

From: V. Nguyen, G. Abi-Ghanem, M. Villars/EWA Inc.  
Subject: Review of Packing Material Performance  
Date: July 15, 1985

## I. INTRODUCTION

A multiple barrier approach is proposed to limit radionuclide release from the Nuclear Waste Repository in Basalt to the accessible environment. These barriers will consist of: 1) multiple engineered barriers (e.g., waste form, waste canister, packing material, etc.), and 2) the natural barriers (e.g., geologic strata). This report focuses on the packing material, its properties and its performance relative to Nuclear Regulatory Commission and Environmental Protection Agency regulations.

NRC has required that 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 (10 CFR Part 60). EPA has issued similar requirements in EPA 40 CFR Part 191. During the containment period (first 300-1,000 years), substantially complete containment of all radionuclides is required by the NRC. To meet this requirement, the waste canister must not be breached during this period. Although no

specific performance requirements are delegated to the packing material, it is included in the engineered barrier system to assure that radionuclide release rates are maintained below prescribed limits. To this effect, it serves four major functions. These functions are to:

- 1) limit canister corrosion,
- 2) limit the release rate of radionuclides by creating conditions of diffusion-controlled flow,
- 3) act as a sorbing medium and cause precipitation of radionuclides into new mineral phases to retard radionuclide transport to the host rock and accessible environment, and
- 4) act as a reducing medium to ensure that low solubilities of some radionuclides (e.g., actinides) are attained.

The physical and chemical properties of the packing material must be able to perform these functions. In addition, the packing material must possess sufficient bearing strength to physically support and redistribute the loads and stresses exerted onto it by the surrounding medium, and acceptable thermal conductivity characteristics to assure that it will not irreversibly dehydrate or undergo significant chemical alteration. The material must be able to perform its objectives under a range of pH, redox, temperature, and pressure conditions expected in the repository. Figure 1 gives a summary of anticipated values for these parameters. There is some question about the reliability of these values; as testing and modeling efforts continue, they will require revision. As can be seen, the maximum temperature expected is approximately 300°C, although this will depend

on waste loading and canister spacing (Lane et al., 1983). Figure 2 illustrates the physical arrangement of the packing material in relation to other components of the repository.

## II. PHYSICAL CHARACTERISTICS OF THE PACKING MATERIAL

The DOE has chosen a candidate packing material comprised of crushed native basalt (75%) and Na-bentonite clay (25%) (DOE, 1982). They believe that optimum performance is achieved with a 75%/25% combination. Bentonite is included for its low permeability, high swelling capacity, and high sorption capacity for some radionuclides. Basalt is included because of its high thermal conductivity and bearing strength. In addition, the DOE expects it to be able to maintain a reducing environment and precipitate certain elements (released from the waste form) as secondary minerals. The availability of basalt also makes it an attractive candidate material. The packing material's properties with respect to its four major functions are discussed in more detail below.

### II.1. Ability to Limit Canister Corrosion

The extent to which a basalt/bentonite packing material will limit canister corrosion depends on the packing material's ability to inhibit groundwater from reaching the canister and on its ability to provide the canister with a chemically non-corrosive environment (i.e., high pH and low Eh). As discussed in the next section, there is a large variance in predictions of the time required for groundwater to penetrate the packing material. Some canister

materials, such as carbon steel, possess only limited corrosion resistance to a humid bentonite environment; whereas, titanium and its alloys have excellent resistance (NRC, 1984). Basaltic groundwater does not appear to be highly corrosive to titanium, iron, nickel, or copper (Anderson, 1981).

## II.2. Barrier for Groundwater Flow

The design life for packing material in contact with groundwater can be divided into two periods, pre-saturation and post-saturation. Saturation refers to the state in which the maximum amount of water is absorbed for a given compaction density (NRC, 1983c). Unsaturated bentonite will accommodate water in the interlayer spaces of its microstructure and expand, filling any cracks in the packing material. If the clay is confined, the expansion can produce a swelling pressure, which will inhibit further groundwater migration.

For bentonite, the time required to become fully saturated is a complex function of clay properties, including bulk density and clay composition. The time for water to penetrate and saturate a given thickness of bentonite or packing material has not been well quantified. PNL estimates the time required for water to penetrate 20 cm of bentonite will be on the order of several thousand years (in NRC, 1983c). DOE has estimated a minimum time range of 15 to 50 years to completely saturate 30 cm of backfill, using Darcy's Law and assuming a hydrostatic pressure gradient across the barrier (Smith et al., 1980). Westinghouse predicts the time for full saturation of

packing material to range from a few years to  $10^5$  years (in NRC, 1983a). Not enough information is given about the conditions for which these calculations were made in order to explain the large variance in results. Obviously, experimental operating conditions need to be more clearly defined, and more accurate data on the hydraulic properties of packing material is needed.

One method of obtaining this information is by conducting laboratory experiments to measure saturation time under the expected repository conditions. Because the saturation time for the complete backfill thickness may be very long, laboratory experiments will necessarily be conducted on a short time frame (with respect to the life of the repository) and possibly at a reduced spatial scale. Under these circumstances, the use of meaningful conceptual models to predict longterm saturation will be required. To that end, all fundamental processes occurring at the microscopic scale, such as particle wetting by diffusion, and swelling by deformation, must be identified for the length of the testing period and used as a basis to investigate their manifestation at the macroscopic scale and for longer time scales. This will help to better characterize the packing material over a large time period, to verify whether it will be saturated before the onset of canister failure, and to accurately assess its long-term performance.

