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WBS 1.1.3

**Project:** Development of Backfill Material

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## Objectives

The objectives for this project are described separately for the individual tasks below.

## 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 geologic 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; the space between the surface of the canister or overpack and the geologic medium is then backfilled with a suitable material or combination of materials. Emplacement of properly engineered backfill materials can significantly reduce the possibility of radionuclide migration into the geologic medium by increasing the redundancy of barriers. A properly placed multicomponent backfill could provide a high-integrity seal against ground-water penetration and could ensure selective sorption of specific radionuclides such as $^{129}$I and $^{99}$Tc in the event that ground-water penetration and leaching of the waste does eventually take place. Backfill components can improve the chemical stability of the canister or overpack by controlling the Eh and pH of the immediate environment.

## Activities During the Reporting Period

### PRELIMINARY DEMONSTRATION OF A MULTIFUNCTIONAL BACKFILL MATERIAL

### Objectives

The objectives of this activity are to provide an early laboratory-scale demonstration of a multifunctional backfill material and, in doing so, to gain experience for use in designing the subsequent screening experiments employed in the laboratory evaluation of selected backfill materials.

Initial laboratory experiments include evaluation of pure bentonite clay plus clay mixed with other selected exchange materials. The compaction properties, the permeability of the compacted forms in a dynamic-flow system, and the migration properties of selected ions of interest (e.g., $^{129}$I, $^{137}$Cs, $^{90}$Sr, and $^{99}$Tc) will be determined.

### Procedures

Pure bentonite pellets 1 inch in diameter have been pressed to densities of 2 $g/cm^3$ at ~2 $tons/in.^2$. The pressed pellets are mechanically stable and do not crack or crumble with reasonable handling. Additional pellets are being evaluated as a function of pressing conditions, water content, and clay-exchange material ratio.

In initial experiments, a heavy-wall stainless steel cylinder with a fritted plate in the bottom was used to investigate the effects of pumping water through pressed bentonite clay. It quickly became evident that the clay must be constrained by a fritted plate on top of the bed. The equipment is being redesigned to include this feature and hopefully, to provide a measure of the pressure exerted by the bentonite against the confining cylinder and end plates as water is forced through the pressed clay bed at high pressures.

Techniques have been developed to determine batch distribution (Kd's) for a number of candidate backfill exchange materials as a part of the screening process to select a manageable number of materials (6 to 12) for a more comprehensive evaluation.

## DEFINITION AND PRIORITIZING OF BACKFILL REQUIREMENTS

### Objectives

The objectives of this activity are to define the "reasonable purposes" of a backfill (what functions it should fill) and then convert the "reasonable purposes" to a prioritized and quantified list of backfill performance attributes. Backfill material could be used simply to fill in the space between the waste canister and the host geologic media. A much more meaningful purpose would be to provide a redundant barrier(s) to the migration of radioactive species from the waste to the biosphere. Any emplaced backfill material will be subjected to five influences:

(1) Mechanical pressure and stress

(2) A high radiation field

(3) Above-ambient temperature

(4) Hydraulic flow of ground water

(5) Materials dissolved in the ground water.

The backfill material must continue to fulfill its function(s) while subjected to these five influences for time periods ranging up to $10^6$ years.

### Procedures

Two backfill functions being examined that do provide redundant barriers are (1) minimizing the penetration of ground water to the encapsulated waste and (2) controlling the chemical environment surrounding the encapsulated waste.

If ground water cannot reach the canister or if the rate of migration is so slow that the amount of water that can come into contact with the waste canister even in $10^6$ years is small, canister corrosion will be significantly reduced and the principal mechanism for transporting radioactive species to the biosphere eliminated. Results of initial experiments described in the Procedures section imply the possibility that compacted bentonite clay can provide such a barrier.

Chemical control of the environment surrounding the encapsulated waste can be accomplished by placing carefully selected materials in the backfill that will buffer the pH to a desired range and reduce the oxygen fugacity to near zero. Such careful control will minimize corrosion of the canister system and greatly reduce the migration rate of most cationic radioactive species. Additional chemical control is achieved by using backfill material such as bentonite clay with high cation-ion-exchange capacity or by adding specialized absorbing material for selected nuclides such as $^{129}I$.

