

## Chemical Compatibility of Sand-Bentonite Backfills Amended with Biopolymer in Vertical Barriers

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### ABSTRACT

This study presents the chemical compatibility and microscopic properties of unamended sand-bentonite (SB) backfill and xanthan gum (XG)-amended SB (XG-SB) backfill, for use in vertical barriers to control lateral movement of lead contaminated groundwater. Liquid limit and hydraulic conductivity tests were carried out on the unamended and XG-amended backfills to assess their chemical compatibility when exposed to lead nitrate ( $\text{Pb}(\text{NO}_3)_2$ ) solutions. The findings of this study present that XG-amendment greatly enhanced the liquid limit of unamended backfill. The hydraulic conductivity ( $k$ ) of unamended and XG-amended backfills increased as the concentrations of lead solutions increased. The  $k$  of XG-amended backfill was found to be less than the acceptable threshold (i.e.,  $10^{-9}$  m/s), whereas that of unamended backfill was higher than  $10^{-9}$  m/s when exposure to lead solutions. Scanning electron microscope (SEM) images presented that polymer hydrogels (i.e., XG hydrogels) blocked the intergranular pores of backfill and the sand particles were covered by hydrated bentonite and XG hydrogel, which results in narrower and more tortuous flow paths that were responsible for low  $k$  of XG-amended backfills as compared to unamended backfills. An extensive comparison of  $k$  for unamended and polymer-amended SB backfills on the basis of this study and previous studies showed that type and content of bentonite, polymer amendment, and cation concentration of testing liquid greatly affected the hydraulic performance of the SB backfills.

*Keywords: Sand-bentonite backfill; Biopolymer; Hydraulic conductivity; Scanning electron microscope*

### 1 INTRODUCTION

Sand-bentonite (SB) backfills containing sodium (Na)-bentonite are widely applied as in-situ vertical barriers to minimize the movement of contaminated groundwater at contaminated sites (Sharma and Reddy, 2004). High-quality Na-bentonite possesses superior swelling property and hydraulic performance, which is responsible for low hydraulic conductivity of SB backfills (Evans 1993; Ruan and Fu, 2022; Fu et al., 2023a). However, the sources of excellent Na-bentonite are relatively rare in a number of Asian countries including China (Fu et al., 2021a, 2021b; Yang et al., 2018a). Instead, Na activated calcium (Ca)-bentonite is commonly used in the SB backfills in these countries. However, long-term exposure to aggressive solutions with relatively high concentrations yields a significant loss in the hydraulic performance of Na activated Ca-bentonite (Ni et al., 2022). To address this concern, use of bentonite enhanced by polymer amendment/modification appears as a feasible method to improve its chemical compatibility in aggressive solutions (Fu et al., 2022, 2023b; Yang et al., 2018b; Bohnhoff and Shackelford, 2014).

Xanthan gum (XG) is one of type of biopolymer, which possesses numerous hydrophilic functional groups. The free water molecules will be bound by these groups via hydrogen bonding, which become immovable (Latifi et al., 2017). Previous research found that the XG could significantly reduce the hydraulic conductivity of silty sand (Bouazza et al., 2009). Moreover, XG is not easy to decompose in acid and alkali solutions and is relatively stable (Rosalam and England, 2006). Meanwhile, XG is a commercial water-soluble fluid loss additive in the field of oil drilling. Based on all these considerations,

XG may have great potential as a bentonite amendment, to enhance chemical compatibility of SB backfills.

This study has evaluated the chemical compatibility of SB and XG-amended SB (XG-SB) backfills when exposed to lead solutions with the concentration ranging from 20 to 500 mmol/L. Liquid limits of XG-amended and unamended bentonites were assessed using distilled water and  $\text{Pb}(\text{NO}_3)_2$  solutions. Scanning electron microscopy (SEM) analyses were applied to evaluate the effect of XG on the microstructure of SB backfill. Furthermore, an extensive comparison of hydraulic conductivity for unamended and polymer-amended backfills on the basis of this study and previous studies was conducted.

## 2 MATERIALS AND METHODS

### 2.1 Unamended and amended backfills

Two backfill specimens were prepared in this study. The one consisted of commercial bentonite (i.e., Na activated Ca-bentonite) and sand, and the other one consisted of commercial bentonite, XG and sand. The commercial bentonite was purchased from Mu-Feng Co. Ltd. (Zhenjiang, China). The XG was purchased from Sinopharm Chemical Reagent Co., Ltd. (Shanghai, China). The sand and bentonite were passed through No. 18 (1 mm) and No. 200 (0.075 mm) sieves, respectively. The properties of XG are shown in Table 1. Moreover, the specific properties of sand and bentonite can be found in Fu et al. (2021a).

