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Experimental Studies on Penetration of Pulverized Clay-Based Grout

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Abstract: For the geological disposal of high level radioactive wastes, an excavation damaged zone (EDZ) having high hydraulic conductivity resulting from the development of fractures in the rock adjacent to the tunnels will be one of the potential pathways for radioactive contaminant transport. The potential pathways will be sealed by closure components, that is, a combination of tunnel plug, backfill and grout, the latter material being a clay-based mixture in consideration of the need for long-term stability of the seals. Clay-based grout is one of the effective candidate materials that can be used to interrupt the migration of radionuclides through an EDZ. Laboratory testing of clay-based grout using pulverized bentonite, with the objective of improvement in grout penetration into a rockmass, was conducted. The results showed that the pulverization of clay-based grout had a positive effect on filtration.

Key words: High-level radioactive waste, geological disposal, repository, sealing, grout, clay-based, bentonite, pulverization.

1. Introduction

High-level radioactive waste (HLW) management in Japan is based on the multi-barrier concept. The manufactured components in the multi-barrier concept constitute the engineered barrier system (EBS), consist of a stable waste form (vitrified waste), a rigid vessel (overpack) for containment of the waste form, and the buffer and backfill placed between the overpack and the surrounding geological formations during emplacement. In the closure phase of the repository, it is important to cut off the potential pathways of the radioactive contaminants. These pathways include the shafts or disposal tunnels used in the operation phase and the excavation damaged zone (EDZ) around these drifts. The potential pathways will be sealed by the sealing components, a combination of tunnel plugs, backfill and grout, the latter material would be a mixture taking into consideration the long-term stability of the seals. Especially important, long-term sealing performance is expected from clay-based grout. Therefore, it is important to characterize the advective transport properties through and around the sealing systems utilizing clay plugs.

Examples of the application of clay-based grout are extremely limited (e.g. [1] Börgesson et al., [2] Miyanaga and Ebara). JAEA (then JNC) carried out a clay-based grouting experiment in two single fractures in granodiorite at Kamaishi Test Field in Japan [3]. The results obtained indicated that clay-based grout (average particle diameter of 6.26 m, maximum particle diameter of 33.08 m, and minimum particle diameter of 1.50 m) injected by static injection into the fractures produced a decrease in hydraulic conductivity that persisted for two years. In the Tunnel Sealing Experiment (TSX) funded jointly by AECL of Canada, ANDRA of France, the US DOE and JNC of Japan, the

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application of clay-based grout using bentonite to seal the EDZ was tested around a tunnel excavated in granitic rock. Modeling of the reduction in permeability was carried out based on the results of the *in situ* test. The filtration model of bentonite slurry was able to replicate the results of the *in situ* test and the modeling indicated the most effective concentration needed to reduce the permeability of the EDZ [4].

It is essential for developing reliable technology to be used in a repository to further improve the penetration of clay-based grout. This paper presents the results of experimental studies on the penetration of clay-based grout, i.e., pulverized bentonite. Clay-base grouting of a limited number of boreholes has the ability to penetrate widely into the target domain (penetrability), to improve the permeability around the rock (permeability), and to help maintain the improved quality (stability). Because the permeability and stability of the clay-based grout was studied in a previous study [3], a penetration test focused on the effect of viscosity, grain size and composition of materials on the penetrability was carried out in this study.

2. Rheological Properties of Clay-Based Grout

2.1 Grout Material

The clay-based grout material examined here has been referred to variously as Japanease bentonite, Kunigel V1 and Kunibond. Table 1 lists the mineral composition and physical properties of the bentonites in this study [5, 6]. Montmorillonite contents of Kunigel V1 and Kunibond are respectively 46-49 wt% and 80 wt%. Kunibond are generally characterized by high montmorillonite content. In addition, finely powdered bentonites were used in this study.

Fig. 1 shows the grain size distribution of the bentonites and the silica sand No. 6 used in a penetration test described later in this paper. The grain size of Kunigel V1 smaller than that of Kunibond is similar with that of finely powdered Kunibond. The physical properties shown in Table 1 are little affected by fine particle. The grout material consists the

Table 1 Mineral compositions and physical properties of Kunigel V1 and Kunibond (JNC, 2000 : Kurimine Industries).

