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MEMORANDUM FOR: John B. Martin, Director
Division of Waste Management

FROM: Michael J. Bell, Chief
High-Level Waste Licensing
Management Branch
Division of Waste Management

SUBJECT: 1982 OPERATING PLAN PRODUCT; DRAFT TECHNICAL POSITION ON WASTE PACKAGE PERFORMANCE AFTER REPOSITORY CLOSURE; COMPLETION OF

Attached hereto is the completed Draft Technical Position on Waste Package Performance After Repository Closure, milestone 312312D due September 30, 1882.

Original Signed by
MICHAEL J. BELL

Michael J. Bell, Chief
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Management Branch
Division of Waste Management

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DRAFT STAFF TECHNICAL POSITION
SUBTASK 1.1: WASTE PACKAGE PERFORMANCE AFTER REPOSITORY CLOSURE

M. S. Davis and D. G. Schweitzer

DATE PUBLISHED -

Prepared by the Nuclear Waste Management Division
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NOTICE

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ABSTRACT

This document provides guidance to the DOE on the issues and information necessary for the NRC to evaluate waste package performance after repository closure. Minimal performance objectives of the waste package are required by proposed 10 CFR 60. This preliminary DSTP describes the various options available to the DOE for compliance and discusses advantages and disadvantages of various choices. Examples are discussed dealing with demonstrability, predictability and reasonable assurance. The types of testing, modeling and statistical analyses that can be used to demonstrate performance are considered. The document summarizes presently identified high priority issues needed to evaluate waste package performance after repository closure.

DSTP on Waste Package Performance After Repository Closure

EXECUTIVE SUMMARY

The primary objective of this Draft Staff Technical Position is to offer guidance to the DOE on the major issues associated with developing a waste package that can be demonstrated to comply, with reasonable assurance, with the performance objectives given in the Code of Federal Regulations proposed Part 60.111. This Position outlines the major issues and problems associated with evaluating the performance of candidate components and package design options. It is based on current knowledge and as such should be viewed as an evolving document.

It is recognized, at this time, that the DOE has many alternatives for developing a waste package that offers reasonable assurance of compliance with the proposed performance objectives. A judgement that a package will in fact provide containment and aid in a controlled release of radionuclides will depend on the quality and quantity of data, test procedures and models submitted during licensing and therefore on the materials and design(s) chosen.

The first part of this Technical Position addresses the options available as well as presently identified major problems associated with demonstrating compliance. It is concluded that a primary issue is to determine the environment experienced by the waste package and how the waste package will interact and alter that environment. This underlying problem affects all aspects of package development from the use of accelerated tests to defining the performance of components and packages to the models used to project the long term performance.

The second half of this Technical Position outlines some of the high priority issues associated with evaluating the performance of candidate package materials. This part of the Position is meant to illustrate the need for early material and design choices and to illustrate some of the concerns surrounding the demonstration of component and package behavior. Several design options are discussed which could, in principle, narrow the scope of work required to demonstrate the ability of a waste package to comply with the performance objectives. These include the use of a discrete backfill, the use of shielding and a low outer container temperature.

1. MAJOR ALTERNATIVES FOR LICENSING

1.1 Introduction

The proposed Code of Federal Regulations (10 CFR 60) on the Disposal of High Level Radioactive Waste in Geologic Repositories[1] requires the licensee to provide the information needed to determine whether a waste package will meet the requirements outlined in Sections 60.111 (Performance Objectives), 60.135 (Waste Package Requirements), and 60.143 (Monitoring and Testing Waste Packages). Section 60.143 is addressed in a separate Staff Technical Position which outlines the type of program required to adequately monitor waste package performance prior to closure. This Staff Technical Position outlines the major issues and problems associated with evaluating the ability of the waste package to comply with the performance objectives and design requirements given in proposed 10 CFR 60.

1.2 Major Alternatives

The proposed requirements on waste package performance after repository closure can be addressed through several major alternatives which allow flexibility in meeting regulatory criteria. The proposed regulatory criteria of 1000-year containment and an annual release rate of less than one part in 10^5 of the inventory present after 1000 years can be met by individual components of the waste package or by the total package.

Thousand-year containment is required of the waste package while the controlled release rate criterion is on the engineered system. The waste package as defined in 10 CFR 60[1] is "the airtight, watertight sealed container which includes the waste form and any ancillary enclosures, including shielding, discrete backfill and overpacks". The engineered system includes the waste packages and the underground facility.

In principle, either or both of the proposed performance objectives can be met by individual component(s) or the whole package as an entity. The components of the waste package can be categorized as: waste form(s), container system, and discrete backfill. The waste form can be simple (e.g., borosilicate glass) or complex (borosilicate glass with a sacrifical layer of non-radioactive glass). The container system is expected to consist of component(s) which provide structural integrity and component(s) which provide corrosion resistance. The corrosion resistance may be achieved by individual or multiple barriers consisting of metallic and non-metallic materials. Similarly, the discrete backfill, which is expected to remain in place, may consist of single or multiple components.

The proposed performance criteria may be met by several components, each of which complies with the criteria, a single component which alone satisfies one or both of the criteria, or partial contributions from all of the components in the waste package. The primary issue to be addressed in considering these alternatives is the assignment of reliability in demonstrating, with reasonable assurance, compliance to the criteria.

2. ACHIEVABILITY, REASONABLE ASSURANCE AND DEMONSTRABILITY

Licensing involves judgements based on definitions and requirements. In a licensing decision, it is important to make definitions and requirements as operational as possible so that subjective decisions are minimized and the uncertainty associated with the decision making process is reduced. The reduction in uncertainty is directly related to the degree with which concepts are made operational since confidence limits in statistics are totally operational. Through operational definitions, decisions can be made on the basis of test results, that is, the reliability and confidence limits are determined from the experimental data.

2.1 Achievability

Achievability denotes the ability of the waste package to comply with proposed regulatory criteria and is based on judgement. The quality of the judgement depends on the level of understanding of the mechanisms involved in the behavior of the waste package, the validity of comparing the behavior to a similar system which already exists, and the ability to predict behavior over extended periods of time.

In assessing the ability of a waste package to comply with the criteria, these approaches play an important role. The need to understand the processes involved in failure or degradation of the waste package is evident. Comparison with the behavior of known systems is important in deciding what short term tests can put conservative limits on long term performance. These concepts are considered when specific examples are discussed in the rationale for acceptance of short term tests used to estimate long term performance.

2.2 Reasonable Assurance

Reasonable assurance is a concept that will be used to determine whether the data, models, and rationale submitted justify the performance claimed. It is expected that the most important use of the concept will be in evaluating the validity of extrapolating short term tests to long term performance. Because of the large number of very different design options available for complying with the proposed criteria of 10 CFR 60, it is not useful to suggest specific statistical methods or confidence limits for analyzing raw data. The use of reasonable assurance in terms of reliability and confidence is discussed in the section on statistics. It is evident that the larger the number of samples measured, the better the statistics and the more the uncertainty in the results can be minimized. The larger the test range in variables around the expected values that are used in tests, the more likely it is that the performance and degradation behavior will be understood. On the most elemental level, reasonable assurance is a judgement, the validity of which depends on the quality of the information submitted.

2.3 Demonstrability

To demonstrate compliance with the performance criteria the quality and quantity of evidence, the test methods used to obtain the evidence, the statistical analysis of the data, the predictive models, and the rationale for the conclusions must be judged acceptable.

When specific waste package designs become available or when choices of materials and the conditions (temperature, water, environment, etc.) under which they are to exist become known, guidance to the applicant can be provided by listing the tests, test procedures, and ranges of acceptable and unacceptable results that should be used in preparing for license application.

Achievability combines elements of demonstrability with methods of prediction and extrapolation. Methods for prediction and extrapolation require an understanding of the processes and mechanisms occurring under realistic and accelerated conditions. Reasonable assurance is the degree of confidence one has that a design will achieve a defined level of performance and that the evidence submitted to support a claim is adequate. The confidence will depend on statistical aspects of the supporting data and/or the level of understanding of the mechanisms involved.

3. LICENSING OPTIONS

3.1 Compliance from a Single Component: Compliance from Several Components Performing Cooperatively

In order to evaluate the performance of a waste package in which several components work together to achieve containment or controlled release, it is necessary to conduct tests and develop data bases on single component behavior, bicomponent behavior and whole package behavior.

If compliance with either criterion is to be demonstrated by the use of a single component, the information required from the licensee is likely to be minimal, well defined, and easier to evaluate. The information required on its behavior, its reliability, and its failure modes may be better defined, more restricted, and the statistical data base required for licensing may be simpler. This alternative also implies that this component, in combination with other non-interacting components which also individually comply with the criteria, clearly forms a demonstration of redundant compliance.

The second alternative, where compliance is achieved by several package components performing cooperatively, requires as a minimum an understanding of the behavior of individual components as well as bicomponent behavior.

3.2 Concurrent and Sequential Behavior

Degradation processes and/or failure modes which occur simultaneously are defined as <u>concurrent</u> while those failure modes which occur at different times, and which depend upon a sequence of events in which components are breached one after another, are defined as <u>sequential</u>.

Two methods for utilizing waste package components are by concurrent single component behavior and/or sequential component behavior. As an example of current single component behavior, assume a controlled release from a given discrete backfill is below 10^{-5}/yr only when the source term (waste form) has a release of $<10^{-3}$/yr. The level of performance assignable to the backfill is then a function of the choice of waste form and that of the waste form depends on the choice of backfill.

An example of performance based on sequential behavior occurs in a design using a series of corrosion resistant overpacks. An assessment of the ability of these overpacks to meet 1000-year containment depends on the time to breach, in sequence, each of the overpacks. If, for example, the outermost overpack fails in 200 years, then a series of four identical non-interacting overpacks may perform adequately for 800 years if one assumes simple sequential behavior.

In the above example, there is an advantage to components made of a single material whose behavior depends upon sequential events. For example, consider a corrosion barrier comprised of four concentric containers constructed from the same material, each container having an average "time to breach" of 200 years. Since the barrier is made of a single material the data base needed to assess the performance and to qualify the lifetime is more restricted and requires shorter test verification times than would be required for the same barrier constructed from different materials or for a barrier in which two or more components may fail simultaneously.

Compliance with the performance objectives by several components may be achieved by a combination of sequential and concurrent behavior. For example, the release rate from the backfill depends not only on the source term (waste form) but also on its volume and the radionuclide inventory. It may take a long time to load the backfill to a level where the release rate reaches its maximum allowed value.

3.3 Preferred Approach

At this time, NRC's preferred approaches for assuring compliance of a waste package with the NRC criteria are, in decreasing order of acceptability:

1. Combinations of independent high integrity components which, by their own behavior, each satisfy the NRC criteria (i.e., redundant compliance).

2. A single component which, by itself, can satisfy the NRC criteria, in combination with other barriers that may not individually meet these criteria (single compliance).

