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Yucca Mountain Site Characterization Project

A Simplified Radionuclide Source Term for Total-System Performance Assessment

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A Simplified Radionuclide Source Term for Total-System Performance Assessment

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Abstract

A parametric model for releases of radionuclides from spent-nuclear-fuel containers in a waste repository is presented. The model is appropriate for use in preliminary total-system performance assessments of the potential repository site at Yucca Mountain, Nevada; for this reason it is simpler than the models used for detailed studies of waste-package performance. Terms are included for releases from the spent fuel pellets, from the pellet/cladding gap and the grain boundaries within the fuel pellets, from the cladding of the fuel rods, and from the radioactive fuel-assembly parts. Multiple barriers are considered, including the waste container, the fuel-rod cladding, the thermal "dry-out," and the waste form itself. The basic formulas for release from a single fuel rod or container are extended to formulas for expected releases for the whole repository by using analytic expressions for probability distributions of some important parameters.

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1 Introduction

A total-system performance assessment consists of calculations that estimate the long-term behavior of a repository system in which highly radioactive waste is isolated from the environment. The ultimate use for such assessments is in determining whether a repository system meets the standards set by the U. S. Environmental Protection Agency (40 CFR Part 191; EPA, 1985). Preliminary assessments are, however, indispensable in repository design, in the characterization of a repository site, and for early determinations of the suitability of a site. In particular, assessments are important to the work of the Yucca Mountain Site Characterization Project of the U. S. Department of Energy, and this report is a part of the ongoing program to improve and extend the assessments made for that project.

Repository systems are complex collections of subsystems, many of which contribute to the isolation of the waste. An assessment of total-system performance combines estimates of the behavior of these subsystems, taking into account the interactions that may occur among them. The mathematical models that are used for making the total-system calculations therefore include models of the subsystems as well as of the entire system.

One of the principal subsystems consists of the containers that, emplaced deep underground, will hold the radioactive material. To assess total-system performance requires an estimate of the amount of radioactive material released from the containers. This estimate can then be used as a source term in the equations that calculate the movement of radionuclides through the rest of the repository system: it describes the amount of radioactive material available for transport through the rock surrounding the repository. For assessments of the total system, it is necessary to use simplified models of the complex processes that govern the behavior of the containers, the transport through the rock near the containers, and the subsequent transport through much greater thicknesses of rock to the environment. Such simplification is necessary because it is impractical to model the multitude of complex processes all at the same time – as they must be modeled in a total-system assessment. For this reason, a total-system assessment generally treats the containers and the rock in their immediate vicinity as a “black box”: the equations do not model in detail the processes that happen there, but simply describe what comes out.

A detailed release model called PANDORA (Performance Assessment of NNWSI Design Omitting Random Aspects) is being developed for the Yucca Mountain Site Characterization Project at Lawrence Livermore National Laboratory (O’Connell and Drach, 1986; O’Connell, 1990). Another existing computer model is Pacific Northwest Laboratory’s AREST (Analytical REpository Source Term) program (Liebetrau *et al.*, 1987; Apted *et al.*, 1989). Both of these source models contain much more detail than the model presented here. Some recent total-system work has been done using the full PANDORA and AREST source terms (Barnard and Deckery, 1991; Apted *et al.*, 1991), but when large numbers of simulations are to be made, simpler models are desirable.

This report presents formulas for a radionuclide source term suitable for use in preliminary total-system performance assessments. The various processes taking place in

and around waste containers will be discussed in general terms, and a set of analytic formulas will be derived that incorporate simple approximations for most, though not all, of the important processes. This represents a step forward from the even simpler approximations that have been used for most total-system performance assessment in the past, such as those used by Sinnock *et al.* (1984), Sinnock *et al.* (1986), DOE (1986, Section 6.4.2), DOE (1988, Section 8.3.5.13), and McGuire *et al.* (1990). The model presented here is at the same level of abstraction as the source models used in those works, but more processes are included, so the formulas have many more adjustable parameters. In fact, the number of parameters for which values are needed and the complexity of some of the formulas (see Equation 23, for example) may seem quite intimidating at first. However, if desired, many of the parameters may be set to zero to get simpler formulas. Often, the simpler formulas may even be more conservative. Conservative formulas, in this report, are formulas that predict higher release rates and/or earlier release times, thus leading to greater releases to the accessible environment within the regulatory period of 10,000 yr than might be predicted by less conservative, but possibly more realistic, formulas. Conservatism is generally considered to be desirable when there is not enough information to be sure of realism.

Sections 2 through 4 of this report go through some details of the release processes and the derivation of the simplified source-term model. For those not interested in details, Section 5 contains a summary of the preceding material, including all of the formulas for the release model. To understand the material in the summary, it may occasionally be necessary to refer back to the earlier sections. It should be noted that the release formulas in Section 3 are more general than those in Sections 4 and 5 because the formulas in the later sections assume a particular form for some of the probability distributions of the model parameters. The last section contains a discussion of the parameters for the release formulas. There is very little information available for many of the parameters, so there is a very wide range of possible values. This is one of the best reasons for using relatively simple release models. As the release processes become better understood and as more data become available, the bounds of uncertainty will draw in, and more-detailed source models will become more appropriate for total-system assessments. There is additional material in three appendices: Appendix A contains a table of the notation used, for quick reference, Appendix B contains radionuclide data of use in total-system performance assessment, and Appendix C contains a listing of a computer implementation of the release formulas in Section 5.

As this report is written, the EPA is reconsidering its standards for radioactive waste disposal and expects to issue a revised version. The revised version will probably have little effect on the discussions in this report, because they do not depend on details of the standards. So long as the standards rely on performance assessment for the demonstration of compliance, they will require the development of radionuclide source terms.

2 The radionuclide inventory

The nation's first repository for high-level nuclear waste is expected to have 70,000 metric tons of nuclear waste: about 54% spent nuclear fuel from pressurized-water reactors (PWRs), 35% spent fuel from boiling-water reactors (BWRs), and 11% glassified high-level waste. These numbers are derived from Tables 6-8 and 7-2 of the Yucca Mountain Site Characterization Plan (DOE, 1988; hereafter referred to as the SCP). This report will be concerned only with source release models for spent fuel; glass waste will not be mentioned again. Most experimental work is being done with PWR fuel, but BWR fuel is very similar. The release models should be valid for either, but the input parameters for the models may be different for the two types of fuel.

The EPA limits in 40 CFR Part 191—the total-system release limits—pertain only to radioactive nuclides with half-lives greater than 20 yr, so the discussion in this report will be concerned solely with such nuclides. In addition to the EPA regulations, which concern radioactivity in the far-field, there are U. S. Nuclear Regulatory Commission regulations governing release rates of radioactivity from the engineered-barrier system in 10 CFR Part 60 (NRC, 1983). The NRC containment requirements in 10 CFR 60.113 are *not* limited to long-lived radionuclides, so a complete discussion of performance assessment of the engineered-barrier system would require including additional species.

Two sample radionuclide inventories are listed in Appendix B, one for PWR fuel and one for BWR fuel. Both inventories are for spent fuel 10 years out of the reactor and only list species with half-lives greater than 20 yr. The PWR inventory is for fuel with a burnup of 33,000 MWd/MTHM (megawatt-days per metric ton of heavy metal) and the BWR inventory is for fuel with a burnup of 27,500 MWd/MTHM. These burnups are considered to be typical of the spent fuel to be stored in a waste repository. A realistic inventory would be a combination of PWR and BWR inventories, with burnups ranging from a few thousand MWd/MTHM up to 60,000 MWd/MTHM and with ages ranging from 10 yr to over 50 yr. For preliminary performance-assessment work, it should be adequate to use one of the two inventories from Appendix B or a combination of them. The two inventories are similar enough that most results should not be sensitive to the choice. Note that the age of the waste will not be a large factor in these inventories because only the long-lived radionuclides are included. The age of the waste does, however, affect other aspects of the repository environment such as the temperature.

The radionuclide inventories in Appendix B were taken from the Nuclear Waste Terminal Storage Program (NWTSP) database (Roddy *et al.*, 1986). They were calculated using the ORIGEN2 computer program. There are 83 nuclides in the tables in Appendix B. For most performance-assessment calculations, only a small subset of those nuclides will make significant contributions. It is desirable to eliminate as many as possible from consideration at the outset to simplify the calculations of the releases and subsequent transport. For total-system performance assessment, releases to the accessible environment are to be calculated in terms of the *EPA sum*. The EPA sum is defined as follows: The releases of radioactivity to the accessible environment

are calculated for all radionuclides with half-lives greater than 20 yr. The amount of release for each nuclide is then divided by the EPA limit for that nuclide (see Appendix B) to form the *EPA ratio* for that nuclide. The EPA sum is the sum of all the EPA ratios. The EPA regulations place limits on the probability that the EPA sum is greater than 1 and on the probability that the EPA sum is greater than 10.

Clearly, then, the inventory of a radionuclide divided by its EPA limit is a measure of its importance to the total-system performance assessment. This quantity will be called the potential EPA ratio, as it is the EPA ratio that would result if the entire inventory of that nuclide were instantly released to the accessible environment. The inventory of each nuclide changes with time because of radioactive decay and generation, and so the potential EPA ratio is also a function of time. For example, the potential EPA ratio for ^{137}Cs is very high initially, as can be seen from Tables B.1 and B.2, but it decays to a low value within a few hundred years. On the other hand, the potential EPA ratios for some nuclides, such as ^{230}Th , increase with time because of decay of parent nuclides. As a first criterion to limit the number of nuclides to only the most important ones, we could consider only those that have potential EPA ratios higher than 1 at some time during the 10,000-yr regulatory period. There are about 25 such nuclides. We really need to consider more nuclides than that, though, because the regulation governs EPA sums rather than EPA ratios. To be safe, we will include all nuclides that have potential EPA ratios greater than 0.02 in the following discussion. Also, because some total-system calculations may use a time period greater than 10,000 yr, all nuclides with potential EPA ratios greater than 0.02 at any time during the first 100,000 yr will be considered here. For the sample inventories in Appendix B, 39 nuclides for the PWR inventory and 38 nuclides for the BWR inventory meet this criterion. The 39 nuclides are shown in Table 1. In addition to the PWR and BWR inventories from Tables B.1 and B.2, a third inventory is shown in Table 1. This third inventory is a linear combination of the other two, given by 60% of the PWR inventory plus 40% of the BWR inventory. This is the approximate ratio that is expected for the first spent-fuel repository (DOE, 1988).

Table 1 is organized by decay chains. To simplify calculations, decay can be computed using simple decay chains with no branching. The breakdown into chains is indicated in the table by lines between the chains. Within each chain, the progression is downward—that is, the first one decays to the second one, etc.

Let $I_0^i(t)$ be the inventory as a function of time for nuclide i . The subscript 0 is meant to indicate that this is the time variation taking only radioactive decay into account (i.e., not subtracting releases as they occur). For nonbranching decay chains, $I_0^i(t)$ is the solution of

$$\frac{d}{dt}I_0^i = -\lambda^i(I_0^i - I_0^{i+1}), \quad (1)$$

where the second term on the right-hand side represents generation of daughter nuclide i as its parent nuclide $i+1$ decays, and the first term represents decay of nuclide i . For Equation 1 to be valid, the inventory I_0^i must be given in terms of radioactivity (e.g., curies). Units of mass or number can be used for the inventory, but then a factor containing the activities of nuclide i and nuclide $i+1$ must be included in the decay

Table 1: Species with potential EPA ratio greater than 0.02, grouped by decay chain.

Species	PWR 33,000 inv. (Ci/MTHM)	BWR 27,500 inv. (Ci/MTHM)	Combination* (Ci/MTHM)	Location
²⁴⁶ Cm	3.12×10^{-2}	1.73×10^{-2}	2.56×10^{-2}	matrix
²⁴² Pu	1.72	1.42	1.60	matrix
²³⁸ U	3.17×10^{-1}	3.20×10^{-1}	3.18×10^{-1}	matrix
²⁴² Am m	6.93	8.33	7.49	matrix
²³⁸ Pu	2.33×10^3	1.78×10^3	2.11×10^3	matrix
²³⁴ U	1.19	1.04	1.13	matrix
²³⁰ Th	1.33×10^{-4}	1.23×10^{-4}	1.29×10^{-4}	matrix
²²⁶ Ra	3.69×10^{-7}	3.64×10^{-7}	3.67×10^{-7}	matrix
²¹⁰ Pb	4.66×10^{-8}	4.79×10^{-8}	4.71×10^{-8}	matrix
²⁴³ Cm	1.66×10^1	1.34×10^1	1.53×10^1	matrix
²⁴³ Am	1.71×10^1	1.29×10^1	1.54×10^1	matrix
²³⁹ Pu	3.13×10^2	3.00×10^2	3.08×10^2	matrix
²³⁵ U	1.72×10^{-2}	1.63×10^{-2}	1.68×10^{-2}	matrix
²³¹ Pa	1.92×10^{-5}	1.98×10^{-5}	1.94×10^{-5}	matrix
²²⁷ Ac	5.08×10^{-6}	5.37×10^{-6}	5.20×10^{-6}	matrix
²⁴⁵ Cm	1.46×10^{-1}	9.25×10^{-2}	1.25×10^{-1}	matrix
²⁴¹ Am	4.28×10^3	3.88×10^3	4.12×10^3	matrix
²³⁷ Np	3.15×10^{-1}	2.46×10^{-1}	2.87×10^{-1}	matrix
²³³ U	2.86×10^{-5}	2.04×10^{-5}	2.53×10^{-5}	matrix
²²⁹ Th	1.51×10^{-7}	1.22×10^{-7}	1.39×10^{-7}	matrix
²⁴⁰ Pu	5.27×10^2	4.78×10^2	5.07×10^2	matrix
²³⁶ U	2.56×10^{-1}	2.15×10^{-1}	2.40×10^{-1}	matrix
²³² U	2.98×10^{-2}	1.74×10^{-2}	2.48×10^{-2}	matrix
¹⁵¹ Sm	3.31×10^2	2.98×10^2	3.18×10^2	matrix
¹³⁷ Cs	8.21×10^4	6.79×10^4	7.64×10^4	matrix, gap
¹³⁵ Cs	3.45×10^{-1}	3.59×10^{-1}	3.51×10^{-1}	matrix, gap
¹²⁹ I	3.15×10^{-2}	2.64×10^{-2}	2.95×10^{-2}	matrix, gap
¹²⁶ Sn	7.76×10^{-1}	6.24×10^{-1}	7.15×10^{-1}	matrix, gap
¹²¹ Sn m	6.83×10^{-1}	1.25	9.10×10^{-1}	structural
¹⁰⁷ Pd	1.12×10^{-1}	9.46×10^{-2}	1.05×10^{-1}	matrix, gap
⁹⁹ Tc	1.31×10^1	1.11×10^1	1.23×10^1	matrix, gap
⁹³ Mo	2.59×10^{-2}	5.97×10^{-4}	1.58×10^{-2}	matrix, gap, structural
⁹⁴ Nb	1.28	3.15×10^{-2}	7.81×10^{-1}	structural
⁹³ Zr	1.93	1.80	1.88	matrix, cladding
⁹⁰ Sr	5.72×10^4	4.70×10^4	5.31×10^4	matrix, gap
⁷⁹ Se	4.09×10^{-1}	3.38×10^{-1}	3.81×10^{-1}	matrix, gap
⁶³ Ni	6.52×10^2	1.47×10^2	4.50×10^2	structural
⁵⁹ Ni	5.15	1.07	3.52	structural
¹⁴ C	1.55	1.53	1.54	matrix, gap, cladding, structural

*Combination inventory is 60% PWR and 40% BWR.

equation. The radioactive decay rate λ is related to the half-life $\tau_{1/2}$ by $\lambda = \ln 2 / \tau_{1/2}$. The inventories listed in Table 1 (multiplied by 70,000 MTHM if the entire repository inventory is desired) are the initial values. For a single-member chain, the solution is simple exponential decay, $I_0^i(t) = I_0^i(0) \exp(-\lambda^i t)$. For chains with more than one member, the solution can be expressed as a linear combination of exponentials (Bateman, 1910).

As an illustration, Figure 1 shows the normalized inventories (referred to previously as potential EPA ratios) as a function of time for all 39 nuclides in Table 1, calculated using the nonbranching-chain approximation. The combination inventory was used in generating this figure. At the right of the figure the nuclides are listed in order by one measure of "importance," the maximum value of the normalized inventory during the 100,000-yr period of the figure. There is too much information in Figure 1 for the reader to be able to decipher it easily, but the figure shows that initially the dominant contributors to the potential EPA sum are ^{137}Cs , ^{90}Sr , and ^{241}Am . After 10,000 yr, those species have mostly decayed away and the dominant contributors are ^{239}Pu and ^{240}Pu . By a time of 100,000 yr, most of the ^{240}Pu has also decayed away and the dominant nuclides are ^{239}Pu and ^{230}Th . The case of ^{230}Th is particularly interesting because there is very little of it initially, as can be seen in Table 1. Figure 1 also shows clearly the challenge of waste isolation: The potential EPA sum is over 100,000 initially, so the combined engineered-barrier and natural-barrier systems must ensure that less than 0.001% of the radioactivity is released to the accessible environment within 10,000 years.

Very little accuracy is lost by the nonbranching-chain approximation. This statement can be verified by comparison with the solutions to the general decay equations (that is, including all species and the correct branching ratios), as tabulated in the NWTSP database (Roddy *et al.*, 1986). Up to 100,000 yr the only two species that have significant inaccuracy are ^{241}Am at early times and ^{232}U at late times. The inventories shown for ^{241}Am in Table 1 are different from those shown in Appendix B because in Table 1 the ^{241}Pu inventory has been lumped with ^{241}Am . ^{241}Pu decays into ^{241}Am with a half-life of only 14.4 yr; because of its short half-life, ^{241}Pu is not listed in any of the tables, but it has a large initial inventory that cannot be ignored. Lumping its inventory with ^{241}Am makes the amount of ^{241}Am too high for about the first 100 yr in Figure 1. Since no releases are expected for the first 100 yr, this should not be a problem. Also, this approximation is conservative in that the model overpredicts any early releases. The amount of ^{232}U shown in Figure 1 is too low after 100 yr or so because decay of ^{236}Np into ^{232}U , with a branching ratio of about 9%, has been neglected. However, the amount of ^{232}U is small enough that it is not very important; its entire inventory has a potential EPA ratio of less than 0.01 after about 300 yr.

Some other species are off by small amounts. The daughters of ^{241}Am (^{237}Np , ^{233}U , ^{229}Th) are high for a while because of the extra ^{241}Am at the beginning, but only by a few percent. After 1000 yr or so, the amount of ^{238}Pu is high because in reality only 83% of ^{242}Am decays into it, not 100%. However, there is much less ^{242}Am than ^{238}Pu initially, and the potential EPA ratio for ^{238}Pu is very small by the time it is off by very much.

Figure 1: Normalized inventories (potential EPA ratios) as a function of time for the combination inventory.

The decay scheme shown in Table 1 is only appropriate for about 100,000 yr. After that, it is necessary to take into account the decay of ^{238}U into ^{234}U . This decay scheme is acceptable for total-system performance assessment because the time period governed by the EPA regulations is only 10,000 yr and very few calculations are continued for more than 100,000 yr.

