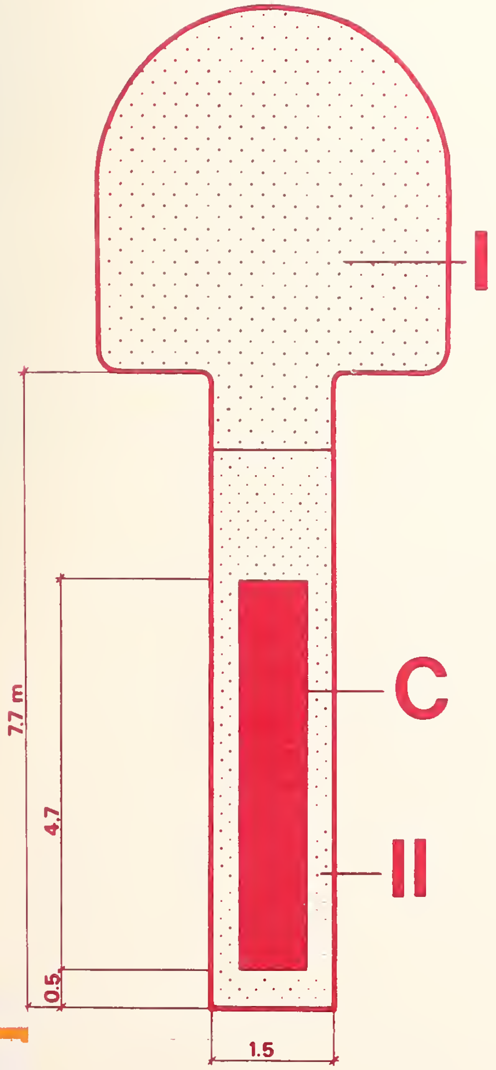
NBS SPECIAL PUBLICATION **668**

U.S. DEPARTMENT OF COMMERCE/National Bureau of Standards



Nuclear Waste Management Program

Summary Report

NATIONAL BUREAU OF STANDARDS

The National Bureau of Standards[1] was established by an act of Congress on March 3, 1901. The Bureau's overall goal is to strengthen and advance the Nation's science and technology and facilitate their effective application for public benefit. To this end, the Bureau conducts research and provides: (1) a basis for the Nation's physical measurement system, (2) scientific and technological services for industry and government, (3) a technical basis for equity in trade, and (4) technical services to promote public safety. The Bureau's technical work is performed by the National Measurement Laboratory, the National Engineering Laboratory, and the Institute for Computer Sciences and Technology.

THE NATIONAL MEASUREMENT LABORATORY provides the national system of physical and chemical and materials measurement; coordinates the system with measurement systems of other nations and furnishes essential services leading to accurate and uniform physical and chemical measurement throughout the Nation's scientific community, industry, and commerce; conducts materials research leading to improved methods of measurement, standards, and data on the properties of materials needed by industry, commerce, educational institutions, and Government; provides advisory and research services to other Government agencies; develops, produces, and distributes Standard Reference Materials; and provides calibration services. The Laboratory consists of the following centers:

Absolute Physical Quantities[2] — Radiation Research — Chemical Physics — Analytical Chemistry — Materials Science

THE NATIONAL ENGINEERING LABORATORY provides technology and technical services to the public and private sectors to address national needs and to solve national problems; conducts research in engineering and applied science in support of these efforts; builds and maintains competence in the necessary disciplines required to carry out this research and technical service; develops engineering data and measurement capabilities; provides engineering measurement traceability services; develops test methods and proposes engineering standards and code changes; develops and proposes new engineering practices; and develops and improves mechanisms to transfer results of its research to the ultimate user. The Laboratory consists of the following centers:

Applied Mathematics — Electronics and Electrical Engineering[2] — Manufacturing Engineering — Building Technology — Fire Research — Chemical Engineering[2]

THE INSTITUTE FOR COMPUTER SCIENCES AND TECHNOLOGY conducts research and provides scientific and technical services to aid Federal agencies in the selection, acquisition, application, and use of computer technology to improve effectiveness and economy in Government operations in accordance with Public Law 89-306 (40 U.S.C. 759), relevant Executive Orders, and other directives; carries out this mission by managing the Federal Information Processing Standards Program, developing Federal ADP standards guidelines, and managing Federal participation in ADP voluntary standardization activities; provides scientific and technological advisory services and assistance to Federal agencies; and provides the technical foundation for computer-related policies of the Federal Government. The Institute consists of the following centers:

Programming Science and Technology — Computer Systems Engineering.

[1] Headquarters and Laboratories at Gaithersburg, MD, unless otherwise noted; mailing address Washington, DC 20234.

[2] Some divisions within the center are located at Boulder, CO 80303.

Nuclear Waste Management Program Summary Report

Workshop on Research and Development Needs in Backfill for Long Term Storage of Nuclear Waste in Deep Geologic Repositories, held at the National Bureau of Standards, Washington, DC, April 13-14, 1981

Riley M. Chung, Editor

Center for Building Technology
National Engineering Laboratory
National Bureau of Standards
Washington, DC 20234

Prepared for:

Office of Nuclear Waste Management
Department of Energy
Washington, DC 20585

U.S. DEPARTMENT OF COMMERCE, Malcolm Baldrige, Secretary
NATIONAL BUREAU OF STANDARDS, Ernest Ambler, Director

Issued December 1983

Library of Congress Catalog Card Number: 83-600607

National Bureau of Standards Special Publication 668
Natl. Bur. Stand. (U.S.), Spec. Publ. 668, 228 pages (Dec. 1983)
CODEN: XNBSAV

U.S. GOVERNMENT PRINTING OFFICE
WASHINGTON: 1983

For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington, DC 20402

Table of Contents

Papers presented to the Workshop and Published in this Report

Paper Presented to the Workshop but withdrawn from Publication

Effect of Hydrothermal and Cyclic Wetting - Drying Treatment on Properties and Characteristics of Montmorillonitic Clay, A.J. Selti, R.N. Yong, McGill University; and G.W. Bird, Atomic Energy of Canada Limited, Canada

Paper not Presented to the Workshop but Included in this Report

ABBREVIATIONS OF ORGANIZATIONS

AECL	Atomic Energy of Canada Limited
ANL	Argonne National Laboratory
BNL	Brookhaven National Laboratory
CBT/NBS	Center for Building Technology, National Bureau of Standards
DoE	Department of Energy
LASL	Los Alamos Scientific Laboratory
LBL	Lawrence Berkeley Laboratory
LLL	Lawrence Livermore Laboratory
MCC	Material Characterization Center
NBS	National Bureau of Standards
NRC	Nuclear Regulatory Commission
ONWI	Office of Nuclear Waste Isolation
ORNL	Oak Ridge National Laboratory
PNL	Battelle Pacific Northwest Laboratory
RHO	Rockwell Hanford Operation
SNL	Sandia National Laboratory
SRL	Savannah River Laboratory

AGENDA

Monday, April 13, 1981

8:30 - 9:00	Arrival at NBS, Lecture Room B Administration Building
9:05 - 9:15	Welcome - Riley M. Chung, NBS
9:15 - 9:25	NBS Nuclear Waste Program - William P. Reed, NBS
9:25 - 9:35	Structures and Materials Programs of CBT/NBS, Edward O. Pfrang, NBS
9:45 - 10:10	Pacific Northwest Laboratory's Backfill Development Project, E.J. Wheelwright, PNL
10:10 - 10:35	Design of Backfill (Overpack) Materials, Rustum Roy and S. Kamarneni, Pennsylvania State University
10:35 - 11:00	Backfill Barrier Research and Development for Radioactive Waste Isolation in Salt, E.J. Nowak, SNL
11:00 - 11:25	The Development and Testing of Waste Package Backfill Materials for a Nuclear Waste Repository Located in Basalt, M.I. Wood, RHO
11:25 - 1:00	Lunch at NBS Senior Lunch Club
1:00 - 1:15	Conditions Affecting the Backfill in a Waste Package for Repository in Tuff, Part I, J.K. Johnstone, SNL
1:15 - 1:40	Conditions Affecting the Backfill in a Waste Package for Repository in Tuff, Part II, E.N. Vine, LASL

1:40 - 2:05	Critical Factors Affecting the Use of Montmorillonite as a Nuclear Waste Backfill Material, G.W. Beall, A.G. Lamkin, and K.L. Kelly, Radian Corp.
2:05 - 2:20	Selection and Evaluation of Buffer and Backfill Materials for Use in a Nuclear Fuel Waste Disposal Vault: The Canadian Research program, G.W. Bird, AECL
2:20 - 2:35	Coffee Break
2:35 - 2:50	Evaluation of Thermal Properties of Buffer Materials for a Deep Underground Nuclear Waste Disposal Vault, H.S. Radhakrishna, Ontario Hydro
2:50 - 3:15	Effect of Hydrothermal and Cyclic Wetting - Drying Treatment on Properties and Characteristics of Montmorillonitic Clay, A.J. Sethi, R.M. Yong, McGill University, and G.W. Bird, AECL
3:15 - 3:40	Bentonite Shielding of Rock-Deposited Radioactive Wastes, R. Pusch, University of Lulea, Sweden
3:40 - 3:55	Coffee Break
3:55 - 4:50	Open Discussion and Adjournment, R.M. Chung, F.Y. Yokel, and L.A. Salomone, NBS

Tuesday, April 14, 1981

9:00	Arrival at NBS, Building 226, CBT Conference Room
9:00 - 12:00	Informal Discussions on Backfill with Geotechnical Engineering Staff
12:00 - 1:30	Lunch at NBS Senior Lunch Room

1:30 - 3:00	Return to CBT Conference Room to continue discussion
3:00 - 4:00	Preparation of Written Recommendations, E.J. Nowak and F.Y. Yokel
4:00	Adjournment

ABSTRACT

An international workshop on the backfill component of a multi-barrier underground isolation system for nuclear waste disposal/storage was conducted on April 13 and 14, 1981, at the National Bureau of Standards, Washington, D.C. This report includes the papers presented and/or submitted to the workshop for publication. It also contains the surveyed results of a questionnaire distributed to the participants at the workshop. The workshop held a one-day informal in-depth discussion on the subject and a summary of these discussions is included in this report, which states areas where further research and development needs are required to better understand the fundamental mechanisms of the backfill in the waste repository.

Key words: backfill; compressibility; geology; nuclear waste; permeability (hydraulic conductivity); research and development; shear strength; sorption; swelling; thermal conductivity.

INTRODUCTION

In 1979, the Interagency Review Group (IRG) Report, presented to then President Carter, stated that there were about 76 million cubic feet of long lived radioactive nuclear wastes stored at various interim sites around the United States. In addition to these wastes generated by military and civilian programs, there are large quantities of uranium mill tailings, transuranic wastes, and spent fuel discharged from commercial nuclear power reactors. Nuclear wastes will be continuously generated by the military weapons and propulsion programs and by the operation of over 65 nuclear power reactors and associated fuel service facilities. Therefore, no matter how uncertain the future that the nuclear industry is facing, nuclear wastes do exist and their environmentally safe disposal will remain as a critical public concern in both the States and overseas.

Major classes of nuclear wastes include high level wastes (HLW), transuranic wastes (TRU), low level wastes (LLW), uranium mine and mill tailings, and gaseous effluents. The NBS program was directed toward high-level and transuranic wastes which pose the greatest hazard to society, and thus require the greatest technical input for their safe management and disposal.

A number of alternative methods for disposing of high-level wastes have been proposed such as placing them into an orbit around the sun, transmutation by neutron bombardment, or deposition into subseabed. In the IRG report these wastes are recommended to be put into a stable form and disposed of by burial in deep geologic sites using a multi-barrier system. As a simplified example, the multi-barrier system consists of a glass waste form, a metal canister containing the waste form which is lowered into a borehole drilled from the floor of a tunnel in the repository, backfill placed between the canister and the borehole wall, and the repository rock.

The glass is only one of the possible waste forms. Others including ceramics and composites (e.g., synthetic rocks) have been considered. Metal canisters made of stainless steel, copper, and titanium alloys were included in the study. It is probable that the selections between waste package and geologic settings are inter-related and the choice of one could influence the optimum choice of the other. Thus, it is logical to tailor a waste package to the requirements of the geohydrologic environment of a geologic site.

Up to the present (1981), geologists have not been able to reach a consensus on the ability to adequately predict long-term geologic stability. For this reason, the Nuclear Regulatory Commission (NRC) is currently (as of 1981) proposing regulations which rely heavily on the selection of waste form and the design of a total waste package for providing a safe geologic storage/disposal system. The proposed NRC regulations focus on four different time periods for the management of both high-level and TRU wastes. The first time period includes waste processing, transportation, and repository operation. The second time period, which is 50 to 100 years after waste emplacement, is the interval proposed by NRC for possible retrieval of waste from the repository if difficulties are detected. The third (up to 1000 years after emplacement) and fourth (beyond 1000 years) time periods follow from ingestion toxicity of fission products as a function of decay. For the third time period, the proposed NRC regulations would require that there be reasonable assurance that there will be no release of radionuclides from the waste package for the 1000 years time period. The proposed regulations would require the release rate for radionuclides not to exceed 10^{-5} of its radioactivity per year for the fourth time period beyond 1000 years.

In view of the above consideration, a great deal of new scientific and engineering information is needed to arrive at a technical public consensus. A large technical program has been developed by the Department of Energy (DoE) to provide the necessary information for waste containment and the program ranges from scientific research to building processing plants and studying candidate repositories. It is conceivable that the credibility of the properties measurements program of DoE to the regulatory agencies of the government, the scientific community, and the general public will be based upon a referenceable basis for test methods and long term predictive capability to determine materials and systems performance. And it is in this capacity that NBS was invited by DoE in 1979 to participate in the national nuclear waste storage/disposal program.

The NBS program covers all the components of the multi-barrier system except the repository rock. The study of the backfill component was assigned to the Geotechnical Engineering Group of the Structures Division in Center for Building Technology. The objectives of the NBS backfill program can be summarized as follows:

- To formulate performance criteria against which backfill materials can be evaluated.
- To develop or adopt methods for measuring the performance of backfill materials.
- To evaluate the consistency and accuracy of available data on backfill materials.
- To select and characterize research materials.
- To determine, and to the extent possible, standardize the tests by which backfill materials are characterized and evaluated.

One major activity in our first year program was to conduct an international workshop with respect to the specific needs of the backfill in a waste package geohydrologic environment. This led to the gathering of thirty-eight people on April 13th and 14th, 1981 at the NBS Campus. Experts from national laboratories and universities as well as from Canada and Sweden participated in the program. On the first day, eleven papers were presented. The second day was devoted to informal in-depth discussions on the subject. Discussions were focused on the current state of backfill evaluation and the research and development needs in research materials, test variables, test methods and test equipment, and fundamental mechanisms of the backfill in a repository environment.

This report includes the papers presented to the workshop and also one paper which was submitted later for publication. Unfortunately, three papers were withdrawn by the authors because of the lack of time for completing the manuscripts. The report also presents the surveyed results of a questionnaire distributed to the participants at the workshop. Finally, a summary of the discussions generated during the workshop states areas where further research and development needs are proposed to better understand the fundamental mechanisms of the backfill in a waste repository; thus leading to a better prediction of the long term behaviors of the backfill material.

PNL BACKFILL DEVELOPMENT PROJECT(1)

E. J. Wheelwright
F. N. Hodges
J. H. Westsik, Jr.
L. A. Bray
Pacific Northwest Laboratory
Richland, Washington 99352

ABSTRACT

Preliminary results of the first year backfill development project of the Pacific Northwest Laboratory (PNL) were presented. A consultant committee which consists of five members was formulated at the inception of the project. All of them are from outside the nuclear community with complementary areas of expertise. The desired material properties of backfill were evaluated and its functions were divided into two categories: the primary and supporting attributes. Primary attributes are those which can have a direct effect upon one or more nuclide transport mechanisms. The supporting attributes do not directly affect a nuclide transport mechanism but provide support to the primary attributes. Of course, there is some overlap and interdependency.

A test program has been formulated. The materials selected for study include Nabentonite, Ca-bentonite, and illite with quartz sand mixtures. Some small amount of other materials will also be added to the mixture to study the sorption properties of anions or for Eh/Ph control. The test program will address several areas including compaction of materials, measurement of hydraulic conductivity, measurement of ion retardation, measurement of thermal conductivity, determination of mechanical properties, and determination of chemical stability under repository conditions. Only the test results of compaction and hydraulic conductivity are presented in this paper. The compaction density has been shown to be a function of compaction pressure and the water content of the

---

1/ Work Sponsored by Office of Nuclear Waste Isolation.

sample during compaction. Proper degree of hydration of the clay materials is important in order to obtain maximum density. Hydraulic conductivity measurements are being carried out in a test set-up developed at PNL. Preliminary test results indicate that values of hydraulic conductivity of less than 10^{-12} cm/sec can be expected from pure Na-bentonite clay. It should be pointed out that all hydration conductivity tests were conducted under rather high hydraulic gradients which were in the range of 3.5×10^4 to 2.1×10^5. These hydraulic gradients are considered much higher than those the backfill will be actually subjected to in the field. Extrapolation of experimental data to the actual gradient region will thus overestimate the flow. Therefore, the laboratory results on hydraulic conductivity are conservative.

INTRODUCTION

Concepts currently under investigation for the disposal of spent nuclear fuel or separated high-level nuclear waste involve deep emplacement of the waste in continental geological formations. In such disposal concepts, primary canisters or primary canisters contained within one or more overpacks are placed in an oversized hole bored into the geologic medium, and the space between the surface of the canister or overpack and geologic medium is backfilled with a suitable material or combination of materials. In such a concept, the waste form, the canister, the overpack, the backfill, and the geology itself each constitute a barrier to transport of the radioactive materials to the biosphere. The use of multiple, well-engineered, independent barriers significantly increases the probability of containment for the required time. We recognize that PNL is not the first to recognize the value of using an engineered backfill and we do want to make maximum use of the work of others.

The Backfill Development Project at PNL is sponsored by the Office of Nuclear Waste Isolation (ONWI). The project has been funded for just over 12 months. ONWI has directed PNL to develop backfill materials for use in repository systems in basalt and tuff. We will do a very limited number of tests of material in salt. The current PNL program is limited to materials development and testing.

SCOPE

The project scope is given below. The first two items are largely completed and our attention is now focused on the third item.

- A definition and prioritization of the desired attributes of the backfill material-quantitative specifications where possible.
- A screening of potential backfill materials and the selection of a manageable number of candidates, i.e., 6 to 12, for laboratory evaluation. This will include multi-component material to satisfy multi-functional requirements.
- Laboratory evaluation of the candidate backfill materials aimed at measuring their performance against the defined requirements.

COMMITTEE OF CONSULTANTS

At the inception of the project, a consultant committee was formed by selecting people of recognized stature. A deliberate effort was made to secure experts from outside the nuclear community with complementary areas of expertise. The committee consists of five members and their names and affiliations are given in table 1. Dr. Lindsay's interests are in the application of electrochemistry to soils, minerals and groundwater. Dr. Low is preeminent in the area of clay-water interactions. Dr. Berner's areas of interest include geochemical processes and rock-water interactions. Dr. Lerman's field of interest is geochemical modeling. Dr. Rollins was selected because of his engineering experience in the structural aspects of clays.

The committee has functioned to review our research plans, and to review and comment on published work from other backfill programs. They will critique our experimental results and conclusions and we hope that the work will result in some jointly authored publications.

FUNCTIONS OF BACKFILL

On the basis of a literature search and a brainstorming session with the consultant committee, the list of backfill attributes was selected as shown in table 2. The list is divided into primary and secondary attributes. Primary attributes are those which can have a direct effect upon one or more nuclide transport mechanisms. The supporting attributes do not, by themselves, directly effect a nuclide transport mechanism but provide support to the primary attributes. There is, of course, some overlap and interdependency.

MATERIALS

The materials selected for study are shown in table 3. The principal materials are Na-bentonite-Quartz sand mixtures. Tests are planned in which small percentages of other materials will be added in an attempt to enhance specific additional properties, i.e., absorption of ^{129}I or Eh/pH control, without seriously detracting from the primary function of providing a barrier to groundwater migration. Calcium-bentonite and illite are included in the list so we can study the effects upon hydraulic conductivity of the conversion of Na-bentonite to one of these phases.

TESTING PROGRAM

Experimental work has been initiated in the four areas shown in table 4. Most comments will be made about work in the first two areas.

Compaction of Materials

Clays, quartz sand, zeolites, minerals, and metal powders are being mixed in various ratios and pressed in an air-hydraulic press to study the relationship of compaction density to material composition and pressing conditions. The objective is to obtain a backfill material composition that will maintain its physical shape during placement, and will then prevent water intrusion, provide enhanced radionuclide sorptive capacity, provide adequate thermal transport, and buffer the Eh-pH of the backfill environment.

The results of compaction experiments with undiluted Na-bentonite (Volclay CS-50[2]) are shown in figures 1 and 2. The solid lines in figure 1 show the relationship between applied pressure and final density for Na-bentonite containing differing amounts of absorbed water. A reference point of 0.0 wt percent absorbed water was assumed for clay that was heated at 110°C for 16 hours. Vacuum drying at the same temperature for the same time gave comparable dehydration. On this basis, the as-received clay contained 7.5 wt percent absorbed water. Clays of higher hydration were obtained by equilibration with water vapor in a sealed system. After the pressed pellets were weighed and measured, each was dehydrated by heating in an oven at 110°C for 16 hours. The dehydration results are shown by the dashed lines in figure 1. The four pellets pressed from previously dehydrated clay powder did not change during the pellet dehydration step.

In figure 2, pressed pellet density is shown as a function of the water content of the unpressed clay for compaction pressed at four different pressures. The importance of using clay with the proper degree of hydration is clearly shown.

Measurement of Hydraulic Conductivity

A schematic of the equipment assembled for measuring hydraulic conductivity of compacted materials is shown in figure 3.

Details of the constant-volume flow through cells are shown in figure 4. The backfill material to be tested is compressed inside the heavy-wall cylinder by a hydraulic press in a separate system. When assembled as shown, the tie bolts hold the components together and resist expansion of the clay as it is hydrated. The difference in pressure on the load cell before and after pumping gives the clay swelling pressure.

Preliminary results from experiments completed and still in progress are shown in table 5. In these tests, the materials were compacted at 138 MPa. Synthetic

---

[2] Registered trademark of American Colloid Company, Skokie, Illinois.

Grande Ronde (Hanford basalt) groundwater was used as the permeating fluid. As the ratio of clay to sand is increased, the conductivity decreases by several orders of magnitude to less than 10^{-12} cm/sec for pure Na-bentonite clay. The hydraulic conductivity of the 25 wt percent clay-75 percent sand mixture shows a decrease by a factor of two over the 189 hour period following the initial breakthrough.

Because of the low hydraulic conductivities of these backfill materials, experimental hydraulic gradients must be significantly higher than those expected in a geologic repository in order to obtain data in a reasonable time period. The data given in table 6 shows that for a 50 percent clay-50 percent sand mixture, the hydraulic conductivity is reasonably constant over a hydraulic gradient range 3.5×10^4 to 2.1×10^5. For these gradients the backfill materials appear to follow Darcy's Law. Deviations from Darcy's Law are not expected until the hydraulic gradient drops below 100. Extrapolations of experimental data to this region will usually overestimate the flow. Therefore, the laboratory results are conservative.

Table 1

DBM Consultants

Dr. W. L. Lindsay	Colorado State University
Dr. P. F. Low	Purdue University
Dr. R. A. Berner	Yale University
Dr. A. Lerman	Northwestern University
Dr. R. Rollins	Brigham Young University

Table 2

Backfill Attributes

Primary

- Exclude groundwater from the waste canister
- Retard migration (immobilization) of selected radionuclides
- Control groundwater pH and Eh

Supporting

- Be compatible with geology and other engineered barrier materials
- Self-seal minor cracks or fractures in backfill and interfacing geology
- Retain performance criteria at all repository temperatures
- Retain performance criteria during and after gamma radiation
- Maintain mechanical stability
- Provide adequate thermal conductivity

Table 3

Backfill Materials

o Clay	Na-bentonite
	Ca-bentonite
	Illite
	Treated Na-bentonite
o Sand	SiO_2 (10-230 mesh)
o Zeolites	Clinoptilolite
	13X
	Zelon 900
o Metal powder or fibers	Iron
	Aluminum
	Lead Oxide
o Minerals/Rocks	Pyrite (FeS_2)
	Ferrosand
	Basalt
	Tuff

Table 4

Current Materials Testing Program

- Compaction of materials
- Measurement of hydraulic conductivity
- Measurement of ion retardation
- Measurement of thermal conductivity

Table 5

Hydraulic Conductivity of Clay/Sand Mixtures

Percent Na-Bentonite	Percent Sand (80-120 Mesh)	Initial Density (g/cm^3)	Hydraulic Conductivity (cm/sec)	Time to Migrate One Centimeter (years)
25	75	2.1	2×10^{-9}*	15
50	50	2.1	3×10^{-12}	11,000
100	0	2.1	7×10^{-13}	45,000

* Hydraulic conductivity decreasing with time

Table 6

Pressure Effects on Hydraulic Conductivity of 50-50 Clay/Sand Mixture

Hydraulic Gradient	Equivalent Pressure Drop (psi)	Time at Pressure (hours)	Total Time (hours)	Hydraulic Conductivity (cm/sec)
1.55×10^5	2200	169*	169	4.1×10^{-12}
7.03×10^4	1000	72	241	--
3.52×10^4	500	62	309	3.5×10^{-12}
7.03×10^4	1000	72	381	3.1×10^{-12}
2.11×10^5	3000	97	478	3.1×10^{-12}
1.58×10^5	2250	334	812	2.9×10^{-12}

* Includes 64 hours before initial water breakthrough

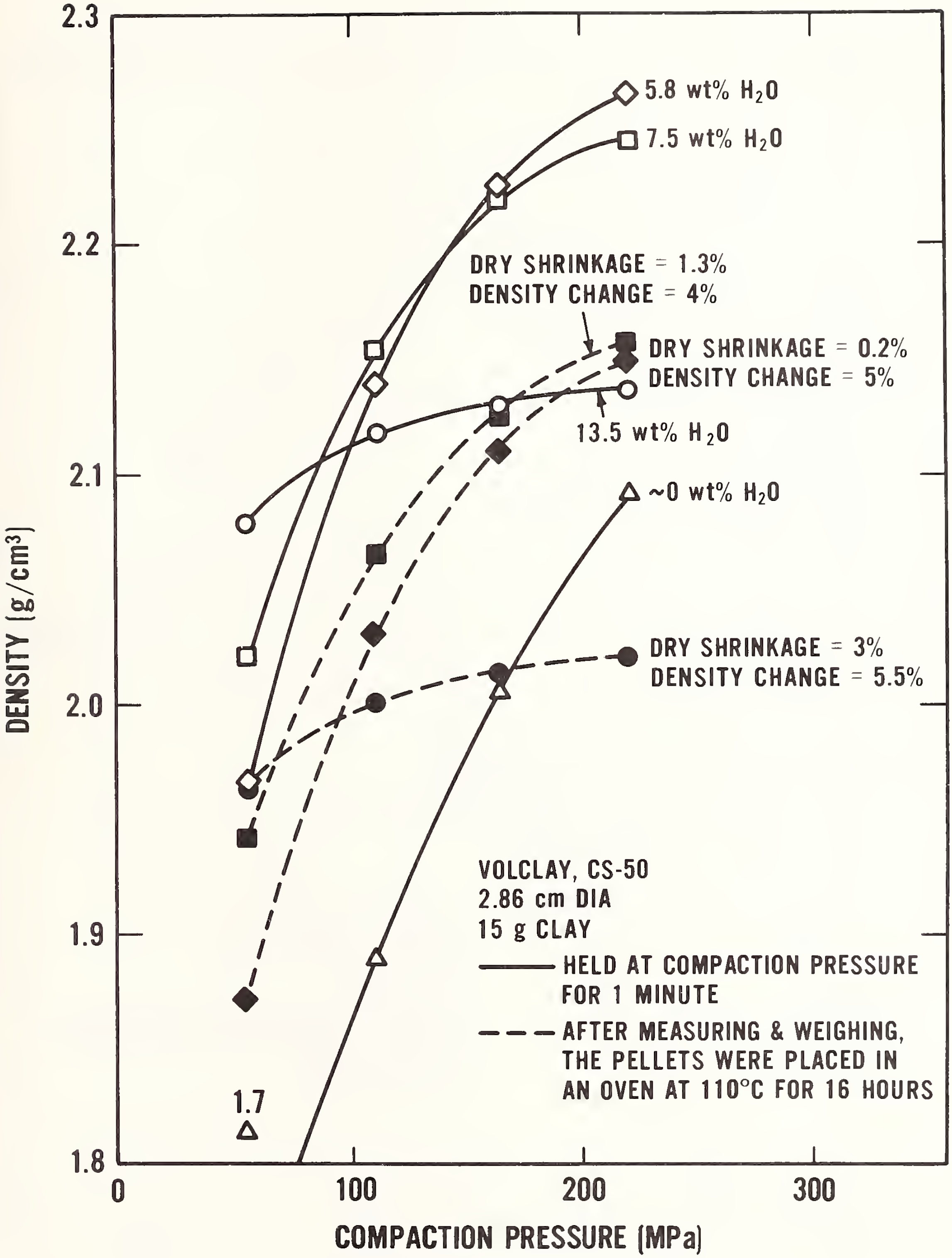


Figure 1. Density of Na-Bentonite as a function of compaction pressure and water content

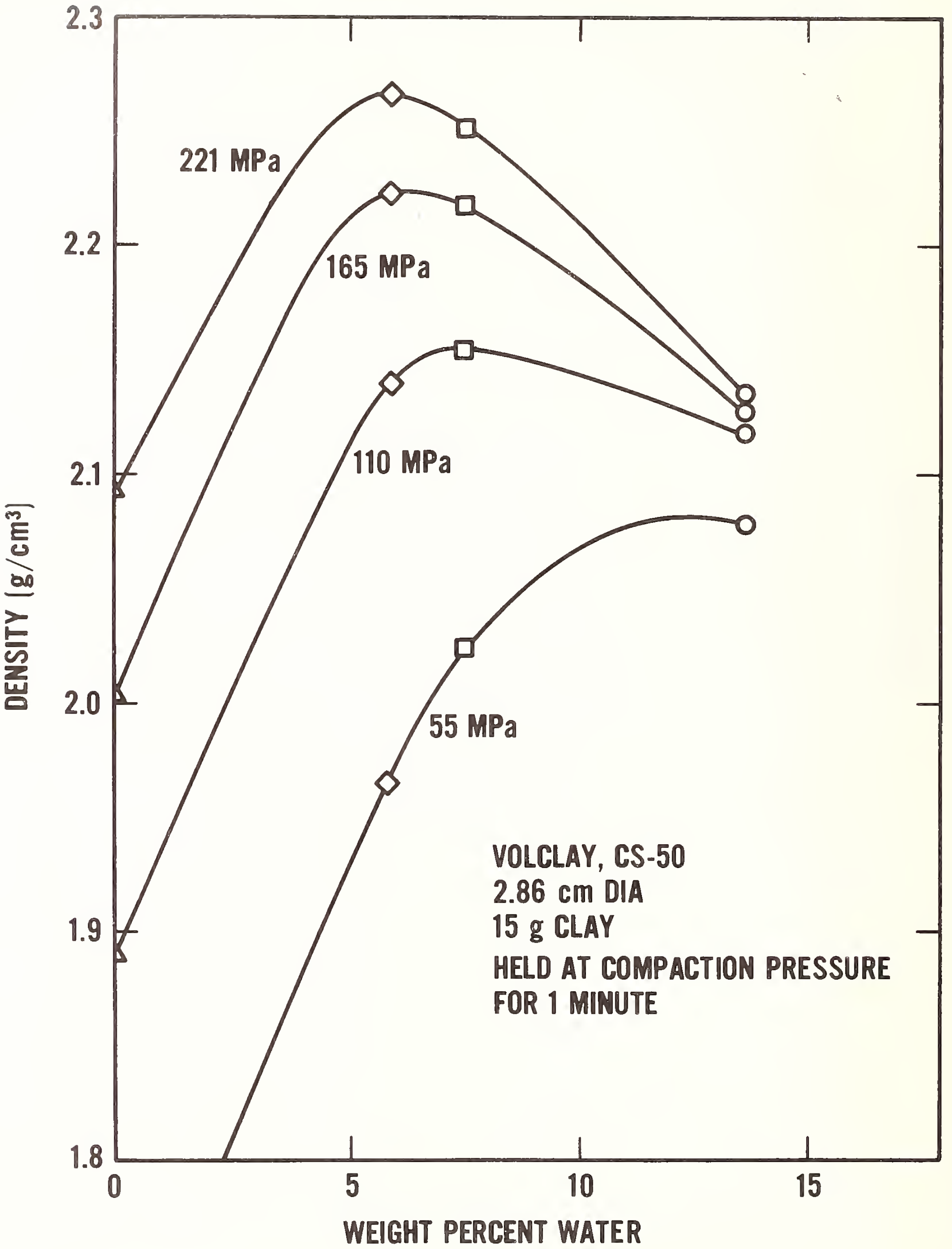


Figure 2. Density as a function of compaction pressure and H_2O content

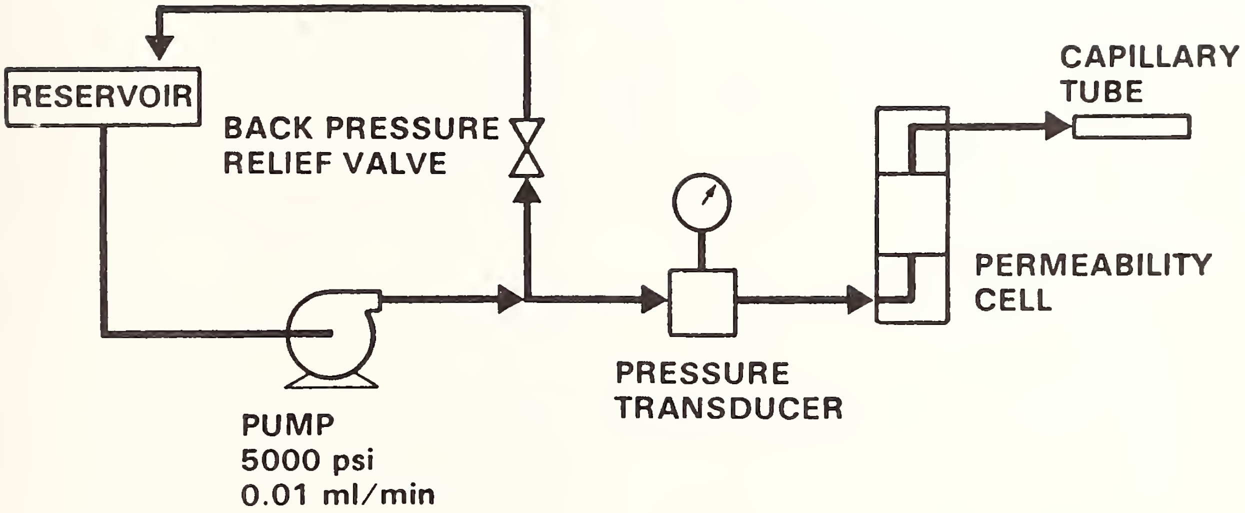


Figure 3. System set-up for hydraulic conductivity testing

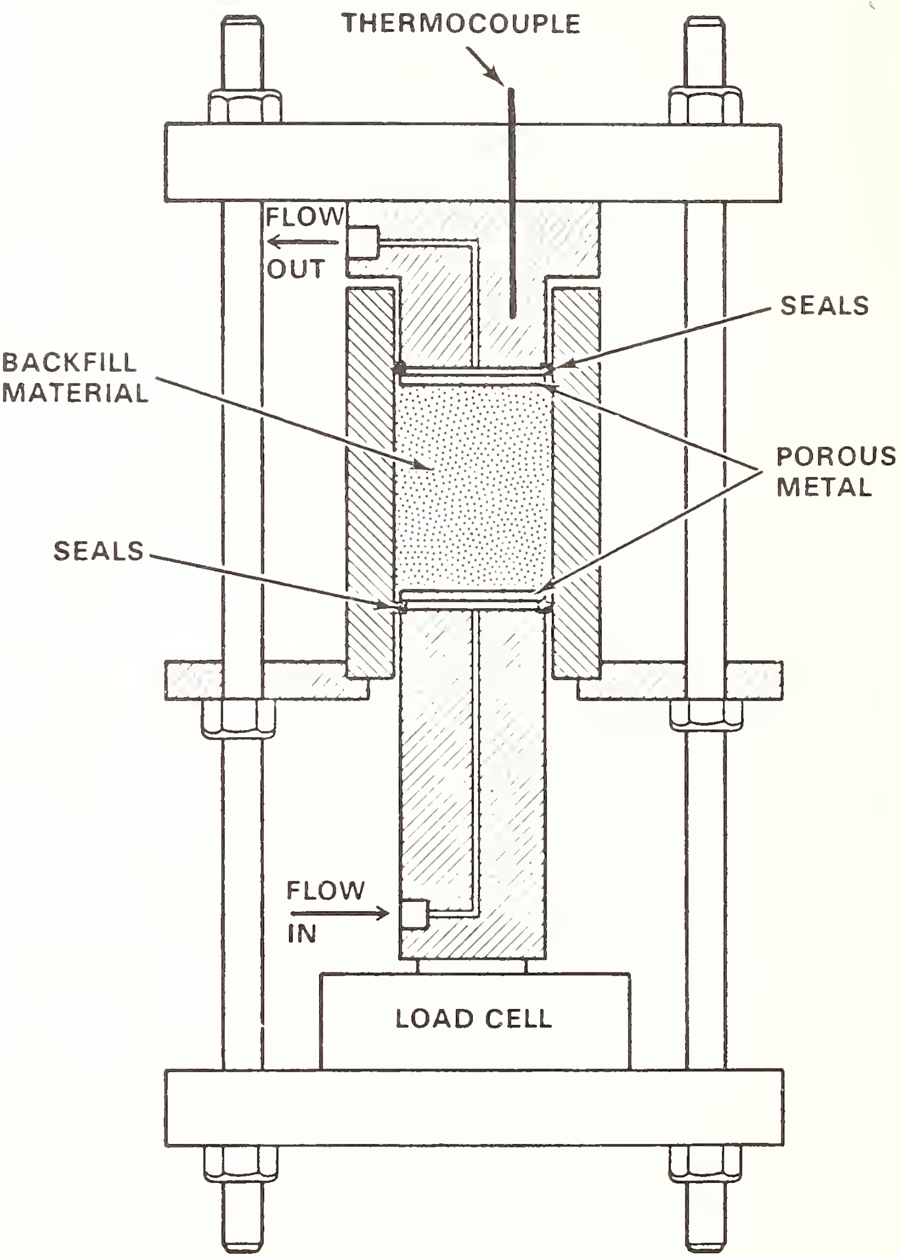


Figure 4. Schematic drawing of equipment for hydraulic conductivity measurement

DESIGN OF OVERPACK (= BACKFILL) MATERIALS

Rustum Roy and S. Komarneni
Materials Research Laboratory
The Pennsylvania State University
University Park, PA 16802

ABSTRACT

Research on backfill (= overpack) design at The Pennsylvania State University has focussed on the stability of such materials under repository conditions, the interactions of these materials and waste under repository conditions, and the sorption and fixation properties of backfill materials. Montmorillonite among clay minerals and mordenite and clinoptilolite among zeolites have been identified as the best backfills based on the present studies. Areas for further research include thermal stability studies of backfill materials under repository conditions in conjunction with natural analogs, waste-backfill interactions and design of backfill materials for purposes such as maintaining a reduced environment and keeping the repository dry.

