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A Study on the Hydraulic Properties of Domestic Clay/Crushed Rock Mixture for the Backfill Material in a Radioactive Waste Repository

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방사성폐기물 처분장 되메움재를 위한 국산점토/분쇄암석 혼합물의 수리특성에 관한 연구

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Abstract

The hydraulic properties of domestic natural clay/crushed rock mixture suggested as a candidate backfill material for the low and intermediate level waste repository were investigated. The dry density-water content relationship was studied to define an optimum water content that gives a maximum attainable dry density at constant compaction pressure. The hydraulic conductivities of clay/crushed rock mixture as a function of clay content were also measured. As the clay content decreased, the maximum attainable dry density increased and the optimum water content became more distinct. However the attainable density is not significantly sensitive to water content. The hydraulic conductivities of the mixture increased from 5×10^{-12} m/s to 7×10^{-10} m/s with clay content decreasing from 100 wt.% to 25 wt.% at dry density of 1.2 Mg/m^3. In case of dry density of 1.5 Mg/m^3, they maintain the lower values of 5×10^{-12} m/s even at 25 wt.% clay content. The concept of effective clay dry density was suggested to estimate the hydraulic conductivity of the mixture. It was shown that the effective clay dry density concept can explain well the hydraulic conductivities of the mixtures with various dry density and crushed rock content.

요 약

중저준위 방사성폐기물 처분장 되메움재 후보물질로 제안되고 있는 국산 천연점토와 분쇄암석의 혼합물의 수리특성을 조사하였다. 혼합물의 수분함량 변화에 따른 혼합물의 밀도 변화를 조사하여, 동일 압축력 하에서 최대밀도 를 얻을 수 있는 최적수분함량을 찾고자 하였으며, 혼합물 중의 점토함량에 따른 수리전도도 변화를 조사하였다. 혼합물 중 점토함량이 감소할수록 얻어 지는

최대밀도가 증가하였으며, 최적수분함량도 보다 명확해졌으나, 혼합물 의 밀도는 수분함량에 그다지 민감하지 않았다. 혼합물의 수리전도도는 점토 함량이 감소할수록 증가하여 건조밀도 1.2 Mg/m^3 일 때 100% 점토인 경우의 3 x 10^{-12} m/s 에서 25% 점토함량의 경우에는 7 x 10^{-10} m/s 로 증가하였으나, 건조밀도가 1.5 Mg/m^3 일 때에는 25% 점토함량의 경우에도 5 x 10^{-12} m/s 의 낮은 값을 유지하였다. 혼합물의 수리전도도 추성을 위한 유효점토건조밀도 개념이 제안되었으며, 이 개념은 다양한 건조밀도 와 분쇄암석 함량 을 가진 혼합물의 수리전도도를 잘 설명할 수 있었다.

1. Introduction

The safety of the radioactive waste disposal can be assured through the function of multi-barriers such as waste form, engineered barrier, and geosphere. The engineered barrier is a near-field component that is closely related to the design of waste repository. The study on the characteristics of engineered barrier is considered to be important because its future behaviours can be predicted and controlled.

The backfill is a major component of engineered barrier and has two important functions on the viewpoint of control of radionuclide release : (1) to minimize contact between waste container and the host environment, and (2) to restrict the release of radionuclides into the host environment in the event of waste container failure. To perform these functions, the required properties of backfill material are as follows : low hydraulic conductivity, efficient compactability, high swelling potential, negligible shrinkage upon drying, low swelling pressure, low segregation tendency, and adequate compressibility [1]. Among them, low hydraulic conductivity is considered to be one of the most important requirements.

Bentonite has been considered as a potential backfill material because of its low permeability, high sorption capacity, self sealing characteristics , and durability in nature [2]. In the repository for the low-level wastes in Korea, the bentonite will be used as a backfill material not a buffer material. Also high quality bentonite might not be available in sufficient quantities to be used as a backfill material in the case of a large repository, and the economic aspect should be considered. Therefore the natural bentonitic clay instead of pure Na-bentonite has been considered as a backfill material. As the bentonitic clay has poor mechanical strength, and generates high swelling pressure, the addition of inert materials such as sands or crushed rocks to clay has been proposed as a potential alternative. Thus the mixture of natural clay and crushed rock is being suggested as a candidate backfill material [3]. It is however possible that the addition of crushed rock to clay would increase the hydraulic conductivity, and the major properties of the mixture might be varied with the ratio of rock and clay.

