PERFORMANCE OF BENTONITE/CRUSHED TUFF SEALS FOR NUCLEAR WASTE REPOSITORIES

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ABSTRACT

Mixtures of bentonite and crushed rock are potential sealing materials for high level nuclear waste repositories. To allow for overall repository performance assessments, evaluations of the sealing performance under diverse conditions are needed. American Colloid c/s granular bentonite and Apache Leap tuff have been mixed to prepare samples for laboratory flow testing. Bentonite weight percent and crushed tuff gradation are the major variables studied. The sealing performance assessments include high injection pressure flow tests, polyaxial flow tests, high temperature flow tests, and piping tests. The results indicate that an appropriate composition would have at least 25% bentonite by weight mixed with well-graded crushed rock. The sealing performance of bentonite/crushed rock mixtures can be enhanced by increasing the amount of bentonite to 35%. The piping damage to the sealing performance is small if the maximum hydraulic gradient does not exceed 120 and 280 for 25 and 35% bentonite content, respectively. The hydraulic gradients above which flow of bentonite takes place are deemed critical. The pressure required to generate a critical gradient probably is related to the yield stress of the bentonite in the mixture. A difference up to one or two orders of magnitude has been observed between the vertical and horizontal permeabilities. The high horizontal permeability results from the uneven bentonite distribution in the pores between the crushed rock particles due to particle segregation. Temperature seems to have no negative effects on the sealing performance within the test range from room temperature to 60°C. Recommendations for future research are included.

INTRODUCTION

Penetrations of a high-level nuclear waste repository must be properly sealed to reduce radionuclide migration to an acceptable level. Bentonite is considered as a sealant because of its desirable swelling and self-healing characteristics, low permeability, sorptive qualities, and longevity in nature (1,2). Mixtures of crushed rock or sand and bentonite are also repository sealing candidates (3-6).

Studies of the sealing performance of bentonite/crushed rock mixtures (7,8,9) show that they can be engineered to have a low permeability similar to that of bentonite. The performance, however, was evaluated at room temperature and under low hydraulic gradients. The sealing performance at elevated temperatures and under high hydraulic gradients is needed to allow for overall repository performance assessments (10,11). The seal components may be required to retain adequate sealing capacity for a long period of time (12). It is therefore important to investigate the effect of piping on the long-term sealing performance, especially if the seal locations are intercepted by joints and/or fractures.

In this study, American Colloid c/s granular bentonite and crushed Apache Leap tuff have been mixed to prepare samples for laboratory flow testing. Bentonite weight percent and crushed tuff gradation are the major variables in sample construction. The sealing performance assessments include high injection pressure flow tests, polyaxial flow tests, high temperature flow tests, and piping tests. For a carefully engineered bentonite/crushed rock plug, a low permeability can be achieved and maintained under the diverse conditions imposed.

MATERIALS

Bentonite

American colloid c/s granular bentonite consists primarily of sodium montmorillonite, with traces of quartz, feldspar and biotite. The bentonite has an average moisture content as-shipped of 9.56% over a range of 9.41 to 9.69%. Approximately 80% of the air-dried bentonite particles fall in a size range of 0.84 to 0.42 mm (U.S. meshes #20 and #40, respectively). This clay product has an average specific gravity of 2.92, liquid limit of 433%, and plastic limit of 50% and is basically dust-free. The moisture-density relation gives the optimal moisture content as 23.5% and a maximum dry density of 12.21 kN/m3. Some properties of c/s granular bentonite are well known (13).

Crushed Rock

The rock tested is from the densely welded brown unit of the Apache Leap tuff from Superior, Arizona. The tuff shows low porosity and permeability and resembles Topopah Spring tuff, in which a high level nuclear waste repository might be located. The specific gravity of the tuff is 2.61. Tuff chunks, approximately 15x15x20 cm in size, are crushed in a conventional jaw crusher and an adjustable roller crusher. An opening of 1.3 cm is set for the roller crusher. The crushing produces less than one percent by weight of particles smaller than 0.075 mm. Crushed tuff is sieved and stored in air-tight plastic containers.

