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NNWSI PHASE II MATERIALS INTERACTION TEST
PROCEDURE AND PRELIMINARY RESULTS

by

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ABSTRACT

The Nevada Nuclear Waste Storage Investigations (NNWSI) project is investigating the volcanic tuff beds of Yucca Mountain, Nevada, as a potential location for a high-level radioactive waste repository. This report describes a test method (Phase II) that has been developed to measure the release of radionuclides from the waste package under simulated repository conditions, and provides information on materials interactions that may occur in the repository. The results of 13 weeks of testing using the method are presented, and an analog test is described that investigates the relationship between the test method and expected repository conditions.

I. INTRODUCTION

The Nevada Nuclear Waste Storage Investigations (NNWSI) project is investigating the volcanic tuff beds of Yucca Mountain, Nevada, as a potential location for a high-level radioactive waste repository. One of the topics to be addressed during this investigation is the performance of the waste package components in the repository at the termination of the containment period, i.e., following the 300/1000-year period during which containment of high-level waste to within the waste package will be substantially complete.

The containment period is followed by the isolation period, during which the potential for canister breach and subsequent groundwater/waste form contact exists. Radionuclide release from the waste package would then be possible, although, in accord with Nuclear Regulatory Commission [NRC] regulations, the total release rate must be less than one part in 10^5 per year of the total repository inventory of a particular radionuclide after the 1000-year period.

To measure this radionuclide release, a test method is required that incorporates the interactions between the waste package components and the groundwater under realistic repository conditions. Release results obtained in this fashion could then be used as source term data in modeling the long-term behavior of the repository. NNWSI is developing such materials interaction tests and is using them to produce waste package release data.

The first test (Phase I) is a rock cup, total submersion test; the second test (Phase II) is a more realistic simulation of conditions expected in the unsaturated zone in tuff. This report describes the development of the Phase II test, provides a test procedure to use when performing the test, and reports results from up to 13 weeks of testing obtained using the test method.

II. NNWSI PHASE II MATERIALS INTERACTION TEST DEVELOPMENT

The guiding principle behind the development of the Phase II test is to provide reliable data using a test matrix and procedure that are simple enough to be performed and interpreted by different laboratories, and that can produce results within a useful time period. Considerable emphasis is placed on reliability, which implies that the data must be reproducible, site-relevant, and good enough quality to be used in further applications.

High-level waste testing under unsaturated conditions is unique to NNWSI, and procedures developed previously for saturated testing are not completely applicable to unsaturated testing. Also, the decision to use an unsaturated medium is relatively recent, and a complete description of the repository horizon is evolving [OVERSBY, BALLOU]. Some parameters necessary to develop a test are not completely understood and may possibly change as more information is developed. The test method must be able to accommodate such changes.

It is apparent that the NNWSI Phase II test involves a potentially large number of experimental parameters and could easily be unwieldy and complex if its goal were to obtain a complete understanding of all mechanisms associated with radionuclide release. Instead, the test has been simplified by severely restricting the experimenter's flexibility in choosing and varying test parameters. Only radionuclide release from the waste package under the restricted set of experimental conditions make up reportable data that can be used directly in further repository modeling. The experimenter is given freedom to study other effects, but the use of this information is solely to guide or interpret other experiments or to suggest modifications to the Phase II test procedure.

The utility of the data depends on the selection and restriction of the repository conditions and on the materials interactions that are incorporated into the test. These interactions could affect release, in perhaps an unpredictable fashion. The effect of selected parameters will be studied in parametric testing. The results of the parametric tests will be used to suggest changes that should be made in the Phase II test procedure, to help interpret Phase II test results, or to provide information concerning the effect of that parameter as related to radionuclide release.

Also, in order to demonstrate the relationship between the Phase II test and the repository, an analog test is being performed. The analog test attempts to more closely simulate actual repository conditions than does the Phase II test and is necessary because some of the constraints required in a regulated test procedure may not exist in a repository. If these test constraints cause a significant deviation between the analog and Phase II test results, then the data obtained by use of the Phase II test may not be a reasonable repository simulation and should be questioned if used in repository modeling. The analog test is described in detail in Section V.

The Phase II test was developed based on the best current knowledge of the repository and on the premise that some radionuclide release would occur sometime during the isolation period. These restrictions are detailed in Section III and result mainly from the repository and waste package design,

the hydrology, and the aged nature of the components; basically, they include assumptions used to estimate the amount of water available to contact the waste form and the manner in which it will contact the waste form.

The test procedure was developed using a series of experiments, with each experiment being more sophisticated than the preceding one. The initial question was whether enough material was released from the waste form to be measured. This question was answered positively [STEINDLER-1], and the test apparatus was refined to produce more reproducible results.

This refined test vessel and test procedure were used in a series of parametric tests [STEINDLER-2]. These tests did not duplicate repository conditions, but used materials on hand, and were designed to demonstrate the utility of the test procedure. The tests were encouraging, indicating that fairly reproducible results could be obtained, and modifications necessary to increase reliability were suggested.

Based on the parametric test results, two prototype vessels were constructed. These vessels were to be the actual Phase II test vessels and were designed for either bench-top or hot cell use. These vessels were tested and produced results similar to those obtained from the parametric tests [STEINDLER-2]; thus, the Phase II test procedure was finalized and a set of Phase II interaction tests was initiated.

The test procedure is given in Section III. It describes the apparatus, procedures, and test matrix in detail. It generally follows the format required by the Materials Review Board (MRB) for submission of procedures. Bracketed sections are provided as explanatory information and may not be included in a final test procedure.

This test method will be used by several laboratories to produce data. These data will then be used to document the reliability of the test method and to suggest changes that may be required. Phase II testing has been completed through 13 weeks, and these results are presented in Section IV.

III. NNWSI PHASE II MATERIALS INTERACTION TEST METHOD

1.0 SCOPE

The test procedure described herein is a Materials Interaction Test Method developed to address conditions relevant to the NNWSI project. The test will provide information to be used by the NNWSI project in licensing the NNWSI repository site. The test procedure will (1) provide data that describe the release of radionuclides from a specifically designed waste package under strictly controlled test conditions, and (2) provide information concerning synergistic effects that may occur between waste package components.

The purpose of the test is to obtain data on the release of waste components from the waste package in the repository environment as it is expected to be after the currently envisioned 300/1000-year containment period has elapsed. Inherent in this goal is that a specific and reasonable description of the waste package and repository at this time period is available, so that aging effects can be incorporated into the test procedure.

Considerable focus is placed on the principle of being specific and reasonable. These constraints allow the test to be limited in scope by controlling the range to which variables need to be addressed, yet still allow the test to produce relevant information because all conditions currently recognized to affect the release are addressed. The test will be performed using selected values for important variables, and the final result will be a reflection of how realistically the values were chosen. This approach was chosen to produce data that are reproducible and adequately simulate NNWSI site-related conditions.

[The NNWSI repository site is located in the unsaturated zone. This introduces a degree of variability to establishing an experimental design for which there is limited experience to use as a reference. It is recognized that the effect of any variable could affect the behavior of the waste package in an unanticipated manner. Yet, to study the effect of each variable in the test procedure would not be practical. The best approach was believed to require identification of important variables and physical processes, assignment of reasonable, perhaps worst-case, values, and performance of the test. The effect of each individual variable could be studied in parametric testing and, if necessary, the test method could be amended in response to these results.]

[The rationale for selecting test conditions and variables was to address those materials interactions that may exist in a repository during the isolation period. The materials consist of the waste form and the canister/overpack. No backfill is included because it is not part of the primary repository design [BALLOU]. The interactions are promoted by contact with water and air. The test provides for several interactions to occur and allows the experimenter to assess the importance of each. Since during actual repository storage each waste package may not be subjected to all possible materials interactions, the experimental design simulates interactions, but does not attempt to model the current waste package design. More detail concerning the selection of test conditions and reaction variables is presented in later sections.]

Another requirement for the test is that the same procedure and equipment be used in testing simulated and actual radioactive wastes, and that the procedures are applicable to interactive testing. The test can be performed with commercial or defense waste glass forms, spent fuel, or an Approved Test Material.

[To demonstrate the relevance of this test to repository conditions, a series of analog tests is being performed. These tests duplicate the waste package used in the interaction test procedure but attempt to be a closer simulation to the actual repository setting. The analog test uses a tuff core instead of a stainless steel reaction vessel and groundwater transport with flow through the tuff core instead of a regulated water drop. The waste package degradation has been compared for the two different simulations after three months of testing and found to be nearly identical. This is preliminary evidence that the repository conditions may be closely simulated in the interaction test. It needs to be demonstrated that the analog test is a good repository simulation.]

2.0 SUMMARY

Specimens representing the anticipated NNWSI waste package (waste form and canister) are contacted intermittently by dripping repository water. The nature and degree of radionuclide release from the waste package is determined by collection and analysis of the water that has contacted the package and by surface analysis of the waste package components. Materials interactions are noted.

The test schedule incorporates batch and continuous testing. In the batch mode, tests are terminated at selected time intervals. The test apparatus is disassembled, and analyses of both the solution and components are performed. In the continuous mode, the waste package (including adhering liquid) is transferred to a new test vessel, and the test is continued. Analyses can be done on the solution in the old vessel. Using the continuous testing matrix, replication of solution analysis can be achieved; investigation of the test components is possible at the termination points, yet the test can continue indefinitely or until information most useful to repository evaluation is obtained.

[At the end of a test period it is necessary to measure the amount of selected elements that have been released from the waste package. By definition this includes species in solution in the test vessel or on the waste package, or species that have sorbed onto the test vessel or other non-waste package components. The total of each element release is obtained by summing the amounts in various sources.]

