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Dear Mr. Wick:

Biannual Report for FIN A-3167 Program on "Review of Waste  
Package Verification Tests" (NUREG/CR-3091, Vol. 5, 1984)

Attached please find a draft of the subject report. Your comments would  
be greatly appreciated.

Sincerely,

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REVIEW OF WASTE PACKAGE VERIFICATION TESTS  
BIANNUAL REPORT

H. Jain  
E. Veakis  
P. Soo

August 1984

NUCLEAR WASTE MANAGEMENT DIVISION  
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REVIEW OF WASTE PACKAGE VERIFICATION TESTS  
BIANNUAL REPORT

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Manuscript Completed August 1984

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## ABSTRACT

This ongoing study is part of a task to specify tests that may be used to verify that engineered waste package/repository systems comply with NRC radionuclide containment and controlled release performance objectives. Work covered in this report includes crushed tuff packing material for use in a high level waste tuff repository. A review of available tests to quantify packing performance is given together with recommendations for future testing work.

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## Executive Summary

This Biannual Report describes an on-going effort to review performance verification tests needed to characterize high level radioactive waste packages and their components. The purpose of the present study is to identify generic types of tests that would be needed in order to quantitatively assess the behavior of packing materials in a waste package under conditions likely to be encountered in a tuff repository environment.

Generic test methodologies identified for the purpose of quantifying tuff repository packing material performance in the unsaturated zone include the following:

1. Tests to determine the mineralogic stability of the packing material under high temperature steam-water environments, including the characterization of alteration products.
2. Tests to determine the rate of steam or water migration, as well as gaseous or volatile fission product migration through tuff packing material due to spent fuel cladding failure.
3. Radionuclide sorption tests under steam and aqueous conditions in order to address the contribution of packing material to the controlled release criterion.

The high-temperature-steam conditions that will exist for the first several hundred years in the vicinity of the waste package will result in changes in the chemical composition of tuff groundwater. Tuff alteration products, deposited salts and thermal and radiolysis effects will result in an altered groundwater chemistry. Tests are recommended to bracket the anticipated tuff repository water chemistry and this would serve to identify water chemistries to be used in waste package test programs. This approach would result in a more realistic assessment of groundwater composition effects than the simple high-temperature tests involving tuff and J-13 well water currently in use by DOE subcontractors.

In the early years after repository closure, the packing material will be exposed to a high-temperature steam environment. Mineralogic stability for this period can be determined by exposing the packing material to steam at the anticipated repository temperature. To accelerate the kinetics, fine-powder samples of the packing material may be heated in a closed vessel to higher temperatures. In subsequent periods of time, when liquid water will be in contact with the packing material, an autoclave system may be used to accelerate alteration by testing at higher temperatures.

Test methods are required to determine the permeability of steam through tuff packing material. In principle, a vertical column-type system can be used to determine the flow of steam through packing material of varying compaction density. Radionuclide transport during this steam phase also needs to be addressed. Sorption tests may incorporate the use of permeability cells using cylinders of compacted tuff/clay mixtures. Test conditions should be designed to provide long-term exposure of the packing material to high-temperature steam-air environments using both closed (sorption) and open (flow-through) system tests.



## 1. INTRODUCTION

The NRC Rule for the Disposal of High Level Waste in Geologic Repositories (10 CFR 60) dated June 1983, specifies two main performance objectives for the engineered barrier system:

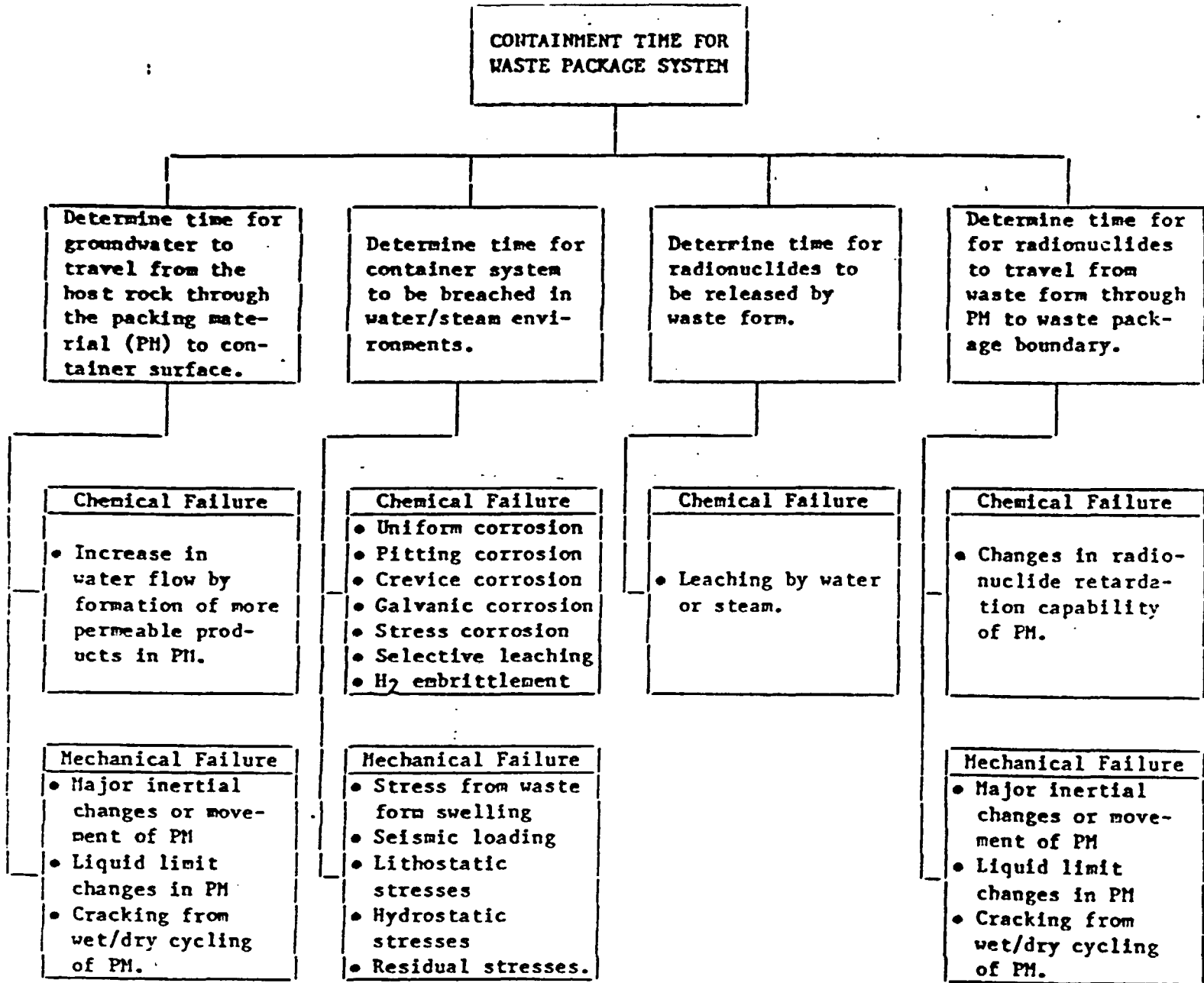
Containment of HLW within the waste packages will be substantially complete for a period to be determined by the Commission taking into account the factors specified in subsection 60.113(b) (of 10 CFR 60) provided, that such period shall be not less than 300 years nor more than 1,000 years after permanent closure of the geologic repository; and

The release rate of any radionuclide from the engineered barrier system following the containment period shall not exceed one part in 100,000 per year of the inventory of that radionuclide calculated to be present at 1,000 years following permanent closure, or such other fraction of the inventory as may be approved or specified by the Commission; provided, that this requirement does not apply to any radionuclide which is released at a rate less than 0.1% of the calculated total release rate limit. The calculated total release rate limit shall be taken to be one part in 100,000 per year of the inventory of radioactive waste, originally emplaced in the underground facility, that remains after 1,000 years of radioactive decay.

