stuck open safety relief valve	Early liner melt
6	Mark I	Long-term station blackout	RCIC operates; DC power lost after 8 hours; stuck open safety relief valve	Late head flange leakage
7	Mark I	Long-term station blackout	RCIC operates; DC power lost after 8 hours; stuck open safety relief valve	Late over-pressure of torus
8	Mark I	Small break; relief valve 'tee'	No coolant injection; low vessel pressure	Early head flange leakage
9	Mark I	Recirculation suction line break	No coolant injection; low vessel pressure	Early dry-well liner melt
10	Mark III	Short-term station blackout	No DC power; no coolant injection; stuck open safety relief valve	Early; H_2 burn at vessel breach
11	Mark III	Short-term station blackout	No DC power; no coolant injection; high vessel pressure	Early; H_2 burn at vessel breach
12	Mark III	Short-term station blackout	No DC power; no coolant injection; stuck open safety relief valve	Late over pressure
13	Mark III	Long-term station blackout	RCIC operates; loss of DC power at 8 hours; stuck open safety relief valve	Early; H_2 burn at vessel breach
14	Mark III	Long-term station blackout	RCIC operates; loss of DC power at 8 hours; stuck open safety relief valve	Late over pressure
15	Mark III	ATWS	All coolant injection fails following containment failure	Prior to core damage
16	Mark III	Recirculation suction line break	No coolant injection; low vessel pressure	Late overpressure

Table 4. Radionuclide inventories for BWR accident analyses.

Chemical Class	Peach Bottom (3514 MW_{th})		Grand Gulf (3833 MW_{th})	
	LBU (kg)	HBU (kg)	LBU (kg)	HBU (kg)
Noble Gases	362	876	472	868
Halogens	14	34	18	33
Alkali Metals	208	506	274	497
Tellurium Group	33	82	43	80
Alkaline Earths	154	372	202	367
Noble Metals	498	1274	649	1246
Lanthanides	486	1240	642	1219
Cerium Group	1213	2281	1462	2254

Table 5. Accidents considered in the development of the source term for high burnup fuel in PWRs.

#	Containment	Accident Initiator	Other Failures
1	Subatmospheric	Station blackout	No ECCS; No auxiliary feedwater; induced RCP seal LOCA
2	Subatmospheric	Small break	No ECCS; Auxiliary feedwater available; late containment failure
3	Subatmospheric	Large break	ECCS injection
4	Subatmospheric	Station blackout	No ECCS; No auxiliary feedwater
5	Subatmospheric	Small break	No ECCS; auxiliary feedwater available; early containment failure
6	Ice condenser	Reactor coolant pump seal failure	No ECCS; auxiliary feedwater available; reactor cavity flooding
7	Ice condenser	Reactor coolant pump seal failure	No ECCS; auxiliary feedwater available
8	Ice condenser	Reactor coolant pump seal failure	ECCS injects; auxiliary feedwater available
9	Ice condenser	Station blackout	No ECCS
10	Ice condenser	Station blackout	No ECCS; no auxiliary feedwater
11	Ice condenser	Large break	No ECCS; auxiliary feedwater available
12	Ice condenser	Small break	No ECCS; no auxiliary feedwater

Table 6. Radionuclide inventories for PWR accident analyses.

Chemical Class	Sequoyah (3411 MW_{th})		Surry (2441 MW_{th})	
	LBU (kg)	**HBU** (kg)	**LBU** (kg)	**HBU** (kg)
Noble Gases	294	514	186	410
Halogens	11	20	7	16
Alkali Metals	164	289	102	372
Tellurium Group	27	47	17	37
Alkaline Earths	126	215	79	169
Noble Metals	210	367	132	145
Lanthanides	391	704	243	561
Cerium Group	973	1444	625	1085

3.2 Accidents Used for the MOX Source Term

In contrast to the NUREG-1465 source term and the source terms for high burnup fuel developed here, the MOX source term is developed for a particular type of plant and a particular application. MOX fuel is to be used in the Catawba reactors as part of a national initiative to dispose of excess weapons-grade plutonium. Consequently, release analyses were focused on these particular plants which are ice-condenser containment PWRs.

MOX fuel to be used in the reactors is prepared by the “MIMAS” or “micronized master” blend method. The product of this fuel production method consists of a refractory oxide fuel composed of mostly the dioxide of ^{238}U enriched with about 0.25% ^{235}U and 2 to 5% plutonium dioxide. The nominal isotopic composition of the plutonium dioxide is:

- ^{239}Pu 93.6%
- ^{240}Pu 5.9%
- ^{241}Pu 0.4%
- ^{242}Pu 0.1%

The blending process leaves most, though not all of the plutonium dioxide atomically dispersed in the uranium dioxide matrix rather than concentrated in isolated “islands” in fuel pellets. The fuel is to be clad with the M5 alloy which is a proprietary alloy of zirconium with niobium.

The reactor core has 193 assemblies of the 17x17 design. Each assembly contains 264 fuel pins. The specific power level is 38.7 kW/kg-heavy metal. The fuel cycle duration is 18 months.

At most 40% of the fuel assemblies will have MOX fuel pins. A proposed loading pattern for the MOX pins is shown in Figure 3 [Duke Power, 2001]. This loading pattern was assumed for the accident analyses used here. For the development of the MOX source term, end of cycle fuel is assumed. That is, there will be three levels of burnup among the assemblies:

- First cycle fuel burnup: 19.16 GWd/t
- Second cycle fuel burnup: 38.31 GWd/t
- Third cycle fuel burnup : 57.47 GWd/t

Expected radionuclide inventories at the end of cycle and immediately after shutdown are listed in Table 7. Also shown in the table for comparison are radionuclide inventories for a low enrichment uranium dioxide fuel at the end of cycle. These inventories were calculated using the ORIGEN2 computer code [Ludwig and Renier, 1989]. Because the mix of radioisotopes is not the same in the MOX core and the low enrichment uranium dioxide core, there are differences in the decay heat. Decay heats are compared in Table 8. Differences in decay heat will affect the progression of accidents and radionuclide release.

Accidents considered in the analysis are summarized in Table 9. Together the first 11 of these accidents represent about 73% of the risk estimated for the Catawba reactor [Duke Energy Corporation, 1992]. The large break loss of coolant accident is not a high frequency event, but figures prominently in the regulatory process that makes use of source term descriptions. Intermediate breaks of 6 and 10 inches are being considered as alternatives to the large break loss of coolant accident used in design basis accident analyses.

Details of the accident analyses using the MELCOR 1.8.5 computer code are provided by Ashbaugh *et al*. [2005]. For the analysis of radionuclide release from MOX fuel, the radionuclide release model in MELCOR was modified using data obtained from the VERCORS test RT-2 of radionuclide release from MOX fuel pellets [Ashbaugh *et al*., 2005]. Similar modifications of the MELCOR modeling to account for differences in the core degradation processes involving MOX relative to low enrichment uranium dioxide fuel could not be made because there are not data to form the basis for such modeling changes. Large differences in degradation of cores containing a fraction of MOX fuel and degradation of cores fueled with low enrichment urania fuel are not expected especially if some of the fuel has reached elevated levels of burnup (> 40 GWd/t). If differences in core degradation exist and if they affect fission product release, they are not reflected in the source term derived here.

