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Original Article

Transport of heavy metals and chemical compatibility of hydraulic conductivity of a compacted sand-bentonite mixture

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Abstract

Clayey soils are usually used as barrier material in landfill liners because of its low hydraulic conductivity and high sorption capacity. Bentonite, which consists mainly of montmorillonite, has a high cation exchange capacity resulting in a high retention capacity of heavy metals. Sand is a permeable material but its hydraulic conductivity decreases significantly when mixed with bentonite. However, using a sand-bentonite mixture as landfill liners is questionable, because the hydraulic conductivity of the sand-bentonite mixture may increase when permeated with heavy metal solutions, which are normally found in landfill leachates. In this paper, transport of heavy metals through a compacted sand-bentonite mixture and its chemical compatibility were studied through the batch adsorption test, the column test, and the hydraulic conductivity test. Experimental results indicate that the sorption capacity of the bentonite, ranked in descending order, was Cr^{3+}, Pb^{2+}, Cd^{2+}, Zn^{2+}, and Ni^{2+}, respectively. The diffusion coefficients of the sand-bentonite mixture were in the order of 10^{-5} cm^2/s and the retardation factors were 130, 115, 111, and 90 for Pb^{2+}, Ni^{2+}, Zn^{2+}, and Cd^{2+}, respectively. The hydraulic conductivity of the sand-bentonite mixture was only compatible with a chromium solution having a concentration not greater than 0.001 M.

Keywords: sand-bentonite mixture, hydraulic conductivity, heavy metals, adsorption, diffusion coefficient, retardation factor

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1. Introduction

Clays and clay minerals play an important role as constituents of geological or technical landfill liners (Shackelford, 1991; Yong *et al.*, 1992; Roehl and Czurda, 1998; Chapuis, 2002). The landfill liners act as a hydraulic as well as geochemical barrier to prevent the breakthrough of hazardous contaminants like heavy metals. Clay minerals have a high adsorption capacity on heavy metal ions because of their large specific surface area and their high number of negatively charged bonding sites (Yong *et al.*, 1992; Helios Rybicka *et al.*, 1995; Roehl and Czurda, 1998; Leroy *et al.*, 2006).

Bentonite refers to any material that is primarily composed of the montmorillonite group of clay minerals. Characteristics of montmorillonite minerals include large cation exchange capacity, large specific surface area, high swelling potential, and low hydraulic conductivity to water (Grim and Guven, 1978; Gleason *et al.*, 1997). Properties of bentonite are greatly affected by interactions between its particles and the surrounding pore fluid. The net electrical charge of bentonite particles is negative, which causes dissolved cations in the pore water to be attracted to the surfaces of the bentonite. The layer of water and absorbed ions that surrounds a bentonite particle is referred to as the diffuse double layer, DDL (Mitchell, 1993; Shackelford, 1994; Yong, 2001).

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Sand is a pervious material and its hydraulic conductivity ranges from 1 to 1x10^{-5} cm/s (Freeze and Cherry, 1979). The performance of sand as a contaminant barrier is enormously improved if an appropriate amount of bentonite is added (Chapuis, 2002). Hydraulic conductivities of the compacted sand mixed with as low as 3% sodium bentonite content are less than 1x10^{-7} cm/s (Gleason *et al.*, 1997; Li and Li, 2001; Chalermyanont and Arrykul, 2005). Sorption behavior and hydraulic conductivity of sand-bentonite mixtures have been studied by many researchers (Cabral, 1992; Roehl and Czurda, 1998; Li and Li, 2001; Chalermyanont and Arrykul, 2005; Kaya and Oren, 2005; Ouhadi *et al.*, 2006). However, transport and chemical compatibility of heavy metal contaminants to compacted sand-bentonite mixtures have not been greatly emphasized.

The objectives of this study are to determine the transport parameters of a compacted sand-5% bentonite mixture permeated with mixed heavy metal solutions and to evaluate the chemical compatibility of the hydraulic conductivity of the compacted-sand bentonite mixture when permeated with a chromium solution with different concentrations. A mixed heavy metal solution was used to simulate real leachate, which consists of many heavy metals (Ou, 1989). The batch adsorption test was conducted to determine the sorption capacities of the bentonite. The column test was used to determine transport parameters of the sand-bentonite mixture namely, hydraulic conductivities, diffusion coefficients, and retardation factors. The falling head hydraulic conductivity test was conducted using different concentrations of an influent solution in order to determine the chemical compatibility of the sand-bentonite mixture.

