CONF-801209--7

SAND80-2190C

STATUS OF SANDIA BACKFILL-GETTER DEVELOPMENT STUDIES*

Martin A. Molecke
E. James Nowak

Sandia National Laboratories
Albuquerque, New Mexico 87185

October 1980

MASTER

Presented at the 1980 NWTS Information Meeting
Columbus, Ohio
December 9-11, 1980

*This work is supported by the U. S. Department of Energy.

DISCLAIMER

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

STATUS OF SANDIA BACKFILL-GETTER DEVELOPMENT STUDIES

Martin A. Molecke
E. James Nowak

Sandia National Laboratories
Albuquerque, New Mexico 87185

A backfill-getter development program has been conducted by Sandia National Laboratories since 1977. The purpose of this program is to develop backfill materials suitable for emplacement in a deep geologic salt repository, adjacent to either high-level (HLW) or transuranic wastes. The backfill-getter serves as an engineered barrier within the waste package; it has multiple chemical and physical functions that can significantly delay or minimize or delay the extent of waste package degradation and subsequent radionuclide release.

In this discussion, ~~we will describe~~ specific functions desired of the backfill as well as various recent experimental studies, results, and current plans for further studies. Modeling of the backfill's effectiveness for delaying radionuclide release ~~will~~ also ~~be~~ presented.

The important chemical and physical functions of the backfill are (1) to act as a barrier to hydrologic intrusion, (2) to act as an actinide and fission product radionuclide sorptive barrier or getter, (3) to chemically modify any intruding groundwater/brine, i.e. for pH, Eh, or ionic composition, (4) to provide a mechanical stress buffer and thereby reduce deformation of the waste package, and (5) to serve as a thermal transport medium.

Initial screening studies on candidate backfill-getter materials have been completed. The smectite swelling clay, bentonite, has been selected as a major component in the backfill because of its favorable properties in contact with salt and brine solution: good sorptive capacity for the actinides and rare earths; retention of sorptive properties in mixtures with other components; capability to buffer brine pH in a near-neutral range; low liquid permeability; favorable swelling properties in brine; adequate thermal conductivity and high-temperature performance; mechanical buffer properties; and reasonable cost for bulk materials. Other materials having desirable backfill properties were also identified.

Advanced chemical and materials performance testing of various backfill mixtures is in progress. The sorptive properties for Pu and Am in batch and column-type experiments are being measured; empirical batch Kd values of 500-30000 (ml/g) for Pu(IV) and 4000-16000 for Am(III) have been obtained under various experimental conditions.[1,2]

Various materials such as synthetic zeolites, titanates, and charcoal show promise for sorbing fission products in brine.[1,2,3] Representative empirical Kd values measured are 20-120 for Cs^{+}, 20-500 for Sr^{++}, 20-50 for I^{-}, and 300-380 for TcO_4^{-}. This advanced phase of the program is continuing.

Overtesting and accelerated test methods are being utilized to investigate the mechanisms of backfill alteration or degradation which may cause the backfill barrier to lose some of its chemical and physical effectiveness. Parameters of interest include pH, Eh, temperature, pressure, radiation, and backfill-getter overall composition. Evaluation of the mechanisms and degradation data will also be useful for further modifications in the overall backfill-getter mixture.

Geochemical analyses are necessary to aid in predicting the long-term stability of backfill components--i.e., the effect of mineralogic phase changes on sorptive and physical properties of the backfill. This program phase involves analyses of applicable natural analogues, theoretical thermochemical modeling, and experimental studies of hydrothermal backfill-brine reactions. After bentonite was contacted with brine B at $250^{\circ}C$ for 11 days, its montmorillonite content (85-90%) was better crystallized, and initially present chlorites and illites were, at the conclusion of the tests, undetectable by x-ray diffraction.[2] These results favor the long-term maintenance of bentonite's desired backfill properties in brine at elevated temperatures. More extensive and detailed experiments are planned.

Geotechnical measurements on bentonite and bentonite-sand mixtures, have yielded brine and water permeabilities in the microdarcy range. This is adequate to essentially eliminate convective transport through the backfills; liquid transport will be essentially by diffusion only. Thermal conductivities of bentonite, bentonite/salt, and bentonite/sand mixtures are in the range of 0.35 to 0.72 W/m^{o}K, similar to that of crushed rocksalt, 0.49 W/m^{o}K. All these mixtures were dry and either loosely packed or tamped. Tests are in progress to measure the thermal conductivities of backfill materials that are compacted to a higher density and/or are saturated with brine. These thermal conductivities will be compared with those of virgin rocksalt, about 5-6 W/m^{o}K.

