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REPOSITORY ENVIRONMENTAL PARAMETERS AND MODELS/METHODOLOGIES RELEVANT TO ASSESSING THE PERFORMANCE OF HIGH-LEVEL WASTE PACKAGES IN BASALT, TUFF, AND SALT

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ABSTRACT

This document provides specifications for models/methodologies that could be employed in determining postclosure repository environmental parameters relevant to the performance of high-level waste packages for the Basalt Waste Isolation Project (BWIP) at Richland, Washington, the tuff at Yucca Mountain by the Nevada Test Site, and the bedded salt in Deaf Smith County, Texas. Guidance is provided on (1) the identity of the relevant repository environmental parameters (groundwater characteristics, temperature, radiation, and pressure), (2) the models/methodologies employed to determine the parameters, and (3) the input data base for the models/methodologies. Supporting studies included are (1) an analysis of potential waste package failure modes leading to identification of the relevant repository environmental parameters, (2) an evaluation of the credible range of the repository environmental parameters, and (3) a summary of the review of existing models/methodologies currently employed in determining repository environmental parameters relevant to waste package performance.

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1. EXECUTIVE SUMMARY

1.1 PURPOSE AND SCOPE

The purpose of this document is to provide specifications for models/methodologies to be employed in determining postclosure repository environmental parameters relevant to the performance of high-level waste (HLW) packages in repositories constructed in the basalt at the Hanford Site, Richland, Washington, the tuff in Yucca Mountain at the Nevada Test Site, and the bedded salt in Deaf Smith County, Texas. The specifications provide guidance on (1) the identity and determination of the repository environmental parameters, (2) the considerations that the models/methodologies must take into account, and (3) the input needed to determine the output parameters within the specifications for implementing the models/methodologies in the context of the overall waste package performance analysis.

It is emphasized that no attempt is made to describe a particular model or modeling approach to be used to demonstrate compliance with regulations. The choice of models and modeling approaches to be made must be dependent on waste package and repository designs, the degree of conservatism assumed, the site specific environment, and the performance allocated to the waste package that is required to satisfy the overall repository performance.

1.2 REGULATORY PERSPECTIVE

The Environmental Protection Agency (EPA) has published an environmental standard with respect to releases of radionuclides from HLW disposal sites (EPA 1985). The Nuclear Regulatory Commission (NRC) develops and issues regulations that cover all aspects of HLW disposal which will implement the standards; subsequently, it also licenses the repository. The Department of Energy (DOE) is primarily responsible for developing methods of disposal of HLW and has determined that national policy should focus on the disposal of HLW in mined geologic repositories.

These standards and regulations clearly require that the performance of a waste package be analyzed over a time frame of several thousand years to show compliance with the NRC containment and release rate criteria for a waste package and to provide source terms for the repository regions surrounding the waste package (near-field) and the regions away from the immediate vicinity of the repository (far-field) analyses. The environment inside and around the waste package is a critical determinant of its performance, thus making the repository environment a subject of concern.

1.3 TECHNICAL PERSPECTIVE

Implicit in the purpose of the document is the assumption that the model/methodology will acquire input parameters from some source, perform certain internal functions, and then generate output values for subsequent use in the performance assessment of a waste package. An approach to a waste package reliability analysis has been given by Sastre and co-workers (1986) and in a more recent Federal Register notice (NRC 1985). In this guidance, the failure probabilities are to be predicted based on the application of simple phenomenological models, with the input parameters sampled from distribution functions by some probabilistic scheme. According to the suggested guidelines, we would expect a performance model to be composed of the following component models:

1. a time-dependent temperature model;
2. a time-dependent heat source model;
3. a time-dependent radiation model to predict gamma dose rates in the packing;
4. a time-dependent water flow model;
5. a water chemistry model capable of predicting the parameters of interest such as pH, redox conditions, and salt concentrations as a function of temperature, radiation, and time;
6. a corrosion model capable of predicting corrosion rates as a function of temperature, water chemistry, and radiation doses;
7. a mechanical failure model capable of predicting damage to a canister due to stresses;
8. a solubility leach model for the release rate of radionuclides as a function of time, temperature, water chemistry, and radiation field; and
9. a packing material transport model for radionuclides.

It is emphasized that these component models must consider synergistic and other feedback effects. For example, a change in water chemistry could affect the corrosion rate which can change the time to mechanical failure of the waste package. The time to failure of the waste package and release rates of radionuclides result from the interaction of the individual component models. All this must be considered or else demonstrated that an assumed independence of these component models produce a conservative result.

1.4 SPECIFICATIONS FOR A MODEL/METHODOLOGY TO PREDICT REPOSITORY ENVIRONMENTAL PARAMETERS

The specifications and guidance in the NRC Draft Technical Position on waste package reliability provide a brief overview of the environmental conditions involved in the performance assessment of a waste package. More-detailed specifications for the repository environmental parameter model/methodology are developed in this report. Specifications are comprised of guidance concerning the output parameters, the subsidiary models, and the input parameters.

1.4.1 Environmental Model/Methodology Output Parameters

A large number of possible waste package degradation/failure modes were characterized to (1) identify the environmental parameters that affected each mode, and (2) to prioritize the degradation/failure modes by comparing the relevant environmental parameters with the anticipated range of the parameter. This procedure led to the development of a prioritized list of repository environmental parameters. In ranking the relevant environmental parameters in order of importance, the more critical parameter was determined based on the following considerations: (1) effects on the life of the waste package, (2) effects on radionuclide release, (3) effects of the degree of uncertainty of the values of the parameter on the performance assessment, and (4) design controllability of the values of the parameters in the engineered barrier system. The necessary output parameters from an environmental model/methodology, in decreasing order of importance, were judged to be: groundwater characteristics (including composition and flow rate), temperature, radiation, and pressure. It is emphasized that these ratings represent a subjective evaluation based on reference designs for the repositories in basalt, tuff, and salt and on the state of the art.

1.4.2 Subsidiary Environmental Models

All subsidiary environmental models should be capable of accommodating time-dependency, nonlinearities, and feedback effects, and should be adequately documented. Unless it can be shown that an alternative model is conservative or has insignificant impact on the calculated results, these models should possess the following attributes:

1. The model for water characteristics, whether calculational, experimental, or a combination of these, should
    - (a) predict pHs, redox conditions, and the concentrations of all significant species;
    - (b) account for kinetic effects;
    - (c) account for water flow through the waste package;
    - (d) predict radiation and temperature effects on water chemistry;
    - (e) allow for boiling effects; and
    - (f) account for supersaturation and colloid formation for the pertinent radionuclides.
2. The temperature model should
    - (a) possess the capability for three-dimensional calculations;

   (b) predict both the waste package and the far-field temperature histories;

   (c) account for heat conduction through multiple layers;

   (d) account for radiative heat transport calculations; and

   (e) allow for heat transport by groundwater convection.

3. The radiation transport model should

   (a) provide for determining the alpha-particle, gamma-ray, and beta-particle (probably insignificant effect except for heat generation) sources;

   (b) possess the capability for two-dimensional calculations; and

   (c) account for transport and absorption of radiation in multiple layers of different materials.

4. The pressure model should provide for determining the hydrostatic head, the lithostatic pressure, and self-induced pressures (e.g., from swelling of the packing material).

The use of alternative models that do not account for some of the aspects given above is acceptable if the applicant has demonstrated, using experimental evidence, theoretical analyses, or sensitivity analyses, that the aspect is unimportant.

1.4.3 Input Parameters for Environmental Mode

The input parameters should be probability density functions (PDFs) since the overall system model is assumed to be probabilistic in nature. A number of methods of sampling from these PDFs could be acceptable if the proper statistical methods are used and documented.

The input data base (thermophysical properties, thermal and radiation source terms, radiation transport properties, water chemistry, etc.) should be sufficient for the requirement of the models employed.

1.4.4 Validation of the Repository Environmental Parameter Model/Methodology

Validation and documentation of the repository environmental parameter model/methodology and the individual submodels contained therein are critical aspects of the model/methodology that is employed by an applicant.

Documentation of the computer codes employed must follow the guidelines in NUREG-0856 (SILLING 1983). In general, validation of a model should rely on comparison with well-conceived, well-conducted experimental results that account for all potentially pertinent effects.

This position leads to the identification of two potentially acceptable methods for validation of the environmental parameter component and subsidiary models. The first approach is to conduct large, integral experiments (laboratory or in situ) simulating the interaction of various groundwaters with a waste package under appropriate temperature, radiation, and pressure conditions. Interpretation of these integral tests will be enhanced by concurrently conducting more fundamental validation tests that address fewer parameters and processes. A second potentially acceptable approach is to employ natural analogs as a comparative basis for the results of the environmental parameter component.

2. INTRODUCTION

2.1 PURPOSE AND SCOPE

The purpose of this document is to provide specifications for models/methodologies to be employed in determining repository environmental parameters relevant to HLW package performance in repositories constructed in the basalt at the Hanford Site, Richland, Washington; the tuff in Yucca Mountain at the Nevada Test Site; and the bedded salt in Deaf Smith County, Texas. Specifically, this entails developing specifications for the following aspects:

1. guidance concerning the repository environmental parameters needed to analyze waste package performance,
2. attributes of models/methodologies that are acceptable for determining these repository environmental parameters,
3. the input necessary to determine the required output parameters, and
4. an approach to implementing the models/methodologies in the context of the overall waste package performance analysis system.

These specifications describe (1) the repository environmental parameters relevant to waste package performance considerations, (2) the considerations and effects that must be taken into account in determining them, (3) the possible use of the models/methodologies by DOE to allocate the contribution of individual waste package components to radionuclide containment and controlled release, (4) the identification of some controllable parameters, and (5) the attributes of acceptable methods for determining these parameters while avoiding a level of detail that would unduly restrict DOE's development of a suitable waste package performance analysis.

The function of the repository environmental parameter model/methodology is to determine the environmental conditions (e.g., temperature, groundwater composition) adjacent to the waste package that affects the performance of the waste package (i.e., waste package degradation and the radionuclide source term). Waste package performance is defined as the behavior of the waste package relative to its ability to meet applicable regulations (see Sect. 2.2).

The following bounding assumptions have been made in this document:

1. the technical considerations will be limited to those representative of the proposed sites for the basalt on the Hanford Reservation near Richland, Washington; the tuff of the Yucca Mountain at the Nevada Test Site; and the bedded salt in Deaf Smith County, Texas;
2. the model/methodology will consider only postclosure conditions and parameters; and
3. the most recent conceptual designs for waste packages (at the three proposed sites) that have been published are taken as reference designs.

The restriction of the model/methodology to postclosure conditions is necessary since preclosure occurrences will be heavily dependent on procedures and events occurring during the operational phase. Variability in these considerations may be handled by treating their cumulative result as the initial state in the time-dependent postclosure determination of repository environmental parameters.

In addition, no effects of highly unlikely natural catastrophic events such as major tectonic episodes, volcanic activity, or meteorite strikes are considered here. Such catastrophic occurrences can destroy a waste package by common cause (i.e., multiple barriers fail simultaneously as the result of a single event). Although the methodology to be used in assessing the overall system performance for the geologic repository after permanent closure must address such unanticipated processes and events (Sect. 60.112 of 10 CFR 60), the objective of this report is to provide the specifications for the repository environmental parameters which will be used in assessing the performance of the waste package, assuming anticipated processes and events. Consideration of catastrophic events is outside the present scope of work.

It should be noted that this report constitutes guidance in meeting the applicable regulations and criteria based on the current state of knowledge concerning waste package performance analysis. Thus, it is expected that the models/methodologies employed by DOE to show compliance with the regulations and criteria will meet, or exceed, the specifications contained herein. However, it is not to be construed that the attributes of the guidance provided in this document cannot, or should not, be modified as future developments in the DOE repository projects occur.

It is also emphasized that no attempt is made to describe a particular model or modeling approach to be used to demonstrate compliance with regulations. The choice of models and modeling approaches to be made must be dependent on waste package and repository designs, the degree of conservatism assumed, the site specific environment, and the performance allocated to the waste package that is required to satisfy the overall repository performance.

The steps used to develop the specifications for the repository environmental parameter model/methodology are as follows:

1. define the proposed repositories and waste packages in sufficient detail to permit subsequent analysis (Appendix A);

2. analyze each of the potential failure/degradation modes for the waste package to determine its effect on waste package performance, resulting in a prioritization of the failure/degradation modes (Appendix B);

3. analyze each of the failure/degradation modes having a significant impact on waste package performance to determine the repository environmental parameters that affect it (Appendix B);

4. develop a consolidated list of repository environmental parameters and prioritize them (Appendix C);

5. estimate the numerical range of each of the significant repository environmental parameters (Appendix D);
6. develop a set of assumptions concerning the nature of the system of models/methodologies and approaches that would be employed to analyze the performance of the waste package, thus providing a framework for the environmental parameter model/methodology and approach (Sect. 2.3);
7. develop specifications for the information that must result from the subject model/methodology to satisfy the needs identified in item 4 above (Sect. 3.1);
8. develop specifications for acceptable methods for determining the information in item 7 (Sect. 3.2);
9. identify the input parameters that are necessary to determine the output parameters in item 7, using the methods described in item 8 (Sect. 3.3); and
10. review the repository environmental parameter models/methodologies and approaches in the context of the specification developed under items 7 through 9 (Appendix E).

The appendixes to this report are included (1) to provide background for the main body of this report (Appendixes A through C) and (2) to provide information for use in other NRC studies and evaluations (Appendixes D and E).

2.2 REGULATORY PERSPECTIVE

Three federal agencies have major roles in the national program for disposal of HLW. The EPA has developed an environmental standard that will serve as the overall performance objective concerning releases of radionuclides from HLW disposal sites (EPA 1985). The NRC develops and issues regulations that cover all aspects of HLW disposal and will implement the EPA standard. The NRC will then consider each license application for HLW disposal to determine whether the application will conform to all applicable regulations. The DOE is primarily responsible for developing methods for disposing of HLW and has determined that national policy should focus on disposal of HLW in mined geologic repositories (DOE 1981; NWPA 1983). Furthermore, DOE is responsible for collecting the data needed for site characterization and for constructing and operating a waste disposal facility in accordance with NRC regulations.

The Nuclear Waste Policy Act of 1982 (NWPA 1983) defines the national program for disposal of civilian (commercial) nuclear waste. In the Act, it is stipulated that the NRC must issue a license before the DOE may construct a geologic repository for the HLW. In order to license a nuclear waste repository, the NRC must have reasonable assurance that the repository will comply with EPA standards (EPA 1985) and NRC technical criteria (NRC 1983).

Four portions of 10 CFR 60 (NRC 1983) are particularly relevant to environmental parameters and their relationship to waste package performance. These parameters are briefly discussed in the following sections.

2.2.1 Sect. 60.135(a)(1): HLW Package Design in General

This part particularly cautions about deleterious effects of the repository environment on HLW packages, and it is stated as follows:

> Packages for HLW shall be designed so that the in situ chemical, physical, and nuclear properties of the waste package and its interactions with the emplacement environment do not compromise the function of the waste package or the performance of the underground facility or the geologic setting.

2.2.2 Sect. 60.112: EPA Standards

The statement of this section is as follows:

> The geologic setting shall be selected and the engineered barrier system and the shafts boreholes and their seals shall be designed to assure that releases of radioactive materials to the accessible environment following permanent closure conform to such generally applicable environmental standards for radioactivity as may have been established by the Environmental Protection Agency with respect to both anticipated processes and events and unanticipated processes and events.

The waste package is a primary component of the engineered barrier system, and its performance will be directly related to the environmental conditions. The waste package could provide an additional basis (other than the geochemistry and geology of the formation) for showing that releases are sufficiently low that the EPA standards for dose rate limits in the accessible environment as prescribed in 40 CFR 191 (EPA 1985) are not exceeded.

The second relevant part of 40 CFR 191 is Sect. 191.13, which requires that reasonable expectation be provided such that specified cumulative radionuclide release limits would not be exceeded under conditions involving reasonably forseeable releases and very unlikely releases over a 10,000-year time frame. Considerations involving waste package performance may be important in two ways with respect to this requirement:

1. a waste package that can be shown to have sufficiently long containment and/or sufficiently low radionuclide release rates could meet this requirement without having to resort to other arguments (i.e., hydrology and/or geochemistry), and

2. even if DOE elects not to employ such an argument, the waste package performance defines the source term (radionuclide flux, concentration, and speciation) that is necessary for modeling radionuclide transport to the accessible environment.

2.2.3 Sect. 60.113(a)(ii)(A): Containment Criteria

This section reads as follows:

> . . . the engineered barrier system shall be designed, assuming anticipated processes and events, so that (A) containment of HLW within the waste packages will be substantially complete for a period to be determined by the Commission . . . provided that such period shall be not less than 300 years or more than 1,000 years after permanent closure of the geologic repository . . .

This portion of 10 CFR 60 specifically addresses the containment of the waste package for a 300- to 1000-year period, thus requiring that the performance be predicted with reasonable assurance over this time. It follows that knowledge of the range of the environmental parameters and effects are required with a reasonable degree of certainty.

2.2.4 Sect. 60.113(a)(ii)(B): Release Rate Criteria

The pertinent parts of this section read as follows:

> . . . the engineered barrier system shall be designed, assuming anticipated processes and events, so that (B) the release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure or other such fraction of the inventory as may be approved or specified by the Commission provided that this requirement does not apply to any radionuclide which is released at a rate less than 0.1% of the calculated total release rate limit.

Thus, the waste package performance analysis must provide reasonable assurance of compliance with this section, which will require continuation of the waste package containment analysis for times well beyond 1000 years, as well as analysis of the radioactive source that is released from the waste package to the near-field environment.

2.2.5 Relationship of Repository Environmental Parameter Determination to Waste Package Performance Assessment

Sections 2.2.1 through 2.2.4 clearly require that the performance of waste packages over a period of at least several thousand years be analyzed to show compliance with the containment and release rate criteria and to provide a source term for other near-field and far-field analyses. Consequently, the repository environmental parameters are also required since it is these parameters that influence the rate at which the waste package is degraded, the release rate after package failure, and the characteristics of the source term. Thus, the repository environmental parameters must be determined for all standards and criteria that require analysis of waste package performance, and the accuracy of the results of the analysis will depend on the quality of these parameters.

2.3 TECHNICAL PERSPECTIVE

As noted earlier, the primary purpose of this report is to provide specifications for a model/methodology to predict repository environmental parameters relevant to the assessment of waste package performance. Implicit in this purpose is the assumption that this model/methodology will acquire input parameters from some source, perform certain internal functions, and then generate output values for subsequent use in the performance assessment of a waste package. The objective of this section is to outline the assumptions concerning the portions of the waste package performance system that interface with this report.

2.3.1 Waste Package Performance Analysis

The approach to a waste package reliability analysis that has been proposed (SASTRE 1986) as an NRC technical position on the performance assessment of a waste package, which has generally become the NRC position (NRC 1986), has been adopted for this report and is briefly outlined here. The proposal discusses a general method of probabilistic reliability analyses as an acceptable framework to identify, organize, and convey the necessary information to satisfy the standard of reasonable assurance of waste package performance according to the regulatory requirements during the containment and controlled release or isolation periods. On the basis of 10 CFR 60, Stephens and co-workers (1986) also concluded that probabilistic analyses should be included in the performance assessment models used in the licensing process. In the proposed methodology by Sastre, predictions of failure probabilities are based on the application of simple phenomenological models, with the input parameters sampled from distribution functions by some probabilistic scheme.

2.3.1.1 Steps in the Proposed Approach for Evaluating the Reliability of a Waste Package

The proposed approach for evaluating the reliability of a HLW package consists of the following steps (SASTRE 1986):

1. Identifying the types of known failures that, on the basis of engineering judgment, are physically possible for the waste package for a given repository system in the sense of not violating physical laws.

2. Evaluating the available approaches and eliminating those processes which are physically possible under some circumstances but physically impossible under the repository conditions. This process is complete when all failure types previously identified are either eliminated or explicitly retained for further analysis. The reasons for elimination in each case are documented with defensible arguments and are presented in sufficient detail so as to facilitate subsequent reviews and possible reevaluations.

3. Constructing a model for each of the failure types retained for further analysis. The model describes the conditions that may lead to the failure, predicts when the failure may occur, and the immediate results of the failure. The nature of the failures, the state of knowledge, and the role of the individual failure in the overall failure of the repository dictates the level of detail required and the model uncertainty that is tolerable. This process is complete when there is a

model for each of the failure modes and the model is documented, not only from the standpoint of nominal values but also with regard to statistical uncertainty and distribution forms of the predictions.

4. Analyzing the properties that describe the environmental conditions of the repository relevant to the selected models and subsequently measuring or calculating their values. This process is complete when the links between observable and measurable properties and parameters of the repository system are identified, their values and uncertainties obtained, and their probability distributions ascertained and documented.

5. Combining the set of system properties, models, and parameters in a scheme that serves to explore all interactions modeled and predict failure probabilities. Because failures tend to be mainly due to a combination of unfavorable circumstances that may occur in nature, a scheme to predict failure probabilities such as Monte Carlo simulation would be desirable and could also be practical and acceptable. Other probabilistic schemes might be acceptable as well.

2.3.1.2 Component Models

In performing a quantitative reliability analysis of a waste package design, the applicant for a license should combine the various models for design failure modes, material properties changes, and evolution of the waste package environment into a composite or overall performance assessment model (SASTRE 1986). The component and overall assessment models must consider synergistic and other feedback effects unless it is demonstrated that neglect of these effects produces conservative results or that these effects are of no consequence.

The design of HLW packages is not sufficiently defined to permit a complete specification of the performance model, but guidelines suggested by Sastre (1986) indicate that a performance model should be composed of the following component models:

1. a temperature model capable of predicting the temperature at any point in the waste package as a function of time;

2. a heat source model capable of predicting the rate of heat generation in the waste as a function of time;

3. a radiation model capable of predicting gamma dose rates in the packing material;

4. a water flow model capable of predicting groundwater flow as a function of time;

5. a water chemistry model capable of predicting the parameters of interest such as pH, redox conditions, and salt concentrations as a function of temperature, radiation, and time;

6. a corrosion model capable of predicting corrosion rates as a function of temperature, water chemistry, and radiation dose rates;

7. a mechanical failure model capable of predicting damage to the canister due to stresses;

8. a solubility-limited release model capable of predicting release rates of radioisotopes as a function of time, temperature, and water chemistry; and

9. a packing material transport model capable of predicting concentrations of radionuclides as a function of time, water flow, temperature, water chemistry, and radiation field.

2.3.1.3 Environmental Conditions

Sastre (1986) states that in predicting the reliability of a waste package, the applicant should show the extreme range of the environmental conditions that the waste package may be subjected to throughout its lifetime. The environmental parameters were listed in the following order, without comments regarding the relative importance or possible ranges: temperature fields, groundwater chemistry (includes flow rate and quantity), radiation fields, and pressure and stress fields. Using a list of general failure modes for the waste package, it is pointed out that these environmental parameters will influence, singly or concurrently, all degradation modes of the waste package components. The relative importance of these parameters and their credible ranges are discussed in Sect. 3 and Appendixes C and D.

2.3.1.4 Numerical Data and Constants

The basic criterion being proposed for acceptance of numerical data to be used in the models for waste package reliability analyses is the reproducibility of the results for both experimental and derived data (i.e., other qualified practitioners can produce essentially the same results, based on the documentation of the experimental methods and data sources). All constants and parameters derived from experimental measurements that are used in the analysis should be presented with an estimate of the error, or confidence interval, along with an estimate of the expected error distribution when the data have uncertainties greater than a few percent. Data in the form of plots for model justification without accompanying tabulations of the numerical values and references to reports are not acceptable (SASTRE 1986).

2.3.1.5 Performance Model and Quantitative Reliability Analysis

A waste package performance model will be composed of component models that address the basic functions, or processes, involved in the degradation and subsequent failure of the components of the waste package. The acceptability of the performance model depends on the completeness with which the individual component model describes all phenomena of importance and their interactions, and the successful validation of the model. The applicant should supply predictive equations or equivalent (SASTRE 1986) for each failure mode (see Appendix B for discussions of possible failure modes) and for each basic process determining the evaluation of the environmental conditions and material property changes. The use of these predictive equations in a computer model (e.g., the WAPPA code as described in Appendix E) that determines the overall reliability of a waste package by analyzing the behavior of the individual waste package or barrier components (i.e., waste form, canister, overpack, and packing) acting as a system should permit allocation of the contribution of each barrier component to the containment or controlled release of radionuclides. This procedure will assist in evaluating the effectiveness of the individual components of the engineered barriers. Such evaluations in the early stages of the licensing processes should be useful in guiding research and development and final designs. By using the performance model and the statistically sampled variables that represent the data and the uncertainty of each component model, the applicant should be able to determine the probability distribution of the times to containment failure and the release of radionuclides in comparison with the standard (SASTRE 1986).

3. SPECIFICATIONS FOR A MODEL/METHODOLOGY TO PREDICT REPOSITORY ENVIRONMENTAL PARAMETERS AFFECTING WASTE PACKAGE PERFORMANCE

In the performance assessment of a nuclear waste repository by an applicant, a model is required that can demonstrate reasonable assurance by quantitatively predicting the release of radionuclides to the accessible environment. In the overall system assessment, the release of radioactivity from the waste package represents the source term for the model describing the transport of radionuclides successively through the repository, the geologic medium, and the accessible environment. Consequently, the integrity of the waste package becomes of paramount importance in meeting the performance objectives of the proposed regulations. Radionuclide containment by the waste package is to be substantially complete for a period of not less than 300 years or more than 1000 years. An isolation period, in which the fractional release rate is less than 10^{-5} per year (based on the inventory present at 1000 years) from the engineered barrier system, follows the containment period.

As discussed earlier in Sect. 2.3, some of the component models/methodologies necessary to implement the waste package performance assessment are concerned with identifying those repository environmental parameters which affect the performance of the waste package. The specifications and guidance that are in the proposed NRC technical position on waste package reliability (SASTRE 1986) generally cover the environmental conditions involved in the performance assessment of a waste package (Sect. 2.3.1.3). This section is concerned with the more-detailed specifications for the repository environmental parameter model/methodology. These component model specifications are comprised of specifications for the output parameters, the subsidiary models, and the input parameters.

3.1 REPOSITORY ENVIRONMENTAL MODEL/METHODOLOGY OUTPUT PARAMETERS

A qualitative evaluation involving, in most cases, the judgment of the current state of the art was undertaken to determine the output parameters that must result from the repository environmental parameter model. Many possible waste package degradation/failure modes were characterized (1) to identify the repository environmental parameters that affected each, and (2) to prioritize the degradation/failure modes by comparing the relevant environmental conditions with the anticipated credible ranges of the parameters (see Appendix D for discussion and rationale) in the proposed repositories.

The term "credible range" was used in place of "expected range" since the expected range is usually thought of in terms of deviations from the norm, whereas "credible range" is not likely to be confused with other conventional terminology. Here, we use "credible range" as an imprecisely defined quantity that is meant to include deviations from the norm, uncertainties, future small design changes, and other local perturbations that could reasonably occur in a sealed repository barring unusual tectonic activity or other catastrophic events. Admittedly, defining a credible range is somewhat subjective and must be based on assumptions that are subject to change. However, the authors believe that the assigned credible ranges represent a reasonable attempt to limit the possible range of the environmental parameters, based on current published information. The intent is certainly not to set absolute limits, and it is recognized that arguments can be made for changing these values based on other assumptions or subjective viewpoints. Nevertheless, they can serve as guidelines in three important ways: (1) to limit the range of environmental parameters for use in preliminary waste package performance analyses, (2) to limit the range of these parameters in R&D work, and (3) to provide a "feel" for the physical realities involved in the performance of a waste package in a repository.

Individual failure modes are discussed in Appendix B and rated as to whether the particular failure mode could be dominant, significant, insignificant, or unknown (i.e., the answer to some critical question is required before a rating can be determined). It is emphasized that these ratings represent an evaluation based on the reference design and the state of the art. Consequently, it does not follow that all the possible failure modes are very probable; however, they do represent a rather exhaustive list of possibilities that were considered in determining the environmental parameters requiring consideration. While this evaluation should not be construed to be an NRC position, it is in line with the suggested procedure for waste package reliability analysis by Sastre (1986), which is quoted below:

> In the SAR, the applicant should list all possible, identified failure modes of each waste package component and their retention or dismissal for further analysis. This preliminary analysis, generally called Failure Mode and Effects Analysis — FMEA, is qualitative in nature. It is expected to result in the reduction of the set of possible failure modes to only those which are relevant under the range of repository conditions identified in Section 2.1.2 [of the Sastre report and in Appendix C of this report]. This set of significant failure modes will be called design failure modes. . . . The dismissal of any given failure mode should be discussed and documented.

Consideration of a consolidated list of environmental parameters with respect to the prioritized degradation/failure modes yielded a prioritized list of repository environmental conditions that have a significant impact on waste package performance (see Appendix C for discussion and rationale). In ranking these parameters in order of importance, the more critical parameter was determined based on the following considerations: (1) effects on the life of the waste package, (2) effects on radionuclide release, (3) effects of the degree of uncertainty of the values of the parameters on the performance assessment of the waste package, and (4) design controllability of the values of the parameters in the engineered barrier system. This analysis produced the following list of repository environmental parameters, in decreasing order of importance: groundwater characteristics, temperature, radiation, and pressure. Each of these parameters is discussed in detail in the following section.

3.1.1 Groundwater Characteristics

The groundwater characteristics that must be determined by the repository environmental parameter model/methodology are: (1) concentration of all significant dissolved species, (2) pH, (3) redox conditions, and (4) flow rate.

These output parameters may be based on calculations with a model/methodology using input values obtained from PDFs or by sampling from bounding experimental results.

3.1.2 Temperature

The temperature of each waste package material should be determined by a repository environmental parameter model/methodology that has been verified and validated.

3.1.3 Radiation Levels

The radiation level in each waste package component and the immediate surrounding should be determined by a repository environmental parameter model/methodology that has been verified and validated.

3.1.4 Pressure

The pressure exerted on, or by, all waste package components must be determined via a repository environmental parameter model/methodology. This includes pressures resulting from hydrostatic head, lithostatic head, and swelling of materials. This pressure becomes an input to a stress model for the reliability analysis of a waste package as mentioned in Sect. 2.1.3.2.

3.1.5 Dimensionality

3.1.5.1 Spatial

Analysis indicates that, in general, a two-dimensional representation of the above data will be required; an R-Z geometry is preferred. This representation will allow any effects resulting from parameter differences occurring radially or axially to be subsequently taken into account. More complex three-dimensional representations do not appear to be necessary, except possibly for some heat transport calculations. Lower-order dimension models are acceptable if the applicant can show that the results are conservative or that the dimensional effects and any potential adverse effects resulting from parameter differences are small.

3.1.5.2 Time

The output parameters must be given as a function of time over a span of at least 10,000 years. The methodology may be based on analytical solutions, numerical solutions with time incremented as an independent variable, or combinations of these. Time dependency for some conditions may be ignored if the applicant can demonstrate that the procedure is conservative or has no significant effect on the performance analysis.

3.1.6 Design Controllability of Output Parameters

The environmental conditions, which are the output parameters of the environmental model, are controllable to a reasonable degree by site selection and design that are based on laboratory data and in situ tests.

While the characteristics of the undisturbed groundwater are obviously not controllable environmental parameters, the components (except perhaps the redox condition) are measurable. The regional hydrology of the disposal site will control the migration rate of any radionuclide in the far field and will influence groundwater flow inside and around the waste package. The difficulty arises in predicting the long-term characteristics of the groundwater after alteration because of its interaction with the waste package. For a detailed discussion of these difficulties and the subsequent uncertainties, see Davis (1983).

The design control of temperature seems straightforward since the canister and areal thermal loadings can be adjusted to limit temperatures to the desired values based on predictions using validated heat-transfer models. In addition, these temperature predictions for the very near field or near field can be confirmed by measurements in a few years or less after waste emplacement prior to repository sealing — a condition that will permit remedial action if necessary.

The gamma radiation field external to a canister can be controlled by the canister design and loading and may be easily and accurately validated by measurements prior to and after emplacement. The alpha-particle source in the waste form should be calculable with reasonable accuracy if the actinide loading of the canister is known.

Hydrostatic pressures that develop with time after repository sealing in basalt (which normally cannot develop in salt or tuff) will depend on the depth of the repository horizon. Swelling pressures that develop from bentonite in packaging material can be controlled with the compaction density or percentage of bentonite in the packing materials. See Sect. 3.3.5 for a discussion of pressure development in a repository.

3.2 REPOSITORY ENVIRONMENTAL PARAMETER MODEL/METHODOLOGY

This section discusses attributes of the models/methodologies that might be used to determine the output parameters described in the previous section. Two general approaches are discussed, the calculational and experimental approaches. These general approaches represent methodology limits. It is more probable and desirable that an acceptable approach will be a combination of these two extremes.

3.2.1 Groundwater Chemistry

3.2.1.1 Calculational Approach

Conceptually, it is possible to calculate all of the output parameters related to groundwater or brine chemistry in an acceptable manner (except for the flow rate) by using thermodynamic and kinetic considerations. Thermodynamic data supply the information necessary to determine the equilibrium state of the system under consideration, and the kinetic data define the rate at which this equilibrium is approached. However, currently available groundwater chemistry models do not appear to be adequate for this task, particularly in the case of the saturated brines to be expected in a salt repository. The following are some of the attributes that are necessary for a calculational approach to groundwater chemistry to be acceptable, unless additional knowledge can be developed to show that the effects they take into account are unimportant or conservative with respect to performance assessment:

1. completeness of the thermodynamic data bases with respect to many radionuclides and engineered materials (especially at elevated temperatures);

2. ability of the models to account for kinetic effects;

3. ability of the models to account for the effects of water flow through the waste package and convection/mixing within the package;

4. ability to account for the effects of alpha particles, gamma rays, and beta rays (which are probably of no significance) on the groundwater chemistry;

5. ability to account for the effects of boiling or vaporization of water on the groundwater or brine chemistry, which will occur initially in a tuff repository above the water table and may occur during the resaturation/repressurization period in a basalt repository or the early times after emplacement in a salt repository; and

6. ability of the models to account for supersaturation, colloids, etc.

Additionally, it should be noted that this approach would require some type of comparison with an integral experiment or natural analog for validation purposes.

3.2.1.2 Experimentally Based Approach

In an experimentally based approach, the values input to the repository environmental parameter component would be the result of experiments conducted at some earlier time and specially designed to provide information concerning groundwater or brine (in the case of salt repositories) chemistry in the vicinity of the waste package. These data could be in a number of discrete sets, with each set having an associated probability or organized so as to eliminate the possibility of violating physical laws.

In this case, the repository environmental parameters obtained by the sampling methodology would be simply passed through to the waste package degradation/failure component for subsequent use. However, if this approach is selected, a method must be devised to accommodate the feedback effects on the groundwater or brine chemistry unless they can be shown to be negligible. This approach might be accomplished with a validated calculational methodology as described above, but other methodologies could be used (e.g., semi-empirical approaches) as long as their validity can be demonstrated.

3.2.2 Temperature

The heat-transport model of the repository environmental parameter component should possess the following attributes unless it can be shown that an alternative model has an insignificant impact on the calculated results:

1. capability for three-dimensional calculations;

2. provisions for modeling the very near field (includes waste, overpack, packing, and nearby rock temperatures), the near field (nearby rock backfill and room pillars), and the far field (shaft temperatures and the vicinity beyond the repository) and a method interfacing the models;

3. provisions for modeling the conduction of heat through multiple layers (primary mechanism of heat transport);

4. provisions for radiative heat transport (may be important between waste package layers); and

5. provisions for accounting for heat transport via groundwater convection, both within the waste package and in the far field.

It is assumed that the time dependence of the case will be handled by either analytical or numerical methods and that feedback effects will be handled by adjusting relevant dimensions and thermal properties as required.

3.2.3 Radiation

The subsidiary radiation transport model of the repository environmental parameter component should possess the following attributes unless it can be shown that an alternative model has an insignificant impact on the calculated results:

1. provisions for determining (obtaining) the alpha-particle, gamma-ray, and beta-particle radiation source as a function of energy (defines input to attenuation calculation);

2. capability for two-dimensional calculations; and

3. provisions for modeling the transport and absorption of radiation in multiple layers of different materials (necessary to account for the expected multilayer waste package).

Discrete-ordinates radiation transport codes, such as DOT (RHOADES 1973) and ANISN (ENGEL 1967), are generally acceptable in this application. Approximate methods (buildup factors, etc.) can also be used if they can be shown to produce sufficiently accurate or conservative results.

3.2.4 Pressure

The pressure model in the repository environmental parameter component should possess the following attributes unless it can be shown that an alternative model (or ignoring a particular effect) is conservative or has an insignificant impact on the results:

1. provisions for determining pressure resulting from hydrostatic head in basalt (hydrostatic heads will not develop in the salt and tuff repositories);

2. provisions for determining pressure resulting from lithostatic pressure, which is a long-range possibility in the case of basalt and tuff but a relatively short-range possibility for salt because of its creep characteristics;

3. capability for determining self-induced pressures (e.g., packing material swelling and thermal expansion of the various materials); and

4. capability for determining pressure distributions in two dimensions.

3.3 INPUT PARAMETERS

This section provides guidance on those independent parameters which constitute the input to the repository environmental parameter model/methodology to determine the outputs described earlier.

3.3.1 Character of Input Parameters

As is evident from the previous discussion, the input parameters for the environmental parameter model/methodology are assumed to be PDFs since the overall system model is considered to be probabilistic in nature. It is recognized that establishing these PDFs may have large attendant uncertainties. The following paragraphs discuss some aspects of these uncertainties and acceptable ways to handle them.

3.3.1.1 Justification of PDFs

It is expected that establishing PDFs using generally accepted methods that require large numbers of samples or trials may be impossible in cases involving physical data. However, the use of assigned PDFs is generally acceptable as long as (1) a rationale is presented for the PDF (shape and range), and (2) some sensitivity analyses have been performed to ensure that results are not inordinately affected by the PDF shape employed. Since the uncertainties in many of the input parameters are expected to increase with time, the time dependence of the PDFs will need to be taken into account.

The use of single-value input data is acceptable if it has been previously shown via techniques such as sensitivity analysis that the variation of a parameter within its credible range has a small impact on the results of the waste package performance analysis.

3.3.1.2 Level of Model Definition

The model/methodology developer is permitted considerable flexibility in the structure of the waste performance systems analysis model/methodology for the purposes of computational efficiency or convenience. As a result, the input parameters to the repository environmental parameter component of the model may vary, depending on the scope of the subsidiary models. For example, the three-dimensional heat transfer problem might be reduced to two coupled, one-dimensional cases encompassing a far-field/near-field calculation and a very-near-field calculation, with the former supplying the temperature at the boundary between the models (i.e., the interface between the waste package and the surrounding rock).

Such methods can be acceptable within the following limitations: (1) sampling from independent input parameter PDFs must not yield a set of input values that violate physical laws (e.g., an input pH must correspond to the input temperature), and (2) the effects of calculated changes in waste package characteristics must have a negligible impact on any modeling performed separately from the package performance analysis (e.g., feedback effects on a separate calculation of near-field/far-field heat transport must not be significant).

3.3.1.3 Independence

If the PDFs cannot be shown to be independent from other input parameters, then special measures must be taken to accommodate the dependence. Alternatively, as discussed in Sect. 3.3.1.2, the applicant may elect to define the input parameters at a level such that they are independent and include a model to account for the dependence in the environmental parameter component.

3.3.1.4 Time at Which Input Parameters are Defined

In general, it is assumed that all of the input parameters will represent the initial state of the system, including any changes that have occurred during packaging, emplacement, and retrievable storage. It should be recognized that as the performance analysis model/methodology proceeds, some of these parameters will be altered (e.g., dimensions, chemical composition) while others will remain fixed (e.g., thermal conductivity of a material at a particular temperature).

3.3.2 Groundwater Chemistry

As mentioned in Sect. 3.2.1, there are two general approaches to determining the groundwater chemistry in and around the waste package: calculational and experimental. It is doubtful that the calculational approach is usable for the saturated brines that are expected in salt repositories. The input information that would be needed for the calculational approach includes:

1. thermodynamic data (e.g., free energies and enthalpies of formation, activity coefficients) for all significant radionuclide and chemical species inside and around the waste package;

2. parameters related to the rate at which the various possible reactions between groundwater constituents, waste package constituents, and the host rock/secondary minerals proceed (i.e., data on kinetics);

3. the unperturbed groundwater composition and the groundwater flow rate into the waste package;

4. composition of all materials inside and around the waste package;

5. the temperature inside and around the waste package; and

6. some information concerning the formation rate and stability of colloids, supersaturated conditions, and so forth.

If the experimentally based approach is employed, then the input parameters are sets of the following information as a function of space, time, and probability of occurrence: (1) concentrations of all significant dissolved species in the groundwater, (2) pH, (3) redox conditions, and (4) groundwater flow rate.

Additionally, some type of information will likely have to be supplied to allow the parameters listed above to be adjusted to compensate for feedback effects.

The calculational and experimental approaches represent methodology limits. It is more probable and desirable that an acceptable approach will be a combination of these two extremes.

3.3.3 Temperature

In general, there are two major aspects to determining temperatures inside and around the waste package: a near-field heat transport calculation to determine the temperature at the waste package-host rock interface, and the very-near-field heat transport calculation that determines the temperature distribution in detail inside and around the waste package. The input parameters for the far-field model would be as follows:

1. thicknesses of the rock layers inside and around the repository;

2. engineering design data for the repository (emplacement hole diameter, pillar thicknesses, disposal room dimensions);

3. diameter of the overpack and the packing material;

4. thermophysical properties (heat capacity, density, and thermal conductivity) for the homogenized waste package (through the overpack), packing material, and rock layers;

5. the rate at which the waste form is generating heat; and

6. parameters (e.g., volumetric flow rates) pertinent to the calculation of convective heat transport in the far field.

The input parameters for the waste package temperature determination would be as follows:

1. dimensions of all of the waste package components (lengths, thicknesses, etc.), including gap thicknesses between layers;

2. thermophysical properties of all of the waste package components, including heat capacities, densities, thermal conductivities, and emissivities;

3. the rate at which the waste form is producing heat and the distribution of the heat source; and

4. parameters related to the calculation of convective heat transport immediately around the waste package if sufficient water is present.

3.3.4 Radiation

If a radiation transport model employs a relatively sophisticated methodology such as a discrete-ordinates calculation, then the input needed would be as follows:

1. the source term gamma rays (alpha and beta particles can, for all practical purposes, be absorbed at the point of assumed emission);

2. the multigroup energy spectrum of the gamma rays;

3. the compositions and dimensions of all of the waste package materials and the immediately surrounding host rock; and

4. multigroup gamma-ray attenuation and absorption cross sections for all of the elements present in significant concentrations.

The use of the older "shielding handbook" approach or buildup factor method that involves a ray-tracing technique (ROCKWELL 1956) would require the same general types of information except that (1) considerably fewer cross sections would be required, and (2) buildup factors (or equivalent) would be needed to account for scattered photons.

3.3.5 Pressure

The pressure history around a waste package in a repository will depend primarily upon the rock type and swelling pressures that develop from expandable clays if used in the packing. In the case of basalt, pressures on an overpacked waste canister can be expected to vary from atmospheric to lithostatic (see Sect. A.1.1 for a description of the current designs for the waste package and BWIP repository). After the sealing of a repository, water will gradually fill the void spaces and the pressure exerted will increase to hydrostatic in a few to hundreds of years, depending on the specific site. This hydrostatic pressure (or head) will depend on the repository depth and the height of the water table above the disposal horizon, an easily measurable quantity. The rate of rise of the hydrostatic pressure, however, seems to be an uncontrollable parameter that is not easily determined. Development of full lithostatic pressure because of rock creep or subsidence is unlikely for several thousand years, barring unexpected tectonic activity; however, pressure around the canister can increase because of the swelling pressure exerted by the bentonite. In addition, unlike repositories in tuff and salt, the undisturbed in situ rock stress at the basalt site is highly anisotropic; the ratio of the horizontal to the vertical stress is about 2.7. This condition could create a horizontal shearing force on the waste package before full lithostatic pressure is attained. Pressures exerted by the rock that are significantly above lithostatic cannot exist because the rock will fracture and relieve the stresses. The swelling pressure from the bentonite or any additional stresses that may arise due to thermal expansion and rock fracturing should be controlled via design.

The pressures are expected to remain at approximately atmospheric in a tuff repository for a long period of time since the site at Yucca Mountain is above the water table and a hydrostatic head cannot develop (see Sect. A.2.1 for a description of the previously published designs of the waste package and the repository). Tuff, which is a hard rock, will resist room closure and, as in the case of basalt, the full lithostatic pressure may take thousands of years to develop. Swelling pressures from the packing can develop if bentonite is used in the packing, which is an alternate design.

Unlike the other rocks, a salt repository can be expected to attain full lithostatic pressure in tens of years depending on the creep properties of the salt (see Sect. A.3.1 for a description of the waste package and the repository). It is possible, however, that pressures on the waste package that are above lithostatic could develop in the first few years because of thermal expansion of the surrounding salt bed and, consequently, this should be considered even though it is expected that salt creep will alleviate these expansion effects. Since groundwater is not present in a salt formation, a hydrostatic head cannot develop, barring an unanticipated tectonic event that creates a vertical opening that connects with an overlying aquifer.

To account for these possible scenarios and their potential effects, the minimum requirements for input data for the environmental model/methodology are:

1. hydrostatic head changes with time,

2. swelling pressure changes with time,

3. rates of development of the horizontal and vertical components of the local lithostatic pressure, and

4. rock tensile and compressive strength in horizontal and vertical directions.

3.4 OPERATION OF REPOSITORY ENVIRONMENTAL PARAMETER MODEL/METHODOLOGY

Several important aspects of the approach to operations performed by the repository environmental parameter model/methodology are discussed below.

3.4.1 Nature of Models

As discussed previously, it is assumed that the probabilistic nature of the overall model/methodology results from the use of PDFs as input and the accumulation of statistical output.

Any type of model (theoretical, empirical, or semi-empirical) can be employed at the developer's discretion, subject to the other limitations described herein. In this regard, Sect. 3.5 should be noted.

3.4.2 Scope

In parallel with the discussion on the level of input data, the scope (near field, far field, etc.) of the subsidiary models is at the developer's discretion, subject to the model's capability for accommodating feedback effects. Interdependence between the individual models and between the components of the waste package performance analysis are assumed to be handled by using appropriate coupling techniques.

3.4.3 Use of Experimental Results

It is generally acceptable to produce experimentally based output parameters in lieu of calculated output parameters; however, certain precautions must be taken. The input parameters constituting the experimental data must (in general) be expressed as PDFs while not violating physical laws. This procedure might be handled by developing several consistent sets of data (e.g., chemical compositions) and assigning each a probability of occurrence. Finally, if feedback effects are important, then the applicant must provide a defensible method for accounting for the effects of the feedback on the input data obtained by sampling the experimentally based PDFs.

3.5 VALIDATION OF THE REPOSITORY ENVIRONMENTAL PARAMETER MODEL/METHODOLOGY

Validation and documentation of the repository environmental parameter model/methodology and the individual models contained therein are critical aspects of the approach to this model/methodology. It is strongly urged that the validation strategy be made a part of the initial development of the environmental parameter model/methodology and its subsidiary models in order to ensure an acceptable result.

3.5.1 Documentation Guidelines

Guidelines have been proposed (SILLING 1983) for the documentation of models and computer codes used in support of licensing activities.

A brief general outline of these guidelines is given below.

1. Software Summary - This summary is a one-page document that identifies the code and version number and provides other basic information.

2. Mathematical Models and Numerical Methods - This work includes a derivation and justification for the model, an explanation of its capabilities and limitations, and a description of the numerical solution strategy, stability, accuracy, and computational sequence.

3. User's Manual — The user's manual, together with the code listing, should be sufficient to instruct the user on how to set up and run problems as well as to resolve possible difficulties.

4. Model Assessment and Support — This document must discuss all work that sheds light on the adequacy of the model, provide evidence that the model has been extensively reviewed and validated, and describe the quality assurance and maintenance programs for the computer code.

5. Continuing Documentation and Code Listings — Continuing documentation includes the development of new capabilities, the detection of repair of errors, and application-oriented modifications.

3.5.2 Methods of Validation

In general, the validation of a model should rely on a comparison with well-conceived, well-conducted experimental results that account for all potentially pertinent effects. The validation of complex calculational results with other calculated results or with simple experiments that only account for one process or effect may not be acceptable. All computer codes written to describe the physical model must be verified with regard to computational accuracy and capability of representing the physical model.

Two potentially acceptable methods for validating the environmental parameter component and subsidiary models can be identified. The first approach is to conduct large, integral experiments (laboratory or in situ) simulating the interaction of various groundwaters with a waste package under appropriate temperature, radiation, and pressure conditions. These integral experiments need to be supported by more fundamental tests that address fewer variables and processes. Otherwise, the results of the integral tests may be difficult to interpret in a meaningful manner. A second potentially acceptable validation approach is to employ natural analogs as a comparison basis for the results of the environmental parameter component. While this provides a more realistic geologic environment than the integral test, it suffers in that many of the conditions expected in the repository (radiation, elevated temperatures, engineered materials) have not been present.

3.5.3 Existing Validation

Subsidiary models have already been developed, documented, and validated for some of the repository environmental parameters such as temperature and radiation transport. If validation of the existing models can be demonstrated by reference in these cases, additional documentation and validation will not be required. It is acceptable to augment existing documentation which lacks some of the required information (SILLING 1983). However, it should be noted that validation concerning accommodating feedback effects and interfaces with other models will still be required.

3.6 FEEDBACK EFFECTS AND OTHER NONLINEARITIES

This section is added to emphasize the importance of "feedback" and other nonlinearities (such as temperature dependence of material properties and synergistic effects) in the performance assessment of a waste package. Feedback and temperature dependence of some parameters cause nonlinearity in the system model, which can make analytical solutions (closed form) extremely difficult or impossible and can considerably increase the computational time for numerical solutions.

The word "feedback" is used in this document to denote the fact that, as the waste package components are affected by their environments, both the components and the environmental conditions are altered, subsequently affecting the repository environment. For example, if it is postulated that a carbon steel overpack is degraded by being oxidized, the product of this process might be a loose layer of iron oxides. The presence of these oxides would change the dimensions and thermal conductivity of the package, which would affect the temperatures inside and in the immediate area surrounding the waste package. In addition, the formation of the iron oxides, coupled with the changes in temperature, will alter the water composition. The obvious method for taking these effects into account is to adjust the appropriate parameters and data during the calculation to reflect the changes that have just been calculated to occur (feedback), although other methods are not precluded. The problems can be linearized when the effect of neglecting nonlinearities can be shown to be insignificant or conservative.

4. REFERENCES

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5. APPENDIXES

APPENDIX A

DESCRIPTION OF WASTE PACKAGE AND REPOSITORY ENVIRONMENT

The time-to-failure of a waste package (i.e., a breach that permits contact between the waste form and ambient water) and its subsequent behavior in limiting or controlling the release of radionuclides are dependent on the waste package and repository designs and the environment in the immediate vicinity. This environment is both strongly site- and design-dependent and changes with time. Thus, many combinations of waste package designs, repository types and designs, and radionuclide release scenarios may ultimately have to be considered. It is anticipated that the model/methodology specifications developed by this project will be sufficiently general to be adaptable to many designs and degradation models and repository types.

The failure scenario that is considered is the postclosure degradation of the waste package as the result of anticipated, long-term processes, which is consistent with the requirements of 10 CFR 60 (see Sects. 2.2.2 and 2.2.3). No effects of highly unlikely natural catastrophic events (e.g., major tectonic episodes, volcanic activity, or meteorite strikes) are considered here. Occurrences of this nature can destroy a waste package by common cause (i.e., multiple barriers fail simultaneously as the result of a single event). These events are not considered here because they are unanticipated processes and events as defined in 10 CFR 60, whereas Sect. 60.113(a)(1) only addresses the performance requirements of engineered barriers assuming anticipated processes and events. If these events are not truly catastrophic, they can be factored into a waste package degradation model by parameter changes, such as increasing the water flow and pressure on the waste package.

Potential repository sites have been selected by DOE for detailed characterization in the basalt at Hanford, Washington; in the tuff at Yucca Mountain, Nevada; and in the bedded salt in Deaf Smith County, Texas. Some aspects of the current conceptual designs and degradation and radionuclide release scenarios are discussed in the following sections of the appendix. An earlier report (CLAIBORNE 1985) discussed the basalt repository only. This report includes discussions of the salt and tuff repositories and the current basalt repository design.

A.1 NUCLEAR WASTE REPOSITORY IN BASALT

The reference repository is located near the center of the DOE-controlled Hanford reservation in the south-central portion of the state of Washington. This area lies within the Pasco Basin, the structural and topographical low part of the Columbia Plateau (DOE 1984a, 1986a).

A.1.1 Repository Design and Initial Conditions

The repository horizon is currently planned to be around 975 m deep in the Cohassett basalt flow in the Sentinel Bluffs Sequence of the Grande Ronde Formation on the Hanford Reservation. The earlier 1982 reference conceptual design (RKE/PB 1983) placed the repository horizon in the Umtanum flow, nearly 200 m lower than the disposal horizon in the Cohassett. Intraflow structures exist that can be simply classified as fractured and jointed, with varying degrees of secondary mineralization present. Higher-porosity interbeds, which are sources of water (GUZOWSKI 1983) also exist. The potential sources of groundwater for contacting the waste canisters are the unconfined aquifer in the sediments lying above the sequence of basalt flows and the numerous confined aquifers at the greater depths within the basalt flows. There are three dominant pathways for groundwater movement in the basalt: (1) breccia zones between basalt flows and in the sedimentary interbeds; (2) vertical faults and fracture zones between adjacent basalt flows; and (3) stratigraphic discontinuities within the basalt flows. The shallow basalts are thought to recharge in outcrop areas and to discharge to the overlying unconfined aquifer and the Columbia River. The deeper basalts appear to be recharged from interbasin groundwater movement originating northeast and northwest of the Pasco Basin. The discharge location is not known but is speculated to be south of the Hanford Site (DOE 1984a, 1986a).

In view of this geologic structure, the available water, and the potential perturbations of the hydrologic system, it had been commonly accepted that a repository in basalt would "fill" or resaturate with water after closure within tens to a few hundreds of years. However, in a study by Pruess (1982) for a repository in the Umtanum, he estimated that the host rock will completely resaturate before the end of the 50-year open period and prior to backfilling. Resaturation of the repository after backfilling would be less than 2 years based on the most probable values of the pertinent rock parameters.

The repository is to be of the conventional pillar-and-room mine design that will facilitate multiple waste package emplacement [both commercial high-level waste (CHLW) and spent fuel (SF)] in horizontal boreholes contained in the pillars. Based on the available information at the time that the previous publication on the relevance of repository environmental parameters in assessing waste package performance was prepared (CLAIBORNE 1985), the reference design consisted of long horizontal boreholes containing several waste packages (see Fig. A.1). The backfill material around the overpacked waste canister, henceforth called the packing material (although technically a part of the waste package), was not to be emplaced simultaneously with the overpacked waste canister. Instead, it was to be pneumatically distributed in the emplacement hole around the overpacked canister prior to repository decommissioning. The operating period is defined by BWIP as 90 years, divided into three phases: emplacement, during which all emplacement operations take place (20 years); observation (50 years); and either backfilling-decommissioning (20 years) or retrieval (20 years).

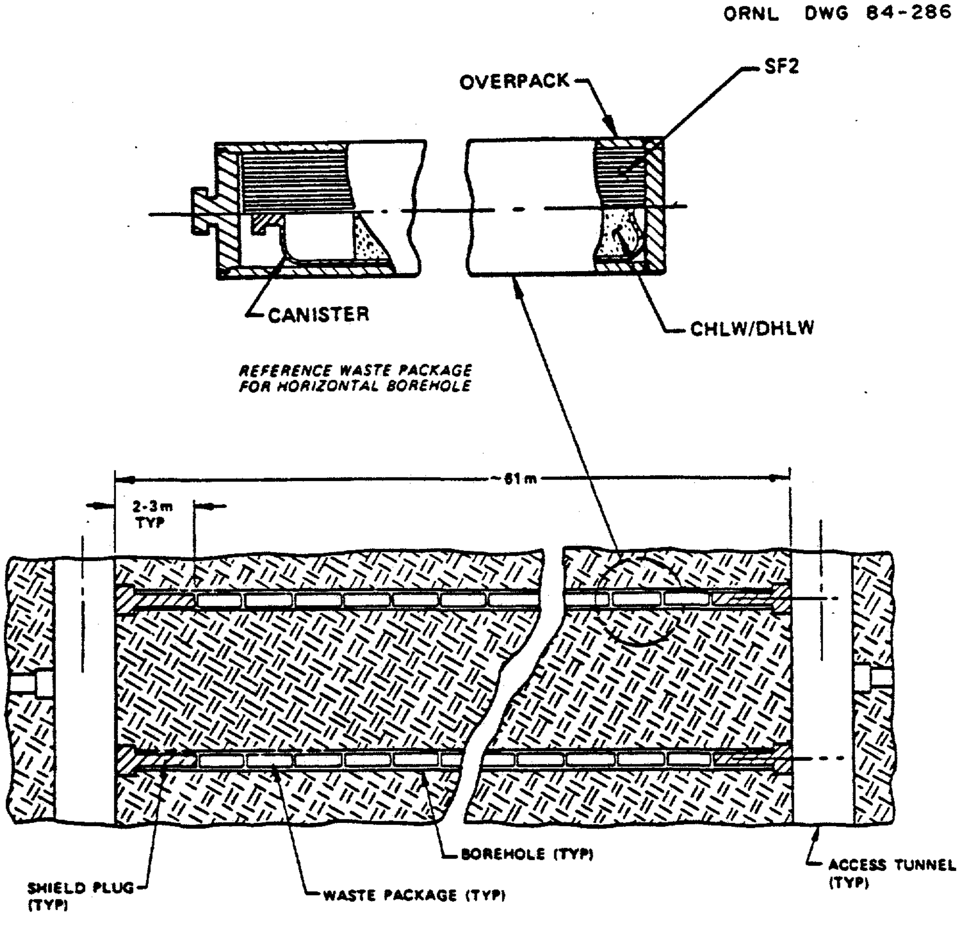

Fig. A.1. Reference package and emplacement scheme for the long horizontal borehole concept. Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Waste Package Conceptual Designs for a Nuclear Repository in Basalt, RHO-BW-CR-136P/AESD-TM-3142, 1982.

By the time the report was issued, the reference conceptual design had been changed to the short horizontal borehole (SHB) concept (see Fig. A.2). The long horizontal borehole concept and the pipe-in-tunnel (PIT) concept, which involves waste canisters placed in horizontal pipes within a disposal room, became potential alternative designs (HENRY 1984). In the SHB concept, the boreholes extend from the walls of the emplacement room and each contains only one waste package (McCALL 1986). Design studies involving these concepts with many variations are available (WEC 1984 and RKE/PB 1983).

Design studies are continuing, and changes can be expected to occur in proposed designs prior to a license application. Actually, an updated conceptual design of the repository that incorporates all of the current functional requirements and criteria will only be completed in 1987. Currently, it is assumed that construction of a repository will proceed in two stages. Stage I provides construction sufficient for acceptance of the equivalent of 400 metric tons of heavy metal per year (MTHM/year) for five years beginning in 1998. In Stage II, the remaining facilities are to be constructed for acceptance of 500 MTHM starting in the year 2001, 1400 MTHM in 2002, and the full operating capacity of 3000 MTHM/year starting in 2003, for a total of 70,000 MTHM in the form of SF and CHLW and, if required, defense high-level waste (DHLW).