INTRODUCTION

For most of the life of the research and development program for nuclear waste management, the role of the waste form--the source of radionuclides--was considered secondary if of any significance at all. As late as in the WIPP documents, absurd "assumptions" were made such as the total 'dissolution' of the waste forms. It does not, therefore, come as any surprise that the concept of the entire waste package itself as a subsystem of redundant barriers has had a difficult time gaining acceptance and understanding. Since 1973 we have advocated this approach (7), and figure 1 shows a late model of our concept of the waste package.

We have consistently used the term "overpack" for the silicate-oxide material to be packed around the canister; however, since different terminology has been adopted in this conference, we shall henceforth use the term "backfill" for what in all our referenced work is called "overpack."

The roles of a backfill barrier in any repository can be envisaged as follows: (a) to selectively adsorb and fix (retain) radioactive ions, in the case of a leakage; (b) to interact with waste to form new crystalline phases under repository conditions; (c) to maintain a reducing environment; (d) to keep the water from reaching the canister. The composition and structure of backfill materials should vary depending upon whether salt or other geological formations are used. There is, however, very little published work on the design of backfill. The Swedish KBS study in 1977 (1) was the first to propose a specific tailor-made set of materials. Research on backfill design at The Pennsylvania State University has examined several facets of the materials problem: (a) hydrothermal stability of the minerals under repository conditions (3); (b) interactions between waste and backfill under repository conditions (4, 6); (c) high sorption and fixation capacity for radwaste ions by materials treated in different ways (2). In the following we present first a summary account of the several preliminary research projects on backfill conducted at The Pennsylvania State University. Then we analyze the role of the backfill in the evaluation of waste forms.

TRANSFORMATION OF CANDIDATE BACKFILL MATERIALS UNDER RADWASTE REPOSITORY CONDITIONS

Although there will no doubt eventually be a wide variety of repositories for different wastes (defense and commercial), of different ages, in different countries, there is no doubt that mild "hydrothermal" conditions will exist in a nuclear waste repository during the thermal period. Such exposure to T, pH_2O, etc., can significantly alter the minerals in a backfill prior to leakage of radioactive ions because the backfill is in the immediate vicinity of the nuclear waste containing canisters. If the backfill materials are easily altered (i.e., thermally unstable), the very purpose of using them may be lost. Therefore, several candidate backfill materials have been screened for their stability (3). Clay minerals such as montmorillonites and vermiculites and zeolites such as clinoptilolite, chabazite, phillipsite, mordenite, and erionite which are candidate backfill materials were treated under hydrothermal conditions of 200 and 300°C with a pressure of 30 MPa for 28 days. Results of X-ray diffraction (XRD) analysis revealed that montmorillonites did not seem to alter while vermiculites were partially chlorotized by hydrothermal treatment at 200 and 300°C. Natural zeolites excepting phillipsite did not seem to have transformed at 200°C but transformed to various extents at 300°C. Selective sorption of Cs and Sr decreased to different degrees in all the above minerals excepting two montmorillonites and mordenites after hydrothermal treatment because of the nature and extent of mineral transformation under these hydrothermal conditions as revealed by XRD and Cs and Sr sorption measurements. For example, Cs sorption K_d (ml/g) values decreased from 8100 to 1000 and 4600 to 2800 in erionite and chabazite respectively, while they decreased drastically from 2600 to 90 and 3900 to 70 in vermiculite and phillipsite respectively after hydrothermal treatment at 300°C (table 1). Montmorillonites among clay minerals and mordenites among the zeolites studied here seem to be very resistant to hydrothermal alteration and therefore may be preferred in a backfill <u>on this account</u>. These results point out that the effects of temperature and water pressure on mineral transformations must be experimentally determined before the selection of proper backfill materials is made. A study of mineral alteration under natural conditions of burial diagenesis or metamorphism may give additional clues in the selection of proper backfill materials. While we have examined several candidate materials in silicate rock environments in a preliminary way, the stability of backfill materials in the brine of a salt repository has yet to be so investigated. No detailed studies have appeared.

SORPTION OF IONS BY CLAYS, ZEOLITES, AND THEIR MIXTURES

Any material that is to be used as a backfill should have a maximum useful capacity for sorption and fixation of the specific sets of radwaste ions during and after the hydrothermal period. This presents the clay and zeolite mineralogist with an intriguing challenge. What is the nature of the adsorption equilibria between a clay + zeolite when several ions are present together? Reality is even more complicated; and we need to study such equilibria with both multiple ions and also with multiple adsorbent phases present. In our study along these lines (2), we measured K_d on several clays, zeolites, and oxides for their sorption and fixation properties, individually and in various mixtures, using a simulated waste solution (table 2). These studies show that mixtures of such phases cannot be treated as simply additives; both synergistic and negative interactions are observed. Among those studied mixtures, zeolites such as clinoptilolite plus mordenite or clinoptilolite plus phillipsite are the best for sorption of ions. Phillipsite, however, is very unstable (table 1) and therefore might not be suitable where fixation is important and the material is heated in the repository prior to adsorption.

Backfill materials in a salt repository should have high specific sorption in brine in order to be effective. Zeolites were found to be the most effective for Cs uptake from saturated NaCl solutions as shown in table 3.

THE QUESTION OF "FIXATION" OF IONS

These same backfill materials also serve as adsorbents in waste water clean-up and possibly as precursors to a final waste form. Hence it is of considerable interest to determine how strongly ions which are adsorbed on these phases are actually held on the phase. For example, certain zeolites used in clean-up at TMI would release 100 percent of their Cs to groundwater, while others may not. Furthermore, simple heat treatments could radically alter the fixation of the radionuclides by the adsorbent. Fixation of Cs was measured by extraction in 0.1N KCl because K is a similar ion to Cs and is effective in replacing Cs.

Our study of these phenomena shows that there exists an enormous range in the fixation by rather similar zeolites. Table 4 summarizes these results. It shows, for example, that while analcite has a low exchange capacity, it will fix virtually 94 percent of the Cs by heating to 300°C. On the other hand, the zeolite actually being used in Harrisburg, is one of the poorest for fixation, since even after heating at 800°C almost 89 percent of the Cs washes out. Concentration of the radionuclide is also an important variable. At the maximum loading level of zeolites at TMI, the Cs can be adsorbed on phillipsite and the latter heated to (a maximum of) 1000°C to make an excellent radiophase with nearly 100 percent fixation. This, if incorporated into a concrete matrix, could make an effective waste form having the same range of leachability as glasses.

INTERACTIONS OF WASTE ELEMENTS WITH POTENTIAL BACKFILL MATERIALS UNDER REPOSITORY CONDITIONS

When we first pointed out the absolute necessity for evaluating waste package performance by simulating repository conditions and studying what we called "waste-rock interaction" (8), we used this shorthand expression principally to stress the fact that the simple leach tests in water were of no significance whatsoever in determining the fate of the waste form in a repository environment after container failure. It was essential we argued as we do now, to take the first order approximation (that given the range of volumes of fluid flow projected) that the compositional system which will be brought to near-equilibrium will be waste-container-overpack (= backfill)-rock-fluid (WCORF). Thus while the waste-rock-fluid research studies now in full swing in many institutions, including our own laboratory, are ultimately much more informative regarding the limits of what can happen at the waste package edge, they do not represent the best approximation to reality at all.

Since the volume and mass of the backfill can be at least as large as that of the waste form, and the canister may be a reasonable fraction thereof, it is quite certain that the WCORF system should be studied as a whole. White et al. (10) have indeed launched a major systematic study of several waste forms, several containers and several rock types. If one considers silicate host rocks and corrosion resistant ceramic or metallic canisters, then the ternary systems of greatest interest are certainly those consisting of combinations of various overpacks (= backfills), various waste forms and water. Thus a series of waste-container-overpack interaction studies are obviously of vastly greater significance for waste package evaluation than "leach tests." We have begun in a preliminary way to study ternary waste-overpack-fluid interaction under simulated repository conditions for various waste forms. Thus Cs, Mo and U ions in Cs_2MoO_4 and β-$Cs_2U_2O_7$ phases of spent fuel elements have been shown to react with potential backfill materials to form pollucite, powellite, and uraninite respectively (5, 6). Figure 2 shows the concentration and time dependence of the reaction. Figure 3 shows the reaction of clinoptilolite with sintered ceramic to form analcime containing Cs (9). The reaction of waste elements with backfill barriers will of course change with the nature of groundwater (table 5) if the latter is a concentrated solution. Backfill materials fix the largest percentages of Cs in deionized water by forming pollucite or analcime (figures 2 and 3), but not in bittern brine. However, montmorillonites, vermiculites, and zeolites are good fixers of Cs, both in deionized water and bittern brine. Since montmorillonites and some zeolites are quite stable under repository conditions (table 1) they may be preferred for a backfill.

The various lines of the authors' preliminary research reported herein support the following general theses:

- That compositional tailoring of the backfill (= overpack) materials is without doubt the most cost effective way of improving the waste package performance. Compared to the complexity of remote and high temperature processing required for making glass or ceramics, the normal cold manipulations of composition and emplacement of backfill are of trivial cost.

- That zeolite and clay minerals and their interactions in the mild hydrothermal regime with the principal radionuclide threats in radwaste will provide the data to assess the limits of what the backfill can do to immobilize the radionuclides in new radiophases not present in the original waste form.

- That waste package evaluation will only be meaningfully simulated from geochemical-petrological views when the waste-canister-overpack-rock-fluid interactions have been studied. These kinds of data should shortly eliminate the entire family of "leach tests" as totally meaningless.

ACKNOWLEDGEMENTS

The studies of waste-rock interaction and the waste-canister-rock stability referred to in this work, have been supported by the U.S. Department of Energy through sub-contract E512-3400 with the Office of Nuclear Isolation, Battelle. Overpack research has been supported by the Pennsylvania Research Corporation, The Pennsylvania State University.

REFERENCES

1. Jacobson, A., and R. Pusch, "Deposition of high-level radioactive waste products in bore-holes with buffer substance," KBS Teknish Rapport 03, Stockholm (1977).

2. Komarneni, S., and R. Roy, "Superoverpack: Tailor-made mixtures of zeolites and clays," in Scientific Basis for Nuclear Waste Management, Vol. 2, C. J. Northrup, Jr. (Ed.), pp. 411-418 (1980).

3. Komarneni, S., and R. Roy, "Hydrothermal transformations in candidate overpack materials and their effects on Cs and Sr sorption," Nucl. Tech., 54: pp. 118-122 (1981).

4. Komarneni, S., and R. Roy, "Interactions of Backfill Materials with Cesium in a Bittern Brine Under Repository Conditions," In Preparation for Nucl. Tech., 56: pp. 575-579 (1982).

5. Komarneni, S., and B. E. Scheetz, "Hydrothermal Interactions of Basalts with Cs and Sr of Spent Fuel Elements: Implications to Basalt as a Nuclear Waste Repository," J. Inorg. Nucl. Chem., 43: pp. 1967-1975 (1981).

6. Komarneni, S., and W. B. White, "Hydrothermal Interactions of Clay Minerals and Shales with Cesium Phases from Spent Fuel Elements," Clays & Clay Minerals, 29: pp. 299-308 (1981).

7. Mendel, J. E., J. L. McElroy and A. M. Platt, "High-Level Radioactive Waste Management Research and Development Program at Battelle Pacific," Adv. in Chem. Ser. 153, M. H. Campbell (Ed.), Amer. Chem. Soc., Washington, DC, pp. 106-107 (1976).

8. Roy, D. M., et al., "Borehole Cement and Rock Properties Studies," Annual Progress Report to Office of Nuclear Waste Isolation, Union Carbide Corporation, Oak Ridge, TN 37830, Y/OWI/SUB-4091/6 (1977).

9. Sasaki, N., S. Komarneni, B. E. Scheetz, and R. Roy, "Backfill-Waste Interactions in Repository Simulating Tests," Scientific Basis for Nuclear Waste Management, Vol. 6, S. V. Topp (Ed.), North-Holland, New York, pp. 397-404 (1982).

10. White, W. B., W. P. Freeborn, S. Komarneni, D. S. Minser and G. D. Cox, "Chemical Interactions of Shale Rock, Prototype Waste Forms, and Prototype Canister Metals in a Simulated Waste Repository Environment," Proceedings of the 1980 National Waste Terminal Storage Program Information Meeting, ONWI, Columbus, OH (1980).

Table 1

Effect of Hydrothermal Treatment on Cs Sorption by Candidate Backfill Materials

Sample name, state	Cs sorption, K_d (ml/g)		
	Untreated	200°C, 30 MPa 28 days	300°C, 30 MPa 28 days
Montmorillonites			
Na - montmorillonite, WY	150±20	140±10	170±20
Ca - montmorillonite, TX	190±20	200±20	160±20
Ca - montmorillonite, AZ	590±60	510±50	280±30
Vermiculites			
Poole vermiculite, SC	2600±300	60±10	90±10
Williams vermiculite, SC	2900±300	100±10	60±10
Zeolites			
Phillipsite, CA	3900±400	3200±300	70±10
Chabazite, AZ	4600±500	4800±500	2800±300
Erionite, CA	8100±800	6900±700	1000±100
Erionite, OR	4300±400	---	3600±400
Clinoptilolite, CA	2900±300	3200±300	2600±300
Clinoptilolite, ID	3800±400	3500±400	3200±300
Mordenite, AZ	4600±500	4200±400	4500±500
Mordenite, NV	4100±400	5100±500	4800±500

± Denotes 90% confidence limit on the mean.

Table 2

Sorption* and Fixation* of Ions from Simulated Waste Solution by Individual and Mixtures of Minerals

Sample	Total mg of Cs sorbed/g	% Cs fixed	Total mg of Rb sorbed/g	% Rb fixed	Total mg of Sr sorbed/g	% Sr fixed
Conasauga shale, TN	1.56	23	0.168	45	0.330	0
Montmorillonite, AZ	4.24	33	0.378	21	0.913	0
Vermiculite, SC	4.76	65	0.218	55	1.213	0
Gibbsite, heated	0.00	--	0.000	--	0.675	0
Clinoptilolite, AZ	5.35	81	0.643	77	1.490	26
Clinoptilolite, ID	5.35	82	0.648	82	1.590	75
Mordenite, AZ	5.43	92	0.558	61	0.618	53
Mordenite, NV	5.37	89	0.558	69	0.410	70
Erionite, OR	5.28	89	0.650	63	0.550	7
Erionite, CA	5.51	91	0.648	75	1.346	9
Chabazite, AZ	5.47	88	0.598	68	0.945	0
Phillipsite, CA	5.44	89	0.663	94	0.775	0
Linde, 3A	2.40	38	0.073	0	1.778	87
Linde, 4A	4.27	70	0.323	40	1.783	70
Linde, 5A	5.06	69	0.345	41	1.670	30
Mixtures of Natural Zeolites						
Clinoptilolite, ID + Mordenite, AZ	5.34	85	0.598	63	1.428	80
Clinoptilolite, ID + Mordenite, NV	5.32	83	0.623	66	1.541	80
Clinoptilolite ID + Erionite, OR	5.39	82	0.605	66	1.445	68

Table 2 (Continued)

Mixtures of Natural Zeolites	Total mg of Cs sorbed/g	% Cs fixed	Total mg of Rb sorbed/g	% Rb fixed	Total mg of Sr sorbed/g	% Sr fixed
Clinoptilolite, ID + Erionite, Ca	5.42	82	0.635	67	1.590	68
Clinoptilolite, ID + Chabazite, AZ	5.33	79	0.620	63	1.482	62
Clinoptilolite, ID + Phillipsite, CA	5.34	83	0.655	84	1.532	72
Erionite, CA + Mordenite, AZ	5.39	89	0.605	62	1.242	47
Erionite, CA + Mordenite, NV	5.44	86	0.628	67	1.330	52

* Sorptions of Cs, Rb, and Sr were measured from a simulated waste solution prepared by dissolving PW-4b waste (Komarneni and Roy, 1980). Fixation here is simply the percent remaining in solid after 1 extraction for 24 hours with 15 ml of 0.1N NaCl + 0.1N $CaCl_2$ solution.

Table 3

Cs Sorption by Backfill Materials from Saturated NaCl*

Sample name	Cs sorption K_d (ml/g)
Montmorillonite, WY	6
Montmorillonite, TX	6
Montmorillonite, AZ	12
Vermiculite, Poole, SC	0
Vermiculite, Williams, SC	2
Clinoptilolite, ID	14
Clinoptilolite, CA	18
Phillipsite, AZ	24
Chabazite, AZ	28
Mordenite, AZ	30

* Contained 8 ppm Cs, 4 ppm La, 4 ppm Nd and 2 ppm Sr. Sorption of Sr, La, and Nd could not be detected.

Table 4

Fixation of Cs in Cs-Loaded Zeolites After Heat Treatment as Determined by One 0.1N KCl Extraction

Zeolite	Exchangeable Cs from unheated zeloites with one 0.1N KCl extraction, meq/100 g	Cs Fixation, %		
		350°C	600°C	800°C
Clinoptilolite, ID	143	0.0	0.0	0.0
Mordenite, AZ	114	0.0	0.0	7.9
Chabazite, AZ (similar to Ionsiv IE 95 proposed for TMI)	137	0.0	0.0	11.3
Erionite, CA	131	6.6	11.4	16.8
Phillipsite, CA	171	0.0	85.0	89.9
Phillipsite, NV	197	0.0	69.1	87.2
Analcite, AZ	6	94.0*	99.5	99.8

* Heated to 300°C instead of 350°C.

Table 5

Analyses of Cs in Solution and XRD Characterization of Reaction Products* in Water vs Brine

Backfill Barriers	Total mg of Cs added/ 100 mg sample	Cs uptake in deionized water, %	Pollucite formation by XRD	Cs uptake in bittern brine, %	Pollucite formation by XRD
Shales					
Conasauga	15.2	82.1	V.St¶	6.8	ND
Salona	13.1	55.7	M	11.0	ND
Basalt Phase					
Labradorite	14.8	99.7	V.St	9.7	ND
Clay Minerals					
Illite	17.9	31.7	Sl	12.0	ND
Mica	12.8	22.3	ND	9.6	ND
Montmorillonite, AZ	17.4	44.4	ND	46.7	ND
Vermiculite, Poole	17.4	--	--	93.6	ND
Vermiculite, Williams	17.4	--	--	97.2	ND
Zeolites					
Phillipsite, CA	17.4	--	--	50.0	Tr
Mordenite, AZ	17.4	--	--	39.6	ND
Clinoptilolite, ID	17.4	--	--	30.6	ND

---

* Hydrothermally treated at 200'C/300b for 2 months.

V.St. = Very strong; ND = None Detected; M = Medium; Tr = Trace; Sl = Slight.

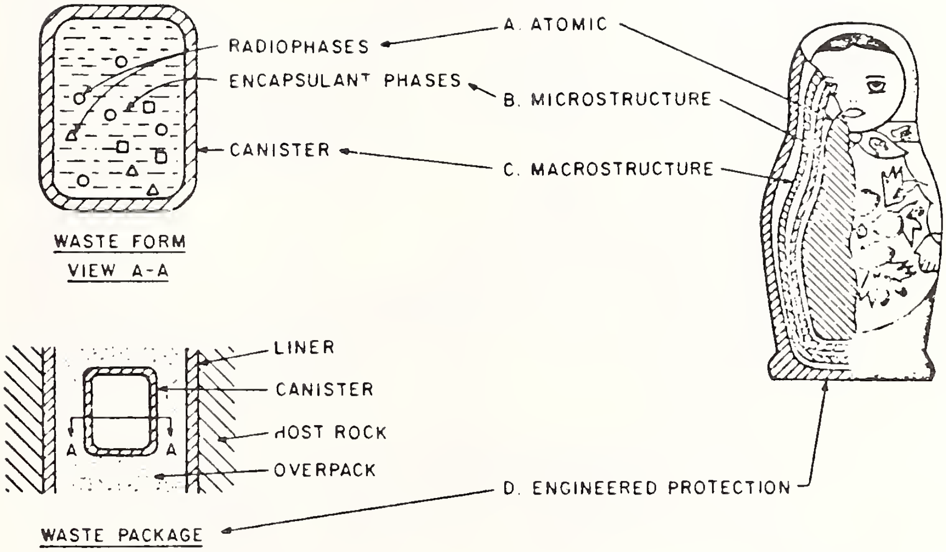


Figure 1. A concept of the waste package

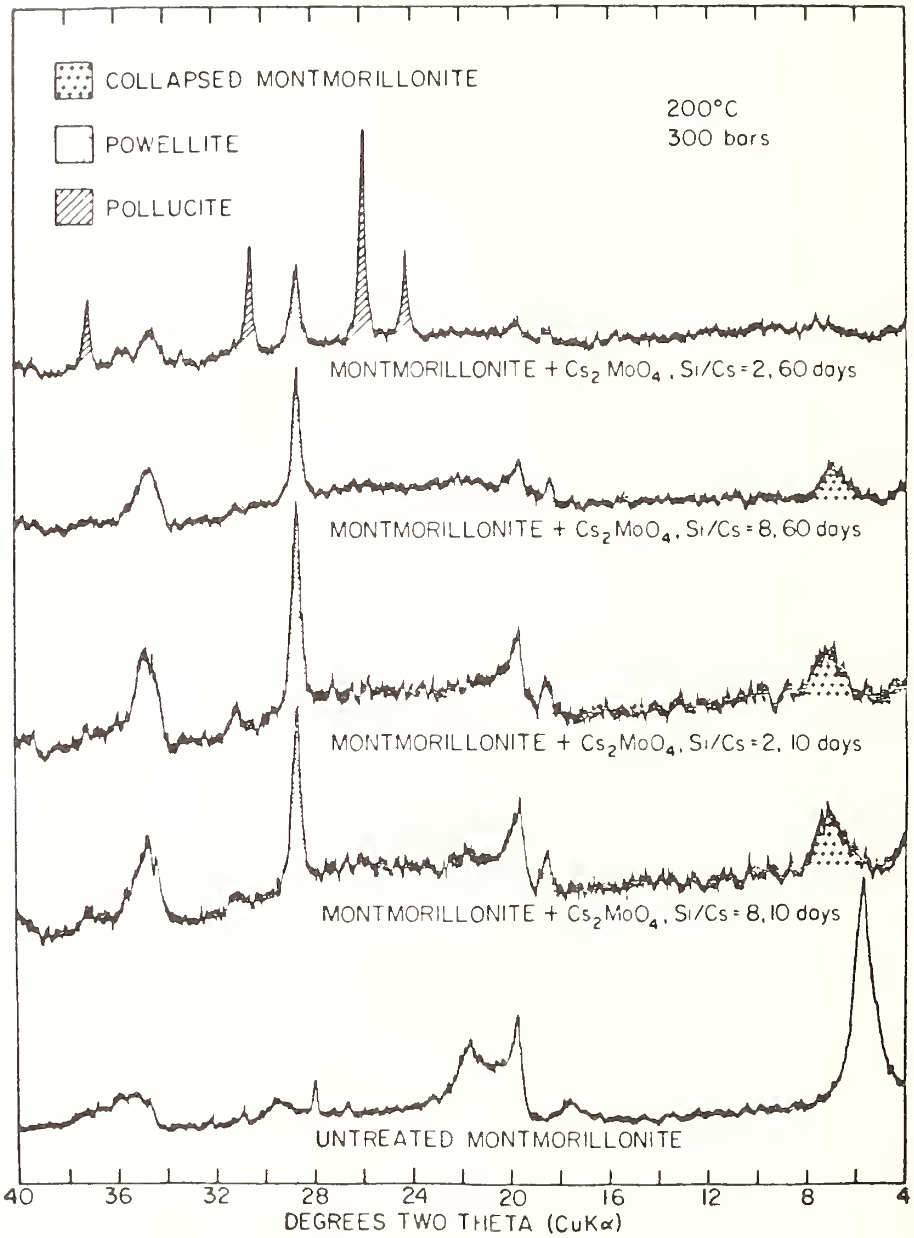


Figure 2. X-ray diffractograms of the products of hydrothermal interactions between Cs_2MoO_4 and Ca-montmorillonite

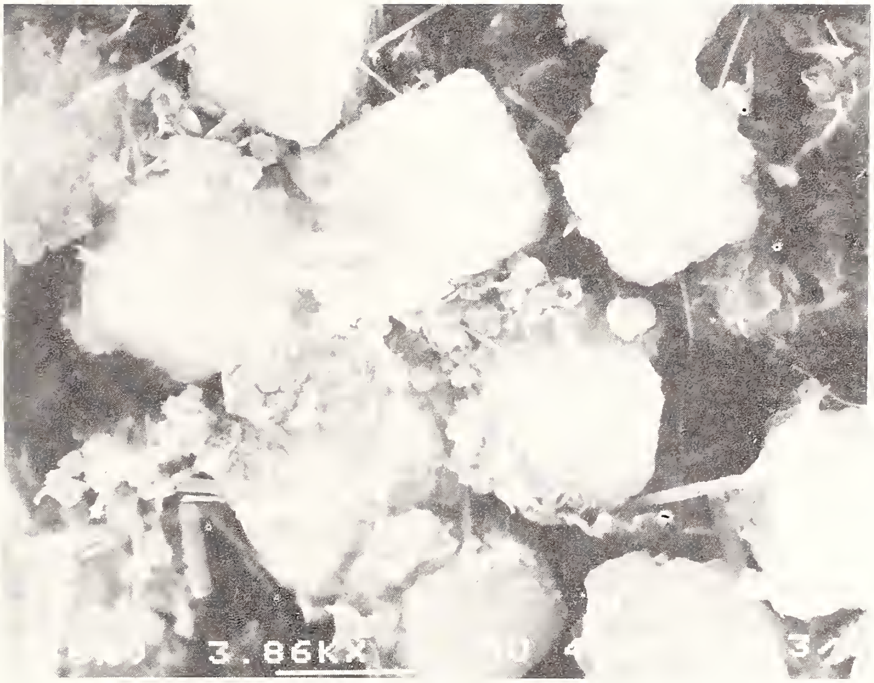


Figure 3. Scanning electron micrograph of analcime (containing Cs) formed from interaction of clinoptilolite with sintered ceramic at 300°C under a confining pressure of 30 MPa for 28 days

BACKFILL BARRIER RESEARCH AND DEVELOPMENT FOR RADIOACTIVE WASTE ISOLATION IN SALT[1]

E. J. Nowak
Sandia National Laboratory[2]
Albuquerque, New Mexico 87185

ABSTRACT

Tailored backfill material emplaced around waste forms and other waste package components can provide a hydrologic barrier to brine flow and a sorptive barrier to radionculide migration. The backfill can also chemically modify brine (e.g., modify Eh and pH) to minimize its corrosivity and to maximize the sorption of radionuclides. Mixtures of bentonite (K_d = ~2000 ml/g for Pu and ~1000 ml/g for Am, permeability = <1 microdarcy, swelling pressure = 18 MPa, all in concentrated brine; thermal conductivity = ~1.0 W/mk for dry bentonite-sand mixtures) with other sorptive materials(mordernite with K_d = ~30 ml/g for Cs, and sodium titanate with K_d = ~130 ml/g for Sr, all in concentrated brine) are promising candidate materials for a repository in salt. Measurements of radionuclide migration rates in backfill mixtures, permeabilities to brine, swelling pressures in brine, thermal conductivities, hydrothermal reactions in brine, and conditions for pre-compaction of backfill solids by isostatic pressing are underway. Among research and development needs are measurements of migration rates of species in leachates from waste forms and of particulates; measurements of radiation effects; engineering assessment of backfill manufacturing processes, emplacement techniques, and in situ physical behavior; and measurements of chemical interactions among backfill components, other waste package components and salt at elevated temperatures in a radiation environment.

---

[1] This work was supported by the U.S. Department of Energy under Contract DE-AC04-76-DP00789.

[2] A U.S. Department of Energy facility.

THE DEVELOPMENT AND TESTING OF WASTE PACKAGE BACKFILL MATERIALS FOR A NUCLEAR WASTE REPOSITORY LOCATED IN BASALT

M. I. Wood

Rockwell Hanford Operations
Richland, Washington 99352

ABSTRACT

A deep repository located in Hanford basalt has been proposed for the disposal of high-level nuclear waste. Backfill materials are required to make up the backfill component of a multiple barrier waste package and to fill the rooms, tunnels, and shafts of the repository sealing system. The major emphasis of this paper is the backfill component of the waste package. However, much of the discussion is applicable to the repository sealing system. The functions, material properties, candidate materials, and testing methods for backfill are discussed. Because of the variety of backfill functions, physical, mechanical, and chemical properties are required of an adequate backfill. Therefore, a multicomponent backfill will probably be needed. The geochemical environment of the basalt site and the physical and chemical reactions that are likely to occur in the near-field environment have been used to derive the essential testing needs.

INTRODUCTION

The Basalt Waste Isolation Project (BWIP) is currently assessing the feasibility of developing the technology for a deep geologic repository for the disposal of high-level nuclear waste in the basalt flows of the Columbia River Plateau. One important aspect of the development of a qualified repository in basalt is the development of optimum backfill materials. Backfill materials are required to make up the backfill component of the waste package and to fill the rooms, tunnels, and shafts excavated prior to the emplacement of the waste. This discussion is focused primarily on the backfill component of the waste package. However, the tests being performed now and those planned in the future for the waste package component are applicable to the repository sealing system as well.

The development of backfill materials is being achieved through the following logical sequence of steps:

1. The functions of a backfill component are defined.
2. The chemical, physical, and mechanical properties required to perform these functions are defined.
3. Candidate materials are chosen which are characterized by these properties.
4. Tests are designed and implemented to select the best backfill materials. Two key elements which are needed to design useful experiments are (a) an understanding of the near-field geochemical repository conditions throughout the service life of the repository, and (b) identification of the chemical and physical properties which will involve the backfill in this environment and may adversely affect its functional capacity.
5. Suitable backfill materials are chosen on the basis of these data and data collection from other National Waste Terminal Storage (NWTS) and international sources.

BACKFILL FUNCTIONS, PROPERTIES, AND CANDIDATE MATERIALS

A fundamental premise of the multiple barrier concept is the development of a barrier system in which one barrier not only promotes the physical and chemical stability of another barrier or set of barriers, but also provides redundant functional ability in the event of the failure of one or more barriers. The current waste package concept designed by the Basalt Waste Isolation Project is given in figure 1. In practical terms, the outer component protects and provides redundant functional support for the inner components. In this case, the backfill component supports and protects the overpack and canister. The backfill has two major functions: (a) to minimize contact between these components and the host environment; and (b) to control the release of radionuclides into the host environment within the recommended guidelines in the event of canister and overpack failure. In table 1, the functions of the backfill are correlated along with the chemical, mechanical, and physical properties of candidate materials which will be used to determine their relative acceptability.

The first function of the backfill can be divided into two parts. First, the backfill component is required to minimize contact between the waste package and groundwater because of the corrosive reaction which will take place, perhaps to a significant degree, over the first 1,000-year period. The most important parameter here is the permeability of the material. Swelling pressure measures the ability of the backfill to fill voids in the space between the waste package and the bedrock. Second, the backfill must act as a shield to protect the canister from the impact of falling rock and undue stress resulting from minor bedrock movement. The important parameters are bearing capacity, plasticity, and shear strength. These properties measure the ability of the backfill to keep the waste package in its emplacement position and to retain its physical integrity. Thermal conductivity is an important parameter since chemical alteration of backfill materials could result if the temperature buildup is excessive within the backfill. A change in mineral phase assemblage could affect both the physical integrity and water-excluding ability of the backfill.

Retardation of radionuclide migration can be accomplished by the backfill via three principal mechanisms: (1) sorption; (2) reaction; and (3) diffusion control. Sorption and reaction capacities are strongly dependent on chemical properties of the backfill (backfill composition, reactivity of backfill materials to specific radionuclides, temperatures). Diffusion controls the rate of radionuclide transport only under certain physical conditions. These conditions are most likely to occur in a dry impermeable backfill or in a saturated backfill where the rate of groundwater flow is essentially zero. Due to the small thickness of the proposed backfill component around the waste package, diffusion controlled transport may prove to be a most effective means of radionuclide retardation. Consequently, the water-excluding properties of backfill components (permeability, swelling pressure, etc.) are significant parameters to consider in terms of radionuclide containment within the backfill.

PRELIMINARY BACKFILL MATERIAL CANDIDATES

An optimum backfill material is likely to be made up of several materials since no single material has been found in the course of a literature search which can satisfy all of the functional requirements listed in table 1. It is unlikely that a satisfactory single component will be found.

Two component types of candidate backfill materials have evolved on the basis of the functional requirements, a major component and a minor component. The major component makes up the bulk of the backfill and can be composed of more than one material. The best major component will satisfy a maximum number of the functional requirements. In addition to the functional properties listed above, two general constraints were applied to make up a preliminary candidate list. First, since backfill will be in direct contact with the geological environment (basalt, secondary minerals, groundwater), it must remain in its functional state in that environment under ambient conditions during the time period of geologic control. Furthermore, the backfill must be compatible with host rock and groundwater during the thermal period when various chemical parameters are disturbed by the introduction of waste into the system (e.g., temperature, pH, oxygen fugacity). The consequences of this constraint are that synthetic materials are eliminated as potential major components. Further preference is given to those natural materials presently found in the basalt environment. Second, potential major component materials must be readily available in quantities sufficient to fill the bulk of the repository volume.

Three potential major component candidates satisfy these two criteria and are presently the subject of extensive investigation. They include: (1) bentonite, (2) crushed basalt, and (3) zeolites. Bentonite has been studied in depth primarily by Swedish scientists and shown to have a variety of useful properties. In particular, the lower permeability and swelling capacity of this clay makes it an attractive material for minimizing contact between the groundwater and the overpack or canister metal. In addition to its water excluding potential, bentonite has a very high sorption capacity relative to other clays and geologic materials. Crushed basalt is an obvious candidate in a repository located in basalt, because of its availability following the excavation of the repository and the need to dispose of this material. Preliminary hydrothermal and sorption tests indicate that altered basalt is highly reactive with uranium, plutonium, americium, cesium, and strontium (2). Zeolites are considered as a potential backfill component because of their high cation exchange capacity. Also, minor amounts of zeolites are found in the fracture mineralization in the Columbia Plateau Basalts.

There is also a need for a minor backfill component which is specifically tailored for transport retardation (either by sorption or reaction) of key long-lived and potentially mobile radionuclides (e.g., ^{129}I and ^{99}Tc). Attention is focused on single phase and the minor backfill component may be composed of several phases.

PHYSICAL AND MECHANICAL TESTING

Perhaps the most critical physical parameter of a backfill component is its permeability. Ideally speaking, if a backfill component existed which would remain perfectly dry, then the inner components of the waste package would remain intact and contain the waste completely for an indefinite period of time. This material probably does not exist. However, if the time required for saturation is as long as several hundred years, the design of a waste package will be affected significantly. Therefore, it is important to examine the sequence of events which lead up to the saturation of a backfill.

The physical state in which groundwater exists in the near-field environment of the waste package depends on the temperature and pressure conditions of the system. These parameters will change with time as the three major periods of repository service life evolve. During the operational period, the waste packages are loaded into boreholes and the rooms above the boreholes will be left open. The backfill volume will be characterized by high temperature

(250° to 300°C) and low pressure (nearly atmospheric). During the thermal period after the repository system is sealed, temperature will decrease slowly and pressure will increase slowly because the room backfill has been emplaced and is slowly saturating and swelling. During the period of geologic control, the temperature eventually will return to the ambient value of 65°C and pressure will rise to its maximum value (100 to 300 bars). Thus, the overall trend is that of decreasing temperature and increasing pressure in the backfill component of the waste package. This trend is consistent with water first being in the vapor phase, then in the vapor and liquid phases, and finally in the liquid phase. Further, it is reasonable to assume that the backfill will be dehydrated when water is in the vapor state, partially saturated when water is present as both liquid and vapor, and fully saturated when water is liquid.

The length of time in which a backfill will remain dehydrated is most sensitive to the pressure increase of the waste package system. Bentonite is probably the only major backfill component which will contain significant amounts of water when it is emplaced (10 to 15 wt%). This interlayer or intergranular water should be the only source of water in the backfill during the operational period because the rock will be pumped dry. Water should also vaporize and escape into the empty room above unless the rate of vapor diffusion through the backfill is slow enough to allow a buildup in effective pore pressure that is sufficient to induce the stability of the liquid phase. Saturation would then begin as soon as groundwater resaturates the host rock and contacts the backfill.

A far more likely scenario is that vapor diffuses easily through the backfill up to the room and the effective pore pressure is not increased. This mechanism may also continue after the room is backfilled and pumping of the repository ceases. The room backfill will be relatively porous and dry, thus presenting avenues of escape for vaporized groundwater. A stable liquid water phase will not exist until the backfill temperature decreases to low enough temperatures (100° to 150°C) where small pressures (1 to 5 bars) are sufficient to promote the stability of liquid water. At this point, a wetting front will be established and the saturation process will begin.

Estimation of the time required to saturate the backfill after water is stable in the liquid phase is a difficult task. Both heat and mass transfer (liquid and vapor) will be occurring simultaneously. Parameters which will affect the saturation process include the temperature of the backfill, the absence or presence of a temperature gradient, the degree of compaction of the backfill, the availability of groundwater from the surrounding fracture system, and the swelling of bentonite as it comes into contact with the groundwater.

If the total time involving dehydration and rehydration of the backfill is on the order of hundreds of years, it is likely that the waste package can be simplified considerably. For example, if corrosion of canister metal through interaction with groundwater is delayed for this period of time, the reduction of oxygen content and tempeature should significantly reduce the corrosion rate. Also, if a metal is used which is thermodynamically stable in the ambient basalt environment (such as copper), the canister is likely to remain physically intact indefinitely. Thus, a waste package consisting of a canister and a backfill may be sufficient to contain the waste within recommended guidelines.

After the backfill is saturated, contact between groundwater and the overpack or canister must be kept to a minimum. This goal is achieved by engineering a backfill which is of low permeability when it is finally saturated. The permeability must be low enough for diffusion to be the rate controlling mechanism of transport. In this manner, the concentration of corrosive agents in the groundwater (oxygen, chlorine, etc.) coming in contact with the metal surfaces are minimized. Also, if the inner components of the waste package are breached, and radionuclides are transported through the backfill, diffusional movement allows the maximum time for precipitation and/or sorption to occur.

Given these considerations, a number of tests must be conducted to determine the behavior of a bentonite-based backfill. They include the following:

1. Determination of Backfill Compaction and Porosity

Prior to experimentation, backfill materials would be compacted and analyzed to determine the porosity of the material. A three-dimensional picture of porosity can be achieved by characterizing thin sections parallel and perpendicular to the direction of compaction. This work would be completed for all types of proposed backfill materials (bentonite, bentonite-basalt, bentonite-basalt-zeolite).

2. Dehydration of Bentonite

Compacted bentonite is dehydrated at 300°C for varying lengths of time. Four factors should be measured: (a) the rate of vapor discharge, (b) pore pressure, (c) the degree of fracturing the steam channeling that may result from vapor discharge, and (d) analysis of the solid phases following discharge. These data will indicate the effects of dehydration on the chemical and physical integrity of bentonite.

3. Rehydration of Bentonite

Compacted bentonite which has been dehydrated is rehydrated over an appropriate temperature range. Simulated Hanford groundwater is used in the test. Data to be collected include (a) the permeability as a function of different temperatures, the absence or presence of a thermal gradient, and the degree of compaction; (b) analyses of the groundwater at the completion of the experiment; and (c) analyses of the bentonite following the experiment. These data will be used to estimate the time required to saturate the backfill, the effect of altered groundwater chemistry on the corrosion rates of the canister or overpack, and the effect of chemical alteration on the sorptive and reactive capacity of the backfill.