Also shown in Table 1 is the location within the spent fuel of the listed species. Figures 2 and 3 show the various locations pictorially. Each container will have 2 to 3 MTHM of radioactive waste, including the spent fuel rods and assembly hardware (Apted *et al.*, 1991, assumed 2.1 MTHM per container). *Matrix* refers to the spent fuel pellets themselves. The matrix is primarily uranium dioxide, with the other species present as small impurities. It will be assumed that most of the impurities are uniformly distributed throughout the UO_2 matrix and are released only as the matrix is dissolved (congruent leaching). Many of the fission products are not distributed completely uniformly within the fuel matrix, but can be concentrated in the pellet/cladding *gap* and the grain boundaries within the pellets. This fraction of the inventory can be released more quickly than the part that is in the matrix. The grain-boundary inventory is not released as fast as the gap inventory, but the two will be lumped together here. There is also radioactivity within the Zircaloy *cladding* (a small fraction of the fuel rods are clad with stainless steel, a minor detail at this level of abstraction). There are small quantities of several radionuclides in the cladding, including some activated ^{93}Zr , but most of the radioactivity in the cladding comes from ^{14}C . Lastly, there are various *structural* metal (stainless steel and Inconel) components present in addition to the fuel rods, and they contain some radioactive species. (In Figure 2 the structural metals are shown as “fuel assembly hardware.”)

3 Release models

Having defined the radionuclide inventory to be used, next we need a model (or models) for the release of these radionuclides from the waste containers. The release model to be described is summarized schematically in Figure 4.

The necessary first step to get any release of radioactivity is container failure. Failure of a container is taken to mean that it develops a hole, thereby making it possible for radioactive waste to escape. Let us focus our attention on a single container that fails at time t_c . Upon container failure, the contents of the container are exposed to air and it becomes possible for water to enter the container. Previously, four radionuclide locations were defined (see Table 1, Figure 2, and Figure 3). When the container fails, two of the locations, the cladding and the structural metals, become exposed and may begin to release radioactivity. The other two locations, the matrix and the gap/grain-boundaries, are still protected by the cladding until it fails. So, the next important time is the time of cladding failure, t_f . As shown in Figure 4, the cladding-failure time will be measured from the time of container failure. This is because the cladding is assumed to be intact at the time of container failure and to start breaking down at that time. There will be some fuel rods with cladding already breached before emplacement, but these are assumed to be negligible (Woodley, 1983,

Figure 2: Spent-fuel-container designs (taken from Figure 7-2 of the SCP).

Figure 3: Schematic cross-section of a spent fuel rod (taken from Figure 1.1 of Apted *et al.*, 1989).

Figure 4: Sequence of times for the simplified release model. (a) Container and cladding failure before resaturation. (b) Resaturation after container failure but before cladding failure. (c) Container and cladding failure after resaturation. Dashed lines indicate releases of gaseous ^{14}C but no aqueous releases. A $\otimes$ indicates a pulse release. The time (t_r , t_m , etc.) refers to the length of the arrow, not the position of the tip of the arrow.

reports a failure level of about 0.01%). The rods that are intact initially (which is the vast majority of rods) are expected to remain intact because the containers are to be filled with inert gas, thus preventing oxidation until the containers are breached and air can enter. Any fuel rod with cladding failure before container failure will be assigned a cladding-failure time of $t_f = 0$.

The "water re-entry time" or "resaturation time," t_r , is also included as a parameter. This represents the amount of time that the containers are hot enough to exclude liquid water. This time is not taken into account in determining container failures (that is, containers may fail during the time t_r because of corrosion in the air-steam mixture present or for other reasons), but it is assumed that waste cannot be released until after t_r . The sole exception is ^{14}C . It is well-known that some of the ^{14}C in the cladding can be released in gaseous form (as carbon dioxide) when water is not present (Van Konynenburg *et al.*, 1985, 1987).

The remaining three times from Figure 4 represent processes rather than events. That is, t_s , t_z , and t_m are time periods rather than being points in time at which something happens like t_c , t_f , and t_r . Each of the three time periods represents one of the radionuclide locations and is the amount of time it takes for all the radionuclides in that location to be released. Radionuclides in the fourth location, the gap/grain-boundaries, will be assumed to be released instantly, so there is no time scale for that release. The time for radionuclides to get out of a waste container and into the groundwater flow system after dissolution (which could be thousands of years under some circumstances) is neglected in this report, but another parameter could be added to represent such a time lag. Neglecting this time lag is conservative since the model then predicts releases earlier than they are actually expected.

Release rates will be assumed to be constant during the release periods, except for variations caused by radioactive decay. In fact, of course, there may be considerable time variation in the release rates. Part of the variation is probably random or unpredictable, and the assumption being made here is that the random fluctuations are not important over the time scales of interest. There are also nonrandom variations in the release rates that could be simulated with a more sophisticated model. For example, there is a strong temperature dependence for many corrosion processes. Since the temperature as a function of time is known (at least approximately), the temperature dependence of the corrosion time scales could be included. This is not done for the simple release model described in this report. The time variation of the high-level parameters used in this release model is not known in most cases, and inclusion of such effects would undoubtedly make the release formulas substantially more complicated. Also, temperature effects are probably most important in the first few hundred years, and of decreasing importance thereafter.

The cladding and the structural metals are assumed to start corroding as soon as the container fails. They should not corrode significantly before container breaching because of the inert gas filling the container. It is assumed that after container failure there is steady corrosion, characterized by the amount of time to completely corrode away. This corrosion time is denoted by t_s for the structural metals and by t_z for the fuel-rod cladding. As the cladding or structural metals corrode, the radionuclides contained in them are assumed to be made available for release congruently; that is,

the fraction of a given nuclide in the structural metal available for release over some period of time is equal to the fraction of the structural metal that corrodes in that period of time. For all nuclides except ^{14}C , release from the container cannot take place until after the resaturation time t_r because until then no liquid water is present to dissolve the waste. However, for simplicity it is assumed that corrosion takes place at the same rate before and after $t = t_r$. Under the relatively dry conditions that are likely in a partially saturated repository, that may even be a fair assumption, as the predominant environment before and after $t = t_r$ is moist air, with little liquid present. There may be conditions that allow for immersion of the waste in water. In such a case the assumption of constant corrosion rate would not be very realistic because the corrosion rate in water is significantly higher than in moist air (Oversby and McCright, 1985). However, the constant-rate assumption is conservative in that, if the higher rate is used to determine t_s then the calculated releases at earlier times (before the immersion) are higher than they would actually be.

It will be assumed that, as the cladding or structural metal corrodes, the ^{14}C is immediately released as carbon dioxide but that the other radioactive species are not released until after $t = t_r$. To be consistent with the assumptions stated so far, it is necessary to account for the amount of the other species in the corrosion layer at $t = t_r$. This will be assumed to be released as a pulse at the time t_r . This is presumably a very small fractional release because t_r should be much smaller than t_s . It will also be assumed that there is a pulse release of ^{14}C at the time of container failure, t_c . The ^{14}C in the cladding is not distributed uniformly. A significant fraction is in a thin oxidation layer on the outside of the cladding (labelled "crud" in Figure 3). This fraction can be released as soon as the container is breached and the cladding is exposed to air (Van Konynenburg *et al.*, 1985, 1987). The "quick-release" fraction of the ^{14}C inventory will be considered a model parameter, f_q . Other fraction parameters associated with the cladding and structural metals are f_s , the fraction of the ^{14}C inventory contained in the structural metals; f_c , the fraction of the ^{14}C inventory contained in the cladding (not counting the quick-release fraction); f_m , the fraction of the ^{93}Mo inventory contained in the structural metals; and f_z , the fraction of the ^{93}Zr inventory contained in the cladding. Of course, all the fractions for a given radionuclide must add up to one.

The discussion in the preceding few paragraphs may be condensed into the following equations for the source terms (release rates) associated with the cladding and structural metals. For a container that fails at time t_c , the source term for releases from the structural metals is given by

$$\Sigma_s = \begin{cases} I_0(t)t_s^{-1}H(t-t_c)H(t_c+t_s-t) & \text{for } t_r < t_c, \\ I_0(t)t_s^{-1}H(t-t_r)H(t_c+t_s-t) \\ \quad + I_0(t_r)t_s^{-1}(t_r-t_c)\delta(t-t_r) & \text{for } t_c < t_r < t_c+t_s, \\ I_0(t_r)\delta(t-t_r) & \text{for } t_r > t_c+t_s. \end{cases} \quad (2)$$

In this equation, Σ_s is the release rate of radioactivity for one of the radionuclides present in the structural metals, I_0 is the inventory of that nuclide, H is the Heaviside

step function, and δ is the Dirac delta function. I_0 was introduced previously in Equation 1; the superscript i has been dropped for convenience. All the nuclides present in the structural metals in significant quantity are single-member chains, so I_0 is just an exponential function. Since only a portion of ^{93}Mo and ^{14}C are in the structural parts, I_0 should be replaced by $f_s I_0$ for ^{14}C and by $f_m I_0$ for ^{93}Mo . Also, ^{14}C does not have to be dissolved in water to be released, so the first case in Equation 2 is always applicable to ^{14}C , regardless of the value of t_r . Except for the third case in Equation 2, the release is such that the inventory is completely depleted at time $t_c + t_s$, or a time t_s after container failure. The third case is probably not needed, as it seems very unlikely that t_r could be greater than $t_c + t_s$.

For cladding releases, only two nuclides are being considered: ^{14}C and ^{93}Zr (see Table 1). The source term for ^{93}Zr is the same as Equation 2, with t_s replaced by t_z :

$$\Sigma_c = \begin{cases} f_z I_0(t) t_z^{-1} H(t - t_c) H(t_c + t_z - t) & \text{for } t_r < t_c, \\ f_z I_0(t) t_z^{-1} H(t - t_r) H(t_c + t_z - t) \\ \quad + f_z I_0(t_r) t_z^{-1} (t_r - t_c) \delta(t - t_r) & \text{for } t_c < t_r < t_c + t_z, \\ f_z I_0(t_r) \delta(t - t_r) & \text{for } t_r > t_c + t_z. \end{cases} \quad (3)$$

Recall that f_z is the fraction of the ^{93}Zr inventory contained in the cladding. As before, the source term for ^{14}C is always of the first form in Equation 3, regardless of the value of t_r . However, in addition there is the quick-release fraction of ^{14}C , released from a thin layer on the surface of the cladding. This part of the ^{14}C inventory is assumed to be released immediately upon container failure, so the cladding source term for ^{14}C is given by

$$\Sigma_c = f_c I_0(t) t_z^{-1} H(t - t_c) H(t_c + t_z - t) + f_q I_0(t_c) \delta(t - t_c). \quad (4)$$

Now let us consider the species in the uranium-dioxide matrix and the gap and grain boundaries. Species in the gap/grain-boundary fraction of the inventory will be assumed to be released immediately when the cladding is breached if liquid water is present, and at the time of resaturation if water is not present. The fraction of the inventory in the pellet/cladding gap and the grain boundaries will be denoted by f_g , for all the nuclides that have the “gap” notation in Table 1. This fraction may not be exactly the same for all relevant species but it is probably fairly close, and for simplicity in this report it will be taken to be the same for all. The exceptions to this are ^{14}C and ^{93}Mo . Since those two nuclides have a substantial portion of their inventories in the cladding and/or structural metals, there is no reason to expect that the fraction of their inventories in the gap and grain boundaries would be the same as for the other nuclides. Instead, the simplest assumption would be for them to have the same ratio of gap to matrix inventories. Thus, for ^{14}C , $f_g(1 - f_s - f_c - f_q)$ should be used in the formulas derived below instead of f_g , and for ^{93}Mo , $f_g(1 - f_m)$ should be used instead of f_g .

The vast majority of the radioactivity of the spent fuel is in the matrix, so in that sense the matrix release model is the most important. It may not turn out to be the most important in the end, though; the quick-release fraction of ^{14}C and

the gap fraction of several other nuclides are critical because they may be released very quickly. The fuel matrix, on the other hand, can start dissolving only after container and cladding failure and after water re-entry; thus, three factors contribute to preventing early releases from the matrix. Details of the matrix dissolution will be ignored, and it will simply be assumed that after the cladding is breached and it is possible for water to contact the fuel pellets, the fuel matrix will be dissolved at a constant rate, with congruent release of all component species. The release rate will be parameterized by the time it takes the matrix to dissolve completely, t_m , a quantity analogous to t_s and t_z . Some recent work (Wilson and Bruton, 1989; Apted *et al.*, 1991) has suggested that releases from the fuel matrix are not limited by uranium dissolution, but rather by the rate of alteration of the uranium from one state to another. This process may be thought of as dissolution and re-precipitation of uranium, which allows the more soluble elements to escape even though uranium remains in the solid phase. Laboratory measurements of spent-fuel dissolution have measured fractional release rates for soluble nuclides such as ^{99}Tc and ^{129}I as high as 10^{-3} yr^{-1} (Wilson, 1987). The interpretation of these results is unclear at this point; the high release rate may be due to dissolution of those species from the grain boundaries or it may be due to progressive alteration of the UO_2 matrix. The latter interpretation requires that the matrix-release time scale t_m be given by the matrix alteration rate rather than by the matrix dissolution rate.

For a fuel rod that fails at time $t_c + t_f$ within a container that fails at time t_c , the source term for matrix and gap releases as just described may be written as

$$\Sigma_m = (1 - f_g)I_0(t)t_m^{-1}H(t - t_x)H(t_x + t_m - t), \quad (5)$$

$$\Sigma_g = f_g I_0(t_x)\delta(t - t_x), \quad (6)$$

where t_x is the greater of t_r and $t_c + t_f$; i.e., $t_x = \max\{t_r, t_c + t_f\}$. Release begins at time t_x , and the inventory is completely depleted at time $t_x + t_m$. In Equations 5 and 6, f_g should be taken to be zero for those species that have no gap fraction. For ^{14}C , $(1 - f_g)$ should be replaced with $(1 - f_g)(1 - f_s - f_c - f_q)$ and f_g should be replaced with $f_g(1 - f_s - f_c - f_q)$; for ^{93}Mo , $(1 - f_g)$ should be replaced with $(1 - f_g)(1 - f_m)$ and f_g should be replaced with $f_g(1 - f_m)$; and for ^{93}Zr , $(1 - f_g)$ should be replaced by $(1 - f_z)$ and f_g should be replaced with 0.

It is useful to consider the dependence of t_m on other variables. A similar analysis could be carried out for t_s and t_z ; the issues are slightly different in each case. This discussion will focus on the case in which t_m represents a matrix-dissolution time. The dependence of matrix-alteration time on other quantities, such as the amount of water present, is not known. The matrix-dissolution time t_m depends on the amount of water contacting the fuel pellets, how long it contacts the fuel pellets, and how fast the UO_2 matrix dissolves in the water. Clearly, these issues depend on a great many factors, including how the containers fail, how water flows through the containers, and the details of the chemistry in the containers. For the first part of the discussion, the following model of release will be assumed: A container has failed and water is able to flow into it. Either the water flows through the container or it fills the container with water until it starts overflowing. When water leaves the container, it

carries with it some dissolved radionuclides. This basic model of releases has been used by many authors, including Sinnock *et al.* (1984) and Apted *et al.* (1991). The dissolution time can be written as

$$t_m = M_U / \dot{V}_w C_U, \quad (7)$$

where M_U is the number of moles of uranium initially in the fuel matrix, $\dot{V}_w$ is the volume of water per unit time contacting M_U , and C_U is the effective release concentration of uranium (in moles per unit volume). That is, C_U is the average uranium concentration of the water after it leaves the waste. C_U may be equal to the solubility limit of uranium in groundwater, but not necessarily; it depends on whether water is in contact with the fuel pellets long enough to become saturated. The values M_U and $\dot{V}_w$ could be for a single container, in which case t_m would be the dissolution time for that container, or they could be summed over some or all of the containers, in which case t_m would be an average release time for that set of containers.

During the period of release as given by Equation 7, the molar concentration of element i in the water leaving the waste is given by

$$C_i = C_U M_i(t) / M_U, \quad (8)$$

where M_i is the number of moles of element i in the fuel matrix, which would be given by $(1 - f_g)I_0/a$, where I_0 is the inventory and a is the molar activity. If more than one isotope of an element is present, all contribute to the concentration (including nonradioactive isotopes). M_i varies with time because the inventory I_0 varies with time, but the formulas above assume that M_U and C_U are constants. It is a good approximation to consider M_U as constant because most of the uranium is ^{238}U , which has a half-life of over four billion years; C_U is only constant insofar as the physical environment is unchanging. Several elements have very low solubility limits and may not be able to leach congruently (Kerrisk, 1984, 1985). The release rates for those elements may be overestimated by the model outlined above for matrix releases. A better estimate of the release rate for elements with low solubilities may be obtained by using

$$\Sigma_m = a \dot{V}_w S \quad (9)$$

for them whenever $S < C_U M / M_U$, with S the solubility limit of the element. If there is more than one isotope of the element, the releases should be divided among the isotopes in proportion to the number of moles of each one (once again, nonradioactive isotopes would have to be included). If t_m is an alteration time rather than a dissolution time, the criterion for solubility-limited release would be $S < M / \dot{V}_w t_m$. From this expression it can be seen that the lower t_m is, the more species will be solubility-limited. If a low value of t_m is used, consideration of solubility limits for the elements may be more important. If a high value is used for t_m (that is, slow release), it may be acceptable to ignore the fact that releases of low-solubility elements such as plutonium and americium should be even slower, but if the alteration time scale is as low as 1000 yr (or even if it is 10^5 yr), it will probably be necessary to include the effects of solubility limits for many elements. As a compromise, two time scales could be used, a dissolution time t_d and an alteration time t_a , with $t_m = t_a$

being used to calculate releases of the most soluble elements and $t_m = t_d$ being used to calculate releases of the less soluble elements (including the actinides). The latter philosophy will be used in this report. Additional formulas are developed in the next section, starting from Equation 5. Release rates are given as a function of t_m ; it is implicitly understood that there could be more than one value of t_m , applying to different elements.

As an aid to the reader, the solubility limits from Kerrisk (1985) are reproduced in Table 2. This table does not show solubilities for all the elements in Table 1, and the ones that are shown are not really reliable. The chemistry of dissolving spent fuel is not yet well understood, even under controlled laboratory conditions. There are measured (and calculated) solubilities that are very different from the values in Table 2.

Some authors have also considered diffusive release from containers (e.g., Kerrisk, 1984; Apted *et al.*, 1991). This type of release is more appropriate to a saturated repository, but it is possible under some circumstances in a partially saturated repository. Preliminary plans call for an air gap between the container and the wall of the emplacement borehole. In order to have diffusive releases, not only does the container have to be breached, but the air gap must be bridged somehow—the gap must be at least partially filled by rubble or by water, or the container must be knocked over so that it contacts the wall.

For high water flux, the release time scale can still be given by Equation 7, but with the water-volume rate now defined by the model:

$$\dot{V}_w \simeq \pi q \left[R + 2\sqrt{\theta D_e L / \pi q} \right]^2, \quad (10)$$

where q is the volume flux of water, R is the container radius, L is the container length, θ is the volumetric moisture content of the surrounding rock, and D_e is the effective diffusion coefficient. Equation 10 gives the volume of water per unit time for a single container; it assumes vertical container emplacement and is taken from McGuire *et al.* (1990). A similar formula, for horizontal emplacement, was given by Chambré *et al.* (1982).

Table 2: Solubility limits of important waste elements (from Kerrisk, 1985).