[It is also of interest to investigate the method by which the species are released from the waste package. Release could be by water dripping from the waste package, by transport through a thin film of water that is present on all surfaces, or by an aerosol transport mechanism. Thus, the test matrix includes experiments that differ somewhat in design and will, it is presumed, provide some useful information on the transport mode.]

3.0 SAFETY PRECAUTIONS

Tests are performed at 90°C, atmospheric pressure, and involve metal test vessels. Care should be exercised when handling the thermally hot test vessels.

Established safety procedures and regulations concerning the use and handling of radioactive materials are to be followed. Depending on the type (alpha, beta, or gamma) and amount of radiation, special precautions such as lead shielding, glove boxes, or protective equipment may be required.

4.0 USES AND LIMITATIONS

The test is designed to provide site-relevant, reproducible data that can be used as input to the licensing of the NNWSI repository site. To achieve this goal, the test has been designed as a simplified, controlled simulation, but not a duplication, of expected repository conditions. The simulated conditions used in designing the test are listed below.

Parameter Choices and Constraints

The anticipated repository conditions will impose constraints on several parameters that could affect release from the waste package. The test conditions were chosen to simulate conditions that may exist in the repository after 300/1000 years of burial, attempting to anticipate worst-case, but realistic bounds [OVERSBY, BALLOU]. The test conditions so chosen are the following:

- 1) temperature is 90°C;
- 2) water contacting the waste package is actual J-13 well water that has been reacted with tuff at 90°C;
- 3) the amount of water that contacts the waste package is a function of the type of waste being tested and the associated repository design (a contact rate will be given for defense and commercial glass and for spent fuel);
- 4) canister material is Type 304 L stainless steel, aged as required;
- 5) contact mode is dripping water with a drop volume of ~0.1 mL per drop, based on in situ measurements from tuff;
- 6) repository rock is tuff;
- 7) pressure is set at 1 atm (100 kPa) for the simulated repository, which is saturated with water vapor; and
- 8) temperature differential between the waste package and the test vessel is based on the thermal equilibration of test components.

Additional conditions have been incorporated into the test procedure to provide the experimental simplicity necessary to obtain reproducibility. These constraints are mentioned and more detail is given in later sections:

- 1) waste form shape, dimension, and physical description;
- 2) waste form/canister contact; and
- 3) degree of component aging.

The parameters described above are limitations placed on the test to provide a worst-case scenario and to ensure data reliability.

The tests are used to provide the following:

- 1) Data on release of radionuclides and selected matrix elements from the waste package by means of solution analysis as a function of time. These data will be used to determine whether approach to constant release from the waste package can be established and what the limits of this release are. The data may be used in modeling repository behavior, subject to the restrictions listed below.

- 2) Data on alteration of the waste form as a function of time. These data will corroborate those from the solution analysis to determine whether constant release has been established and provide insight into the method of the waste form/water reaction. The test matrix is not extensive enough to directly provide data for determination of a detailed mechanism of the release process.
- 3) A description of the interaction between the waste form and canister under the specific test conditions, useful for a more detailed investigation of canister corrosion and reaction.
- 4) A description of the interaction between the released radionuclides and the repository rock under specific test conditions, serving to aid in interpreting more detailed radionuclide-migration testing.
- 5) Information concerning the method of species transport from the waste package to the test vessel and other non-waste-package components of the test apparatus.

[An example as to what No. 5 might encompass is: if aerosol and liquid film transport are important and are equilibrium processes, and if tuff acts as a sink for selected elements, then a driving force to transport a species from the waste package to the tuff would exist and would affect the actual waste form degradation. Aerosol or liquid film transport may be an artifact of the experimental design and may not occur in a repository. A portion of the test procedure addresses this issue.]

Test limitations are as follows:

- 1) The test conditions are not an exact duplication of the repository conditions, and the test results should not be taken out of context and used as such. However, in conjunction with parametric tests, it may be possible to demonstrate whether differences or similarities exist between the test and the repository. If the differences are negligible, then the results may be used as source term data to model repository behavior, provided the data are used in an appropriate model. An appropriate model is one that uses source term data of the type provided by the test. For example, the test may produce data that (i) show an approach to a constant release rate, (ii) actually indicate that a constant release rate has been reached, (iii) are completely random, or (iv) something else. The model must use algorithms that match the type of data produced by the test. In summary, the data provided by this test can be used to model long-term repository behavior, providing that the test can be shown to adequately simulate the repository, the data are reproducible, and the data are appropriate for the model.
- 2) The test will not provide detailed canister corrosion rates that can be used to evaluate canister integrity.
- 3) The test will not provide detailed radionuclide-migration information that can be used to predict transport through tuff.

5.0 APPARATUS, EQUIPMENT, AND ANALYTICAL REQUIREMENTS

[During the isolation period it is assumed that the canister will be breached and that air/water/waste form/metal interactions will occur. These interactions may consist of the waste form being contacted by either water or water saturated air. The contact could be made by either standing or flowing water and could be at a metal/waste form interface or mainly isolated on the waste form. The metal could contain weld-affected and nonaffected regions, and the waste form (glass) will likely be stressed. All the components will have undergone aging during the containment period and will undergo further changes due to reaction during the isolation period. The design of the test apparatus and associated solid materials and procedures creates opportunities for types of interactions to occur, but are specifically not designed to be a reduced copy of the waste package. If focus is needed on a particular interaction, the effect of altering a physical parameter or test procedure can be studied in parametric testing. If appropriate, the test method may be altered according to those results.]

The test apparatus is shown diagrammatically in Fig. 1. The components are the test vessel, which provides for collection and containment of liquid and support of the waste package; the waste package, which consists of the waste form and metallic components representing the canister; the tuff cup, which collects the solution that drips directly from the waste package and which interacts with vapor; the tuff cup holder, which separates the tuff cup from the test vessel; the tuff cup cover, which, when used, keeps the solution from directly dripping into the tuff cup; and a solution feed system, which allows injection of test water. The glass and tuff are discussed in the section on solid materials.

[The eventual release of radionuclides from the waste form/waste package will largely depend on the contact between the liquid and the waste package components. In a leach test this contact is usually not a variable since the waste form is totally immersed in the leachate and there is continuous, uniform contact. In the present test the contact has the potential to be extremely variable and will depend on the flow of liquid through or over the waste package and the forces causing the liquid to be removed from the waste package.]

[The flow will depend on the physical arrangement of the waste package. For the flow to be reproducible in every test, each waste package and its configuration in the test vessel must be identical.]

[Forces causing removal of liquid from the waste package will also depend on temperature differentials in the test vessel that could cause evaporation.]

[The rate of evaporation can be calculated, assuming the evaporative forces are convection and conduction. The importance of convection can be estimated based on the experimental results of [KREITH]. The presence of free convection currents in an enclosed air space can be based on the magnitude of the Grashof number, Gr, which is the ratio of buoyant force to viscous force:

$$Gr = \rho^2 g \beta (T - T_\infty) L^3 / \nu^2$$

where $\beta = (\rho_\infty - \rho) / [\rho(T - T_\infty)]$, coefficient of thermal expansion

ρ = density of air

Fig. 1. NNWSI Interaction Test Apparatus
(a) Cutaway view
(b) Schematic
(c) Photos of Selected Parts

g = gravitational acceleration

ν = viscosity of air

L = characteristic length, here taken as width of the air space

$T - T_{\infty}$ = temperature difference between the two surfaces, test vessel and waste form, as the worst case.

Substituting some typical properties for air, at 90°C, and simplifying the expression for the Grashof number yield

$$\begin{aligned} Gr &= \rho^2 g \frac{(\rho_{\infty} - \rho)}{\rho(T - T_{\infty})} (T - T_{\infty}) L^3 / \nu^2 \\ &= \rho g (\rho_{\infty} - \rho) L^3 / \nu^2 \end{aligned}$$

Now $g = 9.8 \text{ m/s}^2$

$\nu = 2 \times 10^{-5} \text{ Pa}\cdot\text{s}$

$L = 1/2 (2.54 \text{ cm} - 1.5 \text{ cm}) \approx 0.5 \text{ cm}$, from dimensions of the test apparatus.

Density can be calculated from $p = \rho RT$ where $R = 287 \text{ J/(kg}\cdot\text{K)}$ for air. At 90°C,

$$\begin{aligned} \rho_{\infty} &= p / RT_{\infty} = \frac{1.01 \times 10^5 \text{ Pa}}{287 \text{ J/(kg}\cdot\text{K)} 363 \text{ K}} \\ &= 0.969 \text{ kg/m}^3 \end{aligned}$$

The density difference $\rho_{\infty} - \rho$ can be calculated as follows:

$$\begin{aligned} \rho_{\infty} - \rho &= \frac{p}{RT_{\infty}} - \frac{p}{RT} = \frac{pT}{RT_{\infty}T} - \frac{pT_{\infty}}{RTT_{\infty}} \\ &= \frac{p}{RT_{\infty}T} (T - T_{\infty}) = \rho_{\infty} \frac{(T - T_{\infty})}{T} \approx \rho_{\infty} \frac{(T - T_{\infty})}{T_{\infty}} \end{aligned}$$

$$\begin{aligned} \text{Then } Gr &= \left[(0.969 \text{ kg/m}^3)^2 (9.8 \text{ m/s}^2) (5 \times 10^{-3} \text{ m})^3 / \right. \\ &\quad \left. 363 \text{ K} (2 \times 10^{-5} \text{ Pa}\cdot\text{s})^2 \right] (T - T_{\infty}) \\ &= 7.92 (T - T_{\infty}) \end{aligned}$$

For a vertical air space, if $Gr < 8000$, heat flows by conduction only and there is no convection. For horizontal air spaces heated from below, if $Gr < 2000$, heat likewise flows by conduction only and there is no convection. Obviously, a very large temperature difference would be required to set up convection currents, orders of magnitude larger than what would exist in the experiment. The main reason for this result is the small dimension of air space.]