2. Transport of Chemicals in Soil

Transport of chemicals in soil consists of three main processes namely, advective transport, diffusive transport, and chemical reaction between the chemicals and the soil. A concentration profile of a chemical having an initial effluent concentration C_o in a soil is shown in Figure 1. The concentration profile is governed by 1-D transport equation which can be expressed as

$$\frac{\partial C}{\partial t} = \frac{D}{R}\frac{\partial C}{\partial z^2} - \frac{v_s}{R}\frac{\partial C}{\partial z} \qquad (1)$$

Where C is the concentration of the chemical, v_s the seepage velocity (v_s = Ki/n), K the hydraulic conductivity of soil, i the hydraulic gradient, n the porosity of the soil, D the diffusion coefficient, R the retardation factor, t the time of transport and z the direction of transport.

Solution of the 1-D transport equation was proposed by Ogata and Bang (1961) and rearranged by Shackelford (1990) for landfill liner application. The solution is expressed in term of a relative concentration, which is a ratio of concentration as a function of depth and time to the initial concentration of the solution, as expressed in Equation 2.

$$\frac{C(z,t)}{C_0} = \frac{1}{2}\left\{ \text{erfc}\left[\frac{1-T_R}{2\sqrt{\frac{T_R}{P_L}}}\right] + \exp(P_L)\,\text{erfc}\left[\frac{1+T_R}{2\sqrt{\frac{T_R}{P_L}}}\right]\right\} \qquad (2)$$

where T_R (Time factor) = $v_s t/Rz$, P_L (Peclet number) = $v_s z/D$, R = 1+ $r_d K_p/n$, r_d is the dry density of soil, and K_p is the partitioning coefficient.

3. Materials and Methods

3.1 Properties of Sand, Bentonite, and Sand-Bentonite Mixtures

The bentonite used in this study was powdered sodium bentonite manufactured by Wyo-Ben, Inc. This bentonite is generally used as drilling mud. Properties of the bentonite are tabulated in Table 1. The air-dried bentonite had a percent passing No. 200 sieve of 65%, a liquid limit (LL) of 487% and a plasticity index (PI) of 450%. The bentonite had a low organic content and a high cation exchange capacity of 1.65 (%W/W) and 75.22 meq/100g, respectively. The high PI value of the bentonite indicated that the bentonite was a very highly plastic material and had a high swelling poten-

Table 1. Physicochemical properties of sodium bentonite

Property	Value
Liquid Limit (L.L.) (%)	487.00
Plasticity Index (PI) (%)	450.00
P200 (%)	65.00
Color	Light gray
Maximum Dry Density (g/cc)	1.95*
Optimum Water Content (%)	10.50*
Classification (USCS)	CH
pH	8.58
Organic content (%w/w)	1.65
SO_4^{2-} (mg/kg)	3,807.11
Cl^- (mg/kg)	280.12
Exchangeable Cations	
Na^+ (meq/100g)	22.64
K^+ (meq/100g)	1.53
Ca^{2+} (meq/100g)	24.45
Mg^{2+} (meq/100g)	2.52
CEC (meq/100g)	75.22
Mineralogical Composition	
SiO_2 (%)	63.97
Al_2O_3 (%)	15.66
Fe_2O_3 (%)	7.74
K_2O (%)	0.75
SO_3 (%)	0.75
CaO (%)	2.62

* Sand-5% bentonite mixture

tial.

tial. X-ray diffraction test results showed that the bentonite consisted of montmorillonite, which is a very highly plastic material that is consistent with its high PI and cation exchange capacity (CEC) values. X-ray fluorescence spectrometry test results showed that the main chemical compounds of the bentonite were silicon oxide and aluminum oxide.

The sand used in this study was local Songkhla sand, which is typically used as a construction material. The coefficient of uniformity (C_u) and the coefficient of curvature (C_c) were 11.7 and 1.0, respectively. Percent passing No. 200 sieve was 6.5% and the fine content of the sand was found to be nonplastic. Thus, the sand was classified as SW-SM according to the Unified Soil Classification System (USCS).

Sand-bentonite mixtures with 5% bentonite content were used in this study. Compaction test results showed that for the sand-bentonite mixture used, the maximum dry unit weight and optimum water content were 1.95 g/cc and 10.5%, respectively. Hydraulic conductivity of the sand-bentonite mixture was about 5×10^{-9} - 9×10^{-9} cm/s (Chalermyanont and Arrykul, 2005).

