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The Advantages of a Salt/Bentonite Backfill for Waste Isolation Pilot Plant Disposal Rooms

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THE ADVANTAGES OF A SALT/BENTONITE BACKFILL FOR WASTE ISOLATION PILOT PLANT DISPOSAL ROOMS

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Abstract

A 70/30 wt% salt/bentonite mixture is shown to be preferable to pure crushed salt as backfill for disposal rooms in the Waste Isolation Pilot Plant. This report discusses several selection criteria used to arrive at this conclusion: the need for low permeability and porosity after closure, chemical stability with the surroundings, adequate strength to avoid shear erosion from human intrusion, ease of emplacement, and sorption potential for brine and radionuclides.

Both salt and salt/bentonite are expected to consolidate to a final state of impermeability (i.e., $\leq 10^{-18}\mathrm{m}^2$) adequate for satisfying federal nuclear regulations. Any advantage of the salt/bentonite mixture is dependent, therefore, upon bentonite's potential for sorbing brine and radionuclides. Estimates suggest that bentonite's sorption potential for water in brine is much less than for pure water. While no credit is presently taken for brine sorption in salt/bentonite backfill, the possibility that some amount of inflowing brine would be chemically bound is considered likely. Bentonite may also sorb much of the plutonium, americium, and neptunium within the disposal room inventory. Sorption would be effective only if a major portion of the backfill is in contact with radioactive brine. Brine flow from the waste out through highly localized channels in the backfill would negate sorption effectiveness.

In summary, although the sorption potentials of bentonite for both brine and radionuclides are not ideal, they are distinctly beneficial. Furthermore, no detrimental aspects of adding bentonite to the salt as a backfill have been identified. Then two observations are the major reasons for selecting salt/bentonite as a backfill within the WIPP.

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1.0 INTRODUCTION

This report summarizes the available information about the consolidation characteristics of two backfill materials, pure crushed salt and 70% by weight salt/30% by weight bentonite, and justifies selection of a design-basis backfill for the Waste Isolation Pilot Plant (WIPP) disposal rooms. These two materials were the principal backfills proposed for WIPP (U.S. Department of Energy [DOE], 1980) prior to 1990; this report constitutes completion of a milestone in the WIPP Test Phase Plan (DOE, 1990a) in regard to their evaluation. The report does not include evaluation of the usefulness of the alternate backfills presently under examination by the Engineered Alternatives Task Force (EATF) (DOE, 1990b).

The disposal concept for transuranic (TRU) waste at the WIPP is to excavate rooms in a bedded salt formation 650 m below the surface of the earth and fill them and adjacent access drifts with waste. As much of the remaining void volume as possible will then be filled with a suitable backfill. Initially, pure crushed salt was considered the likely candidate for backfill because it can consolidate around the waste with time to form a dense, impermeable barrier (Sandia National Laboratories [SNL], 1979). Pure crushed salt is chemically and physically compatible with the salt surrounding the repository and is readily available. Major unknowns were (1) how dense the backfill would become with time, (2) how long it would take the backfill to achieve this state, (3) how impermeable the backfill would become to brine flow, and (4) whether it would have any sorption potential for brine or radionuclides.

Crushed salt mixed with bentonite was selected as a second candidate material for backfilling the disposal rooms as concern about release of radionuclides by excessive amounts of brine inflowing into the repository increased (see Lappin et al., 1989). The use of bentonite was considered advantageous because of its ability to sorb water, because of its radionuclide sorption properties, and because it is sufficiently plastic not to interfere with consolidation processes. The consensus was that a mixture of 70% by weight salt/30% by weight bentonite was considered optimum because 30% filler is about the smallest portion that assures continuous paths of sorbent (bentonite) throughout the mixture. Less bentonite will become isolated in individual pores, reducing its effectiveness for wicking brine from other regions of the backfill; more bentonite will reduce the quasi-static rigidity provided by the salt skeleton within the mixture and increase the cost of the backfill. Rigidity of the backfill is important to prevent formation of a slurry-like material with high mobility.

In comparison to pure crushed salt, several additional unknowns were associated with salt/bentonite backfill. Testing was needed to establish (1) whether bentonite's chemical affinity for water would be reduced by the large amount of impurities, such as magnesium, present in WIPP brine or in the waste, and (2) what swelling pressures might develop as a consequence of brine sorption. Swelling pressure is both advantageous and disadvantageous. If the bentonite is confined, swelling pressure can reduce the permeability of the mixture. However, swelling pressure may also represent a backstress that may oppose closure of the room by salt creep and thereby limit densification of the backfill.

A possible disadvantage of salt/bentonite mixtures was the observation from early consolidation tests (Pfeifle, 1990) that the consolidation rates of the 70% salt/30% bentonite mixture were much slower than the rates for consolidation of moist, pure crushed salt. An explanation for the decreased rates was that while a small amount of water is necessary for rapid consolidation of pure salt, bentonite simply sorbs all available free water in the mixture and reduces the salt component to a dry, less easily consolidated condition. Fortunately, as will be discussed later in this report, the decrease in consolidation rates, caused by addition of bentonite to the backfill, is offset by the fact that less consolidation is required to assure a given state of permeability in salt/bentonite mixtures; although densification proceeds more slowly, a less dense final state is required, and the time to reach a given permeability remains relatively constant.

In 1989, results from preliminary performance studies of the repository suggested that the high porosity and permeability of unprocessed waste might make compliance with the regulatory standards (specifically 40 CFR 191, U.S. Environmental Protection Agency [EPA], 1985) difficult.[1] These observations challenge the prior performance assessment assumption that extensive reprocessing of the waste would never be required; they suggest instead, that some degree of modification of the waste form might be required. The EATF was subsequently established by the DOE's WIPP Project Office to respond to these concerns. Their objective is to identify changes to the waste form or the repository design that could be beneficial, should the need arise, and to recommend which waste forms need to be considered for further study. As might be expected, alternative backfill material candidates are also under consideration by the EATF, such as pouring grout or concrete around the waste after emplacement in the disposal room.

---

[1] Unprocessed waste is material that is placed in containers without precompaction to reduce void volume or other treatments to transform the waste to a form more acceptable for storage at WIPP. However, some unprocessed wastes are cemented for stability.

The recommendations for alternate backfill materials under consideration by the EATF will not be considered in this report since their exact nature has yet to be defined (DOE, 1990b). Thus, this report is limited to the technical considerations in progress prior to formation of the EATF, which involved comparison of the two salt-based materials. It is organized as follows: (A) some of the reasons for backfilling a repository are described in the first part of Section 2.0, followed by statements of the criteria used here in backfill selection; (B) room closure estimates are described in Section 3.0, because they provide much of the basis for the selection process; (C) the required selection criteria are applied to the two backfill materials in Section 4.0; (D) desirable criteria are applied in Section 5.0; and (E) the overall results of the assessment are summarized in Section 6.0.

2.0 BACKFILL SELECTION CRITERIA

2.1 Reasons for Backfilling the WIPP

The current concept of waste emplacement in the WIPP assumes that almost all empty access-ways, drifts, and waste-filled rooms will be backfilled with materials tailored to improve the results of the closure process, i.e., to reduce long-term flow of brine through the repository and subsidence above its excavated regions. Additions of various other components may also be desirable to alleviate other aspects of performance. These additives presently fall into materials that: (1) adsorb water; (2) getter, i.e., sorbers or materials altering the production of decomposition and corrosion gases; (3) sorb radionuclides and/or buffer the local pH within a relatively narrow range.

Once waste is emplaced, the disposal rooms and access-ways will be backfilled to eliminate as much of the initial void volume as possible to accelerate the transition (closure) of the repository to a consolidated state. Rapid consolidation of the backfill is desirable because the possibility of the voids filling with brine and causing termination of closure is less likely. If the voids fill with brine, consolidation ceases in most cases. Present predictions of the amount of brine inflowing into the repository (described later in this report) suggest, however, that consolidation is not likely to be limited by voids filling up with fluid; low enough porosities will be attained before brine saturation. Reduction of void volume or porosity is also desirable because the less porous the backfill, the lower its permeability. The permeability of a material is directly related to how little porosity it contains. Finally, gas present within the voids will retard brine inflow by exerting a back pressure and introducing capillarity effects that must be overcome. The generation of back pressures may also have the adverse effect of enhancing void volume by limiting or even reversing closure.

Another reason for limiting void volume within the backfill is related to the escape of soluble radionuclides from the immediate vicinity of the repository. In this case, smaller void volumes lead to both smaller volumes of brine as the repository saturates and less fluid being available for the solution of radionuclides. Backfilling the repository also limits damage to the surrounding salt formation by reducing the extent of closure needed to reach mechanical equilibrium. With less closure, large cracks in the surrounding salt become less likely, reducing subsidence effects such as the tendency of the disposal room roof to fracture and cave in, and limiting the cracks' role as conduits of brine to the waste. Backfilled regions can also function as barriers to inflowing brine. All of these effects depend upon achievement of low porosity within the backfill.

2.2 Criteria for Backfill Selection

Development of backfill-selection criteria is dependent on the observation that consolidation must proceed to the point where the backfill has a sufficiently low enough permeability to reduce radioactive brine flow to levels that satisfy regulatory requirements. The state of consolidation of the backfill is emphasized in this statement because almost all of the properties of the backfill depend on its final density. The approach to backfill evaluation thus appears deceivingly simple: we need only to determine how much radioactive brine flow is permitted and then select a backfill that will restrict flow to below the allowable amount. Implementation of evaluation is thwarted, however, by the realization that the allowable brine flow can only be established from very complex and detailed performance-assessment analyses. These calculations will not be completed for several years. Another constraint on the evaluation process is that performance-assessment analysis tends to be a "forward direction only" process where a state of the repository is assumed, and compliance or lack of compliance is established. Analysis in the reverse direction, using the regulatory requirements to determine unique property values for the backfill, is not currently feasible.

The elusiveness of the relationship between the state of the repository and satisfactory performance requires that less-exact criteria be developed. Ideal criteria would be in the form of quantitative statements that a given property of the backfill, such as permeability, not exceed a given value. These statements are not always possible to define, but if more quantitative statements for some of the criteria adopted in this report are possible in the future, re-evaluation of the given aspect of backfill performance will be required.

Selection of either pure crushed salt or salt-bentonite for backfilling the disposal room will be on the basis of two criteria levels. Required criteria describe those conditions that are considered necessary to satisfy federal nuclear regulatory requirements. If a material cannot meet these criteria, it should not be considered as a backfill for the WIPP. Desirable criteria are those that would be useful to achieve, in the sense of good engineering practice. Failure to meet desirable criteria does not mean rejection of a material as backfill. However, it would be advantageous, but not essential, to satisfy them.

2.2.1 REQUIRED CRITERIA

2.2.1.1 Low Permeability

The permeability criteria are divided into two parts: one defines how low the permeability of the backfill must be, a second part considers aspects of performance related to the porosity of the backfill.

Permeability: Performance-assessment studies have shown that the permeability of the consolidated backfill to brine should be within three orders of magnitude (less than 10^{-18} m^2) of the permeability of intact salt (10^{-21} m^2) (Rechard et al., 1990, Figure 4-2) in order to restrict brine flow through the backfill to an intruding borehole. The basis for this assumption will be described later in the report.

Porosity: Either the backfill must have the prescribed low permeability upon emplacement without further consolidation, or it must achieve this state by creep consolidation before the repository has time to saturate with brine. The condition on porosity relates to the saturation restriction and requires verification that the voids do not fill with brine before low permeability is achieved. Additional closure after saturation is possible, but would be almost impossible to demonstrate. We assume, in defining this criterion, that permeability and solubility considerations, with regard to the backfill, outweigh void-volume considerations related to gas storage.

Conditions on permeability and porosity are combined in evaluation of the two backfills because these variables are not mutually exclusive.

2.2.1.2 Chemical Stability

The backfill should not be chemically reactive with brine and/or various other elements within the waste to the extent that gas is generated or permeability enhancement above 10^{-18} m^2 occurs.

2.2.1.3 Shear Strength

The consolidated backfill should have sufficient shear strength to resist wellbore enlargement (breakout) at the time of a human intrusion. The backfill should have a strength at least three orders of magnitude greater than the 6 Pa (0.006 MPa) critical shear strength for borehole enlargement by scouring assumed for the "erosion" analysis described in Lappin et al. (1989, pp 5-9).

2.2.1.4 Ease of Emplacement

Any reasonably effective backfill that can be emplaced without significant risk or major expense should be considered. Specification of what constitutes reasonable cost is beyond the scope of this evaluation.

2.2.2 DESIRABLE CRITERIA

2.2.2.1 Effect of Additives

The performance of the backfill should not be significantly degraded by the inclusion of additives such as gas getters.

2.2.2.2 Brine Sorption

A backfill with brine sorption properties is desirable as long as the hydrous products have acceptable stability.

2.2.2.3 Radionuclide Sorption

Sorption of radionuclides by the backfill is desirable.

2.2.2.4 Deformability (ability to fill voids)

The backfill should be able to intrude or flow into adjacent voids. If disturbed, the backfill should have the ability to deform and partially or totally recover its low-permeability state.

2.2.2.5 Thermal Conductivity

The backfill should have sufficient thermal conductivity to minimize any localized buildup of heat. No buildup of heat within unprocessed TRU waste is likely; therefore, this criterion does not apply to the present evaluation. However, it is included in the list of criteria to cover the future possibility that processing of the waste could concentrate sufficiently to create localized hot spots.

3.0 CLOSURE RATE AND TIME ESTIMATES

3.1 Configurations and Assumptions

The consolidation characteristics of pure crushed salt backfill and 70% salt/30% bentonite backfill must be examined because some of the performance criteria depend on how dense the backfill becomes as a function of time. Two calculations for each type of backfill are presented: (1) two-dimensional closure of a room entirely filled with backfill, and (2) two-dimensional closure of a room filled with backfill and TRU waste. Internal gas pressurization is ignored. The results of both types of calculations describe what happens in planes perpendicular to the longitudinal axis of infinitely long rooms. In almost all cases, results from two different numerical codes are reported for the same configuration: SNL results were calculated with the finite-element, finite-strain code SANCHO (Stone et al., 1985); the RE/SPEC results were obtained with the finite-element, small strain code SPECTROM-32 (Callahan et al., 1989). Although the codes appear to be identical in their content, they are actually quite different in several respects, as shown in Table 3-1. Therefore, comparison of results from the two different codes for identical configurations has the additional benefit of providing a measure of the differences that are to be expected between results from independent calculations.

The analyzed room configurations were approximately the same as the currently accepted configuration of the disposal room with the exceptions that a 2-ft air gap at the top of the room was omitted because its presence occasionally caused numerical-stability problems, and stratigraphy within the salt was ignored. These omissions are not likely to influence the results greatly. Ignoring the gap simply means that simulated consolidation in the calculations presented here starts immediately rather than after the short time predicted for closure of the 2-ft gap. Gap closure is estimated to occur within less than ten years. Similarly, the inclusion of stratigraphy in past calculations greatly increased problem running time on the computer without improving the agreement between predicted and observed initial closure rates. Another major assumption was that the room was symmetric with regard to both its vertical centerline and its horizontal centerline (Figure 3-1). The symmetric condition was adopted to minimize computer running time (usually of the order of hours of CRAY time).

The results of the backfill closure analyses are shown in Figures 3-2 through 3-7. The computed closure histories for a room completely filled with crushed-salt backfill are shown in Figure 3-2 (Weatherby, 1989) (Callahan and DeVries, 1991). The average void fraction in the room, although equivalent in

