

PROGRESS IN THE DEVELOPMENT OF WASTE PACKAGE PERFORMANCE

REQUIREMENTS FOR A REPOSITORY LOCATED IN BASALT

M. J. Smith, M. S. Bensky, T. B. McCall
Rockwell Hanford Operations
Richland, Washington 99352

ABSTRACT

The Basalt Waste Isolation Project (BWIP) waste package reference conceptual design consists of three components: the waste form, the canister, and the backfill. The waste package system is an engineered barrier in series with two barriers which are in parallel, i.e., the geologic site barrier and the repository seal system barrier (shaft seal, tunnel backfill, and borehole seals). Preliminary analyses of radionuclide transport and release through the waste package system and site geology are presented herein. The effect of a range of postulated groundwater travel times on radionuclide release to the accessible environment is shown. The required values for two waste package performance parameters are shown as a function of groundwater travel time and potential radionuclide release to the accessible environment.

INTRODUCTION

The Basalt Waste Isolation Project (BWIP) under the U.S. Department of Energy's National Waste Terminal Storage Program was established in 1978 to determine the feasibility of constructing a nuclear waste repository in basalt (NWRB). A major project task is the development of designs for site-specific, multiple barrier waste package systems for spent fuel and processed high-level waste.

The waste package is one of the three major systems necessary for the safe isolation of nuclear waste. The other systems are the repository seal system (e.g., borehole seals, tunnel backfill, and shaft seals) and the geologic site. The performance requirements to be imposed on the waste package system depend upon the overall performance objectives for the waste isolation system and the capabilities of the other systems. This paper describes the BWIP's progress in the development of waste package performance requirements to meet Environmental Protection Agency (EPA) draft standards (EPA, 1982) and Nuclear Regulatory Commission (NRC) draft standards (NRC, 1981) applicable to the repository post-closure period. The waste package, geologic site, and repository seal system characteristics described herein are based on the latest available data. However, the results presented herein should be considered preliminary. An extensive research effort in this area by the BWIP is continuing.

SUMMARY OF KEY REGULATORY REQUIREMENTS

The primary concern of the NRC and the EPA is that reasonable assurance of acceptable isolation of radioactive waste in a repository can be demonstrated despite the complexities of a geologic site and the long periods of time (i.e., 10,000 yr) over which it must perform. The regulatory approach taken by these agencies calls for the development of an engineered barrier system to complement the geologic site, as indicated by the following excerpt from 10 CFR 60 (NRC, 1981) and 40 CFR 191 (EPA, 1982):

10 CFR 60:

"It has chosen to address this difficulty by requiring that a Department of Energy (DOE) proposal be based upon a multiple barrier approach. An engineered barrier system is required to compensate for uncertainties in predicting the performance of

the geologic setting, especially during the period of high radioactivity. Similarly, because the performance of the engineered barrier system is also subject to considerable uncertainty, the geologic setting must be able to contribute significantly to isolation."

40 CFR 191:

"Disposal systems shall use several different types of barriers to isolate the wastes from the accessible environment. Both engineered and natural barriers shall be designed to provide substantial isolation."

Having stated the necessity for the development and emplacement of an engineered barrier system, the NRC specifically identifies a waste package as a major part of that system and proposes two criteria which the engineered barrier system should meet:

10 CFR 60:

"The engineered barrier system shall be designed so that assuming anticipated processes and events, (a) containment of HLW will be substantially complete during the period when radiation and thermal conditions in the underground facility are dominated by fission product decay; (b) any release of radionuclides from the engineered barrier system shall be a gradual process which results in small fractional releases to the geologic setting over long times."

In 10 CFR 60, the numerical values suggested for the containment requirement and the release requirement are 1,000 yr and one part in 10^5 per yr, respectively. It is also stated that other values may be proposed on a "case by case" basis, but the BWIP staff is presently utilizing the NRC containment time and slow release guidelines as design objectives for the waste package. The draft EPA requirements on isolation system performance include: (1) specific limits (L_i) to the cumulative release (A_i) at the accessible environment over 10,000 yr on individual isotopes such that $A_i \leq L_i$; and (2) a limit on the cumulative release over 10,000 yr of these same radionuclides such that

$$\sum_i A_i / L_i \leq 1 \quad (1)$$

The EPA limits (L_i) and cumulative releases (A_i) are expressed in curies per 1,000 metric tonnes of heavy metal (Ci/1000 MTHM).