3.2 Batch Adsorption Tests

Batch adsorption test was conducted according to the USEPA (1987) standard method to assess heavy metal sorption of the bentonite. The heavy metals used in this study were Cd^{2+}, Pb^{2+}, Zn^{2+}, Ni^{2+}, and Cr^{3+}, which were prepared by mixing $CdCl_2$, $PbCl_2$, $ZnCl_2$, $NiCl_2.6H_2O$, and $CrCl_3.6H_2O$ with deionized (DI) water, respectively. For each test, 0.5 g of the bentonite was added to 25-ml heavy metal solution and shaken with a speed of 300 rpm for 24 hours. The solution then was centrifuged with a speed of 6,000 rpm for 10 minutes and a supernatant was collected. The concentration of the supernatant was then determined using Atomic Absorption Spectrophotometer (Varian 220A). A series of batch adsorbed tests were conducted following the procedure with different concentrations of heavy metal solutions. The concentration of heavy metals varied from 1 to 4,000 mg/L as recommended by Li and Li (2001). In this study, for each batch adsorption test, 12 various concentrations were used for each heavy metal solution, thus, altogether, 60 tests were performed.

3.3 Column Test

A column test was conducted to determine the transport parameters of the compacted sand-5% bentonite mixture. A sand-bentonite sample was prepared by mixing sand, bentonite, and tab water and cured in plastic bags for 24 hours. The water content of 2% wet of the optimum water content (i.e., 12.5%) was used. Subsequently, the samples were compacted in rigid wall columns in accordance with an adaptation of the Standard Proctor Compaction Test (ASTM D698) using a mold of 10 cm in diameter and 6 cm in height. An ELE automatic compactor was used for compacting the samples. The column test set up is shown schematically in Figure 2. The sample was permeated first with DI water. The DI water was permeated first in order to achieve the first exposure effect on the hydraulic conductivity as recommended by Fernadaez and Quigley (1985) and Quaranta *et al.* (1997). Induction of flow was conducted by increasing the pressure head in the influent chamber for 10 kPa in every 24 hours until reaching the pressure of 80 kPa, which is equivalent to a hydraulic gradient of 88. Effluent was collected periodically and used for calculating the hydraulic conductivity of the soil sample following the constant head test method (ASTM D5084). This process was conducted until the hydraulic conductivity of the soil sample was steady and subsequently, the DI water was replaced by the mixed heavy metal solution.

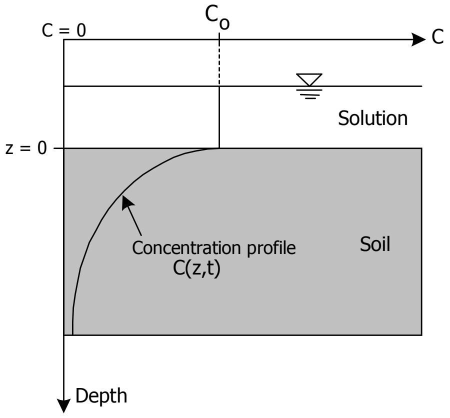

Figure 1. Concentration profile of a chemical in a soil

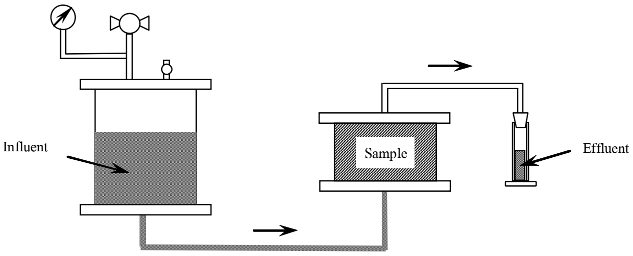

Figure 2. A schematic set up of a column test

The mixed heavy metal solution was used to simulate the leachates. It was prepared by mixing $CdCl_2$, $PbCl_2$, $ZnCl_2$, and $NiCl_26H_2O$ powders with DI water. A concentration of 0.001 M was used for each heavy metal, which corresponded to 112.42, 207.26, 65.39, and 58.87 mg/L for Cd^{2+}, Pb^{2+}, Zn^{2+}, and Ni^{2+}, respectively. The mixed heavy metal solution of Cd^{2+}, Pb^{2+}, Zn^{2+}, and Ni^{2+} was chosen because all the heavy metals used had two positive charges. While transported in the sample, the masses of heavy metals in the solution were adsorbed by the sand-bentonite mixture sample. When the adsorption capacity at the top of the sample was reached, the deeper parts of the sample would adsorb the oncoming heavy metals, meanwhile, at the bottom part of the sample, no adsorption occurred. This process continued until all parts of the sample reached their adsorption capacity.

