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Investigation of the Leachate Effect on Permeability and Geotechnical Characteristics of Fine-Grained Soil Modified Using Nanoclay–Nanofiber Composites

Mehdi Nikbakht [1,*], Fariba Behrooz Sarand [1,*], Rouzbeh Dabiri [1] and Masoud Hajialilue Bonab [2]

[1] Department of Civil Engineering, Islamic Azad University, Tabriz Branch, Tabriz 5157944533, Iran
[2] Department of Civil Engineering, University of Tabriz, Tabriz 5166616471, Iran
* Correspondence: mahdinikbakhteng@gmail.com (M.N.); sarand@iaut.ac.ir (F.B.S.)

Abstract: Using low permeability soils in landfill liners is a guaranteed leachate control and prevents leakage which causes environmental pollution. In this matter, the application of new technologies such as nano provides more capable filters that are used for reducing leachate pollutants and modifying the geotechnical properties of liners. The presented study attempted to conduct experimental research on nanoclay–nanofiber composite usage to control landfill liner permeability and observe its impact on the geotechnical characteristics of liners which provide a strong barrel for leachate leakage prevention and increase the liner durability for crack generations. In this regard, a total of 120 different geotechnical experiments were performed on mixed improved fine-grained soil samples which were categorized into four groups including nanoclay additives, nanofiber additives, nanocomposite additives, and control samples (without additives). According to the experimental results, permeability decreased, and geotechnical properties (e.g., Atterberg limits, unconfined compressive strength, cohesion, and friction) were increased with increasing nanocomposite content in the soil.

Keywords: nanoclay; nanofiber; environments; landfill liners; permeability

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1. Introduction

Landfilling is the final destination for solid waste and the main disposal method for controlling waste worldwide [1]. Landfills are used for waste management aims, such as storage, consolidation, and transfer, or processing of waste that is generated in cities, industries, medical sites, or chemical factories [2–4]. In the meantime, municipal landfills are the most extensive sanitary landfill type in the world which have a strong impact on environments and ecosystems [5,6]. Landfills have the potential to cause large-scale environmental pollution such as the contamination of groundwater and/or aquifers and/or soil contamination by leachate. Leachate contaminations lead to huge damage to different ecosystems and species [7]. Control of leachate generation and isolation in landfills is a great duty of liners and has an important impact on the environment and human health. Therefore, in a landfill's liner design, there was an attempt to use fine-grained soil with low permeability to prevent leachate immigration and groundwater contamination [8]. Clayey soils are widely used in landfill liners due to their low permeability and suitable geo-engineering characteristics [9]. Clayey soil (especially bentonite or montmorillonite) properties are directly related to their clay type, cation exchange capacity, dissolution precipitation, redox reactions, and chemical components which lead to increased durability and reduced permeability against leachate [10,11]. Clayey soils have received success in landfill liner designs with scientific and practical implementations. In general, the materials used in the liners must have very low permeability and high capacity to absorb pollutants, good flexibility against changes in humidity, settlements for landfill, and also these materials must have sufficient strength against forces and loads, which is mainly satisfied by clayey soils [12]. In several cases where the local clay used as a material in these coatings is not of sufficient quality and the transportation of clay with suitable conditions is costly, improving soil conditions by adding suitable additives or synthesizing existing materials is cost-effective and changes some properties, such as reducing the permeability coefficient and eliminating some problems in the soil [13]. Therefore, several scholars provide alternative procedures that help to improve the geo-engineering features as well as environmental properties [14–17].

