



Effect of Adsorbents on Resistance Parameters of Heavy Metal-contaminated Clayey Sand Soils

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PAPER INFO

Paper history:

Received 13 June 2022

Received in revised form 23 July 2022

Accepted 31 July 2022

Keywords:

Shear Strength

Heavy Metals

Rice Husk Ash

Zeolite

Soil Structure Modification

ABSTRACT

Combining soil with some adsorbents improves soil structure and shear strength. Thus, an optimal ratio of some adsorbents in the soil composition enhances soil adsorption capacity, reduces the possibility of groundwater contamination with these hazardous compounds, and leads to increase soil resistance parameters. This paper investigates the effect of heavy metals and adsorbents on the geotechnical behavior of clayey sand soils contaminated with lead and zinc as heavy metals as well as zeolite and rice husk ash as adsorbents. Clayey sand (mixture of sand with 20% kaolinite or bentonite) was considered as a base composition and the behavior of it was studied in both contaminated and uncontaminated states. Then, for increasing the heavy metal adsorption capacity of clayey sand, two types of adsorbents (zeolite and rice husk ash) were added to the base composition and their behavior were investigated in the case of Lead and Zinc contamination. The results revealed that replacing the rice husk ash and zeolite adsorbents in the sand combination with 20% kaolinite clay significantly reduced the concentration of lead and zinc nitrate in the solution. Replacing 15% of rice husk ash with kaolinite heightened the absorption of lead nitrate and zinc nitrate by 228.8% and 291.6% in kaolinite sand. It was also found that adding nitrate to kaolinite increased the liquid and plastic limits. According to the results, in kaolinite, the value of the liquid limit rose from 49.8 to 59.1 upon elevating the concentration of lead to 5000 ppm, while the plastic limit also increased from 31 in the non-contaminated state to 36.4 in 5000 ppm in the infected state. According to obtained results, the dispersed structure is formed by increasing the concentration of lead nitrate in composition of sandy clay with low plasticity adsorbent; thus, shear resistance decreased. Changing in type of clay minerals to high plasticity cause the different trend in shear resistance parameters. Increasing the concentration of lead nitrate in bentonite composition, lead to flocculated structure be formed and shear strength increased.

doi: 10.5829/ije.2022.35.11b.09

1. INTRODUCTION

The expansion of industrial areas over the past few decades, followed by an increase in industrial effluents, has led to the spread of soil contaminations. The leakage of heavy and toxic metals is one of the most important causes of soil contamination. Heavy elements enter the environment through wastewater contaminated with these metals or through leakage from their storage tanks, which results in contamination of soil and groundwater with these toxic compounds [1]. Note that one of the components of crude oil is heavy metals. Thus, in addition to industrial areas, oil-rich areas are also exposed to this pollution from various aspects, from the

stage of oil extraction to the transfer and refining of petroleum products [2]. Soil pollution, in addition to being one of the environmental crises, also leads to changes in its geotechnical parameters [3].

Soil type, initial concentration of heavy metals, and their extent in the soil cause changes in soil resistance parameters. As such, studying the behavior of soils contaminated with heavy metals and finding methods to deal with these contaminants is also of great interest in terms of geotechnical issues [4]. One of the most important processes affecting the cause of pollutants in the soil is their absorption by soil particles or adsorbents in the soil composition [5]. The absorption of heavy metals in fine-grained soils is higher than in granular soils

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[5]. Also, the presence of heavy metals in the soil alters the structure and fundamental behavior of the soil, causing variations in the mechanical, physical, and chemical parameters of the soil. These changes can improve or weaken the geotechnical parameters of the soil [6].

Combining soil with some adsorbents improves soil structure and enhances soil shear strength [7, 8]. Thus, soil composition with an optimal ratio of some adsorbents, in addition to reduce the mobility of heavy elements in the soil and boosting the soil adsorption capacity, reduces the possibility of groundwater contamination with these hazardous compounds and results in increased soil resistance parameters [9, 10]. Also pH of pore fluids has important effect on the physico-chemical and mechanical behavior of clays [11].

Given economic considerations as one of the basic principles in projects, the use of natural and inexpensive adsorbents with sufficient abundance in nature has been presented as one of the solutions to improve contaminated soils [12, 13]. By studying the effect of heavy metals on the properties of kaolinite, it was found that shear strength has been affected by heavy metal contamination [14]. The results showed that the heavy metal contaminants increased the intermolecular space and caused more movement of particles in soils containing kaolinite. Thus, shear strength decreased in contaminated kaolinite by divalent heavy metals [15]. According to reported data in literature [16], due to the number of tanks containing heavy metals as well as pressurized pipelines to transport these compounds to other industrial areas, a huge amount of pollutants always enter through leaks from pipes or tanks. Thus, a significant amount of heavy metals has been reported in the soils around refineries and industrial areas in Iran as well as in different countries [17]. In addition to the possibility of contamination under the beds of reservoirs and pipelines, there is also a chance of contamination by transportations, road communication and road construction sites in such locations. Notably, heavy metal leakage causes bedrock instability and leaves detrimental impacts on the environment. Heavy metals in soils not only induce considerable alterations in resistance parameters, but also yield changes in the soil structure (especially fine-grained soils). However, few studies have been performed on changes in resistance parameters in soils contaminated with heavy metals. The presence of heavy metals in soils containing clay fine grains causes the water thickness of its double layer to decline and due to the presence of heavy metals, a flocculent structure forms in clay soils [18]. One of the important issues in flocculated structures is the possibility of high-pressure subsidence. Thus, it is important to study the behavior of soils contaminated with heavy metals that contain clay (generally charged fine grains). The use of heavy metal adsorbents is one of the appropriate methods to prevent

the spread of pollutants and to improve the behavioral performance of soils. Accordingly, the use of a combination of soil and adsorbent can be considered as one of the suitable methods for bedding reservoirs from which contaminants can leak. An important point in this regard is to study the structural changes and variations in soil resistance parameters [19].

Oni [20] studied the CFD Behavior of Transition Flow in Distinct Tubes of Miscellaneous Tape Insertions. They used three different tubes with with crossed-axes-circle-cut tape insert (C-C tube), plain tube with crossed-axes-triangle-cut tape insert (C-T tube), and plain tube with crossed-axes-ellipse-cut tape insert (C-E tube). Results showed that Reynolds number of C-T tubes is fewer than the other tubes [20]. In a study led by Nikhhah Nasab and Abdeh Keykha [21] on the geotechnical characteristics of clayey sand contaminated with lead(II), it was found that the presence of heavy metal cations in the mixture caused a reduction in the shear strength of sand and kaolinite samples. On the contrary, due to the shrinkage of diffuse double layer of bentonite in the presence of lead ion, shear strength of sand and bentonite mixtures increased. Also, the consolidation behavior of both the mixtures contained kaolinite and bentonite was decreased [21].

According to studies conducted by Shang [22] and given the lack of enough data to explore the effect of heavy metals and adsorbents on the geotechnical behavior of soils, the main question that arises is whether addition of adsorbents such as zeolite and rice husk ash (RHA) to soil contaminated with heavy metals such as lead and zinc can alter the resistance parameters of this soil. For this purpose, this paper will investigate the effect of the presence of heavy metals and adsorbents on the geotechnical behavior of clayey sandy soils. Thus, 20% kaolinite is used in combination with sand. A mineral adsorbent called zeolite and an organic adsorbent called rice husk ash (RHA) are also used. The reason for choosing zeolite is its high adsorption power [9]. Further, the impact of zeolite on soil resistance parameters is greater than that of other organic adsorbents (RHA, sawdust, and the like), since organic adsorbents decompose and lose their functionality within a short period. Additionally, lead and zinc pollutants are employed in this research as they have the largest impact on the environment and are present in most industrial effluents. Note that due to the presence of clay in the soil composition, the CU test is conducted in soils containing fine grains.

2. MATERIALS AND METHODS

2. 1. Sand In order to provide sand materials, Firoozkooh siliceous sand, called 161 sand, was used. This sand was exploited from mines located in

Firoozkooh, 130 km northeast of Tehran, and has a golden yellow color profile. The unified classification system categorizes it as poorly graded sand (SP). Due to non-plastic fine grains in this sand (about 2% of soil weight), the sand was washed using a 0.075 mm diameter sieve to remove all fine grains so that only sand grains would remain. The specifications of the sand and its granulation diagram are presented in Table 1 and Figure 1.