This study intended to examine the dry density-water content relationship for clay/crushed rock mixture, and hydraulic conductivities of the mixtures with various clay contents and dry densities. An attempt was also made to present the methodology for the estimation of hydraulic conductivities of the mixtures with various crushed rock content and dry density from that of clay.

2. Experiment

Materials

Clays and rock aggregates used in this study were selected as follows. The clay was a Ca-bentonite from Kampo, Kyungsangbuk-do, Korea, and contains approximately 53.2% SiO_2, 22.1% Al_2O_3, 8.4% F_2O_3, and some minor elements (Table 1). Its detailed mineralogical composition

Table 1. Chemical Composition of the Domestic Natural Clay from Kampo, Kyungsangbuk-do

Chemical composition	wt. %
SiO_2	53.20
Al_2O_3	22.05
Fe_2O_3	8.37
FeO	0.32
CaO	2.63
MgO	1.98
K_2O	0.96
Na_2O	1.36
MnO	0.11
IG-Loss	9.02

and some basic properties were determined by Moon et al.[4]. The rock aggregate was a crushed granite from Daeduk, Taejeon. A distilled water was used for the sample preparation and subsequent testing.

Dry Density-Water Content Relationship

Particle Size Distribution

The initial step taken to acquire the proper gradation of the candidate backfill materials was to evaluate the gradation that would lead to maximum compaction dry density. When the dry densitty is at its maximum by using the same compaction effort, the hydraulic conductivity of the material should be close to its lowest value. This would satisfy one of the primary requirements of the backfill materials.

When the rock was crushed to prepare rock aggregates using a rock crusher and a ball mill, the wide range of rock particle size distribution was obtained and the shape of rock aggregates was irregular sphere. The use of clay/crushed rock mixture would lead to the maximum dry density with proper particle size distribution.

The particle size distribution of the crushed rock was specified using the relationship proposed by Talbot and Richart for an ideal gradation curve to yield a maximum compacted density of mixture [5]. The ideal gradation curve to maximize the density of the mixture can be expressed in a simplified form as

$$P = 100(d/D)^n \qquad (1)$$

where P is the percentage by weight of material that passed a given sieve with opening of width d. D is the maximum particle size of given aggregates, and n is a variable exponent.

The value of exponent n was determined by the clay content in the mixture. If the d in Eq.(1) for clay was set at 0.074 mm (No.200 sieve size), and D for rock aggregates was set at 4.70 mm (No.4 sieve size), the value of n was varied between 0.09 and 0.72, which corresponded to clay content of from 15% to 70%.

Compaction Tests

The dry density-water content relationship for the clay mixed with up to 85 wt.% crushed rock was determined by a small scale compaction apparatus. This apparatus produces specimens, 70 mm in height and 50 mm in diameter. The procedure used for the compaction of the mixture is as follows; The desired crushed rock-clay mixture was prepared from oven-dried crushed rock and clay. The water was added to reach the desired moisture content of the mixture, and the mixtures were left covered overnight to equilibrate. The pre-determined amount of the mixture was poured into a stainless steel cylindrical mold and compacted to investigate the dry density-water content relationship. The density of specimen was determined by weighing.

Hydraulic Conductivity

The apparatus used to determine the hydraulic conductivity of the samples is designed to supply water to the sample at the pressure of 1.3 Kg/cm^2 (Fig.1). The cylindrical chamber has an inner diameter of 50 mm. The mixture samples are rigidly confined by using a restrainig ram. The compacted mixture samples with pre-determined dry density were placed in the sample chamber, and the water flows from the bottom to the top of the sample chamber. The hydraulic conductivities of the samples were then determined at room temperature using distilled water. The penetrated water volumes were measured by weighing. The hydraulic conductivity at a given hydraulic gradient was monitored until eqilibrium was established. The flow of water through the mixture was assumed to obey Darcy's law.

3. Results and Discussion

Dry Density-Water Content Relationship

The dry density-water content relationships for the clay/crushed rock mixture are shown in Fig.2 and Fig.3.

As the clay content increased, the maximum attainable dry density decreased and the corresponding optimum water content increased. Also in the mixture with less clay content, the optimum water content to give maximum dry density is relatively more distinct. Dixon et al. [6] and Pusch [7] reported a similar behaviour in the Na-bentonite and the Na-bentonite/sand mixture system, respectively. On the other hand, Yong et al. [1] reported the distinct maximum dry density at an optimum water content was observed over wide range of clay content for the mixture of the crushed granite and the Lake Agassiz clay that is a natural clay containing about 20% illite and 35% montmorillonite [8].