Hermann et al. [18] investigated the hydraulic conductivity of fine-grained volcanic ash mixed with bentonite to control sewage sludge in Sweden. According to the results of the study, it appeared that the hydraulic conductivity of the volcanic ash mixture regarding the control of sewage sludge was reduced from 1.0×10^{-4} m/s to 1.7×10^{-11}. Kananizadeh et al. [19] performed experimental assessments on Kahrizak Landfill's dense clay liner in Tehran to analyze the permeability and swelling variations based on different amounts of nanoclay additives. The results of the study indicate that by adding 4% nanoclay, the permeability of soil changed from 3.66×10^{-9} to 7.74×10^{-11} in normal conditions. This variation was from 3.66×10^{-9} to 7.9×10^{-10} for the acidic condition and was from 3.25×10^{-9} to 5.24×10^{-10} for the alkali condition. Concerning this research, it can be mentioned that using nanoclay has a good impact on permeability. Debnath et al. [20] conducted a review study on nanomaterial and nanofiber applications in different aspects of the engineering field. The authors stated that in geotechnology, the application of nanomaterials and nanofibers provides a strong impact on improvements in the geotechnical characteristics of soils. Li et al. [21] examined the effect of leachate contamination on dense clay's mechanical properties which was utilized on clayey soils taken from a Wuhan metro site in China. Samples were used in different geotechnical tests with nanoclay additives after preparation in standard density tests with 95% density and 19.5% moisture. From the results of the assessment, it appeared that the nanoclays caused some improvements in clayey soil, such as hydraulic conductivity, density, durability, Atterberg limit, and unconfined compressive strength (UCS). Bahari et al. [22] used nanoclay to modify the soil structures for agriculture in Abbandan. Experiments were conducted on submerged mixed fine-grained soils with nanoclay additives. According to the results, soil permeability was reduced from 1.58×10^{-4} to 2.88×10^{-5} by adding 0.5% nanomontmorionite. Jafari and Abbasian [23,24] conducted studies on soil permeability reduction by using different amounts of nanoclay (montmorillonite) for leachate control generated from industrial facilities. Based on this research, it was shown that the permeability was reduced with the increase in nanomontmorionite rate. Derakhshani and Naghizadeh [25] state that the application of nanofibers (as inexpensive and non-toxic materials) can improve the geotechnical features of soils.

Almasri et al. [26] evaluated the performance of clay-based nanocomposite fiber prepared using a simple wet-moisture synthesis method. The researchers assessed the impact of groundwater contamination with arsenic and the role of the prepared nanocomposite in the inhibition and uptake of arsenic (As^{III}). Using pure montmorillonite and hydroxypyrene nanofiber led to an increased adsorption capacity of arsenic-contaminated groundwater. Dlamini et al. [27], by preparing mixed fibrous cellulose-based cellulosic composites, tried to analyze the performance of the water treatment of effluents. The researchers used clay-based nanofibers as ultrafiltration (UF) membranes to purify salt and distill salt water. To purify water from salt-containing effluents, used nanofiltration (NF) membranes conducting fiber-based nanocomposites (illite and montmorillonite) were mixed with soils. Qasaimeh et al. [28] investigated the effects of nanobentonite additives to reduce the environmental impact on fine soils. Referring to the results of the study, it can be stated that nanoclay 0.6% increased the soil strength up to 315 kPa and the potential for swelling was significantly reduced. Mehrabi et al. [29] conducted an experimental study on nanofiber's impact on material strength. Results of the study showed that nanofibers can improve the UCS and density significantly and it reduced permeability and porosity.

The presented article relying on the literature review results has tried to develop a new experimental method base on nanoclay–nanofiber composite application in landfill liners to reduce the permeability and improve the geo-engineering of fine-grained soil.

Most researchers are focused on nanoclay or nanofibers' effects on soils individually. The presented study tries to consider both impacts on the geotechnical aspect of landfill liner design.

2. Materials and Methods

2.1. Landfill Leachate

The study was performed on nanoclay–nanofiber composite application to improve the landfill liner performance against leachate leakage. The used study was obtained from the Tabriz Landfill site (Tabriz landfill site is +10 km from Spiran road, northwest of Tabriz) located in northwestern Iran. The leachate samples were taken and isolated in order to prevent changes in the leachate, then samples were transferred to the laboratory and chemical tests were performed on them. Table 1 illustrates the results of the chemical analysis for the studied leachate. To estimate the chemical composition of the Tabriz landfill leachate, several standard chemical laboratory tests were performed on the leachate samples that were taken from the landfill site, such as pH [30], thermometer, total dissolved solids or TDS meter, atomic absorption spectroscopy [31], high-performance liquid chromatography (HPLC), and X-ray fluorescence [32]. Additionally, the fine-grained soil samples were taken from the landfill site as well, which is used for mixed filter design. The soil samples were isolated as well and transferred into the geotechnical laboratory to conduct various geoengineering tests. These samples were divided into four different groups with each group having a specific mixture prepared for certain tests.