In order to show compliance with these performance objectives the license applicant will need to provide a data base and analyses to quantify anticipated behavior of the waste package/repository system after permanent closure. This will necessarily involve research and testing programs to evaluate the likely modes by which engineered system components will degrade or fail by chemical or mechanical means. Knowledge of the ways in which the engineered barriers fail will permit estimates to be made regarding the containment capability of the waste package and the radionuclide release rate from the engineered system. Below is listed a sequence of events leading to loss of containment and the release of radionuclides:

- a. Groundwater enters the engineered barrier system
- b. Groundwater penetrates the geologic packing material
- c. Groundwater penetrates the container/overpack system
- d. Groundwater leaches radionuclides from the waste form
- e. Radionuclides are transported through the failed container/overpack system, packing material and disturbed host rock to the near field environment.

Figures 1.1 and 1.2 outline the failure/degradation modes which may occur during the sequence of events leading to loss of containment and the release of radionuclides. Chemical and mechanical failure modes are given for each of the barriers in the engineered system. The ones ultimately found to be applicable will depend on specific waste package/repository designs, temperatures, water chemistry and flow rate, design geometries and sizes, and materials selected, etc.



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Figure 1.1. Chemical and mechanical failure/degradation modes affecting containment of radionuclides by the waste package system.

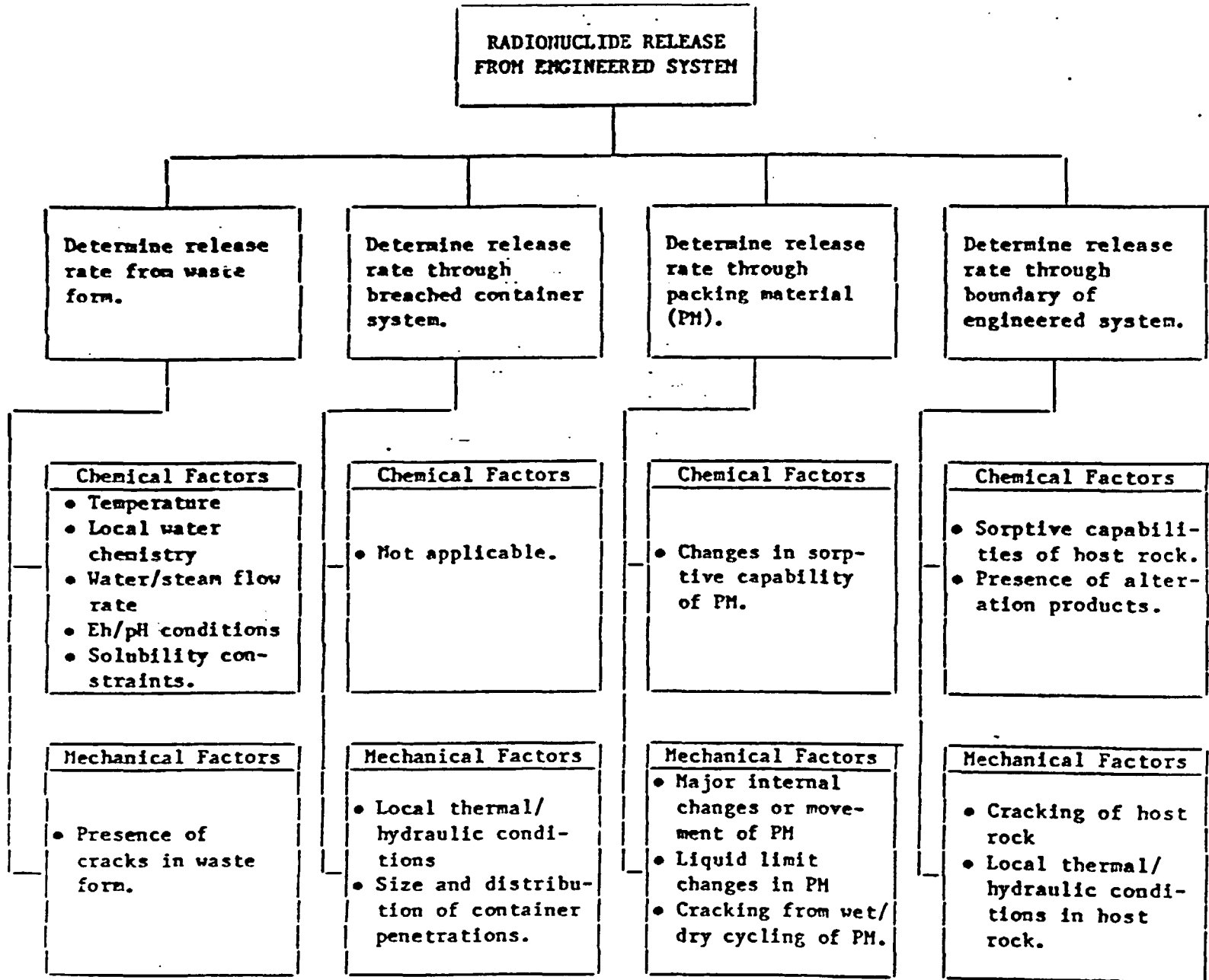


Figure 1.2. Factors affecting radionuclide release from the engineered repository system.

The objective of the current study is to identify the types of test that may be used to demonstrate compliance with the containment and radionuclide release performance objectives for engineered barrier systems. In particular it will be necessary to address how groundwater chemistry, pH and redox potential change as a result of high temperature reaction with package components and on irradiation since these parameters influence the nature of container corrosion. After containment has failed, and radionuclides are released from the waste form, the effects of radionuclide sorption by packing material, water chemistry, water flow rate, and temperature, etc. on the rate of radionuclide release from the engineered barrier system need to be quantified to determine whether regulatory criteria can be met.

Figure 1.1 serves as the basis for specifying containment verification tests for waste package components. Currently, borosilicate glass and spent fuel waste forms, carbon steel, titanium-based and Type 304L stainless steel containers, and bentonite-based, tuff-based, and crushed salt packing materials are emphasized for evaluation in basaltic, tuffaceous, and salt repositories. The types of tests recommended to show compliance with NRC criteria outline the test variables to consider and the test methodology. Where appropriate, tests being developed by the Materials Characterization Center (MCC) are evaluated and any limitations specified. Future research that may be necessary to develop additional testing methodologies is given also.

In the current report, test procedures for evaluating the performance of crushed tuff packing material will be discussed. Because the tuff repository horizon is in the unsaturated zone, there is a need to evaluate packing behavior in the steam/air environment which will be present during repository operations, the retrieval period, and the early post-closure period.

## 2. WASTE FORM TESTING REQUIREMENTS FOR DEMONSTRATION OF COMPLIANCE WITH THE CONTAINMENT AND CONTROLLED RELEASE CRITERIA

This part of the program for borosilicate waste glass has been completed and is reported in a prior Biannual Report entitled "Review of Waste Package Verification Tests" (NUREG/CR-3091, Vol. 2, BNL-NUREG-51630, 1983).

## 3. CONTAINER SYSTEM TESTING REQUIREMENTS FOR DEMONSTRATION OF COMPLIANCE WITH THE CONTAINMENT CRITERION

This part of the program has been completed for titanium alloy Grade-12 (TiCode-12) and carbon steel and is reported in prior Biannual Reports entitled "Review of Waste Package Verification Tests" (NUREG/CR-3091, Volumes 1 and 3, 1983, respectively).

#### 4. CRUSHED TUFF PACKING MATERIAL TESTING REQUIREMENTS TO DEMONSTRATE COMPLIANCE WITH THE RADIONUCLIDE CONTROLLED RELEASE CRITERION

The Yucca Mountain area in Nevada is the principal site for a HLW tuff repository. The geology, hydrology and geochemistry of this area have been described in prior work (NUREG/CR-2937, 1983). The information on design and other characteristics of this repository is sparse compared to basalt and salt repositories. In early designs, the repository horizon was situated in the saturated zone. However, it was recently decided to place HLW packages in the unsaturated zone where container corrosion and waste form leaching is expected to be much lower. Specifically, the proposed repository horizon is located 400 m below the surface and 170 m above the static water table, which is in the lower portion of the densely welded (but highly fractured) Topopah Spring Member of the Paintbrush tuff formation and just above the basal vitrophyre layer (Bish, D. L., 1983). Therefore, some of the data obtained earlier for the saturated zone may no longer be relevant.