2. 2. Clay Fines There are two main factors such as the bond between the constituent of clay and double-layer water around particles that can affect clay behavior. As for the difference between the constituents and the arrangements of particles within both kaolinite and bentonite, considering the effects of clay mineral type on the characteristics of contaminated soil, in the present study two different clay minerals (with high and low plasticity) were used. For the low plasticity clay, kaolinite was used which gained from eastern Azerbaijan (Iran). The used kaolin is white powder. It is noteworthy that the main mineral of this type of soil is kaolinite. Also, for the providing the high plastic clay, Iran Barit Bentonite was purchased. In The properties and particle size distribution curves for kaolinite are shown in Table 2 and Figure 2, respectively.

2. 3. Zeolite With Iran's extensive resources of zeolite and by increasing exploitations of these resources

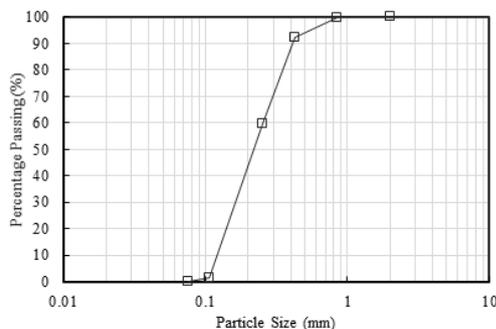


Figure 1. Grain size distribution curve for soil

TABLE 1. Physical and mechanical properties of sand

Property	Value
D_{50} (mm) (ASTM D2487, 2017) · Medium grain size	0.23
C_u (ASTM D2487, 2017) · Coefficient of uniformity	1.9
C_c (ASTM D2487, 2017) · Coefficient of curvature	0.89
G_s (ASTM D854, 2014) · Specific gravity	2.65
e_{min} (ASTM D4253, 2016) · Minimum void ratio	0.59
e_{max} (ASTM D4254, 2016) · Maximum void ratio	0.91
(ASTM D2487, 2017) USCS classification	SP

TABLE 2. Physical and mechanical compositions of the clay sample (kaolinite, montmorillonite)

Properties	Value (%)	
	Kaolinite	Montmorillonite
USCS	CL	CH
Plasticity index PI (%)	18.8	86.1
Plastic Limit PL (%)	31	53.9
Liquid Limit LL (%)	49.8	140
Specific Gravity	2.58	2.62

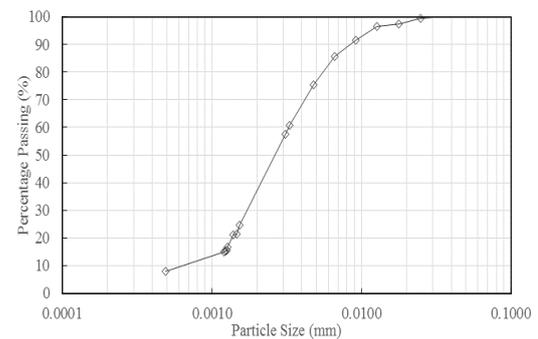


Figure 2. Particle size distribution curves for Kaolinite

in different regions, such as Semnan, this resource has considerable potential to remove heavy metals and even organic pollutants [3, 13]. Although zeolites (as adsorbents) can absorb a range of heavy metals, including lead, copper, zinc, cadmium, nickel, and silver, from contaminated water, their ability to absorb organic contaminants is limited [18]. In this study, to highlight the role of zeolite in the adsorption of heavy metals, zeolite under 1500 sieve has been used. This zeolite sample, which contains particles with diameters of less than 7 micrometers, is one of the newest products of Negin Powder Company of Semnan. Due to its tiny particles, it has greater absorption capacity as well as plasticity than the company's other products. The characteristics and granulation curve of consumable zeolite are outlined in Table 3 and Figure 3, respectively.

TABLE 3. Chemical and mechanical composition of Zeolite

Property	Value
Component	Zeolite
USCS classification	CL
Plasticity index-PI(%)	15.1
Plastic limit-PL(%)	25
Liquid limit-LL(%)	40.1
Specific gravity- G_s	2.58

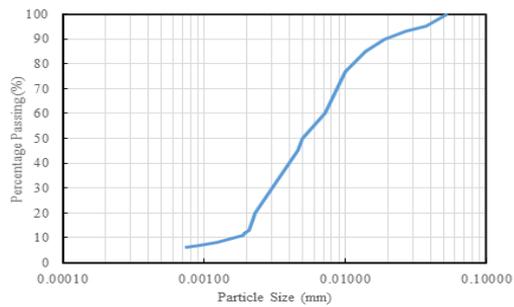


Figure 3. Particle size distribution curves for zeolite

The lattice structure in zeolite allows heavy metals in the solution to pass through infra-structural channels and adsorb reactive surfaces [11]. Figure 4 depicts an overview of the three dimensional structure of the natural zeolite [8].

2. 4. Rice Husk Ash Iran has vast resources of rice husks, with extensive lands, for instance, in Guilan, which are dedicated to cultivating this product. Thus, this resource can be widely used to remove environmental pollutants, including heavy metals and organic pollutants. The rice husk ash (RHA) utilized in this study is a Gilan Kesht product with more capabilities than the company's other products due to its fine particles. The chemical composition of RHA is shown in Table 4. For RHA to burn well and generate good results, the appropriate burning temperature is determined by X-ray diffraction and lime activity measurement tests, whose results are either crystalline or non-crystalline. It is also possible to identify the produced ash.

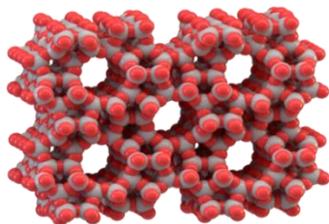


Figure 4. Three-dimensional view of zeolite particles

TABLE 4. Chemical characteristic of rice husk ash.

Component	Value (%)
Calcium oxide (CaO)	0.67
Silicon dioxide (SiO ₂)	88.32
Magnesium oxide (MgO)	0.44
Aluminium oxide (Al ₂ O ₃)	0.46
Iron oxide (Fe ₂ O ₃)	0.67
Sulfate (SO ₄)	-
Potassium/Sodium oxide	3.13

2. 5. Heavy Metals Lead and Zinc The chemical compounds of lead nitrate (Pb(NO₃)₂) and zinc nitrate (Zn(NO₃)₂) were used in this study to induce lead and zinc contamination. These chemical substances were imported from the German company Merck, with their specifications presented in Table 5. Lead nitrate and zinc nitrate both have purity levels exceeding 99.99%.

2. 5. Specimen Preparation and Experiments

In this paper, sampling involved sand combined with different clay minerals. Some compounds also contained RHA and zeolite. The names of the required compounds are given below, and the reasons for choosing these compounds and the percentages of clay minerals are discussed in the subsequent sections.

- Sand with 20% kaolinite (K20)
- Sand with 10% kaolinite and 10% zeolite (K10-Z10)
- Sand with 10% kaolinite and 10% RHA (K10-R10)
- Sand with 20% montmorillonite (M20)
- Sand with 10% montmorillonite and 10% zeolite (M10-Z10)
- Sand with 10% montmorillonite and 10% RHA (M10-R10)

A standard projector test as per ASTM D-698 was used to obtain the maximum specific gravity. Further, soil ether boundaries were calculated according to the ASTM D4318 standard. The values obtained from this test for soil have been expressed in both contaminated and non-contaminated conditions. Also, a static triangle device was installed to evaluate the impact of clay particles with low and high plastic properties, as well as the effect of zeolite and RHA adsorbents, on the static behavior of soils polluted with heavy metals under saturated conditions. Microstructural experiments determined the structure of the micro-results. Atomic absorption spectroscopy is an analytical technique for measuring the concentration of elements. It is so precise and sensitive that it can measure even minute quantities of contaminants per gram of the sample. In order to determine the concentration of lead and zinc adsorbed in

TABLE 5. Chemical characteristic of Lead (II) and Zinc nitrate

Properties	Value (%)	
	Lead (II) nitrate	Zinc nitrate
Assay (complexometric)	99.5>	99.3>
Insoluble matter	0.005<	0.03<
Cl (Chloride)	0.0005<	0.005<
Ca (Calcium)	0.005<	0.005<
Fe (Iron)	0.0003<	0.0002<
K (Potassium)	0.005<	0.0005<
Na (Sodium)	0.02<	0.001<
Pb (Lead)	-	0.006<

each of the desired compounds, the equilibrium test of soil saturation suspension was performed.