During Stage I operations, spent fuel assemblies from both BWRs and PWRs will be received and packaged without disassembly into carbon-steel containers. In Stage II operations, spent fuel elements that are received will be disassembled and consolidated prior to packaging into carbon-steel containers. Any CHLW (or DHLW, if received) will be shipped to the site where it will be overpacked with carbon-steel containers. The waste containers will be individually transported and emplaced in the short horizontal boreholes (one package per hole) at the repository horizon. At least 10-year-old spent fuel will be initially received, but spent fuel five years out of the reactor may also be received. In either case, the same waste containers can be used by limiting the thermal loading to 2.2 kW.

The temperature history will depend on the areal thermal loading, the rate of canister emplacement, and the time interval prior to backfilling and final closure and decommissioning of the repository. The ambient temperature of the undisturbed rock is 51°C at the repository horizon. Calculated maximum rock temperatures are significantly lower for sequential loading (RICKERTSEN 1982) as compared with those assumed for instantaneous loading. Depending on the detailed arrangement, the ventilation system could remove a large amount of the heat liberated (SAI 1977). The estimated BWIP peak temperatures and design limits for each component of the waste package are discussed later in Sect. D.1.3.

The pressure exerted on the canister will be atmospheric prior to backfilling. Although slight local increases can be observed after backfilling, significant increases cannot occur until after decommissioning and water begins to accumulate. Except for (possibly) some local thermal stresses and swelling pressure exerted by the packing, which could be controlled by proper design, the maximum pressure should not greatly exceed hydrostatic (~11 MPa) in the first few thousand years. For a properly

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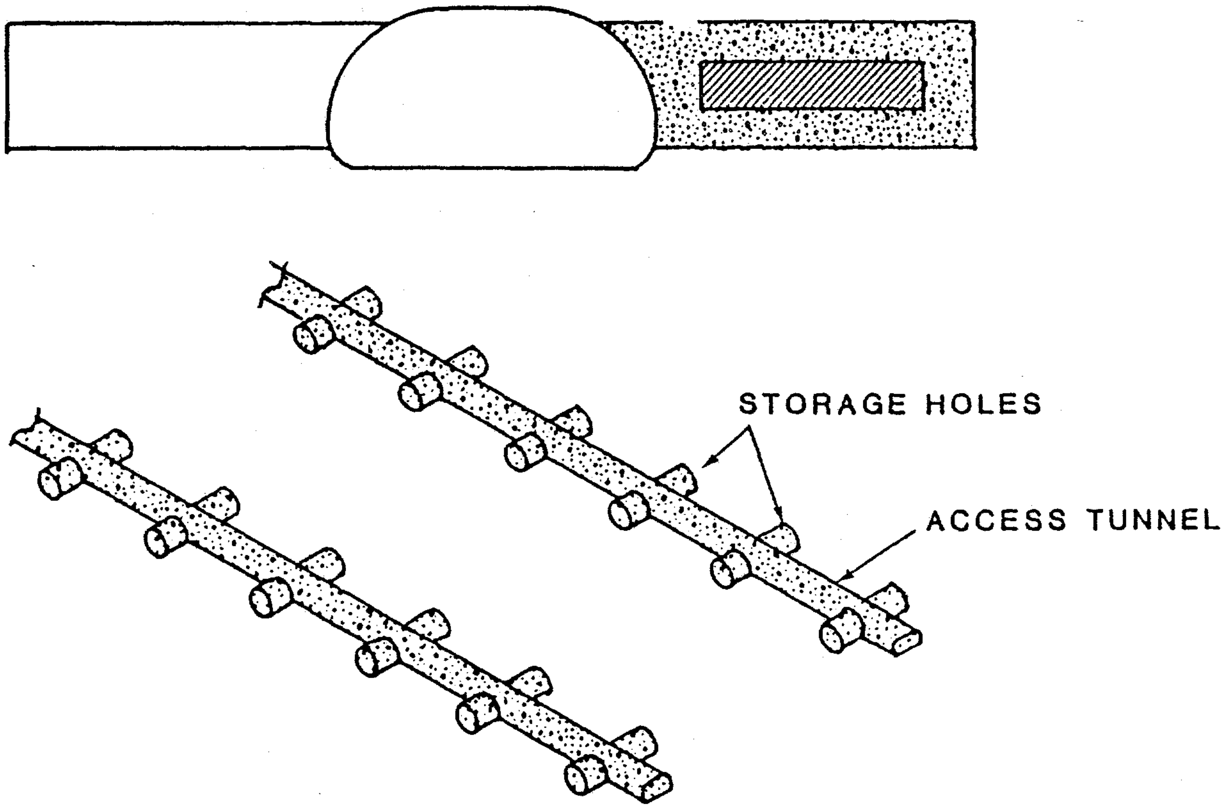

Fig. A.2. Short horizontal borehole emplacement concept. Source: K. H. Henry, Decision Memorandum for Waste Emplacement Concept, SD-BWI-PD-012, 1984.

designed basalt repository, it is not expected that full lithostatic pressure (~30 and ~60 MPa, respectively, for the vertical and horizontal stress components) will be attained for many thousands of years.

The pH and redox conditions will strongly influence corrosion rates, water chemistry, and the various potential chemical reactions within the system. The unperturbed pH is approximately equal to 10, and the Eh has been estimated as low as −0.5 V, which is an anoxic condition and favorable to long canister lifetime. The pH will decrease as the temperature increases. The air in the freshly sealed repository will produce oxic conditions with an Eh up to +0.5 V. A crude estimate of the time for the repository to be restored to geologic equilibrium and slightly anoxic conditions is several hundred years (SMITH 1980). Demonstration with reasonable assurance that such favorable anoxic conditions will exist in the first few hundred years, however, is yet to be accomplished.

A.1.2 Waste Package Designs

Some details of the current conceptual design for the waste packages are not available. The major changes from the previous conceptual design that were discussed in the earlier report (CLAIBORNE 1985) were the increase in the thermal loading of the waste canister from 1.65 kW to a maximum of 2.2 kW for spent fuel, along with some dimensional changes and the use of prepackaged, encapsulated packing instead of pneumatic emplacement of the packing, which was (and still is) considered to be a mixture of 25% sodium bentonite and 75% crushed basalt. The package design for CHLW is essentially unchanged except for the use of prepackaged packing. The packing consists of >15.2-cm-thick preformed annular sections of the bentonite-basalt mixture that are stacked together and encased in an inner and outer sheath of 0.5-cm-thick low-carbon steel. The packing assemblies are to be emplaced in the short horizontal boreholes first; then the waste canisters are inserted and the shield plug is installed.

As pointed out in the previous section, in Stage I operations, the spent fuel assemblies that are received intact (unconsolidated) in either stainless or low-carbon steel canisters will be overpacked with thick low-carbon steel prior to disposal. In Stage II, the spent fuel assemblies will be disassembled and the fuel rods will be tightly repacked into low-carbon steel canisters, which are then overpacked with low-carbon steel prior to insertion in the emplaced packing assembly (see Fig. A.3). The thickness of the overpack is based on the structural requirements necessary to withstand hydrostatic pressures to be developed, the additional thickness for a corrosion allowance, and any additional thickness that may be required to reduce groundwater radiolysis to acceptable levels. The internal diameter of the spent fuel canister is sized to accept the spent fuel rods from four PWR assemblies (heat generation rate, 2.2 kW) or nine BWR fuel assemblies, and the rods are to be installed in a tight fit for the largest expected number and size of the fuel rods (DOE 1984a). The internal length is sized to accept the longest expected fuel rods with a small clearance between the container head and the fuel rod ends. Some overall characteristics are shown in Table A.1. The more-detailed characteristics for spent fuel shown in Table A.2 are for the earlier

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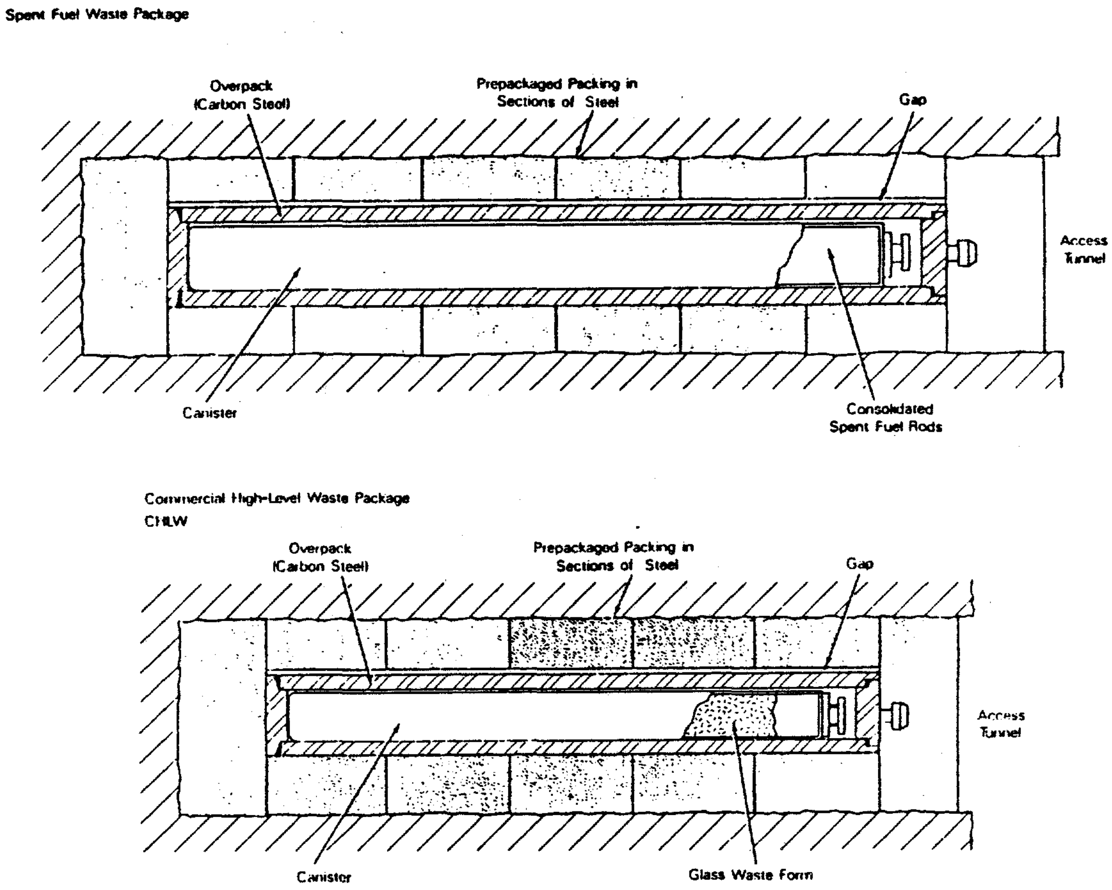

Fig. A.3. Reference waste package conceptual designs in basalt (short borehole horizontal emplacement). Source: S. Stephens et al., Methodologies for Assessing Long-Term Performance of High-Level Radioactive Waste Packages, NUREG/CR-4477 [ATR-85 (5810-01)-1ND], 1986.

Table A.1. Overall waste package and borehole characteristics

Parameter	Spent fuel Stage I	Spent fuel Stage II	Commercial high-level waste
Waste package			
Outside diameter, m	0.852	0.503	0.456
Borehole			
Diameter, m	1.24	0.890	0.840
Length, m	6.4	6.1	6.1
Waste container			
Max. length, m	4.8	4.44	3.25
Loaded container			
Weight, MT[a]	11.7	7.6	2.7
Maximum thermal loading, kW	2.20	2.20	2.21

[a] MT = metric tons.

Table A.2. Summary of BWIP alternative waste package design features

Conceptual design feature	SF[a]	CHLW
Waste form features		
Waste form diameter, cm	30.5	32.5
Waste form length, cm	385	305
Waste form weight, kg	1980	845
Waste content per package, kg	1380[b]	595[c]
Package heat load, W	1650	2210
Package features		
Canister wall thickness, cm	—	0.95
Canister outside diameter, cm	—	32.4
Overpack inside diameter, cm	30.5	35
Overpack wall thickness, cm	5.6	5.3
Overpack outside diameter, cm	41.7	45.6
Overpack head thickness, cm	8.5	8.6
Overpack length, cm	405	325
Overpack empty weight, MT	2.19	1.84
Overpack loaded weight, MT	4.17	2.68
Packing features		
Packing thickness, cm	15.2	15.2
Packing weight, kg[d]	1380	1170
Emplacement features		
Borehole diameter, m	0.73	0.76
Borehole pitch, m[e]	18.3	32.6
Package pitch, m[f]	4.25	3.45

[a] Dimensions shown for the consolidated rods from three PWR assemblies. Length, weight, and waste loading will be slightly different for BWR rods. For spent fuel, a separate canister is not employed. The overpack is the only metallic barrier between the spent fuel and the geology.

[b] kg of uranium.

[c] This amount of CHLW results from the reprocessing of 2280 kg of uranium.

[d] Based on 75% basalt and 25% bentonite (by weight), with 50% voids. Dry particle density is 2.3 g/cm^3.

[e] Distance between borehole centers.

[f] Distance between package centers in a borehole (package length plus 0.15 m for packing).

Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Waste Package Conceptual Design for a Nuclear Repository in Basalt, RHO-BW-CR-136P/AESD-TME-3142, 1982.

Westinghouse design (WEC 1982) and do not apply for the current concept; they now represent a potential alternative concept. The conceptual design of the waste package for CHLW has not significantly changed with regard to dimensions, even though the packing is now preformed and contained in a steel sheath.

The conceptual design (WEC 1982) for a reference waste package containing borosilicate glass is also shown in Fig. A.3. Some overall characteristics are summarized in Table A.1; more-detailed characteristics are given in Table A.2. The reference canister is fabricated by welding together preformed sections of 0.95-cm-thick 304 L stainless steel. The canisters have ~15% void volume at the top to prevent overflow during glass pouring. For design purposes, the canister is considered only as an aid in processing and handling; protection against crushing and corrosion in the repository is provided by the overpack.

The packing material is required to be a low-permeability barrier with a hydraulic conductivity that is sufficiently low to limit transport of water to the overall pack in the containment period by diffusional processes and to also limit radionuclide transport after a breach of the waste canister. When the packing becomes saturated with water, it is expected to expand and reduce the void fraction and permeability to a very low value.

A.1.3 Degradation and Nuclide Release Scenario

Barring unexpected severe tectonic events, water flow through the region of the repository horizon can be expected to be very slow — on the order of a few centimeters per year. A long time will be required to fill the repository voids, saturate the backfill, and start the degradation process on the overpacked waste canister. This expected scenario for the waste package degradation has been selected for the initial study on long-term waste package behavior in a waste repository in basalt.

The design life of a waste package can be divided into two periods (WHEELWRIGHT 1981). The first is one of nearly complete water exclusion; the second occurs after saturation of the backfill.

The water exclusion period is the time interval when the dry,* bentonite-crushed basalt (i.e., structurally bound water is primarily present) is in place prior to water intrusion into the packing. It can be expected that the waste package would remain dry prior to backfilling the rooms since it is reasonable to assume that any water collected in horizontal drill holes will drain toward a room. The overpacked waste canister will be protected from liquid water and steam by the steel-sheathed packing. No significant corrosion of the low-carbon steel should occur for these conditions. When the overpacked waste canister is inserted into the previously emplaced

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*Bentonite does not exist naturally in the dry state. In compacting bentonite, ~10% water is necessary for structural stability. The specifications for the water content for the pneumatic emplacement have not been determined yet, but it will probably be in the range of 5 to 10 wt %.

packing assembly, the temperature of the packing will begin to rise and a slow release of the interlayer water as vapor would begin. This vapor could cause degradation of the packing (see Sect. B.3.1 for a discussion) and supply a source of water for corrosion of the low-carbon steel sheath enclosing the packing. The duration of the "dry" period for the overpacked canister will be determined by the time required for water to migrate through the host rock to the packing material and cause significant breaching of the packing sheath by corrosion. This water migration depends on the site geology, as well as the thermal and hydraulic gradients, around the waste package.

When sufficient water enters the waste package, the second period begins with a slow penetration and hydration of the packing material (rehydration of interlayers since the processes have been generally considered reversible for dehydration under 400°C). However, more recent evidence may indicate otherwise (see Sect. B.3.1). The mechanisms and kinetics by which groundwater hydrates montmorillonite clays and migrates through are not clearly understood. Two concepts for water migration in bentonite packing materials were examined by Wheelwright (1981), and estimates of saturation times were in the thousands of years. In one concept, the packing material is viewed as a system of hypothetical concentric layers. Initially, the groundwater will migrate from the host geology into the interfacing layer of the packing material, and the clay of that layer will then begin to hydrate and exert a swelling pressure. Before the water can reach the second layer, it must migrate through the first layer, which has a reduced permeability because of the swelling that causes the second layer to require a longer time for saturation. By similar reasoning, each successive layer requires a longer time to attain saturation. For this model, it was concluded that the time required to saturate 20 cm of intact packing material will be on the order of several thousand years. The second concept involves the time required for a significant volume of water to reach the waste package, equilibrate with the system, and then continue migration down the hydraulic gradient within the packing material. This model indicated that the order of 10^5 years was required for a significant amount of water to reach the overpack surface.

These time estimates are based on crude models, and it was assumed that the bentonite was highly compacted as compared with the less dense packing (1700 kg/m^3) that has been proposed, or that resulting from pneumatic emplacement as in the older design.

Water movement through the packing material after saturation will be controlled by the hydraulic conductivity of the packing material and the regional hydraulic gradient. Since bentonite swells on hydration, the hydraulic conductivity will decrease to the point that diffusion will probably be the controlling transport mechanism, particularly in view of the low hydraulic gradients (<0.01) that can be expected.

With the availability of liquid-phase water, the corrosion of the overpacking can be expected to accelerate. The various mechanisms and rates for the corrosion of low-carbon steel are dependent on the pH, redox conditions, water composition, temperature, and the accumulation of corrosion products (e.g., pitting corrosion can be retarded as the result of blockage of the pits by accumulated corrosion products). See Appendix B, Sect. B.1.2, for more-detailed discussion of potential corrosion mechanisms.

A.2 NUCLEAR WASTE REPOSITORY IN TUFF

The Yucca Mountain site that has been proposed for detailed characterization is located in Nye County, Nevada, adjacent to the southwest portion of the Nevada Test Site (NTS) and about 100 miles northwest of Las Vegas. Yucca Mountain is in the southern part of the Great Basin, where all surface waters drain into closed basins rather than flowing into the ocean.

A.2.1 Repository Design and Conditions

The regional setting, geology, hydrology, and geochemistry of the Yucca Mountain location have been discussed in a report prepared by the Sandia National Laboratories (GUZOWSKI 1983).

The rock unit selected for the repository at Yucca Mountain is the Topopah Spring Member of the Paintbrush tuff unit, which is about 350 m thick at the site and is characterized by four distinct zones, from top to bottom: (1) nonwelded to densely welded, generally vitric tuff; (2) a moderately to densely welded, devitrified tuff; (3) a basal vitrophyre; and (4) a vitric tuff varying from welded to nonwelded. The densely welded, devitrified portion of the second zone (from the top) is currently being considered for the waste repository (DOE 1984b, 1986b).

The reference horizon is assumed to lie completely in the unsaturated, or vadose, zone at a depth of 350 to 400 m below the surface and more than 100 m above the water table (O'NEAL 1984). Conceptual designs for a repository and analyses for waste packages in tuff below the water table were developed by the Westinghouse Electric Corporation (WEC 1983a). However, the candidate horizon was changed to the vadose zone in late 1982 (VIETH 1982; DUDLEY 1983). Consequently, changes and additions (which are still being made) to the conceptual designs have been made by the Lawrence Livermore National Laboratory (LLNL) to reflect the change in the repository horizon to a location above the water table. The location of the disposal horizon in the unsaturated, or vadose, zone is a significant change that appears to have advantages with respect to a more conventional and originally planned location below the water table.

Discussions of the aspects of nuclear waste disposal in the vadose zone in arid regions are given by Roseboom (1983) and by Wollenberg (1983). In recognition of the concerns that had been expressed as to the applicability of 10 CFR 60 to disposal about the water table, the NRC has proposed amendments (OSTROWSKI 1984) to the rule to ensure that it is equally applicable to disposal within the saturated and unsaturated zones. The proposed amendments do not directly affect the waste package since the NRC staff concluded that the current rule is adequate for the performance objectives and the specific design criteria for the waste package and its components.

As a result of disposal above the water table, the waste canisters will not become submerged in a continuum of water. They will be subject to constant contact with water vapor and only to intermittent contact with limited amounts of liquid water that seeps down through the tuff after the temperature drops below the boiling point. The water influx to the disposal horizon has been estimated to be <1 mm per year (MONTAZER 1984) of the rainfall at the surface. The pressure exerted on the canister should be near atmospheric since a hydrostatic head cannot develop without a continuum of water above the disposal horizon. A small amount of pressure might develop if localized rock sloughing occurs in the emplacement borehole. The vadose water and atmosphere of the repository are expected to be mildly oxidizing. The temperature of the undisturbed rock at the repository horizon is estimated to be 29°C.

A number of tuff—water interaction tests have been completed utilizing both crushed and intact Topopah Spring tuff samples in contact with water from a nearby well, which is completed in the Topopah Spring Member where it extends below the water table. The test matrices include a range of temperatures, rock-surface-area/water-volume ratios, and test durations. The results of these tests have been reported by Knauss (1983). In general, the tests indicate that (1) changes in water chemistry will be minimal, (2) no significant alteration of the primary mineral phases of the rock is to be expected, and (3) the near-neutral pH of the system should be maintained (BALLOU 1983). Increases in the concentration of dissolved salts in the groundwater that will ultimately contact the waste package could result from redissolving the salts precipitated by the evaporation of the water contacting the waste package in the earlier stages (O'NEAL 1984).

The repository is to be of the conventional pillar-and-room mine design, which will facilitate multiple waste package emplacement. Both vertical and horizontal configurations are currently being studied for waste emplacement. Vertical emplacement, currently the reference design (see Figs. A.4 and A.5), consists of emplacing the waste packages in vertical holes bored in the floor of the mine drifts. The horizontal boreholes for the horizontal emplacement layout (see Figs. A.6 and A.7) have a carbon-steel liner that is assumed to be required for reliable emplacement and retrieval operations. For this configuration, multiple waste packages (∿40) are emplaced in long horizontal holes that are drilled in the walls of mined drifts. The waste package design is the same for either waste emplacement configuration (O'NEAL 1984; JACKSON 1984).

Horizontal emplacement will involve the removal of less rock, which will result in a lesser degree of disruption of the natural geology and lower costs for the excavation. However, special horizontal boring technology will require development, which introduces the uncertainty as to whether the horizontal holes are technically feasible to bore. The equipment and methods required to bore the vertical holes are standard (O'NEAL 1984). Studies of the evaluation of horizontal emplacement are being continued, but vertical emplacement remains the reference design pending further developments.

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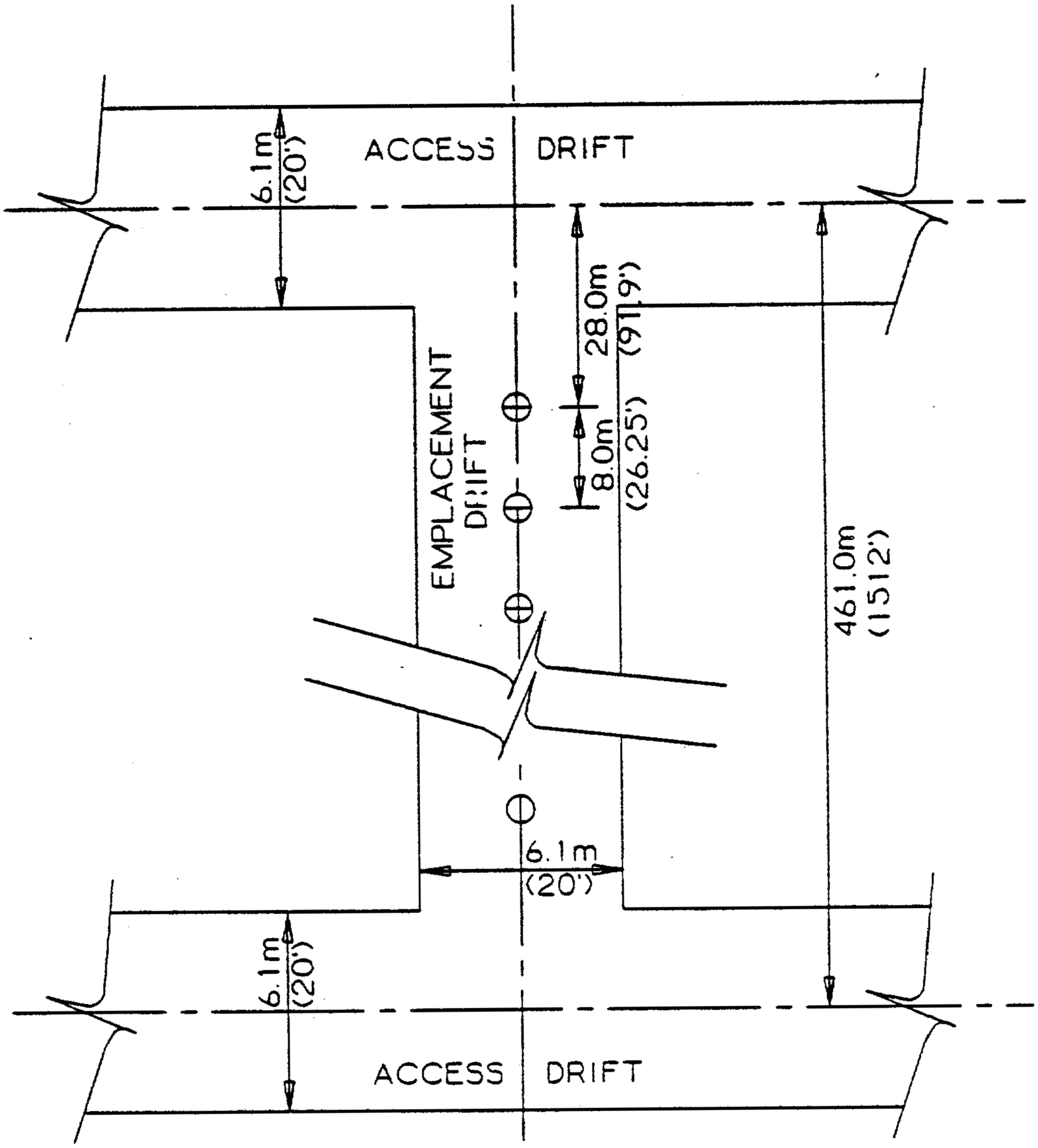

Fig. A.4. Plan view of the reference drift and borehole geometry for vertical emplacement of spent fuel. Source: J. L. Jackson, Nevada Nuclear Waste Storage Investigations — Preliminary Concepts Report, SAND 83-1877, 1984.

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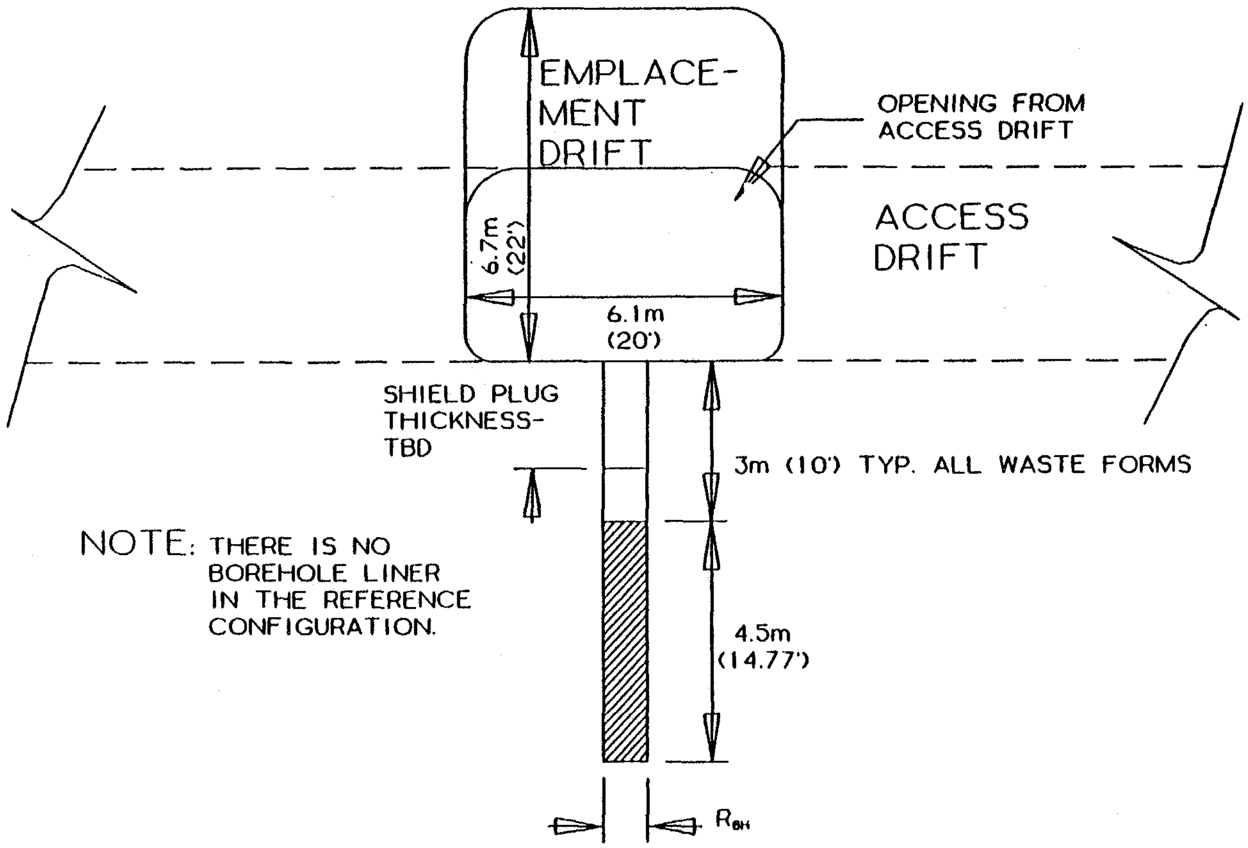

Fig. A.5. Cross-sectional view of the reference vertical emplacement drift and borehole geometry. Source: J. L. Jackson, Nevada Nuclear Waste Storage Investigations — Preliminary Concepts Report, SAND 83-1877, 1984.

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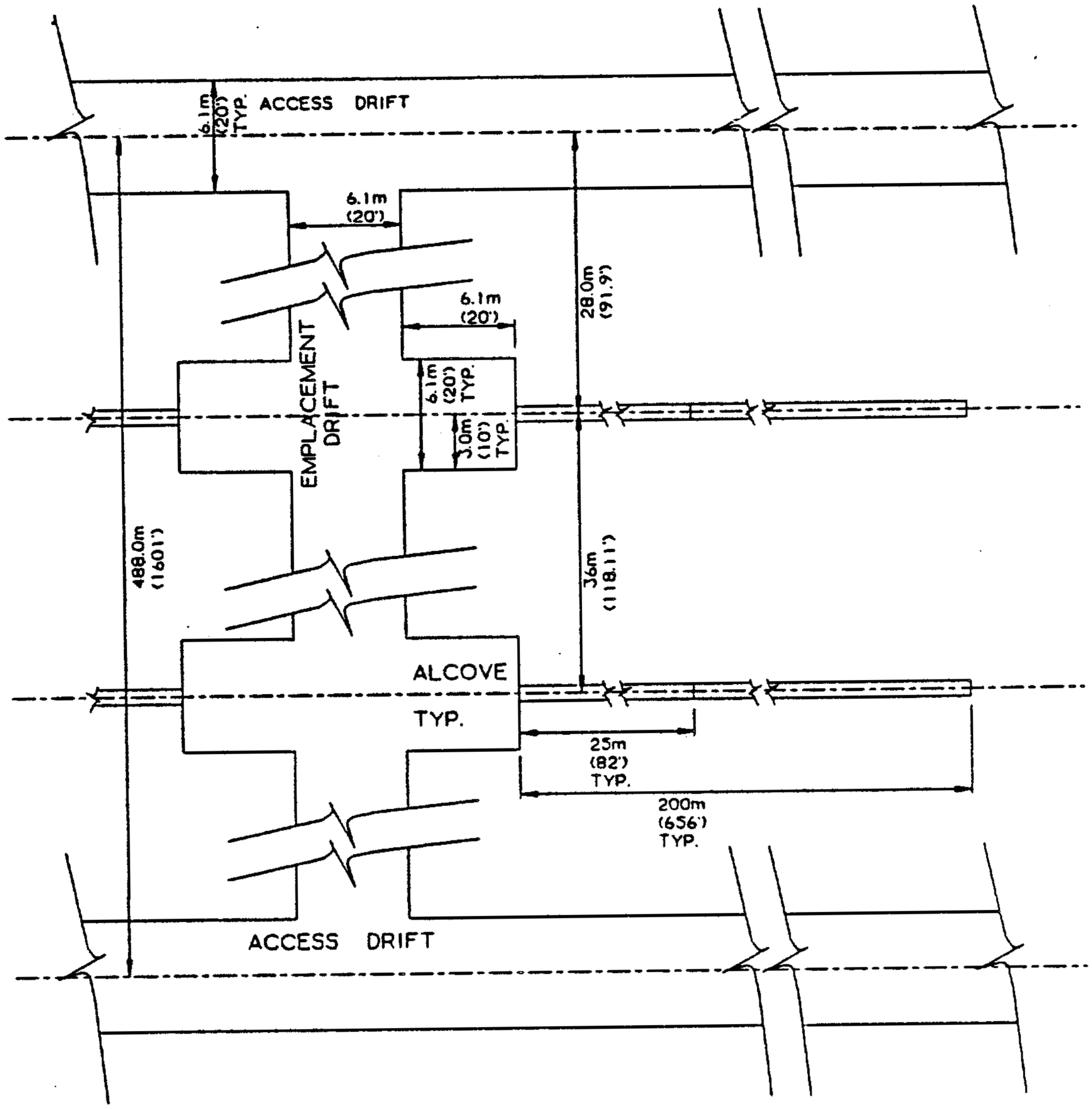

Fig. A.6. Plan view of the reference drift and borehole geometry for horizontal emplacement of spent fuel. Source: J. L. Jackson, Nevada Nuclear Waste Storage Investigations — Preliminary Concepts Report, SAND 83-1877, 1984.

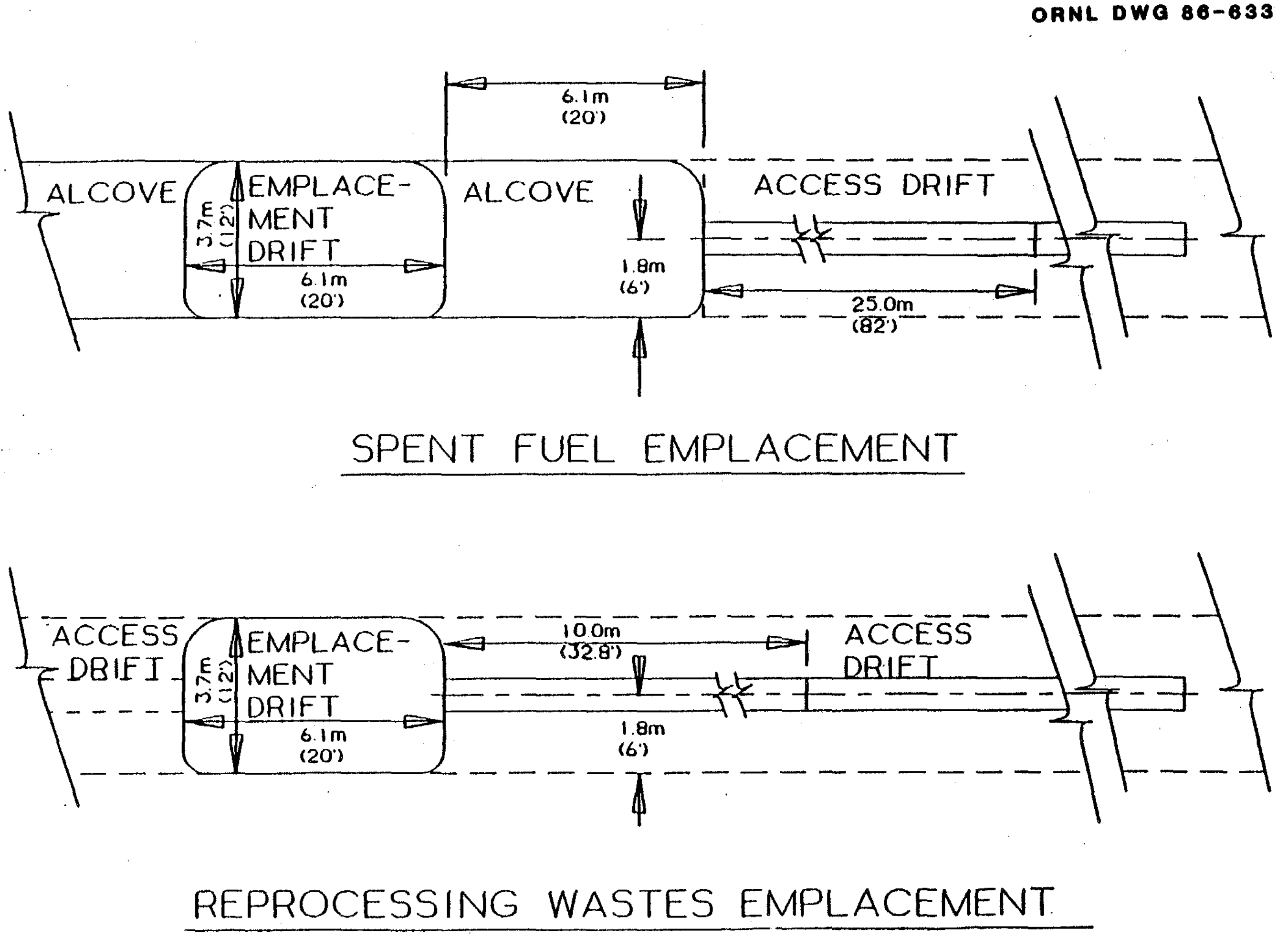

Fig. A.7. Cross-sectional view of the reference horizontal emplacement drift and borehole geometry. Source: J. L. Jackson, Nevada Nuclear Waste Storage Investigations — Preliminary Concepts Report, SAND 83-1877, 1984.

A.2.2 Waste Package Designs

A number of uncertainties have yet to be resolved with respect to the performance of waste package components; consequently, the conceptual package designs include alternatives to accommodate vertical or horizontal emplacement, and spent fuel canisters emplaced either with or without a preformed tuff packing material (BALLOU 1983).

The reference conceptual designs (which do not include packing) for both spent fuel and CHLW are shown in Fig. A.8. Spent fuel canisters of three different lengths, 4.00, 4.50, and 4.75 m (plus pintle), are used to accommodate spent fuel rods of various lengths; each is 70 cm in diameter. The internal space-frame for the internal canisters or containers for the spent fuel provides mechanical stability and enhances heat transfer; it consists of a 1-cm-thick pigeonhole array of 304L stainless steel square receptacles with end plate and pintle. For BWR assemblies, the arrangement may contain up to 7 intact or 14 consolidated fuel assemblies (i.e., fuel rods removed from fuel assemblies and clamped together), which produces a maximum heat generation rate of 3.36 kW. The PWR contains three intact or six consolidated fuel assemblies, which produces a maximum heat generation rate of 3.30 kW (O'NEAL 1984). The spent fuel canisters are to be fabricated from an austenitic stainless steel with a wall thickness of 1 cm. The austenitic steels under consideration for this purpose include: 304L, 316L, and 321, with the Incoloy 825, a nickel-based alloy, as a backup material (RUSSELL 1983). More recently copper alloys [CDA102(OFHC)], CDA613 (8% Al bronze), and CDA715 (70/30 Cu-Ni) were added to the list of backup materials for overpacking (McCRIGHT 1985). Using the 70-cm-diam waste package for consolidated boxed assemblies is contingent on a successful 2/1 volume reduction that has been demonstrated in tests of preconsolidation of fuel elements (ANDERSON 1982).

An alternative design (see Fig. A.9) utilizing a precompressed tuff packing that is encased in a steel container is also being considered to reduce potential release rates (O'NEAL 1984). Currently, the packing material being considered (if needed) is approximately 15 cm thick and consists of either crushed and pressed tuff, or crushed and pressed tuff containing 5 to 15% iron-bearing smectite clay as a binder (GREGG 1983). One reason for the alternate design is that segregated phases within the spent fuel are considerably more soluble in water than the glass waste forms and, therefore, less likely than CHLW waste to meet the postcontainment period release requirements if the Zircaloy cladding does not perform satisfactorily.

The reference CHLW form is borosilicate glass, which is poured to about 85% capacity into 304L stainless steel canisters which are 32 cm in diameter, 300 cm long, and 1 cm thick (SLATE 1981). The pour canister is overpacked (see Fig. A.8) with a 1-cm-thick 304L stainless steel container. The overall length is 324.5 cm (including the overpack pintle), and the outer diameter of the overpack is 36 cm (O'NEAL 1984).

Although the reference canister and overpack metal is currently AISI 304L stainless steel, alternative metals such as 316L and 321 stainless steels and Incoloy 825 and some copper alloys are still under consideration for the following reasons:

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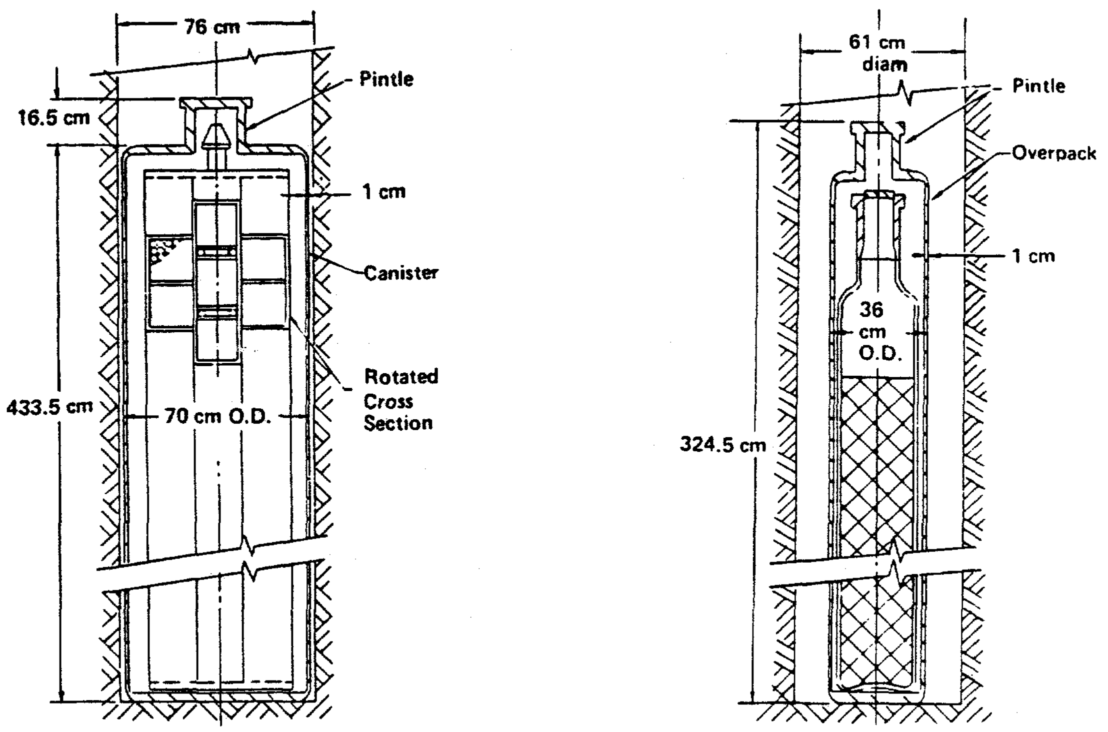

BWR (boiling water reactor) spent fuel waste package emplaced in vertical borehole PWR same except for 3 holes instead of 7.

CHLW (commercial high-level waste) package emplaced in vertical borehole

Fig. A.8. Reference waste package designs. Source: W. C. O'Neal, D. W. Gregg, J. N. Hockman, E. W. Russell, and W. Stein, Preclosure Analyses of Conceptual Waste Package Designs for a Nuclear Waste Repository in Tuff, UCRL-53595, 1984.

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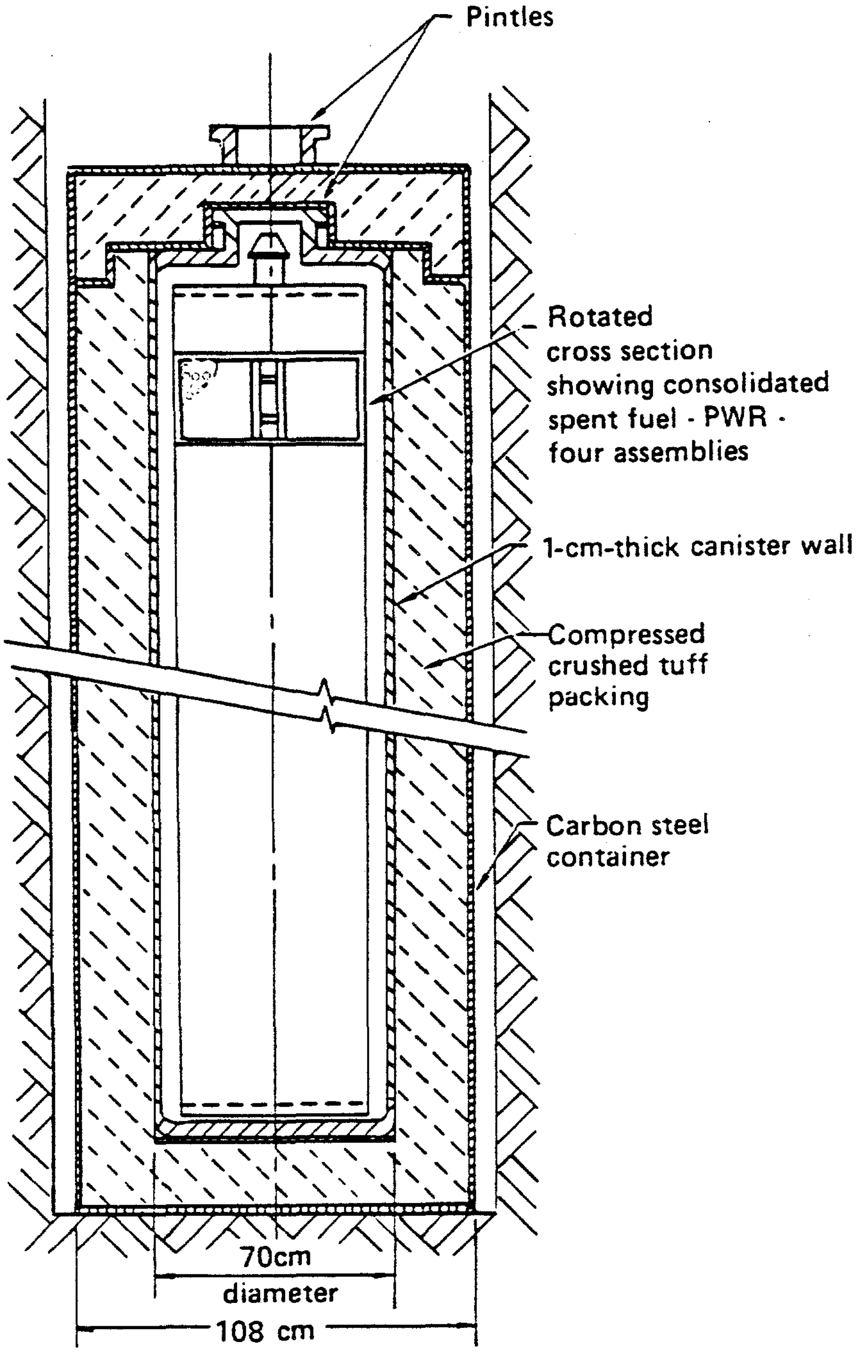

Fig. A.9. Alternative design for consolidated spent fuel waste packages if packing is needed to reduce release rate. Source: W. C. O'Neal, D. W. Gregg, J. N. Hockman, E. W. Russell, and W. Stein, Preclosure Analyses of Conceptual Waste Package Designs for a Nuclear Waste Repository in Tuff, UCRL-53595, 1984.

(1) to provide a potential replacement metal until the reference metal is confirmed by testing under site-specific conditions, and (2) to provide comparative data to support the choice of AISI 304L stainless steel as the reference metal during the regulatory review (RUSSELL 1983).

A.2.3 Degradation and Nuclide Release Scenario

Barring unexpected severe tectonic events along with severe changes in the hydrology of the region, a continuous flow of water around waste packages located in the unsaturated zone is not possible. It has been estimated that downward infiltration from the surface and overlying unsaturated media will supply <1 mm per year (MONTAZER 1984) of the rainfall at the surface as water to drip, or seep, around the waste packages. With the small quantities of water involved and the lack of a water continuum above the waste packages, the pressure should remain at near atmospheric. A small pressure could develop if localized rock sloughing occurs in the emplacement boreholes. It is difficult to envision a return to the original lithostatic pressure by the natural and expected geologic processes within the first thousand years or so in a backfilled repository in the densely welded tuff of the Topopah Spring Member.

At the elevation (~900 m) of the proposed repository, the boiling point of water is ~95°C. Consequently, the waste package will be exposed to air-water vapor mixtures in the first few hundred years until the surface temperature of the waste canister or the overpack (if utilized) falls below 95°C, which is ~300 years for reprocessed waste for the reference design without crushed tuff packing (~400 years with packing) and, in the case of spent fuel, >1000 years (STEIN 1984). After these times, liquid-phase water will be intermittently in contact with the waste package and the aqueous corrosion mechanisms should be the predominant degradation process at an accelerated rate. (See Appendix B, Sect. B.1.2, for a discussion of the potential corrosion mechanisms.) During the period when the temperatures around the waste package exceed the boiling point, evaporation of groundwater that seeps will occur and deposit salts. When the temperatures permit the liquid phase to exist, these salts will dissolve and produce a liquid phase that is probably more aggressive from a corrosion viewpoint than the original groundwater.

Under the expected conditions in the proposed repository, it appears that the waste package will be highly resistant to corrosion but may be susceptible to transgranular corrosion cracking (RUSSELL 1983). A backup material, AISI 316L or 317L stainless steel or Incoloy 825, is planned for use if future testing cannot provide the data to ensure that the 1000-year containment requirement can be achieved.

It can be expected that significant breaching of the waste canisters will occur during the isolation period (10^3 to 10^4 years) and that leaching of the waste will begin. Assuming that leaching of radionuclides will not exceed the NRC limit ($<10^{-5}$/year of the radionuclides present at 1000 years), the transport time of the contaminated water then becomes the important issue. Densely welded tuffs are highly fractured and jointed, with vertical jointing being the most common type. Many welded tuffs also show a horizontal plate jointing in or near the zone of maximum compaction (RAWLINGS 1984). In the unsaturated zone, any contaminated water can be expected to seep down to the water table through these vertical joints and fractures. Tuff, having generally good ion-exchange properties along with the phenomenon of matrix diffusion, can be expected to cause retardation of some radionuclides. These effects are subject to large uncertainties and must be sufficiently substantiated for use in performance assessments that must be made to determine the time of travel and amounts of radionuclides released to the accessible environment.

A.3 NUCLEAR WASTE REPOSITORY IN SALT

As the result of preliminary investigations and site-screening activities, a number of salt sites were examined for possible use as a waste repository. Of these, the site in Deaf Smith County, Texas, has been proposed for detailed characterization. The potential site is located in the north-central part of the county, which is situated in the Southern High Plains of the Texas Panhandle, approximately 56 km west of Amarillo.

A.3.1 Repository Design and Conditions

The Deaf Smith County site is located in the Palo Duro Basin, a structural sub-basin within the larger Permian Basin. The potential repository host rock is a thick sequence of bedded salt (halite) within Unit 4, the second unit from the top of the five units, of the Lower San Andres Formation (see DOE 1984c or DOE 1986c for a complete description of the geology of the region and stratigraphy of the site).

In the vicinity of the site, the depth to the top of Unit 4 ranges from about 700 to 760 m with a thickness of about 48 m. The geothermal gradient is 0.4 to 0.6°C per 30 m (0.8 to 1.0°F per 1000 ft), and the temperature is estimated at 27°C at the proposed repository horizon.

Salt deposits are free of circulating groundwater; however, water is present in the form of small saturated brine inclusions within the halite crystals, the interlayer water in clay impurities, and other minerals that may contain waters of crystallization. When a thermal gradient is applied, solubility differences between the hot and the cool sides of a brine inclusion can cause a migration of a brine inclusion up the thermal gradient toward the heat source. The quantity of brine that would contact a waste package by this or other mechanism depends on the thermal parameters and the properties of the bedded salt, which are discussed in Sect. A.3.4.

The Lower San Andres Unit 4 host horizon contains small amounts of brine as fluid inclusions and intergranular water in halite and as hydration and pore water in the mudstone layers and chaotic salt/mudstone mixtures. The maximum amount of brine available was calculated (DOE 1986c) from data on the water content of halite and mudstone in the Lower San Andres Unit 4 host horizon.

The results of these calculations as reported by DOE were as follows:

> Water in Halite: Based on the analysis of over 150 samples from various Lower San Andres Unit 4 cores, fluid inclusions and intergranular water average 0.4 wt % and vary between 0.1 and 0.8 wt %.

Assuming 0.5 wt % water in clay-free halite and 88% halite in the Unit 4 host horizon strata, 0.44 wt % water could be derived from halite.

Water in Mudstone: The maximum water content of host horizon mudstone was estimated to be 15.0 wt %, but the actual value is likely closer to 7.5 wt %. This includes both pore water in the mudstone and water in hydrous minerals such as clays. The average mudstone content of Unit 4 host horizon is 8%. Therefore, 1.20 wt % water can be derived from Unit 4 mudstone.

Sum of Water from Halite and Mudstone: The 0.44 wt % water from halite plus 1.20 wt % water from mudstone equals 1.64 total wt % water.

Conversion from Wt % Water to Vol % Water: Assuming a density of water of 1.00 g/cm and average density of Unit 4 salt (mostly halite) of 21.16 g/cm, then 1.64 wt % water converts to 3.48 vol % water and 4.14 vol % brine.

The effects of geochemical processes on radionuclide sorption and rock strength are probably small, and there is no clear indication as to whether the effects will be positive or negative. There is little direct evidence on the oxidation state of the fluids in the halite formation, but the mineral assemblages present would be unlikely to create an oxidizing environment.

The lithostatic pressure of the undisturbed rock at the proposed repository horizon will be approximately 17 MPa, which will be essentially isotropic because of the salt plasticity. This lithostatic pressure will produce a resultant closure force on the mined-out rooms, even with crushed salt backfilling, since the crushed salt will contain ~40% voids. Hard rocks can resist these forces; however, salt (halite), which is a plastic material, creeps under stress. No hydrostatic pressure can be exerted because a pressure-producing water column will not exist unless a severe tectonic event creates one.

After the repository has been backfilled with crushed salt, the disposal room margins will slowly move in and squeeze out entrapped brine until the crushed-salt backfill consolidates into a solid mass or the brine becomes trapped. As a result of the salt creep, the pressure after sealing of the repository will slowly increase from atmospheric to the stress state of the surrounding rock, 17 MPa.

It is difficult to estimate the time required for a repository to regain the regional lithostatic pressure. Room-closure calculations are complex and are very specific for the repository design and local geology. For example, the constitutive law (creep rate as a function of stress, temperature, and petrofabrics of the salt) can vary widely for different salt formations.

Room-closure-rate calculations involving thermo/viscoelastic analyses of a room-and-pillar configuration were made by Wagner (1980) for several thermal loadings and repository depths, and the effects of crushed salt backfill on room closure were examined by the IT Corporation (ITC 1984). Wagner's calculations were for open rooms and would hold only for a few tens of years since the model would break down when the strains become large and slabbing and floor heaving begin. The backfill, however, would tend to prevent rock failure by the resistance that is offered. On the basis of the calculated results, a few tens of years seems to be a reasonable guess for time required for the entire repository horizon to approach the full lithostatic stress.

In a more recent study, Ratigan (1985) has predicted that the salt surrounding the waste package will creep and consolidate to a monolithic structure within a few years after emplacement. The foregoing discussion primarily refers to near-field conditions. In the very near field adjacent to the waste package, borehole closure can result in the development of local thermal stresses that cause a rapid increase in the pressure on the waste package. Conservative calculations by Callahan (1975) for a steel sleeve in close contact with the host salt indicate that localized thermal stresses exceeding lithostatic in the form of radial compressive stresses on the waste package may develop in the first year and then decay as the result of salt creep. The results are very dependent on design, salt properties, and the model used. Whether such high stresses are realistic or not is open to question.

A number of non-site-specific conceptual designs have been made for salt repositories. The most recent and detailed designs include those by Kaiser (1978), Stearns-Roger (1979), Bechtel (1981), and Stearns-Roger (1983). All designs were for the conventional room-and-pillar mine arrangements; vertical emplacement of the waste packages was similar to the tuff conceptual design illustrated in Figs. A.4 and A.5. Backfilling of the disposal rooms was to be done with crushed salt.

A.3.2 Waste Package Designs

Conceptual designs for waste packages for the disposal of spent fuel and high-level waste in salt were developed by Westinghouse (WEC 1983b). A reference design and two alternatives for both waste forms were described for disposal in vertical emplacement holes in a room-and-pillar type of salt mine. In a more recent report (WEC 1986), new reference designs have been developed from the earlier designs (WEC 1983b) for commercial high-level waste and consolidated spent fuel. In all cases, it was assumed that crushed salt would be used as packing in the space between the waste package and the host rock salt.

A.3.2.1 Spent Fuel

In the case of the original reference design for spent fuel (which is no longer the reference design), disassembled spent fuel rods from 6 PWR (or 18 BWR) are consolidated in a close-packed circular array (see Fig. A.10) with a heat load of 3.3 kW/package at emplacement. The Alternate I design has the same heat load but utilizes a larger-diameter and longer length container in order to accommodate three stainless steel boxes containing two consolidated fuel elements (see Fig. A.11). For these designs, the canister (called overpack reinforcement by Westinghouse) is made of 7-cm-thick carbon steel. The overpack for these designs is a thin corrosion-resistant shell of a titanium alloy (TiCode-12) nominally 0.25 cm thick.