Although the composition of the packing material to be used with a spent fuel waste package has not been defined, presumably it will consist of crushed tuff with a small fraction of binding material such as clay. Typically, this may be a mixture of crushed tuff and 15 percent or less of iron-bearing smectite clay consolidated to about 75 percent of theoretical density (UCID-19926, 1983). To evaluate the effect of clay addition, it will be useful to compare available results on tuff/clay compositions with those on crushed tuff alone. In an earlier report (NUREG/CR-3091, Vol. 2, 1983), packing material testing methods pertinent to a basalt repository environment have been discussed in detail. Due to some similarities in the properties of basalt and tuff, much of the discussion regarding basalt repository packages may also be applicable to the tuff repository waste package, although the test conditions may be different in the two cases. Therefore, the present report emphasizes the differences expected for packing material tests for tuff systems, and only outlines other pertinent tests which have been discussed in detail in earlier BNL work (NUREG/CR-3091, Vol. 2, 1983).

##### 4.1 Repository Conditions Relevant to Packing Material Testing

The repository parameters which should be known to conduct tests on packing material are temperature, gamma radiation dose and dose rate, pressure, hydraulic properties and the groundwater chemistry. Information regarding these parameters has been reviewed in a prior Biannual Report (NUREG/CR-3091, Vol. 4, 1984).

##### 4.2 Geochemistry and Water Interactions for Tuff/Clay Packing

Mineral composition, compaction density/pressure, water content, hydraulic conductivity, thermal conductivity, swelling pressure, thermal and radiation stability, and sorption of radionuclides are the properties of a packing material which should be known to evaluate its performance. These have been outlined in a prior Biannual Report (NUREG/CR-3091, Vol. 4, 1984).

#### 4.3 Test Methods for Evaluating Tuff Packing Material

In order to quantitatively determine the characteristics of crushed tuff/clay packing material, a series of generic tests have been identified that are specific to the conditions likely to be encountered in a tuff repository. The properties of tuff required for any assessment of packing material performance have been addressed in a previous BNL report and will not be repeated here (see NUREG/CR-2482, Vol. 5, 1984). The focus of this effort is to outline test methodologies capable of generating quantitative data on several parameters of importance in predicting packing material performance during a single test. Tests for mineralogic stability, for example, can also be used to generate data on alteration products resulting from water-steam-rock interactions, radiation-thermal effects, and wet-dry cycling. Test methods considered for water-steam migration can also be designed to evaluate such parameters as hydraulic conductivity, steam-gas transport and the effects of packing material compaction density.

The primary areas in the development of test methodologies required for a quantitative assessment of tuff packing material behavior include:

- (1) Tests to determine repository water chemistry.
- (2) Tests designed to assess the mineralogic stability of packing material. These tests should take into account the formation of alteration products due to high temperature interaction with steam-water-air mixtures in the presence of a radiation field which may cause acid generation (see below).
- (3) Tests designed to assess rates of steam-water transport through packing material. Breakthrough times obtained from these tests may be used to determine probable times of contact between steam-water and the waste container. These migration tests can also address changes in transport rate due to alteration products formed in the packing material, density effects, radiolysis effects, and the effects of thermal gradients.
- (4) Radionuclide sorption tests for the purpose of addressing NRC's controlled release criterion. On the assumption that containment fails during the period when steam conditions prevail, testing should address the possibility of steam-phase transport of radionuclides (e.g. gaseous and volatile fission products due to spent fuel cladding failure) through packing material. More conventional sorption tests are available to address aqueous transport of radionuclides as steam condenses at some point down the thermal gradient or as the repository cools below the boiling point of groundwater. Additional parameters, such as the presence of alteration products or variations in compaction densities mentioned earlier, may be incorporated.

The test methods presented in the following sections attempt to address the unique conditions likely to be encountered in a tuff repository. In instances where test procedures for basalt repository conditions are applicable to tuff, the similarities are noted and the reader is referred to an earlier report for details (see NUREG/CR-3091, Vol. 2, 1983).

#### 4.3.1 Tests to Determine Repository Water Chemistry

Earlier work showed that high temperature reaction with tuff can alter groundwater chemistry significantly, as shown in Table 4.1. However, Oversby (UCRL-90045, 1983) has recently concluded that there is little change in water composition when J-13 groundwater is reacted with pre-washed tuff for 64 days at 150°C. These tests were conducted by reacting crushed tuff with groundwater in a teflon reaction vessel or a Dickson type gold cell rocking autoclave. The latter system was preferred because in the former case teflon absorbs CO<sub>2</sub> and, therefore, gives erroneous pH results. Only a limited range of tuff compositions have been investigated to date. It will be necessary to continue similar tests on tuff specimens which cover a more complete range of host rock compositions.

Table 4.1. Interaction of tuff with groundwater to determine change in groundwater after reaction with hot rock (150°C, 4+ days). (DOE/NWTS-30, 1982)

Element	Initial Water Concentration (wg/mL)	Results to Date, µg/mL
Si	31	140
Na	48	100
Ca	12	5
K	5	6-14
Al	0.01	0.2
pH	7.0	8.5

The autoclave type tests mentioned above give the anticipated groundwater chemistry from simple high temperature rock-water interaction without considering any concentration effects arising from steam formation. Water percolating towards the hot repository horizon will boil at about 95°C since the tuff is porous and pressures just below atmospheric prevail. Steam formation will deposit constituents previously dissolved as the water percolates down from the surface. As the repository cools, however, fresh liquid water will probably redissolve the precipitated salts and may yield water more concentrated than the J-13 composition. These water chemistries, which have been

influenced by the degree of dissolution of deposited salts, high temperatures, and gamma irradiation, will need to be specified for meaningful container corrosion, packing material, and waste form evaluations.

One might expect that when all the deposited salts are eventually redissolved, (this may not be complete if they chemically interact with the tuff) the water chemistry could approach that anticipated for a simple tuff/water reaction. Thus, the anticipated tuff repository water chemistry to be used in waste package test programs would have a range of chemical compositions. One testing approach would be to use groundwater in equilibrium with the host rock at repository temperatures and, also, a highly concentrated solution of this groundwater whose composition has been determined by prior host rock-water tests which quantify the dissolution of deposited salts.

To obtain experimental data on the solute concentration effects, a test design could be employed in which J-13 water is allowed to percolate down a column of crushed tuff. The column could be subjected to a thermal gradient such that the water encounters higher-temperature rock as it descends. Under atmospheric pressure conditions, steam would be formed at the 100°C isotherm in the column where it would deposit salts. Long-term testing will be required to allow significant salt precipitation and alteration in the tuff/salt column. The column temperature could be cooled periodically to a temperature just below the boiling point to collect liquid effluent for analysis. This could help determine how the water chemistry and pH change with time.

Gamma irradiation should also be considered and could be incorporated into this type of column test. The radiolysis of a steam/O<sub>2</sub>/N<sub>2</sub> system could yield nitric oxides and then nitric acid. Low pH is generally known to deleteriously affect container corrosion rates, waste form leaching, and clay packing material integrity.

#### 4.3.2 Tests for Mineralogic Stability

Mechanical as well as chemical stability of packing material under the range of repository conditions is a basic requirement. The principal chemical failure mode for tuff packing material is mineralogic alteration assuming that the alteration products have reduced sorption and hydraulic properties (NUREG/CR-2482, Vol. 5, 1984). In general, fracturing, hydrologic erosion, creep and liquefaction are considered the major mechanical failure modes for a clay-containing packing material (NUREG/CR-3091, Vol. 2, 1983). Testing methods for these failure modes are discussed below with specific consideration for a tuff repository. A more general review can be found in earlier reports (NUREG/CR-3091, Vol. 2, 1983; NUREG/CR-2482, Vol. 4, 1982; Eastwood, D., 1983).