[Another estimate of the conditions where convection would be important can be obtained by considering the instability problem of a fluid layer heated from below, as may be the case in the lower part of the test apparatus (or at the top of the glass if it is hotter than its surroundings). The instability can be determined based on the value of the Rayleigh number (very similar to the Grashof number):

$$Ra = \frac{g\alpha\beta}{K\nu} d^4$$

where g = gravitational acceleration

$\alpha = |dT/dz|$, the adverse temperature gradient

β = coefficient of volume expansion

$K = k/(\rho c_v)$, where k = thermal conductivity, ρ = density,
 c_v = heat capacity of air at constant volume

$\nu = \mu/\rho$, the kinematic viscosity of air

d = characteristic length, about 1 cm.

Using typical values for air at 90°C,

$$Ra = \left[\frac{(9.8 \text{ m/s}^2)(2 \times 10^{-2} \text{ m})^4 / (363 \text{ K})(2.064 \times 10^{-5} \text{ m}^2/\text{s})}{\left(\frac{3 \times 10^{-3} \text{ W}/(\text{m}\cdot\text{K})}{0.969 \text{ kg/m}^3 \cdot 722 \text{ J}/(\text{kg}\cdot\text{K})} \right)} \right] \left| \frac{dT}{dz} \right| = (3.05 \times 10^{-1}) \left| \frac{dT}{dz} \right|$$

The critical Rayleigh number is at least 1700 or so, which means that dT/dz must be greater than 56 K/cm for convection to occur (or a temperature difference of at least 56°C between the glass and the stainless steel container).]

[For these reasons, it does not seem possible that convection currents would be present in the test apparatus.]

[Without convection currents, the only other significant heat transfer mechanism is conduction. The question now is how much heat is required to vaporize the water drop every 3-1/2 days and what temperature gradient would be required to transfer the heat?]

[The water drop is ~0.1 mL, or $1 \times 10^{-7} \text{ m}^3$. The heat of vaporization for water at 90°C is $2.28 \times 10^6 \text{ J/kg}$, so we would need to transfer 228 J in 3-1/2 days (assuming the water density is 1000 kg/m^3), or 2.72 J/h.]

[Assuming that most of the heat is transferred in the annular region, the heat flux can be written as

$$q_k = \frac{(T_i - T_o)2\pi kl}{\ln(r_o/r_i)}, \text{ or } T_i - T_o = q_k \cdot \frac{\ln(r_o/r_i)}{2\pi kl}$$

where q_k = heat transferred

r_0 = vessel diameter

r_1 = glass diameter

k = thermal conductivity of air

l = characteristic length for heat conduction, here given by the height of the glass sample

$T_1 - T_0$ = temperature difference between vessel wall and glass.

Using the values for the test apparatus and the required heat flux,

$$T_1 - T_0 = 2.72 \text{ J/h} \cdot \frac{\ln(1.25/0.75)}{2\pi(3 \times 10^{-2} \frac{\text{W}}{\text{m} \cdot \text{K}})(0.02 \text{ m})} \times \left[\frac{1 \text{ h}}{3600 \text{ s}} \right] = 1.02 \times 10^{-1} \text{ K.}$$

A temperature difference of 0.1 K is all that is needed to transfer the required heat. If the temperature differences in the apparatus are kept below 0.01 K, or if an adverse temperature gradient is set up (very slight) to ensure saturated conditions, evaporation due to conduction would probably be eliminated or controlled.]

[These calculations, combined with laboratory experience, indicate that if temperature gradients greater than 0.1°C exist in the test vessel, there is a potential for significant liquid evaporation. For these reasons, considerable concern exists whether the test results will be reproducible, and concerted emphasis has been placed on test vessel/waste package configuration, and on temperature control.]

5.1 Test Vessels

The test vessel consists of a body and a cap (Fig. 2) and, except for dimension A, must be made to the specifications shown in Fig. 2. Dimension A, which is the control for the maximum amount of liquid collectable without contacting the tuff, is nominally set at 4 cm for either defense or commercial glass but can be varied to accommodate greater flow rates or longer collection times as may be required. At the nominal dimension of 4 cm, ~30 mL of water can be collected before wetting the tuff. This is the volume of liquid anticipated for a commercial glass waste form in a one-year test.

The vessel cap is designed, based on experiment, so that condensed water will flow to the sides of the vessel and no spurious drops will contact the waste package.

The vessel cap and body are mated through a gasket seal that must maintain a positive pressure inside the vessel during the test. While the test conditions call for ambient pressure, a small positive pressure will exist due to the vaporization of the injected liquid. This pressure can be determined upon injection of liquid (see procedure section).

BODY

CAP

Fig. 2. Engineering Drawing of NNWSI Phase II Test Vessel
(Dimensions are in inches.)

The gasket must be made of Teflon[™] that has been cleaned according to approved MCC procedures unless testing is done in a radiation field that degrades the Teflon[™]. As of now the magnitude of the maximum permissible total dose has not been defined. If the total dose exceeds the maximum limit, use an inert metal gasket.

The test vessel must be constructed of Type 304 L stainless steel or the current reference canister material. The vessel cap and body may be constructed from stock from different heats, but the specifications of the stock must be documented.

The vessels are reusable and must be pretreated before initial use and before reuse. The vessel pretreatment and cleaning procedures will be documented when they have been determined. Complete (>99%) removal of activity must be demonstrated before reuse. After cleaning, the mass of the top and bottom (with gasket) of the vessel is weighed and recorded.

5.2 Waste Form Holder

The waste form holder is designed to incorporate into the procedure materials interactions that may occur sometime during the isolation period. The waste form holder is shown in Fig. 3 and consists of four circular pieces of perforated stainless steel, two connecting studs, and four support rods. During the test the waste form holder

- 1) maintains the waste form in position to assure uniform initial contact with the incoming water,
- 2) provides the basis for the water/waste form/metal interaction, and
- 3) keeps the contact between the waste package and the test vessel minimized.

The waste form holder must be made to the dimensions indicated and must be made of 20-gauge Type 304 L stainless steel (or reference canister material). The top and bottom discs of the holder must be made by a punch process using 1/8-in. holes on 1/4-in. centers. Before further fabrication, the perforated stainless steel must be deburred.

The top and bottom assemblies are made to accommodate the waste form and to hold it in place. This is accomplished by spot welding two circular pieces together, one of which has an internal hole sized to tightly accommodate the waste form (Fig. 3). The top and bottom sections are aligned and joined by two stainless steel studs that are tungsten-inert gas welded to the bottom. The rods that are used to support the waste form holder in the test vessel are spot welded to the bottom section. The waste form holder must be manufactured by Argonne National Laboratory following the procedure established by the CMT/ANL shops and as documented in ANL quality assurance guidelines, or by equivalent means.

Before use, the holder must be cleaned according to the stainless steel cleaning and pretreatment procedure. The holders may not be reused. The bottom and top must be parallel and must be horizontal when placed in the test vessel.

Before use, the general appearance of the holder must be inspected with a low-power microscope, and the top and bottom sections must be photographed in color with adequate detail to document changes that may occur during testing. After cleaning, the top and bottom sections must be weighed and the masses recorded.

RETAINER

RETAINER TOP

Fig. 3. Waste Form Holder for NNWSI Phase II Test

5.3 Tuff Support Ring

The test is designed to minimize contact between the tuff and the vessel. The tuff must be supported in a tuff support ring (Fig. 4) that holds the tuff cup in position and minimizes contact. The ring must be made of punched 20-gauge Type 304 L stainless steel. The ring must be cleaned and pretreated according to procedures and its mass taken and recorded before testing.

Fig. 4. Diagram of Tuff Support Ring Used in NNWSI Phase II Test

5.4 Tuff Cup Cover

The purpose of the tuff cup cover is, when used, to keep the solution from dripping directly into the tuff cup, thereby potentially providing information concerning aerosol or liquid film transport.

The dimensions of the tuff cup cover are shown in Fig. 5. The cover must be made of Type 304 L stainless steel and must be cleaned and pretreated according to procedures. It must be weighed and its mass recorded prior to testing.

5.5 Feedthrough

The feedthrough is the system used to introduce the liquid to the system. The feedthrough (see Fig. 6) consists of a fitting that must be centered in the vessel lid and must be tapered with the inside of the lid to eliminate areas for dripping onto the waste package. The drop must be introduced through a 0.01 in. ID and 0.0625 in. OD stainless steel tubing whose end is placed 1/2 in. above the waste form and in the center of the waste form. The 1/16-in. tubing must be set up as shown in Fig. 6, i.e., coiled inside the oven and terminated outside the oven to accept liquid. The tubing must be valved so that during the test period there is no direct access to the test vessel. No condensation of liquid is permitted in the feedthrough system.

The solution must be introduced by a method capable of forcing 0.075 ± 0.005 mL of it and 0.5 mL of air through the tube and onto the waste form. Performance of the feedthrough systems must be verified for each test apparatus before testing begins.