Element	Solubility (mol/m ³)	Element	Solubility (mol/m ³)
C	large	Sm	2×10^{-6}
Ni	10	Ra	3×10^{-4}
Sr	8×10^{-1}	Th	1×10^{-6}
Zr	1×10^{-7}	U	4
Tc	large	Np	1
Sn	1×10^{-6}	Pu	1×10^{-2}
I	large	Am	1×10^{-3}
Cs	large	Cm	1×10^{-3}

A distinguishing feature of diffusive release is that there is release even when there is no water movement. The limiting formula for small water flux is

$$t_m \simeq \frac{\ln(L/R)M_U}{2\pi D_e L C_U} \quad (11)$$

(Chambré *et al.*, 1982). As with Equation 10, this equation is written for a single container.

There are three things to keep in mind about these diffusive-release formulas. (1) Equations 10 and 11 were derived assuming that waste is diffusing out from the entire surface of the container. The container and fuel-rod cladding would both have to be almost completely disintegrated for this assumption to be true. A more realistic assumption would be for diffusion to take place only through holes in the container and the cladding. This assumption would reduce the releases considerably below those given by Equations 10 and 11. (2) The effective diffusion coefficient through rubble filling the air gap could be several orders of magnitude below the normal diffusion coefficient in the host rock (Apted *et al.*, 1991). (3) Equations 10 and 11 are for releases after a steady state has been achieved. It may take hundreds or even thousands of years to reach steady state (Chambré *et al.*, 1982).

4 Probability distributions

The formulas derived in the previous section assumed specific values of the model parameters. Clearly, however, the containers will not all fail at the same time, the fuel rods will not all fail at the same time, etc. There will be a distribution of container failures over time, which means that we need to consider a probability distribution for the parameter t_c , and similarly for the other model parameters. Note that having a distribution of parameter values is different from having time variation of parameters. The distribution referred to here is a distribution that reflects the differences among containers and fuel rods; the parameters for particular containers or fuel rods are still considered to be constant in time. Including time variation of the parameters is more difficult than allowing a probability distribution of parameter values and is beyond the scope of this report.

One way to introduce parameter variability into a total system performance assessment is by use of a Monte Carlo method, in which multiple "realizations" are calculated, with each realization using different values of the parameters, drawn from predetermined probability distributions. Parameter variability can also be included directly by taking the release formulas from the previous section and integrating them with probability distribution functions of the parameters. This can be difficult, especially if the probability distributions are not simple mathematical functions. It is also possible to combine the two methods, perhaps including some parameter distributions directly and other parameter distributions by Monte Carlo methods.

In this section, some examples will be given of the direct method, in which the release submodels are explicitly integrated over simple probability distributions. The model parameters introduced in the previous section are of three kinds. There are

times at which events take place, t_c , t_f , and t_r ; there are time scales that characterize processes, t_s , t_z , and t_m ; and there are fractions that tell how certain radionuclides are distributed within the spent fuel, f_q , f_s , f_c , f_m , f_z , and f_g . The examples presented in this report only involve integration over distributions of parameters of the first kind (t_c , t_f , t_r), but the same procedure can be used with distributions of the other parameters. Because t_c , t_f , and t_r characterize starting times of the release processes, integration of their probability distributions is not trivial and requires consideration of numerous cases depending on the order of the events. (Is t_c greater than t_r or is it less? Is $t_c + t_f$ greater than or less than t_r ? And so on.) Including probability distributions for these parameters is more straightforward than for t_s , t_z , and t_m , though, because they represent discrete events. The process time-scales are treated as constants in this report, but in reality they should be functions of time as well as having distributions of values over the ensemble of containers. They can be given distributions in the same way as will be demonstrated for the event times, but making them functions of time would require additional techniques. Time variation should not be a problem for the fraction parameters (all the f s) and they appear linearly in the release formulas, so it would be easy to put in probability distributions for them, but it is not done in this report.

Another issue in assigning probability distributions to the parameters is their interdependencies. In the examples below, distributions are assigned to t_c , t_f , and t_r independently. But in a more realistic approach the distribution of t_c would depend on the distribution of t_r because the container environment is different before and after t_r , and the distribution of t_f would depend on the distribution of t_c because the temperature will be higher at the time of container failure for the ones that fail earlier and the cladding corrosion is strongly temperature dependent. And, of course, all the parameters depend on other unknowns such as how wet the repository environment is and what the geochemistry is like. Clearly, these dependencies could be spatially variable as well. In fact, spatial variability can be included even in the simple approach followed below. The probability distributions can be made functions of position so that, for example, the resaturation time occurs earlier for the containers at the edge of the repository than for containers in the center of the repository. Whether such spatial discrimination is useful depends on the detail of the overall modeling effort. If flow and transport modeling is being done with only one spatial dimension, then one would not include any spatial variability in the source model, but if modeling is being done in two or three dimensions then the effects of spatial source variability could be explored.

Before going on, let us define what is meant by a probability distribution function. A probability distribution of a variable x is a function $P(x)$ (not necessarily an ordinary function, but including "generalized" functions such as the Dirac delta function) such that the probability of x being in the infinitesimal interval from x' to $x' + dx'$ is given by $P(x') dx'$. The probability that x is in any given interval is found by integrating $P(x)$ over the interval; for example, $Pr(x \in [a, b]) = \int_a^b P(x') dx'$. The function $P(x)$ is referred to as the probability density function (PDF), and its integral from the bottom of its domain ($\int_{-\infty}^x P(x') dx'$ for a variable defined on the whole real line) is called the cumulative distribution function (CDF).

Consider the model for releases from the structural metals given in Equation 2. The release rate Σ_s is given as a function of time t and of the parameters t_c , t_r , and t_s : $\Sigma_s = \Sigma_s(t; t_c, t_r, t_s)$. Suppose now that we have probability distributions for the parameters: $P_1(t_c)$, $P_2(t_r)$, and $P_3(t_s)$. The expected release rate given those distributions is as follows:

$$\bar{\Sigma}_s(t) = \int_0^\infty P_1(t_c) dt_c \int_0^\infty P_2(t_r) dt_r \int_0^\infty P_3(t_s) dt_s \Sigma_s(t; t_c, t_r, t_s). \quad (12)$$

Let us first consider the distribution of container-failure times. For purposes of demonstration, an exponential distribution will be used. That is, for any given container, the probability of failure between time t_c and time $t_c + dt_c$ is given by

$$Pr(\text{container failure}) = P_1(t_c) dt_c = \tau_c^{-1} e^{-t_c/\tau_c} dt_c, \quad (13)$$

where τ_c is the mean container lifetime. It is certainly of interest to explore the consequences of different failure-time distributions, but this will not be done here. For comparisons with the EPA limits, this distribution is fairly conservative because it allows releases at early times. Also, an exponential distribution is the natural choice if mean lifetime is the only information used to characterize the containers (Tribus, 1969). Note, however, that the exponential distribution will not be conservative under all circumstances because, to balance out the fairly high number of containers that fail early, some containers do not fail until quite late. For comparison with the NRC containment requirements in 10 CFR 60.113, the conservatism is mixed. Since the exponential distribution has a relatively high failure rate at early times, it would be considered conservative for comparison with the requirement that there be substantially complete containment for a period of 300 to 1000 years. However, for comparison with the requirement that the fractional release rate be less than 10^{-5} , a distribution in which all containers fail in a short period of time would be more conservative than the exponential distribution, which has failures spread out over a relatively long time span.

Combining Equations 2, 12, and 13, and assuming for the moment that t_r and t_s are constants, the intermediate result is

$$\Sigma'_s = \begin{cases} I_0(t) t_s^{-1} (1 - e^{-t/\tau_c}) & \text{for } t_r < t < t_s, \\ I_0(t) t_s^{-1} (e^{-(t-t_s)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_s, \end{cases} \quad (14)$$

plus a pulse release at $t = t_r$,

$$\Sigma'_{sp} = I_0(t_r) t_s^{-1} [t_r - \tau_c (1 - e^{-t_r/\tau_c})] \delta(t - t_r). \quad (15)$$

These equations assume that $t_s > t_r$; the formulas for $t_s < t_r$ are slightly different and will not be presented since that case seems very unlikely. (It will also be assumed that $t_s > t_r$ and $t_m > t_r$ for the same reason.)

Next, consider the distribution of resaturation times. The resaturation time will depend on environmental conditions such as how much water flows through the repository and how the water is distributed, and on the heating caused by the waste

containers. Altenhofen and Eslinger (1990) studied the distribution of container temperatures assuming conductive heat flow and including a distribution of burnups for the spent fuel. The resulting distribution of resaturation times (in their Figure 7) is remarkably close to a distribution that is uniform on the interval from 150 yr to 1650 yr. The containers around the edge of the repository resaturate fairly quickly (in fact, in their simulation some containers never got hot enough to drive away all water in the first place) while the containers in the middle of the repository stay hot for a much longer time. Following this result, let us take the probability distribution for t_r to be uniform on the interval from τ_a to τ_b . That is,

$$P_2(t_r) = (\tau_b - \tau_a)^{-1} H(t_r - \tau_a) H(\tau_b - t_r). \quad (16)$$

Note that the letter t is consistently being used to represent the time parameters of the release model while the letter τ is being used for the probability-model parameters.

If Equation 16 is integrated with Equation 14, the next intermediate result is

$$\Sigma_s'' = \begin{cases} 0 & \text{for } t < \tau_a, \\ I_0(t) t_s^{-1} (\tau_b - \tau_a)^{-1} [t + (t - \tau_a - \tau_c)(1 - e^{-t/\tau_c})] & \text{for } \tau_a < t < \tau_b, \\ I_0(t) t_s^{-1} (1 - e^{-t/\tau_c}) & \text{for } \tau_b < t < t_s, \\ I_0(t) t_s^{-1} (e^{-(t-t_s)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_s. \end{cases} \quad (17)$$

This example could be completed to obtain $\bar{\Sigma}_s(t)$ by including a distribution for t_s , but it would be more difficult because of the complicated functional dependence of Equation 17 on t_s . The analysis will not be carried further here. Before going on to releases from the cladding, recall that I_0 should be replaced with $f_m I_0$ for ^{93}Mo . Also, the structural-metal releases for ^{14}C are different from the other species because ^{14}C releases do not depend on t_r . The analog to Equation 17 for ^{14}C is

$$\Sigma_s'' = \begin{cases} f_s I_0(t) t_s^{-1} (1 - e^{-t/\tau_c}) & \text{for } t < t_s, \\ f_s I_0(t) t_s^{-1} (e^{-(t-t_s)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_s. \end{cases} \quad (18)$$

Aqueous releases from the cladding have the same form as Equation 17, with t_s replaced by t_z . Assuming that t_z and f_z are constant, the expected release rate from the cladding is given by

$$\Sigma_c'' = \begin{cases} 0 & \text{for } t < \tau_a, \\ f_z I_0(t) t_z^{-1} (\tau_b - \tau_a)^{-1} [t + (t - \tau_a - \tau_c)(1 - e^{-t/\tau_c})] & \text{for } \tau_a < t < \tau_b, \\ f_z I_0(t) t_z^{-1} (1 - e^{-t/\tau_c}) & \text{for } \tau_b < t < t_z, \\ f_z I_0(t) t_z^{-1} (e^{-(t-t_z)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_z. \end{cases} \quad (19)$$

This equation is assumed to apply only to ^{93}Zr , and f_z is the fraction of the ^{93}Zr inventory located in the cladding (see Equation 3).

Gaseous releases (i.e., ^{14}C) from the cladding are somewhat different from gaseous releases from the structural metals because of the addition of the quick-release ^{14}C on the surface of the cladding. If Equation 4 for releases of ^{14}C from the cladding is integrated with the container-failure distribution, Equation 13, the following expression for the expected release rate of ^{14}C is obtained:

$$\Sigma_c'' = \begin{cases} f_c I_0(t) t_z^{-1} (1 - e^{-t/\tau_c}) + f_q I_0(t) \tau_c^{-1} e^{-t/\tau_c} & \text{for } t < t_z, \\ f_c I_0(t) t_z^{-1} (e^{-(t-t_z)/\tau_c} - e^{-t/\tau_c}) + f_q I_0(t) \tau_c^{-1} e^{-t/\tau_c} & \text{for } t > t_z. \end{cases} \quad (20)$$

There is no effect on Equation 20 from integrating with the resaturation-time distribution, since there is no dependence on t_r .

Next, let us consider releases from the fuel matrix. The formula for the release rate was given in Equation 5, and it depends on t_c , t_f , t_r , t_m , and f_g . We wish to include probability distributions for t_c , t_f , and t_r . The formulas will be more complicated than those for the cladding and structural metals because there are more parameters involved (the cladding and structural-metal releases do not depend on t_f). If Equation 13 is used for the distribution of t_c and the other parameters are regarded as constants, the following is obtained:

$$\Sigma_m' = \begin{cases} (1 - f_g) I_0(t) t_m^{-1} (1 - e^{-(t-t_f)/\tau_c}) & \text{for } t_* < t < t_* + t_m, \\ (1 - f_g) I_0(t) t_m^{-1} (e^{-(t-t_f-t_m)/\tau_c} - e^{-(t-t_f)/\tau_c}) & \text{for } t > t_* + t_m, \end{cases} \quad (21)$$

where $t_* = \max\{t_r, t_f\}$.

If the cladding-failure time t_f is now given an exponential distribution in addition to the exponential distribution for container failure, we arrive at

$$\Sigma_m'' = \begin{cases} (1 - f_g) I_0(t) t_m^{-1} \left[1 - (\tau_c - \tau_f)^{-1} (\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f}) \right] & \text{for } t_r < t < t_r + t_m, \\ (1 - f_g) I_0(t) t_m^{-1} (\tau_c - \tau_f)^{-1} \left[\tau_c (e^{-(t-t_m)/\tau_c} - e^{-t/\tau_c}) - \tau_f (e^{-(t-t_m)/\tau_f} - e^{-t/\tau_f}) \right] & \text{for } t > t_r + t_m. \end{cases} \quad (22)$$

The parameter τ_f is the mean cladding-failure time, analogous to τ_c for the containers.

If a uniform distribution for resaturation time t_r is folded in, as was done previously for structural and cladding releases, the only effect is to add regions of linear interpolation between the cases in Equation 22. The final form of the expected release

rate from the fuel matrix, with t_m and f_g assumed to be constants, is

$$\Sigma_m''' = \begin{cases} 0 & \text{for } t < \tau_a, \\ \left[\begin{aligned} &[(t - \tau_a)/(\tau_b - \tau_a)](1 - f_g)I_0(t)t_m^{-1} \\ &\left[1 - (\tau_c - \tau_f)^{-1}(\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f})\right] \end{aligned} \right] & \text{for } \tau_a < t < \tau_b, \\ (1 - f_g)I_0(t)t_m^{-1} \\ \left[1 - (\tau_c - \tau_f)^{-1}(\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f})\right] & \text{for } \tau_b < t < \tau_a + t_m, \\ \left[\begin{aligned} &[(\tau_b + t_m - t)/(\tau_b - \tau_a)](1 - f_g)I_0(t)t_m^{-1} \\ &\left[1 - (\tau_c - \tau_f)^{-1}(\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f})\right] \\ &+ [(t - \tau_a - t_m)/(\tau_b - \tau_a)] \\ &(1 - f_g)I_0(t)t_m^{-1}(\tau_c - \tau_f)^{-1} \\ &\left[\begin{aligned} &\tau_c(e^{-(t-t_m)/\tau_c} - e^{-t/\tau_c}) \\ &- \tau_f(e^{-(t-t_m)/\tau_f} - e^{-t/\tau_f}) \end{aligned} \right] \end{aligned} \right] & \text{for } \tau_a + t_m < t < \tau_b + t_m, \\ (1 - f_g)I_0(t)t_m^{-1}(\tau_c - \tau_f)^{-1} \\ \left[\begin{aligned} &\tau_c(e^{-(t-t_m)/\tau_c} - e^{-t/\tau_c}) \\ &- \tau_f(e^{-(t-t_m)/\tau_f} - e^{-t/\tau_f}) \end{aligned} \right] & \text{for } t > \tau_b + t_m. \end{cases} \quad (23)$$

As in Equation 5, $(1 - f_g)$ should be set to 1 for those species with no gap/grain-boundary inventory; for ^{14}C , $(1 - f_g)$ should be replaced with $(1 - f_g)(1 - f_s - f_c - f_q)$; for ^{93}Mo , $(1 - f_g)$ should be replaced with $(1 - f_g)(1 - f_m)$; and for ^{93}Zr , $(1 - f_g)$ should be replaced with $(1 - f_z)$.

Lastly, here are the formulas for gap/grain-boundary releases. Equation 6 was the formula for releases with no distribution of parameters. With an exponential container-failure distribution, expected releases are given by

$$\begin{aligned} \Sigma_g' &= f_g I_0(t) \tau_c^{-1} e^{-(t-t_f)/\tau_c} H(t - t_*) \\ &\quad + f_g I_0(t_r) (1 - e^{-(t_r-t_f)/\tau_c}) H(t_r - t_f) \delta(t - t_r), \end{aligned} \quad (24)$$

where $t_* = \max\{t_r, t_f\}$, as before. With exponential cladding-failure distribution included as well, expected releases are given by

$$\begin{aligned} \Sigma_g'' &= f_g I_0(t) (\tau_c - \tau_f)^{-1} (e^{-t/\tau_c} - e^{-t/\tau_f}) H(t - t_r) \\ &\quad + f_g I_0(t_r) \left[1 - (\tau_c - \tau_f)^{-1} (\tau_c e^{-t_r/\tau_c} - \tau_f e^{-t_r/\tau_f})\right] \delta(t - t_r). \end{aligned} \quad (25)$$

And, finally, with a uniform distribution of resaturation times, expected releases are

given by

$$\Sigma_g''' = \begin{cases} 0 & \text{for } t < \tau_a, \\ f_g I_0(t) (\tau_b - \tau_a)^{-1} \left[1 + (\tau_c - \tau_f)^{-1} \right. \\ \quad \left. \left((t - \tau_a - \tau_c) e^{-t/\tau_c} - (t - \tau_a - \tau_f) e^{-t/\tau_f} \right) \right] & \text{for } \tau_a < t < \tau_b, \\ f_g I_0(t) (\tau_c - \tau_f)^{-1} (e^{-t/\tau_c} - e^{-t/\tau_f}) & \text{for } t > \tau_b. \end{cases} \quad (26)$$

As before, f_g should be set to 0 for those species with no gap/grain-boundary inventory; for ^{14}C , f_g should be replaced with $f_g(1 - f_s - f_c - f_q)$; and for ^{93}Mo , f_g should be replaced with $f_g(1 - f_m)$.

5 Release-model summary

For convenience, the final results derived in the previous section will be reproduced here. The primary physical assumptions are discussed in Section 3 and summarized in Figure 4. Probability distributions are included for the three “event” times, the time of container failure t_c , the time of cladding failure t_f , and the time of resaturation t_r . To keep the formulas from being too complicated, simple probability distributions are used: an exponential distribution for t_c , with mean value τ_c ; an exponential distribution for t_f , with mean value τ_f ; and a uniform distribution for t_r , between τ_a and τ_b . The “process” time scales— t_s , the corrosion time for the structural metals; t_z , the corrosion time for the cladding; and t_m , the dissolution time for the fuel matrix—are taken to be constants. The inventory fractions are also taken to be constants. These include f_q , the fraction of the ^{14}C inventory available for quick release upon container failure; f_s , the fraction of the ^{14}C inventory in the structural metals; f_c , the fraction of the ^{14}C inventory in the cladding (not counting the quick-release part, which is in a layer on the cladding surface); f_m , the fraction of the ^{93}Mo inventory in the structural metals; f_z , the fraction of the ^{93}Zr inventory in the cladding; and f_g , the fraction of the inventory in the pellet/cladding gap and the grain boundaries within the fuel pellets. The gap/grain-boundary fraction applies only to certain nuclides, which are marked in Table 1. More complicated distributions could be considered for all these parameters using numerical methods, but not in closed form. These probability distributions represent a number of factors, including natural variability and spatial effects. In a two- or three-dimensional calculation, it may be desirable to explicitly include spatial variability based, for example, on the fact that the repository will be cooler around the edges than in the middle.