IMPORTANT BARRIER PROCESSES

The Nuclear Waste Repository in Basalt (NWRB) is a waste isolation system composed of three major barrier systems. Figure 1 schematically shows the three major barrier systems and lists the processes which affect the isolation of radionuclides from the accessible environment. The engineered waste package barrier is in series with both the natural geologic site barrier and the engineered repository seal system barrier, which are in parallel. This configuration has the following important implications with respect to establishing performance requirements for the waste package system:

- (1) The common properties and isolation mechanisms of the two engineered barriers must not be subject to common-mode failure.
- (2) The performance requirements for the waste package system must be integrated with the performance requirements for the repository seal system barrier and the expected performance of the site geologic barrier to meet the isolation system performance criteria.
- (3) The margin-of-safety and the reliability of the NWRB isolation system are directly affected by the waste package performance because the performance of the geologic barrier is fixed.

CHARACTERISTICS OF THE NWRB WASTE PACKAGE REFERENCE CONCEPTUAL DESIGN

Figure 2 illustrates commercial high-level waste (CHLW) and spent fuel (SF) waste package emplaced in basalt based on the present BWIP waste package reference conceptual designs. The waste package for CHLW consists of the waste, a stainless steel container, a carbon steel canister, and a backfill composed of a mixture of crushed basalt and sodium bentonite. The waste package for spent fuel consists of the fuel rods, a carbon steel canister, and the backfill.

The NWRB waste package design is expected to provide an effective barrier to radionuclide releases through the following three mechanisms:

- (1) Containment within the carbon steel canister
- (2) Control of releases from the waste form
- (3) Sorption/precipitation of radionuclides and control of water flow by the backfill.

The low Eh (chemically reducing) and moderate pH (basic) conditions of the groundwater in the natural geochemical environment are expected to result in a low corrosion rate for the carbon steel canister. Furthermore, the basalt component of the backfill should maintain the low Eh conditions and the bentonite component of the backfill should ensure that groundwater movement around the canister is very slow, minimizing removal of corrosion products and the exposure of more active surfaces. Thus, it appears that the conceptual waste canister will prevent contact between the waste matrix and groundwater during the first few hundred years after