3. Combinations of components that cooperatively comply but individually do not completely satisfy the proposed NRC criteria. These components acting together can be assigned, with some level of assurance, credit for complying with the performance objectives (composite compliance). The package constructed from these components should satisfy 1000-year containment.

Achieving compliance through behavior based on sequential rather than concurrent events has a distinct advantage. Multicomponent barriers constructed from similar materials such as a container system of identical canisters may also be more advantageous than ones constructed from dissimilar materials. In this instance the data, model etc. required for licensing may be more restricted.

While redundant compliance is a preferred approach to insure the conformance of the waste package with the performance objectives in proposed 10 CFR 60 (60.111), the rule is structured to give the licensee maximum flexibility in demonstrating compliance. It is the licensee's responsibility to submit for evaluation a convincing data base, analyses and rationale to support the particular performance claimed.

4. MAJOR ISSUES ASSOCIATED WITH DEMONSTRATING COMPLIANCE: MINIMIZATION OF UNCERTAINTIES

Since the proposed regulation 10 CFR 60 requires performance objectives on the overall waste package and on the engineered system, there are many means by which long term containment and controlled release can be attempted. There are, nevertheless, major issues independent of material choices, design choices and repository site properties that address the problem of reducing uncertainties in the waste package performance after closure. These are noted here and are discussed in detail in following sections.

1. Repository/Groundwater Characterization

   Any uncertainties in the repository water properties (composition, temperature, pH, Eh, etc.) will be magnified as uncertainties in corrosion rates, leach rates and backfill properties. Corrosion rates, leach rates and backfill retardation, each depend in complex manners on water composition, temperature, pH and Eh. It is obvious that the better these parameters are known the less will be the uncertainties in understanding the other phenomena.

2. Temperature

   In general, the spread and uncertainties in chemical kinetic reactions increases with increasing temperature. From this point of view keeping the surface temperature of the package below 100°C will tend to minimize uncertainties in all other reactions (corrosion, leaching, ion-exchange) involving package performance.

3. Accelerated Testing - Predictability

As discussed later, existing theory would favor isothermal acceleration techniques when possible, although the potential for using elevated temperatures for accelerating kinetic reactions is recognized. The problems associated with this means of attempting predictability are discussed in Section 5.

4. Radiation Effects

Radiation can alter waste package performance in two general ways. Radiation effects can change the structure and chemical reactivity of the waste forms and backfill, and it can by radiolysis change the composition, pH and Eh of the groundwater. In shielded packages the reduction of radiation effects should make major contributions to reducing the uncertainties in many of the above areas.

5. Total Package Testing

The uncertainties associated with specific component performance cannot, in general, be used to predict uncertainties in total package performance. The more extensive and realistic are the test programs for total packages, the more the uncertainties in performance are reduced.

6. Statistics

The general means of quantifying uncertainties and evaluating them, is through the use of statistics. Although statistical analyses are expected to play an important role in demonstrating compliance, the specific types of statistics used will depend on what part or parts of the waste package are used to achieve containment and controlled release. Some examples are considered in the section on statistics.

7. Modeling

Modeling will be an important portion of the information submitted to demonstrate compliance. Appropriate modeling can deal with quantitative attempts at predictability and the uncertainties in predictability and should be in the form of theoretically and empirically based equations dealing with a realistic range of site and package parameters. Minimization in the uncertainties associated with modeling can be attained by the methods recommended in Section 9.

5. ACCELERATED TESTING AND PREDICTABILITY

One of the major issues to be resolved is the use of short term (accelerated) tests for predicting the long term behavior of man made barrier materials.

Theoretical justification for predicting the long-term performance of a material from shorter term experiments requires that the mechanisms by which the material is degraded remain constant over the time required for the prediction. Furthermore, the mechanism must be experimentally validated by the determination of an explicit, isothermal rate expression which accounts for the correct functional dependence on all the parameters involved.

For example, to have complete assurance that a corrosion barrier will last 1000 years the mechanisms of corrosion leading to failure of the barrier should be determined and validated by explicit rate expressions. The following section illustrates the problem by considering the question of predicting corrosion behavior over extended periods of time.

5.1 Isothermal Predictions

Corrosion is a kinetic, non-equilibrium process. The rate and mechanisms of corrosion are dependent upon, among other parameters, the reactants and products of the various corrosion processes. The initial problem then is to determine what the reactions are, the stoichiometries, and if there is more than one, whether the reactions are simultaneous, sequential, catalytic, or inhibiting. One of the major obstacles in determining corrosion mechanisms results from observations that rates of corrosion can be seriously altered by the corrosion products and how they are distributed or removed from the corroding surfaces.

In general, there are more requirements for heterogeneous systems than for homogeneous systems on the use of isothermal rates to predict for times much longer than the time over which the rates were measured. Homogeneous kinetic systems tend toward equilibrium states that are usually well defined. If the isothermal rate expression is rigorously correct and truly represents the mechanisms of the reaction, it will include all the chemical, physical and geometric factors that are known to alter the rate. For homogeneous systems this can be tested by using the known equilibrium values and showing that the rate goes to zero.

This test is generally not feasible for complicated heterogeneous reactions such as solids corroding in liquids. Here too, it is mandatory in determining the mechanism to show that the isothermal rate expression includes the correct functional dependence of the corrosion rate on all the chemical and physical variables known to affect the rate of corrosion. For heterogeneous reactions, however, it is also necessary to prove that neither the metal nor the metal-solution interface undergoes any structural, physical, or chemical change that can alter the corrosion rate over the total time for which the prediction is intended.

For metals such changes might include:

- isothermal annealing

- formation and subsequent breakaway of a surface film

- stresses developed or removed by formation or cracking of a corrosion product or film
- diffusion of corrosion products into or out of the metal surface
- diffusion of bulk components into surface depleted zones
- initial selective attack at a site that eventually is depleted
- grain boundary precipitation, etc.

Any or all of these might lead to a change in the corrosion rate with time. In general, the overall corrosion rate of most metals represents contributions from several different mechanisms with different temperature dependencies. These can interact with each other sequentially. For example, consider a corrosion process where an oxide layer is formed by one component in an alloy, eventually builds up, spalls, and continues to form depleting the zone beneath the surface. Over periods of time long compared to the time necessary to form the film, the component will diffuse from the bulk to the surface depleted zone. If this component is in equilibrium with its own carbide for example, depletion of the bulk concentration by formation of a surface oxide will eventually cause the carbide to decompose increasing the carbon activity of the alloy. This, in turn, could lead to formation of a new carbide with a different component of the alloy so that short term corrosion rates will not represent the corrosion rates that would occur over longer times. Even in the case of uniform corrosion, it is often not safe to extrapolate a reported rate to times of exposure far exceeding the test period.

Altering concentrations of reactants and products to test if a single rate expression accounts for the observed corrosion behavior may be more significant for uniform corrosion than for pitting corrosion. Nevertheless, it is, even for uniform corrosion, a necessary but not sufficient condition for justifying time predictions.

In the cases of pitting corrosion, the basic properties of the pitting phenomena indicate that several mechanisms operate simultaneously. For a single mechanism, pits should occur uniformly in space, proceed with the same depth dependence on time, and develop the same shapes.

In real systems, pitting generally occurs at structural irregularities or chemical inhomogeneities that vary from sample to sample. Measurements within the pits, of solution compositions and corrosion product concentrations, show that each pit can be a different chemical system with different mechanisms operating at different times. This is also supported by the wide variability in induction periods, the variability in rates of penetration, and the variabilities in shape development and pit morphology.

5.2 Temperature Acceleration

The use of elevated temperatures to increase corrosion rates for the purpose of predicting long term corrosion behavior at the lower temperature can be theoretically unjustified and technically unsound. The effects of changing temperatures in chemical kinetics are covered by the Arrhenius concept which is valid only if it is assumed that the same mechanism occurs over the temperature range studied. If this assumption happens to hold, the ratio of the rates at different temperatures can be used to obtain the numerical value of the activation energy. This numerical value can then be used to speculate on what processes are involved in delaying the reactants from forming the thermodynamically more stable products instantaneously. If several rate measurements are made at several temperatures and if they are precise enough to demonstrate a single numerical value for the activation energy, this type of result can be used to claim that the data are consistent with the assumption that the mechanism did not change over the temperature range.

Rigorously, however, these measurements do not prove the existence of a single mechanism. Thermal barriers (activation energies) for different mechanisms can have similar values. An additional necessary test to verify a constant mechanism is to demonstrate that the individual rate constants at all temperatures are identical in functional form with respect to all the variables involved in the corrosion process. This requirement implies that temperature acceleration is subject to all the problems and uncertainties of isothermal prediction techniques in addition to many complications that can be introduced by the temperature variation itself. It should be clear that a temperature change that causes a phase change in any reactant (i.e., water going to steam) has the potential to drastically alter the corrosion process. If the overall corrosion rate is the result of several mechanisms with different activation energies, changing the temperature, in principle, will change the mechanism and will invalidate any use of the high temperature rate in predicting long term behavior of the low temperature corrosion process.

This discussion also can be extended to the long-term behavior of, for example, borosilicate glass. The glass waste is a dynamic, nonequilibrium chemical system. Fission product decay results in a thermal and a radiation flux which may cause changes in valence states of species present in glass, alteration of glass properties by the buildup of decay products with different ionic radii, different valence states, and different chemical properties. Since the decay is a function of time, the glass at any one point in time is a unique chemical system in which the mechanism(s) of leaching may be different from that at any other point in time.

Furthermore, from data presently available on the leaching of glass, it is increasingly apparent that the ability to determine a rigorous mechanism and rate expression for the leaching process is severely limited by the observation that leaching is a sensitive function of:

- the chemical properties of the matrix and the incorporated waste
- the environment, including temperature, pH, Eh, ionic composition, and flow rate of the leaching medium
- the physical characteristics of the waste form (e.g., exposed surface area, phase separation, and degree of devitrification).

There is no rigorous equation which accurately predicts the leaching of glass under all pertinent conditions at any single temperature. The proposal to increase temperature to accelerate the leaching process and subsequently predict the long term leach rate at lower temperatures is subject to all the problems associated with the use of temperature acceleration to study corrosion phenomena.

5.3 Conclusions

The only rigorous means of predicting long term behavior is through a validated isothermal rate expression, one obtained for all pertinent environmental and material parameters and at each temperature expected during the time required for the prediction. It is recognized that the probability of acquiring such data is small and may be an unreasonable requirement. However, the use of temperature to accelerate a degradation process and determine average time to failure should be viewed with caution. It is expected that the applicant will have demonstrated, at least empirically, the average behavior of component(s) under a range of conditions (e.g. reactants) and over the temperature range expected. This data in the form of a range of average corrosion rates or average leach rates should be accompanied by a description of the test conditions (e.g. range of temperatures, time of observation, range of reactants, etc.) and statistical spread in the data, a rationale for the way in which the tests were conducted and the data analyzed and a model predicting the long term performance. It is understood that the amount of data, the quality of the data, the analysis accompanying the data, the rationale and the models used will be dependent on the design, materials choice, degradation/failure modes and on the level of performance assumed to be assignable to the component.

