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ASSOCIATING CHARACTERIZATION OF BENTONITE-BASED BUFFER/BACKFILL MATERIALS BY DISTRIBUTION RATIO (R_d) AND PLASTIC INDEX (PI)

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Key words: Plastic index (PI), distribution coefficient (R_d), buffer material, additivity, seawater, groundwater.

GW. These results provide useful information on the choice of the composition of the buffer material, and for use in the evaluation of the overall R_d and PI of the mixture from the R_d and PI of each buffer component.

ABSTRACT

Choosing buffer materials with the required engineering and sorption characteristics for radwaste disposal is very important. This work investigates the relationships between engineering characteristics (plastic index; PI) and sorption properties (distribution ratio; R_d) for two mixed buffer materials that are composed of bentonite/quartz sand and bentonite/laterite, to provide an overall functional evaluation of buffer materials. Se is the nuclide of interest, and both synthetic groundwater (GW) and seawater (SW) were employed in batch sorption experiments. Deionized water (DIW) was used in engineering property tests. SW and GW were also used to evaluate the effects on PI . The results indicate that PI was proportional to the bentonite content of the tested buffer materials, independently of the solution used. The coagulation and flocculation effects lead to the ordering $PI_{DIW} > PI_{GW} > PI_{SW}$. The sorption of Se increased with the bentonite content in bentonite/quartz sand mixtures, and the sorption fraction increased with the laterite content in bentonite/laterite mixtures. These findings demonstrate that the sorption of Se on soil follows the order, laterite soil > bentonite > quartz sand, regardless of whether GW or SW was used. Hence, adding laterite to the mixture improved the sorption of the anionic form of Se. A mixed material with a PI value of approximately 50 to 70 in GW and SW effectively balances the engineering needs with the desired chemical properties. A bentonite content of 50% and the addition of both laterite and quartz sand may be optimal for buffer materials, and is thus worthy of further study. This work further indicates that the PI values of both mixtures followed the additivity rule in SW and GW, but not in DIW. The R_d s value of Se indicated the good additivity of the bentonite/laterite mixtures in both SW and GW and of the bentonite/quartz sand mixtures in SW, but not of the bentonite/quartz sand mixtures in

INTRODUCTION

Buffer materials for high-level radwaste disposal must exhibit particular engineering (hydraulic conductivity, thermal conductivity, mechanics, swelling and other properties) and chemical (sorption, diffusion, migration transport, and others) characteristics [10, 23]. Recent studies of buffer materials focused on physical properties [2, 13, 19]; others focused on chemical properties [3, 14, 22], and a few addressed both engineering and sorption properties [9]. Learning about both the engineering and the sorption properties of potential buffer-backfill materials, mixed with various soil materials, is important.

Bentonite clay and quartz sand have been selected as potential buffer and backfill materials [11]. Bentonite is associated with a high cation exchange capacity, a high swelling potential and a low hydraulic conductivity, while quartz sand is associated with high thermal conductivity and strong mechanical properties. However, radioactive anionic species in the buffer materials mentioned above are not expected to exhibit high sorption capacity. Accordingly, composite materials that have a high capacity for absorbing both radioactive cation and anionic species in groundwater must be developed.

Some investigations have pointed out that the sorption characteristics, R_d values, of the radionuclides (RN) with respect to particular mineral components of the mixtures can be used to evaluate the R_d values of the mixtures, providing additional benefits in choosing effective composite materials [8, 15]. Many studies on the additivity sorption behavior of radionuclides on mixtures have been conducted. Some studies have demonstrated that actinides such as Np and Am exhibit

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quite similar additivity, but the R_d values of Cs and Sr on mixtures tend to be overestimated [15]. The additivity rule for the sorption of Eu on red earth, bentonite and alumina mixtures is not generally applicable [6]. Furthermore, sorption of Se and Cs exhibited a good additivity on goethite, smectite and calcite mixtures, but Am did not [8]. Other studies for the additivity behavior of nuclides on different compositions of minerals focused on soil and their components [16, 17, 18]. Recently, the additivity rule and its applicability to soil were analyzed [21]. They pointed out that the additivity rule was formally valid based on the following assumptions; (a) sorption reaction is reversible and independent; (b) sorption reaction is in equilibrium; (c) surfaces are homogeneous; (d) ion exchangers or sorbents are insoluble; (e) no new phase is formed, and (f) no interactions occur among individual minerals of the mixture. Mixed buffer materials and complex solutions, such as seawater, are a few concerns in the cited studies.