Since a material's microstructure is linked with its physical and mechanical properties, the study of structural changes is as important as resistance changes as it can reveal the origin of behavioral changes. The photography was done at Razi Metallurgical Research Center. In this research, the concentration of all elements in zeolite, montmorillonite, and kaolinite fine particles from sodium to uranium was determined using X-ray fluorescence (XRF) analysis based on the predominant cations used in cation exchange capacity specification. Further, the CEC of fine particles would serve as the basis of the overall technique in the study. Note that the concentration of various elements in the particulate matter was determined by Spectro XRF at Sharif University of Technology. A consolidation test was used to determine the speed and extent of soil compaction. Laboratory studies are often performed using reconstructed samples.

This research used wet compaction—one of the proposed methods in ASTM-D5311 standard—to make laboratory samples. This procedure is optimal for making compacted wet samples or soil samples in general. Because of the capillary effect and the formation of surface tension among soil grains, this sampling method is deemed beneficial for producing semi-dense and dense samples.

This study analyzed the behavior of two sets of samples: those contaminated with heavy metals and uncontaminated ones. As for non-contaminated samples, the dry weight of each compound's components (namely, sand, kaolinite, montmorillonite, zeolite, and RHA), with their percentages indicated, was first computed based on the dimensions of the mold and 95% of the maximum density of each compound. Different lead and zinc nitrate concentrations were added to each compound to contaminate the samples with heavy metals. First, lead nitrate or zinc nitrate was dissolved appropriately into water to prepare 1000, 2000, and 5000 ppm solutions. The concentrations were selected based on relevant studies reviewed. For contaminated samples, the optimal humidity in each composition comprised varying lead and zinc nitrate concentrations. After fabrication, the samples were processed for seven days at room temperature and inside a zipper bag. They were subsequently placed inside the unconfined or three-axial apparatus for testing once they had been made and processed.

3. ANALYSIS OF RESULTS

3. 1. Changes in the Concentration of Heavy Metals

In this paper, adsorbents of RHA and zeolite were used in the studied soil compositions. A batch analysis was

undertaken to determine the optimal quantity of adsorbents to be utilized with the soils examined in this study. The analysis was conducted by passing a solution containing 5,000 ppm of lead nitrate and zinc nitrate from primary soil samples through an atomic absorption apparatus and combining it with 2, 5, 7, 10, 12, and 15% of RHA and zeolite. In the original composition, a portion of the adsorbent was replaced with clay mineral, where the results were variations in the concentrations of lead nitrate and zinc nitrate in the soil effluent solution (Cf), as depicted in Figures 6-9. As shown, replacing RHA and zeolite adsorbents with kaolinite in the sand composition with 20% kaolinite clay significantly reduced lead and zinc nitrate concentrations in the output solution. In other words, this replacement significantly enhanced the absorption of contaminants by the soil composition. For example, according to Figure 6, replacing 15% of zeolite with kaolinite has boosted the adsorption of lead nitrate and zinc nitrate in kaolinite sand by 241.8 and 325.4%, respectively. The increased absorption rate of heavy metal pollutants by replacing zeolite with kaolinite can be attributed to the higher specific surface area of zeolite, and thus the ability to absorb more zeolite rather than kaolinite. Also, the reason for the higher adsorption of lead nitrate than zinc nitrate in the composition of primary kaolinite sand and all amounts of zeolite substitution is the greater tendency of divalent cations (Pb^{+2}) to exchange with monovalent cations (H^{+}) than cations (Zn^{+2}).

RHA has a significant porous surface. Thus, it has a considerable capacity to absorb and retain contaminants. When applied to the soil, it boosts the absorption of heavy metal pollutants. Thus, replacing 15% of RHA with kaolinite increased the absorption of lead nitrate and zinc nitrate in kaolinite sand by 228.8% and 291.6% (Figure 6). In addition, Figures 5 and 6 show that zeolite performed better in the adsorption of lead and zinc heavy metals in kaolinite-adsorbent clayey sand mixture than RHA. This better performance could be attributed to the specific surface area and the higher cation exchange capacity of zeolite than RHA. Figures 6 and 7 reveal that the composition of sand with 20% montmorillonite, due to its specific surface area and higher cation exchange capacity of montmorillonite than kaolinite, had a far higher capacity to adsorb heavy metals than the composition of sand with 20% kaolinite. For example, 4077 ppm lead nitrate was adsorbed by sand containing 20% montmorillonite. On the other hand, in the sand with 20% kaolinite, the adsorbed concentration was 850 ppm, representing 81.5% and 17% absorption of lead nitrate, respectively. Also, Figures 6 and 7 indicate that, unlike kaolinite sand, the replacement of RHA and zeolite adsorbents with montmorillonite in combination of sand with 20% montmorillonite reduced the absorption of lead and zinc nitrate by the soil composition. This indicated increased contamination in the output solution. For

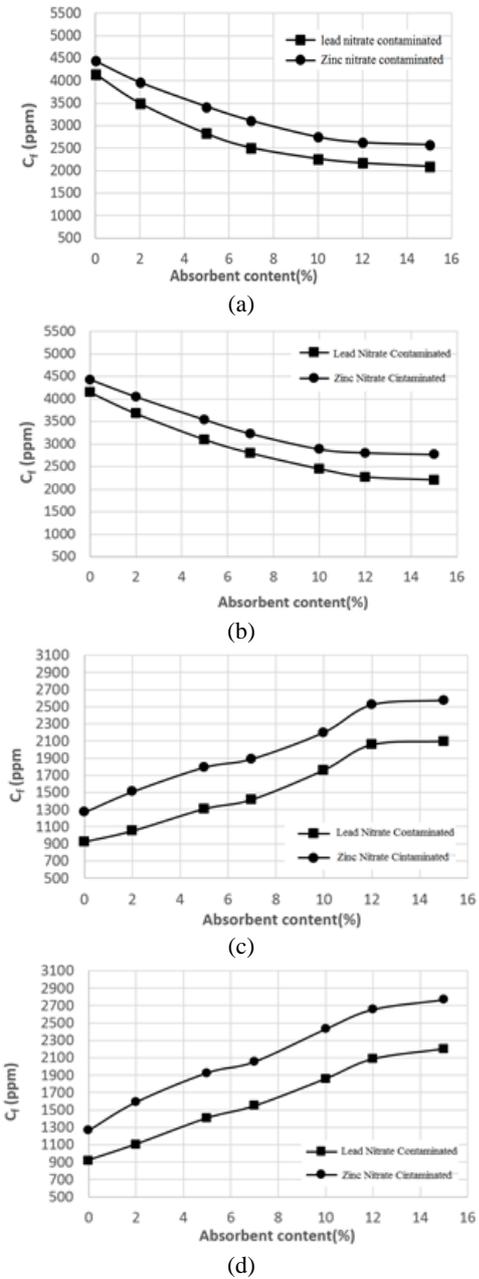


Figure 5. Heavy metal concentrations in extracted solution vs. absorbent content for: A) combination of kaolinite sand with different percentages of zeolite, B) combination of kaolinite sand with different percentages of rice bran ash, C) combination of montmorillonite sand with different percentages of zeolite, D) combination of montmorillonite sand with different percentages of rice bran ash

example, replacing 15% of zeolite with montmorillonite reduced lead nitrate and zinc nitrate adsorption in montmorillonite sand by 29.7% and 35.1%, respectively (Figure 7). This diminished absorption could be related to the specific surface area and the lower cation exchange capacity of zeolite than montmorillonite. The

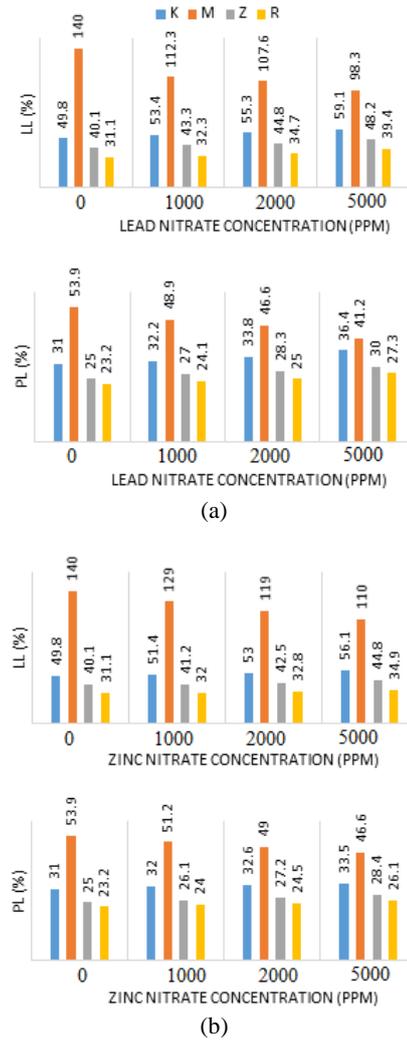


Figure 6. Liquid limit (LL) and plastic limit (PL), and plasticity index (PI) for a) contaminated by lead nitrate and b) contaminated by zinc-nitrate