Table 1. Chemical composition of the Tabriz landfill leachate.

Samples	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**
pH	7.22	7.13	7.20	7.22	7.17	7.22	7.13	7.13	7.13
T (°C)	118.1	17.7	18.3	18.0	17.2	17.8	17.7	18.1	18.3
TDS (ppm)	496.12	498.33	492.25	489.36	496.11	490.45	486.70	450.12	476.42
As (pmm)	12.2	7.3	9.9	5.7	9.6	6.3	10.7	9.6	12.2
Cd (ppm)	1.10	1.14	1.06	1.03	0.98	1.17	1.02	0.97	0.97
Co (ppm)	47.6	45.4	47.3	45.6	47.3	45.5	47.3	47.3	45.6
Cr (ppm)	63.14	71.1	75.63	79.64	63.35	17.17	66.38	65.97	71.49
Cu (ppm)	96.83	96.85	97.10	96.37	97.45	96.17	96.75	97.58	96.19
Mn (ppm)	91.3	102.7	95.5	97.3	95.6	91.3	91.7	95.5	97.3
Ni (ppm)	87.30	87.42	85.96	87.74	87.35	86.91	87.50	87.63	87.33
Pb (ppm)	192.1	189.7	196.3	190.0	196.4	196.4	192.5	231.9	238.0
Zn (ppm)	234.7	248.2	239.7	237.0	237.0	235.4	239.1	231.9	238.0
Hg (ppm)	7.47	6.03	7.17	7.41	7.10	6.65	6.85	7.10	6.36
Ca (ppm)	65.71	67.77	65.63	97.12	65.45	97.36	65.22	67.47	65.60
Na (ppm)	15.9	12.5	15.4	14.9	15.2	12.9	17.2	14.9	12.5
Mg (ppm)	15.23	15.03	14.56	17.73	14.81	15.20	14.65	17.71	15.02
HCO_3^- (ppm)	25.41	27.33	25.45	27.12	25.63	27.92	25.40	25.45	27.12
Cl^- (ppm)	6.39	6.35	7.10	6.89	6.33	7.17	6.63	7.25	6.32
NO_3^- (ppm)	17.20	16.96	17.15	17.12	16.85	17.20	16.74	15.56	17.31
SO_4^{2-} (ppm)	18.85	18.63	18.74	18.52	18.25	19.12	18.78	18.45	19.02

2.2. Experimental Procedures

The samples soils taken from the Tabriz landfill site were prepared and categorized in the geotechnical laboratory in order to perform various geo-engineering tests which concluded particle size, hydrometry, permeability, Atterberg limits, UCS, and direct-shear tests. In this regard, the samples were prepared and we waited for 1, 7, 14, and 28 days before the testing stage. The tests were repeated for all groups 3 times and the average of the measurements was reported as the main value. This repetition helped to reduce technical and human errors during the tests. Some of these tests were conducted on the main soils (without any operations) such as particle size and hydrometry with a 1-day waiting duration. Figure 1 presents the grain size analysis result of the studied soil. Figure 2 provides the plasticity chart [33] for the main soil samples. According to this figure, the studied soil is classified as CL based on the unified soil classification system (USCS). CL stands for clayey soil with low plasticity.