6. TOTAL WASTE PACKAGE TESTING

In principle, a waste package will be designed for containment for about the first 1000 years after emplacement. It will be a multicomponent system designed to provide reasonable assurance of compliance with this criterion. If it has been properly designed, then there are two generic tests which can be required for demonstrating compliance, null tests and tests which realistically simulate a breached package.

6.1 Null Tests

A null test would require a reproducible demonstration of non-detectable release of radionuclides under repository conditions expected during the containment period. These test conditions should include a range in variables such as temperature, radiation levels, pressure, groundwater chemistry, etc. Null tests would also include a range of expected events and processes such as partial and full saturation and wet and dry cycling.

6.2 Degradation/Failure

It is useful to distinguish between degradation and failure. Degradation is an impairment of a property by chemical or physical processes. It does not necessarily denote failure. [illegible] formation degrades a corrosion barrier, but failure of that barrier does not occur until the pit completely penetrates the container exposing the next barrier to the environment. Hydrogen embrittlement may degrade the barrier and result in failure by enhancing another degradation mode, e.g., crack propagation. (The metal may be extremely brittle, but as long as the embrittled and degraded container does not expose the next barrier, it has not failed.)

6.3 Simulated Failed Packages

Null tests demonstrate the lack of radionuclide release under expected repository conditions and therefore demonstrate the potential for a waste package to meet 1000-year containment. Since the consequences of expected failure modes bear directly on the degree of achievability and reasonable assurance associated with a waste package design, these can be addressed by reproducibly demonstrating the behavior of a failed waste package. A failed package should be studied under the same conditions required for a null test. Moreover, the simulated failure should realistically reproduce the most probable failure modes of the specific design.

Failure of a waste package is defined with respect to the NRC criteria. For 1000-year containment, a failed package occurs when the first radionuclides are released from the last engineered boundary (e.g., discrete backfill) and are detectable by state-of-the-art techniques. Failure of a package component occurs when, through some process or mechanism, the designed function of that component is compromised. For example, a failed container system may be one in which the barrier has been completely breached by pitting and the next component is then exposed to the repository water. A failed component does not necessarily imply a failed package. If the next component were another corrosion resistant container, then the waste form would not be exposed to the leaching medium and radionuclides would not migrate to the outermost boundary. Even if the next component were the waste form, failure of the container and subsequent leaching of the waste form does not constitute failure of the package until some level of radionuclides is detectable at the outermost boundary of the waste package.

6.4 Realistic Failed Package Tests

Realistic failed package tests would fall into two categories: (1) tests on packages where single barrier(s) have failed, and (2) tests where the package itself has failed. The aim of the first type of test is to confirm package behavior under conditions where one or more of the barriers have failed. For example, if a package contains a single corrosion resistant barrier whose only failure mode is through pitting, then the first type of test would artificially breach the container with "average" pits. The outermost boundary would then be studied to determine if and when release is observed under typical repository conditions including wet and dry cycles. The aim is to determine when and if a failed component leads to a failed package. These types of studies by their very complexity should be done with specific package designs in which the components have been selected after extensive work to define their behavior and interactions with other candidate components. The objective of such tests would be to define which barrier failures or combination of barrier failures constitute a failed package, the consequences of specific barrier failure on the package performance and to obtain information in support of models that may be used to predict waste package performance.

The next stage in "realistic" package tests would be to look at the effects of a failed package in terms of release to the engineered system. This type of test would follow logically from barrier failure tests and help determine the ability of the waste package to contribute to the controlled release rate criterion and aid in modeling the migration of radionuclides.

These types of failed package tests involve such complex interplay between the various components of the waste package and the interaction (or additivity) of component behavior that the information obtained will be primarily qualitative in nature. It is highly unlikely that rigorous equations describing the behavior of a failed component in a failed or degraded package can be written to predict the behavior over long periods of time. The information obtained from these tests will demonstrate how a failed package releases radionuclides or how failed components affect the behavior of the total package.

6.5 Conclusions

It is expected that whole package tests will be primarily qualitative in nature and design specific. The choice of components and design should be based on data on single component and as a minimum bicomponent behavior. The package submitted for licensing must have strong supporting data to qualify its behavior with respect to compliance with the performance objectives. The types of data, tests, models, etc. that should be submitted will depend on the alternative(s) chosen by the licensee to demonstrate compliance with the criteria.

7. REPOSITORY CONDITIONS

One of the issues pertinent to all aspects of demonstrating compliance with the NRC performance objectives is a definition of the environment experienced by the waste package as well as the effects of the waste package on its environment.

The waste package must function to contain radionuclides for about the first 1000 years in the dynamic environment of the repository. It is anticipated that at some time after the first 1000 years, when the waste form will be exposed to the environment, the release of radionuclides from the waste package to the environment will be controlled by transport in the ground or repository water. In order to qualify waste package components or the waste package for complying with one or both of the performance objectives, the behavior of the components/package in the range of expected repository conditions should be known.

7.1 Pre-Emplacement/Pre-Closure Conditions

Prior to licensing a waste package and a repository, the applicant will have conducted extensive site characterization studies. These studies may include information on:

- the "average" water chemistry at the location expected for the repository
- the "average" water flow rates, or a model used to estimate water flow rates from site specific information
- ambient temperature
- mechanical properties of the host rock.

The mechanical properties of the host rock are important in determining if, for example, the waste package will experience excessive forces that will damage or destroy the package. The temperature profile, flow characteristics, and composition of the groundwater is important information needed to determine the behavior of the waste package under conditions which lead to failure by corrosion or leaching.

7.2 Post-Closure

The primary problem in determining the ability of the waste package/or package components to meet the 1000-year containment criterion and to contribute to a controlled release of radionuclides from the engineered system, is demonstrating the behavior of components alone and in combination (as a package) under a range of repository conditions spanning thousands of years. While models exist which, based on a given waste loading, repository design, thermal conductivity, etc., allow one to calculate the thermal history of packages and the repository,[2] no such model exists for predicting the complex chemical reactions which will occur when a thermally hot, radioactive waste package comprised of many different materials is exposed to "typical" ground or repository water. It is movement of the water to and from the waste package that will result in breach of containment and the release of radionuclides. The ground/repository water is another example of a dynamic chemical system. For example, one way to visualize potential changes is to follow the pathway of typical groundwater from the outermost boundary of the waste package to the source term (waste form) and then out to the outermost boundary. The groundwater will reach the outermost boundary of the waste package with a chemical (ionic, pH, Eh, etc.) composition that is determined to a major extent by the host media, and the temperature. Typical groundwater chemistries have been determined for salt, basalt, shale, and tuffs, but their composition as a function of the possible thermal history have not been extensively evaluated. Increases in temperature may:

- alter solubilities of many species including dissolved gases
- result in reactions of species within the groundwater as well as with host rock or other materials in the host rock
- result in changes not only in ionic composition, but in pH and Eh as well.

Therefore, without adding any possible effects from radiation that may be present if a package contains no shielding or enough discrete backfill to act as shielding, the thermal effect alone may alter the character of the groundwater at the outermost boundary of the waste package as a function of time. When this time/temperature dependent groundwater reaches the waste package, two generic cases are possible: a shielded package with or without discrete backfill, and an unshielded package with or without discrete backfill.

7.2.1 Shielded Package: No Discrete Backfill

In this instance the groundwater will directly contact a metallic container/overpack. The effects of radiation on the groundwater chemistry and the corrosion processes may depend on the residual dose rate and the total dose, the rate at which the water is removed and replenished, and the depth of corrosion (e.g., hot spots) etc. The chemistry of the water will be in a state of flux as the temperature changes, the outer containers corrode and corrosion products build up in the water or react with species in the water as the depth of penetration increases. When the container is finally breached and the waste form is exposed to the "leaching medium," the problems increase. The time at which the waste form is leached will determine whether fission products are predominantly released, actinides are predominantly released or a combination of fission products and actinides are predominantly released. The chemistry, flow rate, and temperature of the leaching medium will affect the release of radionuclides. The composition of the matrix and waste as well as its physical properties will also determine what is "dissolved" in the groundwater. If transport is slow to the outermost boundary of the package, the composition may again be altered by passage through residual metallic materials, changes in temperature, and exposure to radiation.

7.2.2 Shielded Package: With Discrete Backfill

The addition of a discrete backfill to the waste package will affect:

- the time to breach the integrity of the metallic overpack
- temperature profile with time
- the character of the groundwater at the outermost boundary of the waste package compared with that at the interface of the discrete backfill and the next package component
- the flow rate of the water.

The effect of the backfill on the temperature of the groundwater would be to alter in time the physical and the chemical (e.g., reaction, pH, Eh) properties of the groundwater. In other words, if these could be determined for a shielded package with no backfill, they may no longer be applicable to a shielded package containing a backfill with different thermal characteristics and the ability to interact with the groundwater. Those species and products present at the metallic boundary in shielded packages with no backfill need not be the same products present at the interface of the backfill and host rock or the backfill and the next package component.

The backfill will presumably serve in some capacity to limit both the flow of the incoming water as well as filter or condition the water prior to its contact with the container system. The composition of the groundwater contacting the backfill may change because of thermal perturbations of the waste package.

Once the groundwater has passed through the backfill, all the complications cited above (Section 7.2.1) apply to determining the character of the water interacting with the metallic components and ultimately leaching the waste form.

7.2.3 Unshielded Package: With and Without Discrete Backfill

It is obvious that waste packages containing no shielding add yet another level of complication to specifying repository/groundwater conditions as a function of time. Radiation will result in radiolysis of the groundwater. The thermal effects may enhance recombination of radiolysis products, foster formation of new species by reaction of radiolysis products with chemicals present in the groundwater, at the host rock, or backfill surfaces. Radiation has the potential to alter the character of the backfill or host rock (e.g. accumulation of nascent sodium in salt)[2] and in so doing alter the character of the groundwater and its subsequent effects on corrosion and leaching.

7.3 Conclusions

While some existing models explain, for example, the thermal history of the repository, no model exists to predict the changes in groundwater with time under the influence of a waste package and the perturbations it will cause, or the effects of changes in the groundwater on the integrity of the waste package components and release of radionuclides. Furthermore, it should be apparent that if these changes could be determined they would be sensitive functions of repository design, package material choices and package design.

While it is highly improbable, that a reliable model will be developed to predict the complex conditions that will occur in a repository over long periods of time, there are alternatives available for gaining an understanding of how the repository affects a waste packages over a period of time.

