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Hydraulic conductivity and removal rate of compacted clays permeated with landfill leachate

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ABSTRACT

Compacted clay soils are widely used as a barrier to protect environment from leachate migration. The suitability of clay soil for liner material, depends on resistibility to increase in hydraulic conductivity and contaminant transport. In this article, the influence of compaction energy and permeated leachate properties on the hydraulic conductivity were investigated. Natural attenuation capacities of clays compacted both standard and modified compaction methods were also evaluated. With this purpose, a series of laboratory tests were conducted with soil and real leachate samples obtained from Şile-Kömürcüoda Organized Landfill Site. DI and real leachate samples were percolated through the laboratory-scale column reactors that were filled with compacted clay samples prepared according to Standard and Modified Proctor method. During percolation, the hydraulic conductivity and natural attenuation capacity of the clay liner were determined by hydraulic conductivity calculation and chemical oxygen demand (COD), suspenden solids (SS), total Kjeldahl nitrogen (TKN), total phosphorus (TP) monitoring, respectively. According to the hydraulic conductivity measurements using leachate, it is shown that hydraulic conductivity decreased in both compacted clay prepared by Standard and Modified Proctor methods, possibly associated with biological and chemical clogging mechanisms. It is thought that clogging formed due to biofilm growth and/or suspended solids accumulation between the particles of the clay soil. When the variations of the COD, SS, TKN, and TP were examined, it was observed that the removal efficiency of the clay compacted by the modified compaction method was greater than the one compacted by the standard compaction method, especially for SS.

Keywords: Hydraulic conductivity; Landfill leachate; Compacted clay liner; Removal efficiency

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1. Introduction

Over 10,000 tonnes of municipal solid waste (MSW) are generated in Istanbul daily. The quantity of MSW gradually increases depending on the population growth. The problems such as leachate, landfill gas, and landfill area take place during the landfilling of wastes. Hence, disposal of MSW constitutes one of the most important environmental problems in

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Istanbul [1,2]. MSW landfills in Istanbul might receive a large amount of materials generated by the industrial facilities being the potential sources of hazardous contaminants in MSW landfills. Moreover, hazardous contaminants may originate from the small capacity generators of hazardous wastes, household hazardous wastes, and biological and/or chemical transformation products of placed wastes. Hence, landfill leachates represent a serious environmental concern with regard to trace priority pollutants introduced into the aquatic environment. Furthermore, landfills constitute a continuous source of atmospheric and groundwater pollution because of the uncontrolled degradation of the organic matter [2].

Because of the low hydraulic conductivity of clays, they are used with increasing frequency as liners or slurry walls to contain hazardous wastes and organic contaminants within fixed subsurface boundaries. The suitability of clay soil for liner material, depends on resistibility to increase in hydraulic conductivity and migration of contaminants. Hydraulic conductivity is the primary criteria to evaluate the suitability of clay soils for lining of the waste impoundments. According to the design criteria of Turkish Solid Waste Management Legislation, the liner component is compacted to achieve a hydraulic conductivity no greater than $10^{-8}\,\mathrm{m\,s^{-1}}$. But because clay liners are subject to change due to interaction with leachate, clay material characteristics are deteriorated and to obtain this value is getting harder for the *in situ* conditions. Therefore, transport of contaminants through synthetic and mineral liners remains the most serious long-term problem.

The mechanism of contaminant migration involves numerous transformation and transport processes such as advection, dispersion, diffusion, sorption, biodegradation or chemical transformation. Mass transport through clay soil occurs by the pressure driven movement of leachate (advection) and/or the concentration driven movement of contaminants (diffusion). In most cases, compacted clay liners minimize the advective flux of contaminants resulting in diffusion becoming the dominant contaminant transport mechanism [3].

The transport of contaminants through both natural and compacted clay has been examined by many researchers [3–23]. Xie et al. [11] investigated migration of leachate pollutant at a landfill after it had been operated for 13 years. Concentrations of chloride, chemical oxygen demand (COD), and the heavy metals in the soil samples were analiyzed. It is determined that the chloride was migrated to more than 10 m, while the maximum migration depth of COD varied between 1 and 3.5 m. This phenomenon was explained by the variation in diffusion rate and leachate–soil interaction. The chloride profiles also indicated that advection might be the dominant contaminant transport mechanisms at this site.