Mineral compositions, (wt%)		Kunigel V1	Kunibond
Montmorillonite		46-49	80
Quartz/chalcedony		29-38	5
Cristobalite		-	15
Feldspar		2.7-5.5	-
Calcite		2.1-2.6	-
Dolomite		2.0-2.8	-
Analcite		3.0-3.5	-
Pyrite		0.5-0.7	-
Organic matter		0.31-0.34	-
Physical properties			
True specific gravity, (-)		2.7	2.64
Liquid limit, (%)		416	144.5
Plastic limit, (%)		21	63.9
Plasticity index, (-)		395	80.6
Cation exchange capacity, (meq/100 g)		52	86.5
Leach cation	Na^+	54.6	18.1
(meq/100 g)	K^+	1.3	2.4
	Ca^{2+}	41.9	74.2
	Mg^{2+}	6.6	8.1

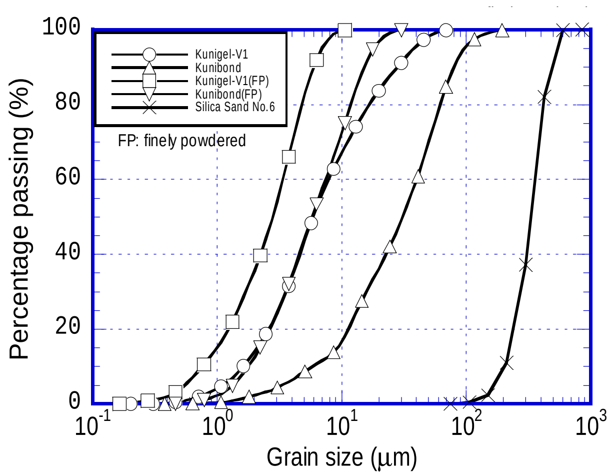


Fig. 1 Grain size distribution of bentonites and silica sand No. 6.

bentonite mixed with 600 mL distilled water by Hamilton-beach mixer for 20 minutes .

2.2 Rheological Properties

Flow properties of grout can be described in terms of rheological properties. Such rheological properties are: viscosity, yield value and thixotrophy. The lower viscosity a fluid has, the better it will flow and penetrate into fractures. Based on flow properties, fluids can be divided into Newtonian (A) and non-Newtonian (B) fluids (Fig. 2). In this context Bingham (B) fluids will be considered as non-Newtonian fluids. The most remarkable difference between Bingham and Newtonian fluids is that Newtonian fluids are described only with viscosity and Bingham fluids are described both with viscosity and yield strength. Funnel viscosity is time in seconds for 500 mL of material to flow through a Marsh funnel. The funnel viscosity is useful only for relative comparisons.

Fig. 3 presents the relationship between funnel viscosity and concentration for each bentonite material. The greater the concentration of bentonite, the greater the funnel viscosity of Kunigel V1. The funnel viscosity is very different between Kunigel V1 and Kunibond, but identical between original bentonite and finely powdered one in the same bentonite. The results suggest that the bentonite type significantly influenced the viscosity and the pulverization of material did not influence the funnel viscosity.

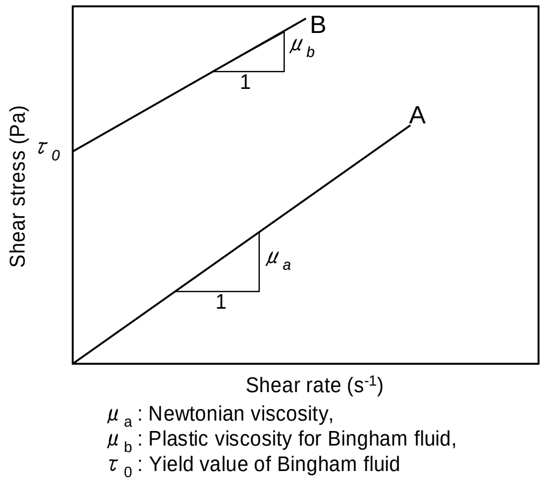


Fig. 2 Rheological behaviour of different fluid types.