After the bentonite packing material is fully saturated, water movement will be mainly controlled by the hydraulic conductivity of the packing material and by the regional hydraulic gradient. The hydraulic conductivity is mainly dependent upon the composition,

porosity, and degree of void interconnection (NRC, 1983c).

Experiments by Wood et al. (1983) to measure the hydraulic conductivity of packing material showed that the conductivity is greatly affected by the material density and the clay composition.

Wood found that:

- 1) hydraulic conductivity decreases as a function of density,
- 2) hydraulic conductivity appears to be independent of temperature up to 90°C, and
- 3) at densities above 1.7 g/cm<sup>3</sup>, hydraulic conductivity values are below  $4 \times 10^{-9}$  cm/sec.

However, Wood had a great deal of scatter in data, and up to one order of magnitude uncertainty in results. In addition, the experiments were conducted only at temperatures less than 100°C. Westinghouse (1981) expects a minimum hydraulic conductivity of  $10^{-14}$  cm/sec for Na-bentonite, based on a density of 2.1 g/cm<sup>3</sup> (after Pusch, 1979). Again, this value was not determined as a function of temperature. Westsik et al. (1982) measured hydraulic conductivity of  $5 \times 10^{-13}$  cm/sec for pure Na-bentonite.

Determination of hydraulic conductivity is especially important because low conductivity is expected to inhibit groundwater flow such that radionuclide transport will be diffusion controlled. Bida and Eastwood (1983) state that for reasonable bulk diffusion coefficients (approximately  $10^{-6}$  cm/sec), diffusion is the dominant process of mass transfer through the packing material if the hydraulic conductivity is less than  $10^{-11}$  cm/sec, under the very low hydraulic gradient conditions ( $5 \times 10^{-4}$  to  $5 \times 10^{-5}$ ) expected at the BWIP site.

Due to the variability of measured values, more effort should be made by DOE to determine the hydraulic conductivity of the packing material under the expected repository conditions. They must show that the hydraulic conductivity of the packing material is lower than  $10^{-11}$  cm/sec in order to justify diffusional transport.

As mentioned before, a laboratory study to measure the hydraulic properties of the packing material may be difficult due to the very slow flow rates expected. However, the importance of verifying that diffusional flow will occur cannot be overstated. The packing material is the last barrier in the waste package, before the radionuclides reach the host rock. It is imperative to understand the radionuclide movement through this region in order to assess the performance of the waste package as a whole, in terms of meeting NRC release requirements.

A further consideration in evaluating packing material performance is that the groundwater flow and chemical transport mechanisms will likely be affected by the heat introduced from waste emplacement. The packing material will experience a temperature cycle from an initial repository ambient temperature of 60°C, to as high as 250-300°C, and then undergo a slow return to ambient conditions as fission products decay and the surrounding host rock cools. During the first few hundred years, the water content of the bentonite may be close to zero due to loss of interlayer water; the water which is present may exist as a vapor. An increase in temperature will influence water flow by decreasing water viscosity, increasing diffusion rates, and possibly changing swelling pressures

(Westinghouse, 1981). An understanding of long term groundwater flow requires that water flow rates be known over the temperature ranges of interest. The increased temperature may also affect chemical transport rates, because radionuclide solubility and sorption are temperature dependent (Early et al., 1982; Maest et al., 1984). This will be especially important if radionuclide releases occur before the repository has returned to ambient temperatures.

In summary, there are several significant areas in which information is insufficient or nonexistent concerning fluid flow and chemical transport through the packing material. Among them are: 1) the length of time it will take for the packing material to become completely saturated, 2) the hydraulic conductivity of the packing material once it has become saturated, 3) the method of chemical transport through the packing material once radionuclides are released from the breached canister, 4) the effects of thermal gradients on the hydraulic properties of the packing material, and 5) the effect of the full range of groundwater chemistries, Eh, and pH on the hydraulic properties of the packing material.

### II.3. Sorption Characteristics

There are two major geochemical processes which will retard radionuclide transport in the packing material. They are precipitation of radionuclides into new mineral phases, and sorption. Precipitation is considered secondary to sorption in retarding most radionuclide migration (DOE, 1982). To fulfill its function as a sorbing medium, the packing material must be able to sorb a number of



key radionuclides. Because of their toxicity and potential mobility, and the quantity available in the stored waste, the following radionuclides are of greatest importance (Smith et al., 1980):

Actinides

Radium-226  
Neptunium-237  
Uranium-233, 234  
Plutonium-239, 240, 242  
Americium-241, 243

Fission Products

Iodine-129  
Selenium-79  
Technetium-99  
Cesium-135  
Strontium-90  
Tin-126  
Palladium-107  
Zirconium-93

BWIP has conducted a series of experiments to determine the sorption performance of the host basalt and interbed materials (in which there are clays similar to bentonite) (Salter et al., 1981a, 1981b; Barney, 1981, 1982). Information was gathered under both oxic and reducing conditions, although the DOE expects conditions to be strongly reducing in the repository. A review of these experiments shows that:

- anionic radionuclides such as I, Se, and Tc are sorbed weakly by basalt and secondary minerals (a third packing material component may be necessary to sorb these radionuclides),
- altered ("weathered") basalt and interbed materials sorbed cationic radionuclides much more strongly than fresh basalt,
- in their oxidized valence states, Am is strongly sorbed and U, Np, and Pu are weakly sorbed by basalt,
- in their reduced valence states, sorption of Tc, Np, U, and Pu on basalt is much greater than under oxidizing conditions.