Mine and process engineering is another phase of the backfill development program. This involves primarily engineering-scale work on backfill emplacement forms and techniques to be used in a repository. It is envisioned that high-density, precompacted backfill forms will be emplaced adjacent to HLW canisters, while a granular backfill mixture will probably be emplaced adjacent to transuranic wastes or be used as a mine tunnel fill material. The mine engineering phase will culminate with an actual demonstration of backfill emplacement in a field test.

Another major phase of this backfill-getter development program involves advanced testing of backfill effectiveness and properties with bench-scale and hot-cell testing, followed by large engineering-scale field tests, then in situ backfill emplacement demonstrations and qualification. Portions of this program phase are in the planning stage[4] or in progress, and include HLW total-system test being conducted in a hot cell at the Pacific Northwest Laboratory.[5]

Mathematical modeling of backfill effectiveness has been performed concurrently with the other program phases described. Effectiveness is calculated in terms of "breakthrough at 1% of the entering radionuclide concentration." Breakthrough has been calculated as a function of empirical Kd, effective porosity, and backfill thickness.[6,7] Results are summarized in Table 1. Breakthrough or radionuclide release delay times of 10^5 to 10^6 years and 10^3 to 10^4 years can be expected for the actinides and fission products, respectively.

Our available data and effectiveness modeling show that the backfill-getter materials appear to be an excellent means for minimizing or significantly retarding the release of radionuclides from the waste package, even in the presence of concentrated brine. It is a valuable component of the multibarrier concept of radioactive waste isolation and, as such, is receiving an increased development effort.

References:

1. Nowak, E. J., Radionuclide Sorption and Migration Studies of Getters for Backfill Barriers, SAND79-1110, Sandia National Laboratories, July 1980.

2. Nowak, E. J., Composite Backfill Materials for Radioactive Waste Isolation by Deep Burial in Salt, SAND80-1377C, to be published in: Scientific Basis for Nuclear Waste Management, Vol. 3, J. G. Moore, ed., Plenum Press, New York (1981).

3. Winslow, D. C., The Sorption of Cesium and Strontium from Concentrated Brines by Backfill Barrier Materials, SAND80-2046, Sandia National Laboratories, in preparation.

4. Molecke, M. A. and Nowak, E. J., Applicability of a Backfill-Getter Barrier for Radioactive Waste Isolation: Chemical and Physical Functions, SAND80-0838C, presented at the Second Chemical Congress of the North American Continent/American Chemical Society Meeting, Las Vegas, Nevada, August 1980.

5. Bradley, D., Shade, J. W., and Molecke, M. A., Joint PNL-Sandia High-Level Waste Total System Test, presented at the 1980 NWTS Annual Information Meeting, Columbus, Ohio, December 1980.

6. Nowak, E. J., The Backfill Barrier as a Component in a Multiple Barrier Nuclear Waste Isolation System, SAND79-1109, Sandia National Laboratories, May 1980.

7. Nowak, E. J., The Backfill as an Engineered Barrier for Nuclear Waste Management, in: Scientific Basis for Nuclear Waste Management, Vol. 2, C. J. Northrup, ed., Plenum Press, New York (1980).

TABLE 1

BREAKTHROUGH TIMES FOR VARIOUS BACKFILL-GETTER PARAMETERS

Kd (ml/g)	Effective Porosity	Breakthrough Time (Years) 0.3 m Thick	0.9 m Thick	3 m Thick
2000	0.01	1×10^5	1×10^6	1×10^7
200	0.01	1×10^4	1×10^5	1×10^6
20	0.01	1×10^3	1×10^4	1×10^5
2000	0.1	1×10^4	1×10^5	1×10^6
200	0.1	1×10^3	1×10^4	1×10^5
20	0.1	1×10^2	1×10^3	1×10^4

Breakthrough was defined as that time when the concentration at the outer surface of the backfill equals 1% of the entering concentration. The interstitial flow velocity was assumed to be equal to 3cm/year, approximately the maximum value for which molecular diffusion predominates over convective transport; this is significantly larger than expected for measured backfill permeabilities. Calibrations are for three different thicknesses of backfill (0.3 m, 0.9 m, and 3 m).