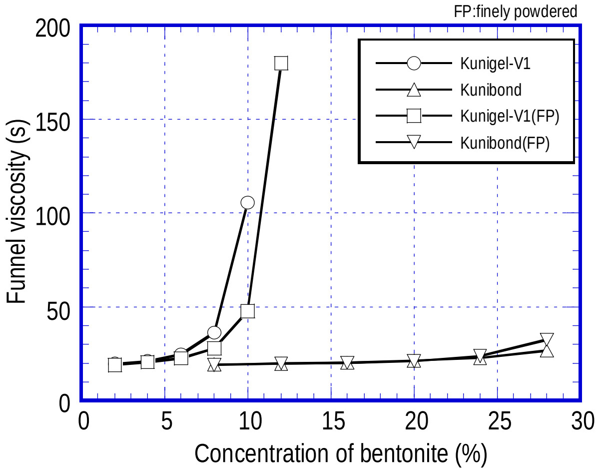


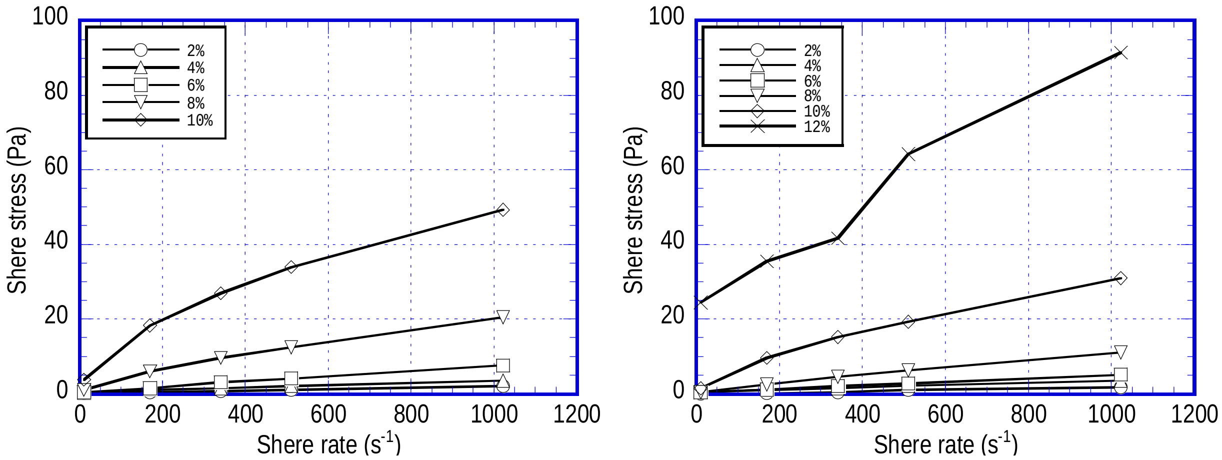
Fig. 3 Relationship between funnel viscosity and concentration for each bentonite material.

The direct-indicating viscometer was used to measure viscosity and gel strength of grout. The direct-indicating viscometer is a rotational cylinder and bob instrument, also known as a V-G meter. The test fluid is contained in the annular space or shear gap between the cylinders. Rotation of the outer cylinder at known velocities is accomplished through precision gearing. The viscous drag exerted by the fluid creates a torque on the inner cylinder or bob. This torque is transmitted to a precision spring where its deflection is measured and then related to the test conditions and instrument constants. This system permits the true simulation of most significant flow process conditions encountered in industrial processing. Model 35 Viscometer manufactured by Fann Instrument Company was used in this study.

