

Impact of lead and sodium carbonate on consolidation and hydraulic properties of clayey sand

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ABSTRACT

Leakage of lead as poisonous heavy metal from pipelines, chemical storages, and industrial waste disposal is of great concern to geo-environmental engineers due to industrialization and development of suburban districts of megacities. Despite its growing environmental concerns, lead contamination can change the geotechnical properties of clayey soils, such as compressibility and hydraulic conductivity with wide application in design of foundations and buffer systems, respectively. Therefore, the main objective of this study is to explore the consolidation characteristics of different sandbentonite mixtures contaminated with three concentrations of lead nitrate. More importantly, the influence of sodium carbonate additive as a remediation technique for the contaminated soil is investigated. Results revealed that the addition of lead nitrate caused a reduction in compressibility coefficient and a rise in consolidation coefficient. However, the sodium carbonate treatment adversely affects the two consolidation properties. On the other hand, the addition of 10% sodium carbonate reduced the hydraulic conductivity of the contaminated soil about tenfold. Form practical point of view, the studied countermeasure is not recommended for load bearing materials but recommended for enhanced isolating liners.

RÉSUMÉ

La fuite de plomb, métal lourd toxique, par les pipelines, le stockage de produits chimiques et le traitement des déchets industriels fait partie des principales préoccupations des ingénieurs géoenvironnementaux en raison de l'industrialisation et du développement des zones périurbaines des mégalopoles. Malgré les préoccupations croissantes sur les questions environnementales, la contamination par le plomb peut altérer les propriétés géotechniques des sols argileux, comme la compressibilité et la conductivité hydraulique qui ont un large champ d'application, respectivement dans la conception de fondations et de systèmes tampons. Par conséquent, l'objectif principal de cette étude est d'explorer les caractéristiques de consolidation de différents mélanges sable-bentonite contaminés par trois concentrations différentes de nitrate de plomb. Plus important encore, l'influence de l'adjuvent carbonate de sodium en tant que technique de réhabilitation du sol contaminé a été étudiée. D'un côté, les résultats révèlent que l'ajout de nitrate de plomb a entraîné une réduction du coefficient de compressibilité et une hausse du coefficient de consolidation. Inversement, le traitement par carbonate de sodium a eu un effet néfaste sur les deux propriétés de consolidation. D'un autre côté, l'addition de 10 % de carbonate de sodium a divisé la conductivité hydraulique du sol contaminé par dix environ. D'un point de vue pratique, les contremesures étudiées ne sont pas recommandées pour les matériaux porteurs, mais plutôt pour les revêtements isolants.

1 INTRODUCTION

Heavy metal pollution is a major environmental problem worldwide. Primary heavy metals in the soil that are harmful to human health and the existing ecosystem include lead, zinc, chromium, cadmium, nickel, and copper. As a consequence of urban planning and development, construction on contaminated lands may be inevitable. Heavy metals are considered among the most important contamination agents. Therefore, it is essential to determine the behavior of soils contaminated by heavy metals. More importantly, proper understanding of countermeasure against polluted soils will help making the right decision during remediation process. Consequently, research on understanding the geotechnical properties of clayey soils contaminated with different heavy metals has received growing attention recently. For example, Hosseinzadeh et al. (2019) assessed the effect of lead nitrate on the behavior of clayey sand using consolidated undrained triaxial tests. The results revealed that the addition of heavy metals to soil significantly increases the cohesion and reduces the internal friction of contaminated soils in comparison to the uncontaminated ones. In addition, changing the base clay mineral from bentonite to kaolinite showed different behavior in terms of shear strength. According to the results, addition of heavy metals increases and decreases the shear strength of sandbentonite and sand-kaolinite mixtures, respectively. Du et al. (2014) carried out a series of tests on leadcontaminated kaolinite clay. Results confirmed that with an increase in lead concentration, the specific gravity increase and the liquid limit, plastic limit, specific surface, elasticity modulus, and pH of the soils decrease. Consolidation characteristics are categorized among the most significant geotechnical properties of contaminated soils for both serviceability and ultimate state analyses. It is well recognized that compressibility of fine-grained soils can be varied under both saturated and unsaturated conditions (Ng et al., 2017) although the focus of current study is on fully saturated conditions. Du et al. (2015) conducted several oedometer tests on bentonite-sand and kaolinitesand mixtures with various lead concentrations. The results revealed that a linear trend can be established between the compression index and the liquid limit. Furthermore, the compression index ratio, which is the ratio between the compressibility coefficients of contaminated and uncontaminated soils, changed considerably in lower lead concentrations and became almost constant in higher concentrations. Fan et al. (2014) proposed a unique relationship between the compressibility coefficient and the void ratio of lead-contaminated bentonite-sand mixtures. However, the proposed relationship is not applicable when the bentonite content of the mixture is as low as 5% of the sand.