Recent work has focused on tuff taken from drill holes at Yucca Mountain for the purpose of determining the alteration of welded glassy tuff following deposition (Levy, S. S., 1984). Alteration due to naturally occurring conditions was viewed as providing a natural analog to the processes likely to occur as a result of waste-induced thermal effects. Possible waste-induced



alteration, based on evidence obtained on natural alteration products, include: "(1) glass dissolution without in situ crystallization of secondary minerals, (2) in situ crystallization of glass to feldspar and cristobalite, (3) in-situ crystallization of glass to smectite and/or heulandite, and (4) crystallization of heulandite and smectite as fracture filling" (Levy, S. S., 1984). The formation of zeolites and smectites, along water flow paths, as a result of elevated temperatures arising from the emplaced waste, may enhance sorption of radionuclides. Crystallization of the above-mentioned minerals may also act to decrease porosity and permeability within rock fractures. While this study of natural alteration processes adds useful information to an overall assessment of long-term repository behavior, alteration processes and alteration product behavior should be confirmed under appropriate experimental conditions. In addition, experimental investigations of tuff alteration need to address the effects of steam/air/gamma irradiation environment on the formation and composition of these products.

A laboratory procedure for evaluating tuff packing alteration might involve the exposure of a prototypic packing to maximum anticipated temperatures in a tuff repository ( $\approx 230^{\circ}\text{C}$  for the steam/air period, and  $\approx 95^{\circ}\text{C}$  in the liquid water period) for a sufficiently long period of time to correlate the amount of alteration with time. Periodic removal of samples for examination would permit a determination of the type and quantity of alteration products, changes in moisture content, and porosity, etc. Such tests should also include the presence of gamma irradiation since, in the presence of air, nitric acid is likely to form and this may have a significant effect on surface alteration.

Since it is very likely that the kinetics of alteration under repository conditions is too slow to be quantified within a laboratory time scale, accelerated testing may become necessary. In general, increasing temperature will accelerate the kinetics, but it should be shown that it does not cause a change in the alteration mechanism since this would invalidate extrapolation of results to repository temperatures. To study the effects of radiation, higher dose rates may be used to accelerate irradiation effects. Preliminary tests on bentonite clay show that gamma radiation causes changes in its hydraulic conductivity and swelling properties, but they tend to saturate at high total doses (PNL-4452, 1983).

Assuming that clay minerals, which swell by absorbing water, are only a minor constituent of tuff packing, swelling is unlikely to cause any detrimental effects on the waste package. In fact, such swelling would tend to fill pores and crevices in the packing without exerting large pressures on the waste package itself. This will improve compaction density and, therefore, may increase sorption and decrease hydraulic conductivity. However, if a large portion of the packing consists of clay, swelling pressures will need to be measured to determine their effect on waste package design. Pusch (KBS-TR-73, 1978), Chan (1981) and LeBell (1978) have developed test cells to determine swelling and extrusion properties of bentonite under ambient conditions. These methods have been reviewed earlier (NUREG/CR-3091, Vol. 2, 1983) and can probably be adopted to test tuff packing without major modifications.

Due to the small hydraulic head and groundwater flow rate in the proposed tuff repository, hydrologic erosion of packing would not be expected to be a major problem. However, under a driving force of 260 cm of hydraulic head, Chan (1981) found severe erosion of bentonite. Similar testing for tuff packing could also be conducted over a range of hydraulic conditions so that extrapolation to tuff repository conditions can be achieved. The test method developed by Pusch (SKBF-KBS-79-01, 1979), and schematically shown in Figure 4.1, can also be used to determine the rate of erosion of packing materials under fixed test conditions. Both of these methods have been reviewed in an earlier BNL report (NUREG/CR-3091, Vol. 2, 1983). It is suggested that these tests be performed at temperatures as high as the boiling point of groundwater ( $\approx 95^{\circ}\text{C}$  at the repository horizon).

#### 4.3.3 Tests for Groundwater Migration

It is important to determine the time taken by groundwater to migrate through the packing material to the overpack. The major parameters to be included in the determination of the flow rate are temperature, compaction density, hydraulic gradient, radiation, and composition of packing material.

As discussed in an earlier BNL report (NUREG/CR-3091, Vol. 2, 1983), the flow of water can occur due to a hydraulic gradient (Darcy's law) or by molecular diffusion under a concentration gradient (Fick's law). The second mechanism is believed to be dominant at low rates of fluid flow and, therefore, may be important for the case of a tuff repository waste package. However, due to capillary forces the flow of water may deviate from these laws during initial wetting of the packing material until pores are filled with groundwater. Because of the possibility of wet-dry cycling of the packing material in a tuff repository, a test method should also be developed to determine how such cycling will influence cracking of the clay component of the packing and associated water transport rates.

In principle, a dynamic test methodology used for sorption measurements, as described in Section 4.3.5.2, could be adapted to measure hydraulic conductivity, although no such measurements on tuff packing seem to have been reported. Also, the permeability cells developed for measuring hydraulic conductivity of basalt/bentonite packing (PNL-3873, 1981; Pusch, R., 1979) could be used for tuff packing tests. In fact, the test apparatus for tuff can be considerably simplified because, in this case, it is not necessary to test under high pressure conditions. Alternatively, a multi-sectional column (schematically shown in Figure 4.2) developed by Pusch (SKBF-KBS-79-01, 1979) can be used to study the rate of water flow during initial wetting of the packing material. In this case the ingress of water may be determined by examining individual sections of the column periodically.

In a repository the packing material will be exposed to thermal gradients which may influence the rate of water migration to the container. Siskind and others (NUREG/CR-3091, Vol. 3, Appendix D, 1984) have studied the flow of groundwater through a mixture of basalt and bentonite under both hydraulic and