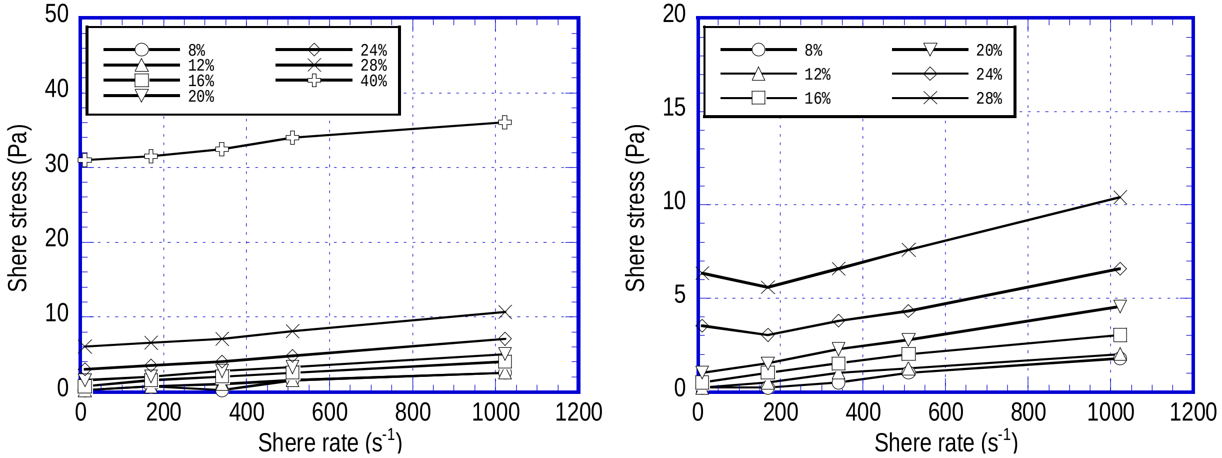
It is called "direct-indicating" because at a given speed, the dial reading is a true centipoise viscosity. Bingham plastic rheological parameters are easily calculated from direct-indicating viscometer readings. The plastic viscosity represents the slope of a straight line between the two dial readings. The yield value represents the theoretical point at which the straight line, when projected, will intercept the vertical axis. The shear stress-shear rate curve for four bentonite types is given in Fig. 4. These data were characteristic of these non-Newtonian fluids and reveal that, at low shear rates, the stress builds up in the fluid until eventually the material "yields". The shear stress of Kunigel V1 and Kunibond rose rapidly over concentrations of bentonite of 10% and 40%, respectively. Table 2 presents viscosity and apparent yield values. Plastic viscosity and yield value are identical between Kunibond and finely powdered Kunibond, but different according to grain size in Kunigel V1. The tendency of these values is similar to the results of funnel viscosity tests.

The pressure filtration test was carried out in order to determine filtration rate and filter-cake properties when a grout slurry was forced against the medium under pressure. The device is shown in Fig. 5.

In the measuring process the grout was pressed into the pressure vessel with a volume of about 300 mL, a



(1) Kunigel V1 (2) Finely powdered Kunigel V1



(3) Kunibond (4) Finely powdered Kunibond

Fig. 4 Shear stress-shear rate curve.

Table 2 Plastic viscosity and yield value (FP: finely powdered).

Concentration (%)		2	4	6	8	10	12	16	20	24	28
Kunigel V1	Plastic viscosity, (mPa s)	1.99	2.98	6.95	15.89	29.79					
	Yield value, (Pa)	0	0.51	0.51	4.32	18.8					
Kunigel V1 (FP)	Plastic viscosity, (mPa s)	1.49	2.48	4.47	9.43	22.8	53.1				
	Yield value, (Pa)	0.25	1.02	0.51	1.52	7.62	37.1				
Kunibond	Plastic viscosity, (mPa s)				1.99	1.99	1.99	2.98	3.48	4.47	4.97
	Yield value, (Pa)				0.51	0.51	0.51	1.02	1.52	2.54	5.57
Kunibond (FP)	Plastic viscosity, (mPa s)				1.49	1.49	1.49	1.99	3.48	4.47	5.46
	Yield value, (Pa)				0.25	0.40	0.51	1.02	1.02	2.03	4.82