It is assumed that the applicant will have characterized the ground or repository water during the site characterization. Using this "average groundwater composition" the effects of temperature and radiation on the groundwater could be elucidated. Species detrimental to the performance of the container system, could be identified and included in testing programs to qualify the performance of waste package materials. The design of the waste package will to some extent also determine the range of conditions which should be tested for demonstrating the behavior of the waste package. For example, a package design containing no shielding, or one which results in a large thermal perturbation will require a wider range of test conditions and thus a more comprehensive determination of the effects of radiation and temperature on the ground/repository water.

8. APPLICATION OF STATISTICS

A demonstration of compliance with the NRC's proposed performance objectives, for the reasons discussed, may not be through the use of rigorous predictive equations. Rather, the most likely type of data and arguments submitted will involve a statement of probability, confidence, and uncertainty associated with the data bases on single components, bicomponents, and packages. In addition, the types of statistical arguments that may be submitted for licensing can be component dependent, design related and may be tied to a cost-benefit analysis. For the purposes of this discussion, it will be assumed that from site characterization reports the applicant will attempt to identify those aspects of repository conditions which would detrimentally affect the waste package and that components chosen will have been studied within this range of conditions.

8.1 Component Dependence

The type of statistical arguments acceptable for licensing are dependent on the material (component) choice and the type of degradation processes which lead to failure of that component. For example, if a material fails under a range of repository conditions in a relatively short period of time, it is possible in principle to test many samples and establish a statistically sound data base to justify an "average time to failure." If a component is subject to only a few failure modes, the number of tests required to statistically verify its performance will be smaller than that required for a component subject to many failure modes.

Components whose failure/degradation rates are very slow can be treated in two different fashions. If during the time of measurement, failure/degradation rates are measurable then a spread in the rate and an average rate for time to failure can be predicted. If, however, the rate is not measurable during the time of observation, then only an estimate of the minimum time to failure can be made.

8.2 Design Dependence

The type of statistical arguments presented for licensing may be design dependent. The quantity and quality of the statistical data would depend upon whether the applicant claims full compliance, partial compliance, or no compliance with the performance objectives. In addition, if the applicant does claim compliance, either partial or full, the types of arguments acceptable will depend on whether the performance claimed is based on sequential, concurrent or a combination of sequential and concurrent behavior of the component(s).

If, for example, the applicant indicates no compliance from one or more components, the data base and accompanying statistical analysis may be limited to demonstrating that this component(s) does not seriously alter the behavior of the component(s) for which the applicant does wish credit for demonstrating compliance (i.e., the applicant must demonstrate by an appropriate analysis that that component does not adversely affect performance).

If the applicant wishes to demonstrate full compliance with the performance objectives by a single component or several components, then the data base and accompanying statistical analysis will be dependent on the material choice and should also include a demonstration that as a minimum, the nearest neighbor components do not detrimentally alter performance. Furthermore, the statistical arguments presented for a licensing decision will also be dependent on which criteria the applicant is trying to comply with. For containment, one is essentially trying to demonstrate the absence of an event over some period of time. In controlled release, one is trying to demonstrate that the behavior (release) occurs within a defined range over some period of time. The length of time is determined by whether the applicant claims complete or partial compliance. In a situation where the applicant wishes to demonstrate partial compliance with the criteria, the statistical data base will be a function of the criteria for which the applicant desires credit, the time over which credit is to be assigned, and the behavior on which credit might be assigned. In claiming partial compliance, the applicant may claim that:

1. The data justify performance for a time shorter than the criteria require, i.e., the statistical spread in the data for time to failure only allows a prediction for a shorter period of time.

2. It is cheaper and more convenient for partial compliance or technically not feasible to demonstrate full compliance, i.e., there may be some advantage in employing components whose individual behavior does not completely satisfy one or both of the performance objectives.

Again, the data base and statistical justification required for licensing depends on the component (modes of failure), the criteria for which partial compliance is demonstrated, the time for which partial compliance is demonstrated, the absence of adverse affects from other components, and the way in which the component behaves. For components of the same material which behave in a sequential manner, the data base and statistical analysis may be more limited than that required from components which behave in a concurrent fashion. For example, in a corrosion barrier constructed from a series of containers of the same material designed to fail in sequence, it may only be necessary to determine average time to failure for the first container which is exposed to the environment and a demonstration that the average time to fail of the following containers are no greater than that of the outermost container. If the same barrier were constructed from dissimilar materials, it would be necessary to verify the time to failure for each container in the sequence. If a barrier relied on the additive performance of components, then the average behavior of each must be known as well as the average combined behavior. The data base and statistical analysis could in principle be more extensive and require a large number of samples to quantify.

8.3 Cost-Benefit Dependence

It is possible that in the process of developing packages for licensing, the acceptability of packages/components will be tied to a cost benefit analysis. For example, it may be argued that in order to build a statistically sound data base for licensing components and/or packages the cost of R and D exceeds the additional benefit derived from conducting such a program. The decision as to what constitutes a sound analysis on which to make a licensing decision will depend on an evolving decision by the NRC of what will constitute assurance of the public's health and safety. In all instances however, the applicant should submit a data base, statistical analysis, rationale for the types of analysis performed, a predictive model and the thorough cost benefit analysis justifying the decision not to proceed with further testing.

8.4 Conclusions

A statistical analysis should be provided for empirical evidence submitted to demonstrate the performance of the waste package. The types of analyses and the rationale for employing a specific analysis will be component dependent, design related, and probably accompanied by a cost benefit analysis. The information that will be acceptable for licensing will depend on the component (failure modes), the criteria being addressed, the length of time for which compliance is being demonstrated, and the behavior for which compliance is demonstrated. In all instances, an important concern is based on the clear distinction between the mathematical theory of probability and its application to a real problem. Mathematical probabilities begin with a set of assumptions. If the assumptions are correct, then the results follow. Determination of whether the assumptions are correct is assured by experiments and an understanding of the processes involved. Thus, separating waste packages into the categories of failed and not failed and using binomial statistics may not adequately describe the occurrence of various states of degradation and partial compliance that are expected when large numbers of containers are emplaced in repositories.

9. MODELING

The applicant should submit models that predict how a package will perform over time in the repository environment and the reliability of the prediction. The description of the model should include the data on which the model is based, the rationale for the model, the procedure used to validate the model, and the reliability of the model. A report, "Draft Technical Position on Documentation of Models" (NUREG-0856) exists. Where applicable, the applicant should attempt to follow these guidelines as closely as possible.

9.1 Design Considerations: Predictive Equations

The model(s) will contain mathematical statements which predict the performance of package components. The model should be designed so that:

- all pertinent degradation/failure modes have been included
- the mathematical statements used to describe the failure/degradation are valid
- the range of input parameters is adequate and applicable.

This implies that isothermal equations should be developed for the range of conditions expected and the temperature range of interest. That is, there exists a series of equations:

$$\text{rate } (T_1) = f\ (a_1, b_1, c_1, \ldots)$$

where a_1, b_1, c_1 are the pertinent variables (e.g., reactant concentrations, geometric factors) which affect the rate of a given degradation process and T_1 is the temperature at which the rate is determined.

If such a set of rate expressions cannot be developed, empirical statements which describe the observed behavior under a specified range of conditions should be used. For example, it may be observed that within a range of environmental conditions, the rate of uniform corrosion, measured as weight loss or weight gain, can be expressed as:

$$\frac{dw}{dt} = a\sqrt{t} + bt + \ldots$$

These mathematical statements would not be predictive equations, but would be empirical statements of the corrosion behavior of a container or container system. Similar empirical statements may be used to describe the leach behavior of the waste form, and the sorptive behavior of the backfill. These equations may have been developed from data obtained on the behavior of single components, on the behavior of combinations of components and on the behavior of "simulated failed packages" under a range of expected repository conditions. In some instances, where the degradation mode may be a stochastic (random) process such as pitting, approximations may have to be employed in which the induction period is estimated from experimental data (which may include accelerated tests) and the propagation rate is estimated from the extreme or deepest pits observed. Therefore, evaluation of the model requires an assessment of the completeness of the data base used to generate an empirical set of equations, the design of the waste package being modeled as well as the option(s) under which the DOE is applying for a license. For example, a waste package consisting only of a waste form and container, will be modeled differently from one which consists of a waste form, shielding, container and discrete backfill. A package in which performance is based on the sequential behavior of package components will be modeled differently from one in which performance is based on the concurrent behavior of components. The model of a package in which corrosion occurs by only one or two mechanisms which have very low scatter in the input (corrosion rates) parameters will be easier to assess than one in which the container material is subject to many corrosion processes whose combined effects may not be well understood.

9.2 Coding Considerations

The applicant should state whether an empirical set of equations has been modified or deleted to make a calculation easier and should demonstrate that such a modification or deletion is justified. Thus, the applicant should show that models "designed" to mimic the behavior of a physical system do not introduce nonrealistic aspects when finally put together or constructed.

9.3 Validation/Verification

It is necessary that the applicant demonstrate that the code or model has been verified. There are three generic ways to do this:

1. Validation by comparison of calculated results with experimentally observed results

2. Validation by comparison with other models (consistency)

3. Validation by modeling an accepted (standard) problem.

The most acceptable means of verifying a model would be by comparison of calculated results with experimental results. Since the waste package is in principle a multicomponent system, this may not be feasible. As an alternative, it would be necessary to validate sections of a model by comparison with experimental results. For example, a section of a model which calculates the corrosion behavior of a container could be verified by comparing with experimental data. This would be necessary particularly if simplifying assumptions were made in coding the model.

9.4 Conclusions

The applicant should assess the model and the results obtained from it. A model in which the degradation and predicted failure rates are based mostly on theoretically rigorous and experimentally verified rate equations will have a sounder base for predicting the long term performance of components and packages. However, when this type of information cannot be developed, the applicant should submit other types of evidence justifying the validity of the simplifying assumptions.

The applicant should assess the reliability of the model because the model will be used to assess the reliability of the waste package. These issues will be addressed in more detail in the Draft Staff Technical Position on Quality Assurance, which will contain a section on Reliability.

10. HIGH PRIORITY ISSUES ASSOCIATED WITH EVALUATING THE PERFORMANCE OF CANDIDATE WASTE PACKAGE MATERIAL

10.1 Introduction

This section of the Branch Technical Position enumerates a limited number of issues which should be resolved if the NRC is to evaluate the performance of a high level waste package. There are design options which may facilitate the evaluation of a waste package and require a minimal amount of research and development to demonstrate compliance with reasonable assurance. One such design option is a waste package that includes, along with a waste form and container system, a discrete backfill. The term discrete backfill denotes any backfill other than crushed host rock that is emplaced as part of the waste package so as to contribute directly or indirectly to the performance of the package.

In reviewing the performance of individual components and the package in toto it is also concluded that restricting water flow around the waste package offers many advantages to favorable performance with no significant disadvantages. In the discussions that follow it is assumed that all packages will be emplaced with some backfill to at least restrict water flow around the container system and therefore around the waste form when the container system is breached. This is compatible with views in the DOE community which would make their solubility limited degradation models and the MCC leach tests more realistic.