The time for radionuclides to get out of the waste container and into the groundwater flow system after dissolution (which might be termed the delay time) has not been included as a parameter in the release model. Neglecting this delay is conservative, since the model then predicts that radionuclides get released sooner than they actually would. If it were desired, it would be fairly easy to generalize the release formulas to include the delay time. It is also possible to include the delay time to some extent by adding it into one of the other times, for example by increasing the container-failure time to include the delay time.

As shown in Table 1 and Figures 2 and 3, the inventory is separated into four main parts: the structural metals, the fuel-rod cladding, the matrix of the fuel pellets, and the pellet/cladding gap plus the grain boundaries within the fuel pellets. There is a release formula for each of these parts. With the above assumptions on the parameter probability distributions, the expected release rate from the structural metals is given by (see Equation 17)

$$\bar{\Sigma}_s = \begin{cases} 0 & \text{for } t < \tau_a, \\ I_0(t)t_s^{-1}(\tau_b - \tau_a)^{-1} \left[t + (t - \tau_a - \tau_c)(1 - e^{-t/\tau_c}) \right] & \text{for } \tau_a < t < \tau_b, \\ I_0(t)t_s^{-1}(1 - e^{-t/\tau_c}) & \text{for } \tau_b < t < t_s, \\ I_0(t)t_s^{-1}(e^{-(t-t_s)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_s. \end{cases} \quad (27)$$

In this equation and the succeeding equations, $I_0(t)$ is the solution to Equation 1; that is, I_0 is the inventory of the given nuclide, including the time variation due to decay but not including decreases to the inventory because of releases. Note that this formula is only valid if $t_s > \tau_b$, which is almost certainly valid. Because only part of the ^{93}Mo inventory is in the structural metals, I_0 should be replaced with $f_m I_0$ for ^{93}Mo . This release formula assumes aqueous releases; for ^{14}C , which is released in gaseous form, expected releases from the structural metals are given by (see Equation 18)

$$\bar{\Sigma}_s = \begin{cases} f_s I_0(t)t_s^{-1}(1 - e^{-t/\tau_c}) & \text{for } t < t_s, \\ f_s I_0(t)t_s^{-1}(e^{-(t-t_s)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_s. \end{cases} \quad (28)$$

As with ^{93}Mo in the previous equation, I_0 is multiplied by f_s to take into account the fact that only part of the ^{14}C inventory is contained in the structural metals.

Aqueous releases from the cladding are given by essentially the same formula as for the structural-metal releases, except for a different time scale (see Equation 19):

$$\bar{\Sigma}_c = \begin{cases} 0 & \text{for } t < \tau_a, \\ f_z I_0(t)t_z^{-1}(\tau_b - \tau_a)^{-1} \left[t + (t - \tau_a - \tau_c)(1 - e^{-t/\tau_c}) \right] & \text{for } \tau_a < t < \tau_b, \\ f_z I_0(t)t_z^{-1}(1 - e^{-t/\tau_c}) & \text{for } \tau_b < t < t_z, \\ f_z I_0(t)t_z^{-1}(e^{-(t-t_z)/\tau_c} - e^{-t/\tau_c}) & \text{for } t > t_z. \end{cases} \quad (29)$$

This equation only applies to ^{93}Zr , and f_z is the fraction of the ^{93}Zr inventory located in the cladding. As with t_s , it is assumed that $t_z > \tau_b$, which should always be valid. The expected ^{14}C release rate from the cladding is given by (see Equation 20)

$$\bar{\Sigma}_c = \begin{cases} f_c I_0(t)t_z^{-1}(1 - e^{-t/\tau_c}) + f_q I_0(t)\tau_c^{-1}e^{-t/\tau_c} & \text{for } t < t_z, \\ f_c I_0(t)t_z^{-1}(e^{-(t-t_z)/\tau_c} - e^{-t/\tau_c}) + f_q I_0(t)\tau_c^{-1}e^{-t/\tau_c} & \text{for } t > t_z. \end{cases} \quad (30)$$

This formula includes releases from the cladding itself (the part containing the factor f_c) and releases from the layer on the surface of the cladding (the part containing the factor f_q).

Expected releases from the fuel matrix are given by (see Equation 23)

$$\bar{\Sigma}_m = \begin{cases} 0 & \text{for } t < \tau_a, \\ \left[\begin{aligned} &[(t - \tau_a)/(\tau_b - \tau_a)](1 - f_g)I_0(t)t_m^{-1} \\ &\left[1 - (\tau_c - \tau_f)^{-1}(\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f})\right] \end{aligned} \right] & \text{for } \tau_a < t < \tau_b, \\ (1 - f_g)I_0(t)t_m^{-1} \\ \left[1 - (\tau_c - \tau_f)^{-1}(\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f})\right] & \text{for } \tau_b < t < \tau_a + t_m, \\ \left[\begin{aligned} &[(\tau_b + t_m - t)/(\tau_b - \tau_a)](1 - f_g)I_0(t)t_m^{-1} \\ &\left[1 - (\tau_c - \tau_f)^{-1}(\tau_c e^{-t/\tau_c} - \tau_f e^{-t/\tau_f})\right] \\ &+ [(t - \tau_a - t_m)/(\tau_b - \tau_a)] \\ &\quad (1 - f_g)I_0(t)t_m^{-1}(\tau_c - \tau_f)^{-1} \\ &\quad \left[\tau_c(e^{-(t-t_m)/\tau_c} - e^{-t/\tau_c}) \right. \\ &\quad \quad \left. - \tau_f(e^{-(t-t_m)/\tau_f} - e^{-t/\tau_f})\right] \end{aligned} \right] & \text{for } \tau_a + t_m < t < \tau_b + t_m, \\ (1 - f_g)I_0(t)t_m^{-1}(\tau_c - \tau_f)^{-1} \\ \left[\begin{aligned} &\tau_c(e^{-(t-t_m)/\tau_c} - e^{-t/\tau_c}) \\ &\quad - \tau_f(e^{-(t-t_m)/\tau_f} - e^{-t/\tau_f}) \end{aligned} \right] & \text{for } t > \tau_b + t_m. \end{cases} \quad (31)$$

This formula requires $t_m > \tau_b - \tau_a$ to be valid. The corresponding formula for $t_m \leq \tau_b - \tau_a$ will not be presented here, but it is included in the computer program in Appendix C. There is no distinction between ^{14}C and the other nuclides for matrix releases, except that $(1 - f_g)$ should be replaced with $(1 - f_g)(1 - f_s - f_c - f_q)$ to accurately reflect the fraction of the ^{14}C inventory contained in the matrix. Also, for ^{93}Mo , $(1 - f_g)$ should be replaced with $(1 - f_g)(1 - f_m)$, and for ^{93}Zr , $(1 - f_g)$ should be replaced with $(1 - f_z)$. For other species with no gap/grain-boundary fraction, $(1 - f_g)$ should be set to 1.

Lastly, the expected release rate from the gap and grain boundaries is given by (see Equation 26)

$$\bar{\Sigma}_g = \begin{cases} 0 & \text{for } t < \tau_a, \\ \left[\begin{aligned} &f_g I_0(t)(\tau_b - \tau_a)^{-1} \left[1 + (\tau_c - \tau_f)^{-1} \right. \\ &\quad \left. \left((t - \tau_a - \tau_c)e^{-t/\tau_c} - (t - \tau_a - \tau_f)e^{-t/\tau_f} \right) \right] \end{aligned} \right] & \text{for } \tau_a < t < \tau_b, \\ f_g I_0(t)(\tau_c - \tau_f)^{-1}(e^{-t/\tau_c} - e^{-t/\tau_f}) & \text{for } t > \tau_b. \end{cases} \quad (32)$$

Once again, ^{14}C and ^{93}Mo need special consideration. For ^{14}C , f_g should be replaced with $f_g(1 - f_s - f_c - f_q)$; and for ^{93}Mo , f_g should be replaced with $f_g(1 - f_m)$.

The preceding release formulas are complicated, but still are straightforward to program for a computer simulation. The more serious problem is that in order to cover all the important processes, even in a simple manner, requires quite a few

poorly known parameters. The source-release model outlined here has seven time parameters and six inventory-fraction parameters. The majority of the species are contained entirely in the fuel matrix and require only τ_c , τ_f , τ_a , τ_b , and t_m to define their releases (or t_c , t_f , t_r , and t_m if the probability distributions are not used). This is really too many parameters to vary exhaustively. The species that depends on the most parameters is ^{14}C , since it is represented in several of the release formulas above; but the most critical part of its release is the quick release from the cladding surface, and that only depends on f_q and τ_c (or t_c).

There is an additional complication for low-solubility species. As discussed in Section 3, if the solubility of a species is low enough, the preceding formulas will overestimate its releases. The formulas could be used anyway, since it is conservative to have higher releases, but if more realistic release rates are desired for the low-solubility elements a solubility correction must be applied. Details are not given here because there is no simple mathematical formula for the correction, but in a computer program the correction can be made by putting in a check of concentrations against solubility limits. A compromise would be to use more than one value for t_m —perhaps a low one for the more soluble, rapidly released, species and a higher one for the less soluble, slowly released species.

6 Parameter values

In this section, the model parameters introduced above, and what might be reasonable values to use for them, will be discussed.

Let us begin with the mean container time-to-failure, τ_c . Oversby and McCright (1985) considered the probability of failure within 10,000 yr to be small, and gave an estimate of 67,000 yr for the failure time, assuming uniform corrosion. When other processes are taken into account, including the effects of disruptive scenarios, the container failure time should be smaller, but how much smaller is not known. McGuire *et al.* (1990) calculated a probability distribution for the frequency of container failures due to seismic effects. Their mean frequency was 4×10^{-4} container failures per year. This is only one container failure every 2500 years, or a mean container lifetime of almost 10^8 yr. Apparently seismic effects are not a large contributor to the probability of container failure. McGuire *et al.* also calculated thermal stresses on the container borehole walls and found a significant probability (38% in 10,000 yr) for “slabbing” of the walls. The breakdown of the borehole wall may occasionally cause container failure; the probability of container failure due to this effect was not calculated, but McGuire *et al.* assumed it to be 1% in their calculations.

NRC regulations (10 CFR 60.113) require “substantially complete” containment of radioactivity for 300 to 1000 years, assuming undisturbed conditions. Thus, a conservative choice for τ_c might be a value of 300 years. Note, however, that the exponential distribution has about 63% of its probability below the mean, so choosing $\tau_c = 300$ yr does not provide containment for 300 years, but instead implies that most containers fail within 300 years. The exponential distribution of container failures is not really consistent with complete containment at early times, but for that reason it

is a conservative distribution, allowing early failures. It would be easy to generalize the release formulas derived earlier to allow for a period of no container failures and an exponential distribution of failures afterward. Such a generalization would increase the complexity of the formulas slightly and add an additional time parameter. Tables 8.3.5.9-1 and 8.3.5.10-2 in the SCP (DOE, 1988) list several design goals for container development. There are criteria based on whether containers are wet or dry and how many are expected to be wet and dry but, all taken together, the goal is for less than 0.012% per year of the containers to fail up to $t = 100$ yr, less than 0.05% per year for $100 \text{ yr} < t < 300 \text{ yr}$, and less than 0.1% per year for $300 \text{ yr} < t < 1000 \text{ yr}$. There are goals for $t > 1000 \text{ yr}$ also, but the limits already stated imply a goal of less than 81% container failure in the first 1000 yr. An exponential distribution of container failures with mean failure time $\tau_c > 600 \text{ yr}$ would be consistent with that number, but $\tau_c > 8000 \text{ yr}$ would be necessary to be consistent with the goal for the first 100 yr.

The situation is similar for the mean cladding time-to-failure, τ_f . Rothman (1984) concluded that cladding failure within 10,000 yr is unlikely, but there are not good estimates for mean lifetime taking everything into account. The failure time assuming uniform corrosion is even longer than the above estimate of 67,000 yr for the containers. Most performance-assessment calculations that have been made to date have ignored the cladding, thereby assuming that $\tau_f = 0$. There are design goals for the cladding in SCP Table 8.3.5.9-3. For a time of 1000 yr, the goal is for less than 50% of the claddings to have failed in the containers that have failed. If exponential distributions are used for container-failure time and cladding-failure time, then this criterion is met if $\tau_c > 600 \text{ yr}$ and $\tau_f > 800 \text{ yr}$. As before, though, the goals for earlier times are not met with these distributions.

It was already noted, in the discussion of the probability distribution for resaturation time, that Altenhofen and Eslinger (1990) simulated repository thermal conditions for an ensemble of containers, and their resulting distribution of resaturation times is approximately uniform on the interval from 150 yr to 1650 yr. That is, $\tau_a = 150 \text{ yr}$ and $\tau_b = 1650 \text{ yr}$. Their simulation included only a portion of the expected variability in repository conditions—in particular, effects due to the presence of water, such as convective heat transport, were neglected—but this range seems like a reasonable choice until further results are available. Simulations including convective “heat pipes” would be expected to show somewhat faster cooling, since the rate of heat transport away from the containers is increased. For example, Nitao (1988) found water returning after about 1000 yr with a percolation rate of 0.5 mm/yr or 1 mm/yr, and after about 1400 yr with a percolation rate of 0.1 mm/yr. Nitao’s simulations assumed an infinite lattice of waste containers, so these results may represent conditions in the middle of the repository but not at the edges. The most conservative choice would be to ignore the epoch of desaturation entirely, i.e., $\tau_a = 0$, $\tau_b = 0$. The goals in SCP Table 8.3.5.9-1 dealing with resaturation say that there should be no liquid water contacting 90% of the containers at 1000 yr. The uniform distribution above has a cumulative probability of 57% resaturation by 1000 yr. However, this number is not really comparable to the 10% goal in the SCP because it could be that after all the containers are “resaturated,” only 10% of them are actually in a wet location.

The time t_s , the amount of time for the structural metal parts to completely disappear, can be obtained from estimates of the corrosion rate of stainless steel. This rate has been measured for the materials being considered for the waste containers, and presumably the rate for the structural metals will be similar. Oversby and McCright (1985) reported a corrosion rate of $0.15 \pm 0.036 \mu\text{m/yr}$ in J-13 water, with little temperature dependence. They also reported a corrosion rate of $0.16 \pm 0.03 \mu\text{m/yr}$ in 100°C saturated steam and values ranging from $0.001 \mu\text{m/yr}$ to $0.008 \mu\text{m/yr}$ in 150°C unsaturated steam. Just for the sake of estimation, suppose that the average thickness of the steel parts is 1 cm; then $0.16 \mu\text{m/yr}$ translates to a lifetime of about 60,000 yr. At the lowest rate of $0.001 \mu\text{m/yr}$, the lifetime would be 1×10^7 yr. In an actual repository, there will probably be a wide variety of conditions, with some containers wet and others completely dry. If conservatism is desired, a low value should be used.

The time for the Zircaloy cladding to corrode away, t_z , can be estimated from calculations by Rothman (1984) of the rate of oxidation of Zircaloy under repository thermal conditions. For a container breach at a time of 300 yr, he calculated an oxidation depth of $0.03 \mu\text{m}$ after 10,000 yr. For container breach at 1000 yr, the oxidation depth at 10,000 yr was calculated to be only $0.006 \mu\text{m}$. Apparently, in the first calculation there was $0.024 \mu\text{m}$ of oxidation between 300 yr and 1000 yr and then an additional $0.006 \mu\text{m}$ between 1000 yr and 10,000 yr. The oxidation rate is quite temperature dependent, so the oxidation rate steadily decreases. There would be a higher oxidation rate in the first 300 yr, when temperatures are higher, but presumably container failures that early would be rare, and anyway the amount of oxidation would still be small compared to the cladding thickness. The cladding is about $600 \mu\text{m}$ thick so, assuming $0.006 \mu\text{m}$ of oxidation in 9000 yr, we could estimate 9×10^8 yr for the cladding to be completely oxidized. Unless it is less than 10,000 yr, which appears to be highly improbable, t_z will not be at all important to calculations of EPA ratios. None of the processes considered by Rothman (1984) were found to increase the Zircaloy oxidation rate substantially. A value of 9×10^8 yr is effectively infinite because it would mean a cladding release of radioactivity in 10,000 yr that is much smaller than the EPA limit. Note also that Zircaloy cladding is almost pure zirconium, and zirconium solubility is extremely low (see Table 2). If t_z were based on dissolution rather than corrosion, the estimate would be $t_z > 10^{11}$ yr. Once again, for conservatism a lower value could be used.

The very long cladding corrosion time may mean that during all times of interest in this study the cladding will be mostly intact, helping to slow down releases from the fuel matrix. However, it could be that swelling of the UO_2 pellets will cause the cladding to split in which case it would not be an effective barrier. The swelling is caused by oxidation of UO_2 to U_3O_8 , which has a lower density. Oxidation of UO_2 to U_3O_8 (and further to UO_3) is favored, energetically, but it proceeds slowly at the expected temperatures. Oxidation of spent fuel has been studied by Einziger and Woodley (1985), and they concluded that the rate of oxidation after container breach is low enough that cladding splitting is not expected to occur within 10,000 yr.

In Section 3, several release mechanisms were discussed for the fuel matrix. Let us first look at the case in which the matrix-release time scale is determined by uranium

dissolution, as described by Equation 7. The water-volume rate can be expressed as $\dot{V}_w = qA_e$, where q is the water volume flux and A_e is an effective water-collection area. With this substitution, Equation 7 can be rewritten as

$$t_m = M_U / qA_e C_U. \quad (33)$$

M_U is the number of moles of uranium initially in the fuel matrix and C_U is the effective release concentration of uranium (in moles per unit volume).

The effective water-collection area depends on what the water flow in the repository is like—it could be the geometrical cross-section area of the containers, or it could be more if water is being funneled into the containers, or it could be less if water tends to avoid the containers. The maximum value would be the total repository area A_{rep} , if all water flowing through the repository comes into contact with the waste, and the minimum value would be zero, if all the water flows around the mined areas without entering. From Equation 10 it can be seen that, for diffusive release, A_e is a function of q , large for small q and approaching the geometrical area of the containers for large q .

The maximum value of C_U would be the solubility limit for the fuel matrix, which might be reached in a “bathtub” situation, in which water collects in a container and leaks out very slowly, so that the waste material sits in the water for a long time. The solubility limit for uranium is rather uncertain. On the basis of theoretical calculations, Kerrisk (1984) lists 0.2 mol/m³ and Kerrisk (1985) lists 4 mol/m³. The experimental values in Wilson (1987) are lower, ranging from 0.004 mol/m³ to 0.02 mol/m³; the lower value may be the solubility limit for UO₂, while the higher value is for one of the higher oxidation states mentioned previously, present only as a thin layer in the experimental situation.