5.6 Temperature Control

The test apparatus is maintained at a preset temperature during the test by containment in an environmental chamber. This chamber must control the temperature of the test vessels to within 0.5°C or 0.5% (whichever is greater) of the set temperature. A temperature recorder or other monitoring device

Fig. 5. Diagram of Tuff Cup Cover Used in NNWSI Phase II Test

must be provided to ensure that the desired temperature has been maintained for the duration of the test. Additionally, the temperature variation within the chamber must be such that the variation in temperature in the volume occupied by each test vessel is less than 0.05°C . This latter variation must be measured with the vessels in place before the beginning of the test but need not be verified during testing. When radioactive specimens are used, the self-heating of the waste form must be reported, and its effect on the test must be explained. Brief fluctuations from the desired temperature are undesirable. Only when specimens are removed or added to the chamber should the chamber be disturbed, and any intrusion should be as expedient as possible. Temperature fluctuations and durations during intrusion must be recorded.

The chamber must accommodate the test vessel so that it rests with its bottom horizontal and is immovable during the test. Also, the vibrations of the surface must be less than $5\text{ }\mu\text{m}$.

5.7 Balances

Balances must provide the accuracies indicated below, depending on the materials being weighed:

Waste package components (waste form and holder)	$\pm 20\text{ }\mu\text{g}$
Test apparatus and components	0.01% of the total mass*
Chemical reagents and final solutions	$\pm 1\%$ of the mass

*[This is required to measure the amount of water collected during a test period and the amount of water retained on the waste package. Typical values are 1000 g total, 1 g water added, and 0.2 g on waste package.]

Fig. 6. Feedthrough System Used in NNWSI Phase II Test. The top photo is the inside of the oven, and the bottom photo is the top outside of the oven.

5.8 Volume Measurement

Test solution volumes are generally introduced with a calibrated syringe. This syringe must be accurate to within 5%. Volumes used for taking analytical samples or preparing solutions are calibrated as described in Table 1 and are to be accurate to $\pm 2\%$ or better.

Table 1. Required Calibration Schedule

Measurement	Device	Frequency Check and Methods
Temperature	Thermocouple or thermometer	6 mo NBS standard or ice/boiling water
	Thermocouple probe	6 mo Against a calibrated millivolt source
Length	Micrometer	6 mo Standard foils, gauge blocks
Mass	Balance	3 mo NBS standard masses
Chemical concentration	Analytical method	3 mo NBS standards 2 times daily Secondary standards
pH	pH meter	Daily with commercial buffer solutions After every 25th sample
Volume	Volumetric flasks	Use certified flasks 3 mo by measuring the mass of pure water contained
Syringe and pipettes		3 mo by measuring the mass of pure water transferred
Activity	Counting techniques	Twice a day NBS standard isotope Source of interest Secondary standards Background (weekly)

5.9 Solution Analysis

Measure solute concentrations using equipment standardized with standards traceable to NBS. Determine and report precision and accuracy as defined in ASTM E 177-71. Although analytical results should normally be accurate to within $\pm 10\%$, this may not be possible when concentrations in the solution approach detection limits. The detection limits for each analysis must accompany the reported result. When the result is less than a factor of 10 greater than the detection limit, use the following reference as a guidelines for determining detection limits: Anal. Chem. 52:2242-2249 (1980).

5.10 pH Measurement

Measure the pH to an accuracy of ± 0.1 units using a calibrated meter. Use ASTM D 1293-78 Method A and commercial buffers to make this measurement.

5.11 Surface Science Equipment

Requirements will be developed.

5.12 Calibrations and Standards

Calibrate all instruments used in these tests initially, and periodically, to minimize possible errors due to drift. Table 1 shows the methods and the minimum frequency of calibration for the various devices used. Use standardization procedures that are published by recognized authorities such as the NBS or the ASTM.

6.0 PREPARATION AND STORAGE OF SOLUTIONS AND CHEMICAL

6.1 General Chemicals

Use chemicals of reagent grade or better that conform to the specifications of the Committee on Analytical Reagents of the American Chemical Society.

Waste forms having very low leach rates may require the use of ultrapure chemicals in the makeup of reagents and in the cleaning of reagent storage containers, accompanied by appropriate modifications in laboratory technique. The results for the leaching of a given element are considered to be adversely affected and may not qualify for entry in the Nuclear Waste Materials Handbook (U.S. Dept. of Energy Report DOE/TIC-11400) if the amount of that element introduced to the leachate by reagent impurities exceeds 10% of the amount of that element leached from the specimens.

6.2 Equilibrated J-13 Water

The test solution is equilibrated J-13 water. This must be prepared and stored as follows:

- 1) Obtain a rock core from the repository horizon and demonstrate that it contains no caliche.

- 2) Crush the rock in a manner demonstrated to add no contaminants, and collect the fraction that passes through 100-mesh screen. Do not remove the fine fraction.
- 3) Place 10 g of crushed rock in a clean 1-L Teflon™ container.
- 4) Add ~100 mL of J-13 water (obtained from NNWSI) and shake by hand for two minutes. Allow the rock to settle for one hour, decant, and analyze the water for anions. If the anion content shows evidence of caliche in the rock, start the process over with fresh rock.
- 5) Fill the bottle with J-13 water allowing room for expansion, cap tightly, and place upright in a 90°C oven.
- 6) Shake the bottle for one minute twice a day for one week (five working days).
- 7) Allow the bottle to remain in the oven undisturbed for a second week.
- 8) After two weeks, remove the bottle from the oven and allow to air cool for one hour, then filter through filter paper known to add no measurable impurities during filtering. Discard the filter.
- 9) Refilter the solution through a 0.1- μ m filter known to add no measurable impurities to the solution.
- 10) Store the filtered solution in the dark in a non-reactive container (see section 6.4). The solution should be stable for several months but should be analyzed before use or every two weeks, whichever is appropriate.
- 11) The equilibrated water must have a composition for each species within 20% of the values shown in Table 2.

Table 2. Composition of Equilibrated J-13 Water

Comp., μ g/mL		Comp., μ g/mL	
F ⁻	2.3	Al	0.7
Cl ⁻	7.2	B	0.18
NO ₃ ⁻	7.6	Ca	6.0
SO ₄ ²⁻	17.3	Fe	0.02
		Li	0.05
		Mg	0.60
		Na	45
		Si	37
		Sr	0.04

6.3 Water

Other water used in rinsing the waste package or in diluting test solutions is reagent water Type I or II in ASTM D 1193-77, which has a total impurity level, including organics, of less than 0.1 mg/L. Analyze the water every 6.5 weeks when it is used in preparing solutions for analysis.

6.4 Solution Storage

Use polyethylene or polypropylene bottles with tight-fitting lids to store J-13 water. Before use, rinse these bottles with three volumes of 6M HNO₃, each rinse equal to 0.2 of the container volume; three volumes of water, each equal to 0.2 of the container volume; and two volumes of freshly prepared solution, each equal to 0.1 of the container volume. Store the solution in the dark.

Check the pH after the final rinse to ensure that there is no residual acid. If the pH is not within 0.1 unit of the water, continue rinsing until it is.

7.0 PREPARATION AND CHARACTERIZATION OF SOLID MATERIALS

The test method is applicable to inorganic monolithic waste forms made according to specifications described below. Emphasis has been placed on glass waste forms. Spent fuel will be addressed in FY 85, and the procedure will be modified accordingly.

[Because of the emphasis the test places on investigating materials interaction, the manufacture of waste form and other solid materials are strictly controlled. For the waste form (glass), the chosen description applicable after 300/1000 years of confinement includes: (1) surfaces that are representative of fracturing and/or casting that occurred. These surfaces would be smooth and stressed. Both parameters have been shown to influence the interaction between glass and water [BATES, MCC]; (2) surfaces that are in contact with stainless steel in the vicinity of the breached area. In this region, there may be a glass/stainless steel interface available for water intrusion between the glass and metal. It is also likely that the composition of the glass and metal would be altered due to contact during the molten glass pouring stage (the consequences of this interaction will be considered); and (3) devitrification that could occur during cooling or storage. Each of these effects will be considered when manufacturing the waste form.]

7.1 Preparation of the Waste Form Specimen

For a glass waste form, the composition studied must be representative of aged glass (radioactive decay) that would occur during the containment period. The glass must be made to the following specifications:

Sample Dimensions - The sample must be a tapered right circular cylinder 2.0 ± 0.1 cm in length. The diameter of one face must be 1.5 ± 0.03 cm and the diameter of the other face must be 1.6 ± 0.03 cm. The faces must be parallel. Determine the geometric surface area of each sample after preparation.

Method of Manufacture - The samples must be cast in a Pt-5% Au mold. The glass used in the final casting must have been homogenized prior to casting to ensure homogeneity of the melt (follow the MCC glass preparation procedure, for example). Pour the glass into a preheated mold ($\sim 700^{\circ}\text{C}$, just slightly red) that is completely enclosed in a mold/firebrick assembly. Immediately transfer the assembly to an annealing oven (500°C) for 15 min; then cool at $\sim 100^{\circ}\text{C/h}$ to 350°C ; then remove the mold from the firebrick and allow it to cool to room temperature. The mold can be reused until the glass removal becomes difficult and the cast glass surfaces become scratched. This method is used to produce smooth, stressed surfaces for testing.

Method of Cutting - The top and bottom faces of the cylinder must be cut on a Buehler low speed cut-off saw with a low-concentration diamond blade #11-4254, using a speed setting of six, a balance weight of 125 g, and water as the lubricant.

[This procedure produces the two types of desired surfaces. The composition of these surfaces may be different. Spinels, typical of glass devitrification, normally form on the surface in contact with the Pt-Au mold but are not noted on the cut surface.]

Cleaning - Clean the specimens by the following procedure:

- Complete one 5-min ultrasonic wash in high-purity water.
- Complete three 5-min ultrasonic washes in fresh absolute ethanol.
- Dry to constant mass using the appropriate balance and record mass. Use a drying technique that has been demonstrated to be applicable to the specific waste form being tested. One hour at 110°C is sufficient for most nonporous waste forms. Porous waste forms may require higher temperatures and longer times.