In this study, the heavy metals were allowed to be transported through the sample for a certain time, so that the adsorption capacity at the top of the sample was reached and the concentration profile of the adsorbed chemicals could be obtained. After that, the sample was extruded from the column and divided horizontally into seven layers. Each layer was oven-dried and then acid digestion technique was employed for determining the masses of the adsorbed heavy metals. The concentration of the adsorbed heavy metal (C_s), which was defined by the ratio of adsorbed heavy metal mass to the mass of the sample, was determined. An adsorbed concentration of the top layer (C_{sm}) was defined by the ratio of the adsorbed heavy metal mass with the adsorption capacity reached to the mass of the sample of the top layer. A relative adsorbed concentration (C_s/C_{sm}) was used as the right side of Equation 2, while the transport parameters can be determined.

An acid digestion technique for determining the concentration of the adsorbed heavy metals, as recommended by Tanchuling *et al.* (2003), was conducted by first adding 2 mL of HNO_3 and 4 mL of HCl to 0.5 g of the sample. A hot plate was used to heat the sample up to a temperature of 100° for 2 hours. Filter paper number 42 was used to separate the solution and the solid sample. The solution obtained then was diluted with 1% HNO_3 and the volume was adjusted to 50 mL. The concentration of the solution was determined using a flame atomic adsorption spectrophotometer.

3.4 Chemical Compatibility Tests

Chemical compatibility of the sand-bentonite mixture was evaluated by measuring its hydraulic conductivities with respect to testing time, when permeated with various concentrations of chromium solution. The sand-bentonite mixture was considered to be compatible to the chromium solution if its hydraulic conductivity did not significantly increase with time. A hydraulic conductivity test was performed on six sand-bentonite mixture samples using falling head test method (ASTM D2423). For each sample, the sand-bentonite mixture was compacted in a standard 4-inch mold with a water content of 2% wet of the optimum water content. The mold with the compacted sand-bentonite sample inside was then used as a rigid wall permeameter for the hydraulic conductivity determination. DI water was first permeated through the sample and the hydraulic conductivity was measured. When the hydraulic conductivity of the sample was constant, the DI water was replaced by the chromium solution as an influent. Five concentrations of the chromium solution were used, 0.0001, 0.001, 0.01, 0.1, and 1 M. Chromium solutions (+3 ions) were used because of the fact that solutions having higher positive ions would more effectively decrease the diffuse double layer surrounding the clay particles, resulting in a higher reduction potential of the hydraulic conductivity (Mitchell, 1976; Mathew and Rao, 1997; Jo *et al.*, 2001). The hydraulic gradient used in the test was set to be less than 20 for all tests. Hydraulic conductivity of the samples was measured every few days with a total testing time of about 700 days.

4. Results and Discussions

4.1 Adsorption of Heavy Metal on Compacted Sand-Bentonite Mixture

Batch adsorption test results (Figure 3) are expressed as isotherms showing the relationships between the ratio (q) of mass adsorbed to the mass of bentonite and the equilibrium concentration (C_f). Heavy metal adsorption on the bentonite ranked in descending order was Cr^{3+}, Pb^{2+}, Cd^{2+}, Zn^{2+}, and Ni^{2+}, respectively. Lungmuir isotherm was found to provide a best fit to the batch absorption test results. Lungmuir parameters are shown in Table 2.

At a low concentration, the isotherms can be assumed to be linear (Fetter 1993). The slope of a linear isotherm is called partitioning coefficient (K_p). Batch adsorption test results (Figure 3) show that for equilibrium concentrations that are lower than 20 mg/L, the isotherms are practically linear. The partitioning coefficients obtained by fitting a linear isotherm to the batch adsorption test results only in the linear part, were tabulated in Table 2. Adsorption capacities determined from q_{max} were also tabulated in Table 2.