The specimens were prepared as follows: First, dry mixtures were obtained by mixing prearranged amounts of air-dried constituents directly. The dry mass ratio of bentonite and sand in the mixture were selected as 1:9. The contents of XG, bentonite and sand in the XG-SB mixture were selected as 0.6%, 10% and 89.4%, respectively. Then, a slurry containing 8% (wt.) bentonite was obtained by mixing bentonite and tap water together, which was used to adjust the water contents of dry mixtures until the slump height of mixtures was  $125 \text{ mm} \pm 5 \text{ mm}$ . Meanwhile, additional sand and XG were added accompanied by the bentonite slurry addition to keep the contents of constituents unchanged in the backfill specimens. The water contents were 36.9% and 52.5.% for the resulting SB and XG-SB backfills, respectively.

**Table 1.** Properties of XG polymer

Property	Details
Charge	Anionic
Particle size	< 0.15 mm
Average molecular weight	2510000
Viscosity	1600 mPa·s
Specific gravity	1.24
Molecular formula	$(\text{C}_{35}\text{H}_{49}\text{O}_{29})_n$

### 2.2 Testing liquids

In this study, distilled water, tap water, and  $\text{Pb}(\text{NO}_3)_2$  solutions were used as testing liquids. The target  $\text{Pb}(\text{NO}_3)_2$  concentrations were selected as 20, 50, 200, and 500 mmol/L, which were measured using an atomic absorption spectrophotometer (ICE3000, Thermo Fisher Scientific Company, Massachusetts, US). The densities, pH, and electrical conductivities (*EC*) of testing liquids were measured by a hydrometer 151H and a pH/*EC* meter (Thermo Fisher Scientific Company, Massachusetts, US), respectively. All densities, pH, and *EC*, concentrations of testing liquids were measured in triplicates. The properties of testing liquids were summarized in Table 2.

**Table 2.** Properties of testing liquids

Liquid type	Target $\text{Pb}(\text{NO}_3)_2$ concentration (mmol/L)	Measured $\text{Pb}(\text{NO}_3)_2$ concentration (mmol/L)	Density ( $\text{g}/\text{cm}^3$ )	Electrical conductivity ( $\mu\text{S}/\text{cm}$ )	pH
Tap water	NA	NA	1.00	145	7.35
Distilled water	NA	NA	1.00	3.18	6.91
$\text{Pb}(\text{NO}_3)_2$ solution	20	19.9	1.01	2590	4.83

50	50.1	1.02	5550	4.79
200	199.7	1.07	16060	4.38
500	500.2	1.17	29960	4.22

Note: NA = not applicable.

### 2.3 Liquid limit test

The specimens used for liquid limit tests were prepared as per the following procedures: First, the dry mixtures were prepared by mixing prearranged amounts of air-dried constituents directly. The contents of bentonite and sand in the unamended backfill were selected as 10% and 90%, respectively. The contents of XG, bentonite and sand in the amended backfill were selected as 0.6%, 10% and 89.4%, respectively. Then, distilled water and  $\text{Pb}(\text{NO}_3)_2$  solutions were used to adjust the water contents of mixtures until the slump heights of mixtures were  $125 \text{ mm} \pm 5 \text{ mm}$ . Finally, these mixtures were put in a plastic bag to cure for 20 days. This curing process ensured chemical equilibrium between specimens and testing liquids. After curing, these specimens were air-dried and then milled with mortar grinder. Liquid limit tests were conducted on these air-dried specimens based on ASTM D4318 (ASTM 2018).

### 2.4 Hydraulic conductivity test

Hydraulic conductivity tests were carried out on unamended and XG-amended backfill specimens using constant head method in accordance with ASTM D5084 (ASTM 2016) and ASTM D7100 (ASTM 2011). These backfill specimens met the slump height of 125 mm. The effective confining stress and hydraulic gradient were selected as 38.3 kPa and 200, respectively. First, the specimens were infiltrated with tap water. After the hydraulic equilibrium was met according to ASTM D5084 (ASTM 2016), the permeant liquid was replaced by 20 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solutions. The outflow specimens were collected at various time intervals, and their pH, EC, and Pb concentration were analysed. After the chemical equilibrium was met according to ASTM D7100 (ASTM 2011), the permeant liquid was sequentially changed to 50, 200, and 500 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solutions.

