



Lead adsorption and transport in loess-amended soil-bentonite cut-off wall



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ARTICLE INFO

Article history:

Received 17 June 2016

Received in revised form 8 September 2016

Accepted 6 November 2016

Available online 8 November 2016

Keywords:

Cut-off wall

Soil-bentonite

Loess

Lead

Adsorption

Breakthrough time

ABSTRACT

Soil-bentonite slurry-trench cutoff walls using backfill consisting on-site sandy or silty soil and bentonite are extensively used as engineered barriers for the containment of groundwater and soil pollution. Chinese loess has been shown to have large adsorption capacity regarding heavy metals. There is a great potential to use Chinese loess as amendments to improve the adsorption capacity of the wall. Batch and column tests were carried out to investigate the adsorption and transport of Pb(II) in loess modified soil-bentonite (LSB). Batch tests were conducted to study the effects of contact time, initial Pb(II) concentration, loess proportion in LSB, and pH on Pb(II) adsorption. The rate constant of pseudo-second-order kinetic adsorption model of LSB is two times greater than that of soil-bentonite (SB), which indicates that the adsorption rate of LSB is much faster than that of SB. The mean free energy of adsorption evaluated by D-R model is 15.56 kJ/mol and 18.89 kJ/mol for SB and LSB, respectively. This indicates the adsorption mechanisms of SB and LSB are mainly ion exchange and chemical reaction. The adsorption capacity of LSB increases linearly with the increase of the loess amount in LSB. The maximum adsorption amount of LSB containing 20% loess can be 2 times greater than that of SB. Results also indicate that Pb(II) is first adsorbed onto loess until the adsorption amount Pb(II) onto loess reaches its adsorption maximum. An equation considering this effect was proposed for prediction of the adsorption capacity of LSB. A set of column tests were carried out to evaluate the tortuosity, mechanical dispersion and retardation factor regarding lead transport in SB and LSB vertical engineered barrier. The retardation factor of LSB and SB are determined to be 38 and 15, respectively, by the column tests. The K_d values obtained from batch tests are significantly larger than those obtained from column tests due to the difference of soil-water ratio used for these two types of tests. Using the obtained parameter values, the breakthrough time of Pb(II) through LSB wall is evaluated to be about 2 times greater than that of the SB wall when the hydraulic head difference is <3 m. This may be due to the increase of adsorption capacity of the soil-bentonite wall amended with the loess.

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1. Introduction

Vertical barriers are used to reduce the movement of contaminated groundwater or uncontaminated groundwater through a contaminated area (Mulligan et al., 2001; Sharma and Reddy, 2004). Soil-bentonite (SB) wall is a typical type of vertical barrier, which is constructed by the slurry trench excavation method, and is used extensively as vertical engineered barrier to control the migration of contaminants in groundwater due to its low permeability and cost-effectiveness (Evans, 1993; Devlin and Parker, 1996; Britton et al., 2005; Yeo et al., 2005a, 2005b; Malusis and McKeehan, 2013; Du et al., 2015a). There are four main processes which control contaminants transport through cut-off walls, i.e., advection, dispersion, diffusion and adsorption (Shackelford, 1995; Malusis and Shackelford, 2004). It is commonly assumed that the migration of contaminants through cut-off walls can be effectively curtailed by

restricting the barrier hydraulic conductivity to $<10^{-7}$ cm/s (Khandelwal and Rabideau, 2000). However, contaminants can readily transport through conventional SB wall by diffusion (Mott and Weber, 1991; Khandelwal et al., 1998; Rowe et al., 2004). In addition, the hydraulic conductivity of SB cutoff wall has been shown to increase after long-term contact with contaminants (Kashir and Yanful, 2001; Malusis and McKeehan, 2013; Scalia et al., 2013). Thus, methods to decrease the transport of contaminant through cut-off walls are of high interests.

The addition of adsorptive amendments is often identified as a strategy for enhancing barrier performance (Krol and Rowe, 2004; Malusis et al., 2010). Currently, the proposed adsorptive amendments include fly ash (Mott and Weber, 1992), activated carbon (Malusis et al., 2009, 2010), organically modified clays (Sreedharan and Puvvadi, 2013), natural humus (Khandelwal and Rabideau, 2000), zero-valent iron (Rabideau et al., 1999; Castelbaum et al., 2011) and zeolite (Hong et al., 2011; Du et al., 2015b).