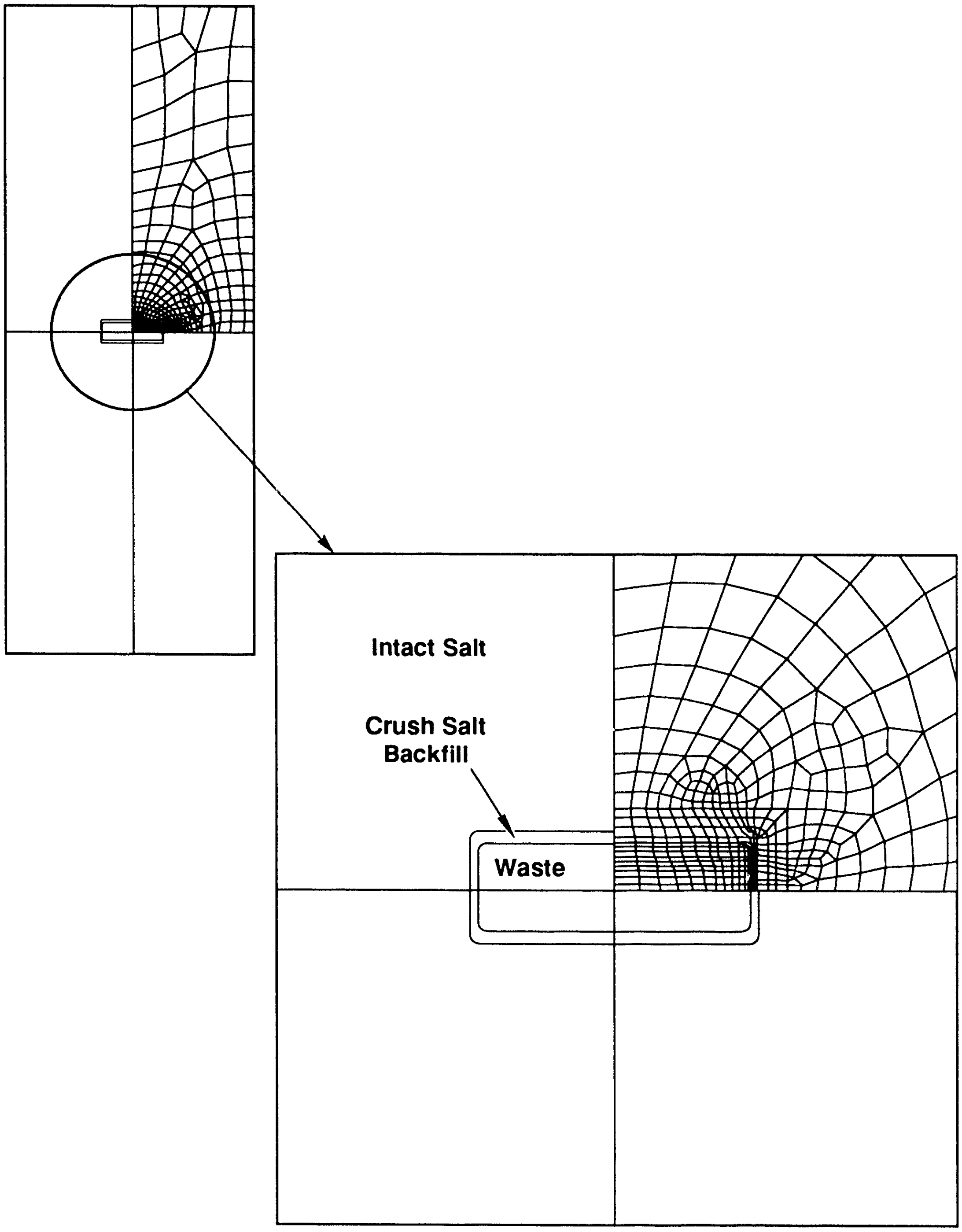

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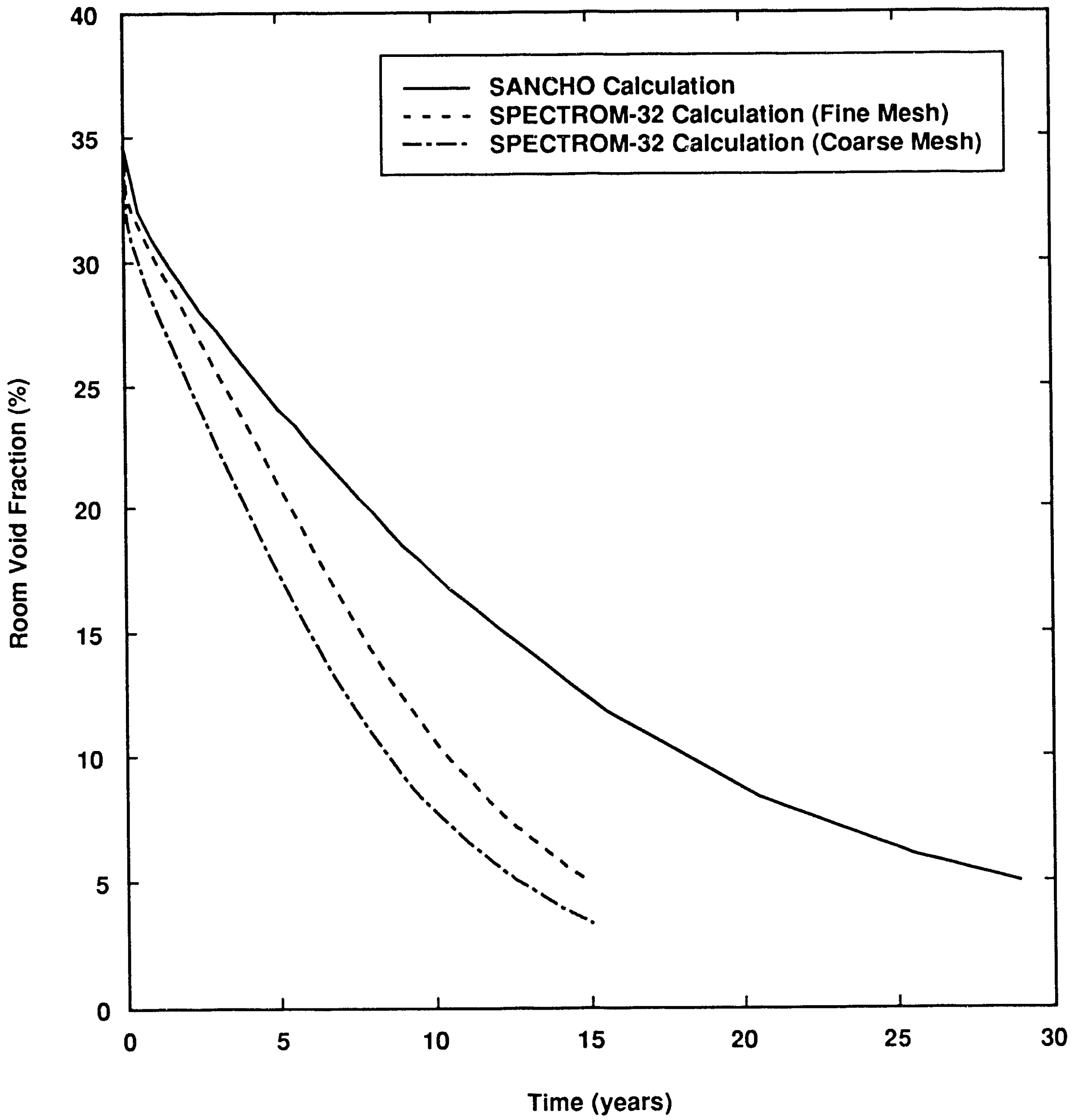
Figure 3-1. Plane strain-finite element model of a TRU storage room.



TRI-6345-1-0

Figure 3-2. Predicted void fraction history for a disposal room entirely filled with pure crushed-salt backfill.

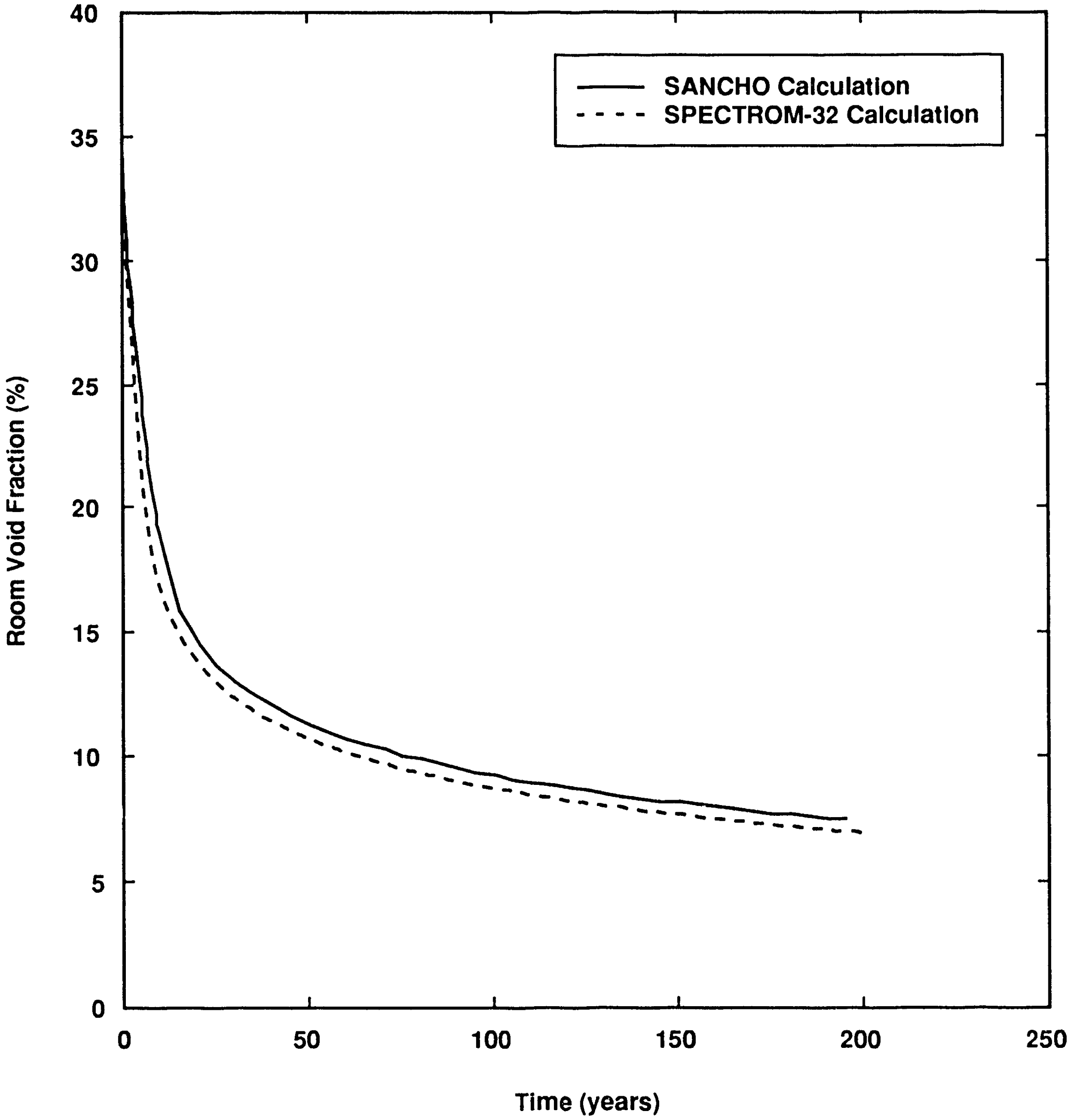


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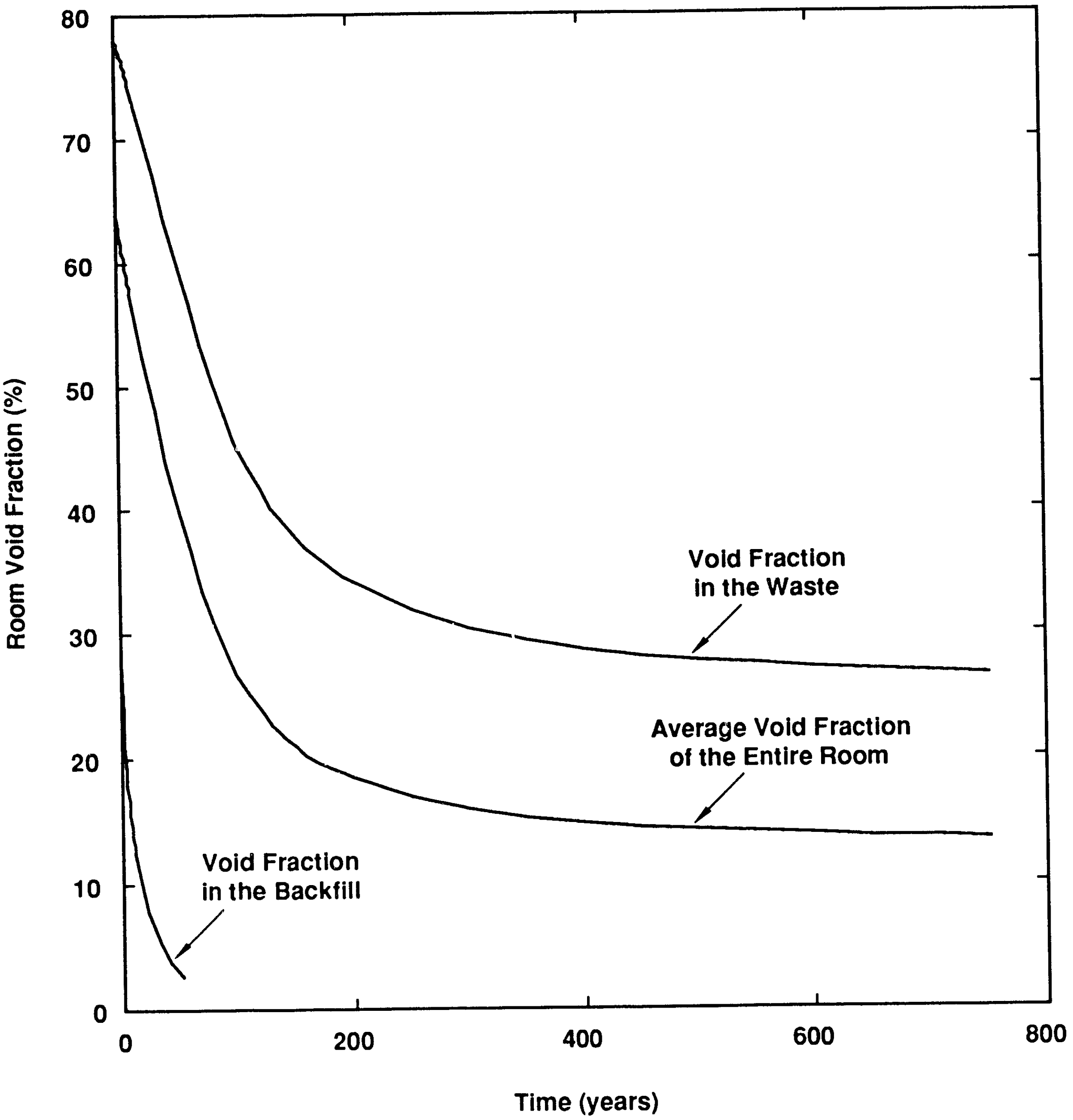
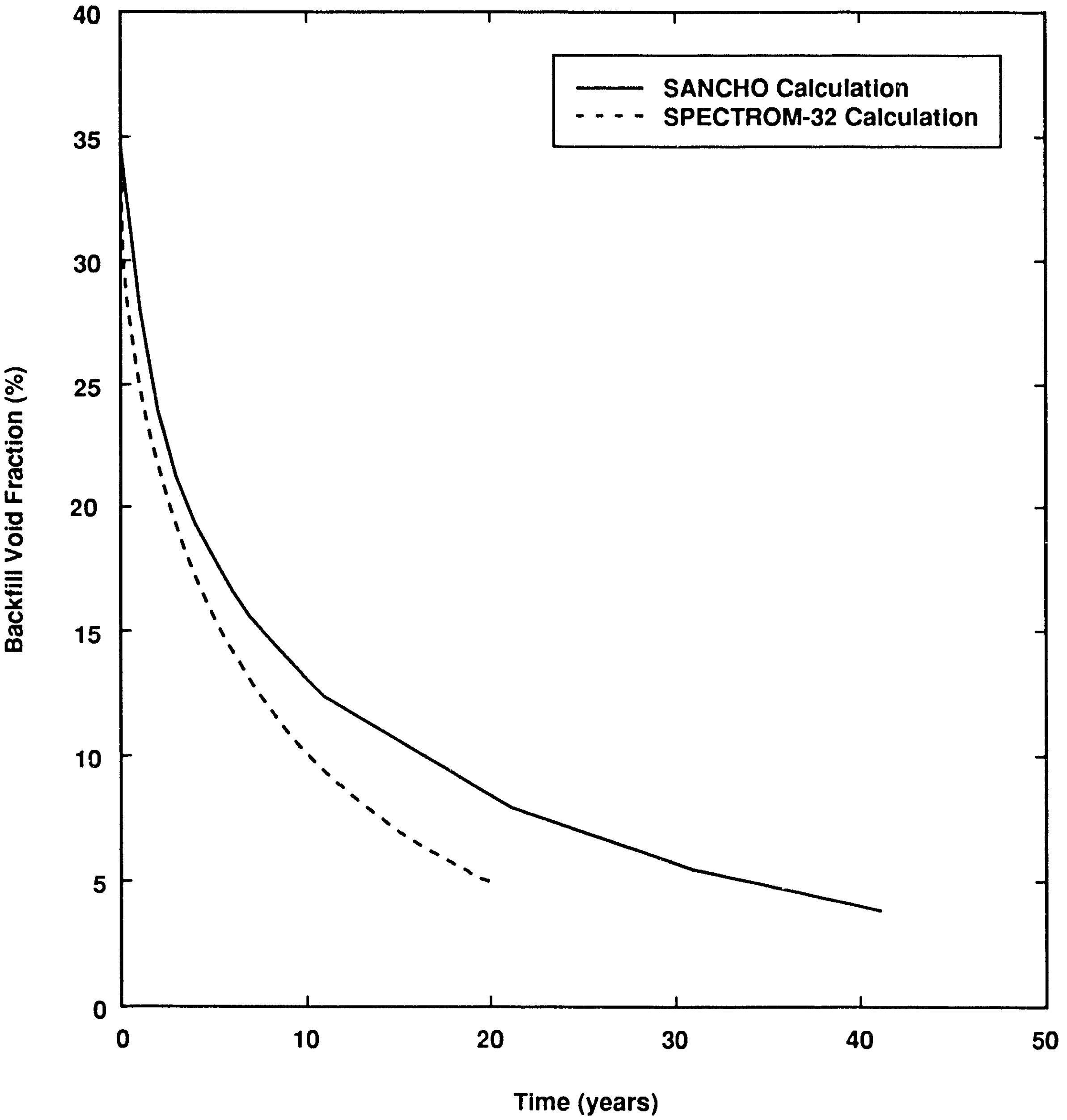
Figure 3-3. Predicted void fraction history for a disposal room entirely filled with 70% salt/30% bentonite backfill.



TRI-6345-3-0

Figure 3-4. Predicted average void fraction-time history of a disposal room filled with TRU waste and pure crushed-salt backfill using SANCHO. The room void fraction curve represents the sum of the backfill void volume and waste void volume divided by the volume of the entire room.



TRI-6345-4-0

Figure 3-5. Predicted average backfill void fraction for a disposal room filled with TRU waste and pure crushed-salt backfill.