4. Field Tests of Backfill Dehydration and Rehydration

On the basis of laboratory data, full scale tests of backfill dehydration and rehydration processes will be done in the field at various temperature and pressure conditions expected to occur during the service life of the repository. The performance of these tests will require development of backfill fabrication and emplacement techniques as well as instrumentation to measure dehydration and rehydration processes. These tests are advantageous because variations in the groundwater backfill interaction can be determined which may occur due to the spatial arrangement of the backfill component of the waste package.

CHEMICAL TESTING - SORPTION AND HYDROTHERMAL TESTING

If the inner components of the waste packages are breached, the backfill component must be capable of reducing rate of radionuclide release to the host rock. This is accomplished through sorption processes or through precipitation of radionuclides in low solubility phases as transport through the backfill occurs. In order to design an accurate and efficient testing program to evaluate the chemical reactivity of backfill materials with radionuclides, two factors must be considered:

1. The tests must simulate the geochemical environment at the repository site. The range of conditions expected to occur in a repository located in the Hanford basalt is summarized in table 2. A reference site groundwater is listed in table 3. The most critical parameters are temperature, groundwater composition, Eh and pH. Changes in temperature affect reaction kinetics and phase solubilities. The pH and Eh values are directly related to the oxygen content of the system which dictates the stable oxidation state of polyvalent radionuclides. This is an important factor for radionuclides such as uranium, which is highly soluble in the hexavalent state (oxidizing environment) and nearly insoluble in the quadrivalent state (reducing environment).

2. A choice of which radionuclides to use in these experiments must be made because an attempt to investigate all of the isotopes in the nuclear waste inventory is expensive, time-consuming, and unnecessary. Simple one-dimensional transport modeling has been used to determine which radionuclides are most likely to reach the biosphere in hazardous amounts (3). The results of this analysis indicate that the choice of which radionuclides are most hazardous is somewhat dependent on the time of release. If the canister is breached in the

first thousand years, then the short-lived fission products, i.e., cesium and strontium, present a hazard. If the canister remains intact for the thousand year period, then these elements no longer exist in hazardous amounts as a result of radioactive decay. The alternate group of radionuclides which are most hazardous include both long-lived cations (uranium, neptium, plutonium, and americium) and long-lived anions (iodine, selenium, and possibly technetium). Because of their long half-lives, these radionuclides are hazardous regardless of their release time. The anionic radionuclides, iodine, and selenium, are given the most immediate priority because few geologic phases are capable of reacting with them. Cesium and strontium are given the lowest priority because experiments completed on the sorptive and reactive interaction between basalt and these elements suggest that they can be adequately retarded with the addition of crushed basalt to the backfill.

Two areas of chemical testing have been defined on the basis of temperature. Backfill sorption studies are being completed in the range of 65° to 100°C. A test matrix has been designed to account for the variety of geochemical conditions discussed above. Potential sorptive and reactive backfill materials and the corresponding radionuclides to be sorbed are listed in table 4. The following elements make up the sorption test plan:

1. All experiments are being run with simulated repository site groundwater (table 3).

Parallel experiments are being completed under oxidizing and reducing conditions. In order to create the reducing environment, hydrazine (N_2H_4) is added as an Eh poising agent.

2. Static tests are currently being conducted at 65°C. The reactants are placed in a polyethylene bottle which is agitated and kept in a heated water bath (65°C). Similar tests will be completed at 90°C. On the basis of static test data, flow-through experiments will be initiated to simulate groundwater flowing through a backfill material surrounding a breached waste package.

3. Isotherms are conducted by doing experiments at several concentrations for each element in order to determine the effects of radionuclide concentration on sorption coefficients.

4. One element and one potential backfill material are tested in one experiment. The experiments are run for up to 60 days and replicated. This experimental procedure is necessary not only to determine the most effective sorptive materials, but also to isolate specific sorptive reactions which will occur in a multi-component backfill and involve many radionuclides. On the basis of single radionuclide-single backfill material tests, multiple radionuclide-single material tests will be completed to determine the degree of competition for sorption sites among various nuclides.

Hydrothermal testing of backfill materials must satisfy two major goals. First, the chemical stability (i.e., solubility) of these materials in the basalt environment must be determined. Second, the ability of these materials to reduce radionuclide release rates (primarily through precipitation of radionuclide-bearing solids) under near-field repository conditions must be determined. As with the sorption experiments, the variety of geochemical conditions that will occur during repository life must be included in the test matrix. The following parameters make up the test matrix:

1. The experimental reactants include (a) groundwater-backfill materials, (b) waste form-groundwater-backfill materials, or (c) groundwater-backfill materials-basalt. The progression of experiments moves from single component systems to multicomponent systems. This sequence of testing enables the investigator to determine which backfill materials are most reactive with key radionuclides. Assuming that the reactions occurring in the single component systems will also occur in the multi-component systems, the data will be used to project the degree of fixation of hazardous radionuclides in the backfill over long periods of time.

2. Temperatures of the experiments are 150°C and 300°C and the pressure is 300 bars. These values represent the upper temperature and pressure limits expected in the near-field repository environment.

3. Parallel experiments will be run under oxidizing and reducing conditions. Experimental methods for controlling Eh at hydrothermal temperatures are currently being developed.

4. Experimental methods for controlling the pH are currently being developed. A pH buffer must be developed in those experiments where a reactant (i.e., basalt) is not present to buffer the solution (a pH value of approximately 9.4 at 65°C and decreasing with increased temperature).

5. Parallel backfill experiments will be conducted with simulated and actual waste forms. If experiments involving simulated wastes are duplicated by experiments with actual wastes, then fewer hot cell experiments need to be done and extra time, experimental difficulty, and expense can be avoided.

6. The experimental apparatus that is being used for the backfill hydrothermal tests is a Dickson-type rocking autoclave. With this pressure vessel, liquids can be periodically withdrawn and analyzed without interrupting the experiment. Continuous sampling at temperature and pressure enables the investigator to determine when steady state conditions are reached. Solubilities of backfill materials and their capability to react with key radionuclides will be determined.

7. As with the sorption experiments, flow-through experiments will be conducted on the basis of the data gathered from the Dickson apparatus and modified to permit flow-through measurements.

Most of the backfill chemical testing completed to data has been done to evaluate basalt as a reactive material capable of precipitating solids. These data indicate that basalt has an excellent potential for sorbing or precipitating stable solids of the radionuclides, cesium, strontium, rubidium, uranium, plutonium, and americium. Thus, the data indicate that basalt is an excellent backfill material and host medium for the disposal of nuclear waste.

SUMMARY

Analysis of the functions of a backfill component within a multiple barrier waste package system indicates that a wide range of physical, mechanical, and chemical properties must characterize an acceptable backfill. Consequently, a variety of tests on several materials must be completed. The types of site-specific tests which need to be designed and completed have been discussed above. Our current data base indicates that an adequate backfill material can be specified for use in a repository located in basalt.

REFERENCES

1. Coons, W. E., Moore, E. L., Smith, M. J., Kaser, J. D., The Functions of an Engineered Barrier System for a Nuclear Waste Repository in Basalt, RHO-BWI-LD-23, Rockwell Hanford Operations, Richland, Washington (January 1980).

2. Smith, M. J., G. J. Anttonen, G. S. Barney, W. E. Coons, F. N. Hodges, R. G. Johnston, J. D. Kaser, R. M. Manabe, S. C. McCarel, E. L. Moore, A. F. Noonan, J. E. O'Rourke, W. W. Schulz, C. L. Taylor, B. J. Wood, and M. I. Wood, Engineered Barrier Development for a Nuclear Waste Repository Located in Basalt, RHO-BWI-ST-7, Rockwell Hanford Operations, Richland, Washington (May 1980).

3. Wood, B. J., Estimation of Waste Package Performance Requirements for a Nuclear Waste Repository in Basalt, RHO-BWI-ST-10, Rockwell Hanford Operations, Richland, Washington (July 1980).

Table 1

Functions and Functional Properties of the Backfill Component (Smith and Others, 1980).

Functions	Isolation of Waste Package From Host Environment		Retardation of Radionuclide Transport to the Host Environment		
	Water Exclusion	Stress and Impact Protection	Reaction	Sorption	Transport Rate Control by Diffusion
Functional Properties	Permeability	Bearing Capacity	Reaction* Capacity	Sorption* Capacity	Permeability
	Swelling Pressure	Plasticity			Swelling Pressure
	Bearing Capacity	Shear Strength			Bearing Capacity
	Plasticity	Thermal Conductivity			Shear Strength
	Thermal Conductivity				Plasticity
					Thermal Conductivity

* Measured by distribution coefficients for radionuclides between liquids and solids.

Table 2

Range of Geochemical Conditions in the Near-Field Repository Environment (at 1,000 m in depth)

	Ambient Conditions	Thermal Period
Temperature	65°C	65°C-250°C (canister)
Pressure	30 MPa (lithostatic) 10 MPa (hydrostatic)	Atmospheric to 30 MPa
Groundwater pH ($\underline{H_4}Si\underline{O_4}$ control)	9.4 - 10.0	5.5 - 9.4
Groundwater Eh (QFM to Ni - NiO control)	-0.3 to -0.5 volts	-0.5 to +0.54

Table 3

Measured Grande Ronde Groundwater Composition (Smith and Others, 1980)

Anionic Concentrations	mg/ℓ	epm*
CO_3^{-2}	30	1.003
HCO_3^-	36	.589
OH^-	3	.182
$H_3SiO_4^-$	137	1.438
Cl^-	148	4.174
SO_4^{-2}	108	2.249
F^-	37	1.947
		TOTAL epm = 11.582

Cation Concentrations	mg/ℓ	epm
Na^+	250	10.875
K^+	1.9	.049
Ca^{+2}	1.3	.065
Mg^{+2}	.04	.033
SiO_2	121	
		TOTAL epm = 11.022

---

*epm = equivalents per million

Table 4

Candidate Backfill Materials for Reducing the Rate of Release of Key Radionuclides in a Basalt Repository

Radionuclide(s)	Potential Backfill Material
I, Se	Bornite: Cu_5FeS_4 Chalcopyrite: $CuFeS_2$ Ultramarine: $Na_8(AlSiO_4)_6S_2$ Graphite: C Copper: Cu
Np	Apatite: $Ca_5(F, Cl, OH)(PO_4)_3$ Azurite: $Cu_3(CO_3)_2(OH)_2$ Basalt:
Tc, U, Pu, Am	Basalt Bentonite

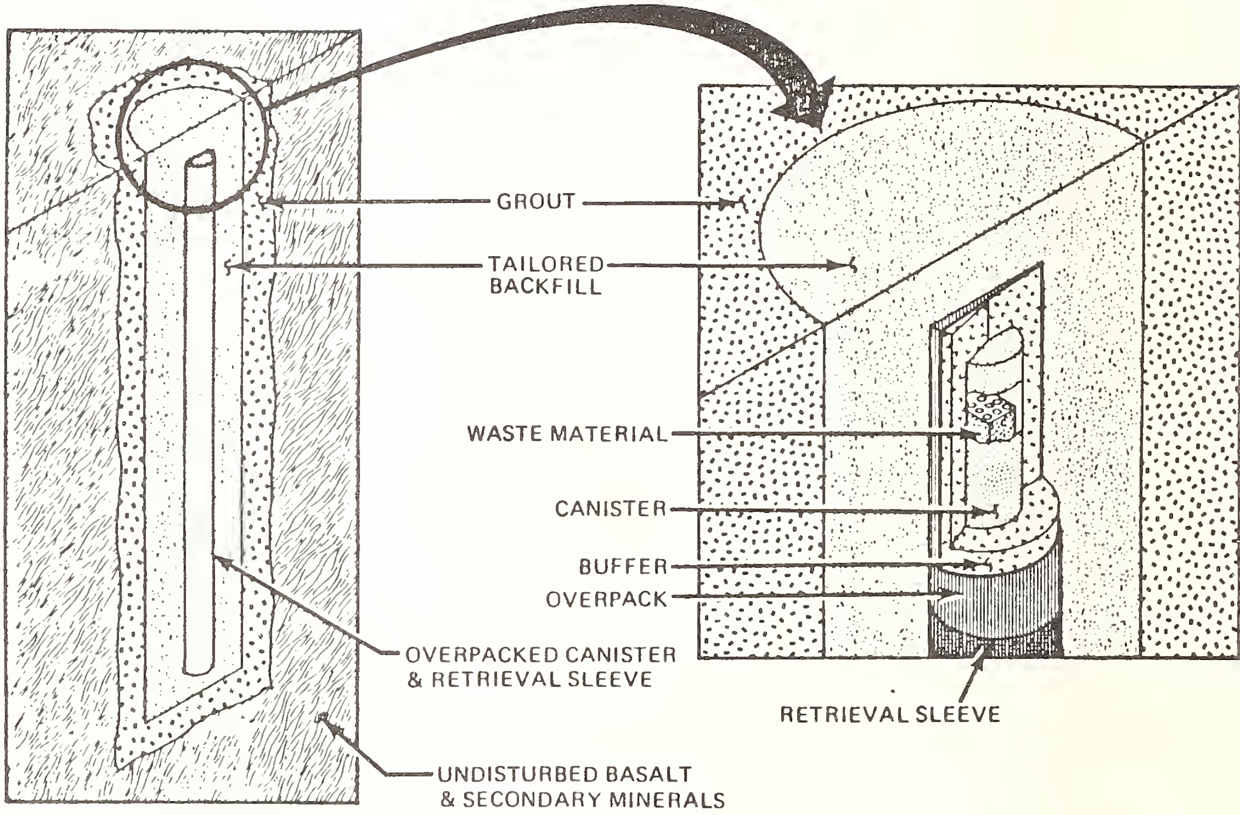


Figure 1. BWIP conceptual waste package in storage hole (Coons and Others, 1980)

CONDITIONS AFFECTING THE BACKFILL IN A WASTE PACKAGE FOR A REPOSITORY IN TUFF

J. K. Johnstone, Sandia National Laboratories, Div. 4537,
Albuquerque, N.M. 87185

E. N. Vine, Los Alamos National Laboratories, Mail Stop 514,
Los Alamos, N.M. 87545

ABSTRACT

We have initiated a program to support waste package development and design for a repository in tuff. Initial efforts have focused on definition of a reference waste package, determination of the repository thermal and geochemical environment and publication of a research plan. The components of the reference waste package are the waste form, canister, overpack and backfill. For commercial high level waste (CHLW), the backfill thickness is 41.3 cm. The reference backfill composition is 70 to 90 percent crushed, welded tuff and 30 to 10 percent Na-montmorillonite clay.

A parametric study has been completed to evaluate the time-dependent thermal environment in a waste package with most emphasis placed on its effect on the backfill. The study included both CHLW and spent fuel. The parameters that were varied included canister power (2.16 and 1.08 kW/canister for CHLW and 0.55 kW/canister for spent fuel), areal power density (100 and 50 kW/acre), backfill thermal conductivity (0.30, 0.75 and 1.1 W/m°C), tuff thermal conductivity (1.55 and 2.40 W/m°C), and time (100 to 1000 years). The emplacement hole diameter was 122 cm or 61 cm. Thermal properties of the rock and waste package were assumed temperature independent. We used a finite element, 2-dimensional, axisymmetric heat conduction code to calculate the radial temperature gradient along the canister midplane. The calculations show that, for a given heat loading, the thermal profiles are most sensitive to the thermal conductivity of the backfill. In the most severe environment (CHLW, 2.16 kW/canister, 100 kW/acre, $k_{backfill}$ = 0.3 W/m°C, and k_{tuff} = 1.55 W/m°C) temperatures in the backfill may exceed 535°C. The Na-montmorillonite and

some components of the crushed tuff in the backfill would be unstable in the potential hydrothermal environment and ground water surrounding the waste package. The conditions imposed by the spent fuel are much less severe and do not appear to affect the stability of the backfill components.

Geochemical studies include investigation of the mineralogy, major and minor trace element rock composition and ground water conditions in a welded tuff horizon. The reference composition for the ground water suggests that it is chemically benign under ambient conditions. The pH is neutral to slightly basic. The Eh remains uncertain. The potassium to sodium weight ratio is about 0.1 which classifies the system as potassium rich. The high potassium content accounts for the potential instability of the Na-montmorillonite at temperatures in excess of 100°C.

A research program has been developed to support the waste package development. The heart of this program is a long-term series of "chemical vector" experiments in which the interactions between the various waste package components is evaluated in a sequence reflecting the flow of water through the waste package. The experiments provide for control of temperature, pressure, radiation, water flow rate, and the number of components in the system.

CRITICAL FACTORS AFFECTING THE USE OF MONTMORILLONITE AS A NUCLEAR WASTE BACKFILL MATERIAL

G. W. Beall, A. G. Lamkin, and K. L. Kelly
Radian Corporation
8500 Shoal Creek Blvd.
Austin, Texas 78758

ABSTRACT

The use of montmorillonite as a component in nuclear waste backfill mixtures was first suggested by Swedish KBS (1) and later in the United States (2). The advantages of montmorillonite in such nuclear waste backfill largely stem from its large cation exchange capacity and from its swelling properties, which result in extremely low permeabilities. This paper details the effects of compaction and common groundwater cations such as calcium, magnesium, potassium, and sodium on the properties of montmorillonite, and what consequences these effects have for nuclear waste disposal.

INTRODUCTION

The suggestion that montmorillonite would make an ideal component in a nuclear waste backfill mixture was made in Sweden (1) and later in the United States (2). The characteristics that make montmorillonite suited for a nuclear waste backfill component are its large cation exchange capacity and its swelling properties. The cation exchange reactions for a large number of radionuclides have been studied (3). It has been shown that in the pH range of interest in most groundwater systems these cation exchange reactions are overshadowed by sorption reactions involving hydrolysis and carbonate complexation. These facts have been incorporated into a backfill mixture that still retains montmorillonite as a component to act as a water barrier, but that contains additional minerals which undergo specific irreversible sorption of radionuclides of importance in nuclear waste (4). The fact that the swelling properties of montmorillonite are critical to its inclusion as a backfill component makes it imperative that a thorough understanding of these properties be obtained.

The majority of work in the past on the physical properties of montmorillonite have centered on the use of the sodium form of montmorillonite. That emphasis has prompted this paper to address some factors that are critical to the proper application of montmorillonite as a water barrier in a backfill material for nuclear waste disposal. The critical factors are cation effects and clay orientation due to montmorillonite morphology upon swelling.

EXPERIMENTAL PROGRAM

Weighed samples of Wyoming montmorillonite obtained from the Source Clay Mineral repository, The University of Missouri at Rolla, were equilibrated with various molar concentrations of Ca, Mg, K, and Na. In all cases the counter ion was chloride. The molar concentration employed in each set of equilibrations were 1, 0.5, 0.25, 0.125, 0.0625, and 0.03125. The samples were equilibrated for four hours with continuous agitation. The samples were then centrifuged for ten minutes in a clinical centrifuge at approximately 3000 rpm. The supernatent was then decanted and the volume determined accurately.

Another equilibration study was conducted using a synthetic groundwater solution containing the following molar concentrations: Ca--.005, Mg--.001, K--.00025, and Na--.004. Again the anion was chloride. This was compared with a solution of only .004 M Na. Weighed samples of the Wyoming montmorillonite were equilibrated with these solutions for 30 minutes with continuous agitation and then centrifuged for 30 minutes. The supernatent was then decanted and the volume determined.

RESULTS AND DISCUSSION

The majority of work on the use of montmorillonite in nuclear waste backfill mixtures has been predicated on the cation exchange and swelling properties of the sodium form of montmorillonite. These studies have not dealt with the effect of various cations common in groundwaters or with how various physical processes affect montmorillonite cation exchange or swelling properties.

The concern with cation effects on montmorillonite stems from the ubiquitous nature of several cations in groundwaters. Table 1 gives the general range of concentrations for major ions in undisturbed groundwater in contact with igneous rocks. It can be seen that Ca^{++}, Mg^{++}, and K^{+} are of the same magnitude of concentration as sodium. In the case of many brines the concentration of Ca^{++}, Mg^{++}, and K^{+} can reach 1 molar range. The fact that Ca^{++} and Mg^{++} are of approximately the same concentration as sodium does not in itself present problems with the cation exchange and swelling properties of montmorillonite. The problem arises from the fact divalent Ca^{++} and Mg^{++} are preferentially taken up by montmorillonite versus sodium. In general the exchanging power of cations on montmorillonite follow Na § K § Ca § Mg § NH_4. This fact again would not in itself cause problems with the properties of montmorillonite. The problem arises from the fact that both the Ca and Mg strongly affect the swelling of montmorillonite. This arises largely from the hydration behavior of the cations involved, Ca and Mg being more highly hydrated than sodium. It has been calculated (5) that the ordered water layers on the Na montmorillonite extend approximately 7.5 Å and for the calcium form 10 Å. It is further calculated that the transition from highly ordered water at the surface out to completely random water is a gradual one extending to 100 Å in the case of the sodium form in contrast to the calcium form, where the transition appears to occur abruptly at about 15 Å. This difference strongly affects the swelling properties of montmorillonite as illustrated in figure 1, where the effect of concentrations of Ca^{++}, Mg^{++}, Na^{+}, and K^{+} upon swelling of the Na form of montmorillonite is presented. It can easily be seen that both Ca and Mg strongly affect swelling of Na montmorillonite. As an example, the swelling volume of the Ca form is a factor of 2.5 less than the Na form at 0.1 M. This appears to be less of a problem with brines since all the curves coalesce at close to one molar. To illustrate the magnitude of the effect on swelling that calcium and magnesium can have at levels common in groundwaters, the sodium form of montmorillonite was equilibrated in a single equilibration, with a synthetic groundwater containing 60 ppm Ca, 25 ppm Mg, 10 ppm K, and 100 ppm Na. This groundwater caused 4 percent less swelling compared with a solution containing only sodium. It must be emphasized that this effect was found after only one equilibration. In a real groundwater situation the effect would be much greater since there would be an infinite supply of equilibration groundwater compared to a finite amount of backfill. These facts then open the possibility that in a real nuclear waste repository, the montmorillonite would initially expand as primarily a Na montmorillonite and then, as it comes to equilibrium with the groundwater, could shrink and actually create fissures through which groundwater migration could be quite rapid. This is what occurs quite frequently to ponds utilized for watering cattle where the ponds were lined with montmorillonite. These ponds with time begin to leak because of the Ca, Mg, and Na equilibration. This effect will most likely be worsened because the groundwater will tend to have higher concentrations of cations than those used in our experiments. This is largely due to the higher temperature around the nuclear waste canisters.

The second factor that has been largely overlooked is the anisotropic nature of montmorillonite. Montmorillonite's morphology can be described as sheet-like. The sheets are quite thin with indistinct and irregular edges. This morphology tends to cause montmorillonite to orient strongly parallel to the C-axis. This orientation can be caused by physical handling such as compacting and extruding or by simple gravity settling. This tendency to orient must be strongly considered in any backfill design to ensure that the swelling properties observed in the laboratory can indeed be obtained in the field.

CONCLUSIONS

The fact that calcium and magnesium have such a pronounced influence upon swelling properties of montmorillonite hints that it would be prudent either to employ the Ca form of montmorillonite in nuclear waste backfill, or to tailor the ratio of Ca, Mg, and Na to fit the groundwater composition to which it will be exposed. If indeed tailoring of montmorillonite occurs, the possibility of adding small amounts of Fe^{++} would eliminate the need to add ferrous-containing minerals to the backfill as an oxygen getter. This tailoring might lend itself to other substitutions that could be of benefit in a backfill.

The second factor of importance is that the tendency for montmorillonite to orient and thus swell anisotropically must be considered in field emplacement of montmorillonite as a backfill material.

REFERENCES

1. Pusch, R. Highly Compacted Bentonite as a Buffer Substance, KBS Technical Report 74, Karnsbranslesaherhet, Stockholm (1978).

2. Allard, B. and G. W. Beall. "Actinide Retaining Backfill Materials in Underground Repositories for Alpha-Contaminated Waste." Proceedings of the IAEA meeting, Vienna (1981), p. 667-677.

3. Serne, R. J. "Waste Isolation Safety Assessment Program," Progress Reports 1977 and 1978.

4. Beall, G. W. and B. Allard. "Chemical Aspects Governing the Choice of Backfill Materials for Nuclear Waste Repository," Nucl. Tech. (in press).

5. Grim, R. E. and F. L. Cuthbert. "The Bonding Action of Clays, I, Clays in Green Molding Sands," Illinois State Geological Survey. Rept. Invest. 12 (1945).

Table 1

General Composition of Granitic Groundwater

Species	Concentration in Nature, mg/L[1]
HCO_3^-	60-400
SiO_2 (total)	5-60
SO_4^{2-}	3-40
Cl^-	5-50
Ca^{2+}	10-60
Mg^{2+}	2-25
K^+	1-10
Na^+	10-100
F^-	0.01-5
HPO_4^{2-}	0.01-0.5
Fe^{2+}	0.5-20
pH	7.2-8.5

---

[1] Probable concentration range for undisturbed deep groundwater in contact with igneous rocks.

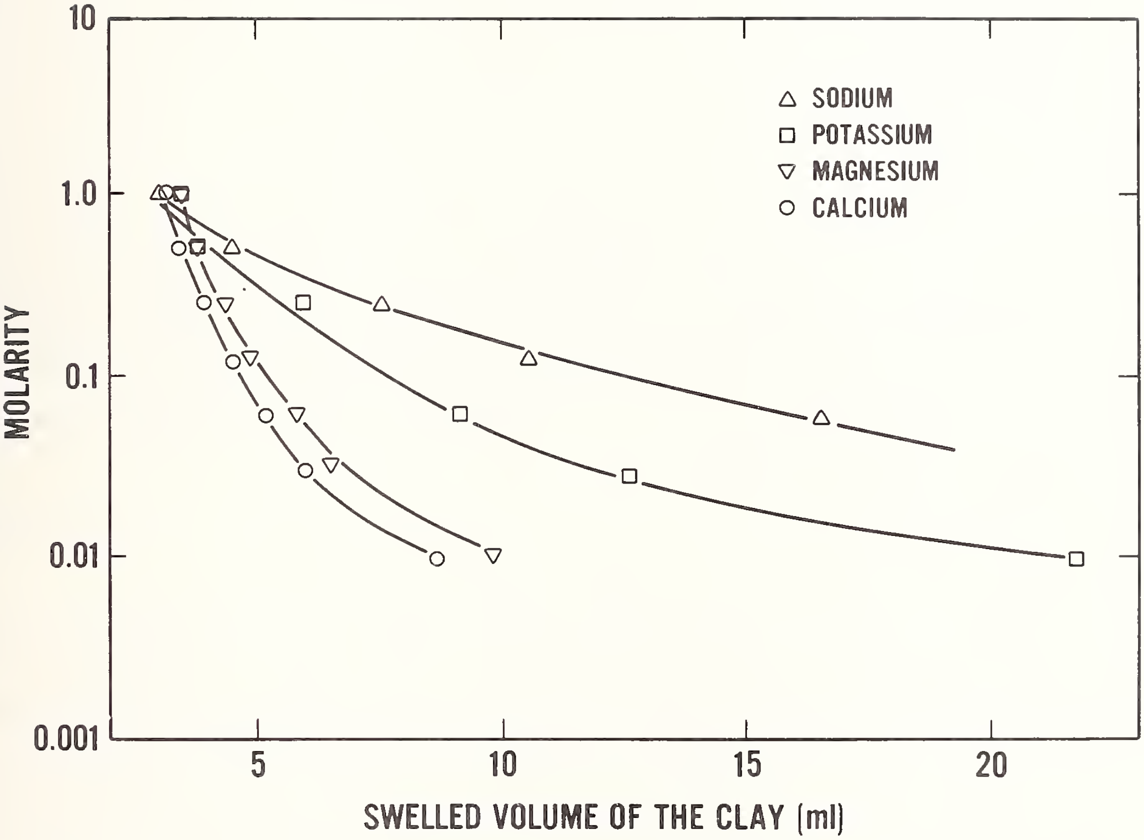


Figure 1. The effect of various cations on the swelling of montmorillonite

SELECTION AND EVALUATION OF BUFFER AND BACKFILL MATERIALS FOR USE IN A NUCLEAR FUEL WASTE DISPOSAL VAULT: THE CANADIAN PROGRAM

G. W. Bird

Fuel Waste Technology Branch
Atomic Energy of Canada Limited
Whiteshell Nuclear Research Establishment
Pinawa, Manitoba, R0E 1L0

ABSTRACT

The selection and evaluation of backfill materials for use in a nuclear fuel waste disposal vault have been divided into two parts based on the physical location of the materials in the vault. Materials in the immediate vicinity of, and in contact with, the waste containers are termed buffer. Backfill includes all materials which fill the remainder of the vault after the waste and buffer have been emplaced.

Potential buffer materials are being evaluated by:

1. Chemical studies which include (a) ion exchange capacities and selectivities for various radionuclides, (b) hydrothermal stability in groundwater at the pressure and temperature conditions in the vault, (c) reactivity to the waste container, waste form and host rock (d) radiolysis effects, (e) assessing the role of organic materials and biological processes and (f) a study of possible additives which could be used to control redox potentials or to trap particular radionuclides.

2. Physical studies which include thermal conductivity and diffusivity, swelling pressures, compressibility and shrinkage, mechanical strength, density, hydraulic conductivity, and diffusivity for water and specific radionuclides.

3. Conceptual engineering studies where equipment and techniques for preparation, emplacement and quality control are being developed.

4. Development of mathematical models to describe the physical and chemical behavior of the buffer within the vault and to provide an analysis of potential pathways and distances of radionuclide transport.

The backfill will occupy a much larger volume than the buffer and will require a different technology for emplacement. These differences lead to additional constraints on material selection and evaluation. As much of the required physical and chemical data will come from the buffer development program, backfill research will be concentrated on engineering aspects such as handling, emplacement and quality control technologies, and on the development of mathematical models for water transport in the vicinity of and within the vault.

The schedule for the buffer and backfill development program calls for preliminary evaluations of the materials to be completed by the end of 1983. From these evaluations, materials will be selected for engineering-scale testing in an underground laboratory.

INTRODUCTION

Safe permanent disposal of radioactive waste requires the isolation of a number of diverse chemical elements from the environment for a long period of time. The Canadian approach to nuclear fuel waste management is disposal in vaults mined deep into crystalline rocks for the Precambrian Shield (4). A vault will consist of a number of mined chambers some 500-1000 meters below the surface, the shafts leading to it and any other underground facilities. These facilities are described in general terms in reports AECL-6415 (2) and AECL-6416 (3). After disposal operations have been completed, the vault must be backfilled and sealed in such a manner as to return the land to a natural condition where no further maintenance or supervision is necessary.

In the post-closure period, the vault and surrounding rocks will become heated from the decay heat of the radioactive waste. The maximum temperature reached will depend on the amount of radioactivity emplaced per unit area. Currently the maximum temperature is assumed to be not more than 150°C (1). The elevated temperature period will last from tens to several thousands of years depending on the waste form.

After closure, the only significant mechanism of radionuclide release which can be identified is that deeply circulating ground water may penetrate to the waste, leach out the radionuclides, and carry them back to the surface. A number of protective barriers, including the waste form itself, its container, the buffer material surrounding the container, the backfill and sealing materials, the massive geological formation and, finally, dilution, dispersion and retention in the geosphere, minimize the probability of radionuclide escape to the biosphere.

The Canadian Nuclear Fuel Waste Management Program has separate research programs on each barrier. This paper describes the research programs on the buffer and backfill barriers.

The term buffer refers to that material which immediately surrounds the waste containers and isolates them from the surrounding rock and backfill (see figure 1). The ideal buffer would have the following chemical and physical properties:

1. Low hydraulic conductivity to groundwater and very low porosity for ionic diffusion.

2. Thermal conductivity equal to or greater than that of the surrounding rocks.

3. Long-term chemical and physical stability at the temperatures, pressures and groundwater conditions of the vault.

4. No unfavorable chemical reactions between the buffer, the waste container, waste or the surrounding rock.

5. Good sorption properties to retard the movement of radionuclides, should the containers be breached.

6. Ability to condition incoming groundwaters so that they are benign with respect to the container materials.

The term backfill refers to that material used to fill any part of the nuclear waste vault other than filled by the waste form or buffer. By this definition (see figure 1), the backfill never comes into direct contact with the waste containers and hence cannot react directly. The remaining physical and chemical specifications for the backfill remain similar to those for buffer. The volume of the backfill is much greater than that of the buffer, and this somewhat restricts the choice of materials on grounds of effective resource utilization and costs.

The buffer and backfill development programs are described separately below.

BUFFER DEVELOPMENT PROGRAM

A number of possible buffer materials have been identified, and a composition of 10 - 20% of bentonite clay in crushed granitic rock and fine aggregate has been suggested in our conceptual design studies (4). Additional materials which will be investigated include kaolinite- and illite-based mixtures and natural clays, which are mixtures of illite, montmorillonite and kaolinite. A short list of potential clay-based buffer materials is being prepared on the basis of existing literature and on our preliminary research results.

Bauxite, carbonaceous shales, fly ash, mine tailings and finely crushed rock such as peridotite and dunite (which will hydrate readily and increase in volume) are also being tested, although these materials are of more interest as backfill.

Two alternative concepts suggested for waste form and buffer emplacement are shown schematically in figure 2. Each concept requires different handling technology, and different physical properties would be expected for each type of emplacement. For these reasons, the buffer development program has been divided into an investigation of: (1) the physical properties, (2) the chemical properties and (3) engineering technology. The logic of the buffer development program is described and shown schematically in figure 3.

Physical Properties of Buffers

The physical properties of greatest concern in selecting a buffer material are thermal conductivity and diffusivity, hydraulic conductivity to groundwater, load bearing capacity, thermal expansivity, thermal stability (5), and shrinkage by drying.

Ideally, the thermal conductivity of the buffer should be similar to or greater than that of the rock; otherwise heat dissipation from the irradiated fuel containers to the surrounding rock may not be adequate and high temperature gradients could result. Maintaining a thermal conductivity equal to or greater than its surrounding rock may not be achievable in practice since thermal conductivity is highly dependent on mineralogy and water content.

Moreover, the thermal gradient within the buffer could induce water and salt movement from the hotter zone to the cooler and vice versa. Each buffer material will be stable with respect to moisture migration for certain ranges of water saturation and in this condition it is described as "thermally stable" (5). Currently an experimental program is underway to test the thermal properties of various buffers. The main thrust of the initial work is to characterize materials in terms of thermal conductivity and moisture migration as a function of temperature, temperature gradient, etc. The moisture content of a thermally stable buffer will depend on water pressure, grain size, hydraulic conductivity, suction pressure of the buffer, density and groundwater chemistry. The overall buffer program must address all these parameters and assess some of the interactive effects.

The load bearing capacity of any potential buffer must be adequate to support the mass of the containers so that they do not sink through the buffer and make direct contact with the rocks. At the same time the buffer material should not be brittle or susceptible to shrinkage cracking by water loss. The bentonite-based buffers recommended in the Swedish KBS-concepts (6, 7, 8) appear to meet these physical requirements, although considerable research is required to confirm and supplement the existing data and develop engineering-scale techniques for handling and emplacement.

Thermal expansion and swelling pressures, generated as the buffer becomes heated and takes up water, are also of concern. The total pressure from thermal expansion and swelling must not exceed the crushing strength of the containers. The current design for the waste container requires it to withstand crushing at the hydrostatic head of water for the vault depth with a safety factor of two (9). If pressures are generated in the buffer or backfill which exceed this value, the waste containers could be deformed and premature failure could occur. One of the research goals of the buffer and backfilling program is to ensure that such a high pressure transient does not occur.

The rate and mechanism of water, salt, radionuclide and oxidant transport through the buffer to/from the waste containers are two of the most important parameters in buffer selection as they control both the physical and chemical behavior of the system by contributing to both heat and mass transport. Empirical and theoretical relationships have been suggested for calculating mass transport rates in soils at room temperature and below, at atmospheric pressure (10, 11, 12). An experimental program is planned to test the usefulness of these relationships at higher temperatures and pressures, and for temperature gradient conditions. Salt and water migration will be measured for a range of buffer densitities, water chemistries, pressures, temperatures and temperature gradients in order to develop empirical or theoretical models which can be used to simulate underground disposal conditions. This program will be integrated with the buffer-chemistry research program and will provide experimental input to the near-field mass transport modelling (see figures 3 and 4).

Chemical Properties of Buffers

The chemical parameters which are of concern in choosing a buffer composition are: (a) total ion exchange capacity, (b) hydrothermal stability under the temperature and pressure conditions in the vault, (c) rates and types of chemical reactions between the buffer, groundwater, waste container, waste form and host rock, (d) the effects of the radiation field on the chemistry, (e) potential for organic and biological reactions to alter the chemistry (f) the use of buffer additives for redox control or selective sorption of radionuclides. For the buffer-chemistry studies, other research groups will provide data on the chemistry of the groundwater and how this chemistry changes as the water reacts at elevated temperatures with the rock surrounding the disposal area (13, 14).

Ion exchange capacity of a number of potential buffer materials is being studied and work is being carried out on diffusion of non-reactive solutes through buffers. The results of these programs should give us a preliminary estimate of the rate of movement of solutes through the buffer and will be supplemented by elevated temperature and pressure experiments. The data will supplement the results obtained by Nowak (10) and Neretnieks (11) and will be useful for transport modelling.

The hydrothermal stability of the buffer in groundwater at elevated temperatures will be studied for various groundwater compositions. The information existing to 1979 has been summarized by Weaver (15) who notes that clay-mineral stabilities are highly dependent on composition of the interstitial water. With unfavorable water compositions, hydrothermal alteration can begin at temperatures as low as 40°C for smectite-type clays. Hydrothermal reactions of the buffer are of most concern if they lead to increased hydraulic conductivity and increased mass transport of radionuclides. Hydrothermal reaction between the buffer, waste container and/or the waste form must not lead to degradation of the waste and enhanced release of the radionuclides.

An assessment of the effect of radiolysis is included in the study of buffer/waste form/container/water interactions. Some preliminary studies have been reported on radiolysis effects (16) and further work is underway, but it is unclear whether the experimental conditions are relevant to the conditions of disposal. Currently there is little evidence to suggest that radiolysis effects will be significant.

Another aspect of the buffer chemistry which is being investigated is the role of trace amounts of organic material which are present in practically all clay deposits. This material could prove detrimental through the effects of biological activity. Some organic materials can act as nutrients for bacteria, and the bacteriological reactions can alter the pH and redox conditions in the groundwater. This could accelerate container corrosion or increase reaction rates between various buffer components. In addition, some organic molecules can complex a variety of radionuclide anions and cations by increasing their solubility and hence transportation rates in groundwater (17).

In addition to using natural materials for buffers, it may be possible to add components which would create desirable chemical conditions. For example, it is suggested in the KBS review (5, 6) that 0.5 percent ferrophosphate (vivianite) be added to the buffer as an oxygen getter. Other suggestions have included adding materials which could trap radionuclide anions (18). In evaluating these proposals, it is necessary to demonstrate that not only do the chemicals function as predicted but also that no unfavorable reactions result. For example, if ferrophosphate is used, we must show that it will not serve as a nutrient for biological activity or buffer the pH of the water-backfill system at unfavorable values, e.g., highly alkaline or highly acidic. In addition, the material must not result in complexing and enhanced transport of the radionuclides should the waste containers be breached. Our research on these topics is continuing.