7.2 Characterization of Waste Forms

If a complete test matrix is run, 12 waste form specimens are required. During the waste form manufacturing process, an additional waste form must be made to be used for characterization and must be saved as an archive sample. Complete characterization must be done on this archive sample. The remaining samples after cleaning must be examined with a low-power optical microscope to document the appearance of both the cut and as-cast surfaces. A color photograph of a typical sample and all samples showing variations is required. Variations may exist in the as-cast surface appearance due to degradation of the mold. These variations are cause for rejection of the sample only if the surface no longer has an "as-cast" appearance. The variations are generally signified by scratches. Samples must also be rejected if chips exist at the as-cast/cut surface interface. Air bubbles are allowed on either the as-cast or cut surface provided their contribution to the total surface area is $< 5\%$.

The archive sample, which must have an appearance similar to the other samples, must undergo characterization prior to the beginning of testing. Differences in surface microstructure, morphology, composition, and porosity between the as-cast and cut surface must be documented. For α -emitting radioactive samples, total α -counting must be done on both types of surfaces to determine the distribution of α -emitting radionuclides.

7.3 Preparation of Tuff Cup

The tuff cup must be made to the dimensions shown in Fig. 7, using core drills and cutoff wheels with 200-grit or finer diamond impregnated cutting surfaces. Use water as the cutting fluid. The tuff cup must be made of caliche-free tuff from the NNWSI repository horizon. Before manufacturing the tuff cup, test the tuff for caliche by shaking the rock in J-13 water and analyzing for anions. Evidence for caliche is anion levels greater than are present in J-13 water. After manufacture, test each rock cup in the same manner. If caliche material is found in the rinse solution, either discard the tuff cup or continue cleaning it in deionized water until no caliche is present.

Prepare the tuff cup for testing by soaking it in J-13 water overnight. Remove the cup from the water immediately before use and weigh and record its mass while wet.

Fig. 7. Diagram of Tuff Cup Used in the NNWSI Phase II Test

7.4 Characterization of the Tuff Cup

No characterization of the cup prior to use other than described above is necessary.

7.5 Specimen Handling

All handling of specimens after preparation and cleaning must be done with tongs, tweezers, or lint- and dust-free plastic or rubber gloves.

8.0 PROCEDURE

The test matrix consists of three different types of tests and a blank. Procedures are given for the different types of tests in Section 8.2. The experimenter must run the entire test matrix.

8.1 Quality Assurance Requirements

The procedures and methods used in this test and the personnel performing the test must conform to all applicable quality assurance requirements of the laboratory performing the test.

8.2 Test Matrix

The NNWSI interaction allows no variability in the parameters used in the test. These parameters are strictly defined and depend on the waste form being tested. Only by varying waste forms can other test parameters be altered. Defense waste processing facility (DWPF) glass has been used in preliminary testing of the test method, and the test matrix described below is for use with DWPF glass.

The test matrix contains three different types of tests, depending on the purpose of the test and the components that are included in this test:

- 1) This test contains a waste package, a tuff cup, and a tuff cup holder. Its purpose is to determine the release of radionuclides and other selected elements from the glass, to provide selected information about radionuclide/tuff interaction or migration, and to provide selected information about waste form/ canister interaction. This is a batch-type test.
- 2) This test contains a waste package. Its purpose is to determine the release of radionuclides and other selected elements from the glass, to provide selected information about the waste form/canister interaction, and to provide means of continuing the test for an indefinite period.

[The inclusion of tuff in the test method complicates sampling, yet it is necessary to study migration and is necessary if the tuff has any, at present unknown, effects on the degradation of the waste form. Tuff does influence the composition of the J-13 water, but this is accounted for by preequilibration. If it can be shown that tuff has no influence on the release from the waste package then, except for migration studies, tuff could be eliminated. If it can be shown that continuous testing results in the same release as batch testing, then the continuous test can be continued with confidence.]

- 3) This test contains a waste package, a tuff cup, a tuff cup support ring, and a tuff cup cover. Its purpose is to measure the release from the waste package and to investigate transport modes that may exist in the test vessel.

[Presumably the only radionuclides that are not collected during each sampling period are those associated with the waste package or the tuff. Only indirect contact between the tuff and the solution is possible, i.e., aerosol transport and thin liquid film transport. It is hoped that this test will provide some information about these two possibilities.]

The test is:

Conditions for DWPF Glass Testing

Glass composition:	Determined by experimenter
Glass dimensions:	Defined in section 7.1
Temperature:	90°C
Pressure:	Slight positive pressure
Solution:	Reacted J-13 water
Solution volume:	0.075 mL
Solution introduction interval:	3.5 days
Test vessel:	Defined in section 5.1
Matrix:	Table 3

8.3 Test Procedure

This procedure also applies to blanks, except that the waste package and tuff are omitted. The volume of solution introduced in the blank is exactly the same as used in the actual tests. The test procedure calls for either terminating a test after a specified period or sampling and continuing the test. Instructions are given for both scenarios.

1. Prepare the materials and components used in the test as described in the previous sections. The components must be cleaned, weighed, and prepared prior to testing. Each vessel lid and bottom must be identified and matched to a specific test in the matrix.
2. Take an aliquot of J-13 water for cation, anion, and pH analysis. Submit the aliquot for analysis.
3. Add 0.5 mL of J-13 water to each test vessel.

Table 3. Test Matrix for Phase II Tests

Test No.	Components	Test Duration, weeks								
		0	6.5	13	19.5	26	32.5	39	45.5	52
1	tuff, waste package	_____								
2	tuff, waste package	_____								
3	tuff, waste package	_____								
4	tuff, waste package	_____								
5	tuff, waste package	_____								
6	tuff, waste package	_____								
7	tuff, waste package	_____								
8	tuff, waste package	_____								
9	tuff, waste package, cup	_____	+	+	+	+	+	+	+	+
10,11	waste package	_____	+	+	+	+	+	+	+	+
12	blank	_____	+	+	+	+	+	+	+	+
13	blank	_____								

+ = periodic change
| = termination

Analyses Required^a

Total radionuclide (solution and tuff when present)	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓	✓✓✓✓	✓✓	✓✓✓✓✓
Total matrix element (solution Li, B, Cs)	✓✓✓	✓✓✓✓✓	✓✓✓	✓✓✓✓✓	✓✓✓	✓✓✓✓✓	✓✓✓	✓✓✓✓✓	✓✓✓	✓✓✓✓✓
Wt loss and surface analyses		✓✓		✓✓		✓✓		✓✓		✓✓✓✓✓
Corrosion of stainless steel		✓✓		✓✓		✓✓		✓✓		✓✓✓✓✓
Migration in tuff		✓		✓		✓		✓		✓
"Vapor phase" transport (includes partial radionuclide and tuff interaction information)	✓	✓	✓	✓	✓	✓	✓	✓	✓	✓

^aA check means this analysis is required; the number of checks indicate replicate analyses possible.

4. Add the components appropriate to each test to the test vessel, making sure each component fits as required. The glass must be oriented with the larger diameter surface facing up. The necessary components are indicated in the test matrix.
5. Weigh the assembled test vessel without the closure fitting.
6. Seal the test vessel with the closure fitting.
7. Place the test vessel in the environmental chamber (preset and operating at 90°C) and connect the feedthrough to the drop insertion system. Record the time (h:min). The testing period starts when all the test vessels for a given matrix have been placed in the chamber.
8. After two hours, add the required amount of solution to the vessel. For defense glass add 0.075 mL of solution. After injecting the solution, inject four times the solution volume in air. Close the vent in the injection system.
9. Every time period, inject the required amount of solution. The time period for injections depends on the waste being tested. For defense waste the time period is every 3.5 days. The time period for other wastes, i.e., commercial glass or spent fuel, will be provided later. Insert the syringe into the addition port before opening the vent. Open the vent and note whether there is positive pressure in the test vessel. Inject the solution and remove the syringe with the vent open. Inject four times the solution volume in air into the vessel. Remove the syringe and close the vent. Restrict the liquid loss on opening the vent by making sure the syringe is in place before opening the vent. If the vessel is not under positive pressure, note this and retighten the lid and the connect fittings once. Control the sample injection time to the designated time period to within $\pm 3\%$ of the time interval.
10. Control the test period to within ± 4 hours of the time that the test was scheduled to end or be sampled. Do not inject more solution before the test is sampled or terminated. At the conclusion of the test period, either terminate or sample the test. Record the time and date. The method of removing the test vessel from the oven is the same for both. Disconnect the feedthrough from the injection system and place a tared sealing nut on the feedthrough. Place the test vessel in an ice bath, being careful not to tilt the vessel. Cool the bottom of the vessel to room temperature for 1-1/2 min. Do not ice the top.
11. Remove the closure nut and weigh the test vessel.
12. To terminate the test, carefully remove the vessel lid. Note: there may be water condensed on the lid. Retain this water. Carefully document observations concerning the location of water and condition of the test vessel. Immediately inspect the waste-form holder and the waste form itself for the location and approximate amount of water present. Note any obvious changes in the appearance

of the waste package. Retain all water on the waste package during this process. Remove the tuff cup from the vessel and weigh immediately. If there is water present in the tuff cup, pipette it into a clean, tared container. Place the tuff cup in dust-free storage for later investigation. Rinse the waste package with 10 mL of water. The object of this rinse is to transfer the solution on the waste package to the test vessel. Everything that can be rinsed from the waste package is considered as having been removed from the waste package. Note and retain any solids that are collected during this rinse or that have accumulated during the test. Inspect the solution and test vessel for residual solids. If solids are present, remove them by filtration through a 0.45- μ m or finer membrane filter, and return the clarified solution to the test vessel for acidification. The solids must be weighed, analyzed, and reported with the test data. After rinsing, separate the waste package components and place them in a dust-free environment and dry at room temperature to a constant weight. Weigh each waste package component and maintain the components in a dust-free environment for future investigation.