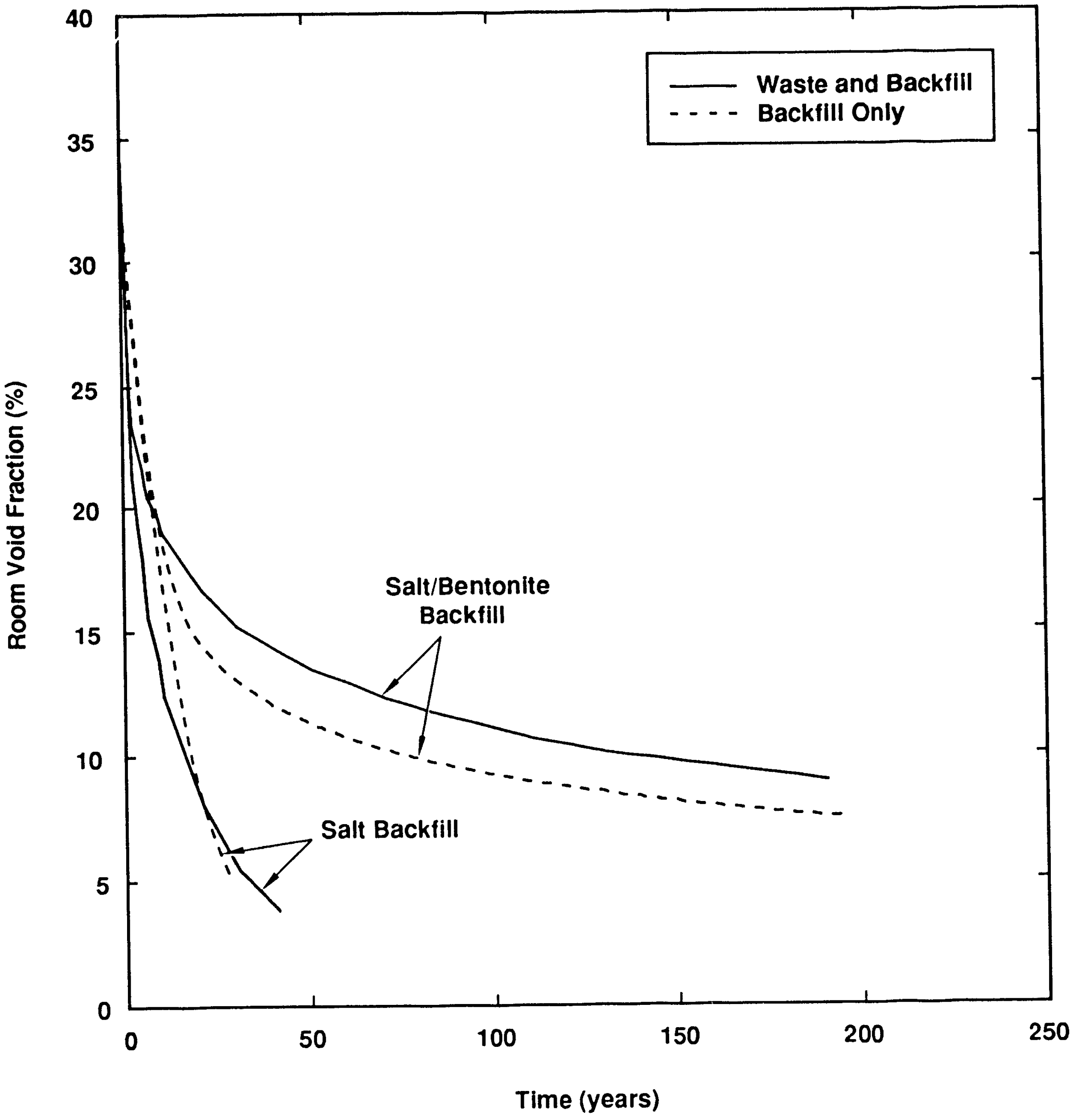


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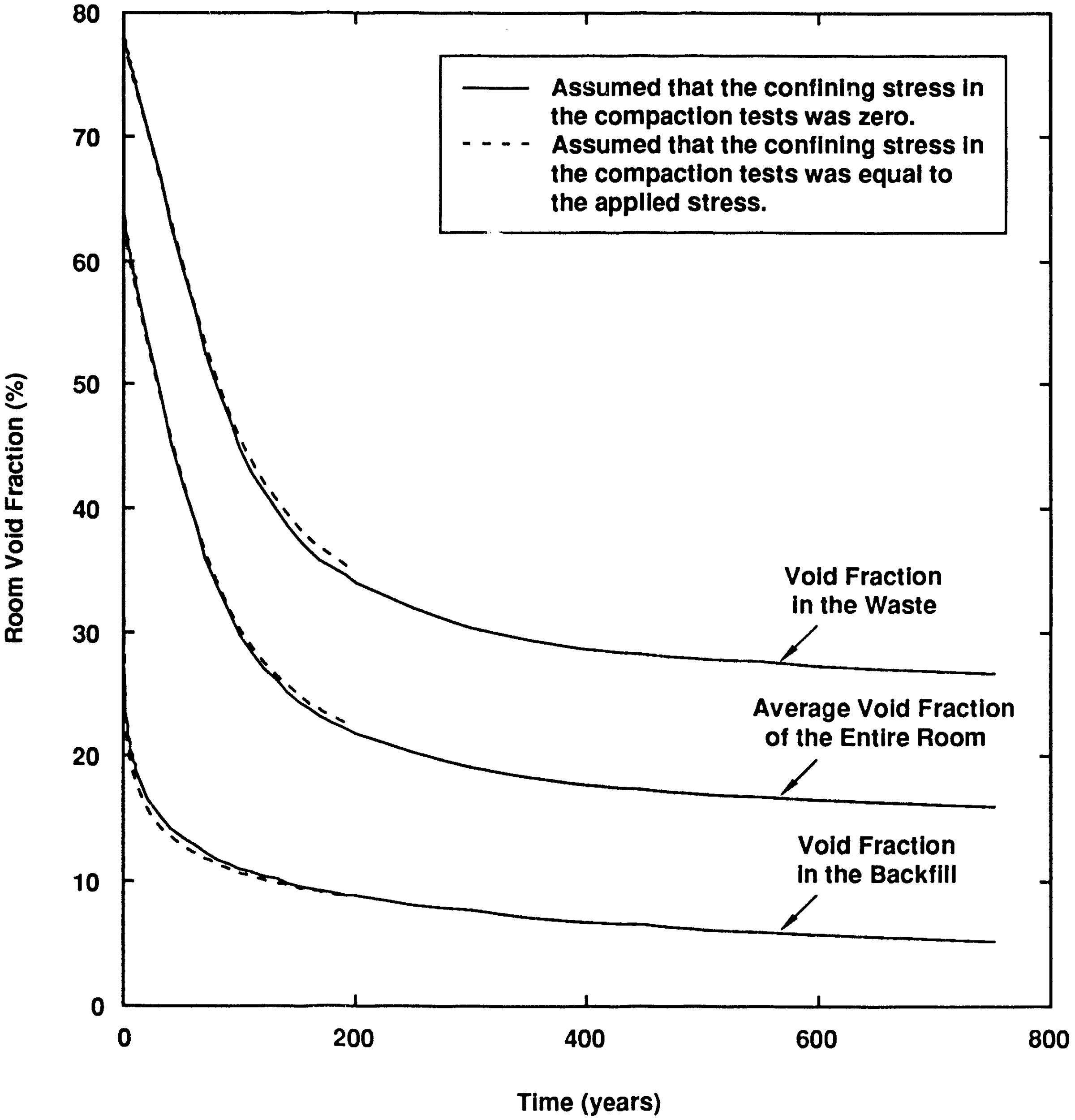
Figure 3-6. A comparison of backfill consolidation in rooms completely filled with backfill and rooms filled with both waste and backfill as computed with SANCHO.



TRI-6345-6-0

Figure 3-7. Predicted average void fraction-time history of a disposal room filled with TRU waste and 70% salt/30% bentonite backfill.

TABLE 3-1. CLOSURE ANALYSIS FINITE-ELEMENT CODE CAPABILITIES

	SANCHO[1]	SPECTRUM-32[2]
Finite strain capability	yes	no
Salt creep model	E/12.5 approximation,[3] Von Mises flow[4]	Munson-Dawson model, Tresca flow[5]
Backfill creep model		
Moist crushed salt	Sjaardema-Krieg model[6]	Sjaardema-Krieg model, with modifications[7]
Moist salt-bentonite	Sjaardema-Krieg model[6]	Sjarrdema-Krieg model, with modifications[7]
TRU waste	Volumetric plasticity model[1], using the average repository consolidation curve[8]	Nonlinear elastic models, using the average repository consolidation curve[8], with modifications[9]

---

[1] Stone et al., 1985
[2] Callahan et al., 1989
[3] Calculations were indicated before Munson-Dawson model and the Tresca flow options were available in SANCHO. Differences between the two models are considered secondary because both descriptions profess to reproduce the same experimental room closure data.
[4] Morgan and Krieg, 1990
[5] Munson et al., 1989
[6] Sjaardema and Krieg, 1987
[7] Morgan, 1987
[8] Butcher et al., 1991
[9] Nieland, 1990

value to average room porosity, is used as a variable in this plot to emphasize that it assumes that void volume is distributed uniformly throughout the backfill. Values ascribed to "room void fraction" are computed from the total void volume within the room at any given time divided by the volume of the room at the same time. The term "porosity" will be used as a variable to describe more subtle variations of void volume, such as how the void volume within the backfill varies from location to location within the room at any given time.

All of the consolidation curves for pure crushed-salt backfill in Figure 3-2 and subsequent figures have been edited to remove data below 5% void volume. This procedure was considered necessary because experimental data are not available for determining how accurately the constitutive equation reproduces the mechanical response of crushed salt at such low porosities. Omission of this part of the data will have no effect on the backfill selection process described here, because acceptable permeabilities in all cases already have been achieved before the time the salt has consolidated to less than 5% porosity, as shown later in this report.

3.2 Closure of A Room Entirely Filled With Crushed-Salt Backfill

The computed closure histories for a room completely filled with backfill in Figure 3-2 illustrate the difference in closure histories predicted by the two numerical codes. Computed times for consolidation to 5% void fraction are 12.5 to 15 years, according to the the SPECTROM-32 calculation (Callahan and DeVries, 1991), and 29 years according to the SANCHO calculation (Weatherby, 1989). Closure, according to the SPECTROM-32 calculations, is more rapid than the SANCHO results, an observation that is also true for all other calculated histories presented in this report. These discrepancies are not considered significant, even though the time to reach a given void fraction, according to the two calculations, can differ by as much as 16 years, because much longer times would be required for brine saturation to impede closure, as discussed later in this report. Both the coarse-mesh and fine-mesh results from SPECTROM-32 are included in Figure 3-2 to provide an indication of uncertainties introduced by the trade-off between problem complexity and program running time. Computer time for solution of the coarse-meshed problem was less than for the fine-meshed problem, but the fine-meshed result was considered a more accurate prediction.

3.3 Closure of A Room Entirely Filled With Salt/Bentonite Backfill

The computed history for closure of a disposal room completely filled with salt/bentonite backfill is shown in Figure 3-3 (Weatherby and Brown, 1990; Callahan and DeVries, 1990). When compared to the closure histories for pure crushed salt (Figure 3-2), the salt/bentonite curves show the retarding effect of bentonite on salt consolidation (Pfeifle, 1990). Evidence that less consolidation is required to assure a given state of low permeability in salt/bentonite mixtures will be discussed later in this report. Although densification of salt/bentonite is known to proceed more slowly, this aspect of its response would not automatically eliminate it for consideration as a backfill material.

3.4 Closure of A Room Filled With Waste and Pure Crushed-Salt Backfill

Results of calculations to estimate closure of a room filled with waste and pure crushed-salt backfill are shown in Figure 3-4 (SANCHO calculation, Weatherby and Brown, 1991). Currently, these calculations represent the most realistic prediction of repository response because they include the effects of backstress exerted by both the backfill and the waste. Although curves for the backfill are the only information needed for this report, the other curves in the SANCHO calculation (Figure 3-4) show how the waste responds and the average state of room consolidation. Of great importance is the observation that the backfill consolidates before the waste because of its relative stiffness.

A comparison between the SANCHO backfill void-fraction results and SPECTROM-32 backfill void-fraction results for waste-filled rooms is shown in Figure 3-5. (Only the backfill results are shown; curves for the waste and the average room void-fractions have been deleted from this figure.) These results confirm the observation in Section 3.2 (room entirely filled with backfill) that backfill consolidation according to the SPECTROM-32 calculation is more rapid than the SANCHO results, but the difference is not considered significant.

There is little difference between the consolidation history for a room entirely filled with crushed salt (Figure 3-2) and the backfill-consolidation curve for a room filled with waste and pure crushed salt (Figure 3-4), as shown in Figure 3-6. This agreement occurs even though the volume of the backfill in the filled room is almost three times the volume of the room filled with waste and backfill and may be simply a consequence of the fact that very little backstress is exerted by the backfill during the early stages of closure.

3.5 Closure of A Room Filled With Waste and Salt/Bentonite Backfill

The final calculations are an estimate of the closure of a room filled with TRU waste and salt/bentonite backfill (Figure 3-7) using SANCHO (Weatherby and Brown, 1991). SPECTROM-32 results for this configuration were not available. Waste-compaction data for these calculations were obtained from samples of simulated waste, which were laterally confined within rigid dies during the tests. Two compaction models had to be considered (Weatherby and Brown, 1991) to compensate for the fact that the simulated waste forms were too heterogeneous to permit direct measurement of lateral stresses: (1) a model based on the assumption that the confining stress during uniaxial laboratory compaction tests on the various waste types was zero, and (2) a model based on the assumption that the confining stress in the compaction tests was equal to the applied stress. The magnitudes of any shear stresses generated during consolidation will lie between these two bounds. Assumption 2 represents more fluid-like response, with very low shear stresses developing during consolidation; assumption 1 represents a material that can support large shearing stresses. Figure 3-8 shows that the results are not strongly dependent upon the particular assumption used for the prediction.

As with pure crushed salt, the results for salt/bentonite backfill show that the backfill-consolidation curve for a room completely filled with salt/bentonite is very similar to the curve for a room filled with waste and salt/bentonite. This agreement is in spite of the fact that the volume of the backfill in the backfill-filled room is almost three times the volume of the room with both waste and backfill and partly a consequence of the fact that the void fraction represents normalization of the void volume with respect to current room volume.

4.0 PERFORMANCE ASSESSMENT: REQUIRED CRITERIA

4.1 Permeability

4.1.1 EXPERIMENTAL PERMEABILITY RESULTS

Changes in backfill permeability during closure are the single most important factor in selection of a backfill: (1) a sufficiently low permeability state in the disposal room must be achieved, within three orders of magnitude of the permeability of intact salt (Rechard et al., 1990, Figure 4-2); (2) this state of low permeability must occur before the backfill becomes fully saturated with brine. Investigations of the effect of permeability were initiated by summarizing published test data on the permeability of each backfill as a function of fraction, f_s, of (theoretical) solid density. The fraction of (theoretical) solid density, is computed from $1 - f_v$, where f_v is the void fraction; f_s is useful as a convenient parameter for examining how changes in input parameters alter the Sjaardema-Krieg (1987) constitutive model for backfill used for the closure calculations. This model predicts that a semi-logarithmic plot of consolidation rate versus fraction-of-solid-density data, for a given stress, should define an approximately straight line. The fractional density values for Figure 4-1 were computed using the theoretical solid densities: $f_s = 2140$ kg/m^3 for salt, and $f_s = 2700$ kg/m^3 for bentonite, which combine to give a value of $f_s = 2282$ kg/m^3 for the salt/bentonite mixture. Another assumption was that an upper bound of 10^{-21} m^2 was a reasonable permeability for salt/bentonite tests (Pfeifle, 1990) for which flow could not be established within 160 days.

The results of the data summary, shown in Figure 4-1, were used to construct two standard curves for permeability versus fraction of (theoretical) solid density. The solid curve is for pure crushed salt, based on Holcomb's data (Stormont, 1988), and the dashed curve is for salt and bentonite. Last-minute additions of salt/bentonite data to Figure 4-1 were made after most of the calculations in this report were completed, which suggested that some upward adjustment of the upper (high permeability) part of the standard curve for salt/bentonite might be appropriate. However, no calculations were redone because the results of the evaluation were controlled by the lower part of the curve, which did not change.

4.1.2 UNCERTAINTIES INTRODUCED BY DIFFERENCES BETWEEN THE WIPP AND THE GERMAN PERMEABILITY DATA

Figure 4.1 shows that the permeability results from different investigations are in general agreement with each other, with one major exception: according to data reported by the Germans, greater consolidation is

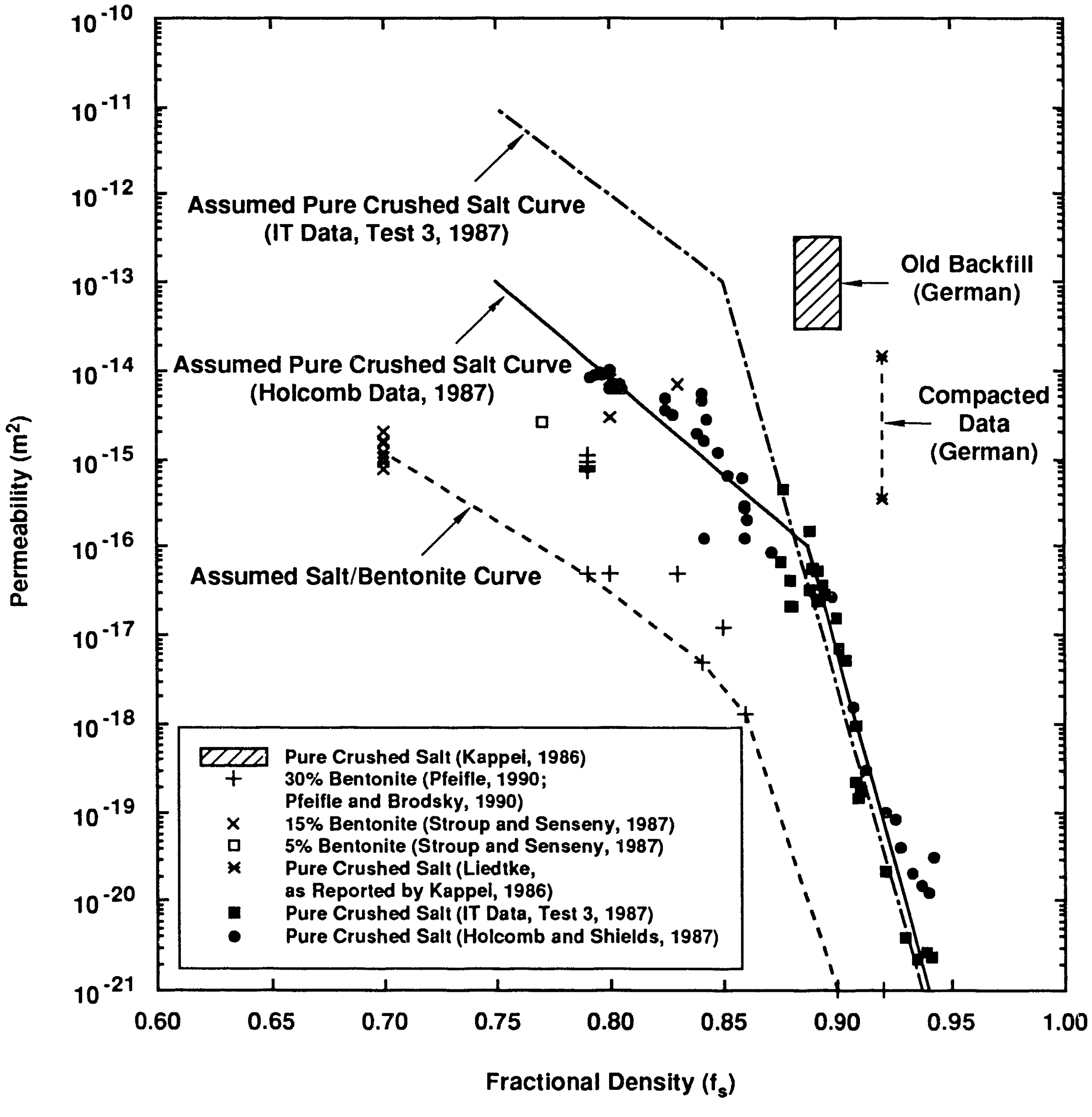


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Figure 4-1. Salt and salt/bentonite permeability data.

required to achieve a given permeability than other permeability data (Holcomb and Shields, 1987; IT, 1987) would indicate. The comparison is troublesome because greater consolidation implies a longer time to reach an acceptable state of the backfill, which is counter to the expectation that closure time should be as short as possible.