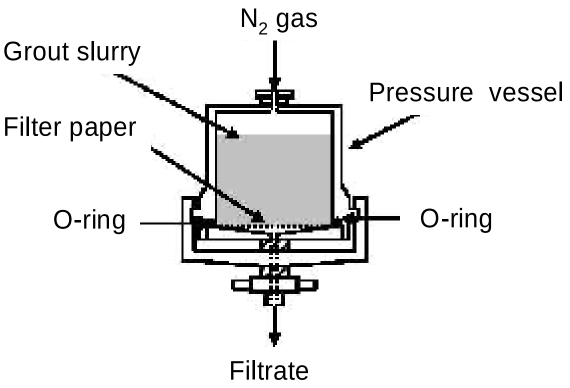
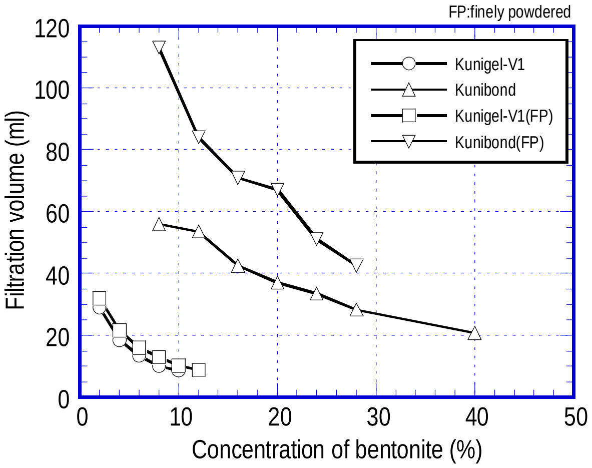
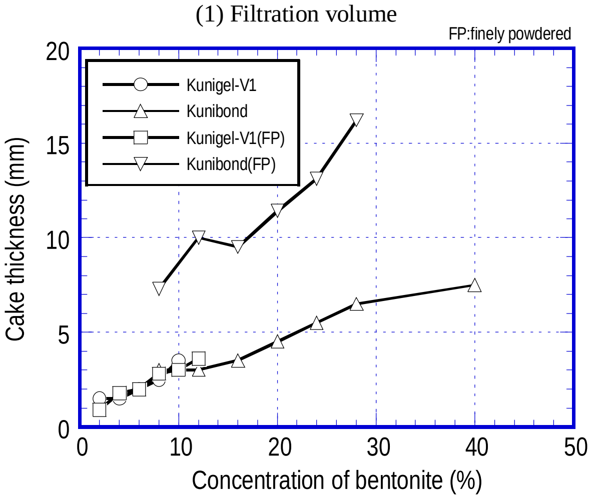


Fig. 5 Filter press.



(1) Filtration volume



(2) Cake thickness

Fig. 6 Results of pressure filtration tests.

diameter of 90 mm and a height of about 70 mm . The grout was pressed through the filter at 0.3 MPa pressure. Whatman filter paper No. 50 was used with a particle retention of 2.7 μm. The filtration volume was measured for 30 minutes. After that, filter cake on the filter paper was measured. The filtration volume and cake thickness are shown in Fig. 6.

The filtrate volume and cake thickness at the same concentration of Kunigel V1 were similar without recourse to pulverization. In the case of Kunibond, the discrepancy of these properties with the finely powdered material was greater. Bleeding means water separation from grout particles. Bleeding in the grout is dependent largely on particle size, concentration and admixtures. Graduated cylinder is used for determining bleeding. Water separation is measured with a tin and pipette in the laboratory. 1,000 mL of the grout is poured into a 1,000 mL graduated cylinder. Observations will be made after 2 hours setting the grout to the graduated cylinder: the amount of the settled water is measured with a pipette. Bleeding is given in percent. Fig. 7 presents the bleeding of each grout used in this study. Bleeding occurs when the Kunibond sets, which depends largely on concentration and particle side of bentonite. However, the Kunigel V1 resulted in zero bleeding.

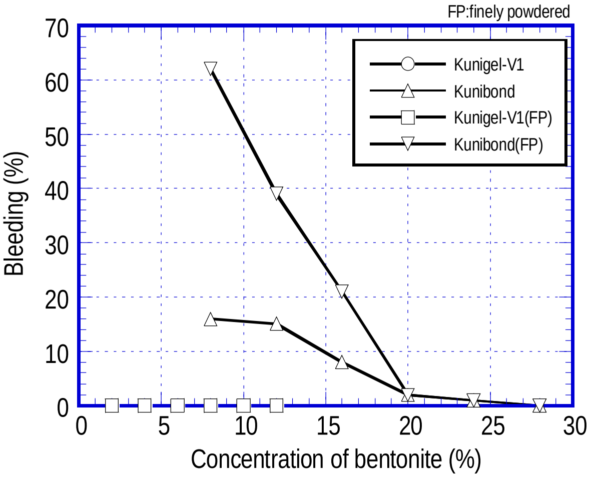


Fig. 7 Results of bleeding tests.