Besides, in order to reduce the consolidation rate and permeability of the contaminated clays, some researchers proposed to add carbonate to the soils. The CO3-2 ions in sodium carbonate, which are the source of potential determining ions (PDI) (Yong et al., 2001), increase the van der Waals forces between clay particles and soil resistance against settlement (Ouhadi et al., 2007). Ouhadi et al. (2011) evaluated the interactions between clay, calcite, and heavy metals in mixtures. According to the results, the effect of coppers and carbonate on microstructural evolution of clayey soils was highlighted. The consequences of a tighter aggregation of clay particles and stiffer consolidation characteristics after soil-contaminant caused the restructuring or development of microstructures into tighter microstructural units. Similar phenomenon was also observed in interaction between salt and clay particles in a natural dispersive loess, where a stiffer aggregation of clay assemblies in presence of salt water resulted in 4% reduction in collapsibility compared to pure and distilled water (Sadeghi et al., 2019). However, there are few types of research conducted on the behavior of clay-heavy metals remediated with sodium carbonate mixtures. The main objectives of current study is to systematically examine the variations in consolidation properties as well as saturated hydraulic conductivity of sand-bentonite mixtures contaminated with different concentrations of lead and treated with sodium carbonate accordingly.

2 MATERIALS AND METHODS

The coarse-grained soil used in this study is Firuzkuh No. 161 soil. Figure 1 shows the grain size distribution of the Firuzkuh No. 161 soil, which has been determined using the sieve analysis (ASTM D 422). According to the grain size distribution curve, the soil lies in the range of sand with a relatively uniform distribution based on the coefficients of uniformity and curvature. Consequently, the soil is classified as poorly graded sand (SP) according to the Unified Soil Classification System (ASTM D 2487). It is also noted that the specific gravity of the Firuzkuh No. 161 soil was determined as 2.67 (ASTM D 854).

The fine-grained soil used in this study is bentonite. Liquid limit, plastic limit, and plasticity index of bentonite are 136%, 52%, and 84%, respectively (ASTM D 4318). The bentonite with the specific gravity of 2.67 is classified as clay with high plasticity (CH). In addition, the mineral components in the bentonite, as determined by X-ray diffraction, are as follows: 52% SiO2, 26% Al2O3, 7% Fe2O3, and 2% CaO. It is to mention that sodium carbonate (NaCO₃) is used as a carbonate based additive in this study.

In order to prepare the specimens in a consistent manner for fair comparison, the maximum dry density obtained from the modified Proctor Effort (ASTM D 1557) for the mixture of Firuzkuh No. 161 sand and 20% bentonite was used for preparing all the specimens. According to the results, the maximum dry unit weight, and the optimum water content of the soil mixture were 17 KN/m3 and 16.5%, respectively. In addition, lead nitrate was used as the heavy metal for contamination. The reason for adding lead as a contaminant in this study is its high presence in the industrial wastewater and its destructive effects on the environment.



Figure 1. Grain size distribution of the Firuzkuh No. 161 soil

2.1 Sample preparation

During the preparation of samples for all the tests, three levels of lead concentration as 0.1, 0.2, and 0.5 mol/lit were sprayed to the oven-dried sand-bentonite mixture to reach the desired weight compositions of the soil and contaminant. In almost half of the specimens, 10% by mass sodium carbonate was added to the pore fluid. After adding the solution to the soil and the mixing process, the mixture was kept in Ziploc plastic bag for seven days for proper lead absorption and moisture equalization. The samples are prepared at 0.95 of the maximum dry unit weight and the optimum water content.