If, instead, the concentration were limited by the dissolution rate and residence time, the effective concentration would be given by

$$C_U = R_l t_d A_d / V_d, \quad (34)$$

where R_l is the uranium leach rate, t_d is the water contact time, A_d is the contact area, and V_d is the volume of water involved. Braithwaite (1985) considered a situation where the fuel matrix is not immersed in water at all, but rather water droplets run down the surface of the fuel and so are in contact with the fuel for only a short time. In this case, V_d represents the volume of a water droplet, A_d is the area of contact between a water droplet and the spent fuel, and t_d is the time the droplet takes to run down. Braithwaite estimated the combination $t_d A_d / V_d$ to be approximately 2×10^{-4} yr/m for vertical container emplacement (he assumed that the cladding was not acting as a barrier to water, but that there was still enough of it present to hold the fuel pellets in a vertical arrangement). Katayama *et al.* (1980) reported uranium leach rates in the range of 3×10^{-3} mol/m²-yr to 2 mol/m²-yr. The higher value is the initial leach rate and the lower value is the (approximately steady-state) rate after a time of order one year. The initial leach rate inferred from the data of Wilson (1987) is comparable to the initial leach rate of Katayama *et al.* Wilson and Gray (1990) gave a leach rate under steady flow conditions in an NaHCO₃ solution as

2×10^{-2} mol/m²-yr, with lower rates in other solutions, including deionized water and J-13 well water. These leach-rate numbers, combined with Braithwaite's estimate of the other parameters, give C_U in the range of 4×10^{-7} mol/m³ to 3×10^{-4} mol/m³.

The water percolation rate at the repository horizon could be as low as zero, but the upper limit is not really known. Montazer and Wilson (1984) summarized several attempts at estimating the percolation rate. At the high end, the average recharge below Yucca Mountain was estimated to be between 0.5 mm/yr and 4.5 mm/yr. At the low end, some calculations of thermal flow suggested that the net water flow within Yucca Mountain may be upward—as water vapor. In this case, there would be essentially no flow of liquid water. Capillary-barrier studies¹ suggest that the percolation rate in the Topopah Spring unit may be no higher than its saturated matrix conductivity, about 0.6 mm/yr, because of lateral diversion at the interface between Topopah Spring and the Paintbrush unit.

Putting together the estimates from the preceding paragraphs gives a low value for t_m of about 2×10^6 yr, using 2.8×10^8 mol for M_U , 1 mm/yr for q , 5.6×10^6 m² for $A_c = A_{rep}$, and 0.02 mol/m³ for C_U (this is the high value from the Wilson experiments). This value is not really an absolute minimum, since the water percolation rate could conceivably be higher than 1 mm/yr and the solubility limit could turn out to be higher under repository conditions. On the other hand, the effective area is almost certainly much smaller than the repository area. The upper bound for t_m is infinity. Practically speaking, a value of $t_m = 1 \times 10^9$ yr is infinite because such a slow rate would result in releases from the containers below the EPA limit. It is easy to get very high values for t_m with values of q , A_c , and C_U that are not unreasonable. For example, consider $M_U = 2.8 \times 10^8$ mol (same as before), $q = 0.1$ mm/yr (one tenth of the previous value), $A_c = 5.6 \times 10^4$ m² (one percent of the repository area), and $C_U = 3 \times 10^{-4}$ mol/m³ (the value from Braithwaite, using the initial leach rate). These values imply a t_m of 2×10^{11} yr. If conservatism is desired, a low value should be used, perhaps even lower than 2×10^6 yr.

As discussed in Section 3, there is some evidence that supports a matrix-release time scale based on chemical alteration rather than dissolution. The alteration time scale may be as low as 1000 yr (Apted *et al.*, 1991), which would be much faster than uranium dissolution. Considerations such as the amount of water contacting the waste and the amount of wetted surface area would presumably increase the average t_m , but releases from containers that become inundated with water may proceed with the very low value of $t_m = 1000$ yr for those containers. As noted previously, it is only the very soluble species that could be released with this low alteration time scale. Most species would be solubility-limited, and would have release times better represented by the uranium dissolution time discussed in the previous paragraphs.

The SCP goals related to source releases are oriented toward compliance with the NRC release-rate limits in 10 CFR 60.113. For times greater than 1000 yr, the only requirement is for annual releases to be less than one part in 10^5 of the 1000-yr inventory (see Appendix B, Tables B.1 and B.2, in this report). Within the present

¹B. Ross, "Quasi-Linear Analysis of Water Flow in the Unsaturated Zone at Yucca Mountain, Nevada, USA," presented at the International Conference on Water Resources in Mountainous Regions, Lausanne, Switzerland, August 27 to September 1, 1990.

model, for the fractional release rate to be below 10^{-5} after all containers and fuel rods are breached, the *average* release time scales t_s , t_c , and t_m need to be greater than 100,000 yr. The goal stated in the SCP (Table 8.3.5.10-2) is for only 10% of the waste containers to be exposed to water, and for that 10% to have fractional releases less than 10^{-4} /yr. The 10% coupled with the 10^{-4} gives the required 10^{-5} . Put into the terms used in this report, the goal can be stated as follows: For 10% of the containers, t_s , t_c , and t_m should be greater than 10,000 yr; for the other 90% of the containers, t_s , t_c , and t_m are effectively infinite since there is no water to carry off the waste. There are several subsidiary goals stated in the SCP, dealing with the amount of water per year contacting the waste form (qA_e/M_L in Equation 34), the solubilities of the primary actinides, etc., but we will not go into those here.

In addition to the time parameters, we also need values for the fraction parameters that tell how the various nuclides are distributed in the different locations within the spent fuel and its associated cladding and structural metals. Unfortunately, Roddy *et al.* (1986) did not break the inventories down by location. There is some breakdown in Croff and Alexander (1980). They list spent-fuel totals and also amounts for the so-called "cladding waste," which includes the cladding and the structural metals. For f_z , the fraction of ^{93}Zr contained in the cladding, Croff and Alexander show about 7% for PWR fuel with 33,000 MWd/MTHM burnup and about 17% for BWR fuel with 27,500 MWd/MTHM burnup. For f_m , the fraction of ^{93}Mo contained in the structural metals, the numbers are 97% for PWR and 91% for BWR. These nuclides are not among the most important nuclides (in fact, ^{93}Mo is the very lowest on the list in Figure 1), so for most purposes it probably would suffice to neglect the distribution between different locations and treat them as though all the ^{93}Zr were in the matrix and all the ^{93}Mo were in the structural metals. That is, f_z could be taken to be 0 and f_m could be taken to be 1, with little effect on the overall results.

The value of the gap/grain-boundary fraction f_g is important because it represents rapid releases. Most of the fission products are listed as having "gap" fractions in Table 1. As mentioned previously, it seems reasonable to use the same fraction f_g for all of these nuclides, but the release formulas derived previously do not depend on this assumption. Each of the gap/grain-boundary nuclides could be given its own value of f_g and the only complication would be the introduction of several additional parameters to the model. The main reason for not doing this is the sparsity of data. Estimates of f_g are quite variable. Oversby and Wilson (1986) suggested a value of 0.5%. Apted *et al.* (1989) used a value of 2%. Garisto *et al.* (1989) used a distribution of $f_g = 8.1\% \pm 0.6\%$, but that was for CANDU (Canada deuterium uranium) spent fuel, which has a higher gap fraction because CANDU reactors run at higher temperatures than either PWRs or BWRs. If a conservative estimate is desired, a value from the higher end of the range should be used. The SCP goal, from Table 8.3.5.9-3, is for f_g to be less than 2%.

Lastly, there are the three inventory fractions for ^{14}C : f_q , f_c , and f_s . It is convenient also to have a symbol for the matrix fraction; it will be denoted by f_x , and is equal to $(1 - f_g)(1 - f_s - f_c - f_q)$. The fraction of ^{14}C actually in the gap/grain-boundaries will be denoted by $f'_g = f_g(1 - f_s - f_c - f_q)$. With these notations, $f_s + f_c + f_q + f_x + f'_g = 1$. The ^{14}C inventory is more variable than for most other

nuclides because ^{14}C primarily results from activation of ^{14}N and ^{17}O present as impurities, and the impurity level may vary significantly from one fuel assembly to another. This means that the inventory values for ^{14}C in Appendix B and Table 1 may have significant uncertainty, whereas the other nuclides are probably reasonably well characterized by the computer calculations on which the tables are based. For that reason, for some calculations it might be desirable to add another variable parameter to represent the initial ^{14}C inventory, while treating the other initial inventories as fixed.

Park and Pflum (1990) listed the following numbers for the breakdown of ^{14}C between different locations: For PWR spent fuel with burnup of 33,000 MWd/MTHM, a total ^{14}C inventory of 1.55 Ci/MTHM, with 0.6 Ci/MTHM in the matrix, 0.35 Ci/MTHM in the cladding, and 0.6 Ci/MTHM in the structural metals. This means that $f_x + f'_g = 0.6/1.55 = 39\%$, $f_c + f_q = 0.35/1.55 = 22\%$, and $f_s = 0.6/1.55 = 39\%$. For BWR spent fuel with burnup of 27,500 MWd/MTHM, a total ^{14}C inventory of 1.53 Ci/MTHM, with 0.54 Ci/MTHM in the matrix, 0.76 Ci/MTHM in the cladding, and 0.23 Ci/MTHM in the structural metals. Thus, $f_x + f'_g = 0.54/1.53 = 35\%$, $f_c + f_q = 0.76/1.53 = 50\%$, and $f_s = 0.23/1.53 = 15\%$.

Van Konynenburg *et al.* (1987) tabulated a number of measured ^{14}C inventories, with some breakdown into the different locations. The numbers for the matrix inventory varied from 0.081 Ci/MTHM to 0.56 Ci/MTHM; the low values are from spent fuel with a low burnup of only 11,000 MWd/MTHM, as opposed to 33,000 MWd/MTHM for the reference inventory in Table B.1. The measured values were all for PWR fuel, and would include any gap/grain-boundary fraction. The high value is close to the calculated value above. The Van Konynenburg *et al.* table shows values for the cladding inventory for ^{14}C from 0.11 Ci/MTHM to 0.14 Ci/MTHM; this amount includes the quick-release portion on the cladding surface. A value of 0.14 would mean that $f_c + f_q = 0.14/1.55 = 9\%$. It may not be correct, though, to use 1.55 Ci/MTHM when taking the ratio; the actual experimental value for the total inventory is not known because the amount in the structural metals was not measured. The highest value listed for the cladding inventory, 0.14 Ci/MTHM, is still well below the theoretical estimates above.

The most important question concerning the ^{14}C inventory is what portion of it is released quickly upon container failure; that is, what is f_q ? Van Konynenburg *et al.* (1985) reported an experimental release of 0.26% of the ^{14}C inventory at a temperature of 275° C. Smith and Baldwin (1989) measured releases three times higher at the same temperature with a different experimental setup, and higher releases at higher temperatures. Both of these experiments were conducted with PWR spent fuel. According to Park and Pflum (1990), the quick-release fraction of the ^{14}C inventory could be as high as 5%. For conservatism, higher values of f_q should be used. The SCP, in Tables 8.3.5.9-3 and 8.3.5.10-3c, shows goals of $f_q < 1\%$ for $t < 1000$ yr and $f_q < 0.1\%$ for $t > 1000$ yr. The latter goal was based on the expectation that the quick-release $^{14}\text{CO}_2$ is formed by oxidation after exposure to air, and the oxidation rate is much slower at lower temperatures. However, according to Park and Pflum there is more recent evidence suggesting that the $^{14}\text{CO}_2$ can form in the absence of air and accumulate inside a sealed container, ready to be released when the container

is breached.

The limits on f_g for ^{14}C are presumably similar to the generic limits discussed previously. However, since f_g and f_q both represent relatively fast releases, it might make sense for many calculations to simply lump them together. That is, increase f_q by the amount f'_g (not f_g) and then use $f_g = 0$ in the release formulas for ^{14}C . This substitution would be conservative because the quick-release fraction is released faster than the gap/grain-boundary fraction.

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A Notation

a	Molar activity of a nuclide. Also used as the lower bound of an arbitrary interval of the variable x .
A_d	Contact area for a water droplet.
A_e	Effective water-collection area.
A_{rep}	Area of the potential repository.
b	Upper bound of an arbitrary interval of the variable x .
C_i	Concentration of nuclide i in water leaving a container.
C_U	Effective release concentration of uranium.
D_e	Effective diffusion coefficient.
e	The number e (base of the natural logarithm).
f_c	Fraction of the ^{14}C inventory in the cladding (not counting the quick-release fraction).
f_g	Fraction of the inventory of several nuclides in the pellet/cladding gap and the grain boundaries.
f'_g	Fraction of the ^{14}C inventory in the gap/grain-boundaries.
f_m	Fraction of the ^{93}Mo inventory in the structural metals.
f_s	Fraction of the ^{14}C inventory in the structural metals.
f_q	Quick-release fraction of the ^{14}C inventory.
f_x	Fraction of the ^{14}C inventory in the fuel matrix.
f_z	Fraction of the ^{93}Zr inventory in the cladding.
H	Heaviside step function (zero when the argument is less than zero, one when the argument is greater than or equal to zero).
I_0^i or I_0	Total inventory of nuclide i (including the portion of the inventory that has already been released).
L	Length of a waste container.
M_i	Number of moles of nuclide i in the fuel matrix.
M_U	Initial number of moles of uranium in the fuel matrix.
q	Volume flux of water (volume of water flowing through a unit area in a unit time).
P	An arbitrary probability density function.
P_1	A probability density function for t_c .
P_2	A probability density function for t_r .
P_3	A probability density function for t_s .
R	Radius of a waste container.
R_l	Uranium leach rate.
S	Solubility limit of an element.
t	Time variable. Normally measured from repository closure.
t_a	Alteration time of the spent-fuel matrix.
t_c	Container-failure time: amount of time it takes a container to fail.
t_d	Dissolution time of spent-fuel matrix. Also, contact time for a water droplet.
t_f	Cladding-failure time: amount of time it takes a fuel rod to fail after the

	container has failed (if cladding fails before container, $t_f = 0$).
t_m	Amount of time it takes for releases from the spent-fuel matrix after the container has failed and after resaturation.
t_r	Resaturation time: end of the thermal period, when temperature has decreased enough for liquid water to be present around the containers.
t_s	Amount of time it takes for releases from structural metals after the container has failed.
t_x	The greater of t_r and $t_c + t_f$ (the time at which matrix releases begin).
t_z	Amount of time it takes for releases from the cladding after the container has failed.
t_*	The greater of t_r and t_f (time at which matrix releases may begin if $t_c = 0$).
V_d	Volume of a water droplet.
$\dot{V}_w$	Volume of water per unit time that comes in contact with waste.
x	An arbitrary variable.
x'	Another arbitrary variable.
δ	Dirac delta function.
θ	Volumetric moisture content (fraction of rock volume filled with water).
λ^i or λ	Radioactive decay rate ($\ln 2/\tau_{1/2}$).
π	The number π (ratio of circumference to diameter of a circle).
Σ_c	Release rate of radioactivity from the cladding.
$\bar{\Sigma}_c$	Expected release rate of radioactivity from the cladding.
Σ'_c	Release rate of radioactivity from the cladding, averaged over t_c and t_r .
Σ_g	Release rate of radioactivity from the pellet/cladding gap and the grain boundaries.
$\bar{\Sigma}_g$	Expected release rate of radioactivity from the gap/grain-boundaries.
Σ'_g	Release rate of radioactivity from the gap/grain-boundaries, averaged over t_c .
Σ''_g	Release rate of radioactivity from the gap/grain-boundaries, averaged over t_c and t_f .
Σ'''_g	Release rate of radioactivity from the gap/grain-boundaries, averaged over t_c , t_f , and t_r .
Σ_m	Release rate of radioactivity from the fuel matrix.
$\bar{\Sigma}_m$	Expected release rate of radioactivity from the fuel matrix.
Σ'_m	Release rate of radioactivity from the fuel matrix, averaged over t_c .
Σ''_m	Release rate of radioactivity from the fuel matrix, averaged over t_c and t_f .
Σ'''_m	Release rate of radioactivity from the fuel matrix, averaged over t_c , t_f , and t_r .
Σ_s	Release rate of radioactivity from the structural metals.
$\bar{\Sigma}_s$	Expected release rate of radioactivity from the structural metals.
Σ'_s	Release rate of radioactivity from the structural metals, averaged over t_c .
Σ'_{sp}	Pulse component of release rate of radioactivity from the structural metals averaged over t_c .
Σ''_s	Release rate of radioactivity from the structural metals, averaged over t_c and t_r .
τ_a	Lower limit of the probability distribution for t_r .

τ_b	Upper limit of the probability distribution for t_r .
τ_c	Mean container lifetime.
τ_f	Mean cladding lifetime.
$\tau_{1/2}$	Radioactive half-life.
∞	Infinity.

B Sample PWR and BWR spent-fuel inventories

This appendix lists two sample radionuclide inventories, Table B.1 for PWR spent fuel with burnup of 33,000 MWd/MTHM and Table B.2 for BWR spent fuel with burnup of 27,500 MWd/MTHM.

The figures for initial inventory were taken from the Nuclear Waste Terminal Storage Program (NWTSP) database (Roddy *et al.*, 1986). The inventories shown are for spent fuel ten years out of the reactor.

The half-lives were taken from the ORIGEN2 decay database (DOE, 1987). The tables include all 83 nuclides from that database with half-lives greater than 20 years.

Molar activity is the amount of radioactivity per mole, given by $3.575 \times 10^5 / \tau_{1/2}$, where $\tau_{1/2}$ is the half-life, in years, and radioactivity is measured in curies.

The NRC limits are based on 10 CFR 60.113(a)(1). The release-rate limits in that regulation are based on the inventory 1000 years after repository closure. The numbers given were calculated assuming that all waste is emplaced at once and the repository is closed immediately, so the inventory 1010 years out of the reactor was used. The NWTSP database gives the inventory at age 1000 yr, and the only change necessary to go to an age of 1010 yr is to reduce the ^{241}Am inventory slightly. The NRC release limit is then the greater of 10^{-5} of the inventory of the given species or 10^{-8} of the total inventory. The latter limit applies to all but eight of the nuclides listed in Table B.1 and to all but seven of the nuclides listed in Table B.2.

The EPA limits were taken from Table 1 in Appendix A of 40 CFR Part 191, and pertain to cumulative releases to the accessible environment over a period of 10,000 years after emplacement in the repository.

Table B.1: Sample PWR spent-fuel inventory.