EXPERIMENTS

Permeameter Design

PVC permeameters have nominal inside diameters of 10.16 and 30.5 cm, stainless steel permeameters 10.16 and 20.3 cm. PVC permeameters are designed for flow testing under low injection pressures, stainless steel permeameters for flow testing under high injection pressures or at elevated temperatures. Circular openings in the walls of some PVC permeameters simulate openings in the walls of holes where seals are emplaced. The openings are plugged during sample installation and saturation and are unplugged for flow testing. No porous stone or filter materials (e.g. sand) are emplaced at these side openings, because they might impede particle movement. The modified permeameters are designed to study the conditions under which bentonite may flow or may be eroded out of the crushed tuff matrix, and to study the significance of piping and erosion on the sealing performance of bentonite/crushed tuff mixtures.

A rectangular plexi-glass permeameter constructed for polyaxial flow testing is 12.5x11.4x11.4 cm in size. The walls are detachable such that thin porous plates can be installed at inflow and outflow ends.

Sample Preparation

Mixtures of nominal 15, 25, and 35 bentonite weight percentages and crushed rock gradation types A, B, and C form the majority of samples for the flow testing. The three tuff gradations are derived by first removing particles smaller than 0.075 mm and then particles larger than 9.42, 5.58 and 4.75 mm, in sequence, from the "coarse" gradation obtained by crushing tuff chunks. Crushed tuff of FA and FC gradations is used to study the effect of gradation on sealing performance. Gradations FA and FC are obtained from the ideal Fuller-Thompson grading equation (Eq. 1), using n = 0.5 and D = 9.42 and 19.05 mm, respectively. Fig. 1 shows the grain size distributions.

$$P = 100(d/D)^n \qquad \text{(Eq.1)}$$

where P = weight percent passing sieve aperture d,

D = maximum particle size,

n = exponent.

The bentonite mixed with the crushed tuff has an initial water content of 23.5%. After mixing, the samples are covered with wraps and are cured for 72 hours before installation.

Sample Installation

The thoroughly mixed bentonite and crushed tuff are transferred to a permeameter chamber using a small plastic shovel. The samples are compacted in the permeameters in three layers. The number of blows per layer is adjusted according to the sample diameter such that a compactive effort equivalent to the standard Proctor compaction is delivered. The 20.32 cm diameter samples mixed with type