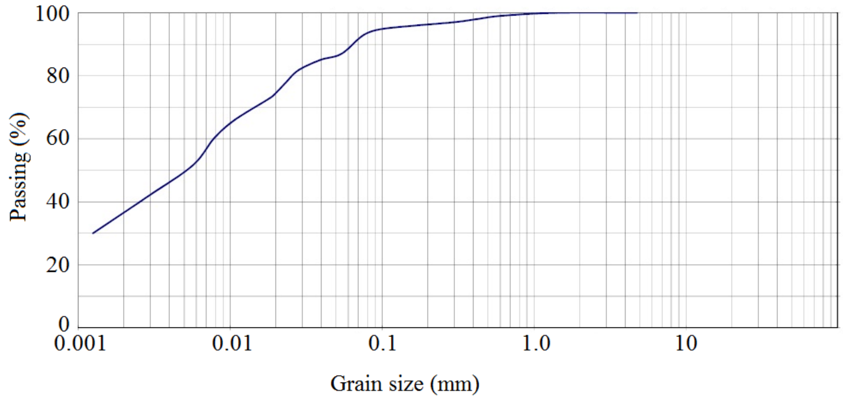

Figure 1. The Tabriz landfill soil's grain-size analysis result.

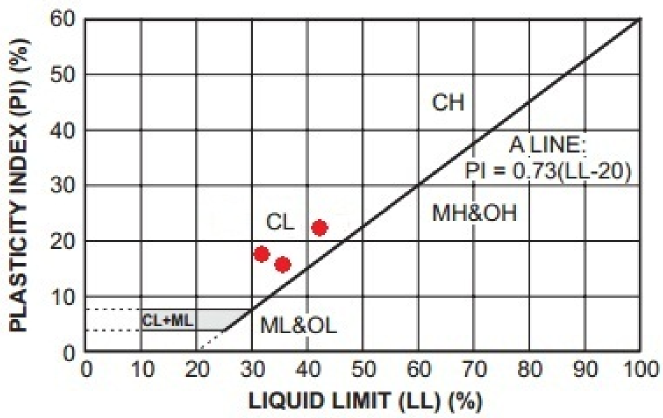

Figure 2. The plasticity chart for Tabriz landfill soil.

After providing the basic tests on main soils, which were conducted based on the American Society for Testing and Materials (ASTM) instructions [34–36], the samples were divided into four groups: concluded nanoclay additives, nanofiber additives, nanocomposite additives, and control samples (without additives), which reached a total of 120 soil samples. Each additive group was mixed with 3%, 6%, and 9% weight (%w) nanoparticles and prepared based on 1, 7, 14, and 28 days of waiting before testing. The aim of these preparations was investigating possible changes over time during periods of 1, 7, 14, and 28 days in isolated storage cells. If any changes were observed over time in the soil samples, it should be recorded. Each tests had specific preparation method that were recommended by ASTM and were described in their standards properly [33]. The control samples were used for observational purposes and each group was measured regarding the control group.

2.3. Nanoparticles Specifications

The nanoparticles used in this research are divided into nanoclay and nanofibers. Montmorillonite-type nanoclay was utilized in this study and the specification of the nanoclay is presented in Table 2. Nanomontmorillonite was chosen due to its high specific surface, special absorption ability, and compatibility with the environment. Additionally, the nanofibers used in this study are cellulose nanofibers (CNF), which were synthesized in the laboratory using the electrospinning method. Electrospinning is a fiber production method that uses electric force to draw charged threads of polymer solutions or polymer melts up to fiber diameters in the order of some hundred nanometers. Electrospinning shares characteristics of both electrospraying and the conventional solution dry spinning of fibers. The presented study used the dual-pump electrospinning machine from Fanavaran Nano-megyas to synthesize nanofibers. This device has a 60–50/V AC 240–100 H input power, 1000 watt 4 amp heater, and a HMI panel for ventilation. These materials were mixed with the tested soil in an ultrasonic mixer with a rotor speed of 25 rpm, for 24 h and were isolated in a dry state for 72 h. After these preparation steps, they were used in tests and sample preparations. Table 3 provides the CNF nanofiber specification used in this task. Figure 3 provides a view of the nanoclay and nanofiber that were used in this study. Nanomontmorillonite additives were prepared from Temad Kala/Nano Sadra Company with IDs Closite 15A/MJ-48 and synthesized cellulose nanofibers were also provided from Nanosani Service Company with IDs NS-CNF-001.