4.2 Transport Parameters

1) Hydraulic Conductivity

Relationships between hydraulic conductivity of two sand-bentonite mixture samples and time or pore volume of effluent (PVE) obtained from the column test using the

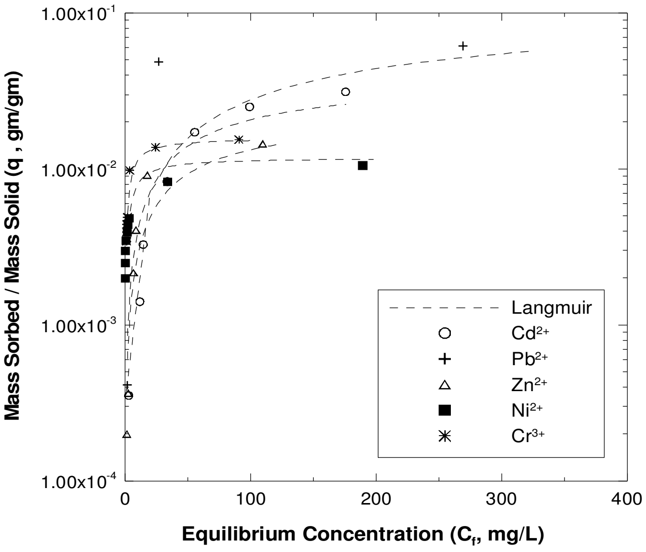

Figure 3. Batch adsorption test results

Table 2. Isotherm parameters from batch adsorption tests

Heavy metal	Langmuir Isotherm*			K_p**(L/kg)	Adsorption Capacity*** (meq/100 g)
	X_m (mg/g)	b (L/mg)	R^2		
Cd	39.370	0.011	0.933	82.848	55.473
Pb	106.383	0.004	0.955	64.596	81.591
Zn	23.148	0.013	0.804	69.320	53.308
Cr	15.848	0.243	0.999	170.039	88.841
Ni	11.834	0.201	0.995	55.592	35.772

* Langmuir isotherm $\frac{C_f}{q} = \left(\frac{1}{bX_m}\right) + \left(\frac{C_f}{X_m}\right)$, b = sorption constant, X_m = maximum adsorbed mass per unit soil mass

** K_p corresponds to the slope of the linear isotherm

*** Adsorption capacity was determined from maximum q

Table 3. Adsorbed heavy metal concentrations on the sand-bentonite mixture sample at 64 days of permeation

Layers	Depth (cm)	Adsorbed Concentration (mg/g)			
		Cd	Ni	Pb	Zn
1	0.25	1.01	0.74	1.46	0.64
2	0.7	1.03	0.71	1.44	0.65
3	1.7	0.82	0.47	0.86	0.42
4	2.7	0.52	0.25	0.36	0.13
5	3.7	0.34	0.15	0.24	0.02
6	4.7	0.00	0.00	0.00	0.00
7	5.7	0.00	0.00	0.00	0.00

constant head test method are shown in Figure 4. One sample was permeated with DI water only whereas another sample was permeated first with DI water and later with the mixed heavy metal solution. For the first sample (i.e., blank sample), the hydraulic conductivity was about 3×10^{-9} cm/s for a testing time of 190 days.

For the second sample, while permeated with the DI water for about 3 weeks, the hydraulic conductivity of the sample was about 4.0×10^{-9} cm/s. After the DI water was replaced by a mixed heavy metal solution, the hydraulic conductivity of the sample was approximately steady at 4.0×10^{-9} cm/s for 40 days. However, after 40 days or about two PVEs of the mixed solution permeation, the hydraulic conductivity of the sample started to increase to 9.0×10^{-9} cm/s and was stable at this value for the rest of the test.

2) Diffusion Coefficient and Retardation Factor

The sand-bentonite mixture sample was taken for a concentration determination after 64 days of mixed heavy metal permeation. Adsorbed heavy metal concentrations and corresponding depths are tabulated in Table 3. It is clearly seen that for the top two layers, the adsorption capacity was reached as the concentration of both layers are fairly equal. On the other hand, for deeper layers, the concentration decreased with increasing depth indicating that the adsorption capacity was not reached. The concentrations at the bottom layer were equal to zero for all heavy metals.