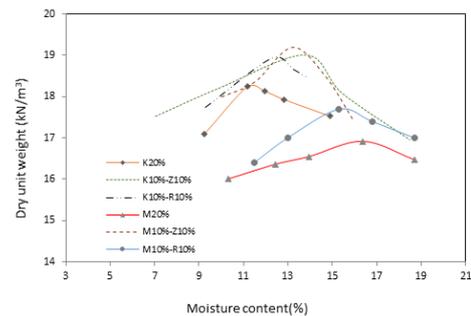


Figure 7. Dry density versus water content for various mixtures

replacement of 15% of RHA with montmorillonite lowered lead nitrate and zinc nitrate adsorption in montmorillonite sand by 31.4% and 40.2%, respectively

(Figure 8). Regarding RHA, it can be stated that despite its porous texture, which can absorb and retain contaminants, it reduces the absorption of heavy metal contaminants if replaced with montmorillonite due to its significantly lower cation exchange capacity than montmorillonite clay. Also, as with kaolinite sand, zeolite has a better performance in adsorption of lead and zinc in the sand-montmorillonite-adsorbent mixture than RHA due to its special surface area and higher cation exchange capacity.

3. 2. Determining the Atterberg Limits for Contaminated with Lead and Zinc Nitrate

In this paper, fine grains of kaolinite, montmorillonite, zeolite, and RHA were used. Due to the different plasticity of clay minerals in the study compounds, Atterberg limits tests, including liquid limit (LL), plastic limit (PL), and plasticity index (PI), were performed on these fine particles (Figure 6). In the non-contaminated state, montmorillonite had the highest liquid and plastic contents due to its higher specific surface area; it thus displayed a higher water absorption capacity than kaolinite. The significant plasticity of montmorillonite could be attributed to the high thickness of the double water layer (DDL) among the minerals. Lead nitrate and zinc nitrate were used as contaminants at concentrations of 1000, 2000, and 5000 ppm and were added to the desired soil composition. In order to investigate the effect of these two contaminants on the plastic properties of different fine-grained ether sheets, tests were performed on soil contaminants contaminated with these heavy metals. Adding lead nitrate to kaolinite would enhance liquid and plastic limits. The results indicated that in kaolinite, upon raising the concentration of lead nitrate to 5000 ppm, the liquid limit value increased from 49.8 to 59.1. The plastic limit also rose from 31 in the non-contaminated state to 36.4 in the contaminated state (5000 ppm).

The increment in the plastic limit was less than the increase in the liquid limit in contaminated kaolinite. In kaolinite, elevation of the concentration of heavy metal pollutants weakened the hydrogen bond between kaolinite particles due to replacing divalent cations of lead and zinc metals with monovalent hydrogen, which causes a slight increase in soil fluidity. The results indicated that, similar to kaolinite, raising the concentration of lead nitrate increased the liquid limit of zeolite and RHA, which was attributed to their minerals' dissimilar behavior toward heavy metal contaminants. For instance, in zeolite, the yield point and the plastic limit in the non-contaminated state were 40 and 25, respectively, and with the entry of lead nitrate at a concentration of 5000 ppm, the liquid limit and the plastic limit increased to 48 and 30, respectively. Note that the changes of Atterberg limits in zeolite and RHA were far less than in kaolinites, to which the greater

relative stability of these two fine grains than kaolinite could be attributed.

The paste properties of high plastic clay soils are primarily related to layer thickness (DDL). By increasing concentration of heavy metals and cation exchange would lower the soil pH, thus reducing the thickness of the double layer (DDL) in montmorillonite clay, as well as the liquid limit, plastic limit, and plasticity index of the montmorillonite. According to the results obtained in non-contaminated montmorillonite, the liquid limit and the plasticity limit were 140 and 53, respectively. Following the introduction of lead nitrate at its maximum concentration, the value of Atterberg limits decreased to 98 and 41, respectively. Also, lead nitrate was found to have a greater effect on increasing the liquid and plastic content of kaolinite, zeolite, and rice husk than zinc nitrate. This more significant effect could be attributed to the higher liquid limit of soils contaminated with lead nitrate than zinc nitrate contaminants can be considered stronger covalent bonds formed between lead cations (Pb^{2+}) than zinc cations (Zn^{2+}).

3. 3. Soil Consolidation Behavior

The sand density curve and its parameters with different fine grains (different percentages of montmorillonite, kaolinite, RHA, and zeolite) are shown in Figure 7 and Table 6, respectively. In order to explore the effect of clay mineral types on settlement changes as well as t_{50} and c_v parameters at different solidification pressures, the values of these parameters for combining sand with 20% kaolinite and montmorillonite were obtained from logarithm-logarithm curves of time at each solidification pressure.

As can be seen in Figure 8, the sedimentation rate of clayey-coated sand samples has grown with increasing compaction pressure, but the rate of increment has been higher, especially at high compaction pressures for montmorillonite sand. As displayed in Figure 8, the settlement rate of kaolinite specimens has grown with increasing compression pressure. This rise is more prominent in the montmorillonite specimens, especially at high compression pressures.

TABLE 6. Parameters obtained from compaction tests for sand with fine grains

Specimen	$\gamma_{d \max}$ (kN/m^3)	ω_{opt} (%)
K20	18.2	11.1
K10-Z10	19.05	13.8
K10-R10	17	16.5
M20	18.5	11.7
M10-Z10	18.88	12.6
M10-R10	19.17	13.2

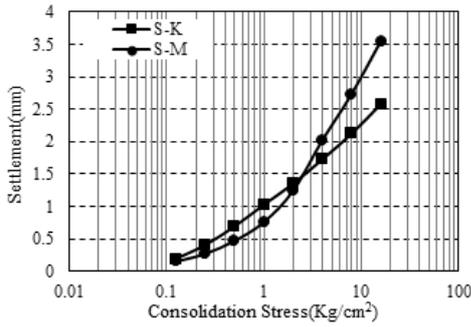


Figure 8. Sedimentation changes for different sand compositions with clay minerals at different stresses

As presented in Figure 9, the changes of t_{50} are different from the increment of stress for kaolinite and montmorillonite sands. In the combination of sand with 20% kaolinite, due to greater drainage and less water absorption as well as storage of kaolinite than montmorillonite, the time to reach 50% of the sample solidification is shorter at all solidification pressures and often within the first minutes of sample loading to reach 50%. In general, since 80% of the study compounds were sand, the values of t_{50} for sand compounds with 20% kaolinite and montmorillonite were very low. The value of the consolidation coefficient depends on several parameters such as t_{50} and the length of the drainage path, which is affected by the amount of sample subsidence at each consolidation stress. The values of the hardening exponent for kaolinite sand samples were higher than those of montmorillonite sand due to the lower settlement rate, which would increase the length of the drainage path and high drainage capacity, thus reducing the t_{50} value of the sample. In addition, the results of these tests are presented in Figures 10 and 11. They display the changes in the porosity ratio of the sample during the loading and unloading stages for sand with 20% kaolinite and 20% montmorillonite. As shown, the decline in the porosity ratio of the sample with the increase of consolidation stress was greater in the montmorillonite sand than in the kaolinite sand due to the higher amount of consolidation settlements. The compaction coefficient (C_c) and the swelling coefficient (C_s) are the important parameters that can be extracted from the porosity-logarithm of the reinforcement stress diagram. These parameters are very useful in identifying the behavior of clay soils and are equal to the slope. The smooth part (fixed slope) of the porosity-logarithm curve is the reinforcement stress and the slope of the bearing part of the curve. According to Figure 10, the values of compaction coefficient and the swelling coefficient for kaolinite sand are 0.113 and 0.004, respectively. Also, these values are obtained have been 0.194 and 0.03 for montmorillonite sand respectively, according to Figure 12.