### 2.5 Image analyses

Scanning electron microscopy (SEM) analyses were applied to evaluate the effect of XG on the microstructure of SB backfill, which were conducted on the unamended and XG-amended backfills under exposure to tap water and 50 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solution after hydraulic conductivity testing. The  $1 \text{ cm}^3$  unamended and XG-amended backfill specimens were obtained using a fine saw, which were freeze-dried prior to SEM (SU3500, Hitachi High-Tech, Tokyo, Japan) analyses.

## 3 TEST RESULTS AND ANALYSES

Figure 1 presents the relationship between the concentration of testing liquids and the liquid limit of unamended and XG-amended backfill. As expected, the liquid limit decreased with growing concentration in spite of the type of backfill. This trend was attributed to the fact that  $\text{Pb}^{2+}$  compresses the thickness of diffuse double layers of bentonite particles (Mitchell and Soga, 2005). It is seen from Figure 1 that the liquid limits of amended backfills were higher than those of unamended backfills at a given concentration of testing liquid. The XG-amendment improved the liquid limit of unamended backfill. The observation is due to the numerous hydrophilic functional groups on the XG molecules, which can bind free water molecules via hydrogen bonding (Fu et al., 2022).

The variations of hydraulic conductivity with different testing times and concentrations of testing liquids for unamended and XG-amended backfills are displayed in Figure 2. It should be noted that the hydraulic and chemical equilibriums between the testing liquids and the specimens were met. The hydraulic conductivity of the two specimens increased with growing concentration of  $\text{Pb}(\text{NO}_3)_2$  solutions. The hydraulic conductivities of unamended and XG-amended backfills were  $4.72 \times 10^{-11}$  and  $4.51 \times 10^{-11}$  m/s, respectively, which were at the same order of magnitude. The hydraulic conductivity of XG-SB backfill was less than that of unamended backfill when permeated with  $\text{Pb}(\text{NO}_3)_2$  solutions with the concentration varying from 20 to 500 mmol/L. Importantly, the hydraulic conductivity of unamended backfill was greater than the acceptable threshold (i.e.,  $10^{-9}$  m/s) under exposure to 20 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solution, whereas that of amended backfill was less than the threshold even if exposed to 500 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solution, which presented excellent chemical compatibility.

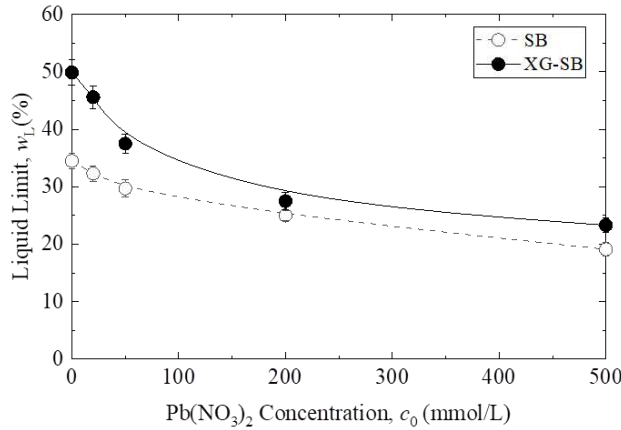


Figure 1. Variation of liquid limit with concentration

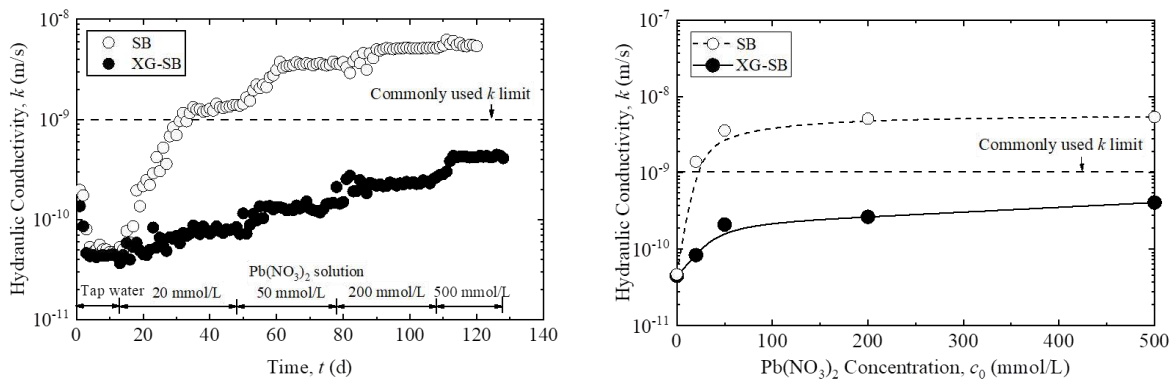


Figure 2. Variation of hydraulic conductivity with different testing times (left). Variation of hydraulic conductivity with different  $Pb(NO_3)_2$  concentrations