2.2 Consolidation test

The one-dimensional consolidation test is used to determine the rate and amount of soil volumetric contraction in response to a rise in the applied effective stress (ASTM D 1557). The rate of consolidation depends on the applied stress level and the corresponding permeability of the specimen. In addition, this study aims at investigating the influence of two other relatively new parameters on consolidation properties, including the lead contamination level and the selected carbonate based additive.

2.3 Test program

In this study, the SP typed sand was mixed with different percentages of bentonite, including 5%, 10%, and 20% of oven-dried sand. The product test materials were used to explore the effect of lead contamination on the sandbentonite mixtures. The soil mixtures were then contaminated by 0, 0.1, 0.2 and 0.5 mol/lit lead nitrate. In addition, sodium carbonate corresponding to 10% of pore fluid was added to nearly half of the specimens to examine its remediation performance. The detailed test conditions for all consolidation tests described in the previous part are reported in Tables 1. In summary, a total of 21 consolidation tests were performed on the contaminated and uncontaminated specimens with different percentages of bentonite, lead nitrate, and sodium carbonate.

Table 1. Specifications of the samples used in this study

Test	Lead nitrate	Bentonite	Sodium
no.	concentration	(%)	carbonate
	(mol/lit)		(%)
1	0	5	-
2	0.1	5	-
3	0.2	5	-
4	0.5	5	-
5	0	10	-
6	0.1	10	-
7	0.2	10	-
8	0.5	10	-
9	0	20	-
10	0.1	20	-
11	0.2	20	-
12	0.5	20	-
13	0.1	5	10
14	0.2	5	10
15	0.5	5	10
16	0.1	10	10
17	0.2	10	10
18	0.5	10	10
19	0.1	20	10
20	0.2	20	10
21	0.5	20	10

3 RESULTS

In order to analyze and compare the consolidation parameters of the contaminated and uncontaminated samples and to observe the effect of sodium carbonate on the samples containing different concentrations of lead nitrate, 21 consolidation tests were conducted. In this section, the consolidation behavior of contaminated soils is explained and discussed. Afterwards, the effects of influencing factors on output parameters including the coefficient of compression (C_c), the coefficient of consolidation (C_V), and the permeability will be interpreted. Finally, the variations in the Atterberg limits with contamination level is presented and a correlation between the coefficient of compression and the liquid limit will be proposed based on the new experimental data.

3.1 The effect of lead nitrate and sodium carbonate on the consolidation curve

Compressibility among other geotechnical properties has been proved to depend on soil microstructure (Ng et al., 2020). Therefore, any factor affecting the soil microstructure can alter the consolidation characteristics as well. The consolidation curves in terms of void ratio versus the effective stress are presented in semi logarithmic scale in Figures 2 to 4 for various lead concentrations with and without sodium carbonate. As shown in Figure 2, the initial void ratio is higher for amended samples with sodium carbonate in comparison to the samples with only lead nitrate contamination. The consolidation curves shift generally downward with a rise in the concentration of contaminant. For instance, the initial void ratio of the sample with 0.5 mol/lit lead nitrate is 0.58. For the sample with 0.5 mol/lit lead nitrate and 10%, sodium carbonate is 1.08, which shows 55% and 16% reduction in the initial void ratio of uncontaminated soils (e=1.28), respectively. In addition, uncontaminated specimen undergo increase porosity changes during the loading process, but in lead nitrate contaminated specimens, porosity changes during loading will be less due to the increased flocculated structure of clay particles. This observation is also consistent with the study of Ouhadi et al. (2011) who reported a stiffer structure would be obtained at higher heavy metal concentrations. On the other hand, the presence of sodium carbonate neutralizes the significant influence of heavy metal on compressibility. As a result, sodium carbonate amended contaminated soils are generally more compressible than the contaminated soils with no sodium carbonate addition. Similarly, the observed trends are also valid for other two admixtures with 10% and 20% bentonite constituent according to figures 3 and 4, respectively. However, there is a clear enhancement of compressibility at all conditions examined due to a rise in the bentonite percentage.