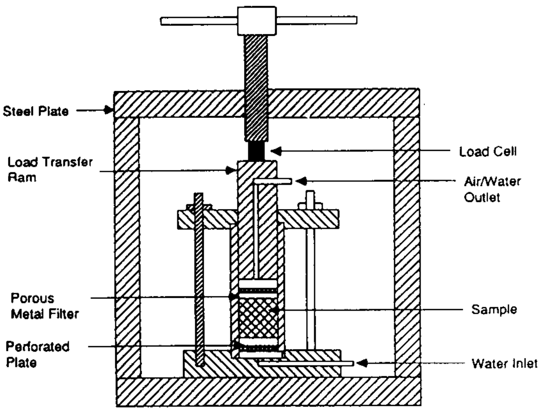

Fig. 1. Schematic Diagram of Apparatus for Measuring Hydraulic Conductivity of Clay/Crushed Rock Mixture

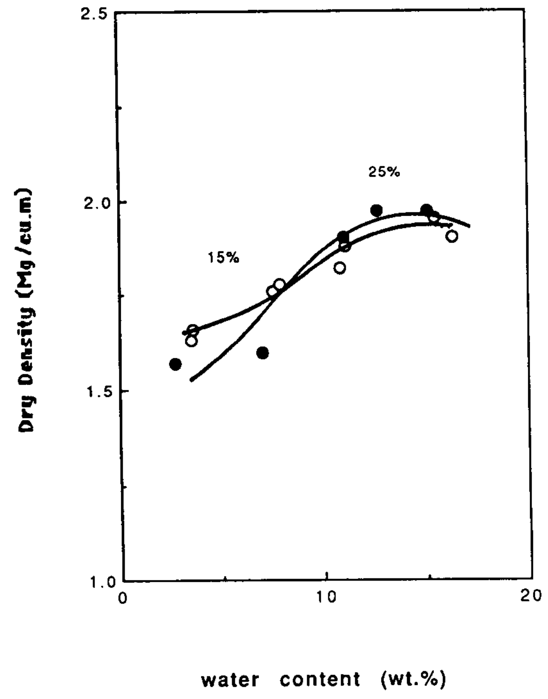

Fig. 2. Relationship Between Dry Density and Water Content for the Mixture of Clay/Crushed Rock (I)

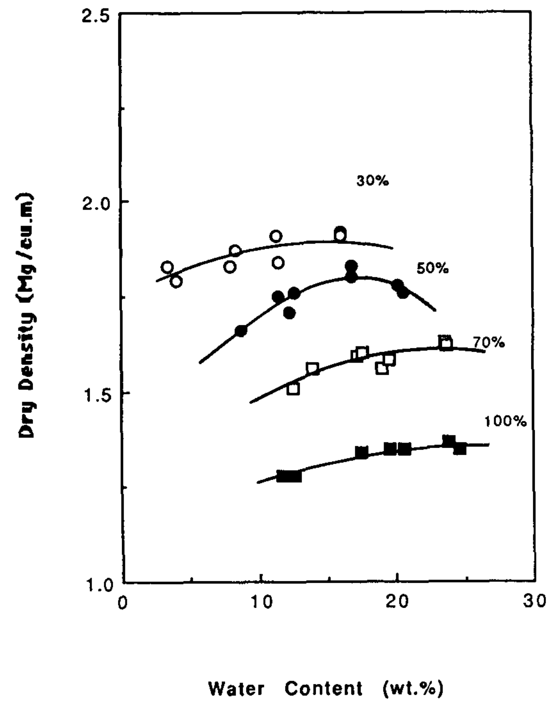

Fig. 3. **Relationship Between Dry Density and Water Content for the Mixture of Clay/Crushed Rock (II)**

This disagreement is likely due to the difference of smectite content in the clay. The smectite has a high affinity for water, and the attached water on smectite shows higher viscosity than free water [9], Therefore for the mixture with higher bentonite content, the compaction energy might be insufficient to overcome the higher shearing resistance of the attached water. At the optimum water content, mixture with low bentonite content possesses a higher ratio of water to clay than those with higher bentonite content, and water is probably more mobile in the mixtures with low clay content. Hence the applied compaction energy is sufficient to overcome internal shearing resistance. This is supported by Dixon's results [6] that for the illite/sand mixture, the optimum water content is evident. The crushed rock may also contribute to compaction by allowing more efficient distribution of energy through the system. However, even if water content is less than the optimum value, the attainable density of mixture is not significantly sensitive to water content.