BWIP concludes from these experiments that except for some anionic radionuclides, sorption by basalt and clay minerals in the near and far field will provide a significant barrier to radionuclide migration. However, Kelmers (1984) shows this data to be inadequate for accurate modeling of radioactivity releases from the repository due to data collection under conditions dissimilar to expected repository conditions and other experimental or informational deficiencies.

#### II.4. Reducing Characteristics

Since the DOE expects solubilities of most radionuclides to be much lower under reducing conditions, and since canister corrosion of some metals may be inhibited under reducing conditions, it is important that reducing conditions be maintained in the repository. The near field redox conditions will be controlled by oxygen reaction with the basaltic groundwater (when oxygen is available) and, perhaps by radiolysis. Radiolysis may induce dissociation of water and create oxidizing conditions (DOE, 1982). The DOE expects that the basalt environment will keep the Eh low enough such that there may be enough hydrogen present to inhibit the radiolysis of water (DOE, 1982). DOE expects the Eh to range from an initial value of +.54 to a value of -.43 during geologic control (DOE, 1982). The NRC (1983b) has criticized the methods used by DOE in determining Eh values and questions the validity of their results. Prediction of redox conditions in the repository (i.e., the time required for Eh to return to its unperturbed value, Eh range, and radiolysis effects) are very

preliminary as the kinetics of the redox reactions have not yet been sufficiently defined, and as definitive methods for determining repository redox conditions have not been devised (NRC, 1983b, 1985b).

### III. FAILURE MODES

There are at least two major possible failure modes for the packing material, chemically-induced failure and thermomechanically-induced failure. Chemically-induced failure includes loss of hydrothermal stability, aging, decrease of sorptive capacity by chemical reaction or poisoning, and selective dissolution or leaching of the packing matrix. Radiolysis will affect the Eh and Ph of the groundwater and thus will also affect chemically-induced failure. Radiolysis is discussed in more detail later in this report. Thermomechanically-induced failure includes crack or fissure formation. Chemically-induced failure may increase the likelihood of fissure formation due to volume shrinkage.

#### III.1. Chemically-Induced Failure

The most significant mode of chemically-induced failure, by far, is loss of hydrothermal stability or early diagenesis (NRC, 1983a). Both basalt and bentonite have been shown to be unstable under conditions similar to those expected in the near field (Weaver, 1979; Eberl and Hower, 1976). Basalt-seawater hydrothermal experiments (Seyfried and Bischoff, 1981; Seyfried et al., 1984; Seyfried and Mottl, 1982) and the BWIP data available (Lane et al., 1983; Wood, 1983; Allen et al., 1983), suggest that basalt is unstable and can

degrade to a wide variety of alteration minerals. Studies on the alteration of bentonite and clay minerals (Eberl, 1978; Eberl and Hower, 1976; Weaver, 1979), show that bentonite may alter first to a mixed layer illite-smectite mineral (I/S) and then to illite by removal of interlayer water and replacement of  $\text{Ca}^{+2}$  or  $\text{Na}^{+}$  by  $\text{K}^{+}$ . Other alteration products can include zeolites, albite, quartz, smectite, and K-feldspar. The extent to which these transformations will occur depends on temperature, ionic concentration of the groundwater, available reaction time, and to a lesser extent, on pressure. The formation of illite is of primary concern because of its inferior swelling and sorption properties relative to bentonite.

Although the upper limit of temperature stability for bentonite is usually given as  $300^{\circ}\text{C} \pm 50^{\circ}\text{C}$ , transformation of montmorillonite (the main component of bentonite) to illite is known to have occurred at temperatures well below  $100^{\circ}\text{C}$  under conditions where high  $\text{K}^{+}/\text{Na}^{+}$  ratios and low pressures exist. Although the basalt groundwater of the Hanford site has very low  $\text{K}^{+}/\text{Na}^{+}$  ratios, the influence of thermal reactions and the presence of the engineered barrier on the groundwater chemistry have not been fully evaluated. It is possible that potassium for the conversion of montmorillonite to illite may be derived from breakdown of K-feldspar in the bentonite and from dissolution of basaltic glass (Wood et al., 1983; NRC, 1983d). It has been shown that the mesostasis basalt phase is the most reactive basalt component (Wood, 1983). In addition, it is possible that even with a low  $\text{K}^{+}/\text{Na}^{+}$  ratio, there could be enough potassium for bentonite conversion to illite. This is because illite tends to preferentially

concentrate ions such as  $K^+$  (with lower hydration energies) over  $Na^+$  (with higher hydration energies) (Eberl, 1978a, 1978b). Sodium has a high hydration energy and does not dehydrate as easily; whereas, potassium is the most naturally abundant, easily dehydrated cation. Therefore, a low concentration of potassium in a basalt-bentonite system can possibly have a disproportionately large effect, and simple analysis of  $Na^+/K^+$ ,  $Mg^+/K^+$  ratio does not yield adequate information about the reaction capability of the system.