Anderson et al. [12] reported laboratory-measured values for the hydraulic conductivity (K_s) of different solvents and solutions, including neat acetic acid, aniline (a base), methanol (hydrophilic), and xylene (hydrophobic) through four clays (two smectites, a kaolinite, and an illite). For these solvents, the K_s was at least 10–100 times greater than that of water. Brown et al. [13] reported similar results for the flow of a paraffin oil, diesel fuel, gasoline, and motor oil through micaceous clay. In addition, the hydraulic conductivity of neat aniline, carbon tetrachloride, or acetic acid for a sand and bentonite slurry mixture was observed to exceed that of water by two to four orders of magnitude [14]. However, Acar et al. [15] reported that the hydraulic conductivity of neat acetone or phenol through a kaolin clay was greater by an order of magnitude or less, while that for neat benzene or nitrobenzene was less by three orders of magnitude.

The differences in the values of hydraulic conductivity reported in the previous studies for water and the solvents cannot be explained by the differences between the viscosities and densities of water and solvents. This is not surprising since the flow of the solvents was probably mainly through cracks in the clays and was not governed by the theory of flow through a porous medium. Furthermore, the difference in the experimental results for the flow of solvents through clays could be due to the type of permeameters used [15].

Daniel [19] determined the permeation of compacted clay soil with diluted, real–world organic waste liquids did not cause any significant effects on the hydraulic conductivity of compacted clay soil. The liquids were from chemical waste landfills and impoundments. Francisca and Glatstein [7] measured the long-term hydraulic conductivity of compacted silt–bentonite mixtures with distilled water, landfill leachate and nutrient solution. They found that the hydraulic conductivity decreased significantly with time when the permeating liquid-contained microorganisms, indicating that other mechanisms (i.e. pore clogging) were controlling the liquid displacement through the soil pores. Other studies [4,13,16,19,20] have indicated that pure, reagent-grade organic chemicals can cause large increases in hydraulic conductivity of compacted clay soil. Nayak et al. [4], determined a small reduction in maximum dry density and an increase in hydraulic conductivity due to leachate contamination. Chalermyanont et al. [22], assessed the potential of a lateritic soil and marine clay for use as a landfill liner material. Hydraulic conductivity test results indicated that the hydraulic conductivity increased as the concentration of the heavy metal species in solution increased.

The objective of this study was to investigate the effect of compaction energy on the hydraulic conductivity and contaminant transport. With respect to this objective, laboratory-scale column reactors were filled with compacted clay samples prepared in accordance with Standard and Modified Proctor method. DI and real leachate samples were percolated through reactors. Natural attenuation capacities of clays that were compacted with both Standard and Modified compaction methods were also evaluated.

Table 2
The chemical analysis of the clay soil used in Kömürcüoda Solid Waste Landfill [23]

Chemical analysis (%)	
SiO_2	51–54
Al_2O_3	27–29
Fe_2O_3	2.5–2.7
TiO_2	1.1–1.2
CaO	0.1–0.2
MgO	0.7–0.8
Na_2O	0.0–0.1
K_3O	2.7–2.9
SO_3	–

2. Materials and methods

2.1. Materials

The clay soil were sampled from Kömürcüoda Landfill Site which is situated in partially or totally abandoned mine quarry areas with damaged native soil surfaces. The site is in a slightly sloped valley covered with Neogene-aged layers of clay soil, sand, gravel, and coal lenses. The clay soil is chemically compatible with the fill area. Typical clay liners have been constructed with natural soils having low permeabilities and they have been built up with heavy soil compaction equipments or cylinders. The clay liner underlying domestic solid wastes stored in the Kömürcüoda solid waste landfill site is 60 cm's thick with a hydraulic conductivity coefficient varying from 1×10^{-5} to 1×10^{-7} m/s [24].

Şile-Kömürcüoda Landfill Site has been operated since 1995. The properties of the leachate and the clay soil were determined by different researchers. The results of the characterization studies which were conducted on the leachate and soil taken from the Şile-Kömürcüoda Landfill Site are presented in Tables 1 and 2.

The Şile-Kömürcüoda Landfill Site soil samples contain 68–71% kaolinite, 6–9% free quartz, 15–18% illite, and 2–5% others. Their color is brownish-gray. Kaolinite and illite have been considered to be true clay soil minerals. The soil samples have a coefficient of hydraulic conductivity $k = 1 \times 10^{-8}$ m/s, a discharge loss of 8.5–9%, and a water absorption of 0.2–0.4% [23].