Engineering Aspects of Buffering

The two irradiated fuel disposal concepts under consideration are illustrated in figure 2. Each concept puts different restraints on the engineering and emplacement of the buffer. In the first concept (figure 2a), the waste containers are placed on a layer of compacted buffer covering the disposal room floor. Additional buffer is spread over and around the containers, using radiation-shielded equipment, and compacted. The remaining free volume of the disposal room would then be backfilled.

In the second concept (figure 2b), the radioactive waste containers are placed in holes drilled into the floor of the disposal room. Two methods have been suggested for emplacing the buffer around these waste containers. In one method the container is placed on a pad of buffer at the bottom of the hole, and powdered buffer is poured around the container and compacted by vibration. An alternative method for buffer emplacement in boreholes has been proposed in the Swedish KBS-project (8). They propose to compress blocks and rings of 90 percent bentonite - 10 percent water to a density of 2.1 - 2.2 Mg/m^3, and place these blocks and rings around the waste containers.

Each waste emplacement concept has a number of advantages and disadvantages. For example, borehole disposal is particularly appealing from the point of view of radiation safety. Moreover, a highly compressed buffer such as bentonite has the advantage of creating a zone with very low hydraulic conductivity (8) as the buffer will swell by taking up water with which it comes in contact. Swelling pressure built up should lead to the extrusion of some bentonite into any pores and fractures in the rock, further inhibiting access of water. However, additional rock mechanics research effort beyond that required for the backfilled drift may be necessary to establish the stability of the boreholes and room floors.

If laboratory testing shows a buffer material and engineering concept to be suitable, a number of practical aspects must be addressed. These include development of methods of material preparation, handling, and quality control on a suitable scale of operation, development of emplacement techniques for the proposed disposal option, and inspection and testing of the emplacement method. Some engineering feasibility studies are planned including a review of material and transportation costs, ease of material handling, storage and quality control. These are not particularly difficult engineering operations and hence are not priority items in the research program. However, the results must be available in order to plan the testing program for the underground research laboratory proposed for construction in the Lac du Bonnet batholith near the Whiteshell Nuclear Research Establishment. This program, which will be described in more detail by Simmons and Soonawala (19), is scheduled to begin around 1985/86.

Mathematical Modelling

Mathematical models are being developed for both buffer and backfill. The models are designed to describe the physical and chemical behavior of the components within the vault. The chemical and physical models are being developed separately.

a. Physical Modelling

The physical modelling has three objectives: (1) to assess the significance of each component of the vault in terms of mass transport of radionuclides from the vault, (2) to assist in developing materials performance specifications and (3) to assist with developing environmental assessment models for the entire vault.

To develop a mass transport model for each component requires knowledge of:

1. The vault thermal-mechanical response, including short-term and steady-state temperature and stress distributions.

2. The rate of water transport into and out of the vault.

3. The major pathways taken by water moving into, through and out of the vault.

4. The mechanism of water movement (e.g., by diffusive or advective flow) within each vault compartment.

Sketches of three geometries we are trying to model are shown in figure 5. The mathematical analysis has been developed for a two-dimensional diffusion model for the borehole emplacement concept (see figure 5a and reference 20). The analysis will be developed initially for a saturated buffer, and will later be extended to account for the unsaturated-flow regime which would be present during vault flooding. As data become available on the diffusion coefficients for various solutes in the groundwater, mass transport of these will be incorporated into the model.

During the later stages of the modelling work, it will be necessary to supplement the diffusion model to include any advective component of mass transport in the regions where advection dominates the flow pattern. To do this, it will be necessary to have better estimates than are currently available of the hydraulic conductivities of the buffer, backfill, host rock and damaged zones surrounding the vault (figure 5b and c). Preliminary estimates of the hydraulic conductivities of potential buffers and backfills will be available from this program, and we are relying on the hydrogeology and rock mechanics research to supply the information on the host rock and the damaged zone. The program schedules require that a preliminary mass transport model be available for the vault by the end of 1983.

b. Geochemical Modelling

Both thermodynamic (equilibrium) and kinetic data must be incorporated into the geochemical modelling for the vault.

In the chemical treatment, the vault could be divided into a series of compartments. The components of a particular compartment could be computed to come to a thermodynamic or kinetic "equilibrium". Water of "equilibrium" composition could then be transported by advection or diffusion, using the physical mass transport model described in the last paragraph, to another compartment, and a computation of a new "equilibrium" composition could be carried out. Using this procedure to transport water through a number of vault compartments would give a time-dependent chemical transport model.

The buffer/backfill development program will provide chemical equilibrium and/or kinetic data, mass transport mechanisms, pathways and rates in support of geochemical modelling. In some cases, sub-models may be developed for individual vault components.

BACKFILL DEVELOPMENT

Much of the physical and chemical characterization of probably backfill components is being carried out through the buffer development program, and backfill research will concentrate on engineering and mathematical modelling. Supply and quality control of the backfill material will be more difficult because of the large volume requirements. The materials will have to be mined, crushed, dried and transported to the repository site; crushed rock or aggregate will be added and the mixture homogenized and tested before removal to the backfilling site. The type of equipment and techniques required for backfilling will depend on whether the area being backfilled is a horizontal room, a drift or a vertical shaft.

Room and Tunnel Backfilling

Several aspects of backfilling emplacement will require research and development once the actual backfill composition has been selected. Emplacement techniques and quality control will probably be the most important considerations. For example, if fine-grained material is used as a part of the backfill, then some water may be required to minimize dust hazard and to ensure good compaction. It will be necessary to know the optimum percentage saturation and how to control this percentage within the required range during emplacement. If the backfill can be emplaced with a suitable water content, it may be feasible to utilize the swelling properties of materials such as bentonite to ensure that the backfill penetrates voids and small fractures in the rock.

Control of porosity and hydraulic conductivity during emplacement will be important as these factors could control the rate of water penetration to the disposal zone. Moreover, if the backfill is emplaced wet and if the hydraulic conductivity of the surrounding rock is low, the volumetric expansion of the trapped water during the heating period following emplacement may control the pressure regime. Because the irradiated-fuel waste containers are designed to withstand a pressure of 10 MPa (9), care must be taken to ensure that pressure does not exceed this value.

Regardless of the method backfilling, we will need to be able to predict the physical response of the backfilling system and of the vault. A mathematical model of vault behavior will have to be developed and be available prior to full-scale testing of backfill emplacement techniques in the URL. This model can then be used to compare predictions and observations in a carefully instrumented full-scale backfilling operation in an appropriate mined room (19).

Shaft Backfilling

Shaft backfilling will differ from room and tunnel backfilling in several significant ways. A different emplacement technology can be developed as the shaft will be accessible to machinery at all levels and the work will be done on a horizontal surface with adequate headroom at all times. As the shafts will not be close to the waste disposal rooms radiation hazards will not be present and temperatures will not rise significantly above ambient. This means that there will be no thermal restrictions on the choice of materials. Ease of compaction, long-term settling properties, load bearing capacity and hydraulic conductivity of the compacted mass will be the most important parameters. Low hydraulic conductivity will be required, not only through the backfill itself but also along the contact between the backfill and shaft liner, or the surrounding rock if the liner has been removed. If it is not possible to obtain a sufficiently low hydraulic conductivity along this interface, separate seals may be required in some sections of the shaft.

Emplacement of the shaft backfill will require sufficient compaction to prevent long-term settling which could leave voids in the shaft.

SUMMARY

The Canadian nuclear fuel waste management research program on buffer and backfill development has four principal objectives:

1. To develop materials and technology for backfilling and sealing a waste vault.

2. To develop and execute a research program to provide experimental data and theoretical support for objective 1.

3. To develop detailed mathematical models of buffer, backfill and sealing barriers and use these models to assess the significance of each barrier.

4. To contribute to the development of a mathematical model for the long-term safety assessment of the entire disposal vault.

The research program has been divided into evaluation of the physical, chemical and engineering properties and mathematical modelling. Logic diagrams for the programs are shown in figures 3 and 4. Initially the work is concentrating on buffer development because the buffer will control the source terms for any radionuclide transport from the waste. The studies described in this paper will lead into a large-scale testing program in an underground research laboratory.

REFERENCES

1. Boulton, J., (Editor) "Management of Radioactive Fuel Wastes: The Canadian Disposal Program", Atomic Energy of Canada Limited Report, AECL-6314 (1978).

2. Acres Consulting Services Ltd. and Associates, "A Disposal Centre for Irradiated Nuclear Waste: Conceptual Design Study", Atomic Energy of Canada Limited Report, AECL-6415 (1980).

3. Acres Consulting Services Ltd. and Associates, "A Disposal Centre for Immobilized Nuclear Waste: Conceptual Design Study", Atomic Energy of Canada Limited Report, AECL-6416 (1980).

4. Acres Consulting Services Limited, "Irradiated Nuclear Fuel and Immobilized Waste Vaults: BAckfilling System", Atomic Energy of Canada Limited Technical Record, TR-66 (1980).

5. Radhakrishna, H.S., "Heat Flow and Moisture Migration in Cable Backfills", Ontario Hydro Research Quarterly 20, 10 (1968).

6. Kärnbränslesäkerhet, "Handling and Final Storage of Unreprocessed Spent Nuclear Fuel, Volume II, Technical", Stockholm (1979).

7. Kärnbränslesäkerhet, "Handling of Spent Nuclear Fuel and Final Storage of Vitrified High Level Reprocessing Waste", Stockholm (1977).

8. Pusch, H., "Highly Compacted Sodium Bentonite for Isolating Rock-Deposited Radioactive Waste Products", Nuclear Technology 45, 153 (1979).

9. Cameron, D.J., "Fuel Isolation Research for the Canadian Nuclear Fuel Waste Management Program", Atomic Energy of Canada Limited Report, AECL-6834 (1982).

10. Nowak, E.J., "The Backfill Barrier as a Component in a Multiple Barrier Nuclear Waste Isolation System", Sandia National Laboratories Publication, SAND 79-1109 (1980).

11. Neretnieks, I., "Transport of Oxidants and Radionuclides Through a Clay Barrier", KBS-Teknisk Rapport 79 (1978).

12. Taylor, S.A. and Cary, J.W., "Linear Equations for the Simultaneous Flow of Energy and Matter in a Continuous Soil System", Proceedings of Soil Science Society of America, 28 (1964).

13. Betcher, R.N. and Pearson, R., "Hydrogeological Research for the Canadian Nuclear Fuel Waste Management Program", Atomic Energy of Canada Limited Technical Record, TR-151 (1982).

14. Sargent, F.P., "Applied Chemistry and Geochemistry Research for the Canadian Nuclear Fuel Waste Management Program", Atomic Energy of Canada Limited Technical Record TR-149 (1982).

15. Weaver, Charles E., "Geothermal Alteration of Clay Minerals and Shales: Diagensis", Office of Nuclear Waste Isolation Technical Record, ONWI-21 (1979).

16. Allard, B., Kipatsi, H., Rydberg, J., "Radiolysis Effects in Packing Material", KBS-Teknisk Rapport 56 (1977), (AECL miscellaneous translation 464).

17. Mean, J.L. and Hastings, D.W., "Status Report on the Importance of Natural Organic Compounds in Groundwater as Radionuclide-Mobilizing Agents", Office of Nuclear Waste Isolation Technical Report, ONWI-84 (1979).

18. Bird, G.W., "Geochemistry of Radioactive Waste Disposal", Geoscience Canada 6, 199 (1979).

19. Simmons, G.R. and Soonawala, N.M., (Editor) "Underground Research Laboratory: Underground Experimental Program", Atomic Energy of Canada Limited Technical Record, TR-153 (1982).

20. Cheung, S., Bird, G.W., and So, C.B., "Diffusion Modeling of the Borehole Emplacement Concept for a Nuclear Fuel Waste Disposal Vault," Proceedings of the NEA Workshop on Near Field Phenomena in Geological Repositories for Radioactive Waste, NEW/OECP, Paris, pp. 375-388 (1981).

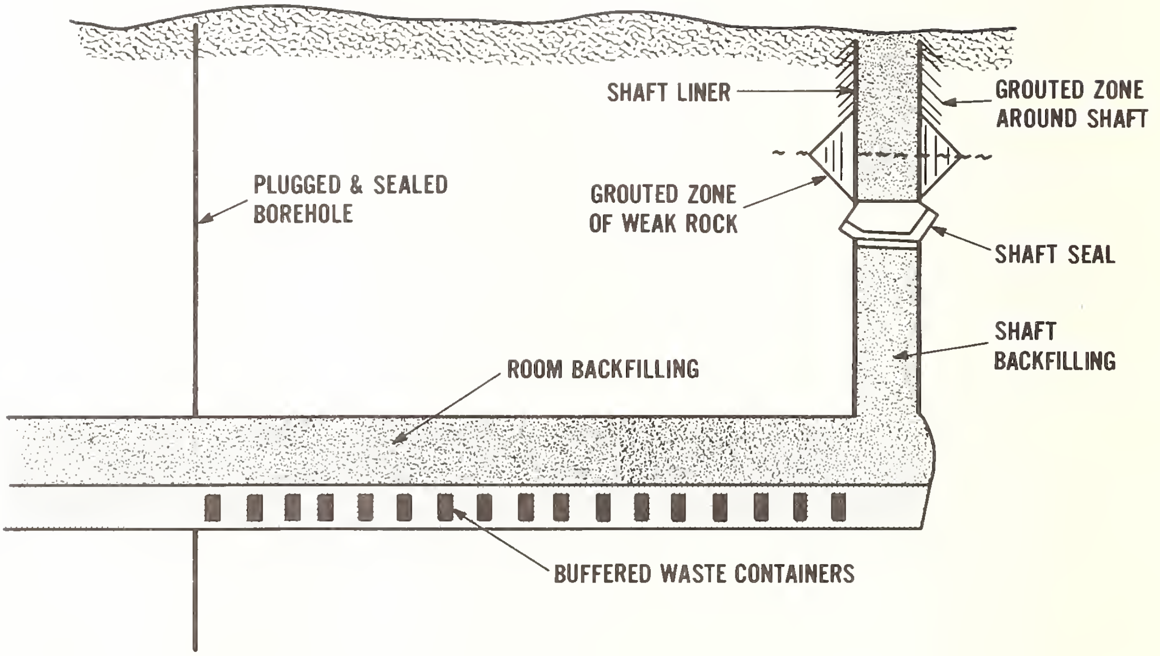


Figure 1. Illustration of components of a backfilled nuclear fuel waste disposal vault (not to scale). See figure 2 for illustrations of two possible waste container/buffer/backfill layouts

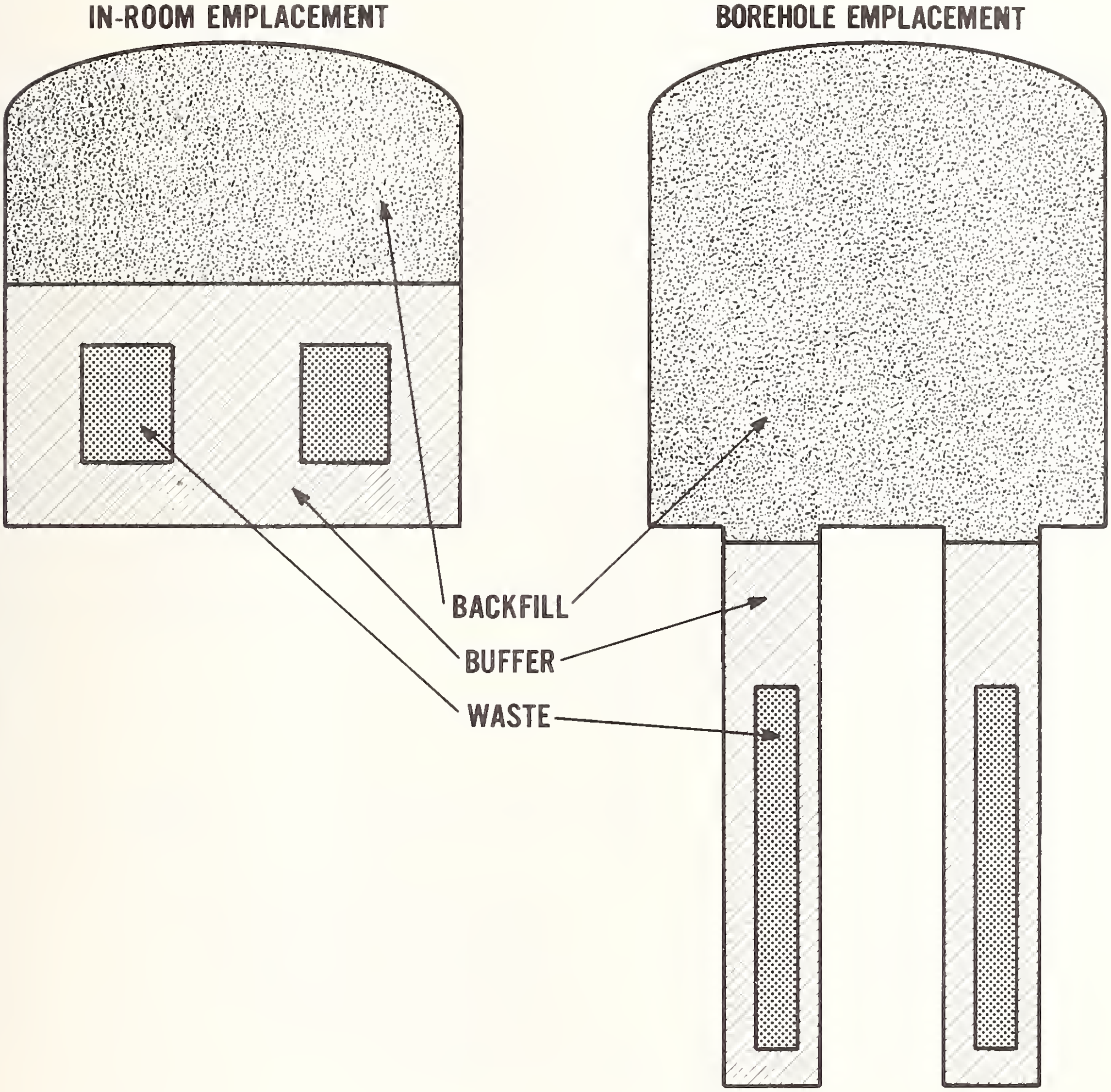


Figure 2. Schematic illustration of the two disposal concepts (not to scale)

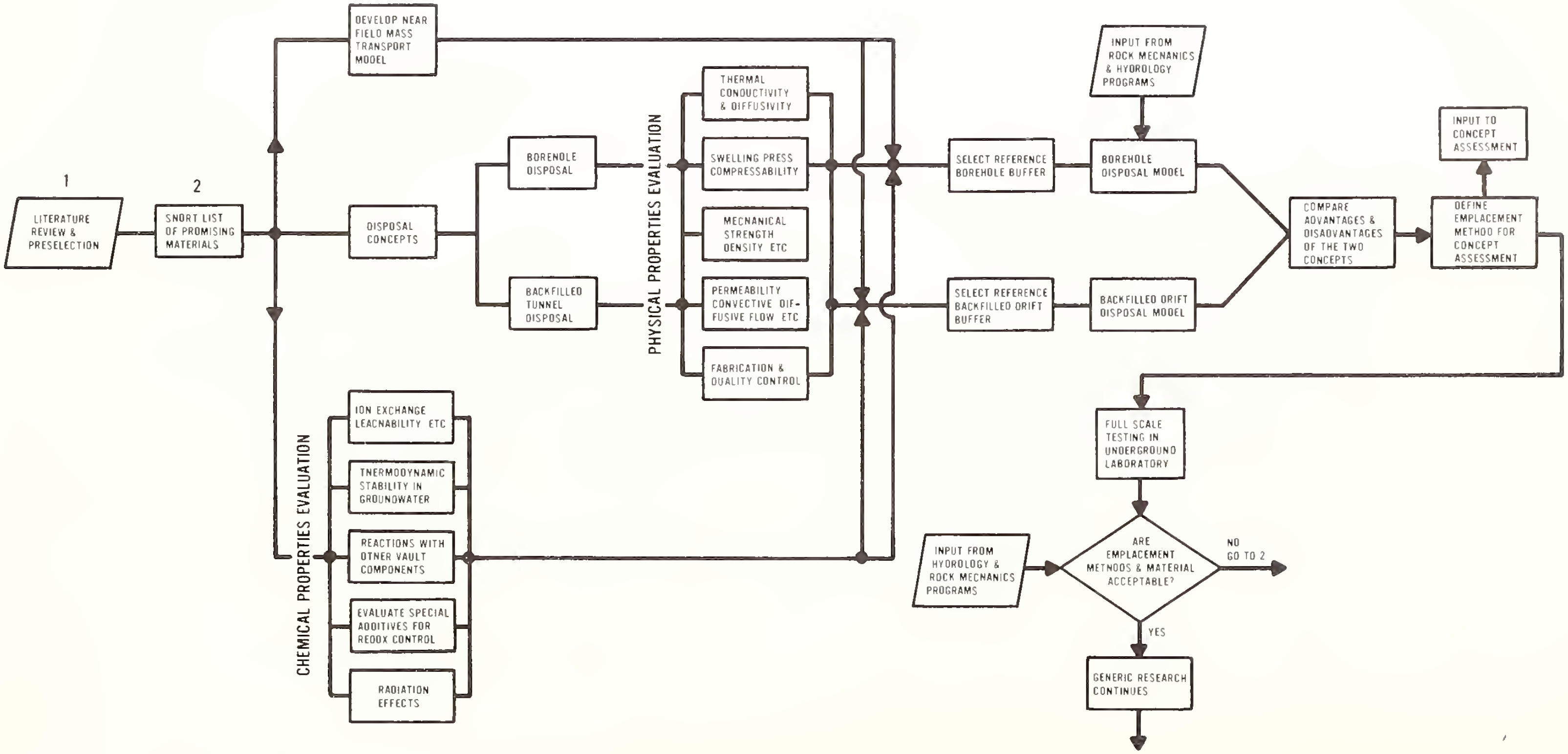


Figure 3. Logic diagram for the buffer development program

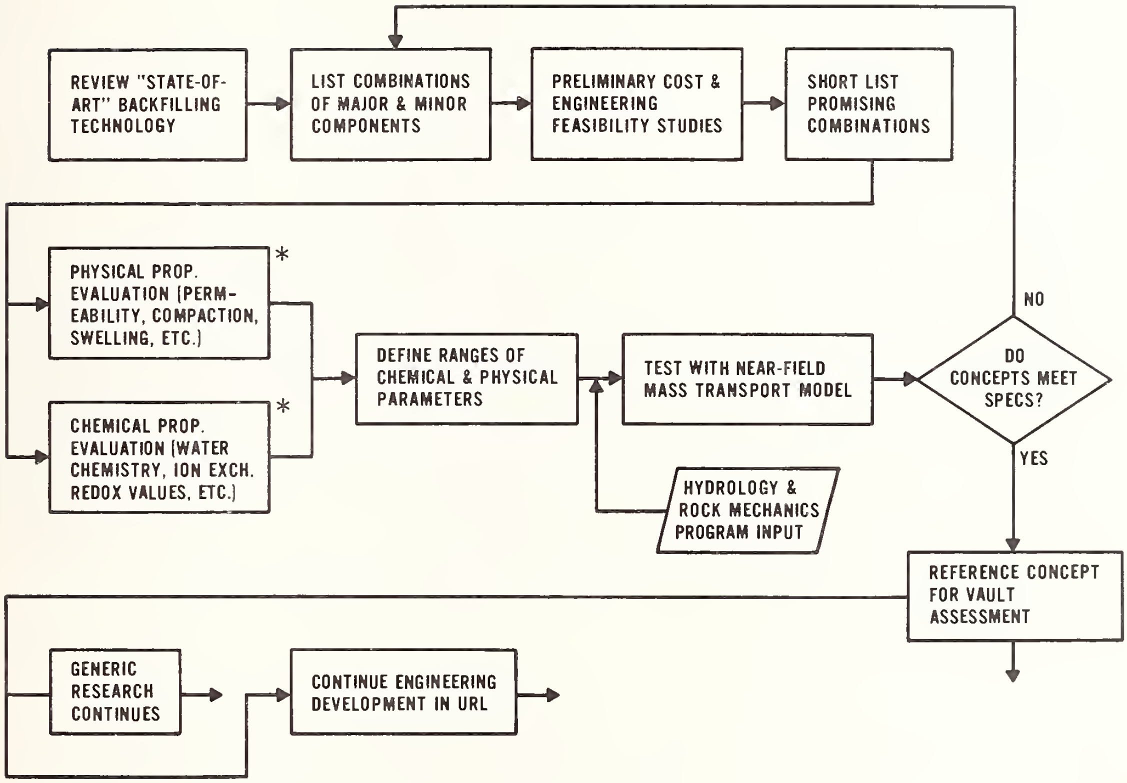


Figure 4. Logic diagram for the backfill development program

* Refers to activities which are part of buffer development program (see figure 3)

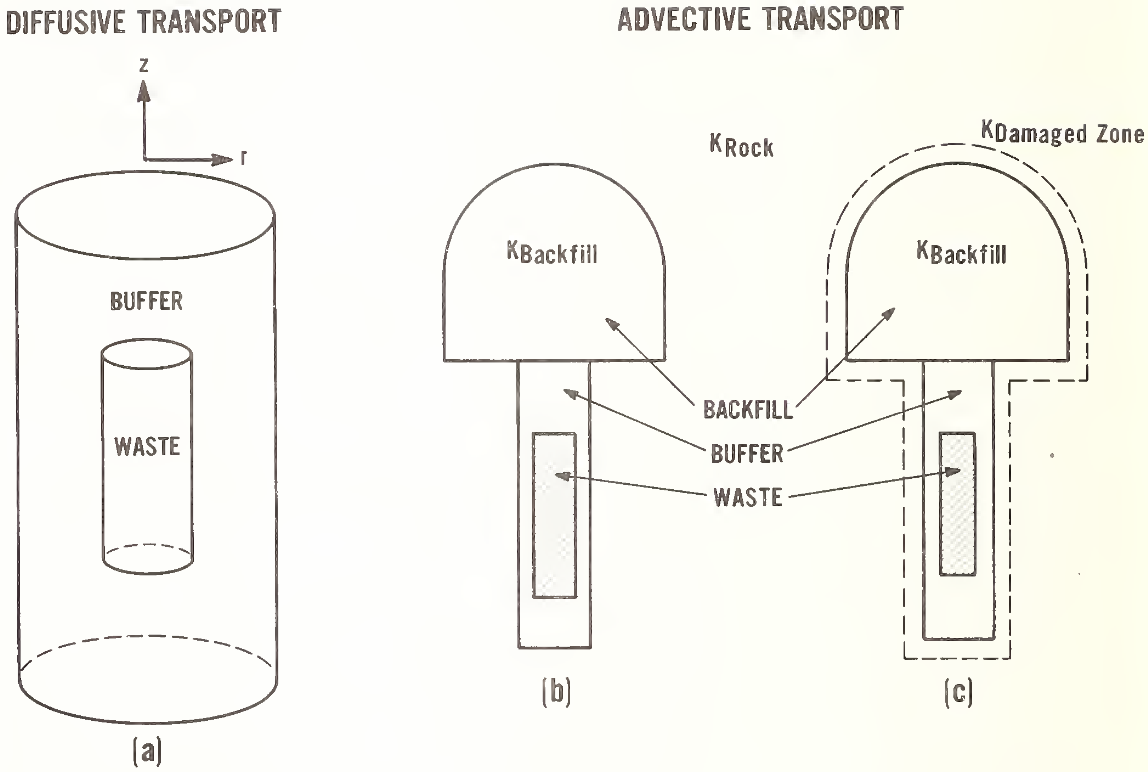


Figure 5. Illustration of the geometry of waste, buffer and backfill emplacement concepts being modelled. In (A) all mass transport is by diffusion and flux gradients can be calculated for the vertical (Z) and radial (R) directions. In (B) and (C) both diffusion and advection are considered. The relative contribution of diffusion advection can be calculated by varying the hydraulic conductivities of the buffer, backfill, host rock, and the thickness and hydraulic conductivity of the damaged zone which exists around the room and borehole

EVALUATION OF THERMAL PROPERTIES OF BUFFER MATERIALS FOR A DEEP UNDERGROUND NUCLEAR WASTE DISPOSAL VAULT

H. S. Radhakrishna
Research Division
Ontario Hydro,
Toronto, Canada

ABSTRACT

Thermal properties of four types of sand/clay mixtures were investigated to evaluate their suitability for buffer material. Tests were done on mixtures of sand/clay over a range of mix proportions, moisture content and inter-particle density and temperature, using a transient heat probe method. Sealbond (illite) and kaolin exhibited better thermal-conducting properties than the swelling clays, namely bentonite and zeolite. Graded silica and crushed granite with about 15 to 25 percent clay added to it appeared to produce a workable material with good compactability, strength and thermal properties. Tests for evaluating the temperature and hydrostatic pressure on the permeability, strength and volume change characteristics of the materials are underway.

INTRODUCTION

Safe and permanent disposal of radioactive waste requires isolation of a number of diverse chemical elements from the environment for a long time. The Canadian approach to nuclear waste management is disposal in vaults mined deep into crystalline rocks of the Precambrian shield (1). This underground disposal vault will consist of a number of chambers, some 1000 meters below the surface, interconnected by a system of tunnels and access shafts as described in the Conceptual Design Reports by Acres Consulting Services Ltd. (2, 3). In the Canadian concept the nuclear waste, packaged in canisters, will be emplaced in boreholes drilled from the floor of the chamber about 4.7 meters deep, as shown in figure 1. The annular space between the host rock and the canister will be filled with a suitable material called the 'buffer'. The repository vault itself is filled with a material called 'backfill'. Thus the primary difference between the buffer and the backfill is in the fact that the buffer is in direct contact with the canister while the backfill is not.

An alternative concept to the borehole emplacement is the room emplacement in which the canisters will be embedded in a buffer mass within the rooms also shown in figure 1.

After disposal operations have been completed, the design criteria requires backfilling and sealing of the entire vault in such a manner as to isolate any harmful chemical elements that may be released from the waste either by accidental or natural breakdown of the canister.

In the post-closure period, the only plausible failure mechanism which can be identified is the prospect that the deeply circulating ground water may penetrate to the waste, leach out radionuclides and carry them back to the surface. As a defense mechanism against such a transport, a series of engineered barriers are proposed in the vault design concept. These are the waste form itself, the canister, the buffer, backfills and the seals or plugs for the mined shafts and boreholes. These, in consort with the geological barrier, should perform the job of isolating the waste from the human environment.

The buffer and backfill, as engineered barriers, are required to perform the following functions: buffer the chemistry of ground water to enhance the life of the canister; capture and retard the migration of radionuclides if they are released from the waste form; provide mechanical support to the canister and also absorb any excessive stresses induced by movements of the rock; and dissipate the heat generated by the waste form, in the process of its decay, to the surrounding rock mass.

The physical and chemical attributes that the buffer material should possess in order to perform the above mentioned functions effectively over long periods of time are discussed in detail by Bird (4). A research program to develop buffer materials has been planned (5).

The three phases in the buffer/backfill development program are: (i) preliminary selection and evaluation of materials, (ii) research into their physical and chemical properties, (iii) engineering development.

The study described in this paper deals with the heat transport aspects of the buffer/backfill materials. The objectives of this study are to evaluate the thermal properties of the candidate materials and to assess the heat transport mechanism and the associated moisture migration phenomenon in the buffer/backfill media. The results presented in here will supplement the results from other studies in the buffer development program to enable a selection of appropriate materials and installation techniques.

CANDIDATE MATERIALS

For reasons of longevity and durability, geological materials such as rock flours and clays are the most favored candidates for buffer/backfill material. Swedish programs have investigated the bentonite-based materials for buffer, backfill and other seals for the deep underground disposal vault. Laboratory-scale experiments by Pusch (6) indicated several positive attributes of highly compacted bentonite, namely its low permeability, self-sealing ability by swelling upon wetting and good ion exchange capacity.

Some of the disadvantages of using bentonite are its low thermal conductivity, high swelling potential and chemical instability at temperatures of 100°C and above. In the Swedish conceptual design, the maximum temperature in the buffer mass is limited to 100°C, however, in the Canadian reference design, the temperature at the canister surface could exceed 100°C. The limitations on maximum permissible temperatures in the buffer mass and host rock (for reasons of thermal spalling) will obviously limit the amount of radioactive waste that can be emplaced per unit floor area.

Research programs in the United States are currently investigating a range of materials (including cement, chemical resins and geological products) for various vault seals. The chemical behavior of clays and crushed rock under the anticipated vault environment is being evaluated (7). Specially formulated backfills designed to adsorb actinides are suggested by Bell and Allard (8). Ground basalt and salt beds are also being examined for the backfill material (9).

THERMAL PROPERTIES OF BUFFER/BACKFILL MATERIALS

Since the decay process of the emplaced radioactive waste generates heat, the buffer backfill and host rock surrounding the canister will act as both heat sink and heat transfer media. The heat stored in the buffer will be relatively insignificant in comparison to that in the backfill and host rock because of its small volume. However, the buffer/backfill will be the only channel for heat flow between the canister and the host rock. A buffer and backfill with good thermal conductivity would prevent excessive temperature build up in the canisters and would also help distribute the heat more uniformly in the rock mass.

Since the buffer and backfill substance is likely to be a particulate material placed in a moist condition, its effective thermal conductivity will be a function of parameters such as composition, thermal properties of the components and their geometrical arrangements. Typical thermal conductivities of component materials in a soil-like medium are summarized in table 1.

From this table it is apparent that air has the lowest thermal conductivity of all the components in a soil, water has a thermal conductivity about 25 times that of air, and solids have thermal conductivities varying from one to 15 times that of water. Because of this wide variation in the thermal conductivities of the component materials, the effective thermal conductivity of the medium is dependent on the volumetric proportions of each component and their arrangement.

Heat transport through soil media has been a subject of investigation for more than a decade. The basic quantitative physics involved in the prediction of the effective thermal conductivity of granular material is contained in the fundamental theoretical work of Maxwell (10), which was extended and modified by DeVries (11). Attempts have also been made by a number of investigators, notably Kersten (12), Knutsson (13) and Jackson (14), to calculate the thermal conductivity of soil-like materials, with different degrees of success. Work carried out by these investigators has resulted in a better understanding of the factors affecting the heat transport qualities of soil. Some of the highlights of the current understanding on this subject that are pertinent to the evaluation of thermal properties of the candidate buffers and backfills are summarized below:

1. At normal temperatures the convective heat flux in soil media is negligible for pore spaces smaller than 0.15 mm. However, the radiative flux is a function of the cube of the mean ambient temperature and, therefore, it may not be negligible at very high temperatures (15).

2. The effective thermal conductivity increases with the size of particles; thus coarse sands have higher thermal conductivities than silts and clays (16).

3. The thermal contacts or bridges created by the liquid film at the solid particle interfaces are the principal contributors to the significant dependence of the effective thermal conductivity of soil to its liquid (or water) content (17).

4. Effective thermal conductivity of the composite soils will be greatest when the continuous matrix is the component with the highest thermal conductivity, and volume fractions of the discrete, lower thermal conductivity components are lowest. To increase the thermal conductivity of sand-like materials it is necessary to couple adjacent particles with a binder whose thermal conductivity is high with respect to that of air. This is most effectively done by using a fluid binder with a heat-conducting filler which remains adsorbed to the particles even under high thermal gradients (14).

5. In practical terms the effective thermal conductivity of granular fill material can be adequately improved by selecting quartz-based solid particles as the major component with a clay binder such as kaolin or calcium carbonate (limestone dust) in proportion of eight to ten percent by weight, and compacting to a high degree of interparticle density (19).

In summary no analytical models are available that can predict the effective thermal conductivity of a wide range of moisture content and temperature for the composite materials including clays. As a result, experimental techniques to measure the thermal conductivity of the candidate buffers have been employed for this study.

EXPERIMENTAL TECHNIQUES

Thermal conductivity of solids and particulate materials is determined by either steady-state heat flow measurements or transient heat dissipation measurements. The steady-state methods have the disadvantages of producing errors in measurements due to moisture migration in the direction of thermal gradients in moist soils and creating convection currents in saturated soils. Furthermore, these steady-state experiments are time consuming.

Transient heat dissipation methods are generally free of the above disadvantages and the measurement techniques are relatively simple and fast. Thus the transient heat dissipating method is favored for routine measurement of the thermal conductivity of particulate materials. In the transient heat dissipating method, which was used in this study, a slender probe simulating a line heat source is installed into the test sample and the temperature rise of the probe under a constant heat input is monitored over a period of 15 minutes. The slope of the slight line portion of the temperature vs log time plot is used for calculating the thermal conductivity of the material. Thermal conductivity, k, of the test material is calculated as:

$$k = \frac{2.303\ W \log_{10} \left(\frac{t_2}{E_1}\right)}{L(T_2 - T_1)}$$

where W = input thermal power for the probe in watts

L = length of the probe (m)

T_1 and T_2 = temperatures of the probe (°C) at times t_1 and t_2 respectively

A microprocessor-based instrument was used to control the constant rate of heat input to the sample, to collect the temperature data at specified time steps and to fit a straight line to the data points by using the least-squares technique. Figure 2 illustrates the logic diagram for this thermal property analyzer.

MATERIALS TESTED

The buffer materials tested consisted of mixtures of coarse-grained and fine-grained materials in different proportions. The coarse-grained materials were:

- uniform graded Wedron silica sand
- graded silica sand
- crushed granite

The fine grained materials (clays) used:

- Blackhills bentonite
- air-floated Georgia kaolin from Harrison and Crossfield (Can) Ltd, Montreal
- Sealbond (air-floated shale) from Domtar Inc, Toronto
- zeolite from Occidental Minerals, Wyoming

The materials were mixed in different proportions to form the test specimens. The physical properties of these materials are summarized in table 2. The chemical composition of these materials is summarized in table 3.

The silica sand used in this study was procured from local suppliers and was blended to produce a particle size gradation similar to the one used in Swedish studies (20). The crushed rock was produced from the granite of the Lac du Bonnet batholith near Pinawa, Manitoba. This rock outcrop is considered to be similar in geochemical composition to the rock at depth. Results of detailed chemical analyses of this rock are described in reference (21).

The following mixes were tested:

1. Wedron sand + Avonlee bentonite (WSAB)
2. Graded silica + Blackhills bentonite (GSBB)
3. Graded silica + sealbond (GSSB)
4. Graded silica + kaolin (GSK)
5. Graded silica + zeolite (GSZ)
6. Crushed granite + Blackhills bentonite (CGBB)
7. Crushed granite + sealbond (CGSB)
8. Crushed granite + kaolin (CGK)

Clay proportions in the above mixes were varied from 15% to 50%. The grain size distribution curves for individual components are shown in figures 3 and 4.

SAMPLE PREPARATION AND TESTING

The sand/clay mixtures were blended dry in proper proportions by dry weight and then mixed with water in a Hobart mixer to produce a uniform and homogeneous material.

Several methods of mixing and different periods of curing after mixing were tried to select a procedure could produce homogenous material and consistency in test results. Mixtures with clay proportions greater than 25% by weight, especially those with bentonite, were generally difficult to blend. They also produced the same degree of inhomogenity in the moisture content.

The mixed materials were compacted into standard Proctor moulds (102 mm diameter by 115 mm high) using standard Proctor compaction energy conforming to the ASTM procedure (D-1557).

THERMAL CONDUCTIVITY OF CANDIDATE MATERIALS

Thermal conductivity measurements were made on each of the Proctor-compacted samples, both in its as-packed condition and over-dried (at 105°C) condition. The results of these measurements are summarized in table 4. For mixes 2, 3 and 4 the thermal conductivity as a function of moisture content at a constant dry density was established by making thermal conductivity measurements on large samples (200 mm by 350 mm) subjected to stage drying. The results of these tests are presented in figures 5 and 6. Thermal conductivity measurements were made on bentonite and sealbond based mixes at elevated temperature (100°C) to examine the temperature effects on thermal properties of candidate materials.