A distribution ratio, R_d , was used to characterize materials to retard the transport of nuclides. If sorption is a surface process, then a proportional relationship must exist between sorption sites and RN. Additionally the R_d of a mixture can be written as

$$R_d \text{ (mL/g)} = \Sigma(m_i/M) \times R_{di} \quad (1)$$

where m_i is the mass of mineral i in the mixture; M is the total mass of the mixture, and R_{di} is the distribution ratio of RN on mineral i .

Plastic index (PI) values are typically applied to classify soil and evaluate various engineering properties such as compressive and swelling characteristics [4]. Plastic index (PI) was calculated by the Atterberg liquid limit (LL) and also by the plastic limit (PL) of soil as follows:

$$PI = LL - PL \quad (2)$$

However, investigation on the additivity of the plastic index of mixtures will be contributive to predict the engineering properties of mixtures.

Laterite, a progressively weathered soil, exhibited better anion sorption capability as the aluminum and iron oxide content increased [5,12]. Various mass ratios of bentonite/quartz sand and bentonite/laterite mixtures were selected in this work as potential buffer materials. Batch experiments were conducted to measure the R_d values of ^{75}Se on mixtures. Atterberg limit tests were conducted to measure LL and PL , to evaluate PI and analyze its additivity, yielding information about the elementary engineering properties of mixtures. Synthetic seawater (SW) and groundwater (GW) were used as the liquid phases to simulate the possible conditions

that may be encountered during geologic disposal in an island. This study aimed primarily at establishing the relationship between sorption and the engineering properties in composite materials should, however, be beneficial in selecting an optimal composite material mixture. Additionally, the additivity of PI and K_d can also be considered to improve the performance assessment of mixed buffer/backfill materials.

MATERIALS AND METHODS

1. Liquid phase

The solutions used in the batch sorption tests were synthetic compositions of GW and SW (Table 1) in which a trace amount of ^{75}Se added (Table 3), to simulate the possible geochemical conditions of a deep geological repository. Deionized water (DIW) was used as the solution in the Atterberg limit test to simulate the conditions for the *in-situ* buffer materials during the construction phase. GW and SW were also used to evaluate the effects on PI as they intrude into buffer/backfill materials.

2. Solid phase

The solid phase material used for both the Atterberg limit tests and the batch sorption tests were composed of varying proportions of bentonite/quartz sand and bentonite/laterite mixtures. The composite ratios of bentonite to quartz sand and bentonite to laterite used for the Atterberg limit tests were as follows; 10:0, 9:1, 7:3, 5:5, 3:7, 1:9 and 0:10 (by dry weight). The ratios of bentonite to quartz sand and bentonite to laterite in the

Table 1. The compositions of groundwater (GW) and seawater (SW)

Compositions	GW	SW
$\text{CaCl}_2 \cdot 2\text{H}_2\text{O}$ [g]	138.6634	30.3008
$\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ [g]	7.0260	215.8030
Na_2SO_4 [g]	16.5704	79.8975
NaHCO_3 [g]	0.2756	---
KCl [g]	0.3086	15.2157
NaBr [g]	1.0311	1.7267
$\text{SrCl}_2 \cdot 6\text{H}_2\text{O}$ [g]	2.1330	0.4639
LiCl [g]	0.1221	0.0212
NaF [g]	0.0672	0.0571
NaCl [g]	92.2581	482.2667
H_3BO_3 [g]	---	0.5145
Total solution volume [L]	20	20
pH	~ 7.5	~ 7.02