3. Grout Penetration Test with Pulverized Clay-Based Grout

3.1 Test Procedure

The penetration test is used to determine the outflow volume or penetration length of grout in the laboratory. The standard is based on the method of measuring the penetrability of a product in a capillary network. The test was originally done to measure the flow rate of injection material through a column of graded sand. The aim of the test was to avoid difficulties occurring while recognising and measuring cracks in a medium. A simplified block diagram of an acrylic mould 5 mm thick, 50 mm inside diameter and 205 mm long is shown in Fig. 8. Sand for grout penetration was put into the mould. The grout injection pressure in the pressure tank was set at a constant pressure of 0.3 MPa with air pressure controlled by a regulator, based on the injection experience in the TSX. The time from initial penetration to discharge, the evolution of discharge with time was monitored and after the test was completed, the performance of grout penetration into sand was observed. The sand used is silica sand No. 6 produced in Yamagata Prefecture (http://www.catvy.ne.jp/~ktsangyo/data1.htm), with the average particle diameter of 0.34 mm shown in Fig. 1, specific gravity of 2.6, and approximate hydraulic conductivity of 2.20×10^{-4} to 8.90×10^{-3} m/s.

The test cases of penetration test are as follows: For

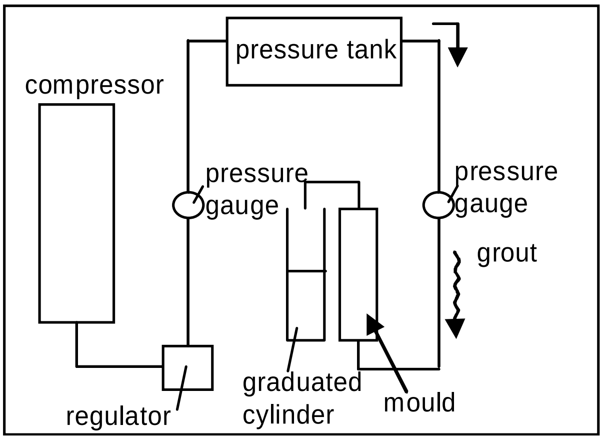


Fig. 8 Simplified block diagram of penetration test.

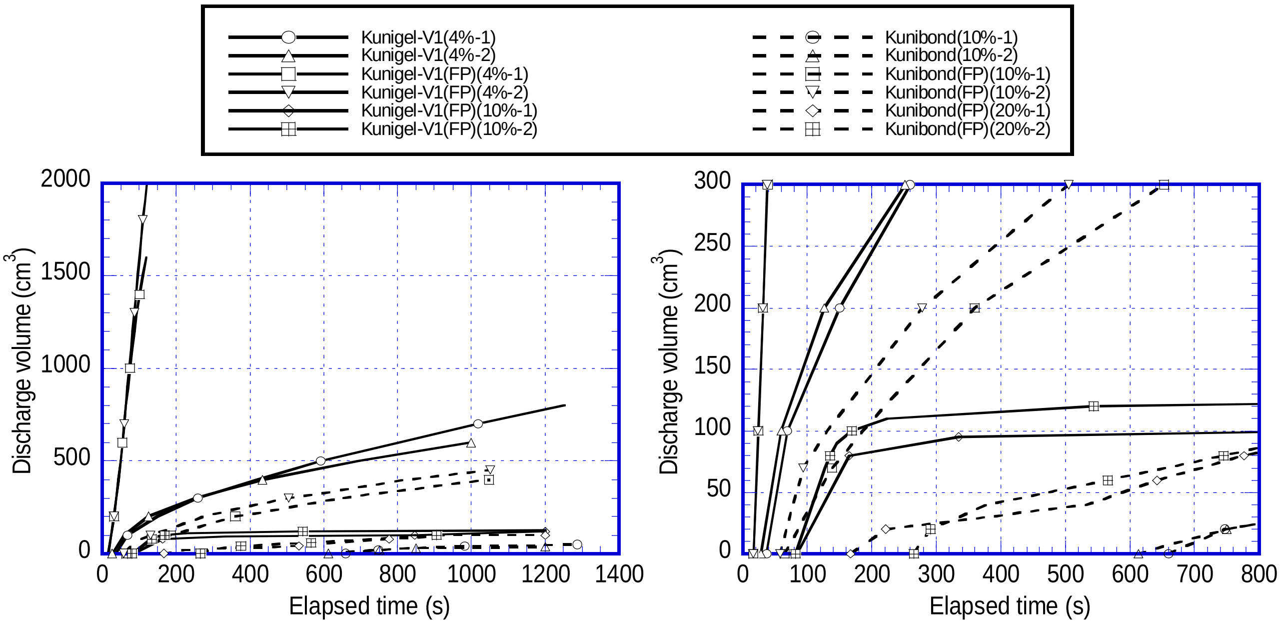
Kunigel V1, bentonite concentrations of 4% and 10% were selected in consideration of cases with differences in viscosity. For Kunibond, since the difference in viscosity for bentonite concentration was smaller, the bentonite concentrations of 10% and 20% were selected in consideration of the case of difference in bleeding. Two runs were conducted for each case.