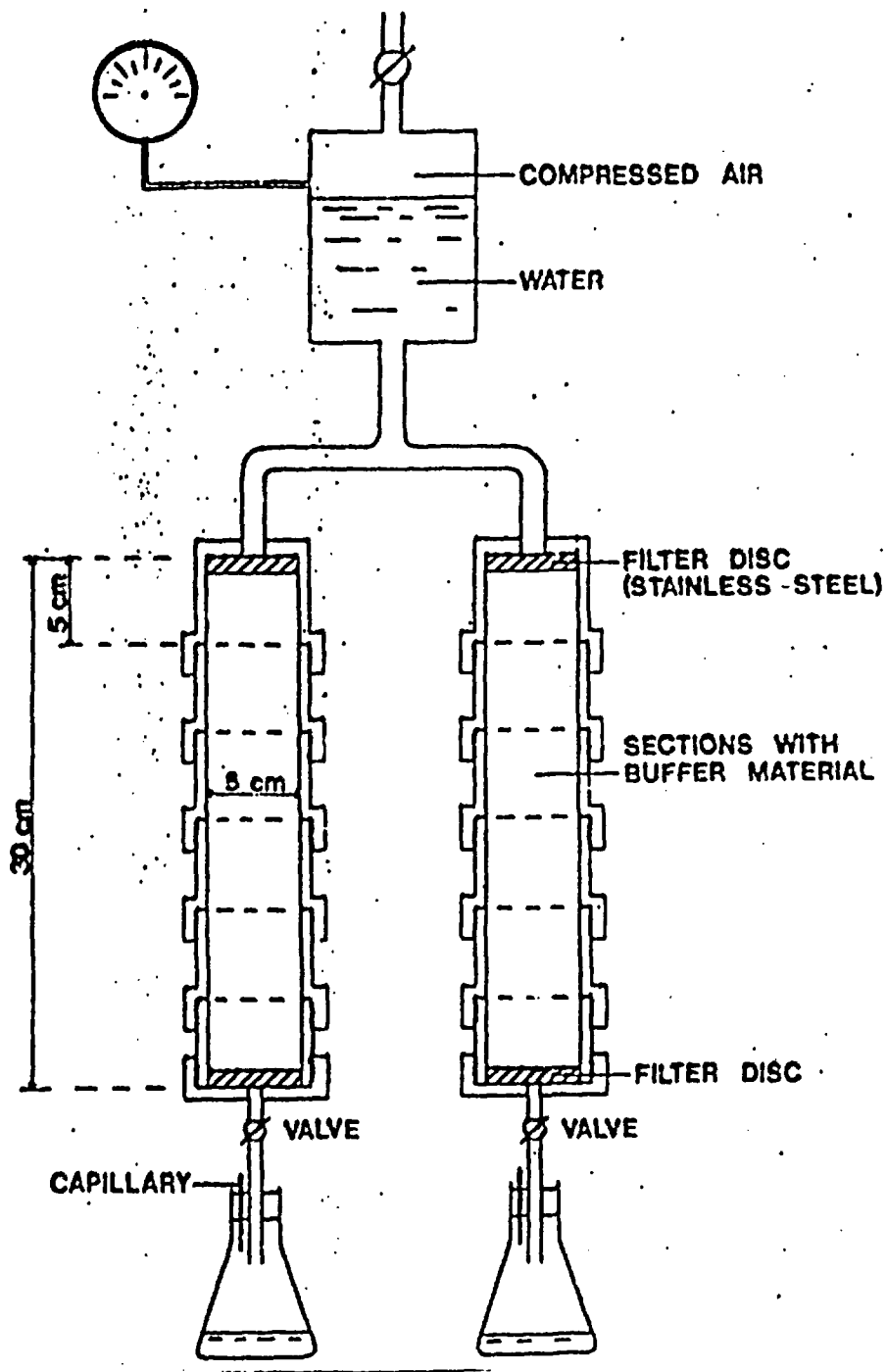


Figure 4.1. Schematic diagram of the apparatus used in hydraulic measurements (Pusch, R., 1979).

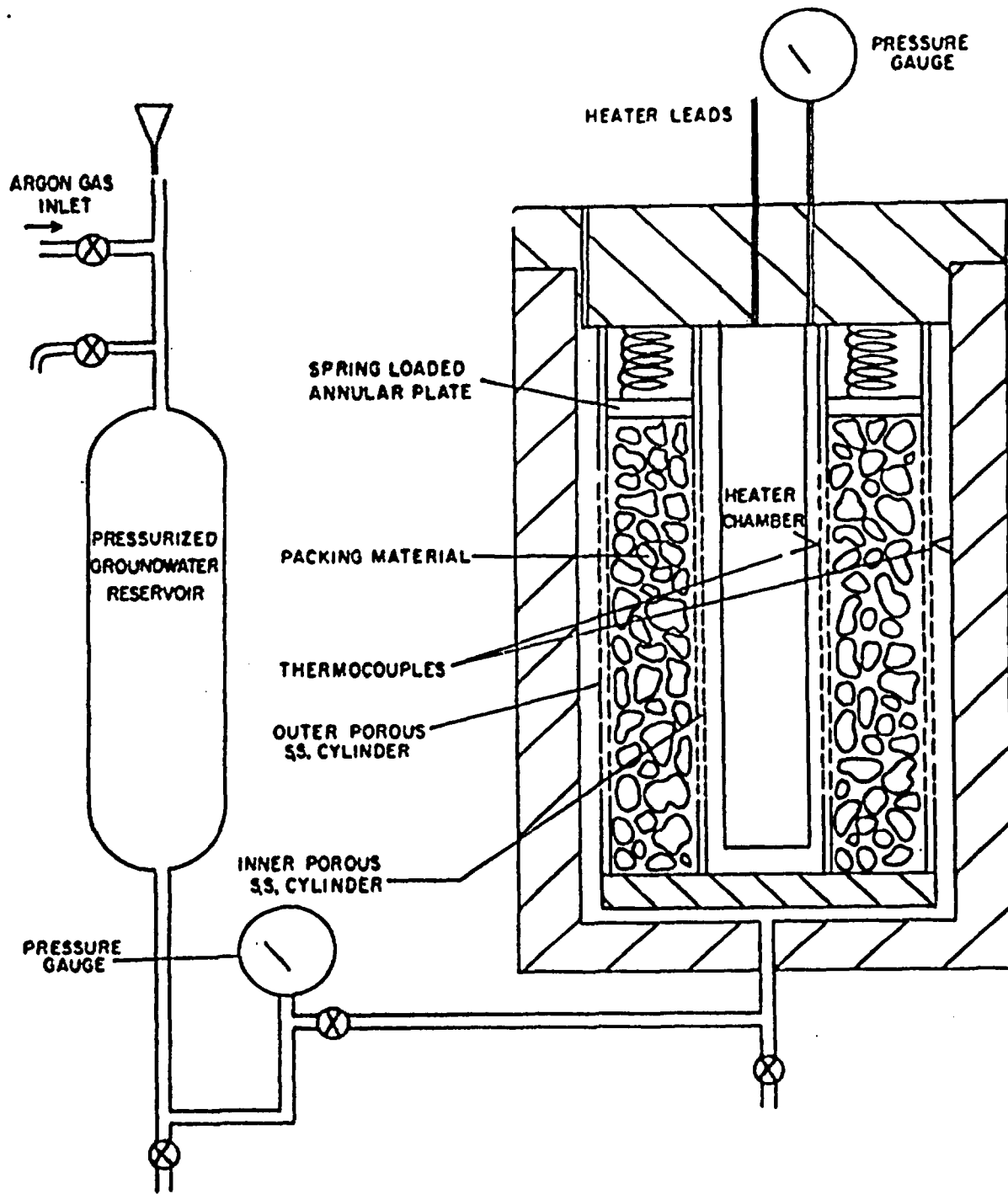


Figure 4.2. Schematic of the test system to study ingress of groundwater under thermal gradients (NUREG/CR-3091, Appendix D, 1984).

thermal gradients. Their test apparatus is shown schematically in Figure 4.2. The authors concluded that their apparatus is not ideally suited for the measurement of hydraulic conductivity, mainly because there was no technique for monitoring the precise rate of water migration. However, with suitable modifications, this kind of test could be conducted on tuff packing.

The test method developed by Byerlee and others (1983) to measure permeability of tuff in a temperature gradient is shown schematically in Figure 4.3. They have determined the time dependence of permeability of solid samples, assuming Darcy's law. It is interesting to note from their results that the permeability of granites and quartzite decreased with time by two orders of magnitude because of blocking of small pores and microfractures by mineral growth. However, there was little change in the permeability of tuff cylinders which had larger pores and vug spaces. The extent of pore blocking in tuff packing will depend on its packing density, which needs to be characterized in order to quantify long term permeability values. Note that in this method (Figure 4.3), the water flows from the hot end of the specimen to its cold end. To represent the situation where groundwater will initially enter the packing from the outside, the water flow direction should be reversed.

#### 4.3.4 Tests for Steam Phase Migration

The groundwater percolating through the pores and fractures in the tuff host rock will form steam on approaching the boiling point isotherm. In the early stages of repository life the host rock temperature will be sufficiently high so that steam will be produced outside the engineered barrier system. Eventually, it will come in contact with the external surface of the tuff/clay packing. To ascertain the time at which the container will be subjected to steam corrosion, a test could be conducted to determine the permeability of steam through the packing.

At present there are no test methods developed to determine the permeability of steam through tuff packing. However, in principle, a vertical column-type system can be used to determine the flow of steam through a prototypic packing. For example, the central part of the column would contain the packing material which may be maintained under a thermal gradient to simulate waste package conditions. High temperature steam would enter the upper part of the column at a given pressure which may be controlled by adjusting the temperature of a sealed water bath which is used to generate the steam. Steam permeation through the packing can be determined by monitoring the breakthrough time at the base of the column. Another method to monitor this flow rate is to spike the water bath with tritiated water and measure the radioactivity along the column for different test periods.