For heavy metal contaminants such as Pb(II), Cu(II), Zn(II) and Cd(II), loess exhibits superior adsorption capability relative to the

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amendments listed above (Tang et al., 2008a, 2008b; Li et al., 2009; Tang et al., 2009; Yang et al., 2012). The adsorption is carried out by means of chemical precipitation with calcite in loess (Li et al., 2009). In addition, loess is a natural soil widely distributed in Middle Asia, Russia, Middle East, North America and China (Tang et al., 2009). It is a cheap, easily available and environmental friendly amendment. Consequently, there is a great potential to use loess as an amendment to increase the heavy metal adsorption capability of SB vertical wall. Although the adsorption mechanisms of heavy metals onto pure loess (Li et al., 2009; Tang et al., 2009), bentonite (Eren, 2009; Ouhadi et al., 2006; Wang et al., 2009; Hamidpour et al., 2010; Chen et al., 2012) and SB backfill matrix (Daniels et al., 2004) have been studied extensively, the mechanism of adsorption of heavy metals onto SB backfill matrix containing loess is still unknown. The feasibility of loess amended soil-bentonite (LSB) to be used as a potential engineered vertical barrier for groundwater pollution control also needs to be evaluated in an aquifer/LSB wall system.

This study aims to demonstrate the adsorption characteristic of Chinese loess amended soil-bentonite as a potential barrier material for heavy metal contaminated soil remediation. Pb(II) was used as a typical type of heavy metal contaminant. Both equilibrium and non-equilibrium adsorption batch tests were carried out. The influence of various experimental conditions including initial solution concentration, contact time, and pH on the adsorption process was discussed. Laboratory consolidation-permeability tests and column tests were also conducted to determine the transport mechanisms of Pb(II) in the LSB. Numerical analysis was carried out to evaluate its feasibility as a potential barrier material based on the obtained transport parameters. In addition, scanning electron microscope (SEM) and X-ray powder diffraction (XRD) test were conducted to analyze the Pb(II) adsorption mechanism of LSB. The findings of this study would be useful for the development of a new vertical engineering barrier containing loess. The results of this study would also provide guidelines and parameters for assessing and designing of LSB vertical barrier.

2. Materials and methods

2.1. Solid and liquid materials

LSB was prepared with three types of soils i.e., silt (S), loess (L) and Na-bentonite (B). The silt was sampled from the shore side of Qiantang River, southeast of China, which is typically a type of silty soil. The loess was sampled from Xi'an, west of China. The bentonite was a highly qualified natural Na-based bentonite and was commercially available by Wyo-Ben, Inc., Billings, Montana. Silt and loess were dried at 105 °C for 24 h, grounded into powder, sieved through a 2 mm mesh, and then put into plastic bags for storage. Na-bentonite was also dried at 105 °C for 24 h and afterward stored in a two layered plastic-oil paper bag. The XRD pattern of silt, loess and Na-bentonite were recorded on a D/MAX-RA apparatus (Rigaku Corporation, Japan) with CuK α radiation ($\lambda = 0.15406$ nm) in Zhejiang University. Grain size distribution of silt and loess were determined. Particles larger than 0.075 mm were tested by using a series of sieves and the remaining soil was tested using the sedimentary methods. The cation exchange capacity (CEC) of silt, loess and bentonite were determined using the ammonium exchange method. The specific surface area was determined by N₂ adsorption method and the results were analyzed by Brunauer-Emmett-Teller (BET) adsorption theory. The natural pH (pH_{na}) of silt, loess and bentonite were determined by mixing 50 mg of soils with 25 mL of distilled water (DI water). The pH of Pb(II) solution was measured by a glass electrode potentiometer (multi 3240 SET B, made in Germany).