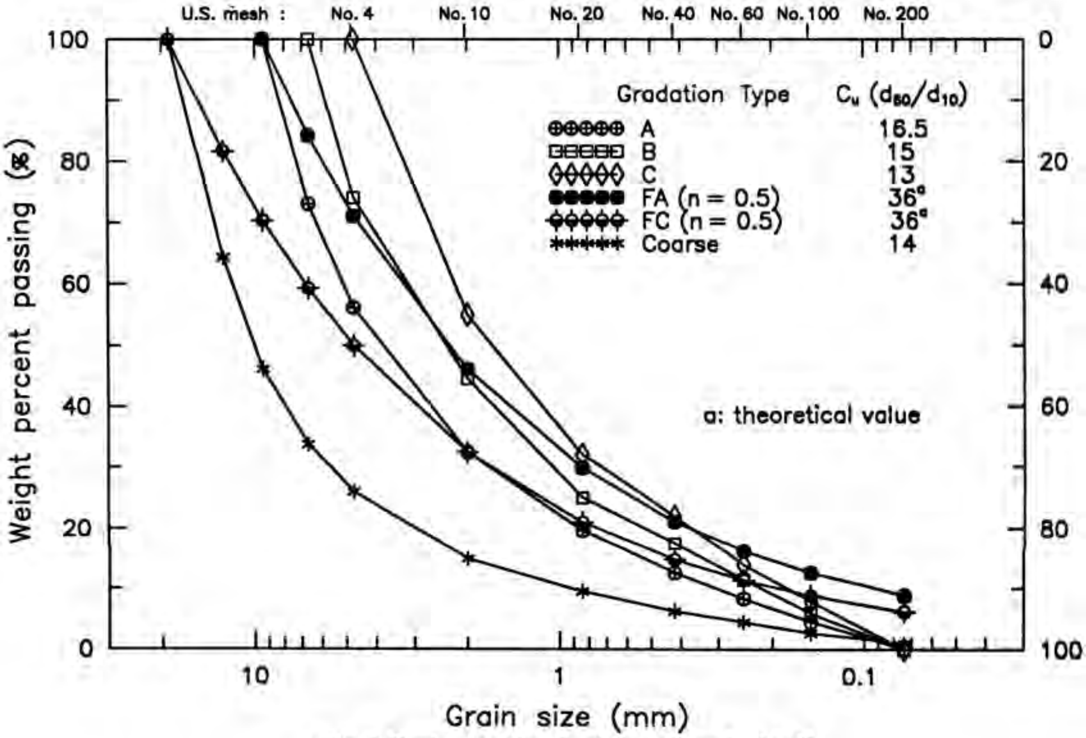

Fig. 1 Grain size distributions of crushed tuff.

FC crushed tuff, however, received only one quarter of the standard compaction. The majority of the plugs are restrained at the top and bottom by sand and/or porous stones. The samples installed in the stainless steel permeameters are confined at the top by porous stones and a piston. The remaining space between the piston and the top cap plate is filled with water to minimize the vertical movement due to swelling.

PERMEABILITY TESTING

The samples are subjected to an approximately 2.5m water head from the bottom port for saturation. The saturation is aided intermittently by applying vacuum to the top port, at minus 103.5 kPa for 30 to 45 minutes. This saturation proceduregenerally lasts for six weeks before the flow testing is started. The samples are believed to be highly saturated in view of the good mass balance obtained during flow testing.

Permeabilities of the plugs are determined using the constant head, falling head, or double-pipette falling head method [e.g. 13]. High injection pressure flow testing is performed under constant head driven by compressed gas(helium). To determine the permeabilities at elevated temperatures, the samples are immersed in a water bath equipped with an immersion heater/circulator. A double-pipette system is used in the high temperature testing, and the hydraulic gradients induced are less than 10.

SUMMARY OF RESULTS

Variables studied include sample size, crushed rock gradation, bentonite weight percent, flow direction, hydraulic gradient, and temperature, The effect of piping on the sealing performance of the mixture plugs containing 25% bentonite and 75% type A crushed tuff by weight has been investigated. Great efforts have been made to obtain good mass balance. For the majority of the plugs tested, the inflow and outflow differs only by 5 to 10%.

Longitudinal Flow Tests

Average permeabilities obtained from the downward flow testing under low hydraulic gradients(< 35) are summarized in Fig. 2. The permeability decreases with increasing bentonite content. The permeabilities of the plugs containing 25% or more of bentonite are close to the permeability of plugs constructed of bentonite only. Fig. 2 also demonstrates the effect of crushed tuff gradation on the sealing performance: the greater the uniformity coefficient (d_{60}/d_{10}) of crushed tuff, the lower the permeability. The effect of sample size on the sealing performance is not clear. The inconsistency in the permeabilities measured for different plug sizes may be due to variations in the stiffness of the permeameters, compaction, and the ratio of grain size to permeameter diameter. Upward flow testing was performed to study the action of an upward seepage force. The upward permeability is about three times higher than the downward permeability.