1					L-2	M1	L-2	M1	L-2	M1	L-2				
2			L-2	L-1	M0	L-0	M0	L-0	M0	L-0	M0	L-1	L-2		
3		L-2	M0	M0	L-0	M1	M1	L-0	M1	M1	L-0	M0	M0	L-2	
4		L-1	M0	L-1	L-1	M1	L-0	L-1	L-0	M1	L-1	L-1	M0	L-1	
5	L-2	M0	L-0	L-1	M1	M0	L-1	L-0	L-1	M0	M1	L-1	L-0	M0	L-2
6	M1	L-0	M1	M1	M0	L-1	L-0	L-1	L0	L-1	M0	M1	M1	L-0	M1
7	L-2	M0	M1	L-0	L-1	L-0	L-1	L-0	L-1	L-0	L-1	L-0	M1	M0	L-2
8	M1	L-0	L-0	L-1	L-0	L-1	L0	L2	L-0	L1	L0	L-1	L0	L0	M1
9	L-2	M0	M1	L-0	L-1	L-0	L-1	L-0	L-1	L-0	L-1	L-0	M1	M0	L-2
10	M1	L-0	M1	M1	M0	L-1	L-0	L-1	L-0	L-1	M0	M1	M1	L-0	M1
11	L-2	M0	L-0	L-1	M1	M0	L-1	L-0	L-1	M0	M1	L-1	L-0	M0	L-2
12		L-1	M0	L-1	L-1	M1	L-0	L-1	L-0	M1	L-1	L-1	M0	L-1	
13		L-2	M0	M0	L-0	M1	M1	L-0	M1	M1	L-0	M0	M0	L-2	
14			L-2	L-1	M0	L-0	M0	L-0	M0	L-0	M0	L-1	L-2		
15					L-2	M1	L-2	M1	L-2	M1	L-2				

L-n = low enrichment uranium dioxide fuel burned n cycles
Mn = MOX fuel burned n cycles

Figure 3. Proposed loading pattern for MOX fuel assemblies in the reactor core.

Table 7. Comparison of radionuclide inventories for a 40% MOX core to those of a low enrichment uranium dioxide core.

Radionuclide Group	**Low enrichment uranium dioxide core** (kg)	**40% MOX core** (kg)
Noble Gases	517.8	488.2
Halogens	23.46	25.81
Alkali Metals	291.0	279.6
Tellurium Group	48.65	50.48
Barium, Strontium Group	220.0	190.1
Noble Metals	732.0 Mo = 372.8 Ru = 359.2	759.5 Mo = 350.0 Ru = 409.5
Lanthanides	699.6	641.0
Cerium Group	1489	2213

Table 8. Comparison of decay heat in a low enrichment uranium dioxide core and a 40% MOX core.

Time after shutdown (hours)	**Decay Heat** (MW)	
	Low enrichment uranium dioxide core	**40% MOX core**
0.0	204	197
1.0	45.1	43.7
2.0	36.4	35.3
12	22.6	22.2
24	18.6	18.3

Table 9. Accidents at an ice-condenser containment PWR considered in development of a source term for a reactor with a 40% MOX core.

#	Accident Initiator	Other Failures	Containment Failure
1	Station Blackout	Reactor coolant pump seal failure; loss of auxiliary feedwater after 3 hours	Late
2	Station Blackout	Reactor coolant pump seal failure; loss of auxiliary feedwater after 3 hours	Early
3	Station Blackout	Reactor coolant pump seal failure; loss of auxiliary feedwater immediately	Late
4	Station Blackout	Reactor coolant pump seal failure; stuck open safety relief valve; loss of auxiliary feedwater after 3 hours	Late
5	Station Blackout	Loss of auxiliary feedwater immediately	Late
6	One inch break in cold leg	ECCS recirculation fails	Late
7	One inch break in cold leg	ECCS recirculation fails	Early
8	One inch break in cold leg	Prompt ECCS failure	Late
9	One inch break in hot leg	ECCS recirculation fails	Late
10	Two inch break in cold leg	ECCS recirculation fails	Late
11	Two inch break in cold leg	Prompt ECCS failure	Late
12	Six inch break in cold leg	Prompt ECCS failure	Late
13	Six inch break in hot leg	Prompt ECCS failure	Late
14	Ten inch break in cold leg	Prompt ECCS failure	Late
15	Ten inch break in cold leg	Prompt ECCS failure	Late
16	27.5 inch break in cold leg	Prompt ECCS failure	Late

4. RESULTS

4.1 High Burnup Fuel

Source term results derived from calculations of accidents at plants with high burnup fuel are summarized in Tables 10 and 11 for boiling water reactors and pressurized water reactors, respectively. Also shown for comparison purposes in these tables are the results obtained for the same plants and accidents assuming that the fuel was of lower burnup. Values listed in these tables are the medians of cumulative distribution functions derived from the accident analysis results. Uncertainties associated with entries in the tables refer only to the uncertainty of locating the medians of distributions derived using nonparametric statistics applied to a small set of accident analyses. That is, the uncertainties listed in the tables do not refer to overall distribution of source terms calculated for the various accidents.

The most striking feature of the results is that there are not large differences between results for high burnup fuel and lower burnup fuel. What differences are noticeable can be explained by differences in decay heating and uncertainties in the locations of the medians of the distributions.

There are, however, definitely differences between the results calculated for this work and what would be obtained applying the NUREG-1465 source terms for BWRs and PWRs. These differences can be seen in the comparisons presented in Tables 12 and 13. These differences are discussed for the various phases of the accidents in the subsections that follow. The differences are due predominantly to the improved understanding of severe accident progression and radionuclide release that has developed since the NUREG-1465 Source Terms were derived from accident analyses using the Source Term Code Package [Gieseke *et a*l., 1986]. The accident analysis model used for work reported here (MELCOR) has benefitted from substantial progress in both analytic and experimental research since publication of NUREG-1465.

4.1.1 The Gap Release Phase

Modern analyses of core degradation do not produce an identifiable time period that can be exclusively ascribed to “gap release”. A period of “gap release” can be identified in experiments involving the degradation of a single rod or a few closely spaced rods. When a single fuel rod or a small bundle of fuel rods is heated in steam, a point is reached at which the zirconium alloy cladding on the fuel balloons and ruptures due to the internal pressurization and loss of cladding strength at elevated temperature. The gap inventory of noble gases (Kr, Xe) and vapors (typically various chemical forms of iodine and cesium) vent when the

clad ruptures. Venting of the gap inventory is not quite instantaneous along the fuel rod because of flow resistance in the fuel/cladding interface. The venting depletes the gap and the fuel rod plenum as well as the voids and pores in the fuel pellets that are open to the fuel/cladding gap.

Following this burst of radionuclide release associated with clad rupture, rates of radionuclide release from the fuel are low until the cladding heats to a temperature high enough that the exothermic reaction of steam with the zirconium alloy becomes rapid. Typically, this temperature for rapid reaction is a few hundred degrees above the temperature at which clad rupture takes place. Rapid, exothermic reaction of the cladding raises fuel temperatures and prompts rapid diffusive release of radionuclides characteristic of the In-vessel Release Phase of an accident as defined in NUREG-1465.