3.2 Results

Table 3 presents the results of the penetration tests. Fig. 9 presents the evolution of discharge volume with time. In the case of Kunigel V1 with a 4% concentration, the average time from initiation of penetration to discharge was 32.5 sec for normal and 18.0 sec for finely powdered grout. In the case of Kunibond with a 10% concentration, the average time from initiation of penetration to discharge was 636.5 sec for normal and 61.5 sec for finely powdered grout. Discharge was not observed for Kunigel V1 with a 10% concentration and Kunibond with a 20% concentration. The average time from initial penetration to discharge of finely powdered Kunigel V1 with a 10% concentration was 83.0 sec and 216.0 sec for finely powdered Kunibond with a 20% concentration. Comparisons of the same bentonite type at the same concentrations, indicated that the time from initial penetration to discharge of the finely powdered material was faster than that of the normal material. It suggests that the particle size probably

Table 3 Results of penetration tests (FP: finely powdered).

	Concentration (%)	Times	Initial volume (g)	Discharge time (s)
Kunigel V1	4	1st	669.3	37
		2nd	663.0	28
	10	1st	666.2	No discharge
		2nd	662.5	No discharge
Kunigel V1 (FP)	4	1st	664.3	17
		2nd	661.8	19
	10	1st	668.1	84
		2nd	667.5	82
Kunibond	10	1st	663.0	613
		2nd	664.3	660
	20	1st	669.2	No discharge
		2nd	664.0	No discharge
Kunibond (FP)	10	1st	663.2	65
		2nd	664.8	58
	20	1st	666.8	167
		2nd	662.9	265



(a) Normal view (b) Enlarged view

Fig. 9 Evolution of discharge volume with time (FP: finely powdered).

(The numbers within parentheses identify the concentration and number of times.)

dominated behaviour of grout in the sand, because the grout injection pressure of 0.3 MPa was sufficiently larger than the apparent yield stress (Fig. 4) and the differences in funnel viscosity between normal and finely powdered materials were less than 3%. Consequently, viscosity of bentonite in the present study had no effect on the time from initiation of penetration to discharge.

In order to understand the effect of grain size of materials on penetration behavior, the relationship between the non-dimensional time computed by dividing elapsed time by discharge time and the non-dimensional volume computed by dividing injection volume by that at discharge for some materials having similar viscosity is shown in Fig. 10. Though Kunigel V1 (4% concentration), Kunibond