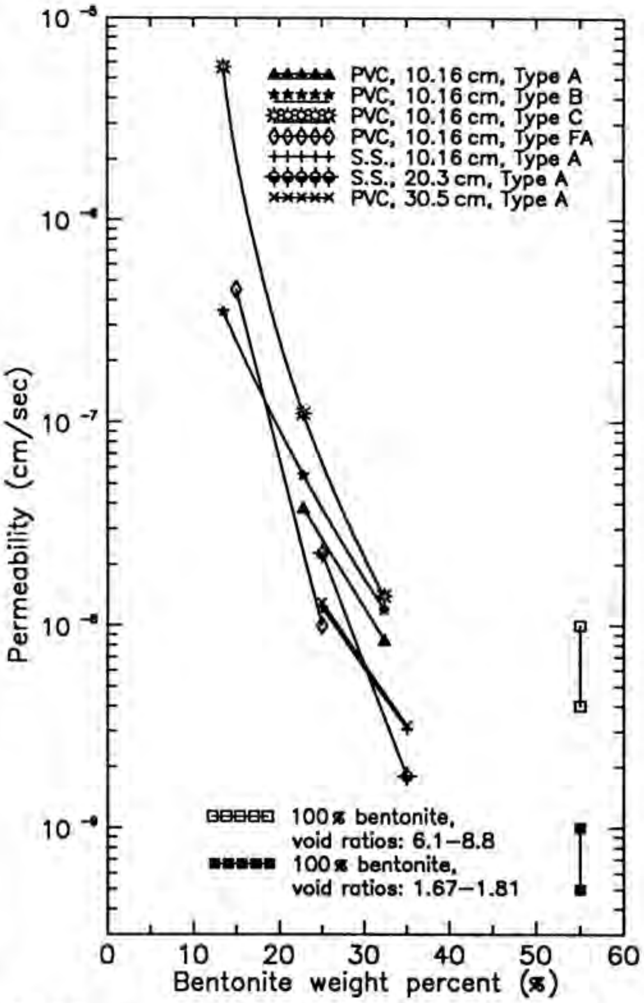

Fig. 2 Permeability of bentonite/crushed tuff mixtures as a function of bentonite content. Hydraulic gradients lower than 35.

Permeability of the mixtures appears to be relatively constant for the low range of imposed hydraulic gradients. As the hydraulic gradient increases, permeability decreases, causing a breakdown of the linear relationship between flow rate and hydraulic gradient. This behavior has been observed for all high injection pressure flow tests. An example is shown in Fig. 3. The sudden jumps in permeability may result from the radial expansion of pores due to the quick large increases in injection pressure, 390 kPa for the first jump, 762 kPa for the second one. No permanent damage in permeability can be detected, which may be indicative of the excellent healing capability of bentonite. The departure from the early linear flow rate-hydraulic gradient relationship is abrupt and can not be explained by small-strain deformation. The sudden change may indicate

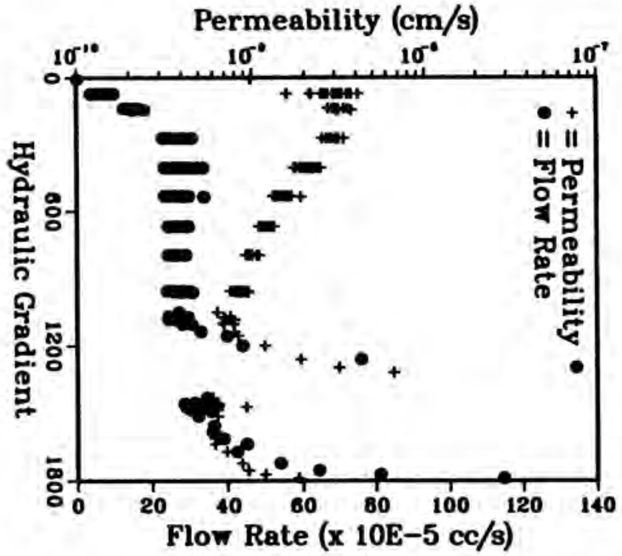

Fig. 3 Typical example of flow rate and permeability vs. hydraulic gradient. Mixture of 35% bentonite and 65% crushed tuff of gradation A.

Fig. 4 Top of a sample (25% bentonite and 75% type A crushed tuff) in which the bentonite has flowed downward as a result of applying an excessive injection pressure.

the onset of bentonite flow in the pores between crushed tuff particles. This assumption was confirmed when two samples were examined upon completion of the testing. For the sample containing 25% bentonite, the clay has flowed downward, leaving the top quarter of the sample comprised of primarily crushed tuff(Fig. 4). Bentonite flow was less developed in the sample containing 35% bentonite, and can only be discerned by examining the subtle difference in texture between top and bottom of the sample. The hydraulic gradient at which bentonite flow takes place appears to vary with bentonite content. Such an hydraulic gradient ranges from 120 to 150 and 280 to 300 for mixture samples of type A crushed tuff containing 25% and 35% bentonite, respectively. The pressure required to initiate bentonite flow likely relates to the yield stress of the bentonite in the mixture.