There is a distinct phenomenological and temporal distinction between gap release and in-vessel release when only a single rod or a small bundle of fuel rods is considered. The temporal distinction disappears when an entire reactor core is considered. The onset of gap release will occur when the first fuel rod ruptures. This first rupture will occur at the hottest part of the core – typically along the core centerline somewhat above mid-height. This hottest part of the core will go through the sequence of phenomenological events as outlined above for a single rod or small bundle of rods. But, as the gap release phase nears an end at this hottest location, other, cooler, regions of the core will just be reaching temperatures sufficient to cause cladding on the fuel to balloon and rupture to start the gap release. Indeed, it can be observed in accident analyses that hotter regions of the core have gone through gap release and major portions of in-vessel release to the point that fuel is melting and relocating from the core region before cooler regions near the core periphery have even started gap release. Thus, for a core as a whole, there is not a distinct period which can be exclusively categorized as the Gap Release Phase of an accident.

Gap release is taken to start when coolant levels fall to the top of the active fuel. Because there is no phenomenological boundary to mark the end of the Gap Release Phase, a figure of merit was adopted in this work to define the end of the Gap Release Phase. This phase of release was judged to have ended when 5% of the initial, total core inventory of xenon had been released from the fuel. Note, this is release from the fuel and not release to the containment. In many of the analyzed accidents, release of this amount of xenon did not take the full half hour allowed in the NUREG-1465 source term for the Gap Release Phase. The median duration of gap release in the case of BWR accidents involving high burnup fuel was only 0.16 hour. In the case of PWR accidents, the median duration was 0.22 hour. Modestly longer durations of gap release were calculated for both BWRs and PWRs using low burnup fuel.

There is no particular physical significance to the figure of merit taken to mark the conclusion of the gap release. The figure of merit was selected largely because it is a metric easily identified in the computer code calculations.

Because of the abbreviated duration of the Gap Release Phase, there was not time for all the fission gases and radionuclide vapors to escape the reactor coolant system and enter into the containment. Consequently, the gap releases to the containment are much lower than indicated in the NUREG-1465 source term where transport of the gap releases to the containment was taken to be quite rapid.

At the same time, the Gap Release Phase was calculated to be long enough that some core degradation characteristic of the In-vessel Phase of release as prescribed in the NUREG-1465 Source Term did take place. This is indicated by small amounts of tellurium release during the Gap Release Phase and in the case of PWR accidents small amounts of alkaline earth release. Ordinarily tellurium and alkaline earths are not thought to be contributors to the gap release. They contribute here because some portions of the core had entered into what would be categorized phenomenologically as in-vessel release before the criterion to terminate gap release had been reached.

Table 10. Proposed source term for BWRs using high burnup uranium dioxide fuel. Durations and release fractions for high burnup fuel are shown in bold. Parenthetical quantities are for lower burnup fuel and are included for the purposes of comparison. A dash entry means that a negligible release of the group was predicted to occur during the indicated phase of an accident.

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration (hours)	**0.16 ± 0.01** (0.20 ± 0.03)	**8.0 ± 1.1** (8.8 ± 1.1)	**2.9 ± 0.8** (1.6 ± 0.5)	**12 ± 2** (12 ± 2)
Release Fractions of Radionuclide Groups				
Noble Gases (Kr,Xe)	**0.008 ± 0.002** (0.008 ± 0.002)	**0.96 ± 0.01** (0.92 ± 0.01)	**0.009 ± 0.001** (0.064 ± 0.006)	**0.016 ± 0.006** (0.006 ± 0.002)
Halogens (Br,I)	**0.002 ± 0.001** (0.003 ± 0.002)	**0.47 ± .19** (0.54 ± 0.13)	**0.013 ± 0.002** (0.048 ± 0.007)	**0.39 ± 0.18** (0.27 ± 0.12)
Alkali Metals (Rb, Cs)	**0.002 ± 0.001** (0.003 ± 0.002)	**0.13 ± 0.04** (0.14 ± 0.03)	**0.010 ± 0.002** (0.065 ± 0.005)	**0.050 ± 0.0023** (0.046 ± 0.017)
Alkaline Earths (Sr, Ba)	-	**0.005 ± 0.002** (0.005 ± 0.002)	**0.029 ± 0.013** (0.028 ± 0.010)	**0.005 ± 0.001** (0.005 ± 0.002)
Tellurium Group (Te, Se, Sb)	**0.002 ± 0.001** (0.003 ± 0.002)	**0.39 ± 0.15** (0.39 ± 0.12)	**0.002 ± 0.001** (0.006 ± 0.002)	**0.33 ± 0.16** (0.33 ± 0.12)
Molybdenum (Mo, Tc, Nb)	-	**0.02 ± 0.01** (0.03 ± 0.01)	**0.003 ± 0.001** (0.017 ± 0.002)	**0.0055 ± 0.0032** (0.0058 ± 0.0024)
Noble Metals (Ru, Pd, Rh, etc.)	-	**0.0027 ± 0.0008** (0.0026 ± 0.0005)	[0.0025]	**1.0 ± 0.4 x10^{-4}** (1.7 ± 0.8 x10^{-5})
Lanthanides (Y, La, Sm, Pr, etc.)	-	**1.4 ± 0.2 x10^{-7}** (2.0 ± 0.5 x10^{-7})	**5 ± 3 x10^{-5}** (6 ± 4 x10^{-5})	-
Cerium Group (Ce, Pu, Zr, etc.)	-	**1.3 ± 0.2 x10^{-7}** 1.6 ± 0.3 x10^{-7}	**0.0021 ± 0.0015** (0.0019 ± 0.0011)	-

Table 11. Proposed source term for PWRs using high burnup uranium dioxide fuel. Durations and release fractions for high burnup fuel are shown in bold. Parenthetical quantities are for lower burnup fuel and are included for the purposes of comparison. A dash entry means that a negligible release of the group was predicted to occur during the indicated phase of an accident.