2.2. Methods

Representative clay samples were obtained from Şile-Kömürcüoda Landfill Site of 1–2 m. depth. Column tests were conducted to determine the transport parameters of clay soil permeated with real leachate sample. After 24 h air-drying period, clay samples were subjected to compaction and hydraulic conductivity tests. Clay samples have been hydrated with DI water, compacted within a Proctor mold using standard or modified energies and then the hydraulic conductivities of the DI water and leachate have been investigated. Standard (ASTM D698/AASHTO T99) and Modified Proctor (ASTM D1557/AASHTO T180) methods were applied in the laboratory at different water contents using a mold of 0.102 m diameter and 0.117 m high to determine the maximum dry density and optimum moisture content (OMC) [26]. The

Table 1
Properties of the landfill leachate

Parameter/ date	pH	COD (mg/L)	BOD (mg/L)	TKN (mg/L)	Total P (mg/L)	SS (mg/L)	VSS (mg/L)
Calli et al. [25]	6.2–8.4	5,850–47,800 (20,700)	3,500–28,500 (12,200)	1,550–3,590 (2,510)	0.65–20.9 (9.8)	670–2,720 (2,170)	–
Tufekci et al. [23]	6.8	10,000	1,010	1,635	5	1,010	855
This study	6.7	13,526	6,235	2,876	7	1,283	987

height of the compacted clay soil was 110 mm (Fig. 1). The soil was constrained against swelling. The clay soil has been water-saturated under a 30 kPa pressure. Outlet was covered to prevent evaporation. The reactor tests have been performed by flowing the liquid through compacted specimens at a gradient i ($\Delta H/L$) of 26.3 and calculating the hydraulic conductivity when the volume of the effluent reached 100 ml [27,28].

Constant Head Tests, have been performed using the following equation to find the coefficient of hydraulic conductivity of the clay soil:

$$k = \frac{QL}{At(h_1 - h_2)}$$

where k is coefficient of hydraulic conductivity, cm/s, A is surface area of the specimen, cm^2, L is distance between the manometers, cm, ($h_1 - h_2$) is differential head across the sample, cm, Q is total discharge, cm^3/s, t is elapsed time, s.

In order to determine the natural attenuation capacity of the compacted clay, analyses (COD, suspenden solids (SS), total Kjeldahl nitrogen (TKN) and total phosphorus (TP)) have been performed according to Standard APHA Methods both in the influent and effluent of the continuous reactor [29]. These analyses have been conducted on the effluent samples when the effluent reached a volume of 100 ml. Scanning electron microscopy (SEM) analyses have been performed on the clean and contaminated soil samples.

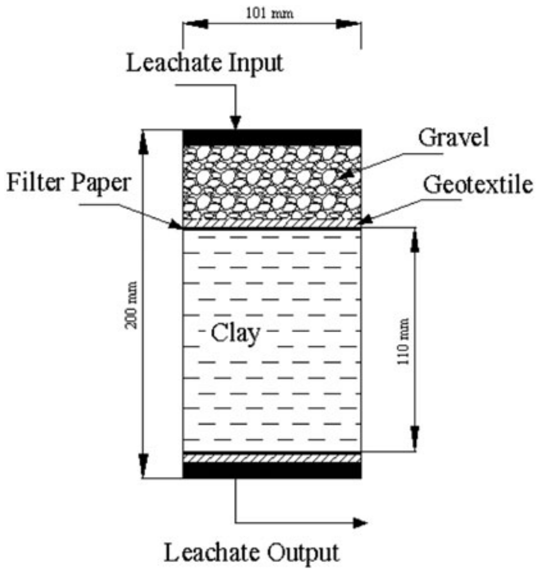

Fig. 1. Experimental setup.

3. Results and discussion

3.1. Hydraulic conductivity

Fig. 2 shows the OMCs determined by Standard and Modified Proctor methods. The hydraulic conductivity coefficients have been determined at the OMC and ±3% wet and dry side of this value. According to Fig. 2, at OMC, the hydraulic conductivity lower than the dry side as it expected. On the wet side of optimum, the permeability remained almost same as the hydraulic conductivity at optimum or increased slightly with increasing water content. It is not surprising that near optimum, hydraulic conductivity decrease, but on the wet side of optimum, it may increase or decrease, but generally remains within same order of magnitude as the hydraulic conductivity at optimum [30]. A slight decreasing trend was observed on the wet side of OMC in the case of leachate percolation through modified compacted soil.

When compacted clay permeated with DI water, the hydraulic conductivities of the clays were measured between $k = 5.2 \times 10^{-8}$ and 6.45×10^{-8} m/s. The hydraulic conductivity was decreased ($k = 2.53 \times 10^{-9}$ m/s) when compacted clay permeated with leachate, which clearly can be seen from Fig. 2. These findings are similar with Daniel [19] and Francisca and Glatstein [7]. They showed the permeation of compacted clay soil with leachate did not changed or reduced the hydraulic conductivity. Surprisingly, the difference between hydraulic conductivities of