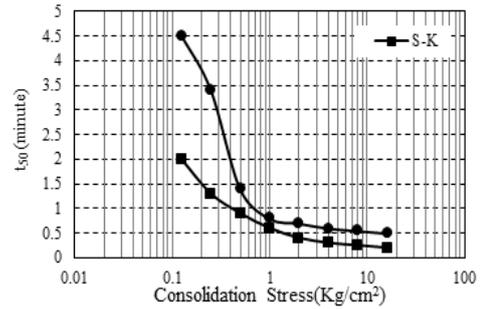


Figure 9. Changes of t_{50} for Specimen of clayey sand minerals at different stresses

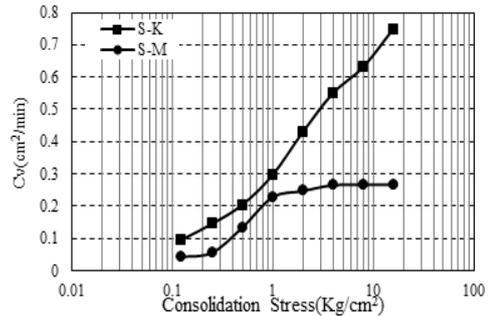


Figure 10. Variation coefficient for different combination of sand with clay minerals in different stress

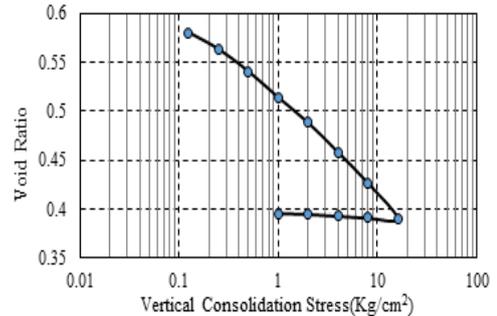


Figure 11. Changes in the porosity ratio of sand composition with 20% kaolinite in different stresses

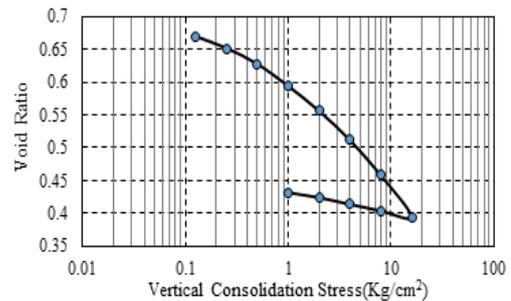


Figure 12. Changes in the porosity ratio of sand composition with 20% montmorillonite in different stresses

3. 4. Sand-clay-adsorbent Composition in Non-contaminated State

In this section, in order to investigate the effect of the presence of zeolite adsorbents and RHA on changes in the consolidation parameters of sand composition with clay minerals, an odometer consolidation experiment was performed.

As revealed in Figure 13, replacing zeolite with kaolinite enhanced the settlement of the kaolinite sand sample, which could be attributed to the higher specific surface area of zeolite and more significant ability to absorb water than kaolinite and increased settlements due to drainage of excess water pressure. Unlike zeolite, the addition of RHA elevated the settlement rate of kaolinized sand at low setting pressures due to its high porosity and the lower specific gravity of RHA than kaolinite.

Regarding changes in the time to reach 50% of the setting (t_{50}) (Figure 14), replacing zeolite with kaolinite increased the t_{50} of kaolinite sand. Also, replacing kaolinite with RHA reduced the t_{50} of the sample. Replacing the zeolite and rice husk adsorbents with montmorillonite in the montmorillonite sand composition reduced t_{50} .

The solidification coefficient (c_v) by replacing RHA and zeolite with clay mineralization in samples is indicated in Figure 15.

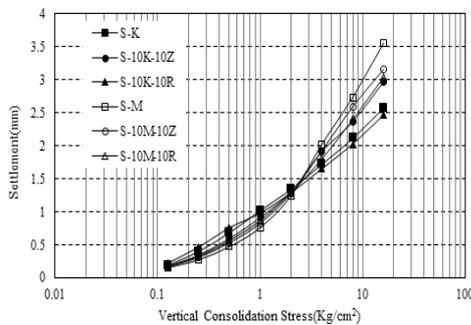


Figure 13. Sedimentation changes of different sand-clay-adsorbent specimens in non-contaminated state at different stresses

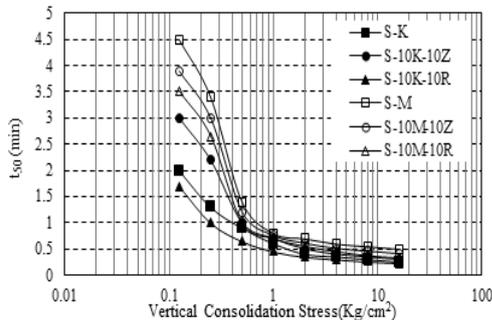


Figure 14. Changes of t_{50} for different sand-clay-adsorbent specimens in uncontaminated state at different stresses

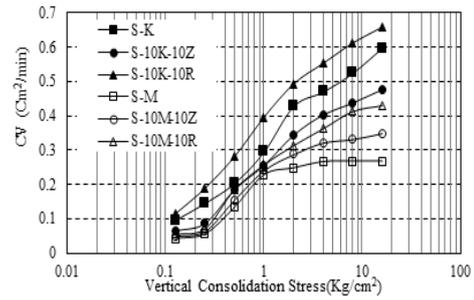


Figure 15. Consolidation coefficient (c_v) of different sand-clay-adsorbent specimens in non-contaminated state at different consolidation stresses

According to Figure 16, the replacement of RHA and zeolite lowered the swelling of the soil during the loading stage due to lower swelling properties of the adsorbents used than montmorillonite clay. It is believed that in the case of kaolinized sand, the use of adsorbents instead of kaolinite generally would have little effect on the swelling behavior of kaolinite sand.

3. 5. Sand-clay-adsorbent Composition in Contaminated State

This section investigates the audiometric solidification test results for different sand-clay-adsorbent compounds in both clean and contaminated states with different concentrations of lead nitrate and zinc nitrate in terms of variations in leakage parameters, solidification coefficient (c_v), compaction coefficient (C_c), and swelling coefficient (C_s).

As reported in Table 7, the contamination of all sand-kaolinite-adsorbent compounds with lead nitrate and zinc nitrate increased settlement and reduced the coefficient of consolidation at different reinforcement stresses, augmenting the compression coefficient and lowering the swelling coefficient. It was observed that the contamination of kaolinite sand with 5000 ppm zinc nitrate elevated the compaction coefficient by 13.5% and reduced the swelling coefficient by 15.9%. Elevation of the nitrate concentration of lead and zinc from 1000 to 5000 ppm increased the settlement increment rate at different reinforcement stresses and compaction

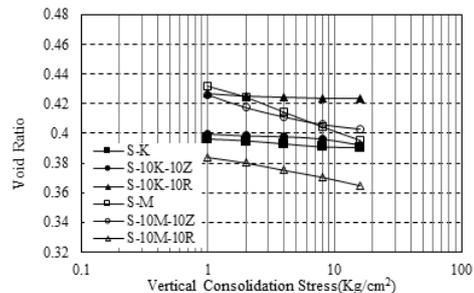


Figure 16. Changes in the porosity ratio of different sand-clay-adsorbent specimens in non-contaminated state

coefficients and accelerated the descending rate of swelling coefficient of sand-kaolinite-adsorbent compounds. The increased settlement in kaolinized sand with raising the concentration of heavy metals could be attributed to the breakdown of strong hydrogen bonds between kaolinite particles and creating a more porous and more structure settlement in the combination of sand

and kaolinite. Despite increasing the time to 50% solidification (t_{50}) by replacing the divalent cations of Pb^{2+} and Zn^{2+} with monovalent cation H^+ , increased settlement in kaolinite sand due to elevated nitrate concentration of lead and zinc contents augmented the consolidation coefficient at different consolidation stresses.