These results for the domestic clay-crushed granite mixture as a candidate backfill material suggest that the stringent control of water content would not be required during the in-situ compaction of backfill in the waste repository.

Hydraulic Conductivity

When the mixtures of clay and inert material such as crushed rock are used as backfill in a waste repository, their hydraulic conductivities should be as low as possible.

The results of the hydraulic conductivity test are shown in Fig.4 and Fig.5. The typical flow characteristics of the mixture are presented in Fig.4. For a short period of time immediately after starting experiment, the outflow is high because of the unsaturated nature of mixture, and then the amounts of flow start to decrease substantially until a steady state is attained after about 10 days. The hydraulic conductivities were obtained under a steady state condition, and are presented in

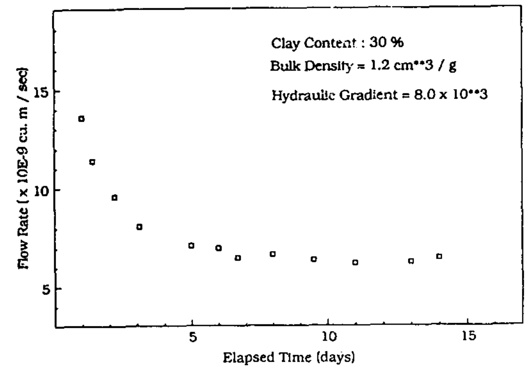

Fig. 4. **Typical Flow Rate-Time Relationships for the Clay/ Crushed Rock Mixture**

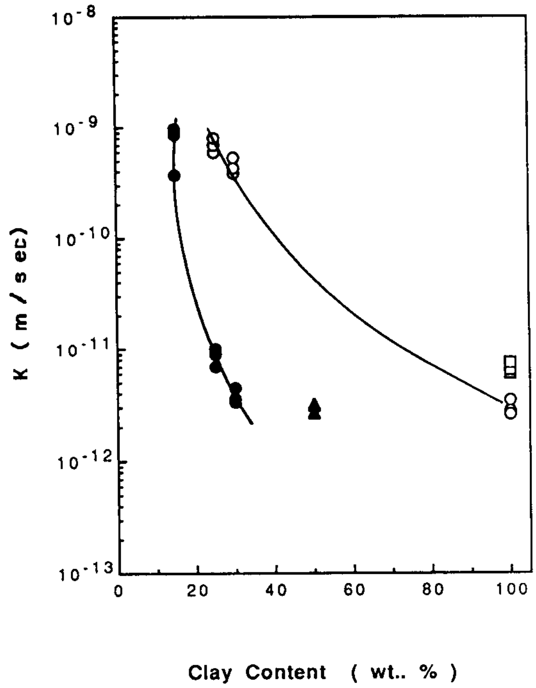

Fig. 5. **Hydraulic Conductivity of Clay/Crushed Rock Mixture as a Function of Clay Content (from this study, —□— : ρ_d=1.0 Mg/m^3, —○— : ρ_d=1.2 Mg/m^3, —●— : ρ_d=1.5 Mg/m^3, —▲— : ρ_d=1.7 Mg/m^3)**

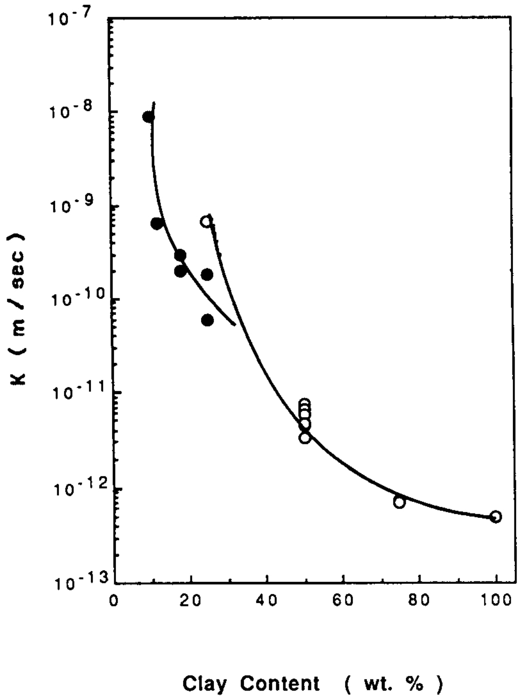

Fig. 6. **Hydraulic Conductivity of Clay/Inert Material Mixture as a Function of Clay Content (—○— : from Westsik et.al [10], —●— : from Yong et.al [1])**