The following observations can be made from the test data:

1. Use of graded silica instead of uniform sand improves both the compaction and the thermal conductivity of the mix materials. However, the crushed granite mixes produce slightly lower thermal conductivity values than the graded silica mixes.

2. Sample density and thermal conductivity decreases significantly with the increase in clay content. This effect is more pronounced in the case of bentonite based mixes (figure 7).

3. The thermal conductivity of the bentonite based mixes varied from 1.0 W/(°C m) to 3.0 W/(°C m) depending on the percentage of bentonite and the type of sand used. The lowest thermal conductivity values occurred in the dry state of materials which ranged from 0.5 W (°C m) to 1.0 W/(°C m).

4. Of the four sand/clay mixtures tested, the mixes containing 15% kolin or 15% seal bond had the best thermal conductivities. These were in the range of 1.5 W/(°C m) in totally dry state to 4.0 W/(°C m) in moist state.

5. Zeolite based mixes had somewhat better thermal conductivities than the bentonite based mixes but the samples appeared to be somewhat porous due to the large percentage of silt size particles in zeolite.

6. A marked reduction in thermal conductivity occurred upon drying for all of the mixes. The thermal conductivity appeared to be far more dependent on the soil moisture than the ambient temperatures of up to 100°C (figure 8).

7. For all of the mixes, there appeared to be a critical moisture level below which a marked change in the thermal conductivity would occur, and for the mixtures with 15 percent clay the critical moisture content was about 2 percent. In earlier studies of backfills for underground cables, it has been demonstrated that the critical moisture content at which a sharp change in thermal conductivity occurs also corresponds to the condition of highest moisture migration in vapor phase and thermal instability (22). Moisture migration experiments to be performed next will examine the potential for thermal instability in the candidate mixtures.

CONCLUSIONS

Buffer/backfill materials made up of sand/clay mixtures will have adequate heat transport qualities with sealbond based mix having the best thermal properties of the four sand/clay mixes tested. None of the mixes will, in dry state, be as conductive as the host rock. Good compaction and moisture control will be necessary to achieve the desired thermal properties. Preliminary tests suggest that a high ambient temperature does not alter the thermal properties significantly. The high degree of moisture dependency of the heat conduction properties of the buffer/backfill should be considered in modelling the thermal transients in the repository vault. A preliminary thermal analysis with a thermal conductivity value of 0.5 W/°C m for buffer should give the upper limit for canister temperature.

REFERENCES

1. J. Boulton (Editor), "Management of Radioactive Fuel Wastes" The Canadian Disposal Program", Atomic Energy of Canada Limited Report, AECL-6314 (1978).

2. Acres Consulting Services Ltd. and Associates, "A Disposal Centre for Irradiated Nuclear Fuel" Conceptual Design Study", Atomic Energy of Canada Limited Report, AECL-6415 (1979).

3. Acres Consulting Services Ltd. and Associates, "A Disposal Centre for Immobilized Nuclear Waste" Conceptual Design Study", Atomic Energy of Canada Limited Report, AECL-6416 (1979).

4. G. W. Bird, "Possible Buffer Materials for Use in a Nuclear Waste Vault", Atomic Energy of Canada Technical Record, TR-72 (1979).

5. G. W. Bird, "Vault Sealing for the Canadian Nuclear Fuel Waste Management Program", Atomic Energy of Canada Limited Technical Record, TR-145 (in preparation).

6. R. Pusch, "Highly Compacted Sodium Bentonite for Isolating Rock-Deposited Radioactive Waste Products", Nuclear Technology 45, 153 (1979).

7. E. J. Nowak, "Radionuclide Sorption and Migration Studies of Getters for Backfill Barriers", Sandia National Laboratories, Sand 79-1110 (1980).

8. G. B. Bell and B. Allard, "Chemical Aspects Governing the Choice of Backfill Materials for Nuclear Waste Repositories", Proceedings of Symposium on Backfill as an Engineered Barrier for Radioactive Waste Management, American Chemical Society, 1980, August 24-29.

9. M. I. Wood and W. E. Coons, "UMTANUM Basalt as a Potential Backfill Component in a Repository Located Within the Columbia River Basalt", Proceedings of Symposium on Backfill as an Engineered Barrier for Radioactive Waste Management, American Chemical Society, 1980, August 24-29.

10. J. C. Maxwell. A Treatise on Electricity and Magnetism. Oxford University Press, London, 1892.

11. D. A. De Vries. Thermal Conductivity of Soil, Mededel, Landbouuhogeschool, Wageningen, Netherlands, 1952.

12. M. S. Kersten, "Thermal Properties of Soils", Engineering Experimental Station, University of Minnesota, Tech., Bulletin 29 (1949).

13. S. Knutsson, "Thermal Conductivity of Highly Compacted Bentonite", Karn-Bransle-Sakerhet Teknisk Rapport 11 (1977).

14. K. W. Jackson, "Enhancement of Thermal Energy Transport Through Granular Media", Ph.D Thesis, Georgia Tech, 1980.

15. H. W. Russell, "Principles of Heat Flow in Porous Insulators", J. Amer. Ceram. Soc. 18, 1 (1935).

16. W. Woodside and J. H. Messmer, "Thermal Conductivity of Porous Media. I. Unconsolidated Sands", J. Appl. Phys. 32, 1688 (1961).

17. A. Gemant, "The Thermal Conductivity of Soils", J. Appl. Phys. 21, 750 (1950).

18. L. D. Bauer, W. H. Gardner and W. R. Gardner, Soils Physics, Wiley, 1972.

19. J. I. Adams and A. F. Baljet, "Thermal Behavior of Cable Backfill Materials", IEEE Transactions on Power Apparatus and Systems, PAS-87, 1149 (1968).

20. S. Knutsson, "Heat Conduction Experiments with Buffer Substance of Bentonite/Pite Silt", Karn-Bransle-Sakheret Teknisk Rapport 28 (1977). (Atomic Energy of Canada Limited Miscellaneous Translation, AECL-tr-MISC-611).

21. Analytical Science Branch, Whiteshell Nuclear Research Establishment, "Analysis of Surface Outcrop Rocks in the Lac du Bonnet Batholith, Field Samples 1977", Atomic Energy of Canada.

22. H. S. Radhakrishna, "Heat Flow and Moisture Migration in Cable Backfills", Ontario Hydro Research Quarterly 20, #2 (1968).

23. S. P. Clark, Jr., "Handbook of Physica l Constants, revised edition", Geological Society of America, Momoir 97 (1966).

24. S. Knutsson, "Thermal Conductivity Tests on Highly Compacted Bentonite", Karn-Bransle-Sakerhet Teknisk Rapport 72 (1977). (Atomic Energy of Canada Limited Miscellaneous Translation, AECL-tr-MISC-468 (1978).

25. R. C. Weast (Editor), CRC Handbook of Chemistry of Physics, 55th edition, Cleveland, Ohio, CRC Press, 1975.

Table 1

Thermal Conductivity of Geological Materials

Material	State	Moisture Content (%)	Dry Density (kg/m^3)	Thermal Conductivity W/(°C m) Moist	Thermal Conductivity W/(°C m) Dry	Information Source
Quartzite Rock	natural	1.5	2400-2700	9.0	8.0	23
Granite Rock	natural	2.0	2600	4.0	3.3	23
Limestone	natural	4.0	2400-2600	3.5	2.5	23
Sandstone	natural	2.0	2300-2700	4.0	3.0	23
Shale	natural	4.5	2000-2300	1.8	1.0	23
Crushed Quartz	compacted	6.0	2200	3.0	1.6	*
Crushed Limestone	compacted	6.0	2100	2.2	0.8	*
Crushed Shale	compacted	10.0	2100	1.3	0.6	*
Silty Clay	compacted	14.0	1800	1.3	0.6	*
Ottawa Sand	compacted	5.0	1700	2.0	0.5	*
Glacial Till	compacted	12.0	1900	2.3	0.8	*
Bentonite Clay	compacted	25.0	2020	0.9	0.8	24
Coal Ash	compacted	15.0	1500	1.0	0.3	*
Water	non-convective	-	-	0.63	-	25
Air	non-convective	-	-	0.025	-	25

* Ontario Hydro, unpublished data

Table 2

Physical Properties of Components in Candidate Buffers

Component	Specific Gravity	Size Fractions (%)			Air Dry Moisture Content (%)
		Sand	Silt	Clay	
Wedron Sand	2.65	100	0	0	0.3
Graded Silica	2.70	90	10	0	0.5
Boundary Dam Fly Ash	2.60	11	87	2	-
Blackhills Tower Bond Bentonite	2.18	0	18	82	8.2
Avonlea Bentonite	2.45	0	39	61	8.8
Avongel Bentonite	2.21	0	12	88	9.3
Sealbond	2.76	0	65	35	1.4
Kaolin	2.60	0	30	70	1.1
Zeolite	2.33	35	60	5	4.4

Table 3

Chemical Characteristics of Buffer Materials Tested

	Chemical Composition (%)									Loss on Ignition	
Material	SiO_2	Al_2O_3	Fe_2O_3	MnO	TiO_2	CaO	MgO	K_2O	Na_2O	(%)	pH
Wedron Sand*	99.8	0.10	0.02	-	0.015	0.015	0.005	-	-	0.08	7.0
Graded Silica*	99.8	0.10	0.02	-	0.015	0.015	0.005	-	-	0.08	7.0
Blackhills Tower Bond Bentonite	65	20.2	2.30	1.0	0.10	1.40	1.00	0.80	3.10	7.0	8.4
Avongel Bentonite*	60	18.0	2.0	-	-	0.8	2.0	0.4	2.3	5.5	9.3
Sealbond**	59.1	15.8	5.30	0.12	0.53	4.96	3.99	4.0	0.81	4.1	-
Kaolin**	44.0	86.67	0.31	-	1.0	0.24	0.19	0.17	0.13	13.14	-

* Data supplied by manufacturer

** AECL unpublished data

Table 4

Thermal Conductivity of Candidate Buffer/Backfills

Mix Composition[1]	Moisture Content Range (%)	Dry Density Range (kg/m^3)	Thermal Conductivity in Moist Condition W/°C m	Thermal Conductivity in Dry Condition W/°C m
WSAB-15	6 - 17	1100 - 1770	1.3 - 2.8	0.7 - 1.0
WSAB-25	5 - 16	1200 - 1760	1.2 - 2.7	0.6 - 0.9
WSAB-50	15 - 29	1330 - 1400	1.2 - 2.0	0.5 - 0.6
GSBB-10	6 - 10	1850 - 2000	2.0 - 3.0	0.4 - 0.7
GSBB-25				
GSBB-50				
GSSB-15	4 - 9	2050 - 2175	3.0 - 4.0	1.5 - 1.7
GSK-15	4 - 9	2000 - 2200	2.5 - 3.5	1.8 - 2.0
GSZ-15	5 - 12	1725 - 1850	0.8 - 3.5	1.8 - 2.0
CGBB-15	5 - 19	1750 - 2000	1.3 - 2.6	0.8 - 1.0
CGBB-25	6 - 12	1620 - 1710	1.0 - 1.5	0.6 - 0.8
CGBB-50	12 - 17	1500 - 1550	0.8 - 1.0	0.5 - 0.6
CGSB-15	6 - 10	1890 - 2200	2.4 - 3.4	1.4 - 1.8
CGSB-25	7 - 12	1600 - 2050	1.4 - 2.4	0.8 - 1.0
CGSB-50	9 - 15	1850 - 1980	1.8 - 2.4	0.9 - 1.0
CGK-15	6 - 10	1900 - 2050	2.2 - 3.0	1.7 - 2.0
CGK-25	9 - 12	1850 - 1980	1.7 - 2.5	0.9 - 1.2
CGK-50	9 - 15	1770 - 1860	2.1 - 2.2	1.0 - 1.2

[1] Notations for Mix Compositions:

WS = Wedron Sand
GS = Graded Silica
CG = Crushed Granite
AB = Avonlea Bentonite
BB = Blackhills Bentonite
SB = Seal Bond
K = Kaolin

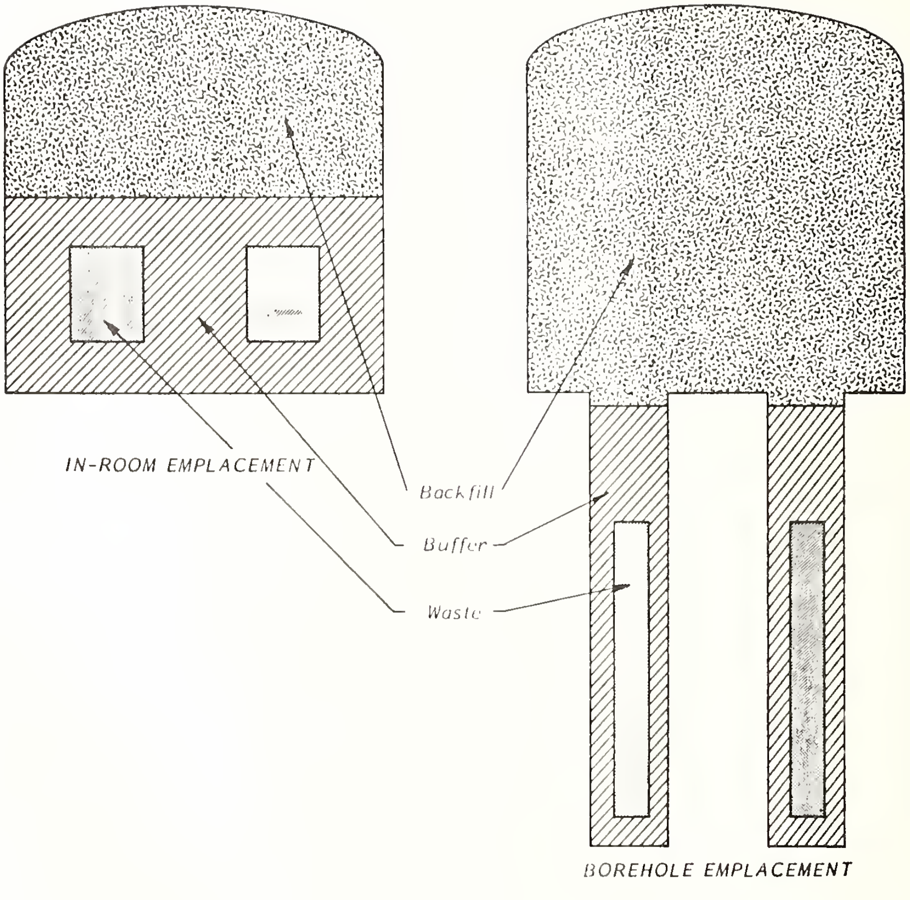


Figure 1. Alternative concepts of waste emplacement in the repository

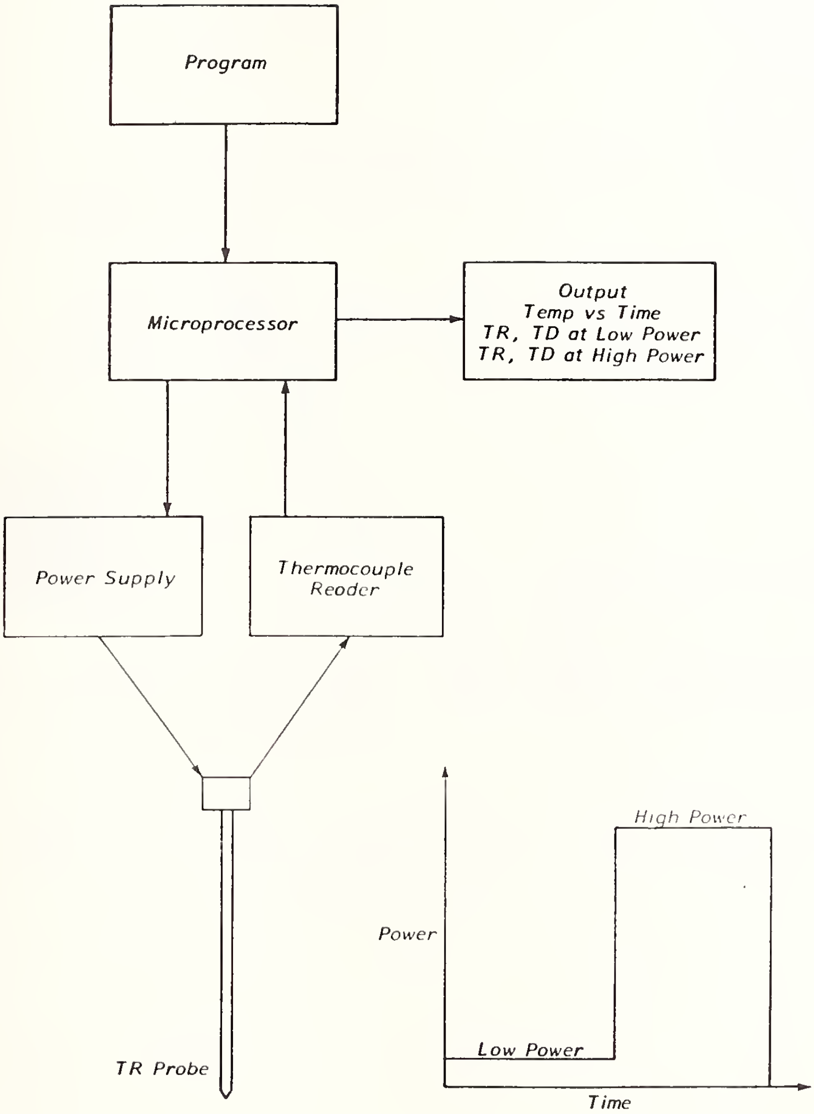


Figure 2. Logic diagram of the thermal properties analyzer

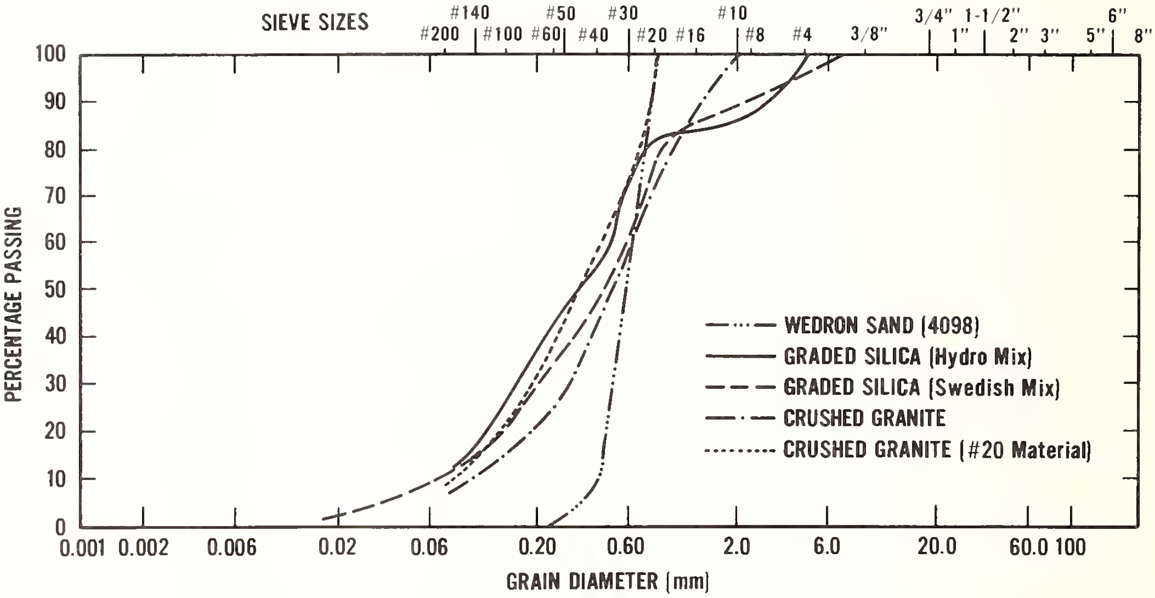


Figure 3. Grain size distribution curves for the coarse fraction of buffer mixes

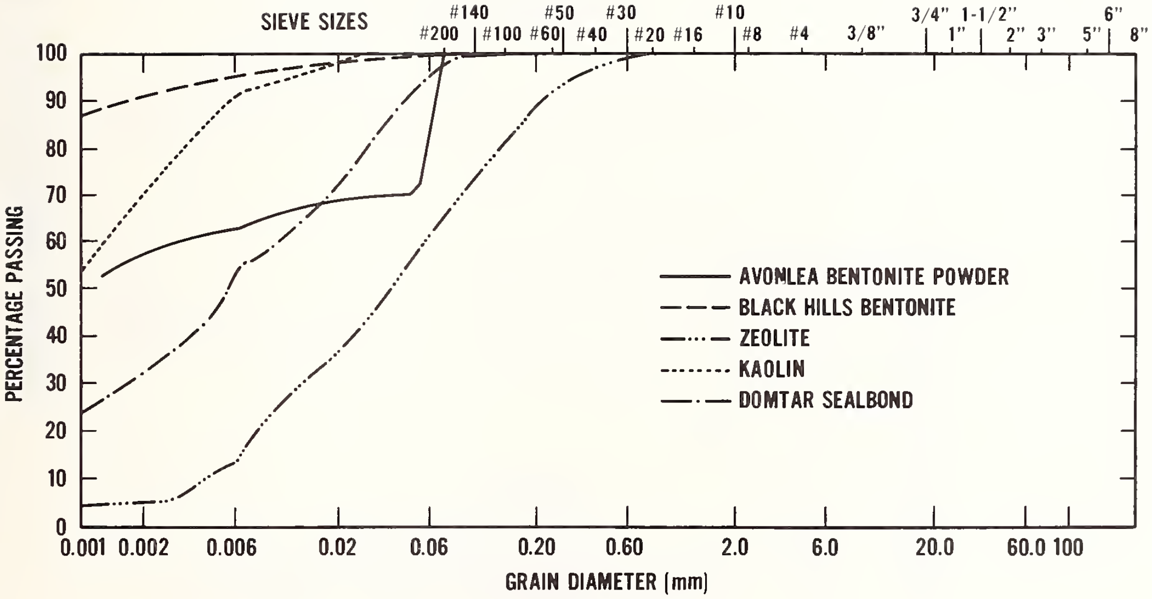


Figure 4. Grain size distribution curves for the fine fraction of buffer mixes

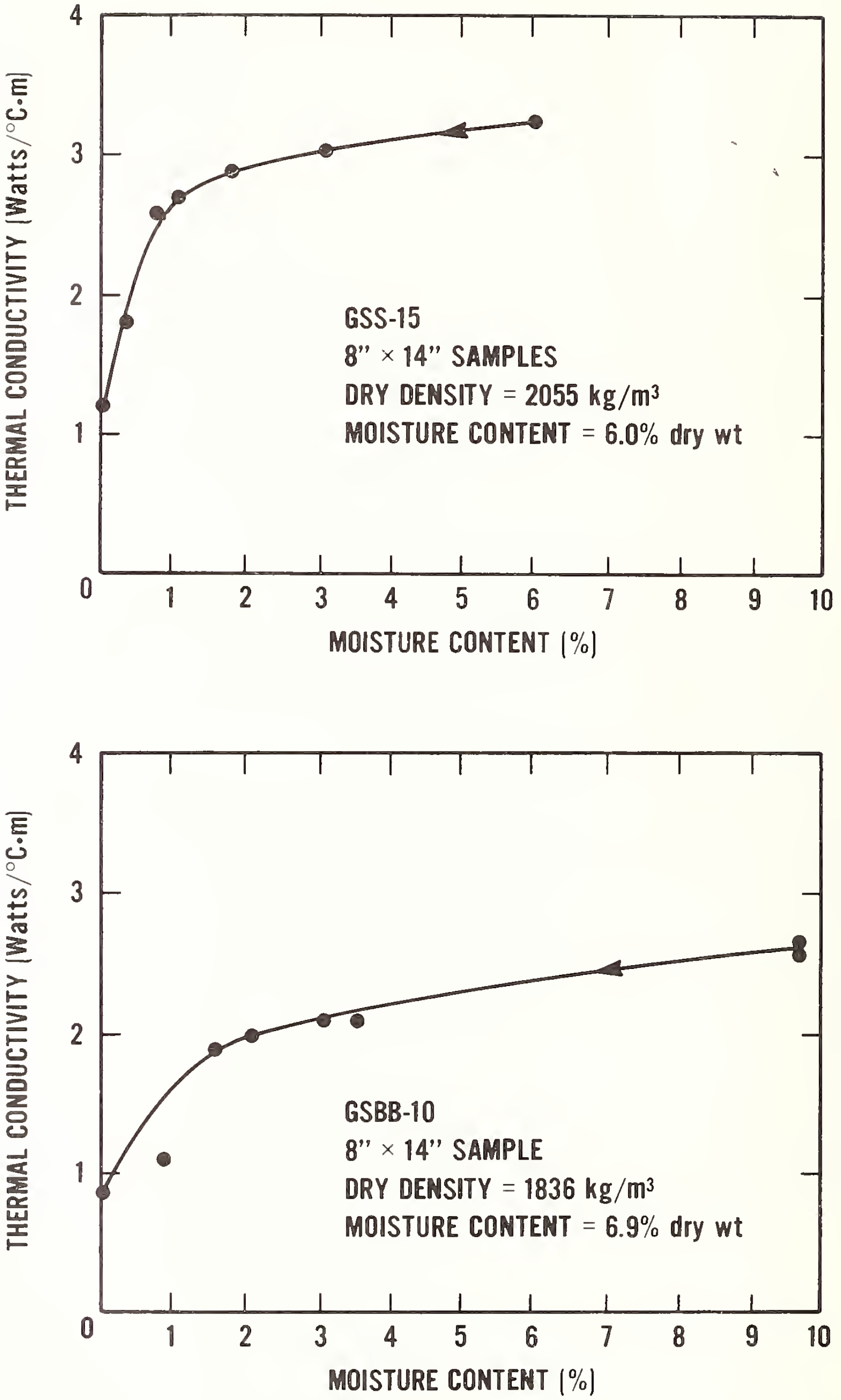


Figure 5. Thermal conductivity vs moisture content relationship at constant density from stage drying tests for

A) 85% Graded Silica + 15% Sealbond Mix

B) 90% Graded Silica + 10% Black Hills Bentonite Mix

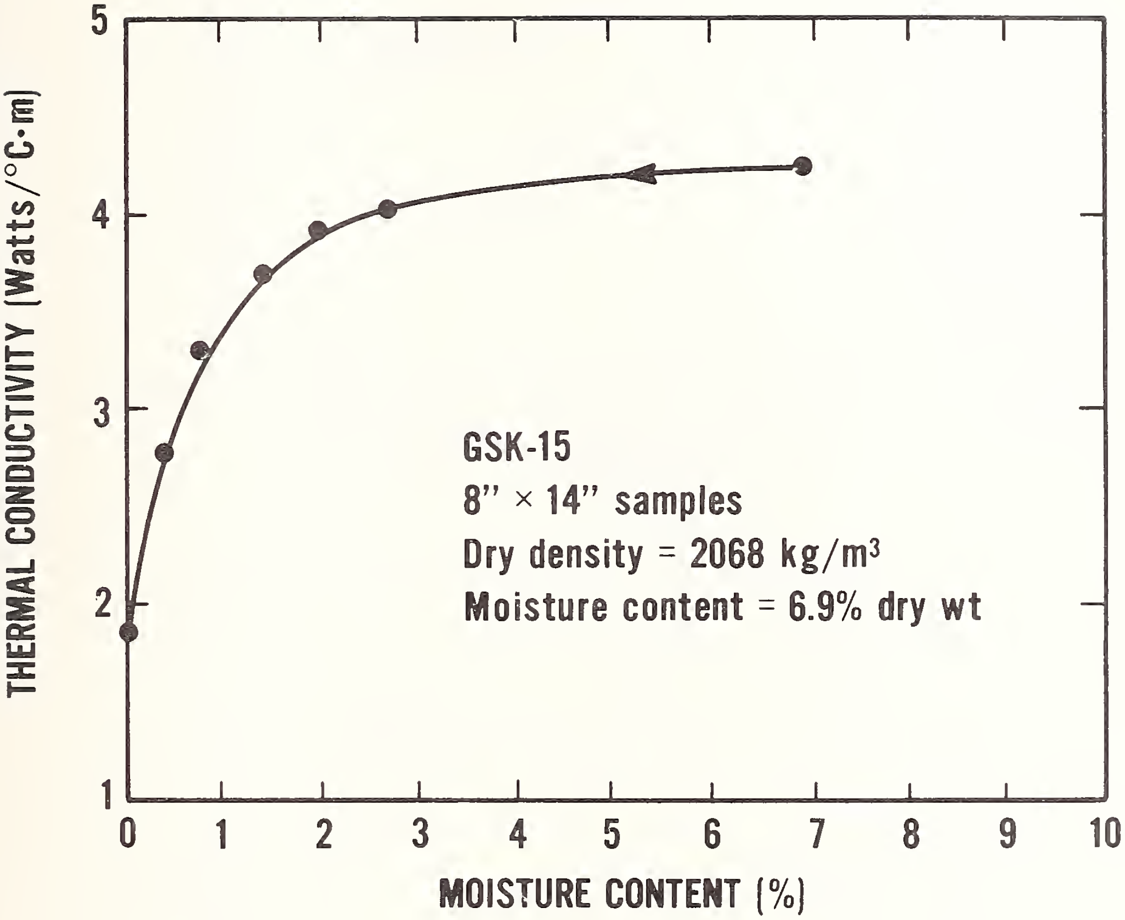
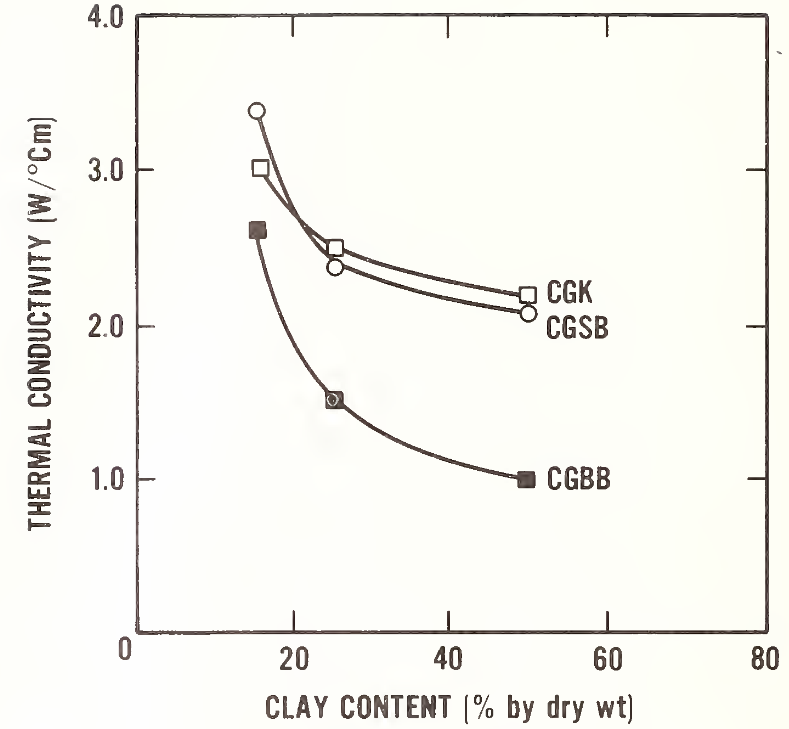


Figure 6. Thermal conductivity vs moisture content relationship at constant density from stage drying tests for 85% graded silica + 15% kaolin mix



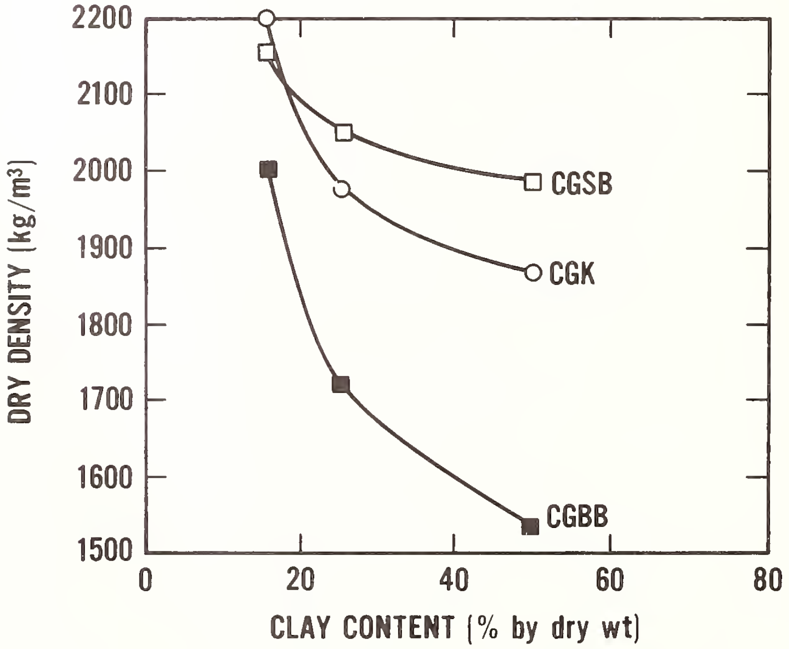


Figure 7. Thermal conductivity and density as a function of clay content in crushed granite and clay mixes

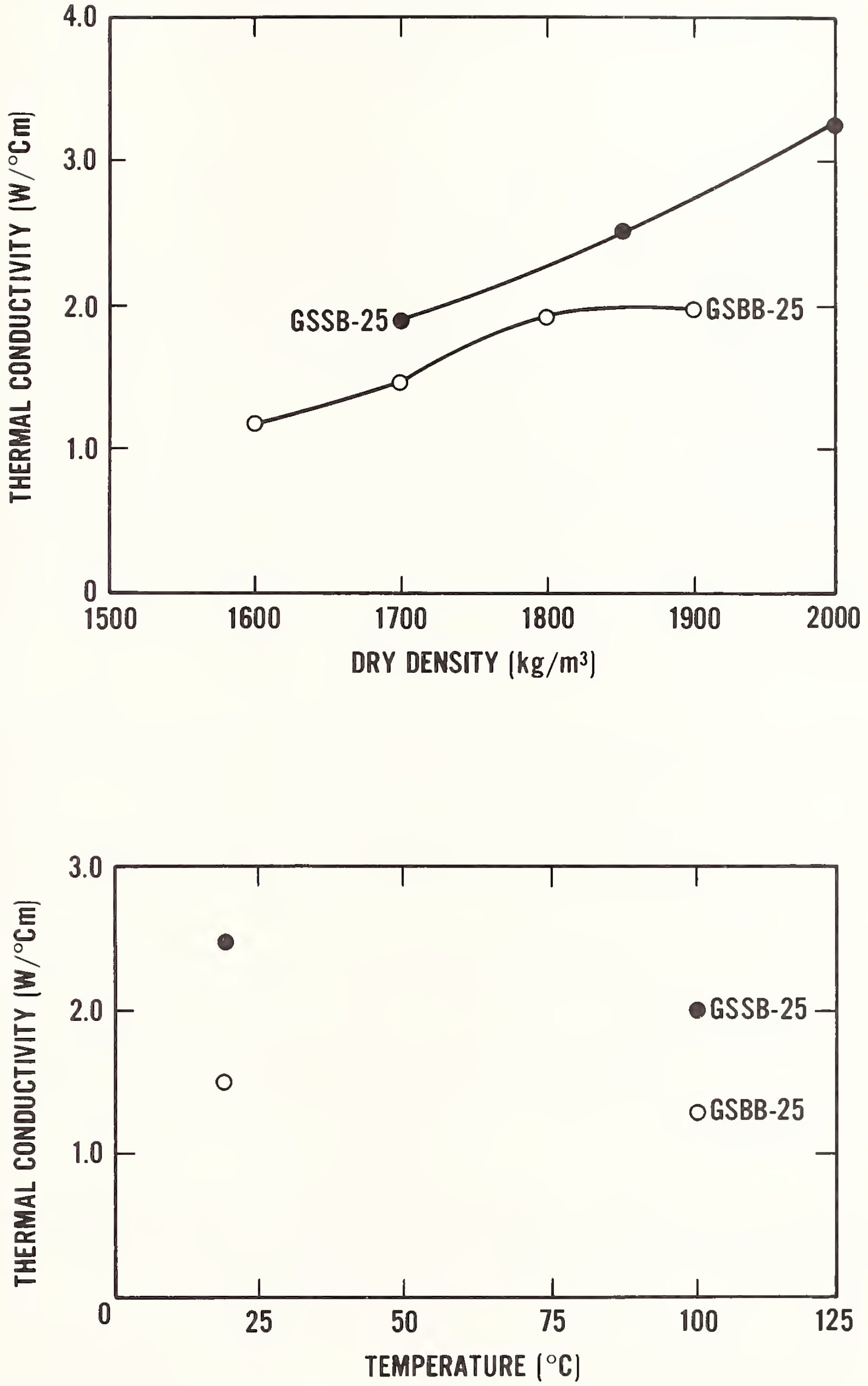


Figure 8. Influence of density and temperature on thermal conductivity of crushed granite - clay mixtures

BENTONITE SHIELDING OF ROCK-DEPOSITED RADIOACTIVE WASTES

Roland Pusch
Division of Soil Mechanics
University of Lulea
Lulea, Sweden

INTRODUCTION

The Swedish KBS concepts for final deposition of canisters with nuclear wastes from reactors, imply that the canisters be placed in deposition holes which are bored from the floors of tunnels at about 500 m depth in Swedish crystalline bedrock. The canisters are isolated from the rock by a "buffer mass" consisting of pure bentonite in the case of unreprocessed wastes, and the deposition plant is finally sealed by backfilling shafts and tunnels with bentonite/sand mixtures (figure 1). The design, involved techniques, and expected barrier effects have been described in various KBS reports. A short description of the properties of highly compacted bentonite has been published by Pusch in 1979 (1).

The bentonite, which surrounds the canisters, is applied in the form of blocks of highly compacted bentonite. The blocks are produced by isostatic compaction of commercial bentonite powder under pressures in the range of 50-100 MPa, which yields block densities of 2.1-2.2 t/m^3 when using Wyoming Na bentonite MX-80 powder. The compacted bodies are cylindrical and they can be manufactured by sawing, drilling etc. so that cylindrical discs, rods, annuli, and practically any shape can be obtained (figure 2). This means that the canisters can be enclosed in a tightly fitting stack of such blocks which are easily applied in each deposition hole.

The particularly attractive properties of the highly compacted Na bentonite, some of which will be commented in this report, are:

- Extremely low permeability
- Low diffusivity
- Considerable cation adsorption capacity

- Sufficient bearing capacity to support canisters
- Sufficient ductility to sustain minor rock displacements without transferring very high stresses to canisters
- High swelling and self-sealing capacities

THE BENTONITE MATERIAL

Bentonite is the term for smectite-rich clay formed by devitrification of the natural glass component of volcanic ash deposited in prehistoric lakes and estuaries. The crystal lattice of montmorillonite[1/] is characterized by a variable c-axis spacing (figure 3) which provides the large internal mineral surface area for cation adsorption, and the strong swelling ability.

The commercial bentonite powder for the production of compacted blocks is obtained by simple grinding and drying of crushed raw material. The grains are silt-sized and consist of face-to-face-grouped stacks of lamellae, i.e., the microstructural order of the natural bentonite beds (figure 4).