Differences in sample-moisture contents and methods of measuring permeability may explain the discrepancy between the WIPP and the German data. The measurements on the WIPP salt were made with argon gas on samples with an initial moisture content of 2.4 to 2.5%. This moisture content can impede gas flow because of saturation threshold and/or relative permeability effects (Davies, 1990) when the backfill becomes dense, but is still partly saturated. Simply stated, water trapped in sufficiently small voids because of capillarity forces can block the flow of gas until the gas pressure driving flow is large enough to force the brine out. While detailed information about the method used by the Germans to measure the permeability of consolidated salt is not available, it is thought to have involved the use of brine flowing through saturated samples (Kappei, 1986). With no threshold effect present, these tests might have produced higher permeabilities, for a given density, than were observed for the partially saturated samples of WIPP salt. Disturbance of samples during acquisition could be another reason for the higher observed permeabilities. The measurement of crushed WIPP salt to brine under fully saturated conditions will be the objective of future experiments.

4.1.3 PERMEABILITY HISTORIES

Changes in average permeability of a given backfill as a function of time were determined from a given closure calculation by cross-plotting the closure results with the appropriate solid-line or dashed-line permeability curve in Figure 4.1. Closure histories for these predictions were limited to SANCHO predictions because, in all cases examined, SANCHO predictions gave greater times for closure to a given porosity than were obtained using SPECTROM-32. The SANCHO results represented, therefore, a conservative estimate of time for closure, i.e., they represent the largest time required for a given permeability to result from closure.

As an example of the analytical method, the computed permeability as a function of time for a room completely filled with crushed-salt backfill is shown in Figure 4-2. This curve was obtained by cross-plotting the curve from SANCHO, in Figure 3-2, with permeability relationship (Holcomb and Shields et al., 1987) for pure crushed salt (solid line curve), shown in Figure 4-1.

Because of the uncertainty about the experimental permeability information for pure crushed salt, it was also instructive to investigate the extent to which evaluation results were sensitive to variations of the permeability data.

This check was made by constructing a different permeability versus fraction-of-theoretical-density relationship for pure crushed salt by shifting the WIPP salt-permeability curve from the data in Figure 4-1 to higher fractions of solid density. The shift in curve position was enough to make it pass through the German results on old backfill (Figure 4-3), but not enough to make it pass through the German "compacted data." This procedure was adopted because no information was available about the distribution of particle sizes in the "compacted data" samples. Many of the German tests have been on samples of single-sized particles, which are much harder to compact than samples containing distributions of particle sizes, and the concern was that if the compacted data fell into this category, their use would bias the evaluation.

Also shown in Figure 4-2 is the cross-plot of adjusted permeability results with the consolidation history for a room filled with crushed salt. Comparison with the unaltered permeability-time curve shows that the change in the permeability history of the room is slight, largely because consolidation of pure crushed-salt backfill is so rapid. The implication is that uncertainty in the permeability data can be quite large for pure crushed salt and still cause little change in the time to reach acceptable permeabilities.

The same conclusion for salt/bentonite backfill may not be valid, however, because salt/bentonite does not consolidate as rapidly as pure crushed salt does. Fortunately, the data on salt/bentonite from different sources were more consistent than the pure-salt data; therefore, the uncertainty in its application was less. Data consistency was probably better because all of the permeability measurements for material reported here were made with brine on fully brine-saturated samples.

4.1.4 BASIS FOR EVALUATION OF PERMEABILITY HISTORIES

Before proceeding with the closure analyses, it is useful to describe the first part of the permeability criterion defined in Section 2.2.1.1. The significance of the permeability histories is interpreted as follows. We know that consolidation must proceed to the point where the backfill becomes sufficiently impermeable to prevent unacceptable brine flow. Further, how much brine flow is acceptable can only be determined from detailed future performance-assessment analyses. Therefore, less stringent criteria must be developed in order to anticipate the results of the final performance calculations. The criterion that will be used is based on calculations by Rechard et al. (1990, Figure 4-2), which show that retardation of brine flow by the backfill, in the event of human intrusion, is unlikely unless the backfill permeability is of the order of 10^{-18} m^2 or less. This value of permeability and the corresponding state of consolidation will be used as an upper bound (maximum permeability) for the condition that must be achieved to make the performance of the backfill acceptable.

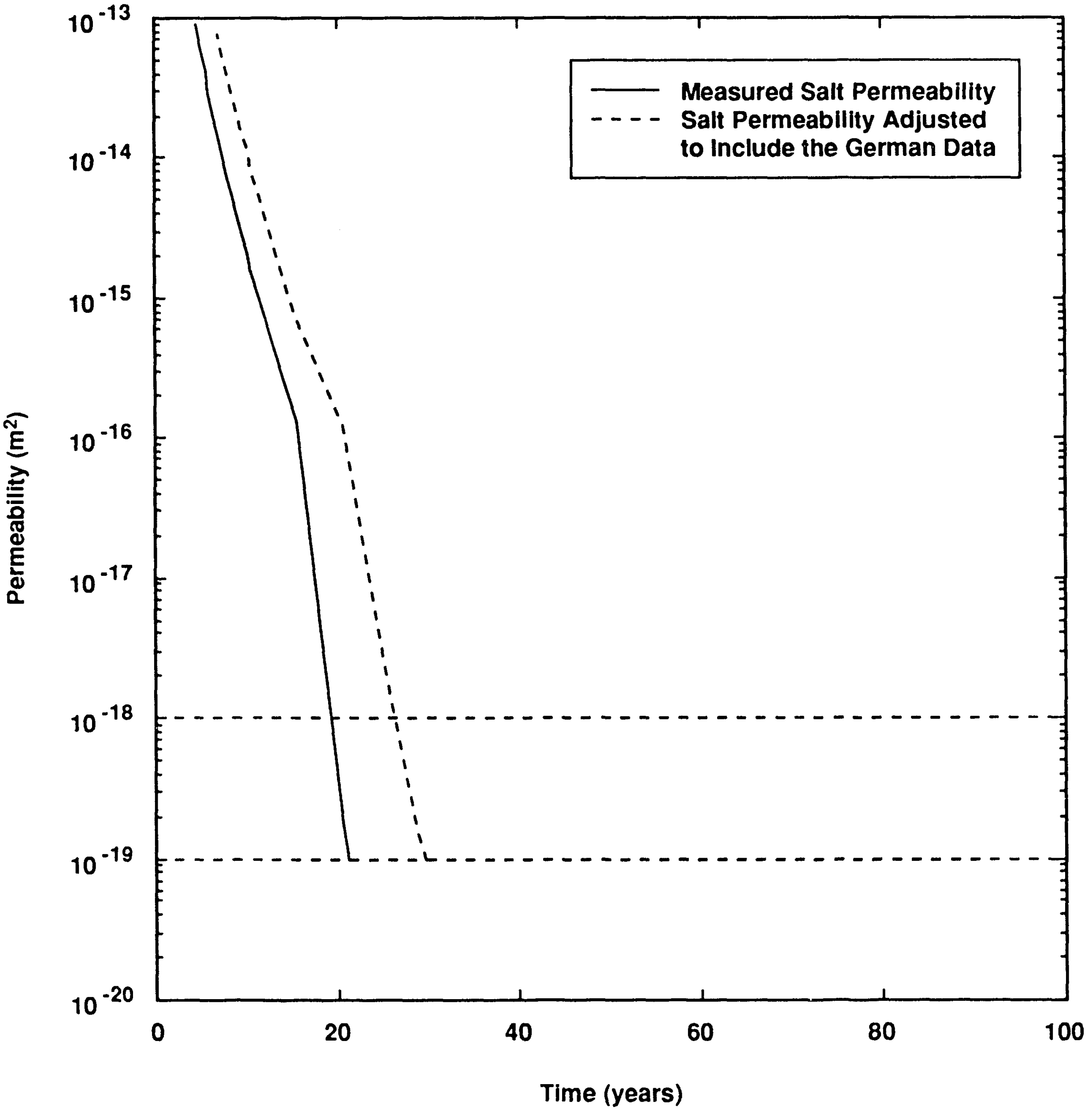

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Figure 4-2. Disposal room closure permeabilities for rooms completely filled with pure crushed-salt backfill. The portions of the curves below 10^{-19} m^2 are not shown because the predictions at such low permeabilities are considered inaccurate.

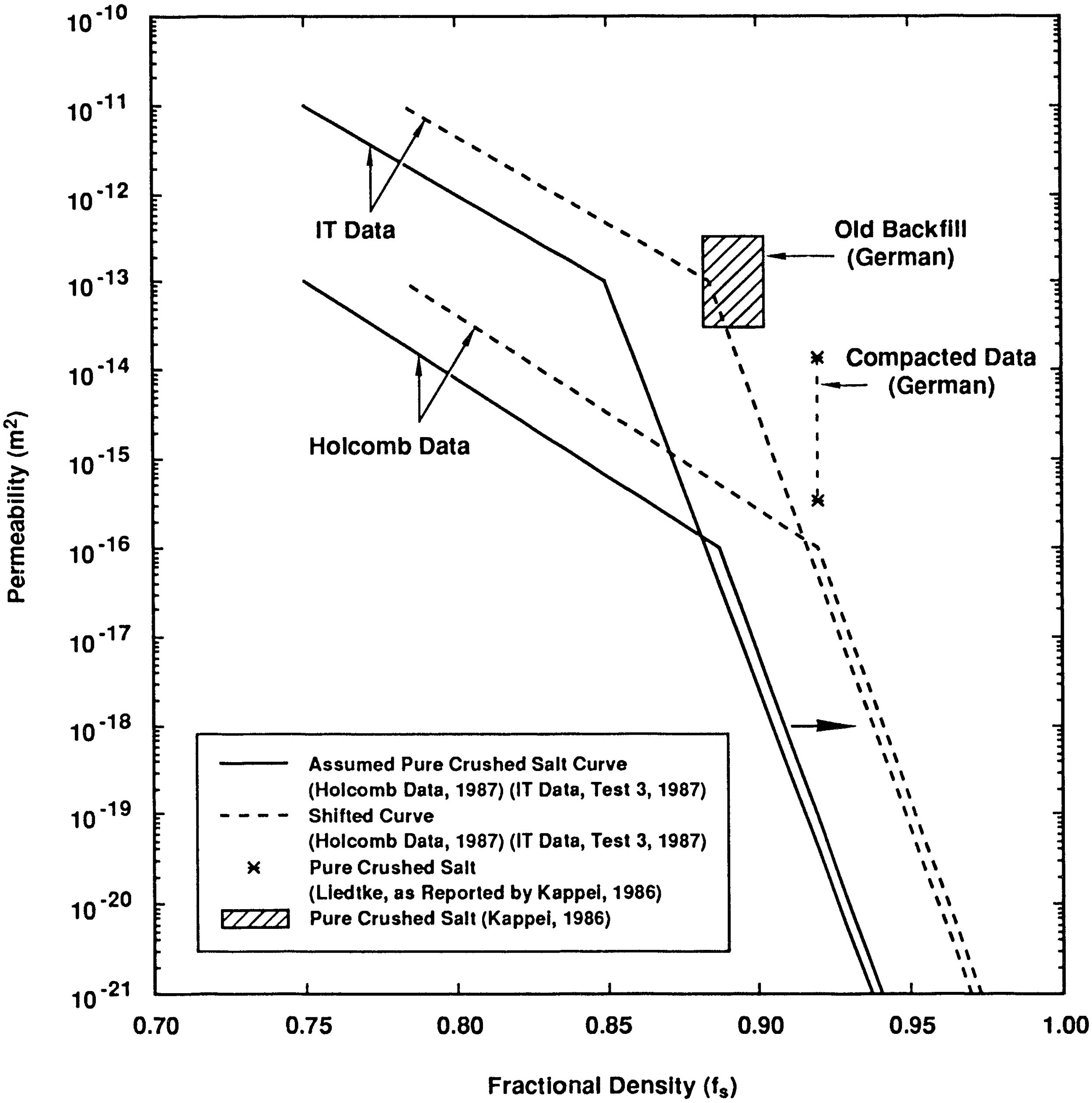

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Figure 4-3. The method used to adjust the salt permeability curves to include the German "old backfill" data.

The permeability criterion can be used to determine the merit of different backfills by (1) estimating the time to reach the required state of low permeability, and (2) estimating whether adverse factors, such as the flow of large amounts of brine into the repository, have time to occur prior to consolidation to an acceptable permeability. Thus, backfill acceptability has been evaluated here by estimating the time required to achieve two permeability values, 10^{-18} m^2, the upper bound of permeability considered acceptable for the backfill, and 10^{-19} m^2, which would be more desirable. Even lower permeabilities may be physically achievable for a particular backfill, but the value of 10^{-19} m^2 is considered sufficient to eliminate substantial flow of brine through backfill as a short circuit for the migration of soluble radionuclides in response to human intrusion.

4.1.5 EVALUATION OF A DISPOSAL ROOM CONTAINING PURE CRUSHED SALT

Estimated times for pure crushed-salt backfill to achieve the required low permeabilities have been determined from the results shown in Figure 4-2 to be from 19 to 26 years for 10^{-18} m^2 permeability and 21 to 30 years for 10^{-19} m^2 permeability. These results are listed in Tables 4-1 and 4-2. The longer times of 26 and 30 years in the ranges quoted in Tables 4-1 and 4-2 correspond to the arbitrary shift in permeability values that was used to force agreement with the German data, described previously. The shorter times correspond to direct use of the WIPP data without adjustment. The difference between the two sets of values is not considered enough to influence backfill selection.

4.1.6 EVALUATION OF A DISPOSAL ROOM CONTAINING BOTH TRU WASTE AND PURE CRUSHED SALT

Estimated backfill permeabilities as a function of time for a room completely filled with both waste and crushed-salt backfill are shown in Figure 4-4. This curve was obtained by cross-plotting the consolidation curve for the backfill, from the SANCHO results in Figure 3-5, with the permeability results shown in Figure 4-1. Predicted times for backfill consolidation for this configuration are from 18 to 29 years for 10^{-18} m^2 permeability (Table 4-1) and 21 to 35 years for 10^{-19} m^2 permeability (Table 4-2). These times are almost identical to predicted times for consolidation of a room entirely filled with backfill, even though waste was included in these calculations.

4.1.7 EVALUATION OF A DISPOSAL ROOM COMPLETELY FILLED WITH SALT/BENTONITE BACKFILL

The estimated permeability as a function of time for a room completely filled with 30% salt/70% bentonite backfill is shown in Figure 4-5. This curve was obtained by cross-plotting the curve for SANCHO in Figure 3-3 with the permeability results shown in Figure 4-1. Unlike the results for crushed salt,

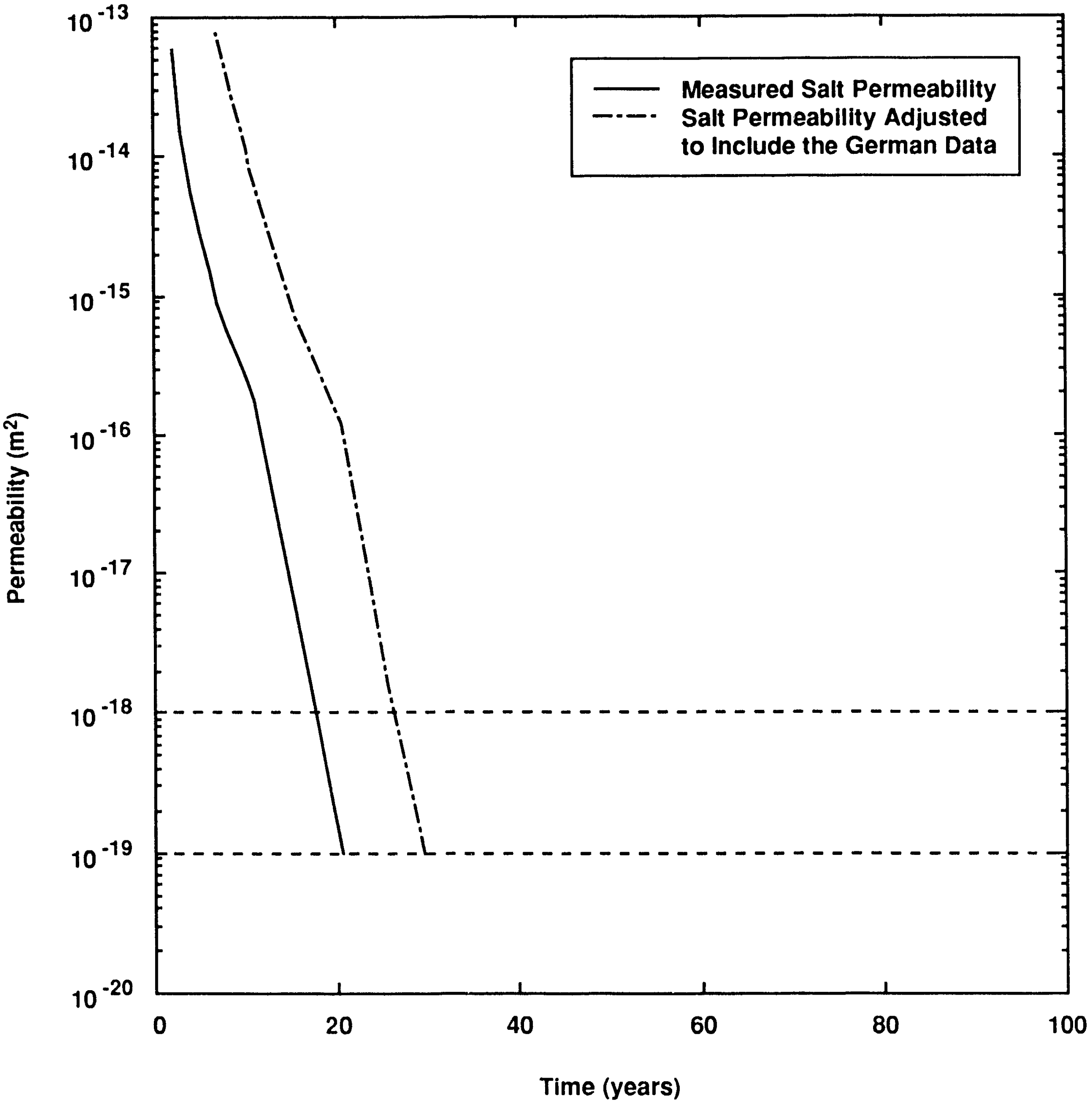

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Figure 4-4. Disposal room closure permeabilities for rooms filled with TRU waste and pure crushed-salt backfill. The portions of the curves below 10^{-19} m^2 are not shown because the predictions at such low permeabilities are considered inaccurate.