Table 2. Physical-chemical properties of the nano montmorillonite.

Parameter	Unit	Value
	Physical properties	
Clay type	-	Montmorillonite
Particle size	mn	1–2
Density	g/cm^3	0.5–0.7
Specific surface area	m^2/g	220–270
Electrical resistivity	MV	−25
Inter-particles distance	A°	60
Ion exchange coefficient	meg/100 g	48
Color	-	Pale yellow
Moisture	%	1–2
	Chemical properties	
Na_2O	%	0.98
MgO	%	3.29
Al_2O_3	%	19.60
SiO_2	%	50.95
K_2O	%	0.68
CaO	%	1.97
TiO_2	%	0.62
Fe_2O_3	%	5.62
LOI	%	15.45

Table 3. The index properties of the CNF nanofiber.

Parameter	Unit	Value
Fiber type	-	Carbon nanofiber
Solids	w%	Aqueous gel (3.0) + dry powder (98)
Fiber dimensions	nm	50
Surface property	m^2/g (BET)	31–33 (Hydrophilic)
Density	g/cm^3	Aqueous gel (1.0) + dry powder (1.5)
Fiber length	A°	4.5
Specific surface area	m^2/g	149.5
Electrical resistivity	MV	−253
Color	-	Pale yellow
Moisture	%	1
Yield of fibrillation	%	91.25
Transmittance at 600 nm	%	65
Water Retention	g/g	4.8

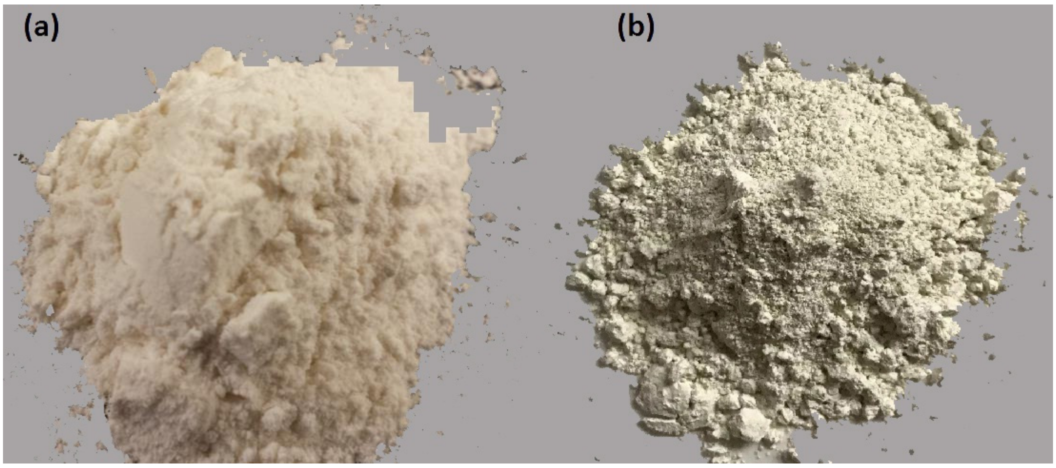

Figure 3. A view of (**a**) nanofiber and (**b**) nanoclay applied in this study.

2.4. Permeability Analysis

The permeability tests were performed on selected samples from the mentioned groups based on ASTM instructions [37]. In this regard, the samples after preparations were filled with compacted soil mixed with nanofiber and nanoclay particles. The cell was installed in a standard constant-head permeability test device, and to avoid the washed-away problem, porous papers were used at the top and bottom of the cell. Additionally, cleaned gravel was used in the top of the cell in a thin layer in order to prevent swelling and help the flow of leachate in the cell. Figure 4 illustrates the scheme of the permeability tests conducted on the different group of mixtures. The leachate was collected in the output point after passing through the filters and samples which were used for the calculation of permeability by using the following equation [33]:

$$k = \frac{QL}{At(h_1 - h_2)} \quad (1)$$

where k is permeability (cm/s), Q is the total discharge (cm^3/s), L is the distance between the manometers (cm), A is surface area (cm^2), t is the elapsed time (s), and $h_1 - h_2$ are the leachate head in cell columns. The results of the permeability variations were controled by the observational group and reported accordingly.