The Alternate II design, which was considered the reference design for a short period (DOE 1984c), is no longer a reference. It was replaced as

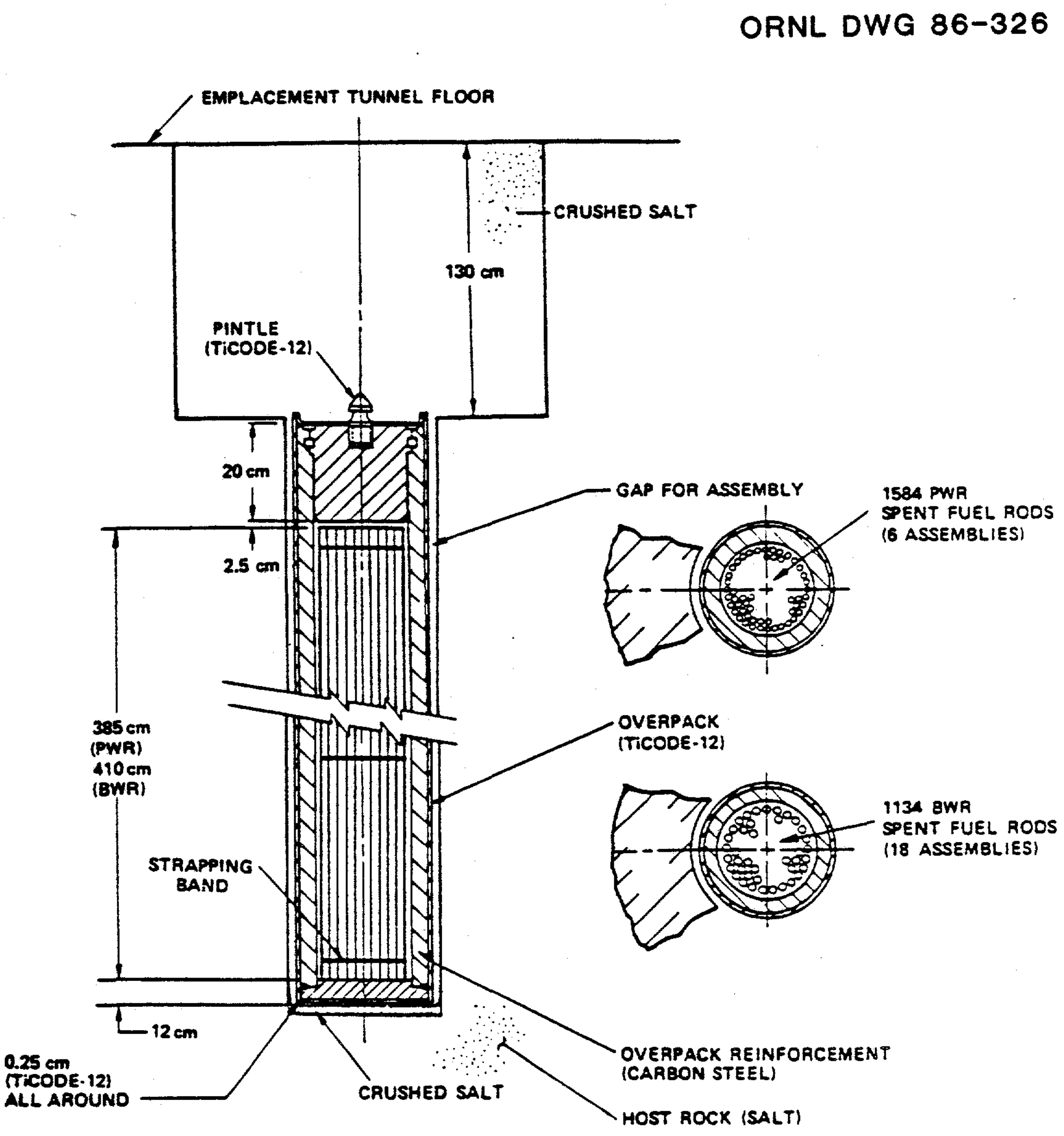

Fig. A.10. Reference spent fuel package for borehole emplacement. Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, 1983.

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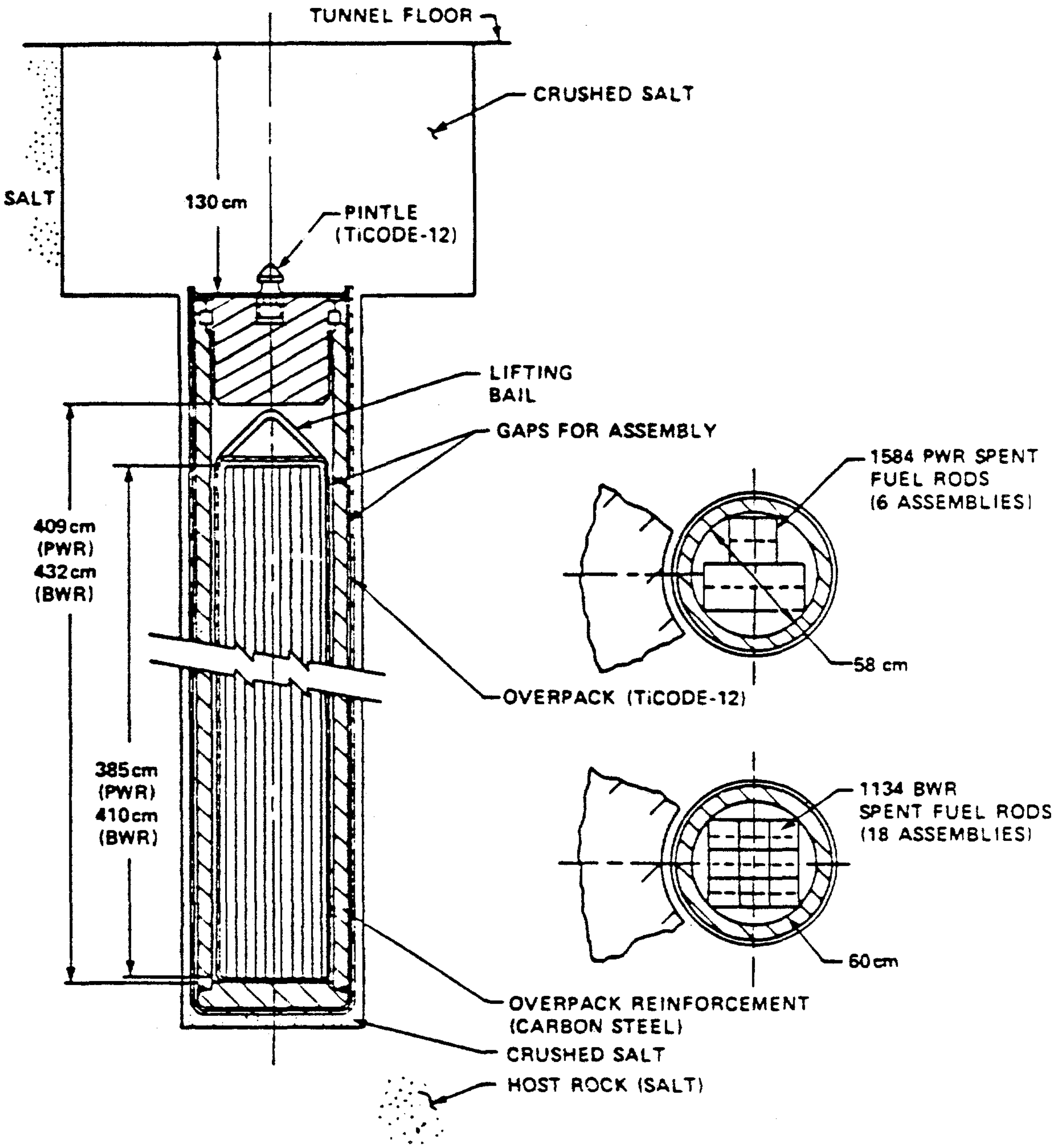

Fig. A.11. Boxed spent fuel rod package concept for borehole emplacement. Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, 1983.

the result of continuing design studies (WEC 1986). In the Alternate II design, the TiCode-12 cladding was eliminated and the thickness of the low-carbon steel container (or overpack) was increased to 12 cm to allow for corrosion. The loading per waste package was increased to the number of fuel rods from ten PWR fuel assemblies, which produces a heat generation rate of 5.5 kW at emplacement. The fuel canister of stainless steel was divided into six inner, six middle, and six outer compartments with respect to the radius. Each inner compartment contains 44 rods; each middle compartment holds 135 rods; and the outer compartments hold 264 rods. The dimensions and some performance characteristics of the Alternate I, Alternate II, and the original reference designs are summarized in Table A.3.

The current reference design (WEC 1986) for a waste package containing spent fuel evolved from the previous design studies (WEC 1983b). It specifically represents a variation of the early Alternate II design; the all-steel overpack concept eliminated the need for a TiCode-12 protective layer on the overpack steel reinforcement.

The major differences are the elimination of the boxed concept, the increase in the heat load by increasing the number of disassembled and consolidated fuel assemblies to be contained from 10 PWRs or 24 BWRs to 12 and 30, respectively, and changing the internal arrangement of the fuel rods. Sizing studies showed that canisters containing these numbers of rods will have very similar minimum diametral envelopes and that a standardized canister envelope is viable. A schematic of the overpack design is shown in Fig. A.12. A cross section of a typical canister for consolidated spent fuel is shown in Fig. A.13. The canister is segmented into six equal pie-shaped compartments that each hold the rods from either two PWR or five BWR fuel assemblies. The vacant space at the center of the canister accommodates a bar that extends from the canister baseplate for handling purposes. The characteristics of the current waste package design are listed in Table A.4, along with those for the current conceptual design for high-level waste, which is discussed in the next section. The emplacement method is schematically indicated in Fig. A.14.

A.3.2.2 Commercial High-level Waste

As in the case of spent fuel disposal, a reference conceptual design for a waste package and two alternate designs for the disposal of CHLW in vertical boreholes in a salt repository were developed in the earlier study (WEC 1983b). In these alternate designs, CHLW is processed into a borosilicate glass form and poured into stainless steel canisters, which were to be overpacked prior to disposal. The major difference in the original reference design for the canister and the alternate designs is the size, as illustrated in Fig. A.15. The larger canisters of the Alternate I and II designs have a heat generation rate of 9.5 kW each at emplacement, while those of the smaller original reference design have a rate of 2.21 kW. The Alternate II canister design is now the current reference design for the canister, as shown in Fig. A.15. The canister for the original reference design is overpacked with 0.25 cm of TiCode-12; reinforcement is provided by 6 cm of low-carbon steel. A schematic of the emplaced waste package is shown in Fig. A.15. The method of emplacement and components of the waste package for the Alternate I conceptual design are as shown in Fig. A.14, except for the increased diameter and length to accommodate the larger waste canister.

Table A.3. Early waste package design characteristics[a] for spent fuel

	Original reference		Alternate I		Alternate II	
	PWR	BWR	PWR	BWR	PWR	BWR
Waste form						
Diameter, cm	43	49	58	60	57	60
Length, m	3.85	4.11	4.09	4.32	4.10	4.35
Weight of waste, kg U	2766	3402	2766	3402	4610	4536
Total waste form weight, kg	4850	6260	4950	6360	8590	8750
Waste package						
Overpack material	TiCode-12		TiCode-12		Carbon steel	
Outside diameter, cm	57.5	64.5	76.5	80.5	83.5	88
Length, m	4.2	4.45	4.44	4.78	4.48	4.78
Empth weight, tonnes	4.0	4.8	7.7	8.6	10.4	11.9
Loaded weight, tonnes	8.9	11.1	12.7	15.0	19.0	20.7
Heat load, W	3300	3420	3300	3420	5500	4560
Repository						
Package pitch, m	10.0	10.0	10.0	10.0	10.4	8.5
Tunnel height, m	7.2	7.2	7.4	7.4	7.4	7.4
Tunnel width, m	4.0	4.0	4.0	4.0	4.0	4.0
Borehole diameter, cm	64	68	84	86	89	93
Borehole depth, m	5.6	5.6	5.8	5.8	5.9	5.9
Radiation						
Surface of overpack, mrem/h	1.1x10^{6}	<PWR	2.6x10^{5}	<PWR	8.5x10^{4}	<PWR
Surface of tunnel, mrem/h	1.4	1.3	1.3	1.3	1.4	1.4
Corrosion						
Allowance, cm	0.25	0.25	0.25	0.25	2.5	2.5
Predicted depth at 10^3 years for:						
Unlimited brine, cm	0.011	0.011	0.011	0.011	2.5	2.5
Expected brine, cm	<0.011	<0.011	<0.011	<0.011	~0.075	<2.5
Structural component thickness						
Cylinder wall, cm	7	~7	9	~9	12	12
Actual top head, cm	20	20	20	20	20	20
Actual bottom head, cm	12	~12	16	16	18	18
Peak temperatures						
Waste form, °C	372	372	372	372	258	258
Overpack, °C	110	110	110	110	146	146
Salt, °C	110	110	110	110	146	146
Local areal load, W/m^2	12	12	12	12	18.75	18.75

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[a] Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, 1983.

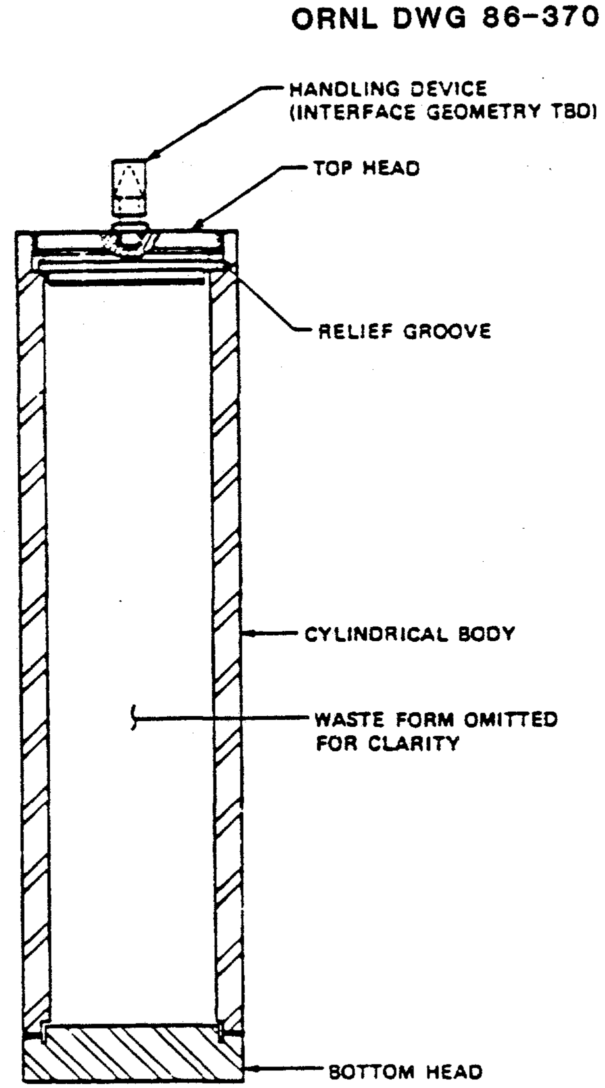

Fig. A.12. Current conceptual waste package overpack design. Source: Westinghouse Electric Corporation, Waste Package Reference Conceptual Designs for a Repository in Salt, BMI/ONWI-517, 1986.

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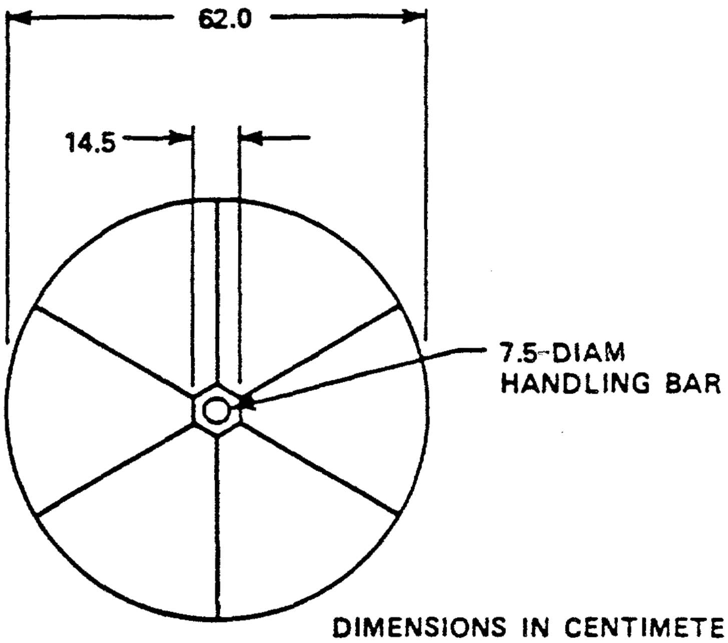

DIMENSIONS IN CENTIMETERS

Fig. A.13. Typical consolidated spent fuel canister cross section for current reference design. Source: Westinghouse Electric Corporation, Waste Package Reference Conceptual Designs for a Repository in Salt, BMI/ONWI-517, 1986.

Table A.4. Current conceptual waste package design characteristics[a] for spent fuel and commercial high-level waste

	Spent fuel		High-level waste
Design parameter	PWR	BWR	(CHLW)
Waste canister:			
Outer diameter, cm	62	62	56
Thickness, cm	0.2	0.2	6.95
Length,[b] cm	400	435	409
Material[c]	Low-carbon steel	Low-carbon steel	Stainless steel
Overpack:			
Center diameter, cm	84.5	84.5	76.3
Thickness, cm	10	10	8.9
Length,[b] cm	446.5	481.5	446.8
Material[c]	Low-carbon steel	Low-carbon steel	Low-carbon steel
Component weights:[b]			
Waste form, kg	7920	8310	2560
Canister, kg	470	500	865
Overpack, kg	9250	9890	7285
Total package, tonnes	17.6	18.7	10.7
Heat loads:			
Canister, W	6.6	5.7	9.5
Local areal, W/m^2	14.9	12.8	15.0
Repository dimensions:			
Package pitch, m	22.14	22.14	24.17
Tunnel height, m	6.40	6.40	5.79
Tunnel width, m	4.57	4.57	4.57
Rib width, m	20.1	20.1	26.2
Borehole diameter, m	0.90	0.90	0.82
Borehole depth, m	5.86	6.22	6.37
Peak temperature:			
Waste form, °C	348	<PWR	480
Overpack, °C	175	<PWR	230
Salt, °C	175	<PWR	230
Waste form at 10^3 years, °C	99	<PWR	60
Radiation (max.):			
Canister surface, mrem/h	2.85×10^7	2.80×10^7	1.51×10^8
Overpack surface, mrem/h	1.9×10^5	<PWR	1.6×10^8
Tunnel floor, mrem/h	1.8	<PWR	1.6

Table A.4 (continued)

Design parameter	Spent fuel		High-level waste (CHLW)
	PWR	BWR	
Corrosion:			
Allowance, cm	2.30	NA[d]	1.92
Penetration at 10^3 years for:			
Expected brine, cm	0.07	<PWR	0.14
Unlimited brine, cm	2.30	<PWR	1.92

[a] Source: Westinghouse Electric Corporation, Waste Package Reference Conceptual Designs for a Repository in Salt, BMI/ONWI-517, 1986.

[b] Representative values only. This value is design-specific and will vary somewhat.

[c] Equivalent to AISI 1018-type steel.

[d] NA = not available.

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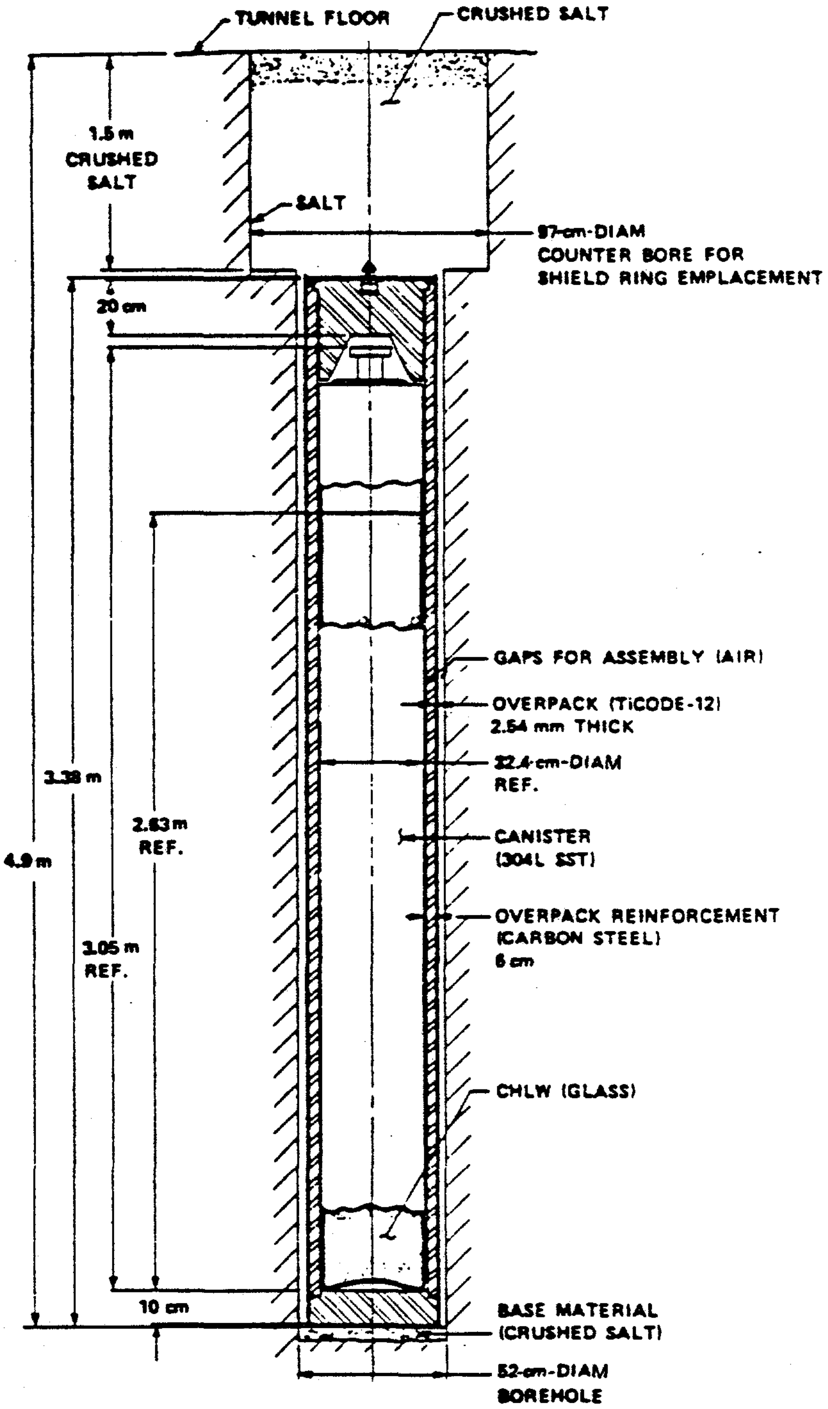

Fig. A.14. Original reference design for CHLW with borehole emplacement. Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, 1983.

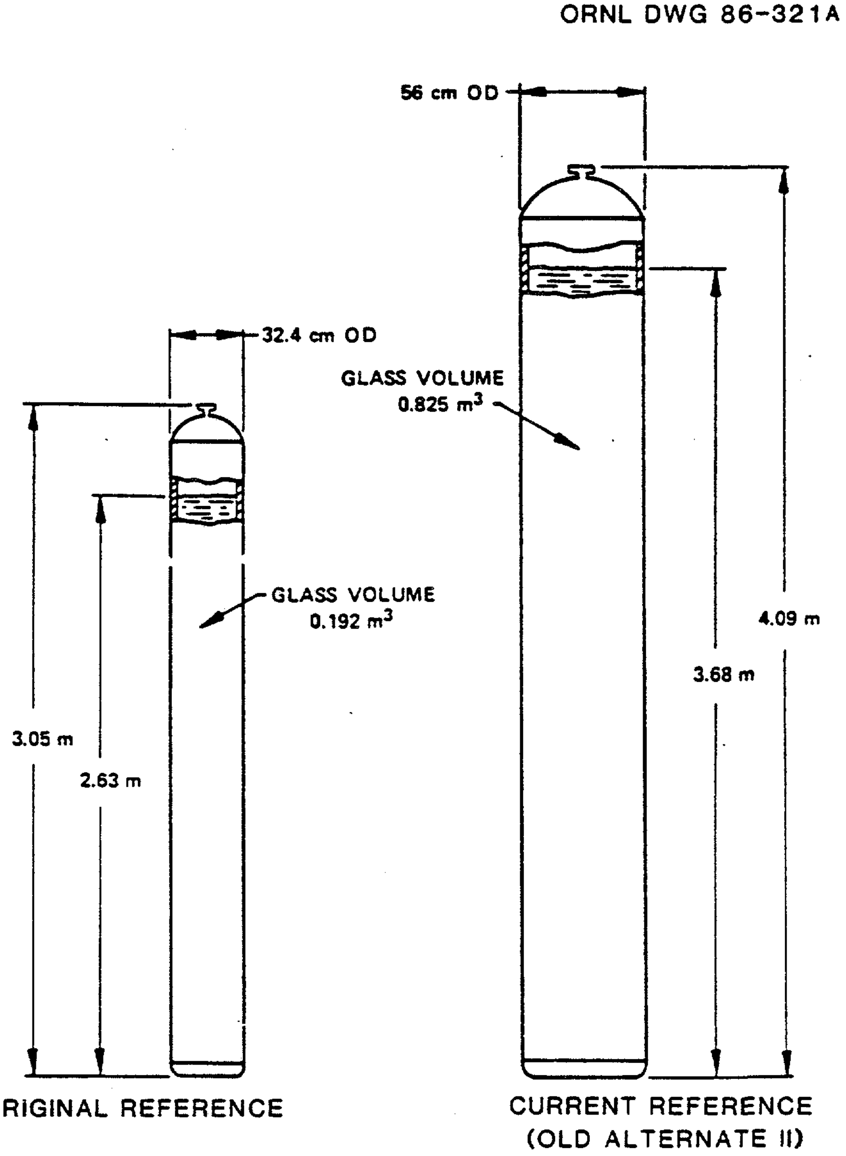

Fig. A.15. Schematics of the original and current reference designs for CHLW. Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, 1983.

The original Alternate II conceptual design is similar to the Alternate I design except that the TiCode-12 cladding has been eliminated and the steel overpack has been increased to 15 cm in thickness to provide a corrosion allowance. The dimensions and some characteristics of the original reference and the Alternate I and II designs are summarized in Table A.5. The characteristics of the current conceptual designs for the canister and the overpack are shown in Table A.4.

A.3.3 Degradation and Nuclide Release Scenarios

Barring unanticipated events, groundwater cannot be expected in a salt repository in the time frame of concern. Water is available, however, in the form of brine inclusions that can migrate up the thermal gradient toward the waste package and collect in the space between the waste package and host rock that is filled with crushed salt. This brine is very corrosive, and eventually sufficient metal will be converted to the oxide, which will weaken the overpack to the point where the developing lithostatic pressure (see Sect. C.4) can cause a large breach in the waste canister and significant reaction between the glass waste or spent fuel will begin.

Using the Jenks equation (JENKS 1972 and Appendix F) as the basis for calculating brine migration rates, the total brine accumulations per waste package for the current reference designs have been estimated at 0.85 and 0.41 m^3 for CHLW and PWR spent fuel, respectively, when assuming a threshold temperature gradient for brine migration and 0.95 and 0.75 m^3, respectively, without a threshold gradient (DOE 1984c). It was also assumed that the concentration of the brine inclusions was 5 vol %, a factor of 2.8 greater than the estimated total water content of 1.8 vol %. (The brine migration rate is considered to be directly proportional to the water content.) The rate of brine flow decreases with time, but the highest initial rate that was calculated was only 0.06 m^3/year. For these relatively small brine migration rates, the supply of brine to the surface of the waste package should govern both the corrosion rate and the rate of release of the solubility-limited radionuclides from the waste canister.

In the postclosure period after the entrapped oxygen has been consumed in the oxidation reactions, the only oxygen available for corrosion is in the in-migrating brine. There is a possibility that this supply could be insufficient to consume enough of the steel overpack to produce a significant breach in the waste package. However, an acceptable demonstration of such a condition may not be possible.

Assuming that contaminated brine has collected around a waste package, diffusion along grain boundaries is the only process available for transporting the brine away from the repository area without flow pathways being opened by shaft seal failures or tectonic events. Theoretically, a brine inclusion with sufficient vapor phase can migrate down a thermal gradient (see Appendix F), but it is difficult to visualize the initiation of this transport process with contaminated brine. It is even more difficult to visualize the maintenance of the movement in the cooler temperatures and smaller thermal gradients that exist away from the repository.

In the unexpected event of an intrusion scenario, a possible source of groundwater is the Ogallala/Dockum aquifer. The piezometric potentials are such that any flow into the repository would come from the aquifer above. In view of the possibility of such an event, corrosion calculations were made based on an unlimited supply of brine with a composition

Table A.5. Early conceptual waste package design characteristics[a] for commercial high-level waste

	Original Reference	Alternate I	Alternate II
Waste form			
Diameter, cm	32.4	56	56
Length, m	3.05	4.09	4.09
Weight of waste, kg	595	2560	2560
Total waste form weight, kg	845	3425	3425
Waste package			
Overpack material:			
Outer	TiCode-12	TiCode-12	Carbon steel
Inner	Carbon steel	Carbon steel	--
Outside diameter, cm	47.5	79.5	89
Thickness:			
TiCode-12, cm	0.25	0.25	12
Carbon steel, cm	6	6	--
Length, m	3.38	4.48	4.57
Empty weight, tonnes	2.4	8.6	13.4
Loaded weight, tonnes	3.2	12.0	16.9
Heat load, W	2210	9500	9500
Repository			
Package pitch, m	2.48	10	10
Tunnel height, m	6.1	7.2	7.2
Tunnel width, m	3.8	4.0	4.0
Borehole diameter, cm	52	82	94
Borehole depth, m	4.9	6.3	6.3
Radiation			
Surface of overpack, mrem/h	5.2×10^5	9.3×10^5	1×10^5
Surface of tunnel, mrem/h	1.7	1.5	1.5
Corrosion			
Allowance, cm	0.25	0.25	5.0
Predicted depth at 10^3 years for:			
Unlimited brine, cm	0.05	0.03	4.9
Expected brine, cm	<0.05	<0.03	0.07
Structural component thickness			
Cylinder wall, cm	6	10	15
Top head, cm	20	20	21
Bottom head, cm	10	16	21
Peak temperatures			
Waste form, °C	322	476	476
Overpack, °C	233	250	250
Salt, °C	233	250	250

Table A.5 (continued)

	Original reference	Alternate I	Alternate II
Local areal load, W/m^2	37.5	30	30

[a]Source: Westinghouse Electric Corporation, Advanced Energy Systems Division, Engineered Waste Package Conceptual Design: Defense High-Level Waste (Form 1), Commercial High-Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Salt, ONWI-438, 1983.

determined by the dissolved halite (DOE 1984c). For transport of the contaminated brine to the accessible environment by means other than diffusion, some sink has to develop to complete a flow path, a condition that should be evaluated in a safety assessment of the repository.

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APPENDIX B

IDENTIFICATION AND PRIORITIZATION OF WASTE PACKAGE FAILURE MODES

In considering the failure modes of the materials in a waste package, the reference design is assumed to be the current concept for disposal in basalt, tuff, and salt, as described in Appendix A. It is also assumed that the waste package will not be subjected to extremely unlikely catastrophic events that can destroy the waste package (e.g., volcanism, large meteorite impacts, and severe tectonic events) because of the emphasis of NRC regulations [10 CFR 60, Sect. 60.113(a)(ii)] on anticipated processes and events.

The potential failure modes that have been postulated for each component of the waste package and their relevance in meeting the requirements of 10 CFR 60 are discussed individually below. It does not follow that all of these modes are very probable; instead, they represent a rather exhaustive list of possibilities to consider in a safety analysis report (SAR). The discussion and analysis of failure modes that follow should not be construed as a final NRC position with regard to waste package reliability, but it is in line with the current NRC generic technical position (NRC 1986) and with the suggested procedure on Waste Package Reliability by Sastre (1986), which is quoted as follows:

> In the SAR, the applicant should list all possible, identified failure modes of each waste package component and their retention or dismissal for further analysis. This preliminary analysis, generally called Failure Mode and Effects Analysis — FMEA, is qualitative in nature. It is expected to result in the reduction of the set of possible failure modes to only those which are relevant under the range of repository conditions identified in Sect. 2.1.2 [of the Sastre report]. This set of significant failure modes will be called design failure modes. In the dismissal of potential failure modes, the applicant should consider the natural variability of environments to which the package will be exposed. The dismissal of any given failure mode should be discussed and documented.

B.1 DEGRADATION, OR FAILURE, MODES FOR THE OVERPACK AND STAINLESS STEEL CANISTER

The degradation, or failure, modes for the metallic overpack, or the nonoverpacked stainless steel canister in the case of disposal of spent fuel elements in tuff, can be classified as thermally/mechanically, chemically, or biologically induced corrosion. It is assumed that a quality control and assurance program will generally preclude poor workmanship and materials that do not meet specifications.

B.1.1 Thermal/Mechanical Failure Modes

In considering the various mechanical failure modes, it is assumed that corrosion has not substantially degraded the overpack or the non-overpacked stainless steel canister in the case of disposal in tuff. After substantial corrosion has occurred, breaching of the canister can be expected and the mechanism (pitting, cracking, shearing, etc.) becomes irrelevant.

B.1.1.1 Force and/or Temperature-Induced Elastic Deformation

Elastic deformation is not a failure mode per se, but excessive deformations under certain conditions could lead to a failure mode even though the average stresses remain in the elastic range. In discussing the conditions that could lead to a failure mode, it is assumed that deformations occur as a result of an application of force (external and/or thermally induced) and that the removal of this force will usually permit the member to return to its original shape (i.e., behave elastically). It is customary to design structural members with average stresses sufficiently below the yield point to generally preclude excessive deformation and failure in the elastic range. For such a design condition, localized stresses exceeding the yield point could exist at notches, bolt-holes, stringers, or other flaws. Ductile materials can redistribute these localized stresses without failure occurring. With brittle materials, however, localized stresses can continue to increase, even when there is no local yielding, until a crack forms that can spread rapidly over the section. Failure in the elastic range is also possible from sudden deflection or buckling under conditions of instability, or from fatigue and fretting when the load becomes cyclic. These potential failure modes in the elastic range and brittle fracture are discussed below under the appropriate headings.

B.1.1.2 Yielding

Yielding, or plastic, deformation occurs when the applied load causes the elastic limit of the material to be exceeded. When the load is relieved, the recovery is along a path different from the loading one, which results in a permanent set or elongated shape. Additional loading will cause an increase in the yield point, a phenomenon known as strain or work hardening. Yielding of the material could lead to improper functioning of the component and subsequent failure of the structure. However, in a ductile metal under conditions of static loading at moderate temperatures, yielding rarely results in fracture in properly designed structures because the metal strain hardens as it deforms. At temperatures significantly higher than room temperature, metals no longer exhibit strain hardening and can continue to plastically deform by the mechanism denoted as "creep" (discussed under same heading below). While the temperature at which strain hardening stops is not accurately known, it is generally about one-third of the melting-point temperature (in absolute units, K or R). However, for ordinary mild steel it is about 450°C (DAVIS 1982a).

Sufficiently large stresses will cause failure after large deformations either via a ductile tensile or shear rupture, brittle fracture after strain hardening, or an accelerating or tertiary creep. However, barring unexpected strong tectonic activity, it is difficult to foresee that a well-designed and sited repository will develop stresses that can cause rupture or fracture of an overpack before considerable degradation has occurred through corrosive action. However, for disposal in tuff, the stainless steel canister may be subject to stress corrosion cracking before significant degradation occurs as the result of general corrosive action (see Sect. B.1.2.2 for discussion).

B.1.1.3 Ductile Rupture

Any metal that remains ductile while being strained will undergo ductile rupture when stressed beyond its tensile, or ultimate, strength. Typically, ductile rupture or failure results after gross plastic deformation. With ductile rupture a number of possible structure patterns can develop on the fracture surface, whereas a brittle fracture produces little or no plastic deformation and is always characterized by a lustrous appearance. The customary design technique of limiting stresses to less than the yield stress generally eliminates the likelihood of a ductile rupture during the expected lifetime of a structure or equipment. In the case of a waste package, ductile rupture (although not credible in the first few hundred years) will become highly probable after considerable degradation by corrosive processes.

B.1.1.4 Brittle Fracture

It is well known that low-carbon steel under load is subject to brittle fracture at low temperatures under certain conditions of stress application. The primary factors that influence brittle fracture in steel include average and localized stress levels, temperature, and flaw size. All commercial steel products have microcracks or inclusions that act like cracks from the viewpoint of stress concentration. Since flaw size is somewhat imponderable and stress level can be controlled by design practice, service temperature has become the criterion for the avoidance of brittle fracture. Stainless steel, however, is not subject to brittle fracture in the classical sense at repository temperatures, or even significant hydrogen embrittlement.

A particular mild steel can exhibit both ductile and brittle behavior, depending on the temperature. The origin of the classical brittle fracture problem (rapid propagation of a crack across a loaded member) is the decrease in fracture toughness resulting from a change in the fracture mode from high-energy ductile tearing to low-energy absorption cleavage fracture in a rather narrow range of temperature. The full span of the transition is in the order of 67°C. However, the transition range of engineering interest for conventionally loaded members usually occurs over a range of 6 to 34°C (PELLINI 1964).

It is possible to construct a crack arrest temperature (CAT) curve from laboratory test data by plotting nominal stress vs temperature beyond which fractures will not propagate regardless of the flaw size (PELLINI 1964). The reference is the "nil ductility temperature" (NDT), which can be established directly by drop-weight tests with good accuracy. Below the NDT, a fracture becomes 100% cleavage and a minute crack or flaw, irrespective of size, could initiate a brittle fracture (provided that the yield point stress was reached at the flaw) that could propagate through the surrounding region even though the general stress level was below the yield point. As the flaw size increases, there is a decrease in the stress required to initiate brittle fracture and an increase in the temperature required to prevent propagation. Below a nominal stress of about 3.4 to 5.5 MPa (5000 to 8000 psi), however, brittle fractures probably will not occur, since there is insufficient elastic strain energy available to support propagation of fractures (DIETER 1976).

The lowest temperature for which fracture is 100% fibrous, or totally ductile, is called the "fracture transition plastic" (FTP) temperature, which occurs at the intersection of the tensile strength curve and the CAT curve. Cracking does not occur above this temperature; failures result in large deformations with subsequent-ductile tearing. For practical purposes the FTP temperature is equal to the NDT + 67°C.

Between the NDT and the FTP temperatures, the cracks exhibit a mixture of cleavage and fibrosity or shear. A number of transition temperature criteria are possible in this range [e.g., the 50% cleavage—50% shear point called the "fracture appearance transition temperature" (FATT) and the "fracture transition elastic (FTE) temperature"]. This latter temperature, which is the intersection of the yield strength curve and the CAT curve, is approximately equal to the NDT + 34°C. It represents an important design point for most engineering structures. It has been established experimentally that a brittle fracture cannot propagate across any region that is loaded within the elastic range (average stress below yield point). Since design practice generally requires that average stresses not exceed the yield point in any extensive region, only small localized areas of excessive stress (assuming no long stringers are present) would exist due to some stress riser, such as a flaw. A fracture might be initiated under such a condition, but it would not propagate if the region temperature was above the FTE temperature. Consequently, maintaining the service temperature above the FTE temperature (which should present no problem for a basalt repository) will provide almost certain protection against the classical type of brittle fracture.

It is possible that ductile steel can be embrittled (increase in the NDT) by work hardening, as the result of successive stress-deformation cycles, and by neutron irradiation, which occurs in reactor pressure vessels; however, these conditions will not exist in a waste repository. A possibility does exist, of course, that hydrogen embrittlement could occur (see Sect. B.1.2.8), thus leading to brittle fracture or stress corrosion cracking. In the authors' opinion, however, it does not appear likely that fracturing could occur prior to extensive degradation of the overpack, with subsequent embrittlement by some mechanism.

B.1.1.5 Fatigue

Repetitive, or fluctuating, stress applied to metals in a sufficient number of cycles can cause fracture even though the maximum stress is far below the ultimate strength and in the elastic range. Such fractures are called fatigue failures because they generally occur under dynamic loading after a considerable period of service. Physically, a minute crack originates at a stress riser such as a surface scratch or other flaw. The crack then enlarges during the cyclic loading until there is insufficient material left ahead of the crack front to support the load; subsequently, failure occurs.

The three basic factors necessary to cause fatigue failure include: (1) a sufficiently high maximum tensile stress, (2) a sufficiently large variation in the amplitude of the applied stress, and (3) a sufficiently large number of cycles. Other variables, such as stress concentration, corrosion, temperature, overload, metallurgical structure, residual stress, and combined stresses that tend to alter the conditions for fatigue failure, are also involved (DIETER 1976).

Although the basic causes of fatigue in metals are not completely understood, a mass of empirical data that permit safe engineering design exists. However, even with careful design and testing materials, a large percentage of actual failure in structures and machinery can be traced to some type of fatigue. The general types of fatigue failure, which are discussed below, include: high cycle, low cycle, thermal, corrosion, and fretting.

The conventional method for presenting high-cycle engineering fatigue data is by the S-N curve, which is a plot of stress range (S) vs the number of cycles to failure (N). Most laboratory determinations of fatigue properties have been made for reverse stressing (bending or torsional), where the mean stress is zero. However, nonzero mean stress data are also obtained, depending on the engineering application. Characteristically, the S-N curve shows an increasing fatigue life with decreasing stress. For some engineering materials, such as steel, the S-N curve becomes horizontal at a limiting stress. Below this stress, which is called the fatigue (or endurance) limit, the material presumably can endure an indefinitely large number of cycles without failure. In high-cycle fatigue tests ($N = >10^5$ cycles) for engineering applications, the stress on a gross scale is elastic, but the metal deforms in a localized way and failure occurs without any obvious warning.

Low-cycle fatigue ($N = <10^4$ cycles) failure occurs at relatively high stress and a low number of cycles. This type of failure must be considered in the design of steam turbines and other types of power machinery since low-cycle fatigue conditions are frequently created in which the repeated stresses are thermal stresses resulting from expansion and contraction of the metal. Consequently, low-cycle fatigue failure results from cyclic strain rather than from cyclic stresses, as in the case of high-cycle fatigue failure. In low-cycle fatigue testing, this is started by pulling the specimen into the inelastic range and then cycling the load under strain control. The equivalent S-N curve is constructed using the plastic strain range in place of the stress range.

In determining S-N curves, considerable random scatter is found in the data. Consequently, fatigue life and fatigue limit are frequently treated as statistical quantities that require considerably more data for obtaining the necessary statistical parameters than are required for constructing the usual S-N curves. The statistical problem of determining the fatigue limit is complicated by the destructive method of testing. If a specimen fails, it cannot be retested. It is only known that the fatigue limit is lower, or greater, than the stress employed. Consequently, groups of specimens are tested at several stresses to find out how many fail at each level. Two statistical approaches, called "probit analysis" and the "staircase method," are used (DIETER 1976). The procedures for applying these approaches are well established and reasonably standardized (ASTM 1963).

Fatigue failure can be produced by fluctuating thermal stresses even when no stress is produced by mechanical causes. Thermal stresses result when the thermal expansion of a member is restrained in some fashion. When restrained metallic members are exposed to cyclic heat sources or sinks, fatigue failure can result from mechanisms similar to those involved in low- and high-cycle fatigue failure. Such conditions for failure are frequently present in high-temperature equipment. Austenitic stainless steels are particularly sensitive to thermal fatigue because of their relatively low thermal conductivities and high thermal coefficients of expansion (DIETER 1976).

Corrosion fatigue is defined as the behavior of metals when they are subjected to cyclic stress and simultaneously exposed to a corroding environment. The separate damage caused by static corrosion or by fatigue offers little indication of the greatly accelerated damage that can result when these two phenomena act simultaneously. With ordinary corrosion, the corrosion products often retard, or suppress, further reactions. When cyclic stress is superimposed, cyclic strains tend to disrupt any protective film, and the surface pitting or roughening caused by corrosion tends to reduce the mechanical strength and susceptibility to cracking as the pits become sharp enough to produce a high stress concentration. In this sense, cyclic stress accelerates the corrosion by a more or less mechanical action, but the influence on the chemical corrodibility of the material is usually small (LESSELS 1954).

Fretting is the surface damage that results when two hard surfaces in contact experience some relative periodic motion. It produces a wearing action on metal and can also simulate corrosion fatigue. Prolonged fretting will produce surface cracks that can grow and cause a high-cycle fatigue failure (WATERHOUSE 1981).

Fretting damage and failure frequently occur on shafts with press-fitted hubs or bearings. The damage is caused by a small amount of slip deformation (in the order of 10^{-6} in.) that is virtually impossible to eliminate completely. Examples of fretting in other than machinery include damage to wire ropes and cables, flanges, and pin-lug combinations. A thorough treatment of fretting damage, including theory, methods of mitigation, and examples of in-service failures is given by Waterhouse (1981).

In basalt, tuff, or salt repositories, stress cycling of a waste package by external mechanical means is not important. Some slight thermal cycling may occur during the operational period due to repository temperature fluctuations, but this would be insignificant with respect to inducing cyclic stresses. After backfilling of the repository is complete, a more likely (but still improbable) scenario for inducing thermal cycling or mechanical vibration would be a slugging action caused by water vaporization, condensation, or motion. However, such a phenomenon could not exist for more than a few tens of years because boiling would be suppressed by the increasing hydrostatic head in the case of a basalt repository and the decreasing temperature at the surface of the overpack or canister after backfilling transients have been damped out in all three repository types.

B.1.1.6 Creep

Creep is the continuing plastic deformation that occurs when a material is subjected to a load or stress. In determining the engineering creep curve of a metal, a constant uniaxial tensile load is usually applied to a specimen maintained at a constant temperature and the strain is measured as a function of time. Three distinct periods can often be observed: (1) transient creep, with the rate decreasing with time; (2) secondary or steady-state creep; and (3) tertiary or accelerating creep leading to rupture. Creep rates in metals increase both with temperature and with applied stress, which, at high levels, can result in tertiary creep. This third creep stage occurs when there is a localized reduction in cross-sectional area because of necking or void formation. In metallic structures and machinery, steady-state creep could lead to instabilities and mechanical weakening. Tertiary creep is usually not possible in structures or machinery since other failure mechanisms would take precedence or sufficient constraints would be present. Tertiary creep can be a problem in metalworking, which involves high temperatures and stresses.

The understanding of the high-temperature behavior of material is usually insufficient to predict creep and stress-rupture behavior from theoretical models; good empirical data are necessary. It is a common practice to speak of "creep strength" or "rupture strength." Creep strength is the stress at a given temperature that produces a given steady-state creep rate, usually taken as 10^{-5} or $10^{-3}\%$ per hour. Alternatively, the creep strength may be defined as the stress to cause a creep strain of 1% at a given temperature. Rupture strength refers to the stress at a given temperature to produce a rupture in a predetermined time, usually 10^3, 10^4, or 10^5 h.

At the expected temperatures, pressures, and induced stresses that a waste package will experience, metal creep should be insignificant and, in fact, may be beneficial in that residual and other local stresses would tend to be relieved.

B.1.1.7 Thermal Relaxation

Thermal relaxation is important when a component or structural member is intended to carry a preload by means of a fixed deformation (e.g., torqued bolts, spring loading, and shrink fits), but the preload stress is relieved during operation because of a change in stress and strain from a temperature change or creep. A waste package design that requires preloaded members cannot be visualized by the authors; consequently, it seems improbable that such a design would be a consideration in performance assessment.

B.1.1.8 Stress Rupture

The term "stress rupture" is misleading since it encompasses all the failure mechanisms. Stress rupture is usually applied to creep-rupture situations. The stress-rupture test is basically similar to creep tests except that it is always carried out until the material ruptures. Thus, it represents an accelerated creep test. The higher stresses and creep rates of the stress-rupture test cause structural changes to occur in shorter times than are observed for ordinary creep tests. It has direct application in design where creep deformation can be tolerated but fracture must be prevented (e.g., high-temperature strength of alloys for jet-engine applications). Stress rupture tests are designed to provide data that can be used to guard against rupture at high temperatures when creep is significant (DIETER 1976).

B.1.1.9 Spalling

Spalling, a term usually applied to rocks and ceramic materials, is a brittle-type fracture that results in chipping, fragmentation, or flaking from a material. Spalling of metals can occur under only unusually severe conditions (e.g., creating shock waves with explosive charges), which would not exist in the operational or postclosure phases of a repository. Spalling of hard rocks such as basalts and granite, and possibly welded tuff, can occur if the existing temperature gradients are high enough to produce thermal stresses that exceed the fracture strength of the rock. Compressive stresses applied to the surface will inhibit spalling.

B.1.1.10 Buckling

Buckling is a failure mode that involves the geometry and mechanical stability of the system. Failure occurs when the deflections of a member become too large for structural stability even though deformations in the metal are within the elastic limit. Classical buckling conditions involve long beams, columns, thin-walled cylinders, and thin plates with an axial load that exceeds the critical load (i.e., the load that causes the column to deflect beyond the stability region with subsequent buckling or collapse). Basically, instabilities result from large bending moments on application of a load with a small eccentricity. Application of a transverse load will increase the critical load and prevent buckling if the transverse load is sufficiently high. Buckling in torsion can also occur. When the critical torque is reached, one or more wrinkles appear (theoretically on a 45° diagonal) due to buckling failure on compression. Crushing of a hollow geometric configuration (such as a submarine) by traverse forces is also considered as a buckling phenomenon.

An overpacked canister (except for the void located at the top 15% of the canister volumes, which could be braced or filled with some inert material such as sand) represents a solid cylinder that is relatively stocky (i.e., has a low slenderness ratio) and, as such, is not susceptible to the classical buckling-type failure prior to excessive degradation of the waste canister with subsequent removal of significant quantities of the internal support (i.e., the waste material). If thin cladding (such as 100 mils of TiCode-12), which is under consideration for a salt repository, were wrapped around the steel overpack and some critical clearance developed between the cladding and the overpack as the pressure increased, a buckling failure in the form of wrinkling or dimpling of the cladding could occur. Such a failure should be preventable by proper fabrication procedures that preclude the formation of a critical clearance.

B.1.1.11 Creep Buckling

Geometries with low slenderness ratios that will not buckle in the elastic range can fail in the inelastic range (i.e., after some yielding). With sufficient compressive stresses and sufficiently high temperatures, cumulative creep deformation due to long-term loading can create geometric instabilities that lead to failure of the structure. As previously discussed, creep should not be significant and may be beneficial in relieving stresses.

B.1.2 Chemical Failure Modes (Corrosion)

All possible chemical failure modes of overpacks and canisters involve oxidation of a metal by the environment, which, by definition, is corrosion. One possible exception is hydrogen attack or embrittlement, but even this form of degradation may have its origin in the corrosion process.

In this section, corrosion in geologic repositories constructed in three different rock types is considered: basalt, tuff, and rock salt. The environment to which the canisters/overpacks are exposed includes the host rock, groundwater, backfill, and corrosion products — all in the presence of ionizing radiation and at elevated temperatures and pressures and differential stresses. Because the environment is different in the various rock types, the materials used to enclose the waste may also differ. Regardless of the repository, however, two classes of materials are considered: those that are relatively inexpensive and would be expected to corrode at an appreciable rate, and those that are nearly passive in the environment but are expensive. The former type includes the steels and cast iron and would be used as thick-wall containers, whereas titanium and high-nickel alloys are examples of the latter and would probably be used as relatively thin cladding on a less-resistant structural material.

Only the one or two materials that are currently favored for overpacks will be considered here. In all repositories, a low-carbon austenitic stainless steel is specified for the canister. Either steel or cast iron is the reference material for the overpack in a basalt repository (WEC 1982). The overpack for canisters in salt will probably be made of either steel or steel clad with a titanium alloy (WEC 1983). In tuff, the repository location will be 300 to 400 m below grade, but at least 100 m above the permanent water table (O'NEAL 1984). Spent fuel element canisters will probably be used without an overpack in tuff, but the canisters containing glass may be overpacked; a low-carbon austenitic stainless steel is the material of choice for the overpack (McCRIGHT 1984a). To facilitate retrievability from the tuff repository during the first 50 years, the boreholes will be fitted with a liner, made of mild steel or possibly an austenitic stainless steel (O'NEAL 1984).

General descriptions of the different types of corrosion that could lead to failures of overpacks and canisters are presented below. Following each, a discussion of corrosion of the materials specified for overpacks in the three environments is included. While the present report will repeat some of the information presented earlier by Claiborne (1985) concerning corrosion in a basaltic environment, it will also incorporate information generated since that time. Canister corrosion is discussed in Sect. B.1.2.9.

Table B.1 lists analyses of representative groundwaters in repositories. The compositions of the brines are taken from those given by Molecke (1983). It is noted that Brine A is representative of brines that would result from intrusion of water into a bedded salt repository in the vicinity of magnesium and potassium minerals and that Brine B is representative of brines that would be expected if water entered domed salt or bedded halite. Brine A is similar to brine inclusions in bedded salt crystals. It has been estimated that these inclusions contain about 64,000 ppm Mg, 34,000 ppm Na, 257,000 ppm Cl, and 4000 ppm Br (JENKS 1972). The tuff groundwater listed in Table B.1 is from Well J-13 at Jackass Flats, Nevada, and is

Table B.1. Representative compositions of solutions in salt, basalt and tuff repositories

	Ionic composition (ppm)			
	Salt[a]			
Ion	Brine A	Brine B	Basalt[b]	Tuff[c]
Na	42,000	115,000	250	51
K	30,000	15	1.9	4.9
Mg	35,000	10	0.4	2.1
Ca	600	900	1.3	14
Cl	190,000	175,000	148	7.5
SO_4	3,500	3,500	108	22
HCO_3	700	10	70	120
CO_3			27	0
Br	400	400		
F			37	2.2
NO_3				5.6
SiO_2			65	61
pH	6.5	6.5	9.8-10	7.1

[a] From Molecke (1983).

[b] From Wood (1980).

[c] From Westerman (1982). Water from Well J-13, Jackass Flats, Nevada Test Site. Water also contains 5 to 6 ppm oxygen.

expected to be representative of the water underlying much of the tuff in Nevada (WESTERMAN 1982). The composition of the basaltic groundwater is that for the water in the basalt at the Hanford, Washington, reservation (WOOD 1980).

B.1.2.1 General Corrosion (SHREIR 1976; UHLIG 1948)

Corrosion failures seldom occur as the result of general corrosion; they are usually the result of localized forms of attack. However, for waste repositories where container integrity is expected for periods as long as 1000 years, general or uniform corrosion must be considered and an appropriate corrosion allowance must be designed into the overpack and/or canister. This is particularly important for the less-resistant materials such as steel.

Since all practical engineering metals and alloys are thermodynamically unstable in repository environments (copper may be stable under some conditions), the successful use of these materials requires that the kinetics of the degradation process be controlled. Control may consist of the development of protective films or limiting the quantity of the aqueous corrosive that reaches the metal. In the former case, it must be remembered that films are not completely protective and thicken as corrosion proceeds. The possibility exists that stresses within these films or scales will ultimately cause breakup, and corrosion rates higher than predicted on the basis of short-term tests will result. In the latter case, the presence of a packing can limit the amount of solution reaching the overpack. For example, hydraulic conductivities as low as 10^{-11} to 10^{-13} cm/s have been reported for compacted bentonite (WESTIK 1983). Also in bedded salt, where migration of brine inclusions within the crystalline salt to the hot waste package is expected to be the principal source of water, corrosion rates may be limited by the rate at which the brine reaches the overpack rather than by the corrosiveness of the brine. It has been calculated (JENKS 1979) that the rate of brine migration to an overpack 35 years after emplacement of 10-year-old waste could be so small that the corrosion rate would be <<25 μm/year if the brine corroded steel uniformly.

The most important variables that affect metal corrosion are the composition of the solution to which they are exposed and the temperature. In a waste repository, it is possible to control the temperature by adjusting the quantity of radionuclides in the waste or by spacing the waste packages (i.e., area thermal loading); however, the chemistry of the groundwater in a given repository cannot be changed. The following are some general statements about how specific components of solution influence corrosion:

1. pH - In the absence of oxygen, many corrosion reactions are controlled by the reduction of hydrogen ions. Thus, corrosion of most metals is much less at pH values in the alkaline range (8 to 13) than in acid solutions. Some waters contain appreciable concentrations of multivalent cations (Mg^{2+}, Fe^{2+}) that undergo significant hydrolysis at high temperatures to yield hydrogen ions. Thus, it is possible that a water that is virtually nonaggressive at the natural repository temperature can become much more aggressive in the vicinity of a waste package.

2. Oxygen Concentration - Reduction of dissolved oxygen, if present, constitutes the primary cathodic process in neutral or alkaline solutions. Most groundwaters are devoid of oxygen, but air will be in the repository during operation. Radiation can also produce oxidizing species, including oxygen and hydrogen peroxide, that may affect corrosion.

3. Chloride Ion Concentration - Chloride ions are usually associated with localized attack, although their presence can accelerate uniform corrosion. However, in alkaline solutions, chloride ions have minimum effect on the general corrosion of most metals.

4. Other Ions - Most groundwaters contain sulfate ions, and some contain low levels of fluoride ions. The sulfate salts are generally less corrosive than chloride ions and do not usually contribute to localized attack, except perhaps in the case of crevice corrosion. Fluoride ions, particularly in acid solutions, are detrimental to the corrosion resistance of iron-base alloys, and they are particularly corrosive to metals like titanium and zirconium. In the latter cases, stable fluoride ion complexes are formed and the usual thin protective oxides do not form. Fortunately, when fluoride ion is present in groundwater, the concentration is usually low; and the groundwater is typically neutral or alkaline, where its effect is minimal.

5. Hydrogen Sulfide Concentration - Most naturally occurring groundwaters are reducing, and low concentrations of hydrogen sulfide exist in some of them. Certain anaerobic bacteria are also capable of reducing sulfate ions to hydrogen sulfide. Hydrogen sulfide usually causes an increase in the corrosion rate of steel; it can also cause an increase in the hydrogen pickup of some steels and lead to embrittlement. However, to increase the hydrogen pickup by a metal, the sulfide must be present as undissociated molecules (H_2S) that exist only in acid solutions; at pH levels above 8 or 9, hydrogen sulfide does not accelerate hydrogen uptake because the formation of HS^- begins to be favored. As the pH is increased further, complete ionization to S^{2-} occurs. The general corrosion resistance of titanium, zirconium, and austenitic stainless steels is unaffected by the presence of hydrogen sulfide.

6. Radiation Effects - The absorption of radiant energy by aqueous solutions leads to the formation of a variety of products, some of which are oxidizing in character and can influence corrosion processes. The yields and compositions of the radiolysis products are strongly dependent on the chemistry of the irradiated solution, the radiation type and intensity, and the capability of the radiolysis products for escaping from solution. In a given closed environment from which radiolysis products cannot escape, the steady-state concentrations of the products are established in short times relative to the flow of groundwater. If radiation causes an increase in the potential of the metal, the corrosion rate of steels will probably increase. With other metals, an increase in potential may lead to passivation. As discussed later, changes in potential may affect pitting and stress corrosion cracking of certain alloys. Although radiation will probably not influence the uniform corrosion of titanium in repository environments, it may adversely affect the corrosion resistance of steel and low-alloy

steels; however, the effects cannot be predicted in a quantitative fashion. It is clear, however, that tests aimed at determining the behavior of materials in repositories should be conducted in the presence of radiation.

The irradiation of moist air produces nitric acid and lesser amounts of other nitrogen oxides (LIND 1961). The steady-state concentration of nitric acid is very low; however, if a process exists for the removal of the acid, such as the reaction with a metal susceptible to attack by nitric acid, nitric acid is continuously produced. Such a situation has been observed in beam tubes associated with a nuclear reactor (PRIMAK 1954). There appears to be a critical relative humidity below which the nitric acid formed in the vapor will not attack a metal. Whether such attack will occur on heated waste packages during handling or in a repository before flooding needs to be established.