Relationships between the adsorbed heavy metal concentrations (C_s), normalized by the adsorbed heavy metal concentration of the top layer (C_{sm}), so called "relative adsorbed concentration" (C_s/C_{sm}) and the depths are plotted using data from Table 3 and shown in Figure 5. The corresponding transport parameters were determined by fitting the relative adsorbed concentration profiles to Equation 2. Porosity of the sample was 0.26. The hydraulic conductivity of the sample used in Equation 2 was the average hydraulic conductivity for 64 days of the mixed solution permeation of 6.0×10^{-9} cm/s. An Optimization tool (i.e., Microsoft Excel's Solver) was used in trial and error process for determining diffusion coefficients and retardation factors of the sand-bentonite mixture. The best estimates of the diffusion coefficients and the retardation factors were achieved when a mean square error (MSE), which is showing the difference between a relative adsorbed concentration profile obtained from column test results and the corresponding values calculated from Equation 2, was minimized. The calculated diffusion coefficients and the retardation factors were tabulated in Table 4. The diffusion coefficients of the sand-bentonite mixture for different heavy metals were fairly similar at about 10^{-5} cm^2/s. The similarity of diffusion coefficients can be verified by the similar shape of the relative adsorbed concentration profiles.

The retardation factors of the sand-bentonite mixture were 130, 115, 111, and 90 for Pb^{2+}, Ni^{2+}, Zn^{2+}, and Cd^{2+}, respectively (Table 4). Lower retardation factor (i.e., Cd^{2+}) indicated that Cd^{2+} was transported relatively fast as seen at the position of the concentration profile. The concentration profile of Cd^{2+} (Figure 5) is at a deeper position compared to the other profiles.

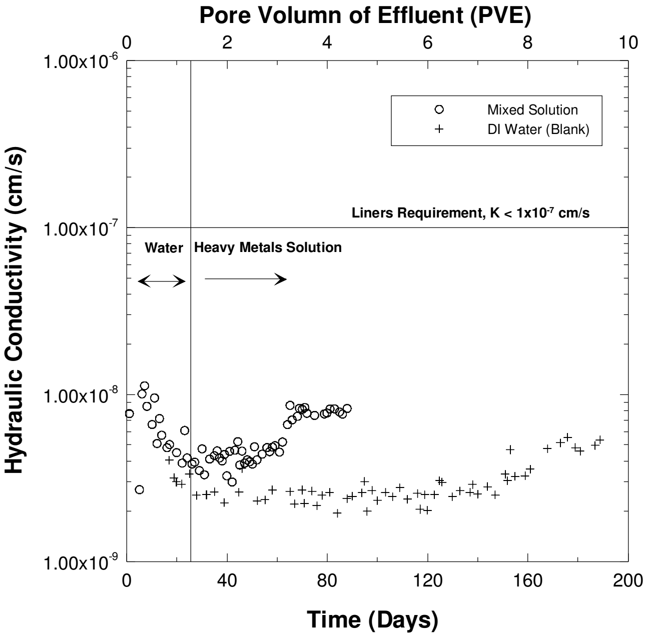

Figure 4. Hydraulic conductivity of compacted sand-bentonite mixture permeated with mixed heavy metal solution and DI water

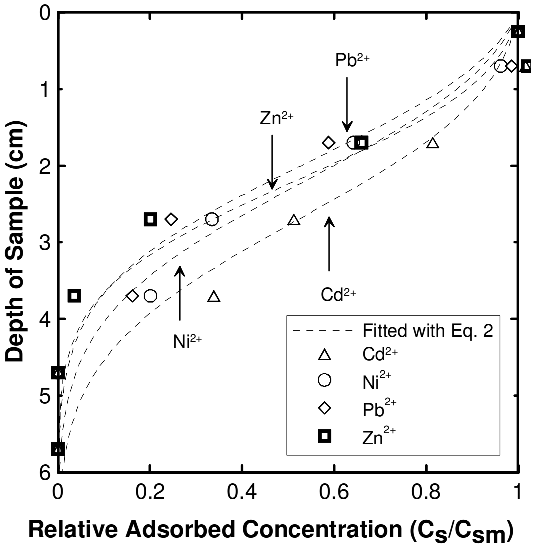

Figure 5. Relative adsorbed concentration profiles of heavy metals in the sand-bentonite mixture sample

4.3 Chemical Compatibility

The chemical compatibility of the hydraulic conductivity of the sand-bentonite mixture to chromium solutions was evaluated by permeating five sand-bentonite mixture samples with various concentrations of the chromium solution. Concentrations of chromium solutions, ranged from 0.0001, 0.001, 0.01, 0.1, and 1.0 M. The longest testing time was about 700 days. A very long testing time was used to ensure that there was enough pore volume of chromium solution permeating through the samples.