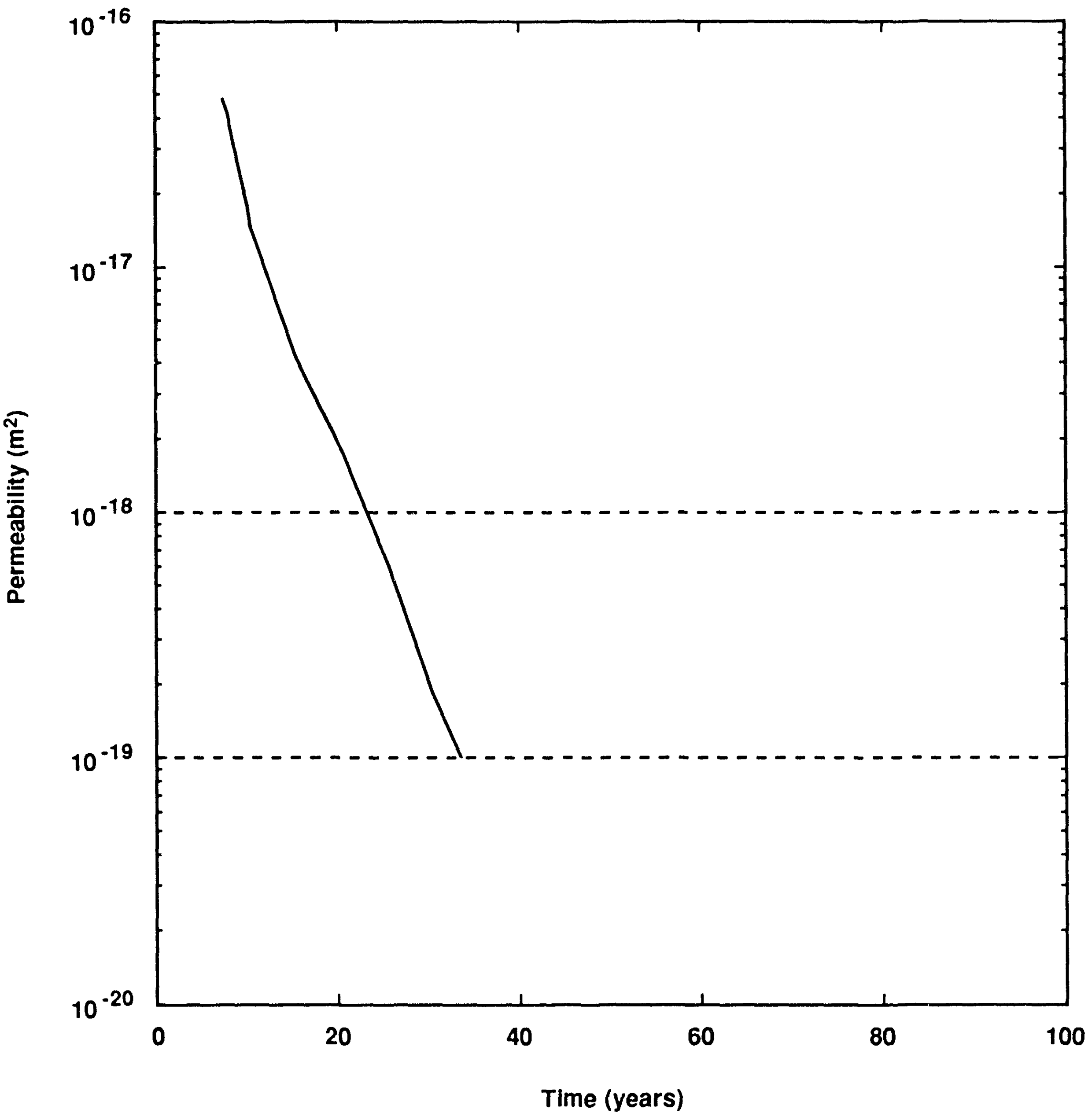

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Figure 4-5. Disposal room closure permeabilities for rooms filled with 70% salt/30% bentonite backfill. The portions of the curves below 10^{-19} m^2 are not shown because the predictions at such low permeabilities are considered inaccurate.

TABLE 4-1. BACKFILL CHARACTERISTICS AT 10^{-18} M^2 PERMEABILITY

	Pure-Salt Backfill	Salt/ Bentonite Backfill	Salt Backfill and Waste	Salt/Bentonite Backfill and Waste
Closure time corresponding to a permeability of 10^{-18} m^2	19-26 years	23 years	18-29 years	44 years
Void fraction corresponding to a permeability of 10^{-18} m^2	0.093-0.058	0.140	0.093-0.058	0.140
Related Parameters				
Backfill void volume*	245-147 m^3	364 m^3	85-51 m^3	127 m^3
Saturation (43 m^3 brine in 100 years)	3%, 8%	3%	9%, 24%	15%
Saturation (43 m^3 brine in 100 years +) 10% water in as-received bentonite		45%	9%, 24%	57%
Bentonite-saturated density**	-	2112 kg/m^3	-	2112 kg/m^3
Bentonite swell pressure	-	1.9-3.0 MPa	- 1.9-3.0 MPa	
Lower bound volume of chemically bound water	0	32 m^3	0	11 m^3
Upper bound volume of chemically bound water	0	149 m^3	0	51 m^3
Shear Strength	>1.9 MPa	0.9 MPa	>1.9 MPa	0.9 MPa

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* The total room volume is assumed to be 3646 m^3; backfill volume in a room filled with waste and backfill is 1268 m^3; the volume of the waste is 1817 m^3; and the emplacement density of the backfill is 1400 kg/m^3 for both types of backfill.

** The saturated density of bentonite, ρ_{sat}, for dry density ρ, is: $\rho_{sat} = 1200 + (1 - 1200/2700)\rho$, assuming 1200 kg/m^3 density for WIPP brine and 2700 kg/m^3 for the theoretical solid density of bentonite.

TABLE 4-2. BACKFILL CHARACTERISTICS AT 10^{-19} M^2 PERMEABILITY

	Pure-Salt Backfill	Salt/ Bentonite Backfill	Salt Backfill and Waste	Salt/Bentonite Backfill and Waste
Closure time corresponding to a permeability of 10^{-19} m^2	21-30 years	34 years	21-35 years	64 years
Void fraction corresponding to a permeability of 10^{-19} m^2	0.081-0.048	0.127	0.081-0.048	0.127
Backfill void volume*	210-120 m^3	325 m^3	73-42 m^3	113 m^3
Related Parameters				
Saturation (43 m^3 brine in 100 years)	4%-11%	3%	12%-36%	24%
Saturation (43 m^3 brine in 100 years +) 10% water in as-received bentonite		52%	9%, 24%	70%
Bentonite-saturated density**	-	2151 kg/m^3	-	2151 kg/m^3
Bentonite swell pressure	-	2.3-3.5 MPa	-	2.3-3.5 MPa
Lower bound volume of chemically bound water	0	28 m^3	0	10 m^3
Upper bound volume of chemically bound water	0	130 m^3	0	47 m^3
Shear Strength	>2.2 MPa	1.6 MPa	>2.2 MPa	1.6 MPa

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* The total room volume is assumed to be 3646 m^3; backfill volume in a room filled with waste and backfill is 1268 m^3; the volume of the waste is 1817 m^3; and the emplacement density of the backfill is 1400 kg/m^3 for both types of backfill.

** The saturated density of bentonite, ρ_{sat}, for dry density ρ, is: $\rho_{sat} = 1200 + (1 - 1200/2700)\rho$, assuming 1200 kg/m^3 density for WIPP brine and 2700 kg/m^3 for the theoretical solid density of bentonite.

a second curve, showing the effect of adjusting permeabilities to compensate for differences in data from different sources, was not considered because of data consistency and the fact that the permeability measurements were made with brine on fully saturated samples. Therefore, the calculated times should be upper bounds. The estimated times of 23 and 34 years to achieve the required low permeabilities, listed in Tables 4-1 and 4-2, are almost the same as the estimated times for closure of rooms containing crushed salt. The reason for the similarity is that although compaction of salt/bentonite is slower, it requires less consolidation to reach the required permeability, a factor which compensates for the much slower rate of consolidation of this material.

4.1.8 EVALUATION OF A DISPOSAL ROOM CONTAINING BOTH TRU WASTE AND SALT/BENTONITE BACKFILL

The computed permeability as a function of time for a room filled with TRU waste and salt/bentonite backfill is shown in Figure 4-6. This curve was obtained by cross-plotting the curve for SANCHO in Figure 3-7 with the permeability results shown in Figure 4-1. The estimated times of 44 and 64 years to achieve the required low permeabilities, listed in Table 4-1, are within a factor of two of the estimated times for closure of rooms backfilled with salt/bentonite (23 and 34 years).

4.1.9 POROSITIES

The second part of the permeability criterion for backfill evaluation is that the void volume remains unsaturated with brine. The void volume of the backfill at a given state of compaction determines how much brine would have to flow into that region of the backfill to saturate it and impede consolidation. The porosity of the backfill at a given time is also important with regard to the spread or transport of soluble radionuclides. In this case, smaller void volumes lead to smaller volumes of radioactive brine as the repository saturates.

The significance of the porosities of the two backfills, when the required permeabilities of 10^{-18} m^2 and 10^{-19} m^2 are achieved, is demonstrated by the backfill void volume at these times. Interpretation of results is facilitated by the fact that backfill porosity results for a room with waste will be the same as for a room completely filled with backfill because the permeability of the backfill is assumed to depend only on the void fraction of the backfill at a given time (Tables 4-1 and 4-2). On the other hand, there is approximately three times as much backfill in a room completely filled with backfill as in a room containing waste plus backfill, so that the void volume associated with a completely filled room will be greater. The results in Table 4-1, show that when a room completely backfilled with crushed salt reaches a permeability of 10^{-18} m^3, its void volume is estimated to be between 147 and 245 m^3. If as

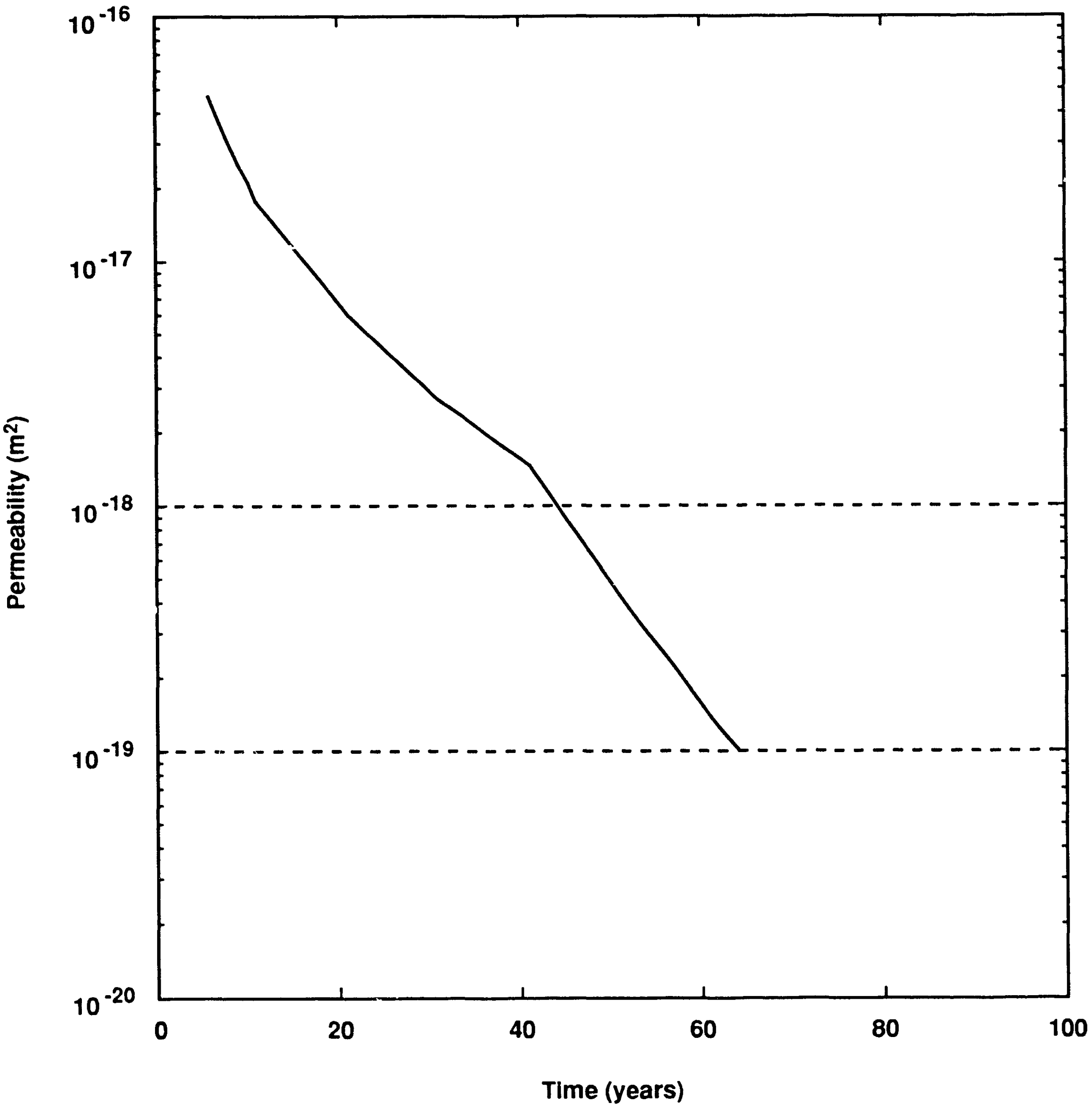

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Figure 4-6. Disposal room permeabilities for rooms filled with TRU waste and 70% salt/30% bentonite backfill. The portions of the curves below 10^{-19} m^2 are not shown because the predictions at such low permeabilities are considered inaccurate.

much as 147 m^3 of brine enters the backfill within the prescribed time, then additional consolidation of any significance is unlikely. The amount of brine potentially available for the solution of radionuclides is equal to the brine volume causing saturation.

Observations about the effect of various brine inflow rates on consolidation are also possible. If the early estimate of 43 m^3 brine inflow into a room over 100 years is accepted, and all of this brine went into salt backfill, then, using simple ratios, the degree of saturation (brine volume/void volume) of the backfill for a void volume of 147 m^3 would be approximately 8% after the 26 years required for closure. Further, the small degrees of saturation observed for all of the other void volumes given in Table 4-1 suggest that if the early brine-inflow estimate is correct or an upper bound, consolidation of neither of the backfills under consideration is likely to be retarded because of brine saturation, even if all available brine goes into the backfill porosity. In fact, consolidation retardation by brine saturation would be even less likely than estimated above, either because some of the brine entering the disposal areas may be used up by corrosion of iron in the waste or because brine is fixed by bentonite added to the backfill.

Saturation estimates for salt/bentonite are also given in Table 4-1 and assume all of the inflowing brine went into the backfill. Unlike salt, however, bentonite may contain, according to supplier specifications, an upper bound of 10% moisture in the as-received condition. This moisture could occupy as much as 153 m^3 of water in a room completely filled with bentonite, or 53 m^3 in a room filled with backfill and waste. When these volumes are included in the saturation estimates, the percent saturations increase to as much as 70% (Table 4-2). Thus, the salt/bentonite backfill does not saturate within the time required to consolidate to a state with the prescribed permeability. In reality, however, the percent saturation of the backfill will be even less than predicted because the actual as-received water content for bentonite stored at the WIPP has been less than 6%; this content can be ensured by specifying the allowable moisture content of the bentonite at the time of emplacement.

4.1.10 EFFECT OF BENTONITE SWELLING ON SALT/BENTONITE CONSOLIDATION

Saturated densities of the bentonite corresponding to the required permeabilities of 10^{-18} m^2 and 10^{-19} m^2 are also summarized in Table 4-1. These values were used to determine swelling pressures from the experimental results of Pfeifle and Brodsky (1990), shown in Figure 4-7. Since the experimental results are given in terms of the dry density of the compacted bentonite, dry densities were computed by dividing the weight of bentonite in a given volume, V, of backfill by the difference ($V - V_{ss}$), where V_{ss} is the theoretical solid

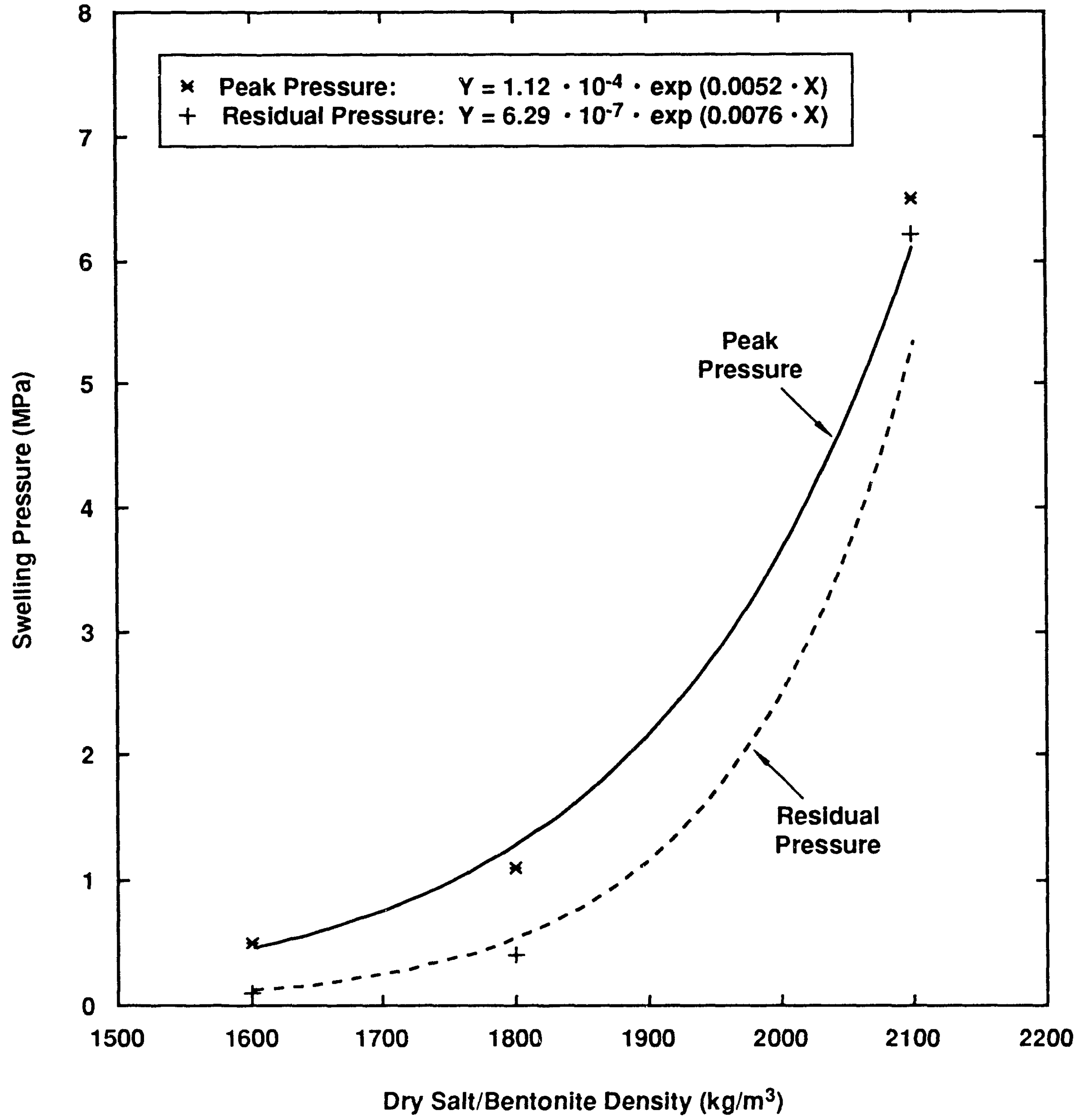

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Figure 4-7. Swelling pressures as functions of dry densities for 70% salt/30% bentonite mixtures (Pfeifle and Brodsky, 1990).

volume of the salt. Saturated densities of bentonite are obtained by assuming that all voids are completely filled with brine (Table 4-2).