Temperature also plays an important part in determining the corrosion rate of waste packages. Like most kinetic processes, the rate of corrosion usually increases with temperature; and, as previously noted, groundwaters may also become more corrosive as the temperature increases. On the other hand, protective scales may form more readily, or have a different composition, at high temperatures than at lower temperatures. Under some conditions, the average long-term corrosion rates may be lower at high temperatures than at lower temperatures. However, experience indicates that most degradation rates increase as the temperature increases. In particular, localized forms of attack (pitting, crevice corrosion, and stress corrosion cracking) are more likely at the higher temperature.

B.1.2.1.1 General corrosion in a basalt repository

A number of studies have been conducted to determine the general corrosion resistance of iron-base alloys in simulated basaltic environments, but tests of the longest duration (up to 20 months) have been reported by Westerman (1984b). Alloys tested were 2-1/4 Cr—1 Mo cast steel, 1-1/4 Cr—0.5 Mo cast steel, 1025 cast steel (ASTM A-27, Grade 60-30), ductile cast iron (ASTM A536-77, Grade 60-40-18), 1020 wrought steel, and 9 Cr-1 Mo wrought steel. The latter alloy was only tested for 5 months. The tests were conducted in oxic Grande Ronde groundwater in refreshed autoclaves at 150 and 250°C. At 250°C, corrosion rates were high initially but decreased markedly; after 1 month, the corrosion rates were reasonably uniform among the materials and low [i.e., <8 μm/year (<0.3 mil/year)], and there was little sign of nonuniform attack. However, pits began to appear on all specimens during the period between 12 and 17 months. Similar pitting was also observed on the specimens exposed at 150°C during the 17- to 20-month interval. The average corrosion rates (based on weight loss) observed at 150°C were somewhat higher than those observed at 250°C. As the author points out, pitting of the ferrous alloys is of much greater concern than the modest penetration rates associated with general corrosion.

Tests identical to those described above were also conducted at 250°C in a gamma field with an intensity of 3×10^5 rads/h (WESTERMAN 1984b). The results showed somewhat higher corrosion rates in the presence of radiation than in the unirradiated tests. The scatter of the data and the presence of pits made quantitative comparison difficult; however, after 12 months of the same test, the presence of radiation increased the corrosion rates by factors of 2 to 3 (NELSON 1984).

Basaltic groundwater at the Hanford reservation contains measurable quantities of dissolved methane. It has been shown that irradiation of such waters produces high-molecular-weight organic compounds (GRAY 1983). The presence of these organic compounds may influence the corrosiveness of such waters either by the action of newly formed organic compounds themselves or by changing the steady-state concentrations of other radiolysis products. That such a situation can occur is illustrated by the results described briefly by Davis (1985a). Carbon steel in simulated basaltic groundwater at 150°C in a radiation field of 3.8×10^4 rads/h developed pits when the water contained methane, but no pits were detected when methane was absent. No other corrosion results were reported.

The data discussed above illustrate some of the problems involved in modeling general corrosion to predict long-term corrosion damage. Most modelers usually assume that the corrosion rate increases with temperature, but this is not always true. Furthermore, existing models have been based on short-term data and do not account for the fact that the mode of attack may change with time. It is obvious that a much larger data base than presently exists must be established before any meaningful model can be developed.

B.1.2.1.2 General corrosion in a salt repository

Ferrous alloys. Several studies have suggested that mild steel, cast irons, and low-alloy steels may have adequate corrosion resistance for use as overpacks in salt repositories. Results from some recently reported investigations are presented below. Westerman (1984a) investigated cast and wrought 1025 cast steel, 2-1/4 Cr—1 Mo cast steel, and ductile cast iron in brine solutions (122 to 134 ppm Mg) at 150°C both in the absence of radiation and at two radiation levels, 1×10^5 and 2×10^3 rads/h. In the tests without radiation, the brine contained 0.5 to 1.5 ppm oxygen; however, it was free of oxygen in the irradiation tests. Attack was not severe in the absence of radiation; the results suggested that less than 25 mm of material would be lost during a 1000-year period. Gamma irradiation at a dose rate of 1×10^5 rads/h enhanced corrosion by a factor of ~6, whereas a dose rate of 2×10^3 rads/h produced little or no effect. There was little difference among the materials tested. A similar increase in corrosion rate as a result of gamma irradiation was noted by Koster (1983).

The corrosion of materials in brine represents an extreme case for a salt repository. It is much more likely that the vessel will be in contact with either damp or dry salt. Tests at 150°C in salt moistened with a low-magnesium brine showed low corrosion rates (~5 μm/year) for cast mild steel, wrought mild steel, and ductile cast iron (WESTERMAN 1984c). However, when the salt was moistened with a high-magnesium brine, such as that present in brine inclusions in rock salt, much higher corrosion rates

(>700 μm/year) were noted. It appeared that the presence of magnesium interfered with the formation of normally protective Fe_3O_4 films. Attack on mild steel was very low in dry salt, at least up to 225°C (RANKIN 1980).

The corrosion rate of mild steel and 2-1/4 Cr—1 Mo steel in bentonite moistened with brine (low magnesium content) at temperatures up to 250°C without radiation was very low, although quantitative rates were not measured (MOLECKE 1984). The 2-1/4 Cr—1 Mo steel, however, experienced some pitting and, in this regard, was inferior to mild steel.

It is obvious that the use of steels or low-alloy steels in salt repositories may be acceptable, but more work is required to define the conditions that will exist in the specific repository and to evaluate the corrosion of ferrous alloys in these environments. For example, it has recently been shown that brines made from gamma-irradiated salt have high pH values (AHN 1985), and the corrosion of iron alloys in these brines should be less than that in the nearly neutral brines used in most tests. Existing data do not permit modeling of corrosion of ferrous alloys in salt repositories.

Titanium alloys. The uniform corrosion resistance of titanium and its alloys to brines is very high, but localized attack is of much more concern than general attack in repositories. The average corrosion rate of TiCode-12 (0.6 to 0.9% Ni, 0.2 to 0.4% Mo) in Brine A at 250°C was 3 μm/year during a 28-d test. In a much longer test (190 d) at 200°C, the corrosion rate decreased by as much as eightfold between 21 and 190 d and the maximum measured value observed was 0.35 μm/year during this interval (MOLECKE 1983). The effect of radiation on the corrosion of TiCode-12 in Brine A appears to be low; a dose rate of 10^7 rads/h increased the corrosion rate by a factor of 2 (BRAITHWAITE 1980). In alkaline brine at 150°C, formed from salt that had been irradiated, corrosion rates of TiCode-12 were factors of 5 to 8 greater than in brine made from the same unirradiated salt (AHN 1985). The high pH of the irradiated brine was presumably responsible for the higher, but still low, corrosion rate. Thus, it seems clear that the general corrosion rate of TiCode-12, even under the worst conditions envisioned in a waste repository, will be low and a uniform corrosion allowance of a relatively few millimeters will be adequate for a waste package expected to last for 1000 years.

B.1.2.1.3 General corrosion in a tuff repository

The environments expected to be encountered in a tuff repository are hot, "dry" air with limited humidity levels at 95 to 300°C, corresponding to (1) initial conditions when the hot waste package drives moisture away from its vicinity; (2) "moist" air with humidity at or near saturation, corresponding to the period when the rock has cooled to about 95°C and water can reenter the vicinity of the waste package; and (3) water exposure that corresponds to possible episodic intrusion of water into the repository environment. Although the first environment cannot be summarily dismissed, the last two environments are likely to be more aggressive to waste packages than the first and, consequently, have received more attention. It should be noted that the waste package will be exposed to an environment consisting of air plus water vapor until the temperature of the canister or overpack is 95°C or less (95°C is the boiling point of water at the repository elevation). Actually, it is possible for capillary condensation to occur in cracks and crevices at temperatures somewhat higher than 95°C, depending on the size of cracks and crevices and the relative humidity of the gaseous environment.

The corrosion rate of 304L stainless steel exposed to tuff-conditioned water at 100°C and the vapor above it during a 1000-h test was very low, ranging from no detectable attack to 0.25 μm/year. In the same environment, mild steel experienced rates of 26.8 and 12.5 μm/year in the aqueous and vapor phases, respectively, during 5000-h tests at 100°C (McCRIGHT 1984a).

The corrosion of 304L stainless steel and mild steel in tuff-conditioned water at 105 and 150°C in a gamma field was also examined during a 2-month test. At 105°C, the radiation dose rate was 3×10^5 rads/h; at 150°C, it was 6×10^5 rads/h. The 304L stainless steel corroded at 0.3 μm/year under each condition, whether the alloy was exposed in water above a bed of tuff or embedded in the tuff. On the other hand, mild steel corroded at rates of 40 μm/year, regardless of exposure location or temperature (McCRIGHT 1984a). Both of the above conditions are more severe than would be expected in a tuff repository.

Tests aimed specifically at determining the corrosion resistance of mild-steel borehole liners indicated that a wall thickness of 1.25 cm would be sufficient for a life of more than 50 years in either steam or aqueous environments (McCRIGHT 1984b). However, one area not investigated was the effect of a radiation field on corrosion of steel in moist air. The canister and/or overpack will be at a higher temperature than the gas and will probably not be subject to attack by dilute gaseous nitric acid. The temperature of the liner, on the other hand, may be lower than that of the gas; thus, any nitric acid formed in the moist air may condense on, and react with, the steel.

In general, the uniform corrosion rate of an austenitic stainless steel under tuff repository conditions would be predicted to be low, and experimental results verify this. In such an environment, localized attack is of much greater concern than uniform corrosion. A uniform stainless steel corrosion allowance of only a few millimeters should be adequate for the 1000-year life of a waste canister or overpack.

B.1.2.2 Stress-Corrosion Cracking (STAEHLE 1969, 1977; SCULLY 1971)

Stressing a metal or alloy has no significant effect on its general corrosion resistance but can cause cracks to form in some environments. With a sufficiently high sustained tensile stress (either applied or residual) and in the presence of certain corrosive species, stress-corrosion cracks can develop in most metals and alloys. Stress-related cracks caused by cyclic stresses in corrosive environments are called corrosion fatigue cracks; they are not considered here.

Stress-corrosion cracking has two successive stages: an incubation period and a period of crack growth. For engineering materials used in practical environments, either the incubation period is very long or the material is not susceptible to cracking in the environment. In most practical cases, it is not possible to measure or estimate the length of the incubation period. On the other hand, crack growth is more predictable; once a threshold stress at the tip of the crack is exceeded, fracture mechanics can be used to estimate its growth rate. Thus, crack growth lends itself to analysis, whereas no known method of predicting incubation periods exists. The likelihood of cracking in waste packages is difficult to estimate for two reasons: (1) phase changes within the alloy may occur over long periods at relatively high temperatures (up to 300°C) and produce a susceptible structure, whereas the original structure was not susceptible; and (2) unpredictable changes can also occur in the composition of groundwater during a 1000-year period. An example of the former is the long-term, low-temperature sensitization and subsequent cracking of austenitic stainless steel in boiling-water-reactor environments.

Because of the incubation period, stress-corrosion cracking tests that use constant-load specimens can give inconclusive results. When a test is discontinued after a given period without evidence of cracking, the question remains as to whether cracks would have developed if the test had been continued longer. It is generally assumed that if a material can crack in a given environment, cracking will ultimately occur, regardless of the incubation period. Consequently, a type of test that essentially eliminates the incubation period has found favor with many investigators of stress-corrosion cracking. This test, referred to as the constant extension rate test (CERT), involves straining a tensile-type specimen at a very low constant rate in the selected environment until it fractures. The results obtained (time to failure, elongation, reduction in area, and appearance of fracture surfaces) are compared with those of an identical specimen strained under the same conditions but in the absence of the corrosive fluid. A measure of susceptibility of a material to cracking is obtained from the difference in behavior with and without the corrosive fluid. By straining the specimen at a low constant rate, the protective film on the surface is constantly disrupted and the stress level is continuously increased so that any tendency toward cracking should be apparent. Evidence of cracking does not necessarily mean that the material will fail in service; however, it does indicate that the material has a susceptibility to cracking in the environment tested. Particularly with waste packages, it would be prudent to avoid materials that show any susceptibility to cracking in simulated repository environments when using CERT.

There is one case in which CERT results may be misleading. Cracks sometimes originate at the base of pits because the pits serve as stress risers; however, it has been alternately suggested that the solution that develops in a pit, which is different from the bulk solution, is the cracking agent (BROWN 1972). If the latter view is correct, the CERT approach with unpitted specimens could lead to a nonconservative conclusion.

Another way of getting around the induction period is to use precracked specimens, and this is the technique used in the fracture mechanics approach to the study of stress corrosion cracking. Fracture mechanics establishes crack growth rates as a function of stress intensity and allows the development of relationships between the two. Using this approach, a model predicting the time to failure of waste containers may be possible. One disadvantage of this method for waste containers is the very long lifetimes involved, 300 to 1000 years; crack growth rates as low as 10^{-11} to 10^{-12} μm/s are important, and measurements of such low rates present significant experimental difficulties.

In most aqueous environments in which stress-corrosion cracking has been observed, cracking occurs over only a limited range of electrode potentials. The potential of the material is determined, to a large measure, by the redox potential of the solution in which it is immersed; consequently, the chemistry of the solution (in addition to the presence of the cracking agent) is of prime importance in determining whether the material will crack.

Controlling the stress level within a material is one way of avoiding stress-corrosion cracking. All materials require a minimum stress in order for stress-corrosion cracks to start and grow. This stress is near the yield stress of the material in some cases, but is well below that level in others. While it is customary to design for average stresses below the threshold stress, it is difficult to fabricate vessels that have all parts below that level. Examples are residual stresses caused by welding and stresses caused by deformation during handling.

B.1.2.2.1 Stress-corrosion cracking of steel in a basalt repository

Until a few years ago, carbon steels were believed to be susceptible to stress-corrosion cracking in only strong caustic or nitrate solutions. However, more recent investigations, largely using those CERT methods, have shown that a number of other ions or compounds can cause cracking in mild steel. The literature on agents reported to be capable of producing stress-corrosion cracking in low-strength carbon steels has recently been reviewed (BEAVERS 1985a). In most cases, cracking occurs more readily as the concentration of the corrosive reagent and/or the temperature increases. Cracking has not been observed in steel when the individual ions are present at concentrations found in basalt groundwater, but CERT-type testing has apparently not been performed in simulated basalt groundwater at high temperatures in the presence of a radiation field. In addition, the effect of concentrating the solution (e.g., by loss of steam) must also be considered. Of the components present in basalt groundwater, only the carbonate/bicarbonate mixture has been demonstrated to crack steel; however, a combination of other ions at sufficiently high concentrations could possibly induce cracking.

Long-term stress-corrosion cracking tests with mild steel and 9 Cr—1 Mo steel in basaltic groundwater containing bentonite at 150 and 250°C have been started, and preliminary data for a 2000-h exposure have recently become available (JAMES 1985). Modified wedge-open-loading specimens that had been precracked in air were used in these tests. No signs of crack growth were found with mild steel under any of the conditions used, but the results suggest that slight crack extension may have occurred in the 9 Cr—1 Mo steel specimens at the two highest stress intensities at 250°C.

Longer tests (up to 20,000 h), presently in progress, will be needed to confirm these results. In addition, cyclic tests with the same materials are planned (JAMES 1983). Although cyclic stresses are not expected in a repository, the information obtained from these tests and the application of linear elastic fracture mechanics can lead to conservative estimates of the minimum stresses necessary to produce crack growth for given flow sizes in steel containers.

The stress-corrosion cracking of steel in basaltic repository environments, including radiation and concentration effects, has not yet been investigated. Such tests in the absence of radiation have been started, but only preliminary results are available at this time. Initial tests with very low concentrations of ferric chloride in water produced cracking in carbon steel at 300°C (BEAVERS 1985b). Since the presence of radiation may increase the redox potential of the groundwater in the same manner as ferric ions, there is at least a suggestion that mild steels may be susceptible to cracking in irradiated basaltic groundwaters.

The stress on an overpack is increased, and thereby the probability of stress-corrosion cracking is increased, by the force exerted on the waste package by corrosion products. When a volume of steel corrodes, the volume of the corrosion product is more than twice that of the corroded metal. Thus, the pressure on a packed waste package may increase as corrosion proceeds. The importance of this phenomenon on corrosion in general, and on stress-corrosion cracking in particular, needs to be considered.

Our overall understanding of stress-corrosion cracking is poorly developed, and the susceptibility of steels to cracking in basaltic groundwaters has not been determined. It seems unlikely that a mathematical model based on an understanding of the stress-corrosion cracking process will be developed in time for licensing the first repository. However, a model based on measured cyclic crack growth rates as proposed by James (1983) may provide a means of conservatively predicting times to failure.

B.1.2.2.2 Stress-corrosion cracking in a salt repository

Titanium and TiCode-12 do not appear susceptible to stress-corrosion cracking in any environments associated with a salt repository. Experiments conducted under a wide variety of conditions, both with and without radiation and at temperatures up to 250°C, show no evidence of loss of ductility on a macro scale (MOLECKE 1983; BRAITHWAITE 1980; ABREGO 1981). However, on a micro scale, a very thin surface layer (up to 50 μm) showed a nonductile failure that may have been related to hydrogen embrittlement and will be discussed more fully in Sect. B.1.2.5.2.

As was the case in basaltic environments, there appear to be no reported studies of the cracking of steels in brine solutions. If steel overpacks are to be considered in salt repositories, information on the stress-corrosion cracking susceptibility of these materials is essential. It is possible that the amount of brine migrating to a hot overpack surface will be too small to cause a significant general attack, but even small amounts of brine could produce stress-corrosion cracking.

B.1.2.2.3 Stress-corrosion cracking in a tuff repository

Since waste packages will be stored above the water table, the austenitic stainless steel canisters or overpacks will be exposed to a gaseous environment until the temperature of the waste package drops to <95°C. It seems reasonable to assume that stress-corrosion cracking will not occur until the stainless steel is exposed to an aqueous solution; consequently, most cracking tests have been performed in solutions at <100°C and, in some cases, in the vapor above such solutions.

Both U-bend and beam-type specimens of 304L stainless steel exposed to tuff groundwater at 50 to 100°C in the presence of radiation showed no evidence of stress-corrosion cracking after 3 months. Absence of cracking was also observed in related CERT tests at 150°C (JUHAS 1985). Similar results were obtained with V-notched, C-ring specimens exposed at 100°C in and above tuff groundwater and the same water concentrated tenfold (DAVIS 1985b). In later tests, however, microcracks were reported on notched C-ring specimens exposed to the aqueous phase of synthetic J-13 well water and the vapor above it (ABRAHAM 1986). In these tests, the specimens in both locations were packed in crushed tuff and during the test, the solution increased in salt content as components were leached from the tuff. The cracks has a maximum depth of 27 μm and the depth of penetration was about the same for 6- and 12-month exposures. Cracks were observed on the smooth part of the C-ring as well as in the root of the notch. From the micrographs shown in the report, it is not certain that these slight penetrations should be classified as stress corrosion cracks.

While the above tests do not completely establish the susceptibility of 304L stainless steel to cracking in tuff groundwater, a scenario exists whereby a dilute chloride solution percolating through the tuff could concentrate on the stainless steel surface and produce a chloride ion concentration high enough to produce cracking. Additional testing under all conceivable conditions appears justified in view of the high susceptibility of austenitic stainless steels to stress-corrosion cracking in chloride environments.

Mild steel will be used as a borehole liner in the tuff repository. A crack in this material is of less consequence than a through-wall crack in the waste package; nevertheless, it could interfere with the retrievability of the package. Tests have shown the absence of cracks in welded 1020 steel specimens stressed to 90% of the yield stress in tuff groundwater at 90°C (McCRIGHT 1984b). On the other hand, 9 Cr—1 Mo steel specimens cracked in the weld metal after 3500 h, and the investigators suggested that hydrogen was responsible. The weld metal was martensitic. Based on these few data, it seems that stress-corrosion cracking of the borehole liner will not be a problem during its 50-year life.

B.1.2.3 Pitting (SHREIR 1976; STAEHLE 1974; KRUGER 1982)

Pitting is a form of localized corrosion in which random deep penetrations (pits) develop in a metallic substrate. Pitting primarily occurs when the metal is passive over most of its surface and the solution contains aggressive ions such as chloride and bromide. Such ions can penetrate films and start pits at weak spots in the passive film. Once attack starts, the solution at the pit site develops a lower pH than that of the bulk solution because the metal ions from the initial reaction hydrolyze to produce an acidic environment. The low pH accelerates attack, and the pitting process becomes autocatalytic. Pitting is much more likely to occur in static than in flowing solutions, where the corrosion products may be washed away. For example, austenitic stainless steels pit badly in quiet seawater but show no tendency to pit in flowing seawater. Also, as the pits get wider, the solution in them becomes easier to wash out — a fact that accounts for the frequent self-passivation of pits after they reach a certain size.

After a pit has started, the formation of cations in the solution within the pit is balanced by reduction of an oxidizing agent on the surfaces adjacent to the pit. The formation of cations in the pit requires the migration of anions (usually chloride ions) into the pit to maintain charge neutrality. This fact, combined with hydrolysis of the corrosion-product cations, makes the solution within the pit more corrosive than the bulk solution. Soluble corrosion products diffusing from the pit may precipitate at the mouth of the pit, where they meet the bulk solution that has a higher pH. Thus, pits are often marked by deposits of corrosion products, which are referred to as tubercles. Tubercles tend to keep the pit active by restricting mixing of the pit solution with the bulk solution.

A minimum electrode potential — the pitting potential — is required for pits to remain active and depends on the specific environment. Many studies have been conducted to determine the pitting potential of various metals and alloys in specific environments, and results generally indicate that the potential where pitting begins becomes lower as the temperature and aggressive ion concentration increase; in other words, pitting is easier. As in most types of localized attack, the initiation of pitting requires an induction period. Even though this period may be long, it is generally believed that pitting will ultimately begin at the pitting potential, or above, but not at lower electrode potentials. The required potential for pitting may be supplied by an external source or by the redox potential of the solution.

When a pit starts, oxidation at the base of the pit keeps its potential in the active dissolution potential range, and reduction on the surface surrounding the pit cathodically protects that surface. Because of the difference in potential between the base of the pit and the adjacent surface, the pit continues to grow; and because of restricted mixing, the solution within the pit increases in salt concentration and becomes depleted in oxidant (usually oxygen).

The above discussion is based on conventional pitting studies that usually stop while the pits are small. For waste packages, the behavior of deep pits is important. Some questions peculiar to waste packages have apparently not been addressed. For example, as noted above, it is usually postulated that the solution at the base of an active pit is depleted in oxidizing agent; however, in a radiation field the concentration of oxidizing species may be similar both inside and outside the pit. Will a pit grow under such circumstances? Or when a pit is deep in an overpack wall and the radiation intensity to which the solution is exposed is greater at the base than at the surface, will the pit continue to grow? What happens to the corrosion products generated in pitting when the overpack is embedded in a plastic material such as wet bentonite? Does a pit which is started under these conditions get so impacted with bentonite and solid corrosion products that the electrical conductivity of the aqueous phase within the pit becomes too low to support a pit? What effect does heat flux have on pitting?

Answers to these questions need to be determined experimentally and under conditions simulating as closely as possible those anticipated in a repository before the significance of pitting in overpacks can be assessed.

B.1.2.3.1 Pitting in a basalt repository

Measurements of pitting potential on carbon steel at temperatures as high as those in waste repositories and with solutions comparable to those expected in basalt formations have apparently not been made. It has been shown, however, that steel pits in aerated groundwater at 150 and 250°C (WESTERMAN 1984b) and at 150°C in gamma-irradiated deaerated basalt groundwater containing methane (DAVIS 1985a); pitting does not occur in the absence of methane. The pits observed in these tests were small and shallow and, as such, do not threaten the integrity of the overpack. The question of how, or if, these pits continue to grow to the point of penetration of the overpack wall (at least several cm thick) remains to be answered.

A recently initiated experimental program will study pit growth kinetics in steel under basalt repository conditions, but results on growth rates are not presently available (LUMSDEN 1985). Results have shown, however, that pits cannot be started at 50°C; however, at 100 to 200°C pitting can be started by anodic electrochemical polarization after a protective layer of magnetite has formed.

B.1.2.3.2 Pitting in a salt repository

Problems of pitting in carbon steel in a salt repository might be expected to be similar to those in basalt groundwater that also contains chloride ions, but at much lower levels. However, short-term tests in pure deaerated 4 M sodium chloride solutions at temperatures up to 200°C showed high corrosion rates but no evidence of pitting (POSEY 1979). Similarly, a 3-year exposure to aerated wet salt in the salt mine at Avery Island, Louisiana, at temperatures near 200°C showed heavy corrosion of steel but no evidence of pitting (GRIESS 1982). These data, combined with the absence of pitting of cast iron in brine (MOLECKE 1983), suggest that pitting of steel in a salt brine will not be a major problem.

Titanium and its alloys passivate easily and might be expected to become pitted in concentrated chloride environments; however, experience indicates that pitting seldom causes failures in titanium equipment. It has been shown, however, that pure titanium and its alloys can be pitted in chloride solutions, but the pitting potentials are very high. The pitting potential decreases with temperature, but even at 200°C the pitting potential for each alloy examined (except for one containing tin) is about +1.0 V with reference to the saturated calomel electrode (POSEY 1967). TiCode-12 had not been developed at the time of the above investigation. Potentials of titanium containers in a salt repository will undoubtedly be less than +1.0 V;

therefore, pitting should not be expected. In view of the above considerations, failure of either steel or TiCode-12 overpacks in salt repositories by pitting seems very unlikely.

B.1.2.3.3 Pitting in a tuff repository

In the usual sense, pitting requires the development of specific anodes surrounded by cathodic regions that are connected by an aqueous solution. Since the repository in tuff will be above the water table and the temperature of the waste packages will be higher than that of the surroundings, an aqueous phase will not exist on the surface of the waste package for a significant part of its life. However, the possibility exists that at some time an aqueous solution containing low levels of chloride will contact the waste package and concentrate on it. Pitting of the austenitic stainless steels is possible under these conditions. How fast pits will grow under these conditions depends on the temperature and chloride concentrations.

The austenitic stainless steels (304L, 316L, and 317L) showed no evidence of pitting in a 1000-h test at 100°C in water from well J-13 at the Nevada Test Site, as well as in the vapor above it. In these same tests, carbon steel (possible borehole liner materials) showed evidence of localized attack, although the depth of attack was not measured (McCRIGHT 1984a). During a 2-month test in J-13 well water in a radiation field at 105°C (3×10^5 rads/h) and at 150°C (6×10^5 rads/h), pits also failed to form in stainless steel (McCRIGHT 1984a). In other tests in boiling J-13 well water, carbon steel experienced pitting; however, both the generalized and pitting corrosion rates decreased with time (McCRIGHT 1984b). Pitting in borehole liners is not of major consequence, provided the liner remains in place for the 50-year period of retrievability.

Thus, the data presently available indicate that pitting of stainless steel is not likely to be a means of failure for either an overpack or a canister; of course, this conclusion is based on tests conducted in J-13 well water that is very low in choride content. It seems probable that early in the repository life rain water percolating through the rock above the canister will dissolve salts and evaporate in the hot rocks around the waste package. Later, after the temperature of the package and surrounding rocks has become <95°C, percolating water will dissolve the deposited materials and the resulting water will have a greater salt concentration than the present J-13 well water. Pitting of stainless steel may be possible under these conditions.

B.1.2.4 Crevice Corrosion (SHREIR 1976; STAEHLE 1974; KRUGER 1982).

As the name implies, crevice corrosion is a form of attack that sometimes occurs on metal parts located in the confines of narrow crevices. The solution within the crevice cannot mix freely with the bulk solution, and its composition gradually becomes different from, and more corrosive than, the bulk solution.

When a metal object that has a creviced area on its surface is initially exposed to a solution containing dissolved oxygen, or perhaps some other oxidizing agent, the solution in contact with all surfaces has the same composition; consequently, all surfaces have the same electrode potential. Corrosion initially consumes oxygen on all surfaces; however, because of the very small volume of the crevice solution and its inability to mix with the bulk solution, the oxygen in the crevice is soon consumed. As a result, the potential of the metal within the crevice becomes less noble than the freely exposed surface that has access to oxygen. Hence, the crevice region becomes an anode with metal ions being solubilized and anions migrating into the crevice; to complete the cell, oxygen is reduced on the surface outside the crevice. Hydrolysis of metal ions in the crevice produces acid that accelerates crevice attack until the diffusion of corrosion products and acid out of the crevice is balanced by their rates of formation. Once started, the corrosion mechanism in a crevice is identical to that in an active pit.

Both pitting and crevice corrosion usually occur on passive metals and become more severe as the temperature and the salt concentration of the solution increase. Pitting, however, usually requires specific ions for initiation, whereas crevice corrosion can occur with different salts. Under normal conditions both types of attack occur by electrochemical action, involving clearly defined anodic and cathodic areas.

Assessment of the likelihood of crevice corrosion that is damaging to waste packages requires knowledge of the effects of radiation, heat flux, and restricted access of solution to all surfaces. Based on the theory of crevice corrosion, as it is usually interpreted, one would conclude that when the entire surface is a "crevice," such as a buried waste package, crevice corrosion could not occur. Similarly, if the solution is totally free of oxygen when it comes in contact with the package or if radiation produces adequate concentrations of oxidizing radicals in the solution at all locations, corrosion should not occur even if discreet crevices exist on the overpack. Tests to verify the above suppositions have not been carried out in repository environments.

B.1.2.4.1 Crevice corrosion in a basalt repository

As a result of a literature survey, Charlot and Westerman (CHARLOT 1981) concluded that crevice corrosion was an unlikely failure mode for cast iron (and presumably mild steel) in a basalt repository. In view of the low salt content of the groundwater, this seems to be a reasonable assumption; however, no consideration was given to the possible concentration of salts in the water. Short-term tests in which mild steel was embedded in a 75% basalt—25% bentonite including either aerated or deaerated groundwater at 150 and 250°C seem to have confirmed the conclusion of Charlot and Westerman (ANANTATMULA 1983); no crevice corrosion (or other forms of localized attack) was noted. On the other hand, steel coupons exposed in oxygen-containing groundwater in the presence of radiation at 250°C showed crevice corrosion when the coupons contacted the support rod (NELSON 1984).

Based on the above information, it is clear that additional testing under realistic repository conditions is necessary before the importance of crevice corrosion of steel in a basaltic repository can be assessed.

B.1.2.4.2 Crevice corrosion in a salt repository

Crevice corrosion of titanium and most of its commercial alloys is recognized as a major cause of failures of titanium equipment. Two commercially available alloys that show great resistance to crevice corrosion are Ti-0.2% Pd and TiCode-12. TiCode-12 and steel are the materials most likely to be used as overpacks in salt repositories.

Little information appears to be available on the crevice corrosion of steels in salt-related environments. In tests in the Avery Island salt mine, steel specimens bolted to a steel sleeve and in contact with wet salt showed no more attack on the creviced surface than on the other surfaces, suggesting the absence of a crevice effect (GRIESS 1982). Also, tests in bentonite, in bentonite-sand mixtures (both wet and dry), and in crushed salt in field tests in New Mexico at temperatures of up to 272°C failed to show evidence of crevice attack on mild steel (MOLECKE 1984). Thus, on the basis of these results, crevice corrosion of steel in salt repositories does not appear to be a likely failure mode, but further testing to confirm this tentative conclusion is warranted.

Although commercially pure titanium is highly resistant to neutral chloride solutions, it is susceptible to crevice corrosion; for that reason, it has not been considered as a material for overpacks in salt repositories. Because of its high resistance to general attack by acid salt solution, its resistance to crevice corrosion, and its lower cost as compared with the Ti-0.2% Pd alloy, TiCode-12 is the preferred titanium alloy for overpacks in a salt repository.

Tests made with TiCode-12 in deaerated Brine A at 200°C showed no evidence of crevice attack for up to 3 weeks (MOLECKE 1983); apparently, no tests were performed using aerated brine. In a completely deaerated salt solution containing no oxidizing agents, crevice corrosion is unlikely even for pure titanium (GRIESS 1968). Crevice corrosion of TiCode-12 was observed when the brine was acidified to a pH of 2 and contained oxygen, the TiCode-12 had an adherent layer of salt on its surface, and the temperature was 150 to 200°C (DIEGLE 1984). Also, crevice corrosion was reported in Brine A under both aerated and deaerated conditions at 150°C, but attack was greater in the aerated solutions (AHN 1983). No significant damage to the alloy was noted in these latter tests; only a thickening of the oxide on surfaces within the crevice was observed.

Whether crevice corrosion of TiCode-12 will occur in a salt repository remains to be determined. Attack can occur if the conditions are severe enough and, once started, is likely to penetrate the thin titanium overpack or cladding on the overpack. Useful information in this regard will be obtained from long-term (5-year) crevice corrosion tests in progress at Sandia National Laboratories (MOLECKE 1983).

B.1.2.4.3 Crevice corrosion in a tuff repository

Crevice corrosion requires a continuous aqueous phase on the creviced surfaces, as well as on the freely exposed surfaces. Since the repository will be located above the water table and the temperature of the waste package will be above the boiling point of dilute solutions for hundreds of years, crevice corrosion is impossible for that period. By the time that intruding solutions could exist on the surface for any duration, the temperature of the overpack would be <95°C and the concentration of salts in the solution would probably be low. Crevice corrosion, while possible under such conditions, seems highly unlikely and, therefore, is considered to be a very improbable route for breaching stainless steel canisters or overpacks in a tuff repository.

B.1.2.5 Hydrogen Embrittlement (STAEHLE 1977; BERNSTEIN 1974; API 1977)

Hydrogen can degrade the tensile properties of certain metals and alloys, including steels and titanium and its alloys. Embrittlement of high-strength steels is a time-dependent function that causes cracks to form and grow when the steel is subjected to sustained tensile stresses below their yield strength. The problem becomes less severe as the strength of steel decreases, and steels with yield stresses below about 550 MPa (80,000 psi) are not subject to this form of attack, although other hydrogen-related attack is possible (see Sect. B.1.2.5.1). In the case of high-strength steels, only very low concentrations of dissolved hydrogen produce embrittlement; however, in the case of titanium, a loss of tensile properties occurs only when the concentration of hydrogen exceeds the solubility of titanium hydride and a brittle hydride phase exists.
In order to enter a metal, hydrogen must first be present on the metal surface as hydrogen atoms. In a waste repository, the only sources of atomic hydrogen are radiolysis of groundwater and corrosion. In the former case, the energy absorbed by the water dissociates water molecules to yield a low steady-state concentration of hydrogen atoms throughout the solution. In the latter case, the reduction (or cathodic) half reaction of the overall corrosion reaction produces atomic hydrogen on the surface of the metal. Most of the atomic hydrogen on the metal surface combines to form molecular hydrogen, which escapes from the surface. Certain substances, such as hydrogen sulfide and arsine (AsH_3), interfere with the recombination of hydrogen atoms and thereby facilitate the entry of hydrogen into metals.

B.1.2.5.1 Hydrogen embrittlement in a basalt repository

The embrittlement of high-strength steels is of no direct consequence in waste repositories since these materials are not considered for overpacks. Embrittlement of steels with normal tensile strengths has been reported, but all documented cases of this type have been initiated in localized regions of high hardness with martensitic or bainitic microstructures (FESSLER 1977). Some of these failures have originated in hard welds and heat-affected zones of welds. The above form of embrittlement is temperature dependent; maximum susceptibility occurs at ~25°C, while no cracking was observed in tests at 82°C (TOWNSEND 1972). Since the temperature of the overpack in a repository is projected to be >75°C for about the first 1000 years (ALTENHOFEN 1981), these results suggest that this form of failure is not likely for most, if not all, of the required life of the overpack, even if hard spots exist in and around welds. Whether the above temperature limit for embrittlement is applicable for long times and for the particular steel used in a repository needs to be determined.

In addition to the classical hydrogen embrittlement of high-strength steels, hydrogen in steel, including low-carbon steels, can produce other detrimental effects such as (1) pressure buildup in the cavity between the canister and the overpack, (2) surface blistering, (3) hydrogen attack (methanation), and (4) hydrogen-induced cracking or stepwise cracking, which is a linking up of parallel inclusions aligned in parallel planes, after hydrogen has expanded the void spaces around the inclusions. In addition, should the cladding of carbon steel with an austenitic stainless steel be considered, the formation of molecular hydrogen at the interface between the two materials may lead to disbanding.

Pressure buildup in the overpack should not present a problem at the temperature of the steel. However, at temperatures lower than those expected to exist in the repository (~25°C), substantial pressures could build up and overstress the overpack.

Surface blistering occurs primarily at low temperatures and should not pose a problem with the steel overpack. Atomic hydrogen entering the metal may recombine at inclusions and form bubbles of hydrogen at high pressure. Since these inclusions are always very small, only those close to the surface can generate a sufficiently high pressure to cause yielding of the metal, which is manifested as small blisters. However, as noted in (4) above, linking up of such blisters could lead to the formation of cracks parallel to the surface. Using a clean steel with few inclusions can minimize this type of damage.

At the highest temperature expected in the overpack walls (300°C), hydrogen attack (also referred to as methanation) may occur, as illustrated below, if the hydrogen pressure is sufficiently high:

$$Fe_3C + 4H \rightarrow 3Fe + CH_4.$$

The methane collects at both grain boundaries and inclusions and embrittles the steel. Hydrogen attack of carbon steel is possible at temperatures as low as ~250°C, provided the pressure of the hydrogen gas is sufficiently high. This type of problem has been most frequently encountered in petroleum refining, where high-pressure hydrogen is handled at high temperatures; however, carbon-steel boiler tubes in fossil power plants have also experienced brittle failure in cases where the source of hydrogen is corrosion-product hydrogen. Temperatures in boiler tubes are usually in the range of 300 to 350°C.

Hydrogen produced electrolytically on the steel surface (either by cathodic charging or as part of the corrosion process) acts in the same manner as high-pressure gaseous hydrogen; consequently, corrosion is one method of introducing hydrogen into steel. Corrosion in the waste repository environment could conceivably lead to embrittlement of the overpack; the probability of this occurring increases as the corrosion rate and/or the temperature increases.

It should be noted that hydrogen attack of carbon steel does not appear to be a problem below about 200°C. This observation, however, is based on short-term data relative to the 300 to 1000 years in a waste repository where temperatures of 200°C or greater will exist for 50 to 200 years. Consequently, before licensing of a repository that uses carbon steel containers can be expected, the absence of methanation must be demonstrated. However, the addition of small amounts of carbide-forming elements (Mo, Cr, Ti, etc.) to the steel can mitigate the problem (API 1977). Carbides of molybdenum and other elements, such as titanium and chromium, are very stable and tie up the carbon so that it is unavailable to react with hydrogen.

In summary, hydrogen embrittlement of steel overpacks in a basalt repository does not appear to be a likely failure mode. If hydrogen attack is a problem, it is one that can easily be remedied.

B.1.2.5.2 Hydrogen embrittlement in a salt repository

The considerations of embrittlement of steel in a salt repository are the same as those in a basalt repository; thus, they are not repeated here. However, it is possible that corrosion of steel may be more extensive in salt than in basalt and that more hydrogen may be available for attack. In general, the probability of hydrogen-related failures of steel overpacks in salt repositories is very low.

On the other hand, titanium and its alloys can absorb hydrogen and become embrittled. For example, a TiCode-12 specimen exposed to brine at 250°C showed a normal ductile failure during slow straining, but fractographic analysis revealed a 50-μm-deep layer around the perimeter of the specimen that showed a less ductile fracture (BRAITHWAITE 1980). Such results indicate that the environment can affect the fracture process of TiCode-12.

CERT tests in Brine A at 150°C at an applied voltage of −1.000 V with respect to the silver/silver chloride electrode showed no signs of embrittlement. Also, straining under the same conditions with a constant cathodic current density of 1.4 mA/cm^2 produced no embrittlement; however, a significant loss of ductility was observed when the current density was increased to 7 mA/cm^2 (MOLECKE 1983). A similar loss of ductility in TiCode-12 after cathodic charging was noted at Brookhaven National Laboratory (AHN 1985).

Results of weight-loss tests in basaltic groundwater showed that both titanium and TiCode-12 absorbed more hydrogen in the presence of radiation than in its absence (NELSON 1984). In fact, the amount of hydrogen absorbed was greater than could be explained by corrosion. It was also noted that the hydrogen absorption in alkaline brine (pH ~11.5) was greater than in neutral brine (AHN 1985).

The above data at least hint that TiCode-12 may become embrittled in a salt repository, and additional long-term testing is definitely needed to determine if this is the case. The results suggest that the use of fracture mechanics may shed light on the cracking/embrittlement of titanium and its alloys. Since TiCode-12 would probably be used as a cladding on a structural material such as steel, it is also necessary to determine whether hydrogen has any effect on the bond between titanium and the substrate.

B.1.2.5.3 Hydrogen embrittlement in a tuff repository

An austenitic stainless steel is the material of choice for the canister and overpack in a tuff repository. Since this class of materials is not subject to hydrogen embrittlement, failure will not occur by this means. The borehole liner will be made of mild steel, and the comments made for basalt (see Sect. B.1.2.2.1) are applicable here; however, since the environment will be moist air during the design life of the liner corrosion should be low and the quantity of hydrogen entering the steel should be much smaller than in basaltic groundwater. At the high temperatures expected during the 50-year life, only hydrogen attack seems possible. If this proves to be a problem, the incorporation of low concentrations of chromium and/or molybdenum in the steel will prevent attack.

B.1.2.6 Microbial Corrosion (TILLER 1982)

Microbial corrosion refers to the degradation of metallic structures resulting from the activity of microorganisms. The attack may be initiated by the products generated during the metabolic process of the organisms or by colonies of organisms that partially block the surface of a metal, thereby facilitating differential concentration cells. It is important to note that microbial involvement in corrosion produces no new corrosion processes.

Although microorganisms of a wide variety can cause deterimental effects on metals under specific conditions, the most important types are bacteria that utilize sulfur and its compounds in their metabolism. The Thiobacillus genus of bacteria oxidizes sulfur to sulfuric acid under aerobic conditions. These bacteria are destroyed at temperatures of 55 to 60°C and should represent no threat to waste packages since the temperature of most, if not all, repositories will be >60°C for at least the first 1000 years.

In terms of underground corrosion, the genera of bacteria called Desulfovibrio and Desulfatomaculum are the most damaging because they reduce sulfate ions to hydrogen sulfide. These bacteria, which are strict anerobes, can survive for long periods under well-aerated conditions. Hydrogen sulfide would likely have its greatest effect on carbon-steel overpacks, but it may also facilitate the entry of hydrogen into titanium alloys. The effect of hydrogen sulfide on annealed austenitic stainless steel should be nil. In the presence of hydrogen sulfide, carbon steel achieves a low potential at which water can be reduced to hydrogen. The microbes use this hydrogen to reduce sulfate ions, which in turn depolarizes the cathodic reaction and increases the corrosion rate.

Tiller (1982) has reported that sulfate-reducing bacteria are killed at 70°C, but other information shows that at least certain strains can still produce hydrogen sulfide at 104°C (WEST 1982). Therefore, from a thermal standpoint, it seems possible that the temperature of waste that has decayed for some period will be low enough to permit sulfate-reducing bacteria to grow. However, the pH will increase on cooling (less hydrolysis), and the same authors show that sulfate-reducing bacteria do not function at pH values of ~9, regardless of the redox potential of the solution. Some waste packages may have high enough radiation fields associated with them to kill most bacteria in the immediate vicinity of the overpack, but it is known that certain bacteria can function in high radiation fields.

Most repositories are likely to have some concentration of bacteria present at the time of waste emplacement. Whether the types and numbers of bacteria are likely to lead to significant corrosion must be determined for each repository.

B.1.2.7 Intergranular Corrosion (SHREIR 1976)

Intergranular corrosion is a form of attack in which the corrosion rate at the grain boundaries is very much higher than on the grains per se. Thus, even though very little metal may be oxidized, intergranular penetration into the metal can be extensive and the loss of load-carrying ability can be substantial.

Grain boundaries are regions of high energy because of the disarray of atoms where crystals with different orientations meet and because precipitates preferentially form at grain boundaries. Under some conditions, the formation of grain boundary precipitates can impoverish the edges of the grains of an essential alloying element, leaving that portion of the grains much less corrosion resistant than the remainder. A classic example is the sensitization of austenitic stainless steels in which the precipitation of chromium carbide at grain boundaries results in such a low chromium concentration at the edge of the grains that these regions no longer have a stainless character. Consequently, sensitized austenitic stainless steels experience severe intergranular attack in a variety of solutions (e.g., acidified copper sulfate and nitric acid solutions).

In the case of carbon steel, intergranular attack seems to be of minor importance. For example, Shreir (1976) in his bibliography on intergranular attack lists 94 references, none of which concern carbon steel. In at least one case, however, intergranular attack occurred under deposits in an alkaline nitrate solution (ONDREJCIN 1978). The lack of intergranular attack in carbon steels is probably related to the fact that Fe_3C, which is an integral part of most steels, is more noble than iron itself in most solutions, and precipitates at grain boundaries are likely to be at least as corrosion resistant as iron. Also, carbon steels contain no passivating element (e.g., chromium) that could become depleted at the edges of grains and provide a means of selective attack at grain boundaries. Because of the above considerations, steel overpacks in waste repositories are unlikely to fail from intergranular corrosion.

Heat-treating of TiCode-12 does not appear to promote intergranular attack in acid chloride solutions, although the corrosion rate was increased somewhat as the result of sensitizing heat treatments (MOLECKE 1983). The austenitic stainless steels are subject to sensitization if the alloy contains sufficient carbon (usually regarded to be >0.03%), and in the sensitized condition they may be susceptible to intergranular attack. Although the stainless steel chosen for use in a tuff repository contains $\leq$0.03% carbon, long times in the sensitizing temperature range (~500 to 750°C) may still produce grain boundary precipitates that lead to intergranular attack. Stainless steels with carbon contents >0.03% may be sensitized at much lower temperatures if nuclei of carbides exist in the alloy. An overview of the low-temperature sensitization phenomenon has recently been prepared (FOX 1983), and tests are being conducted at Lawrence Livermore National Laboratory to study the sensitization of 304L stainless steel (McCRIGHT 1984a).

In most cases, intergranular attack requires the presence of an aqueous phase for an extended period. For most of the first 1000 years, canisters/overpacks in the proposed tuff repository will be contacted by water only occasionally and for short periods. Consequently, there appears little likelihood that intergranular attack will cause failure of low-carbon austenitic stainless steel in the tuff repository.

B.1.2.8 Stray-Current Corrosion (SHREIR 1976; PEABODY 1967)

Stray currents are direct currents flowing in the earth and are associated with some source other than the object affected by them. To cause corrosion, stray currents must enter the metallic object at one area, flow through the metal, reenter the earth, and complete the circuit to its ultimate destination. Corrosion occurs only in locations where the current leaves the metal. Such sources of current may be dc transit systems, welding machines, high-voltage dc transmissions systems, and others. In past years, stray currents originating from dc transit systems created serious corrosion problems with underground piping systems. In these cases, the current was returned to the generator through the tracks; however, the tracks were not always completely insulated from the earth and part of the current passed to the ground and was carried by underground pipes for some distance. Where the current left the pipe, the severe.

In reported cases of stray-current corrosion, affected pipelines were near the earth's surface (buried no more than 1 to 2 m deep). Waste packages will be buried 700 to 1000 m deep in specific geologic formations. It seems highly probable that stray-current attack will not occur under these conditions since it is generally assumed that such currents flow primarily near the earth's surface. When an overpack is breached, the cause is much more likely to be some other type of corrosion than stray-current attack.

Although unrelated to stray-current corrosion, the establishment of a galvanic cell after a steel overpack is breached could prolong the life of the stainless steel canister in a basalt or salt repository. After the overpack is breached, probably by some form of localized attack, the space between the overpack and the canister will become partially filled with a solution containing chloride ions. If the canister were not in electrical contact with the overpack, the stainless steel wall would probably be penetrated by stress corrosion cracks and/or pitting in times in the order of 10 years or less. However, if the stainless steel is in electrical contact with the steel, corrosion of the steel will cathodically protect the stainless steel. How long the steel will be effective depends on how much steel exists at the time the overpack is penetrated and its corrosion rate, assuming, of course, continued electrical contact between the steel and stainless steel.

B.1.2.9 Other Corrosion Considerations

Certain aspects of corrosion are common to all repositories. In each case, the canister material is specified as a low-carbon austenitic stainless steel that will be closed by welding; the overpacks that contain the canisters will also be welded shut. Canisters will enclose either glass that incorporates radioactive wastes or spent fuel elements. Thus, the corrosion of canister, weld, and fuel element cladding is a potential problem in all repositories and is briefly dicsussed below.

B.1.2.9.1 Canister corrosion

Once an overpack is breached, groundwater will enter the cavity between the overpack and the canister and corrosion of the canister can begin (except in the proposed tuff repository). If the canister is filled with glass, it will have been subjected for some extended period to temperatures high enough to produce sensitization in stainless steels containing more than 0.03% carbon. Furthermore, if the glass is liquid when poured into the canister, the stainless steel will be under a high hoop stress after the glass solidifies because the coefficient of expansion is greater for stainless steel than for glass. Recognizing that all groundwaters contain chloride ions, stainless steel under the conditions described above is highly susceptible to stress-corrosion cracking and pitting. Consequently, it is highly probable that the canister wall will be penetrated in a few years. As noted in Sect. B.1.2.8, some protection may be afforded by a less noble overpack — but only if good electrical contact between the overpack and the canister is maintained. Therefore, it seems appropriate to take no credit for such canisters as barriers against the environment, and this assumption has been made for the basalt repository (WEC 1982).

If the canister contains fuel elements, it will not have been subjected to the thermal treatment and will not have a high hoop stress, but there will be residual tensile stresses in the weld region; and even with fully annealed stainless steel, pitting in chloride-containing solutions is probable. Thus, while the metallurgical condition of a canister with spent fuel elements in it is more favorable for corrosion resistance than the glass filled canisters, penetration is still likely to occur in a relatively short time once the overpack has been breached.

B.1.2.9.2 Corrosion of fuel rod cladding

The cladding on spent fuel elements could be considered an additional metal barrier that separates radionuclides from the environment if the cladding retains its integrity when contacted by groundwater. However, the outside surfaces of fuel elements will probably contain some more or less adherent radionuclides, including fission products and fuel from leaking fuel elements and activated corrosion products. These must be considered in terms of radionuclide release after the overpack and canister have been breached. Most of the fuel elements stored in a waste repository will be clad in either Zircaloy-2 or Zircaloy-4, but a few will be clad in an austenitic stainless steel.

The Zircaloy cladding has a dark oxide on its surfaces on removal from the reactor, and the part that was in the cooler regions of the reactor may be embrittled. In general, zirconium has very good corrosion resistance to many types of solutions, and one would expect a very low corrosion rate in repository groundwaters. The presence of an oxide film should only enhance its corrosion resistance; however, Zircaloy-2 is subject to stress-corrosion cracking at 25°C (and presumably at higher temperatures) in neutral dilute sodium chloride solutions if the potential is increased only slightly above its corrosion potential (COX 1973). In this case, the presence of an oxide film increases the tendency to crack. Either an applied anodic current from an external source or coupling to platinum can polarize Zircaloy-2 into the potential range where cracking is likely to occur. The above observations led Jenks to speculate that the stainless steel canister in the presence of groundwater and a radiation field could polarize Zircaloy-2 into the crack-ing potential range (JENKS 1979). Assuming the above scenario, cracking would soon follow breaching of the overpack and canister. If credit for radionuclide retention is to be taken for Zircaloy cladding, experiments must be conducted to determine the life of the cladding under repository conditions.

A comprehensive review of the general corrosion and stress corrosion cracking of Zircaloy and stainless steel fuel claddings has been reported by Soo (1985).

Zircaloy-clad fuel elements are also subject to stress-corrosion cracking from the fuel side, and this type of cracking may progress and penetrate the cladding while in the repository. This failure mode is not discussed here, but it needs to be considered if the Zircaloy cladding is to be regarded as an effective barrier. A recent publication discusses the effects of selected fission products and metal iodides on such cracking (SHANN 1983).

The corrosion of stainless steel cladding on spent fuel elements in various repository groundwaters should be similar to that already discussed for canisters. However, the cladding has been subjected to fast-neutron irradiation that produces changes in the alloy. For example, it has been shown that annealed 304 stainless steel exposed to a minimum neutron fluence of about 2×10^{20} neutrons/cm^2 is subject to intergranular stress-corrosion cracking during CERT testing in high-temperature water containing low levels of oxygen; in the absence of radiation damage, cracking does not occur (CLARKE 1983). It seems probable that the stainless steel cladding may also have an increased susceptibility to other forms of corrosion. Since unirradiated stainless steel canisters with even thicker walls were judged to remain intact for only a relatively short time, it seems appropriate to assume that the stainless steel cladding will be penetrated very soon after contact with groundwater; therefore, no credit should be taken for the cladding as a metal barrier.

B.1.2.9.3 Weld-related corrosion

Welds are areas of metallurgical inhomogeneity and, as such, are potential areas of enhanced corrosion. If the composition of the weld metal is different from that of the base metal, galvanic attack of the less noble metal is possible. In addition, changes in the microstructure of the base metal adjacent to the weld may result in lower corrosion resistance in that region. An example of the latter is the formation of chromium carbides in the grain boundaries in the heat-affected zone of austenitic stainless steels containing more than 0.03% carbon — a fact that makes the material susceptible to intergranular attack in that area.

Welds in carbon steel that are not subsequently heated are usually harder than the base metal and are likely to be more susceptible to stress-corrosion cracking and hydrogen embrittlement than the base metal. In the welding of titanium, both metallic and atmospheric impurities must be avoided in order to obtain ductile welds. For that reason, titanium is usually welded in a dry box under carefully controlled conditions. Welding of large highly radioactive waste packages clad with a titanium alloy has apparently not been fully considered; nor has corrosion associated with such welds. In all types of canisters, weld shrinkage during solidification produces residual tensile stresses that increase the probability of stress-corrosion cracking.

A review of the available literature indicates that inadequate attention has been given to the behavior of welds in repository environments. Some tests have been conducted with welded coupons, but the welds in this case probably have different characteristics than those made in the field on heavy-walled vessels. Full evaluation of the integrity of waste packages will require a better knowledge of the behavior of welds than currently exists.

B.2 MODES OF WASTE FORM DEGRADATION

B.2.1 Glass Waste Form

Glass has long been the medium of choice for immobilizing radioactive wastes. The vitrification process is relatively simple and capable of accommodating a wide range of chemical elements. Also, glass has adequate chemical, thermal, and radiation stability for the expected repository conditions. Borosilicate glass, the preferred type, is the most thoroughly studied of the possible waste forms. Typically, a borosilicate glass with incorporated waste might consist of 20 to 30% waste oxides, 30 to 40% silica, 5 to 10% boron oxide, and 10 to 15 wt % alkali oxides, plus additives.

The degradation of glass forms is reflected by increased leachability of radionuclides when in contact with groundwater. The mechanisms that lead to increased leachability include: (1) matrix dissolution, or corrosion of the glass, by physical and chemical processes; (2) hydration of silicates, which leads to swelling and disruption of protective films that usually form; (3) devitrification with subsequent cracking; and (4) radiation damage.

Considerable information is available in the literature; detailed reviews of the properties and behavior of glass and other waste forms have been made by Mendel (1978, 1981, 1984), Rusin (1980), Dayal (1982), Davis and Schweitzer (DAVIS 1982b), and Kircher (1983). Recently, Westsik (1983) performed a number of leach tests on simulated HLW glass (PNL 76-68 glass) doped with Tc, U, Np, Pu, Am, and Cm using a salt brine simulating the WIPP site brines, a bicarbonate solution representing potential silicate rock repositories, and deionized water at temperatures of 150 and 250°C. His general conclusions were as follows: Normalized releases of the actinides tended to be higher in the sodium bicarbonate solution than in the other leachants. Technetium releases were highest in the deionized water and bicarbonate leachants. Increasing the temperature increased the releases of technetium and the actinides. All of the actinides showed less leaching than silicon, but the technetium releases were higher than those for silicon. Many of the nonradioactive components of the glass had higher releases in the WIPP brine than in the other leachants; however, silicon, boron, and molydenum behaved more like technetium in that their releases were lowest in the brine. Except for the alkaline earths, the releases of nonradioactive species increased with temperature and leaching time. Solubility constraints dictated by the pH and species that can complex with those leached from the glass appear to control the release of many species from the glass.

B.2.1.1 Matrix Dissolution

Matrix dissolution is akin to corrosion, which results in chemical reactions and/or the physical removal of surface material. The terms "corrosion" and "leaching" are generally used interchangeably, even though it is possible to have a particular nuclide leached from glass with no significant removal of surface material. Although evaluating waste forms in terms of a leach rate is convenient, the radionuclides involved should be specified. Initially, individual nuclide leach rates are frequently different (incongruent); as dissolution proceeds, however, the leach rates usually become congruent.

The corrosion of glass is complex and involves a number of possible mechanisms. The corrosion and subsequent leach rates are influenced by many variables, including solution and glass composition, pH, redox conditions, groundwater flow rate, temperature, pressure, surface area-to-volume ratio, and glass structure.

Many years of research have established that there are two general stages of surface reactions in alkali-silicate glasses exposed to water. The first is dealkalization of the surface by exchange of a hydrogen or hydronium ion with an alkali ion, which tends to be the dominant mechanism for pH <9. The overall result of this reaction is a depletion of cations from the surface, leaving a hydrous silica layer. It has been postulated that this layer functions as an increasingly protective barrier until the second-stage mechanism becomes rate controlling, but this has not been well established. In stage two, for pH >10, network dissolution and sloughing off by hydroxyl-ion breakdown of silicon-oxygen bonds tend to dominate. However, it is known that both processes can occur simultaneously over very wide pH ranges (HENCH 1977).

In view of the complexities and the number of variables involved in glass corrosion, the development of models for predicting long-term behavior is fraught with uncertainty. However, there is general agreement that under static conditions, reactions in stage 1 occur with square-root time dependence at low to intermediate pHs, while the network dissolution in stage 2 occurs with a linear time dependence at high pHs. With this view, the amount removed with time per unit area may be expressed as $(at^{1/2} + bt)$, where a and b are empirical constants. No comprehensive mechanistic studies are currently available to explain the relationship, but the square root generally indicates a diffusion-controlled reaction and the linear term reflects an interface-controlled reaction for test conditions. Most leaching experiments with borosilicate glass are reconcilable with an overall rate process that varies with $t^{1/2}$ at short times and low temperatures, and with t at longer times and higher temperatures (DAVIS 1982b). There has been considerable controversy over the transition in time regimes from square-root to linear, but it is generally acknowledged that the transition primarily depends on the time required for the pH to reach 9 to 10

(HENCH 1977). However, in nuclear waste glass studies, the most important factor is the long-term behavior, and in most cases it should be sufficient to consider the linear term bt for prediction of long-term durability in geologic disposal service.

B.2.1.2 Hydration

The aging, or weathering, of natural glasses by contact with humid atmospheres has been studied for many years in relation to the archaeological problem of dating natural artifacts. It seems unlikely that water vapor could exist in a repository by the time that the waste canister has been penetrated and the waste form becomes exposed to ambient conditions, but contact with a liquid phase could promote hydrothermal reactions. The results of past studies, as well as data from a recent investigation of accelerated aging of simulated waste glass, (BATES 1983) are beneficial to understanding the long-term alteration of glasses under hydrothermal conditions.

The hydration effects that may occur in liquid-phase experiments are part of the overall dissolution mechanism and are usually not distinguished as such. Phenomenologically, partial hydration in the glass leads to a more open structure, which permits ions and water molecules to penetrate the partially hydrated or gel layer with greater mobilities than the intact glass network. Hydration sometimes involves stress generation in the hydrated layer, causing swelling, contraction, cracking, or peeling of the layer (DAVIS 1982b).