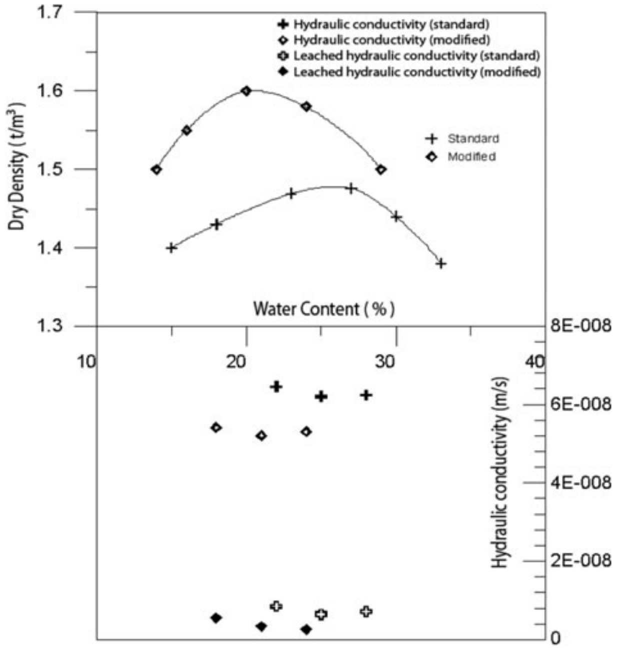

Fig. 2. Results of standard (opt.) and modified (opt.) proctor compaction and hydraulic conductivity.

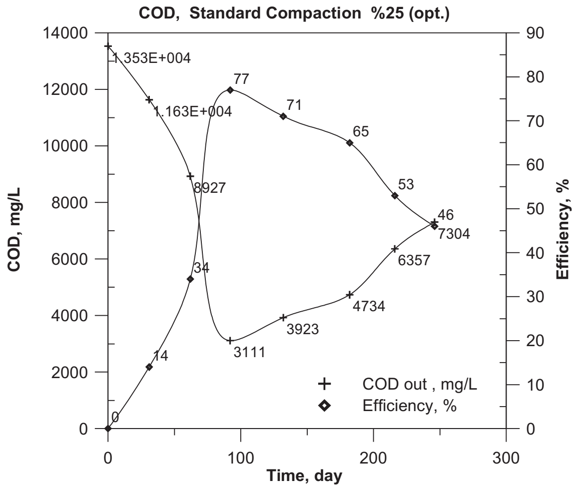

Fig. 3(a). Variation and the removal rate of COD (Reactor 1: standard compaction, water content 25% (opt.).)

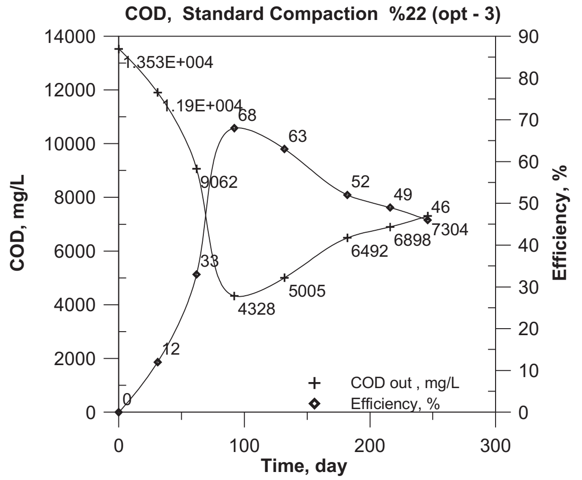

Fig. 3(b). Variation and the removal rate of COD (Reactor 2: standard compaction, water content 22% (opt. −3).)

compacted clay using Standard and Modified Proctor were reduced notably, when the leachate permeated. This phenomenon is explained by the chemical and physical interactions due to the contamination in the soil.

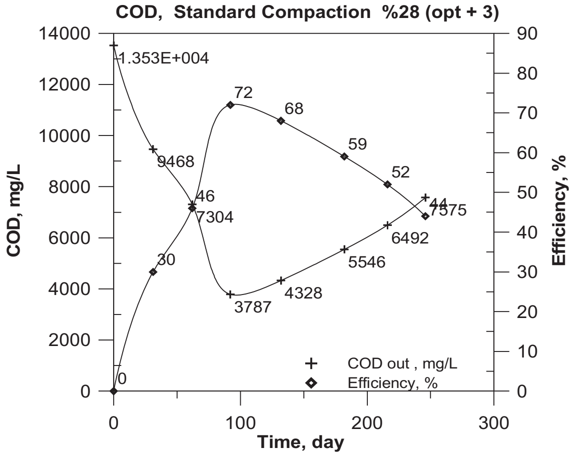

Fig. 3(c). Variation and the removal rate of COD (Reactor 3: standard compaction, water content 28% (opt. +3.)