Compaction of the powder under high pressure welds the aggregates together and yields a strongly coherent, talk-like product. A convenient water content of the bentonite powder is 10% for the production of blocks with a bulk density of about 2.15 t/m^3. This water content corresponds to a degree of water saturation of 50-60%.

MICROSTRUCTURAL PROCESSES ASSOCIATED WITH WATER UPTAKE AND SWELLING

The water uptake and swelling are associated with particle rearrangement meaning that properties like permeability and diffusion should only be determined after the establishment of internal microstructural equilibrium. The driving forces when partially saturated highly compacted bentonite takes up water are due to the strong affinity of the montmorillonite to water, and to capillary action. The aggregates start to take up water in their outer parts and thereby expand by

---

[1/] The major smectite clay mineral is Wyoming bentonite, the KBS reference clay.

which the interaggregate voids are successively filled with a clay gel. As long as there are differences in water content in various parts of the system, there is a tendency of adjustment so that a homogeneous microstructural state is finally achieved.

The migration of water leading eventually to water saturation can be considered as a diffusion process governed by a "concentration" gradient. The "concentration" equals to the water content, which has a fairly low value (approximately 10 percent at a bulk density of 2.1-2.2 t/m^3) at the start of the water uptake, and which finally reaches a certain maximum value (20-25 percent) under completely confined conditions. The isotropic character of the highly compacted bentonite suggests a simple form of the diffusion equation for uniaxial migration:

$$\frac{\partial w}{\partial t} = D \frac{\partial}{\partial x} \left(\frac{\partial w}{\partial x}\right)$$

where w = water content

$$D \sim 3 \cdot 10^{-10}\ m^2/s$$

This finding is of great practical importance since it offers a simple way of predicting the rate of water intake and the distribution of the water that has been taken up by the clay blocks.

SWELLING PRESSURE

If a partly water saturated, compacted bentonite body has access to external water and is confined so that its total volume remains constant, i.e. in principle the conditions in figure 1, the individual crystallites take up water in connection with an expansion in the c-axis direction. When this expansion can no longer proceed as a consequence of the limited space, a "swelling pressure" is exerted on the confinement. Figure 5 illustrates a generalized relationship between the swelling pressure p_s and the bulk density MX-80 bentonite at complete water saturation.

A close examination of the influence of pore water chemistry on the swelling pressure has shown that the concentration and composition of dissolved salts (maximum concentration corresponding to that of the oceans) did not have any noticeable effect on p_s when the bulk density was higher than about 2 t/m^3. This is because electrical double layers are not, or incompletely developed at interlamellar distances less than 10-15 Å. Instead, the high swelling pressure at such high densities is related to the pressure of strongly fixed, "steric"-type interlamellar water molecules. The matter is dealt with in various KBS reports (2).

PERMEABILITY

The presence of largely immobilized interlamellar water leaves only narrow, tortuous passages for water migration between stacks of montmorillonite sheets. The resulting permeability is therefore extremely low (figure 6).

The interaction of mineral lattices and water molecules - mainly through hydrogen bonding - is strong in the narrow passages, such as interlammellar space, and less intense in wider passages, the variation in interaction being stochastic. While water flow through clays of low density can be considered as the motion of a fluid with a definite and constant viscosity, it should rather be regarded as a shear-induced displacement of a "structured" medium in the case of dense bentonites. This suggests a close resemblance to the creep of clay. It is in fact illustrated by the obvious retardation of the flow rate after changing the hydraulic gradient (figure 7), by the non-Darcy behavior, and by the very obvious influence of temperature (3).

SELF-HEALING EFFECTS

The unique relationship between bulk density and swelling pressure suggests that homogeneous conditions and uniform water distribution are attained in confined, sufficiently dense bentonite bodies if given enough time for the establishment of internal equilibrium. The strong ability to swell and become homogeneous is illustrated by figure 8 which shows how 3 loose pieces of precompacted bentonite in a steel cylinder absorbed water from one end and turned into one, coherent, swollen, and fairly homogeneous piece of clay after only 3 weeks. Such self-healing requires, however, that the average density is of the order of 1.5 t/m^3. Lower densities imply non-associated water and thus a possibility of the formation of permanent local zones with "excess" water.

The property of very dense bentonite to swell out and fill voids and form, ultimately, a uniform clay body is the main principle of a new technique of sealing boreholes (4). Perforated, closely fitting copper tubes filled with highly compacted bentonite cylinders are inserted in the hole, its outermost part then being sealed with cement. Water is taken up from joints which intersect the hole and a completely homogeneous state of the bentonite is expected after a sufficiently long time. Wider joints require sealing by pretreatment with bentonite, cement, or some other suitable substance. Pilot tests in 6 m long holes in granite are presently being run in the Stripa mine.

REFERENCES

1. Pusch, R., 1979, Highly Compacted Sodium Bentonite for Isolating Rock-Deposited Radioactive Waste Products, Nuclear Technology Vol. 45, Sep. 1979, pp. 153-157.

2. Pusch, R., 1980a, Swelling Pressure of Highly Compacted Bentonite, KBS Technical Report 80-13.

3. Pusch, R., 1980b, Permeability of Highly Compacted Bentonite, KBS Technical Report 80-16.

4. Pusch, R. and Bergström, A., Highly Compacted Bentonite - A Self-Healing Substance for Nuclear Waste Isolation, Proc. Mat. Res. Soc. Ann. Meet. Nov. 16-21 1980 Boston, Mass.

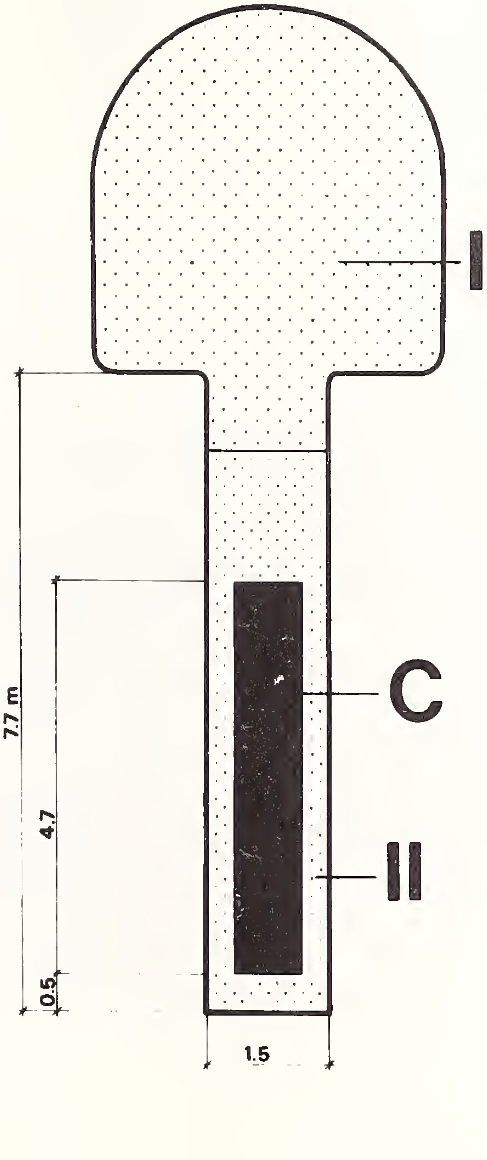


Figure 1. Schematic view of tunnel and deposition hole containing canister with unreprocessed reactor wastes. I) bentonite/sand, II) highly compacted bentonite, C) copper canister

Figure 2. Example of precompacted ring of bentonite

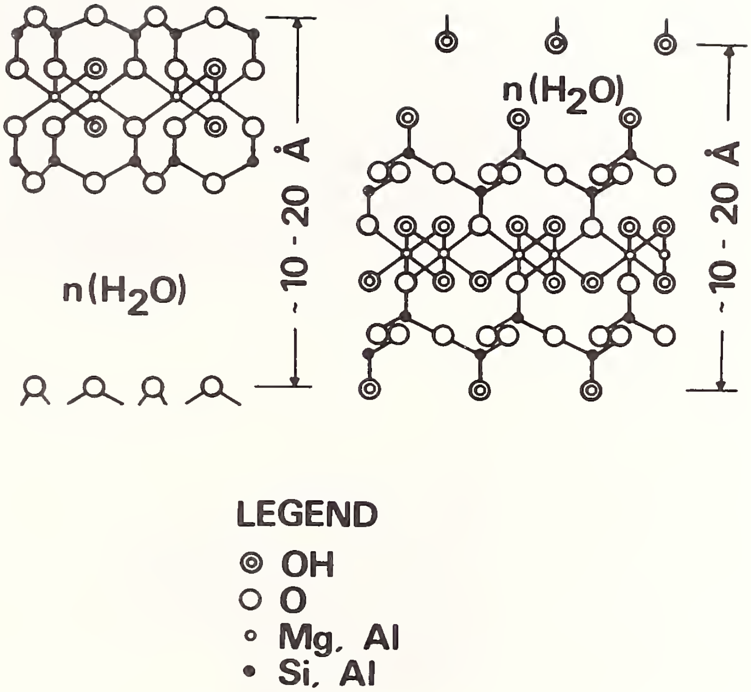


Figure 3. Probable crystal structures of montmorillonite; left model according to Hofmann, Endell and Wilm; right model according to Edelman and Favejee. The intrastructural space contains water molecules and cations

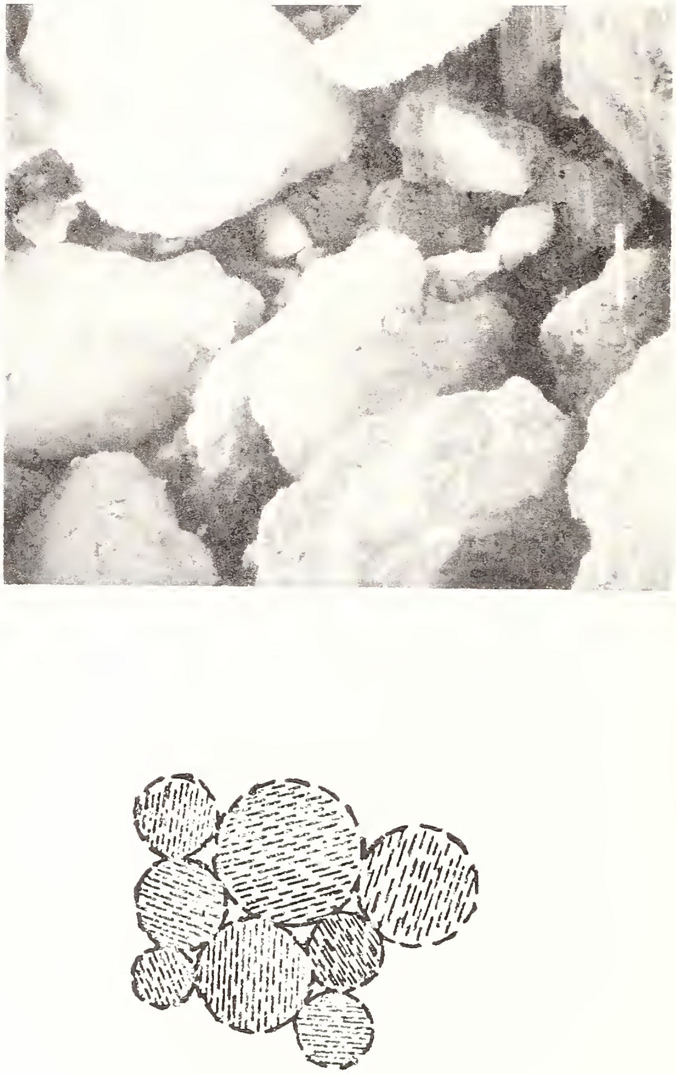


Figure 4. Upper picture shows scanning micrograph of MX-80 aggregates. Notice the tapered shape of the aggregates (100x). (Photo D.M. Anderson, New York State University at Buffalo)

Lower picture illustrates, schematically, the microstructural pattern of bentonite powder

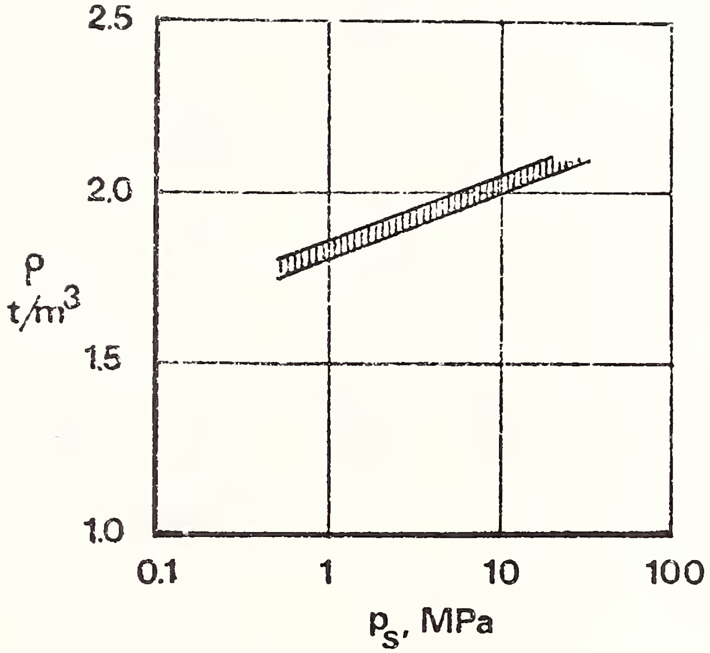


Figure 5. Approximate relationship between swelling pressure, P_s/density of MX-80 bentonite at complete saturation and 20°/C

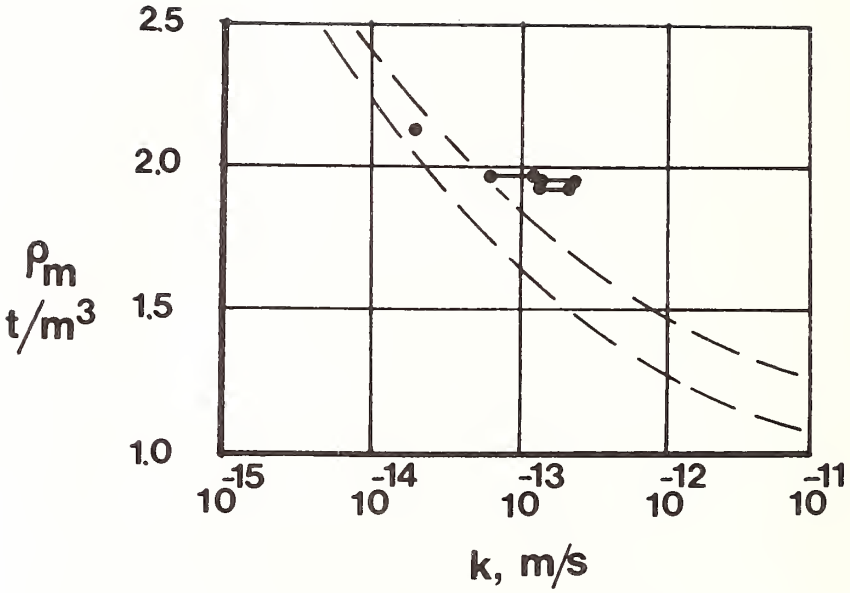


Figure 6. The coefficient of permeability versus bulk density. Where two values are plotted for one and the same density, the left represents $i = 10^3$, and the right 10^4 (i = hydraulic gradient). Broken curves are upper and lower boundaries of literature-derived values

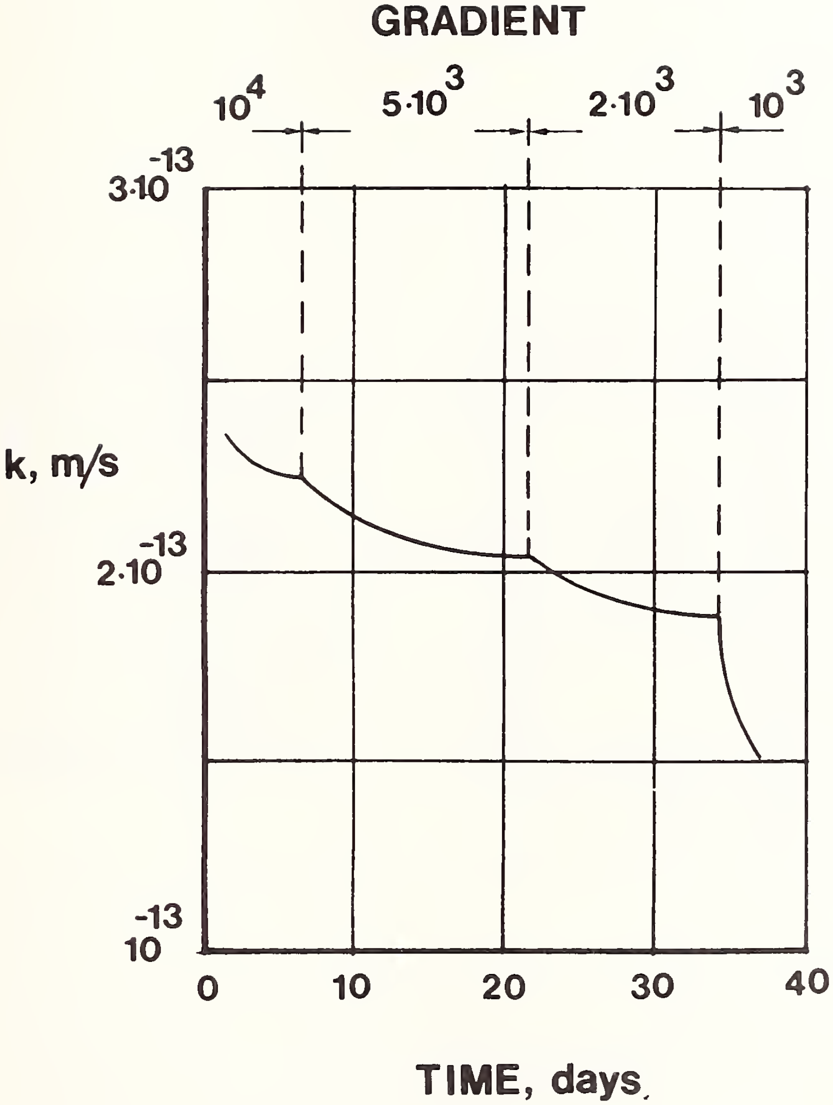


Figure 7. Example of peremability test with smoothened curve for k versus time after the test start

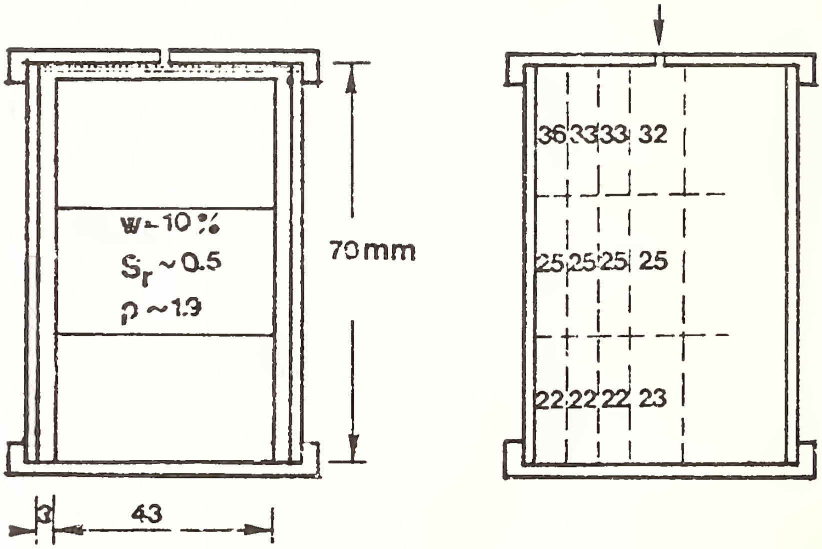


Figure 8. Swelling test with precompacted bentonite. The right picture shows the average water contents of symmetrically situated elements after 3 weeks

EVALUATION OF BACKFILL AS A BARRIER TO RADIONUCLIDE MIGRATION IN A HIGH LEVEL WASTE REPOSITORY[1,2]

T. M. Ahn, R. Dayal, and R. J. Wilke

Department of Nuclear Energy
Brookhaven National Laboratory
Upton, NY 11973

ABSTRACT

The feasibility of using highly sorptive synthetic minerals such as zeolites or titanates as backfill in a HLW repository has been evaluated in terms of the NRC 1,000 yr containment and 10^{-5}/yr controlled release criteria. The results indicate that for groundwater velocities below 1 ft/yr, diffusion and sorption are the dominant processes controlling radionuclide migration in backfill systems. A 3-ft-thick bed of synthetic zeolite backfill can provide total containment of activity released continuously from a 55-gallon glass monolith for a period of up to 1,000 years. For longer time periods of up to 10^6 years, the controlled release rate of 10^{-5}/yr can be met by a >10-ft-thick bed of zeolite backfill, assuming the glass monolith to be the source term. In the absence of the waste form as an engineered barrier, a bed thickness of 3 to 10 ft is required to

---

[1] Support for this work was provided by the U.S. Nuclear Regulatory Commission Contract No. DE-AC02-76CH00016.

satisfy the controlled release criterion for a period of up to 10^5 years. With a bed thickness of 200 ft, synthetic zeolites can be used as backfill to provide containment of the total U.S. waste inventory (10^9 Ci), if it were placed in one repository, for periods up to 10^6 years. Zeolites are known to exhibit radionuclide-specific sorption properties. We believe that the range of sorption coefficients (10^3-10^4 ml/g) considered in the calculations will cover the various zeolite-radionuclides systems. We wish to emphasize that the above estimates are conservative since diffusion occurs radially and we have only considered uni-dimensional transport in our models. In addition, radioactive decay of the nuclides has not been considered in the calculations.

We also wish to note that in this report we have not dealt with radiation and thermal stability of backfill materials. Zeolites have been used to clean radioactivity from water systems and have been loaded to as high as 75,000 curies per MTHM. Although we believe that zeolites should be structurally stable to low radiation loadings (approx. 1,000 curies per cubic foot) and that radiolysis efforts should not be important at these levels of radiation, very little work exists in this area. In addition, we have not addressed the retention of long-lived anionic species such as SeO_4^{2-}, I-, TcO_4^-. It appears that other materials will have to be added to backfill to maximize its effectiveness for retention of all radionuclides of interest.

INTRODUCTION

The operational life of a geologic high level waste (HLW) repository after decommissioning can be divided into two time periods: (a) the period (<1,000 yr) during which the short lived fission products dominate the hazard posed by the waste, and (b) the long term period (>1,000 yr) during which the hazard is dominated by the very long lived isotopes including the actinides [1]. In view of these considerations, the Nuclear Regulatory Commission (NRC) had adopted a strategy for regulatory and licensing the disposal of high level radioactive wastes in geologic repositories which requires that the engineered systems of the underground facility meet the following criteria [2]:

a. Containment of all radionuclides in HLW for at least the first 1,000 years after decommissioning of the geologic repository, assuming expected events and processes. This containment shall result from properties of the waste package.

b. Starting 1,000 years after decommissioning of the geologic repository, the radionuclides present in HLW will be released from the engineered system to the geologic setting at an annual rate that in no case greater than one part in one hundred thousand of the total activity present within the underground facility at that time, assuming expected processes and events.

c. For transuranic waste (TRU), the engineered system shall be designed so that following decommissioning of the geologic repository the annual release rate from the underground facility into the geologic setting is at most one part in one hundred thousand of the total activity present in the underground facility at any time following decommissioning.

Most of the current Department of Energy (DoE) programs in high levels waste research are investigating the waste form and container as potential components of a waste package in meeting the containment requirements outlined above. In this report, we have attempted to evaluate the effectiveness of using synthetic zeolites and titanates as potential backfill materials in an HLW repository. It should be pointed out that a DoE waste package is defined to include everything that is placed in the waste repository emplacement hole, i.e., the waste form, filled container, overpack, sleeve, and backfill. The NRC waste package includes discrete backfill [3] in contrast to non-discrete backfill in DoE definition. Since the 1,000 yr criterion applies specifically to the waste package and the 10^{-5}/yr controlled release criterion refers to the engineered system, we have evaluated the effectiveness of backfill in meeting both performance requirements: discrete backfill for 1,000 yr containment and non-discrete backfill for controlled release.

Zeolites and Titanates as Backfill

Although backfill in an HLW repository has been identified to provide several functions, the most important role is to retard migration of radionuclides in groundwater that has reached the waste in the event of a container breach. It is primarily this property that we have considered in our evaluation of synthetic zeolites and titanates for backfill. In addition to providing a high potential for required retention of radionuclides, synthetic backfill materials of regularly shaped small spheres can be used, through prior selection of the mass, volume, shape, and packing densities, to predetermine and control the groundwater flow rates and patterns into and through the engineered underground facility for varying aquifer pressures and flow rates. Such a modification of the hydrodynamics of an engineered facility could be advantageous in that rapid radionuclide transport by advection would be minimized, resulting in diffusion-controlled transport.

Existing technology, widely used in purification systems for water reactors, has demonstrated that synthetic materials including a wide range of zeolites can be used to quantitatively hold up fission products and actinides. A most prominent example of zeolite application to waste management is the use of IONSIV-IE95 and Zeolon 900 in the Submerged Demineralized System (SDS) to be used to decontaminate water from the Three Mile Island Unit 2 Nuclear Power Plant [4]. Another such example is the storage of krypton-85 in sodalite zeolite [5]. Recently, Nowak [6] and Winslow [7] at Sandia Laboratories and Komarneni and Roy [8] at Pennsylvania State University have examined the sorption properties of zeolites as potential backfill material for use in a HLW repository. Pennsylvania State University is also investigating the use of mixtures of zeolites and clay minerals for backfill material around a waste package and the fixation properties of sorbed radionuclides of various zeolite minerals [9].

Sodium titanate materials have been developed at Sandia Laboratories for quantitative removal of fission products and actinides from commercial liquid wastes [10, 11]. Subsequent pressure sintering of the loaded material results in a dense ceramic waste form resistant to leaching. Based on the high decontamination factors observed in HLW studies, the Sandia titanate materials are also being investigated for the decontamination of defense liquid wastes stored at the Hanford site in Washington.

In this report, we have attempted to evaluate the feasibility of using highly sorptive materials such as synthetic zeolites and titanates as backfill in a HLW repository. The effectiveness of such a barrier with respect to radionuclide retention has been assessed in terms of the NRC release criteria discussed above.

Radionuclide Transport Processes

To evaluate the migration of radionuclides in an ion exchange backfill barrier, we have to first consider the dominate transport mechanisms. The principal mechanisms controlling the nature and extent of radionuclide transport in sorptive materials are ion exchange, sorption, diffusion, precipitation, advection, and other irreversible reactions. Ion exchange and sorption processes are essentially reversible (at least over short time periods), resulting in a net retardation in the diffusion-controlled ion migration. The extent of retardation will be primarily determined by the sorption kinetics and equilibria of solid-fluid interactions. Non-reactive ions such as tritium, however, do not exhibit interactions with particle surfaces. In this case, ion exchange and sorption processes are not significant and, consequently, the migration is primarily controlled by diffusion and advection.

Precipitation of dissolved radionuclides as authigenic minerals will decrease the rate of radionuclide migration. Formation of new minerals, immobilizing radionuclides release from the waste form, as a result of quaternary interactions involving waste/backfill/host rock with hydrothermal fluids has been documented [12]. However, such data are not available for the various backfill materials. Therefore, precipitation, as a retardation process, will not be considered in this report.

Similarly, irreversible reactions such as fixation of sorbed ions with aging of the sorbent can also decrease the rate of radionuclide migration. Recently, it was reported that synthetic zeolites, used for decontamination purposes at Hanford, were found to fix certain radionuclides irreversibly in a period of less than five years [13]. Although this mechanism could be potentially effective in retarding radionuclides in a bed of synthetic zeolites, the available data are insufficient at this time for this mechanism to be considered in our transport model.

At a recent Waste Rock Interaction Technology Meeting in Seattle, a paper was presented describing the role of fine suspended particulates in transporting radionuclides [14]. This process could serve as a potentially important mechanism for radionuclide migration in argillaceous backfill materials which commonly contain extremely small particulates. However, in the case of synthetic materials, the particle size distribution would be very narrow, giving rise to a relatively uniform particle size. Therefore, we believe that this mechanism will not contribute significantly to an increase in the rate of radionuclide migration in backfill systems. However, the significance of this process as a transport mechanism for radionuclides should be investigated.

The principal transport mechanisms which we have considered for evaluating zeolite backfill are diffusion, sorption, and advection. Since groundwater flow rate is relatively low in a realistic repository situation, dispersion associated with advection is considered to be negligible. An example of radionuclide transport by advection, diffusion, and a combination of advection, diffusion, and sorption processes is displayed in figure 1 [15]. As shown in figure 1, pure diffusion corresponds to radionuclide migration as a result of interstitial concentration gradient for non-reactive ions such as tritium. For reactive ions such as Cs^+ and Sr^{2+} and assuming bulk fluid flow to be negligible, the transport is primarily determined by diffusion and sorption, where the sorption equilibrium coefficient determines the extent of ion retardation as compared with diffusional transport for non-reactive ions. For systems having a high groundwater flow velocity, advection becomes the dominant transport process as compared with diffusion (figure 1). Dispersion effects, resulting from the variation in bulk fluid flow velocities between the particles, could give rise to a broadening of the advective front.

MODEL CALCULATIONS

We have developed one-dimensional models that describe quantitatively the transport of radionuclides in a backfill bed. A range of values are considered for groundwater flow rate, sorption coefficient, bed thickness, and transit time.

Phenomenological Model 1

In model 1, the details of which are given in appendix 1, we have assumed a waste glass, having a constant leach rate over a longer period of time, to be the source term. A constant concentration is maintained at x=0, the interface between the waste glass and backfill. In other words, it is assumed that the radionuclide source is non-depleting, time invariant. The transport mechanisms considered are diffusion, sorption, and advection. The groundwater velocity is assumed to be constant with time and uniform throughout the bed. The dispersion term, resulting from localized velocity variations around the particles, is assumed to be negligible. Finally, radioactive decay of the source term and precipitation and fixation of the radionuclide are not considered.

Nowak [6] simplified a material balance equation around a differential volume element in the backfill bed. His formulation is directly applicable to the present analysis. It consists of a linear partial differential equation with three terms: (1) the rates of accumulation (or depletion) of sorbing species in the liquid and on the solid, (2) the net transport of liquid phase species by advection, and (3) the net transport of liquid phase species by ion diffusion. The groundwater flow in a repository must be considered and could serve as a potentially important transport process, especially for non-reactive radionuclides.

Using model 1, the results were calculated for a range of conditions and the concentration-bed thickness profiles for various transit times are displayed in figure 2. In figure 2(a), for a sorption coefficient of 1,000 ml/g and a water velocity of 1 ft/yr, the 1,000 year and 10,000 year profiles show that for relatively short time periods diffusion plus sorption are the rate controlling processes, with the initial concentration dropping by a factor of $>10^6$ within 20 feet of backfill thickness. However, for longer time periods of up to 100,000 years and greater, transport associated with bulk fluid flow becomes dominant, even for reactive ions having a sorption coefficient of 1,000 ml/g. In the absence of bulk fluid flow, the advective term becomes zero. As shown in figure 2(b), the transport is dominated by diffusion and sorption. These findings are in general agreement with those reported recently by Nowak [21]. For materials having negligible sorptive capacity such as quartz, and in a zero groundwater flow stationary system, radionuclide transport is determined by pure diffusion via interstitial water (figure 2(c)), emphasizing the importance of sorption as a retardation process in the transport of radionuclides through a backfill barrier.

Phenomenological Model 2

In this model, we exclude the waste form as an engineered barrier which maintained constant concentration at the interface of the glass and backfill by continuous congruent leaching over a period of time. Instead, at zero time (t=0), the total concentration of the radioactive waste is hypothesized to enter into the backfill instantaneously along the x direction as a sharp step increase (more precisely as a delta function) in its liquid phase concentration c from zero to a constant value C_o. We derived a mathematical expression using the appropriate initial and boundary conditions in appendix 2. The calculated results were obtained using the same range of parameters given in table 1. The assumption of an instantaneous pulse at time t=0 and continuously depleting over a period of time for the source term in model 2 means that no reliance is placed on the waste form as an engineered barrier. Concentration-bed thickness profiles for various transit times, based on model 2 using the parameters given in table 1, are given in figure 3.

Evaluation of NRC Criteria

Although the concentration-bed thickness profiles generated using models 1 and 2 (figure 2 and 3) are useful in terms of identifying the processes that control radionuclide migration in a sorptive backfill system and in determining their relative magnitudes, quantitative information is required to address the performance objectives set by NRC. For this purpose, we assumed the source term to be a glass monolith having a volume of 55 gallons. Assuming 30 percent waste loading in the glass, an activity of approximately 4.2 Ci/cm^3 of glass monolith is calculated. The age of the waste is assumed to be approximately 1.5 years old [21]. The average glass leach rate is considered to be 10^{-3} g/cm^2-yr.

Based on assumed leach rates for the waste glass, surface areas of the glass monolith and the total amounts of radioactivity released from the source for the periods 10^3 and 10^6 years as estimated from the profiles shown in figure 2, we estimated the total amount released from a 3-ft-thick backfill, having a range of sorptive capacities, in a period of 1,000 years at the groundwater flow velocity of 1 ft/yr. As shown in table 2, a 3-ft-thick bed of backfill materials, such as zeolites having a sorption coefficient of 10^4 ml/g, will contain practically all the inventory released from the waste for a period of up to 1,000 years. This implies that a discrete zeolite backfill will satisfy the NRC 1,000 year containment criterion. Table 3 shows calculated results of fractional controlled release rate from a 10- and a 100-ft-thick backfill bed over a period 10^6 years. In these calculations, the assumptions made for the source term and backfill are the same as those described in table 2. Assuming a leach rate of less than 10^{-3} g/cm^2-yr and a surface area of the glass monolith to be 3.6×10^4 cm^2, the NRC controlled release criteria can be met using a >10-ft-thick non-discrete zeolite backfill bed for periods of up to 10^6 years. These calculations show the significance of a waste form having a low leach rate with respect to meeting the controlled release criterion. In such a situation, the sole purpose of the backfill would be to control water ingress in order to minimize the probability of container corrosion. It must be emphasized that bulk fluid flow, based on a groundwater velocity of 1 ft/yr, also contributes to radionuclide transport. In a realistic repository situation, the flow velocities are much smaller than that assumed in our calculations. In that case, the transport would be predominantly diffusion-controlled, resulting in a more effective containment by the backfill barrier.

Using model 2 and making the same assumptions for the source term and backfill barrier, we obtained estimates of the total amount of activity released from a 3-ft-thick backfill barrier in a period of 10^3 years. As shown in table 4, the calculated values indicate that the 1,000 year containment criterion will be met by a 3-ft-thick bed of backfill with sorption coefficients in the range of 10^4 ml/g. Also included in table 4 are values for fractional amounts of activity released from the backfill barrier in 1,000 years.

Using the same model and considering a time period of 10^5 years and bed thicknesses of 3 ft and 10 ft, we obtained estimates of fractional release per year from the backfill bed (table 5). The results show that for backfill materials having sorption coefficients in the range of 10^4 ml/g, the annual fractional release from 3-ft-and 10-ft-thick beds are 10^{-5}/yr and 10^{-6}/yr, respectively. This indicates that the controlled release criterion of 10^{-5}/yr will be met by highly sorptive backfill materials such as synthetic zeolites and/or titanates for long period of time.

Model Calculation for Total U.S. Waste Inventory

In order to demonstrate the effectiveness of backfill as a barrier to radionuclide migration, we assumed the source term to be the total U.S. waste inventory and calculated the total thickness of backfill that would be required to provide controlled release rate over a period of 10^6 years.

Using models 1 and 2, we used a value of 10^9 Ci [23] for the total waste inventory in the U.S. [12] and 100- and 200-ft for bed thicknesses. The calculated estimates of radionuclide release are compiled in tables 6 and 7. Table 6, based on model 1, shows that the annual release from 200-ft-thick backfill, having a sorption coefficient of 10^4 ml/g, is on the order of 10^{-80} Ci/yr. This indicates that the total containment will be achieved during this period.

Table 7, based on model 2, also shows that a 200-ft-thick bed of backfill (K_d=10^4 ml/g) will provide total containment. A bed thickness of 100 ft, however, is not sufficient to obtain the desired containment.

CONCLUSIONS

For groundwater velocities below 1 ft/yr, diffusion and sorption are the dominant processes controlling radionuclide migration in highly sorptive backfill materials such as synthetic zeolites. A 3-ft-thick bed of synthetic zeolite backfill can provide total containment of activity released continuously from a 55-gallon glass monolith for a period of up to 1,000 years. For longer time periods of up to 10^6 years, the controlled release rate of 10^{-5}/yr can be met by a >10-ft-thick bed of zeolite backfill, assuming the glass monolith to be the source term. In the absence of the waste form as an engineered barrier, a bed thickness of 3 to 10 ft is required to satisfy the controlled release criterion for a period of up to 10^5 years. With a bed thickness of 200 ft, synthetic zeolites can be used as backfill to provide containment of total U.S. waste inventory (10^9 Ci), if it were placed in one repository, for periods of up to 10^6 years.

ACKNOWLEDGMENTS

We thank D. G. Schweitzer for initiating this study and C. Sastre for his assistance in computer work. E. J. Nowak, A. Okubo, and G. W. Beall reviewed the manuscript. We thank them for their constructive comments. We are also grateful to Grace Searles, Katherine Becker, Sharon Moore, and Cathy Van Noy for typing and preparation of the manuscript. We also would like to thank Ulesia Gray of the Word Processing Center for final typing of the manuscript.

REFERENCES

1. R. Dayal, K. J. Swyler, and P. Soo, NRC Nuclear Waste Management Technical Support in the Development of Nuclear Waste Form Criteria, BNL Report, BNL-NUREG-27961 (1980), Chapters 4 and 5.

2. Code of Federal Regulations, 10 CFR 60, "Disposal of High-Level Radioactive Wastes in Geologic Repositories," (Proposed Rule), Advance Notice of Proposed Rule Making, Federal Register, May 13, 1980.

3. Code of Federal Regulations, 10 CFR 60, "Disposal of High Level Radioactive Wastes in Geologic Repositories," (Proposed Rule), Advance Notice of Proposed Rule Making, Federal Register, July 8, 1981.

4. E. D. Collins, J. E. Bigelow, D. O. Campbell, L. J. King, and J. B. Knauer, "Evaluation of the Submerged Demineralizer System (SDS) Flowsheet for Decontamination of High-Activity-Level Water at the Three Mile Island Unit 2 Nuclear Power Station," ORNL Report, ORNL-TM-7448 (1980).

5. R. W. Benedict, A. B. Christensen, J. A. Del Debbio, J. H. Keller, and D. A. Knecht, "Technical Feasibility of Krypton-85 Storage in Sodalite," in The Scientific Basic for Nuclear Waste Management - Vol. 2, Plenum Press, N. Y. (1980).

6. E. J. Nowak, "The Backfill as an Engineered Barrier for Nuclear Waste Management," Sandia Report, SAND 79-0990C (1979).

7. C. D. Winslow, "The Sorption of Cesium and Strontium From Concentrated Brines by Backfill Barrier Matreials," Sandia Report, SAND 80-2046 (1981).

8. S. Komarneni and R. Roy, "SUPEROVERPACK: Tailor-made Mixtures of Zeolites and Clays," Presented at Materials Research Societ Annual Meeting, Massachusetts, 1979.

9. R. Roy and S. Komarneni, "Design of Overpack (Backfill) Materials," Paper presented at the National Bureau of Standards Sponsored Workshop on Research and Development Needs Relating to Backfill, Gaithersburg, Maryland, April 1981.