### III.2. Thermomechanically-Induced Failure

Bentonite, like other clays, has a layered platy structure with water contained between the layers and within the structure of individual plates. Reversible dehydration and associated volume reduction occur when interlayer water is lost. This takes place at temperatures between  $100^{\circ}C$  and  $300^{\circ}C$  at atmospheric pressures (NRC, 1983c). If the temperature rises much above  $300^{\circ}C$ , both interlayer and structural water will be lost. The loss of structural water causes irreversible dehydration, loss of swelling capacity, and volume reduction and, if this occurs in the packing material, it could result in the formation of cracks. Cracking of bentonite caused by wet/dry cycling is the most probable cause of thermomechanically-induced failure (NRC, 1983a). The first cycle, which is of greatest importance, would occur soon after emplacement, i.e., water adsorption immediately after emplacement and then reversible dehydration during the thermal peak. As bentonite adsorbs water, swelling and creep are induced. During the subsequent drying cycle, volume will be reduced and cracks will form since there will be no reverse pressure to recompact the

bentonite (the effect of lithostatic pressure on recompaction has not been discussed by the DOE or NRC). Alternatively, if the repository temperatures increase rapidly enough, cracking due to dehydration could occur during the early thermal period before saturation. As long as this is not irreversible dehydration, it is not expected to compromise the performance of the packing material since groundwater will later swell the clay back to its original volume (NRC, 1983a). Another effect that wet/dry cycling may have is anisotropic swelling and separation of multicomponent packing materials (NRC, 1983b).

The DOE is collecting more information concerning expected repository conditions and the hydraulic properties of clay under those conditions to adequately assess the importance of these failure modes.

#### IV. DOE DEHYDRATION AND HYDROTHERMAL EXPERIMENTS

Dehydration and hydrothermal experiments have been conducted for the DOE by Rockwell to determine if bentonite dehydration can be expected under repository conditions and to determine the extent and character of alteration minerals that may be formed in the packing material and host basalt. Simulated repository groundwater and Wyoming Na-bentonite clay were used in these experiments. The Wyoming bentonite was used as-received and consisted of 85 vol% Na-montmorillonite, 5 vol% quartz, 5 vol% feldspars, 2 vol% cristobalite, 2 vol% illite, lesser amounts of nontronite (a zeolite), gypsum/anhydrite, and calcite. It should be noted that the exact composition of the bentonite may vary from one shipment to another.

The extent and effect of this variation remains to be adequately assessed by the DOE.

#### IV.1. Dehydration Experiments

Dehydration experiments were conducted to determine the temperatures at which structural water would be lost. Palmer et al. (1983) has heated bentonite to temperatures of 250°C to 550°C at atmospheric pressure in a dry environment. Results show that no structural water loss occurred within one year at 370°C, although complete structural water loss occurred after only one day at 440°C. As mentioned, if the clay's structural water is lost, fractures could be produced in the packing medium. Since Palmer does not expect repository temperatures to exceed 300°C, he concludes that the dehydration process should not decrease packing material stability in the repository. However, this conclusion is highly conservative in that no rate data is given. Water may be lost at some measureable rate at lower temperatures also. In addition, there are other ways of losing 'structural water', such as a slow change to some non-expanding clay (illite) or zeolite (mordenite), which were not taken into account.

Little information has been published on cracking due to wet/dry cycling and, according to the NRC, this process would seem to warrant more thorough investigation (NRC, 1983a). An analysis of wet/dry cycling requires that the length of time for water to reach the canister and sorption and desorption of water under repository conditions as a function of time be accurately defined.

#### IV.2. Hydrothermal Experiments

In DOE hydrothermal experiments, bentonite-groundwater and crushed basalt-bentonite-groundwater systems were tested to determine bentonite stability (Wood et al., 1983; Allen et al., 1983; Lane et al. 1983). Pressures were maintained at 30 MPa, temperatures ranged from 100°C to 300°C and exposure times from 25 to greater than 458 days (although how much greater was not specified).

The alteration products formed at 300°C in the bentonite-groundwater system included albite, quartz, smectite, Na<sup>+</sup>/K<sup>+</sup> montmorillonite, illite/smectite, illite, zeolites, and K-feldspar. Except for illite, these alteration products were also reported for the basalt-bentonite-groundwater system. Smectite was formed at temperatures of 200°C or less in both systems. Allen et al. (1983) suggests that the transformation of Na-montmorillonite to smectite may be a precursor to formation of illite/smectite. Quantitative estimates of percent conversion of bentonite to alteration minerals is the subject of current research efforts.

As discussed previously, the formation of illite would detrimentally affect the swelling and sorption characteristics of the packing material. Several factors are cited by Wood, (Wood et al., 1983) that indicate that conversion of smectite to illite may not be a significant reaction. These factors are: 1) a limited source of potassium exists in the basalt and bentonite; 2) the potassium



available will also be distributed among other alteration phases such as K-feldspar and K-bearing zeolites; and 3) at expected repository temperatures when saturated conditions are estimated to occur (i.e., less than 150°C) kinetics of the conversion are estimated to be quite slow relative to repository time scales. Howard and White (1981) quote data from Eberl and Hower (1976) which suggests that a 20% conversion would require 1,000 years at 140°C. However, this is based on the slower of the two rate curves which Eberl and Hower published. The other rate curve suggests conversion at rates orders of magnitude faster. Oversby (1982) calculated that an 80% conversion could occur in three years at temperatures between 285 and 330°C.