A preliminary list of backfill attributes (see Table 1) has been compiled.

## SCREENING OF POTENTIAL BACKFILL MATERIALS

### Objective

Potential backfill materials and combinations of materials will be screened to select a small number for exhaustive laboratory testing. Those that show the greatest promise of meeting the performance attributes defined in the activity described in the previous section will be chosen. As noted, major emphasis has been placed upon expanding clays, but other materials have not been excluded from consideration.

### Procedures

Activities to date include an extensive literature search and documentation of materials properties to provide a basis for the ultimate selection.

## LABORATORY EVALUATION OF SELECTED BACKFILL MATERIAL

### Objective

The objective in this task is to experimentally measure the performance characteristics of the selected backfill materials and to evaluate the experimental results in terms of the defined attributes.

## TABLE 1. UNORDERED LIST OF POSSIBLE BACKFILL MATERIALS ATTRIBUTES

| Attribute | Relative Importance | | | | | | | | | Experimentally Measurable Property | References | General Comments |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | High | | | Medium | | | Low | | | | | |
| | 1 | 2 | 3 | 1 | 2 | 3 | 1 | 2 | 3 | | | |
| 1. Water Exclusion | | | | | | | | | | -Porosity ($\phi$)<br>-Permeability ($\epsilon$)<br>-Diffusivity (D)<br>-Low hydraulic conductivity<br>-Capillary pressure | | -Hydrophobic surfaces<br>-Very small pores |
| 2. Radionuclide Retention | | | | | | | | | | -Distribution coefficient ($K_d$)<br>-Dispersion coefficient ($K_f$)<br>-Radionuclides - Specific Loading Capacity | | -Channeling (seams, cracks)<br>-Thickness (boundary effects) |
| 3. Mechanical Stability | | | | | | | | | | -Modulus of elasticity<br>• in shear (G)<br>• in tension<br>• in compression $\}$ (E)<br>-Bulk modulus of elasticity (K)<br>-Modulus of resilience ($U_p$)<br>-Modulus of rupture ($S_R$)<br>- in bending<br>- in torsion | | |
| | | | | | | | | | | -Atterberg limits | | -Atterberg limits are the plasticity index, liquid limit, plastic limit |
| | | | | | | | | | | -Activity | | -Activity = Ratio of Atterberg plastic limit to fraction of clay in the substance |
| 4. Thermal Stability | | | | | | | | | | -Retention of chemical and physical properties during and after thermal stress<br>-Coefficient of thermal expansion ($\beta$) | | -Cracking<br>-Diagenesis |

## TABLE 1. (Continued)

| | Relative Importance | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | High | | | Medium | | | Low | | | | | |
| Attribute | 1 | 2 | 3 | 1 | 2 | 3 | 1 | 2 | 3 | Experimentally Measurable Property | References | General Comments |
| 5. Radiation Stability | | | | | | | | | | -Retention of chemical and physical properties during and after irradiation | | -Diagenesis |
| 6. Satisfactory Heat-Transfer Capability | | | | | | | | | | -Thermal conductivity (k)<br>-Emissivity ($\epsilon$)<br>-Overall heat-transfer coefficient (U) | | |
| 7. Mitigation of Long-Term Intrusion | | | | | | | | | | | | -Possible colored material<br>-Do not utilize materials that are currently, or projected to be, valuable or in short supply |
| 8. Availability | | | | | | | | | | -Resource assessment<br>-Supply capabilities<br>-Resource utilization/conservation | | |
| 9. Cost/Cost-Benefit | | | | | | | | | | | | -Resource available in adequate quantity |
| 10. Homogeneity/Reproducibility | | | | | | | | | | | | -Can the selected backfill be adequately reproduced with the same attributes? |
| 11. Resistance to Biological Degradation | | | | | | | | | | -Retention of physical and chemical properties during and after exposure to biological attack | | |
| 12. Self-Sealing Ability/Ductility | | | | | | | | | | See Item 3 | | |
| 13. Ease of Emplacement/Formability | | | | | | | | | | See Item 3 | | |