B.2.1.3 Devitrification

Devitrification is the formation of crystals resulting from atomic rearrangement in the glassy matrix. Glasses represent a thermodynamically unstable system that naturally strives for an ordered structure with a lower energy state (e.g., crystal formation). The amount and identity of the crystals depend on the initial composition of the glassy matrix and on the annealing time schedule. Below the glass transition temperature, which (in the U.S. borosilicate glasses of interest here) is near 500°C, the viscosity of the glassy matrix is so high that crystallization is limited by diffusion to very low rates (MENDEL 1978), but eventual devitrification can be expected because of the inherent thermodynamic instability of the system.

Devitrification of a glass does not necessarily lead to disintegration. Changes in friability as a result of devitrification are slight with properly formulated waste glasses, but some microcracking can occur from volumetric changes due to the ingrowth of various crystalline phases. The principal problem arising from devitrification is that the overall leach rate of glasses is increased. The increase in leaching of borosilicate waste glass is usually no more than a factor of 2 to 5, although a factor of 10 increase has been observed in some instances for maximum devitrification (MENDEL 1978).

B.2.1.4 Radiation Enhancement of Leaching

Alpha particles, beta particles, gamma rays, and neutrons [from (α,n) and spontaneous fission reactions] can cause radiation damage in waste glass and could also affect the leach rate.

From the results obtained in a number of studies, Mendel (1978) concluded that leach rates of waste glasses containing fission products should not be significantly different from those obtained from nonradioactive simulated glasses of the same composition. The maximum factor of 2 found in his work was judged to be within the normal experimental deviation of leach test data. Similar data were obtained with ^{244}Cm-spiked waste glass.

Similar results were also reported in a more recent study by Walker (1981) and co-workers. In tests with borosilicate glass, he found that the leach rate was increased up to a maximum factor of 2 by gamma irradiation, presumably because of a change in leachant pH. No significant effects were observed for alpha or beta radiation.

After reviewing the available information, Davis and Schweitzer (DAVIS 1982b) concluded that it was difficult to assess the effects of radiation on the integrity of glass over geologic time. It was their opinion that the radiolysis effect was quite minor and any microfracturing that resulted from stress-induced radiation could be mitigated by proper design of the waste form. Similar conclusions were reached by Kircher (1983).

In a separate review, with emphasis on the work in the United Kingdom, the following conclusion was made by Burns (1981):

> The general conclusion is that these effects are not sufficiently large to give grounds for serious concern under the radiation conditions expected for a waste repository. If extrapolated to the doses and dose rates which will be used in practice, all pertinent experimental data obtained so far point to surface leach rates no more than a few times larger than those of unirradiated glasses. The water flow conditions (near stagnant conditions) in repositories should in any case be such that saturation solubility rather than surface leach rate controls the rate at which glass is dissolved into groundwater. The radiation effect on solubility has not been evaluated but it seems not unlikely to cause a significant increase.

B.2.1.5 Materials Interaction Relating to Waste Glass

Much research has been directed at the long-term interactions of borosilicate glass as a waste form in a geologic repository. The results of the research concerning interactions of borosilicate glass as a waste form with materials that will be in a repository are being reviewed and summarized by Bibler (1987).

He relates the material interactions that may occur between borosilicate glass, groundwater, and the components of a waste package to waste repositories in tuff, salt, and basalt and granite. The effects were summarized as shown in Table B.2.

In an overall evaluation, Bibler (1987) states that: "In many cases, the results in the above summary are incomplete and more tests need to be done.

Table B.2. Probable results from material interactions between borosilicate glass, groundwater, and components of waste package in a geologic repository

Repository media	Interaction material	Interaction results
Tuff	Repository rock	No effect on glass durability; sorption of certain radionuclides reduces their concentration.
	Stainless steel	No effect in saturated tuff; possible effect in unsaturated system due to sensitized steel.
	Radiation	In saturated system, buffers pH change caused by leaching; when moist air irradiated, HNO_3 forms that lowers pH.
Salt	Repository rock	No effect.
	Stainless steel	No effect.
	Iron overpack	Increases leaching of glass when O_2 present; forms reducing atmosphere in closed system which decreases solubility of certain radionuclides; forms corrosion products which sorb certain radionuclides.
Basalt and Granite	Repository rock	Causes reducing conditions in closed system which lowers solubility of redox sensitive elements; some sorptive properties indicated; little effect on durability of glass.
	Stainless steel	No effect.
	Iron overpack	Same interactions as in salt; data exist which suggest that iron may not cause increased leaching in reducing environments.
	Radiation	In a closed system, radiolysis appears not to increase the oxidizing potential of the rock-groundwater system.

However, the above results do indicate the substantial data base that has been generated over the past several years to support the complicated and difficult repository interaction tests that are now being performed in many laboratories throughout the world. Based on our review, we conclude that techniques have now been developed so that these tests can closely simulate repository conditions including reducing conditions in crystalline or salt rock repositories and unsaturated conditions in tuff repository. To be of value for the process leading to the safe geologic disposal of radioactive waste, future tests should apply the new techniques and, as much as possible, concentrate on the behavior of the specific radionuclides themselves while still evaluating the performance of the glass. Whenever possible, appropriate radiation levels should be included in these interaction tests."

B.2.2 Spent Fuel

For spent fuel disposal, it is proposed that the spent fuel rods, or pins (uranium oxide pellets clad in Zircaloy tubing), be removed from the fuel assemblies and consolidated in a close-packed array within the waste canister. The Zircaloy cladding will prevent leaching of the fuel as long as it is intact; however, it may become weakened or damaged while in service in a reactor. Some physical restructuring of the cladding will occur from radiation damage, as well as from chemical attack by some of the fission products. Currently, fuel pins have recorded less than one failure per 10,000, or less than 0.01% (SMITH 1980). No evidence of fuel rod degradation or failure after storage in water basins has been observed (JOHNSON 1977).

During service, the fuel pellets become heterogeneous, both physically and chemically, due to the steep temperature gradient. They may deform slightly, becoming bunched toward the center and expanding at the ends (SMITH 1980); significant cracking also occurs.

B.2.2.1 Zircaloy Cladding Failure

Zirconium alloys have outstanding corrosion resistance to pure water, all forms of aqueous salt solutions, and most acids and bases as well; however, they are subject to hydrogen embrittlement and stress corrosion cracking in some environments. Zircaloy-clad fuel elements are also subject to stress-corrosion cracking from the fuel side. A recent publication (SHANN 1983) discusses the effects of selected fission products and metal iodides on such cracking.

Hydrogen can be produced in a repository by corrosion of zirconium (particularly in the absence of an oxidizing agent) and by radiolysis. The solubility of hydrogen in zirconium alloys increases with temperature; and in cases where the zirconium is nearly saturated with hydrogen at high temperatures, an embrittled hydride phase will form on cooling.

If the exposure is at a temperature where the diffusion rate is extremely slow (probably <100°C), embrittlement becomes unlikely for many years (McCOY 1981). However, Zircaloy is subject to stress-corrosion cracking in low-temperature chloride solutions if its potential exceeds the free corrosion potential. Zirconium can be polarized into the cracking potential range by using an applied current from an external source or by coupling to a more noble metal (McCOY 1981). (See Sect. B.1.2.9 for additional discussion on Zircaloy corrosion.)

Zircaloy cladding has the potential to be a significant barrier to the leaching of spent fuel pellets; however, in the past, performance assessments have generally ignored the effect of Zircaloy cladding. Some preliminary assessments for a tuff repository using a nonoverpacked stainless steel canister in the unsaturated zone of the Yucca Mountain tuff indicated that the barrier effect of the Zircaloy cladding may need to be included in order to meet release rate criteria for radionuclides. A testing program for determining the efficacy of Zircaloy cladding in a tuff repository is in place at the Lawrence Livermore National Laboratory (OVERSBY 1983), which is discussed at the end of the next section.

B.2.2.2 Degradation and Leaching

In general, the rate of spent fuel dissolution or leaching will be influenced by the chemical composition of the groundwater, its pH, and the oxidizing or reducing capacity of the environment, particularly for elements such as uranium that can exist in either oxidized or reduced form.

In an early study on leach tests on PWR fuel pellet fragments using deionized water, building distilled water, and typical Hanford groundwater, Katayama (1976) found that the relative leachabilities of the elements decrease in the following order: Cs > Sb > Sr+Y > Pu > Cm. The data were fitted to an equation in the form of

$$F = Bt^n ,$$

where

F = fraction leached,
t = time (d), and
B and n = fit constants.

For Hanford groundwater and deionized water, the long-term leach mechanisms seemed nearly identical for $^{239+240}$Pu, ^{90}Sr + ^{90}Y, ^{244}Cm, and ^{137}Cs since the exponent (n) did not vary significantly with the radionuclide, averaging 0.07 for the groundwater and 0.31 for the deionized water. The exponent for the building distilled water, however, varied from 0.06 to 0.35, indicating different leach mechanisms for the individual radionuclides. This wide range was attributed to the varying quality of the building distilled water and the subsequent variation in the ionic concentration. Since an exponent of 0.5 would indicate a diffusion mechanism with a constant coefficient of diffusion, Katayama concluded that the 0.07 exponent reflected the major role of dissolved chemicals in suppressing leach rates and the 0.31 exponent probably reflected the inverse relationship of the diffusion coefficient for the radioisotope with its ionic concentration in the leachant.

In later studies, Katayama (1979) found that, based on the release of ^{137}Cs, the leach rate of spent LWR fuel was approximately five times lower in sea brine than in deionized water. He also concluded that the burnup of the fuel does not have a measurable effect on the leach rates, based on the behavior of ^{137}Cs, $^{239+240}Pu$, and uranium. It does have an effect on the leach rates of ^{244}Cm, however; results show that the higher the burnup, the lower the leach rate below 350 d.

Wang and Katayama (WANG 1981) performed a number of experiments with UO_2 single crystals and spent fuel in an attempt to develop a clearer understanding of the leaching mechanism. They concluded the following:

> The oxidation and dissolution mechanisms for UO_2 and spent fuel will be quite similar based on this preliminary work with electrochemical leaching of UO_2 and spent fuel. In solutions containing oxygen or other oxidizing species, the UO_2 surface will be rapidly oxidized and dissolved following the transformation of uranium from U(IV) to U(VI). The hydrolysis of dissolved uranyl ions forms solid UO_3 hydrates or related complex compounds deposited onto the UO_2 surface, or other surfaces, as thin or thick coatings. Depending on the pH, temperature and time, the various kinds of porosity and the mechanical properties of the hydrate coatings will control the dissolution rate. The effects of radiation in terms of generation of H_2O_2 will enhance the dissolution kinetics.

Mitchell (1981) studied the leaching of declad, irradiated fuel samples in a borate solution. The effects of temperature and fuel fragment size on the leach rate and released fraction of several fission products were examined. Some of the conclusions reached, after 250 d of leaching, were:

1. The relative order of the rate of leaching is: $Sb > I > {}^3H > Cs > Sr > Ru,Ce,Eu,U > Pu$.

2. Data fit to an equation of the form $F = Bt^n$ gave values for the exponent which varied from 0.01 to 0.59, depending on the leachate and the leaching conditions.

3. No significant differences in leach rates were found at 100°C and 85°C.

4. The leach rates (in units of $g\ fuel/cm^2 \cdot d$) were higher for the larger particle sizes.

5. The fractional releases (in units of fraction of the substance in inventory) were smaller for the larger particle sizes.

Results of the study gave fractional releases and relative leaching rates for the various fission products which were in general agreement with those obtained by Katayama. Differences between the studies may be attributed to the longer leaching time obtained in the study by Mitchell (40 weeks vs 20 weeks) and the difference in leachants (borate solution vs distilled water).

After analyzing the published data of G. L. McVay, of the Pacific Northwest Laboratory, on the comparison of the leaching of spent fuel chunks (no cladding included) from the H. B. Robinson reactor with deionized water, sodium bicarbonate solution, and salt brine B (see Appendix D, Sect. 1.3) at temperatures from 25 to 250°C, Kircher (1983) made the following general comments:

1. At 25°C, the cumulative release of actinides from spent fuel is approximately two orders of magnitude greater than that from PNL 76-68 glass.

2. At 250°C, there is no significant difference in the release from the spent fuel and that from the glass.

3. Spent fuel is as good a waste form as PNL 76-68 glass for retaining actinides at temperatures >150°C for the leachants tested.

In leaching studies of spent fuels from CANDU reactors, Vandergraaf (1980) stated that ~4% of the cesium leached out in a few days using chlorinated river water. The actinides and rare earths leached more slowly. The matrix itself dissolved at a conservatively calculated rate of $\sim 2 \times 10^{-6}$ g $UO_2/cm^2 \cdot d$, and eventually leach rates of the various nuclides approached this value.

In the program (OVERSBY 1983) to evaluate leach rates from wastes in a tuff repository, along with the efficacy of the Zircaloy cladding, tests are being made with PWR spent fuel rods from the H. B. Robinson and Turkey Point reactors using deionized water and J-13 well water under hot-cell ambient air and temperature conditions (WILSON 1985). Four specimen types, which represent a range of fuel physical conditions that may develop in a failed waste canister containing a limited amount of water, were used in the tests. These were: (1) fuel rod sections split open to expose bare fuel, (2) fuel rod sections with a 2.5-cm-long by 150-μm-wide slit through the cladding, (3) fuel rod sections with 200-μm laser-drilled holes through the cladding, and (4) undefected fuel rod segments. The principal conclusions based on the initial results were as follows:

1. The actinides, U, Pu, Am, and Cm, and most likely Np, appear to be released congruently.

2. Both Tc and Cs are preferentially released relative to the actinides.

3. Much greater total releases were observed in both J-13 and deionized water for bare fuel vs the same quantity of fuel contained in defected cladding.

4. Lower actinide releases were observed in the tests with J-13 well water than for the tests with deionized water.

On the basis of these results and the results of others (FORYSTH 1984; JOHNSON 1983; OVERSBY 1984), a preliminary model has been developed (OVERSBY 1985) to estimate the upper-bound release rates of radionuclides from spent fuel under conditions expected for a repository sited at Yucca Mountain. Experiments and tests are in progress to improve the understanding of the processes involved and to provide additional needed data. The most important are: (1) the dependence of uranium solubility on temperature, (2) the rate of failure of the containment barrier, (3) the rate of breach of cladding, and (4) the rate of oxidation of the fuel matrix.

Recently Bunker (1987) has reviewed the development in the release kinetics of nuclear waste glass leaching. He identified the major deficiencies in current models and key areas requiring further research to enable more accurate nuclide leach predictions to be made. He concluded that at the current level of understanding, essentially only qualitative predictions concerning the various factors involved in leaching can be made, which permits only "order of magnitude" or "upper limit" prediction of radionuclide release rates.

B.3 PACKING FAILURE MODES

The primary function of outer packing around a waste container is to control groundwater intrusion to the outer surface by providing a low-permeability barrier. Limiting the ingress and egress of groundwater serves to inhibit corrosion and the migration of radionuclides after leaching begins. Other possible functions of the packing include: (1) acting as a sorptive barrier for radionuclides; (2) chemically buffering or modifying the pH, redox conditions, or ionic composition of the intruding groundwater; and (3) providing a mechanical stress barrier against external forces.

Packing is included in only the reference design for a waste package for disposal in basalt. Currently, it is assumed to be a compacted mixture containing 25% bentonite and 75% crushed basalt. In an alternative waste package design for a tuff repository, packing in the form of crushed and pressed tuff containing 5 to 15% iron-bearing smectite clay as a binder is proposed if it is considered necessary (see Appendix A, Sect. 2.2). The crushed salt used in the annular space around a waste package in a salt repository is not intended to act as packing material in the previously described sense. Its primary function is to enhance heat transfer, particularly after reconsolidation.

Degradation of the packing or barrier is reflected in a shorter waste isolation period and, on breach of the canister, an increased transport rate of radionuclides into the surrounding country rock.

The degradation and failure modes for the packing have been discussed as a prelude to the DOE accelerated test program (DOE 1981). The discussion was further expanded and the modes were thoroughly reviewed by Eastwood (1982) and Dayal (1982).

Failure, or degradation, modes can be divided into two general classes: chemical/physical and mechanical. These are discussed in the following sections.

B.3.1 Chemical Failure, or Degradation, Modes

Chemical failure modes can be defined as chemical or physicochemical processes that degrade the performance of the packing material either (1) by increasing the permeability, decreasing the sorptive capacity, and making mineralogical alterations that contribute to such performance reductions, or (2) by indirectly contributing to mechanical failure.

A number of the processes involved in chemical degradation are not necessarily independent of each other. This makes classification of the failure modes somewhat subjective; however, a reasonable classification includes: (1) thermal stability, (2) diagenetic-like changes, (3) chemical poisoning or chemically induced mineralogic changes, (4) selective dissolution or leaching of the packing material, and (5) radiation effects.

B.3.1.1 Thermal Instability

The smectite clay, sodium bentonite, in the packing for a basalt repository is a rock consisting primarily of a montmorillonite clay mineral, which is an aluminosilicate composed of a two-dimensional, expandable layer lattice. The behavior of the interlayer water and structural water (hydroxyl groups) is important to the properties (swellability, permeability, and some mineralogic changes).

The reaction kinetics for alteration processes are sluggish, and this characteristic, along with other differences in conditions, has apparently contributed to some contradictions with regard to temperature effects and water loss. Wood (1981) suggests that the montmorillonite structure should be thermally stable up to 300°C and presumably retain its properties. This conclusion is based on experiments which indicated that interlayer water is lost reversibly up to 300°C and that structural water is not lost until temperatures >400°C are reached. This general conclusion seems to be confirmed by the results of Komarneni and Roy (KOMARNENI 1981a). Based on later experimental results, Allen and co-workers (ALLEN 1983) concluded that sodium-bentonite would retain its swelling properties on addition of water after initial drying up to 370°C.

In more recent experiments with packed columns containing a mixture of 25% bentonite and 75% quartz or basalt sand, Couture (1985) found that exposure to water vapor caused irreversible increases in the permeability of bentonite by factors up to 10^5 for temperatures up to 260°C. According to Couture, the available data suggest that moisture in limited amounts is more effective than excess groundwater or dry heat in altering the properties of bentonite, and he concludes that even brief exposure of bentonite-based packing material to water vapor at high temperatures in a waste repository could seriously impair its performance. Since the increases in permeability were due to a large decrease in the ability of bentonite to swell in water, Couture (1985) suggests that the problem will be less severe if the proportion of bentonite is higher and the material is highly compacted.

Temperatures in the bentonite should not exceed 400°C (current design limit for basalt is 300°C); consequently, it seems that the bentonite should reabsorb water when resaturation of the repository occurs, providing that no significant mineral alteration has occurred.

However, the more recent results by Couture (1985) creates some doubt as to the efficacy of a packing with only a 25% content of bentonite since some drying with subsequent exposure to water vapor can be expected prior to resaturation of a basalt repository. If a bentonite-crushed tuff mixture were used for the alternative design of a waste package for a repository in tuff, the probability of exposure to water vapor for a significant time would be even greater because of the lower pressure and the lack of a water continuum in the vicinity of the waste package.

These results raise questions as to the efficacy of bentonite as a packing material and seem to be at variance with earlier investigators. Mostly, they found that higher temperatures (in contact with liquid-phase water) were required to produce irreversibility of swelling on rehydration, which was probably due to mineralogical alterations. Couture found little mineralogical change and postulated that the reduced swelling capacity was the result of effects on the basal spacing of hydration layers by a minor redistribution of the changes in the clay structure.

B.3.1.2 Diagenetic-Like Changes

Diagenetic and hydrothermal reactions are lumped together here under diagenetic-like changes since these classifications can be somewhat arbitrary. "Diagenesis" is a term that is usually applied to mineralogical alterations below 200°C in sediments; the term "hydrothermal" generally refers to reactions with water that has been heated by contact with hot rocks or released from cooling magma chambers.

Early diagenesis is the most likely process for producing hydrothermal instability; for example, it is known that bentonites are formed near the surface of the earth at relatively low temperatures (50 to 100°C). Bentonites have limited stability at elevated temperatures, and their thermal stabilities are strong functions of imposed hydrostatic pressure, chemical composition, and ionic strength of the groundwater. Weaver (1979) even contends that montmorillonite alteration can occur at temperatures as low as 40°C, based on observations of long-term diagenetic alteration of clays in Gulf Coast sediments.

High K^+/Na^+ ratios will also contribute to instability by promoting alteration of montmorillonite to illite. Illite would not be a poor packing material, but its swellability and exchange capacity are lower than those for montmorillonite. Reduction in swellability will result in greater permeability of the backfill. Chlorite, which has even less desirable properties than illite, may also be formed as a by-product if magnesium or iron (as would be present for the iron-bearing smectite clay that has been proposed as a possibility in a tuff repository) is readily available when montmorillonite is altered to illite.

Apparently, the kinetics of the various possible reactions are sensitive to a number of conditions, and more carefully controlled experiments are required to fully evaluate the behavior of sodium-bentonite for repository conditions.

Aging is a diagenetic-like process that can be defined as a series of reactions that take place in metastable materials as they are converted to the lowest energy state (DOE 1981). Usually aging and microstructural changes involve recrystallization in amorphous phases such as glasses; however, mineralogic aging of bentonites may also occur. In view of the slow reactions that are frequently involved in the loss of hydrothermal stability, which leaves minerals in metastable states, the distinction between the two processes can become blurred and somewhat arbitrary. For bentonite, aging mechanisms could include mineralogic phase changes (e.g., clay diagenesis hydration or dehydration, lithification, and recrystallization).

Experiments have been conducted (WOOD 1983) in basalt/groundwater, bentonite/groundwater, and basalt/bentonite/groundwater systems at 300°C and 300 bars. A reference Umtanum basalt, a Wyoming sodium bentonite, and a simulated Grande Ronde groundwater were used. No illite formation was observed, and Wood concluded that sodium bentonite would remain sufficiently stable at 300°C under hydrothermal conditions in basalt to permit its use as a packing material. Based on the available information and the results of their work, Komarneni and White (KOMARNENI 1981b) and Sasaki (1982) also concluded that sodium bentonite would remain sufficiently stable up to 300°C for use as a packing material. Further, more recent evidence for bentonite stability in hydrothermal experiments up to 300°C was reported by Couture (1984) and Peacor (1984).

For the expected temperature and other conditions in a nuclear waste repository in basalt, it seems unlikely that mineralogic alterations could occur that would significantly degrade the packing performance; however, there is concern as to whether the available results are sufficient for long-term predictions with reasonable certitude. Concern has been expressed that insufficient experimental data are presently available to make reliable predictions about the kinetics of conversion of pure sodium montmorillonite to mixed layer clays, mainly because the experiments may have been too short to allow some of the possible alteration products to form in measurable quantities because of the sluggish kinetics. Eastwood (1983) is generally in agreement with this position and also points out that Wood (1983) apparently failed to recognize the possible importance of the nonswelling and nonsorption transformation products, such as albite and paragonite, that were detected in the experiments. Longer-term experiments seem necessary to resolve these concerns.

B.3.1.3 Chemical Poisoning or Chemically Induced Mineralogic Changes

Chemical poisoning of the packing would degrade radionuclide sorption capability and, possibly, the chemical conditioning of the groundwater. Possible mechanisms for this chemical degradation mode include: ion-exchange saturation or overload, precipitation, oxidation or reduction of chemical species, and formation of complexes. Physical mechanisms, such as pore plugging by precipitation or by particulates or colloids and microstructural changes with aging, can also decrease the sorption capability. If these reactions occur prior to the presence of radionuclides, the result is nearly always detrimental. However, when reactions involving radionuclides, such as precipitation, trapping, or fixation in the packing material matrix, permanently occur, these irreversible processes represent an enhancement of the sorption process.

The chemical composition of and the concentration of chemical species in the leachant, pH, redox conditions, temperature, radiation dose rate and cumulative dose, pressure, and time obviously affect the possible reaction mechanisms. The redox conditions around the waste package are particularly important for a number of nuclides; different oxidation states of an element can result in significantly different distribution coefficients (e.g., Np^{3+} and Np^{5+}; Tc^{4+} and Tc^{7+}). The effect of interbarrier interaction, if it occurs, could be similar to that caused by chemical poisoning.

The potential for many chemical reactions between the groundwater and packing material is very high. In view of the slow kinetics of many of these reactions and the unknown synergistic effects, long-term predictions require additional information.

B.3.1.4 Selective Dissolution, or Leaching, of the Packing Material

Selective dissolution, or leaching, may result in partial removal of specific packing components and contribute to degradation of the hydrologic and sorption functions of the packing, as well as cause adverse chemical conditioning of the groundwater.

The primary variables controlling selective dissolution, or leaching, of the packing material include: temperature, leachant composition, flow rate, material particle size, pH, redox conditions, and time.

Leach rates are generally increased by increasing the temperature and the leachant flow rate. Low flow-rate conditions decrease the leach rate by allowing the concentration of dissolved species to build up, thus hindering further dissolution. Decreasing the particle size of the packing material increases the surface area that is exposed to the leachant and may increase the leach rate. Conversely, the decreased particle size may reduce the permeability and thus decrease the leach rate. The pH is a major factor in the stability of the aluminosilicate phases since it strongly affects the solubilities of both silicon and aluminum. The stability of bentonites is adversely affected by both very low and very high pH levels; the low level favors the formation of kaolinite, while the very high level, coupled with high concentrations of alkali or alkaline earth metals, favors the formation of zeolites or feldspars — all at the expense of montmorillonite (WHEELWRIGHT 1981).

B.3.1.5 Radiation Effects

The most intense radiation field in the packing material results from gamma rays produced by relatively short-lived fission products (primarily ^{137}Cs) that will essentially have decayed away in 300 years. Neutron radiation is insignificant, and alpha and beta rays will be absorbed within the canister prior to a breaching event. Once leaching of the waste form begins, the packing will also experience some alpha and beta radiation from the radionuclides contained in the leachate.

These ionizing radiations may degrade the performance of the packing material by direct damage of the crystal structure and by radiolysis of the water contained in the bentonite or other packing material. The preponderance of the radiation dose will come from the gamma rays prior to a breach event; the additional effect of alpha and beta radiation after a breach can only be minor in comparison. Although structural damage from gamma radiation is usually minor in silicate or aluminosilicate minerals (WHEELWRIGHT 1981), high gamma-ray doses could cause damage to the crystal structure, which could enhance the degradation processes. Since the highest gamma-ray flux will coincide with the highest temperature, some of the structural damage will be annealed out. The crystal structure damage could, in theory, affect sorptive capacity, permeability, plasticity, swelling pressure, and thermal conductivity of the bentonite.

Radiolysis of the water produces H_2 and O_2, along with some other possible chemical species that change the pH and redox conditions. Such changes could conceivably affect both the sorptive capacity and the chemical stability of the packing materials.

If air is free to diffuse or move through a repository, a significant amount of HNO_3 may be generated. Irradiation of air produces oxides of nitrogen. Nitric acid is formed in the presence of liquid water vapor, and nitrates are formed in the presence of water vapor and bases (COUTURE 1984). The possible production of nitric acid in a repository has been considered based on literature data (STEINDLER 1984), and this study should be examined for a detailed discussion of the radiolytic chemistry of nitrogen. More recent experiments at 200°C appear to confirm the breakdown of montmorillonite in acid solution, and it was concluded that the amount of acid produced may be sufficient to adversely affect the stability of montmorillonite in packing material (COUTURE 1984).

Since radiation can affect the chemical reactions and mineralogy, it can be of importance in long-term predictions of the packing behavior. However, the current concept for a waste package utilizing relatively thick steel overpacks in repositories in basalt and salt will considerably mitigate the effects by reducing the gamma dose rate by an order of magnitude (prior to large metal loss by corrosion). Radiation doses will be higher around the nonoverpacked waste canister in tuff, but the effect of radiation is not as important in this case.

B.3.2 Physical or Mechanical Failure, or Degradation, Modes

Physical or mechanical failure, or degradation, modes include: (1) hydrologic erosion, (2) cracking or fissuring, (3) detrimental changes to mechanical properties, and (4) inadequate swelling of bentonite or other packing material.

B.3.2.1 Hydrologic Erosion

Hydrologic, or hydraulic, erosion is a physical "washaway" of the packing material. In order for this type of failure to occur, the flow rates would have to be increased beyond that expected for the regional hydrology. Thus, hydraulic erosion could result from a change in the regional hydrology or intrusion by man.

B.3.2.2 Cracking or Fissuring

Cracking or fissuring could occur by drying after some degree of saturation of the bentonite or other packing material. If continued wet/dry cycling occurs, cracking could happen when drying begins after initial swelling from water absorption, with subsequent shrinkage of the bentonite. However, continued wet/dry cycling is not a credible scenario. Only one cycle can be visualized, and it is discussed below.

A cracking scenario suggested by Siskind (1982) concerns the emplacement of bentonite in a partially hydrated condition. Initially, heat from the waste may reduce the water content and cause cracking. This possible mode of failure should be of limited importance, however, since the presence of groundwater would eventually cause rehydration and closure of the cracks by swelling prior to a breach of the waste canister.

B.3.2.3 Detrimental Changes to Mechanical Strength of Packing

Exceeding the liquid limit of bentonite by absorption of groundwater could affect the capability of the packing for mechanically supporting the waste container. As the liquid limit is exceeded and the shear strength approaches zero, the waste container could sink to the bottom of the emplacement hole, or segregation of the basalt-bentonite packing could occur and compromise the radionuclide containment function. In order for this scenario to occur, it appears that sufficient water for "washout" should be available.

B.3.2.4 Potential Design Inadequacies of the Packing

The primary function of the packing is to inhibit water access to the overpack. As bentonite absorbs water, swelling occurs and the permeability and porosity are reduced. With an initially dense bentonite-basalt mixture (or other swelling clay and rock mixtures), the swelling pressure that can develop will theoretically seal cracks in the rock immediately surrounding the waste package and reduce the permeability and porosity of the packing to very low values. The previous concept for disposal in basalt that utilized a 25% bentonite—75% basalt mixture with an estimated 50% voids after a pneumatic emplacement may not be adequate at the expected temperatures since the bentonite would need to expand by a factor of 4 to reduce the porosity and permeability to the very low value that is assumed for diffusion-controlled transport through the packing. It is not clear that a significant swelling pressure can develop for such an emplaced mixture based on the extensive information developed by Pusch (1979 and 1980). More recent experimental work by Westsik (1983b), however, indicates that sufficient swelling pressures may develop for such a design. Such concerns in regard to adequate swelling and difficulty of ensuring adequate pneumatic emplacement were apparently instrumental in developing the current concept that utilizes preformed, compacted packing.

Concern as to the efficacy of bentonite packing has also been expressed in reviews by Soo (1984) and Peacor (1984). Peacor points out that in order to have a very low porosity packing it is necessary to have a high proportion of clay minerals forming, when compacted, a continuous structural network. Consequently, Peacor concluded that it is essential for packing materials that include clays to have (1) a high proportion (>50%) of clay; (2) a low proportion of granular material (e.g., basalt which should be as fine-grained as possible since large nonclay grains interrupt the clay network); and (3) a high degree of compaction. The conclusion by Peacor that 75% clay may be required, along with the results of Couture (1985) (see Sect. B.3.1.1 for a discussion), creates doubt about the efficacy of the preformed, compacted packing in the current concept since the use of only 15% bentonite is anticipated. A similar concern can be expressed for the adequacy of the packing, if considered necessary, for disposal in tuff since the bentonite content is proposed to be only 5 to 15%.

In the earlier BWIP design, the uncompacted density of the packing when saturated with water will be less than 1.75 t/m^3 — the lower limit of the suite of densities of compacted, water-saturated bentonite for which Pusch (1980) determined swelling pressures. He found that swelling pressure is a very sensitive function of the bulk density, degree of saturation, and particular properties of the bentonite. In addition, Pusch (1980) estimated that a temperature increase to 70°C reduces the pressure to ~50% of the value at 20°C.

In estimating swelling pressures for saturated bentonite with densities <1.75 t/m^3, Pusch (1980) reinterpreted (method undescribed) the data (presumably for 20 or 25°C) for pure montmorillonite gels from 35 different locations (saturated densities from 1.3 to 1.65 t/m^3) obtained by Low (1980). Pusch concluded that the fresh water-saturated MX-80 bentonite (the type used in the Pusch experiments), expanded to give a density of 1.3 t/m^3, produces a swelling pressure of not less than 0.1 MPa at room temperature. In the experiments conducted by Low (1980), the water content at equilibrium was determined for the clay-water mixture as a function of applied pressure. These data were used to develop a general empirical equation in which the swelling pressure depends exponentially on the water content and a parameter that was an empirical linear function of the specific surface area and the cation exchange capacity of the montmorillonite. Additionally, these data were used to develop semiempirical equations in which the swelling pressure depended exponentially on the distance between the superimposed layers of montmorillonite, which is a function of the surface charge density of the montmorillonite. In view of the assumptions made in developing these low-density equations, the application to a repository may give rise to a large uncertainty because of the difficult mechanism involved in the saturation of the bentonite and development of the swelling pressure. In addition, swelling pressure has been shown to decrease significantly with rising temperatures and to be a rather sensitive function of the particular kind of bentonite. In the more recent experiments by Westsik (1983b), swelling pressures of 5 to 7 MPa developed for pure sodium bentonite with an as-pressed density of 1.7 t/m^3. It is difficult to put a lower limit on the packing density, but densities >2 t/m^3 seem to be desirable.

In view of the current information, experiments more representative of repository conditions will be required before it can be assumed that bentonite will perform as generally anticipated.

B.4 SUMMARY OF THE IDENTIFICATION AND PRIORITIZATION OF FAILURE MODES

The purpose of this section is to summarize the results and evaluations discussed in the preceding sections. To facilitate this goal, four summary tables (Tables B.3 through B.6) have been prepared to present the failure modes and aspects of their prioritization in a condensed form. Each of these tables has the same general format, which is defined, from left to right, as follows:

1. Failure/degradation mode — a short phrase identifying each failure mode considered.

2. Description — a short description of the effect/process involved in the subject failure mode.

3. Limiting environmental conditions — the minimum repository conditions necessary for the subject failure mode to proceed; can be viewed as a threshold for the failure mode.

4. Controlling environmental conditions — the repository conditions that control the rate, or extent, of the subject failure mode.

5. Major interactions — identification of the potential interactions (synergisms) between failure modes that might significantly reduce the package lifetime.

6. Priority and rationale — statement of the assigned priority and the rationale for the assignment. The time frame considered is 10,000 years. The priority scheme employed is as follows:

   a. Dominant — a failure mode that will clearly be important and for which there are no major competing failure modes.

   b. Significant — a failure mode that will be important, but at the same time there are other failure modes of similar importance.

   c. Insignificant — a failure mode which cannot occur or which has a relative probability so low that it is not important compared to other failure modes.

   d. Unknown — a failure mode in which the answer to a critical question was required for resolution and the answer could not be determined; the nature of the question is identified below.

7. Failure mode models — identification of existing models which purport to analyze waste packages with respect to the subject failure mode.

8. Major unknowns — identification of questions that should be resolved to fully address the importance of the subject failure mode (always used if an unknown priority was assigned).

It should be noted that the identification of major interactions only includes the synergism between the effects of the failure modes (i.e., corrosion weakening the package components sufficiently so that they fail by ductile rupture). It does not account for second-order, but potentially important, effects such as the effects of corrosion products on corrosion rates or heat transfer.

As a further summary aid, the failure modes assigned dominant, significant, or unknown priorities are summarized in Table B.7.

Table B.3. Summary of characterization and prioritization of overpack and canister thermal/mechanical failure modes

Failure mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Specific failure mode models	Major unknowns
Yielding	Permanent deformation caused by stresses exceeding yield point	1. Applied stress greater than yield stress	1. Temperature 2. Pressure 3. Rock plasticity	1. Would be affected by prior weakening modes 2. May affect overpack and packing performance	Insignificant; other failure modes will dominate. Yielding not a breach failure	WAPPA BARIER	None
Ductile rupture	Applied force sufficient to rupture or tear metal while in a ductile state	1. Applied shear stress greater than ultimate strength 2. Temperature above ductile-brittle transition	1. Temperature 2. Differential pressure 3. Rock plasticity	1. Would be affected by prior weakening modes 2. Would affect overpack and packing performance	Significant likely after overpack has been sufficiently weakened by other modes	None identified	Potential existence of severe tectonic shearing stresses
Brittle fracture	Rapid propagation of a crack across a stressed metallic component	1. Temperature below ductile-brittle transition 2. Tensile stress >35 MPa	1. Temperature 2. Pressure 3. Rock plasticity	1. Would affect overpack and packing performance	Insignificant; proper choice of steel will eliminate brittle fracture possibility	WAPPA	None
Fatigue – high cycle, low cycle, thermal, corrosion, and fretting	Metal fractures or ruptures after repeated cycling through a stress range	1. Excessive number of cycles of stresses or strains 2. Excessive stress	1. Temperature 2. Temperature changes 3. Pressure 4. Pressure changes 5. Corrosive conditions	1. Would be affected by prior weakening modes 2. Would affect overpack and packing performance	Insignificant; a scenario involving sufficient cycles through a sufficient stress range is not credible	None identified	None
Creep (also stress rupture, an accelerated creep test to rupture)	Creep under stress through tertiary or accelerating creep	1. Temperature and stress > that is required to produce tertiary creep	1. Temperature 2. Pressure 3. Rock plasticity	None	Insignificant; temperatures and stresses are insufficient to produce tertiary creep by design	None identified	None

Table B.3 (Continued)

Failure mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Specific failure mode models	Major unknowns
Thermal relaxation	Preload stress needed to carry a load is relieved during operation	1. Excessive stress and deformation	1. Temperature 2. Pressure 3. Pressure changes 4. Temperature changes	None	Insignificant; the need for pre-load design requirements cannot be visualized	None identified	None
Spalling	Sudden chipping fragmentation, or flaking of material	1. Shock waves	1. Shock waves	None	Insignificant; shock waves sufficient to cause metallic spalling of ductile material cannot be visualized	None identified	None
Buckling	Failure occurs when the deflections of a member became too large for structural stability	1. Critical loading for buckling is exceeded	1. Temperature 2. Stress distribution	None	Possibly significant after severe degradation. Buckling of the relatively short cylinders cannot occur until metal is thin and some glass (or SF) has been leached; wrinkling of TiCode-12 clad is possible if not properly installed	None identified	None
Creep buckling	Failure occurs after creeping increases deflections to point of structural instability	1. Critical loading for buckling is exceeded 2. Temperature and stress greater than that required to produce tertiary creep	1. Temperature 2. Stress distribution	None	Insignificant; large creep deformations for metals at expected temperature and stresses cannot be visualized	None identified	None

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[a] Carbon steel.

Table B.4. Summary of characteristics on prioritization of overpack and canister corrosion failure modes

Failure mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Failure mode models	Major unknowns
1. General corrosion	Uniform wall thinning	1. Presence of aqueous phase	1. Temperature 2. Radiation 3. Salt concentration 4. pH 5. Corrosion product scale	1. Produces H_2 that may produce H_2-related failure 2. Reduces load carrying ability	Significant for steel; wall thinned to point of collapse. Not significant for TiCode-12 and stainless steel	BARIER WAPPA	None
2. Stress corrosion cracking	Development of fine cracks that penetrate wall	1. Presence of certain ions in solution (e.g., $CO_3^{=}$-HCO_3^{-}) 2. Oxidation potential of solution 3. Threshold stress	1. Specific salts in groundwater 2. Redox potential of solution 3. pH 4. Temperature	1. May originate at pits 2. Stresses may be caused by shifts in geologic formation	Significant; experiments may show SCC to be improbable, but for present cracking must be considered likely	None	None
3. Pitting corrosion	Deep penetrations in highly localized spots that may penetrate the wall	1. Presence of specific ions in solution 2. Oxidation potential of solution	1. Aggressive ions in solutions 2. Temperature 3. pH 4. Redox potential	1. May be source of stress corrosion cracking	Significant; experiments may show that pitting is insignificant but until proven otherwise pitting must be considered likely	None	None
4. Crevice corrosion	Localized attack on areas that have restricted access to aqueous environment	1. Ionized salt in solution 2. Presence of oxidizing agent	1. Minimum salt concentration 2. pH 3. Temperature 4. Oxidizing agent	1. Can aggravate all other forms of corrosion	Unknown for basalt and salt; does a "crevice" exist when overpack is backfilled? What does radiation do to crevice corrosion?	None	1. Effects of radiation 2. Is there "crevice" on buried overpack
5. Intergranular corrosion	Selective attack at grain boundaries that reduces load carrying ability of metal	1. Presence of ions that attack grain boundaries in preference to grains	1. pH 2. Temperature 3. Aggressive reagent 4. Susceptible material in grain boundaries	1. Reduce load carrying ability of overpack 2. May lead to intergranular SCC	Insignificant for tuff waste package; steel should have minimum tendency toward general intergranular corrosion. Possibly significant for stainless steel	None	Long-term low-temperature sensitization of low-carbon stainless steel
6. Stray current corrosion	Metal objects may carry d.c. ground currents. Where current leaves metal to return to ground, heavy attack occurs	1. Ground must have some minimum conductivity to carry current 2. d.c. ground currents	1. Conductivity of earth (salt concentration)	1. Could change potential of overpack to stress corrosion cracking range 2. Could facilitate pitting	Insignificant; unlikely based on present use of d.c. and repository depth	None	None
7. Microbial corrosion	Corrosion produced by the metabolic products of bacteria	1. Presence of microorganisms that use sulfur in their metabolic processes 2. Presence of some form of sulfur 3. Temperature <100°C	1. Presence of sulfur assimilating bacteria 2. Temperature 3. Radiation	1. Increase H_2 uptake by production of H_2S 2. Increase general corrosion rate	Insignificant; temperature probably too high for bacteria	None	None

Table 8.4 (Continued)

Failure mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Failure mode models	Major unknowns
8. Hydrogen-related failures							
a. Overpressurization of overpack	Hydrogen either from corrosion or from high pressures of gaseous hydrogen could penetrate through steel and generates high internal pressure	1. Temperature 2. Corrosive environment 3. High H_2 pressure around overpack	1. Temperature 2. Corrosion rate 3. H_2S	1. May affect packing 2. Other corrosion increases the H_2 uptake	Insignificant for steel and stainless steel. Significant for TiCode-12—could lead to hydride formation	None	None
b. Hydrogen blistering	Hydrogen collects at inclusions in steel near the surface and pressure causes blistering	1. Temperature 2. Corrosion environment	Same as above	1. May increase general corrosion by increasing surface area	Insignificant for steel and stainless steel. Significant for TiCode-12—could lead to hydride formation	None	None
c. Hydrogen attack (methanation)	Embrittlement of steel due to formation of methane bubbles in steel from reaction of carbon with hydrogen either from corrosion or from high pressure of gaseous hydrogen	1. Temperature >250°C 2. Corrosion of steel to produce H_2 3. High partial pressure of gaseous hydrogen (>100 psi)	Same as above plus high pressure of H_2 in system	1. Increase stress and increase likelihood of SCC 2. High corrosion rate increase H_2 uptake	Significant for steel only; methane may embrittle steel but small additions of Mo or Cr in alloy can eliminate problem	None	None
d. Hydrogen Embrittlement (TiCode-12 only)	Formation of a titanium hydride leading to embrittlement	1. High pressure of hydrogen	1. Corrosion rate parameters 2. Hydrogen pressure 3. Prescence of H_2S		Significant; as temperature decreases, formation of hydride phase and subsequent embrittlement likely	None	None

Table B.5. Summary of characterization and prioritization of waste form degradation modes

Degradation mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Specific failure mode models	Major unknowns
Matrix dissolution (also called corrosion and leaching)	Matrix dissolution is the physical removal of material at exposed surface	1. Local water unsaturated with respect to matrix material 2. Water in contact with waste form	1. Temperature 2. Local water composition, pH, Eh, and ionic strength 3. Local water flux	1. Effect of leachate on corrosion of other materials 2. Local water containing leachate can affect sorptive properties of backfill	Dominant; eventually all barriers must fail and allow leaching of waste form	WAPPA BARIER	None
Hydration (not applicable to S.F.)	Penetration of water molecules into glass structure creating gel layers that cause swelling and cracking	Water in contact with waste form	1. Temperature 2. Local water composition, pH, Eh, and ionic strength 3. Repository water flux	Affects matrix dissolution and nuclide diffusion within the glass	Insignificant; the effect is included in leach rate determinations	None identified	None
Devitrification (not applicable to S.F.)	The formation of crystals from atomic rearrangement in the glass matrix	1. Temperature	1. Temperature; below 500°C the devitrification rate is very low	Devitrification causes an increase in matrix dissolution	Significant; data indicates increased leach rates by factors of 2 to 10	None identified	None
Radiation enhancement of leach rates	Radiation damage to the glass matrix can increase leach rate or affect leachant pH. No significant effect on S.F. since it has been damage saturated in reactor	1. Presence of radiation 2. Water in contact with waste form	1. Dose rate 2. Cumulative dose	Could increase matrix dissolution	Insignificant; consensus seems to be from a small effect to a maximum of a factor of 2 on the leach rate for borosilicate glass	None identified	None
Zircaloy cladding failure (not applicable to glass waste)	Zircaloy cladding of fuel pellets subject to failure from stress corrosion cracking and hydrogen embrittlement	1. Hydrogen present 2. Coupling with a more noble metal can polarize Zr into the cracking potential range	1. Hydrogen partial pressure 2. Temperature	Cladding will increase the time before leaching begins	Unknown; Cladding could significantly increase time before leaching. The effect of cladding has been ignored in past performance assessments which is conservative.	None identified	Long-term behavior of zircaloy cladding in repository

Table B.6. Summary of characterization and prioritization of packing material degradation modes[a]

Degradation mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Specific failure mode models	Major unknowns
Thermal instability	Water loss causes shrinkage. Water loss above 400°C is irreversible and possibly lower temperatures	1. Temperature above 400°C	1. Temperature 2. Pressure 3. Water characteristics	1. May affect source term 2. May affect temp. 3. Increase in permeability will permit greater water access to overpack and could enhance rates of corrosion	Probably insignificant; bentonite temperature above 300°C is not expected. Initial drying should be compensated by resaturation; although recent tests indicate effects of water vapor at lower temperatures	None identified	1. Long-term effect of water vapor at 200°C on permeability
Diagenetic-like changes	Includes all mineral alterations as the result of chemical and physical geochemical reactions	None	1. Temperature 2. Pressure 3. Water characteristics	1. May affect temp. 2. Will affect permeability 3. Will affect sorption properties 4. May affect source term	Unknown	None identified	1. Effect of alteration on long-term sorptive properties and permeability of the packing 2. Degree of alteration possible
Chemical poisoning or chemically induced mineralogical changes	Chemical degradation modes include ion exchange saturation, precipitation, redox reactions and formation of complexes. Physical mechanisms include pore plugging and microstructural changes	None	1. Temperature 2. Pressure 3. Water characteristics	Increased rates of nuclide transport after breach	Unknown	None identified	1. Effect of chemicals on long-term sorptive properties and permeability of the packing 2. Degree of alteration possible
Selective dissolutioning or leaching of packing material	Selective dissolution may result in partial removal of specific components	None	1. Temperature 2. Pressure 3. Water characteristics	May contribute to the degradation of the hydrologic and sorption functions of the packing and adversely chemically condition the repository water	Unknown	None identified	1. Effect of component removal on long-term sorptive properties and permeability of the packing 2. Degree of alteration possible
Radiation effects	Irradiation of packing damages crystal structure and causes radiolysis of water	Presence of radiation	1. Dose rate 2. Cumulative dose	1. Crystal structure damage could affect sorptive capacity, permeability, plasticity, swelling pressure, and thermal conductivity of bentonite 2. Radiolysis of water produces H_2 and O_2 and other chemical species that change Eh and pH. Irradiation of air to produce nitrogen oxides.	Unknown	None identified	1. Effect of radiation on long-term sorptive properties and permeability of the packing 2. Degree of alteration possible

Table B.6 (Continued)

Degradation mode	Description	Limiting environmental conditions	Controlling environmental conditions	Major interactions with other failure modes	Failure mode priority and rationale	Specific failure mode models	Major unknowns
Hydrologic erosion	Physical "wash away" of packing materials	Water velocity insufficient to entrain packing	Water velocity	1. Creates ready access of water to overpack and enhances corrosion rates and leach rates after breach of the canister 2. Affects temperatures	Insignificant; site selection will eliminate or reduce to very low probability	None identified	None
Cracking or fissuring	Drying of moist, uncompacted packing resulting in openings in the packing that permit direct access of water to the overpack	Insufficient water for resaturation	1. Temperature	Creates ready access of water to overpack and enhances corrosion and leach rates	Insignificant; wet/dry cycling cannot occur; water available for resaturation in hard rocks	None identified	None
Detrimental changes to mechanical properties	Strength of bentonite could be reduced by absorbing water to the point of failure to support waste canister	1. Excessive water present 2. Enough room for packing to expand	None	Mechanical strength could be reduced to where shear strength approaches zero and waste canister sinks to the bottom. Segregation of basalt-bentonite packing could occur and compromise radionuclide containment and enhance corrosion rates	Insignificant; highly improbable because of site selection and assuming adequate QA	None identified	None
Inadequate swelling	Swelling of bentonite may be insufficient to seal gaps, cracks, produce low permeability	None	None	Permeability permits enhanced corrosion rates and rates of nuclide transport	Significant; current design may have insufficient bentonite fraction	None identified	Minimum bentonite fraction to produce desired swelling pressure for proposed emplacement technique and repository conditions

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[a] Assumed to be 75% bentonite/25% basalt. Packing is not included in reference diagrams for tuff and salt repositories. In the alternate design for tuff, which may include a mixture of crushed tuff and bentonite, the comments above will apply.

Table B.7. Summary of dominant, significant, and unknown waste package failure, or degradation modes, for materials used in repositories in basalt, tuff, and salt

Package component	Failure/degradation modes		
	Dominant	Significant	Unknown
Overpack		Ductile rupture (all) General corrosion (steel) Stress-corrosion cracking (all) Pitting corrosion (steel and stainless steel) Hydrogen attack (steel) Hydrogen embrittlement (TiCode-12)	Crevice corrosion (TiCode-12)
Waste form	Matrix dissolution	Devitrification (glass)	
Packing		Inadequate swelling	Diagenetic-like changes Chemical degradation Selective dissolution Radiation effects

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APPENDIX C

IDENTIFICATION AND PRIORITIZATION OF SIGNIFICANT REPOSITORY ENVIRONMENTAL PARAMETERS

The conditions that exist in a waste repository can be classified into environmental parameters that are necessary inputs to the performance assessment of the repository. Environmental parameters have been previously identified and estimated for typical repositories in salt for commercial and defense high-level wastes and spent fuel (RRCIWG 1983 and SASTRE 1986). These parameters include temperature, pressure, groundwater characteristics, and radiation fields. They are shown in Tables B.1 through B.4 in relation to the various failure and degradation modes of a waste package.

In ranking these parameters in order of importance, the more critical parameter was determined based on the following considerations: (1) effects on the life of the waste package, (2) effects on radionuclide release, (3) effects of the degree of uncertainty of the values of the parameters on the performance assessment of the waste package, and (4) design controllability of the values of the parameters in the engineered barrier system. Even though the environmental parameters are not completely independent of one another, they are usually treated as decoupled — a quite reasonable approach in most cases. For example, the water quantity and flow rates can influence the temperature; the temperature and radiation fields can affect the water chemistry; and the temperature determines water vapor pressure and influences thermal stress and waste package and rock deformations. In turn, the pressures and thermal stresses in the rock can affect water flow rates in the vicinity of the waste package and in the packing. In addition to these interrelationships, determination of the importance of each parameter with respect to assessing waste package performance involves both site selection and repository design, which reflects the degree of understanding of the potential mechanisms leading to inadequate performance and their control or mitigation. For example, temperature might be considered the most critical parameter because of its obvious strong influence on corrosion rates (which determine canister lifetime), matrix dissolution leach and radionuclide diffusion rates (which determine the upper limit of radionuclide release rates), and thermal stresses (which influence permeability of the country rock and water access). However, design control of temperature is straightforward since the canister and areal thermal loadings can be adjusted to limit temperatures to the desired values based on predictions using validated heat-transfer models. In addition, these temperature predictions for the near field can be confirmed in a few years after waste emplacement and prior to repository sealing.

Under anticipated conditions, the maximum pressure exerted on the overpacked canister in the first few thousand years will probably be between hydrostatic and lithostatic for the Hanford site and close to atmospheric for the Yucca Mountain site. A salt repository should attain lithostatic pressure in a few tens of years after an initial increase around the canister that could exceed lithostatic because of thermal stress. These thermal stresses could also aid in promoting brine migration along grain boundaries in a direction away from the waste package (see Sect. A.3.4.4). The pressure ranges to be expected should present no significant design problems and should not exert a strong influence on the performance of a waste package designed to withstand the pressure developed.

The major effect of radiation will be the changes in water chemistry as the result of radiolysis and, as such, could be considered a subset of the groundwater characteristics. The radiation field will be controlled by the canister loading and overpack design and can be easily and accurately measured.

The groundwater characteristics (flow rate, composition, pH, and redox conditions) have a large effect on the mechanisms and rates of corrosion and on the waste leach rates. In view of the potential effects of reactions between the groundwater and repository contents on the groundwater characteristics, the long-term prediction of the corrosion and leach rates probably presents the greatest uncertainty in predicting the performance of the waste package.

As a consequence of the previous discussion and our definition of critical parameter, we rank the environmental parameters in descending order of importance as follows: (1) groundwater characteristics (including flow rate), (2) temperature, (3) radiation field, and (4) pressure.

Sastre and co-workers (SASTRE 1986) also recognize the importance of these environmental parameters, but they list temperature ahead of water characteristics with no comment on their relative importance. As previously discussed, it is difficult to rank them precisely; however, the actual ranking becomes immaterial since one cannot be properly considered without the other.

A more-detailed discussion of these individual environmental parameters and rationale for our ranking is given in the following sections.

C.1 GROUNDWATER

Transport of radionuclides to the biosphere by groundwater is generally considered to be an eventual certainty. The waste package criteria of containment for 300 to 1000 years and controlled release after failure are designed to delay and mitigate the consequences of radionuclide release from a repository. The conceptual designs of the waste package for basalt and salt repositories are strongly dependent on the steel overpack as a means of complying with regulations. In the tuff repository, it is currently assumed that the unoverpacked canister is sufficient. The most credible failure mode of the waste package in the first few thousand years appears to be a breach of the overpack and waste canister as the result of the various possible corrosion mechanisms involving the groundwater. In addition, transport of radionuclides by groundwater represents the only credible threat to the biosphere by a waste repository. Consequently, groundwater is the greatest threat to waste package integrity and controlled release after leaching of the waste form begins.

The corrosion and leach rates will depend on the flow rate, composition, pH, and redox condition of the undisturbed groundwater as altered by interactions with the waste package components and radiation effects, such as radiolysis of the water and radiation damage to the waste form. The corrosion and leach and radionuclide diffusion rates are also strongly influenced by temperature; however, as previously discussed, temperature is a design-controllable parameter.

The characteristics of the undisturbed groundwater is obviously not a controllable environmental parameter, but the components are measurable (except perhaps the redox condition). The difficulty arises in predicting the long-term characteristics of the groundwater after alteration due to interaction with the waste package. Davis and Schweitzer (DAVIS 1983) have presented a detailed discussion of these difficulties and the subsequent uncertainties.

C.2 TEMPERATURE

There is no question that temperature is an all-pervading parameter that strongly affects the most important performance characteristics, namely, corrosion and leach and diffusion rates. However, as previously mentioned, it is a controllable environmental parameter and the uncertainty in the predictions will be relatively small and verifiable during the operational phase of a repository. The primary effect is to increase the corrosion and leach diffusion rates since these generally increase with temperature and may create larger uncertainties in the measured values.

C.3 RADIATION FIELDS

As previously mentioned, the radiation field is a controllable parameter that primarily affects the water chemistry as the result of radiolysis and, as such, could be considered a subset of groundwater characteristics. The steel canister and overpack will initially reduce the gamma dose rate at the overpack-packing interface by about an order of magnitude and thus reduce the radiolysis effect. Radiation damage to the waste glass by alpha and beta particles can be expected; however, the effect on the leach rate is apparently minimal (see Sect. B.2.1.4).

C.4 PRESSURE

Pressures on an overpacked waste canister can be expected to vary from atmospheric to lithostatic. After the basalt repository has been sealed, water will gradually fill the void spaces and the pressure exerted will increase to hydrostatic in tens to hundreds of years. Hydrostatic pressures will normally not develop in tuff or salt repositories since a continuous water column cannot exist. Development of lithostatic pressure due to rock creep or subsidence is unlikely for several thousand years under anticipated conditions for either basalt or tuff but can develop in a few tens of years in salt. In the very near field adjacent to the waste package, borehole closure in salt can result in local thermal stresses that cause a rapid increase in the pressure in a waste package. Conservative calculations by Callahan (1975) for an earlier conceptual design that included a steel sleeve in close contact with the rock salt indicated that these localized stresses exceeding lithostatic in the form of radial compressive stresses on the waste package may develop in the first year and then decrease as the result of salt creep. Recently, Cunnane and Pederson (CUNNANE 1986) reported on borehole closure rates for a configuration similar to the current conceptual design. The principal results, shown in Fig. C.1, indicate that closure of the borehole can be expected within two years, except near the top.

In the case of the designs for the Hanford site, the pressure around the canister could also increase as a result of the swelling pressure exerted by the bentonite. This swelling pressure will create a radial compressive stress on the overpack and tend to create a tensile stress on the rock surface, which could cause additional cracking of the rock. Consequently, pressures significantly above lithostatic, in the near and far field, should not exist because of the initially fractured condition of the rock and additional thermal fracturing will tend to relieve the rock stresses. The swelling pressure from the bentonite is controllable through design. The current conceptual design for the Hanford site, which is a 75% basalt—25% bentonite mixture, may not produce sufficient swelling pressures to produce anticipated low permeabilities for the repository conditions since the bentonite may dehydrate irreversibly (see Sect. B.3.1).

Consequently, the maximum pressure exerted should be between hydrostatic and lithostatic in the long term for the Hanford site and should not significantly exceed lithostatic as a result of local stresses. During the isolation period, the pressure should not significantly exceed hydrostatic because of the stability of the backfilled repository. These pressures will not contribute significantly to the uncertainties in other parameters.

Unexpected tectonic activity could produce unequal forces and pressures that could shear the waste package, but this is improbable because of site selection criteria.