Polyaxial Flow Tests

It is anticipated that the horizontal permeability is higher than the vertical permeability. Such a permeability variation should be considered in the assessment of overall repository performance. The variation may allow piping in the lateral direction and may eventually jeopardize the sealing ability in the longitudinal direction. Polyaxial flow testing has been performed on two rectangular samples. One sample consists of 75% crushed tuff and 25% bentonite, the other one of 65% crushed tuff and 35% bentonite. The crushed tuff constitutes the type A gradation. The flow test results are summarized in Table I.

The high permeability is observed in only one horizontal direction, for both samples. Despite the difference in bentonite content between the two samples, the high horizontal permeabilities are very similar. The horizontal permeability differs from the vertical permeability by almost one order of magnitude for the sample containing 25% bentonite. The difference is more than two orders of magnitude for the sample containing 35% bentonite.

High Temperature Flow Tests

Two samples installed in stainless steel permeameters were immersed in a constant temperature water bath for flow testing up to 60° C. The samples were constructed by mixing crushed tuff of type A gradation with 25% and 35% bentonite. Table II summarizes the results of high temperature flow testing. The "measured" permeability is computed directly using the experimental measurements. Assuming the major temperature effects are the changes in viscosity and density of the permeant (i.e. water), permeabilities of the plugs at other temperatures can be predicted based on the average permeability measured at the room temperature(21°C) provided that the permeant properties are adjusted. The results are shown in the sixth column of Table II. The *specific permeabilities* calculated are given in the last column of Table II.

The specific permeability of the two samples changes with temperature, implying an alteration of sample structure. The specific permeability reaches a maximum at 35° C. The specific permeability at 60°C is reduced by 10% for the sample containing 25 percent bentonite, and by 50% for

TABLE I

Summary of Polyaxial Flow Test Results

Wt. Percent Clay/Tuff	Permeability (cm/s) Vertical	Horizontal(1)	Horizontal(2)
25/75	5.2-1.7•10^{-8}	1.8-1.4•10^{-7}	1.8-1.5•10^{-8}
35/65	9.2-2.5•10^{-9}	11-1.9•10^{-7}	12-6.3•10^{-9}

NOTE: The crushed tuff constitutes the type A gradation.

TABLE II

Results of Flow Tests at Elevated Temperature

Temp. (°C)	Test Duration (days)	Total Inflow (cc)	Total Outflow (cc)	Permeability Measured (cm/s) (*10^{-8})	Predicted (cm/s) (*10^{-8})	Specific (m^2) (*10^{-17})
Sample 1: 25% bentonite, 75% type A crushed tuff						
21	28	29.25	28.6	1.28±0.40	1.28	1.28
35	23	23.25	22.6	2.32±0.15	1.72	1.72
45	21	21.5	21.8	2.35±0.12	1.62	1.45
60	6	8.05	8.6	2.39±0.31	2.05	1.16
Sample 2: 35% bentonite, 65% type A crushed tuff						
21	28	8.1	7.25	3.06±1.16	3.06	3.05
35	23	7.25	6.15	5.02±1.38	4.13	3.72
45	21	6.45	6.1	5.26±0.90	3.88	3.24
60	4	0.8	1.2	4.26±1.34	4.92	1.58

NOTE: Hydraulic gradients applied are less than 10.

the sample containing 35 percent bentonite, when compared to the specific permeability at 21°C.

Piping Tests

Piping tests have been performed on two samples installed in perforated 10.16 cm diameter PVC permeameters. The samples contain 25% bentonite and 75% crushed tuff of type A gradation. A 2.25 mm diameter hole drilled through the walls of the PVC permeameters simulates an opening in the walls of boreholes or shafts where seals have been emplaced. The bottom of the samples is approximately 1 cm below the center of the holes.