Fig.5. As shown in this figure, the hydraulic conductivities of clay/crushed rock mixtures decrease with increasing clay content, They range from about 7 x 10^{-10} m/s at clay content of 25 wt.% to less than 3 x 10^{-12} m/s at clay content of 100 wt.% at the dry density of 1.2 Mg/m^3. When the dry density increased to 1.5 Mg/m^3, the hydraulic conductivities of mixtures decreased considerably and even at 25 wt.% of clay content, the hydraulic conductivity of mixture reaches about 5.0 x 10^{-12} m/s. Young et al.[1], and Westsik et al.[10] reported the similar results for natural clay/crushed rock mixture, and bentonite/quartz sand system respectively (Fig.6).

These are largely the results of decreasing clay density with increasing crush rock content at constant bulk density. However even at 70 wt.% of crushed rock content, the hydraulic conductivities of mixtures are considerably lower than about 10^{-9} m/s so that the principal mechanism of radionuclide transport through backfill material will be molecular diffusion. These results, however, show the non-linear and qualitative trend that the hydraulic conductivity increases with increasing inert material content, and it is difficult to estimate the hydraulic conductivities for the mixtures with various clay content and dry density from that of clay. To clarify this relationship, instead of using dry density, the concept of "effective clay dry density" suggested by Gray et al.[11] to explain the swelling pressure of the mixture of clay and sand was introduced.

The effective clay dry density ρ_e (Mg/m^3) is defined as

$$\rho_e = \frac{\text{mass of clay in the system}}{\text{combined volume of clay plus void}} \qquad (2)$$

and for 100 wt.% clay system,

$$\rho_e = \rho_d \qquad (3)$$

where ρ_d is the bulk dry density. The effective clay dry density of mixture decreases with increasing inert material content at the same dry density of mixture. For pure clay, the logarithm of hydraulic conductivity (log K) decreased almost linearly with an increase in the clay dry density [7,10]. According to Darcy's law,

$$Q = K\,A\,(dh/dl) \qquad (4)$$

where Q is volumetric flow rate (m^3/s), A is area normal to flow (m^2), (dh/dl) is hydraulic gradient, and K is hydraulic conductivity (m/s). When the mixture of clay and crushed rock is considered, the area available to flow might be decreased because the rock aggregates can be impermeable, and Eq.(4) becomes

$$Q = K'\,A'\,(dh/dl) \qquad (5)$$

where K' is a hydraulic conductivity of the mixture based on the available cross sectional area to flow, A', and A/A' is given as

$$A/A' = \rho_e/[\rho_d(1-\psi_r)] \qquad (6)$$

where Ψ_r is the weight fraction of rock in the mixture. Therefore if the concept of effective clay dry density is appropriate to explain the hydraulic conductivity of mixture, log [K(A/A')] is decreased almost linearly with increasing effective clay dry density. Also K(A/A') of mixture with high inert material content should show similar value as K for 100 wt.% clay system that dry density is the same as the effective clay dry density of mixture. The relationship between the effective clay dry density and K(A/A') for the mixture is shown in Fig.7. The hydraulic conductivity data shown in Fig,6 obtained by Young et al.[1] and Westsik et al.[10] are also re-plotted as a function of effective clay dry density, instead of clay content to test the availability of the effective clay dry density concept, and the results are shown in Fig.8. As shown in these figures, except for some data points, when the clay content is above 15 wt.%, log [K(A/A')] of mixture decreases almost linearly with increasing effective clay dry density. If the effective clay dry densities are same, K(A/A') for the mixture and 100 wt.% clay system gives a similar value. The deviation of some data points (at clay content of 25, 30 wt.%, ρ_d=1.5 Mg/m^3) from the trend might be due to the characteristics of crushed rock aggregates being used as inert