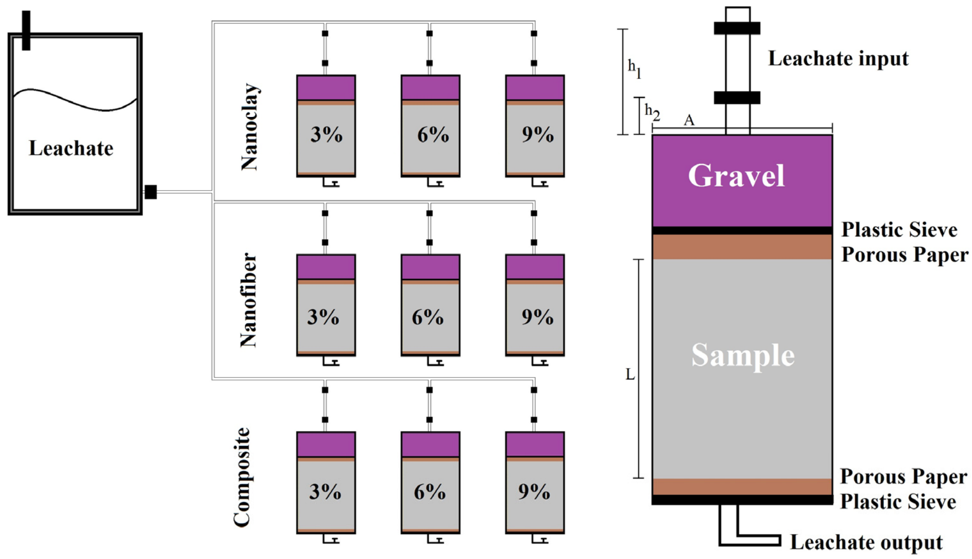

Figure 4. A Scheme of the permeability tests.

2.5. Geotechnical Investigations

To investigate the geo-engineering characteristics of the studied soil in both improved (with additives) and non-improved (without additives) cases, comprehensive experiments were conducted on selected samples. Each test was repeated three times and the average value was considered as the main parameter. In this regard, the Atterberg limits [36], unconfined compressive strength, UCS [38], and direct-shear [39] tests were conducted on samples according to the ASTM regulations [40]. Atterberg limits were used to investigate the plasticity of the soil behavior analysis and estimate the modifications on liquid and plastic limits of the soil with respect to the controlling samples. The UCS and direct-shear tests were used to investigate the soil strength parameters. The geo-engineering characteristics after estimation for each group, and the variation of the measured parameters (e.g., UCS, cohesion, c, friction, φ) were obtained and presented.

3. Results and Discussion

For the preparation of nanoclay–nanofiber-composite-added samples, the selected samples are divided into groups where each group had a specific amount of nanoclay (3%w, 6%w, and 9%w), nanofiber (3%w, 6%w, and 9%w) and composite (3%w, 6%w, and 9%w). The samples of the group during the preparation stage were mixed using a ultrasonic homogenizer sonicator cell disruptor mixer to reach the homogeny mixtures. After the preparation stage, samples were tested and results were reported. There were several types of geotechnical experiments conducted on specimens that represented the permeability and geo-engineering characteristics. Figure 5 shows the permeability results of different groups of samples. According to this figure, permeability was reduced by using nanoclay particles from 4.25×10^{-6} cm/s to 5.25×10^{-8} cm/s with an increasing nanoclay quantity from 0% to 9%. This amount for nanofiber became 3.24×10^{-8} cm/s which was less than nanoclay additives. This event indicated that the nanoclay is more suitable for permeability reduction on landfill liners individually, but paying attention to the changes in the curve of composite samples reveals an interesting point. The nanoclay–nanofiber composite reached 6.34×10^{-9} cm/s permeability. Therefore, the application of the composite form of the nanoparticles is more appropriate than using them separately. This reduction in permeability can be attributed to the optimal design of the landfill liners. Figures 5–11 provide the main compression which is targeted in this study. The aim of the study is understanding the variations of nanoclay, nanofiber, and composite (clay-fiber) for the controlling of permeability, plasticity as well as UCS, C, and internal friction. The first group represented the soil physical properties and the second group was responsible for the strength properties.