C.5 REFERENCES FOR APPENDIX C

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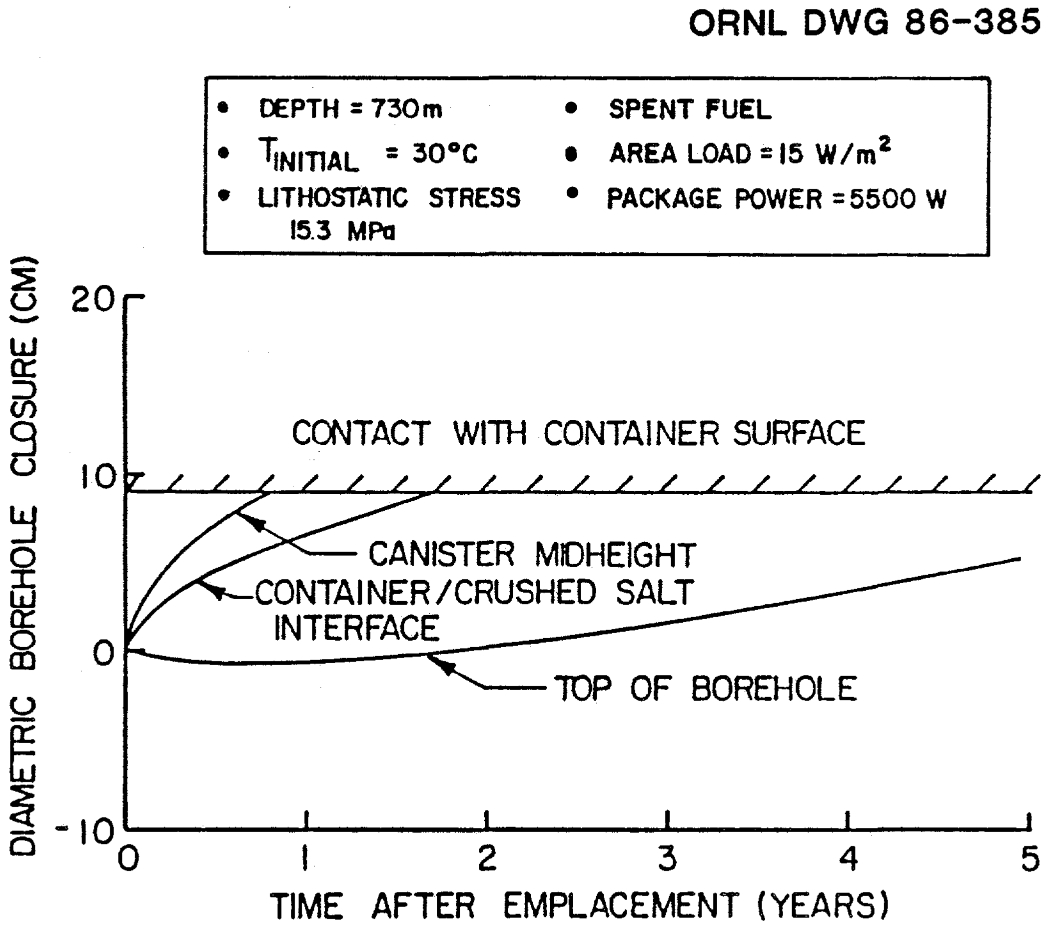

Fig. C.1. Diametric borehole closure rates for a typical design. Source: J. Cunnane and L. Pederson, "Waste Package Environment," presented at the SRP/NRC Waste Package Meeting, Columbus, Ohio, Jan. 22–24, 1986.

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APPENDIX D

RANGE OF ENVIRONMENTAL PARAMETERS

In this appendix, the reasonably possible or credible ranges of environmental parameters (groundwater characteristics, temperature, radiation field, and pressure) are estimated in some cases, and the rationale for the selected values for basalt, tuff, and salt repositories at the proposed sites is briefly discussed.

The term "credible range" is used in place of "expected range" since the expected range is usually thought of in terms of deviations from the norm, whereas credible range is not likely to be confused with other conventional terminology. Here we use credible range as an imprecisely defined quantity that is meant to include deviations from the norm, uncertainties, future design changes, and other local perturbations that could reasonably occur in a sealed repository, barring unusual tectonic activity or other catastrophic events. Admittedly, defining a credible range is somewhat subjective and must be based on assumptions that are subject to change. However, in the authors' opinion, the assigned credible ranges represent a reasonable attempt to limit the possible range of the environmental parameters.

The intent is certainly not to set absolute limits, and it is recognized that arguments can be made for changing these values based on other assumptions or subjective viewpoints. We believe, however, that they can serve as guidelines in three important ways: (1) to limit the range of environmental parameters for use in preliminary waste package performance analyses, (2) to limit the range of these parameters in research and development work, and (3) to provide a "feel" for the physical realities involved in the performance of a waste package in a repository.

D.1 NUCLEAR WASTE REPOSITORY IN BASALT

D.1.1 Groundwater Characteristics

The present discussion includes the chemical properties (dissolved solids, pH, and redox properties) of the ambient repository groundwater along with some expected changes in these properties brought about by temperature, pressure, radiation, and interaction of the groundwater with the backfill material. The results of chemical analyses of samples of groundwater from beneath the Hanford site have identified three distinct groundwater compositions: calcium-magnesium-bicarbonate, sodium-carbonate-bicarbonate, and sodium-chloride. The latter type is characteristic of the Grande Ronde formation, which is the proposed location of a repository system. There are considerable variations (both vertically and horizontally) in the chemistry of water samples collected from the horizons within the Grande Ronde basalt (DOE 1982). The composition ranges for major dissolved solids observed in ambient geologic samples from five boreholes in the Grande Ronde basalt are given in the first column of Table D.1.

Table D.1. Groundwater characteristics — chemical constituents of Grande Ronde groundwater and credible concentration ranges for these constituents in a repository

Component	Ambient Grande Ronde groundwater concentrations[a] (mg/L)	Maximum credible repository concentration[b] range (mg/L)	Synthetic Grande Ronde groundwater (mg/L)	
			GR-3[c]	GR-4[d]
Na^+	161–350	10–500	358	334
K^+	3–16	0–100	3.43	13.8
Ca^{2+}	0.8–10	0–100	2.78	2.20
Mg^{2+}	0–0.17	0–50	0.032	
Fe^{2+}	0–0.06	0–0.5		
B^{3+}	0–1.5	0–5		
Al^{3+}	0–0.18	0–20		
Total Si	30–170	10–2000	35.5	45.0
HCO_3^-	45–118	40–200	100	
CO_3^{2-}	4–55	0–70		
Inorganic C			10.8	18.1
Cl^-	98–297	0–500	312	405
SO_4^{2-}	4–197	0–200	173	4.0
F^-	11–42	0–50	33.4	19.9
S^{2-}	Not detected	0–100		

[a] Compiled from data in DOE 1982.

[b] Range of ambient groundwater and changes which result from groundwater basalt and/or packing interaction at elevated temperature and pressure.

[c] Data taken from JONES 1982.

[d] Data taken from RHO 1984.

Groundwater reaching the waste canister in the closed repository will have to transverse backfill material that surrounds the waste canister and occupies the volume between the canister and the host basalt rock. No publications are available on experiments conducted to determine the chemical interaction between the proposed backfill mixture, 75% basalt—25% bentonite, and the Grande Ronde groundwater. However, hydrothermal experiments have been made with the basalt/groundwater and bentonite/groundwater systems at 30 MPa and temperatures up to 300°C (WOOD 1982); experiments have also been performed with spent fuel/groundwater, doped glass/groundwater, and doped glass/groundwater/basalt systems (SCHRAMKE 1984).

Analysis of solution data from the 300°C, 30-MPa experiments by Wood (1982) as a function of time indicated that the primary reaction was the slow alteration of the basalt glass phase to illite and/or smectite clays and quartz. It was concluded that a waste package backfill containing significant amounts of crushed basalt will provide a geochemical environment which establishes and maintains a highly reducing groundwater. The following trends in specific aqueous species were noted:

1. The sodium content remained constant during the basalt experiments and showed only a slight decrease with bentonite.

2. Potassium demonstrated a net increase in both systems to final values between 3 and 25 mg/L.

3. Aluminum increased in both systems to a maximum value of 17 mg/L in the basalt/groundwater system and then systematically decreased with time to a final value of approximately 3 mg/L.

4. The potentially corrosive aqueous species — fluoride, chloride, and sulfate — were all observed to demonstrate insignificant increases for the basalt/groundwater system. With bentonite, fluoride showed a steady decrease from 32 to 3.4 mg/L, chloride increased from 290 to 375 mg/L, and sulfate decreased from 163 to 115 mg/L.

5. A sulfide odor was detected in both basalt/groundwater and bentonite/groundwater experiments. A single analysis for the basalt system indicated a minimum of 5.0 mg/L of sulfide was present. Experiments suggested that sulfide could result from reduction of dissolved sulfate, but the sulfide could have been from the pyrite in the crushed rock.

6. After an initial rapid increase, the silica concentration leveled off and then decreased slowly in both systems.

7. The pH value was initially ~9.7 but decreased rapidly to a constant value of 6.0 for basalt/groundwater and 5.7 for the bentonite/water system.

The packing material — a mixture of crushed basalt and sodium bentonite — is expected to have ample opportunity to interact with the stagnant or slow-flowing groundwater; however, it is credible that some ambient Grande Ronde formation groundwater might reach the waste overpack unchanged through imperfection (channels, voids left during backfilling, etc.) in the backfill. The maximum credible range for the dissolved solids is shown in the middle column of Table D.1. These values include the projected maximum credible ranges in the concentrations due to interaction of the ambient groundwater with the basalt rock and/or the backfill at the expected temperatures and pressure. The credible range should also include the concentration in any groundwater reaching the waste canister throughout the different major time periods of the repository — during operation, thermal periods, and after the repository returns to geological control.

In the experiments by Schramke and co-workers (SCHRAMKE 1984), a synthetic groundwater that simulates the expected compositions at the proposed site was used. This particular water, designated GR-3, has the composition (JONES 1982) shown in the fourth column of Table D.1. In some earlier experimental work, different compositions of synthetic groundwater had been used and designated GR-1 and GR-2. A more recent composition of the synthetic groundwater (GR-4) has been devised for use in future experiments; the analysis is shown in the fifth column of Table D.1. The composition of the GR-4 is based on the analysis of a Cohassett flow bottom sample from borehole RRL-2 (RHO 1984). The experiments by Schramke et al. (SCHRAMKE 1984) were conducted at 200°C and 3 MPa in Dickson-type autoclaves. They included two runs with simulated high-level waste glass and synthetic groundwater (one with basalt included and one without basalt) and one run with synthetic groundwater and spent fuel (SF) from the Turkey Point Reactor. In the experiments with glass, the concentrations of dissolved silica, ^{99}Tc, ^{75}Se, and ^{137}Cs were less when basalt was included than when it was absent. The actinide concentration in the water was much less than the calculated values (based on the amount of waste form dissolved), which implies that these radionuclides were either absorbed or precipitated in secondary phases. Similar behavior was observed in the experiment with the spent fuel/groundwater system (SCHRAMKE 1984). These experiments were terminated before steady-state concentrations of most of the radionuclides were achieved. It was recommended that future experiments with these systems be of longer duration (SCHRAMKE 1984).

Ambient-groundwater pH for the Grande Ronde formation is in the range of 9.4 to 9.9 (SMITH 1980). These elevated pH values are attributed to hydrolysis of silicate minerals and/or silicate glass (KRAUSKOPF 1967; SMITH 1980). In addition, the ambient groundwater is highly buffered by dissolved silica and carbonate species (SMITH 1980). The temperature dependence of pH has been characterized by experimental measurements. The expected pH values shown in Table D.2 are those calculated based on the expected silicic acid dissociation, and the credible range is the maximum range of experimental values observed during hydrothermal experiments.

Unlike pH, direct measurements of the redox properties of natural waters often do not represent reversible redox potentials even when reproducible values are obtained. Another difficulty in this type of measurement is the possibility that the measured "equilibrium" potential may be a composite of two or more redox processes. These problems in operationally measuring redox potentials have been discussed in detail by Stumm and Morgan (STUMM 1981). Since meaningful field measurements of the redox properties of the groundwater have not been possible, the values of the oxygen

Table D.2. Groundwater characteristics — summary of pH and redox properties of Grande Ronde groundwater in a repository

Repository period	Temp. (°C)	pH		Redox properties	
		Expected[a]	Credible range[b]	Expected range[c] (V)	Credible range[d] (V)
Thermal periods	100	8.7	8.4 to 8.9	−0.49 to −0.53	+0.20 to −0.45
	150	7.9		−0.48 to −0.53	
	200	7.2	7.0 to 7.8	−0.47 to −0.52	
	250	6.7		−0.47 to −0.52	
	300	6.2	5.6 to 6.4	−0.45 to −0.51	
(Max. credible temp.)	400	5.6			
Ambient geological control	55 to 65	9.6	8.5 to 10.0	−0.40 to −0.50	+0.3 to −0.5

[a] Calculated by assuming that pH is controlled by the dissociation of silicic acid (SMITH 1980).

[b] Maximum range of measured pH.

[c] Thermodynamic calculations on assumed mineral assemblages.

[d] Arbitrary maximum range based on uncertainties in calculated values.

fugacity and the redox potential have been inferred either from thermodynamic calculations or from experimental data on appropriate mineral assemblages. The inferred values are given in Table D.2 for the redox potential for a basalt host rock environment (SMITH 1980). Considerable uncertainty exists in these data; however, there is little doubt that the redox conditions are controlled by the geology of the site and that the groundwater system of the Grande Ronde is generally reducing in nature.

Data from the high-temperature basalt rock/groundwater and bentonite/groundwater interaction experiments indicated that the steady-state solution is very reducing (WOOD 1982). The credible ranges shown in the last column of Table D.2 are intended to arbitrarily reflect the uncertainties in the calculated Eh and their applicability to the specific repository site.

The previous discussion has dealt exclusively with estimations of equilibrium redox potential resulting from groundwater contact with the water canister. More oxidizing conditions are certain to result during repository excavation operations and subsequent backfilling. These conditions will be temporary, lasting only until the air trapped between particles of backfill is exhausted by reaction with the rock. The exact rate of oxygen consumption by basalt (or backfill) will be directly dependent on the surface area of the solid and the temperature prevailing at the air-particle interface. However, the lack of detailed knowledge of the specific reaction kinetics prevents making confident estimates of how long the redox potential will be outside of geologic control. A crude guess of the time required to bring repository redox potential from air (0.54 V) to geologic equilibrium (-0.20 to -0.50 V) is hundreds of years. Results from the high-temperature basalt rock/water and bentonite/water interaction experiments indicate that anoxic conditions are expected to prevail by the time groundwater reaches the container at least 100 years after repository decommissioning (LANE 1983).

Radiolytic processes involving the dissolved solids in the basalt groundwater are not expected to be important because of their low concentrations (GLASS 1981). The primary radiolytic process in aqueous solutions is the decomposition of water to form short-lived radicals, e^-_{aq}, H, OH^-, and HO^-, and the long-lived molecular products, H_2, O_2, and H_2O_2 (BURNS 1981). If the radiolytic H_2 is continuously removed from the system in some manner, the oxidation potential of the solution will be increased. Conversely, in a closed system such as a sealed repository located in basalt, where the hydrogen fugacity is expected to remain high (SMITH 1980), the radiolysis reaction products will recombine and the oxidation potential will remain essentially unchanged.

Groundwater within several of the basaltic layers contain methane at concentrations up to 700 mg/L. Gray (1984) found that gamma irradiation of synthetic basaltic groundwater (see Table D.1 for composition) containing 700 mg/L of dissolved methane produced a milky liquid that was a suspension of fine particles of organic polymers similar to polyethylene. Later, hydrothermal waste package interaction tests were made by McGrail (1985) using a mixture of crushed glass, basalt, and steel in methane-containing synthetic groundwater. He found that the colloidal polymers did not form under conditions similar to those used by Gray (1984). It was concluded that the presence of the waste package constituents apparently inhibited the formation of organic polymers, but the mechanism that prevented their formation was not determined.

D.1.2 Temperatures

Table D.3 shows the design limit temperatures (ANDERSON 1982), the calculated maximum or expected maximum temperatures for the 1982 reference conceptual design (WEC 1982) of waste packages for a repository in basalt, and our estimate of the credible maximum temperatures.

Complete thermal analyses for the current conceptual designs for the waste are not available. However, the temperatures listed in Table D.3 for the commercial high-level waste (CHLW) packages should be representative of the current reference design since the thermal loading and dimensions of the overpacked canister are unchanged. The changes due to horizontal emplacement and installation of preformed packaging (see Sect. A.1.2) should not be large for the post-closure period. Actually, the temperatures should be slightly lower for the current reference design (assuming that large gaps

Table D.3. Waste package temperatures (°C) and limits[a] for the 1982 conceptual designs of waste packages in a basalt repository

Waste package component	CHLW			SF		
	Design limit[b]	Maximum		Design limit[b]	Maximum	
		Expected[c]	Credible		Expected[c]	Credible
Waste form	500	328	550	380	303	400
Canister	430	250[d]	375	430	210[d]	375
Overpack	430	247[e]	370	430	208[e]	370
Packing	300	196[e]	370	300	208[e]	370
Basalt	450	218	250	450	190	220

[a] The geothermal temperature or the unperturbed repository horizon temperature was assumed to be 59°C.

[b] Data taken from ANDERSON 1982.

[c] Data taken from WEC 1982.

[d] The canister temperature would equal the maximum overpack temperature if estimated with no gap or contact resistance.

[e] The apparent anomaly is the result of not adding backfill until 50 years after emplacement when the overpack temperature has decreased from the maximum. The effect is not as great with SF because of a smaller heat source and less rapid decay as compared with CHLW.

will not be left between the packing containers and the wall of the drill hole) because of the lower temperature in the Cohassett (~51°C) as compared with the earlier design temperature for the Umtanum (~59°C) and the presumably higher thermal conductivity of the preformed and compacted bentonite-basalt packing as compared with the pneumatically emplaced packing of the earlier design.

The temperatures for the current design for the waste package for spent fuel would be higher if the same package spacing is used because of the increase of the thermal loading from 1.65 to 2.20 kW. However, the higher temperatures could be compensated for by an increase in the package spacing if necessary. As an example of the possible temperature increase, the expected temperature for the earlier design was 210°C (Table D.3) for the maximum canister temperature, or 151°C rise; for a 2.20-kW loading, the temperature rise (all conditions equal, with constant thermal properties) would be 201°C and the canister temperature would be 252°C for the Cohasset, with a geothermal temperature of 51°C. The table shows that the expected maximum temperatures are well within the design limits. In a recent paper (YUNG 1986) on the current conceptual design for a waste package containing spent fuel with a heat load of 2.069 W for the short horizontal borehole concept (see Sect. A.1.1), maximum temperatures of 255 and 163°C were reported for the spent fuel and outer surface of the packing (or inner surface of the borehole), respectively. The maximum temperature of the overpack was 200°C at the midplane and 170°C at the base. This demonstrates a need for three-dimensional models for accurate temperature predictions.

As previously discussed, the design limits could be increased or unexpected perturbations (if they occur) could cause the expected temperatures to be higher. In general, it seems unlikely that design temperatures could be exceeded (except in the case of a few odd canisters) because of the validation program that will be in force during the retrieval period.

The glass waste form limits seem adequately high, but we will arbitrarily assume that 550 and 400°C may occur in the glass and SF, respectively. The 430°C limit for the canister was determined on the basis of intergranular corrosion and appears to be high on the basis of the sensitization of 304L stainless steel that will occur at this temperature. An earlier suggested limit (RUSSELL 1979) of 375°C seems more reasonable with regard to sensitization of the stainless steel. A limit of 300°C on the packing is probably conservative since temperatures up to 400°C are generally not expected to cause serious degradation of the bentonite packing. Based on recent experimental results, Allen and co-workers (ALLEN 1983) concluded that sodium-bentonite would retain its swell properties on addition of water after initial dehydration at temperatures up to 370°C. To allow for a possible design limit increase and other perturbations, 370°C was selected as the credible maximum. The findings of Couture (1985), however, that exposure to water vapor can cause large increases in the permeability of bentonite for temperatures up to 260°C (see Sect. B.3.1.1) create some doubt that 300°C is conservative. On the other hand, it is not clear that the conditions of the experiments relate to the actual repository conditions. The limit of 450°C for the basalt, which is based on decrepitation, is obviously much higher than what could occur because the inner package temperatures will be much lower than 450°C. The values of 250 and

220°C, respectively, for maximum credible basalt temperatures for the CHLW and SF cases represent about a 20% increase in the heat source term or the equivalent in reduction of the effective thermal conductivity of the basalt. The overpack temperature limit is not a controlling one, and the 370°C specified for the credible maximum is based on contact with the packing at its credible maximum, which allows for a small temperature drop due to a gap or contact resistance at the interface with the canister. It is obvious that the credible maximum temperatures for all components could not be attained, but they represent possibilities that could occur if design changes were made.

With the passage of time, the temperature distribution across the waste will "flatten out," and all temperatures will approach the geothermal temperature of the disposal horizon (i.e., 50 to 60°C, depending on the particular horizon selected). After ~1000 years, the waste temperatures will drop to around 100 and 150°C for the CHLW and SF, respectively (WEC 1982).

D.1.3 Radiation Fields

Published information on the gamma-ray, alpha-particle, and beta-particle radiation fields to be expected in the emplaced reference waste package is very meager, which is surprising since such calculations are straightforward with documented and well-used computer codes such as ANISN (ENGEL 1967) and ORIGEN2 (CROFF 1980).

The maximum dose rates at the surface of the overpack from 10-year-old waste were reported by WEC (1982) to be 7.1×10^3 and 1.4×10^3 rem/h for gamma emissions from CHLW and SF, respectively, and correspondingly 7.1 and 2.4 rem/h from neutrons for the earlier 1982 conceptual design. For practical purposes, the gamma-ray values may be assumed to be numerically equal to the dose rate in rads/h for gamma energy absorbed in the packing; the neutron absorbed dose rate is negligible in comparison. The dose rates for the current reference design for the CHLW will not be significantly different. An estimate of the maximum dose rates at emplacement (assuming 10-year-old waste) for the current reference design for SF can be obtained by multiplying the dose rates for the earlier design by a ratio of the thermal loadings for the two designs (1.20/1.65). This results in dose rates of 1.9×10^3 and 3.2 rem/h for gamma rays and neutrons, respectively.

In view of the sparse information, we arbitrarily assumed that the credible dose rate range at the overpack-packing interface at emplacement is about ±50% of the reported values (i.e., 4×10^3 to 10^4 rads/h for the CHLW and 10^2 to 3×10^3 rads/h for SF). The dry packing can be expected to reduce these dose rates to the basalt exponentially by about a factor of 2. Based on previous calculations and the Westinghouse values for the emplaced dose rates for a salt repository using similar packages, we estimate that the cumulative absorbed dose after 10,000 years will be 1.1×10^9 and 4.3×10^8 rads for CHLW and SF, respectively, with the cumulative dose being ~10% less at 100 years after emplacement.

Published information on the alpha- and beta-particle radiation is not available for the reference designs. The primary effect of alpha particles is radiation damage to the glass by atomic displacements and its impact on glass properties. Beta-particle and gamma-ray radiation damage will be small in comparison to the alpha-particle damage (SOO 1983). Radiation damage produced by different radioactive sources saturates in a manner that implies that dose rates are not important; however, direct information is lacking. In silicate glasses doped with alpha emitters, saturation effects are commonly observed due to the internal alpha decay. A typical saturation dose is $\sim 10^{18}$ alpha disintegrations per gram, which is roughly 10% of the expected million-year dose that is anticipated to the glass waste form (DAYAL 1982).

D.1.4 Pressure

The pressure exerted on an emplaced canister will rise as water floods the repository until the regional hydrostatic pressure is reached. Basalt will not exhibit significant creep at repository temperatures; consequently, attainment of the regional lithostatic pressure should not occur for many thousands of years, and it will probably develop rather suddenly as the result of noncatastrophic shifts in the rock structure.

The repository horizon is assumed to be at a depth of ~975 m with a regional hydrostatic pressure of 11 MPa; an early estimate assumes a vertical rock stress of 23 MPa and a horizontal rock stress in the range of 30 to 58 MPa (DOE 1986a). In 1985, DOE's peer review group recommended that the repository design be based on a vertical stress of 24.4 MPa and a maximum and minimum horizontal stress range of 50 to 75 MPa and 30 to 40 MPa, respectively (KIM 1986).

Other pressure changes on the waste canister could occur from the swelling of the bentonite and the vapor pressure of the water. As previously discussed, however, swelling pressures can be controlled through design, and it is unlikely that the swelling pressure will greatly exceed the design value. The vapor pressure in the emplacement hole is not expected to rise significantly above the ambient pressure at the repository horizon because the reference repository design has no provision for hermetically sealing the emplacement holes. Even if provisions were made for hermetically sealing the emplacement hole, it is unlikely that the hole would remain sealed because of the joints and fractures present in basalt.

In view of the previous discussion, a reasonable scenario for the development of pressure on the overpacked canister seems to be as follows: The pressure remains near atmospheric for 50 years during the retrieval phase and rises linearly to hydrostatic (11 MPa) in 100 years after sealing. The pressure exerted remains hydrostatic until a few thousand years after emplacement, at which time the maximum lithostatic pressure obtains.

D.2 WASTE REPOSITORY IN TUFF

D.2.1 Groundwater Characteristics

Analyses of groundwater sampled directly from the proposed repository horizon in the tuff of Yucca Mountain are not available. However, a number of samples have been taken from various producing wells in the study area and analyzed for the various chemical constituents. These results have been summarized by Guzowski, Daniels, and their co-workers (GUZOWSKI 1983; DANIELS 1982). The groundwater in the vicinity of the proposed repository is classified as the sodium-potassium-carbonate-facies type, but the composition of the dissolved chemical constituents will vary depending on a number of factors, including well usage. The water from Well J-13, which is nearest to the potential site, was chosen as the reference groundwater to be used in experiments and preliminary performance assessments. The chemical analysis of the reference water that was established was based on multiple analyses of J-13 well water by the U.S. Geological Survey (DANIELS 1982); the results are shown in Table D.4. References to the analyses of the individual samples and the method of averaging for determining the reference composition are not available. As an example of the variation in composition with time, the range in the analyses for various chemical constituents are shown for a number of samples taken over a period of several years from Well J-13 (GUZOWSKI 1983).

In order to define the changes in water chemistry that would result from temperature changes caused by the emplacement of high-level nuclear waste in a repository in tuff, Oversby (1984a, 1984b) performed a series of

Table D.4. Chemical analyses of water from Well J-13: ranges for some samples compared with reference composition

Component	Range in composition[a] of some samples (mg/L)	Reference composition[b] (mg/L)
Li^+	0.004–0.16	0.05
Na^+	44–52	51.0
K^+	4.7–6.8	4.9
Mg^{2+}	1.7–2.5	2.1
Ca	12.2–14	14.0
Sr^{2+}	0.04–0.09	0.05
Ba^{2+}	0.002–0.020	0.003
Fe^{2+}	0.001–0.04	0.04
Al^{3+}	0.003–0.008	0.03
SiO_2	57–64	61.0
F^-	1.7–2.4	2.2
Cl^-	5.4–7.7	7.5
$HCO_3^- + CO_3^{2-}$	124–170	120.0
SO_4^{2-}	18–21	22.0
NO_3^-	9.0–9.9	5.6
PO_4^{3-}		0.12
pH	6.9–7.3	7.1

[a] Data taken from GUZOWSKI 1983.

[b] Data taken from DANIELS 1982.

hydrothermal experiments using crushed tuff from the Topopah Spring member and natural groundwater from Well J-13. These experiments were conducted for four water/rock ratios at 90, 120, and 150°C. The major conclusion drawn from the work was that changes in water chemistry due to heating of the tuff-groundwater system can be expected to be minor. There was no significant source of anions from the rock. The major changes in cations were an increase in silica to approximately the level of cristobalite solubility, supersaturation of aluminum followed by slow precipitation, and fairly rapid precipitation of calcium and magnesium carbonates. The pH of the solutions increased. This was attributed to CO_2 uptake by the Teflon reaction vessels since it did not occur in other experiments by Knauss et al. (KNAUSS 1984) using Dickson autoclaves with a gold cell.

None of the experiments reached steady-state concentrations in the solution. Since it is expected that percolation of the water through the tuff matrix would be slow enough to achieve steady-state concentrations at the ambient temperatures, Oversby (1984b) estimated the concentrations of the various species at steady state based on the results of the hydrothermal experiments. These estimated concentrations are shown in Table D.5.

The water that eventually contacts the waste canister will probably be much more concentrated than the water from Well J-13 (Tables D.4 and D.5), for several reasons. It is expected that groundwater approaching the package will boil and deposit salts along the flow paths until the temperature drops below 95°C (boiling point at the proposed elevation). After that time, dissolution of the earlier precipitated salt will begin. No estimates of the concentration increases or their effects are available (SOO 1985a).

Table D.5. Estimated steady-state concentrations (mg/L) for the Topopah tuff—J-13 water system at various temperatures[a]

	Temperature (°C)		
Component	90	120	150
Al^{3+}	0.4	1.2	1
B^{3+}	0.1	0.1	0.1
Fe^{2+}	0	0	0
Si^{2+}	49	81	122
Ca^{2+}	8	3.5	3
Mg^{2+}	0.2	0.1	0.1
K^{+}	9	9	9
Na^{+}	40	45	40
F^{-}	2	2	2
Cl^{-}	7	7	7
NO_3^{-}	9	9	9
SO_4^{2-}	18	18	18

[a] Data taken from OVERSBY 1985b.

At present, the Eh of groundwater in the potential horizon for a repository is unknown. The Eh measurements that have been made for wells in the vicinity of Yucca Mountain are subject to doubt because of possible sample contamination and water being poorly poised [i.e., the water lacks a sufficiently high concentration of oxidizable or reducible species to permit accurate measurements (GUZOWSKI 1983)]. As pumped, water from Well J-13 contains ~5.5 ppm oxygen, which makes it slightly oxidizing. It has been suggested that deep groundwaters in the tuffs of the NTS area might always contain dissolved oxygen (WINOGRAD 1982).

A possibility that is rather unique to a tuff repository located above the water table is the formation of gaseous reaction products during an initial period when only steam and air are present. The formation of nitric oxides in the relatively strong radiation fields around a nonoverpacked canister (or with thin overpacking) which would form nitric acid in the probable humid atmosphere or on contact with groundwater might be of particular importance. The presence of nitric acid should not have a significantly deleterious effect on the stainless canister, but the effect could be serious for low-carbon steel (or copper) if used for an overpack or hole liner.

D.2.2 Temperatures

Thermal analyses of a large number of design concepts for waste packages were made by Stein and co-workers (STEIN 1984) in which they considered variations in package dimensions, thermal loading, package spacing, internal fin arrangements, and overpack arrangements for SF and CHLW in vertical and horizontal emplacement holes. In general, peak temperatures were reached in several years after emplacement around the waste package and within 50 years in the tuff within several meters of the package. These calculations were also reported and reviewed by O'Neal et al. (O'NEAL 1984), with the addition of two PWR cases that involved calculations using an unnamed 2D/3D code (2D infinite cylinder inside borehole, 3D outside borehole). All the other calculations were made with the finite-element code TACO2D (BURNS 1982).

Currently, design temperature limits are only set for the waste form — 400°C for the glass waste and 350°C for SF (STEIN 1984; O'NEAL 1984). These limits are lower than those usually taken for design limits (see Sect. D.1.2 for limits set for BWIP) and seem to be conservative.

Of the various cases considered, only one (a CHLW case) by Stein et al. (STEIN 1984) and the two additional spent fuel (PWR) cases with different areal thermal loadings as calculated by O'Neal et al. (O'NEAL 1984) match the current reference package designs discussed in Sect. A.2.1. Table D.6 shows the maximum temperatures attained for the CHLW case (which included a borehole liner) and the SF case with higher areal thermal loading of the two as calculated by O'Neal and co-workers (O'NEAL 1984). The current conceptual design does not call for a borehole liner for the CHLW package unless needed. The effect of the borehole liner is to act as a radiation shield (a thermal resistance) and to slightly increase the temperatures for the components internal to it.

Table D.6. Maximum temperatures (°C) expected for the reference conceptual design of a waste package in a tuff repository[a]

Waste package	CHLW[b]	Spent fuel (PWR)[c]
Waste form	330	322
Canister	275	245
Overpack	259	
Borehole liner	228	
Tuff	205	233

[a] The geothermal temperature or the unperturbed repository horizon temperature was assumed to be 29°C.

[b] Areal thermal load, 44.9 kW/acre; package thermal load, 2.21 kW. Data taken from STEIN 1984.

[c] Areal thermal load, 48.4 kW/acre; package thermal load, 3.3 kW. Data taken from O'NEAL (1984).

These calculated temperatures are within the waste form limits, and other component temperatures seem sufficiently low to be acceptable. The results, however, are very tentative since the repository layout has not been set and the waste package designs are still subject to change. In addition, no consideration was given to the effect on temperatures of backfilling the rooms several years after emplacement of the waste packages. The design status is too premature for estimating maximum credible temperatures, but a guess of 25 to 30% greater than the values listed in Table D.6 appears reasonable.

D.2.3 Radiation Fields

The comments on alpha- and beta-particle radiation included in Sect. D.1.3 also apply for a tuff repository.

Results of calculations of the gamma dose rates at the surface of the waste canister or overpack are not available for the current reference designs; however, it is possible to estimate dose rates based on previous designs. In an earlier reference design prepared by Westinghouse (WEC 1983a), for the CHLW, the only significant difference was the thickness of the overpack — 2 cm for the Westinghouse design vs 1 cm for the current design described in Sect. A.2.1. The estimated maximum dose rate (10-year-old waste) at the surface of the overpack for the Westinghouse design was 4.2×10^4 rem/h. Since an additional 1 cm of stainless steel will reduce the dose rate by a factor of nearly 2, the estimated dose rate for the current reference design for CHLW becomes 8.4×10^4 rem/h and, assuming a 100% possible increase because of the greater uncertainty than in the basalt case, the maximum credible dose rate is 1.7×10^5 rem/h. Cumulative doses over time periods were not reported; however, since the same radioactive materials and thermal loadings are assumed for the basalt and tuff repositories, the cumulative doses will be proportional to the emplacement dose rate. Therefore, the cumulative absorbed dose for 10,000 years at the surface of the overpack for CHLW is estimated as $1.1 \times 10^9 (8.4 \times 10^4/7.1 \times 10^3) = 1.3 \times 10^{10}$ rads. In the case of spent fuel (PWR), the reference Westinghouse design (WEC 1984) has an overpack* (or canister, in this case) diameter of 53.5 cm and a wall thickness of 2.5 cm, which result in a dose rate at emplacement of 350 rem/h at the overpack surface. The current reference design has an overpack* (or canister, in this case) diameter of 70 cm and a wall thickness of only 1 cm. The difference of 1.5 cm in wall thickness will result in a dose-rate increase of almost 3, but the larger diameter will cause a decrease in the maximum dose rate in inverse ratio to the diameters. Therefore, the estimated maximum dose rate at the overpack surface for the current design for PWR fuel elements is $350 \times 3 \times (53.5/70) = 800$ rem/h. As in the case of CHLW, where an additional correction was made for the thermal loading, the cumulative absorbed dose is $4.3 \times 10^8 (800/1900)(3.3/2.2) = 2.7 \times 10^8$ rads. The neutron doses will be insignificant in comparison.

D.2.4 Pressure

Location of the repository horizon above the water table will preclude development of hydrostatic pressure. One could postulate a seal developing around a repository because of evaporation and precipitation of salts in locations where the temperature exceeds 95°C; however, such a possibility seems very remote in view of the porosity and fracture pattern of the tuff. Vitrified tuff is a reasonably competent rock, and the development of full lithostatic pressure can be expected to take hundreds to thousands of years, barring unusual tectonic activity. At a repository depth of 350 to 400 m, the lithostatic pressure would be 8 to 9 MPa.

D.3 WASTE REPOSITORY IN THE SALT OF THE PALO DURO BASIN

D.3.1 Brine Characteristics

Groundwater does not circulate in salt deposits, nor can the groundwaters lying above the repository horizon be expected to reach the disposal horizon in the time frames of interest. Any groundwater that reaches the disposal horizon through failure of shaft seals or other events (barring catastrophism) will dissolve salt and probably become saturated prior to interacting with a waste package. Water will be available, however, in the form of brine inclusions that can migrate up the thermal gradient toward a waste package and possibly other water from dehydration of minerals in the salt and bedding interlayers that can collect in the space between the waste and the host rock (see Sect. A.3.4 for a discussion of brine migration). In addition to the brine inclusions and the possible brine that would be formed by dissolution of salt by inflowing groundwater, Hubbard and co-workers (HUBBARD 1984) claim that there are two other possibilities: (1) a "desiccation brine" that could result if magnesium-rich fluid inclusion brines should become largely desiccated by the heat from the waste package; and (2) a brine

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*Actually, the overpack is considered the canister or container since the fuel elements are stacked in a honeycomb box structure within the canister (see Sect. A.2.1 for a description).

that is present in areas of impure, commonly clay-rich salt. The chemical composition of the latter brine cannot be estimated from the currently available data; however, Hubbard and co-workers (HUBBARD 1984) point out that there are good arguments that it lies between those of fluid inclusions and dissolution brines. Possible compositions of the other three types of brines are shown in Table D.7.

Hubbard et al. also point out that all brines are saturated with $CaSO_4$ and that the inverted solubility of this compound (its solubility decreases with increasing temperature) could result in the deposition of a $CaSO_4$ scale on the waste package that might inhibit corrosion. The dissolution brine composition shown in Table D.7 is based on a series of brines made by dissolving, in distilled water, a blend of ten rock-salt cores taken from the lower San Andreas Unit 4 (MOLECKE 1983).

Table D.7. Palo Duro brine compositions

Component	Dissolution brine[a] (g/L)	Recommended fluid inclusion brine[b] (g/L)	Desiccation brine[b] (g/L)
Ca^{2+}	1.56	1.336	
Mg^{2+}	0.134	50.0	138.72
Na^+	123.46	25.29	
K^+	0.039	15.69	
SO_4^{2-}	3.197	3.20	
Cl^-	191.38	200.0	406.26

[a] Data taken from MOLECKE 1983.
[b] Data taken from HUBBARD 1984.

Hubbard et al. (1984), after examining the effects of evaporating seawater and the concomitant chemical reactions, developed a recommended composition for brine inclusions (see Table D.7) in the region of the proposed site.

The "desiccation brine" shown in the fourth column of Table D.7 represents an extreme that could result from extensive desiccation of a high-magnesium brine. The concentrations given in the table are for a saturated solution of $MgCl_2$. Such a brine could form in the open spaces around an emplaced canister. Loss of water from the inflowing brine inclusions, which are $MgCl_2$-rich and saturated in NaCl, results in the precipitation of NaCl and an increase in the concentration of $MgCl_2$. With the continued loss of water, the solutions will become saturated in $MgCl_2$ and a solid hydrate will precipitate. This hydrate could be either $MgCl_2 \cdot 6H_2O$ and/or $MgCl_2 \cdot 4H_2O$, depending on the temperature of the system. These hydrates exert vapor pressures of water and can be converted to lower hydrates at suitably low partial-pressures of water vapor. The hydrates can also undergo hydrolysis to form solid magnesium hydroxy chloride, MgOHCl, and HCl gas. This hydrolysis reaction is likely to be slow for the temperatures that will exist around the waste package. The $MgCl_2$-rich solutions can also undergo hydrolysis to some extent, which will be accomplished by the formation of soluble hydroxy complexes and hydrogen ions (JENKS 1972).

On examining the phenomenon of brine migration into the emplacement hole, Hubbard and co-workers (HUBBARD 1984) concluded that the actual composition of brines that first contact the waste package will be somewhere between that of a dissolution brine, a fluid inclusion, and a desiccated brine which is nearly a saturated $MgCl_2$ solution. They also concluded that the actual composition will depend on the temperature field, the rate and temperature at which fluid inclusion exits the halite crystals, and the physical details of the evaporation-condensation cells around the waste package.

In addition to the hydrothermal reactions possible in a salt repository, significant amounts of gamma energy will be absorbed by the crushed salt, the nearby rock, and the brine in the spaces of the emplacement hole. Radiolysis of the brines and the rock salt will produce changes in the brine chemistry and the composition of the surrounding rock salt. An early review of radiolysis and hydrolysis in salt-mine brines was made by Jenks (1972). Later, comprehensive reviews that considered more additional information on radiation effects were made by Panno and Soo (PANNO 1984; SOO 1985b).

Gas evolution occurs on heating salt and from radiolysis of the brine. Gases released on heating salt include HCl, SO_2, CO_2, H_2S, and H_2O. Some of these will dissolve in the brine to give acidic solutions. In a repository, the pH could increase and possibly become alkaline as a result of the interaction of migrating brine inclusions with the radiation-induced colloidal sodium (see below for a discussion) that is formed in the salt crystals and subsequently reacts with water to produce NaOH (SOO 1985a).

Other gases could also undergo various reactions and/or be liberated from rock salt and brines as the result of radiolysis. Gamma irradiation of rock salt causes damage in the form of "F-centers" (Cl^- ion vacancies) that aggregate and form colloidal sodium in the crystals with the rate of formation depending on the dose rate and total dose in a complex manner. The generation of colloidal sodium is accompanied by the production of Cl_2 or Cl_2^- aggregates in the crystal. When a salt crystal containing these species comes into contact with water or brine, OH^-, OCl^-, HOCl, etc., are generated in solution and H_2 and Cl_2 might escape as gases. If Cl_2 cannot escape and reacts with the water, an increase in pH due to the formation of OH^- will be neutralized by HOCl and HCl resulting from Cl_2 reactions, and the final pH will be close to neutral. With escaping Cl_2, however, the brine pH can increase to >10 (SOO 1985b). Radiolysis of brines will also produce H_2 and O_2 from the radiolytic decomposition of H_2O and possibly ClO_3^- and BrO_3^-. Most of the ClO_3^- and BrO_3^- will decompose to halides and O_2 at the relatively high temperatures around a waste canister, and $Mg(BrO_3)_2$ (if present) may produce some Br_2 (JENKS 1972).

The formation of colloidal sodium and chlorine in rock salt has been the subject of investigation by Levy and co-workers (1983) for several years. In his recent work, Levy (1983) subjected 14 natural rock salt samples from several locations to dose rates of 1.2×10^8 rads/h and to total doses of 2 to 4×10^8 rads at 150°C, which is the temperature at which colloid formation rates are near maximum. At higher temperatures, annealing effects on the damage apparently become more pronounced with respect to the production rate of colloids. Important conclusions were that (1) colloid formation was related to the salt impurity level; (2) on a unit-dose basis, lower dose rates produce more colloids than higher dose rates; and (3) colloid formation rates vary by a factor of >10 between samples from different localities.

Levy (1983) extrapolated his results based on a simple power-law fit to the data (i.e., fraction of salt converted to sodium as a function of time or cumulative dose for the constant dose rate of the experimental plots to a straight line on log-log paper) to the cumulative doses in salt that may be expected in a repository. He estimated that a dose of 10^{10} rads will convert between 0.1 and 10%, while 2×10^{10} rads will convert between 1 and 50% of the salt to colloidal sodium. The results of his experiments for the U.S. rock salts, when extrapolated to a dose of 10^{10} rads, predicted between 0.8 and 1.5% conversion to sodium, and between 2.9 and 4.7% conversion for a dose of 2×10^{10} rads.

Levy (1983) claims that these values may be low because the dose rate to the salt in the experiments was $\sim 10^4$ times larger than that expected in a repository, which would produce fewer colloids on a unit-dose basis than the lower repository rate. However, other factors could make the predictions conservative, including: (1) extrapolation to doses 100 times larger than the experiment is a dubious procedure; (2) the 150°C temperature produces peak damage rates; (3) back reactions with chlorine will occur as the sodium content increases beyond the small quantities that existed in the experiment; and (4) extrapolation of a 2- to 3-h experiment to several hundred years is a very dubious procedure because the annealing kinetics (reversing the damage) may be more important in view of the much longer time involved and the much lower formation rates that will exist in a repository.

Prediction of the brine characteristics that interact with a waste package is very difficult because colloidal sodium will affect the brine characteristics and the complexity of the possible hydrolytic and radiolytic reactions. In addition, these reactions can be sensitive to some of the parameters involved. Even the concentration of only minor components is of apparent importance in the radiolysis of brines and rock salt. For example, small changes in the Br^- concentration (a scavenger for the OH radical) of the brine can cause large changes in the equilibrium hydrogen and oxygen pressures. In the radiolysis of rock salt, the presence of impurities can stabilize radiation-induced defects and yield a wide range of sodium colloid formation rates for different salts (PEDERSON 1984). Therefore, it seems that characterization of a brine in contact with rock salt in a radiation field requires site-specific testing under repository conditions.

D.3.2 Temperature

Parametric thermal studies were made by Westinghouse (WEC 1983b), along with thermal analyses for the original reference designs of the waste package and the alternate ones using an unnamed finite-element computer code (WEC 1983b). The unit-cell concept was used to reduce the calculations to 2D types. The assumptions made were reasonable and in accordance with the usual ones made in calculations of these types; however, future thermal analyses for more specific repository designs should include some 3D calculations and the use of documented computer codes. Later calculations using the undocumented TEMPV5 code were made by McNulty and co-workers (McNULTY 1984) and reported in the DEA for Deaf Smith County (DOE 1984). The few results from these calculations that were available are shown in Table D.8.

Table D.8. Maximum temperatures (°C) expected for the original reference conceptual designs of waste packages in a salt repository

Waste package	Design limits	Spent fuel (PWR)		CHLW	
		WEC 1984[a]	DOE 1986[b]	WEC 1984[c]	DOE 1986[d]
Waste[e]	500	258		476	
Overpack	375	146	130	250	228
Salt	250	146		250	

[a] Assumed geothermal temperature, 34°C; local areal thermal loading, 18.75 W/m^2.

[b] Assumed geothermal temperature, 31.8°C; local areal thermal loading, 15 W/m^2.

[c] Assumed geothermal temperature, 34°C; local areal thermal loading, 30 W/m^2.

[d] Assumed geothermal temperature, 31.8°C; local areal thermal loading, 30 W/m^2.

[e] Thermal load for SF(PWR), 5.5 kW; for CHLW, 9.5 kW. Design limit is 375°C for spent fuel.

The two sets of calculations do not agree very well. The overpack temperatures for the spent fuel are based on different areal thermal loads (although the geometry is apparently the same), but correcting the temperature rises by the ratio of the areal thermal loads and accounting for the 2°C difference in the geothermal temperature still leave about an 11°C difference in the two calculations. In the case of the CHLW, the areal thermal loads are the same, and correcting for the geothermal temperature leaves a 20°C difference. It is possible that the differences result from the treatment of the gap between the overpack and the salt. In the calculations made by Westinghouse (WEC 1983b), it was assumed that the gap between the overpack and the rock salt would close in a matter of months; hence, no temperature difference was taken between the maximum overpack and salt temperatures. The details are not available for the other calculations.

Maximum temperatures expected for the current conceptual designs for waste packages described in Sect. A.3 have been reported by Westinghouse Electric Corporation (WEC 1986). These are listed in Table D.9. In the calculations, it was assumed that the gap between the rock salt and the overpack did not exist. A simple one-dimensional calculation was used to obtain the CHLW results. For the spent fuel calculations, the WECAN code (STILLMAN 1984), a biaxial plane of axisymmetric finite-element model with two-dimensional conduction capability, was used. The effects of waste from canister dividers were included. An effective thermal conductivity for the rod bundle, which included radiation heat transfer effects, was determined as a function of temperature.

Table D.9. Maximum temperatures (°C) expected for the current reference designs of waste packages on a salt repository[a]

Waste package	Design limit		Spent fuel (PWR)[b]	CHLW[c]
	SF	CHLW		
Waste form	375	500	348	480
Overpack	375	375	175	230
Salt	250	250	175	230
Waste at 10^3 years	d	100	99	60

[a] Assumed geothermal temperature, 34°C.
[b] Package heat loading, 6.6 kW; local areal heat loading, 14.9 W/m^2.
[c] Package heat loading, 9.5 kW; local areal heat loading, 15 W/m^2.
[d] To be determined.

Adding 25 to 50°C because of uncertainties would put the salt in the generally unacceptable temperature range because of possible salt decrepitation, as well as causing the design limit for CHLW to be exceeded. Better-documented calculations on a more-detailed conceptual design would be required to evaluate the temperature ranges and limitations of the design.

D.3.3 Radiation Fields

The comments on alpha- and beta-particle radiation included in Sect. D.1.3 also apply to a salt repository.

The radiation dose-rate calculations were made with some unspecified type of hand calculations (WEC 1986). The dose rates at the overpack surface at emplacement (10-year-old waste) for the current reference conceptual designs (see Sect. A.3.1) were 1600 rem/h from gamma radiation and 7.7 rem/h from neutrons for CHLW. The corresponding values for spent fuel (PWR) were 180 and 2.7 rem/h.

The cumulative dose for the isolation period has not been published for the reference designs; however, it can be estimated using the same technique described in Sect. D.2.3, which is based on the Westinghouse design (WEC 1982) for the basalt repository. Therefore, the cumulative absorbed dose in 10,000 years at the surface of the overpack for CHLW is:

$$1.1 \times 10^9 \ (1600/7100) = 2.5 \times 10^8 \text{ rads;}$$

the corresponding absorbed dose for SF is

$$4.3 \times 10^8 \ (180/1900) = 4.1 \times 10^7 \text{ rads.}$$

D.3.4 Pressures

By nature of the salt deposits, a waste package will not be accessible to groundwaters in the times of interest unless an unusual and catastrophic event or intrusion occurs. Consequently, hydrostatic pressure cannot develop. The salt will creep and gradually exert more and more pressure on a waste package, which could exceed lithostatic for a few years because of thermal expansion of the salt. As the thermal stresses are relaxed, the pressure will attain the lithostatic value after a few additional years. The maximum thermal stress obtained and the length of time required for these processes will depend on the repository design, the region geology, and the constitutive properties of the salt. A reasonable expectation would be tens of years. Full lithostatic pressure for the proposed horizon for the repository will be ~17 MPa.

D.4 REFERENCES FOR APPENDIX D

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APPENDIX E

COMMENTS ON THE DOE REPOSITORY ENVIRONMENTAL PARAMETER MODELS/METHODOLOGIES AND APPROACHES

The environmental parameter models/methodologies for use in the performance assessment of a waste package are still in the developmental stage for waste disposal in basalt, tuff, and salt. The assessment for BWIP is the most detailed, and tuff the least detailed, on the basis of the available information. A BWIP performance assessment plan (SONNICHSEN 1984) has been published that defines the approach to the analyses and the use of performance assessment activities for all phases of the project. The plan is being revised to reflect changes in design and the management and technical needs of the project. Only a brief and overall system performance plan has been published for the Yucca Mountain site (TYLER 1984). A preliminary performance assessment plan that includes anticipated methods for the Salt Repository Project (SRP) has been published; a more-detailed plan is being prepared for publication in the near future (ONWI 1984).

E.1 BWIP PERFORMANCE ASSESSMENT PLANS

Details of the plan for the waste package performance assessment have not yet been formulated; the only published program plan states (SONNICHSEN 1984) that the BWIP modeling methodology is based on the use of a suite of deterministic numerical models describing the coupled processes of rock stress/strain, heat transfer, groundwater flow, and nuclide transport. It is recognized that the prediction of repository system performance over large space and time scales strongly indicates the need for using mathematical models based on the underlying physical and chemical principles to extrapolate measured system behavior.

Sensitivity and uncertainty analyses will be used to define probabilities to assist in the establishment of levels of confidence or bounds, and statistical sampling techniques (such as the Monte Carlo technique) will be applied where appropriate (see SUTCLIFFE 1981 and STEPHENS 1986 for some applications). The techniques being developed by BWIP seem to be in general agreement with the methodologies that are recommended (STEPHENS 1986) for acceptance by the NRC for assessing long-term performance of high-level radioactive waste packages.

E.1.1 Overview of the BWIP Very-Near-Field Performance Assessment Methodology and Computer Codes

The relationship between the package environmental models/computer codes and the models/computer codes discussed in the published program plan (SONNICHSEN 1984) for use in the very near-field region (waste package and nearby rock) of the system performance model is shown in Fig. E.1. The characteristics of these computer codes and other possible ones that are mentioned and may be used to replace or augment the indicated system are summarized in Table E.1.

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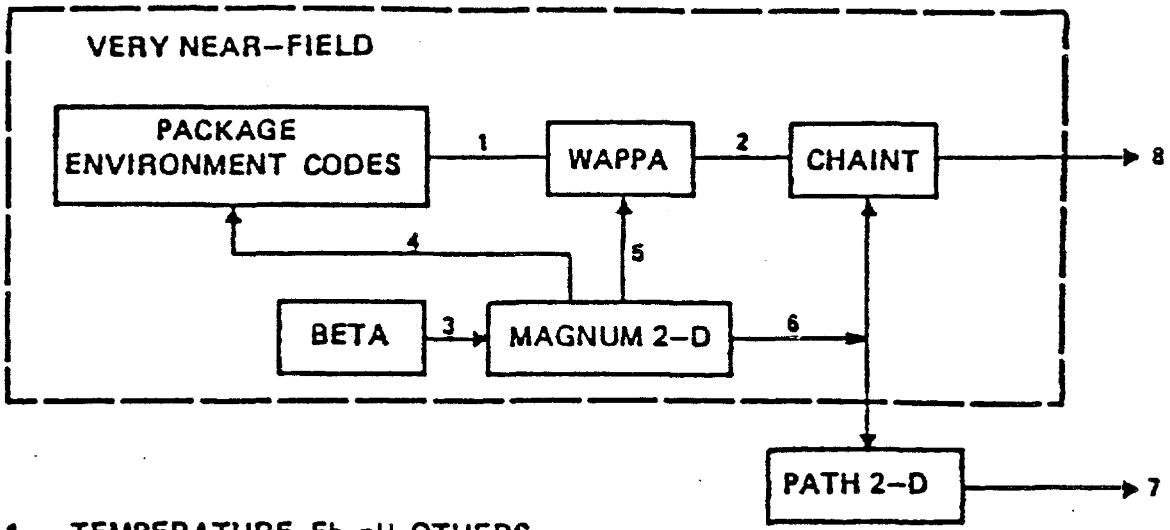

1. TEMPERATURE, Eh, pH, OTHERS
2. CONTAINMENT TIME AND SOURCE TERM
3. FRACTURE PERMEABILITY
4. RESATURATION TIME
5. WATER FLOW RATES
6. GROUNDWATER FLOW FIELDS
7. FLOWPATHS AND WATER TRAVEL TIMES

Fig. E.1. Relationship of waste package (repository) environmental parameter codes to the waste package performance assessment system model. Source: J. C. Sonnichsen, Basalt Waste Isolation Project Performance Assessment Plan, SD-BWI-PAP-001, 1984.

Table E.1. Summary of characteristics of codes for performance assessment involving the very near field[a]

Computer code	Approach		Stress/ strain		Ground-water flow		Heat		Radionuclide transport			Computational method			
	CO	DC	LI	NL	IS	NI	AD	DS	S	MC	DE	FE	FD	AL	DI
ADINA	X		X	X								X			3
ADINAT	X				X	X		X				X			3
BETA	X		X		X	X		X				X			2
DAMSWEL	X		X	X	X	X		X				X			2
ANSYS	X	X	X	X	X	X		X				X			3
HEATING5	X					X		X					X		3
MAGNUM	X	X			X	X	X	X				X			2
CHAINT	X	X							X	X	X	X			2
WAPPA	X					X			X				X		1

[a] Taken from Sonnichsen (1984). Legend for characteristics of codes:

AD = advection
AL = analytical
CO = continuum
DC = discontinuum
DE = decay chains
DI = dimensionality
DS = diffusion
FD = finite difference
FE = finite element
IS = isothermal
LI = linear properties
MC = multicomponent
NI = nonisothermal
NL = nonlinear properties
S = single component

A critical review of the published computer programs that could be used to analyze the potential performance of a high-level radioactive waste repository has been completed (COFFMAN 1984). The computer programs identified in the report address the performance of a waste package, including the areas of thermal analysis (ADINAT, ANSYS, HEATING6, SINDA, STEALTH), structural analysis (ADINA, ANSYS, NONSAP, SAP, STEALTH), and special-purpose codes (BARRIER, WAPPA, BUCKLE, COVE-1, HYDRA-1, IMPAC2, KENO, and BOSOR5). Other codes could be used, and new ones could be developed, for specific applications in a final repository design.

The published program plan is being revised to reflect changes in the design and technical needs of the project. The WAPPA code (see Sect. E.3.4 for a description) included in the methodology of Fig. E.1 is no longer under consideration for use at BWIP. A probabilistic-type code, REPREL (ESLINGER 1985), has essentially replaced it. The methodology for estimating the fractional release rate of each significant radionuclide at the boundary of the waste package subsystem currently involves the following four steps (DOE 1986a):

1. The probability of a typical waste container failing in any specified time interval and the resulting distribution function are determined.

2. The random failure of containers is represented by a nonhomogeneous Poisson process, with the input parameters being estimated from the probability distribution obtained in step 1.

3. Samples of the fractional release rate versus time are obtained for a single container failing at time zero using random sampling of the distribution functions for radionuclide solubility and adsorption coefficients.

4. The fractional releases from the large set of containers in the repository are estimated by combining the releases of step 3 with the random failure sequence of step 2 in the form of a convolution integral from which the means and standard deviations of releases can also be estimated as functions of time.

A flow diagram illustrating the procedure is shown in Fig. E.2. Sagar (1984) provides a detailed description of the methodology and application to a repository (SAGAR 1984).

Currently, the REPREL computer code is used to perform the calculations with a modified CHAINT-MC (BACA 1984a, 1984b; KLINE 1986) code and the undocumented PCM.STAT code used as subroutines.

The latter code performs Monte Carlo simulations on the corrosion model (FISH 1983), which consists of a set of empirical equations that give the corrosion penetration as a function of time under air—steam and under aqueous conditions at various temperatures (see Sect. E.1.7). The values of the coefficients in the corrosion equations for each simulation are obtained by random sampling from specified parameter probability distributions. The probability distribution of failure time of a typical container is constructed from the individual results of a simulation (i.e., time at which the specified corrosion allowance is depleted).

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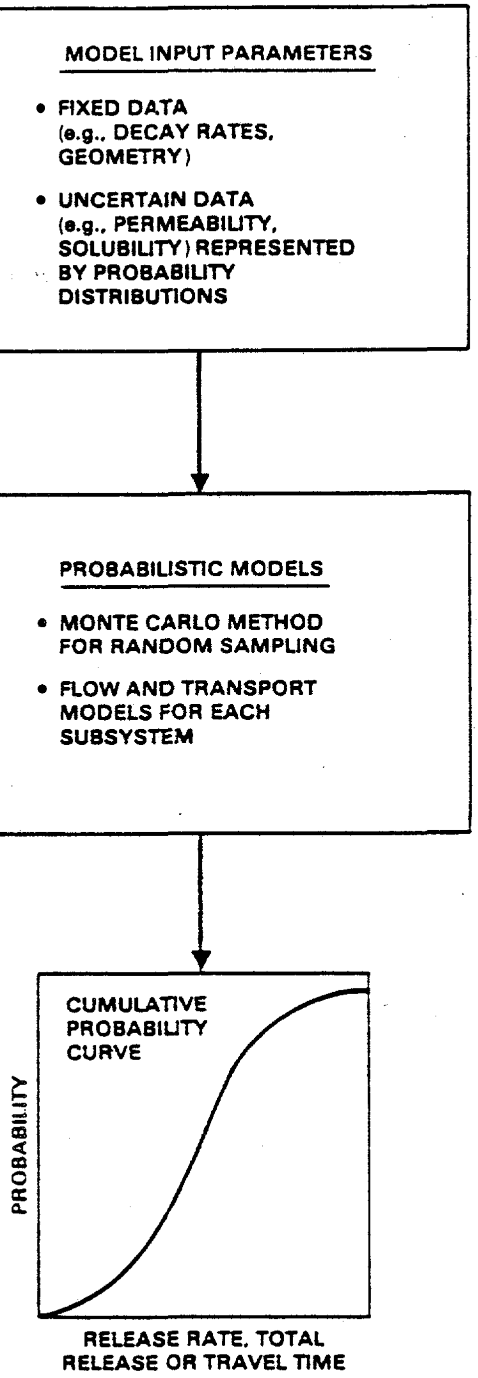

Fig. E.2. Stochastic approach for parameters applied to subsystem performance assessment. Source: U. S. Department of Energy, Environmental Assessment — Reference Repository Location, Hanford Site, Washington, DOE/RW-0070, 1986.