The chemical stability of the packing material may be overestimated by the DOE. Information deficiencies exist concerning the reaction rates and processes under the full range of repository conditions, i.e., fluctuating groundwater composition, conditions involving groundwater flow, fluctuating pH, and long time periods. More importantly, the thermodynamic properties of the bentonite packing material cannot be quantified because they are not available for materials of such undefined composition. Work by Tardy and Garrels (1976, 1977) attempts to estimate free energy values for many minerals, but is not reliable for such mixed composition minerals where there are varying cationic components. In choosing minerals which are not thermodynamically characterized, and which seem to be metastable under repository hydrothermal conditions, DOE can have little quantitative understanding of the packing material performance.

The type of alteration product formed during hydrothermal reactions is greatly influenced by the ionic composition of the groundwater. Groundwater migration patterns through the complex basalt and interbed layers, and the effects that high temperatures may have on these patterns, have not been well defined. Thus it can not be shown that hydrothermal reactions have been studied for all relevant groundwater compositions. Also, interactive effects between the packing material and components of the waste package must be considered. After failure of the waste canister, the packing material may react with both the borosilicate glass and the waste elements to form new mineral phases such as analcime, oligoclase, and powellite (NRC, 1983a). The effect that these would have on the performance of the packing material has not been well documented, although it may, in part, be beneficial due to stabilization of certain waste elements such as Cs, Mo, and Sr.

All of the hydrothermal experiments cited have been performed as "static" tests in which there was zero fluid flux. This condition does not simulate all repository conditions. Lane et al. (1983) states that "alteration assemblages in the packing material and host basalt may be a function of flow rate and time-integrated fluid flux at a given temperature-pressure condition". Studies are in progress to compare static results with tests using varying flow rates.

Aluminosilicate clays such as bentonite are adversely affected by both very high and very low pH since pH strongly effects the solubilities of silicon and aluminum. High pH, coupled with high

concentrations of alkali or alkali earth metals (i.e., lithium, sodium, potassium, and cesium) favors the formation of zeolites or feldspars. Unperturbed Grande Ronde groundwater pH values range from 9.1 to 10.6. Conditions are expected to become more acidic during the operating period when the repository is exposed to oxygen. The extent of this pH change has not been well defined. Based on calculations from thermodynamic data at 250°C, Na-montmorillonite stability is best achieved by maintaining the pH between 5 and 8 (NRC, 1983a). The pH range which can be expected during the repository's lifetime is not yet known with any degree of certainty.

Although the rate of conversion from smectite to illite is estimated to be quite slow, this rate may increase if there are changes in the groundwater chemistry. As noted previously, the groundwater composition may vary during repository history from the composition simulated in these experiments. Also, because these rates are slow, the relatively short time spans used in these experiments do not permit thorough reaction analysis.

## V. SUMMARY

To maintain radionuclide release rates below EPA and NRC limits, the packing material is designed to provide the following protections:

- 1) limit canister corrosion;
- 2) limit the release of radionuclides by creating conditions of diffusion-controlled flow;
- 3) act as a sorbing medium and cause precipitation of radionuclides into new mineral phases; and

- 4) act as a reducing medium to ensure low solubilities of radionuclides such as actinides.

The DOE has gathered information on the expected performance of the packing material relative to the above functions, and has investigated probable chemical and mechanical failure modes. In general, the quantity and/or quality of this information has been insufficient to allow accurate assessment of performance. Before performance can be accurately assessed, expected repository conditions (i.e., temperature, pH, redox conditions, and groundwater composition ranges), reactions of the packing material under these specific conditions, and interactions between repository components must all be better defined. The following discussion summarizes packing material performance and failure modes versus their corresponding informational needs.

#### Ability to limit canister corrosion:

The packing material will limit canister corrosion according to its ability to inhibit groundwater flow and its ability to provide a chemically non-corrosive environment. Estimates of the time required for groundwater to penetrate the packing material have a wide variance and must be more narrowly defined. A basalt/bentonite packing material will provide an environment that may either inhibit or induce corrosion, depending on the choice of waste canister material. As yet, the DOE has not chosen the metal to be used in the waste canister.

#### Ability to limit groundwater flow:

Although the DOE believes a basalt/bentonite packing material will limit flow to the extent that transport will be diffusion-controlled, the following factors have not been sufficiently investigated: 1) the effect of increased temperature on the rate of water movement, 2) the effect of thermal gradients across a given packing material thickness, 3) the functional dependence of swelling pressure on hydraulic pressure, 4) the hydraulic properties of bentonite under the full range of groundwater chemistries which may be present, and 5) the effect of Eh and pH on hydraulic properties.

#### Sorption ability:

DOE sorption experiments have indicated that except for anionic radionuclides, sorption by the packing material and host basalt will provide a significant barrier to radionuclide migration. In contradiction, the NRC states that sorption properties of the packing material have not been well defined since most of the sorption experiments were conducted under conditions unlike those expected in the repository or because of other experimental or informational deficiencies.