Figure 2. Effect of lead nitrate and sodium carbonate on the consolidation curves of admixture containing 5% bentonite



Figure 3. Effect of lead nitrate and sodium carbonate on the consolidation curves of admixture containing 10% bentonite



Figure 4. Effect of lead nitrate and sodium carbonate on the consolidation curves of admixture containing 20% bentonite

3.2 The effect of lead nitrate and sodium carbonate on the coefficient of compression (C_c)

According to Figure 5, adding lead nitrate to the soil results in a decrease in the compression coefficient at a reduced rate. In other words, as the concentration of lead nitrate increases, the compression coefficient approaches a threshold value. The delimiting concentration level above which no significant changes in C_c occurred is roughly 0.1 mol/lit. In fact, due to the addition of lead nitrate to the soil and increasing its concentration, the structure of the soil becomes more flocculated, and therefore, with increasing the concentration of pollutants, the compression ratio decreases. However, the results suggest a threshold concentration value that further structural flocculation may not form afterwards. The cause of the flocculated structure is the cation exchange process that occurs as a result of the addition of lead nitrate. Due to the cation exchange process, lower capacity ions of bentonite such as K⁺ and Na⁺ are replaced with higher capacity ions as Pb⁺². As a result of this chemical reaction, the clay particles are brought closer together as Pb⁺² has a lower hydrate radius than Na⁺ or K⁺ and by reducing the thickness of the diffuse double layer, the structure becomes more flocculated (Ouhadi et al., 2011). A similar influence of existing prevailing amount of Na⁺ in the pore water of a natural collapsible soil was documented to reduce wetting-induced collapse compared with the same sample inundated with distilled water in absence of Na⁺ (Sadeghi et al., 2020). The reduction in the compression coefficient is such that, for example, by increasing the concentration of lead nitrate from 0.1 to 0.5 mol/lit in samples containing 5% bentonite. the compression coefficient changes from 0.16 to 0.13 which is insignificant. However, a significant reduction in C_c as much as 0.1 occurred due to addition of 0.1 mol/lit lead nitrate to the uncontaminated soil.

On the other hand, the inverse interaction mechanism at clay particle surface happens as a consequence of sodium carbonate amendment. Adding sodium carbonate to samples contaminated with lead nitrate augments Na+ cation near the clay surface, which prevents the formation of flocculated structure due to its relatively higher hydrated radius compared with Pb⁺². As a result, samples containing sodium carbonate have a higher compression coefficient. The comparison of solid lines with broken lines in Figure 5 confirms this theoretical physico-chemical justification. For example, in the admixture containing 5% bentonite and contaminated with 0.1 mol/lit lead nitrate, the compression coefficient reduces from 0.24 to 0.16 if the sodium carbonate is removed. In addition, the bentonite percentage is also a prevailing factor in enhancing the compressibility. According to Figure 5, if the bentonite percentage increases from 5% to 20% at 0.1 concentration in the absence of sodium carbonate, the compression coefficient increases from 0.16 to 0.23. Figure 6 plotted the variations in the compression index ratio against the concentration of lead nitrate. It is noted that the compression index ratio is defined as the ratio between the compressibility coefficient of contaminated soil to that of uncontaminated one (C_c / C_0) . According to the results of Figure 6, the compression index ratio is less than unity for all testing conditions considered in this study, implying that both lead nitrate contamination and sodium carbonate amendment results in a stiffer microstructure compared to the base soil.

Figure 7 shows the changes in the coefficient of compression versus the initial void ratio defined as the void ratio corresponding to the vertical effective stress of 1 KPa ($\sigma' = 1$ KPa) for contaminated and uncontaminated samples with different percentages of bentonite. According to the results, there is a nonlinear ascending trend for variations in C_c against e_1 , which is also consistent with the observation of Fan et al. (2014). However, the correlation

may not be unique as stated by them for the sand-calcium bentonite mixtures:

$$C_C = 0.056e_1^2 + 0.13e_1$$
 [1]

On the other hand, a minor modification was applied to equation 1 to give the most appropriate correlation for the test materials and conditions in in the present study:

$$C_C = 0.088e_1^2 + 0.0599e_1 + 0.0474$$
 [2]

Figure 7 also compares the two independent dataset and the corresponding best-fit curves according to equations 1 and 2.

3.3 The effect of lead nitrate and sodium carbonate on the coefficient of consolidation (C_v)

Figures 8 to 10 show variations in consolidation coefficient against the average vertical compression stress applied to the sample at each loading interval (σ'_{ave}) for different percentages of bentonite (5, 10, and 20%), including lead nitrate and sodium carbonate contaminated soils.