The modified CHAINT-MC code, a nuclide transport code, is used to estimate the fractional release rates from a single container that fails at a specified time. The code can solve the diffusion or convection-dispersion equation of mass transport in one, two, and quasi-three dimensions using the finite-element method (BACA 1984b). This code incorporates various processes that are important to radionuclide migration (e.g., chain decay, sorption, molecular diffusion, and hydrodynamic dispersion), and it can also be used to model conditions of layering or arbitrary heterogeneity and anisotropy. It is designed to perform Monte Carlo simulations by repeatedly solving the mass transport equation using values of the parameters that have been randomly sampled from specified probability distributions.

In general, it appears that the overall BWIP plan related to waste package performance assessment is in line with the specifications presented herein, namely, that the assessment should be of the probabilistic-deterministic type employing statistically sampled data as input to an interrelated set of deterministic models.

The potential uses of the models listed in Table E.1 and other possible models for determining the environmental parameters necessary for the waste package assessment calculations are briefly discussed in the following sections. As previously pointed out, the methodology for assessing the performance of a waste package is still in the developmental stage; therefore, the following discussion is intended only to provide examples and possible guidelines for the development of the associated environmental models.

E.1.2 Groundwater Characteristic Models/Methodologies

The chemical properties of the groundwater at the specific site and rock horizon for the repository in the Grande Ronde basalt are not yet available. Generally, the chemical properties of Grande Ronde basalt possess considerable variation (both vertically and horizontally), and the proposed site properties can only be inferred from the analyses of various samples taken from different locations (see the discussion in Sect. D.1, Appendix D). The ambient groundwater will react initially with the basalt at the elevated temperatures of the repository and then with the bentonite-basalt packing, which will alter the chemical characteristics of the intruding groundwater that contacts the overpack. Generally speaking, such information is determined either by employing geochemical model calculations or by using experimental results.

E.1.2.1 Geochemical Models

Models to describe the interactions and resulting composition, pH, and redox conditions of the groundwater involved in the corrosion of the overpack and canister (and ultimately the waste glass or spent fuel) have not yet been developed. However, numerous computerized models have been developed for equilibrium calculations in aqueous systems (NORDSTROM 1979). Three of these — EQ3/6, WATEQF, and PHREEQE — appear to be the most commonly used for calculations in complex geologic and hydrologic systems. They have been critically reviewed by Thomas (1982). Each of these models was developed for a somewhat different purpose since there are no general-purpose models which can be used for all of the hydrochemical conditions that might exist in a repository located in the Grande Ronde basalt. In addition, all of these models are based on equilibrium thermodynamic calculations and are not capable of handling the kinetics of the aqueous reactions, which can be important.

The EQ3/EQ6 code, which seems to have the greatest potential for application to waste repositories, is being used in the tuff experimental program (KNAUSS 1984). This code is being extended to include some kinetic effects (OVERSBY 1983) for possible use in the performance assessment program, a development that has applicability to the BWIP.

The EQ3/6 code (WOLERY 1979) was developed to compute equilibrium models of aqueous geochemical systems. The package contains two principal programs, EQ3 and EQ6. EQ3 computes the distribution of chemical species such as ions, neutral species, ion pairs, and complexes in the aqueous solution. This calculation produces a model of the fluid, which specifies the concentration and thermodynamic activity of each chemical species that occurs in the chemical system and is included in the data base. The program then calculates the saturation state of the fluid with respect to all relevant mineral phases in the data base.

EQ6 can be used to compute several mass-transfer models. If the initial fluid composition is supersaturated with respect to any mineral phases, the program first "equilibrates" it by calculating a modified fluid composition plus precipitates. EQ6 then computes the progress of compositional evolution and mass transfer in a closed or open system containing this aqueous solution.

Reaction progress calculated in EQ6 may describe changes in temperature and pressure, irreversible reaction of the fluid with reactants such as minerals or gases, or both of these simultaneously. The calculation predicts changes in fluid composition; the identity, appearance, and disappearance of secondary minerals; and the values of reaction progress at which the fluid saturates with reactants and/or products. Such computations permit modeling of the effects of heating and cooling aqueous solutions and of irreversible reactions in rock-water systems.

The BWIP has an ambitious experimental program in place to develop the necessary geochemical data base (MYERS 1983). The study areas include: redox buffering, reaction mechanisms, alteration and characterization of mineral assemblages, effects of variable flow rates, and natural analogues.

Included in the water characteristics, but distinct from the geochemical aspects, is the flow rate of the altered groundwater around the overpack. Figure E.1 shows the MAGNUM2-D (BACA 1981) as a possibility for use in calculating flow into the waste package. MAGNUM is a two-dimensional, finite-element code designed to simulate transient groundwater flow and heat transport in fractured-porous rock systems. The theoretical framework of the code is based on concepts for a porous continuum and for discrete conduits. In particular, a dual-porosity approach is used to represent the continuous rock mass, where flow-through planar conduits are described by Poiseuille's equation.

E.1.2.2 Experimental Approach

It is possible that an entirely experimental approach to determining the chemical conditions inside and around the waste package could be employed by the BWIP. This approach would involve conducting a large number of hydrothermal experiments to determine the credible range of altered groundwater compositions and characteristics at various spatial points in the very near field. The bounding groundwater compositions and characteristics obtained from these experiments would then be used in subsequent component material and integral testing to evaluate waste package performance.

E.1.3 Temperature Models/Methodologies

As previously mentioned, the HEATING5 (TURNER 1977) and ADINAT (BATHE 1977) computer codes are being reviewed for temperature calculations by the BWIP. Presumably, either of these codes may be used to model the thermal environment surrounding the waste to produce interface temperatures for the waste package temperature calculations.

The HEATING5 program is a finite-difference type that is designed to solve steady-state and/or transient heat conduction problems in one-, two-, or three-dimensional Cartesian or cylindrical coordinates or one-dimensional spherical coordinates. The thermal conductivity, density, and specific heat may be both spatially and temperature dependent. The thermal conductivity may be anisotropic. The thermal conductivity, density, and heat capacity may also be time dependent if they are defined in user-supplied subroutines. Materials may undergo phase changes. Heat-generation rates may be dependent on time, temperature, and position; and boundary temperatures may be time dependent. The boundary conditions, which may be surface to boundary or surface to surface, may be fixed temperatures or any combination of prescribed heat flux, forced convection, natural convection, and radiation. The boundary-condition parameters may be time and/or temperature dependent. A modified version of the code is now available as HEATING6 (ELROD 1981).

ADINAT was designed for the automatic, dynamic incremental nonlinear determination of temperatures using finite-element analysis. It is a heat-transfer program that is compatible with the stress analysis program ADINA (BATHE 1975) and can be used to input the temperature distributions to that program. ADINAT can address heat-transfer and field problems in essentially any geometry or combinations of geometries. The orthotropic thermal conductivity may be a function of position or temperature. The heat capacity may be a function of position. The source terms may be a function of position or time. The boundary conditions include prescribed temperature, forced and natural convection, radiation, and prescribed heat flux.

Both the HEATING5 and the ADINAT codes are well-documented and have been widely used. HEATING5, in particular, has been used more extensively in the application to waste repositories. Altenhofen (1981) used HEATING5 in early studies for the BWIP. In later calculations for the current conceptual design of a nuclear waste repository in basalt, WEC (1982) used an unspecified three-dimensional, finite-element model.

E.1.4 Pressure Models/Methodologies

The pressure exerted on the waste package is not expected to exceed hydrostatic pressure in the first few thousand years. The hydrostatic head data from available borehole data in the vicinity of the potential basalt site have been fitted to a linear regression line. The approximate projected hydrostatic pressures for the middle Sentinel Bluffs sequence (Cohassett flow) and the Umtanum flows are 9.3 and 11 MPa, respectively (RHO 1982). The time required to reach equilibrium pressures depends on the assumed resaturation rates for the repository and surrounding rock.

An alternative approach would be to assume rapid attainment of lithostatic pressure, which would be ~23 and ~58 MPa for the vertical and horizontal components, respectively.

E.1.5 Radiation Field Models/Methodologies

The models to be used for calculating gamma-ray, alpha-particle, and beta-particle radiation fields have not been specified, although the widely accepted codes ANISN (ENGEL 1967) for gamma-ray attenuation and ORIGEN2 for alpha-particle and gamma-ray source terms have been used in unspecified ways by the BWIP (WEC 1982).

An integrated model might involve using the ORIGEN2 code with its associated library of nuclear properties of the various nuclides to calculate the alpha-particle and gamma-ray source terms in the waste and DOT (a two-dimensional ANISN; RHOADS 1973) for the gamma-ray attenuation calculation. Since alpha and beta particles are essentially absorbed at the point of emission, the volumetric energy source term and deposition rate will be the same within the waste canister. After breaching of the canister occurs, the alpha activity will cause some radiolysis (beta effects will be relatively small). The gamma-ray emission rate calculated by ORIGEN2 can be expressed in a convenient energy group structure that can be used in gamma-ray attenuation and energy deposition calculations in the DOT code. The available libraries of nuclear cross sections such as ENDF/B (KINSEY 1979) can be fed into a code such as AMPX (GREENE 1978) to produce the necessary group cross sections, such as FSXEC (FORD 1980) for the DOT calculations. It should be noted that only one-dimensional calculations may be needed since they would generate the equivalent of dose rates along a radius at the midplane, which is conservative.

The approximate Reactor Shielding Manual Method (a kernel technique using infinite media buildup factors), as applied in the WAPPA code (see Sect. E.2.2), could be used as an alternative to the DOT code. However, this method has many limitations, particularly in the case of finite and multilayered geometry with volumetrically distributed source terms [see any shielding treatise, such as that described by Schaeffer (1973)].

E.1.6 Sampling Techniques

A straightforward random or Monte Carlo sampling procedure from a probability density function (PDF) is relatively simple and generally most satisfying. Unfortunately, many practical problems require an unmanageable number of trials to produce worthwhile results (i.e., an acceptably low estimated variance of the mean, which is commonly known as "good statistics"). Consequently, sophisticated sampling techniques are frequently used to reduce the computation time.

The nature of the performance assessment of a geologic repository is apparently a problem that will require more sophisticated sampling procedures than the straightforward Monte Carlo technique in order to produce statistics with a reasonable number of trials. The Latin Hypercube Sampling technique (INMAN 1980) has been most widely used in the nuclear waste area, in particular at Sandia National Laboratories (PEPPING 1983). This technique has several advantages over other sampling procedures. Specifically, it guarantees that the range of each input variable is covered. It can also be shown to yield a smaller variance than other methods when the output variable is a monotonic function of each input variable (HARPER 1983b).

E.1.7 Corrosion Models

Apparently the only corrosion model that is considered for use is the one developed by Fish and Anantatmula (FISH 1983) for low-carbon steels. The model consists of three empirical equations as described below.

The form of the equation for estimating the thickness corroded as a function of time in the air-steam environment that exists during the operational period and early part of the postclosure period is

$$d = at^{b} \exp(-c/T) \ , \tag{E.1}$$

where

d = corroded thickness, mm;

t = time, years;

T = surface temperature, K; and

a, b, and c are empirical fit constants or parameters.

The equation for corrosion under the aqueous conditions that develop in later times is

$$d = Aet \ , \tag{E.2}$$

where

A = a fit constant, and

e = an irradiation enhancement factor that depends on the radiation level associated with elevated surface temperatures that are determined by the following equation:

$$e = 1.75 - 0.0025\ t_s \ , \qquad (E.3)$$

where

t_s = the time, in years, after the container emplacement at which saturation occurs.

The value of e is >1 for t <300 years and is set at unity for t >300 years.

For the purpose of preliminary analyses, the ranges of all the fit constants were generally chosen to be approximately ±3 standard deviations, where the standard deviation was taken to be ~10% of the mean. For the probabilistic analyses, the constants or coefficients of Eqs. (E.1) through (E.3) are assumed to be random variables, and sampling for each Monte Carlo simulation was from a distribution function of assumed shape (uniform or normal) with the ±3 standard deviations from the mean.

The general technique is a reasonable one, but the corrosion equation does not seem defensible since it is based only on uniform corrosion results and data from short-term experiments (on the order of weeks). In addition, no explicit causal relationship between the corrosion-causing correlations and the corrosion rate was established. The limitations of the current corrosion model are recognized, and its parameters are expected to change in the future as better understanding of the corrosion process is obtained (SAGAR 1984).

E.1.8 Stochastic Modeling Studies

Most stochastic modeling studies performed by the BWIP used the straightforward Monte Carlo sampling technique of the input variable in conjunction with numerical models in limited performance assessment studies (BACA 1983; CLIFTON 1984; SAGAR 1984). A preliminary stochastic analysis was made to determine the times to failure for the current conceptual design of a waste package for spent fuel (DOE 1986a). Full details of the calculational method are given by Sagar (1984); a brief description is included in Sect. E.1.1. Container failure was considered to occur when the corrosion allowance (7.5 cm) was removed by uniform corrosion only. Actually, more steel (as well as the Zircaloy cladding) would still be present, and the waste canister would not be breached. For these reasons, the results were considered conservative (DOE 1986a). Whether these results are conservative is immaterial at this point since the calculation served to test the corrosion model and to yield a preliminary estimate of the containment period. Obviously, other corrosion and waste package degradation mechanisms will require consideration before the performance analysis can be judged acceptable. The results obtained by Monte Carlo simulation of the corrosion equations are shown in Fig. E.3 in the form of a cumulative probability distribution curve that shows the probability of container failure (or fraction failed) in a specified number of years and a PDF that gives the fractional failure rate in any given year. In generating these curves, it was assumed that the coefficients of the corrosion equations had Gaussian distributions that were truncated on either tail by the specified range.

Stochastic analysis provides a means of explicitly relating uncertainty in model predictions to uncertainties in the input data. Consequently, on an overall basis, a scheme for predicting failure probabilities such as Monte Carlo simulation seems desirable, and it could be practical and acceptable (SASTRE 1986). Other probabilistic schemes might also be acceptable; however, at this stage, a preferred scheme cannot be identified because of the fluid state of repository design (SASTRE 1986).

E.2 PERFORMANCE ASSESSMENT PLANS FOR THE YUCCA MOUNTAIN SITE (TUFF)

Performance assessment plans for a waste package in the tuff at the Yucca Mountain site are virtually nonexistent in available publications. Methods for performing a probabilistic-deterministic analysis for a waste package are under consideration, as evidenced by the probabilistic model proposed by Sutcliffe (1984) and the considerations discussed by Tyler (1984).

E.2.1 Overall Performance Assessment Plans

The overall performance assessment considerations were examined by Tyler (1984) in a very general way. He divided the program into five tasks that include: (1) total system definition, (2) geologic system analysis, (3) engineered system analysis (includes waste package), (4) computer code models, and (5) total system analysis. The objectives of engineered system analysis are to assess the performance of waste package designs and underground facilities in order to determine compliance with regulatory specifications and to develop simplified models that will provide the radionuclide source term for the total system performance assessment. The total system analysis task involves the development of a special-purpose code incorporating simplified models of the geologic and engineered systems, as well as the coupling between the subsystems. This code is to use statistical techniques for qualitatively addressing uncertainties about the performance at the Yucca Mountain site and the engineered repository system. It is to predict overall performance in the form of probability distributions of release rates and concentrations to the accessible environment.

E.2.2 Computer Codes Under Consideration

The existing computer codes being considered for the performance assessment of the waste package include: for temperature distributions — ADINAT,

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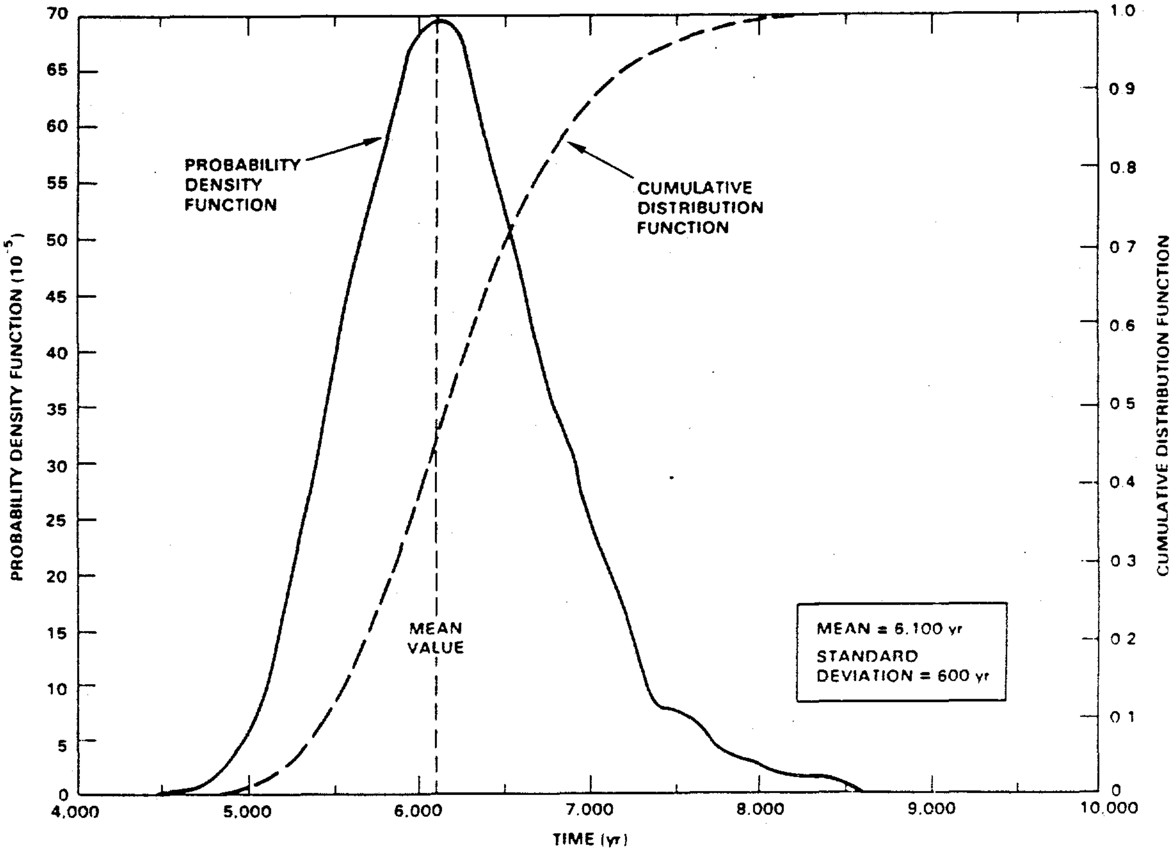

Fig. E.3. Probability curves for failure of a typical container. Source: U.S. Department of Energy, Environmental Assessment — Reference Repository Location, Hanford Site, Washington, DOE/RW-0070, 1986.

SAGUARO COYOTE, and ARRAYF; for waste containment time and radionuclide release rates — WAPPA and ORIGEN; and for groundwater-rock interactions — EQ3/Q6 and PHR81.

E.2.3 Some Results of Preliminary Performance Assessments

Preliminary subsystem and total system analyses have been made (DOE 1986b) for the reference design at the Yucca Mountain site using simple deterministic models implemented by a computer code, which have been described by Sinnock (1984). The results of the calculations indicated that compliance with regulations was attainable. Without discussing the details of the assumptions and the data base, these results can be questioned because only uniform corrosion was considered. It was pointed out, however, that the results were preliminary and the intent was not to show definitively that the subsystems and system will meet the applicable regulations, but only to detect areas that require increased study or emphasis and to develop levels of confidence in the designs as they proceed. It was concluded that the preliminary analyses indicate that the site characterization activities and studies could profitably focus on the following key uncertainties (DOE 1986b):

1. conceptual hydrologic models of flow in the unsaturated zone at Yucca Mountain,

2. the expected physical and chemical environment in the repository after closure, and

3. the conditional waste-package lifetime distributions in the postclosure repository environment.

In addition, the preliminary analyses suggest that refinements in the theory of flow and radionuclide transport in fractured, porous undersaturated rock are needed. In particular, methods for treating the stochastic aspects of flow and transport in the country rock need to be developed in order to estimate the effects of hydrodynamic dispersion and retardation on radionuclide releases to the accessible environment (DOE 1986a, 1986b, 1986c).

E.2.4 Adequacy of the Performance Assessment Plans for the Waste Package

Although the problems involved in assessing the performance of the waste package have been recognized, the conceptual design has not been firmly established and the plans have not evolved sufficiently to judge the adequacy.

A stochastic model for the containment and isolation determinations will be required for demonstrating compliance with regulations if the recommendations of Sastre (1986) and this report become a necessary adjunct to the licensing process. This need is recognized by DOE, as evidenced by item 3 in Sect. E.2.3 and the work of Sutcliffe (1984) and Tyler (1984).

E.3 PERFORMANCE ASSESSMENT PLANS FOR THE DEAF SMITH COUNTY SITE (BEDDED SALT)

A report was issued (ONWI 1984) on the preliminary plan and anticipated methods to be used in the SRP for assessing the postclosure and preclosure performance of a nuclear waste repository in salt. It is intended that the plan be revised on about an annual basis; consequently, an updated version is expected to be issued in the near future. The emphasis in the preliminary report is on the method of conceptually dividing the system into three subsystems: the very-near field, the near field, and the far field. The report (ONWI 1984), which is generic in nature, does not address the proposed site per se. Presumably, this will be covered in future revisions. The very-near field (waste package and immediate environs) is the primary consideration here since this report deals specifically with the waste package environmental parameters.

E.3.1 Overall Performance Assessment Plans

According to the ONWI (1984) report, the postclosure performance assessment for the SRP can be viewed as occurring on two levels. The first level addresses the total system and its uncertainties and lends assurance that the subsystem interactions are well understood. The second level deals with the various subsystems, of which the waste package is one. It is assumed that separate subsystem assessments are necessary (which is a reasonable assumption) because of the complexity of the geologic disposal system and the need for detailed results, primarily for design purposes.

It is pointed out that some type of system analysis is required to demonstrate compliance with all the regulations, but the form that the analysis should take with regard to a single system computer code, or a series of individual process codes, is not clear. Currently, the project is using a series of individual codes to perform preliminary safety assessments, and the merging of these into a single system code is being considered; however, the consensus is that the need for a systems-level code has not yet been established (ONWI 1984).

It was recognized that a great number of models are needed just to perform the assessment on the waste package alone (very-near-field analysis) and that very important factors were the ability of the waste package to withstand lithostatic pressure and to tolerate the effects of the brine that migrates in toward the waste package (see Appendix F for a discussion of brine migration).

Parameters and processes that are to be described by the models for the waste package performance assessment include: temperatures, stresses, local fluid flow (brine migration), geochemistry, canister and overpack corrosion, and local radionuclide transport.

The preliminary report (ONWI 1984) provides a good description of the mechanisms of these processes, coupling effects of the individual models, the general approach to the problem of modeling each process, and possible computer codes that could be used. However, the discussion is general, and a specific approach with interface considerations is still needed; this will apparently be addressed in the soon-to-be issued revision of the report.

The SRP is considering two major approaches to sensitivity and uncertainty analyses: a statistical design approach and a deterministic first-order approach. The project position (a very reasonable one) is that the choice between deterministic or stochastic models is often a question of feasibility. The complexity of stochastic models increases exponentially as more detail is added to the model. A simple deterministic model could reasonably be converted to a stochastic form, but a more complex one might be treated more efficiently with different uncertainty and sensitivity techniques. Most of the models that have been used in preliminary performance assessment by the project were deterministic.

The plan is to utilize the approach that is best suited to a particular model; in some cases, more than one technique will be required. For example, advantage can be taken of the comprehensive screening capabilities of the adjoint sensitivity method to help define and construct a simplified model that can be handled more efficiently by one of the sampling techniques, such as Latin Hypercube Sampling (INMAN 1980).

The statistical design approach may be derived using statistical experimental design theory or Monte Carlo methods. These methods typically treat the model as a black box and randomly generate a set of input values based on probability distributions for use in a computer model run. To reduce computation time, Latin Hypercube Sampling will be used since it seems best suited for the performance assessment model (ONWI 1984). The project plans specifically used such techniques to analyze system models.

The deterministic first-order approach is typified by the adjoint method (INTERA 1983; HARPER 1983b). This approach does not treat the model as a black box but transforms the model equations into a related system of adjoint equations using the mathematics of the calculus of variations and the theory of perturbation. The considerable effort that has been expended in developing the adjoint method should be advantageous in reducing the calculational effort in analyzing sensitivity and uncertainty.

E.3.2 Validation of Models and Data Base

The project plan details the present and future activities involved in enhancing the credibility of performance assessments to be made. These activities are to be accompanied by appropriate technical and peer reviews and are designed to meet NRC requirements. Documentation of all computer codes, which includes both verification and validation of the models, is to follow the requirements given by the NRC in NUREG-0856 (SILLING 1983). Verification is to provide assurance that a computer code correctly performs the operations specified in a numerical model, whereas validation is to provide assurance that the model as embodied in a computer code is a correct representation of the process or system for which it is intended.

The requirements for code verification are straightforward, involving checking with analytical solutions and benchmark problems and comparing the results with those from similar codes. Validation presents a more difficult problem since it involves comparisons with experiments, with the added complication of long time frames.

It is planned that site-specific validation will be used to provide comparisons of model predictions with physical measurements whenever possible. It is recognized that complete validation of many models that simulate very long-term processes will not be possible. Validation in these cases will rely heavily on the peer review process. In addition, scale-model experiments may offer additional evidence of the suitability of some models. Natural analogue comparisons offer another possibility, but they are not specifically included in the project plan.

E.3.3 Computer Codes

The computer codes that have been identified for potential use in assessing the performance of a waste package in the Salt Repository Project (SRP) are listed in Table E.2, with an X indicating the processes that they model. The brine migration process is not listed in the ONWI (1984) report under waste package processes, but under a repository response to waste package heat. In the context of this report, brine migration produces the hydrologic environment for the waste package.

Each code listed in Table E.2 (and others to be used in the overall performance assessment) are discussed in the project plan (ONWI 1984), with emphasis on (1) descriptions of the purpose, capabilities, and limitations of each code; (2) status of the elements of documentation and review essential for code verification and validation; and (3) proposed applications of the model to performance assessment. Areas covered in each code summary include:

1. Code Description. A general description of the purpose and approach of the model is given along with the assumptions, method of selection, and limitations.

2. Development Stage. This refers to distinctions between recently coded models and well-established models in later versions.

3. Documentation. Comparison of the available code documentation is made with the guidelines in NUREG-0856 (SILLING 1983), and the differences are noted.

4. Performance Specification Review. This includes examination of the conceptual design of the model, scoping analysis, adequacy of the mathematical model and derivations, range of model applicability, and input/output requirements.

5. Code Design Review. This includes an examination of the equations of the numerical model, numerical solution technique, computational error and solution stability, system interfaces, and input/output structures.

6. Verification. This includes tests and evaluations conducted to ensure that a computer code correctly performs the operations specified in a mathematical model.

Table E.2. Processes inside and around a waste package, and existing computer codes that can model the processes[a]

	Waste package processes					
Computer code	Heat and radiation generation; isotopic composition of waste	Heat flow within waste package	Thermo-mechanical response and radiation effects	Corrosion rates	Radio-nuclide release rates	Brine migration to waste package[b]
ORIGEN2	X					
HEATING 6/5		X				
SPECTROM-32		X				
WAPPA		X	X	X	X	
ANISNW			X			
SWENT					X	
TEMP[c]						X
BRINEMIG						X
SPECTROM-58						X

[a] From ONWI (1984).

[b] This is considered separately as a repository response to waste heat in ONWI (1984), but it is included here in the waste package processes because the brine migration to the waste package will influence the corrosion rate and radionuclide release rates.

[c] TEMP has been replaced by TEMP V5 for use in repository temperature calculations.

7. Validation. This includes tests and evaluations conducted to ensure that a model, as embodied in a computer code, is a correct representation of the process or system for which it is intended.

8. Proposed Application. The types of performance assessment (waste package, repository, and site) for which application is planned, as well as the particular problems specifically addressed by the code, are identified.

9. Relationship of Model to Other Codes. Links to other codes in terms of output of the subject code are indicated. Constraints on input and output data resulting from such linkage are noted.

10. Application of Code to Other Problems. Documented applications of the code to other (nonsalt or nonrepository) situations are identified, and the relevance of these applications is noted.

11. Suitability of the Model for SRP Application. Limitations of the model that may make it unsuitable for application in performance assessment for various aspects are given.

12. Peer Review. Documented independent peer reviews of model code specifications, design, or performance by recognized experts and peer groups are cited.

13. References. References cited in the code summary are listed.

The reader is referred to the report published in 1984 by ONWI (ONWI 1984) for detailed descriptions and/or evaluations of the above items for each computer code listed in Table E.2 and for all others under consideration. Section E.1 provides a brief description and discusses the potential application to a repository of the computer codes ORIGEN2, HEATING6/5, ANISNW (which is not significantly different from ANISN). A rather detailed discussion of WAPPA is provided in the following section (E.3.4) because of its importance in the current planning. The brine migration codes BRINEMIG and SPECTROM-58 are discussed in Appendix F.

TEMP is a 3-D code that calculates temperature distribution generated by finite-length line sources of heat embedded in a geologic medium using the superposition method to sum contributions from each line source. TEMP V5 (DOE 1986c) is an improved version of TEMP that accounts for the dependence of thermal conductivity on temperature, but it does not account for the temperature dependence of diffusivity. However, the latter thermal property seems to have a small effect for the conditions in a salt repository.

The interfacing of the WAPPA code with BRINEMIG and TEMP V5 is shown schematically in Fig. E.4 according to the current SRP performance assessment plans.

The SWENT code uses a finite-difference technique to simulate steady-state and transient, 3-D transport processes that include fluid flow, heat flow, chemically inert contaminant transport, and radionuclide transport in a

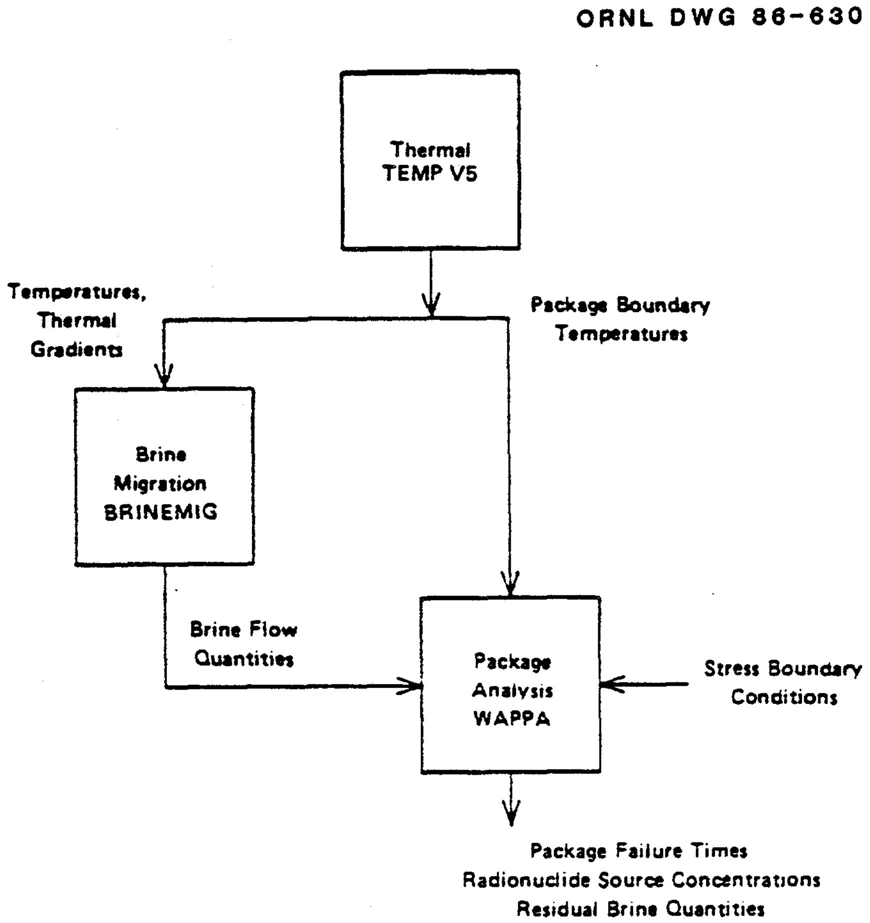

Fig. E.4. Flow diagram for the performance assessment of a waste package. Source: U.S. Department of Energy, Environmental Assessment — Deaf Smith County Site, Texas, DOE/RW-0069, 1986.

heterogeneous geologic medium. The STEALTH program is a package of explicit finite-difference codes that solve time-dependent, nonlinear problems in continuum mechanics and in fluid flow. The STEALTH-2D hydrological model will be used for predicting flow through fractured rock that had low permeability prior to fracturing.

The GRESS code (OBLOW 1983a, 1983b) has been developed for application of the adjoint method in sensitivity studies using first-order deterministic techniques. This code is not specifically designed for performance assessment; it is a tool to facilitate sensitivity studies. GRESS is a FORTRAN compiler language program that enhances conventional FORTRAN programs with analytical differentiation of arithmetic statements so that any standard FORTRAN code can be upgraded to obtain the derivatives required, whether they are used for internal calculations (e.g., for iteration) or for external ones (e.g., sensitivity studies). Although GRESS cannot be used for uncertainty studies to produce a distribution of results based on stochastic input, it can be incorporated into a package for uncertainty calculations that will produce a distribution of results (e.g., waste package failure rates as a function of time).

Most of these computer codes seem to be verified to a degree that may be acceptable; however, validation for them is lacking, with the possible exception of SWENT. A clarification of what constitutes adequate verification and validation is needed; at present, this seems to represent a judgment call which only the NRC can adjudicate. Plans for validation of these codes are scheduled to be published in the next revision of the project plan.

E.3.4 WAPPA Code

A requirement of the overall waste package performance assessment model (see Sect. 2.2.2) is a submodel that predicts the degradation and subsequent failure of the waste package (see Appendix B for a discussion of the various failure modes), the mean time to failure, and the release of radionuclides after a breach of the waste canister. Two waste package performance assessment models have been developed and implemented by the computer codes BARIER (STULA 1981) and WAPPA (IEC 1983). The WAPPA code, which represents a second-generation model, is being considered for use in assessing the performance of a waste package even though it needs some modifications, particularly the various submodels.

Even though WAPPA is primarily a waste package degradation/failure model, it (or similar codes) could implicitly represent part of the repository environmental parameter model/methodology as well as a reasonable technique for assessing the performance of a waste package; therefore, it is the subject of the following discussion. It should be emphasized that the WAPPA code is offered only as an example of a methodology that could be used to satisfy the requirements of waste containment and isolation and as a means of interfacing the environmental parameters and models with the assessment calculations. As the code now exists, some of the submodels (particularly the corrosion submodel) are obviously inadequate.

E.3.4.1 WAPPA Overview

WAPPA is a barrier-integrated and process-sequential degradation code that includes five distinct, coupled degradation process models: radiation, thermal, mechanical, corrosion, and leaching — all of which are driven internally by decay heat and externally by repository stresses and fluids. The penetration of groundwater into the waste package is tracked radially inward. The important outputs are the time-to-failure values of the waste canister and the subsequent radionuclide fluxes to the repository. A system model, which is the executive driver, coordinates the input data files, the use of the process models, and all the I/O requirements (see Fig. E.5 for a block diagram illustrating the structure of the WAPPA code). Each of the WAPPA submodels, as shown in the blocks on the right of Fig. E.5, is briefly discussed below. These submodels receive the greatest criticism when the WAPPA code is examined critically (e.g., STEPHENS 1986). It is possible, of course, to replace these submodels and to retain the main features of the code, which is being done by the salt repository project. The modifications to the WAPPA code are indicated in Fig. E.6.

E.3.4.2 Radiation Process Model

In the calculational procedure for each time step, the radiation process model is executed first. This model has a (1) source-term submodel based on a "look-up" table of time-dependent nuclide inventories and decay characteristics for providing the thermal power input and waste nuclides as a function of time, (2) a gamma-ray attenuation submodel using the simple ray-tracing technique with buildup factors (ROCKWELL 1956), and (3) simple empirical radiation damage and radiolysis submodels for estimating degradation of some waste form physical properties, leach and corrosion rates, and radiation enhancement factors, which will undoubtedly have to be determined from empirical data. Currently, the SRP is using the ANISNW code with a cross-section library of 18 gamma-energy groups; the source terms are calculated by ORIGEN2 (DOE 1986c).

E.3.4.3 Thermal Process Model

The thermal process model is called next since all subsequent process models are dependent on temperature. The current WAPPA model is a quasi-steady-state, one-dimensional, analytical solution for the temperature drops and heat flows across concentric cylindrical annuli based on the power input at the particular time; conductive, convective, and radiative heat-transfer modes are included. The actual temperatures for each time step are determined by combining the temperature drops with the temperature at the midplane of the waste package, which is provided by a "look-up" table or a separate calculation that must interface with the temperature drop calculations. ONWI is considering the TEMP V5 or HEATING5/6 codes for calculating this interface temperature as a function of time.

E.3.4.4 Mechanical Process Model

The next-called, mechanical process model includes three coupled submodels for stress analysis — overpack, canister fracture, and waste-form fracture.

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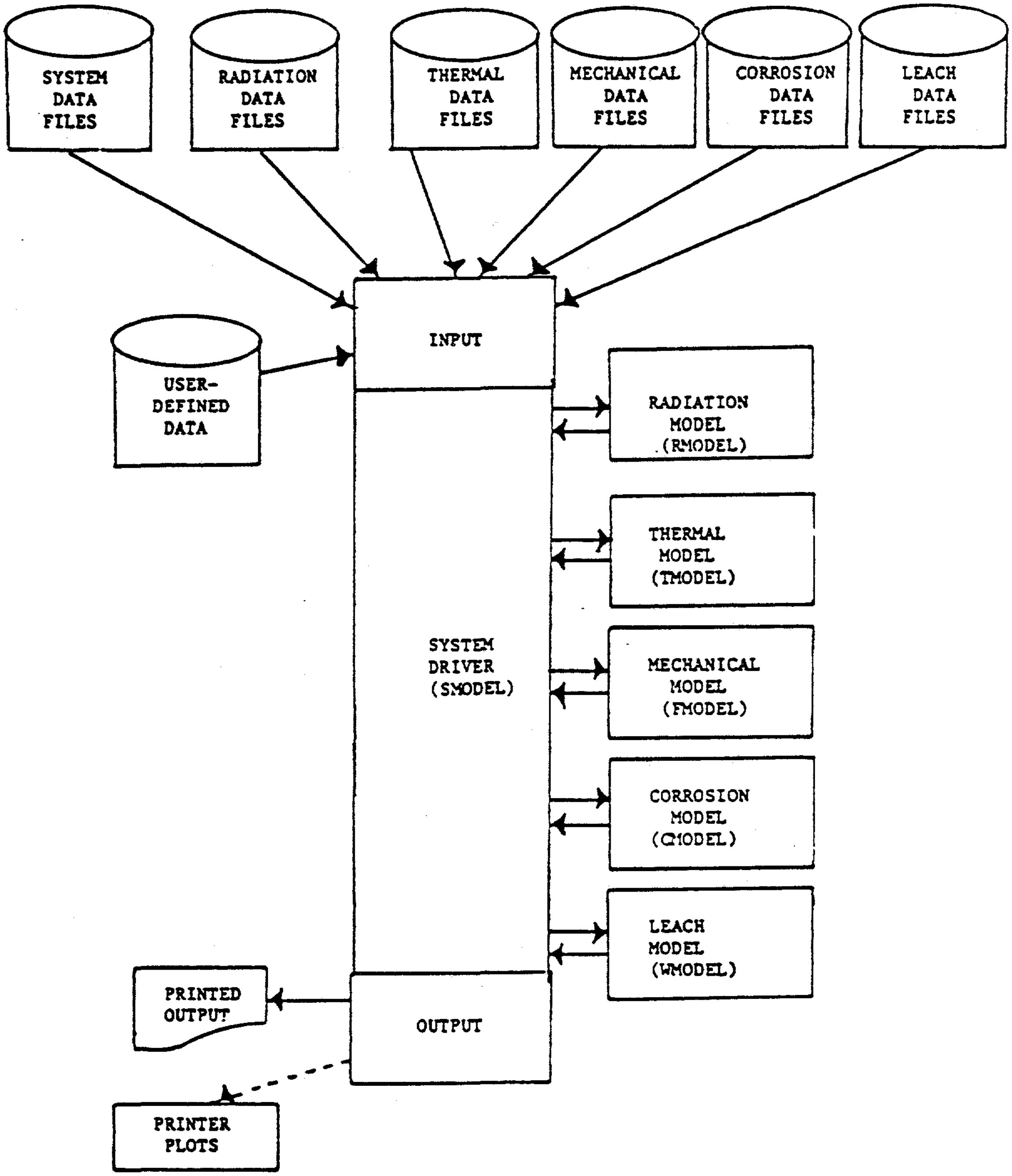

Fig. E.5. Overview block diagram for the WAPPA code, including system structure (SMODEL) and process models. Source: Intera Environmental Consultants, WAPPA: A Waste Package Performance Assessment Code, ONWI-452, 1983.

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1.00 WAPPA A – As received from Intera - in ONWI-452

2.00 WAPPA B – Modified to compute corrosion for EA's and to provide inputs for release calc for EA's

2.01 WAPPA B – Changed to correct minor errors found in verification of 2.00 WAPPA B

3.00 WAPPA C – Modified release model to be used for sensitivity studies for SCP planning

4.00 WAPPA D – Modified stress model

5.00 WAPPA E – Site specific corrosion and release models

Fig. E.6. Stages in waste package code development by ONWI. Source: J. Kircher, G. Raines, V. McCanley, and G. Jansen, "Performance Assessment of Waste Packages," presented at the SRP/NRC Waste Package Meeting, Columbus, Ohio, Jan. 22–24, 1986.

This model allows for plastic yielding of each metallic barrier, but the only source of loss of integrity is a breach, which may be mechanically induced or may result from stress-corrosion cracking. The time at breach of the canister is flagged, and the leach model becomes operative. For the waste form, fracturing causes an increase in surface area, which enhances leaching. Stresses are determined due to displacement incompatibilities, applied vertical and horizontal repository pressures, and thermal gradients.

The current mechanical model is an axisymmetric, plane strain formulation that is consistent with the other process models in the system. For the idealized system, analytical expressions for the stresses are used for both elastic and elastic/plastic responses. ONWI is developing a new stress model, but the details are not available.

E.3.4.5 Corrosion Process Model

The corrosion process model couples the results of the previous three models and accounts for dry oxidation and wet corrosion for both general and localized modes. Five distinct corrosion submodels are considered: generalized corrosion, localized pitting and crevice corrosion, stress corrosion, galvanic corrosion, and dry oxidation (by three possible mechanisms). The models are simple and empirical in nature and, at this point, represent only an example of a methodology that could be used if sufficient data are available. A new model is being developed by ONWI, but the details are not available.

E.3.4.6 Leach Process Model

The leach process model, which is called after a canister breach, calculates the mass removed by leaching from the waste form for both the matrix and specific nuclides, accounts for transport of radionuclides in the waste package (including diffusion, retardation, and advection in the packing), and outputs the radial concentration profile and mass fluxes into the repository rock (source term for repository and pathway transport calculations). The model is based on the solution of a system of radial equations in one dimension for mass transfer and diffusion through breached barriers, coupled with a transient mixing-cell equation for the packing. The mass-transfer equation, or leaching mechanism, is based on physical principles but relies on experimental data characterizing leach enhancement due to radiation and temperature, solution chemistry effects, and nuclide solubilities. As in the corrosion model, all of the environmental parameters and the results of previous model calculations (the corrosion model supplies an areal degradation factor) enter into the leach model calculation.

The intent of ONWI with regard to the use of a leach model is not clear. The emphasis now seems to be on the use of the solubility-limited model (see Sect. E.5) in its place.

E.3.4.7 Comments on WAPPA

The WAPPA approach to the performance assessment of a waste package is a rational attempt to model the gradual loss of integrity of a waste package.

The various processes are modeled individually, and their coupling effects can be accounted for within each time step. Each process is allowed to modify the state of the entire waste package in each time step and, in principle, includes feedback effects. The modular construction with the separate data files for each submodel, or module, allows for flexibility in changing the submodels and data files as more data and better understanding of the various mechanisms become available. Some of the current submodels are obviously inadequate, and more appropriate ones need to, and are being, developed.

The WAPPA model is limited to a one-dimensional calculation, which, for relatively long cylinders, is the equivalent midplane result for a two-dimensional model. The one-dimensional results will obviously be on the high side for the waste package temperatures and radiation fields.

E.3.5 Results of Preliminary Performance Assessments

Functionally, the waste disposal system is considered by the project to be made up of three major subsystems: the waste package; the mined repository, including additional engineered barriers (if any); and the geologic setting, including geohydrologic and geochemical conditions. In a preliminary performance assessment (DOE 1986c) for the proposed repository in Deaf Smith County, it was assumed that no engineering barriers existed in addition to the waste package, since none are planned at this time. The boundary of the engineered subsystem was taken as the outside surface of each overpack of each waste package. Nuclide releases from the engineered barrier subsystem, or the waste package in this case, were assumed to begin at the time of overpack failure and to be limited only by the quantity of each nuclide that would dissolve into the quantity of brine available. Because of the limitations on the data base and analytical methods, it was emphasized that the preliminary performance assessment was not intended to demonstrate satisfaction of the postclosure system guidelines; its objective was simply to supplement the evidence that will be used to establish whether the site is suitable for site characterization (DOE 1986c).

The preliminary assessment (DOE 1986c and JANSEN 1984) of the waste package designs was made with the WAPPA code. Expected values for temperature, stress, brine composition, radiation level, and brine flow rate (see Sect. A.3.1 for results of brine migration calculations and Sect. F.4.5.1 for the calculational method using the BRINEMIG computer code) were used as boundary conditions to calculate the corrosion of the thick-walled overpack of low-carbon steel. Package failure was assumed to occur when the corrosion allowance was exceeded, allowing the brine to contact the waste form; the mechanical submodel in the WAPPA code was not used.

The results of the performance assessment showed that the waste package can be expected to last for more than 10^4 years, assuming uniform corrosion rates and using the calculated brine inflow rates. With the exception of the first 100 years after burial of the high-level waste package, the calculations indicated that all the brine reaching the waste package is used up by chemical reaction with the iron overpack. However, if unlimited brine accumulation (i.e., the waste package is completely immersed in an infinite field of brine) is assumed, failure of the overpacks is calculated to occur at 372 and 232 years for HLW and spent fuel, respectively, for the high-magnesium-content brines that may exist at the site.

If the package should fail prior to 10^4 years, it was pointed out that the solubilities of the radionuclides in the expected total volume of brine that migrates to the waste package would limit the releases to EPA limits, except for ^{137}Cs and ^{14}C. However, decay of these nuclides during the estimated 1500 to 2200 years required for travel to the accessible environments would produce acceptable values for release to the acceptable environment.

Obviously, these results are subject to question simply on the bases of the extrapolation of the corrosion data and the lack of sufficient solubility data. As previously pointed out, DOE does not consider the analyses definitive. The main conclusion by DOE (1986c) is that there is no evidence from the preliminary performance assessments that a repository at the Deaf Smith County site will not comply with the system criteria, or that engineered subsystems would not be able to perform as required.

E.3.6 Adequacy of Performance Assessment Plans for the Waste Package

The overall program plan for the performance assessment of a waste package is not presented in sufficient detail to permit adequate evaluation. However, it will probably provide a good basis for the detailed plans that can be expected in subsequent revisions. The provisions for documenting the computer codes are excellent; however, the actual validation plans are, for the most part, left for future revisions.

At this stage, it appears that a fairly heavy dependence will be placed on the method that was developed for sensitivity studies. However, adoption of this adjoint method to include uncertainties, with subsequent generation of results in the form of occurrence probabilities based on using distribution functions for the input values, will require additional development.

E.4 EVALUATION OF WASTE PACKAGES BY THE WASTE ISOLATION SYSTEMS PANEL OF THE NATIONAL RESEARCH COUNCIL

A study (PIGFORD 1983) was conducted for the U.S. Department of Energy by a Waste Isolation Systems Panel (WISP) of the Board on Radioactive Waste Management, under the National Research Council's Commission on Physical Sciences, Mathematics, and Resources. The panel was charged to review the alternative technologies available for the isolation of radioactive waste in mined geologic repositories, evaluate the need for and possible performance benefits from these technologies as potential elements of the isolation system, and identify appropriate technical criteria for choosing among them to achieve satisfactory overall performance of a geologic repository.

The panel examined and evaluated the plans, information, and the state-of-the-art technology that were available through the year 1982 for respositories in basalt, tuff, salt, and granite. The material for the study was acquired through examination of a large body of technical literature, briefings by representatives of governmental agencies and their industrial and university contractors, in-depth discussions with individual experts in the field, site visits, and calculations by panel members and staff. Deliberations extended over a period of approximately two years. The panel's principal conclusions with regard to the waste packages are quoted below (PIGFORD 1983):

> - The rates of release of radionuclides from candidate waste forms can be affected by the rate of transformation and dissolution of the waste-form matrix, by the rate of diffusion within the waste form, by the solubilities of stable compounds of low-solubility radioelements, and by the rates of diffusion and convection into the groundwater surrounding the exposed waste form. Laboratory data for the release of radionuclides from waste forms have not been shown to be applicable to predicting performance of waste packages in geologic repositories, and suitable laboratory and field experiments for determining releases from waste packages have not been carried out.
> - The theory for solubility-limited dissolution of waste forms used in this study applies to the extent that effects of the formation and transport of colloids and unspecified complexes can be neglected. Based on the experimental and estimated solubilities quoted in this report, the theory predicts very low release rates for low-solubility radionuclides from the waste package. The concentrations of radionuclides later released to surface water are calculated to be low enough that the predicted doses from surface water will be several orders of magnitude below the individual-dose criterion adopted for this study.
> - The effects of uncertainties in waste-package performance, including uncertainties in solubilities and unknown effects of colloids and complex formation, are important. Therefore, additional study needs to be undertaken to determine whether or in which cases the resulting rates of release to groundwater are solubility limited.
> - If the fractional release rate of a candidate waste matrix is less than the solubility-limited release rate of a contained radionuclide, the rate of release of that radionuclide to groundwater will be determined by the rate of dissolution or physical/chemical transformation of the waste matrix, by the rate of diffusion of that radionuclide within the waste solid, or by both processes. It is theoretically predicted that release rates governed by diffusion within the solid could be as low as those governed by solubility.
> - Uncertainties about the physical integrity of borosilicate glass exposed to leaching solutions at high temperatures and uncertainties as to the effect of physical integrity on radionuclide dissolution may require that glass high-level waste be protected from groundwater by a corrosion-resistant overpack when the repository rock is at temperatures greater than about 100°C. There are candidate overpack metals, such as titanium

alloys, that may protect the waste canister from groundwater; but these candidate metals require further testing in an assembled package under the mechanical, chemical, and thermal stresses that are expected in a geologic repository.

- Repositories are likely to be loaded initially with wastes derived from accumulated and aged discharged reactor fuel. With such loadings, maximum rock temperatures in current repository designs are predicted to be low enough, in the neighborhood of 100°C, to warrant confidence in the suitability of borosilicate-glass waste for these initial emplacements.
- There is need for additional information on the performance under repository conditions of 10-year-old high-level waste in borosilidate glass, for which repositories are currently designed. Experiments are needed to verify the predicted dissolution rate of radionuclides, to determine the extent of release by other mechanisms, and to determine the effect of long-term physical integrity on the rate of dissolution of radionuclides.
- There is need for a continuing program to develop new and better alternative waste forms.
- Borosilicate glass may be a suitable waste form for transuranic wastes. Uncertainties are similar to those for high-level waste, except that transuranic waste will be at ambient temperature.
- If the performance of borosilicate glass containing transuranic waste is to equal that of such glass containing high-level waste, it may be necessary to reduce the chemical contaminants normally associated with transuranic waste.
- Carbon-14 recovered when spent fuel is reprocessed, if converted to a concentrated insoluble form, is predicted to dissolve at a rate low enough that appreciable isolation of this radionuclide could be achieved.
- There are alternatives to geologic disposal of separated iodine-129 and carbon-14, such as sea disposal, but they have not been evaluated by the panel.
- Once the fuel cladding, canister, and overpack of a waste package containing unreprocessed spent fuel are breached by water intrusion, the rates of release of many critical radionuclides are likely to be greater than those for waste from fuel reprocessing. The resulting radiation doses from radionuclides transported to the environment are likely to be greater for unreprocessed spent fuel.
- More information is needed concerning the expected performance of a solid backfill surrounding the high-level waste canister and overpack. A dense backfill, such as compressed bentonite clay, may delay and reduce the discharge of radionuclides from the waste to the groundwater but at the expense of increasing waste temperatures. More information is needed on the chemical, physical, and radiation stability of candidate backfill materials under repository conditions and on the effective diffusion properties of backfill materials before backfill can be relied on to retard and reduce significantly the release of long-lived radionuclides to the surrounding groundwater.

- A backfill material may affect the leach rate of the waste form. Further studies are required to evaluate the synergistic and possibly antagonistic interactions among waste form, waste package, and backfill materials.

E.5 COMMENTS ON CONTAINMENT AND THE RELEASE RATES FOR WASTE PACKAGES

The NRC regulations (10 CFR 60) require that a waste package offer substantially complete containment for 300 to 1000 years after permanent closure; and after 1000 years, during the isolation period, the fractional release rate of any individual radionuclide shall not exceed 10^{-5}/year of its 1000-year inventory (as activity) or 10^{-8}/year of the total 1000-year activity, whichever is smaller.

Design of a waste package to meet the containment criterion with a very low probability of failure should not present any large technical problems. It would seem that a waste package of sufficient strength with a conservative corrosion allowance can be designed to preclude most rational concerns in regard to meeting the containment criterion. However, in a practical design, which includes cost considerations, the wall thickness of an overpack needs to be limited and total repository cost considerations favor larger package and areal thermal loadings and the use of the more commonplace materials such as mild carbon steels. All of the factors that tend to reduce the conservativeness of a design will increase concern as to its adequacy; for example, in the current conceptual waste package designs with reasonable wall thicknesses of low-carbon steel, its adequacy can be questioned since the corrosion allowance is based on extrapolating 1000 years of corrosion data that were obtained from experiments lasting on the order of one year or less.

It seems generally accepted that "substantially complete containment" means that only small percentages of the waste packages would fail (i.e., develop random leaks in the first 1000 years). It seems reasonable to assume that the acceptable failure rate is in the 0.1 to 1% range for design and performance assessment purposes until the acceptable failure rate is established by the NRC.

The release rate criterion seems more likely to create licensing problems because of the great uncertainties that could result from using laboratory data and extrapolating short-term, in situ experiments over thousands of years. It has been argued that the numerical criteria for containment and release rate were made without a technically valid basis using invalid assumptions of existing technology, that these criteria are of questionable importance to long-term safety, and that compliance probably could not be verified (PIGFORD 1983). It is certainly true that much greater release rates from the waste packages could occur and still not exceed acceptable releases to the accessible environment for well-sited repositories. Regardless, DOE must demonstrate compliance for all parts of the rule.

The use of sorptive-type packing around a waste container has been investigated for many years as a method of increasing the container lifetime and decreasing the release rates of radionuclides. The current conceptual design of a waste package in basalt includes sorptive packing in the form of a 75% basalt—25% bentonite mixture (Sect. A.1.2). In the tuff repository, packing will be used only if future performance assessments indicate a need (Sect. A.2.2). Currently, the salt repository project assumes the use of essentially nonsorptive crushed salt in the narrow annulus around the waste container (Sect. A.3.2).

After a waste canister is breached, the leached radionuclides will have to traverse the packing and/or gap, which may be liquid filled, into the surrounding country rock. Eventually the release rate from the boundary of the waste form will approach equality with the flux into the country rock; that is, steady state will obtain and sorption will no longer have a significant effect on the release rate from the waste package. The time required to approach steady state depends on the properties of the waste package, each radionuclide, and the country rock. Breakout of the radionuclides will occur prior to steady-state conditions, and the release rate of radionuclides from the waste package could exceed the steady-state value for typical repository conditions during this interim period (CHAMBRÉ 1984b).

The sorptive holdup in any packing will be of small significance in the total releases for long-lived radionuclides, such as plutonium, over the time frame of concern. For example, if breakthrough required 10^4 years, only 25% of the plutonium inventory would have decayed. The sorption in the packing could serve to hold up ^{137}Cs and ^{90}Sr until these nuclides decayed to innocuous levels in the case of premature breaching of the waste containers (e.g., <300 years). Sorptive holdup in the transient phase will also steepen the concentration gradients and increase the diffusion rates from the waste form (PIGFORD 1985b).

Many possible functions have been attributed to packing materials without regard to potential detrimental effects, such as temperature increases in the waste form and waste container (CLAIBORNE 1982). The primary function of packing should be to inhibit hydrologic intrusion to the waste package with concomitant limitation of the transport of radionuclides through the packing to a diffusional process. The use of packing sufficiently thick to promote holdup by sorption processes should be weighed against any detrimental effects that accrue from the increased temperatures in the waste form and container. Similar conclusions were reached by the WISP (PIGFORD 1983).

In addition to the potential problems caused by increased temperatures, Pigford (1980) showed that a thick, sorbing packing could make it difficult to meet the release rate criterion for ^{226}Ra because holdup will cause an increase in the concentrations of the ^{230}Th and ^{226}Ra decay daughters, thereby increasing the later release rates to the country rock.

According to 10 CFR 60.135(a), the synergistic interactions between the various factors that could compromise the waste package function must be considered. It seems that protecting the waste package with thick packing could be interpreted as creating a synergistic effect since the waste container temperatures could be increased substantially, the use of materials such as bentonite in salt repositories (and possibly tuff) could add more water around a waste package than would migrate there (CLAIBORNE 1982), and the release rates of the decay daughters of radium and thorium could be increased in later years.

Over the past three decades, much data have been accumulated for the leaching of borosilicate glass, spent fuel, and other candidate waste forms. The usefulness and applicability of these data and the various empirical correlations have been a subject of continuing controversy. Various proposals have been made (MENDEL 1984) for extrapolating laboratory leach-rate data for long-term radionuclide release calculations at repository conditions by using laboratory data obtained at the same surface-to-volume ratio of a leached specimen as that which is presumed to exist in a repository. However, it has been pointed out that there is no meaning to an equivalent volume of groundwater in contact with the surface area of each waste package (PIGFORD 1983).

According to the WISP (PIGFORD 1983), it is possible that extrapolations based on laboratory leach data represent a conservative upper limit to release rates (particularly when early-time leach data are used); however, the laboratory data cover such a short exposure time that the extrapolations are of uncertain validity, even as upper-limit estimates, since there is little basis for extrapolating to the thousands of years important to the performance assessment of waste repositories. Such extrapolations should have a sound basis in theory that can be verified with reasonable assurance so as to give more credibility to long-term predictions.

The Performance Assessment National Review Group (LIEBERMAN 1985) has also exhibited concern over extrapolating empirical data greatly beyond the time frame of the experiments. In view of the lack of data on spent fuel and the difficulty of extrapolating leaching data for long time periods, they concluded that there was a ". . . need for developing defensible bases for the extrapolation of results from short-term experiments and tests to the long time periods required for postclosure performance assessment. For example, one alternative is to use a verifiable theory for predicting long-term behavior that does not require extrapolation, such as mass transfer analysis . . . ."