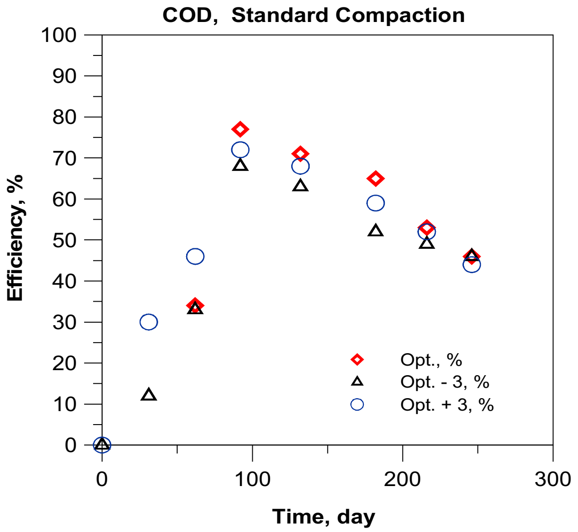

Fig. 3(d). Removal rate of COD (standard compaction).

3.2. Removal rate

The removal rate of clay soil for the COD, SS, TKN and TP has been investigated taking samples from the influent and effluent. Results are presented in Figs. 3–5. Fig. 3 shows the COD removal rates of compacted clay using standard proctor. The beginning COD value of the leachate has been measured as 13.526 mg/L. The first transition of the leachate through compacted clay took 31 days. For the reactor where the sample has been compacted with the water content of 25% OMC, in the day 31, COD removal efficiency of 14% with corresponding effluent COD value of 11.632 mg/l. In the day 62, COD removal efficiency of 34% with corresponding effluent COD value of 8.927 mg/L has been obtained. In the day 92, for the samples with the moisture contents of 25, 22, and 28%, COD effluent values of 3.111, 4.328, and 3.787 mg/L have been obtained. For these moisture contents, removal rates obtained as 77, 68, and 72%, respectively (Fig. 3(a) and (c)). As can be seen from Fig. 3, the removal rate of the COD increased up to the day 92 and afterward started to decrease.

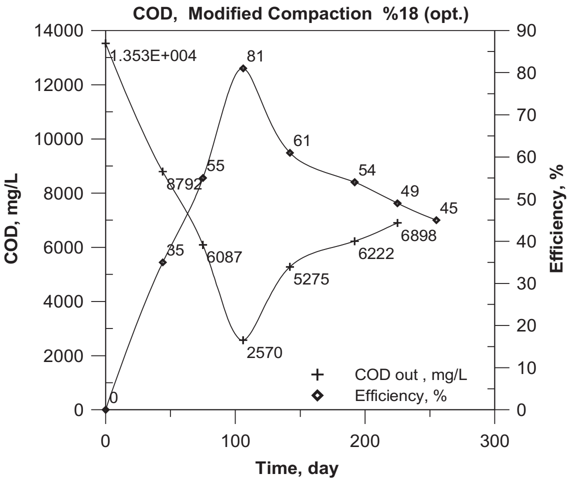

Fig. 4(a). Variation and removal rate of COD (Reactor 1: modified compaction, water content 18% (opt.).)

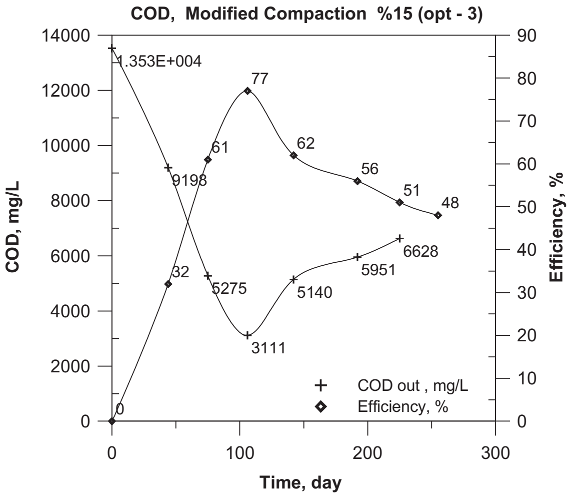

Fig. 4(b). Variation and removal rate of COD (Reactor 2: modified compaction, water content 15% (opt. −3).)

For the clay compacted with modified compaction, the first transition of the leachate through the clay soil has taken 44 days. Similarly, the removal rate of the COD increased up to the day 106 and afterwards started to decrease (Fig. 4(a) and (c)). This variation could be explained as natural attenuation processes for COD worked well up to the day 92 (standard compaction) and 106 (modified compaction), reduced afterward.

Removal rates of SS, TKN, and TP are given in Fig. 5. As can be seen from the figures, the removal rate generally showed similar trend for the SS, TKN, and TP. The removal rate of SS and TP increased up to the day 100 and afterward started to decrease as it seen for COD (see Fig. 5).