TABLE 1. (Continued)

| | Relative Importance | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | High | | | Medium | | | Low | | | | | |
| Attribute | 1 | 2 | 3 | 1 | 2 | 3 | 1 | 2 | 3 | Experimentally Measurable Property | References | General Comments |
| 14. Compactibility | | | | | | | | | | See Item 3 | | |
| 15. Faulting Characteristics | | | | | | | | | | See Item 3 | | |
| 16. Shear Resistance | | | | | | | | | | See Item 3 | | |
| 17. Long-Lived Physical and Chemical Properties | | | | | | | | | | -Retention of physical and chemical properties with time periods up to and beyond 1000 years | | |
| 18. Radiation Attenuator | | | | | | | | | | Shielding properties | | |
| 19. Unconfined Compressive Strength | | | | | | | | | | See Item 3 | | |
| 20. Hydrophobic | | | | | | | | | | See Item 1 | | |
| 21. Swelling Capability | | | | | | | | | | See Item 1 | | |
| 22. Large and Permanent Molecular and Cationic (Anionic) Sorption Capacity | | | | | | | | | | See Item 2 | | |
| 23. High Consolidation Rate | | | | | | | | | | | | -Rate of compression |

TABLE 1. (Continued)

| Attribute | Relative Importance | | | | | | | | | Experimentally Measurable Property | References | General Comments |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | High | | | Medium | | | Low | | | | | |
| | 1 | 2 | 3 | 1 | 2 | 3 | 1 | 2 | 3 | | | |
| 24. Low Sensitivity | | | | | | | | | | -Strength in undisturbed state divided by strength in remolded state | | |
| 25. Compatibility | | | | | | | | | | -Change in physical and chemical properties following exposure to or reaction with (a), (b), (c), or (d) | | |
| a. With Geology | | | | | | | | | | | | |
| b. With Package | | | | | | | | | | | | |
| c. With Media Solutions | | | | | | | | | | | | |
| d. With Other Backfill Components Prior to and Following Any "Reaction" (i.e., Sorption) | | | | | | | | | | | | |
| 26. "Permanent" Retention of Radionuclides | | | | | | | | | | See Items 2 and 17 | | |
| 27. Enhances Package Retrievability | | | | | | | | | | | | |
| 28. Barrier to Corrosion Products/High Ion-Exchange Capacity | | | | | | | | | | -Sorption capacity for possible corrosion products, $H^+$, $OH^-$, radionuclides, etc. | | |

TABLE 1. (Continued)

| Attribute | Relative Importance | | | | | | | | | Experimentally Measurable Property | References | General Comments |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | High | | | Medium | | | Low | | | | | |
| | 1 | 2 | 3 | 1 | 2 | 3 | 1 | 2 | 3 | | | |
| 29. Available in Adequate Purity | | | | | | | | | | -Low organic impurities | | |
| 30. Corrosion Resistance | | | | | | | | | | | | |
| 31. High Oxidation Resistance | | | | | | | | | | | | |
| 32. General Items | | | | | | | | | | a. Diagenesis<br>b. Thixotropy<br>c. Precipitation in reaction<br>d. Transport phenomena<br>e. Filtration capability<br>f. Radionuclide convection<br>g. Particle shape $(\psi)$, $(\eta)$<br>h. Particle size (Do)<br>i. Void ratio<br>j. Swelling versus sorption/ cross effects<br>k. Density $(\rho)$<br>l. Cushion effect<br>m. Fluids content at time of emplacement<br>n. Are any properties directional; isotropy and anisotropy<br>o. What is the effect of density on mechanical stability? | | |
| 33. High Redox Potential | | | | | | | | | | | | |

TABLE 1. (Continued)

| | Relative Importance | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | High | | | Medium | | | Low | | | | | |
| Attribute | 1 | 2 | 3 | 1 | 2 | 3 | 1 | 2 | 3 | Experimentally Measurable Property | References | General Comments |
| 34. Enhances pH and Eh of environment | | | | | | | | | | See Items 29 and 34 | | |
| 35. Gettering properties for $O_2$, etc., are high | | | | | | | | | | See Items 2, 29, and 34 | | |