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration (hours)	**0.22 ± 0.04** (0.33 ± 0.12)	**4.5 ± 2.4** (5.3 ± 1.2)	**4.8 ± 1.3** (9 ± 10)	**143 ± 8** (130 ± 20)
Release Fractions of Radionuclide Groups				
Noble Gases (Kr,Xe)	**0.017 ± 0.003** (0.022 ± 0.002)	**0.94 ± 0.01** (0.85 ± 0.05)	**0.011 ± 0.008** (0.08 ± 0.05)	**0.003 ± 0.003** 0.002 ± 0.002
Halogens (Br,I)	**0.004 ± 0.002** (0.007 ± 0.002)	**0.37 ± 0.13** (0.30 ± 0.13)	**0.011 ± 0.008** (0.08 ± 0.03)	0.21 ± 0.16 (0.15 ± 0.11)
Alkali Metals (Rb, Cs)	**0.003 ± 0.001** (0.005 ± 0.002)	**0.23 ± 0.10** (0.23 ± 0.10)	0.02 ± 0.01 (0.03 ± 0.04)	**0.06 ± 0.04** (0.03 ± 0.01)
Alkaline Earths (Sr, Ba)	**0.0006 ± 0.0003** (0.0014 ± 0.0006)	**0.004 ± 0.002** (0.004 ± 0.001)	**0.003 ± 0.002** (0.002 ± 0.001)	-
Tellurium Group (Te, Se, Sb)	**0.004 ± 0.002** (0.007 ± 0.003)	**0.30 ± 0.12** (0.26 ± 0.11)	**0.003 ± 0.002** (0.03 ± 0.01)	**0.10 ± 0.10** (0.10 ± 0.07)
Molybdenum (Mo, Tc, Nb)	-	**0.08 ± 0.03** (0.10 ± 0.02)	**0.01 ± 0.01** (0.10 ± 0.09)	**0.03 ± 0.03** (0.05 ± 0.06)
Noble Metals (Ru, Pd, Rh, etc.)	-	**0.006 ± 0.006** (0.006 ± 0.004)	[0.0025]	-
Lanthanides (Y, La, Sm, Pr, etc.)	-	**1.5 ± 1.2 x10^{-7}** (1.1 ± 0.9 x10^{-7})	**1.3 ± 0.3 x10^{-5}** (2.6 ± 0.8 x10^{-5})	-
Cerium Group (Ce, Pu, Zr, etc.)	-	**1.5 ± 1.2 x10^{-7}** (1.1 ± 0.9 x10^{-7})	**2.4 ± 0.9 x10^{-4}** (1.0 ± 0.8 x10^{-4})	-

Table 12. Comparison of BWR high burnup durations and release fractions (bold entries) with those recommended for BWRs in NUREG-1465 (parenthetical entries).

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration (hours)	**0.16** (0.5)	**8.0** (1.5)	**2.9** (3.0)	**12** (10)
Release Fractions of Radionuclide Groups				
Noble Gases (Kr,Xe)	**0.008** (0.05)	**0.96** (0.95)	**0.009** (0)	**0.016** (0)
Halogens (Br,I)	**0.002** (0.05)	**0.47** (0.25)	**0.013** (0.30)	**0.39** (0.01)
Alkali Metals (Rb, Cs)	**0.002** (0.05)	**0.13** (0.20)	**0.01** (0.35)	**0.05** (0.01)
Alkaline Earths (Sr, Ba)	-	**0.005** (0.02)	**0.029** (0.10)	**0.005** (0)
Tellurium Group (Te, Se, Sb)	**0.002** (-)	**0.39** (0.05)	**0.002** (0.25)	**0.33** (0.005)
Molybdenum (Mo, Tc, Nb)	-	**0.02** (0.0025)	**0.003** (0.0025)	**0.0055** (0)
Noble Metals (Ru, Pd, Rh, etc.)	-	**0.0027** (0.0025)	[0.0025]	**1.0×10^{-4}** (0)
Lanthanides (Y, La, Sm, Pr, etc.)	-	**1.4×10^{-7}** (2×10^{-4})	**5×10^{-5}** (0.005)	-
Cerium Group (Ce, Pu, Zr, etc.)	-	**1.3×10^{-7}** (2×10^{-4})	**0.0021** (0.005)	-

Table 13. Comparison of PWR high burnup durations and release fractions (bold entries) with those recommended for PWRs in NUREG-1465 (parenthetical entries).

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration (hours)	**0.22** (0.5)	**4.5** (1.5)	**4.8** (2.0)	**143** (10)
Release Fractions of Radionuclide Groups				
Noble Gases (Kr,Xe)	**0.017** (0.05)	**0.94** (0.95)	**0.011** (0)	**0.003** (0)
Halogens (Br,I)	**0.004** (0.05)	**0.37** (0.35)	**0.011** (0.25)	**0.21** (0.10)
Alkali Metals (Rb, Cs)	**0.003** (0.05)	**0.23** (0.25)	**0.02** (0.35)	**0.06** (0.10)
Alkaline Earths (Sr, Ba)	**0.0006** (0)	**0.004** (0.02)	**0.003** (0.10)	**-** (-)
Tellurium Group (Te, Se, Sb)	**0.004** (0)	**0.30** (0.05)	**0.003** (0.25)	**0.10** (0.005)
Molybdenum (Mo, Tc, Nb)	-	**0.08** (0.0025)	**0.01** (0.0025)	**0.03** (0)
Noble Metals (Ru, Pd, Rh, etc.)	-	**0.006** (0.0025)	[0.0025]	-
Lanthanides (Y, La, Sm, Pr, etc.)	-	**$1.5x10^{-7}$** ($2x10^{-4}$)	**1.3x10-5** (0.005)	-
Cerium Group (Ce, Pu, Zr, etc.)	-	**$1.5x10^{-7}$** ($5x10^{-4}$)	**$2.4x10^{-4}$** (0.005)	-

4.1.2 The In-vessel Release Phase

The In-vessel Release Phase starts when the Gap Release is completed and ends when core debris penetrates the reactor vessel and cascades into the reactor cavity where it can interact with accumulated water and structural concrete. The median durations of In-vessel Release Phase predicted for BWRs and PWRs are much longer than specified in the NUREG-1465 Source Terms for this phase of reactor accidents. The longer duration of the In-vessel Release Phase is the most profound difference among accident analyses used here and those that were the basis of the NUREG-1465 Source Term. Prolonged core degradation is not altogether surprising. Since the development of the NUREG-1465 Source Term, modeling of core degradation has been greatly improved largely by identifying and modeling of efficient mechanisms for distribution of heat from the degrading reactor fuel to the reactor coolant system especially by natural convection processes. As a consequence, degrading core material is not predicted to become as hot as rapidly as it was in calculations of reactor accidents using the Source Term Code Package [Gieseke *et al*., 1986] that were the basis of the NUREG-1465 Source Term.

Nevertheless, the degrading core materials do become hot enough to sustain the continued release of volatile radionuclides. Because the period between the onset of core degradation and the penetration of the reactor vessel by core debris is longer, there is ample opportunity for volatile radionuclides to escape the fuel. Release fractions for volatile radionuclides such as iodine are larger than indicated in the NUREG-1465 Source Term. On the other hand, maximum core temperatures are not as high, so there are not similarly large increases in the releases of the less volatile radionuclides such as the Lanthanides or the Cerium Group radionuclides.

Cesium, otherwise recognized as a volatile radionuclide, does not behave like iodine. As noted in Section 2 of this report, the assumed chemical form of cesium released from degrading reactor fuel has changed since the development of the NUREG-1465 Source Term in response to findings especially in the Phébus-FP program, but also in response to analytic investigations. It has been assumed in this work that much of the cesium released from the fuel that is not in the form of cesium iodide (CsI) is present as vapor or particulate cesium molybdate (Cs_2MoO_4) rather than being present as the much more volatile species cesium hydroxide (CsOH).

The assumption that cesium is released from the reactor fuel as Cs_2MoO_4 rather than as CsOH has three consequences for the source term to the reactor containment. Because Cs_2MoO_4 is less volatile than CsOH, more deposition of released cesium takes place in the reactor coolant system and less of the released cesium reaches containment. Cesium that reaches containment as

CsOH will raise the pH of water it contacts and, consequently, could affect the partitioning of iodine from these waters back into the atmosphere as discussed further in Section 4.5. Cs_2MoO_4 has much less impact on the pH of water it contacts. Release of cesium in the form of Cs_2MoO_4 also leads to increases in the releases of molybdenum relative to releases of other refractory metal fission products such as ruthenium and palladium. Because of this third effect, the Refractory Metals class of radionuclides has been divided for this work to create a special class for molybdenum. The redefined chemical classes of radionuclides released to the containment are shown in Table 14. For reasons of chemical analogy, niobium and technetium have been included in the molybdenum category.