The relationship between hydraulic conductivity and liquid limit of unamended and XG-amended backfills is presented in Figure 3. In general, the hydraulic conductivity declined with the increase of liquid limit of backfill. The hydraulic conductivity of unamended backfill presented more sensitive to the variation of liquid limit. The hydraulic conductivity of unamended backfill increased sharply to a stable value with the decrease of liquid limit. The hydraulic conductivities of XG-amended backfills were less than those of unamended backfills at a given liquid limit when exposed to lead solutions.

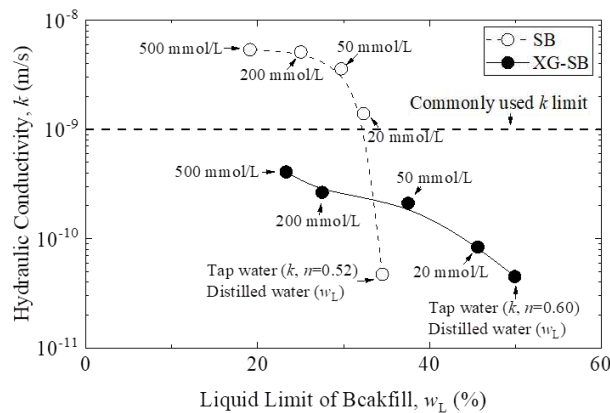
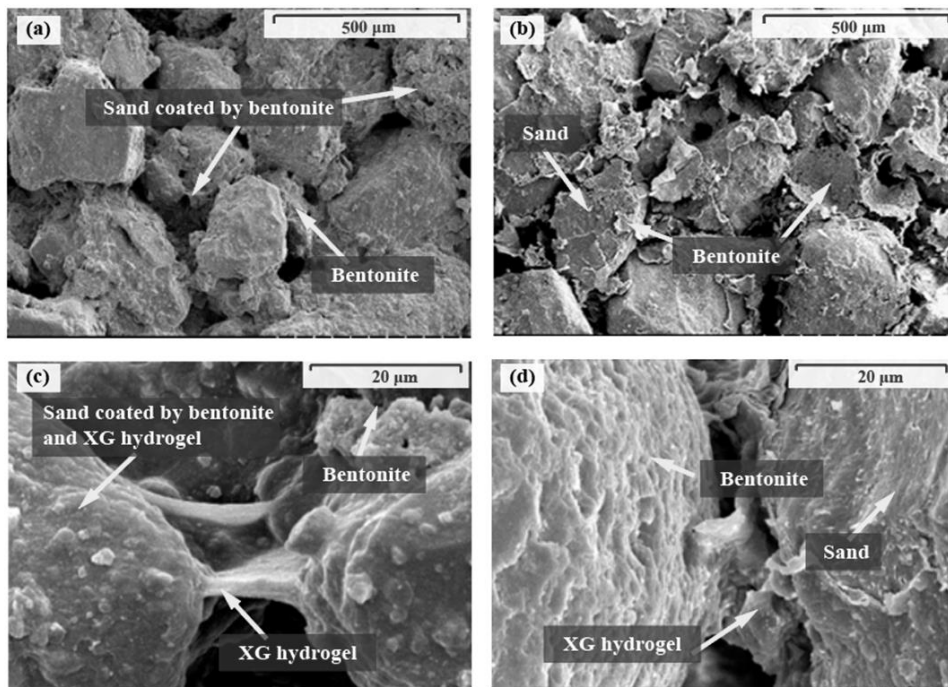


Figure 3. Variation of hydraulic conductivity with liquid limit

Figure 4 presents SEM graphics of XG-amended and unamended backfills under exposure to tap water and 50 mmol/L  $Pb(NO_3)_2$  solution. From Figure 4(a), the bentonite particles swell due to tap water