TABLE 7. Consolidation parameters of K20 Specimen in both clean and lead and zinc nitrate contaminated mixtures

Contaminant concentration (ppm)	Consolidation stress (kg/cm ²)	Lead II Nitrate				Zinc Nitrate			
		Settlement (mm)	C_v (cm ² /min)	$C_c \times 10^{-2}$	$C_s \times 10^{-2}$	Settlement (mm)	C_v (cm ² /min)	$C_c \times 10^{-2}$	$C_s \times 10^{-2}$
0	0.125	0.189	0.096			0.189	0.096		
	0.25	0.401	0.146			0.401	0.146		
	0.5	0.686	0.204			0.686	0.204		
	1	1.022	0.296			1.022	0.296		
	2	1.346	0.428	11.25	0.44	1.346	0.428	11.25	0.44
	4	1.725	0.470			1.725	0.470		
	8	2.119	0.525			2.119	0.525		
	16	2.578	0.598			2.578	0.598		
1000	0.125	0.193	0.092			0.199	0.095		
	0.25	0.410	0.145			0.418	0.150		
	0.5	0.702	0.204			0.723	0.209		
	1	0.046	0.285			1.088	0.293		
	2	1.396	0.426	11.46	0.44	1.438	0.437	11.63	0.43
	4	1.801	0.466			1.855	0.477		
	8	2.221	0.502			2.265	0.515		
	16	2.659	0.570			2.739	0.581		
2000	0.125	0.200	0.097			0.216	0.099		
	0.25	0.425	0.135			0.423	0.139		
	0.5	0.727	0.193			0.759	0.198		
	1	1.083	0.271			1.116	0.278		
	2	1.427	0.378	12.12	0.42	1.460	0.387	12.47	0.42
	4	1.869	0.405			1.925	0.414		
	8	2.316	0.481			2.372	0.493		
	16	2.793	0.540			2.857	0.552		
5000	0.125	0.217	0.084			0.224	0.086		
	0.25	0.461	0.125			0.470	0.129		
	0.5	0.789	0.182			0.813	0.187		
	1	1.175	0.249			1.222	0.256		
	2	1.568	0.372	12.58	0.39	1.615	0.381	12.77	0.37
	4	2.024	0.398			2.085	0.407		
	8	2.477	0.432			2.526	0.433		
	16	2.967	0.476			3.033	0.487		

The effect of zinc nitrate on the changes in the consolidation parameters of various sand-kaolinite-adsorbent compounds was found to be greater than that of lead nitrate: The contamination of sand composition with 10% kaolinite and 10% zeolite with 5000 ppm lead nitrate and nitrate on the compaction coefficient increased the sample by 7.3% and 10.6%, respectively.

3. 6. Shear Strength Parameters of Contaminated Soil

According to the previous sections, about heavy metal absorption and also according to reported data in literature [1-12], it can be concluded that the effect of lead nitrate on shear strength parameters is much higher than that of zinc nitrate. Accordingly, shear strength parameters in lead nitrate contaminated soil were investigated. In order to determine the shear strength parameters, series of consolidated undrained (CU) triaxial test was used. The triaxial device is shown in Figure 17.

In Figure 18, the ultimate shear strength changes for sand with 20% kaolinite in two contaminated and uncontaminated states are shown. Generally, the results show that in the different confining stresses at 1000 ppm concentration of lead nitrate in clayey sand, the amount of the ultimate shear strength compared to the uncontaminated state decreased approximately about 13%. Higher concentration of lead nitrate to 2000 ppm and 5000 ppm caused the ultimate shear strength in the contaminated samples to decrease about 23% and 30% respectively. Changes in shear strength parameters (cohesion and effective internal friction angle) for uncontaminated and contaminated samples are shown in Figure 19. In uncontaminated soil, the internal friction angle and cohesion are 37 degrees and 17 kPa, respectively. Changing the behavior of kaolinite in heavy metal contaminated soil, caused the cohesion to increase and the internal friction angle decrease.

Similar to the previous part, the ultimate strength at different concentrations of lead nitrate in the combination of sand with 20% bentonite is shown in Figure 20. The trend of ultimate shear strength of sand with 20%



Figure 17. Used triaxial device

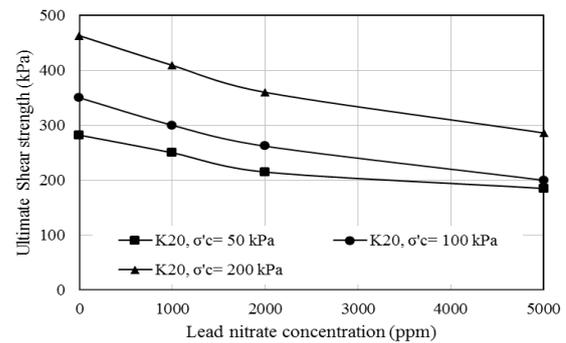


Figure 18. The changes of ultimate shear strength for composition of sand with 20% kaolinite

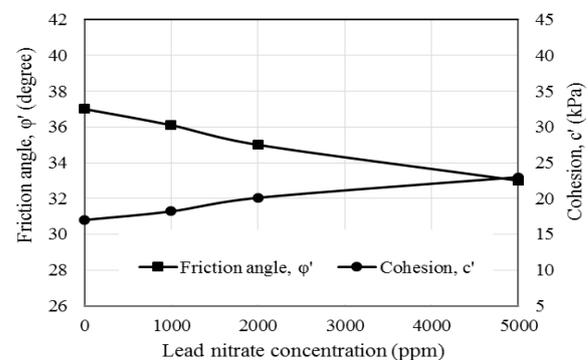


Figure 19. The changes in the internal friction angle and cohesion for composition of sand with 20% kaolinite

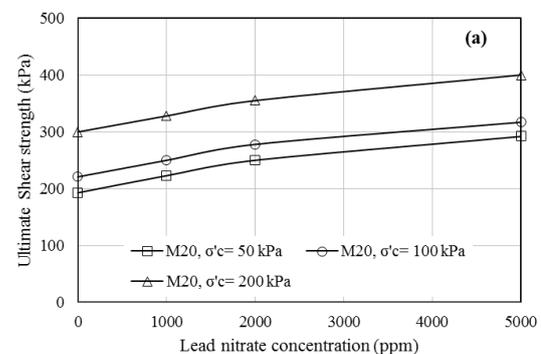


Figure 20. The changes of ultimate shear strength for composition of sand with 20% bentonite

bentonite showed the ultimate shear strength increased by changing of lead nitrate concentration from 0 to 1000 ppm about 11%. The further increase in the concentration to 5000 ppm caused the ultimate shear strength increase to 45. The changes of internal friction angle and cohesion for sand-bentonite samples in two contaminated and uncontaminated states are shown in Figure 21. Increasing the cations of heavy metal into the sand-bentonite mixtures, the structure of the mineral changed and the

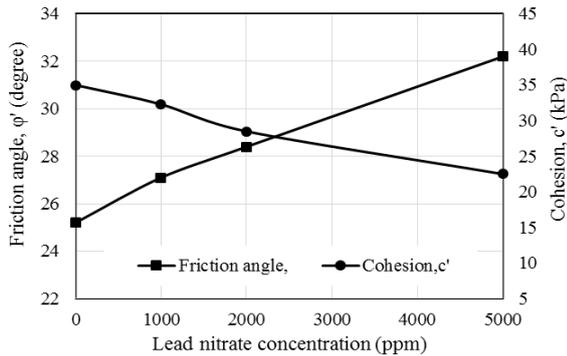


Figure 21. The changes in the internal friction angel and cohesion for composition of sand with 20% bentonite

behavior of the soil changed to flocculated from. Increasing the concentration up to 5000 ppm caused decrease in cohesion to 22 kPa. Although increasing the concentration of heavy metals in sand-bentonite mixtures caused reduction of the cohesion, but it caused the internal friction angle increase. The angle of internal friction for the sample with 20% bentonite is about 25.2 degree, which by increasing in concentration to 5000 ppm, caused the friction angle to raise about 25% compared to the uncontaminated state. It should be noted that the shear strength parameters (in both contaminated and uncontaminated state) are obtained based on Mohr's criteria.

3. 7. Shear Strength Parameters of Contaminated soil-adsorbent Mixture

According to the results provided in the field of absorption capacity of Zeolite is higher than rice husk ash, therefore, the shear strength parameters of the soil with zeolite in two contaminated and uncontaminated states are studied. It should be mentioned that the method in this research is replacing the adsorbent to fine-grained part. Accordingly, the amount of kaolinite or bentonite in the mixtures is decreased as the adsorbent content increased. Figure 22 shows shear strength of the sand- kaolinite composition with 10% zeolite. Comparing the results of the mixtures including 10% kaolinite and 10% zeolite shows that the replacement of zeolite with kaolinite has increased the ultimate strength in both contaminated and uncontaminated soils compared to the soil without adsorbent. Based on this, it can be concluded that the addition of zeolite, as well as increasing the absorption capacity in the soil, increase the ultimate shear strength. The strength value in the soils with kaolinite and zeolite has increased by about 50% compared to the soil without adsorbent in both contaminated and uncontaminated states.