The following discussion and Tables 1-7 are based on what is presently known of the performance of the materials chosen as examples. These examples are a waste form of borosilicate glass, an overpack or container of TiCode-12, a sacrificial container such as cast steel and discrete backfills of either sand-bentonite or synthetic zeolites/titanates. Where possible optimum design alternatives are discussed.

In all instances, it has been assumed that reliable, rigorous predictive equations may not be developed. It is, however, expected that programs to address the basic properties of leaching, corrosion and backfill behavior will develop a statistically significant data base from which an evaluation of the waste package materials can be made.

10.2 Major Issue: Typical Repository Conditions

10.2.1 Introduction

The major factor in determining how the waste form, the container system, the backfill(s) and the total package will behave is the nature of the water environment and how it is affected by the package and, in turn, affects the package.

10.2.2 Groundwater as Repository Water

Groundwater and repository water are defined as two potentially different systems. Groundwater is used to denote the water present at the site and at the depth of the repository. Repository water is the groundwater after it has been altered by the engineered components such as backfill and corrosion products and which has been exposed to thermal and radiation effects (i.e. in the absence of shielding). The differences then between ground and repository water and the effect this has on the package will be greatly affected by the package design. For example, an unshielded commercial high level waste package may subject the adjacent host rock to a total dose of 10^{10} to 5×10^{10} rads[3] during decay of the fission products. When salt at 115 to 170°C experiences a radiation dose of 2×10^{10} rads the amount of colloidal sodium formed may be between 3 and 50%.[1] For very large variations in the brine content of the salt, this amount of colloidal sodium in contact with the brine will result in a solution with a pH of about 14. There is essentially no work available on the performance of backfill material, metallic container material or waste forms in solutions with a high pH. The information that does exist indicates that:

- The backfill materials such as bentonite may dissolve.
- For materials such as Ti alloys, hydrogen pickup is accelerated in a basic medium.[4]
- The rate of matrix dissolution in glass can increase by as much as three orders of magnitude.[5]
- There is no data base for corrosion of metals considered for waste canisters in strong NaOH solutions.

In other repositories the major failure modes of TiCode-12 are associated with hydrogen pickup. In order to evaluate long term performance of TiCode-12 in an unshielded package a great deal of R and D on radiolysis of typical groundwaters and threshold effects on detrimental hydrogen absorption will be required that would not be necessary for a shielded package.

10.2.3 Conclusions

The complexity of the mechanisms of leaching and corrosion indicates that complete understanding of the factors involved probably will not be achievable in the times necessary to license a repository. A more reasonable approach to understanding and predicting the performance of a waste package is to limit the R and D to the range of variables that will occur in a given situation.

As a minimum, in the absence of a package design, studies should be undertaken to determine the effects of radiation and temperature on typical groundwaters equilibrated with the host rock media. These studies should help define the range of conditions necessary for studying the leaching of the waste form, the corrosion of the container and the properties of backfill materials. These studies should be augmented by a program to determine how package components will alter the groundwater. Again, it should be apparent that an early package design could significantly alter the amount of work required to determine the environment experienced by the waste package.

In the discussions that follow, typical repository water indicates that the medium used for leaching, corrosion studies and backfill studies is as a minimum, groundwater equilibrated with host rock.

10.3 Major Issue: Waste Form[5] Matrix Dissolution

The long term leach behavior of borosilicate glass is likely to be determined by the matrix dissolution rates. For monovalent sodium in Pyrex borosilicate glass, one of the most mobile cationic species, at about 400°C the diffusion coefficient would result in a movement of 1 to 2 cm over 1000 years. Higher valence state species are expected to diffuse even more slowly. Radionuclide specific leach rates and some surface phenomena will determine only short term leach behavior. It is, therefore, recommended that matrix dissolution rates and the uncertainties in those rates be determined under typical repository conditions.

10.3.1 Temperature

Matrix dissolution rates and the uncertainties should be determined over the temperature range where the glass is expected to be exposed to the repository water. In the absence of a design this may require a temperature range extending from ambient to the surface temperature of the glass at emplacement. It is apparent that a package design in which the glass temperature is as low as possible (preferably below 100°C) at the time of contact with the water will be easier to evaluate for its performance. At present the leach rates of the best borosilicate glasses approximate the annual one part in 10^5 release criterion at around 30°C in pH values between 5 and 8 and at low flow rates.[5] Not only do the leach rates increase with increasing temperature but the uncertainties in the leach rate become greater. The majority of existing data on the effects of pH and flow rate on leaching are at temperatures below 100°C.[5,7-12]*

In order to evaluate the performance of glass at temperatures above 100°C a great deal more R and D would be required than is necessary to evaluate the performance below 100°C. At high temperatures the chemical compositions of the groundwaters may change in more complex manners. The variations in pH and ionic composition may become larger and evaluation of performance becomes more uncertain. For most silicate glasses, the quantity leached in a given time is nearly doubled for every 8°C to 15°C rise in temperature and the reaction rate increases by a factor of 10-100 for every 100°C increase in temperature, depending on the composition of the glass. Below approximately 80°C near pH 7, a siliceous layer forms on a glass which acts to retard further leaching. Metasomatic reactions, in which new crystalline compounds form from some of the glass constituents, can also occur at the glass surface, particularly at elevated temperatures. Such complications make it difficult to theoretically define a single rate-determining step in a given temperature range. In hydrothermal environments, the complexity is more significant since glass is altered rapidly if the temperature is sufficiently high. Under hydrothermal conditions, alteration is a major variable influencing the enhanced leach rate. Since the alteration is accompanied by complications such as stress generation, a delineation of the mechanisms involved in hydrothermal leaching is not easily achieved.

10.3.2 Simulated 1000-Year-Old Glass

Following initial surface changes, the long term behavior will be governed by the factors that affect matrix dissolution (i.e. chemical composition, possible phase separation, homogeneity, etc.) and the factors that change the leaching environment. If the waste form is to be protected from leaching during the containment period, the composition of the glass and the radionuclides of concern are not represented by much of the existing data. Matrix dissolution rates and the uncertainties in those rates should be determined for simulated aged glass under typical repository conditions. The aging should correspond to the time at which containment is likely to fail.

10.3.3 Radiation Effects

Much of the past work on radiation dealt with radiation effects on the glass. Although such effects may alter the leach behavior, existing evidence indicates the effects are small compared to the effects of temperature or pH. The radiolysis of groundwaters which may produce species that could increase leaching rates is of greater concern. Here little work is available. If it is assumed that the repository will be saturated early in its life, then large quantities of water which may be relatively slow moving will be subjected to high radiation fields if the waste packages are unshielded.

Experiments determining the changes in composition of typical groundwaters and their subsequent effects on leaching (and corrosion) will be necessary if a self shielded package is not developed. Again, in the absence of a self shielded package much of the existing work on matrix dissolution may need to be repeated in the presence of a radiation field if performance of the glass is claimed during the containment period. Such a situation would correspond to a waste package without a canister system where containment could be attempted from combined properties of the glass and backfills. This situation would require a great deal more R and D for performance evaluation than would a package in which a canister system provides reasonable assurance of containment.

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*See also: M. E. Nordberg, "Chemical Durability" Corning Glass Works, unpublished manuscript.

10.3.4 pH and Flow Rate Effects

pH

Along with temperature, glass composition and homogeneity, and radiolysis of the leach media, the factors most affecting leaching appear to be pH and flow rates.[13] pH effects may be more readily evaluated in the absence of radiolysis than in a radiation field. For glasses, data indicate that the enhanced leaching is due to radiolysis effects on leachant chemistry. Studies show that nitric acid,[14] formed by water radiolysis in the presence of atmospheric nitrogen, can significantly enhance leach rates by lowering the leachant pH. It does not appear that acid formation in the presence of air can account for all the irradiation enhanced leaching. The importance of factors other than irradiation induced pH changes depends on the radiation dose rate, and possibly depends on sample type. At repository dose rates, these effects could be less significant than pH effects.

The possible effects of leachant radiolysis have received emphasis in leach testing. Studies have been carried out investigating alpha and beta radiolysis effects, as well as the leaching under gamma radiation*. Effects of gamma radiolysis products on leachant pH were emphasized. It appears that the major effect on leach rates may be due to an irradiation induced decrease of leachant pH. It is not yet clear whether this mechanism can account for all the observed radiolysis changes.

Flow Rates

Contact time variations will affect the release and subsequent movement of radionuclides from the package. In the repository, groundwaters will contact the package components for varying time depending on a number of conditions, such as permeability of the host rock, temporal variations in the thermal field, etc. The contact time, or flow rate, is the most difficult variable to estimate because it will be controlled by site-specific conditions that are not easily predictable.

Designing a package which attempts to restrict flow around the canister and waste form may be a more reasonable approach than developing a program to try to completely understand the effects of flow.

If a backfill with positional stability is placed around the container system the uncertainties in leach behavior due to flow effects may be minimized.

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*D. D. Walker, M. D. Dukes, M. J. Poldenic, N. E. Bibler, Savannah River Laboratory, presented at the 181st National Meeting of the ACS, March 1981.

10.3.5 Modeling

It is improbable that a reaction such as leaching which may depend upon some 20 to 30 complex variables can be predicted from a mathematical model based on measurements in which only one or two variables are tested at a time. The feedback and number of possible interactions is enormous. A necessary requisite for testing any mathematical model purporting to predict long term performance is a test involving leaching in the presence of all the variables.

Empirical relationships developed from measurements based on site characterization information and design dependent materials interactions might be more useful in predicting long term behavior. There are several theoretical arguments which show that under appropriate conditions, short term engineering tests can be conservative and can be used to put limits on long term behavior.

For example, in the case of matrix dissolution, the leach rate versus time decreases. This may be due in part to the buildup of a protective surface layer on the glass, the decrease in surface area to volume as the matrix surface is corroded away, saturation of the leaching medium, etc. Changes in the dissolution rate caused by spallation of a surface film, small changes in leachate chemistry and flow should cause minor perturbations around the average dissolution rate, resulting in an envelope of rates which may best be described by sets of empirical equations, or by defining with some level of certainty, the range in dissolution rates as a function of the range in leachate chemistries, temperature and flow rates.

10.3.6 Conclusions

The major issue to be addressed is the matrix dissolution rates of glass and the uncertainties in those rates under typical repository conditions. These conditions include the variations in dissolution that would occur with temperature, flow rates, changes in leachate chemistry and aging of the glass. It is also apparent from the existing information that major aspects of the leaching behavior will be waste package design dependent. Corrosion products, backfill properties, host rock, etc. may make major contributions to the long term aspects of matrix dissolution. Therefore, in the absence of a specific design, the matrix dissolution rates should be measured in typical groundwater chemistries, over the temperature range anticipated from ambient to emplacement and include the effects of radiation and "aging." Typical groundwaters should be consistent with what is expected for the water in the presence of host rock. The rates should be determined over a period of time required for the rate to level off and long term matrix dissolution tests should also be initiated. These tests should continue until the time of emplacement. Such tests would help insure that the performance claimed from short term engineering tests does, in fact, represent a conservative approach.