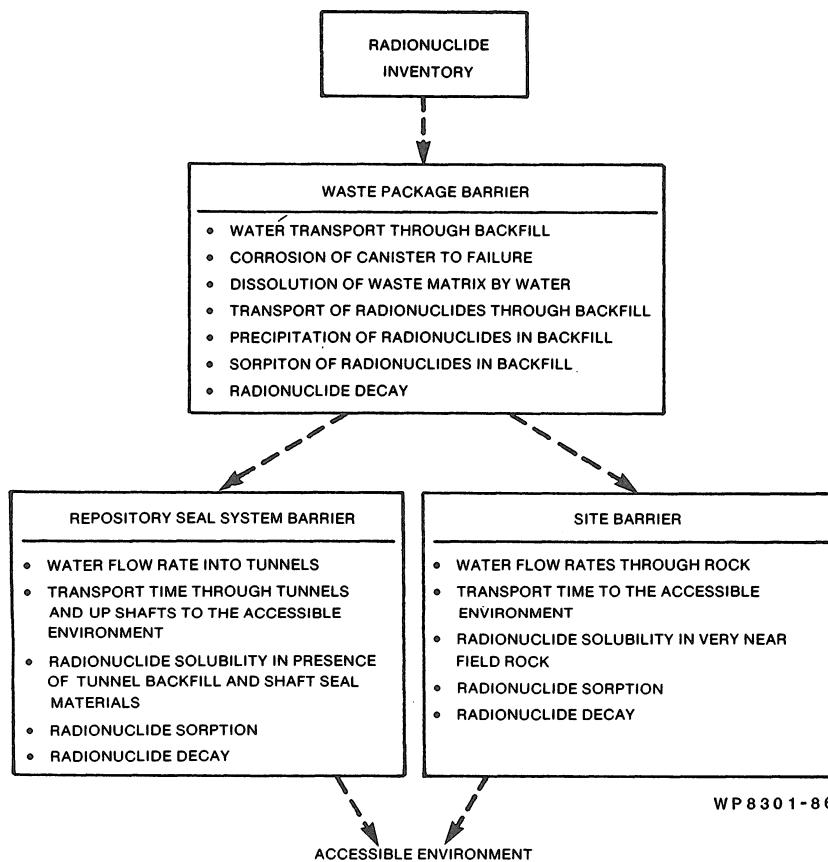
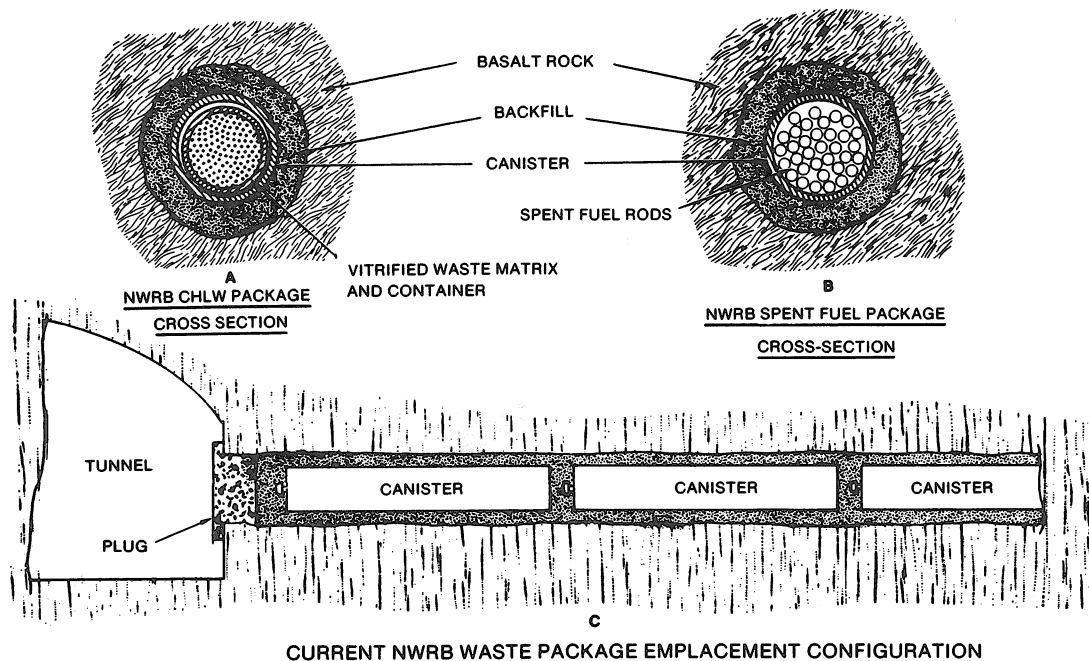


Fig. 1. Processes That Occur Within the Major Barrier Systems for a Nuclear Waste Repository Located in Basalt.



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Fig. 2a. NWRB CHLW Package Cross Section; 2b. NWRB Spent Fuel Package Cross Section; 2c. Current NWRB Waste Package Emplacement Configuration.

repository closure when the temperatures are relatively high and will likely provide this protection for several thousand years. If the canister fails through localized attack, the waste matrix would be contacted by groundwater, but transport rates would be extremely low through small openings due to pitting. General wall thinning leading to catastrophic failure is not expected for thousands of years.

The waste matrix is expected to be either ceramic spent fuel pellets inside fuel cladding for spent fuel or borosilicate glass for processed commercial high-level waste. The release of radionuclides from spent fuel will be limited in the post-closure condition by the cladding, canister, and the waste package backfill. The dissolution rate of borosilicate glass will be a function of chemical composition, temperature, composition of the water to which it is eventually exposed, and other variables. It is presently assumed that the waste package control of radionuclide release from processed waste will be achieved by the waste matrix, canister, and backfill. The major function that the backfill will perform in addition to control of canister corrosion is control of radionuclide transport after canister perforation. Control is achieved by the following mechanisms: (1) diffusion control of mass transport; (2) maintenance of a chemically reducing environment which results in low radionuclide solubility; (3) sorption of radionuclides, and (4) radionuclide chemical reactions to form secondary minerals.