Table 2. The characteristics of solid phase

Characteristics	Bentonite	Laterite	Quartz sand
Particle size (mm)	< 0.074	< 0.074	0.297~0.84
Specific area (m ² /g)	27.96	32.25	1.98
CEC (meq/100g)	68.5	14.3	
Specific gravity	2.75	2.57	2.62
Major minerals	Quartz Montmorillonite	Quartz, hematite Kaolinite, goethite	Quartz
Major chemical composition	SiO ₂ (60.53%) Al ₂ O ₃ (20.12%) Fe ₂ O ₃ (3.14%) MgO (2.65) Na ₂ O (2.13%)	SiO ₂ (63.82%) Al ₂ O ₃ (23.12%) Fe ₂ O ₃ (10.9%)	SiO ₂ (99.5%)

Table 3. pH values of GW and SW in Se sorption on mixtures

Mixture	Solution	Radioactivity (Bq/mL)	Initial pH	Mixed Ratio				
				10/0	7/3 ³	5/5	3/7	0/10
B/Q ¹	GW	15,537 ± 145	6.65	7.75	7.67	7.65	7.56	7.19
	SW	12,776 ± 227	6.66	7.68	7.73	7.65	7.65	7.1
B/L ²	GW	15,537 ± 145	6.53	7.75	7.16	7.08	6.71	6.11
	SW	12,776 ± 227	6.51	7.68	7.35	7.17	7.11	6.05

Note: 1. Bentonite mix with quartz sand.
 2. Bentonite mix with laterite.
 3. 7/3 means bentonite 70% mixed with quartz 30% by dry weight.

batch sorption tests were 10:0, 7:3, 5:5, 3:7 and 0:10.

The laterite, progressively weathered soils, was taken from 3m to 4m below the surface in the Jungli area of northern Taiwan. The laterite soil sample was air-dried and ground to pass through a 0.074 mm sieve, and then heated for 3 hours by 800°C ± 50°C for removing organic matter. The MX-80 bentonite and quartz sand were commercial products. Table 2 lists the characteristics of MX-80 bentonite, laterite and quartz sand. Specific area was analyzed using N₂-BET (Micromeritics ASAP 2000), the sodium acetate method was used to determine cation exchange capacity, and XRD (CuK α radiation, MAC Sci., model MXP-18) and ICP-MS were used to identify the major elemental constituents in the solid phase.

3. Batch sorption tests

Batch sorption tests were conducted using a solid/liquid ratio of 1g/30mL. The liquid-phase solutions (SW and GW) were individually added to the solid phase materials with various composition ratios; they were placed in the centrifugal tubes in triplicate and shaken at 120 rpm. Samples were collected on the 14th day following centrifugalization at 10380 g, and the

residual radioactivity of ⁷⁵Se in the liquid phase was measured using an NaI (TI) γ counter. The pH values were measured using a glass electrode. The distribution ratio (R_d) of RN can be calculated from the following formulae;

$$R_d (\text{mL/g}) = \frac{S}{C} \quad (3)$$

where S is the concentration of RN sorbed on the solid phase (Bq/g), and C is the concentration of RN in solution (Bq/mL).

4. Atterberg limit tests

The Atterberg liquid limit test and the plastic limit test were performed on the solid-phase mixtures described above, using DIW, SW and GW. These tests were performed following the method of ASTM D4318, [1] and the PI value was calculated using Eq. 2.

RESULTS AND DISCUSSION

1. Sorption of Se on Mixtures

Table 3 lists the pH values after 14 days of batch sorption reactions. The buffer capacity of bentonite

caused the samples with bentonite contents from 30% to 100% to have pH values of between 7 and 8. The laterite is an acidic soil [7] with a pH of as low as approximately 6 in both SW and GW.

Figures 1 and 2 indicate that the sorption increased with the bentonite content in the bentonite/quartz sand mixtures. Additionally, the sorption fraction increased with the laterite content in the bentonite/laterite mixtures. The evidence suggests: that the sorption of Se on soils followed the order laterite soil > bentonite > quartz, independently of whether GW or SW was used, perhaps because the Se (SeO_3^{2-} and HSeO_3^- in aerobic aqueous solution) was sorbed primarily on the mineral surface by inner-complexation and outer-complexation; thus, the early sorption of cations on the surface of the soil is followed by an increase in positive charge, which increases the attraction for SeO_3^{2-} and HSeO_3^- . The specific surface area of laterite exceeded that of bentonite (Table 2) might lead to the sorption results. The ionic radius (1.84 Å) of Se does not allow it to replace the smaller ion in the lattice but to be sorbed only on the edges of clay minerals, the surface of oxides or the surface of hydroxides. On the other hand, the lower pH value the higher sorption of Se might lead to the higher sorption on laterite soil [20]. The pHs of the mixed bentonite/laterite solutions, especially that in only laterite soil, is lower than those of mixed bentonite/quartz sand solutions. Hence, adding laterite to the mixture could promote the sorption of the anionic form of Se and slightly decrease the pH buffer capacity.