10.4 Major Issue: Containers

Metallic container systems can provide long term containment in several ways depending upon the waste package design. These may range from a single, relatively thin corrosion resistant overpack to thick sacrificial metals that corrode in a predictable manner. In the latter case, the major issues of concern would involve determining the corrosion rate in the "worst case" repository environment, providing a rationale and model that the worst case corrosion rate either remains constant or decreases with time and then determining the thickness required for the containment period. The reliability and confidence associated with such a design would be related to the uncertainties in corrosion rate and to the excess thickness used in the design. A major concern with such a design may involve the positional, thermal, and chemical instability of the backfill-container interface after appreciable thicknesses of the container have corroded. The mechanical, chemical and thermal changes resulting from the gap formed may seriously affect performance and will require careful evaluation. In the situation where the corrosion products do not spall and a gap is not formed, the corrosion products may occupy a greater specific volume than the original metal. The consequences of this expansion and the possible pressures applied at the container-backfill interface will also require evaluation. Since there appears to be no significant effort in developing a data base on such a design, the discussion at this time is restricted to the above generic comments.

In the following major issues of concern, the first type of container system, TiCode-12, was chosen as an example of the component to provide long term containment.

10.4.1 Temperature

Temperature is one of the most important parameters affecting the lifetime of the container. If the surface temperature of the container system is designed to be low, then a minimal effort at higher temperatures may suffice to determine over what temperature range the corrosion mechanisms may remain the same. Similarly, the corrosion tests at elevated temperatures used to obtain information on failure modes should be fewer and simpler if the design favors a low container temperature.

The point to be stressed is that the mechanisms of corrosion at high temperatures are very likely to be different from those at low temperatures. Any design which has the container initially at a high temperature will require corrosion data over a range of temperatures covering the thermal changes expected during the containment period.

In addition to changing mechanisms at higher temperatures, there are a large number of theoretical and practical reasons why the scatter and uncertainties of kinetic reactions such as corrosion and leaching increase as the temperature increases. At higher temperatures the Maxwell distribution of energies (velocities) of both reactants and products widens so that a wider range of different close energy states exists. The consequences of these effects in corrosion and leach measurements result in a larger spread in final values. In practice, high temperature experiments will be more difficult and more expensive.

Specifically for titanium and TiCode-12,[15] temperature is likely to be one of the most important variables in the corrosion of HLW containers. The following evidence may be cited to support this:

- For titanium, exposed to 20% NaCl solution at 105°C, uniform corrosion, pitting corrosion and crevice corrosion occur. If the temperature is decreased to 80°C then pitting and crevice corrosion failure mechanisms are absent. Therefore, decreasing the temperature greatly reduces the number of possible failure mechanisms.

- The above statement is intimately connected with the widespread observation that in chloride containing solutions temperature changes from approximately 100°C to 200°C can change titanium based materials from the passive (very low corroding) state to the active state.

- In brine at 200°C, measurements show that very high acidity levels may be present ($pH = 2$). At room temperature the pH ranges between 4.0 to 6.5. Thus, by keeping the temperature low in a brine environment, accelerated corrosion from low pH values is minimized.

- Decreasing temperatures will reduce the rate of hydrogen diffusion into a TiCode-12 container, thereby reducing the potential for hydrogen embrittlement.

- A low temperature will greatly minimize the rate of plastic deformation in a container and reduce the possibility of failure associated with creep and stress-corrosion cracking.

- Oxide scales formed at low temperature will be thinner. This will reduce the buildup of stresses at the scale/metal interface and minimize scale spallation. Spallation, if it occurs, would lead to accelerated corrosion which would be extremely difficult to quantify over periods of hundreds of years.

- Because of the fewer failure mechanisms and slower kinetics at lower temperatures it would be expected that scatter in the experimental data would be reduced. This would allow more accurate extrapolation of behavior of container materials to very long times.

In summary, low temperatures will greatly reduce the number of possible corrosion and mechanical failure modes, especially in a brine medium. Reaction kinetics will be far slower and data scatter minimized. This will allow a more accurate estimate to be made of long term corrosion behavior.

10.4.2 Radiation Effects

A second, and equally important, parameter that will be considered in evaluating the performance of a corrosion resistant container like TiCode-12, is the effect of radiation. While direct damage to the container is not expected to be a concern, radiolysis of the groundwater can potentially result in catastrophic failure of the container by delayed hydrogen assisted fracture as well as enhance or accelerate other failure mechanisms.

Radiolysis of the groundwater by gamma radiation will produce hydrogen which can potentially lead to hydrogen embrittlement and the potential for enhanced delayed fracture. The parameters important[16] in hydrogen induced delayed fracture include hydrogen content, temperature, and stress.

Radiolysis effects on the groundwater may also enhance the rates of corrosion mechanisms such as crevice corrosion. For example, the production of an acidic solution by radiolysis within a crack may induce or enhance crevice corrosion particularly in higher temperature packages.[15,17] In the case of uniform corrosion, in TiCode the production of large amounts of hydrogen by radiolysis resulting in a reducing atmosphere, may also enhance the rate of uniform corrosion[18,19] by undermining the protective oxide layer. With both temperature and radiation effects the primary issues are the effects these parameters have on the rates of degradation either directly or indirectly by producing changes in the corrosive environment. It is, therefore, necessary as in the case of matrix dissolution to determine the behavior of the corrosion resistant barrier in "typical repository waters." With a specific design the amount of work required to generate the data necessary to evaluate the performance of the container material, could, in principle, be greatly reduced. For example, a package in which the container temperature is as low as possible and in which shielding has been applied to eliminate the effects of radiolysis should be easier to test and evaluate than one in which there is no shielding and the container temperature is high. Of all practical kinetic systems corrosion is the most difficult to predict or accelerate by obtaining data at elevated temperatures. Here again, not only do the rates increase but new mechanisms may arise and the uncertainties in the results increase with increasing temperatures.

Radiation effects introduce a potentially catastrophic failure mode and the combined effects may lead to enhancement of all potential failure modes (i.e. uniform, crevice, pitting, stress corrosion cracking, and hydrogen embrittlement).

10.4.3 Modeling

If data bases extensive enough for use in predictive modeling cannot be developed, it is expected that for phenomena like uniform corrosion, empirical equations may be developed which describe the behavior of the metal under specific environmental conditions. It is recognized that in most instances these empirical equations may be rudimentary and would assume a constant mechanism (e.g. there is no spallation of the oxide layer).

Threshold values for the expected range of environmental conditions and uncertainties in those values should be supplied for probable corrosion mechanisms for which predictive or empirical equations cannot be developed. An example would be hydrogen induced delayed fracture or crevice corrosion. Empirical relationships used for predictive modeling should be conservative and should include a rationale for their use. The range of conditions over which they approximate behavior as well as the data base and statistical analyses to justify their use should be given.

10.4.4 Conclusions

For a corrosion resistant container, such as TiCode-12, it is expected that the primary degradation modes will be determined for "typical repository waters" and anticipated events such as thermal and radiation induced changes in the repository water. This evaluation should be done for both weld and base metal. In the case of uniform corrosion the rates and uncertainties in the rates should be determined.

Recent evidence[20] also indicates that TiCode-12 may undergo crevice corrosion under certain conditions. For this mechanism as well as for hydrogen induced failure, it is expected that as a minimum the environmental conditions leading to these types of failure will be determined. For degradation modes such as pitting and stress corrosion cracking, it is again expected that the absence or presence of these modes under expected environmental conditions will be determined, and if present, the range of conditions which will lead to these failure modes will be determined.

Since all of the potential failure modes associated with TiCode-12 are influenced to some extent by a package design, a proper evaluation of the performance of the material would be facilitated by an early choice. For example, a package which includes a discrete backfill will result in a different range of repository water chemistries than one which does not. There are package options which would greatly reduce the effort needed to evaluate the performance of the metal. Shielding and low container temperature are obvious examples.

Long term performance tests should also be initiated to insure that information generated in short term accelerated tests truly represents a conservative estimate of the material behavior. Accelerated testing, using temperature as the accelerating parameter should be used cautiously and the data generated in these tests should be accompanied by a statistical analysis and a rationale for why the test represents an empirical extrapolation.

The concern is that the use of elevated temperatures to increase corrosion rates for the purpose of predicting long term corrosion behavior at one lower temperature can be theoretically unjustified and technically unsound and that the R and D needed to evaluate the performance of a container system at high temperatures is much more extensive and complicated than the R and D required for evaluation at a lower temperature.

10.5 Major Issue: Backfills[15]

Backfills can be used to prevent water from reaching the waste package, control the water flow to and from the package and retard radionuclides during and after containment.

In a repository environment the backfill will have to withstand large thermal changes, high radiation fields, changes in groundwater chemistry and flow and large mechanical stress. All of these conditions may influence its ability to control or prevent water flow and retard radionuclide migration. It is also possible that depending on a package design one or more of the possible functions of a discrete backfill may not be utilized. For example, it is possible that only the thermal conductivity of the backfill is an issue, that is, it is utilized only as a heat transfer agent. The data necessary to evaluate its performance will be more limited than in an instance where it is utilized to control water ingress as well as transfer heat.

In the absence of design information there are three major areas of concern associated with demonstrating the performance of backfill materials: positional stability, water permeability and radionuclide retardation properties.

10.5.1 Positional Stability and Water Permeability

In all cases the positional stability and the stability with time of water flow properties are of prime concern under expected events of wet and dry cycling. If the backfill is to perform any function as part of the waste package, it is expected that it will remain emplaced around the package. Alternate wet and dry cycles should not result in any physical alteration such as separation or settling of a component within the backfill. Furthermore, if the backfill is designed to provide for controlled water flow to and from the package, the alternate wet/dry cycling should not, for example, lead to major fissure formation. The changes in properties with wet/dry cycling must include anticipated conditions; for example, a thermal gradient over hundreds of years in typical groundwaters.

The problems associated with positional stability will be material dependent and design dependent. In composite backfills made of several materials with differing densities and particle sizes, repeated flooding and drying can lead to a physical separation similar to the process in which gold is separated from sand. For a backfill such as sand-bentonite repeated flooding and drying can fluidize the bentonite causing separation and possible collapse of the backfill. This particular process is accelerated when the pH of the water exceeds ~9. For homogeneous backfills that are particulate, changes in structure over long times would be expected only if there were large differences in particle size. Such particle size separation would lead to changes in thermal conductivity and water flow. A generic concern in addition to those noted above depends upon the packing fraction or void space in the repository. With large void volumes some settling and densification should occur. Depending upon the repository design, this could either aid or hinder the performance of the waste packages.