#### Ability to create reducing conditions:

The DOE's position that a basalt/bentonite packing material will aid in creating reducing conditions is based on preliminary findings

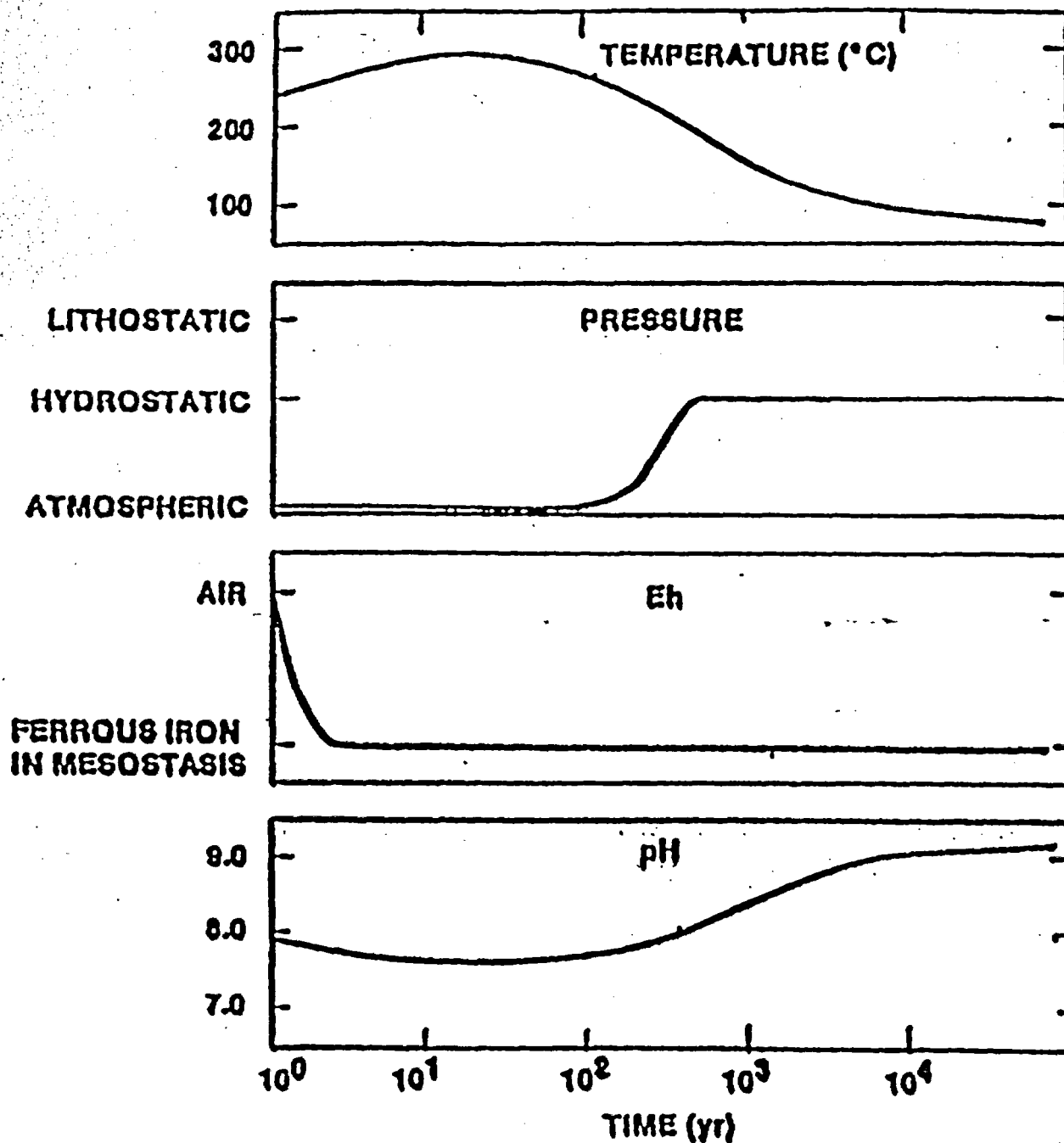
since the kinetics of the redox reactions have not yet been sufficiently defined.

#### **Chemically-induced failure:**

The most important mode of chemically induced failure is loss of hydrothermal stability. Alteration of montmorillonite to illite is of greatest concern due to illite's relatively low swelling and sorption characteristics. DOE experiments show illite may be produced, although some DOE researchers state that the amount produced will be insignificant. This position is based only on preliminary evidence, as hydrothermal testing has not been conducted for all relevant conditions (i.e., varying groundwater compositions, groundwater flow conditions, and long time periods). Also, repository pH and redox condition ranges and their effect on hydrothermal stability have not been well defined.

#### **Thermomechanically-induced failure:**

Wet/dry cycling appears to be the most significant mode of thermomechanically-induced failure. Information on the repositories' thermal and hydrological conditions, and saturation and drying times under those conditions have not been adequately defined.