2. Plastic Index of Mixtures

DIW was used to simulate the liquid phase under construction; while SW and GW were used to simulate the intrusion of water after the construction had been completed. These three solutions were independently used to determine the liquid limit and the plastic limit for bentonite/quartz sand and bentonite/laterite mixtures, and the *PI* values were calculated. The *PI* values, regardless of using the three liquid phases, were proportional to the bentonite content (Figures 3 and 4). As Das [4] pointed out, the high swelling potentials were responsible for high *PI* values of over 35. A higher *PI* value corresponds to lower water permeability. The experimental results demonstrated that the *PI* values for GW were lower than those for DIW, and those for SW were lower than those for GW ($PI_{DIW} > PI_{GW} > PI_{SW}$) in individual soil and bentonite/quartz sand mixtures. In bentonite/laterite mixtures, the *PI* values for GW and SW were lower than those for DIW. For all the mixtures, the *PI* increased significantly with the bentonite content. The bentonite content strongly affected the *PI* value. The relatively high cation concentrations in GW and

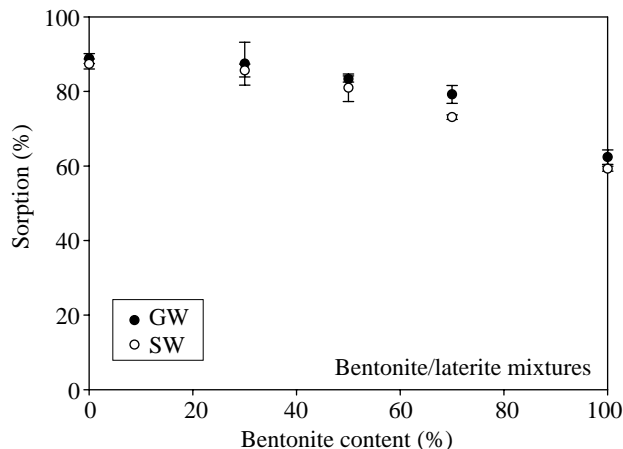


Fig. 1. Sorption percentage of Se in relation to bentonite content on bentonite/laterite mixtures.

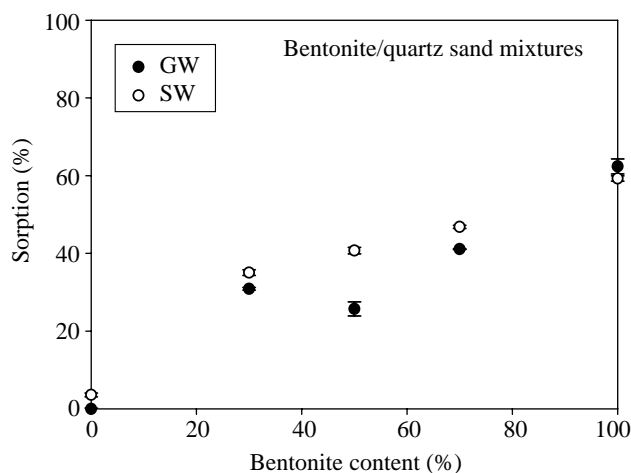


Fig. 2. Sorption percentage of Se in relation to bentonite content on bentonite/quartz sand mixtures.