Table 1 shows the chemical and physical properties of the soil samples comprising the LSB (i.e. silt, loess and Na-bentonite) used in this study. Silt and loess contain the same types of mineral components, i.e. calcite, quartz, clinocllore, muscovite, albite, and orthoclase, with a different amount of each component. However, bentonite contains different mineral components (except quartz) from silt and loess. The

Table 1
Physical and chemical properties of silt, loess and bentonite.

Property	Silt	Loess	Bentonite
Mineral components (% by dry weight)			
Calcite	2.6	14.8	0
Quartz	54.6	38	12.3
Clinocllore	3.6	6.6	0
Muscovite	14.2	22.8	0
Albite	17.3	12.9	0
Orthoclase	7.7	4.8	0
Andesine	0	0	12.7
Illite	0	0	3.6
Na-mont	0	0	71.4
Particle contents (% by dry weight)			
Sand (% 2–0.075 mm)	15	6.7	0
Silt (% 0.075–0.002 mm)	80	81.1	13.2
Clay (% < 0.002 mm)	5	11.2	86.8
CEC (meq/100 g)	1.65	9.94	79.93
Specific surface area (m ² /g)	4.65	23.12	80.23
pH _{na}	8.00	8.05	8.53

main mineral component of silt is quartz (54.6%), the amount of which is 1.5 and 4.5 times larger than that of loess and bentonite, respectively. Loess contains a relatively large amount of calcite (14.8%), while bentonite contains a large amount of Na-montmorillonite (71.4%) and a small amount of illite (3.6%), neither of which was found in silt and loess. The technical specification applied for particle size distribution is the China Standard for Engineering Classification of Soil (MCC, 2007). The silt and loess mainly consists of silt particle, which is 80% and 81.1% by dry weight, respectively. The silt contains more sand particles and fewer clay particles than loess. Most of the soil particles of the bentonite are clay particles, which makes up to 86.8% by dry weight. The CEC of loess and bentonite are 9.94 meq/100 g and 79.93 meq/100 g, respectively, which are 6 and 48 times greater than that of the silt (4.65 meq/100 g). The specific surface area of loess and bentonite is 23.12 and 80.23 m²/g, which is 6 and 19 times larger than that of silt, respectively. The natural pH of the silt, loess and the bentonite are 8.00, 8.05 and 8.53, respectively.

Liquid materials in this study include PbCl₂, NaOH, HCl and NaCl solutions. Pb(II) was used as the heavy metal of interest. Pb(II) solution (1000 mg/L) was prepared by dissolving lead chloride into DI water. Pb(II) solution of other concentrations which was used in this study was diluted from the Pb(II) solution (1000 mg/L). NaOH and HCl (1 M) were used as the pH adjustment to study the adsorption characteristic of Pb(II) onto LSB at different pH values. Cl⁻ (500 mg/L) was used as a non-reactive ion and was prepared by dissolving NaCl into DI water. All chemicals used were of Analytical Reagent (AR) grade.

2.2. Batch tests

In all batch tests, 50 mg soil samples were mixed with 25 mL of Pb(II) solution at a shaking rate 180 rpm at room temperature. The adsorption materials in the batch tests are loess, bentonite, silt, SB, and LSB. Normally SB contains 4%–6% bentonite (Yeo et al., 2005b; Malusis et al., 2009; Hong et al., 2011; Malusis and McKeehan, 2013). In this study, 5% of bentonite was used in both LSB and SB. In LSB, the amount of loess is 20%, determined from preliminary experiments that the maximum adsorption amount of LSB with 20% loess doubles that of SB. In kinetic adsorption batch tests, the initial concentrations of Pb(II) were 100, 200 and 300 mg/L, and shaking time was 10 min, 20 min, 30 min, 1 h, 2 h, 4 h, 8 h, 12 h, 24 h and 48 h, respectively. In equilibrium adsorption tests, the initial concentrations of Pb(II) were 20, 50, 100, 200, 400, 500 and 600 mg/L and the shaking time was 24 h. The 24 h period was determined from preliminary tests to be sufficient for achieving equilibrium conditions. The initial pH values of the solution in kinetic and equilibrium adsorption batch test were 8.0 ± 0.5 and no adjustment was conducted. However, in the tests regarding the investigation of the effect of pH on

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