Two curves for the swelling pressures corresponding to the various degrees of compaction of the salt/bentonite backfill are shown in Figure 4-7. One curve represents the peak pressures observed in samples at various densities; the other curve represents the average values of the residual pressures that eventually occurred in the tests. Dry densities were used because the data were reported in this manner (see Table 4-3 for the equivalent values of saturated densities corresponding to the data). The peak pressures for both the 1600 kg/m^3 and 1800 kg/m^3 samples were observed almost immediately after brine was added to the samples. Once pressures peaked, the swelling pressures in these samples decayed to the residual values shown. In contrast, the swelling pressure history for the highest density samples (2100 kg/m^3) showed no relaxation, even after 120 days, and it is interesting to speculate whether or not decay would have ever occurred had the tests been conducted longer. In the absence of this information, the analytic fit to the residual curve in Figure 4-7 was constructed assuming no decay at 2100 kg/m^3 and is therefore an upper bound of the residual pressures.

Swelling-pressure results were also compared with information published about bentonite. As shown in Figure 4-8, swelling pressures for salt/bentonite in the WIPP brine differ from the results for pure bentonite in salt water reported by Pusch (1980). The source of this discrepancy is thought to be the high cation content of the WIPP brine, which is addressed in Section 4.1.11.

Swelling pressures for the four different cases given in Tables 4-1 and 4-2 were computed from the two analytic curves shown in Figure 4-7 to predict their effect on backfill consolidation. As a basis for comparison, stresses of 3 to 4 MPa are considered to represent the pressure threshold above which backstress has been observed, in past calculations, to impede the calculated creep closure of the disposal rooms. Since none of the pressures quoted in Tables 4-1 and 4-2 exceeds this threshold, we conclude that neglect of swelling pressure did not cause a serious error in the current closure predictions for salt/bentonite backfill. Therefore, neglect of swelling in closure analyses is reasonable.

4.1.11 PERMEABILITY SUMMARY

Test results (Section 4.1) show that both backfills are predicted to consolidate to a sufficiently impermeable state within a time period well under 100 years. For 10^{-19} m^2 permeability, the longest estimated consolidation time for a room containing TRU waste and pure crushed-salt backfill is 35 years; the

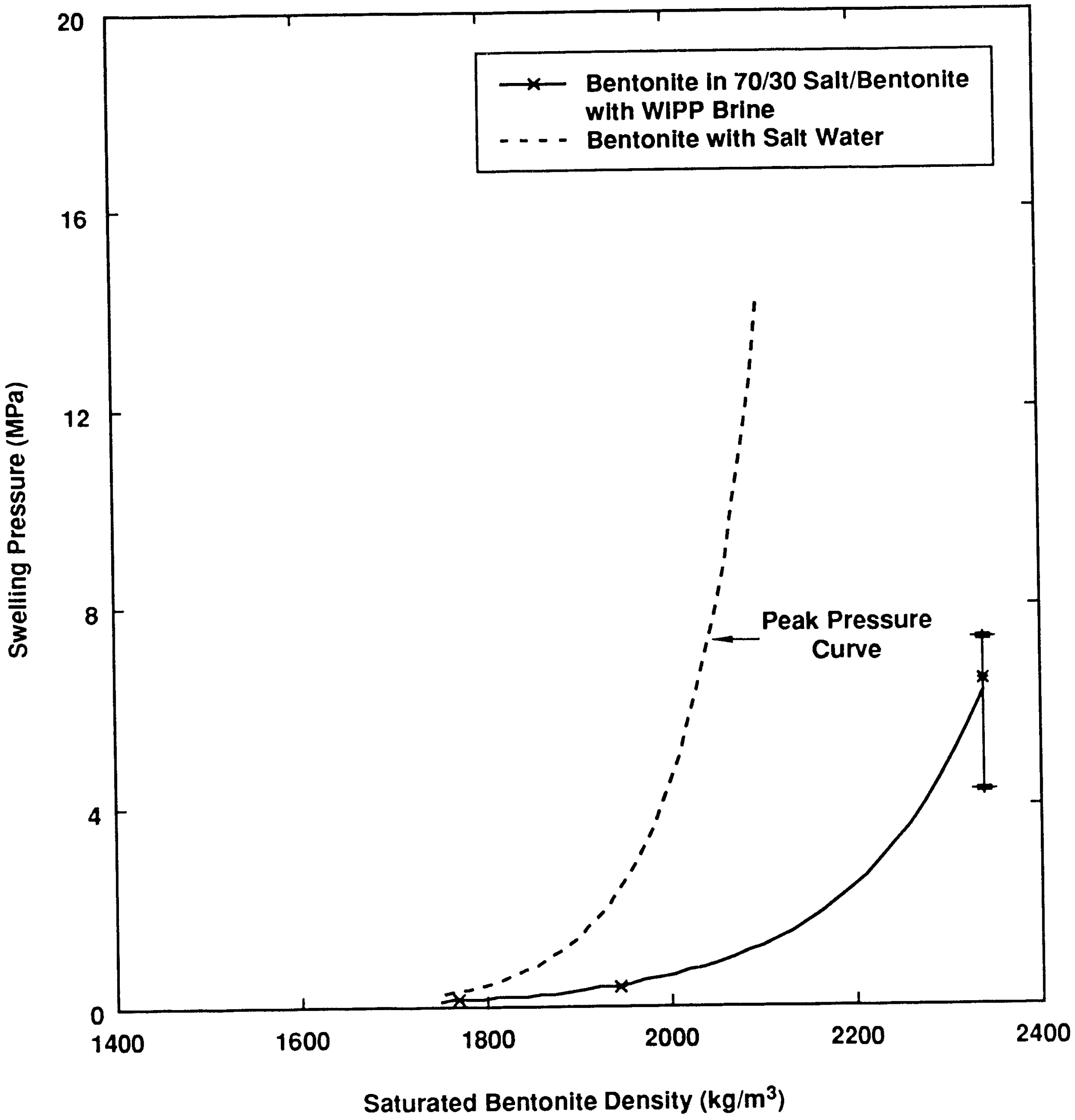

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Figure 4-8. A comparison of the swelling pressures observed in salt/bentonite mixtures in WIPP brine (Pfeifle and Brodsky, 1990) with Pusch's results on bentonite in brine (Pusch and Karnland, 1986).

longest estimated consolidation time for a room containing TRU waste and salt/bentonite backfill is 64 years. Further discussion in subsequent sections establishes that, according to present estimates of brine inflow, these times are not long enough to permit significant amounts of brine to enter the repository. Gas generation over such short times is also not expected to impede backfill consolidation sufficiently (Weatherby, as interpreted by Mendenhall et al., 1990). Finally, back pressure caused by swelling of bentonite is estimated to be small enough to neglect in closure predictions for salt/bentonite backfill.

4.2 Chemical/Mineralogical Stability

In terms of chemical and physical stability, pure crushed salt is the best material that could be used for backfilling WIPP disposal areas because it has the same physical properties as the surrounding salt. Bentonite, in the salt/bentonite mixture, is also considered to be stable in most environments and is therefore widely used for this reason in the construction industry for the erection of low permeability barriers and in other applications that depend on its chemical sorption properties. Because WIPP must be designed to perform satisfactorily over very long periods of time, there are some aspects of the stability of bentonite that would not normally be of concern.

The bentonite recommended for backfilling WIPP disposal rooms must be chemically and mineralogically similar to MX-80 Granular Volclay bentonite produced by the American Colloid Company. Additional information about MX-80 bentonite and reasons for its selection are given by Molecke (1986) and Pfeifle (1987b), and it has been used for almost all of the research supporting this investigation. Its average chemical composition is $(NaCa)_{.35}\ (Al_{1.60}Fe_{.15}Mg_{.25})\ (Si_{3.90}Al_{.10})\ O_{10}\ (OH)_2$, and it has 55-65 milli-equivalents/100 g exchangeable sodium ion content, 15-25 milli-equivalents/100 g exchangeable calcium ion content, and 10-15 milli-equivalents/100 g exchangeable magnesium ion content (Pfeifle, 1987b)[2].

An independent review of the long-term stability of bentonite in the WIPP disposal rooms has recently been completed by Jercinovic (Appendix A.1). The following areas of concern were identified.

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[2] Data provided by the American Colloid Company, Skokie, IL.

(1) MX-80 is considered an aluminous smectite clay, and as such can be expected to gradually convert to illite when exposed to WIPP brine for extended periods of time. Illite will be favored, rather than chlorite, because potassium will tend to be preferentially selected over other available adions onto clay surfaces. The development of illite will be a function of the temperature and reaction time, and it can be expected to parallel illitization reactions observed in natural hydrothermal systems and potassium bentonites.

The consequences of illite formation are that it has a much lower cation exchange capacity than smectite, and therefore, the amount of smectite converted to illite or interlayered illite/smectite will alter the long-term hydrodynamic and sorption properties of the backfill. Initial swelling of the bentonite will generate a maximum pressure, which should eventually decrease in response to illitization. Radionuclides initially sorbed onto bentonite may exhibit some desorption as recrystallization to illite serve to lower the overall surface sorption potential. Illite envelopment over long periods of time will alter the sorption and sealing properties of the backfill, making the material a less effective barrier overall.

(2) Bentonite may consume magnesium from the brine by the precipitation of magnesium chloride or reconstruction of montmorillonite to magnesium saponite or chlorite. A concern has been raised in the past that hydrated magnesium chloride can then decompose to produce hydrochloric acid. In turn, an acidic brine would react with the waste to generate hydrogen. Experimental results by Krumhansl (1986) show that hydrated $MgCl_2$ can decompose <u>above</u> 100°C to produce a solution with pH <7. Krumhansl's results suggest, however, that this reaction will not occur in the WIPP, because temperatures will never reach 100°C, and water vapor pressures will never be low enough to reduce $MgCl_2$ to the anhydride form.

(3) The potential effects of the release of organic compounds as a result of complete dissolution of the waste package are difficult to assess quantitatively, but may be significant. Organic compounds may play a role in both sorption (as organics sorb onto clay surfaces) and in mobilizing some elements (e.g., chelation).

The effect of these stability issues on backfill performance is considered important, but not so serious as to discredit bentonite as a candidate backfill material. While transformation of smectite clay to illite may reduce sorption potential, the increase in permeability, because of the illite, is expected to be less than a factor of 100 (Figure 2-1, Meyer and Howard, 1983) for the worst possible case of saturated bentonite reverting to saturated illite with no change in the total volume of the clay. Considering the permeability increase first, because this condition is of greatest concern, the closure calculations for evaluation of salt/bentonite predict (1) that the backfill can achieve permeabilities lower than 10^{-19} m^2, although consolidation will take a little longer, and (2) that the backfill would still be far from saturated at 10^{-19} m^2. These observations suggest that even complete transformation of bentonite to illite over long periods of time would still make the mixture acceptable as a backfill.

Reduction of sorption potential by either transformation to illite or release of organics is less critical, because a basic assumption of the evaluation described in this report has been that sorptive potential has to be considered a useful, but not necessary requirement for disposal room backfill. Part of the reason for not insisting on a quantitative criterion for sorptive potential is because the chemistry of the waste in its unprocessed state is too complex to expect predictions to have quantitative significance. In other words, the difference between pure crushed salt and salt/bentonite as a backfill material, on the basis of chemical stability, is reduced to (1) consideration of pure crushed salt with no sorptive potential that satisfies all other backfill selection criteria, or (2) consideration of salt/bentonite with some sorptive potential (albeit unknown) that also satisfies all other backfill selection criteria.

4.3 Shear Strength

The shear strength of the backfill at rapid loading rates is important because it is a measure of how much erosion of a borehole can occur during a human intrusion into repository by drilling. While erosion of the waste is really the issue impacting compliance with the regulatory standards, the backfill adjacent to the waste should be sufficiently competent to prevent any unstable borehole enlargement. Shear strengths of salt/bentonite were calculated from unconfined compressive strengths measured at various degrees of consolidation after permeability testing by loading the unconfined samples to failure at a constant displacement rate of 0.002 mm/s (Pfeifle, 1990). The samples were fully saturated. The results of these tests are shown in Figure 4-9. For the permeability conditions of 10^{-18} m^2 and 10^{-19} m^2, the salt/bentonite shearing strengths of 0.9 MPa and 1.6 MPa (one half the failure stress in unconfined compression) quoted in Table 4-1 are many orders of magnitude above the 6 Pa critical shear strength for borehole erosion estimated in Lappin et al. (1989, p 5-9). An increase of the mean pressure on the samples during the test to make conditions more representative of the consolidated state of the repository would increase these salt/bentonite strengths even further.

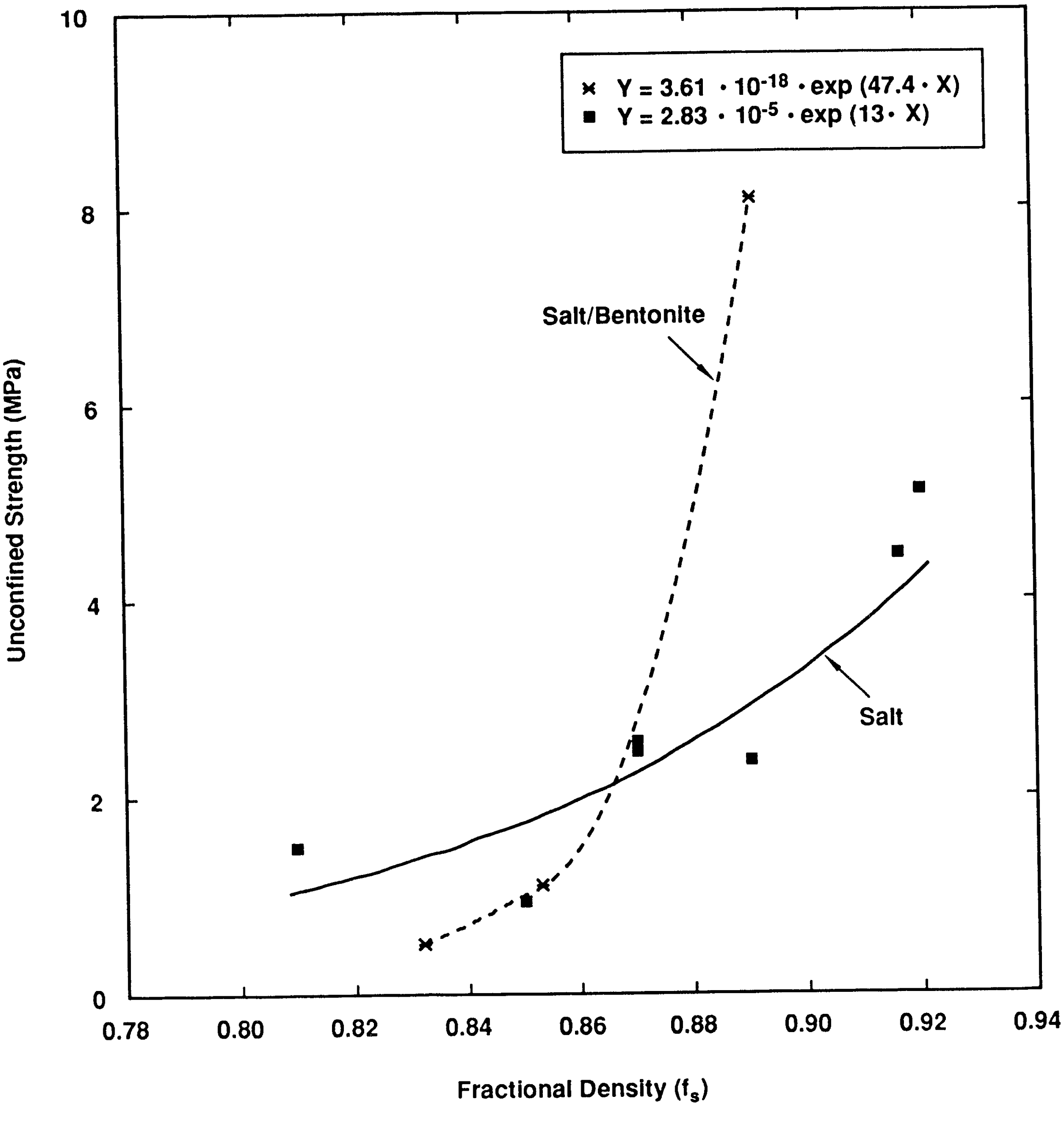

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Figure 4-9. The strength of crushed salt (Pfeifle, 1987a) and brine-saturated and 70% salt/30% bentonite mixtures (Pfeifle, 1990) under rapid loading conditions.