[Because of the small amount of solution generated in this test, especially for the initial testing periods, and because of the low concentration of many constituents, no aliquots are withdrawn for separate analyses. The main goal of the test is to measure the release of radionuclides and selected other elements from the waste package, and it has been demonstrated that conservation of the undiluted test solution best accommodates this goal. If, in tests of long duration, enough solution exists, aliquots can be taken for other analyses. This question will be addressed later.]

- 12a. For those tests that continue, i.e., tests 9, 10, and 11, proceed as follows. Remove and cool the test vessel as described in step 12. Weigh the vessel after removing the closure nut. Carefully open the test vessel and make observations concerning the appearance of the vessel and the waste package. For tests 10 and 11 (waste package only) remove the waste package, make observations concerning appearance, and place the waste package in a clean, tared test vessel to which 0.5 mL of the J-13 water has been added. Place the lid on the test vessel and weigh it. Seal the test vessel, place it in the oven, and connect to the feed system. After two hours add the appropriate amount of J-13 water and continue as indicated in the test matrix. Add 10 mL of water to the original test vessel, acidify to 1% by volume and proceed as in step 12.
- 12b. For test No. 9, proceed as in step 12a up to removing the waste package. Place the waste package in a new tared vessel to which 0.5 mL of J-13 water has been added and weigh. Then remove the waste package and transfer the tuff cup to the new vessel. Use a new tuff cup support ring and a new tuff cup cover. Weigh the vessel including the tuff cup. Add the waste package to the vessel and proceed as in step 12a.
13. Acidify the solution in the test vessel to 1% by volume with nitric acid. The tuff cup support ring must be in the acidified solution, as should the tuff cup cover, if present. Seal the vessel. Shake

the solution in the vessel so that it contacts and wets the entire vessel. Place the vessel in an oven at 90°C for 12 hours. After cooling, transfer the solution to a separate polyethylene container and submit the solution for appropriate analyses. These analyses must include determination of actinide, fission product, and other cations of interest.

14. For tests in which the tuff was removed from the vessel, analyze the radionuclide content of the tuff. The total amount of each species removed from the waste package must include the contribution from the solution and from the tuff. A tuff blank must be analyzed to determine the composition of the tuff. The tuff analyzed must come from remnants of the tuff cup manufacturing process or from tuff identical to that used in testing.

9.0 INTERLABORATORY TESTING

Because of the complex nature of the materials interactions testing, there is a particular need to establish that the data produced by the test method is reproducible. Tests done at different times by a given laboratory or by different laboratories may not be conducted under the same experimental conditions. Interlaboratory tests are needed where the NNWSI interaction test is performed with a specific glass composition by separate laboratories using the NNWSI interaction test procedure. The interlaboratory tests are, in essence, replicate tests except that they are not necessarily run at the same time or by the same laboratory.

Data derived from several properly documented interlaboratory tests should be similar with a distribution that can be statistically treated to determine overall precision. Erratic or widely varying data from interlaboratory tests indicate that the parameters have not been well controlled or that additional unknown parameters are in need of control.

10.0 CALCULATIONS

[The NNWSI interaction test introduces two avenues whereby species can be introduced into the final solution other than by interaction with the waste package. These are via solution/test vessel and solution/tuff interactions. Preliminary testing has demonstrated that only elements unique to the glass or elements whose contribution comes predominately from the glass can be used to assess the degradation of the glass. Elements that are introduced into solution due to both glass/solution and some other solution interaction cannot be profitably studied. This is because the amount of many species introduced by the solution/glass interaction is small compared to that introduced by the other solution interaction. The elements that can be attributed to release from the glass are all radionuclides and B, Li, and Cs. The formalization of this conclusion is discussed later.]

10.1 Use of Blank Data

Blank data are measurements from tests conducted in which only the solution initially present and that which has been incrementally added to the test vessel are included. Conditions, except for the omission of solid

phases, are the same as in the interaction test. The blank, therefore, measures the degree of interaction between the solution and the test vessel. The purpose of the blank is to determine corrections that need to be made to individual element concentrations due to contributions from the solution/test vessel interaction. In tests done where no tuff is present, solution concentrations are corrected by subtracting the corresponding blank concentration.

[Preliminary testing has indicated that the blank contribution results from dissolution of stainless steel presumably during the acid wash step. No radionuclides or unique glass components (B, Li, or Cs) are released because of solution/test vessel interaction. Therefore, only two blanks are required. One blank measures the solution/test vessel interaction during the continuous test (tests Nos. 10 and 11), and one blank measures the solution/test vessel interaction for the longest batch test. Blank corrections to the batch tests cannot be made until the blank is completed, and this will represent a conservative correction.]

For the test runs in which tuff is present in the test vessel, no exact blank correction is possible. This is because, on the scale used in the test, tuff cannot be considered homogeneous. Thus, the elemental contribution to solution from the tuff may be different for each test. Therefore, pending further study, only those elements unique to the glass or those that can be shown to come predominately from the glass can be attributed to release from the glass. The composition of tuff taken from the repository vicinity, but not the exact horizon, is given in Table 4. The composition of J-13 water equilibrated with tuff is also given in Table 4.

Table 4 indicates that tuff and J-13 water contain immeasurable quantities of transuranic actinide elements or of radioactive fission products. Any amount of these elements present in the test samples would have to be released from the glass. Tuff does contain measurable amounts of B, Li, and U. However, J-13 water that has been reacted with tuff under a variety of conditions contains small and constant amounts of B and Li and no U or Cs. Therefore, any U or Cs found in the test solution is attributable to the glass and any B or Li in excess of that found in reacted J-13 water is attributable to release from the glass. The effect of tuff would be to decrease the amount of B, Li, U, or Cs in the test solution but not increase it. Correct the solution concentration by subtracting the corresponding blank concentration.

It may be possible to make similar conclusions about other elements that are released from the waste package, and this will be pursued in further testing.

10.2 Weight Change

Report the weight change for the waste package components. Calculate a normalized mass loss for the waste form using the following equation

$$(NL)_m = \frac{\Delta m}{SA}$$

where Δm is the mass change for the waste form, and SA is the surface area of the waste form.

Table 4. Composition of Tuff and Reacted J-13 Water

	Composition ^a							
	Al	B	Ba	Ca	Fe	K	Li	Mg
Tuff ^b	6.57	0.027	0.005	0.39	0.71	4.42	0.002	0.032
J-13 ^c	0.70	0.18	<0.01	6.0	0.02	NA ^d	0.05	0.60

	Composition ^a							
	Mn	Na	Si	Sr	Ti	Zn	Zr	U
Tuff ^b	0.053	2.85	-	0.002	0.047	0.006	0.007	0.00034
J-13 ^c	<0.005	44.0	37.0	0.04	<0.005	0.020	<0.005	<0.00005

^aNo transuranic actinides of radioactive fission products were detected.

^bComposition by wt %.

^cComposition by ppm in solution ($\mu\text{g/mL}$).

^dNA - Not analyzed.

[The surface area includes the faces that have been in contact with the perforated stainless steel and the as-cast surfaces. The ratio of as-cast to cut surface areas could affect the value of $(NL)_m$, and the degree of any effect is being investigated in parametric testing.]

10.3 Element and Radionuclide Release

Report the concentration of all elements in solution in units of $\mu\text{g/mL}$. Report the blank corrected concentrations of all elements in solution in units of $\mu\text{g/mL}$.

The total amount of elemental release may be a combination of several analysis, e.g., solution and tuff. Report the results from separate analyses and the sum of all analyses.

10.4 Calculations

Calculate and report the average of all replicate data and the standard deviation. Use the average data in plots, and show the standard deviation as error bars.

[It is hoped that results (elemental release from the waste package) for each type of test, i.e., tests with tuff, tests without tuff, and tests with the tuff cover, will be identical and can be used as replicate samples. This will have to be demonstrated. If this proves not to be the case, additional replication may be required.]

Note any experimental errors encountered while performing the experiment and the expected effect on the results. Also, note any sources of systematic errors and biases. Estimates of these quantities must be reported and considered in any statement of accuracy and precision.

It is recommended that one out of every 20 solutions submitted for analysis shall be a blind specimen that is an aliquot of a standard solution. Include a summary of such blind specimen results in the Error Analysis Section of the data report.

11.0 Reports

A standard format for reports will be developed. Although adherence to the format is not required, such a format will allow the MRB to more easily evaluate the data that are submitted.

IV. INTERIM PHASE II TEST RESULTS

A long-term Phase II test using defense glass (165 black frit) doped with uranium, cesium, and strontium was started 6/14/84. The glass composition is given in Table 5. This series of tests is being done according to the test procedure described in Section III. The test matrix and required analyses are shown in Table 3. Test Nos. F-9 and F-10 were sampled at the 6.5-week interval on 7/30/84 and at the 13-week interval on 9/13. Test Nos. F-1 and F-2 were terminated on 9/13/84. It was recently decided to add tests F-11 and F-12 to the matrix, and these were started 9/13/84.

Table 5. Composition, in element wt %, of SRL Glass
Used in Phase II Testing

Al	B	Ca	Fe	Li	Mg	Mn	Na	Ni	Si	Cs	Sr	U
2.17	2.11	1.07	8.60	2.18	0.48	2.25	7.64	0.71	25.29	0.10	0.13	0.99

Selected analyses were done for tests F-1, F-2, F-9, and F-10 and are reported in Tables 6 and 7. Surface analyses of the waste package components are in progress, but have not been incorporated into this report. A final interpretation of the test results cannot be accomplished until all analyses have been done and the test matrix completed. However, some qualitative observations can be made and the data can be compared to earlier results from prototype and parametric testing [STEINDLER-1, -2].