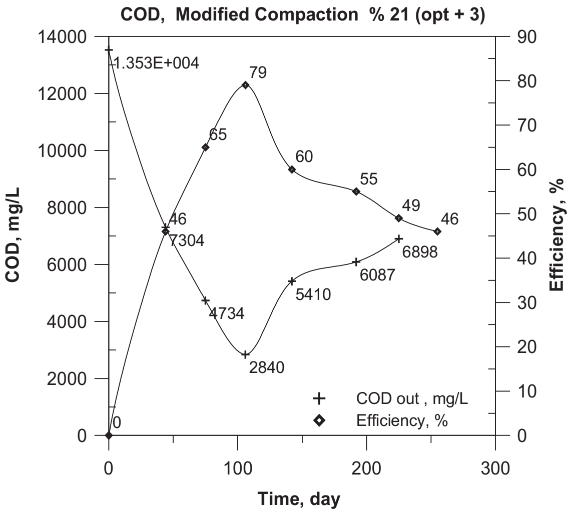

Fig. 4(c). Variation and removal rate of COD (Reactor 3: modified compaction, water content 21% (opt. +3).)

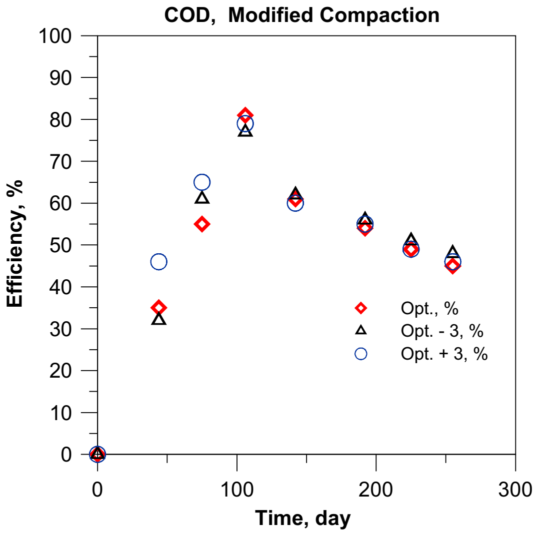

Fig. 4(d). Removal rate of COD (modified compaction).

As we know, when the dimensions of the suspended solid matters are greater than the mesh of the clay material, they are hold. On the other hand, as a result of the fact that some of the particles come into contact with each other during the leaching, greater flocs take shape; hence, the contaminants cannot pass through the layer and cannot interfere the effluent. Certain reactions may take place during the percolation; hence, the dissolved contaminative matters decompose, convert either to less hazardous components or to undecomposable components and move away from the water by sedimentation and adsorption. Interaction between soil and liquid phase is

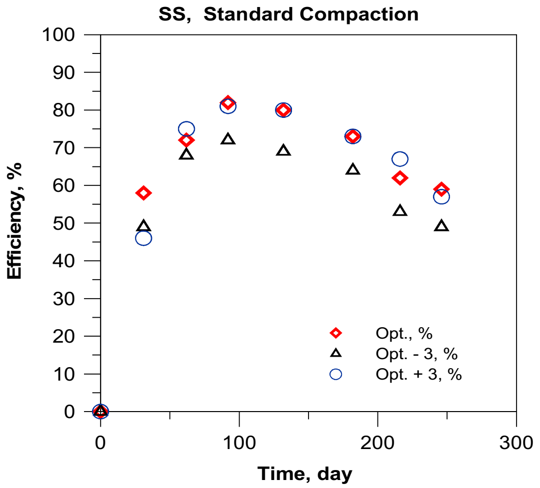

Fig. 5(a). Removal rate of SS (standard compaction).

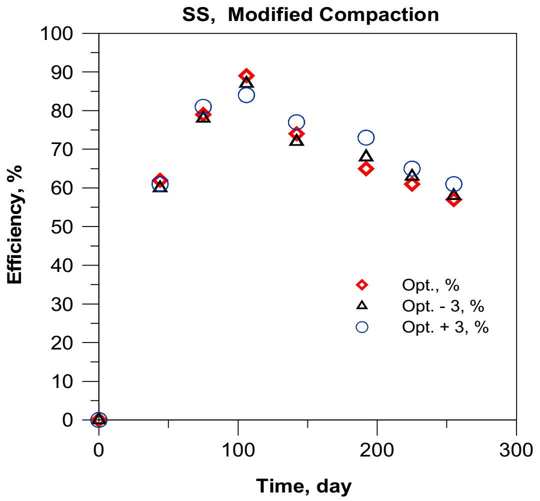

Fig. 5(b). Removal rate of SS (modified compaction).