Similar results for rapid consolidation of crushed salt in the literature (Pfeifle, 1987a) suggest even higher shear strengths, although the tests reported by Pfeifle did not include saturated samples. The shear strengths for pure crushed salt given in Table 4-3 are actually larger than the salt/bentonite backfill at a comparable permeability because the pure-salt backfill has to consolidate to a lower porosity to satisfy the permeability criteria.

Given the magnitudes of the shear strengths determined for both backfills, a conclusion based on the erosion analysis assumed in Lappin et al. (1989) is that the in-place strengths of both backfills should be sufficient to remove the possibility of any excessive enlargement of the borehole during a human intrusion by drilling (at least 100 years after institutional control of WIPP ceases). Further, since the strengths of the two materials are roughly comparable, there is no clear advantage or disadvantage of crushed salt over salt/bentonite as a backfill.

4.4 Ease of Emplacement

Emplacement of either crushed-salt or salt/bentonite backfill is anticipated to be straightforward. Both backfills have been used in large quantities in the WIPP underground experimental rooms T and J (Pfeifle, 1987b; Molecke, 1986), and similar demonstrations have been performed by the Germans in the Asse Repository. Emplacement is facilitated by the wide range of mechanical and pneumatic methods that have been developed for backfilling various types of mines by the mining industry. Salt/bentonite backfill costs more, but it is doubtful that the additional cost incurred by its use would override its more advantageous sorption performance as a backfill.

Initial backfill emplacement densities of salt without vibratory or other mechanical means of densification are between 1300 kg/m^3 and 1400 kg/m^3 (Pfeifle, 1987b). Because closure is so rapid, closure calculations are not sensitive to the exact value used. Other requirements for the backfill are that it should have approximately the particle-size distribution of the mine-run crushed salt indicated by Pfeifle (1987b), with all particles larger than about 12.5 mm removed and recrushed. In addition, as-mined salt is considered to have sufficient moisture to avoid the need for the addition of brine to facilitate consolidation, as long as it is not stored above ground long enough to dry out. A water content of 0.5% or greater, but certainly no more than several percent, is considered desirable. The bentonite used for backfill should be as similar as possible in composition and form to the MX-80 Granular Volclay bentonite produced by the American Colloid Company (Molecke, 1986). (MX-80 bentonite was specified for all research supporting this investigation.) Finally, the bentonite should be well-mixed with the salt and contain less than 10% moisture when emplaced.

TABLE 4-3. CONSOLIDATED DENSITIES OF SALT/BENTONITE MIXTURES

Dry Density of Mixture kg/m^3	Saturated Density of Mixture kg/m^3	Dry Density of Bentonite in Mixture kg/m^3	Saturated Density of Bentonite in Mixture kg/m^3
1600*	1958	1007	1758
1800*	2053	1313	1928
2100*	2195	2012	2316
1963**	2130	1645	2112
1992**	2144	1717	2151

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* Corresponds to swelling pressure tests

** Rooms completely filled with salt/bentonite at permeabilities of 10^{-18} m^2 and 10^{-19} m^2.

5.0 PERFORMANCE ASSESSMENT: DESIRABLE CRITERIA

5.1 Effect of Additives

No formal selection of any backfill additives other than bentonite has been completed prior to the date of this report. However, incorporation of the effect of low-strength additives similar to bentonite into the closure analyses is not expected to be difficult, because these materials do not resist the consolidation process. Incompressible additives are also not expected to present difficulties because the salt will flow around them and decrease void volume. Experimental consolidation data are needed to support both of these hypotheses.

5.2 Brine Sorption

5.2.1 WATER SORPTION BY SALT

The sorption or release of water by sodium chloride has been a topic of considerable research in the past because caking of salt in cartons, bags, or bulk is undesirable (Kaufmann, 1960, p. 531). The ability of an impure sodium chloride to either deliquesce or cake is related to its critical humidity, which for WIPP salt is approximately 70% at atmospheric pressure (Brush, 1990, p. 33) and is almost insensitive to changes in temperature (Kaufmann, 1960). The critical humidity is defined as the value of the humidity when the water vapor surrounding the salt is in equilibrium with water in liquid form. For example, consider a region in the backfill where the air is stagnant. Eventually the humidity within this space will rise (or fall) to the critical humidity if sufficient liquid water is available.

If the humidity of the air space is less than the critical humidity, water will evaporate from brine within the salt and cause dissolved solids to come out of solution; if the humidity of the space is greater than the critical humidity, water will condense and solid salt will go into solution. In the extreme, acceptance of water by the salt when the humidity of the air is always greater than the critical humidity will continue until all of the solid salt is in solution. Solution or precipitation of salt will occur mainly at contact points between grains of salt because capillary forces lead to the concentration of excess water in these regions.

Though salt is frequently referred to as hydroscopic, control of the water content of salt in the manner described in the previous paragraph is not considered sorption in the sense of the chemical affinity for water that bentonite has. The reason for this conclusion is that, in contrast to a sorptive material such as bentonite, which eventually reaches various equilibrium moisture states depending on the humidity, any constant humidity condition in crushed salt that differs from the critical humidity will cause a continual change in moisture content. Eventually, almost all of the liquid water will be removed from the salt, or all the solid salt will be dissolved. In other words, in an open system the amount of water retained in the salt at a given time does not depend uniquely upon the current humidity.

Applying these concepts to salt backfill within a disposal room, if the air space is ventilated, as, for example, before and during emplacement, then some of the moisture will be drawn out of the salt. It is unlikely, however, that the moisture of the salt in a drying environment will ever drop much below 0.5% by weight because of capillary forces at grain contacts and imperfect circulation of air through the backfill.

Once emplacement of the backfill is completed and the disposal room is sealed, the humidity of the voids should rapidly return to the critical value. The amount of liquid water required to increase the humidity is slight and can come from brine in salt surrounding the disposal rooms via cracks in the disturbed rock zone (DRZ), as well as from water in the backfill. These considerations indicate, however, that crushed salt in backfill has no potential for chemical sorption of water and will simply provide storage capacity for excess brine within the disposal rooms.

5.2.2 WATER SORPTION IN SALT/BENTONITE BACKFILL

Loose granulated bentonite is characterized as randomly oriented and roughly equi-dimensioned fragments of natural clay bed material. The size of the fragments corresponds to silt and fine sand fractions, and they are aggregates of flakey particles composed mostly of montmorillonite. The montmorillonite lamellae are approximately parallel to each other (Pusch and Karnland, 1986) in a manner similar to the structure of graphite. When water is added to bentonite, part of it, the "internal" water, is chemically bound between the lamellae of the particles, separating each adjacent crystal surface by as many as three layers of water molecules (Pusch and Karnland, 1986). Up to 0.357 g H_2O/g clay sorption is possible in this manner, according to one lattice configuration model (Pusch and Karnland, 1986).

Additional "external" water is present in voids between the clay fragments (Pusch and Karnland, 1986). Unlike internal water, external water is unaffected by the minerals. Upon wetting, water is drawn into the (interconnected) voids by capillary action as external water, and then some is redistributed to become internal water. The amount of internal, chemically bound water depends on how much of the microstructure is closely oriented and lamellinar in nature (Pusch and Karnland, 1986). Particle reorientation is accomplished by applying pressure; the degree of microstructural order is dependent upon the state of consolidation.

The balance between internal and external water can be illustrated by some examples that bound the range of bentonite densities expected for backfill applications. The lowest density bound of fully saturated pure bentonite of interest is about 1600 kg/m^3, which is calculated to be equivalent to a dry uncompacted emplacement density of about 950 kg/m^3 (assuming pure water is the saturating fluid). The water content[3] of the saturated density is 0.67, with 40-60% of the water bound chemically.[4] A more ordered structure exists at a greater consolidation. For a saturated density of 2300 kg/m^3 (water content of about 0.11), the chemically bound water is as much as 80-90% of the total water content (Figure 4, Pusch and Karnland, 1986), and for a saturated density of 2100 kg/m^3 (Table 4-2), the approximate density that we are interested in, the chemically bound water is 70-85% of the total water content. Therefore, an upper bound of the water in WIPP brine that can be sorbed by the bentonite is obtained by assuming that chemical sorption of water from WIPP brine is 70% of the total water content. These estimates are listed in Table 4-1.

It is not known, however, if the internal, chemically sorbed, water content is preserved at high densities for WIPP brine, which has an exceptionally high impurity content. A basic concern is that impurities, such as magnesium, can inhibit chemical sorption by altering the nature and number of available reactive sites. Evidence of such effects has already been suggested by the recently observed decrease in swelling pressures (Section 4.1.10) observed for bentonite saturated with WIPP brine (Pfeifle and Brodsky, 1990) in comparison to those for bentonite saturated with seawater (Pusch and Karnland, 1986). In view of this new development, the extent of chemical sorption of water from WIPP brine will be determined. Instead, since swelling pressure is considered an indirect physical measure of sorption, the assumption is that a lower bound for the amount of chemically-bound water can be defined. This estimate is obtained from the ratio of the swelling pressure of about 3 MPa, associated with bentonite in WIPP brine at a fully saturated density of 2100 kg/m^3, to the swelling pressure of 15 MPa, which is associated with bentonite in pure water at the same saturated density. The ratio of 1:5 in swelling pressure implies that only about 15% of the total water content might be chemically bound when the source of the water is WIPP brine. While the linear scaling used in this estimate is overly simplistic, this lower bound to the amount of water that can be sorbed will be adopted until additional laboratory data become available to change it.

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[3] The water content is the weight of the water in the bentonite divided by the weight of the (dry) bentonite.

[4] The water content of this mixture is very much less than the water content of more fluid-like bentonite-based drilling "muds or gels." Bentonite drilling muds are highly viscous slurries composed of 16 parts water to 1 part by weight of bentonite (WC = 16), with densities of about 1040 kg/m^3, assuming that the density of water is 1000 kg/m^3.

Estimates of the amount of chemically sorbed water in salt/bentonite mixtures in this report assumed, therefore, that only 15% of the total saturated water content is chemically bound. Considering a room entirely filled with salt/bentonite backfill, the saturated densities of the salt/bentonite mixture for permeabilities from 10^{-18} m^2 to 10^{-19} m^2 are 2130 to 2144 kg/m^3 (Tables 4-2 and 4-3). These densities correspond to dry densities from 1963 kg/m^3 to 1992 kg/m^3 for an initial emplacement density of 1400 kg/m^3. Assuming, in addition, that all of the void volume is filled with brine (density 1200 kg/m^3), that water represents 0.6877 of the brine by weight, and that only 15% of the water is chemically bound, the amount of chemically bound water per disposal room is computed to be about 30 m^3 for rooms filled with salt/bentonite backfill (32 m^3 for 10^{-18} permeability, 28 m^3 for 10^{-19} m^2 permeability, Tables 4-1 and 4-2).[5] For rooms containing waste, the chemically bound water is about 10 m^3. A problem with interpretation of these lower bounds is that water, initially in the bentonite upon emplacement, could exceed these rather small sorption capabilities if it is not controlled.

Water sorption estimates for bentonite in a room containing waste can therefore be interpreted several ways, depending on whether the upper or lower bound estimates are used and what is assumed about the initial water content of the bentonite. If (1) the upper bound estimates are assumed and (2) the initial water content of the bentonite is included (53 m^3 in a room filled with backfill and waste), then the sorption potential (about 50 m^3, Tables 4-1 and 4-2) is just sufficient to take care of the initial water content. Any additional brine entering the backfill will be present as external water. If, on the other hand, (3) the lower bound estimates are assumed, (4) initial water content is ignored, and (5) a brine inflow rate of 43 m^3 in 100 years all of which remains in the backfill is assumed, then approximately 50% of the brine inflowing into a waste-filled room or 10 m^3 is estimated to be chemically sorbable by the bentonite. In reality, a greater amount of inflowing brine is likely to be chemically bound, albeit loosely, than either estimate would suggest, but sorption of inflowing brine by bentonite cannot be considered an advantage of salt/bentonite as a backfill until additional data become available.

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[5] With ρ the dry density of the mixture, an initial mixture density of ρ_0 = 1400 kg/m^3 and a brine density of 1200 kg/m^3 1 m^3 of backfill has a brine volume of $(1400/\rho - 0.614)$.

5.3 Radionuclide Sorption

5.3.1 SORPTION OF SOLUBLE RADIONUCLIDES

The potential for sorption of soluble radionuclides is an important consideration in selection of a backfill. Sorption of radionuclides by the bentonite in salt/bentonite-backfill mixtures will be discussed in this section. The evaluation is based upon the assumption that a linear sorption model is appropriate (Novak, 1990), that the room contents are in equilibrium in the sense that brine is neither entering or leaving the room, and that the backfill is fully saturated. A quite different approach is needed for the analysis of a disposal room with brine flowing through it at a substantial rate.

The sorptive potential of bentonite will be evaluated using radionuclide distribution coefficients (K_d's) for bentonite estimated by Novak (1990). Novak's recommendations are contained in Appendix B. Assumptions for the application are: (1) that the concentration of solute in solution is 10^{-6} Molar (Lappin et al., 1989); (2) the radionuclide inventory of the repository given in Table 5-1 is representative of the repository; (3) the volume of 70% salt, 30% bentonite backfill in a typical disposal room is 1268 m^3 (Table 4.1, Butcher, 1990), its initial density is 1400 kg/m^3; and (4) colloid formation is unimportant.

The mass of the solute, M_s, is computed from:

$$M_s = K_d * Z * 10^{-6} * M_b,$$

where Z is the molecular weight of the radionuclide in kilograms, and M_b is the mass of bentonite.

The results, shown in Table 5-1, predict that if <u>brine containing dissolved radionuclides is distributed throughout all of the backfill (assuming 10^{-6} M solubility)</u>, sufficient bentonite is present to sorb almost all of the plutonium, americium, neptunium, or over 50% of the total inventory in the room. Practically none of the uranium would be sorbed. However, uniform distribution of radioactive brine throughout the consolidated backfill is unlikely, and therefore, not all of the bentonite would be in direct contact with radioactive brine. This observation is supported by the rapidity with which the salt/- bentonite backfill reaches an impermeable state, discussed in Section 4.1 of this report.

An alternative emplacement scheme might be considered to avoid (1) having to demonstrate that uniform radioactive fluid penetration of the backfill is realistic, or (2) having to defend an assumption about how much the estimated sorption should be reduced to account for nonuniformity of radioactive brine, which may be very difficult. In this scheme, spaces between the drums (about 320 m^3 per room) would be filled with pure bentonite at an initial density of 1400 kg/m^3. This volume would be in almost direct contact with the radioelements when the waste containers breach. A calculation shows that the volume of 100% bentonite between the drums would have about the same mass as if it were dispersed uniformly in the 70% salt, 30% bentonite mixture; therefore, bentonite would have about the same potential for sorption. With this option in mind, no reduction of the sorption amounts given in Table 5-1 is planned; in a static environment, bentonite has the potential of sorbing a significant amount of the radioelements in a disposal room.

Potential uncertainties arise, however, when bentonite sorption of radionuclides during flow is considered. As discussed by Novak (1990) and Lappin et al., Appendix D, (1989), estimated K_d values must be multiplied by a utilization factor when used in a porous-medium flow calculation. A factor of 0.001 was applied to account for dilution of the clay by crushed salt and the possibility that the flow will be channeled through localized regions. Localization can limit sorption to a very small portion of the bentonite actually present. While the effect of dilution has already been incorporated into the "static" analysis of this report, flow channeling cannot be discounted. For flowing brine, therefore, the sorptive potential of the bentonite in the backfill is considered greatly reduced.

5.3.2 BENTONITE SORPTION OF RADON

Radon gas is an important product of the radionuclide decay process. The potential of bentonite to sorb radon was not known at the time this report was prepared.

5.4 Deformability (ability to fill voids)

Both salt and bentonite deform with time and will intrude into voids or cracks to the point where the permeability of the backfill becomes very low.

5.5 Thermal Conductivity

No large amounts of heat are generated by CH TRU waste, and the thermal conductivity of salt is high enough to prevent the formation of localized hot spots within the repository. However, the thermal conductivity of salt is important with regard to the thermal loads imposed by RH waste, which is not considered in this report because the canisters will be emplaced in solid salt in the walls of the rooms, remote from the backfill.

TABLE 5-1. RADIONUCLIDE SORPTION BY BENTONITE

Radionuclide	K_d[1]	Repository Inventory[2] (Grams)	Computed Room Inventory[3] (Kilograms)	Mass Sorbed Per Room (Kilograms)	Percent of Room Inventory
Pu-238	0.1 m^3/kg	$2.28*10^5$	2.28	12.7	92%
Pu-239	0.1 m^3/kg	$6.84*10^5$	6.84		
Pu-240	0.1 m^3/kg	$4.61*10^5$	4.61		
U-233	0.001 m^3/kg	$8.15*10^5$	8.15	0.12	1.2%
U-234	0.001 m^3/kg	$1.71*10^5$	1.71		
Am-241	0.1 m^3/kg	$2.26*10^5$	2.26	12.85	100%
Np-237	0.01 m^3/kg	$1.14*10^4$	0.11	1.26	100%
			26.0		

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[1] Novak, 1990

[2] Table 4-2, Lappin et al., 1989

[3] Estimates for the number of equivalent room volumes in a repository range from 100 to 119. A value of 100 was used.

6.0 SUMMARY

6.1 Results

Two materials, pure crushed salt and 70% by weight salt/30% by weight bentonite were compared with regard to their potential for backfilling WIPP disposal rooms. A synopsis of this analysis are given in Table 6-1. Both materials were predicted to consolidate to states with permeabilities of less than 10^{-18} m^2 within times of the order of 50 years. The time to consolidate to a permeability of 10^{-19} m^2 was only slightly greater, with an estimated closure time for salt/bentonite, the slowest consolidating material, of 64 years. Swelling pressure exerted by the bentonite during the later stages of consolidation is small enough to neglect in closure calculations.

The estimated largest void fractions (porosities) that can exist in the backfill after long periods of time are between 0.09 and 0.05 for crushed salt and about 0.14 for salt/bentonite. Assuming the early accepted brine inflow rate of 43 m^3 in 100 years, there is insufficient brine flow into the rooms during consolidation to saturate either backfill. Furthermore, even if the materials were saturated, their shear strength in the consolidated state is sufficient to prevent extensive borehole enlargement during human intrusion.

Since both materials are considered acceptable with regard to their ability to achieve a sufficiently impermeable state in a reasonable time, the major difference is their potential for sorbing either brine or radionuclides. Salt has no demonstrated potential for sorption, but the bentonite in the salt/bentonite material was chosen specifically for this purpose.