Figure 5. Effect of lead nitrate and sodium carbonate on the compression coefficient of samples containing different percentages of bentonite



Figure 6. Effect of lead nitrate and sodium carbonate on the compression index ratio of samples containing different percentages of bentonite.



Figure 7. The correlation between the coefficients of compression and the initial void ratio.

Based on Figure 8 for admixture with 5% bentonite, it is observed that the amount of consolidation coefficient in uncontaminated soil is the lowest and in contaminated soil with lead nitrate is the highest value. Uncontaminated soil has a lower rate of consolidation because it undergoes more changes in porosity during the consolidation process. Increasing the concentration of heavy metals also leads to an increase in the consolidation coefficient. On the other hand, the presence of sodium carbonate in leadcontaminated soils prevents the formation of a flocculated structure and a lower coefficient of consolidation is observed than lead-contaminated samples without sodium carbonate. According to Figures 8 to 10, it is observed that with increasing the bentonite percentages in the mixture, the amount of consolidation coefficient also decreases.

Figure 11 shows the average value of the consolidation coefficient, $C_{V (ave)}$, for each sample during consolidation in terms of lead nitrate concentration and sodium carbonate amounts. It reveals that by adding lead nitrate to the soil and increasing its concentration, the average consolidation coefficient increases. In addition, lead-contaminated samples treated with sodium carbonate have a lower average consolidation coefficient than lead-contaminated samples. It is also observed that samples containing 20% and 5% of bentonite have the highest and lowest consolidation coefficients, respectively.

3.4 The effect of lead nitrate and sodium carbonate on the coefficient of permeability (k)

One of the critical parameters in investigating the effect of heavy metal pollutants on soil behavior is the coefficient of permeability. According to the consolidation theory, the coefficient of permeability can be obtained as:

$$k = C_V . m_V . \gamma_w \tag{3}$$

$$m_V = \frac{\Delta \varepsilon}{\Delta \sigma}$$
 [4]

Where *k* is the coefficient of permeability (m/s), *C_v* is the consolidation coefficient (m²/s), *m_v* is the coefficient of volume compressibility (m²/N), and γ_w is the unit weight of water (N/m³). In addition, $\Delta \varepsilon$ and $\Delta \sigma$ denote changes in the strain and stress, respectively.



Figure 8. The effect of lead nitrate and sodium carbonate on the consolidation coefficient of samples containing 5% bentonite



Average vertical stress (kPa)

Figure 9. The effect of lead nitrate and sodium carbonate on the consolidation coefficient of samples containing 10% bentonite





Figure 10. The effect of lead nitrate and sodium carbonate on the consolidation coefficient of samples containing 20% bentonite



Lead concentration (mol/l)

Figure 11. The effect of lead nitrate and sodium carbonate on the average consolidation coefficient of samples containing different amounts of bentonite

Figures 12 to 14 show the variations in permeability of leadcontaminated samples against the average effective stress during each loading interval (σ'_{ave}). According to the figures, the addition of lead nitrate to different sandbentonite admixture leads to a significant increase in the permeability coefficient. For example, in the first step of loading as shown in Figure 12, the permeability coefficient of uncontaminated soil and 0.5 mol/lit lead-contaminated soil is 3.64E-10 and 6.58E-9 m/s, respectively. Furthermore, as the concentration of heavy metal increases, the permeability also increases. In fact, the addition of lead nitrate to the soil resulted in the formation of a more flocculate structure; hence, the coefficient of permeability increases. On the other hand, the addition of sodium carbonate to the lead-contaminated soil prevents the creation of flocculate structure and reduces the permeability coefficient. It should be also noted that according to the results of figures 12 to 14, the lowest permeability corresponding to the clean material may not be achieved even after amendment of contaminated soil with 10% sodium carbonate for the soil and pollutant conditions at hand. This finding can be of primary importance in safe and reliable design of the barrier mechanism of landfills where the leachate contains certain amount of heavy metals.