EWA

ENVIRONMENT & WATER RESOURCES MANAGEMENT

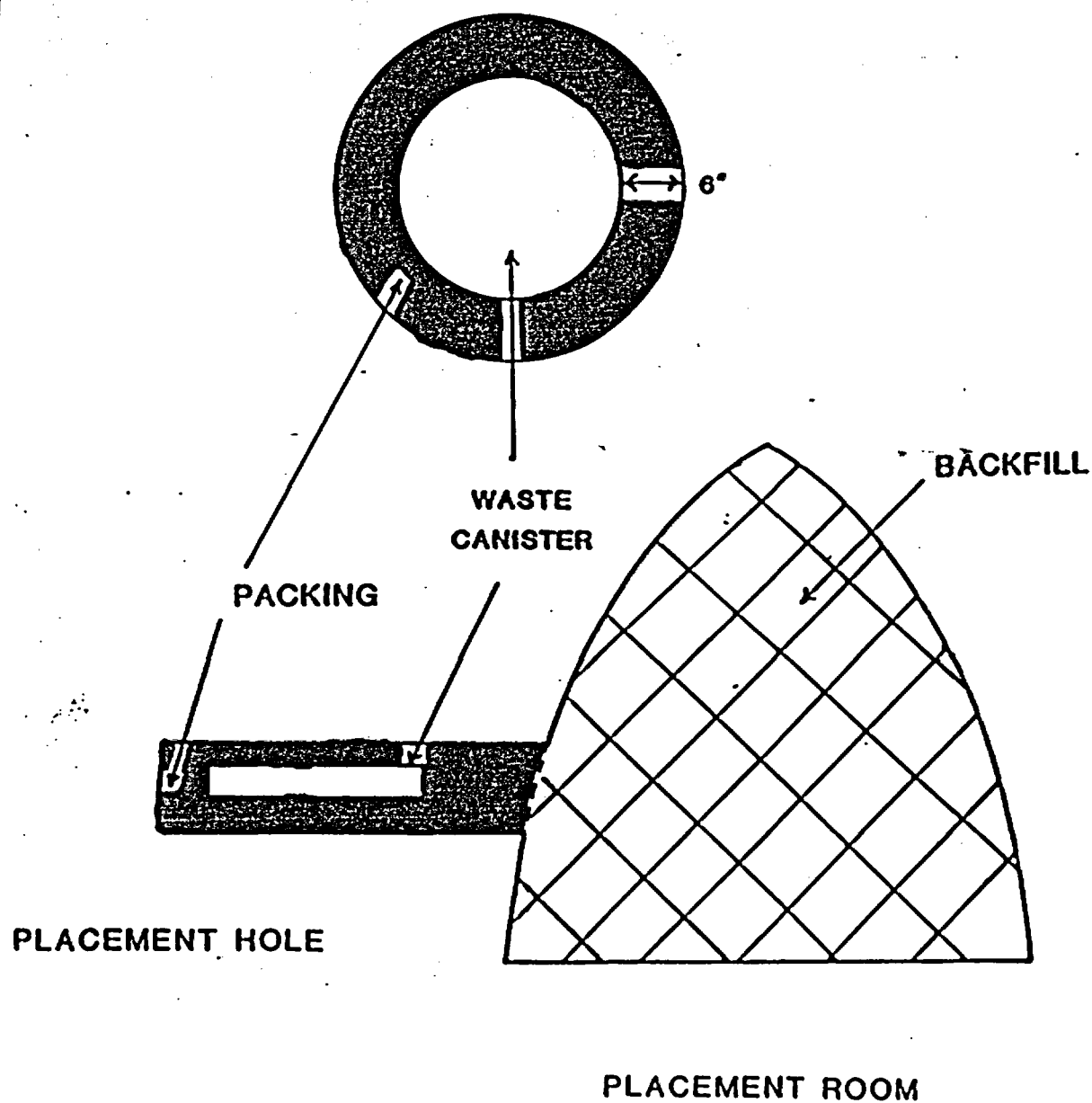
YAKIMA INDIAN NATION

Schematic showing anticipated changes in repository conditions with time (NRC, 1985a)

FIGURE

1

(NOT TO SCALE)





## VI. REFERENCES

- Allen, C.C., D.L. Lane, R.A. Palmer, and R.G. Johnston, 1983. "Experimental Studies of Packing Material Stability," RHO-BWI-SA-313 P, November 1983.
- Anderson, W.J., 1981. "Tests of Canister and Overpack Materials in Simulated Basalt Groundwater," RHO-BWI-ST-15, May 1981.
- Barney, G.S., 1981. "Radionuclide Reactions with Groundwater and Basalts from Columbia River Basalts," RHO-SA-217, June 1981.
- Barney, G.S., 1982. "Radionuclide Sorption of Columbia River Basalt Interbed Materials," RHO-BW-SA-198 P, May 1982.
- Bida, G. and D. Eastwood, 1983. "Packing Material Testing Required to Demonstrate Compliance with 1000-Year Radionuclide Containment," NUREG/CR-2755, February 1983.
- DOE 1982. "Site Characterization Report for the Basalt Waste Isolation Project," DOE/RL 82-3 Vol. 2, 1982.
- Eberl, D., 1978a. "The Reaction of Montmorillonite to Mixed-layer Clay: The Effect of Interlayer Alkali and Alkaline Earth Cations," in *Geochemica et Cosmochemica Acta*, Vol. 42, 1978.
- Eberl, D., and J. Hower, 1976. "Kinetics of Illite Formation," in *Geological Society of America, Bulletin* v.87, p 1326, September 1976.
- Eberl, D., G. Whitney, and H. Khoury, 1978b. "Hydrothermal Reactivity of Smectite," *American Mineralogist*, Volume 63, pp.401-409.
- Kelmers, A.D., 1984. "Review and Assessment of Radionuclide Sorption Information for the Basalt Waste Isolation Site," NUREG/CR-3763 ORNL/TM-9157, September 1984.
- Lane, D.C., M.J. Apted, C.C. Allen, and J. Meyers, 1983. "The Basalt/Water System: Considerations for a Nuclear Waste Repository," RHO-BW-SA-320 P, November 1983.
- Myers, J., et al., 1982. "Comparison of Hydrothermal Stability of Simulated Spent Fuel and Borosilicate Glass in a Basaltic Environment," *Proceedings of the 1982 National Waste Terminal Storage Program Information Meeting*, DOE/NWTS-30, December 1982.
- NRC, 1982. "Nuclear Waste Management Technical Support in the Development of Nuclear Waste Form Criteria for the NRC- Task 1: Waste Package Overview," R.Dyal et al., NUREG/CR-2333, BNL-NUREG-51458, Vol. 4, February 1982.