10. R. G. Dosch, "The Uses of Titanates in Decontamination of Defense Waste," Sandia Report, SAND 78-0710 (1978).

11. D. G. Dosch, "Application of Titanates, Niobates, and Tantalates to Neutralized Defense Waste Decontamination-Materials Properties, Physical Forms, and Regeneration Techniques," Draft Sandia Report, SAND 80-1212 (1980).

12. B. E. Scheetz, S. Komarneni, D. K. Smith, and C. A. F. Anderson, "Hydrothermal Interaction of a Ceramic Waste Form With Basalt," in Alternate Nuclear Water Forms and Interactions in Geologic Media, Proceedings of the Workshop held in Gatlinburg, Tennessee, May 1980.

13. R. E. Barletta, Personnal Communication.

14. G. G. Eichholz, "Subsurface Migration of Radioactive Waste Materials by Particulate Transport," Abstrate of paper presented at Waste Rock Interation Technology Meeting, 13-15 October (1980), Seattle, Washington.

15. I. Neretnieks, "Retardation of Escaping Nuclides From a Final Depository," KBS-TR-30, (1977).

16. R. K. Kibbe and A. L. Boch, Technical Support for GEIS: "Radioactive Waste Isolation in Geologic Formations, Vol. 21, Groundwater Movement and Nuclide Transport," Y 10 WI/FM-36/21 (1978).

17. L. D. Ramspott, "Waste Isolation Project: FY-78", UCRL-50050-78.

18. I. Neretnieks, "Transport of Oxidants and Radionuclides Through a Clay Barrer," KBS-TR-79, (1978).

19. D. W. Breck, Zeolite Molecular Sieves, J. Wiley & Sons, N. Y. (1974), Chapter 9.

20. R. C. Weast, Handbook of Chemistry and Physics, CRC Press, 56th Ed. (1975).

21. E. J. Nowak, "The Backfill Barrier as a Component in a Multiplier Barrier Nuclear Waste Isolation System," Sandia National Laboratories, SAND 79-1109, 1980.

22. Management of Commercially Generated Radioactive Waste, DOE Report, DOE/EIS-0046-D, Volume 2, Appendices (1979).

23. D. Majumdar and J. P. Indusi, "Inventory of HLW in the U.S.," BNL-NUREG-27817 (1980).

Table 1

Parameters Considered in Radionuclide Transport Calculations

Backfill Bed Thickness (ft): 1, 3, 10, 100

Transit Time (years): 1,000, 10,000, 100,000, 1,000,000

Sorption Coefficient (ml/g): 1, 10, 100, 1,00, 10,000

Ion Diffusion Coefficient (cm^2/sec)[6]: 10^{-5}

Groundwater Velocity (ft/yr)[16,17,18]: 0, 1

Bed Porosity[19]: 0.34

Bed Density (g/cm^3)[20]: 0.423

Table 2

Total Release of Radioactivity from a 3-ft-thick Backfill in 10^3 Years (Model 1)

Backfill Sorption Coefficient Kd (ml/g)	Total Assumed Released from Backfill (Ci)	
	SA = $3.6 \times 10^4 (cm^2)$	SA = $3.6 \times 10^6 (cm^2)$
100	4×10^4	4×10^6[a]
1,000	4×10^0	4×10^2
10,000	5×10^{-46}	5×10^{-44}

[a] Artificial number indicating total inventory release.

V_g = 1 ft/yr; backfill thickness = 3 ft; leach rate (LR) = 10^{-3} g/cm^2-yr;

C_o = LR x SA x transit time/area under the concentration curves;

SA = surface area of the glass monolith.

Table 3

Fractional Release Rate from a 10-ft-and a 100-ft-Thick Backfill in 10^6 Years (Model 1)

Backfill Sorption Coefficient Kd (ml/g)	Fractional Release Rate From Backfill (/yr)			
	SA = $3.6 \times 10^4 (cm^2)$		SA = $3.6 \times 10^6 (cm^2)$	
	10-ft	100-ft	10-ft	100-ft
100	6.8×10^{-5}	6.7×10^{-5}	6.8×10^{-3}	6.7×10^{-3}
1,000	6.7×10^{-5}	6.0×10^{-5}	6.7×10^{-3}	6.0×10^{-3}
10,000	6.0×10^{-5}	3.6×10^{-9}	6.0×10^{-3}	3.6×10^{-7}

V_g = 1 ft/yr; leach rate (LR) = 10^{-3} g/cm^2-yr;

SA = surface area of the glass monolith.

Table 4

Fractional and Total Release from a 3-ft-Thick Backfill in 10^3 Years (Model 2)

Backfill Sorption Coefficient Kd (ml/g)	Fractional Release	Total Release (Ci)
100	9.97×10^{-1}	8.80×10^{5}
1,000	3.25×10^{-4}	2.90×10^{2}
10,000	5.93×10^{-50}	5.20×10^{-44}

V_g = 1 ft/yr, backfill thickness = 3 ft.

Table 5

Fractional Release Rate from a 3-ft-Thick Backfill and a 10-ft-Thick Backfill in 10^5 Years (Model 2)

Backfill Sorption Coefficient	Fractional Release Per Year (/yr)	
Kd (ml/g)	3-ft	10-ft
100	$>10^{-5}$	$>10^{-5}$
1,000	$>10^{-5}$	$>10^{-5}$
10,000	9.97×10^{-6}	1.85×10^{-6}

V_g = 1 ft/yr.

Table 6

Annual Release From a 200-ft-Thick Backfill for 10^6 Years, Assuming the Source of Term to be Total U.S. HLW Inventory (Model 1)

Backfill Sorption Coefficient Kd (ml/g)	Annual Release From Backfill (Ci/yr) SA = 2.4×10^5 (cm^2)	SA = 2.4×10^7 (cm^2)
100	4×10^2	4×10^4
1,000	3×10^2	3×10^4
10,000	3×10^{-80}	3×10^{-78}

V_g = 1 ft/yr; backfill thickness = 200 ft; leach rate (LR) = 10^{-3} g/cm^2 yr; SA = surface area of all the glass monolith considered.

Table 7

Annual Release From a 200-ft-Thick Backfill for 10^6 Years, Assuming the Source Term to be Total U.S. HLW Inventory (Model 2)

Backfill Sorption Coefficient Kd (ml/g)	Annual Release From Backfill (Ci/yr)
100	10^3
1,000	10^3
10,000	10^{-79}

V_g = 1 ft/yr.

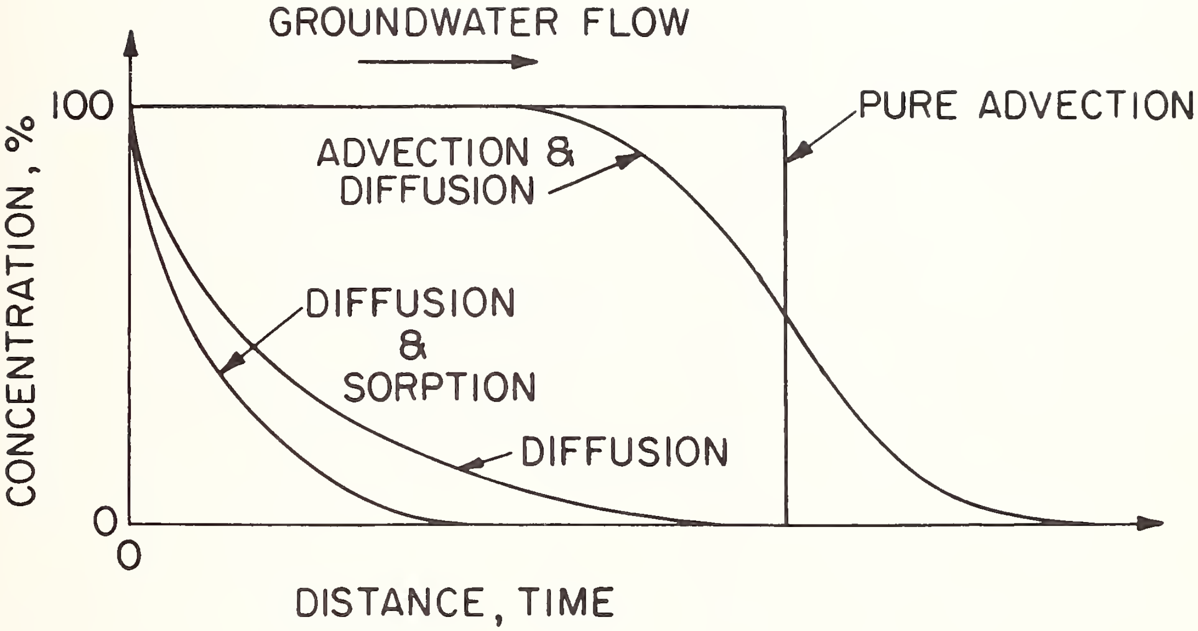


Figure 1. Principal radionuclide transport processes in sorptive backfill material [after Neretnieks [15]]

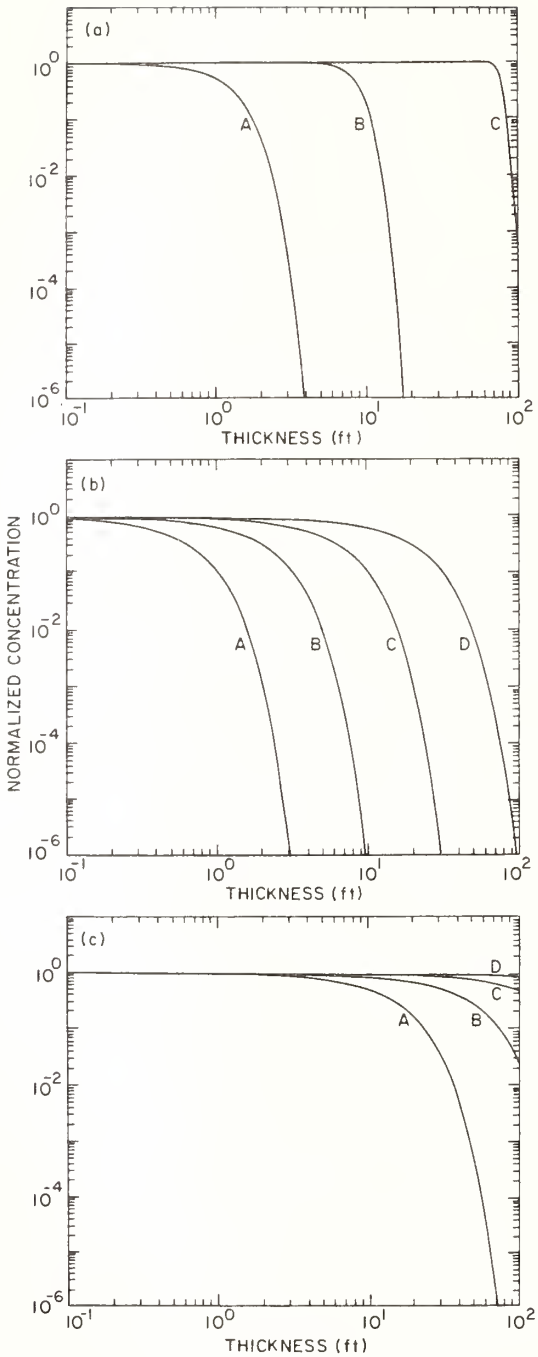


Figure 2. Concentration vs. bed thickness profiles for various transit times based on model 1: (a) K_d = 1000 ml/g, V_g = 1 ft/yr; (b) k_d = 1000 ml/g, V_g = 0 ft/yr; (c) k_d = 1 ml/g, V_g = 0 ft/yr; A = 10^3 yr, B = 104 yr, C = 105 yr, D = 106 yr

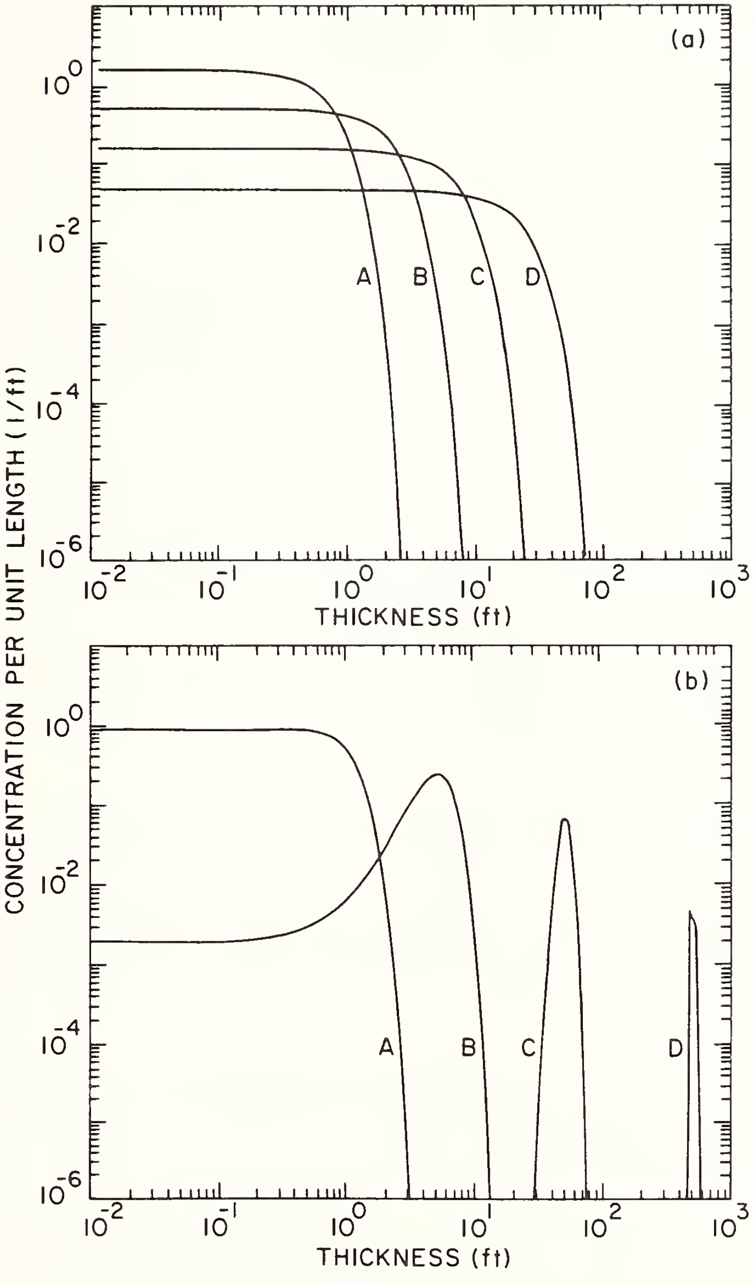


Figure 3. Concentration vs. bed thickness profiles for various transit times based on model 2: (a) K_d = 1000 ml/g, V_g = 0 ft/yr; (b) k_d = 100 ml/yr, V_g = 1 ft/yr; A = 10^3 yr, B = 10^4 yr, C = 10^5, D = 10^6 yr

APPENDIX 1

The simplified equation of the differential mass balance is

$$\frac{\delta c(x,t)}{\delta t} + \frac{V_g}{R_f}\,\frac{\delta c(x,t)}{\delta x} - \frac{D_f}{\sqrt{2}\,R_f}\,\frac{\delta^2 c(x,t)}{\delta x^2} = 0 \tag{1}$$

where

$R_f = \left(1 + \frac{\rho_d\ K_d}{\varepsilon} \right)$, the retardation factor

x = the distance from the interface of the glass and the zeolite along the direction of flow and longitudinal diffusion

t = time

$c(x,t)$ = liquid phase concentration, quantity of sorbing species per unit volume of liquid at distance along flow direction x and time t

ε = effective porosity of bed (fraction of bed volume containing flowing liquid)

V_g = average interstitial velocity of flowing water

$D_f/\sqrt{2}$ = coefficient of longitudinal fluid diffusion with a tortuosity factor to account for the tortuous diffusion path through the porous bed

ρ_b = the bulk packing density of solid sorbent, mass of solid per unit bed volume

K_d = the distribution coefficient for a linear sorption isotherm. The ratio of quantity of sorbed species per unit mass of solids to quantity of mobile species in the liquid phase per unit volume of liquid.

The boundary condition is

$$c(x = 0,\ t > 0) = C_o.$$

The initial condition is

$$c(x > 0,\ t = 0) = 0.$$

Define

$$U(x,t) = c(x,t) \cdot \exp\left[\frac{-V_g\, x}{\sqrt{2}\, D_f}\right] \cdot \exp\left[\frac{V_g^2 \cdot t}{2\sqrt{2} \cdot D_f \cdot R_f}\right]$$

Then, Equation (1) is reduced to a heat equation of U(x,t) as function of x and t. With the rearrangement of boundary and initial conditions, the Fourier transform is applied to the heat equation, and the subsequent convolution gives the following solution

$$c(x,t)/C_o = 1 - \frac{1}{2}\left[\operatorname{erfc}\left\{\frac{V_g \cdot \sqrt{t}}{2^{3/4}\sqrt{D_f \cdot R_f}} - \frac{\sqrt{R_f} \cdot x}{2^{3/4}\sqrt{D_f \cdot t}}\right\} - \exp\left\{\frac{\sqrt{2}\, V_g \cdot x}{D_f}\right\} \cdot \operatorname{erfc}\left\{\frac{V_g \cdot \sqrt{t}}{2^{3/4}\sqrt{D_f \cdot R_f}} + \frac{\sqrt{R_f} \cdot x}{2^{3/4}\sqrt{D_f \cdot t}}\right\}\right] \qquad (2)$$

where

erfc(y) = 1 - erf(y) and erf(y) is the error function of y.

The integration from x = backfill thickness to infinity is done numerically for the total release from backfill.

APPENDIX 2

The same differential equation is used as in model 1. The new constraint instead of the boundary condition is

$$\int_{0}^{\infty} C(x>0,\ t>0)dx = C_{total}$$

and the new initial condition is

$$C(x > 0,\ t = 0) = C_{total}\ \delta(x)$$

where $\delta(x)$ is a delta function of x and C_{total} is a total quantity of sorbing species.

The transformation technique may not be plausible in this case for the closed form solution. However, since we know the response of step function from appendix 1, the response of the delta function should be the derivative of the response of the step function with respect to x: the derivative of equation (2) of appendix 1. Further, we need the integral from x equals the backfill thickness of infinity, which is equivalent to the response function of appendix 1 (equation 2).

PRESENTATION OF SURVEYED RESULTS OF WORKSHOP QUESTIONNAIRE

A questionnaire containing ten questions covering a wide range of problems relating to the use of backfill was handed out to the participants during the workshop for them to fill out and return to NBS. A copy of the questionnaire is included in Appendix A of this report. Out of the twenty-eight non-NBS participants, eleven responded the request. It should be noted that the Canadian representatives only felt able to respond to the questions dealing with experimental techniques and equipment. They felt that all other questions are premature based on the current state of research and development. This point of view was echoed by a few other researchers at the national laboratories, as will be seen later in the report.

The surveyed results of all questions are given in this section. Names of the respondents are not spelled out in the presentation, but are identified by letter designation. Before the first question was asked, it was thought appropriate to ask the respondents to indicate their areas of expertise and the percentage of time which they have devoted to the nuclear waste problem. Table 1 summarizes their responses to this question. For those answered (7 out 11), the percent of time worked on this problem varies from 30 to 100 with 5 respondents working on full time basis. The areas of their expertises reflect a wide range of disciplines including chemistry, geology, civil engineering, seismicity, and metallurgy.

Question 1 asked "What do we expect from the placed backfill?" and proceeded to list seven functions that the organizing committee of the workshop believe the backfill should be able to perform. Four of the respondents did not answer this question. Three respondents completely accepted what was presented and made no comments to either modify the statements or add additional functional requirements for the emplaced backfill. The answers to Question 1 can be summarized as follows:

- It should serve as a hydrologic barrier to groundwater intrusion to the canister-waste form. It should also provide strong retardation of groundwater flow after intrusion.

- Its swelling and plasticity characteristics should enable it to seal cracks within the backfill and localized cracks in the surrounding rock to minimize the potential of groundwater intrusion.

- It should serve to buffer mechanical stresses and deformations.

- It should serve as a chemical conditioner of intruding groundwater by modifying pH, Eh, and chemical composition to minimize the corrosion potential of canisters.

- It should sorb and fix radionuclides to the extent possible.

- It should also be tailored specifically to retard or fix anionic chemical species.

- The backfill should be expected to be stable in the repository environment, i.e., it should be compatible with host-rock-imposed geochemical environment, with other barriers and waste form, and should have long term stability in expected radiation field.

- Finally, the backfill should be expected to perform all the necessary functions across a broad range of pressure, temperature and saturation conditions.

Question 2 requested the participants to list the criteria for the material properties they consider important for selection of the backfill and then distribute 100 points among these criteria based on their relative importance. In Question 3, eight (8) pertinent material properties were listed for the participants to distribute 100 points based on the relative importance. Furthermore, both questions asked the respondents to choose not more than four (4) material properties for preliminary screening purposes.

Nine out of eleven respondents answered Questions 2 and 3 and the results are presented in table 2. The numbers listed under the respondent are the points out of 100 which he/she has assigned to each material property and the numbers in the brackets are the priority of the material properties which the respondent chose for the preliminary screening process. In addition, a scoring system was devised as shown at the bottom of table 2 to tally the total points in the brackets scored on each material property. Consequently, the material properties can be ranked as shown on the last column in table 2. Thus, for preliminary screening purposes, the researchers would choose the backfill properties in the following order: sorption, hydraulic conductivity, thermal conductivity, chemical stability, swelling/shrinkage characteristics and plasticity, bearing capacity/shear strength, and diffusivity. Diffusivity was not on the original list but was brought up by Respondent B. The respondents also provided criticism of the method used in selecting material properties. One comment was that chemical stability does not belong to a list as given in Question 3 because if the material does not survive in a repository environment, all the other properties will then be meaningless. The issue of the interdependent nature of some of these material properties was also raised, e.g., the hydraulic conductivity of clay materials is closely related to swelling and plasticity and obviously the bearing capacity is related to shear strength of the material.

Question 4 was asked in two parts. The first part addressed the advantages and disadvantages of manufactured backfill with artificial additives versus various mixtures of natural materials. The second part asked the participants to indicate whether the location and type of parent material selected for the repository affect the answer to the first part of Question 4 and to the backfill evaluation in general. Nine out of eleven respondents responded to this question and their opinions are summarized in table 3. In short, artificial backfill has the advantages of ease in achieving better quality control and being available to be readily tailored for specific needs. However, it is disfavored for its comparably expensive nature, and most critically, for its lack of proof in providing long term stability in a repository environment for the required time period.

From the information revealed in table 3, it can be concluded that the natural materials are favored as the backfill mainly because they offer a much better chance to achieve long term chemical and thermodynamic stability, for the reasons that these materials have already been seriously tested under a geologic time frame. The answer of part II question to part I is very much divided. First of all, only 5 respondents responded to this part of the question. Among the five, two indicated affirmative and three negative, and none offered reasons to back up their responses. As to the part II question for the backfill evaluation in general, the answer is that the choice of backfill has to be repository dependent, particularly with respect to the overall chemical stability. One respondent indicated that this part of Question 4 was too broad and that the answer could go either way.

Question 5 was also asked in two parts. In the first part (Question 5(a)), the participants were asked to construct the ideal backfill using material such as montmorillionite or bentonite, zeolite, sand, crushed host rock materials, additives for sorption of selected radionuclides, and others that the participants may add to the list. Question 5(b) asked whether they think the design concept should change if factors such as availability, quality control, and cost are considered. Table 4 showed the constituents of the backfill envisioned by the eight respondents who answered this part of the question. Two of the respondents believe that it is too early to make such a selection. For those who made the selection, the consensus is that the backfill has to be site-specific and also to be dependent on design loading conditions. A question on possible thermal instability of zeolites during the thermal period of the repository was brought up. To summarize, it is clear that certain amount of clay materials will be included. Depending upon where the backfill will be placed, the amount of clays may vary from 5 to 100 percent. For example, the highly compacted pure bentonites have been considered for placement around the canister as a buffer, whereas backfills to be used to fill in the tunnels may only need 5 to 10 percent of the fine-grain materials. Because of the inexpensive nature of crushed host rocks and their many inherited favorable properties that they should be blended together with other ingredients to form the backfill.

Only 5 out of the eight respondents went further to answer Question 5(b) and their responses are given in table 5. Apparently, at the present thinking the major components of the desired backfill, i.e., clay, sand, and crushed hot rocks, are abundant in their natural existence and are relatively uniform in their composition, thus will not impose serious problems on availability, cost, and quality control. Of course, in the final design stage all these factors will have to be considered which means that some minor modifications of the backfill composition would be anticipated.

As commented by one respondent, Question 6 may well outline one of the most difficult problems needing to be resolved in order to use the backfill as an effective barrier. The question requested the participants to describe in their vision the placement procedures and anticipated difficulties in the installation of the backfill. One respondent disqualified himself/herself to assess the problem and another believed that the problem has to be site-specific without elaborating his/her comments. For those four who answered the question, their answers can be summarized separately as follows:

- Respondent B:
  - There is no identified major problem of backfilling in relatively dry rock.
  - If inlet of water is high, there may be some difficulties of containing swelling bentonite in the deposition hole before backfilling the tunnel.
  - Sealing water-bearing rock joints and drainage facilities may be necessary.
- Respondent E:
  - Possible procedures are briefly described below but not in the order of probability:
    - Pressed billets (donuts or hemi-cylinders) are inserted around the canisters or positioned in the hole prior to insertion of the canisters, or
    - Backfill is poured in under dry condition and be tamped around the canister, or
    - Backfill is poured in around canister in a slurry state, or

    - Backfill will be dry poured into borehole, tamped, and then core-drilled to accept canister.

  - All of the above four approaches have serious limitations, most of which affect the bulk properties of the backfill, especially homogeneity. Selection and evaluation of an emplacement procedure is one of the most important actions that could be taken at the present time.

- Respondent G:
  - Preformed blocks of compressed bentonite mixer, probably fabricated at the repository site, will be placed in the emplacement hole.
  - The canister/overpack assembly will be emplaced in the backfill line hole.
  - Gaps resulting from necessary tolerances will be filled with powdered bentonite.
  - A section of preformed bentonite mix will be placed over the top of the canister/overpack.
  - A ram, attached to the bottom loading transporter, will be braced against the tunnel roof and will compress the backfill into one solid piece surrounding the canister/overpack.
  - A temporary radiation shield, possibly a block of magnetite concrete, will be placed over the top of the emplacement hole.

- Respondent I:
  - Backfill must be pre-installed due to radiation protection and quality control requirements.

- The backfill also can not be pre-installed too long in advance in order to assure that size (opening for canister/overpack) is maintained, unless
    - If "overpack" becomes a sleeve around which backfill is packed, there could be a longer lag between backfill and waste emplacement.
    - Drilling 48" diameter holes of 20 ft deep that are both plumb and straight is difficult and expensive.
    - Dry, highly compacted backfill as in the Swedish concept will have to be kept away from water - a non-trivial task in an operating mine, even a "dry" one.
    - The simplest field placement would involve backfill outside a sleeve. The backfill would be rodded in place.

Question 7 requested the participants to indicate for a number of test variables the desired range/type as well as those actually used for tests already completed in their projects. These variables included temperature; hydraulic gradient and liquid/solid ratio of groundwater/leachant; the pH, Eh, species and concentration of ionic composition; corrosion products; and radiolysis products of the solution composition; mechanical stresses such as compression, swelling, shear, cyclic wetting/drying; particle size distribution and density of the backfill; and radiation dose rate and total dose. Answers from the respondents were summarized with respect to each variable as presented in tables 6 through 11.

Part (a) of Question 8 asked the participants about the test equipment used in evaluating the backfill properties including swelling, permeability, thermal conductivity, strength, sorption, and chemical analysis. Part (b) of the same question had the participants to describe the test methods and the availability of test procedures for evaluation of the above mentioned backfill properties. Tables 12 through 22 summarize the responses from those who answered the question.

Some general observations can be made from the answers to Questions 7 and 8: (a) there exists a wide range as well as many types of test variables which the respondents considered important considerations in their programs. This is another example of why the backfill probably has to be site-specific to best suit the waste loading as well as the hydrogeologic condition of the repository sites. It was also recommended by the researchers that at this stage of the work, ASTM Standards should be used wherever they are applicable. Some test methods, such as those used for hydraulic conductivity and swelling determinations, will require consensus among the researchers as to how they can be adopted for the nuclear waste standard work. Only a few organizations have started the material testing under high pressure and elevated temperature conditions. The test equipment is unique in each case and the method of testing will have to be developed during the execution of the preliminary test program.

Question 9 requested the participants to briefly discuss the change in backfill properties as a function of time and the participant's vision of the sequence of events happening to the backfill after its placement. They were also asked to identify major problem areas associated with the development of the backfill. Descriptions from the five respondents were quite similar and the focus was to address the mechanism of a hydrothermal interaction region within the emplaced borehole. Detailed description of the nature of the problem will be delayed to the next section where further research and development needs will be discussed.

Finally, table 23 summarizes the reference sources where information with regard to the characteristics of backfill materials are located. It also gives the most commonly used references in their research work. It can be seen that for each researcher who responded has preferences and their preferences are very much dependent upon his/her training, background, and familiarity with the reference sources and their accessibility.

Table 1

Area of Expertise and Percent of Devoted Time of the Respondents

Respondent	Area of Expertise	Percent of Time
A	o Backfill o Primary Waste Form o Radionuclide Migration	30
B	o Performance of Backfill	-
C	o Development of Test Procedures o Corrosion of Engineered Barriers	85
D	o Geochemistry	100
E	-	-
F	-	100
G	o Geology and Geochemistry	100
H	-	-
I	o Geochemistry o Rock Mechanics o Site Exploration o Seismology	100
J	-	-
K	o Geotechnical Engineering o Heat Transfer	50

Table 2

Relative Importance of the Backfill Material Properties

Material Properties	Respondent A	B	C	D	E	F	G	H	I	Total Points	Ranking
Hydraulic conductivity	15(3)		50(1)	(4)		20(1)	40(1)	(1)	5	26	2
Swelling/Shrinkage	10		20(2)	(3)		20(2)				12	5
Thermal Conductivity	10	(4)			(1)	5	10(4)	(3)	50(1)	19	3
Sorption	25(1)		10(4)	(2)	(3)	20(4)	30(2)	(2)	20(2)	28	1
Chemical Stability	25(2)			(1)	(2)	20(3)	(3)		15	17	4
Plasticity	5	(1)	20(3)			5	10		5	12	5
Bearing Capacity/ Shear Strength	10	(3)				10	10		5	7	6
Diffusivity		(2)								4	7

Notes:
a. Number of persons responded the questionnaire = 11.
b. Number of persons answered this question = 9.
c. Scoring system:

Level of Scoring	Scoring points
not scored	0
scored but not within top 4	1
(4)	2
(3)	3
(2)	4
(1)	5

Table 3 Question 4

Part 1: Advantages and Disadvantages of Manufactured Backfill with Artificial Additives versus Various Mixture of Natural Materials

Part 2: Does the Location and Type of Parent Material Selected for the Repository Affect the Answer to (a) Part 1 and to (b) Backfill Evaluation in General?

Respondent	Manufactured Backfill with Artificial Additives		Various Mixtures of Natural Materials		Answer of Question Part 2 to	
	Advantages	Disadvantages	Advantages	Disadvantages	Part 1	Backfill Explanation in General
A	o Good Quality Control o Availability	o Expensive o Problems in Geochemical Stability	-	-	-	Yes
B	o Can be tailored to enhance its role to serve as oxygen getter	o Interaction between different materials may result in reduced capability, e.g., plasticity	-	-	-	Yes
C	No comment	No comment	No comment	No comment	-	Choice of backfill has to be repository dependent in the sense that the same material properties will be considered but their value chosen will be different
D		Not an important issue to discuss	-	-	No	Yes, local conditions are important
E	o Minimize the compositional variation o More well-defined initial properties	o Much more expensive o Unknown long term stability o Inability to achieve some material properties, e.g., swelling	-	-	Yes	Yes
F	-	-	o Has less difficulty in assessing their stability in a host rock environment	-	Yes	Yes, this is most pertinent to the chemical stability of the backfill as well as the effect of groundwater composition, Eh, and pH, on sorption and chemical reactivity with key radionuclides
G	o More readily tailored to fit specific needs o More uniform	o Very few man-made materials have records of longevity exceeding 100 years o There is little evidence that the man-made materials will survive in the repository for the required time period	o Many of the natural materials have survived under conditions similar to those expected in a repository for millions of years.	-	No, the type of host rock, or its location will not change the answer to Question, Part 1	Yes, the specific geologic environment will effect the overall evaluation and selection process
H	-	-	o Materials from site (natural materials) may allow use of historical evidences for chemical stability evaluation	-	-	-
I	o Given sufficient time for testing, a variety of desirable additives could be included	o Extensive testing has to be undertaken to prove the suitability of the man-made material	o If material is known to be thermodynamically stable with respect to host rock and expected environment, nature has done the "long-term" experiment for us	o Some desirable proportions may be given up	No	Question is too broad, may be yes, may be no

Table 4

The Ideal Backfill Envisioned by the Respondents

Material	A	B	D	E	F	G	H	I
Clay - bentonite, montmorillonite	20	100 - 10	(a)	(d)	60	(d)	(e)	5 - 25
Zeolite	-	-	(b)	-	5	-	-	5 - 10
Sand	60	0 - 90	-	-				
Crushed host rock		may contain this	(c)	-	30	-	-	70 - 90
Additives for sorption of selected radionuclides	20 -apatite -ferrous minerals	-	-	-	5	-	-	1 - 5
Others	-	-	-	-	-	-	-	-

Notes:
(a) depend on temperature
(b) probably not stable through the thermal period
(c) certainly the least expensive bulk material
(d) too early for making selections
(e) suggested two commercially available bentonites as Standard Reference Materials

Table 5

Will the Design Concept Change if The Following Factors are Considered?