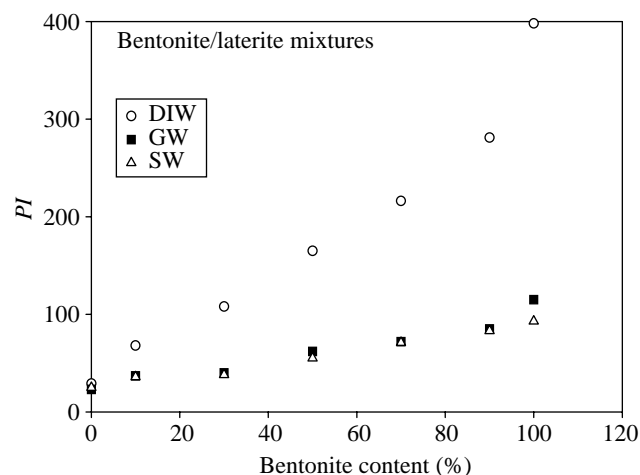


Fig. 3. Plastic index (*PI*) in relation to bentonite content on bentonite/laterite mixtures.

SW, and the coagulation and flocculation that might be caused by them probably were responsible for the compression for surface electrical double layer of the bentonite particle. This fact also reveals that the swelling potential decreased when GW or SW was used, and water permeability increased. However, neither GW nor SW significantly affected the PI values of quartz sand ($PI = 0$) and laterite ($PI = 20$ to 30).

3. Relationship between PI and R_d

The sorption measurements described in section 3.1 can be used to evaluate the sorption characteristics of mixtures. Likewise, the PI measurements made using SW and GW, and provided in section 3.2, can be used to evaluate the engineering properties such as water permeability, swelling potential and compression properties, among others. Therefore, establishing the relationship between R_d and PI provides an advantage in choosing and evaluating the possible compositions of mixture materials. Figures 5 and 6 plot the relationships between R_d s of Se and the PI values of mixtures in SW and GW, respectively. Figure 5 demonstrates that the R_d values decreased linearly as PI increased in both GW and SW, on bentonite/laterite mixtures. Figure 6 showed the opposite trend: the R_d values varied linearly with PI in both GW and SW on bentonite/quartz sand mixtures. The results indicate that adding either quartz sand or laterite to bentonite reduced PI (decreasing the swelling force and the difficulty of construction and compressibility, but increasing the permeability). Adding quartz sand might improve the mechanical and thermal conductivities of mixtures, but it reduces the capacity to absorb Se. Adding only laterite increased the sorption of Se. Consequently, a material with a PI of approximately 50 to 70 is preferred for balancing the engineering needs against desirable chemical properties. A bentonite content of 50% and the addition of both laterite and quartz sand may be optimal for buffer materials.

4. Additivity of PI and R_d

The following equation was used to analyze the additivity of PI from as a weighted average among the various soils;

$$PI(\text{overall}) = \frac{\sum (m_i/M) \times PI_i}{\sum (m_i/M)} \quad (4)$$

where m_i is the mass of soil i in the mixture; M is the total mass of the mixture, and PI_i is the plastic index of soil i .

The physical parameter, PI , is generally highly consistent with the additivity. Figures 7 and 8 indicate

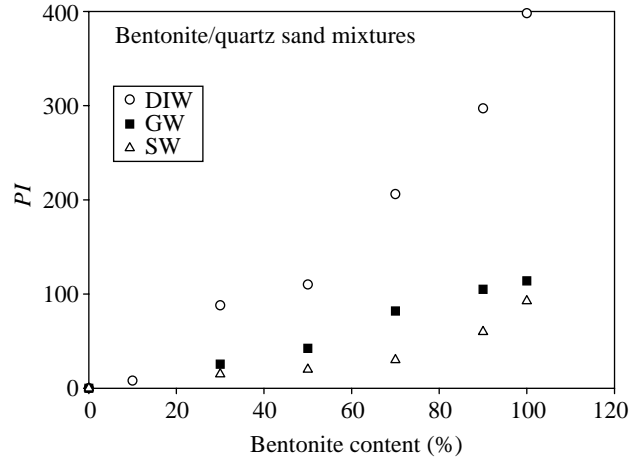


Fig. 4. Plastic index (PI) in relation to bentonite content on bentonite/quartz sand mixtures.