T. H. Pigford, P. L. Chambré, and associates at the University of California have published a number of papers (CHAMBRÉ 1982, 1984a, 1984b, 1985; KIM 1986; PIGFORD 1985a, 1985b; ZAVOSHY 1985) related to developing methods that have a sound basis in the theory of mass transfer for predicting transport rates from the waste to the country rock. This work has led to the development of a technique for predicting the rate of transport of dissolved species (CHAMBRÉ 1984a) from the waste package surface by molecular diffusion and convection. The model assumes that the concentration of each elemental species is at the maximum value determined by its solubility at the surface of the waste form. Any leaching or dissolution of the waste form producing greater than solubility-limited concentrations would result in precipitation of that particular species. The release to the surrounding rock (with or without packing) would then depend on molecular diffusion and convective transport (if any), which is a function of the properties of the packing and the country rock. No leaching data are required, and assumptions and uncertainties in the extrapolation of the leaching data are avoided. The theory requires no arbitrary or adjustable parameters to predict the dissolution rate.

The dissolution rates of some species may not be limited by their solubilities or by the waste matrix. For example, the portion of the fission products such as cesium and iodine in spent fuel that exists in any gap between the cladding and the fuel and in other voids and grain boundaries is available for rapid dissolution. Even though dissolution may be rapid and not limited by solubility, the release rates of these soluble species will be limited by their mass-transfer rates through the surrounding porous media. The solubility-limited concept with mass transfer was extended to the time-dependent release rate of species that can dissolve rapidly from the "gap" activity and more slowly from the waste matrix, as limited by molecular diffusion, into the surrounding porous rock (KIM 1986).

In a recent work, Pigford (1985b) reviews the theory and applications to waste packages and possible extensions of the methodology, as well as further examining the problems involved in the application of leaching data. He also points out that some supposedly conservative estimates for transport of radionuclides from a waste package may not be a bounding result for low flow situations. For example, in the environmental assessment for the Yucca Mountain site (DOE 1986b), it was assumed that an upper limit to the release rate could be obtained by multiplying the saturation concentration by the calculated volumetric rate of groundwater flow through rock equal in cross section to the cross-sectional area of the waste canister. Applying the equation developed for diffusion from the waste form into the moist surrounding tuff, Pigford estimated that the limiting low-velocity fractional release rates were one-fourth of those calculated for a pore velocity of 1 m/year, which illustrated the nonconservatism in neglecting diffusional transport at pore velocities of <1 mm/year as in the Yucca Mountain case. It can be argued that the Pigford-Chambré methodology is not applicable to the Yucca Mountain site because of the unsaturated medium with the attendant discontinuities in the diffusional paths. However, it is not clear whether water flow-paths are continuous or discontinuous (KERRISK 1984) or that a continuous flow path will not develop by the end of 1000 years or so in the vicinity of the waste package. It is not too difficult to envisage circumstances which lead to filling any void space between the waste container and the country rock with corrosion products, rock particles, and accumulated water after the temperature drops below the boiling point of 95°C. In addition, it does not seem unreasonable to postulate the existence of an envelope of saturated rock around a waste package because of a partial seal that could develop from precipitated salt as the result of 1000 years or so of evaporation of the available groundwater.

Kerrisk (1984) applied a diffusion-limited dissolution model (i.e., element saturation is assumed at the waste/water interface, and dissolution is limited by diffusion into the water flowing past the waste) and a saturation-limited model (i.e., water flowing through the repository is assumed to be saturated with each element) to determine release rates for nominal values of the parameters for the Yucca Mountain site. The primary purpose of the work was to estimate the effect of element solubilities in limiting dissolution rates. It was recognized that the quantitative results are strong functions of the many assumptions that were made and significant changes could occur when better site-specific data become available. It was concluded, however, that solubility-limited dissolution models may ultimately provide a powerful tool for estimating source terms for performance assessments of a repository.

As a result of the previously mentioned review, Pigford (1985b) reiterated earlier conclusions more firmly, namely, that:

> Predicting rates that radionuclides are released from waste packages cannot rest upon empirical long-term extrapolations of laboratory leach data. Reliable predictions can be based upon simplified assumptions, such as solubility-limited bulk-flow, if the assumed parameters are reliably known or defensibly conservative, but assuming volumetric flow rates through a waste-package cross section is arbitrary and can be nonconservative.
>
> Wherever possible, performance analysis should proceed beyond simple bounding calculations to obtain more realistic — and usually more favorable — estimates of expected performance. Desire for greater realism must be balanced against increasing uncertainties in prediction and loss of reliability. Theoretical predictions of release rate based on mass-transfer analysis are bounding, and the well-established theory is well adapted to verification. The results from the exact analytical solutions can be used to test predictions from numerical techniques and from less mechanistic analogues. . . .
>
> Several efforts to predict waste-package release rates in a repository, utilizing empirical correlations of laboratory leach-rate data, have invoked postulates of repository analogues to simulate the laboratory leach experiments. The postulated analogues are unrealistic; they introduce fictitious repository parameters, such as volume and volumetric flow rate of groundwater associated with each waste package and groundwater residence time, which are assigned arbitrary values for making predictions. They invoke functional dependence on parameters inconsistent with well-established mass-transfer theory, and they incorrectly assume that the dissolution mechanisms that control release rates observed in laboratory experiments are controlling or important in the repository.

Both the WISP and the Performance Assessment National Review Group endorse the Pigford—Chambré approach for long-term predictions of releases from the waste packages. We also agree that it seems to be a reasonable and more defensible method for use in the licensing process. It is recognized by all concerned, however, that even though using saturation concentrations at the canister wall represents a bounding condition for dissolved species, the question of colloid formation has to be addressed since, in theory, this could cause greatly increased concentrations. Plutonium, in particular, is very prone to forming colloidal suspensions. Such suspensions could be treated as diffusing species, provided that the effective diffusion coefficient was known. The most important parameter is the solubility of each radionuclide of importance. Determination of reasonably accurate solubilities (or perhaps, more accurately, saturation concentrations) for some of the low-solubility radionuclides is not a simple task and is subject to large variations, depending on the exact conditions, particularly in the case of the multivalent actinides.

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APPENDIX F

BRINE MIGRATION IN SALT

The phenomenon of brine migration appears to be the most likely mechanism that can deliver water to the waste package during the containment period, and possibly through the isolation period, depending on the efficacy of the shaft and borehole seals. Consequently, for the expected small brine migration rates, the rate of inflow of brine to the waste package could control the corrosion rate, and the total brine accumulation can determine the effective lifetime of the waste package. This possible control of corrosion by the brine influx is the result of the chemical reactions involved in the corrosion of steel (see Sect. B.1.2). After some initial period, the entrapped oxygen would be consumed, and in a sealed repository the only oxygen available for converting metallic iron to the oxide is the oxygen in the brine that would accumulate around the waste package. However, this is a supposition that must be demonstrated. The brine flow rate could be significantly higher than expected such as occurred in the tests at the WIPP site (NOWAK 1986). The phenomenon of brine migration is discussed in detail in the following sections along with the various models that have been developed to quantify brine migration, the experiments, and the in situ tests that have been performed to develop the necessary data base.

F.1 MIGRATION OF ALL-LIQUID BRINE INCLUSIONS IN CRYSTALS

All-liquid brine inclusions within a crystal of NaCl (or other alkali halides) migrate up a thermal gradient in the crystal by a mechanism in which salt dissolves at the warmer brine-solid interface and crystallizes out at the cooler interface, after diffusing through the inclusion by molecular and thermal mechanisms. Based on laboratory work with KCl, Anthony (1971), who used a thermodynamic approach, was the first to publish a correct derivation of the equation describing the mechanism that tacitly assumes linear interfacial kinetics (i.e., the salt flux at an interface is directly proportional to the supersaturation or undersaturation). Geguzin (1975) later derived essentially the same equation by using a more straightforward mass balance approach that also assumed linear kinetics at the salt-brine interface. The more recent reviews of this phenomenon are by Gnirk (1981), Shefelbine (1982), Olander (1980, 1984), Yagnik (1983), and Jenks (1981). The review by Jenks is the most detailed with regard to comparing the various derivations, as well as demonstrating their equivalency when put on the same basis. Olander also discusses the interfacial interactions from a crystal theory viewpoint.

Olander and co-workers (1980, 1984) have suggested that linear kinetics may not hold for the small departures from equilibrium that will generally exist in moving brine inclusions. Consequently, they suggested that a power law be used for the surface kinetics (i.e., salt flux is dependent on the degree of supersaturation or undersaturation raised to some power). Based on the Burton-Cabrerra-Frank (BCF) theory of crystallization kinetics (BENNEMA 1973), a value of 2 was assumed for the exponent of the power law, which is approached for low supersaturation according to theory. The derived equation for the migration velocity shows a dependence on the square of the temperature, in contrast to the linear dependence of the linear kinetics model. For the case of diffusion control (i.e., mass transfer coefficient at the interface is very high), the Olander equation reduces to the equation derived by Anthony (1971) and others. The experimental data of Olander were in good agreement with his nonlinear kinetics model for the relatively small inclusions and test conditions for KCl crystals, but the NaCl data gave inconclusive results; either model seemed to apply within the limits of experimental error. According to Olander, with the relatively large dimensions (typically 1 mm) of the inclusions in naturally deposited halite, diffusion control (essentially linear kinetics) can be expected because the number of crystal dislocations that would intersect is sufficient to reduce the kinetic resistance to salt dissolution to negligible values.

F.2 MIGRATION OF TWO-PHASE BRINE INCLUSIONS

Brine inclusions that contain two phases (brine and gas/vapor) can move down the thermal gradient. A qualitative description of the mechanism (ANTHONY 1972; OLANDER 1984) is as follows: Water evaporates from the hotter side of the gas/vapor bubble and condenses at the cooler side. The condensed water dissolves salt on the cooler side, which is recycled by backflow of the brine to the hotter side where salt crystallizes out because of the water evaporation that occurs there. Modeling such a mechanism is more difficult than in the case of the all-liquid inclusions because of the additional parameters that involve the gas phase.

Olander (1980, 1984) developed an approximate analytical model to determine the velocity of migration of two-phase inclusions in the presence of a thermal gradient. The predictions of the model were in reasonable agreement with their experimental results for NaCl, but the results for KCl were in poorer agreement. A less sophisticated, but basically similar, theoretical relationship was derived by Anthony (1972).

This motion away from the heat source by two-phase inclusions is of concern to waste disposal because of the potential for transporting radionuclides out of the repository. It has been postulated that this could occur when an all-liquid inclusion contacts the waste canister after it has been breached, some water evaporates, and then the inclusions reseal with some of the insoluble gas (radiolytic or otherwise) trapped inside. Similar inclusions could possibly be formed by resealing of cracks in the salt near the waste canister.

It is difficult, however, to visualize the phenomenon occurring to an extent that will create a significant hazard, particularly when one considers that the thermal gradient decreases as the distance from the repository increases. In addition, Jenks (1981) points out that the down-gradient migration might depend on the composition within the inclusions. For example, the water vapor pressure over a brine containing a high concentration of $MgCl_2$ will be significantly less than that over a pure NaCl brine at the same temperature. Also, the transfer of water by backflow of brine might be reduced by the presence of $MgCl_2$. These considerations have caused Jenks (1981) to conclude that the formation, or occurrence, of two-phase inclusions in a salt repository will not adversely affect either the amount of brine reaching a waste package or the satisfactory isolation of radioactivity.

F.3 THRESHOLD THERMAL GRADIENT FOR BRINE MIGRATION

The existence of a threshold temperature gradient for the thermally induced migration of brine inclusions is supported by the theoretical equations of Anthony (1971) for the rate of migration of these inclusions within alkali halides and by the experimental findings of Cline (1972b). In order for an inclusion to move, the theory requires that a minimum chemical potential be exceeded by the driving force created by the temperature difference across the inclusion. That is, the temperature difference must be sufficient to meet the requirement of critical undersaturation for dissolution at the hotter interface and critical supersaturation at the cooler interface.

The occurrence of a threshold thermal gradient can also be inferred from the presence and distribution of brine inclusions in bedded salt, which could be considered as the geologic analogue for the theoretical and laboratory studies.

Based on the theory developed by Anthony (1971) and Geguzin (1975) and on the assumption that there is no surface tension at the grain boundaries, Pigford (1982) estimated that the brine inclusions in the halite layer in the Delaware Basin would migrate downward roughly 100 m under the influence of the existing geothermal gradient in the >200 million years of the salt deposit if no threshold temperature (or dissolution-nucleation threshold) gradient existed. He points out that no 100-m-thick halite beds exist and that the halite layers are separated by interbeds of less soluble material such as anhydrite, clay, salt, or polyhalite. Consequently, even lesser brine migration rates would have removed all the brine inclusions and caused an accumulation of brine at the interfaces of these layers. If reasonable values for surface tension are assumed to exist at grain boundaries, Pigford states that essentially all of the inclusions should have been trapped there. The lack of evidence for significant brine accumulation at the interbeds or the grain boundaries led him to propose the existence of a finite dissolution-nucleation threshold (chemical potential threshold), which required a temperature gradient greater than some minimum value to overcome resistance to the motion of the brine inclusions.

Assuming a temperature of 25°C, Pigford estimated threshold thermal gradients of 0.109 and 0.39°C/cm for inclusion sizes of 2 and 0.1 mm, respectively. Calculations made by Jenks and Claiborne (1981) indicate a threshold gradient of ~5°C/cm for 1-mm inclusions at 150°C. Lambert's (1979) post-test evaluation of the Salt Block II experiment at Sandia National Laboratories indicated that brine inclusions did not move in locations where the thermal gradient was <3.25°C/cm for the case of a mined natural salt block with an average inclusion size of ~0.1 mm at 25°C. Review of laboratory results for determining temperature threshold gradients in natural rock salt from the Palo Duro Basin indicated that threshold gradients may be much greater than previously hypothesized (RATIGAN 1985a). For example, these studies indicated that a 0.1-mm inclusion at 25°C would only move when the temperature gradient exceeded 4 or 5°C/cm. This led Ratigan to conclude that substantiation of the results by further testing may very well prove that migration of brine inclusions is a nonissue in rock salts with relatively small inclusions on the basis of the developed theory and the available data for brine migration in crystals. This may be the case, but natural salt is not homogeneous and other factors such as dehydration of minerals in stringers and interbeds and liquid and vapor transport along grain boundaries and cracks could predominate, which apparently occurred in tests at the WIPP site (NOWAK 1986).

In spite of the evidence for the existence of a threshold gradient, the acceptance of the concept is not universal. Roedder (1982) commented that the experimental data used to support the concept of a threshold gradient may not be applicable, and some other data suggest that a threshold gradient may not exist. Chou (1983) also disagrees with the threshold concept, based on considerations of the magnitude of interfacial kinetics and grain boundary effects.

F.4 BRINE MIGRATION IN NATURAL SALT DEPOSITS

Naturally deposited formations of salt are composed of tightly bound aggregates of crystals of NaCl in halite form with various amounts of clay minerals interspersed throughout the formation as stringers and interbeds. The halite contains anhydrite impurity and small amounts of other minerals, including clays and brines. The crystals are anhedral in shape, with characteristic dimensions in the range of 3 to 30 mm. The anhydrite and the clays are usually concentrated along the crystal or grain boundaries, but small amounts of these impurities are also found within the halite crystals. Water is present in the form of small saturated brine inclusions within the halite crystals, the interlayer water in clay impurities, and other minerals that may contain waters of crystallization.

Field tests involving brine migration in salt deposits are discussed below. These include Project Salt Vault, the Avery Island heater tests, the experimenta in the Asse II salt mine in the Federal Republic of Germany, and tests at the disposal horizon of the Waste Isolation Pilot Plant (WIPP) in New Mexico.

F.4.1 Project Salt Vault

The first significant experience with thermally induced brine migration in geologically deposited bedded salt occurred in Project Salt Vault (BRADSHAW 1971). The experiment was carried out in the Lyons, Kansas, mine of the Carey Salt Company using 14 irradiated Engineering Test Reactor (ETR) fuel assemblies contained in seven cans for a heat loading of ~1.6 kW/can at emplacement. These cans were placed in a circular array of holes (~10 in. in diameter and 12 ft deep) in the floor with one can in the center and the other six cans located peripherally on 5-ft centers. An identical array using electric heaters only was operated as a control to determine the combined effect of radiation and heat on the salt characteristics. Over the course of the experiments, during which the temperatures in the salt in the immediate vicinity of the emplacement holes reached upwards of 200°C, the discovery was made that small inclusions of brine in the salt were migrating toward the heat sources. As a consequence, several experiments were conducted to determine the rate of moisture inflow into the emplacement holes after installation of an off-gas condensate collection system.

Based on the condensate collected over a period of ~19 months, it was estimated that prior to shutdown the average brine inflow rate ranged from 0.2 to 3.0 mL/d. By utilizing a modification of the theory of migration of liquid inclusions in ice and the results of some experiments on salt specimens from the Carey Salt Company, Bradshaw and McClain (BRADSHAW 1971) estimated the transient rate of brine migration into emplacement holes for a conceptual repository situation for salt having a water content of 0.5 vol %. For a waste age of 3.33 years, an initial heat generation rate of 1.63 kW, and a waste canister spacing of 2.9 m (corresponding to a thermal loading density of ~19.4 W/m^2), the transient temperature field in the salt around a canister was calculated on the basis of an infinite array of line sources. The calculations indicated that a maximum water inflow of ~3.6 mL/d could be anticipated at approximately one year after waste emplacement. This value is in general agreement with the upper-limit estimate for the emplacement arrays in Project Salt Vault. The estimated rate decreased with time, approaching zero at ~20 to 30 years after burial. Very roughly, the first five years would produce a total inflow of ~6 L/hole; the next five years, ~5 L/hole; 10 to 15 years, ~3 L/hole; and 15 to 20 years, ~1 L/hole, with a total inflow of ~15 L/hole. It was stated that the calculations were conservative, so they may be too high by at least a factor of 2, but probably not by an order of magnitude. One might then expect a total inflow per hole of something like 2 to 10 L, taking place over a period of 20 to 30 years.

F.4.2 Avery Island Experiments

Brine migration experiments (KRAUSE 1979; 1983) were made using electric heaters emplaced in the floor of the upper level (depth, 169 m) of the Avery Island Salt Mine in a salt dome near New Iberia, Louisiana, which is operated by the International Salt Company. Three experimental configurations were used, and each had Calrod heaters delivering 1 kW for the heat source that produced heat fluxes into the salt which are significantly larger than typical repository design values. The three configurations included:

1. Test AB. Natural brine movement under ambient-temperature conditions: This was a "control" experiment that was included to evaluate the movement of any natural brine as a consequence of stress perturbations due to drilling.

2. Test SB. Synthetic brine movement under elevated-temperature conditions: This experiment consisted of a centrally located and heated borehole within a pattern of three smaller-diameter boreholes containing synthetic brine that was tagged with deuterium. These holes were filled with glass beads and pressurized (690 kPa).

3. Test NB. Natural brine movement under elevated-temperature conditions: This experiment was designed to evaluate the movement of natural brine in dome salt at elevated temperatures.

During the initial 315 d of heating, the experiments exhibited steady-state moisture collection rates ranging from 0.026 to 0.044 g/d. Apparently, the heating at Sites NB and SB caused the collection rate to be about twice that for the unheated condition at Site AB. Site AB can be considered to yield a background level of moisture collection for the other sites, implying the moisture collected at Site AB can be subtracted (for equal time periods) from that collected at Sites NB and SB to calculate the moisture increase related to heating. The moisture collection data at all sites showed a significant initial collection rate, which can be attributed to removal of the residual moisture in the air included in the heater boreholes and on the heater borehole walls and the heater sleeves (at Sites SB and NB). A detailed examination of the data showed that the final precooldown moisture collection rate at Site SB was 0.037 g/d, while the collection rate at Site NB was 0.044 g/d.

The moisture collection during Site SB cooldown showed significant gains that corresponded with permeability changes measured during the cooldown period. The moisture collection rate at Site SB was ~0.04 g/d prior to cooldown, which was significantly less than the cooldown values. The increased collection rates were postulated to be attributed to microcracking and opening of the grain boundaries in the vicinity of the heater borehole wall. This explanation implies that the large amount of brine (moisture) released during cooling of the salt was caused by the change in tangential stresses created by thermal response of the salt during power reduction. These stresses were relieved during cooling, allowing the trapped water in the salt to break free and enter the heater borehole.

Generally, the results of these experiments are in line with others, but a quantitative application of the brine migration results would be a dubious procedure. Whether the experimental room is representative of the salt at Avery Island can be questioned because of its past history (i.e., flooding; brine puddles; and damp crushed salt, which was scraped away in some places). In the limited area where the heater holes were emplaced, the floor was apparently dry and had not been covered with crushed salt.

F.4.3 Asse II Salt Mine in the Federal Republic of Germany

Elaborate in situ experiments were performed in the Asse II salt mine in the Federal Republic of Germany. The tests were made at the 900-m level in areas specially mined in the Main Halite, which is considered typical of the potential repository salt types.

Four tests were designed to be performed at a maximum salt temperature of 210°C with a 3°C/cm thermal gradient at the borehole wall. Each test configuration consists of a central borehole 43.5 cm (17 in.) in diameter containing sleeve-protected heaters and other internals and, in two of the test holes, ^{60}Co sources. The void between the heated sleeves and the borehole wall, which is 5 cm thick, is filled with alumina beads. In the brine test holes (sites 2 and 4; the latter contained ^{60}Co, but the former did not), the brine vapors migrate to the alumina beads and are transported to the collection points by circulating nitrogen. Eight guard heaters are peripherally located around the borehole to simulate an array of waste packages. A cross section of the brine test assembly is shown in Fig. F.1.

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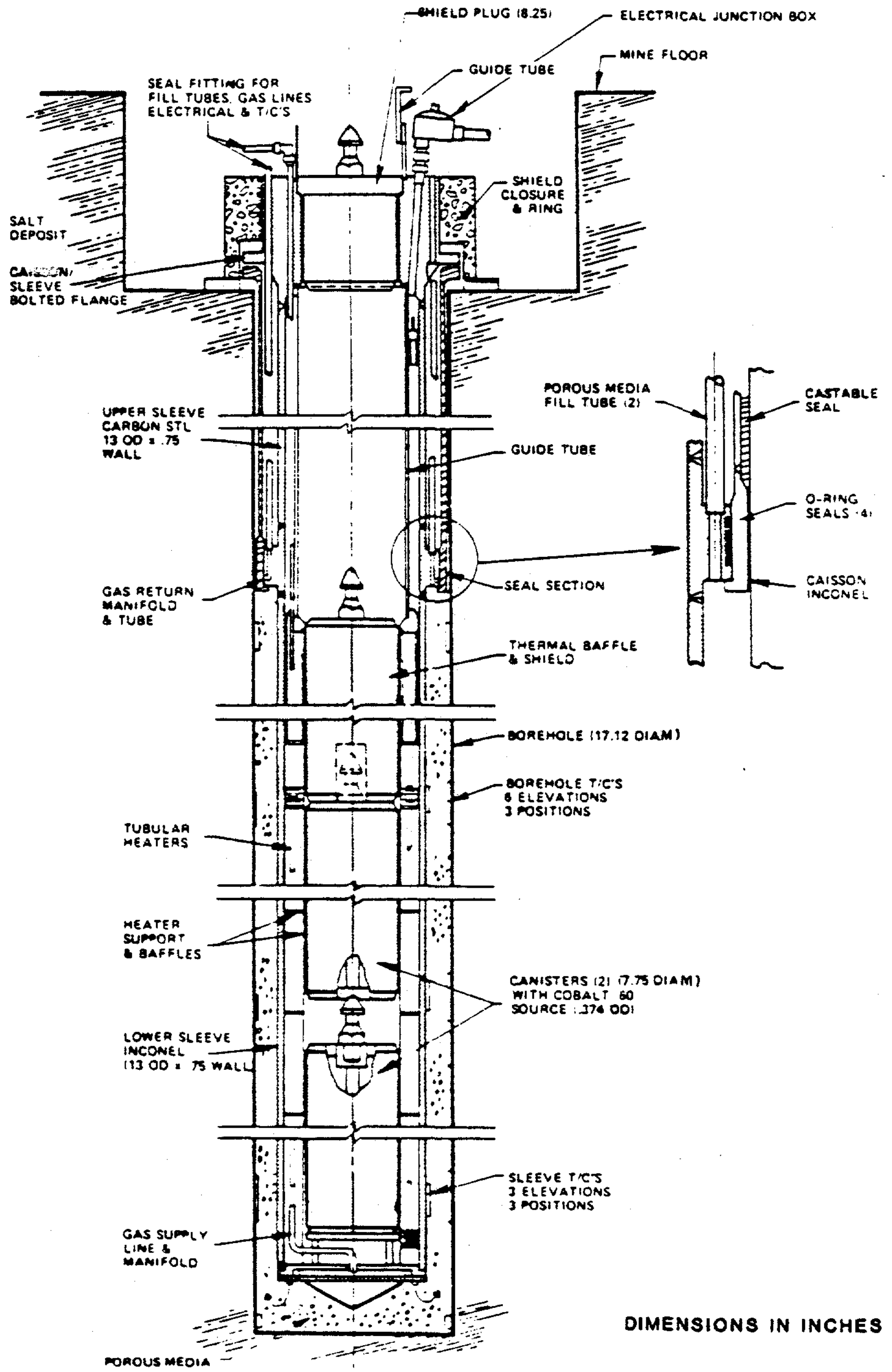

Fig. F.1. Cross section of brine migration test assembly. Source: T. Rothfuchs, D. Lübker, A. Coyle, and H. Kalia, Nuclear Waste Repository Simulation Experiments, Asse Salt Mine, Federal Republic of Germany: Annual Report 1983, BMI/ONWI-539, 1984.

Correlation of the results of the tests in comparison with a pretest estimate made using the Jenks equation (Sect. F.5.1) is shown in Fig. F.2. The effect of radioactivity on brine migration was small. In this particular application, the Jenks equation produced very conservative results.

F.4.4 Waste Isolation Pilot Plant (WIPP)

Experiments to quantify brine migration in full-scale tests simulating repository environments are under way in the Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico. Two vertical boreholes in the floor (655 m below the surface) equipped with 1500-kW electrical heaters and a moisture collecting system were used to simulate high-level defense wastes for overtest conditions (NOWAK 1986).

After 265 d of operation, 17.6 kg of water had collected in one borehole and 16.1 kg in the other. Water collection was characterized by a small, nearly constant rate before the heaters were turned on, which increased rapidly after heat was applied. During the first 4.2 d before the heaters were turned on, 22.4 g of water collected in one borehole and 19.3 in the other, for an average rate of 5.4 g/d. After operation for 160 d with the heat on, water collection rates remained relatively constant at ~75 g/d in one borehole and 65 g/d in the other. These quantities of water were much larger than predicted by the Jenks equation and considerably more than that collected at the Asse II test. In an attempt to reconcile the difference between the Asse II and the WIPP test results, the data from the Asse II tests were scaled to that of the WIPP (i.e., by correcting for the differences in the moisture content of the salt and the borehole wall areas in the heater zone). Application of the scaling factors indicated that 3 kg of water would have been collected from Asse II site 2 in 10 months and 2 kg from site 4 in 3.5 months. These values are significantly smaller than the 16 to 18 kg collected from the two test holes (in room B of the test facility) where the thermal conditions are most nearly equivalent to those for the Asse II experiment.

Another significant difference between the WIPP and Asse II test results is the unmeasurably small water collection rate before the heat was turned on at the Asse II site compared to the 4- to 15-g/d collection rates at the WIPP site.

It was concluded that the unexplained differences between the Asse II and WIPP test results may be due to differences in the transport mechanisms for the brine at the two sites. The salt diapir of the Asse II site is likely to be characterized by tighter grain boundaries, more uniform grain size, and thinner, more generally discontinuous interbeds of clay and other minerals. Consequently, the preferred flow paths for brine or water may be different and may present greater flow resistance than that encountered at the WIPP site (NOVAK 1986). In addition, the numerous thin clay interbeds at the WIPP site could be acting as an additional water source in comparison with the pure salt at the Asse II site.

F.4.5 Trapping of Brine Along Crystal Boundaries

On arriving at a crystal or grain boundary, brine may become trapped there, depending on a number of variables. Trapping is predicted by theory, and experimental evidence of such a phenomenon does exist.

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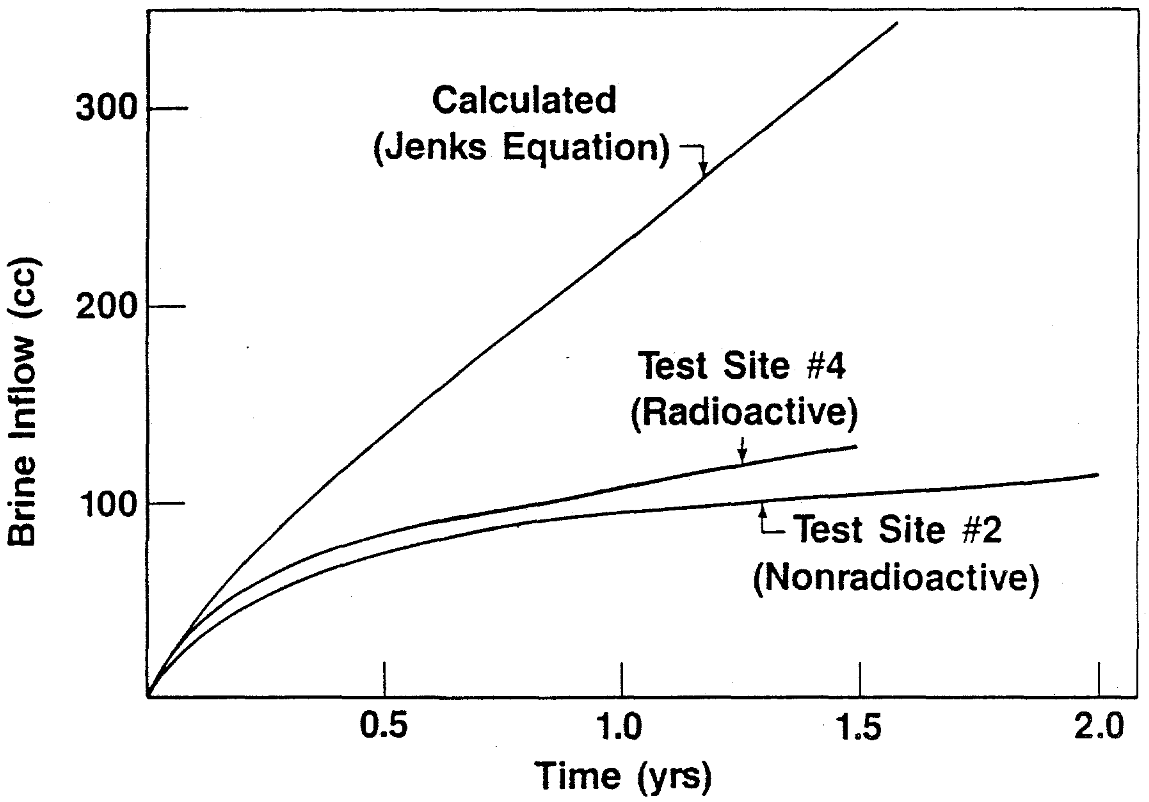

Fig. F.2. Comparison of Asse brine inflow results with pretest estimate using the Jenks equation. Source: J. Cunnane and L. Pederson, "Waste Package Environment," presented at the SRP/NRC Waste Package Meeting, Columbus, Ohio, Jan. 22–24, 1986.

An important result of the Project Salt Vault experiment was that ~90% of the brine inflow to the emplacement hole occurred when the electric heaters were turned off and the salt walls were allowed to cool. The explanation offered for this greatly increased brine release during the cooling of the salt was that the tangential stresses created by thermal expansion of the salt during heating were relieved during heating, thereby allowing the trapped water in the salt to break free and enter the space between the heated canister and the salt formation. A further explanation might be that the brine was trapped on grain boundaries after migrating there during heating and that relieving the tangential stresses allowed the grain boundaries to open sufficiently for the brine to flow into the emplacement hole (JENKS 1979). Another possibility for brine trapping along the grain boundaries based on the effects of the temperature gradient on thermal stresses and the differences in the coefficients of expansion of brine and salt has been suggested by Pigford and Chambré (1985); this is discussed later in this section.

The results of Project Salt, as well as similar results from the Salt Block I and Salt Block II experiments (HOHLFELDER 1980) where the heat was turned off, are good evidence that brine inclusions are trapped at grain or crystal boundaries when naturally deposited salt is exposed to a heat source. These results do not prove that all inclusions are trapped or that some transport, as brine or vapor, does not occur along the grain boundaries. However, because of the poorly soluble materials on the grain boundaries, it seems evident that the mechanism of intercrystalline transport would differ from the intracrystalline transport that has been discussed previously. The latter type of transport is discussed in Sect. F.1.

Cline (1972a) pointed out that brine droplets on crystal boundaries in KCl and other alkali halides are in positions of minimum energy; therefore, the force produced by the thermal gradient that is necessary to drive a droplet across a boundary can exceed the force required for motion through the body of the crystal. A theoretical expression developed for the retarding effects of the grain boundary surface tension was used by Jenks (1981) to make estimates on the gradient required to drive a brine inclusion through a boundary. His calculations indicated that a gradient sufficient to cause movement within a crystal of a >0.2-cm-wide inclusion may drive the same inclusion through a boundary. However, this transfer to adjacent crystals has only been observed in experiments for artificially produced clean contacts. This does not necessarily mean that a brine inclusion would be driven into the adjacent crystal because the insoluble impurities contained between the crystal surfaces could act as a retardant to further migration through a crystal boundary or along the paths between adjacent crystals.

Pigford and Chambré (1985) have developed another theory that predicts that the flow of brine along grain boundaries could be directed away from the waste package and subsequently halted under the influence of the temperature gradient and thermal stress. This theory (as described below) is based on the fact that brine, in regions of higher temperatures, will be at a higher pressure than that present in cooler regions because of the greater thermal stresses and because the liquid brine has a higher coefficient of thermal expansion than the surrounding salt. Consequently, if there is an annular space (either void or filled with crushed salt) that is below the pressure in the salt near the interface, some of the intergranular brine can be expected to flow into the annular space and some will flow outward into the cooler salt. Figure F.3 illustrates the pressure profile that could result with no intergranular flow and the profiles that could result after pressure-induced flow. After a few years, the void space or crushed salt will consolidate into a monolithic structure (RATIGAN 1985a), but the lack of a low-pressure cavity need not affect the brine inflow. In the case of pressure-driven transport of the intergranular brine, the direction of flow will depend on the boundary conditions at the interface. If the salt is heated more rapidly than the thermal stress can be relieved by creep, the salt adjacent to the waste package will be under additional compression because of the thermal expansion of the salt. Since brine has a higher coefficient of thermal expansion than solid salt, the heating can generate a pressure within the confined brine that exceeds lithostatic pressure. The net result is an outflow of brine until a zero-pressure gradient is attained. Surface tension can result in a brine film between the salt and waste package, and the local fluid pressure of this film can temporarily exceed the local compressive stress on the salt until outward flow of brine relaxes the fluid pressure to lithostatic. After that, no mechanism for brine inflow to the waste package will exist other than that resulting from consumption of brine components by chemical reaction and radiolysis.

In summary, the evidence indicates that brine could remain entrapped at crystal boundaries for an indefinite period or could be driven toward the cooler region according to the Pigford-Chambré theory since, unlike the heater experiments, the nuclear heat source cannot be turned off. However, openings along boundaries or cracking due to thermal stresses could provide a path for transport by advective flow for liquids and vapor. It may also be possible for vapor to diffuse along unopened crystal boundaries. These possibilities have led to the development of models that include such phenomena. They are discussed in the following sections, along with the application of the solution-precipitation model, which assumes that the rock salt is a homogeneous mass (i.e., no crystal boundaries; brine migration is intracrystalline).

F.5 BRINE MIGRATION MODELS

A number of models have been developed for calculating brine migration rates in rock salt. The modeling effects can be categorized by either of three different major assumptions, as follows:

1. All migration is governed by the intracrystalline transport mechanisms (i.e., grain boundaries are neglected).

2. All migration occurs on the crystal faces and is governed by pressure-drop transport.

3. The mechanisms of items 1 and 2 are coupled.

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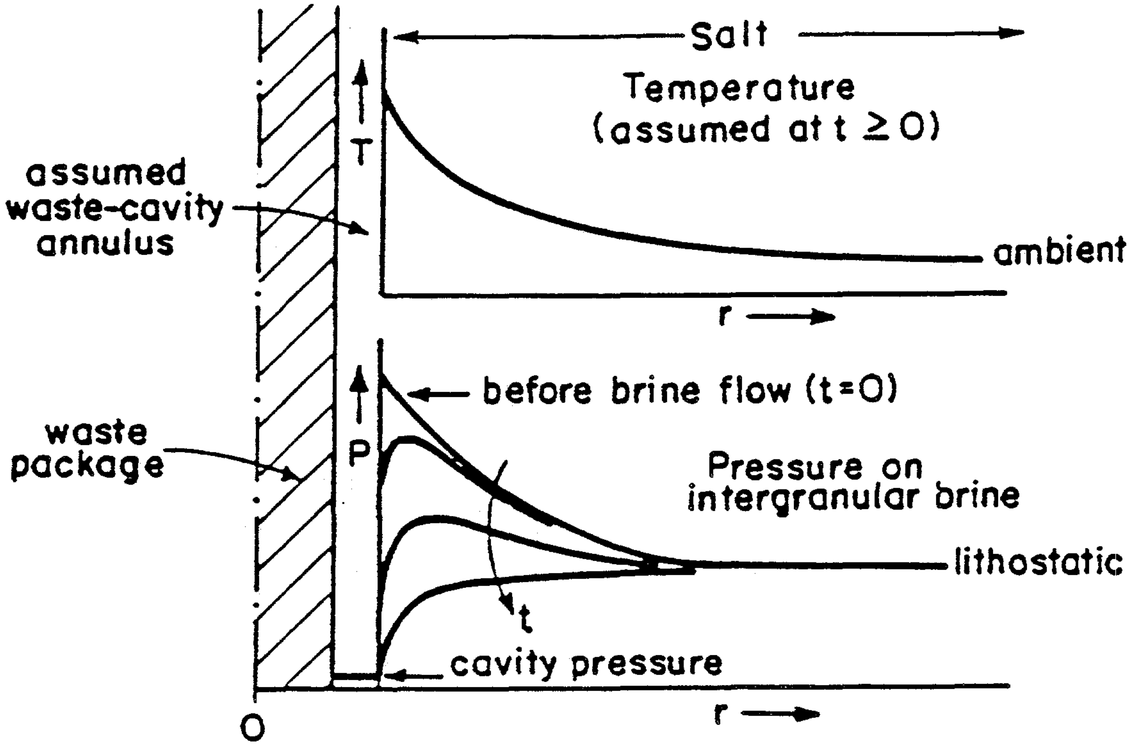

Fig. F.3. Profiles of pressure on intergranular brine, neglecting salt creep (steady-temperature profile assumed for illustration). Source: T. H. Pigford and P. L. Chambré, Mass Transfer in a Salt Repository, LBL-19918, 1985.

F.5.1 Intracrystalline Migration Models

The basic assumption in applying the intracrystalline migration model to a repository is that the salt is homogeneous and no crystal boundaries exist. It has been argued that application of the intracrystalline migration equation (either the theoretical one or the empirical Jenks equation discussed below) may be conservative in that trapping on crystal boundaries is neglected. However, the possibility of cracking and vapor transport along grain boundaries creates doubt as to whether the homogeneous crystalline transport model is conservative.

The theoretical equation derived by Anthony and Cline (ANTHONY 1971), and the equivalent of those derived by others when put on the same basis (JENKS 1981), is

$$V = \frac{DC_\ell}{C_s} \left[\left(\frac{1}{C_E} \frac{\partial C_E}{\partial T} - \sigma \right) \nabla T_\ell - \frac{K}{RTL} \right], \qquad (1)$$

where

V = migration velocity, cm/s;

D = diffusivity of salt in brine, cm^2s;

C_ℓ = concentration of salt in brine droplet, mol/L;

C_s = concentration of salt in solid salt, mol/L;

C_E = equilibrium concentration of salt in brine, mol/L;

T = absolute temperature, K;

∂ = Soret coefficient of salt in brine, $°C^{-1}$;*

∇T_ℓ = temperature gradient in the brine droplet, °C/cm;

K = kinetic potential, $K_d + K_p$, ergs/mol;

K_d = change in chemical potential at the dissolution interface generated by irreversible processes associated with the transfer of salt between the solid and liquid phases, ergs/mol;

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*The Soret coefficient, σ, is conventionally negative when thermal diffusion causes transport of solute from the hot region to the cold region of a solution.

K_p = change in chemical potential at the solidifying interface generated by irreversible processes associated with the transfer of salt between the solid and liquid phases, ergs/mol;

R = gas constants, ergs $\mathrm{mol}^{-1}\ {}^{\circ}\mathrm{C}^{-1}$; and

L = dimension of droplet parallel to thermal gradient, cm.

The kinetic potential term involving the interfacial kinetics, which is probably the most difficult term to quantify, governs the threshold gradient. For motion to occur, the first term must exceed the second, or kinetic potential, term.

In developing his empirical equation, Jenks (1979) evaluated the available theoretical and experimental information. He concluded that crystal stressing, high pressure, and energy storage by radiation damage would have little effect on brine migration in halite crystals and that the kinetic potential was largely responsible for most of the scatter in experimental results. He also concluded that the kinetic potential was of little importance (and the conservative treatment would be to neglect it) in the many experimental results of Bradshaw (1968) for natural salt crystals. He then drew a curve that conservatively represented the migration velocity per unit temperature gradient as a function of temperature. About 80% of the observed points were on, or below, the curve; the remainder were above. Consequently, it can be argued that some conditions could produce higher migration velocities than those represented by the Jenks curve, which was fitted by the following equation:

$$\log V/G = 0.00656T - 0.6036, \tag{2}$$

where

V/G = velocity per unit temperature gradient, cm^2/year, °C; and

T = temperature of salt, °C.

Equation (2), commonly referred to as the Jenks equation, was used in the calculations for salt repositories made by Claiborne (1980) and Jenks (1981) via the MIGRAIN computer code and the calculations made by McCauley (1984) via the BRINEMIG computer code (McNULTY 1984). The BRINEMIG code is a finite-element brine migration model that assumes the salt to be a homogeneous and isotropic continuum and is based on the use of the continuity equation to predict the brine inclusion density in salt. The MIGRAIN code is similar, except that the solution is by finite differences.

The BRINEMIG code was shown to give brine accumulations that agreed with those from the Salt Block II experiments (HOHLFELDER 1980) at low temperatures and overpredicts brine accumulations at higher temperatures. The McCauley (1984) calculations for the current reference design, as reported by DOE (1984), gave 0.85 and 0.41 m^3 for CHLW and PWR spent fuel, respectively, when assuming a threshold temperature gradient for brine migration and 0.95 and 0.75 m^3 without a threshold gradient. It was also assumed that the concentration of the brine inclusions was 5 vol %, a factor of 2.8 greater than the estimated total water content of 1.8 vol %. The rate of brine flow is known to decrease with time; however, the highest rate was only 0.06 m^3/year.

The MIGRAIN code was developed for both Cartesian and two-dimensional cylindrical coordinates. The required input of temperatures and temperature gradients as a function of time and space is supplied by separate heat-transfer calculations. Claiborne (1980), using MIGRAIN, calculated the brine inflow for a number of conditions. For example, assuming 0.5 vol % brine inclusions and no threshold gradient, the brine accumulation for an HLW repository with a 37-W/m^2 (150-kW/acre) thermal loading and 2.16 kW/canister was ~12 L. For spent fuel with a thermal loading of 14.8 W/m^2 (60 kW/acre) and 0.55 kW/canister, the brine accumulation was ~6 L.

Jenks (1981) made upper-bound estimates of water in migration by using a method that involved estimating the volume of rock salt around the waste canister, based on threshold temperature gradient considerations, from which all contained brine inclusions were assumed to eventually reach the waste emplacement hole. To facilitate the calculations, concentric shells were assumed to exist around a waste package with the following shape: a cylindrical surface surrounding the waste package with a height equal to the length of the package with hemispherical surfaces attached to the bottom and top of the cylinder. Previously calculated salt temperatures as a function of time at the midplane were used in obtaining values for threshold temperature gradients [from Eq. (1)]. In determining the salt volume that could contribute brine to the waste package, the total area was found for a shell surface which was required for uniform and instantaneous flow of heat at a particular distance from the waste package for a calculated value of a threshold temperature gradient in the salt located at the shell surface. The volumes of water contained from the largest computed volume of salt with the procedure discussed above were 240 and 60 L for the CHLW container (2.16-kW) and SF container (0.55-kW), respectively, for an assumed 1 vol % water in bedded salt. These values are factors of ~13 and ~8 greater than the results of MIGRAIN calculations for the same conditions.

Such calculations should be useful in establishing an upper-bound estimate for brine migration to a waste package, provided that the uncertainty in the threshold gradient can be reasonably established and the Jenks equation is representative of the particular salt formation, which may not be the case as was apparently demonstrated by the WIPP experiments (NOVAK 1986).

F.5.2 Vapor and Stress-Gradient Transport Models

Investigations of brine migration in blocks of halite at Sandia National Laboratories have led to a proposed model of vapor transport along grain boundaries or cracks, with no consideration being given to migration of the intracrystalline inclusions. The observed behavior of increased flow after the heat source was turned off is accounted for by including stress relaxation effects in the model. The first model was developed on the assumption of Darcy flow of vapor through a porous medium; this was applied to the 1-kg block experiment (HOHLFELDER 1979) and later to the Salt Block II experiments (HOHLFELDER 1980) at Sandia National Laboratories. A second model (HADLEY 1981), which produced a more accurate correlation of the experimental data, assumed that the vapor flow was entirely in the Knudsen regime (the mean-free path of the vapor molecules is on the order of the pore diameter or equivalent spacing) when applied to these experiments.

Both models utilize the concept of a receding evaporation front in a flow channel, and both attempt to predict the effect of varying or turning off the heat source by including the effect of stress changes on the cracks in the salt. Both use fit parameters in correlating the data. The experimental data were correlated by solving five simultaneous differential equations: the steady-state continuity, the ideal gas, the Darcy's or the Knudsen flow, the evaporation front velocity, and the time-dependent heat conduction equations.

Aside from the assumption that Knudsen flow is the most important transport mechanism, the refined model divides the total porosity into two parts: (1) a small connected porosity that reflects the "flow channel" dimensions limiting the vapor flow rate, and (2) a larger "inclusion" porosity that reflects the total water content influencing the rate of recession of the evaporation front. Since residual strains might cause a considerable change in flow channel properties, the connected pore radii were considered to vary as a linear function of the residual strain in the tangential direction. The initial values for the equivalent pore radius and the connected porosity were treated as fitting constants that provided the best match with the experimental data.

A simple stress-gradient transport model was developed by Shefelbine (1982), who assumed that the pressure of brine in connected porosity is determined by the stresses (bulk pressure) in the salt itself. This seems justifiable since the difference between the pressure in a brine-pore and the stress in the adjacent salt would cause the salt to creep until the pressures are equalized. On this basis, the changes in rock stress produced by a heat source in rock salt can be expected to provide a driving force for brine migration.

As an argument against the vapor-phase mechanism and a possible support for a stress-gradient model, Shefelbine points out that the rock stresses near the heater hole in the Salt Block II experiment are much greater than the vapor pressure of the brine and, therefore, any channel filled with water vapor will tend to be closed by salt creep. However, on the other hand, he points out that the stress-gradient mechanism cannot explain the release of water from samples heated in an oven since no stress gradients will exist after equilibrium temperature is reached.

In making calculations for a repository, Shefelbine used the Darcy flow equation in combination with a secondary creep model for salt (TRUCANO 1981). Figure F.4 shows a comparison of Shefelbine's results for the stress-gradient model, the fluid inclusion model (the Jenks equation with a homogeneous mass), and the Knudsen flow model for vapor-phase transport when applied to a repository for CHLW with thermal loadings of 2.16 kW/canister and 37 W/m^2 (150 kW/acre) and with salt containing 0.5 vol %

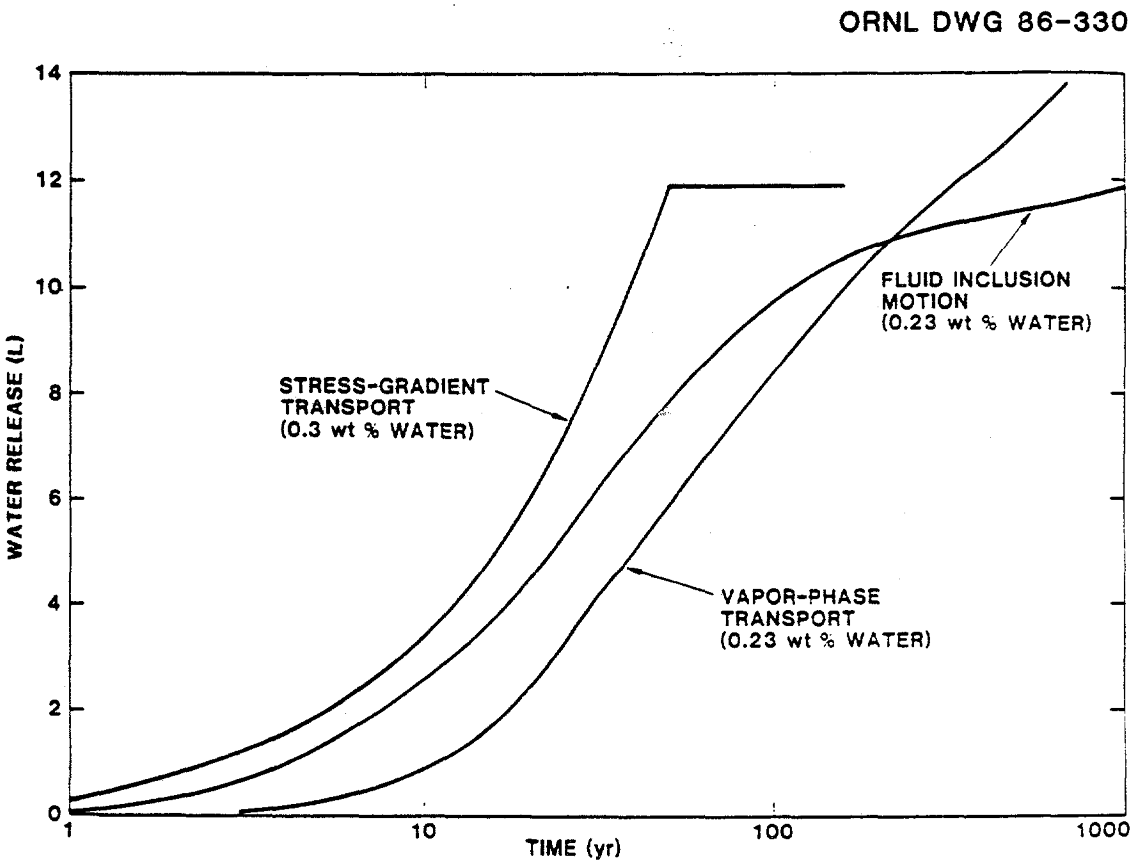

Fig. F.4. Water accumulation vs time for repository conditions. Source: H. C. Shefelbine, Brine Migration: A Summary Report, SAND82-0152, 1982.

(0.23 wt %) water. The results show that the three very different models predict about the same total water accumulation in 1000 years, 12 to 15 L. These results and other observations caused Shefelbine to conclude that an accumulation of 20 L at the emplacement hole in 1000 years was a conservative value for the above repository conditions.

F.5.3 Combined Intracrystalline Transport and Crystal Boundary-Flow Models

Bloom (1979) developed a preliminary model that combined the temperature-dependent, dissolution-deposition, diffusion model with the Darcy vapor-flow model proposed by Sandia National Laboratories. This was applied both to the Salt Block II experiments (HOHLFELDER 1980) and the ORNL experiments (SHOR 1978), which involved drying a salt core by passing dry air over the upper surface held at 190°C and the lower surface at 105°C.

The primary assumptions used for the combined model were that the two mechanisms act independently (the two flows are additive) and that a distinct and moving vapor-liquid interface exists within a uniformly porous medium. The basic equation expresses a mass balance between the vapor loss from the vapor-liquid interface and the brine loss that produces the motion of the interface.

The results of the analysis were three equations describing flow rates for the experiments at ORNL and Sandia National Laboratories (i.e., the hot and cold faces of the former and the centrally heated hole of the latter). Properties of the salt and brine and the vapor-liquid interface positions were "lumped together" into convenient groups, which were treated as arbitrary fitting constants in correlating the theoretical equations and experimental data. The theoretical equation for the cold face of the ORNL experiments contained three fitting constants; the equations for the hot face and the Salt Block II experiments contained four fitting constants. A good fit was obtained, particularly for the Sandia data; however, this does not necessarily validate the model since one would expect a good fit using four arbitrary constants.

Bloom (1979) recommended reexamination of the existing data on brine migration in order to test the proposed model and to develop new data that could lead to an improved model. However, further development of this model has apparently been discontinued.

A recent, more sophisticated model for the combined effects of intracrystalline and grain boundary flow has been developed by Ratigan (1985b) and incorporated into the computer code SPECTROM-58 (RATIGAN 1984a), a finite-element code for brine transport in rock salt for two-dimensional and axisymmetric geometries. In this model, the following significant assumptions were made:

1. All brine inclusions reaching the crystal boundary will exit crystal; that is, no kinematic deterrent exists.

2. All brine inclusions that exit a crystal will never reenter the same, or any other, crystal.

3. The escape of brine inclusions from the crystal is independent of the pressure, temperature, and inclusion density at the surface.

Transport within individual salt crystals is assumed to be driven by the differential solubility of salt resulting from thermal gradients established by thermal calculations. The governing equation for the flow mechanism is a modification of the usual continuity equation, and the code allows for a choice of functions (e.g., the Jenks equation) for the calculation of brine inclusion velocities. Transport of brine within the interconnected pore space is the result of brine pressure gradients; the governing field equation for this flow mechanism is the diffusion equation for pressure-induced flow in a saturated porous medium.

Using this model, Ratigan (1984b), in concert with the Plackett-Burman method of experimental design, determined that the most significant parameter ranges contributing to the total range in brine inflow are: (1) ratio of interconnected porosity to total porosity, (2) initial brine inclusion density, and (3) threshold temperature gradient. In calculations made with SPECTROM-58 utilizing the intracrystalline flow mechanism only, the results were essentially the same as those produced by BRINEMIG (McCAULEY 1984) with and without a threshold gradient of 0.125°C/cm for the 100 years modeled. However, calculations with SPECTROM-58, using the coupled field equations for brine flow, showed a factor of ~3 increase in the maximum brine flow rate to the emplacement hole when a gradient of 0.125°C/cm was assumed as opposed to the relatively small effect found by the BRINEMIG calculations. Consequently, Ratigan concluded that the crystal interface system is a very important flow path that is fundamental to modeling brine migration and should be investigated further.

F.6 COMPARISON OF BRINE MIGRATION MODELS

Neither the vapor-transport model nor the inclusion-migration model completely represents the phenomenon of brine transport up the thermal gradient. The former assumes transport only along cracks or openings in the grain boundaries, while the latter assumes only intracrystalline transport. Obviously, both of these transport phenomena and (probably others) could exist simultaneously with one or the other being dominant, depending on the existing conditions.

The a priori application of either model is not possible because values of some of the basic parameters cannot be determined without resorting to empiricism, which produces some of the parameters on a best-fit basis. The various predictions with the inclusion migration model use the Jenks equation, which is empirical and apparently conservative for the temperature gradients expected in a salt repository provided that brine inclusions are the only significant source of brine and no significant transport occurs along cracks or grain boundaries. The WIPP results indicate that such assumptions are not valid. Other empirical equations relating the migration velocity to the temperature and temperature gradient could be used in the computer codes.

The exhaustive discussions of theoretical and experimental information on inclusion migration in salt crystals by Jenks (1981) showed that the various treatments of theory can be made consistent and demonstrated the compatibility of the Jenks equation with such theory. The behavior of brine movement along the grain boundaries is not as well understood and does not seem to be as amenable to theoretical analysis as intracrystalline transport.

The vapor-transport model has conceptual difficulties with the receding liquid-vapor interface and with the assumption of exclusively vapor transport. Probably the interface is not sharply defined but extends over a region, and two-phase transport likely occurs.

The improved vapor-transport model with Knudsen flow gave results that agreed with the experiment more closely than did the original Darcy flow model, which is not surprising since two additional fit parameters were used to correlate the experimental data. The inclusion migration model using the Jenks equation also gave a very good fit to the heating phase of the Salt Block II experiment when the thermal conductivity of salt was treated as a fit parameter.

A considerable effort has been made to include the stress relationship in developing the vapor-transport model in order to predict the sudden increase in flow that occurs when the heat source is turned off. This will help substantiate the postulate that stress relaxation causes this flow phenomenon, although this information is purely academic since nuclear heating cannot be terminated abruptly.

Development of a combined intracrystalline and grain boundary flow model would be preferable if the model could be demonstrated to predict brine flow rates in repositories with a reasonable degree of uncertainty. Of the two models previously discussed, the one represented by SPECTROM-58 is more sophisticated and useful. The greater effect of the threshold thermal gradient that occurred when the grain-boundary transport was included in the calculations stressed the possible importance of establishing more firmly the concept and the quantitative values for the threshold gradient.

F.7 CONCLUSIONS

On the basis of the available information prior to the WIPP experiments, it seemed that any model could probably be used provided that it is based on, or normalized to, empirical data. In any event, the inflows of brine that had been predicted for repository conditions were relatively small (even in the case of the Asse II experiments) and did not seem to create serious problems. It is quite possible that grain boundary trapping and/or reverse flow (Pigford-Chambré theory) is responsible for the low inflow rates, but additional work is needed to corroborate these mechanisms. However, the much greater than predicted brine inflow rates to the waste canister in the WIPP experiments cast doubt on the general applicability of the Jenks equation and the ability to predict, or even bound, the brine migration rates in all types of salt formations. It is not clear as to why the WIPP results were surprisingly high. It has been postulated by Novak (1986) that the preferred flow paths for brine or water may be different at the WIPP site and present less flow resistance than other salts. In addition, the numerous clay interbeds and stringers could be providing an additional water source. A model that fits the WIPP results needs to be developed, which requires a greater understanding of the transport processes involved. However, if it is considered necessary to have accurate values for brine migration in a repository, only in situ testing at the disposal horizon can produce values with relatively small uncertainties.

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This document provides specifications for models/methodologies that could be employed in determining postclosure repository environmental parameters relevant to the performance of high-level waste packages for the Basalt Waste Isolation Project (BWIP) at Richland, Washington, the tuff at Yucca Mountain by the Nevada Test Site, and the bedded salt in Deaf Smith County, Texas. Guidance is provided on (1) the identity of the relevant repository environmental parameters (groundwater characteristics, temperature, radiation, and pressure), (2) the models/methodologies employed to determine the parameters, and (3) the input data base for the models/methodologies. Supporting studies included are (1) an analysis of potential waste package failure modes leading to identification of the relevant repository environmental parameters, (2) an evaluation of the credible range of the repository environmental parameters, and (3) a summary of the review of existing models/methodologies currently employed in determining repository environmental parameters relevant to waste package performance.

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