Species	Half-life (yr)	PWR 33,000			
		Molar activity (Ci/mol)	Inventory (Ci/MTHM)	NRC limit (Ci/MTHM-yr)	EPA limit (Ci/MTHM)
¹⁰ Be	1.600×10^6	2.23×10^{-1}	4.69×10^{-6}	1.73×10^{-5}	1.
¹⁴ C	5.729×10^3	6.24×10^1	1.55	1.73×10^{-5}	0.1
³² Si	6.500×10^2	5.50×10^2	2.52×10^{-7}	1.73×10^{-5}	1.
³⁶ Cl	3.010×10^5	1.19	1.22×10^{-2}	1.73×10^{-5}	1.
³⁹ Ar	2.690×10^2	1.33×10^3	7.10×10^{-5}	1.73×10^{-5}	1.
⁴² Ar	3.300×10^1	1.08×10^4	4.24×10^{-13}	1.73×10^{-5}	1.
⁴⁰ K	1.280×10^9	2.79×10^{-4}	4.60×10^{-9}	1.73×10^{-5}	1.
⁴¹ Ca	8.100×10^4	4.41	1.93×10^{-4}	1.73×10^{-5}	1.
⁵⁰ V	4.000×10^{16}	8.94×10^{-12}	9.36×10^{-15}	1.73×10^{-5}	1.
⁵⁹ Ni	8.000×10^4	4.47	5.15	5.11×10^{-5}	1.
⁶³ Ni	9.200×10^1	3.89×10^3	6.52×10^2	1.73×10^{-5}	1.
⁷⁹ Se	6.496×10^4	5.50	4.09×10^{-1}	1.73×10^{-5}	1.
⁸¹ Kr	2.099×10^5	1.70	4.68×10^{-7}	1.73×10^{-5}	1.
⁸⁷ Rb	4.696×10^{10}	7.61×10^{-6}	2.13×10^{-5}	1.73×10^{-5}	1.
⁹⁰ Sr	2.912×10^1	1.23×10^4	5.72×10^4	1.73×10^{-5}	1.
⁹³ Zr	1.530×10^6	2.34×10^{-1}	1.93	1.93×10^{-5}	1.
⁹¹ Nb	1.000×10^4	3.58×10^1		1.73×10^{-5}	1.
⁹⁴ Nb	2.030×10^1	1.76×10^1	1.28	1.73×10^{-5}	1.
⁹³ Mo	3.498×10^3	1.02×10^2	2.59×10^{-2}	1.73×10^{-5}	1.
⁹⁷ Tc	2.600×10^6	1.38×10^{-1}	---	1.73×10^{-5}	1.
⁹⁸ Tc	4.199×10^6	8.51×10^{-2}	5.46×10^{-6}	1.73×10^{-5}	1.
⁹⁹ Tc	2.130×10^5	1.68	1.31×10^1	1.30×10^{-4}	10.
¹⁰⁷ Pd	6.496×10^6	5.50×10^{-2}	1.12×10^{-1}	1.73×10^{-5}	1.
¹⁰⁸ Ag m	1.270×10^2	2.81×10^3	1.25×10^{-2}	1.73×10^{-5}	1.
¹¹⁵ In	4.997×10^{14}	7.15×10^{-10}	1.51×10^{-11}	1.73×10^{-5}	1.
¹²¹ Sn m	4.997×10^1	7.15×10^3	6.83×10^{-1}	1.73×10^{-5}	1.
¹²⁶ Sn	1.000×10^5	3.58	7.76×10^{-1}	1.73×10^{-5}	1.
¹²³ Te	1.000×10^{13}	3.58×10^{-8}	2.63×10^{-12}	1.73×10^{-5}	1.
¹²⁹ I	1.570×10^7	2.28×10^{-2}	3.15×10^{-2}	1.73×10^{-5}	0.1
¹³⁵ Cs	2.300×10^6	1.55×10^{-1}	3.45×10^{-1}	1.73×10^{-5}	1.
¹³⁷ Cs	3.000×10^1	1.19×10^4	8.21×10^4	1.73×10^{-5}	1.
¹³⁷ La	5.999×10^4	5.96	---	1.73×10^{-5}	1.
¹³⁸ La	1.350×10^{11}	2.65×10^{-6}	1.08×10^{-10}	1.73×10^{-5}	1.
¹⁴² Ce	1.049×10^{11}	3.41×10^{-6}	2.70×10^{-5}	1.73×10^{-5}	1.
¹⁴⁴ Nd	2.099×10^{15}	1.70×10^{-10}	1.56×10^{-9}	1.73×10^{-5}	0.1
¹⁴⁶ Sm	7.000×10^7	5.11×10^{-3}	2.71×10^{-7}	1.73×10^{-5}	0.1
¹⁴⁷ Sm	1.070×10^{11}	3.34×10^{-6}	4.51×10^{-6}	1.73×10^{-5}	0.1
¹⁴⁸ Sm	8.001×10^{15}	4.47×10^{-11}	5.01×10^{-11}	1.73×10^{-5}	0.1
¹⁴⁹ Sm	9.994×10^{15}	3.58×10^{-11}	8.88×10^{-13}	1.73×10^{-5}	0.1
¹⁵¹ Sm	8.999×10^1	3.97×10^3	3.31×10^2	1.73×10^{-5}	1.

Table B.1 continued.

Species	Half-life (yr)	Molar activity (Ci/mol)	PWR 33,000	NRC limit (Ci/MTHM-yr)	EPA limit (Ci/MTHM)
			Inventory (Ci/MTHM)		
¹⁵⁰ Eu	3.600×10^1	9.93×10^3	1.31×10^{-5}	1.73×10^{-5}	1.
¹⁵² Gd	1.080×10^{14}	3.31×10^{-9}	3.60×10^{-13}	1.73×10^{-5}	0.1
¹⁵⁷ Tb	1.500×10^2	2.38×10^3	---	1.73×10^{-5}	1.
¹⁶³ Ho	3.300×10^1	1.08×10^4	---	1.73×10^{-5}	1.
¹⁶⁶ Ho m	1.200×10^3	2.98×10^2	2.53×10^{-3}	1.73×10^{-5}	1.
¹⁷⁶ Lu	3.000×10^{10}	1.19×10^{-5}	2.64×10^{-11}	1.73×10^{-5}	1.
¹⁸² Hf	9.000×10^6	3.97×10^{-2}	4.03×10^{-7}	1.73×10^{-5}	1.
¹⁸⁰ Ta	1.600×10^{13}	2.23×10^{-8}	---	1.73×10^{-5}	1.
¹⁸⁷ Re	5.000×10^{10}	7.15×10^{-6}	1.98×10^{-8}	1.73×10^{-5}	1.
¹⁹² Ir m	2.410×10^2	1.48×10^3	3.86×10^{-9}	1.73×10^{-5}	1.
¹⁹⁰ Pt	6.000×10^{11}	5.96×10^{-7}	---	1.73×10^{-5}	0.1
¹⁹³ Pt	5.000×10^2	7.15×10^2	2.45×10^{-8}	1.73×10^{-5}	1.
²⁰⁴ Pb	1.400×10^{17}	2.55×10^{-12}	1.72×10^{-16}	1.73×10^{-5}	1.
²⁰⁵ Pb	3.000×10^7	1.19×10^{-2}	1.89×10^{-9}	1.73×10^{-5}	1.
²¹⁰ Pb	2.230×10^1	1.60×10^4	4.66×10^{-8}	1.73×10^{-5}	1.
²⁰⁸ Bi	3.680×10^5	9.71×10^{-1}	2.66×10^{-8}	1.73×10^{-5}	1.
²¹⁰ Bi m	3.000×10^6	1.19×10^{-1}	1.75×10^{-8}	1.73×10^{-5}	0.1
²²⁶ Ra	1.600×10^3	2.23×10^2	3.69×10^{-7}	1.73×10^{-5}	0.1
²²⁷ Ac	2.177×10^1	1.64×10^4	5.08×10^{-6}	1.73×10^{-5}	0.1
²²⁹ Th	7.339×10^3	4.87×10^1	1.51×10^{-7}	1.73×10^{-5}	0.1
²³⁰ Th	7.700×10^4	4.64	1.33×10^{-1}	1.73×10^{-5}	0.01
²³² Th	1.405×10^{10}	2.54×10^{-5}	1.49×10^{-10}	1.73×10^{-5}	0.01
²³¹ Pa	3.277×10^4	1.09×10^1	1.92×10^{-5}	1.73×10^{-5}	0.1
²³² U	7.200×10^1	4.97×10^{11}	2.98×10^{-2}	1.73×10^{-5}	0.1
²³³ U	1.585×10^5	2.26	2.86×10^{-5}	1.73×10^{-5}	0.1
²³⁴ U	2.445×10^5	1.46	1.19	2.03×10^{-5}	0.1
²³⁵ U	7.038×10^8	5.08×10^{-4}	1.72×10^{-2}	1.73×10^{-5}	0.1
²³⁶ U	2.341×10^7	1.53×10^{-2}	2.56×10^{-1}	1.73×10^{-5}	0.1
²³⁸ U	4.468×10^9	8.00×10^{-5}	3.17×10^{-1}	1.73×10^{-5}	0.1
²³⁶ Np	1.150×10^5	3.11	5.43×10^{-6}	1.73×10^{-5}	1.
²³⁷ Np	2.140×10^6	1.67×10^{-1}	3.15×10^{-1}	1.73×10^{-5}	0.1
²³⁸ Pu	8.774×10^1	4.07×10^3	2.33×10^3	1.73×10^{-5}	0.1
²³⁹ Pu	2.406×10^4	1.49×10^1	3.13×10^2	3.05×10^{-3}	0.1
²⁴⁰ Pu	6.537×10^3	5.47×10^1	5.27×10^2	4.78×10^{-3}	0.1
²⁴² Pu	3.869×10^5	9.24×10^{-1}	1.72	1.73×10^{-5}	0.1
²⁴⁴ Pu	8.261×10^7	4.33×10^{-3}	4.22×10^{-7}	1.73×10^{-5}	0.1
²⁴¹ Am	4.322×10^2	8.27×10^2	1.69×10^3	8.79×10^{-3}	0.1
²⁴² Am m	1.520×10^2	2.35×10^3	6.93	1.73×10^{-5}	0.1
²⁴³ Am	7.380×10^3	4.84×10^1	1.71×10^1	1.56×10^{-1}	0.1
²⁴³ Cm	2.850×10^1	1.25×10^1	1.66×10^1	1.73×10^{-5}	0.1

Table B.1 continued.

Species	Half-life (yr)	PWR 33,000			
		Molar activity (Ci/mol)	Inventory (Ci/MTHM)	NRC limit (Ci/MTHM-yr)	EPA limit (Ci/MTHM)
^{245}Cm	8.499×10^3	4.21×10^1	1.46×10^{-1}	1.73×10^{-5}	0.1
^{246}Cm	4.731×10^3	7.56×10^1	3.12×10^{-2}	1.73×10^{-5}	0.1
^{247}Cm	1.560×10^7	2.29×10^{-2}	8.42×10^{-8}	1.73×10^{-5}	0.1
^{248}Cm	3.391×10^5	1.05	1.89×10^{-7}	1.73×10^{-5}	0.1
^{250}Cm	1.740×10^4	2.05×10^1	1.91×10^{-14}	1.73×10^{-5}	0.1
^{249}Cf	3.505×10^2	1.02×10^3	1.84×10^{-6}	1.73×10^{-5}	0.1
^{251}Cf	8.980×10^2	3.98×10^2	5.18×10^{-8}	1.73×10^{-5}	0.1

Table B.2: Sample BWR spent-fuel inventory.

Species	Half-life (yr)	BWR 27,500			
		Molar activity (Ci/mol)	Inventory (Ci/MTHM)	NRC limit (Ci/MTHM-yr)	EPA limit (Ci/MTHM)
¹⁰ Be	1.600×10^6	2.23×10^{-1}	3.43×10^{-6}	1.57×10^{-5}	1.
¹⁴ C	5.729×10^3	6.24×10^1	1.53	1.57×10^{-5}	0.1
³² Si	6.500×10^2	5.50×10^2	1.53×10^{-8}	1.57×10^{-5}	1.
³⁶ Cl	3.010×10^5	1.19	1.10×10^{-2}	1.57×10^{-5}	1.
³⁹ Ar	2.690×10^2	1.33×10^3	5.42×10^{-5}	1.57×10^{-5}	1.
⁴² Ar	3.300×10^1	1.08×10^4	2.03×10^{-13}	1.57×10^{-5}	1.
⁴⁰ K	1.280×10^9	2.79×10^{-4}	3.09×10^{-9}	1.57×10^{-5}	1.
⁴¹ Ca	8.100×10^4	4.41	1.74×10^{-4}	1.57×10^{-5}	1.
⁵⁰ V	4.000×10^{16}	8.94×10^{-12}	7.50×10^{-15}	1.57×10^{-5}	1.
⁵⁹ Ni	8.000×10^4	4.47	1.07	1.57×10^{-5}	1.
⁶³ Ni	9.200×10^1	3.89×10^3	1.47×10^2	1.57×10^{-5}	1.
⁷⁹ Se	6.496×10^4	5.50	3.38×10^{-1}	1.57×10^{-5}	1.
⁸¹ Kr	2.099×10^5	1.70	3.79×10^{-7}	1.57×10^{-5}	1.
⁸⁷ Rb	4.696×10^{10}	7.61×10^{-6}	1.77×10^{-5}	1.57×10^{-5}	1.
⁹⁰ Sr	2.912×10^1	1.23×10^4	4.70×10^4	1.57×10^{-5}	1.
⁹³ Zr	1.530×10^6	2.34×10^{-1}	1.80	1.80×10^{-5}	1.
⁹¹ Nb	1.000×10^4	3.58×10^1	---	1.57×10^{-5}	1.
⁹⁴ Nb	2.030×10^4	1.76×10^1	3.15×10^{-2}	1.57×10^{-5}	1.
⁹³ Mo	3.498×10^3	1.02×10^2	5.97×10^{-4}	1.57×10^{-5}	1.
⁹⁷ Tc	2.600×10^6	1.38×10^{-1}	---	1.57×10^{-5}	1.
⁹⁸ Tc	4.199×10^6	8.51×10^{-2}	3.06×10^{-6}	1.57×10^{-5}	1.
⁹⁹ Tc	2.130×10^5	1.68	1.11×10^1	1.11×10^{-4}	10.
¹⁰⁷ Pd	6.496×10^6	5.50×10^{-2}	9.46×10^{-2}	1.57×10^{-5}	1.
¹⁰⁸ Ag m	1.270×10^2	2.81×10^3	1.14×10^{-2}	1.57×10^{-5}	1.
¹¹⁵ In	4.997×10^{14}	7.15×10^{-10}	1.26×10^{-11}	1.57×10^{-5}	1.
¹²¹ Sn m	4.997×10^1	7.15×10^3	1.25	1.57×10^{-5}	1.
¹²⁶ Sn	1.000×10^5	3.58	6.24×10^{-1}	1.57×10^{-5}	1.
¹²³ Te	1.000×10^{13}	3.58×10^{-8}	2.18×10^{-12}	1.57×10^{-5}	1.
¹²⁹ I	1.570×10^7	2.28×10^{-2}	2.64×10^{-2}	1.57×10^{-5}	0.1
¹³⁵ Cs	2.300×10^6	1.55×10^{-1}	3.59×10^{-1}	1.57×10^{-5}	1.
¹³⁷ Cs	3.000×10^1	1.19×10^4	6.79×10^4	1.57×10^{-5}	1.
¹³⁷ La	5.999×10^4	5.96	---	1.57×10^{-5}	1.
¹³⁸ La	1.350×10^{11}	2.65×10^{-6}	9.33×10^{-11}	1.57×10^{-5}	1.
¹⁴² Ce	1.049×10^{11}	3.41×10^{-6}	2.25×10^{-5}	1.57×10^{-5}	1.
¹⁴⁴ Nd	2.099×10^{15}	1.70×10^{-10}	1.26×10^{-9}	1.57×10^{-5}	0.1
¹⁴⁶ Sm	7.000×10^7	5.11×10^{-3}	1.79×10^{-7}	1.57×10^{-5}	0.1
¹⁴⁷ Sm	1.070×10^{11}	3.34×10^{-6}	4.18×10^{-6}	1.57×10^{-5}	0.1
¹⁴⁸ Sm	8.001×10^{15}	4.47×10^{-11}	4.07×10^{-11}	1.57×10^{-5}	0.1
¹⁴⁹ Sm	9.994×10^{15}	3.58×10^{-11}	7.05×10^{-13}	1.57×10^{-5}	0.1
¹⁵¹ Sm	8.999×10^1	3.97×10^3	2.98×10^2	1.57×10^{-5}	1.

Table B.2 continued.

Species	Half-life (yr)	BWR 27,500			
		Molar activity (Ci/mol)	Inventory (Ci/MTHM)	NRC limit (Ci/MTHM-yr)	EPA limit (Ci/MTHM)
¹⁵⁰ Eu	3.600×10^1	9.93×10^3	1.13×10^{-5}	1.57×10^{-5}	1.
¹⁵² Gd	1.080×10^{14}	3.31×10^{-9}	2.22×10^{-12}	1.57×10^{-5}	0.1
¹⁵⁷ Tb	1.500×10^2	2.38×10^3	---	1.57×10^{-5}	1.
¹⁶³ Ho	3.300×10^1	1.08×10^4	---	1.57×10^{-5}	1.
¹⁶⁶ Ho m	1.200×10^3	2.98×10^2	2.68×10^{-3}	1.57×10^{-5}	1.
¹⁷⁶ Lu	3.000×10^{10}	1.19×10^{-5}	6.18×10^{-11}	1.57×10^{-5}	1.
¹⁸² Hf	9.000×10^6	3.97×10^{-2}	6.28×10^{-7}	1.57×10^{-5}	1.
¹⁸⁰ Ta	1.600×10^{13}	2.23×10^{-8}	---	1.57×10^{-5}	1.
¹⁸⁷ Re	5.000×10^{10}	7.15×10^{-6}	3.57×10^{-8}	1.57×10^{-5}	1.
¹⁹² Ir m	2.410×10^2	1.48×10^3	4.33×10^{-9}	1.57×10^{-5}	1.
¹⁹⁰ Pt	6.000×10^{11}	5.96×10^{-7}	---	1.57×10^{-5}	0.1
¹⁹³ Pt	5.000×10^2	7.15×10^2	2.85×10^{-8}	1.57×10^{-5}	1.
²⁰⁴ Pb	1.400×10^{17}	2.55×10^{-12}	1.72×10^{-16}	1.57×10^{-5}	1.
²⁰⁵ Pb	3.000×10^7	1.19×10^{-2}	1.71×10^{-9}	1.57×10^{-5}	1.
²¹⁰ Pb	2.230×10^1	1.60×10^4	4.79×10^{-8}	1.57×10^{-5}	1.
²⁰⁸ Bi	3.680×10^5	9.71×10^{-1}	1.78×10^{-8}	1.57×10^{-5}	1.
²¹⁰ Bi m	3.000×10^6	1.19×10^{-1}	1.59×10^{-8}	1.57×10^{-5}	0.1
²²⁶ Ra	1.600×10^3	2.23×10^2	3.64×10^{-7}	1.57×10^{-5}	0.1
²²⁷ Ac	2.177×10^1	1.64×10^4	5.37×10^{-6}	1.57×10^{-5}	0.1
²²⁹ Th	7.339×10^3	4.87×10^1	1.22×10^{-7}	1.57×10^{-5}	0.1
²³⁰ Th	7.700×10^4	4.64	1.23×10^{-4}	1.57×10^{-5}	0.01
²³² Th	1.405×10^{10}	2.54×10^{-5}	1.30×10^{-10}	1.57×10^{-5}	0.01
²³¹ Pa	3.277×10^4	1.09×10^1	1.98×10^{-5}	1.57×10^{-5}	0.1
²³² U	7.200×10^1	4.97×10^3	1.74×10^{-2}	1.57×10^{-5}	0.1
²³³ U	1.585×10^5	2.26	2.04×10^{-5}	1.57×10^{-5}	0.1
²³⁴ U	2.445×10^5	1.46	1.04	1.68×10^{-5}	0.1
²³⁵ U	7.038×10^8	5.08×10^{-4}	1.63×10^{-2}	1.57×10^{-5}	0.1
²³⁶ U	2.341×10^7	1.53×10^{-2}	2.15×10^{-1}	1.57×10^{-5}	0.1
²³⁸ U	4.468×10^9	8.00×10^{-5}	3.20×10^{-1}	1.57×10^{-5}	0.1
²³⁶ Np	1.150×10^5	3.11	3.03×10^{-6}	1.57×10^{-5}	1.
²³⁷ Np	2.140×10^6	1.67×10^{-1}	2.46×10^{-1}	1.57×10^{-5}	0.1
²³⁸ Pu	8.774×10^1	4.07×10^3	1.78×10^3	1.57×10^{-5}	0.1
²³⁹ Pu	2.406×10^4	1.49×10^1	3.00×10^2	2.92×10^{-3}	0.1
²⁴⁰ Pu	6.537×10^3	5.47×10^1	4.78×10^2	4.33×10^{-3}	0.1
²⁴² Pu	3.869×10^5	9.24×10^{-1}	1.42	1.57×10^{-5}	0.1
²⁴⁴ Pu	8.261×10^7	4.33×10^{-3}	2.51×10^{-7}	1.57×10^{-5}	0.1
²⁴¹ Am	4.322×10^2	8.27×10^2	1.56×10^3	8.07×10^{-3}	0.1
²⁴² Am m	1.520×10^2	2.35×10^3	8.33	1.57×10^{-5}	0.1
²⁴³ Am	7.380×10^3	4.84×10^1	1.29×10^1	1.18×10^{-4}	0.1
²⁴³ Cm	2.850×10^1	1.25×10^1	1.31×10^1	1.57×10^{-5}	0.1

Table B.2 continued.