The Atterberg limits are a basic measure of the critical water contents of fine-grained soil which represents the plastic behavior of the soil. Improving this behavior can increase the durability of the soil structures. Figures 6–8 show the Atterberg limit results measured for different groups of the samples which are modified via nanoclay, nanofiber, and nanoclay–nanofiber composite. Each group was mixed with 3%, 6%, and 9% of sample weight (%w) additive. According to these figures, the concluded Atterberg limits show the different path of variations in liquid limit (LL), plastic limit (PL), and plasticity index (PI). Generally, the Atterberg limits were increased with the increasing in the nanoparticle additives, but these increases occurred with different slope angles. The nanoclay–nanofiber composites increased with a lower slope compared to nanoclays. The nanofibers acted independently of nanoclays and it can be mentioned that the nanoclays are normative responsible for the plastic behavior of the soil. Based on the results of the study, LL was changed from 37 in the control group to 54 for the 28-day mixed nanoclay. It happened for nanofiber and composite becoming 67 and 75, respectively. PL and PI also increased with nanoparticle additives. In the nanoclay group, PL and PI changed from 20.67 and 16.33 to 29.67 and 25.34. This variation for the nanofiber group was up to 35.67 and 31.34; for the composite group it was 45 and 30.

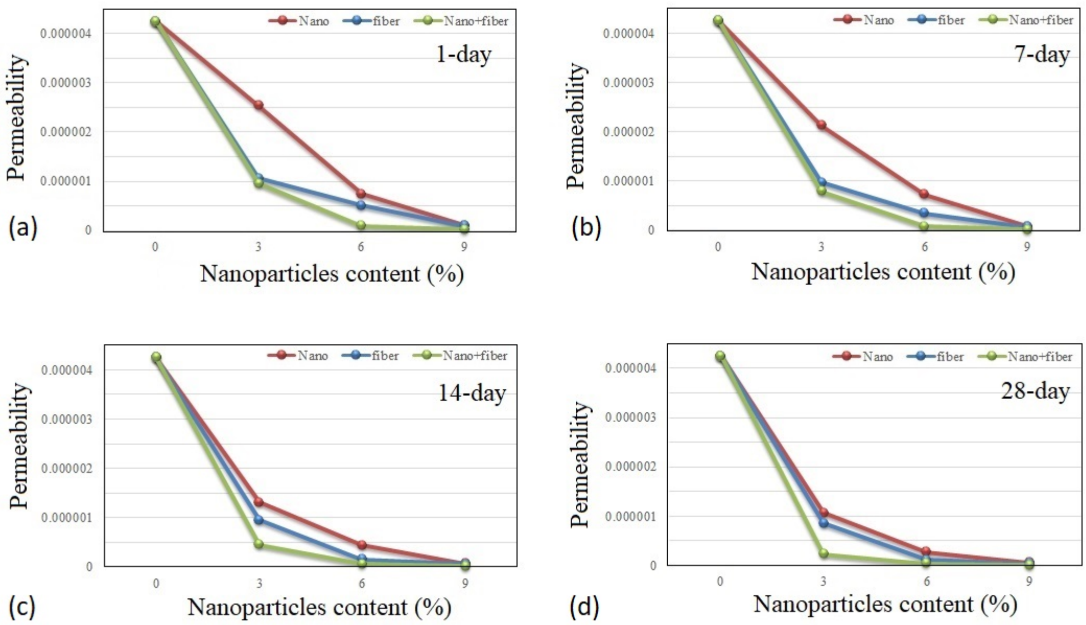

Figure 5. Permeability of studied samples in different groups (unit is cm/s): (**a**) 1-day, (**b**) 7-day, (**c**) 14-day, (**d**) 28-day.