- NRC, 1983a. "Review of DOE Waste Package Program Biannual Report," P. Soo editor, NUREG/CR-2482 BNL-NUREG-51494 Vol. 3, March 1983.
- NRC, 1983b. "Draft Site Characterization Analysis of the Site Characterization Report for the Basalt Waste Isolation Project," NUREG-0960 Vol. 2, March 1983.
- NRC, 1983c. "Review of Waste Package Verification Tests," P. Soo editor, NUREG/CR-3091 Vol. 2, August 1983.
- NRC, 1983d. "Review of the Waste Package Program," P. Soo editor, NUREG/CR-2482, BNL-NUREG-51494 Vol. 4, September 1983.
- NRC, 1984. "Review of the Waste Package Program," P. Soo editor, NUREG/CR-2482, BNL-NUREG-51494 Vol. 5, August 1984.
- NRC, 1985a. "Review of DOE Waste Package Program," P. Soo editor, NUREG/CR-2482, BNL-NUREG-51494, Vol. 7, March 1985.
- NRC, 1985b. "NRC Comments on DOE Draft Environmental Assessment for the Hanford Site," U.S. NRC, March 20, 1985.
- Palmer, R.A., R.G. Johnston, and M.I. Wood, 1983. "Determination of an Upper Temperature Limit for Bentonite as a Backfill Component in a Nuclear Waste Repository in Basalt," RHO-BW-ST-47 P, 1983.
- Pusch, R., 1979. "Highly Compacted Sodium Bentonite for Isolating Rock Deposited Radioactive Waste Products," Nuclear Technology, Vol. 45, pp. 153-157.
- Salter, P.F., L.L. Ames, and J.E. McGarrah, 1981a. "Sorption of Selected Radionuclides on Secondary Minerals Associated with the Columbia River Basalts," RHO-BWI-LD-43, April 1981.
- Salter, P.F., L.L. Ames, and J.E. McGarrah, 1981b. "The Sorption Behavior of Selected Radionuclides on Columbia River Basalts," RHO-BWI-LD-48, August 1981.
- Seyfried, W.E., and J.L. Bischoff, 1981. "Experimental Seawater-Basalt Interaction at 300°C, 500 bars, Chemical Exchange, Secondary Mineral Formation and Implications for the Transport of Heavy Metals", *Geochemica et Cosmochemica Acta*, Volume 45, pp. 135.
- Seyfried, W.E., D.R. Janecky, and M.J. Mottl, 1984. "Alteration of the Oceanic Crust: Implications for Geochemical Cycles of Lithium and Boron", *Geochemica et Cosmochemica Acta*, Volume 48, pp. 557.
- Seyfried, W.E. and M.J. Mottl, 1982. "Hydrothermal Alteration of Basalt by Seawater Under Seawater-Dominated Conditions", *Geochemica et Cosmochemica Acta*, Volume 46, pp. 985.

Smith, M.J. et al., 1980. "Engineered Barrier Development for a Nuclear Waste Repository Located in Basalt- An Integration of Current Knowledge," RHO-BWI-ST-7, May 1980.

Tardy, Y. and Garrels, R.M., 1976. "Prediction of Gibbs Energies of Formation: Relations among Gibbs Energy of Formation of Hydroxides, Oxides and Aqueous Ions", *Geochim. Cosmochim. Acta*, Volume 40, No.9, pp 1051-1056.

Tardy, Y. and Garrels, R.M., 1977, "Prediction of Gibbs Energies of Formation of Compounds from the Elements: Monovalent and Divalent Metal Silicates, *Geochim. Cosmochim. Acta*, Volume 41, No.1, pp. 87-92.

Weaver, C.E., 1979. "Geothermal Alteration of Clay Minerals and Shales: Diagenesis", prepared for Battelle, Office of Nuclear Waste Isolation, ONWI-21.

Westick, J.H. et al., 1982. "Permeability, Swelling and Radionuclide Retardation Properties of Candidate Backfill Materials," in "The Scientific Basis for Nuclear Waste Management, Vol. 4, S.V. Topp ed., North-Holland, New York, PP.329-336.

Westinghouse 1981. "Engineered Waste Package Conceptual Design- Defense High Level Waste (Form 1), Commercial High Level Waste (Form 1), and Spent Fuel (Form 2) Disposal in Basalt," AESD-TME-3113, Section 5 and Appendix A, September 1981.

Wood, M.I., J.F. Relyea, D.L. Lane, and R.A. Carlson, 1983. "Chemical and Physical Properties of Waste Package Packing Materials," SD-BWI-TI-155, September 1983.