REPOSITORY SEAL AND GEOLOGIC SITE BARRIER PERFORMANCE ESTIMATES

Preliminary performance estimates have been completed for both the repository seal barrier and the geologic site barrier. These performance estimates included hydrological analyses of the repository seal system, identification of the important radionuclides based on initial concentrations and half-lives, and

estimation of releases to the accessible environment considering dissolution and sorption processes.

The preliminary hydrological analyses completed by the BWIP show that, of the total water flow which could dissolve radionuclides from the waste package, less than one percent will be transported through the seal system pathway (i.e., tunnel backfill, shafts, and seals) to the accessible environment. Analyses were performed which included the effects of the small fraction of water which would move through the seal system, the transport time of water in the seal system, radionuclide solubility limitations, radionuclide sorption and radioactive decay. These analyses lead to the conclusion that a seal system can be designed which will effectively limit radionuclide releases through that path and that releases through the seal system are small relative to releases from the geologic site barrier.

In contrast with the repository seal system, the geologic site has fixed characteristics and performance which are being evaluated through the BWIP's site characterization activities (Rockwell, 1982). Because the site has fixed characteristics and performance and because it will be the dominant pathway for the movement of radionuclides to the accessible environment, the waste package performance requirements need to be established relative to the performance of the site. Therefore, the remainder of this paper will address the waste package/site processes shown in Fig. 1.

The major processes which occur naturally in the geologic site barrier or will be designed into the waste package barrier are listed in Fig. 1. In the NWRB isolation system, waste package integrity, low radionuclide solubility, radionuclide sorption, and long groundwater travel time combine to ensure very slow release and transport of radionuclides so that decay will reduce the radioactivity to the EPA limits before arrival at the accessible environment. Simple equations

were used in the conceptual design effort to analyze the major processes listed in Fig. 1. Equations 2 and 3 below were used appropriately under conditions where releases from the waste package are controlled primarily by solubility or by the waste package alone. For simplicity the subscript i has been omitted from Equations 2 and 3 and from all of the radionuclide properties given in the equations below (e.g., M, m, s, λ , R, etc.).

Equation 2 is applicable to radionuclides which will form phases that have low solubilities in the basalt geochemical environment. Equation 3 is used for radionuclides which are not solubility limited but are limited by the waste package characteristics.

For Radionuclide Solubility-Limited (RSL) Release:

$$A = mMSWT_R e^{-\lambda R t_w} \quad (2)$$

and

For Waste Package-Limited (WPL) Release:

$$A = r A_0 e^{-\lambda R t_w} \frac{e^{-\lambda t_c}}{\lambda} 1 - e^{-\lambda/r} \quad (3)$$

where:

$$T_R = \frac{1}{\lambda} \ln \frac{F + \lambda}{F} \quad (4)$$

$$F = \frac{mMSW}{A_0 e^{-\lambda t_c}} \quad (5)$$

$$R = 1 + Kd \rho \frac{1 - \phi}{\phi} \quad (6)$$

M = molar solubility limit, mol/L

m = molecular weight, g/mol

S = specific activity, Ci/g

W = water flow, L/yr

T_R = release duration, yr

λ = decay constant, yr⁻¹

t_w = groundwater travel time to accessible environment, yr

A_0 = radionuclide inventory at emplacement, curies

t_c = containment time, yr

r = initial waste package release rate, yr⁻¹, for WPS release

R = retardation factor

F = the initial annual release fraction, yr⁻¹, for RSL release

Kd = distribution coefficient, mL/g

ρ = flowpath rock density, g/mL

ϕ = flowpath effective porosity

In Equation 2, the term mMSW accounts for solubility-controlled release of radionuclide and has units of Curies per year per 1,000 metric tonnes of heavy metal (MTHM). This term is multiplied by the release duration T_R (yr). The product is the total amount of radionuclide which would be released into groundwater during the period of release. The initial fractional release rate due to solubility limits, expressed as a fraction, F, of the inventory is given by Equation 5. In Equation 2, the exponential term, $e^{-\lambda R t_w}$ accounts for the decay of a radionuclide during transport from the waste package to the accessible environment. The amount of time in which decay can occur is the product of the groundwater travel time, t_w , and the retardation factor for the specific radionuclide, R. Equation 6 related the radionuclide retardation factor to the radionuclide distribution coefficient, the flowpath rock density, and flowpath effective porosity. The radio-

nuclide distribution coefficients must be determined experimentally for each radionuclide. In Equation 5, the exponential term, $e^{-\lambda t_c}$, accounts for radioactive decay during the containment period (i.e., while the waste is inside the canister).

Analyses which include the isolation capability of the waste package require the use of Equation 2 for radionuclides which have low solubilities. Equation 3 is utilized to calculate release rates for radionuclides which must be controlled by the waste package because they have high solubilities relative to a specified release rate. Equation 3 has terms similar to those in Equation 2 for decay of each radionuclide during the containment time and the transport time. The term within the brackets accounts for the release of radionuclides during the time interval from t_c to $t_c + 1/r$. The computer program which uses Equations 2 and 3 has an algorithm which allows the user to evaluate partial or total radionuclide release at the accessible environment over any selected interval of time. A preliminary analysis of the geologic site barrier was performed by the BWIP using a computer code which implements Equation 2 using groundwater travel time as the independent variable. It was assumed that the total repository inventory consisted of one-half spent fuel and one-half commercial high-level waste. Conservative values were used for the molar solubility and distribution coefficient of each radionuclide and water flow rates. The containment time was conservatively assumed to be zero. The analysis included selected radionuclides which have sufficient inventories or long enough half-lives to be important. The results were expressed in fractions of the EPA limits for the worst 10,000 yr interval and is, therefore, more conservative than the EPA standard which is only concerned with the first 10,000 yr after repository closure. The results of the calculation are shown in Curve 1 of Fig. 3. Curve 1 shows that for basalt, the amount of radioactivity released to the accessible environment is less than the EPA limit for groundwater travel times greater than 6,000 yr. Current estimates of the groundwater travel times for a NWRB are much greater than 6,000 yr. Curve 1 decreases rapidly as travel time increases because the release is dominated by ¹⁴C which has a half-life of 5,632 yr, is soluble, and is not sorbed. At a groundwater travel time of about 10⁵ yr, ¹²⁹I will contribute to a release of approximately 0.033 because of a half-life of several million years, a high solubility, and low sorption. The important conclusion illustrated by Curve 1 is that even if conservative values are chosen for radionuclide solubilities and radionuclide retardation factors, the geologic site barrier could meet the EPA limit with no isolation contribution from the waste package if the groundwater travel time is 6,000 yr or more. However, an important consideration that is not evident in Curve 1 is the margin-of-safety required in the isolation system which will be discussed in the sections which follow.

CONCEPTUAL WASTE PACKAGE PERFORMANCE ESTIMATES

A preliminary analysis of the isolation performance of the NWRB was made by considering the contribution provided by the waste package barrier in a manner similar to that completed for the site alone except that a waste package release rate of 10⁻⁵/yr was assumed. Values of A were calculated using Equations 2 and 3. The lesser of the two values of A computed for each radionuclide was used in Equation 1. The result is plotted as Curve 2 in Fig. 3. Other parameters were the same for Curves 1 and 2. Curve 2 shows that the release is less than 0.1 (i.e., the release one-tenth of the EPA limit) for groundwater travel times greater than 1,000 yr. As is the case for Curve 1, ¹⁴C and ¹²⁹I are the dominant radionuclides.