Estimates about how much brine flowing into the disposal room during backfill consolidation, which can be chemically sorbable by bentonite, remain inconclusive because the effect of the exceptionally high impurity content of WIPP brine on sorption remains uncertain. Therefore, while no credit is taken, at present, for brine sorption in salt/bentonite backfill, the possibility that some amount of inflowing brine would be chemically bound is considered likely.

Predicted sorption of radionuclides by bentonite under condition of uniform distribution of brine throughout the backfill, and no brine flow through a disposal room, could be as much as 50% of the inventory. However, sorption is limited to plutonium, americium, neptunium, with practically no sorption of uranium. When brine transport by flow-through porous media is considered, the amount of sorbed material will be greatly reduced because flow localization or channeling will limit the amount of sorbent in direct contact with the bentonite.

6.2 Recommendation

Little difference was found between the the consolidation of backfill candidate materials crushed salt and 70% by weight salt/30% by weight bentonite to the final state of impermeability considered necessary to satisfy federal nuclear regulations. Therefore, any advantage of one backfill over the other is in the potential of bentonite to sorb brine and radionuclides. Although bentonite's sorption potential for water in brine may be considerably less than for pure water, sorption of some of the brine inflowing into the disposal room during consolidation is likely. Sorption of radioelements by bentonite under stagnant (no flow) room conditions can also occur, with up to 50% of the radionuclide inventory in the disposal room possible, although emplacement of pure bentonite between waste containers may be required to optimize conditions. In contrast, little or no sorption of any radioelements from brine flowing through the backfill would occur if flow channeling becomes important.

After consideration of the backfill selection criteria presented in this report (Table 6-1), salt/bentonite backfill is recommended for backfilling WIPP disposal rooms. While the sorption potential of bentonite is not ideal, no detrimental aspects of its adding bentonite to the salt as a backfill have been identified. In other words, selection of a suitable material has been made on the basis that having a material with properties of partial benefit, which may even exceed expectations, is better than a material that is totally lacking in such attributes.

TABLE 6-1. COMPARISON OF THE PROPERTIES OF CRUSHED-SALT AND SALT/BENTONITE BACKFILL

	Crushed-Salt Backfill	Salt/Bentonite Backfill
Required Criteria		
Low Permeability	$<10^{-19}$ m^2	$<10^{-19}$ m^2
Low Porosity	<0.08	<0.13
Chemical Stability	greatest	good
Shear Strength	acceptable	acceptable
Ease of Emplacement	good	good
Desirable Criteria		
Effect of Additives on Consolidation	predictable	predictable
Brine Sorption	none	questionable
Radionuclide Sorption	none	good
Deformability, Ability to Fill Voids	excellent	excellent
Thermal Conductivity	acceptable	acceptable

7.0 REFERENCES

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APPENDIX A:

EVALUATION OF BENTONITE AS A BACKFILL COMPONENT

EVALUATION OF BENTONITE AS A BACKFILL COMPONENT

Taken from an unpublished contractor report by

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Summary and Recommendations

Natural bentonite deposits vary considerably in terms of the predominant smectite species and the clay chemistry. In addition, most natural bentonites contain significant amounts of other (accessory) minerals, in particular quartz, cristobalite, feldspar, and mica minerals. In some cases, these accessory minerals may account for as much as 50% of the deposit. Most bentonites are primarily aluminous dioctahedral smectite clays, but they vary in terms of Al, Fe, Mg, and exchangeable cation contents. These compositional variations are related to variations in swelling pressure, hydrodynamics, and adsorption effectiveness.

The mineralogy of the bentonite to be used must be known. This should include knowledge of the types and amounts of accessory minerals, the presence of which will affect the sorption, swelling, and permeability properties of the backfill. Higher contents of accessory minerals should serve to lower the overall effectiveness of the backfill to adsorb solutes, lower the maximum swelling pressure, and increase permeability.

The clay comprising the bentonite must be chemically and mineralogically characterized as properties such as sorption, swelling pressure, and hydraulic conductivity are dependent on clay composition. Cation selectivity is dependent on layer charge density, which is primarily a function of compositional variations in octahedral sites. More Mg-rich varieties (e.g., Chambers (Cheto)-type montmorillonite) have a higher net layer charge and higher resulting cation exchange capacity than less magnesian varieties (e.g., Wyoming-type montmorillonite).

The use or comparison of test data on bentonites from other studies (e.g., Swedish SKB studies) must be done cautiously, keeping in mind the previous two summary points. If there are chemical or mineralogical differences in the bentonites tested, the results of otherwise identical experiments may differ also.

Sorption modeling should include an assessment of physical adsorption, absorption, ionic substitution, and precipitation phenomena. The formation of anion complexes or stable precipitates at high pH may be particularly important in predicting net solution concentrations of radionuclides if the brine becomes alkaline. Empirical models of chemisorption (e.g., Langmuir, Freundlich isotherms) are generally inadequate in real systems. Therefore, sorption should be modeled using surface complexation site-binding theory or a pH-independent isotherm model in combination with aqueous speciation modeling, and should also take into account competitive effects.

The potential effects of the release of organic compounds as a result of complete dissolution of the waste package are difficult to assess quantitatively but may be significant. Organic compounds may play a role in both sorption (as organics may sorb onto clay surfaces) and in mobilizing some elements (e.g., chelation).

Solute filtration efficiencies are due to complex interactions between a number of phenomena, including cation adsorption selectivity and hydrated radius, and hydrodynamic variables. The resulting relative efficiencies increase with temperature and compaction pressure.

Mg may be consumed from the brine by precipitation of Mg-chloride, or reconstruction of montmorillonite to Mg-saponite or chlorite. Mg consuming reactions may serve to drive the pH of the brine down initially.

Aluminous smectite clay will not be stable when exposed to WIPP brine for extended periods of time and can be expected to convert to illite. K will tend to be preferentially selected over other available adions onto clay surfaces; therefore, illite will be favored rather than chlorite. The development of illite will be a function of temperature and reaction time and can be expected to parallel illitization reactions observed in natural hydrothermal systems and K bentonites. A quantitative model for illite development should be developed that will predict how much of the smectite will be converted and the rate at which this conversion can be expected to take place.

Illite is more permeable and has a much lower cation exchange capacity than smectite. Therefore, the amount of smectite converted to illite or interlayered illite/smectite is critical in determining both the long-term sorption properties of the backfill and the hydrodynamic properties. Initial swelling of the bentonite will generate a maximum pressure which should eventually decrease in response to illitization. Radionuclides initially sorbed onto smectite may exhibit some desorption as recrystallization to illite should serve to lower the overall surface sorption potential. Illite development will alter the sorption and sealing properties of the backfill, making the material a less effective barrier overall.

APPENDIX B:

RADIONUCLIDE DISTRIBUTION COEFFICIENTS FOR BENTONITE

Sandia National Laboratories
Albuquerque, New Mexico 87185

Date: 21 August 1990
To: E. D. Gorham, 6344

From: C. F. Novak, 6344

Subject: Radionuclide Distribution Coefficients (K_d's) for Bentonite, and the Validity of the Linear Sorption Model for Multiple Radioelements

SUMMARY

This memo is in response to 1) the 14 August memo by B. M. Butcher containing a query as to appropriate K_d's for bentonite backfill, and 2) informal discussions within the department about modifications to the K_d model. This memo provides recommended values for the distribution coefficients, and presents rationale for using the linear sorption (K_d) model as it was developed, that is, without changes.

"RECOMMENDED" DISTRIBUTION COEFFICIENT VALUES FOR BENTONITE

K_d values for partitioning of radionuclides between brines and "pure clay" are given in Table D-5 of Lappin et al. (1989) as

100 ml/g	for Pu, Am, Cm, and Th
10 ml/g	for Np, and
1 ml/g	for U and Ra.

In the supporting text, they state that "For the drifts and seals, the values for clay must be multiplied by a utilization factor of 0.001 for use in a porous-medium transport equation to account for dilution of the clay by crushed salt and the possibility that fluid flow will be diverted away from the clay particles in the reconsolidated salt." Therefore, the adjusted K_d's are

0.1 ml/g	for Pu, Am, Cm, and Th
0.01 ml/g	for Np, and
0.001 ml/g	for U and Ra.

Approximating the grain density of the backfill material as 2 g/cm^3, with a porosity of 5%, the retardation factors calculated from these K_d's are 4.8, 1.38, and 1.038, respectively.

Experimental work on the sorption of uranium from brine onto Na-Bentonite has yielded preliminary K_d in the 1.5 to 7.6 ml/g range (Meyer, 1990), which is consistent with the "pure clay" values of Lappin et al.

I feel that the adjusted K_d's in the above table are the best estimates for backfill K_d's available, based on the discussion in Appendix D of Lappin et al. (1989).

THE LINEAR ISOTHERM (OR DISTRIBUTION COEFFICIENT, OR K_d) SORPTION MODEL

There has been informal discussion among members of the department about the appropriateness of using K_d's for the backfill, especially with regard to the "unlimited sorption capacity" inherent in the model. We'll address this after reviewing the model.

The linear sorption model states that the amount of an element sorbed onto a solid is proportional to the concentration of the element in solution, period. The model does not indicate how, where, or why the element sorbs, just how much sorbs. Distribution coefficients are valid only at the particular conditions at which they were determined; there is no way to extrapolate/interpolate K_d's to physical conditions (e.g. solution to solid ratio) or to chemical conditions (e.g. different ionic strength, different pH) other than those for which they were measured.

It is well known (see for example Kent et al., 1988; Hayes et al., 1990; and Siegel et al., 1990) that sorption isotherms are in general not linear, but are instead complex functions of solution chemistry. Figure 1 shows that uranium sorption on corrensite is a function of both pH and carbonate concentration; if the linear isotherm model were accurate, pH and carbonate concentration would have no effect on uranium sorption. The K_d model has not been replaced, though it is not very good, because sorption is not well understood, and very little data is available. Various surface complexation models (SCM) such as double layer, triple layer, and constant capacitance models, have been proposed (Kent et al., 1988; Hayes et al., 1990). These models have several fitting parameters which must be determined for each chemical system. In addition to the lack of data for use with SCM's, these models generally require rigorous chemical modeling which can take orders of magnitude more computer time. So, despite all its limitations, the linear sorption model remains the best model available for calculations which do not include rigorous chemistry.

It may never be practical to include the rigorous chemical models in performance assessment calculations. However, it may be possible to calculate "effective retardation coefficients," or delay coefficients, to coin a term, that can be used with approximately the same ease as distribution coefficients. This is an area of active research.

COMPETITIVE SORPTION WITHIN THE K_d MODEL

The question of competitive sorption within the K_d model has arisen. The argument is

> Can K_d's for two or more different elements, which were measured separately, be applied to systems with two or more elements sorbing at the same time? Do the elements sorb to the same sites and therefore compete, or do they sorb to different sites?

To my knowledge, all K_d data has been taken in systems with only one sorbing species, thus the effect of two species sorbing onto one substrate has not been examined experimentally. Since the distribution coefficient model has no theoretical basis, there is no way to determine this other than through experiment.

I feel that arbitrarily limiting the sorption capacity of the substrate (e.g. if element 1 and element 2 are both present and potentially sorbing, reduce the K_d's for each by "some factor" to account for competition) is not a reasonable way to proceed. There is absolutely no data or theory to suggest that setting a maximum sorption capacity will reflect reality any better than the distribution coefficient model. Indeed, it is possible that the sorption of one element would be enhanced, not hindered, by the presence of another sorbed element.

A Thought Experiment

The following thought experiment, in which we hold the K_d model to be completely accurate, argues that having multiple sorbing radionuclides does not violate the linear isotherm model.

Consider sorption of element A on a solid substrate. We add some A to a beaker and let it equilibrate with the solid in the beaker. We add more A and again let it equilibrate. The amount of A sorbed will increase, and we can describe the amount sorbed with the K_d model. If we also assume that element A will never form a precipitate, we could continue to add A to form very high concentrations, and the amount of A sorbed would increase correspondingly. There is no maximum amount of A allowed to sorb.

Now let's assume we have two kinds of element A, A-blue and A-red, which have identical chemical and physical properties, yet we can tell them apart. We bring a solution of A-blue into equilibrium with a substrate in two separate beakers, yielding identical systems. Now let's add x moles of A-blue to the first beaker, and x moles of A-red to the second beaker, and let them equilibrate. There is no question of competitive sorption in the beaker containing all A-blue, but what about the other beaker? When we add A-blue to the first beaker, we assume that the additional A-blue which is sorbed attaches to sites (somehow) without disturbing the amount of A-blue already sorbed. Because A-red has identical properties, it also would sorb without disturbing the already-sorbed A-blue.

Now let's assume that A-red really is a different element than A-blue, call it B, with different physical and chemical properties, and thus a different distribution coefficient. If we repeat the above experiment, would B compete with A-blue for sorption sites? No, at least not according to the K_d model. Generalizing this result, there is nothing inconsistent with allowing any number of elements to sorb to the same substrate **according to the K_d model**. The K_d model does not automatically break down when more than one radionuclide is available for sorption.

There is no reason to require a maximum sorption capacity within the linear sorption model, whether there are one or many sorbing species. In fact, to include such a limit will destroy the consistency of the linear sorption model, because it would no longer be linear.

Conclusions About Competitive Sorption

I do not support altering the sorption capacity of substrates when more than one radionuclide is present and sorbed. Currently, the K_d model is the best method we have to model interactions of dissolved species with the solid phase in the performance assessment framework.

I do support the development of (simple) alternative models which can explain laboratory data better than the linear sorption model, but this should be done according to the Scientific Method. I have seen no theory or correlation which does this, and thus I feel it inappropriate to include a model with limited maximum sorption capacity in performance assessment calculations at this time.

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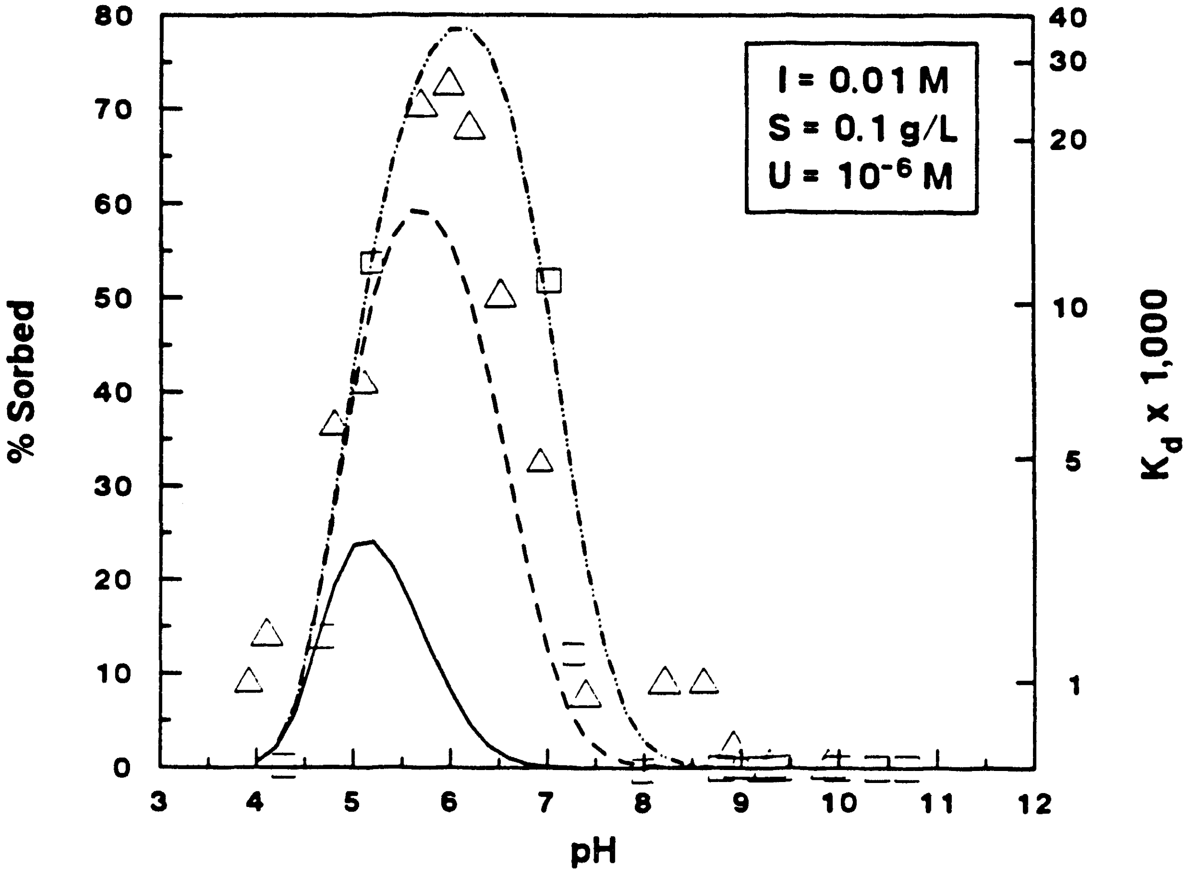

Figure 1 Effect of Carbonate Concentration on Uranium Sorption by Corrensite: Data and Model Fit for CorWa Reference Corrensite. Experimental data: △ = 10^{-4}M CO_3^{2-}; □ = 10^{-5} M CO_3^{2-}. Model calculations: ——— = 10^{-3} M CO_3^{2-}; ----- = 10^{-4} M CO_3^{2-}; - - - - = 10^{-5} M CO_3^{2-}. S = concentration of solid in the suspension.

(from Siegel et al., 1990.)

AUTHOR'S SUPPLEMENT TO APPENDIX B

C. F. Novak would like to emphasize and expand upon some of the issues raised in the preceding memo (Appendix B).

A value for K_d applies only to the physical and chemical conditions under which it was measured. The WIPP room environment is expected to be extremely heterogeneous on a centimeter scale. This heterogeneity precludes the use of a single K_d value for each element because the chemical conditions will vary widely; it would not be unreasonable to expect local sorption ratios to vary by orders of magnitude. However, if each room "micro-environment" were characterized completely, sorption could be integrated over the room to arrive at a "room average" K_d. These are the values provided in the above memo.

The discussion of the appropriateness of the K_d model was not intended to indicate that the K_d model is an accurate representation of the solid-aqueous interactions in the WIPP room environment. At this time and within project constraints, the K_d model for performance assessment calculations is the best available. The purpose was to discourage arbitrary modification of the K_d model in an attempt to improve it, and to encourage the development of better models from first principles.

11 April 1991

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