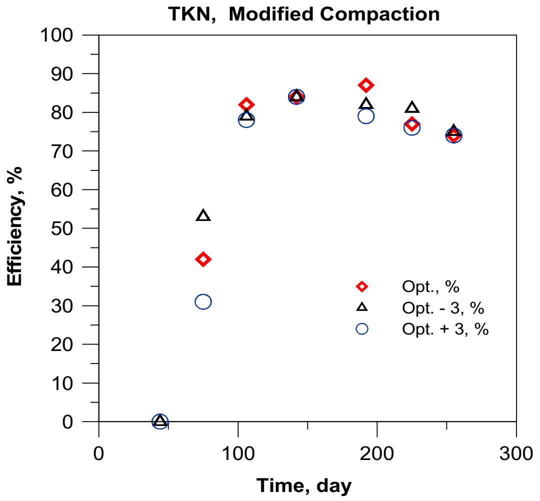

Fig. 5(c). Removal rate of TKN (standard compaction).

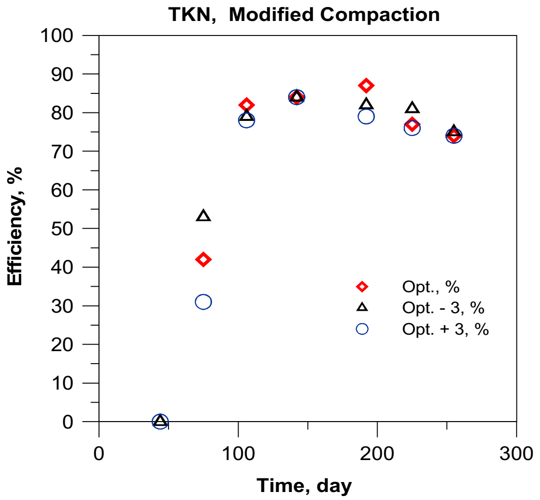

Fig. 5(d). Removal rate of TKN (modified compaction).

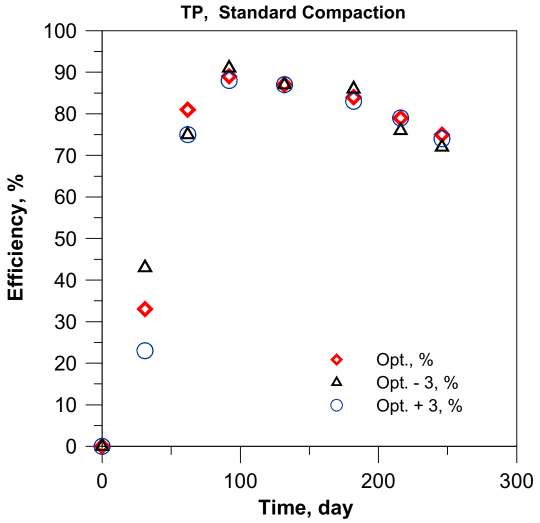

Fig. 5(e). Removal rate of TP (standard compaction).

responsible for ion exchange mechanisms and the development of the diffuse double layer around soil particles, which affects soil fabric and hydraulic conductivity [31]. Together with the fact that the leachate changes the coefficient of hydraulic conductivity, while it passes through the clay soil, these mechanisms can be effective in removing the contaminant of leachate.

As can be seen from Fig. 5(c) and (d), the trend of increase for TKN lasted to 132 days. Removal efficiency of TKN was not reduced significantly, possibly because of lasting transformation or adsorption. According to the results, the efficiency of natural attenuation processes for COD, SS, and TP, increased

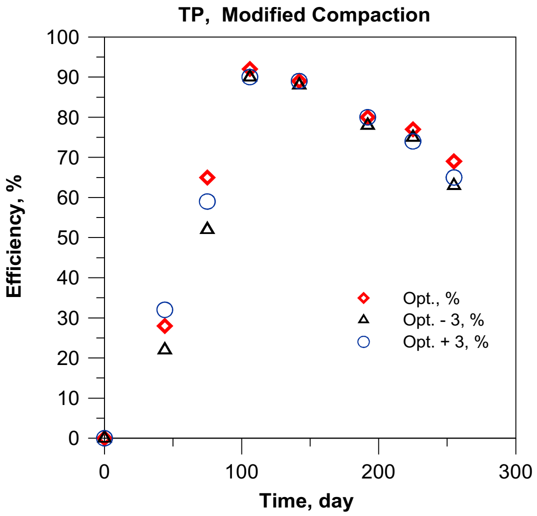

Fig. 5(f). Removal rate of TP (modified compaction.

until the day of 92nd, while this increasing trend in removal efficiency of TKN was lasted to 132nd day. This phenomenon can be explained by the decline of the hydraulic conductivity results from clogging, allows high natural attenuation process efficiency. Clogging was predominantly associated with the deposition of inorganic precipitates (mostly $CaCO_3$), but that the precipitation of $CaCO_3$ was linked to the biological processes and the reduction in COD [32].