Species	Half-life (yr)	BWR 27,500			
		Molar activity (Ci/mol)	Inventory (Ci/MTHM)	NRC limit (Ci/MTHM-yr)	EPA limit (Ci/MTHM)
^{245}Cm	8.499×10^3	4.21×10^1	9.25×10^{-2}	1.57×10^{-5}	0.1
^{246}Cm	4.731×10^3	7.56×10^1	1.73×10^{-2}	1.57×10^{-5}	0.1
^{247}Cm	1.560×10^7	2.29×10^{-2}	4.25×10^{-8}	1.57×10^{-5}	0.1
^{248}Cm	3.391×10^5	1.05	8.52×10^{-8}	1.57×10^{-5}	0.1
^{250}Cm	1.740×10^4	2.05×10^1	5.68×10^{-15}	1.57×10^{-5}	0.1
^{249}Cf	3.505×10^2	1.02×10^3	7.87×10^{-7}	1.57×10^{-5}	0.1
^{251}Cf	8.980×10^2	3.98×10^2	1.99×10^{-8}	1.57×10^{-5}	0.1

C Computer algorithms

As an aid to readers who may wish to use the formulas in this report, this appendix provides FORTRAN routines for calculating them. Included are the functions in Equations 27, 30, 31, and 32 and their integrals, and a subroutine for solving the radioactive-decay equation (Equation 1). The integrals of the functions are recommended for use rather than the functions themselves because they include the effects of possible pulse releases at $t = 0$ and $t = t_r$. The usage is illustrated in the following calling sequence.

```
c
c Example of usage of the functions. The releases are to be
c calculated for a time step of length DT that ends at TIME (that is,
c for the interval from TIME-DT to TIME).
c
c  TAUC      mean container lifetime,
c  TAUF      mean cladding lifetime,
c  TAUA      beginning of "resaturation",
c  TAUB      end of "resaturation",
c  TS        time scale for structural-metal releases.
c  TC        time scale for cladding releases.
c  TM        time scale for matrix releases.
c  FSC       fraction of C-14 inventory in structural metals,
c  FCC       fraction of C-14 inventory in cladding,
c  FQC       fraction of C-14 inventory available for quick release,
c  NUMSPC    total number of species included,
c  I         array index indicating radionuclide species,
c  FS(I)     fraction of inventory in structural metals (0 for C-14),
c  FC(I)     fraction of inventory in cladding (0 for C-14),
c  FM(I)     fraction of inventory in fuel matrix,
c  FG(I)     fraction of inventory in gap/grain-boundaries,
c  FC14(I)   1 for C-14, 0 for other nuclides,
c  SINIT(I)  initial inventory of nuclide I (radioactivity units),
c  STIME(I)  initial inventory decayed to TIME,
c  NSPEC(I)  position of nuclide I in its chain
c             (1 for parent, 2 for 1st daughter, etc. The nuclides
c             for each chain must be grouped together and in order),
c  TMCNST(I) decay constant for nuclide I,
c  RELSTR    release rate for structural metals,
c  RELCLD    release rate for cladding,
c  RELXMT    release rate for fuel matrix,
c  RELGAP    release rate for gap/grain-boundaries,
c  RELC14    release rate for structural-metal and cladding
c             part of C-14 releases,
c  REL(I)    the source term (radioactivity released per unit time).
c
c First, the release rates are calculated without including the
c inventory.
c
      tt1=time-dt
      tt2=time-0.5D+0*dt
      tt3=time
```

```

    relstr=strint(tt3,tauc,taua,taub,ts)
&    -strint(tt1,tauc,taua,taub,ts)
    if((relstr.le.1.D-10).and.(tt3.gt.taua)) then
        relstr=strden(tt2,tauc,taua,taub,ts)
    else
        relstr=relstr/dt
    endif
    relcld=strint(tt3,tauc,taua,taub,tz)
&    -strint(tt1,tauc,taua,taub,tz)
    if((relcld.le.1.D-10).and.(tt3.gt.taua)) then
        relcld=strden(tt2,tauc,taua,taub,tz)
    else
        relcld=relcld/dt
    endif
    relxmt=xmtint(tt3,tauc,tauf,taua,taub,tm)
&    -xmtint(tt1,tauc,tauf,taua,taub,tm)
    if((relxmt.le.1.D-10).and.(tt3.gt.taua)) then
        relxmt=xmtden(tt2,tauc,tauf,taua,taub,tm)
    else
        relxmt=relxmt/dt
    endif
    relgap=gapint(tt3,tauc,tauf,taua,taub)
&    -gapint(tt1,tauc,tauf,taua,taub)
    if((relgap.le.1.D-10).and.(tt3.gt.taua)) then
        relgap=gapden(tt2,tauc,tauf,taua,taub)
    else
        relgap=relgap/dt
    endif
    rel1=c14int(tt3,tauc,ts,fsc,0.D+0)
&    -c14int(tt1,tauc,ts,fsc,0.D+0)
    if(rel1.le.1.D-10) then
        rel1=c14den(tt2,tauc,ts,fsc,0.D+0)
    else
        rel1=rel1/dt
    endif
    rel2=c14int(tt3,tauc,tz,fcc,fqc)
&    -c14int(tt1,tauc,tz,fcc,fqc)
    if(rel2.le.1.D-10) then
        rel2=c14den(tt2,tauc,tz,fcc,fqc)
    else
        rel2=rel2/dt
    endif
    relc14=rel1+rel2

c
c Call DECAY to get the decayed inventory at the middle of the
c time step (time = TT2).
c
    call decay(tt2,numspc,nspec,tmcnst,sinit,stime)

C
C Add up the fractional releases for each nuclide and multiply
C by the inventory.
C
    do 10 i=1,numspc
        rel(i)=stime(i)*(relstr*fs(i)+relcld*fc(i)

```

```

      &      +relxmt*fm(i)+relgap*fg(i)+relc14*fc14(i))
10      continue

```

Note that the integral functions (STRINT, C14INT, XMTINT, and GAPINT) are used unless the difference across the time step is less than 1×10^{-10} , in which case the "raw" release functions (STRDEN, C14DEN, XMTDEN, and GAPDEN) are used. This switch is included to improve stability.

```

      real*8 function strden(time,tauc,taua,taub,ts)
c
c STRDEN is the release rate for structural metals or cladding. It
c calculates the expression in Equation 27 of SAND91-0155
c (not including effects of radioactive decay). Inputs are
c
c      TIME    time,
c      TAUC    mean container lifetime,
c      TAUA    beginning of "resaturation",
c      TAUB    end of "resaturation",
c      TS      time scale for structural-metal releases.
c
c The calculation is only valid if TS .GE. TAUB .GE. TAUA .GE. 0 and
c TAUC .GE. 0, but those conditions are not tested.
c
      implicit real*8 (a-h,o-z)
      if(time.le.taua) then
         strden=0.D+0
      else if(time.le.taub) then
         if(tauc.eq.0.D+0) then
            tex=1.D+0
         else
            tex=1.D+0-exp(-time/tauc)
         endif
         strden=(time+tex*(time-taua-tauc))/(taub-taua)/ts
      else if(time.le.ts) then
         if(tauc.eq.0.D+0) then
            tex=1.D+0
         else
            tex=1.D+0-exp(-time/tauc)
         endif
         strden=tex/ts
      else
         if(ts.eq.0.D+0) then
            if(tauc.eq.0.D+0) then
               strden=0.D+0
            else
               strden=exp(-time/tauc)/tauc
            endif
         else
            if(tauc.eq.0.D+0) then
               tex=0.D+0
            else
               tex=exp(-time/tauc)*(exp(ts/tauc)-1.D+0)
            endif
         endif
      endif

```

```

        strden=tex/ts
    endif
endif
return
end

real*8 function strint(time,tauc,taua,taub,ts)
c
c STRINT is cumulative release for structural metals or cladding. It
c calculates the integral of the expression in Equation 27 of
c SAND91-0155 (not including effects of radioactive decay).
c Inputs are
c
c     TIME    time,
c     TAUC    mean container lifetime,
c     TAUA    beginning of "resaturation",
c     TAUB    end of "resaturation",
c     TS      time scale for structural-metal releases.
c
c The calculation is only valid if TS .GE. TAUB .GE. TAUA .GE. 0 and
c TAUC .GE. 0, but those conditions are not tested.
c
    implicit real*8 (a-h,o-z)
    if(time.le.taua) then
        strint=0.D+0
    else if(time.le.taub) then
        if(tauc.eq.0.D+0) then
            tex=0.D+0
        else
            tex=tauc*(1.D+0-exp(-time/tauc))
        endif
        strint=(time-tex)*(time-taua)/(taub-taua)/ts
    else if(time.le.ts) then
        if(tauc.eq.0.D+0) then
            tex=0.D+0
        else
            tex=tauc*(1.D+0-exp(-time/tauc))
        endif
        strint=(time-tex)/ts
    else
        if(ts.eq.0.D+0) then
            if(tauc.eq.0.D+0) then
                tex=0.D+0
            else
                tex=exp(-time/tauc)
            endif
            strint=1.D+0-tex
        else
            if(tauc.eq.0.D+0) then
                tex=0.D+0
            else
                tex=tauc*exp(-time/tauc)*(exp(ts/tauc)-1.D+0)
            endif
            strint=1.D+0-tex/ts
        endif
    endif
endfunction

```

```

        endif
    endif
    return
end

real*8 function c14den(time,tauc,tz,fc,fq)
c
c C14DEN is the release rate for carbon-14 in gaseous form. It
c calculates the expression in Equation 30 of SAND91-0155
c (not including effects of radioactive decay). Inputs are
c
c     TIME    time,
c     TAUC    mean container lifetime,
c     TZ      time scale for cladding releases,
c     FC      fraction of C-14 inventory in cladding,
c     FQ      fraction of C-14 inventory available for quick release.
c
c The calculation is only valid if TAUC and TZ are .GE. 0, and FC and
c FQ are between zero and one (also, their sum should be .LE. 1).
c These conditions are not tested.
c
    implicit real*8 (a-h,o-z)
    if(time.le.0.D+0) then
        c14den=0.D+0
    else if(time.le.tz) then
        if(tauc.eq.0.D+0) then
            c14den=fc/tz
        else
            tex=exp(-time/tauc)
            c14den=fc*(1.D+0-tex)/tz+fq*tex/tauc
        endif
    else
        if(tauc.eq.0.D+0) then
            c14den=0.D+0
        else
            c14den=fq*exp(-time/tauc)/tauc
        endif
        if(tz.eq.0.D+0) then
            if(tauc.gt.0.D+0)
&                c14den=c14den+fc*exp(-time/tauc)/tauc
            else
                if(tauc.eq.0.D+0) then
                    tex=0.D+0
                else
                    tex=exp(-time/tauc)*(exp(tz/tauc)-1.D+0)
                endif
                c14den=c14den+fc*tex/tz
            endif
        endif
    endif
    return
end

real*8 function c14int(time,tauc,tz,fc,fq)
c

```



```

c C14INT is cumulative release for carbon-14 in gaseous form. It
c calculates the integral of the expression in Equation 30 of
c SAND91-0155 (not including effects of radioactive decay).
c Inputs are
c
c      TIME    time,
c      TAUC    mean container lifetime,
c      TZ      time scale for cladding releases,
c      FC      fraction of C-14 inventory in cladding,
c      FQ      fraction of C-14 inventory available for quick release.
c
c The calculation is only valid if TAUC and TZ are .GE. 0, and FC and
c FQ are between zero and one (also, their sum should be .LE. 1).
c These conditions are not tested.
c

```

```

      implicit real*8 (a-h,o-z)
      if(time.le.0.D+0) then
        c14int=0.D+0
      else if(time.le.tz) then
        if(tauc.eq.0.D+0) then
          tex=1.D+0
        else
          tex=1.D+0-exp(-time/tauc)
        endif
        c14int=fc*(time-tauc*tex)/tz+fq*tex
      else
        if(tauc.eq.0.D+0) then
          tex=1.D+0
        else
          tex=1.D+0-exp(-time/tauc)
        endif
        c14int=fq*tex
        if(tz.eq.0.D+0) then
          if(tauc.eq.0.D+0) then
            tex=0.D+0
          else
            tex=exp(-time/tauc)
          endif
          c14int=c14int+fc*(1.D+0-tex)
        else
          if(tauc.eq.0.D+0) then
            tex=0.D+0
          else
            tex=tauc*exp(-time/tauc)*(exp(tz/tauc)-1.D+0)
          endif
          c14int=c14int+fc*(1.D+0-tex/tz)
        endif
      endif
    endif
  return
end

```

```

      real*8 function xmtden(time,tauc,tauf,taua,taub,tm)
c
c XMTDEN is the release rate from the fuel-pellet matrix. It

```

```

c calculates the expression in Equation 31 of SAND91-0155
c (not including effects of radioactive decay). Inputs are
c
c      TIME    time,
c      TAUC    mean container lifetime,
c      TAUF    mean cladding lifetime,
c      TAUA    beginning of "resaturation",
c      TAUB    end of "resaturation",
c      TM      time scale for matrix releases.
c
c The calculation is only valid if TAUB .GE. TAUA .GE. 0 and
c TAUC, TAUF, and TM are .GE. 0, but those conditions are not tested.
c Note that this subroutine is actually more general than Equation 21,
c because TM is allowed to be less than TAUB - TAUA.
c

```

```

      implicit real*8 (a-h,o-z)
      if(tm.eq.0.D+0) then
        xmtden=gapden(time,tauc,tauf,taua,taub)
      else if(time.le.taua) then
        xmtden=0.D+0
      else
        if(time.le.taub+tm) then
          if(tauf.eq.tauc) then
            if(tauc.eq.0.D+0) then
              ex1=0.D+0
            else
              ex1=(1.D+0+time/tauc)*exp(-time/tauc)
            endif
          else if(tauc.eq.0.D+0) then
            ex1=exp(-time/tauf)
          else if(tauf.eq.0.D+0) then
            ex1=exp(-time/tauc)
          else
            ex1=(tauc*exp(-time/tauc)
&              -tauf*exp(-time/tauf))/(tauc-tauf)
          endif
          ex1=1.D+0-ex1
        endif
        if(time.gt.taua+tm) then
          if(tauf.eq.tauc) then
            if(tauc.eq.0.D+0) then
              ex2=0.D+0
            else
              ex2=((1.D+0+(time-tm)/tauc)*exp(-(time-tm)/tauc)
&              -(1.D+0+time/tauc)*exp(-time/tauc))
            endif
          else if(tauc.eq.0.D+0) then
            ex2=exp(-time/tauf)*(exp(tm/tauf)-1.D+0)
          else if(tauf.eq.0.D+0) then
            ex2=exp(-time/tauc)*(exp(tm/tauc)-1.D+0)
          else
            ex2=(tauc*exp(-time/tauc)*(exp(tm/tauc)-1.D+0)
&              -tauf*exp(-time/tauf)*(exp(tm/tauf)-1.D+0))
&              /(tauc-tauf)

```

```

        endif
    endif
    if(tm.ge.taub-taua) then
        if(time.le.taub) then
            xmtden=ex1*(time-taua)/(taub-taua)/tm
        else if(time.le.taua+tm) then
            xmtden=ex1/tm
        else if(time.le.taub+tm) then
            xmtden=(ex1*(taub+tm-time)
&             +ex2*(time-taua-tm))/(taub-taua)/tm
        else
            xmtden=ex2/tm
        endif
    else
        if(time.le.taua+tm) then
            xmtden=ex1/tm*(time-taua)/(taub-taua)
        else if(time.le.taub) then
            xmtden=(ex1*tm+ex2*(time-taua-tm))/(taub-taua)/tm
        else if(time.le.taub+tm) then
            xmtden=(ex1*(taub+tm-time)
&             +ex2*(time-taua-tm))/(taub-taua)/tm
        else
            xmtden=ex2/tm
        endif
    endif
endif
return
end

real*8 function xmtint(time,tauc,tauf,taua,taub,tm)
c
c XMTINT is cumulative release from the fuel-pellet matrix. It
c calculates the integral of the expression in Equation 31 of
c SAND91-0155 (not including effects of radioactive decay).
c Inputs are
c
c     TIME    time,
c     TAUC    mean container lifetime,
c     TAUF    mean cladding lifetime,
c     TAUA    beginning of "resaturation",
c     TAUB    end of "resaturation",
c     TM      time scale for matrix releases.
c
c The calculation is only valid if TAUB .GE. TAUA .GE. 0 and
c TAUC, TAUF, and TM are .GE. 0, but those conditions are not tested.
c Note that this subroutine is actually more general than Equation 21,
c because TM is allowed to be less than TAUB - TAUA.
c
    implicit real*8 (a-h,o-z)
    if(tm.eq.0.D+0) then
        xmtint=gapint(time,tauc,tauf,taua,taub)
        return
    endif
    if(tm.ge.taub-taua) then

```

```

if(time.le.taua) then
    temp=taua
else if(time.le.taub) then
    temp=time
else
    temp=taub
endif
if(temp.eq.taua) then
    xmtint=0.D+0
else
    if(tauf.eq.tauc) then
        if(tauc.eq.0.D+0) then
            ex1=0.D+0
            ex2=0.D+0
        else
            ex1=tauc*(3.D+0*tauc+taua)*exp(-taua/tauc)
            ex2=(temp*(temp-taua+3.D+0*tauc)
&             +tauc*(3.D+0*tauc-2.D+0*taua))*exp(-temp/tauc)
            endif
        else if(tauc.eq.0.D+0) then
            ex1=tauf*tauf*exp(-taua/tauf)
            ex2=tauf*(temp-taua+tauf)*exp(-temp/tauf)
        else if(tauf.eq.0.D+0) then
            ex1=tauc*tauc*exp(-taua/tauc)
            ex2=tauc*(temp-taua+tauc)*exp(-temp/tauc)
        else
            ex1=(tauc**3*exp(-taua/tauc)
&             -tauf**3*exp(-taua/tauf))/(tauc-tauf)
            ex2=(tauc*tauc*(temp-taua+tauc)*exp(-temp/tauc)
&             -tauf*tauf*(temp-taua+tauf)*exp(-temp/tauf))
&             /(tauc-tauf)
            endif
        xmtint=(0.5D+0*(temp-taua)**2-ex1+ex2)/(taub-taua)/tm
    endif
if(time.le.taub) then
    temp=taub
else if(time.le.taua+tm) then
    temp=time
else
    temp=taua+tm
endif
if(temp.gt.taub) then
    if(tauf.eq.tauc) then
        if(tauc.eq.0.D+0) then
            ex1=0.D+0
            ex2=0.D+0
        else
            ex1=(2.D+0*tauc+taub)*exp(-taub/tauc)
            ex2=(2.D+0*tauc+temp)*exp(-temp/tauc)
        endif
    else if(tauc.eq.0.D+0) then
        ex1=tauf*exp(-taub/tauf)
        ex2=tauf*exp(-temp/tauf)
    else if(tauf.eq.0.D+0) then