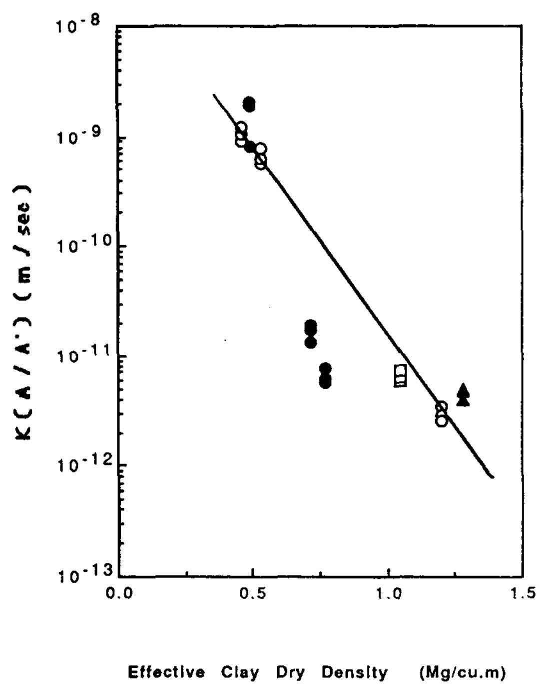

Fig. 7. Hydraulic Conductivity of Clay/Crushed Rock Mixture as a Function of Effective Clay Dry Density (from this study, —□— : ρ_d=1.0 Mg/m^3, —○— : ρ_d=1.2 Mg/m^3, —●— : ρ_d=1.5 Mg/m^3, —▲— : ρ_d=1.7 Mg/m^3)

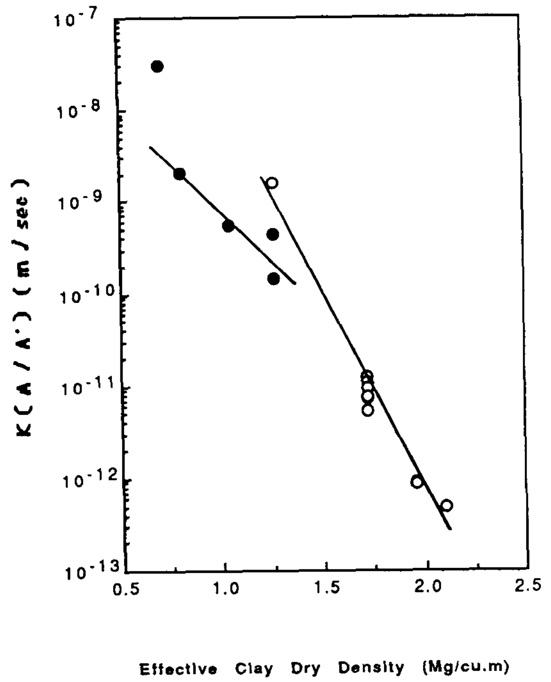

Fig. 8. Hydraulic Conductivity of Clay/Inert Material Mixture as a Function of Effective Clay Dry Density (—○— : from Westsik et.al [10], —●— : from Yong et.al [1])

material. The crushed rock aggregates have fairly irregular shapes, and their size distribution covers a wide range, and average size is much larger than that of clay particle. Therefore it is possible that test sample specimens have non-homogeneous structures which results in the deviation. These results indicate that the concept of effective clay dry density is useful to explain the increase of hydraulic conductivity for the mixture of clay and inert material. Further studies are needed to test in detail the availability and limitation of the effective clay dry density concept.

Therefore the hydraulic conductivities of mixtures with various inert material content can be estimated in order-of-magnitude from the effective clay dry density of mixture and hydraulic conductivity for the 100 wt.% clay system. In the cases of very low clay content (<10 wt.%), this relationship might not be applicable because of the insufficient amount of clay. At very low clay content, the amount of clay in the mixture may not be sufficient to occupy the whole space between the inert material grains.

4. Conclusions

The hydraulic properties of clay/crushed rock mixture suggested as a candidate backfill material for the low and intermediate level waste repository were investigated. As the clay content decreased, the maximum attainable dry density increased, and the optimum water content became more distinct. However, the attainable density is not significantly sensitive to water content, and the stringent control of water content would not be required during in-situ compaction of backfill in the repository.

The hydraulic conductivities of the mixture increased with decreasing clay content. However, even at low dry density of 1.2 Mg/m^3 and clay content of 30 wt.%, the hydraulic conductivities are considerably low, and the principal mechanism of radionuclide migration through backfill would be diffusion. Also the results show that the concept of "effective clay dry density" is useful to estimate roughly the hydraulic conductivities of mixtures with various dry densities and clay contents. Further studies are needed to test in detail the availability and limitation of the effective clay dry density concept, and the additional experimental works will be required.

Acknowledgement

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