Figure 15 shows the average value of the coefficient of permeability, K_{ave} , for each sample in terms of lead nitrate concentration and sodium carbonate amounts. For example, the average permeability coefficient in samples contaminated with 0.1 mol/lit lead nitrate without sodium carbonate and with sodium carbonate are 1.22E-9 and 2.76E-10, respectively. It is also shown that increasing the percentage of bentonite leads to a decrease in the coefficient of permeability as expected.



Average vertical stress (kPa)

Figure 12. The effect of lead nitrate and sodium carbonate on the permeability coefficient of samples containing 5% bentonite



Average vertical stress (kPa)

Figure 13. The effect of lead nitrate and sodium carbonate on the permeability coefficient of samples containing 10% bentonite



Average vertical stress (kPa)

Figure 14. The effect of lead nitrate and sodium carbonate on the permeability coefficient of samples containing 20% bentonite



Figure 15. The effect of lead nitrate and sodium carbonate on the average coefficient of permeability

3.5 The effect of lead nitrate and sodium carbonate on the Atterberg limits

The variation of the plasticity index (PI) of contaminated bentonite is presented in terms of liquid limit (LL) in Figure 16. By increasing the lead concentrations, due to the high cation exchange in bentonite and the replacement of higher capacity cations (Pb⁺²) with lower capacity cations, the thickness of double diffuse layer reduces which in turn results in lower plasticity values. A summary of measured Atterberg limits for both clean and contaminated soils with three levels of lead nitrate concentrations is given in Table 2.



Figure 16. The effect of lead nitrate concentration on liquid limit and plasticity index of bentonite.

Table 2. Summary of Atterberg limits for lead nitrate contaminated bentonite.

	Lead nitrate concentration (mol/lit)				
	0	0.1	0.2	0.5	
LL (%)	79.9	74.8	70.9	65.5	
PL (%)	31.1	28.3	26.6	25	
PI (%)	48.8	46.5	44.3	40.5	

Figure 17 illustrates the variations in the coefficient of compressibility with liquid limit of sand-bentonite mixtures. The data corresponds to lead-contaminated and uncontaminated samples containing 5%, 10%, and 20% bentonite with and without sodium carbonate. The relationships between the compression coefficient and liquid limit proposed by Bowels (1979) as well as Terzaghi and Peck (1948) are also depicted. As shown in this figure, the data corresponding to the test samples more or less lay within the upper bonds and fairly follow the average trend of Bowels (1979).



Figure 17. The relationship between the coefficient of compression and liquid limit for lead-contaminated samples with and without sodium carbonate amendment

4 CONCLUSIONS

In this study, the consolidation behavior of clayey sand in the presence of lead as a heavy metal pollutant was investigated. In order to remediate the contaminated soil, sodium carbonate was added to the specimens for the second series of consolidation tests. According to the results of 21 consolidation tests as well as some complementary index tests, the key conclusions can be drawn as follows:

- The compression coefficient (Cc) decreased with the addition of lead nitrate to the soil. A significant decrease in Cc of polluted soil occurred at lead concentration of 0.1 mol/lit compared with the base soil. However, only marginal reduction in Cc was measured for higher concentrations. On the other hand, the addition of 10% sodium carbonate to the contaminated soil increases Cc. This observation may suggest the initial enhancement of flocculate structure due to the interaction between bentonite and sodium carbonate. Moreover, an increase in the percentage of bentonite resulted in the augmentation of compressibility as expected.
- The consolidation coefficient (C_V) increased with the addition of lead nitrate to the bentonite-sand mixture, implying a more accelerated consolidation process in contaminated soil compared to the base soil. However, the presence of sodium carbonate reduced the average coefficient of consolidation of contaminated soils.
- 3. As the concentration of lead nitrate increases, the average permeability coefficient in samples with different percentages of bentonite also increases. Of particular interest is the positive role of sodium carbonate in reduction of saturated permeability. According to the measurements, a tenfold drop in permeability of contaminated soil treated with sodium carbonate was observed. In other words, the results suggest that the proposed treatment can enhance the performance of buffer liners while the remediated soil may not be considered desirable as a load bearing geo-material.
- 4. From a practical point of view, the hydraulic conductivity of contaminated clayey soil could be improved by sodium carbonate amendment if the material is used as a leachate barrier. However, it is important to keep it in mind that the best performance of uncontaminated soil may not be achieved even after remediation with sodium carbonate.

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