The images obtained from the scanning electron microscope of the clean and contaminated clay specimens are presented below (see Figs. 6 and 7). It is evidently observed from the photographs that clean and contaminated clay specimens have different microstructure. It is worth noting that at the end of the experiment blackish brown color on the surface of the clay sample were observed. The SEM photos illustrate a more disaggregated structure for leachate-permeated clay samples. Hence, the decrease in hydraulic conductivity can be attributed to disaggregated structure. This finding may be the major cause of reduced hydraulic conductivity through pore clogging.

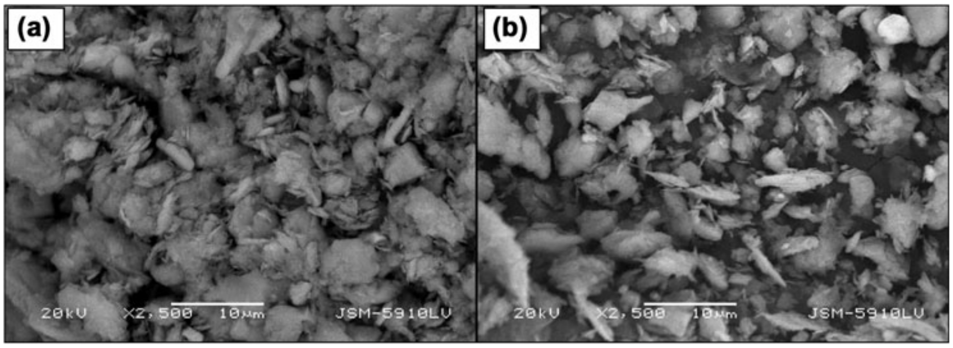

Fig. 6. (a) Clean clay standard compaction, water content 25% (opt.) and (b) contaminated clay standard compaction, water content 25% (opt.).

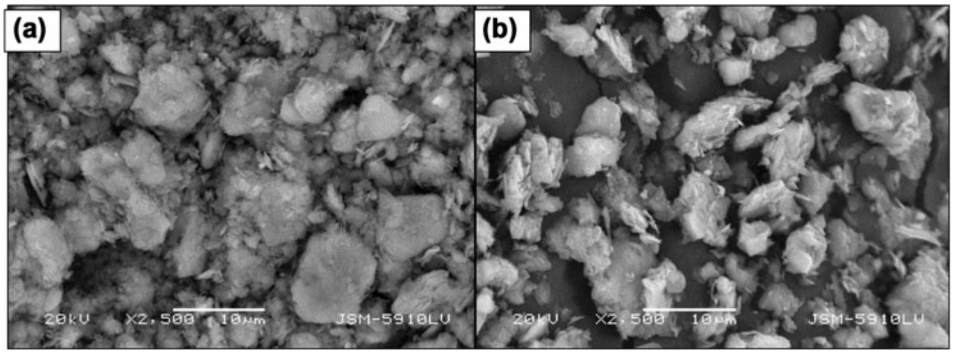

Fig. 7. (a) Clean clay modified compaction, water content 18% (opt.) and (b) contaminated clay modified compaction, water content 18% (opt.).

4. Conclusion

In this study, hydraulic conductivity and the removal capacity of the clay soil taken from the Şile Kömürcüoda Organized Landfill Area were investigated. In the specimens compacted with the standard methods, the first sample outflow took 31 days, whereas this period was observed to be 44 days for the samples compacted with the modified methods. When compacted clay permeated with DI water, the hydraulic conductivities of the clays were measured between $k = 5.2 \times 10^{-8}$ and 6.45×10^{-8} m/s. The hydraulic conductivity decreased ($k = 2.53 \times 10^{-9}$ m/s) when compacted clay permeated with leachate.

When the results of the tests for the COD, SS, TKN, and TP parameters were investigated, the removal rates of the soils compacted with the modified compaction method were observed to be greater than the ones compacted with the standard compaction method, especially for SS parameter.

As mentioned earlier, studied clay comprised mainly kaolinite and illite fraction which have different characteristics. Kaolinite is relatively stable against chemical attack from leachate, and it has small diffuse layer, low-specific surface area and low cation exchange capacity (CEC). However, due to the high-specific surface area of illite mineral, the diffuse layer occupies a large fraction of the pore space and is considered as part of the pore space. Illite has relatively high CEC and adequate adsorption [33]. Although it is not easy to come to a conclusion about the change in hydraulic conductivity and removal efficiencies according to SEM, it is considered that these changes may result from physical interaction of clay and leachate as it is known that main fraction–kaolinite is not sensitive to chemical interactions. This finding supports the reduced infiltration concept, possibly on account of pore clogging.

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