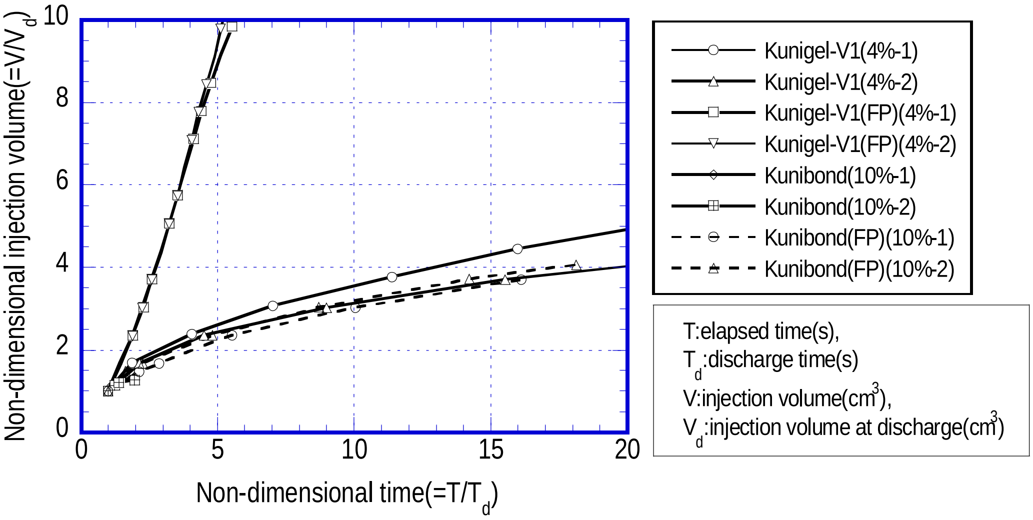


Fig. 10 Relationship between non-dimensional time and non-dimensional injection volume (FP: finely powdered).

(10% concentration) and finely powdered Kunibond (10% concentration) have similar penetration behavior, the rate of injection volume of finely powdered Kunigel V1 (4% concentration) is faster than others. This difference is due to grain side distribution of material and it suggests that pulverized bentonite is resistant to clogging in the host medium and thus that it can penetrate more readily.

3.3 Discussion

It is essential for development of reliable technology for a repository to improve penetration of clay-based grout. This paper presents the results of experimental studies on penetration of clay-based grout by pulverization and the efficacy of pulverized bentonite on penetration.

Considering the geological conditions and environmental standards in Japan, grouting technologies applicable to high water pressure deep underground during the construction of geological disposal facilities for high-level radioactive waste are of great importance. Although cementitious grout materials are commonly used for rock grouting, they have limited penetrability for small fractures and their high pH plume are considered to have an adverse effect on the long term safety of a geological disposal system. Considering the complexity of geological structures in Japan, grouting of smaller fractures, aperture < 100 μm, is expected. Therefore, it is necessary to confirm the applicability of new grout materials, which have better penetrability and are environmentally more friendly than cementitious grout materials. The average particle diameters are 20 μm for ordinary portland cement and a few μm for the ultra fine cement.

The bentonite used, which is predominantly Montmorillonite, has no adverse environmental effect. The average particle diameters are 10~30 μm and few μm of the pulverized one. Notably, the bentonite seems to have better penetrability with pulverization of the clay-based material. But, bentonite has lower strength than cementitious material and is hard to deal with because of remarkable swelling in water. It is necessary to resolve these problems in order to use it as an alternative material to cementitious grout materials. At present, it is preferred that pulverized bentonite grout be used temporarily considered as the post-excavation grout material applied for further reduction of permeability after pre-excavation grouting using other material.

4. Conclusion

For the geological disposal of high level radioactive wastes, an EDZ having a high hydraulic conductivity resulting from the development of fractures in the rock adjacent to tunnels will be one of the major potential pathways for radioactive contaminant transport. The potential pathways will be sealed by the engineered closure components, that is, a combination of tunnel plugs, backfill and grout. The likely candidate material would be a clay-based mixture taking into consideration the long-term stability of the seals. Clay-based grout is one of the effective materials that can be used to interrupt the migration of radionuclides through an EDZ.

The laboratory testing of clay-based grout using pulverized bentonite with the objective of improvement in grout penetration was conducted. The results obtained in this study indicate as follows:

(1) The viscosity of bentonite had no effect on the time from initiation of penetration to discharge and the difference in the penetration behavior is due to grain size distribution of the test material.

(2) Comparisons of the same bentonite type at the same concentrations, indicated that the time from initial penetration to discharge of the finely powdered material was faster than that of the normal material. It suggests that the particle size probably dominated behaviour of grout in the sand.

From the results of the laboratory tests, it has been concluded that pulverized bentonite material is resistant to clogging in the grouted medium and has a positive effect on penetration of grout material.

In addition, further research and development are required to widen the application as an alternative material to cementitious grout materials. In future work, an injection method for clay-based grout will be included.

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