The cohesion changes are shown in Figure 23. According to the results, in the sand- kaolinite and adsorbent composition, internal friction angle decreased

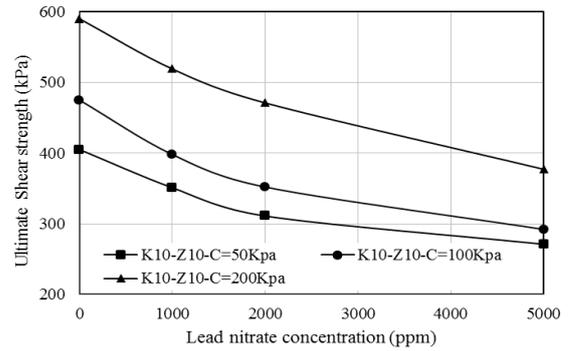


Figure 22. The changes of ultimate shear strength for composition of sand with 10% kaolinite and 10% zeolite

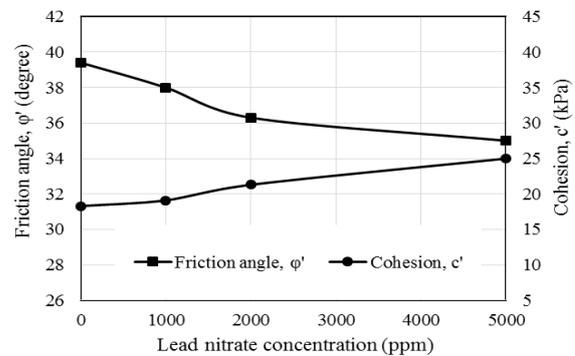


Figure 23. The changes in the internal friction angel and cohesion for composition of sand with 10% kaolinite and 10% zeolite

and cohesion increased, respectively. The amount of cohesion in the contaminated soil with concentrations of 1000, 2000, 5000 ppm, were 19.1, 22 and 25 kPa and the friction angles of soils were about 38, 36 and 34 degrees, respectively.

The ultimate shear strength ,which resulting from the triaxial test in different confining stresses, for the combination of bentonite sand with zeolite adsorbent are shown in Figure 24. Similar to the previous part, the amount of bentonite decrease and zeolite is replaced. It can be seen that by reducing the amount of bentonite and adding zeolite, the trend of changes in the shear strength in contaminated soil is different from the trend that mentioned for the mixtures of sand with 20% bentonite. Accordingly to results , shear strength of samples with adsorbent increase about 50% to 60% in the uncontaminated state compared to sand-bentonite mixtures. Therefore, according to the appropriate absorption capacity of zeolite, the combination of zeolite and bentonite, in addition to provide a suitable environmental composition, is also an ideal mixture in terms of shear strength parameters. Thus this combination is suitable from both environmental and geotechnical aspects. In 1000 ppm concentration, the

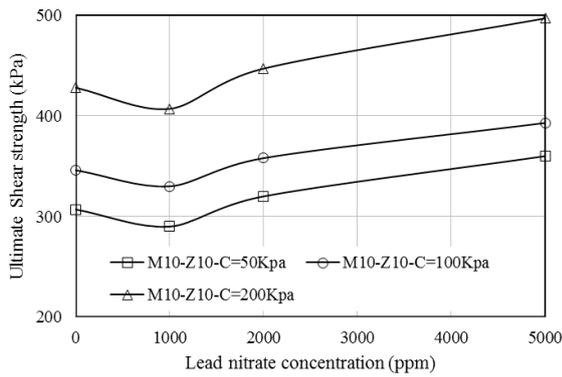


Figure 24. The changes of ultimate shear strength for composition of sand with 10% bentonite and 10% zeolite

ultimate shear strength in the sand-bentonite-absorbent mixtures decreases. One of the reasons is different reaction of two types of fine grains, bentonite and zeolite in contaminated case. Considering that the absorption capacity is high in both bentonite and zeolite minerals, therefore, at low concentrations, they almost neutralize each other, and it caused to shear strength decreases approximately 7 to 10%. By increasing the concentration of lead nitrate, the shear strength of sand-bentonite-zeolite composition increases. The results show that by increasing the concentration of lead nitrate to 5000 ppm, the ultimate shear strength has been increase by about 15-17% compared to the uncontaminated soil. The changes in cohesion and angle of friction are also shown in Figure 25. The cohesion has decreased and the internal friction angle has increased. With the increase of concentration from 1000 ppm to 2000 ppm, the amount of cohesion decreased by about 15%, while the internal friction angle also increased from 32 to 34.7 degree. By further increasing the concentration of lead nitrate to 5000 ppm in the sand with bentonite-zeolite, cohesion decreases to 17kPa, and internal friction angle increases to 19 degree.

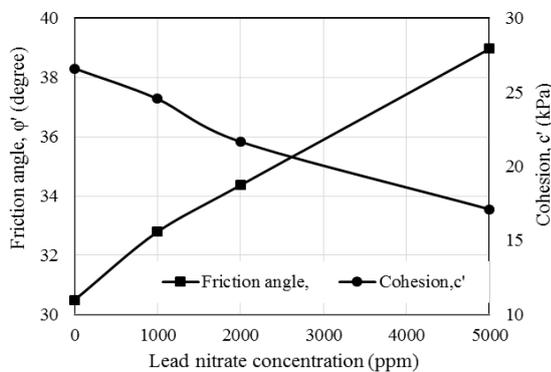
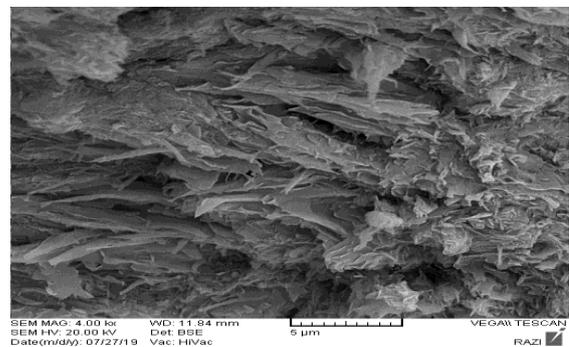


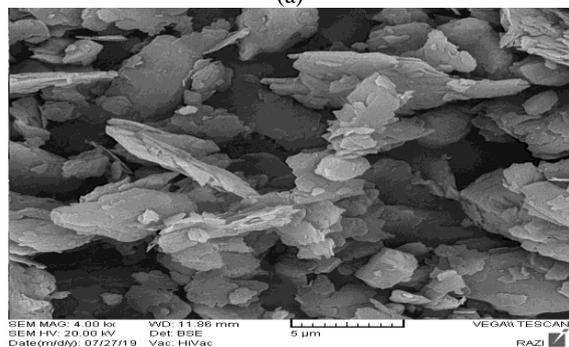
Figure 25. The changes in the internal friction angel and cohesion for composition of sand with 10% bentonite and 10% zeolite

3. 8. Microstructure of Contaminated Soil

To examine the microscopic structure of contaminated and uncontaminated soil, photographic images by using scanning electron microscope (SEM) analyses was conducted. SEM test is a standard test to determine the dispersion and flocculation structure of soils. In flocculated structure, there is strong attractive force between the particles giving a stable structure. While in dispersed structure, the repulsive forces between the particles or between the fabrics units are more. Because of the attractive force at the particle contacts, soils with flocculated structure possess better resistance to external loads and thus have high shear strength. In dispersed structure, the repulsive forces are dominant over attractive forces, thus the shear strength of soils is low and the thickness of DDL around the particle is large [23]. The shear behavior of bentonite is controlled by the DDL, while for kaolinite and zeolite, as the DDL expansion is almost absent, the shearing is controlled by interparticle contacts [24]. The scanning electron microscope (SEM) results for uncontaminated and contaminated kaolin and bentonite are shown in Figure 26. The results show that kaolinite in the uncontaminated state have flocculated structure while the bentonite structure is dispersed in the uncontaminated state. The bond between the kaolinite minerals are hydrogen and ion binding, which led to the formation of flocculated structure in uncontaminated state, while the bond between the bentonite particles attributed to weak Van