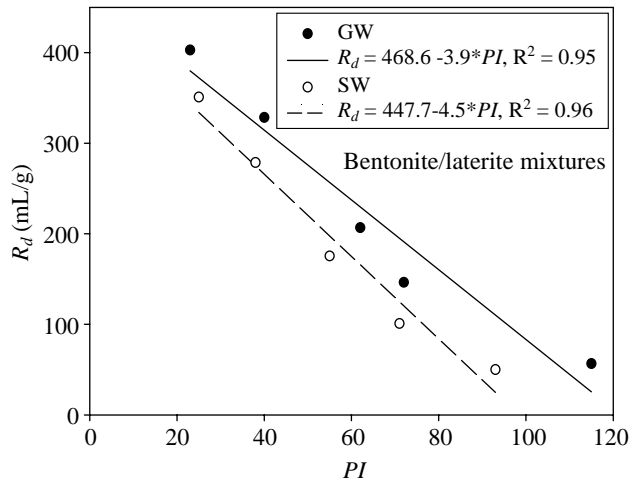


Fig. 5. Relation of R_d of Se and PI on bentonite/laterite mixtures.

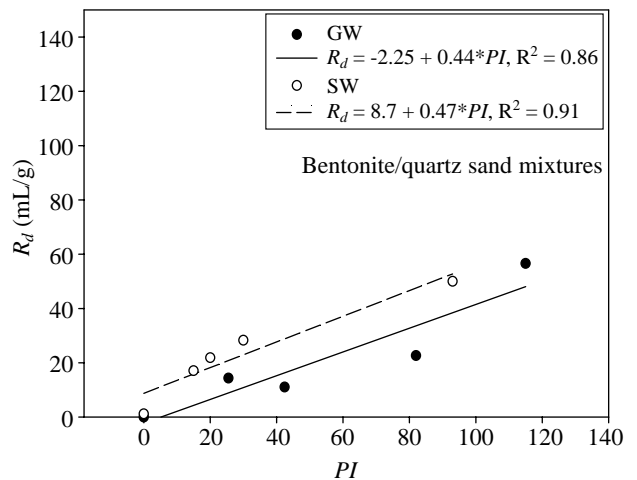


Fig. 6. Relation of R_d of Se and PI on bentonite/quartz sand mixtures.

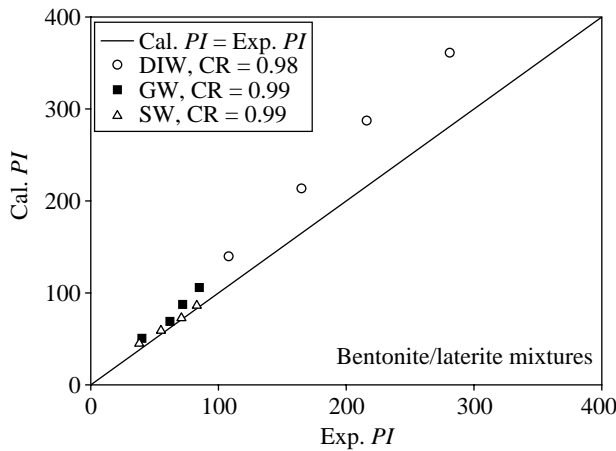


Fig. 7. Additivity of PI on bentonite/laterite mixtures (CR: correlation).

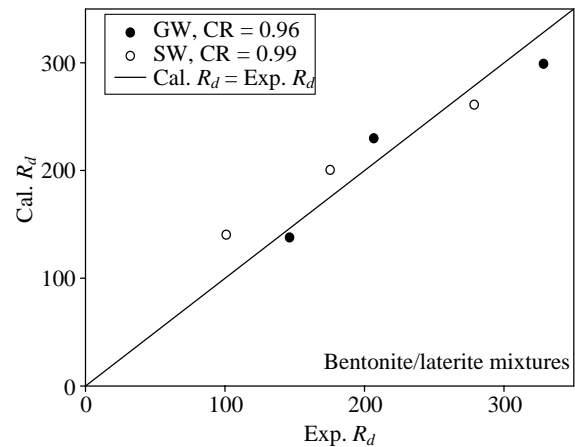


Fig. 9. Additivity of R_d on bentonite/laterite mixtures (CR: correlation).