Tellurium behavior is notable. Releases of tellurium are much higher than indicated in the NUREG-1465 Source Term. It was assumed in the development of the NUREG-1465 Source Term that much of the tellurium released from reactor fuel would react to form stable, solid compounds either with zirconium cladding that had not yet been oxidized or with steel in the reactor coolant system. Consequently, tellurium released from degrading reactor fuel would not reach the containment efficiently. Such extensive trapping of tellurium has not been observed in the Phébus-FP tests of radionuclide release and transport. Indeed, transport of released tellurium to containment has been found to be quite efficient. MELCOR modeling used in this work is consistent with these experimental observations. Consequently, tellurium releases to the containment are predicted to be higher than would have been predicted in the past.

Table 14. Revised chemical classes of radionuclides adopted for this work.

Chemical Group Name	Elements in the Group
Noble Gases	Kr, Xe
Halogens	Br, I
Alkali Metals	Rb, Cs
Tellurium Group	Se, Sb, Te
Barium, Strontium Group (Alkaline Earths)	Sr, Ba
Molybdenum Group	Mo, Nb, Tc
Noble Metals	Ru, Rh, Pd, Co
Lanthanides	La, Nd, Eu, Nb, Pm, Pr, Sm, Y, Cm, Am
Cerium Group	Ce, Pu, Np, Zr

4.1.3 The Ex-vessel Release Phase

The Ex-vessel Release Phase begins when core debris penetrates the reactor vessel and begins to interact with structural concrete in the reactor cavity. Thermal attack on concrete by core debris produces steam and carbon dioxide. These gases will sparge through the core debris. They will react exothermically with metals, especially any metallic zirconium in core debris that escaped oxidation during the in-vessel release phase of the accident. The exothermic reactions of the concrete decomposition gases with metallic zirconium can raise the core debris temperature substantially. Because the sparging gases provide large free surfaces within the core debris, releases of volatile radionuclides from the high temperature debris can be efficient. Some release of even non-volatile radionuclides will take place by entrainment of debris droplets in the sparging gases.

There is not a phenomenological event that marks the end of core debris attack on concrete in accidents that are not terminated by accident management measures. Core debris will continue to thermally decompose concrete to produce gases that sparge through any melt present even after the reactor fuel and structural steel have solidified. Releases become quite attenuated at these lower temperatures and as a result of core debris dilution by the condensed-phase products of concrete decomposition. Consequently, a figure of merit was defined to mark the end of the radionuclide release during the ex-vessel phase of the accident. This phase was taken to be complete when 95% of the releasable cesium inventory in the core debris expelled ex-vessel had been released. When core debris is quite hot, such as will be the case if there are large amounts of unoxidized zirconium in the core debris, this limit can be reached quite promptly. The limit is slow to be reached in cases where the core debris is depleted of unoxidized zirconium and the concrete is easily melted.

It was found for these analyses that the durations of the ex-vessel release phase of accidents were longer than found for the NUREG-1465 Source Term. Despite the longer duration of the Ex-vessel Release phases of accidents, release fractions for volatile radionuclides such as cesium, tellurium and iodine are, on the whole, much lower than indicated by the NUREG-1465 Source Term. This is simply because there is not as much inventory of these volatile fission products left in the core debris that penetrates the vessel following the prolonged In-vessel Release Phases of accidents.

The prolonged In-vessel Release Phase allows more time for oxidation of zirconium alloy cladding on the fuel. Consequently, debris expelled from the reactor vessel marking the onset of the Ex-vessel Release Phase has less metallic zirconium than was predicted in the past. There is less chemical heating of the ex-vessel core debris by reaction of gases from the concrete with the core debris. Releases of more refractory radionuclides such as the Lanthanides and the Cerium Group are somewhat lower because the temperatures of core debris with less metallic zirconium do not become as high for as long as predicted in the past.

Note that there is a known deficiency of the MELCOR model of ex-vessel releases of refractory metal radionuclides such as ruthenium and palladium. Consequently, ex-vessel release fractions for this class of radionuclides shown in the tables are values prescribed in NUREG-1465.

4.1.4 The Late In-vessel Release Phase

Definition of the Late In-vessel Release Phase was an important contribution to reactor accident source term analysis made in the development of the NUREG-1465 source term. It was not explicitly calculated in most reactor accidents done at the time. The predicted durations and release fractions associated with this phase of release in the NUREG-1465 Source Term are estimates. Today, this phase of reactor accidents is much more explicitly calculated. Release during this phase of accidents occurs because of:

- continued escape of radionuclides from fuel within the reactor vessel that has not relocated into the reactor cavity to participate in ex-vessel releases, and
- revaporization of radionuclides that were released from fuel during the In-vessel Release Phase of the accident, but deposited in the reactor coolant system and did not reach containment.

The Late In-vessel Release Phase is taken to begin when the In-vessel Release Phase ends. Late In-vessel Release begins at the same time as the Ex-vessel Release and proceeds in parallel with Ex-vessel Release for a time. In principle,

the Late In-vessel Release Phase of reactor accidents continues as long as there are sufficient deposits of radionuclides present in the reactor coolant system that can be heated enough by decay energy to produce meaningful vapor pressures of volatile radionuclides. For the purposes of the accident analyses done here, the period of Late In-vessel Release was ended once release had occurred for 95% of the cesium that would be released during this phase of reactor accidents. The median predicted duration of Late In-vessel Release for BWRs is rather similar to the duration estimated in the development of the NUREG-1465 source term – 12 versus 10 hours. As will be discussed further in a later subsection of this report dealing with MOX source terms, the median duration of Late In-vessel Release for accidents at an ice-condenser PWR is also similar to that estimated in the development of the NUREG-1465 source term – 16 versus 10 hours. A rather longer period, >100 hours, is predicted here for Late In-vessel Release in other types of PWRs. Note that uncertainties in the durations are large. Uncertainty in the location of the median duration is indicative of a high degree of variability among the various accidents in the duration of Late In-vessel Release. Late In-vessel Release varies especially with details of plant design and the ability to sustain elevated temperatures in the reactor coolant system with decay heating of deposited radionuclides.

Release fractions during Late In-vessel Release are larger than estimated in the development of the NUREG-1465 Source Term. In large part this is because large fractions of the more volatile fission products are predicted to be released during the In-vessel Release Phases of accidents. Consequently, there are larger inventories of the more volatile radionuclides such as iodine and tellurium deposited in the reactor coolant systems. The Late In-vessel Release fractions are larger for BWRs than PWRs which is opposite the ordering indicated by the NUREG-1465 Source Term. Lesser, but still significant, fractions of cesium are predicted to be released during the Late In-vessel Release phase because the cesium is deposited in the form of cesium molybdate which is less volatile than the deposited forms of either iodine or tellurium. Revaporization of cesium molybdate contributes to the Late In-vessel Release of molybdenum which was not anticipated at the time the NUREG-1465 Source Term was developed. Revaporization of deposited alkaline earths is predicted for BWRs which was not anticipated when the NUREG-1465 Source Term was devised.