The vertical permeabilities are in the range of 1.2 to 1.8x10^{-8} cm/s, under hydraulic gradients less than 32. One sample was then subjected to an injection pressure of 32.75 kPa with the bottom outlet closed and the side hole opened. Approximately two days later, bentonite and fine tuff particles along with a small amount of water moved through the hole into the connecting tubing. The bottom outlet was re-opened a week later to allow a determination of the vertical permeability. The permeability has increased by two orders of magnitude, into the lower 10^{-6}cm/s range. In

Fig. 5 Failure of a sample due to piping when outflow is permitted only through a side hole of 2.25 mm diameter.

subsequent flow testing, the bottom outlet was closed again. About 12 hours after the injection pressure had been raised to 113 kPa, a spill of water was observed(Fig. 5) and the inflow reservoir was completely drained. The gross hydraulic gradient induced by the injection pressure is 116.

During flow testing of the second sample, the bottom outlet was left open while the injection pressure was increased. Bentonite and fine tuff particles appeared in the tubing connected to the side hole at an injection pressure of 27 kPa. No signs of piping were detected for injection pressures up to 376 kPa. The amount of water flowing out of the side hole is less than 2% of the amount flowing out of the bottom outlet. The latter outflow was used in calculating the vertical permeability. This sample maintains a relatively constant permeability, in the low 10^{-8} cm/s range, under injection pressures up to 145 kPa. The permeability decreases as the injection pressure increases above 145 kPa. This behavior parallels what has been observed in the high injection pressure flow testing described earlier.

CONCLUSIONS AND RECOMMENDATIONS FOR FUTURE RESEARCH

Mixtures of bentonite and crushed rock can be engineered to yield a low permeability close to that of pure bentonite loosely emplaced. An appropriate composition would contain at least 25% bentonite by weight mixed with well-graded crushed rock. A mixture containing 25% bentonite and 75% crushed tuff of type A gradation appears to be a promising seal material. The sealing performance of bentonite/crushed rock mixtures is enhanced by increasing the amount of bentonite to 35%. A similar effect has been observed if crushed rock constituting a Fuller-Thompson grading curve(e.g. with $n = 0.5$) is used.

For mixtures consisting of type A crushed rock and 25% or 35% bentonite, the piping damage to the sealing performance is small if the maximum hydraulic gradient does not exceed 120 and 280, respectively. Compaction and the amount of bentonite are decisive factors in producing good mixture seals. For a loosely or ineffectively compacted mixture containing 25% bentonite, the performance can be damaged by dynamic disturbances (14). The influence of such disturbances is greatly reduced when more bentonite is added.

A difference of up to one or two orders of magnitude may be expected between the vertical and horizontal permeabilities. The high horizontal permeability results from the uneven bentonite distribution in the pores between crushed rock particles due to particle segregation. The difference may be reduced by introducing a thin layer of bentonite on top of each compacted layer, an approach which deserves further investigation. Temperature seems to have no negative effects on the sealing performance within the test range from room temperature to 60° C. The specific permeability reaches a maximum at 35° C and decreases with increasing temperature. Causes for the decrease are not clear but may relate to fluid-solid surface effects (15). Further research is recommended to study whether the decreasing trend continues for the higher temperatures.

Bentonite-based mixtures should remain integral and preserve sealing ability as long as the clay stays in place. A breakdown of the linear relation between flow rate and hydraulic gradient has been observed in all high injection pressure flow tests. The departure is believed to indicate the onset of bentonite flow in the mixtures. The critical hydraulic gradient at which bentonite flow occurs may depend on the yield stress of bentonite for a given water content. It is postulated that, if the yield stress of bentonite can be established as a function of water content, the critical gradient can be estimated without experiments. Such an investigation is warranted.

ACKNOWLEDGEMENT

This work is part of research on sealing of boreholes and shafts in tuff, Contract NRC-04-86-113, supported by the Division of Engineering, Office of Nuclear Regulatory Research, U.S. Nuclear Regulatory Commission. The American Colloid company supplied the bentonite. Valuable discussions with Mr. J. Philip, Contract Monitor, are gratefully acknowledged.

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