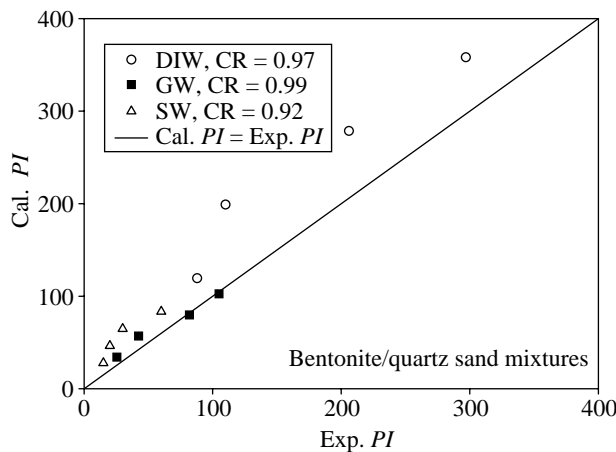


Fig. 8. Additivity of PI on bentonite/quartz sand mixtures (CR: correlation).

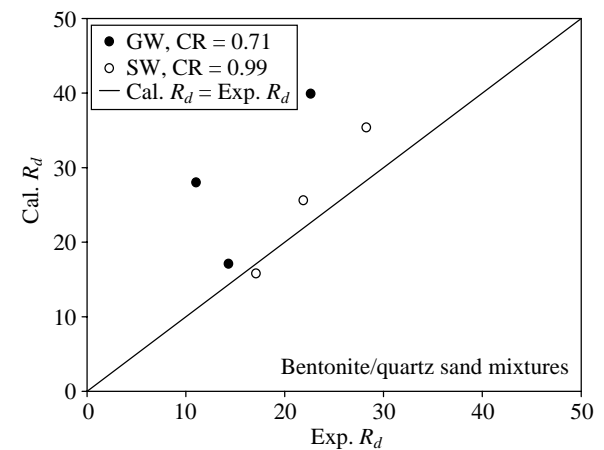


Fig. 10. Additivity of R_d on bentonite/quartz sand mixtures (CR: correlation).

that the PI values for the bentonite/laterite and bentonite/quartz mixtures followed the additivity rule in SW and GW but not in DIW. The results may have followed from the fact that the PI of bentonite in DIW greatly exceeded those of the mixtures, in which neither coagulation nor flocculation occurred.

Another issue was whether the R_d value at various composition ratios could be predicted from the R_d value of the individual materials could. Such prediction is useful in evaluating and choosing buffer-backfill materials. Figures 9 and 10 compare the experimental and calculated R_{dS} (Eq.1) for Se in GW and SW, for the various tested mixtures. The results of this investigation showed that the R_d value of Se exhibited good additivity for the bentonite/laterite mixtures in both SW and GW and for bentonite/quartz sand mixtures in SW, whereas similar results were not obtained for bentonite/quartz sand mixtures in GW. Not only did the calculated and experimental R_d values of Se for bentonite/quartz

sand mixtures in GW not show high additivity, but also, they did not tend to be conservative.

CONCLUSION

Under the experimental conditions herein, the sorption percentage increased with the bentonite content in bentonite/quartz sand mixtures. Additionally, the sorption percentage increased with the laterite content in bentonite/laterite mixtures. The evidence indicated that the sorption of Se by soils followed the order laterite soil > bentonite > quartz, regardless of whether GW or SW was used. Hence, adding laterite to the mixture promoted the sorption of the anionic form of Se. PI was proportional to the bentonite content of the tested buffer material that included bentonite/quartz sand and bentonite/laterite, independently of the solution used (SW, GW, or DIW). The results also indicated $PI_{DIW} > PI_{GW} > PI_{SW}$, because of coagulation and flocculation effects.

A material with a PI of approximately 50 to 70 (in GW and SW) is preferred to balance engineering needs with desired chemical properties. A bentonite content of 50% and the addition of both laterite and quartz sand may be optimal for buffer materials and should be studied further. This work further indicates that the PI values of both mixtures followed the additivity rule in SW and GW, but not in DIW. The R_d value of Se exhibited good additivity on the bentonite/late-rite mixtures in both SW and GW and on bentonite/quartz sand mixtures in SW, but similar results were not obtained on bentonite/quartz sand mixtures in GW. These results provide useful information concerning the choice of buffer material composition, and for evaluating the overall R_d and PI of the mixture, from the R_d and PI of its components.

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