(a)



(b)

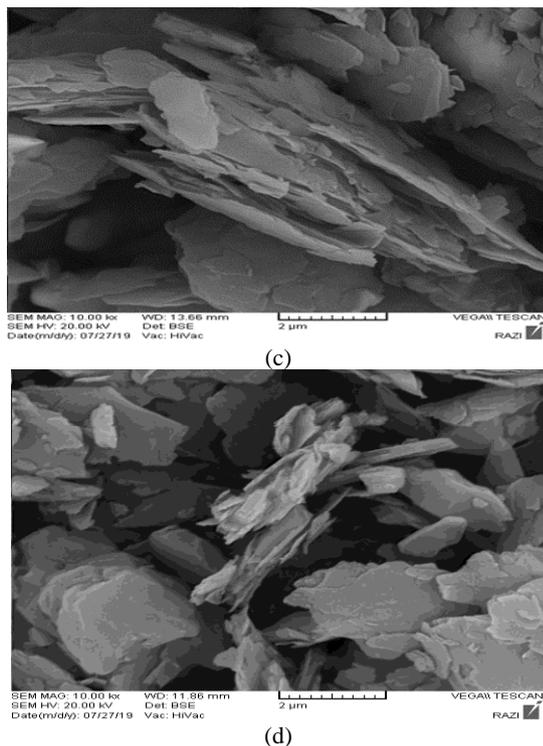


Figure 26. Structural changes for (a) uncontaminated and (b) contaminated bentonite, (c) uncontaminated and (d) contaminated kaolinite

Der Waals type and showed higher water absorption, which caused a dispersed structure in uncontaminated state. The results for kaolinite in the contaminated state show that an increase in the tendency of the soil to form a dispersed structure. The results for bentonite in both uncontaminated and contaminated states reveals that the flocculated structure is occurred in the case of contaminated state. In contaminated bentonite with lead or Zinc nitrate, cation exchange occurs between lead (Zn^{+2} or Pb^{+2}) and sodium (Na^{+}) ions and leading to reduce the thickness of the DDL and flocculate the clay particles. Reducing the thickness of the water double-layer forms a flocculated structure in the bentonite, which increases the shear resistance of the soil.

4. CONCLUSION

By the expansion of the industrial zones, heavy metals entered the environment through industrial wastewater or leakage from storage tanks, which contaminated the soil and groundwater. Heavy metal contamination caused significant changes in shear strength and soil structure. This study aimed to achieve optimal amounts of zeolite adsorbents and RHA to replace clay in the initial composition of the study soil and continue the laboratory study process based on the determined percentages. The optimum amounts of zeolite and RHA to replace the

kaolinite in combination with sand for lead nitrate contaminants were found to be 10%. With greater amounts, the process of reducing the concentration of contaminants in the output solution was almost constant. However, the amounts of zeolite and RHA for absorbing zinc nitrate contamination were 10%. Regarding montmorillonite sand, replacing the studied adsorbents with montmorillonite clay in all amounts lowered the adsorption of lead and zinc in the sand-clay montmorillonite-adsorbent composition. Identical optimal adsorbent amounts in the composition of kaolinite sand were used in the experiments on montmorillonite sand. The results also indicated that the rate of reduction of subsidence and the coefficient of consolidation in different consolidation stresses as well as the rate of reduction of the compression coefficient of the studied adsorbents were lower than those of montmorillonite sand. For instance, contamination of sand composition with 20% montmorillonite, sand composition with 10% montmorillonite and 10% zeolite, along with sand composition with 10% montmorillonite and 10% RHA with 5000 ppm zinc nitrate reduced the compression coefficient by 1.17, 0.16, and 2.15% respectively. This decline in the presence of adsorbents can be related to the lower cation exchange capacity and hence smaller double layer thickness (DDL) in the presence of adsorbents as well as lower sensitivity of RHA and zeolite to increasing cation concentrations.

Also, similar to kaolinite compounds, at all concentrations of lead and zinc heavy metals nitrate, the settlement values in reinforcement stresses were lower than 1 kg/cm^2 for montmorillonite-containing samples with RHA due to more porous and ductile ash texture. Also, the effect of zinc nitrate on reducing settlement, compaction coefficient, and swelling coefficient of different sand-montmorillonite-adsorbent compounds was more pronounced than lead nitrate. The contamination of the sand composition with 10% montmorillonite and 10% RHA with 5000 ppm lead nitrate and nitrate on the compaction coefficient reduced the sample by 13.1% and 15.2%, respectively. Due to the greater tendency of divalent cations (Zn^{2+}) to exchange with monovalent cations of montmorillonite than cations (Pb^{2+}) in soil structures, the adsorption of zinc nitrate was higher than that of lead nitrate in various montmorillonite-containing compounds.

In the composition of sand with 20% kaolinite, in the contaminated state with 1000 ppm and 2000 ppm of lead nitrate, the ultimate shear resistance value decreased on average by 13% and 23% compared to the uncontaminated state. Contrary to the behavior of sand with 20% kaolinite, in the composition of sand with 20% bentonite, the ultimate shear resistance increased by an average of 11% and 22% with the increasing of lead nitrate concentration from 0 to 1000 ppm and 2000 ppm. Also, the further increase of concentration to 5000 ppm

almost caused a 40-50% increase in the ultimate resistance. According to the results, replacing 10% of zeolite with kaolinite increasing the ultimate shear resistance in both contaminated and non-contaminated conditions up to 50% compared to the soil without absorbent. Thus, by adding zeolite, in addition to increasing the absorption capacity in the composition, shear resistance has increased. In uncontaminated state, by reducing the amount of bentonite and adding zeolite, the ultimate shear resistance increased about 50 to 60% compared to only bentonite composition. By considering the proper absorption capacity of zeolite, the combination of zeolite and bentonite, in addition to providing a combination with suitable environmental characteristics, is also an ideal combination in terms of resistance parameters, therefore, this combination is suitable from both environmental and geotechnical aspects.

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Persian Abstract

چکیده

ترکیب خاک با استفاده از جاذب‌ها باعث بهبود ساختار خاک شده و مقاومت برشی خاک را نیز تقویت می‌کند. از سویی دیگر می‌توان اینگونه تفسیر کرد که ترکیب خاک با جاذب، به شرط رعایت طرح اختلاط و نسبت بهینه، قدرت جذب آب خاک را افزایش داده و احتمال نفوذ و نشست آب‌های زیر زمینی حاوی مواد خطرناک را کاهش می‌دهد و از این طریق، مولفه‌های مقاومتی خاک را تقویت می‌کند. در این مقاله، تاثیر فلزات سنگین و جاذب‌ها بر روی رفتار ژئوتکنیکی خاک‌های ماسه-رسی حاوی نیترات سرب و روی به عنوان فلزات سنگین و زئولیت و خاکستر سیوس پسته برنج (پسماند کشاورزی) به عنوان جاذب، مورد مطالعه قرار گرفته است. نتایج نشان می‌دهد که جایگزین کردن خاکستر پسته برنج و زئولیت در ترکیب دانه‌بندی خاک با ۲۰ درصد رس کائولینیتی به شکل قابل توجهی منجر به کاهش نیترات روی و سرب می‌شود. همچنین، سطح متخلخل خاکستر پسته برنج به عنوان یک مزیت تاثیرگذار در افزایش جذب و تعدیل نشست آلودگی، می‌تواند در صورت جایگزینی با کائولینیت، قدرت جذب خاک را ارتقا دهد. جایگزین کردن ۱۵ درصد خاکستر پسته برنج با کائولینیت قدرت جذب نیترات‌های سرب و روی را به ترتیب ۲۲۸۸ و ۲۹۱.۶ درصد افزایش می‌دهد. شایان ذکر است که افزودن فلزات سنگین نیتراتی به کائولینیت حدود روانی و پلاستیک خاک را افزایش می‌دهد همچنین باعث تغییر پارامترهای تحکیم خاک می‌گردد بر اساس نتایج به دست آمده، با افزایش حضور فلزات سنگین به مقدار ۵۰۰۰ ppm مقدار حد روانی از ۴۹.۸ به ۵۹.۱ می‌رسد در حالی که، حد پلاستیک از ۳۱ (در حالت عدم حضور فلز سنگین) به ۳۶.۴ (در حضور ۵۰۰۰ ppm فلز سنگین) میل می‌کند.