The most important factors affecting steam permeability are the local pressure difference between the inner and outer surface, and the density (or porosity) of the packing component. Both of these parameters may change with time as irradiation (radiolysis products) and alteration of the packing occur after waste emplacement. Wet-dry cycling can also cause cracking of the clay

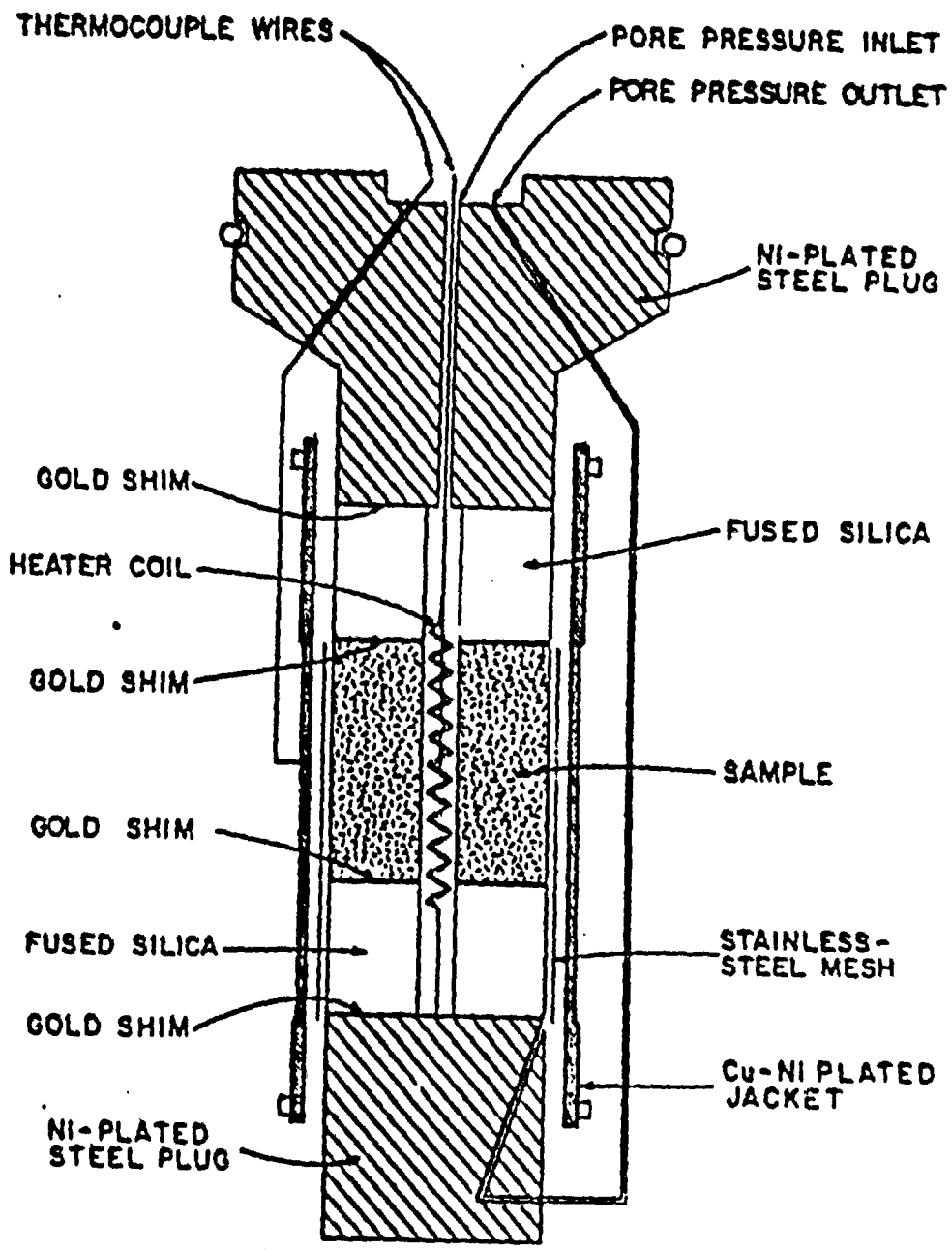


Figure 4.3. Schematic of the test assembly to determine groundwater permeability under thermal gradient (Byerlee, J., 1983).

in the packing and, thus, provide easy paths for steam migration. Therefore, it is suggested that the permeability tests be conducted on prototypic packing that has been subjected to wet-dry cycling at 100°C.

#### 4.3.5 Tests for Radionuclide Migration

Radionuclide transport through cracks or void spaces in packing material can be retarded as a result of molecular diffusion or sorption of the radionuclides. The sorption mechanisms include reversible processes such as ion-exchange and surface adsorption as well as irreversible processes such as precipitation and fixation. Standard test methods developed to determine geochemical retardation of radionuclide migration can be used in testing crushed tuff packing material. At present, there are probably no methods which consider radionuclide (i.e. gaseous or volatile nuclides) migration via the steam phase. It is desirable, therefore, to study the potential for steam phase transport since it may be important for the first few hundred years in a tuff repository. A two-step testing procedure may be used to evaluate this effect. In the first step, the possibility of high temperature steam reacting with the waste form and then releasing radionuclides would be determined. If this possibility is shown to exist, the second step would be to determine the effectiveness of packing material in retarding this process. Sorption mechanisms during steam phase transport may be different from those in the liquid phase and, therefore, test methods may have to be modified to include steam phase transport of volatile or gaseous nuclides.

##### 4.3.5.1 Sorption Under Steam Conditions

Although the predominant pathway for the migration of radionuclides from the waste package is via groundwater transport, it is important to also establish the nature and extent of radionuclide movement under vapor or gaseous states. This is because: (1) the container may fail during the period when steam is present around the waste package; (2) gaseous and volatile fission products may be released as a result of spent fuel cladding degradation due to corrosion.

Previous work conducted at PNL on the volatility of simulated high-level radioactive waste glasses showed that the volatility of radionuclides only becomes significant at temperatures above 1000 K (W. J. Gray, 1980). While such temperatures may be encountered during waste processing, they are not likely to occur under repository conditions. Thus, the release of volatile radionuclides from borosilicate glass waste forms is negligible with respect to transport by steam in a tuff repository. In the case of BWR and PWR fuel rods, the amounts and activity of gases produced based on calculated values, are shown in Tables 4.2 and 4.3. Note that these gaseous radioisotopes may not all be readily released if the cladding fails, since they may be trapped within the fuel pellets or the cladding itself. Gases which are quickly released will be those residing in the space between the fuel and the cladding.

Table 4.2. Amount and activity of gases produced in a BWR fuel rod irradiated to an average burnup of 27,500 MWD/MTU as a function of age.<sup>a</sup>

Gas	At Discharge		300 Years After Discharge	
	Weight (g)	Activity (Ci)	Weight (g)	Activity (Ci)
H <sub>2</sub>	2.153E-04 <sup>b</sup>	2.076E00( <sup>3</sup> H)	9.792E-12 <sup>b</sup>	9.440E-08( <sup>3</sup> H)
He	7.006E-04 <sup>c</sup>	---	3.133E-02 <sup>c</sup>	---
Cl <sub>2</sub>	9.731E-04 <sup>d</sup>	3.124E-05( <sup>36</sup> Cl)	9.722E-04 <sup>d</sup>	3.121E-05( <sup>36</sup> Cl)
Br <sub>2</sub>	5.257E-02 <sup>b</sup>	N.R. <sup>f</sup>	5.259E-02 <sup>b</sup>	N.R. <sup>f</sup>
Kr	8.892E-01 <sup>b</sup>	2.244E01( <sup>85</sup> Kr)	8.317E-01 <sup>b</sup>	8.598E-08( <sup>85</sup> Kr)
I <sub>2</sub>	5.805E-01 <sup>b</sup>	7.527E-05( <sup>129</sup> I)	5.705E-01 <sup>b</sup>	7.619E-05( <sup>129</sup> I)
Xe	1.294E01 <sup>b</sup>	N.R. <sup>f</sup>	1.272E01 <sup>b</sup>	N.R. <sup>f</sup>
Cs	6.784E00 <sup>b,e</sup>	3.230E02( <sup>134</sup> Cs)	3.676E00 <sup>b,e</sup>	7.619E-05( <sup>135</sup> Cs)
		1.035E-03( <sup>135</sup> Cs)		2.502E-01( <sup>137</sup> Cs)
		2.503E02( <sup>137</sup> Cs)		
Rn	3.944E-15 <sup>b</sup>	4.754E-11( <sup>222</sup> Rn)	3.952E-12 <sup>b</sup>	6.084E-07( <sup>222</sup> Rn)
TOTAL		5.978E02 <sup>g,h</sup>		2.504E-01 <sup>i</sup>