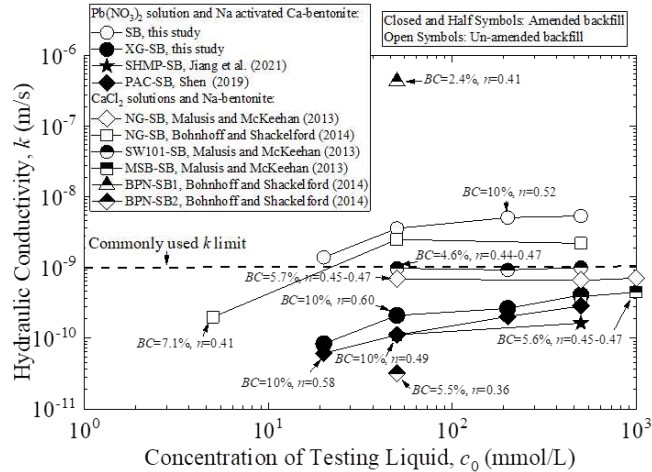
hydration to coat the surface of sand granules and fill the intergranular pores. Similarly, the sand particles in the XG-amended backfill were covered by XG hydrogel and hydrated bentonite, which looked like a “thin coating” (see Figure 4(c)). From Figure 4(c), the XG hydrogel filled the intergranular pores like a viscous film. Therefore, the XG-amended and unamended SB backfills possessed narrow and tortuous flow paths that were responsible for low hydraulic conductivity (Yang et al., 2022a).

Under exposure to 50 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solution, the hydrated bentonite was not enough to fill the intergranular pores in the unamended backfill (Figure 4(b)), which is because  $\text{Pb}^{2+}$  compresses the thickness of diffuse double layers of bentonite (Mitchell and Soga, 2005). The intergranular pores initially filled with hydrated bentonite were opened, causing the hydraulic conductivity was significantly higher than the threshold (see Figure 2). By contrast, the hydrated XG hydrogel exposed to 50 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solution could still fill the intergranular pores in the amended backfill (see Figure 4(d)), forming narrow and tortuous flow paths. Therefore, the XG-amended backfill presented more excellent chemical compatibility than unamended SB backfill.



**Figure 4.** SEM graphics of unamended and XG-amended backfills exposed to different testing liquids: (a) SB backfill exposed to tap water; (b) SB backfill exposed to lead solution; (c) XG-SB backfill exposed to tap water; and (d) XG-SB backfill exposed to lead solution

Figure 5 summarizes the hydraulic conductivities of various polymer-amended and unamended backfills permeated with  $\text{Pb}(\text{NO}_3)_2$  and  $\text{CaCl}_2$  solutions on the basis of this study and previous studies. It should be noted that the hydraulic and chemical equilibria between the testing liquids and the backfills were met. From Figure 5, the hydraulic conductivity of backfill was related to bentonite content and type, polymer amendment, and cation concentration. In general, the hydraulic conductivity of polymer-amended backfills was less than that of unamended backfills, except for SW101-SB backfill. The hydraulic conductivity of unamended backfills increased as the cation concentration increased regardless of types of permeating liquid and bentonite. The hydraulic conductivity of unamended backfills containing Na-bentonite was less than that of unamended backfills containing Na activated Ca-bentonite, which is due to superior swelling performance of Na-bentonite (Yang et al., 2018b, 2022b). The higher bentonite content yielded decrease in the hydraulic conductivities of unamended backfills.



**Figure 5.** Hydraulic conductivity with different testing liquids for unamended and polymer-amended SB backfills

#### 4 CONCLUSIONS

The results of this study indicated that XG-SB backfill might be a promising backfill material for use in SB vertical barriers at sites with Pb-contaminated groundwater. The XG-amendment improved the liquid limit and chemical compatibility of backfill exposed to lead solutions with the concentration varying from 20 to 500 mmol/L. As expected, the hydraulic conductivity increased with growing concentration of lead solutions regardless of the type of backfill. SEM analyses showed the hydrated XG hydrogel exposed to 50 mmol/L  $\text{Pb}(\text{NO}_3)_2$  solution could still fill the intergranular pores in the amended backfill, forming narrow and tortuous flow paths that were responsible for excellent chemical compatibility of XG-amended backfill. An extensive comparison of hydraulic conductivity for unamended and polymer-amended SB backfills on the basis of this study and previous studies showed that the bentonite content and type, polymer amendment, and cation concentration of testing liquid greatly influenced the hydraulic performance of the SB backfills.

#### 5 ACKNOWLEDGEMENTS

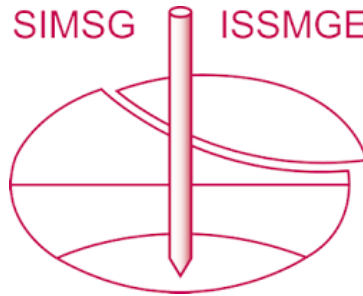
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