10.5.2 Radionuclide Retardation Properties

From a generic point of view, any positional or structural change in the backfill which alters its water flow properties or its thermal conductivity and heat transfer properties will, in principle, alter its chemical retardation properties. In addition, radiation effects on the backfill material, radiolysis of the groundwaters and development of large quantities of corrosion products can also alter retardation performance of the backfill. For certain choices of materials long term stability under the temperature, radiation and wet/dry cycling conditions may require extensive evaluation.

10.5.3 Backfill Performance Modeling

The performance of the backfill should be modeled under the anticipated range of water flow, geometry and other pertinent environmental conditions.

10.5.4 Conclusions

A large number of serious concerns associated with backfill performance can be eliminated if the waste package is shielded and if attempts are made to develop homogeneous, uniform particle size backfills that are emplaced carefully so as to minimize the void volume. Such configurations should be stable to dimensional changes that might occur from large quantities of corrosion products if sacrificial thick self shielding canisters are used. It also seems likely that changes in the thermal conductivity or heat transfer of the canister due to corrosion products will not be serious if the backfill maintains positional stability during these changes.

10.6 Summary of Major Issues for Various Package Options

The following tables are based on what is known of the performance of the materials chosen as examples and are meant to illustate some of the major issues which should be addressed in order to evaluate the performance of the design options listed. The following assumptions were used to develop these tables:

(1) The package materials were assumed to be borosilicate glass (BSG), TiCode-12, and sand-bentonite or zeolite backfills. In some instances a shielding material is assumed. Table 5 is based on a sacrificial container for containment.

(2) The repository considered is a hard rock repository.

(3) The optimum containment is considered to be the time required for the waste form to return to ambient temperatures.

(4) It should be noted that the addition of a new parameter (e.g., radiation) to a test program or the extension of the range on a parameter (e.g., temperature) increases the quantity of work required to address an issue.

(5) Any test program which uses accelerated test methods to address an issue should be accompanied by a rationale justifying the use of the accelerated test procedure.

(6) These tables also assume that a statistical approach to testing is used, the average behavior and uncertainty in the average are determined from a statistically significant number of samples. For those processes where threshold levels are indicated, the range of conditions under which a process does or does not occur should be determined.

TABLE LEGEND

The Δ in parentheses next to each table refers to changes relative to Table 1:

Δ:NS = no shielding
Δ:T↑ = higher temperature
Δ:CWB = containment also achieved by properties of waste form and backfill
Δ:SC = sacrificial container

Example: Δ: NS, T↑, CWB indicates a package with no shielding, higher temperature, container fails before waste form reaches ambient and waste form and backfill provide for containment until waste form returns to ambient.

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Table 1

Reference Case

ASSUMPTIONS

1. Low temperature: outermost container temperature as low as possible (ALAP)
2. Shielded package: radiolysis of groundwaters is minimal.
3. Restricted water flow: controlled by discrete backfill.
4. Containment is achieved by a corrosion resistant barrier: containment is defined as length of time required for waste form to return to ambient.
5. Examples: BSG, TiCode-12, sand-bentonite/zeolite. shielding to minimize hydrolysis.

SINGLE COMPONENT ISSUES - Purpose: To develop a base for assessing the basic performance of package components and for a comparison of the positive or negative interaction of combinations of components.

WASTE FORM	CONTAINER	BACKFILL	REPOSITORY CONDITIONS
1. Effects of compositional variability on matrix dissolution (range and uncertainty) in typical repository water (T near ambient).	1. For both weld and base metal: determine range and uncertainty in uniform corrosion rates in typical repository water chemistries and under thermal changes (T: ALAP).	1. Establish positional stability during thermal changes and partial/full saturation.	1. Determine range in groundwate chemistries as functions of thermal changes: a. alone b. in combination with host rock.
2. Determine range and uncertainty of matrix dissolution rates for simulated "aged" glass in typical repository groundwaters.	2. Crevice corrosion: both weld and base metal (see bicomponent tests).	2. Determine effect of wet/dry cycling on positional stability retardation properties, and ability to restrict or retard water ingress (T: emplacement to ambient).	2. See bicomponent issues: backfill/host rock.
		3. Radionuclide retardation as a function of typical repository water; low flow.	

BICOMPONENT/MULTICOMPONENT ISSUES - Purpose: To define interactions of nearest neighbors and assess possible positive or negative interaction of components in combination under typical conditions.

WASTE FORM/CANISTER	BACKFILL/CONTAINER	BACKFILL/GROUNDWATER (Equilibrated With Host Rock)
1. Define range, uncertainty in matrix dissolution rate of aged BSG in presence of corroded stainless or cast steel containers.	1. Determine if crevice corrosion occurs in range of repository water chemistries present. Thermal changes, conditioning of water by backfill, etc.).	1. Determine range and uncertainties of groundwater chemistries after going through backfill under thermal changes.
	2. Determine range of uniform corrosion rates in presence of backfill and typical repository water.	
	3. Determine ability of backfill to retard radionuclide migration in presence of corroded container material (T~ambient).	

LONG TERM PERFORMANCE DEMONSTRATION - Purpose: To insure long term behavior extrapolated from shorter term tests.

WASTE FORM	CONTAINER	BACKFILL
1. Long term matrix dissolution studies of BSG in typical repository water at temperature at and near ambient.	1. Long term corrosion tests of TiCode-12 base and weld metal cycled from T (ALAP) to ambient (uniform corrosion samples, crevice corrosion samples.	1. Long term test on wet/dry cycling from T (ALAP) to ambient using typical groundwaters with and without corrosion products.

Table 2

Case 2 (Δ:NS)

ASSUMPTIONS

1. Low temperature: outermost container temperature ALAP
2. Unshielded package.
3. Restricted water flow: controlled by discrete backfill.
4. Containment is achieved by a corrosion resistant barrier (containment is defined as length of time required for the waste form to return to ambient).
5. Examples: BSG, TiCode-12, sand-bentonite/zeolite.

SINGLE COMPONENT ISSUES

WASTE FORM

1. Same as Case 1.
2. Same as Case 1.
1. Determine range and uncertainty in matrix dissolution rate for simulated "aged" glass in typical repository water <u>which has been irradiated during containment period</u>.

CONTAINER

1. Same as Case 1: Add radiation effects.
2. Same as Case 1: Add radiation effects.
3. Hydrogen effects* - demonstrate absence or presence of with respect to radiation and thermal changes in typical repository waters. Determine threshold values.

BACKFILL

1. Same as Case 1.
2. Same as Case 1: Add radiation.
3. Establish absence or presence of extensive radiation damage to backfill and its subsequent ability to retard radionuclide migration as a function of typical repository waters including radiolysis of the water (pH changes, new species, etc.)

REPOSITORY CONDITIONS

1. Same as Case 1: Add radiatio effects.
2. Same as Case 1.

BICOMPONENT/MULTICOMPONENT ISSUES

WASTE FORM/CANISTER**

1. Determine range and uncertainty in matrix dissolution rate of BSG in presence of corroded stainless or cast steel containers. Typical repository water, which has been irradiated during containment period, near ambient temperatures.

BACKFILL/CONTAINER

1. Determine if and under what conditions crevice corrosion occurs - range of repository water chemistries that increases the effects of:
   a. radiation
   b. thermal changes (T: ALAP to ambient).
2. Determine range of uniform corrosion rates in presence of backfill:
   a. radiation changes
   b. thermal changes (T: ALAP to ambient).
3. Determine if catastropic hydrogen assisted failure occurs, range of conditions including
   a. radiation changes
   b. thermal changes
4. Determine ability of backfill to retard radionuclide migration (near ambient) in presence of corroded TiCode-12 using typical repository water.

BACKFILL/GROUNDWATER
(Equilibrated With Host Rock)

1. Same as Case 1: Add radiation effects.

LONG TERM PERFORMANCE DEMONSTRATION

WASTE FORM

1. Long term matrix dissolution studies of BSG in typical repository water (includes effect of exposure to radiation) at temperatures near ambient).

CONTAINER

1. Long term corrosion tests on both base and weld metal.
   T: ambient to ALAP:
   a. uniform corrosion samples
   b. crevice corrosion samples
   c. hydrogen embrittled samples.

BACKFILL

1. Long term test on wet/dry cycling using typical repository waters:
   a. thermal and radiation effects.
2. Establish long term tests on retardation properties of irradiated backfills (includes effects of irradiation of repository water).

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* This is a potentially catastrophic failure mode and it is recognized that determining the threshold values may require extensive R D.

**This is the BSG waste form in the cast or stainless steel container. R&D more extensive since shielding in Case 1 separates waste form from overpack.

Table 3

Case 3 (L:NS, CWB)

ASSUMPTIONS

1. Low temperature: outermost container temperature ALAP
2. Unshielded package.
3. Restricted water flow: controlled by discrete backfill.
4. Containment: Partially achieved by the corrosion resistant barrier. The waste form and the backfill are assumed to provide for residual containment until the waste form returns to ambient temperature.
5. Example: BSG, TiCode-12, sand bentonite/zeolite.

SINGLE COMPONENT ISSUES

WASTE FORM	CONTAINER	BACKFILL	REPOSITORY CONDITIONS
1. Same as Case 1.	1. Same as Case 1, add radiation.	1. Same as Case 1.	1. Same as Case 1, add radiation
2. Determine range and uncertainty in matrix dissolution rates in typical repository waters including: a. thermal changes (T: ambient to emplacement) b. radiation fields c. simulated, aged glass.	2. Same as Case 1, add radiation.	2. Effect of wet/dry cycling on positional stability, retardation properties and ability to restrict or retard water ingress (T: emplacement to ambient).	2. See bicomponent issues - backfill/host rock.
	3. Hydrogen effects - demonstrate absence or presence of with respect to radiation and thermal changes in typical repository waters. Determine threshold values.	3. Establish absence or presence of extensive radiation damage to backfill and its subsequent ability to retard radionuclides in typical groundwaters as a function of: a. temperature (T: emplacement to ambient) b. radiation.	

BICOMPONENT/MULTICOMPONENT ISSUES

WASTE FORM/CANISTER	BACKFILL/CONTAINER	BACKFILL/GROUNDWATER (Equilibrated With Host Rock)
1. Same as Case 1, add: a. temperature b. radiation.	1. Same as Case 1, add radiation.	1. Same as Case 1, and as a function of a. radiation b. temperature.
	2. Same as Case 1, add radiation	
	3. Hydrogen effects in presence of backfills, range of conditions.	
	4. Determine ability of backfill to retard migration in presence of corroded TiCode-12 as functions of temperature and irradiated groundwaters.	