The qualitative observations are:

- 1) The waste package appearance was similar, with one exception, to that observed in previous testing. The top surface had a very thin layer

Table 6. Selected NNWSI Phase II Test Results

Test Description	Test No.	Time, weeks	Δ Sample Mass, 10^{-5} g	Δ Top Can Mass, 10^{-5} g	Δ Bottom Can Mass, 10^{-5} g	Δ Tuff Cup Mass, g	Δ Mass Total, ^a g	SA, cm ²
Waste package, tuff	F-1	13	-35	-15	+39	+0.07	-0.35	13.62
Waste package, tuff	F-2	13	-32	+4	+2	+0.15	-0.25	12.86
Waste package	F-9	every 6.5	NA ^b	NA	NA	NA	NA	
Waste package, tuff, tuff cover	F-10	every 6.5	NA	NA	NA	NA	NA	

^aThis value is the weight difference between the "initial" weight of the vessel (vessel + components + J-1.3 water added during the test) and the final weight.

^bNA - not applicable.

of water present, which dried within 30 seconds of opening the vessel. The sides of the glass were watermarked, and a considerable amount of standing water was around the bottom of the waste package. The exception is that in previous prototype and parametric testing much more water was on the top of the package. Associated with the lack of water on the top of the waste package is that no semicircular watermarks are observed around the top of the as-cast/cut interface. These semicircular watermarks are associated with standing water that collected between the canister and glass. In the prototype and preliminary tests, the smaller diameter face of the glass was at the top of the waste package. This created a gap of ~ 1 mm between the glass and the canister at both the top and bottom faces. However, in the final Phase II testing, the large diameter face of the glass was at the top. This created a tight fit between the canister and the glass at the top and an ~ 2 mm gap at the bottom. This was done to lock the glass in place during the test and to promote even flow down the sides of the glass. A photograph of the sample from parametric test No. 10 is shown in Fig. 8, and the watermarked areas are clearly visible. The ramifications of these differing water/glass/canister contact modes are evident in the weight loss and solution analysis data discussed later.

- 2) Considerable reaction occurred between the glass and the bottom part of the canister, as evidenced by discoloration of the glass and the canister. The same type of reaction was also seen in the prototype tests [STEINDLER-2]. This reaction was focused mainly, but not exclusively, around the weld-affected area of the canister. A considerable amount of rust-colored flakes was present in the solution. A before and after photograph of the bottom section of the waste form holder from test F-1 is shown in Fig. 9, and the reacted areas are clearly visible. Examination of these reacted areas in prototype testing indicated that the reacted areas consist of iron and chromium silicates [STEINDLER-2].

Table 7. Solution Analyses from NNWSI Phase II Tests

Test No.	Normalised Mass Losses, g/m ² ^a				Total Mass of Selected Elements in Solution, ng									
	(NL) _{wt}	(NL) _g	(NL) _{Li}	(NL) _U		Al	B	Ca	Li	Mg	Na	Si	Sr	U
F-1	0.26	0.14	0.20	0.008	Initial solution	5360	4290	32500	6070	5360	106000	72700	500	110
					Blank ^b	1910	440	11000	160	950	114000	86500	70	10
					Corrected	3450	3850	21500	5910	4410	(8000) ^c	(13800)	430	100
F-2	0.25	0.22	0.22	0.022	Initial solution	3920	6420	41300	6420	6770	141700	113500	680	285
					Blank	1910	440	11000	160	950	114000	86500	70	10
					Corrected	2010	5980	30300	6260	5820	27700	27000	610	275
F-9					6.5-week analysis	2440	1310	6560	<730	930	28800	25150	<360	<18
					13-week analysis	4620	<1070	15650	<1070	1670	25600	44500	<360	<18
F-10	0.09	0.31	0.005		6.5-week analysis	<1820	1820	5100	<730	1020	24800	25900	<360	<18
					1 st week analysis	3550	1170	15400	9130	2200	74900	61900	<170	71
					13-week total	3550	2990	20500	9130	3220	99700	87800		71
					Blank	2300	530	13280	190	1150	137500	104100		11
					Corrected	1250	2460	7220	8940	2070	(37000)	(16300)		60

^aOnly those elements whose release can be mainly attributed to the glass are considered.^bThe blank is based on the amount of J-13 water added to the system and not on a specific blank analysis.^cValues in parentheses are negative.

Fig. 8.

Photograph of Waste Form Taken
from Parametric Test No. 10a

- 3) In both tests F-1 and F-2, there was standing water in the tuff cups. This had not been previously observed. The standing water was poured into the test vessel for analysis, but the test procedure has been changed so that in the future that liquid will be withdrawn from the tuff cup and analyzed separately.
- 4) Weight loss measurements were made for the waste package components. In previous experiments, weight loss from the waste form was quite reproducible and provided evidence concerning the degree of reaction undergone by the waste form. In the present experiments, the normalized weight losses, $(NL)_{wt}$, for tests F-1 and F-2 are similar (Table 7) and are ~ 8 times less than was observed in 12-week parametric testing. The reduction in weight loss can be attributed to the reduced amount of water/glass contact that occurs at the upper surface, and possibly to the formation of alteration products on the glass surface.
- 5) As demonstrated in previous testing [STEINDLER-2], if tuff is present in the test, the degree of waste form reaction can be measured by the amount of Li, B, and U in solution. Lithium provides the most consistent results likely because it does not interact with any of the test components. Values for Li release from tests F-1 and F-2 gave a normalized elemental loss of $\sim 0.2 \text{ g/m}^2$ based on the total surface

Before Testing

After Testing

Fig. 9. Bottom Section of Waste Form Holder from Test F-1
Before and After Testing

area of the waste form. Boron release, as had been observed previously [STEINDLER-3] was more erratic; nevertheless, the $(NL)_B$ values, 0.14 and 0.22 g/m^2 , are not too different from $(NL)_{L1}$, which would be expected. Uranium release, as based on solution analysis, was quite low, and varied between $(NL)_U$ of 0.008 and 0.022 g/m^2 . Uranium is expected to interact with both tuff and stainless steel and an analysis of these components must be done to fully describe the uranium behavior.

Release of Li, B, and U can be compared to that found in prototype and parametric testing [STEINDLER-2]. As was observed for the waste form mass loss, the release of these individual elements is also lower in Phase II testing. This is rationalized as being caused by less water/glass interaction occurring at the top portion of the waste package in the Phase II tests.

The release of some elements that are present in both the glass and in either the tuff or the stainless steel is presented in Table 7 but not discussed.

- 6) Part of the data replication in the Phase II test matrix is based on identical materials interactions occurring for the tests that are terminated and the tests that are periodically changed. Test F-10 can be compared to tests F-1 and F-2. This comparison, when done for B, Li, and U, indicates that some deviations exist. The best match based on solution analyses is for B and Li. Since there may be some standing water on the waste package that is not analyzed during periodic sampling, the (NL)_i values may be slightly reduced in test F-10. Evidence from previous tests [STEINDLER-2] indicates that U may interact with tuff, so that in tests F-10 and F-11, which contain no tuff, there should be more U in solution than for tests F-1 and F-2. This was not observed. It is expected that further testing will provide better correspondence between the different types of tests.

Based on the above observations, a preliminary conclusion of the Phase II test method is that reproducible materials interactions have been achieved for the terminated tests (F-1 and F-2). For these tests, waste form degradation is similar, based on the overall appearance of the waste package, normalized mass loss from the waste form, and normalized elemental losses for those elements representative of the waste form.

Also, the usefulness of periodic sampling to provide replicate samples for the terminated tests, and to provide a method of continuing the test period indefinitely, without altering the results, can only be judged with further testing.

V. ANALOG EXPERIMENT

The purpose of the analog test is to measure the degradation of the waste package under conditions that closely simulate the actual repository. The analog test is less rigorously controlled than the Phase II test and may provide an indication as to the degree of realism that has been incorporated into the Phase II test.

The analog test utilizes a tuff rock core the shape of a right circular cylinder, 5-in. length and 2-1/2-in. diameter, as a reactor vessel. Several cores were drilled from a large piece of outcrop tuff taken from Fran Ridge (near the repository horizon in Nevada). Each core was cut diametrically to give two sections as shown in Fig. 10. A cavity was machined in the larger section (Fig. 10) to accommodate a waste package similar to that used in the

Fig. 10.

Diagram of Analog Test Apparatus
and Rock Core

Phase II test. The dimensions of the cavity were similar to the inside of the Phase II reaction vessel, except that the second section of tuff, which acts as the vessel lid, had a flat lapped surface.

The test is initiated by placing the waste package in the cavity and placing both sections of J-13 saturated tuff into a Teflon™ sleeve. The mating surfaces of both tuff sections had lapped faces which produced a good seal. The Teflon™ sleeve was placed in a stainless steel holder, and end caps were secured over the top and bottom of the tuff core. The end caps were tapered so they only contacted the tuff along the outside edges. The end caps were connected to inlet and outlet tubing. The entire system was sealed by water, which pressurized the Teflon™ jacket to 1600 psi (11 MPa). This constricted to flow to the tuff core and not to the Teflon™/tuff interface.

The input of water to the tuff core was supplied by a storage reservoir of water that was heated to $\sim 130^{\circ}\text{C}$. The inlet line was also heated. The goal was to supply the tuff core with an evenly distributed supply of water vapor. The rate of flow through the tuff core could be regulated by the temperature (pressure) of the storage reservoir. Water, after passing through the tuff core, was collected in a vented bottle for analysis of radionuclides, cations, and anions.