<sup>a</sup>This table is adapted from ORIGEN-2 calculated values presented in ORNL/TM-6008 (1977) for a 8 x 8 assembly assumed to contain 63 rods. All volatile materials have not been included. For example, ≈8.0E-05 moles of C-14 are produced as fission and activation products. Some of this may be present in the form of gaseous compounds.

<sup>b</sup>Weight shown includes weight of all isotopes present.

<sup>c</sup>This is the amount of He produced. It does not include He added during manufacture.

<sup>d</sup><sup>36</sup>Cl is formed as an activation product. Weight shown is only for <sup>36</sup>Cl.

<sup>e</sup>Cs is a volatile fission product. The vapor pressure P (in torr) of Cs in the range 200-350°C can be calculated using the formula:

$$\log_{10} P = \frac{3833}{T(\text{in K})} + 6.949.$$

<sup>f</sup>N.R. indicates not reported.

<sup>g</sup>This activity represents ≈0.15% of the inventory at discharge.

<sup>h</sup>Total activity at 10 years from discharge due to these elements is 2.230E02 Ci. This represents ≈23.2% of the inventory 10 years after discharge.

<sup>i</sup>This activity represents ≈2.30% of the inventory 300 years after discharge.



Table 4.3. Amount and activity of gases produced in a PWR fuel rod irradiated to an average burnup of 33,000 MWd/MTU as a function of age.<sup>a</sup>

Gas	At Discharge		300 Years After Discharge	
	Weight (g)	Activity (Ci)	Weight (g)	Activity (Ci)
H <sub>2</sub>	1.918E-04 <sup>b</sup>	1.849E00( <sup>3</sup> H)	8.722-E12 <sup>b</sup>	8.408E-08( <sup>3</sup> H)
He	5.103E-04 <sup>c</sup>	---	2.781E-02 <sup>c</sup>	---
Cl <sub>2</sub>	7.828E-04 <sup>d</sup>	2.513E-05( <sup>36</sup> Cl)	7.828E-04 <sup>d</sup>	2.511E-05( <sup>36</sup> Cl)
Br <sub>2</sub>	4.897E-02 <sup>b</sup>	N.R. <sup>f</sup>	4.898E-02 <sup>b</sup>	N.R. <sup>f</sup>
Kr	8.348E-01 <sup>b</sup>	2.141E01( <sup>85</sup> Kr)	7.799E-01 <sup>b</sup>	8.201E-08( <sup>85</sup> Kr)
I <sub>2</sub>	5.422E-01 <sup>b</sup>	6.971E-05( <sup>129</sup> I)	5.309E-01 <sup>b</sup>	7.078E-05( <sup>129</sup> I)
Xe	1.199E01 <sup>b</sup>	N.R. <sup>f</sup>	1.199E01 <sup>b</sup>	N.R. <sup>f</sup>
Cs	6.221E00 <sup>b,e</sup>	3.494E02( <sup>134</sup> Cs)	3.277E00 <sup>b,e</sup>	---
		8.387E-04( <sup>135</sup> Cs)		8.402E-04( <sup>135</sup> Cs)
		2.346E02( <sup>137</sup> Cs)		2.345E-01( <sup>137</sup> Cs)
Rn	2.632E-15 <sup>b</sup>	2.879E-11( <sup>222</sup> Rn)	3.636E-12 <sup>b</sup>	5.598E-07( <sup>222</sup> Rn)
TOTAL		6.073E02 <sup>g,h</sup>		2.354E-01 <sup>i</sup>

<sup>a</sup>This table is adapted from ORIGEN-2 calculated values presented in ORNL/TM-6008 (1977) for a 15 x 15 assembly assumed to contain 204 rods. All volatile materials have not been included. For example,  $\approx 6.0E-05$  moles of C-14 are produced as fission and activation products. Some of this may be present in the form of gaseous compounds.

<sup>b</sup>Weight shown includes weight of all isotopes present.

<sup>c</sup>This is the amount of He produced. It does not include He added during manufacture.

<sup>d</sup><sup>36</sup>Cl is formed as an activation product. Weight shown is only for <sup>36</sup>Cl.

<sup>e</sup>Cs is a volatile fission product. The vapor pressure P (in torr) of Cs in the range 200-350°C can be calculated using the formula:

$$\log_{10} P = \frac{3833}{T(\text{in K})} + 6.949.$$

<sup>f</sup>N.R. indicates not reported.

<sup>g</sup>This activity represents  $\approx 0.13\%$  of the inventory at discharge.

<sup>h</sup>Total activity at 10 years from discharge due to these elements is 2.107E02 Ci. This represents  $\approx 23.6\%$  of the inventory 10 years after discharge.

<sup>i</sup>This activity represents  $\approx 2.44\%$  of the inventory 300 years after discharge.

In order to determine the effectiveness of the packing material under high temperature (steam) conditions with respect to inhibiting vapor/gaseous transport of these radionuclides, test designs should take into account a realistic radionuclide source term (inventory). The sorption tests could involve a column of compacted tuff/clay mixture situated in a thermal gradient to simulate packing conditions. Boiling of a predetermined amount of groundwater would establish steam condition within a bed of compacted tuff/clay packing material. Gases expected to be released from spent fuel would be introduced into the bottom of the cell. Experimental conditions could be used to provide longterm exposure of the packing material to high-temperature-steam-gas environments. Following a period of long term exposure, the packing material would be removed, sectioned, and a determination made on sorption behavior with respect to the source term. The similar procedures could be followed in order to determine the amount and rate of gas/vapor flow through a column of packing material, and breakthrough times under various packing densities. Gamma irradiation should be incorporated in the test designs in order to determine effects on packing material integrity as a result of the radiolysis of a steam-O<sub>2</sub>-N<sub>2</sub> system. The radiolysis could result in the formation of nitric oxides and nitric acid. The resultant low pH environment is known to be deleterious to clay/packing material integrity and could modify sorptive capacity.

#### 4.3.5.2 Sorption Under Groundwater Conditions

Sorption is a measure of the removal of radionuclides by solids from solution and is usually expressed in terms of the distribution ratio  $R_d$ . This is an experimentally determined quantity, given by:

$$R_d = \frac{\text{Activity of radionuclides in solid phase per unit mass of solid phase}}{\text{Activity of radionuclides in solution per unit volume of solution}}$$

Under equilibrium conditions  $R_d$  is equal to the distribution coefficient,  $K_d$ , which is determined by thermodynamic parameters of the system. Equilibrium can be assumed if the same value of  $R_d$  is obtained during sorption and desorption tests. In general, however, desorption experiments gives higher values than sorption experiments although such behavior is highly radionuclide specific (LA-9328-MS, 1982). Temperature, groundwater chemistry, pH, Eh, radionuclide concentrations (as in isotherm studies) and chemistry, liquid to solid ratio, particle size, test time and flow rates are the major contributing factors in the determination of  $R_d$ . The test methods used in various laboratories for the measurement of sorption may be classified as static or dynamic depending on whether the test solution is stationary or flowing through the packing material.