Table 4. Diffusion coefficients and retardation factors of the sand-bentonite mixture sample

Heavy metal	D (cm^2/s)	R	MSE
Cd	1.40 x 10^{-5}	90	0.0026762
Ni	1.77 x 10^{-5}	115	0.0022115
Pb	1.72 x 10^{-5}	130	0.0031274
Zn	1.12 x 10^{-5}	111	0.0015020

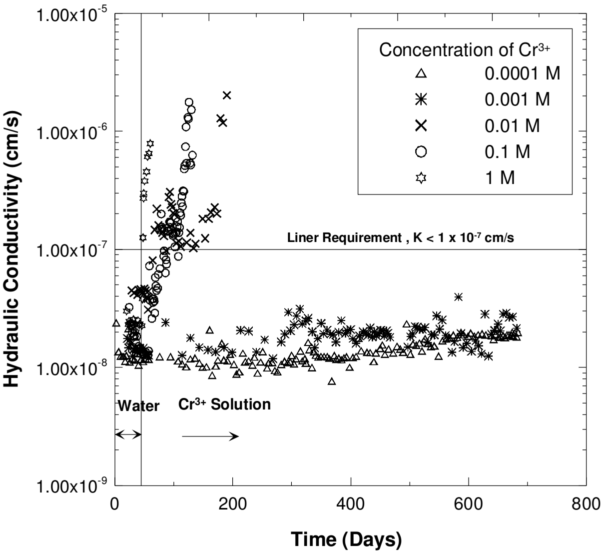

Figure 6. Hydraulic conductivities of the sand-bentonite mixture permeated with various concentrations of the chromium solution.

Relationships between the hydraulic conductivities of the samples and the testing time are shown in Figure 6. The samples were first permeated with DI water and the corresponding hydraulic conductivities were about 9x10^{-9} cm/s. After 60 days of testing time, the DI water was replaced by the chromium solutions and the hydraulic conductivities were found to increase. For the sample that was permeated with 1 M-chromium solution, the hydraulic conductivity of the samples abruptly increased such that the samples cannot hold the solution. The hydraulic conductivities of the samples continuously increased and were greater than 1x10^{-7} cm/s at 13 and 20 days after the chromium solutions of 0.1 and 0.01 M, respectively, were introduced. When permeated with chromium solutions of 0.0001 and 0.001 M, the hydraulic conductivities of the samples insignificantly increased to about 2x10^{-8} cm/s for about 700 days. Thus, the hydraulic conductivity of a sand-bentonite mixture was compatible only with chromium solutions having concentration not greater than 0.001 M.

5. Conclusions

Adsorption, transport parameters, and chemical compatibility of the sand-bentonite mixture were assessed by conducting a series of tests with heavy metal solutions, and the following conclusions can be drawn.

1. Batch adsorption results showing that the heavy metal adsorption capacity of the bentonite, ranked in descending order, was Cr^{3+}, Pb^{2+}, Cd^{2+}, Zn^{2+}, and Ni^{2+}, respectively, and Lungmuir Isotherm was found to provide best fits.

2. For concentration less than 20 mg/L, linear isotherm practically gave best fits to the batch adsorption test results and partitioning coefficients and sorption capacities can be obtained.

3. When permeated with DI water, the hydraulic conductivity of sand-bentonite mixture was 4×10^{-9} cm/s. The hydraulic conductivity increased when permeated with mixed heavy metal solution. The average hydraulic conductivity of the sand-bentonite mixture of 9×10^{-9} cm/s was obtained when it was permeated with mixed heavy metal solution for two PVEs. However, its hydraulic conductivity was still less than 1×10^{-7} cm/s, which was a common regulatory for landfill liners.

4. At 64 days of permeation, adsorbed heavy metal concentrations decreased with depth. The relative adsorbed concentration profiles from column test were used to determine the diffusion coefficients and the retardation factors. The diffusion coefficients were in the order of 10^{-5} cm^2/s, whereas the retardation factors were 130, 115, 111, and 90 for Pb^{2+}, Ni^{2+}, Zn^{2+}, and Cd^{2+}, respectively.

5. The chemical compatibility of the hydraulic conductivity of the sand-bentonite mixture samples was assessed by permeated the samples with various concentrations of the chromium solution. It was found that only chromium solutions having concentrations of 0.001 to 0.0001 M were compatible with the sand-bentonite mixture.

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