Prior to actual testing, the system was assembled without a waste package. This allowed the tuff core to be pretreated to remove caliche and provided experience in adjusting flow rates. The system was run in this fashion for 30 days, and the flow rate through the core stabilized at ~1.2 mL/day using an inlet temperature of 180°C. Upon disassembly standing water was found in the cavity. The amount was ~6 g and would not have directly contacted the waste package, but it was disconcerting enough to suggest that a lower flow rate be used. The outlet water was analyzed and the results indicated that the amount of caliche was declining but still was measurable. However, the core was judged to be usable.

The first analog test was initiated with a projected flow of ~0.2 mL/day. This is roughly the worst-case flow that would be expected through a bore hole containing defense waste processing facility (DWPF) waste. The initial water temperature was set at 140°C. A DWPF glass spiked with ¹³³Ba, ¹³⁷Cs, and ¹⁵²Eu was used as the waste form. ¹³⁷Cs was used to represent highly mobile, easily leached elements, but is not expected to be a hazardous radionuclide during the isolation period. ¹³³Ba was used to represent the alkaline earth group of elements. Again, few of these elements will be a hazard during the isolation period. ¹⁵²Eu was used to represent fission products and actinide elements that are slowly removed from the glass and do represent hazards during the isolation period. The levels of these radionuclides doped into the glass are given in Table 8. These levels were chosen based on previous experience, but with care taken to err conservatively for ¹³⁷Cs, since too much ¹³⁷Cs in the reacted liquid will interfere with the detection of the other radionuclides.

Table 8. Radiotracer Levels^a in DWPF Analog Glass

Radiotracers	Primary γ-Radiation, keV (Photons/100 decays)	Activity/g Glass, μCi
¹³³ Ba	356.0(62), 81.0(31.7), 302.8(18.1)	0.453
¹³⁷ Cs	661.7(85.0)	0.116
¹⁵² Eu	1408.0(21.3), 181.8(29.1), 344.3(27.2)	13.7

^aBased on γ-ray analysis of a 1.287 g sample of glass.

The test began 5/8/84 and was terminated 8/29/84. It took until ~5/29/84 for constant flow to be established through the rock core, so the actual time interval of fairly constant flow was 3 months. The weights of the components at the beginning and at the end of the test are given in Table 9, while the volume of solution collected and flow rates are given in Table 10.

The test was terminated after ~3 months of constant flow. The rock core was not saturated at the beginning of the test (total weight = 1195.6 g; saturated weight = ~1206 g), so it took ~3 weeks for the core to become saturated and flow to begin. The core was carefully disassembled, and observations were

Table 9. Results of Analog Test

Component	Initial ^a Weight, g	Final Weight, g	Dry Weight, g	Δm , g
Tuff top	469.10 (1195.6) ^b	474.10 (1206.7) ^b	(1173.0) ^b	
Tuff bottom	726.48	732.59		
Waste package	17.70	17.72		
Canister top	3.32343		3.32356	+0.00013
Canister bottom	4.19959		4.19983	+0.00024
Glass	10.17473		10.17459	-0.00014

^aDuring pretreatment the core was soaked in J-13 water for 4 days and had weight of 1205.1 g. It was then vacuum dried at 110°C to constant weight of 1173. After 30 days with a flow of 1.2 mL/d, it had a weight of 1207.2 g.

^bOriginal dry weight.

Table 10. Flow Rate Determinations for Analog Test

	Sampling Periods ^{a,b}					
	5/29	6/4	6/8	6/14	6/22	6/29
Liquid volume, mL	2.39	1.75	1.41	1.80	3.15	3.09
Flow rate, mL/h	—	0.012	0.014	0.013	0.016	0.018
	Sampling Periods					
	7/9	7/16	7/23	7/30	8/13	8/29
Liquid volume, mL	6.78	3.51	2.47	2.41	5.63	7.87
Flow rate, mL/h	0.028	0.021	0.015	0.014	0.017	0.020

^aTest begun 5/8/84, terminated 8/29/84.

^bConstant flow appears to begin ~5/29/84.

made of all components. The components were weighed immediately upon disassembly and again after drying. The waste package was rinsed with ~10 mL of high-purity water, which was analyzed for cations and radionuclides. The tuff sections were γ -counted and the bottom section was cut diametrically just above the bottom of the cavity for further counting. Of interest was whether the radioactivity was located at the bottom of the cavity or spread over the walls, bottom, or top of the cavity. The waste package holder was also γ -counted. The analytical results are given in Table 11.

Table 11. Analytical Results from the Analog Test

Component	Species Analyzed												Cl ⁻	Fe ⁻	NO ₃ ⁻	SO ₄ ²⁻
	Al	B	Ca	Li	Mg	Na	Si	Sr	U	¹⁵² Eu ^e	¹³⁷ Cs ^f	¹³³ Ba ^g				
Pretreatment sol'n ^{a,b}	<130	3500	33000	260	2400	95000	140000	260	NA ^h	NA	NA	NA	30	50	52	87
Eluent 5/8-6/4 ^b	<350	8900	39000	270	2100	107000	83000	350	<0.5	NA	NA	NA				
Eluent 6/14-6/29 ^b	<350	5900	21000	150	900	73500	85000	170	<0.5	NA	NA	NA				
Eluent 7/16-7/30 ^b	<350	5400	12000	<150	700	51000	77000	100	<0.5	NA	NA	NA				
Eluent 7/30-8/13 ^b	NA	NA	NA	NA	NA	NA	NA	NA	NA	0	0	0				
Waste package rinse ^c	<1100	<650	3275	7400	420	43660	<2200	<200	238	6E-3	0	8E-4				
Tuff top										2E-4	0	ND ⁱ				
Tuff bottom 1 ^d										1E-3	2E-3	4E-3				
Tuff bottom 2 ^d										4E-4	0	ND				
Canister										7E-3	1.5E-3	5E-3				
Total										1.4E-2	3.5E-3	9.8E-3				
1 mL J-13 water	780	180	4500	64	400	46600	35300	30		0	0	0	2.3	7.2	7.6	17.3

^aSolution collected at the end of the 30-day pretreatment step.^bIn ng/mL.^cTotal ng in solution.^dThe bottom of the tuff core was sectioned into cavity bottom and cavity wall sections. Tuff bottom 1 is the cavity bottom and tuff bottom 2 is the cavity wall.^eγ-counting based on 1408 keV peak, values are blank corrected counts per second, counting data are preliminary.^fγ-counting based on 661 keV peak, values are blank corrected counts per second, counting data are preliminary.^gγ-counting based on 356 keV peak, values are blank corrected counts per second, counting data are preliminary.^hNA = not analyzed.ⁱND = could not be determined.

Several observations can be made:

- 1) The tuff was damp and dark upon disassembly, and the disassembly weight indicated that saturation to those levels obtained on prolonged soaking was achieved. The method of maintaining flow through the core does not cause extra water to be incorporated into the tuff. The degree of saturation is presumably similar to that in the unsaturated zone. This point will be addressed when a complete description of the repository site is available.
- 2) Upon disassembly, the waste package was visually dry on the top and wet on the bottom. The sides of the glass had watermarks similar to those observed in the final version of Phase II testing. Overall, the waste package had an appearance identical to that in a Phase II test.
- 3) The weight loss from the glass was 0.14 mg after 90 days of testing. This value corresponds to a normalized weight loss of 0.1 g/m^2 . In Phase II testing, the weight loss after 90 days of testing is 0.35 mg or $(\text{NL})_{\text{wt}}$ of 0.25 g/m^2 . These values are in remarkable agreement.
- 4) The element releases, as measured by what was rinsed from the waste package (Table 11), are also similar to values for waste package rinses reported in earlier testing [STEINDLER-2]. The divergence for the solution values between the parametric tests and the Phase II tests is greater than the weight loss values because waste package rinse values represent those species on the waste package at a particular moment, which is not reproducible between tests.
- 5) Release of ^{152}Eu , ^{133}Ba , and ^{137}Cs was found to occur. These radionuclides were found either in the waste package rinse or on the bottom of the collection cavity. No radioactivity was found on the top of the cavity or on the walls of the cavity. These results indicate that the radionuclides were transferred from the waste package to the tuff by dripping from the waste package. There is no direct evidence for aerosol transport or liquid film transport that was suggested in previous parametric testing. Normalized release values for these elements are $(\text{NL})_{^{152}\text{Eu}} = 0.03$, $(\text{NL})_{^{137}\text{Cs}} = 0.2$, and $(\text{NL})_{^{133}\text{Ba}} = 0.09$. These values are below the 1 part in 10^5 required by the NRC, and are in the range calculated from Phase II test solution analyses. Of interest is what the release trend is for longer time periods.
- 6) The canister sections, particularly the bottom section, show signs of corrosion. The weight gain is due to reaction with components of the glass and the J-13 water and occurs at the glass/stainless steel interface. This type of corrosion was noted in earlier prototype testing and in actual Phase II testing and may be due to crevice corrosion.

The glass and stainless steel components are undergoing continued inspection using secondary ion mass spectrometry and scanning electron microscope/energy dispersive X-ray analysis techniques. These results will be presented later.

The results of the 90-day analog test are very similar to Phase II test results for similar time periods. However, they are the results of only one test and may represent only a portion of possible materials interaction. Analog tests that will run for six-month and one-year durations are in progress. The results of these tests will be compared to Phase II test results, and if the analog test can be shown to be a reasonable simulation of the repository, then it may be possible to judge the relevance of the Phase II test to the repository site.

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