#### a. Static Method

Due to its simplicity the static method, in which one measures activity of a radioactive element in solid and liquid phases after the test, has been widely used to study the effects of experimental variables listed above. However, no standardized test procedures have been adopted. Details of a typical batch method and the data obtained on crushed tuff may be found in LA-9328-MS (1982). To simulate high temperature conditions in a repository the use of a Dickson-type, gold-cell rocking autoclave has been recommended and gives useful results (Knauss, K. G., 1984). However, the static method does not include the effects of water flow which is anticipated in a repository. Care also needs to be exercised in the interpretation of these test results since agitation may result in new surface formation due to abrasion or grinding, especially in the case of hard materials such as tuff. This phenomenon causes error in the results and is probably the reason for increased sorption with contact time (LA-8747-MS, 1981).

#### b. Dynamic Method

In a dynamic method the effectiveness of a crushed rock in retarding the migration of radionuclides is generally determined by using flow-through columns. Usually, the test solution is passed at a constant rate through a fixed column of sorbing material. A retardation factor,  $R_f$ , is then defined as the velocity of radionuclides in solution relative to that of groundwater flowing through the column. Under equilibrium conditions:

$$R_f = K_d (\rho/c) + 1$$

where  $\rho$  is the bulk density and  $c$  is the porosity of the sorbing material. The effective column porosity is determined in terms of free column volume by measuring the flow of water containing a non-interacting radioisotope such as  $^{131}\text{I}^-$ . Next, the test solution is spiked with isotopes of elements for which a retardation factor is desired (LA-8747-MS, 1981). In these elution studies the effective retardation factor has been calculated from the concentration profile of radioisotopes adsorbed in the test column, which is obtained by sectioning the column and measuring the radioactivity in each section (LA-9329-MS, 1982).

In a modified dynamic test (circulating system), the stationary rock-powder feature of the flow-through column test and the non-renewal of test solution in a batch test are combined as shown schematically in Figure 4.4 (LA-9328-MS, 1982). Unlike a batch test, this setup produces no new particles by grinding, and equilibrium conditions may be expected with greater certainty. A comparison of sorption results using batch, flow-through column, and circulating systems shows reasonably good agreement for some nuclides but poor agreement for others. At present, there is no good explanation for such differences in the behavior, although there is some dependency on the composition of tuff (LA-9328-MS, 1982; SAND80-1464, 1980). Until the mechanisms of sorption are established, the use of the most conservative sorption parameters is recommended.

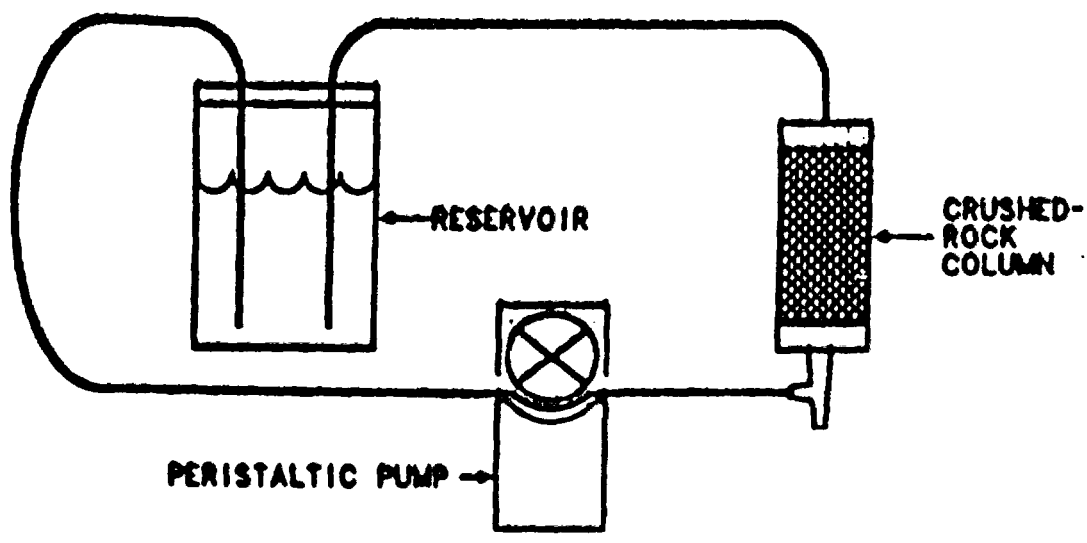


Figure 4.4. Circulating system for crushed-rock system studies (LA-9328-MS, 1984).

The term "sorption" has been used above to describe the general phenomenon of removal of radionuclides from solution by a solid without specifying the underlying mechanism. However, for proper modeling and long term prediction of retardation of radionuclides by packing it is very important to determine the dominant mechanism and whether it changes with time. At present, there are very few tests aimed at determining the principal modes of radionuclide retardation. In a coarse-grain compacted crushed tuff, radionuclides may initially be transported by convection and be adsorbed on the tuff. Further retardation of radionuclides may be governed by their diffusion into the packing material lattice. Walter (LA-9471-MS, 1982) has made use of a simple experimental method to determine the effective diffusivity of various ionic species through disk shaped diaphragms made of tuff. The apparatus described is made of plexiglass, and would, therefore, need modification to obtain data at high temperatures.

Since mechanistic studies may be time consuming, it is suggested that based on the behavior of nuclides in batch-type sorption experiments, they may be classified into groups. Comprehensive testing may then be performed on selected elements in each group.

#### 4.4 Conclusions

- a. At present, a packing material in a tuff repository has been proposed only in one design for a spent fuel package. Preliminary analysis suggests that the packing material could encounter the following repository conditions:

Temperature	- up to $\approx 250^{\circ}\text{C}$ ; steam environment for the first several hundred years.
Gamma radiation	- $10^9$ - $10^{10}$ rads. Total dose.
Pressure	- nearly atmospheric with lithostatic pressure as high as 8.6 MPa.
Water flow	- rainwater percolating at 8 mm/yr at the repository horizon.
Groundwater chemistry	- Oxidic and possibly neutral. However, the radiolysis of a $\text{H}_2\text{O}-\text{O}_2-\text{N}_2$ system could result in the formation of nitric oxides and nitric acid. The resultant low pH environment will be deleterious to packing material integrity. Major dissolved ionic species are $\text{Na}^+$ , $\text{Si}^{+4}$ , $\text{Ca}^{+2}$ , $\text{SO}_4^{-2}$ and $\text{HCO}_3^-$ . High temperature/steam environment may result in highly concentrated groundwater compositions.

- b. Although crushed tuff with small additions of clay as binder is being considered as the packing material for a spent fuel waste package, no definite composition has been formally proposed for this purpose. Accordingly, mineralogic composition for the purposes of testing has not been determined. Other important unknowns are compaction density/pressure, water content, hydraulic conductivity, thermal and radiation stability, sorption of radionuclides and the swelling pressure.
- c. Deterioration in sorption properties due to alteration, and increases in hydraulic conductivity due to cracking of the clay component of the packing appear to be the major modes of packing material degradation. Tests to address the effects of alteration products and crack formation are recommended.
- d. To quantify the contribution the packing will make to the radionuclide controlled release criterion, testing is needed on the migration of radionuclides under steam (e.g. volatile or gaseous nuclides) and groundwater conditions.
- e. In general, the test methods which have been developed or are in the process of development for testing packing for a basalt repository (reviewed in NUREG/CR-3091, Vol. 2, 1983) can also be used for tuff packing exposed to liquid water environments. In fact, the testing

equipment will be much simpler for tuff because high-pressure conditions are not expected in the proposed unsaturated zone. Note that some repository variables such as or wet-dry cycling may be more important in tuff testing than for the case of a basalt repository. In the latter case, a waste package would probably remain wet after the repository has become saturated.

- f. Release of volatile or gaseous radionuclides via the steam phase has not been considered previously. A test is needed to assess the potential for this mode of transport. Consideration should be given to the release of gaseous and volatile fission products due to spent fuel cladding degradation.

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