```

```

        ex1=tauc*exp(-taub/tauc)
        ex2=tauc*exp(-temp/tauc)
    else
        ex1=(tauc*tauc*exp(-taub/tauc)
&         -tauf*tauf*exp(-taub/tauf))/(tauc-tauf)
        ex2=(tauc*tauc*exp(-temp/tauc)
&         -tauf*tauf*exp(-temp/tauf))/(tauc-tauf)
    endif
    xmtint=xmtint+(temp-taub-ex1+ex2)/tm
endif
if(time.le.taua+tm) then
    temp=taua+tm
else if(time.le.taub+tm) then
    temp=time
else
    temp=taub+tm
endif
if(temp.gt.taua+tm) then
    if(tauf.eq.tauc) then
        if(tauc.eq.0.D+0) then
            ex1=0.D+0
            ex2=0.D+0
            ex3=0.D+0
            ex4=0.D+0
        else
            ex1=(2.D+0*tauc+taua+tm)*exp(-(taua+tm)/tauc)
            ex2=(2.D+0*tauc+temp)*exp(-temp/tauc)
            ex3=tauc*(3.D+0*tauc+taua)*exp(-taua/tauc)
            ex4=((temp-tm)*(temp-tm-taua+3.D+0*tauc)
&             +tauc*(3.D+0*tauc-2.D+0*taua))
&             *exp(-(temp-tm)/tauc)
        endif
    else if(tauc.eq.0.D+0) then
        ex1=tauf*exp(-(taua+tm)/tauf)
        ex2=tauf*exp(-temp/tauf)
        ex3=tauf*tauf*exp(-taua/tauf)
        ex4=tauf*(temp-tm-taua+tauf)*exp(-(temp-tm)/tauf)
    else if(tauf.eq.0.D+0) then
        ex1=tauc*exp(-(taua+tm)/tauc)
        ex2=tauc*exp(-temp/tauc)
        ex3=tauc*tauc*exp(-taua/tauc)
        ex4=tauc*(temp-tm-taua+tauc)*exp(-(temp-tm)/tauc)
    else
        ex1=(tauc*tauc*exp(-(taua+tm)/tauc)
&         -tauf*tauf*exp(-(taua+tm)/tauf))/(tauc-tauf)
        ex2=(tauc*tauc*exp(-temp/tauc)
&         -tauf*tauf*exp(-temp/tauf))/(tauc-tauf)
        ex3=(tauc**3*exp(-taua/tauc)
&         -tauf**3*exp(-taua/tauf))/(tauc-tauf)
        ex4=(tauc*tauc*(temp-tm-taua+tauc)*exp(-(temp-tm)/tauc)
&         -tauf*tauf*(temp-tm-taua+tauf)
&         *exp(-(temp-tm)/tauf))/(tauc-tauf)
    endif
    xmtint=xmtint+0.5D+0*(taub-taua

```

```

&          -(taub+tm-temp)**2/(taub-taua))/tm
&          -(ex1-ex2-(ex3-ex4))/(taub-taua))/tm
endif
if(time.le.taub+tm) then
    temp=taub+tm
else
    temp=time
endif
if(temp.gt.taub+tm) then
    if(tauf.eq.tauc) then
        if(tauc.eq.0.D+0) then
            ex1=0.D+0
            ex2=0.D+0
            ex3=0.D+0
            ex4=0.D+0
        else
            ex1=(2.D+0*tauc+temp)*exp(-temp/tauc)
            ex2=(2.D+0*tauc+taub)*exp(-taub/tauc)
            ex3=(2.D+0*tauc+temp-tm)*exp(-(temp-tm)/tauc)
            ex4=(2.D+0*tauc+taub+tm)*exp(-(taub+tm)/tauc)
        endif
    else if(tauc.eq.0.D+0) then
        ex1=tauf*exp(-temp/tauf)
        ex2=tauf*exp(-taub/tauf)
        ex3=tauf*exp(-(temp-tm)/tauf)
        ex4=tauf*exp(-(taub+tm)/tauf)
    else if(tauf.eq.0.D+0) then
        ex1=tauc*exp(-temp/tauc)
        ex2=tauc*exp(-taub/tauc)
        ex3=tauc*exp(-(temp-tm)/tauc)
        ex4=tauc*exp(-(taub+tm)/tauc)
    else
        ex1=(tauc*tauc*exp(-temp/tauc)
&          -tauf*tauf*exp(-temp/tauf))/(tauc-tauf)
        ex2=(tauc*tauc*exp(-taub/tauc)
&          -tauf*tauf*exp(-taub/tauf))/(tauc-tauf)
        ex3=(tauc*tauc*exp(-(temp-tm)/tauc)
&          -tauf*tauf*exp(-(temp-tm)/tauf))/(tauc-tauf)
&          ex4=(tauc*tauc*exp(-(taub+tm)/tauc)
&          -tauf*tauf*exp(-(taub+tm)/tauf))/(tauc-tauf)
        endif
        xmtint=xmtint+(ex1+ex2-ex3-ex4)/tm
    endif
else
    if(time.le.taua) then
        temp=taua
    else if(time.le.taua+tm) then
        temp=time
    else
        temp=taua+tm
    endif
    if(temp.eq.taua) then
        xmtint=0.D+0
    else

```

```

if(tauf.eq.tauc) then
  if(tauc.eq.0.D+0) then
    ex1=0.D+0
    ex2=0.D+0
  else
    ex1=tauc*(3.D+0*tauc+taua)*exp(-taua/tauc)
    ex2=(temp*(temp-taua+3.D+0*tauc)
&      +tauc*(3.D+0*tauc-2.D+0*taua))*exp(-temp/tauc)
    endif
  else if(tauc.eq.0.D+0) then
    ex1=tauf*tauf*exp(-taua/tauf)
    ex2=tauf*(temp-taua+tauf)*exp(-temp/tauf)
  else if(tauf.eq.0.D+0) then
    ex1=tauc*tauc*exp(-taua/tauc)
    ex2=tauc*(temp-taua+tauc)*exp(-temp/tauc)
  else
    ex1=(tauc**3*exp(-taua/tauc)
&      -tauf**3*exp(-taua/tauf))/(tauc-tauf)
    ex2=(tauc*tauc*(temp-taua+tauc)*exp(-temp/tauc)
&      -tauf*tauf*(temp-taua+tauf)*exp(-temp/tauf))
&      /(tauc-tauf)
    endif
    xmtint=(0.5D+0*(temp-taua)**2-ex1+ex2)/(taub-taua)/tm
  endif
if(time.le.taua+tm) then
  temp=taua+tm
else if(time.le.taub) then
  temp=time
else
  temp=taub
endif
if(temp.gt.taua+tm) then
  if(tauf.eq.tauc) then
    if(tauc.eq.0.D+0) then
      ex1=0.D+0
      ex2=0.D+0
      ex3=0.D+0
      ex4=0.D+0
    else
      ex1=((temp-tm)*(temp-tm-taua+3.D+0*tauc)
&      +tauc*(3.D+0*tauc-2.D+0*taua))
&      *exp(-(temp-tm)/tauc)
      ex2=(temp*(temp-taua+3.D+0*tauc)
&      +tauc*(3.D+0*tauc-2.D+0*taua))*exp(-temp/tauc)
      ex3=tauc*(3.D+0*tauc+taua)*exp(-taua/tauc)
      ex4=(tauc*(3.D+0*(tauc+tm)+taua)+tm*(taua+tm))
&      *exp(-(taua+tm)/tauc)
    endif
  else if(tauc.eq.0.D+0) then
    ex1=tauf*(temp-tm-taua+tauf)*exp(-(temp-tm)/tauf)
    ex2=tauf*(temp-taua+tauf)*exp(-temp/tauf)
    ex3=tauf*tauf*exp(-taua/tauf)
    ex4=tauf*(tauf+tm)*exp(-(taua+tm)/tauf)
  else if(tauf.eq.0.D+0) then

```

```

ex1=tauc*(temp-tm-taua+tauc)*exp(-(temp-tm)/tauc)
ex2=tauc*(temp-taua+tauc)*exp(-temp/tauc)
ex3=tauc*tauc*exp(-taua/tauc)
ex4=tauc*(tauc+tm)*exp(-(taua+tm)/tauc)
else
ex1=(tauc*tauc*(temp-tm-taua+tauc)*exp(-(temp-tm)/tauc)
&      -tauf*tauf*(temp-tm-taua+tauf)
&      *exp(-(temp-tm)/tauf))/(tauc-tauf)
ex2=(tauc*tauc*(temp-taua+tauc)*exp(-temp/tauc)
&      -tauf*tauf*(temp-taua+tauf)*exp(-temp/tauf))
&      /(tauc-tauf)
ex3=(tauc**3*exp(-taua/tauc)
&      -tauf**3*exp(-taua/tauf))/(tauc-tauf)
ex4=(tauc*tauc*(tauc+tm)*exp(-(taua+tm)/tauc)
&      -tauf*tauf*(tauf+tm)*exp(-(taua+tm)/tauf))
&      /(tauc-tauf)
endif
xmtint=xmtint+(temp-tm-taua
&      -(ex1-ex2-ex3+ex4)/tm)/(taub-taua)
endif
if(time.le.taub) then
temp=taub
else if(time.le.taub+tm) then
temp=time
else
temp=taub+tm
endif
if(temp.gt.taub) then
if(tauf.eq.tauc) then
if(tauc.eq.0.D+0) then
ex1=0.D+0
ex2=0.D+0
ex3=0.D+0
ex4=0.D+0
else
ex1=(2.D+0*tauc+temp)*exp(-temp/tauc)
ex2=(2.D+0*tauc+taub)*exp(-taub/tauc)
ex3=((temp-tm)*(temp-tm-taua+3.D+0*tauc)
&      +tauc*(3.D+0*tauc-2.D+0*taua))
&      *exp(-(temp-tm)/tauc)
ex4=((taub-tm)*(taub-tm-taua+3.D+0*tauc)
&      +tauc*(3.D+0*tauc-2.D+0*taua))
&      *exp(-(taub-tm)/tauc)
endif
else if(tauc.eq.0.D+0) then
ex1=tauf*exp(-temp/tauf)
ex2=tauf*exp(-taub/tauf)
ex3=tauf*(temp-tm-taua+tauf)*exp(-(temp-tm)/tauf)
ex4=tauf*(taub-tm-taua+tauf)*exp(-(taub-tm)/tauf)
else if(tauf.eq.0.D+0) then
ex1=tauc*exp(-temp/tauc)
ex2=tauc*exp(-taub/tauc)
ex3=tauc*(temp-tm-taua+tauc)*exp(-(temp-tm)/tauc)
ex4=tauc*(taub-tm-taua+tauc)*exp(-(taub-tm)/tauc)

```



```

else
    ex1=(tauc*tauc*exp(-temp/tauc)
&         -tauf*tauf*exp(-temp/tauf))/(tauc-tauf)
    ex2=(tauc*tauc*exp(-taub/tauc)
&         -tauf*tauf*exp(-taub/tauf))/(tauc-tauf)
    ex3=(tauc*tauc*(temp-tm-taua+tauc)*exp(-(temp-tm)/tauc)
&         -tauf*tauf*(temp-tm-taua+tauf)
&         *exp(-(temp-tm)/tauf))/(tauc-tauf)
    ex4=(tauc*tauc*(taub-tm-taua+tauc)*exp(-(taub-tm)/tauc)
&         -tauf*tauf*(taub-tm-taua+tauf)
&         *exp(-(taub-tm)/tauf))/(tauc-tauf)
endif
xmtint=xmtint+(0.5D+0*(tm*tm
&         -(taub+tm-temp)**2)/(taub-taua)
&         +ex1-ex2-(ex3-ex4)/(taub-taua))/tm
endif
if(time.le.taub+tm) then
    temp=taub+tm
else
    temp=time
endif
if(temp.gt.taub+tm) then
    if(tauf.eq.tauc) then
        if(tauc.eq.0.D+0) then
            ex1=0.D+0
            ex2=0.D+0
            ex3=0.D+0
            ex4=0.D+0
        else
            ex1=(2.D+0*tauc+temp)*exp(-temp/tauc)
            ex2=(2.D+0*tauc+taub)*exp(-taub/tauc)
            ex3=(2.D+0*tauc+temp-tm)*exp(-(temp-tm)/tauc)
            ex4=(2.D+0*tauc+taub+tm)*exp(-(taub+tm)/tauc)
        endif
    else if(tauc.eq.0.D+0) then
        ex1=tauf*exp(-temp/tauf)
        ex2=tauf*exp(-taub/tauf)
        ex3=tauf*exp(-(temp-tm)/tauf)
        ex4=tauf*exp(-(taub+tm)/tauf)
    else if(tauf.eq.0.D+0) then
        ex1=tauc*exp(-temp/tauc)
        ex2=tauc*exp(-taub/tauc)
        ex3=tauc*exp(-(temp-tm)/tauc)
        ex4=tauc*exp(-(taub+tm)/tauc)
    else
        ex1=(tauc*tauc*exp(-temp/tauc)
&         -tauf*tauf*exp(-temp/tauf))/(tauc-tauf)
        ex2=(tauc*tauc*exp(-taub/tauc)
&         -tauf*tauf*exp(-taub/tauf))/(tauc-tauf)
        ex3=(tauc*tauc*exp(-(temp-tm)/tauc)
&         -tauf*tauf*exp(-(temp-tm)/tauf))/(tauc-tauf)
        ex4=(tauc*tauc*exp(-(taub+tm)/tauc)
&         -tauf*tauf*exp(-(taub+tm)/tauf))/(tauc-tauf)
    endif
endif

```

```

        xmtint=xmtint+(ex1+ex2-ex3-ex4)/tm
    endif
endif
return
end

real*8 function gapden(time,tauc,tauf,taua,taub)
c
c GAPDEN is the release rate from the gap/grain-boundaries. It
c calculates the expression in Equation 32 of SAND91-0155
c (not including effects of radioactive decay). Inputs are
c
c     TIME    time,
c     TAUC    mean container lifetime,
c     TAUF    mean cladding lifetime,
c     TAUA    beginning of "resaturation",
c     TAUB    end of "resaturation".
c
c The calculation is only valid if TAUB .GE. TAUA .GE. 0 and
c TAUC and TAUF are .GE. 0, but those conditions are not tested.
c
    implicit real*8 (a-h,o-z)
    if(time.le.taua) then
        gapden=0.D+0
    else if(time.le.taub) then
        if(tauf.eq.tauc) then
            if(tauc.eq.0.D+0) then
                ex1=0.D+0
            else
                ex1=(time*(time-taua-tauc)/(tauc*tauc)-1.D+0)
                & *exp(-time/tauc)
            endif
        else if(tauc.eq.0.D+0) then
            ex1=(time-taua-tauf)/tauf*exp(-time/tauf)
        else if(tauf.eq.0.D+0) then
            ex1=(time-taua-tauc)/tauc*exp(-time/tauc)
        else
            ex1=((time-taua-tauc)*exp(-time/tauc)
            & -(time-taua-tauf)*exp(-time/tauf))/(tauc-tauf)
        endif
        gapden=(1.D+0+ex1)/(taub-taua)
    else
        if(tauf.eq.tauc) then
            if(tauc.eq.0.D+0) then
                gapden=0.D+0
            else
                gapden=exp(-time/tauc)*time/(tauc*tauc)
            endif
        else if(tauc.eq.0.D+0) then
            gapden=exp(-time/tauf)/tauf
        else if(tauf.eq.0.D+0) then
            gapden=exp(-time/tauc)/tauc
        else
            gapden=(exp(-time/tauc)

```

```

&          -exp(-time/tauf))/ (tauc-tauf)
      endif
    endif
  return
end

      real*8 function gapint(time,tauc,tauf,taua,taub)
c
c GAPINT is cumulative release from the gap/grain-boundaries. It
c calculates the integral of the expression in Equation 32 of
c SAND91-0155 (not including effects of radioactive decay).
c Inputs are
c
c      TIME    time,
c      TAUC    mean container lifetime,
c      TAUF    mean cladding lifetime,
c      TAUA    beginning of "resaturation",
c      TAUB    end of "resaturation".
c
c The calculation is only valid if TAUB .GE. TAUA .GE. 0 and
c TAUC and TAUF are .GE. 0, but those conditions are not tested.
c
      implicit real*8 (a-h,o-z)
      if(time.le.taua) then
        gapint=0.D+0
      else
        if(tauf.eq.tauc) then
          if(tauc.eq.0.D+0) then
            ex1=0.D+0
          else
            ex1=(1.D+0+time/tauc)*exp(-time/tauc)
          endif
        else if(tauc.eq.0.D+0) then
          ex1=exp(-time/tauf)
        else if(tauf.eq.0.D+0) then
          ex1=exp(-time/tauc)
        else
          ex1=(tauc*exp(-time/tauc)
&          -tauf*exp(-time/tauf))/ (tauc-tauf)
        endif
        if(time.le.taub) then
          factor=(time-taua)/(taub-taua)
        else
          factor=1.D+0
        endif
        gapint=factor*(1.D+0-ex1)
      endif
    return
  end

      subroutine decay(time,numspc,nspec,tmcnst,sinit,stime)
c
c DECAY solves the decay-chain equations by solving for normal
c modes, for which the decay matrix is diagonal. For the normal

```

```

c modes, the solution is simply given by an exponential decay for each
c mode. Then, the normal-mode decomposition is reversed.
c
c      TIME          time,
c      NUMSPC        total number of species.
c
c For species I:
c
c      NSPEC(I)       position of nuclide I in its chain
c                    (1 for parent, 2 for 1st daughter, etc.),
c      TMCNST(I)      decay constant,
c      SINIT(I)       initial inventory (radioactivity units),
c      STIME(I)       initial inventory decayed to TIME,
c      TM(I,II)       coefficient in solution,
c      SNORM(I)       amount in I-th normal mode.
c
c      implicit real*8 (a-h,o-z)
c      dimension sinit(numspc),stime(numspc),nspec(numspc),
c      &          tmcnst(numspc)
c      dimension tm(100,100),snorm(100),expon(100)
c
c Calculate coefficients for decay solution. Note that this algorithm
c does not work if two nuclides in the same chain have the same
c half-life.
c
c      do 30 ii=1,numspc
c      do 20 i=1,numspc
c      if(i.lt.ii) then
c          tm(i,ii)=0.D+0
c      else if(i.eq.ii) then
c          tm(i,ii)=1.D+0
c      else if(nspec(i).gt.1) then
c          tm(i,ii)=tm(i-1,ii)*tmcnst(i)/(tmcnst(i)-tmcnst(ii))
c      else
c          do 10 iii=i,numspc
10      tm(iii,ii)=0.D+0
c          go to 30
c      endif
c      continue
20      continue
30      continue
c
c Find the normal-mode decomposition.
c
c      do 40 i=1,numspc
c      snorm(i)=sinit(i)
c      do 40 ii=i-nspec(i)+1,i-1
c      snorm(i)=snorm(i)-tm(i,ii)*snorm(ii)
40      continue
c
c The preceding would normally be calculated just once, at the
c beginning of the program. The final part is all that has to be
c done at each time step.
c
c And now, reverse the normal-mode decomposition.

```

```

c
do 50 i=1,numspc
  expon(i)=exp(-tmcnst(i)*time)
  stime(i)=0.D+0
  do 50 ii=i-nspec(i)+1,i
    stime(i)=stime(i)+tm(i,ii)*snorm(ii)*expon(ii)
50  continue
  return
end

```

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