Respondent	Availability	Cost	Quality Control
A	No	No	No
E	Possibly	No	No
F	Current materials being considered are available in sufficient quantity and not prohibitively expensive	--	--
G	The final design will, if necessary, consider all of these factors	--	--
I	No	No	No

Table 6

Test Variable - Temperature

Respondent	Desired	Actually Used
A	25° - 200°C	-
B	-	20° - 90°C
C	60° - 250°C	-
D	-	-
E	-	-
F	65° - 300°C	65 - 300°C
G	25° - 350°C	25°
H	Best obtained from reference repository conditions published by OWWI	-
I	Not adequately defined at present in the U.S. program	-
J	25° - 150°	25° - 100°
K	100° - 150°C	100°C (in progress)

Table 7a

Test Variable - Groundwater/Leachant (Site-Specific)

Respondent	Desired	Actually Used
A	Granitic, Tuff, Basaltic groundwater, and Brine	-
B	-	"Allard" synthetic groundwater
C	-	-
D	-	-
E	-	-
F	Grando Ronde (proposed site groundwater)	Same as left
G	-	-
H	-	-
I	-	-
J	-	1. Synthetic granitic groundwater 2. Basaltic groundwater 3. WN-1 Saline solution 4. Standard Canadian sheild saline solution
K	Granitic water	tap H_2O, deionized H_2O

Table 7b

Test Variable, Groundwater/Leachant, Hydraulic Gradient

Respondent	Desired	Actually Used
A	-	-
B	-	None when in diffusivity measurement
C	Very low	-
D	-	-
E	-	-
F	-	-
G	10^{-2} to 10^{6}	10^{4} to 10^{6}
H	-	-
I	-	-
J	Not defined yet	-
K	50 - 500	50 - 100

Table 7c

Test Variable, Groundwater/Leachant, Liquid/Solid Ratio

Respondent	Desired	Actually Used
A	-	-
B	-	Realistic for density of highly compacted bentonite = 2.1 t/m^3 and mixture with 10 to 30 percent bentonite
C	Low under expected condition	-
D	-	-
E	-	-
F	-	-
G	-	-
H	-	-
I	-	-
J	Not defined yet	-
K	-	-

Table 8a

Test Variable, Solution Composition (site specific)

Respondent	Desired	Actually Used
A	-	-
B	-	-
C	° For certain repositories, specific solutions are available, e.g., Hanford GW, Brine A, Brine B, Tuffs related GW, and Granite GW	
	° Must test ambient oxygen pressure to very low oxygen to simulate open repository and conditons often sealing	-
D	-	-
E	-	-
F	-	-
G	-	-
H	-	-
I	-	-
J	-	-
K	-	-

Table 8b

Test Variable, Solution Composition, pH

Respondent	Desired	Actually Used
A	4 - 10	-
B	-	8 - 9
C	-	-
D	-	-
E	-	-
F	6 - 10	6 - 10
G	4 - 10	9.8
H	-	-
I	-	-
J	5.5 - 8.5	5.5 - 8.5
K	-	-

Table 8c

Test Variable, Solution Composition, Eh

Respondent	Desired	Actually Used
A	Aerated to Fe^{+2}/Fe^{+3}	-
B	-	-
C	-	-
D	-	-
E	-	-
F	-0.5 - + 0.5	-0.5 - +0.5
G	-0.2 - atmospheric	atmospheric
H	-	-
I	-	-
J	Fe_3O_4 - Fe_2O_3 Buffered	Air saturated H_2O
K	-	-

Table 8d

Test Variable, Solution Composition, Ionic Composition

Respondent	Species: Desired	Species: Actually Used	Concentration: Desired	Concentration: Actually Used
A	Ca^{+2}, Mg^{+2}, Na^{+}, K^{+}, $CO_3^{=}$, $SO_4^{=}$	-	-	-
B	-	-	-	-
C	-	-	-	-
D	-	-	-	-
E	-	-	-	-
F	-	-	-	-
G	Site-specific ambient groundwater for basalt and tuff plus solutions resulting from rock groundwater reaction at elevated temperature	Ambient groundwater from Hartford Basalt (Grande Ronde)	-	-
H	-	-	-	-
I	-	-	-	-
J	-	-	-	-
K	-	-	-	-

Table 8e

Test Variable, Solution Composition, Corrosion Products

Respondent	Desired	Actually Used
A	-	-
B	-	-
C	-	-
D	-	-
E	-	-
F	-	-
G	To be determined	-
H	-	-
I	-	-
J	Work is undefined	-
K	-	-

Table 8f

Test Variable, Solution Composition, Radiolysis Product

Respondent	Desired	Actually Used
A	-	-
B	-	Hydrogen
C	This can be answered only by radiation tests	-
D	-	-
E	-	-
F	-	-
G	To be determined	
H	-	-
I	-	-
J	Work is undefined	-
K	-	-

Table 9a

Test Variable, Mechanical Stresses, Compression (Lithostatic) Pressure

Respondent	Desired	Actually Used
A	-	-
B	-	-
C	-	-
D	-	-
E	-	-
F	300 bars (30 MPa)	-
G	0 - 30 MPa	-
H	-	-
I	-	-
J	$\leq$ 25 MPa	100 kPa - 20 MPa
K	up to 15 Mpa	None

Table 9b

Test Variable, Mechanical Stresses, Swelling

Respondent	Desired	Actually Used
A	-	-
B	-	Confined w/access to H_2O
C	-	-
D	-	-
E	-	-
F	-	-
G	0 - > 10 MPa	-
H	-	-
I	-	-
J	Undefined	-
K	-	-

Table 9c

Test Variable, Mechanical Stresses, Shear & Cyclic Wetting/Drying

Respondent	Shear		Cyclic Wetting/Drying	
	Desired	Actually Used	Desired	Actually Used
A	-	-	-	-
B	-	-	-	-
C	-	-	-	-
D	-	-	-	-
F	-	-	-	-
G	-	-	To be determined on site specific basis	
H	-	-	-	-
I	-	-	-	-
J	Undefined	-	-	-
K	-	-	-	-

Table 10a

Test Variable, Material Physical Properties, Particle Size Distribution

Respondent	Desired	Actually Used
A	-	-
B	-	-
C	-	-
D	-	-
E	-	-
F	-	-
G	To be determined	-
H	-	-
I	-	-
J	-	-
K	Clays to sands	Mixtures of crushed rock and clay

Table 10b

Test Variable, Material Physical Properties, Density

Respondent	Desired	Actually Used
A	-	-
B	-	2.1 t/m^3 (highly compacted bentonite)
C	-	-
D	-	-
E	-	-
F	-	-
G	To be determined	
H	-	-
I	-	-
J	-	-
K	1.6 to 2.2 g/cm^3	-

Table 11

Test Variable, Radiation Dose Rate for Total Dose

	Dose Rate		Total Dose	
Respondent	Desired	Actually Used	Desired	Actually Used
A	-	-	-	-
B	-	450 krad/hr	-	7 Mrad
C	~ 10^7 R/hr (no radiation tests done on backfill)			
D	-	-	-	-
E	-	-	-	-
F	-	-	-	-
G	4 x 10^4 R/hr	-	cumulative ~ 10^{10} R	-
H	-	-	-	-
I	-	-	-	10^9 R
J	Undefined		Undefined	
K	-	-	-	-

Table 12

Test Equipment Used to Evaluate Backfill Swelling

Respondent	Equipment
A	-
B	Permeameter described by Pusch
C	Dilatometry
D	-
E	-
F	Permeameter
G	Load cell attached to the PNL permeability cell
H	-
I	-
J	Applicable ASTM standards
K	o One dimensional consolidometer o Swelling chamber (Yong & Warkentin, 1975) o Automatic swelling pressure apparatus (Holland, 1968)

Table 13

Test Equipment Used to Evaluate Backfill Permeability

Respondent	Equipment
A	-
B	Permeameter described by Pusch
C	o Water migration - rigid volume flow cell o Radionuclide - modified version of above migration
D	-
E	-
F	Permeameter
G	PNL permeability cell
H	-
I	-
J	Applicable ASTM standards
K	o Constant head permeameter o Pressure decay permeameter

Table 14

Test Equipment Used to Evaluate Backfill Thermal Conductivity

Respondent	Equipment
A	-
B	"Transient hot strip" method by Prof. Lunden
C	Cut - Bar thermal conductivity apparatus
D	-
E	-
F	Thermal probe
G	Under study to determine best equipment and techniques for application to backfill materials
H	-
I	-
J	o Transient heat probe o Guarded hot plate (steady state method)
K	o Transient hot probe o Guarded hot plate (steady state method)

Table 15

Test Equipment Used to Evaluate Backfill Strength

Respondent	Equipment
A	-
B	Described by Pusch
C	Mechanical compression test
D	-
E	-
F	Triaxial test
G	Standard uniaxial and triaxial test equipment
H	-
I	-
J	-
K	Triaxial compression apparatus

Table 16

Test Equipment Used to Evaluate Backfill Sorption

Respondent	Equipment
A	-
B	-
C	-
D	-
E	-
F	Teflon tubes, autoclaves
G	Column (retardation) techniques are being developed for use in PNL permeability cell
H	-
I	-
J	-
K	-

Table 17

Test Equipment Used to Perform Backfill Chemical Analysis

Respondent	Equipment
A	-
B	-
C	-
D	-
F	XRD, STEM, SEM, ICP, Chromatograph, AA
G	ICP, AA, XRF
H	-
J	-
K	-

Table 18

Test Methods and Procedures for Backfill Swelling Evaluation

Respondent	Test Method	Test Procedures
A	-	-
B	Described by Prof. Pusch	Described by Prof. Pusch
C	o Measured by mechanical testing o Creep can also be measured by standard method	- -
D	-	-
E	-	-
F	-	-
G	Use of a load cell attached to the PNL permeameter	Under development
H	-	-
I	-	-
J	Applicable ASTM standards	Applicable ASTM Standards
K	o Stress-controlled method o No volume change method	o To be developed o Available

Table 19

Test Methods and Procedures for Backfill Hydraulic Conductivity Evaluation

Respondent	Test Method	Test Procedures
A	-	-
B	Described by Prof. Pusch	Described by Prof. Pusch
C	Hydraulic conductivity is a "standard" method of assessing; extrapolation to low pressure is overly conservative	Variety of techniques is available in open literature
D	-	-
E	-	-
F	-	-
G	PNL permeability cell	Under development
H	-	-
I	-	-
J	Applicable ASTM standards	Applicable ASTM standards
K	o Steady-state flow method o Flow velocities using tracers	o Available o Available

Table 20

Test Methods and Procedures for Backfill Thermal Conductivity Evaluation

Respondent	Test Method	Test Procedure
A	-	-
B	"Transient hot strip" method by Prof. Lunden	See left
C	Relatively straight forward compared to other properties (importance diminishes with emplacement time, i.e., long term assessment is not that critical)	-
D	-	-
E	-	-
F	-	-
G	Most test method are relatively standard	-
H	-	-
I	-	-
J	-	-
K	Transient line source method	Available

Table 21

Test Methods and Procedures for Backfill Strength Evaluation

Respondent	Test Method	Test Procedure
A	-	-
B	Method described by Prof. Pusch	See left
C	Mechanical testing	-
D	-	-
E	-	-
F	-	-
G	Most test methods are relatively standard	-
H	-	-
I	-	-
J	Applicable ASTM standards	Applicable ASTM standards
K	Triaxial testing	Available

Table 22

Test Methods and Procedures for Backfill Sorption Evaluation

Respondent	Test Method	Test Procedures
A	-	-
B	-	-
C	K_d measurement; its primary difficulty is the specification of proper, relevant boundary conditions, e.g., Eh, pH. Tests will have to be conducted over a range of both variables.	-
D	-	-
E	-	-
F	-	-
G	Column (retardation) technique	Under development
H	-	-
I	-	-
J	-	-
K	-	-

Table 23

Reference Sources for Backfill Properties

Respondent	Reference Sources Regarding Backfill Characteristics	References Used Most Often
A	-	-
B	-	-
C	o No comprehensive data compilation is available o Proc. of Material Research Society Annual Meetings o American Chemical Society Meetings o Nuclear and Chemical Waste Management	o ASTM Standards o ASTM Geotechnical Testing Journal o Technical Reports from ANSL, SNL, ORNL, LLL, SRL, and PNL
D	-	-
E	-	-
F	o Swedish Documents	o Swedish Reports
G	-	-
H	-	-
I	-	-
J	-	-
K	o KBS reports o AECL documents	o Proc. of American Society of Soil Sciences o International Journal of Heat and Mass Transfer o Soil Sciences o Transactions, Geophysical Union o ASCE Geotechnical Engineering Division

SUMMARY OF FUTURE RESEARCH AND DEVELOPMENT NEEDS

Areas in which research and development of the backfill component are required were discussed during the informal discussion sessions on the second day of the workshop. The content of the discussion can be divided into two categories, the test methods used in the laboratory for evaluation of the backfill properties including hydraulic conductivity, swelling pressure, diffusivity, and thermal conductivity; and the study of backfill properties in a hydrothermal region in a repository environment. Measurement of sorption of radionuclides and radiation effects were dropped from the discussion since much research has been focused on the former and for the lack of expertise on the latter subject among those participating in the discussion.

Table 1 summarizes the current status of laboratory testing to determine swelling and hydraulic conductivity, as reported by the workshop participants. In addition, the method of specimen preparation used in hydrothermal and wetting-drying studies by one institution was included in the tabulation. As it can be seen, the tests were conducted under a variety of conditions that make comparisons of the test results difficult. Differences in testing conditions include the type of backfill, the amount of clay used in the specimen, specimen size, initial density, moisture content at the time of compaction, the type of water used, and the method of specimen compaction.

Various types of bentonites, silica or quartz sand (or fine aggregates), and crushed host rocks were used as the candidate backfill. Depending on where the backfill will be placed (either adjacent to the canister in the borehole called buffer in many studies, or for backfilling the tunnels), the amount of clay soils used was varied from 10 to 100 percent. The size of specimens used in testing was closely related to the method of compaction and varied from 25 mm to 102 mm in diameter and 5 mm to 115 mm in height. The height of the specimen is also dictated by the time required to saturate the specimen prior to hydraulic conductivity measurements. For highly compacted samples, very long time periods are required to reach the saturation. This is the reason why the specimen height used in highly compacted materials is very small, 5 to 10 mm. Even with this short dimension, the researchers have reported that the specimen would still need up to 3 weeks to reach complete saturation. Great variation is also shown in the initial density of the specimens used in the testing, which was reported to vary from 1.1 to 2.2 g/cm^3.

In some cases, air-dried materials with moisture content as low as 6 to 10 percent were used in preparing the specimens and in other cases the moisture content was as high as 30 percent. The researchers addressed the difficulties encountered in compacting specimens having moisture content beyond 25 percent. In addition, high initial moisture content can result in a nonhomogeneous moisture distribution within the specimen. The type of water used to wet the soil varies considerably and may be site specific. Tap water, distilled water, deionized water; brine,and granitic, basaltic, tuff, or synthetic groundwater were used.

Four different compaction techniques can be used for specimen preparation. They are impact, vibratory, kneading, and static compaction. Other than the vibratory method, the other three methods were listed in table 1. The ASTM D1557 method, which is the modified Proctor, is an impact method in which a 4.54 kg rammer is dropped from a height of 457 mm for a specified number of drops for each layer placed in a compaction mold. The Harvard miniature compaction is a kneading method. At present, the uniaxial static compaction method is probably the most popular technique used for backfill specimen preparation. It can be conducted by using any press machine.

Another variable which was not included in the table is the temperature. Conflicting results were obtained on the effect of temperature on the properties of swelling and hydraulic conductivity. Lambe in 1960 [1][1/] found that the samples under consolidation in consolidation tests swelled and compressed with decreases and increases in temperature, respectively. However, a study by Yong et al., [2] of the swelling pressure of Na-montmorillonite at below room temperature resulted in the reverse effect. Further studies by Yong [3] on pure clays at and above room temperatures (25 to 45°C) indicated an increase

---

[1/] References are listed at the end of this section.

in swelling pressure when the temperature was increased and thus was consistent with his earlier findings [2]. Recent studies on bentonite by Pusch [4] concluded that at 90°C the swelling pressure is lower than that at room temperature. Although he indicates that the number of tests was too small to give any definite relationship between swelling pressure and temperature, Pusch thinks that a 50 percent reduction in swelling pressure should be expected when the temperature is raised from 20°C to 90°C. Pusch [5] also concluded from his test results that at a 70°C temperature, the hydraulic conductivity is estimated to be 5 to 10 times higher than that at 20°C for both the Na and Ca bentonites used in this studies.

Although it is impossible to unify the methods that should be used for the determination of the swelling and hydraulic conductivity of the backfill, the test variables should be limited in order to make the comparison of test results meaningful. It was the consensus among the participants that for simple routine purposes, standard test methods, such as those specified in the ASTM standards, should be followed. It should be noted that no ASTM standard is available for the evaluation of swelling potential of the material; however, methods such as free swelling testing and swellconsolidation tests have been routinely used in the engineering analysis. A good summary of methods used by the engineering profession can be found in a paper presented by Heinz in 1981 [6].

Due to the intimate relationship between hydraulic conductivity and swelling, it would be desirable to evaluate both properties in one test set up. Some studies have been conducted by coupling these two measurements, such as the work by Pusch [4,5] and Wheelwright et al. [7] presented in this workshop. Testing of these properties at the temperature and pressure comparable to the condition in a real repository should also be conducted. Obviously no standard can be referred to for conducting these special types of testing, therefore, a close working relationship among the researchers should be sought in order to develop meaningful and reliable measurement data for the design of the long term stability of the backfill in a repository environment.

One compromise which has to be made in laboratory determination of hydraulic conductivity is the application of rather high hydraulic gradient across the specimen during testing to reduce the time required for the measurement. Nevertheless, the hydraulic conductivity determined using fictitiously high gradient should result in a conservative measure of the estimated breakthrough time of the multibarrier isolation system.

For backfill constituted of compacted clay materials such as bentonite, the hydraulic conductivity of the barrier will be very low and will probably limit the flow rate to a negligible amount. In this case, the minimum significant flow rate can be reasonably determined by the estimated rate of molecular diffusion in the aqueous phase. In other words, the molecular diffusion of radionuclides rather than convective transport will be the predominant mechanism for migration if the flow rate through the barrier is small enough. Therefore, the mechanisms of diffusion should be thoroughly studied. A one-dimensional linear mathematical model has been developed by Nowak [8] and Pusch [9], however, only bulk diffusion was considered in the simulation. Surface diffusion, which is another contributing mechanism, is identified as one area where further research is required [10].

The backfill surrounding the canister will have to act as a heat transfer medium since the decay of the radionuclides in the waste form generates heat. A backfill with sufficient thermal conductivity would help to distribute the heat faster and more uniformly into the host rock mass, thus would prevent excessive heat buildup in the canister. Designing a backfill with sufficient thermal conductivity to prevent excessive heat buildup under a given waste loading condition is a very intriguing problem since the backfill is also expected to serve as a medium to slow down the flow of groundwater toward the canister.

Several parameters have been cited in the literature to affect the thermal conductivity of a soil (backfill). They include soil composition and the geometrical arrangement of soil particles, density, moisture content, and temperature gradient between the backfill and the surface of the canister. Up to this point, either air was assumed to fill the annulus between the canister and the borehole wall [11], or a low but fixed value of thermal conductivity [12] has been recommended for the heat transfer calculation. Although this approach is conservative, a better simulation should be pursued considering the parameters listed above which have direct effect on the magnitude of thermal conductivity. Obviously, the backfill placed in a repository environment will go through thermal properties changes with time since its composition may change due to chemical alternation; the geometrical arrangement of soil particles and density may change due to the application of lithostatic pressure and swelling when it has the access to groundwater; its moisture content will change from being dried out initially to eventually being saturated; and the decreasing thermal gradient across the backfill throughout the decay of the radionuclides.

At present, two methods are used in the laboratory to determine the thermal conductivity of a soil: steady-state heat flow measurement and transient heat dissipation measurement. The steady-state method has several disadvantages. Firstly, it introduces measurement due to migration of moisture in the direction of thermal gradients in a moist soil. It also creates convection currents in a saturated medium. Finally, the method is time consuming. The transient method is generally free of the disadvantages mentioned above. Furthermore, the measurement method is relatively simple and fast. For these reasons, the transient method is favored for the measurement of the thermal conductivity of the backfill.

It was also the consensus among the participants that both mathematical modeling and experimental work should be carried out to study the backfill properties in a hydrothermal environments generated by the multi-barrier isolation system. The hydrothermal region of the backfill is graphically depicted in figure 1. The region is confined by the metal surface of the canister on one side and borehole wall on the other. The curve shown to the left of the borehole wall is the average hydraulic gradient within the backfill as a function of time. During the operational period of the repository when the repository is still open, the water table around the repository is lowered by pumping. It is only after the closure of the repository that the hydraulic gradient will be gradually restored to the boundary at the borehole wall because of the restoration of the groundwater table. The curve on the right of the surface of the canister depicts the decay of thermal loading with time. The two dashed curves across the backfill indicate the cross-coupling of the thermal gradient and hydraulic gradient. It is conceivable that the backfill will dry out immediately after the emplacement of the wastes. This is considered to be the most critical period for the backfill with regard to its long term capability to function as intended. Remember that no groundwater will be present during the operational period of the repository. Thus the degree of drying out of the backfill would be completely dependent upon the design of waste loading in the canister, and the type of soil used as the backfill, its density, and the moisture content used in placing the backfill.

After the closure of the repository, and dependent upon both site geology and hydrology, groundwater will eventually reach the backfill. This is probably the time when the hydraulic gradient across the backfill is the greatest. The rate at which groundwater will be permeating and saturating the backfill is a very complicated problem because of the existence of the thermal gradient, and is one of the major unsolved problems in the design of using backfill as one barrier. With the gradual reduction of thermal gradient with time, the backfill will eventually be saturated. At that time, the backfill will exist in an aqueous environment influenced by reactions between groundwater and host rock, between groundwater and the metal canister, and between groundwater and the waste forms. What reactions that may affect the properties of the backfill after saturation and the reaction rates remain to be determined.

The following is a summary of the discussions presented above:

1. For routine determination of hydraulic conductivity, ASTM standards should be followed.

2. For evaluation of hydraulic conductivity under repository condition, no standard method is available. Close working relationship among researchers should be sought in order to provide a better basis for comparison of the test results.

3. There is also no standard method on the evaluation of swelling potential of fine-grained soils. Several techniques have been popularly used among

the researchers. Equipment has also been developed to incorporate the measurements of both hydraulic conductivity and swelling. Again, close working relationship among researchers is essential in developing reliable material properties for the design of long term stability of the backfill barrier.

4. The mechanism of diffusion should be further studied. Surface diffusion, which has been a neglected area because most of the studies up to the present have focused on the bulk diffusion, should be evaluated.

5. Thermal properties of the backfill should be better defined and measured as a function of its material type, the geometrical arrangement of its particles, its moisture content at the placement, its density, and the thermal gradient imposed across the backfill under a specific design waste loading.

6. Finally, both mathematical modeling and experimental work should be carried out to study the backfill properties in a hydrothermal environment developed in a multi-barrier waste isolation system.

References

1. Lambe, T.W., Compacted Clay: Engineering Behavior Transaction ASCE, 125, pp. 718-741, (1960).

2. Yong, R.N., Taylor, L.O. and Warkenton, B.P., Swelling Pressures of Sodium Montmorillonite at Depressed Temperatures, Proc. 11th National Conference on Clays and Clay Minerals, (1962).

3. Yong, R.N., The Swelling of a Montmorillonitic Clay at Elevated Temperatures, Proc. 3rd Asian Regional Conference on SMFE, (1967).

4. Pusch, R., Swelling Pressure of Highly Compacted Bentonite, KBS Technical Report 80-13, (1980).

5. Pusch, R., Permeability of Highly Compacted Bentonite, KBS Technical Report 80-16, (1980).

6. Heinz, W.E., A Means of Predicting Swelling Pressures of Soils Found in the Rocky Mountain Area, M.S. Thesis, University of Colorado, (1980).

7. Wheelwright, E.J., Hodges, F.N., Westsik, J.H. Jr., and Bray, L.A., Pacific Northwest Laboratory's Backfill Development Project, This Publication, (1983).

8. Nowak, E.J., The Backfill Barrier as a Component in a Multiple Barrier Nuclear Waste Isolation System, SAND 79-1109, (1979).

9. Pusch, R., Bentonite Shielding of Rock-Deposited Radioactive Wastes, This Publication, (1983).

10. Nowak, E.J., Backfill Barrier Research and Development for Radioactive Waste Isolation in Salt, Abstract Only, This Publication, (1983).

11. Draft Report, "Interime Reference Repository Conditions for Spent Fuel and Commercial High-Level Nuclear Waste Repositories in Tuff," Prepared by The Reference Repository Conditions Interface Workshop Group, (1980).

12. Radhakrishna, H.S., Evaluation of Thermal Properties of Buffer Materials for a Deep Underground Nuclear Waste Disposal Vault, This Publication, (1983).

Table 1

Summary of Test Methods, Specimen Size, Methods of Specimen Preparation, Specimen Composition, and Test Equipment Used in the Study of Swelling, Hydraulic Conductivity, and Hydrothermal Effect of Backfill Materials

Institution	Mat'l Used	Clay Fraction	Method of Compaction	Moisture Content	Specimen Size	Initial Density	Swelling	Hydraulic Conductivity
		%		%	mm	g/cm^3	MPa	cm/sec
A	o Wyoming Montmorillonite	100	No compaction, free swelling	N/A	N/A	N/A	N/A	N/A
B	o Bentonite o Quartz	o 100 o 10-20[2]	Uniaxially and statically compressed	Air-dried 10	o Swelling: 50Dx10-40H o Hyd.Cond: 50Dx5H (1 day to saturate)	1.8-2.2	5-10	10^{-11}-10^{-12}
G	o Na-Bentonite o 80-120 mesh sand	25 50 100	Uniaxially and statically compressed	0 5.8 7.5 13.5	25Dx10H (1-3 weeks to saturate)	2.1	N/A	2×10^{-9}-7×10^{-13}
J	o Bentonite o Crushed o Granitic Rock o Fine Aggregate	10-20	N/A	N/A	N/A	N/A	N/A	N/A
K	o Bentonite o Silica Sand o Crushed Granitic Rock	15-50	ASTM D-1557	5-30	102Dx115H	1.1-2.2	N/A	N/A
L	o 3 types of Bentonites	100	Harvard miniature compaction	Varied but not specified	33Dx72H	N/A	[3]	[3]

1. Types of water used in the study included the following: tap water, distilled water, deionized water, brine, granitic groundwater, basaltic groundwater, tuff groundwater, and synthetic groundwater
2. Clay fraction for backfill around canister
3. For hydrothermal and wetting - drying study

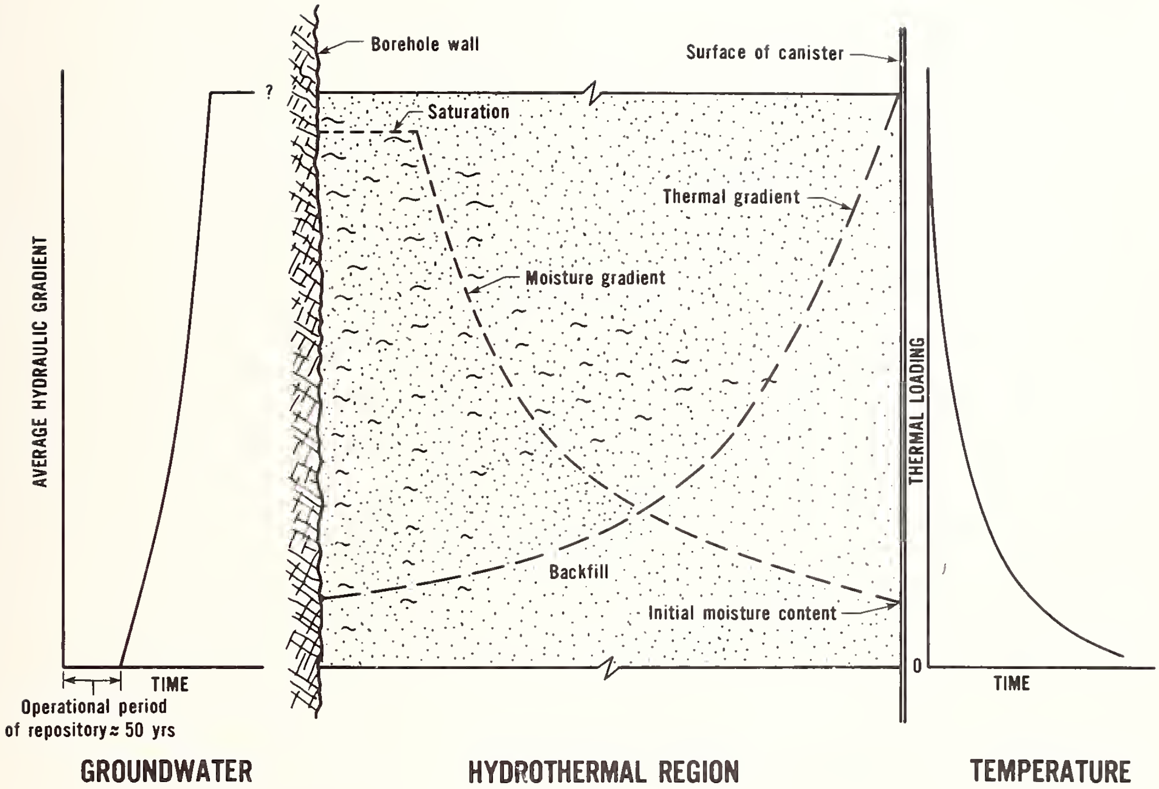


Figure 1. Hydrothermal region of the backfill in a repository environment

APPENDIX A

NATIONAL BUREAU OF STANDARDS

Questionnaire

Nuclear Waste Management
Deep Geologic Repositories
Backfill Selection and Evaluation

To get the maximum benefit from your participation in this workshop, we are soliciting your concise comments on the questions given herein before adjournment on April 14, 1981.

Name: ____

Affiliation: __________________________________

Address: ______________________________________

Telephone Number: ______________________________

Area of Expertise Relating to Nuclear Waste Management:

Percent of Time Devoted to Nuclear Waste Problem:

1. What do we expect from the placed backfill in addition to that given below:

    - Hydrologic barrier to groundwater intrusion
    - Its plastic nature will seal localized cracks in the surrounding rock to minimize the potential of groundwater intrusion
    - To buffer mechanical stresses and deformations
    - To provide suitable bearing support and thus minimize subsidence of the waste package
    - To serve as an adequate thermal transporting medium
    - To serve as a chemical conditioner of intruding groundwater by modifying pH, EH and chemical composition
    - To serve as a radionuclide sorptive barrier
    - Others

2(a) List criteria for the material properties you consider important for selection of backfill and then distribute 100 points among the criteria based on their relative importance.

Criteria	Points

2(b) For preliminary screening purposes, which of the above criteria would you use.

3(a) Distribute 100 points based on the relative importance of the following backfill material properties.

Material Properties	Points
Permeability	
Swelling	
Thermal conductivity	
Sorption	
Chemical Stability	
Plasticity	
Bearing capacity	
Shear strength	

3(b) For preliminary screening purposes, which of the above properties would you use (no more than 4).

4(a) What are the advantages and disadvantages of manufactured backfill with artificial additives versus various mixtures of natural materials?

4(b) Do you feel the location and type of parent material selected for the repository affect your answer to Question 4(a) and to the backfill evaluation in general?

5(a) Combining the materials given below and others, please construct the ideal backfill you can envision:

- Clays - montmerillonite, bentonite
- Zeolites
- Sand
- Crushed host rock materials
- Additives for sorption of selected radionuclides (getters) or Eh, pH control
- Others

5(b) Will the design concept change if you consider such factors as:

Availability:

Quality Control:

Cost:

6. Describe in your vision the placement procedures and anticipated difficulties in the installation of the backfill.

7. What is the ranges in the test variables listed within which laboratory testing should be conducted.

Test Variables	Range/Type	
	Desired	Actually used for tests already completed in your project.
° Temperature		
° Groundwater/leachant (site specific)		
- hydraulic gradient		
- liquid/solid ratio		
° Solution Composition (site specific)		
- pH		
- Eh		
- ionic composition		
species		
concentration		
- corrosion products		
- radiolysis products		
° Mechanical stresses		
- compression (lithostatic) pressure		
- swelling		
- shear		
- cyclic wetting/drying		
° Material physical properties		
- particle size distribution		
- density		
° Radiation dose rate and total dose		
° Others		

8(a) List test equipment, if known, used in evaluating the following backfill Properties:

Properties	Equipment
Swelling	
Permeability	
Thermal conductivity	
Strength	
Sorption	
Chemical analysis	
Others	

8(b) Describe the test methods used in evaluating the backfill properties:

Property	Test Methods	Availability of Test Procedures
Swelling		
Permeability		
Thermal conductivity		
Strength		
Sorption		
Chemical analysis		
Others		

9. Briefly discuss the change in backfill properties with time and your vision of sequence of event happening to the backfill after its placement. Identify major problem areas associated with the development of backfill.

10(a) Please list reference sources that would be helpful in the collection of information regarding the backfill characteristics.

10(b) What references do you use most often?

APPENDIX B

Members of the Organizing Committee:

Mr. William P. Reed

Acting Program Manager
Nuclear Waste Management Program
National Measurement Laboratory

Dr. Riley M. Chung
Dr. Felix Y. Yokel
Mr. Lawrence A. Salomone

Geotechnical Engineering Group
Structures Division
Center for Building Technology
National Engineering Laboratory

List of Participants

Workshop on Research and Development Needs in Backfill for Long Term Storage of Nuclear Waste in Deep Geologic Repositories, April 13 and 14, 1981 at the National Bureau of Standards.

Organizing Committee:

Riley M. Chung	National Bureau of Standards Building 226, Room B162 Washington, D.C. 20234	FTS-921-2137 301-921-2137
William P. Reed	National Bureau of Standards Building 221, Room B320 Washington, D.C. 20234	FTS-921-3868 301-921-3868
Lawrence A. Salomone	National Bureau of Standards Building 226, Room B162 Washington, D.C. 20234	FTS-921-3128 301-921-3128
Felix Y. Yokel	National Bureau of Standards Building 226, Room B162 Washington, D.C. 20234	FTS-921-2648 301-921-2648

Participants:

Name	Organization and Address	Telephone No.
Erik D. Anderson	National Bureau of Standards Building 226, Room B162 Washington, D.C. 20234	FTS-921-3180 301-921-3180
Gary W. Beall	Radian Corporation 8500 Shoal Creek Road Austin, TX 78766	512-454-4797 ext. 5631
Anders Bergstrom	SKBF/KBS Box 5864 S-102 48 Stockholm Sweden	08-67 95 40
D. J. Cameron	Atomic Energy of Canada Limited Whiteshell Nuclear Research Establishment Pinawa, Manitoba ROE1LO, Canada	204-753-2311
Steven Cheung	Atomic Energy of Canada Limited Whiteshell Nuclear Research Establishment Pinawa, Manitoba ROE1LO, Canada	204-753-2311

Name	Organization and Address	Telephone No.
Karen P. Coccia	National Bureau of Standards Building 225, Room A140 Washington, D.C. 20234	FTS-921-3395 301-921-3395
Ramesh Dayal	Brookhaven National Laboratory Department of Nuclear Energy Nuclear Waste Management Div. Upton, NY 11973	FTS-666-4993 516-345-4993
Bruce R. Erdal	Los Alamos National Laboratory CNC7 Mail Stop 514 Los Alamos, NM 87545	FTS-843-5338 505-667-5338
John T. Greeves	U.S. Nuclear Regulatory Commission Mail Stop SS-905 Washington, D.C. 20555	FTS-427-4177 301-427-4177
Floyd Hodges	Battelle Pacific Northwest Laboratory P.O. Box 999 Richland, WA 99352	FTS-444-4467 509-444-4467
J. Keith Johnstone	Sandia National Laboratories Division 4537 Albuquerque, NM 87185	FTS-844-7633 505-844-7633
William D. Kovacs	National Bureau of Standards Building 226, Room B162 Washington, D.C. 20234	FTS-921-2885 301-921-2885
M. D. Merz	Nuclear Waste Material Characterization Center Battelle Northwest P.O. Box 999 Richland, WA 99352	FTS-440-2943 509-373-2943
Donald P. Moak	ONWI Battelle Memorial Institute 505 King Avenue Columbus, OH 43201	FTS-976-7125 614-424-7125
Judith Moody	ONWI Battelle Memorial Institute 505 King Avenue Columbus, OH 43201	FTS-976-5536 614-424-5536
E. James Nowak	Sandia National Laboratories P.O. Box 5800, Div. 5812 Albuquerque, NM 87185	FTS-844-1109 505-844-1109
Edward O. Pfrang	National Bureau of Standards Building 226, Room B370 Washington, D.C. 20234	FTS-921-2196 301-921-2196

Name	Organization and Address	Telephone No.
C. L. Pittiglio	U.S. Nuclear Regulatory Commission Mail Stop SS-905 Washington, D.C. 20555	FTS-427-4177 301-427-4177
Roland Pusch	Division of Soil Mechanics University of Lulea Lulea, Sweden	46-920-91-357
H. S. Radhakrishna	Ontario Hydro Research 800 Kipling Avenue Toronto, Canada M8Z 5S4	416-231-4111 ext. 6695
Larry Ramspot	Lawrence Livermore National Laboratory Mail Code 204 P.O. Box 808 Livermore, CA 94550	FTS-422-4176 415-422-4176
Della M. Roy	Material Research Laboratory Pennsylvania State University University Park, PA 16802	814-865-1196
Rustum Roy	Material Research Laboratory Room 203 Pennsylvania State University University Park, PA 16802	814-865-3421
Tom Schmitt	U.S. Nuclear Regulatory Commission Mail Stop NL-5650 Washington, D.C. 20555	FTS-443-5976 301-443-5976
James Schornhorst	Westinghouse Electric Company Advanced Energy Systems Box 10864 Pittsburgh, PA 15236	412-892-5600
Ronald A. Schrack	National Bureau of Standards Building 245, Room B110 Washington, D.C. 20234	FTS-921-2677 301-921-2677
Trueman Seamans	U.S. Nuclear Regulatory Commission Mail Stop SS-905 Washington, D.C. 20555	FTS-427-4177 301-427-4177
A. J. Sethi	Geotechnical Research Center McGill University 817 Sherbrooke Street, W Montreal, PQ. Canada	514-392-8295

Name	Organization and Address	Telephone No.
Dale Simpson	Lehigh University Dept. of Geological Sciences Williams Hall #31 Bethlehem, PA 18015	215-861-3664
Elizabeth N. Vine	Los Alamos National Laboratory Los Alamos, NM 87545	FTS-843-4845 505-667-4845
Earl J. Wheelwright	Battelle Pacific Northwest Laboratory P.O. Box 999 Richland, WA 99352	FTS-444-3441 509-444-3441
M. I. Wood	Rockwell Hanford Operation Basalt Waste Isolation Project Columbia Bank Building Richland, WA 99352	FTS-444-7833 509-376-7833
Howard T. Yolken	National Bureau of Standards Building 220, Room A259 Washington, D.C. 20234	FTS-921-3775 301-921-3775
Raymond N. Yong	Geotechnical Research Center McGill University 817 Sherbrooke Street, W. Montreal, PQ. Canada H3A 2K6	514-392-4751

NBS-114A (REV. 2-80)

U.S. DEPT. OF COMM. **BIBLIOGRAPHIC DATA SHEET** *(See instructions)*	1. PUBLICATION OR REPORT NO. NBS SP 668	2. Performing Organ. Report No.	3. Publication Date December 1983

4. TITLE AND SUBTITLE

Nuclear Waste Management Program Summary Report
(Workshop on Research and Development Needs in Backfill for Long Term Storage of Nuclear Waste in Deep Geologic Repositories, held at the National Bureau of Standards, Washington, DC, April 13-14, 1981)

5\.

Riley M. Chung, Editor

6\. PERFORMING ORGANIZATION *(If joint or other than NBS, see instructions)*

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, D.C. 20234

7\. Contract/Grant No.

8\. Type of Report & Period Covered

Final

9\. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS *(Street, City, State, ZIP)*

Office of Nuclear Waste Management
Department of Energy
Washington, DC 20585

10\. SUPPLEMENTARY NOTES

Library of Congress Catalog Card Number: 83-600607

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

11\. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here)*

An international workshop on the backfill component of a multi-barrier underground isolation system for nuclear waste disposal/storage was conducted on April 13 and 14, 1981, at the National Bureau of Standards, Washington, DC. This report includes the papers presented and/or submitted to the workshop for publication. It also contains the surveyed results of a questionnaire distributed to the participants at the workshop. The workshop held a one-day informal in-depth discussion on the subject and a summary of these discussions is included in this report, which states areas where further research and development needs are required to better understand the fundamental mechanisms of the backfill in the waste repository.

12\. KEY WORDS *(Six to twelve entries; alphabetical order; capitalize only proper names; and separate key words by semicolons)*

backfill; compressibility; geology; nuclear waste; permeability (hydraulic conductivity); research and development; shear strength; sorption; swelling; thermal conductivity.

13\. AVAILABILITY

☒ Unlimited
☐ For Official Distribution. Do Not Release to NTIS
☒ Order From Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402.
☐ Order From National Technical Information Service (NTIS), Springfield, VA. 22161

14\. NO. OF PRINTED PAGES

228

15\. Price

NBS TECHNICAL PUBLICATIONS

PERIODICALS

JOURNAL OF RESEARCH—The Journal of Research of the National Bureau of Standards reports NBS research and development in those disciplines of the physical and engineering sciences in which the Bureau is active. These include physics, chemistry, engineering, mathematics, and computer sciences. Papers cover a broad range of subjects, with major emphasis on measurement methodology and the basic technology underlying standardization. Also included from time to time are survey articles on topics closely related to the Bureau's technical and scientific programs. As a special service to subscribers each issue contains complete citations to all recent Bureau publications in both NBS and non-NBS media. Issued six times a year. Annual subscription: domestic $18; foreign $22.50. Single copy, $5.50 domestic; $6.90 foreign.

NONPERIODICALS

Monographs—Major contributions to the technical literature on various subjects related to the Bureau's scientific and technical activities.

Handbooks—Recommended codes of engineering and industrial practice (including safety codes) developed in cooperation with interested industries, professional organizations, and regulatory bodies.

Special Publications—Include proceedings of conferences sponsored by NBS, NBS annual reports, and other special publications appropriate to this grouping such as wall charts, pocket cards, and bibliographies.

Applied Mathematics Series—Mathematical tables, manuals, and studies of special interest to physicists, engineers, chemists, biologists, mathematicians, computer programmers, and others engaged in scientific and technical work.

National Standard Reference Data Series—Provides quantitative data on the physical and chemical properties of materials, compiled from the world's literature and critically evaluated. Developed under a worldwide program coordinated by NBS under the authority of the National Standard Data Act (Public Law 90-396).

NOTE: The principal publication outlet for the foregoing data is the Journal of Physical and Chemical Reference Data (JPCRD) published quarterly for NBS by the American Chemical Society (ACS) and the American Institute of Physics (AIP). Subscriptions, reprints, and supplements available from ACS, 1155 Sixteenth St., NW, Washington, DC 20056.

Building Science Series—Disseminates technical information developed at the Bureau on building materials, components, systems, and whole structures. The series presents research results, test methods, and performance criteria related to the structural and environmental functions and the durability and safety characteristics of building elements and systems.

Technical Notes—Studies or reports which are complete in themselves but restrictive in their treatment of a subject. Analogous to monographs but not so comprehensive in scope or definitive in treatment of the subject area. Often serve as a vehicle for final reports of work performed at NBS under the sponsorship of other government agencies.

Voluntary Product Standards—Developed under procedures published by the Department of Commerce in Part 10, Title 15, of the Code of Federal Regulations. The standards establish nationally recognized requirements for products, and provide all concerned interests with a basis for common understanding of the characteristics of the products. NBS administers this program as a supplement to the activities of the private sector standardizing organizations.

Consumer Information Series—Practical information, based on NBS research and experience, covering areas of interest to the consumer. Easily understandable language and illustrations provide useful background knowledge for shopping in today's technological marketplace.

Order the **above** *NBS publications from: Superintendent of Documents, Government Printing Office, Washington, DC 20402.*

Order the **following** *NBS publications—FIPS and NBSIR's—from the National Technical Information Service, Springfield, VA 22161.*

Federal Information Processing Standards Publications (FIPS PUB)—Publications in this series collectively constitute the Federal Information Processing Standards Register. The Register serves as the official source of information in the Federal Government regarding standards issued by NBS pursuant to the Federal Property and Administrative Services Act of 1949 as amended, Public Law 89-306 (79 Stat. 1127), and as implemented by Executive Order 11717 (38 FR 12315, dated May 11, 1973) and Part 6 of Title 15 CFR (Code of Federal Regulations).

NBS Interagency Reports (NBSIR)—A special series of interim or final reports on work performed by NBS for outside sponsors (both government and non-government). In general, initial distribution is handled by the sponsor; public distribution is by the National Technical Information Service, Springfield, VA 22161, in paper copy or microfiche form.