4.2 MOX Fuel

Results obtained from analysis of accidents at an ice-condenser PWR with a 40% MOX core as summarized in Table 15. The first striking finding is that in-vessel releases from a 40% MOX core are not greatly different than what would be expected for in-vessel releases from a moderate burnup, low enrichment uranium dioxide core. That is, there are not greatly enhanced releases of radionuclides from MOX fuel. This is, of course, somewhat surprising since experiments with single MOX fuel pellets indicate more rapid release of volatile fission products from MOX than from uranium dioxide fuels. The higher releases were most pronounced for low temperatures (1000 to 1800 ˚C). Release rates for the two types of fuel tended to converge as temperature increased. MELCOR models for release from MOX had been modified to account for these experimental observations concerning the relative release rates from MOX and uranium dioxide fuels. It appears that under reactor accident conditions, fuel spends so little time at temperatures where release rates of volatile radionuclides in MOX and uranium dioxide differ that the differences do not have great effects on the overall release of radionuclides to the containment.

Other differences between source term characteristics of MOX and low enrichment uranium dioxide cores seem to be explainable in terms of the somewhat lower decay heat in the case of the MOX core. Of course, any differences between the core degradation in a MOX core and core degradation in a low enrichment uranium dioxide core have not been modeled because there are not data showing differences in degradation processes. Since plutonium oxides accumulate in low-enrichment urania fuel as burnup progresses, large differences in the degradation of MOX fuel and high burnup urania fuel would not be expected.

The MOX source terms derived here are compared in Table 16 with the NUREG-1465 Source Term for PWRs. Overall, the calculated releases for the MOX core look much like the releases calculated for cores with high burnup fuel discussed in Section 4.1. Differences from the NUREG-1465 Source Term are predominantly due to differences in the current understanding of core degradation phenomena and fission product chemistry now in comparison to when the NUREG-1465 Source Term was developed. In summary:

- There is no phenomenological end to the gap release. As with high burnup fuel, gap release was taken to end when 5% of the core inventory of xenon had been released from the fuel. The fractions of the core inventory of fission products that reach the containment are smaller because there is not time over the abbreviated duration of the Gap Release Phase for all of the released species to reach containment. Of course, some of the condensable

radionuclides do deposit in the reactor coolant system even during the Gap Release Phase of the accident.

- In-vessel core degradation is prolonged relative to what is specified in the NUREG-1465 Source Term probably because modeling of heat transfer from the degrading core to the reactor coolant system and core structures is better.
- Prolonged core degradation allows more of the volatile radionuclides to be released during the in-vessel release phase of an accident. The release fraction for iodine is higher than in the NUREG-1465 Source Term.
- Chemical forms of tellurium and cesium affect the transport of these radioactive elements from the core to the containment. Cesium is now thought to be released to a significant extent as cesium molybdate which is less volatile than cesium hydroxide and consequently more prone to deposit in the reactor coolant system prior to reaching the containment. On the other hand, tellurium is thought to be released in a form that does not interact strongly with either residual, unoxidized cladding on fuel or with the metal surfaces in the reactor coolant system.
- Release of cesium as cesium molybdate enhances release of molybdenum relative to releases of other refractory metals such as ruthenium and palladium. Consequently, a separate radionuclide class has been created for molybdenum.
- Because so much of the inventory of volatile radionuclides are released from the fuel during the in-vessel stage of an accident, much smaller releases of these radionuclides can take place during the ex-vessel stage of an accident. Consequently, release fractions for the halogens and the alkali metals are smaller than specified in the NUREG-1465 Source Term.
- Because large fractions of the halogens, tellurium group radionuclides and alkali metals are released during the in-vessel core degradation stage of the accident, and these released radionuclides do deposit in the reactor coolant system, there are opportunities for large releases due to revaporization during the late in-vessel release phase. Release fractions predicted for core meltdown accidents in an ice-condenser containment with a 40% MOX core are commensurate when uncertainties are recognized with recommendations for PWRs made in NUREG-1465. Release of alkali metals by revaporization is suppressed relative to the release of halogens because the alkali metals are present on surfaces in a less volatile chemical

form. There is more extensive revaporization of deposited tellurium than anticipated when the NUREG-1465 because of the chemical form adopted by tellurium as observed in tests and assumed in the MELCOR code.

Table 15. Proposed source term for a PWR with an ice-condenser containment using a 40% MOX core. Durations and release fractions for MOX are shown in bold. Values found from analyses of identical accidents for the reactor with a low enrichment uranium dioxide core are shown parenthetically. Uncertainties associated with each entry reflect the uncertainty in locating the median of the distribution of results. A dash entry means that a negligible release of the group was predicted to occur during the indicated phase of an accident.

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration (hours)	**0.36 ± 0.12** (0.34 ± 0.12)	**4.4 ± 1.1** (4.1 ± 0.7)	**6.5 ± 6.1** (6.0 ± 3.7)	**16 ± 15** (15 ± 18)
Release Fractions of Radionuclide Groups				
Noble Gases (Kr,Xe)	**0.028 ± 0.009** (0.029 ± 0.006)	**0.86 ± 0.06** (0.87 ± 0.07)	**0.05 ± 0.02** (0.10 ± 0.06)	**0.026 ± 0.006** (0.019 ± 0.007)
Halogens (Br,I)	**0.028 ± 0.009** (0.014 ± 0.007)	**0.48 ± 0.12** (0.47 ± 0.12)	**0.06 ± 0.020** (0.08 ± 0.03)	**0.055 ± 0.021** (0.060 ± 0.024)
Alkali Metals (Rb, Cs)	**0.014 ± 0.009** (0.014 ± 0.008)	**0.44 ± 0.08** (0.41 ± 0.08)	**0.07 ± 0.03** (0.11 ± 0.05)	**0.025 ± 0.020** (0.032 ± 0.018)
Alkaline Earths (Sr, Ba)	-	**0.0015 ± 0.0005** (0.0015 ± 0.0004)	**0.008 ± 0.003** (0.015 ± 0.015)	**9 ± 5 x10^{-5}** (9 ± 6 x10^{-5}**)**
Tellurium Group (Te, Se, Sb)	**0.014 ± 0.009** (0.014 ± 0.008)	**0.48 ± 0.10** (0.45 ± 0.10)	**0.04 ± 0.01** (0.04 ± 0.02)	**0.055 ± 0.025** (0.058 ± 0.020)
Molybdenum (Mo, Tc, Nb)	-	**0.27 ± 0.06** (0.28 ± 0.07)	[0.0025]	**0.024 ± 0.008** (0.024 ± 0.006)
Noble Metals (Ru, Pd, Rh, etc.)	-	**0.005 ± 0.002** (0.006 ± 0.002)	[0.0025]	**3 ± 1x10^{-4}** (2 ± 1 x10^{-4})
Lanthanides (Y, La, Sm, Pr, etc.)	-	**1.1 ± 0.25 x10^{-7}** (1.1 ± 0.3 x10^{-7})	**3 ± 2 x10^{-5}** (6 ± 16 x10^{-4})	-
Cerium Group (Ce, Pu, Zr, etc.)	-	**1.0 ± 0.25 x10^{-7}** (1.1 ± 0.3 x10^{-7})	**5 ± 4 x10^{-4}** (5 ± 13 x10^{-3})	-

Table 16. Comparison of proposed source term for an ice-condenser PWR with a 40% MOX core (bold entries) to the NUREG-1465 source term for PWRs (parenthetical entries).