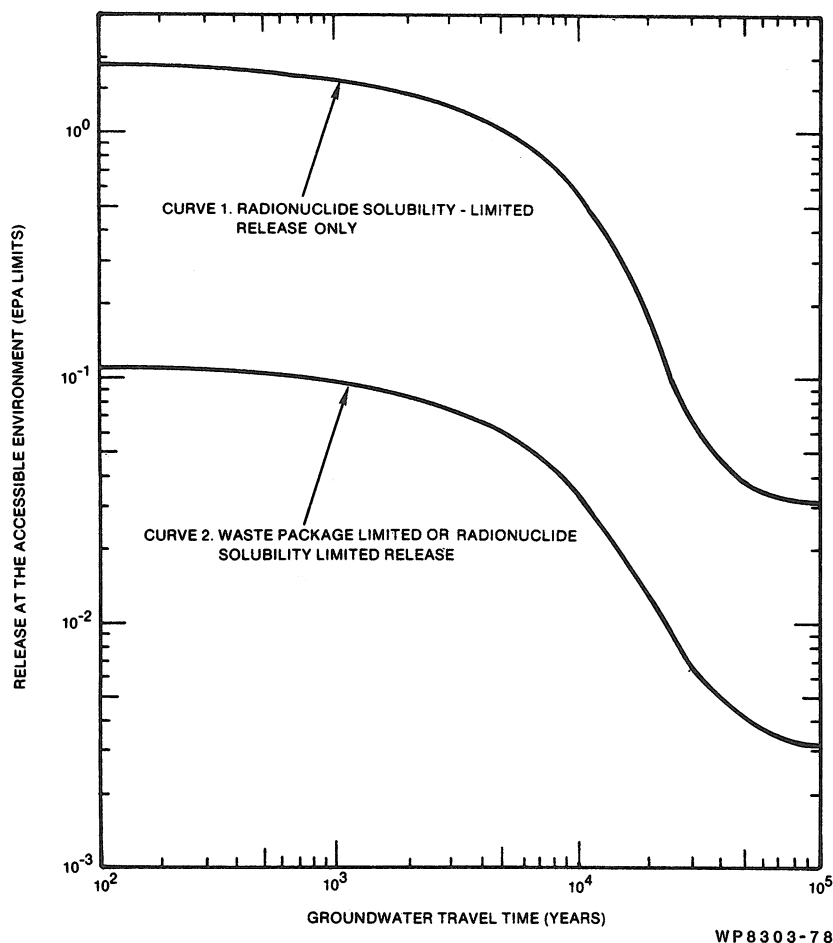


Fig. 3. Radionuclide Release at the Accessible Environment as a Function of Groundwater Travel Time With and Without Waste Package Limited Release Rates.

The improvement between Curves 1 and 2 shows that a waste package with a release rate of 10^{-5} parts per year can contribute more than an order of magnitude reduction in the release to the accessible environment and, thus, provides a margin-of-safety.

The release of ^{14}C and ^{129}I to the accessible environment as a function of waste package release rates, containment time, and groundwater travel time has also been estimated using Equations 3 and 6 and the same assumptions as in the previous cases. The results are shown in Fig. 4. Since the retardation factor, R , is unity for the only two radionuclides of importance, the effect of containment time and groundwater travel time on radioactive decay is the same (see Equation 3). Therefore, the results shown in Fig. 4 allow the reader to select a value of containment plus groundwater travel time and to choose one of the two performance objectives (either the EPA curie limits or one-tenth of the EPA curie limit), and obtain the waste package release rate (fraction per year) which will meet the performance objective.

Some typical tradeoffs can be illustrated using the lower curve ($0.1 \times$ EPA limit) of Fig. 4 which is based on the release in the worst 10,000 yr interval and is, therefore, more conservative than the EPA standard which is only concerned with the first 10,000 yr after closure. As one example, 2,000 yr of containment, 20,000 yr of water travel time ($t_c + t_w = 22,000$), and an initial waste package release rate of $1 \times 10^{-4} \text{ yr}^{-1}$ or less will meet the 0.1 EPA limit. This goal would also be met if the canister were designed for a 10,000 yr containment time to supplement a groundwater travel

time of 12,000 yr.