LONG TERM PERFORMANCE DEMONSTRATION

WASTE FORM	CONTAINER	BACKFILL
1. Long term matrix dissolution in repository waters at temperatures up to emplacement and in the presence of radiation field.	1. Long term corrosion tests of both base and weld metal. T: ambient to ALAP; radiation changes: a. uniform corrosion samples b. crevice corrosion samples c. hydrogen embrittled samples.	1. Long term test on wet/dry cycling using typical groundwaters: a. thermal and radiation effects
		2. Establish long term tests on retardation properties under irradiation and temperature change.

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*R+D more extensive than Case 1 because of leaching during thermal period with radiation.

Table 4

Case 4 (A:SC)

ASSUMPTIONS

1. Low temperature: Sacrifical corrosion barrier: Temperature as low as possible (ALAP)
2. Shielded package.
3. Restricted water flow: controlled by discrete backfill.
4. Containment: Achieved by container until waste form returns to near ambient.
5. Examples: BSG, low carbon steel, sand bentonite/zeolite.

SINGLE COMPONENT ISSUES

WASTE FORM

1. Same as Case 1.
2. Same as Case 1.
3. Repeat 1 and 2 in presence of expected corrosion products.

CONTAINER*

1. Same as Case 1.
2. Establish failure modes other than uniform corrosion: typical repository water.
3. Evaluate 1 and 2 in varying radiation and thermal, i.e., waste is decaying and shielding is corroding.
4. Evaluate thermal, mechanical and chemical instabilities due to gap or expansion caused by corrosion.

BACKFILL

1. Same as Case 1.
2. Same as Case 1.
3. Same as Case 1.
4. Re-evaluate 1-3 in presence of large amounts of corrosion products, and varying radiation field.
5. Re-evaluate positional stability under volume change (see bicomponent issue).

REPOSITORY CONDITIONS

1. Same as Case 1.
2. Same as Case 1.
3. Re-evaluate 1 and 2 in prese varying radiation field and sion products.

BICOMPONENT/MULTICOMPONENT ISSUES

WASTE FORM/CANISTER

1. Same as Case 1.
2. Matrix dissolution in very large amounts of corrosion products.

BACKFILL/CONTAINER

1. Determine failure modes of container in presence of backfill under varying thermal and radiation fields.
2. Re-evaluate backfill performance when backfill-container interface is saturated with corrosion products.
3. Re-evaluate positional stability under volume change induced by corroding container.
4. Re-evaluate thermal and chemical (ion retardation) changes due to changes caused by corrosion.

BACKFILL/GROUNDWATER
(Equilibrated With Host Rock)

1. Same as Case 1.
2. Re-evaluate in presence of corrosion products.

LONG TERM PERFORMANCE DEMONSTRATION

WASTE FORM

1. Same as Case 1, add large quantity of corrosion products.

CONTAINER

1. Long term corrosion test.
   a. varying radiation field
   b. varying temperature.

BACKFILL

1. Long term test on wet/dry cycling using typical groundwaters:
   a. thermal and radiation effects
   b. volume changes as a result of container corrosion.

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*It is unlikely that such a system can avoid pitting corrosion.

Table 5

Case 5 (Δ:T↑)

ASSUMPTIONS

1. High temperature: outermost container T > 100°C.
2. Shielded package.
3. Restricted water flow: controlled by discrete backfill.
4. Containment: Achieved by a corrosion resistant barrier (containment is defined as the length of time required for the waste form to return to near ambient.
5. Examples: BSG, TiCode-12, sand-bentonite/zeolite. shielding

SINGLE COMPONENT ISSUES

WASTE FORM	CONTAINER	BACKFILL	REPOSITORY CONDITIONS
1. Same as Case 1.	1. Same as Case 1: extended temperature range.	1. Same as Case 1: extended temperature range.	1. Same as Case 1: extended temperature range.
2. Same as Case 1.	2. Same as Case 1: extended temperature range.	2. Same as Case 1: extended temperature range.	2. Same as Case 1.
	3. For weld and base metals determine presence or absence of other failure modes (e.g., crevice corrosion, stress corrosion cracking, pitting) in presence of typical repository water under a large temperature range. Average time-to-failure.	3. Same as Case 1.	

BICOMPONENT/MULTICOMPONENT ISSUES

WASTE FORM/CANISTER	BACKFILL/CONTAINER	BACKFILL/GROUNDWATER (Equilibrated With Host Rock)
1. Same as Case 1.	1. Re-evaluate failure modes (uniform, crevice, etc.) in range of repository water chemistries present. (Extended temperature range, conditioning of water by backfill.)	1. Same as Case 1: extended temperature range.
	2. Same as Case 1.	

LONG TERM PERFORMANCE DEMONSTRATION

WASTE FORM	CONTAINER	BACKFILL
1. Same as Case 1.	1. Long term corrosion testing on base and weld metal. Extended temperature range from emplacement to ambient, e.g., uniform corrosion, crevice corrosion, stress corrosion.	1. Same as Case 1: extended temperature range.

Table 6

Case 6 (Δ:NS, T↑)

ASSUMPTIONS

1. High temperature: outermost container (e.g. T >100°C).
2. Unshielded package.
3. Restricted water flow: controlled by discrete backfill.
4. Containment: Achieved by a corrosion resistant barrier (containment is defined as length of time required for waste form to return to ambient).
5. Examples: BSG, TiCode-12, sand-bentonite/zeolite.

SINGLE COMPONENT ISSUES

WASTE FORM

1. Same as Case 1.
2. Same as Case 1.

CONTAINER

1. Same as Case 1: Add radiation effects and extended temperature range.
2. Same as Case 1: Add radiation effects and extended temperature range.
3. Determine if hydrogen assisted failure occurs - range of conditions: Extended temperature range.
4. For weld and base metals determine if other failure modes present in extended temperature range: Add radiation effects. Average time-to-failure.

BACKFILL

1. Same as Case 1: Extended temperature range.
2. Same as Case 1: Extended temperature range.
3. Establish absence or presence of extensive radiation damage to backfill and its ability to retard radionuclide migration as a function of "typical" groundwaters. Include:
   a. radiolysis of groundwater
   b. effects of large thermal changes on backfill.

REPOSITORY CONDITIONS

1. Same as Case 1: Add radiation effects and extended temperature range.
2. Same as Case 1.

BICOMPONENT/MULTICOMPONENT ISSUES

WASTE FORM/CANISTER

1. Determine range and uncertainty in matrix dissolution rate of BSG in presence of corroded stainless or cast steel containers. Typical repository water is that expanded to combine radiation field and large temperature fluctuations

BACKFILL/CONTAINER

1. Same as Case 1. Add:
   a. extended temperature range
   b. radiation
2. Same as Case 1. Add:
   a. extended temperature range
   b. radiation
3. Determine if catastrophic hydrogen assisted failure occurs, range of conditions including:
   a. extended temperature range
   b. radiation changes.
4. For base and weld metals determine if other failure modes occur, range of conditions including:
   a. extended temperature range
   b. radition effects
5. Determine ability of backfill to retard radionuclides in presence of corroded TiCode-12 using typical repository waters (which have been exposed to large radiation and thermal changes).

BACKFILL/GROUNDWATER
(Equilibrated With Host Rock)

1. Same as Case 1.
   a. extended temperature range
   b. radiation effects

LONG TERM PERFORMANCE DEMONSTRATION

WASTE FORM

1. Long term matrix dissolution studies of BSG in typical repository waters (includes exposure to large temperature change and exposure to radiation) at temperatures near ambient.

CONTAINER

1. Long term corrosion tests of base and weld metals. T: emplacement to ambient: radiation:
   a. uniform corrosion
   b. crevice corrosion
   c. stress corrosion
   d. pitting corrosion

BACKFILL

1. Long term tests on wet/dry cycling using typical groundwater:
   a. extended temperature range
   b. radiation effects
2. Establish long term tests on retardation properties of irradiated backfills (includes effects of irradiation of groundwaters and large temperature change).

Table 7

Case 7 (Δ:NS, $T\uparrow$, CWB)

ASSUMPTIONS

1. High temperature: outermost container has emplacement temperature >100°C.
2. Unshielded package.
3. Restricted water flow: controlled by discrete backfill.
4. Containment: Partially achieved by a corrosion resistant barrier. The waste form and backfill are assumed to provide residual containment until the waste form returns to ambient temperature.
5. Examples: BSG, TiCode-12, sand-bentonite/zeolite.

SINGLE COMPONENT ISSUES

WASTE FORM	CONTAINER	BACKFILL	REPOSITORY CONDITIONS
1. Same as Case 1.	1. Same as Case 1: a. extended temperature range b. radiation effects.	1. Same as Case 1: Add extended temperature range.	1. Same as Case 1; a. extended temperature ran b. radiation
2. Determine range and uncertainty in matrix dissolution rate in typical repository waters including: a. large temperature changes (T: emplacement to ambient) b. radiation field c. simulated, aged glass.	2. Same as Case 1: a. extended temperature range b. radiation effects.	2. Effect of wet/dry cycling on positional stability, retardation properties and ability to restrict or retard water ingress (extended temperature range).	2. Same as Case 1.
	3. For weld and base metal hydrogen effects: Demonstrate absence or presence of with respect to radiation and thermal changes in typical groundwater. Determine threshold values.	3. Establish absence or presence of extensive radiation damage to backfill and its ability to retard radionuclides in typical groundwaters as a function of: a. extended temperature range b. radiation effects.	
	4. For weld and base metal: Evaluate presence or absence of other failure modes. Average time-to-failure: a. large temperature range b. radiation effects.		

BICOMPONENT/MULTICOMPONENT ISSUES

WASTE FORM/CANISTER	BACKFILL/CONTAINER	BACKFILL/GROUNDWATER (Equilibrated With Host Rock)
1. Same as Case 1, add: a. extended temperature range b. radiation.	1. Same as Case 1, add: a. extended temperature range b. radiation effects.	1. Same as Case 1 and as a function of: a. extended temperature range b. radiation effects.
	2. Same as Case 1, add: a. extended temperature range b. radiation effects.	
	3. Determine hydrogen effects in presence of backfills, typical repository water. a. extended temperature range b. radiation effects.	
	4. Determine presence or absence of other failure modes in presence of a backfill and typical repository water. Average time-to-failure: a. extended temperature range b. radiation effects.	
	5. Determine ability of backfill to retard migration in presence of corroded TiCode-12: a. extended temperature range b. irradiated groundwater.	

LONG TERM PERFORMANCE DEMONSTRATION

WASTE FORM	CONTAINER	BACKFILL
1. Long term matrix dissolution in groundwaters at temperatures up to emplacement temperature and in presence of radiation.	1. Long term corrosion tests on both base and weld metal: Extended temperature range and in a radiation field: a. uniform corrosion samples b. pitting corrosion samples c. crevice corrosion samples d. stress corrosion samples e. embrittled samples.	1. Long term tests on wet/dry cycling using typical groundwaters including a. extended temperature range b. radiation effects.
		2. Establish long term tests on retardation properties: a. extended temperature range b. radiation effects.