	Gap Release	In-vessel Release	Ex-vessel Release	Late In-vessel Release
Duration (hours)	**0.36** (0.50)	**4.4** (1.3)	**6.5** (2.0)	**16** (10)
Release Fractions of Radionuclide Groups				
Noble Gases (Kr,Xe)	**0.028** (0.050)	**0.86** (0.95)	**0.05** (0)	**0.026** (0)
Halogens (Br,I)	**0.028** (0.050)	**0.48** (0.35)	**0.06** (0.25)	**0.055** (0.10)
Alkali Metals (Rb, Cs)	**0.014** (0.050)	**0.44** (0.25)	**0.07** (0.35)	**0.025** (0.10)
Alkaline Earths (Sr, Ba)	-	**0.0015** (0.020)	**0.008** (0.1)	**$9x10^{-5}$** (0)
Tellurium Group (Te, Se, Sb)	**0.014** (0)	**0.48** (0.05)	**0.04** (0.25)	**0.055** (0.005)
Molybdenum (Mo, Tc, Nb)	-	**0.27** (0.0025)	[0.0025]	**0.024** (0)
Noble Metals (Ru, Pd, Rh, etc.)	-	**0.005** (0.0025)	[0.0025]	**$3 x10^{-4}$** (0)
Lanthanides (Y, La, Sm, Pr, etc.)	-	**$1.1 x10^{-7}$** (0.0002)	**$3 x10^{-5}$** (0.005)	-
Cerium Group (Ce, Pu, Zr, etc.)	-	**$1.0 x10^{-7}$** (0.0005)	**$5 x10^{-4}$** (0.005)	-

4.3 Nonradioactive Releases

Source terms developed here only address the releases of radionuclides to the containment. It is known, however, that volatile but nonradioactive materials will be released to containment during reactor accidents. Important contributors to this nonradioactive release include silver, indium, and cadmium from control rods, tin from fuel cladding alloys, as well as constituents of structural steels heated and melted during core degradation. Boron carbide used as a control material can be oxidized in steam to produce copious quantities of boric oxide and boric acid. Volatile constituents of concrete including sodium oxide, potassium oxide, manganese oxide and the like can be vaporized and released to the containment during the ex-vessel phase of a reactor accident.

Most of the nonradioactive materials released to the containment will condense to form aerosol particles that will affect the behavior of radioactive aerosol particles. Repeated tests, most recently tests in the Phébus-FP program, have shown that nonradioactive and radioactive aerosol particles agglomerate. That is, inclusion of nonradioactive aerosol will promote the growth of all aerosol particles. To the extent that aerosol removal processes, either natural processes or processes due to engineered safety systems such as sprays and suppression pools, depend on aerosol particle size, the nonradioactive aerosol sources can shorten the period released radionuclides remain suspended in the containment atmosphere and available for leakage into the environment.

On the other hand, the additional aerosol mass from nonradioactive sources can affect the performance of engineered safety systems such as filtration systems and sump screens. Agglomeration and reaction of nonradioactive particles with radionuclide particles can complicate analyses of aerosol behavior in containment such as the analysis of hygroscopicity effects on aerosol particle deposition in containment.

Nonradioactive aerosol releases should be recognized in source term analyses. Because these releases are very dependent on reactor design and especially material selections, some care is required in estimating the magnitude of nonradioactive sources for each reactor type.

4.4 Release Rates

The tables of proposed source terms presented in this report provide releases fractions of various classes of radionuclides and the durations over which these release fractions take place. A second-order consideration is the rate at which each class of radionuclide is released during each release phase of an accident. An approximation introduced with the development of the NUREG-1465 Source Term was the assumption of constant release rates of the various radionuclide classes. That is, the release rates were determined by dividing the release fractions by the duration of the release phase.

This was known at the time it was introduced to be a convenient, but very approximate description of the true release rates. The simplification was introduced to facilitate the use of the NUREG-1465 Source Term in the regulatory process.

The approximation of constant release rates is also adopted here for the same reasons it was adopted for the NUREG-1465 Source Term. The approximation is not especially significant for Gap Release or Ex-vessel Release. It is more questionable for the prolonged In-vessel Release Phase found in this work. Typically, but not universally, mechanistic calculations of core degradation predict that release rates are high during the early stages of the in-vessel core degradation process when temperatures spike due to the rapid steam oxidation of cladding. As fuel melts and loses free surface from which radionuclides can vaporize, release rates slow. Experiments show the slowing release is punctuated by episodes of more rapid release thought to be the result of enhanced mass transfer when core debris relocates from the fuel region to cooler regions. Such episodic instances of excursions in radionuclide release rates associated with core debris relocation are challenging to predict accurately.

The critical issue of radionuclide release rates is the impact of particle concentration on aerosol behavior. At higher release rates, aerosol concentrations can be higher and aerosol particle growth will be more rapid. Larger particles are more easily removed from containment by gravitational settling, inertial deposition and by spray droplets.

Figures 4 and 5 show comparisons of containment concentrations of radionuclides based on different assumptions of release rates during the In-vessel Release phase of a reactor accident. Release rates termed realistic are calculated using the MELCOR code. The constant release rates are calculated from the proposed source term tables presented here. The comparisons shown in the figure are intended to be illustrative and not definitive of the effects assumptions about release rate can have on containment concentrations of radioactive aerosol. In Figure 4, radionuclides released to the containment are affected only by natural aerosol agglomeration and deposition processes. In Figure 5, it is assumed that containment sprays are operational. These comparisons suggest that the assumption of constant release rates maintains elevated aerosol concentrations in the containment atmosphere for longer periods than would be predicted using more realistic release rates.

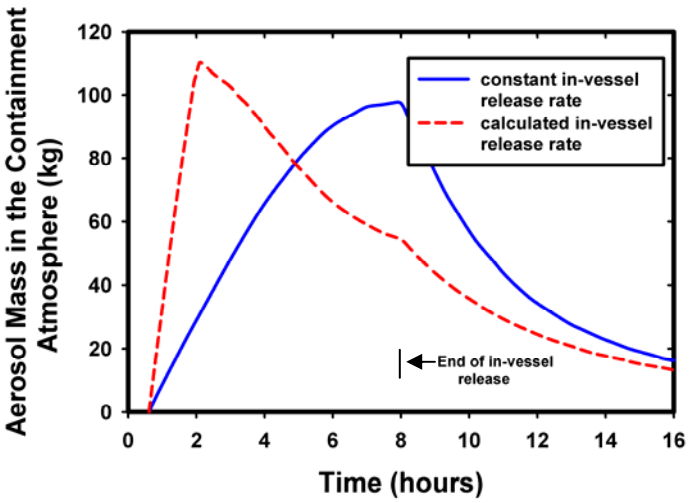

Figure 4. Comparison of containment concentrations of aerosol assuming only natural aerosol deposition processes and realistic release rates of radionuclides to containment or constant rates of radionuclide release to containment.