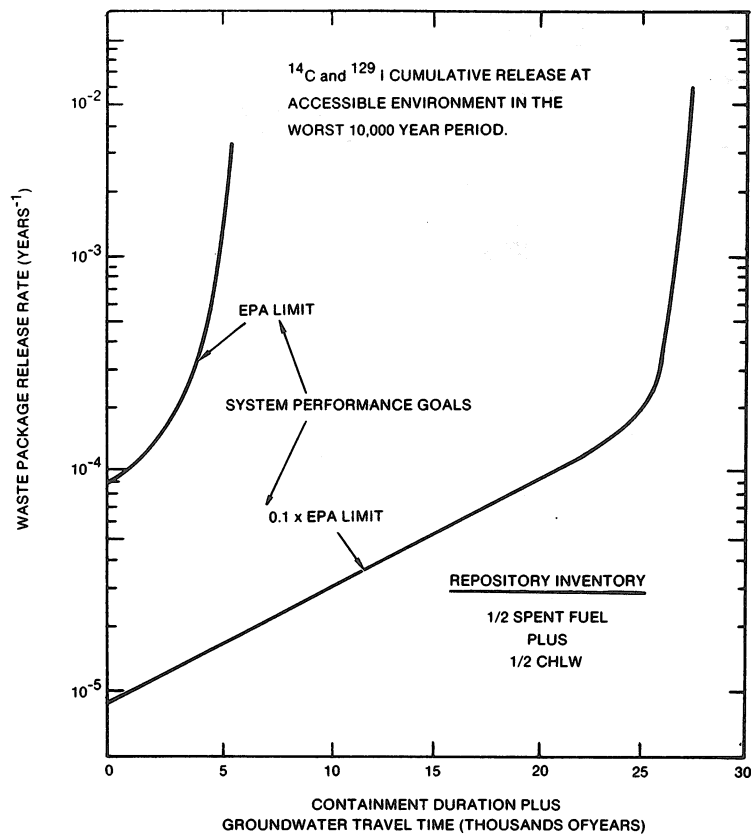
The upper curve of Fig. 4 shows that the EPA limit will be met for a postulated water travel time of 5,000 yr if the waste package release rate is 10^{-3} parts per year or less.

The important conclusion of these preliminary waste package performance analyses is that, once a performance objective for release to the accessible environment has been selected, two waste package design parameters, containment time and waste package release rate, can be selected, consistent with groundwater travel time, to meet the goal.

Analyses are being conducted to better quantify the performance of the waste package system, repository seal system, and geologic site to determine what margin of safety is possible in the performance of the major barriers comprising a NWRB. Experiments are underway to characterize the behavior of potential materials (e.g., waste form release rates, canister corrosion, canister-backfill interactions) and obtain key radionuclide behavior data (e.g., solubility, distribution coefficients, transport through backfill). Two major milestones in waste package design are the development of Waste Package Conceptual Design Upgrade Criteria (9/83) and the completion of the Waste Package Conceptual Design Upgrade (9/84) for a NWRB.

SUMMARY AND CONCLUSIONS

- (1) The present conceptual design for the NWRB waste package consists of: (a) either spent fuel rods



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Fig. 4. Example of Effect of System Performance Objective on Waste Package Requirements as a Function of Groundwater Travel Time Plus Containment Duration.

or vitrified waste in a stainless steel container (for processed commercial and defense wastes); (b) a carbon steel canister; and (c) a backfill composed of crushed basalt mixed with sodium bentonite.

- (2) The performance requirements for the waste package system must be integrated with the performance requirements for the repository seal system and the expected performance of the geologic site to meet the NWRB isolation system performance criteria.
- (3) The margin-of-safety and the reliability of the NWRB isolation system are directly affected by the waste package performance because the performance of the geologic site barrier is fixed.
- (4) Preliminary analyses have led to the conclusion that even if conservative values are chosen for radionuclide solubilities and retardation coefficients, the geologic site barrier could meet the EPA limit with no contribution from the waste package if the groundwater travel time is 6,000 yr or more.
- (5) Preliminary analyses demonstrate that the waste package can be designed to provide an effective barrier to radionuclide release. Once a performance objective for release to the accessible environment has been selected, two waste package performance parameters, containment time and waste package release rate, can be selected consistent with groundwater travel time to meet that goal.

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