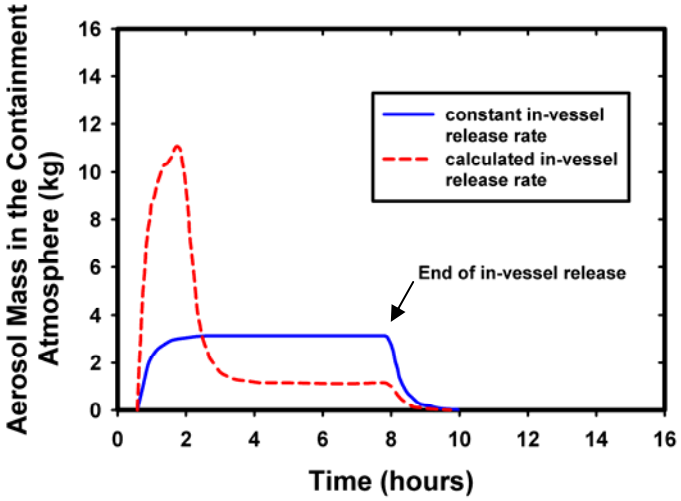

Figure 5. Comparison of containment concentrations of aerosol assuming continuous containment spray operation and realistic release rates of radionuclides to containment or constant rates of radionuclide release to containment.

4.5 Chemical Form of Iodine

The chemical form of iodine released to the containment in core melting accidents has been the topic of continuing debate and research. The TID-14844 source term assumed iodine was released to the containment predominantly as a gaseous species. The Reactor Safety Study questioned this assumption based on thermochemical arguments [Ritzman, *et al*., 1975], but, in the end, retained the assumption for risk assessment. The thermochemical arguments were advanced further following the accident at Three Mile Island. Analyses done for the NUREG-1150 risk assessments assumed iodine was released to the containment predominantly in the form of metal iodide particles – notably cesium iodide (CsI).

The NUREG-1465 source term specifies that most of the released iodine can be considered metal iodide particulate, but a percentage (5%) of the released iodine should be considered to be present as gaseous species (molecular iodine, I_2, and volatile organic iodides such as methyl iodide, CH_3I). Furthermore, the NUREG-1465 source term recommended that sump waters within containment be maintained at basic pH (pH>7) values to assure that dissolved iodine does not partition back into the containment atmosphere as a gaseous species that could leak into the environment.

Treatment of the release of iodine to containment as a mixture of gaseous species and metal iodide particulate has been adequately validated by the results of tests done in the Phébus-FP program. The precise ratio of gaseous and particulate forms of iodine released to the containment and the behavior of iodine within the containment are subjects of ongoing investigation. Simple descriptions of iodine dissolving and partitioning from aqueous solution are being questioned. Investigations are focusing on the thermal and radiolytic chemistry of iodine in the gas phase and the aqueous phase as well as the interactions of gaseous and particulate iodine species with metallic and painted surfaces.

Pending the outcome of these ongoing investigations, it is prudent to retain the prescription of iodine chemical form adopted in the NUREG-1465 source term. That is, 95% of the iodine released to containment is assumed to be in the form of metal iodide particles that will agglomerate with other aerosol particles in the containment atmosphere. The remaining 5% of the iodine is released to the containment as a gaseous species.

Most of the iodine species released to containment will readily dissolve in water. The exception is insoluble silver iodide (AgI) that can be formed by reaction of iodine with silver from silver-indium-cadmium control rods. Iodine dissolved in water at pH less than 7 can react both thermally and radiolytically to form molecular iodine and volatile organic iodides that will partition from the aqueous phase back into the containment atmosphere. Maintenance of basic pH in sump waters within containment cannot rely on the availability of cesium hydroxide as the dominant chemical form of cesium

released to the reactor containment. Cesium molybdate is thought to be a more important chemical form. Cesium molybdate is water soluble, but its hydrolysis will not drive solutions to basic pH.

4.6 In-containment Aerosol Removal Mechanisms

As noted in NUREG-1465, the principal mechanism by which source terms of gaseous and particulate radionuclides to the containment can emerge into the environment is by leakage of the containment atmosphere. The concentrations of radionuclides in the leaking atmosphere of containment depend on both the rates at which radionuclides reach the containment and the rates at which they are removed from the atmosphere. Both natural and engineered processes can contribute to the removal of radionuclides from the containment atmosphere. NUREG-1465 reviewed the capabilities of engineered processes such as:

- Containment sprays
- BWR steam suppression pools
- Filtrations systems
- Cavity flooding and water over debris interacting with concrete
- Ice condenser beds

to remove radioactive aerosols from the containment atmosphere. The state of knowledge of these processes has been refined but not fundamentally changed since NUREG-1465 was written. Recently, the state of knowledge of nuclear aerosols including the understanding of the effects of natural and engineered processes in containment has been reviewed [OECD, 2008]. This review also provides recommendations on additional research that would be of use for prediction of radionuclide behavior in containments.

5. CONCLUSIONS

Source terms patterned after those developed in NUREG-1465 have been developed for high burnup fuel in PWRs and BWRs and for MOX fuel in ice-condenser containment PWRs. The source terms have been developed using nonparametric statistics to examine results of an ensemble of selected accident sequences analyzed with the MELCOR 1.8.5 computer code. In terms of fractional releases, source terms developed for high burnup fuel and for MOX do not differ markedly from source terms developed by similar means for lower burnup fuel or for low-enrichment uranium dioxide fuel. The source terms do differ from those described in NUREG-1465. These differences can be attributed to improved understanding of reactor accident phenomenology and modeling since publication of NUREG-1465.

Some important differences are:

- The predicted duration of the In-vessel Release Phase of reactor accidents is longer because modeling accounts for more heat loss from core debris to the reactor coolant system.

- The prolonged In-vessel Release Phase allows for more extensive release of volatile radionuclides such as iodine and tellurium.

- In-vessel release of cesium is not increased similarly because it is assumed based on experimental studies to be present predominantly in the less volatile form cesium molybdate (Cs_2MoO_4) rather than cesium hydroxide (CsOH).

- Because of cesium release predominantly in the form of cesium molybdate, molybdenum releases are predicted to be higher than in the past and to differ significantly from releases of other nominally metallic fission products such as ruthenium and palladium. Consequently, a new chemical class has been defined for molybdenum.

- More extensive tellurium release is predicted because tellurium is expected to be in a chemical form that does not readily react with residual fuel cladding or metal surfaces within the reactor coolant system.

- The contribution of the Ex-vessel Release Phase to radionuclide release to containment is attenuated. This is because core debris is depleted of volatile radionuclides

during the prolonged In-vessel Release Phase and less metallic zirconium is present in core debris that emerges from the reactor vessel to interact with structural concrete.

- Late In-vessel Release can make a larger contribution to radionuclide release to the containment because more of the volatile radionuclides escape the fuel and deposit in the reactor coolant system during the In-vessel Release Phase of the accident. For PWRs the Late In-vessel Release Phase of an accident can be prolonged.

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