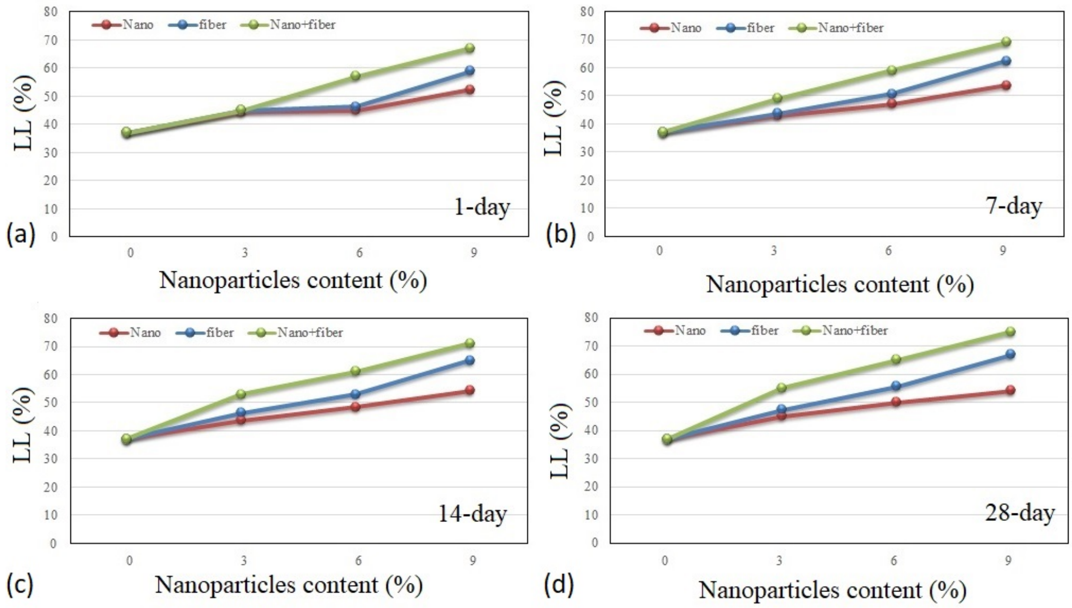

Figure 6. Liquid limit results were measured for different groups of the samples: (**a**) 1-day, (**b**) 7-day, (**c**) 14-day, (**d**) 28-day.

The UCS and direct-shear were used to investigate the strength parameter variations of mixed soils. The UCS, c and φ are the index strength parameters that represent the mechanical properties of the fine-grained soil regarding durability. These mechanical properties were estimated for studied samples in different groups and presented in Figures 9–11. According to Figure 9, the UCS measured for the main soil (control group) was 120 kPa which became 188 kPa in the nanoclay group; 200 kPa in the nanofiber group, and 216 kPa in the nanoclay–nanofiber composite after a 28-day waiting period. These changes indicated that using the nanofiber mixture increased the UCS more than the nanoclay mixture and the composite mixture showed improvement. Therefore, the UCS is modified using a nanofiber mixture in samples, but the composite mixture reaches the highest rate of UCS. It can be concluded that nanoclay–nanofiber composite application significantly improve

UCS and the durability of the soil. Figures 10 and 11 show the c and φ changes in the nanoclay, nanofiber, and composite groups. As seen in this figure, the variation of the various mixtures in the φ does not show a visible change, but c has shown an increase with the increase in waiting time and additives. In the nanoclay group, the c has reached 185 kPa to 270 kPa. In the nanofiber group, it reached 275, and in the composite group, it hit 288 kPa. Regarding the obtained results, the nanoclay–nanofiber composite has provided good improvements in soil durability which can be considered an appropriate alternative to modify landfill liners.

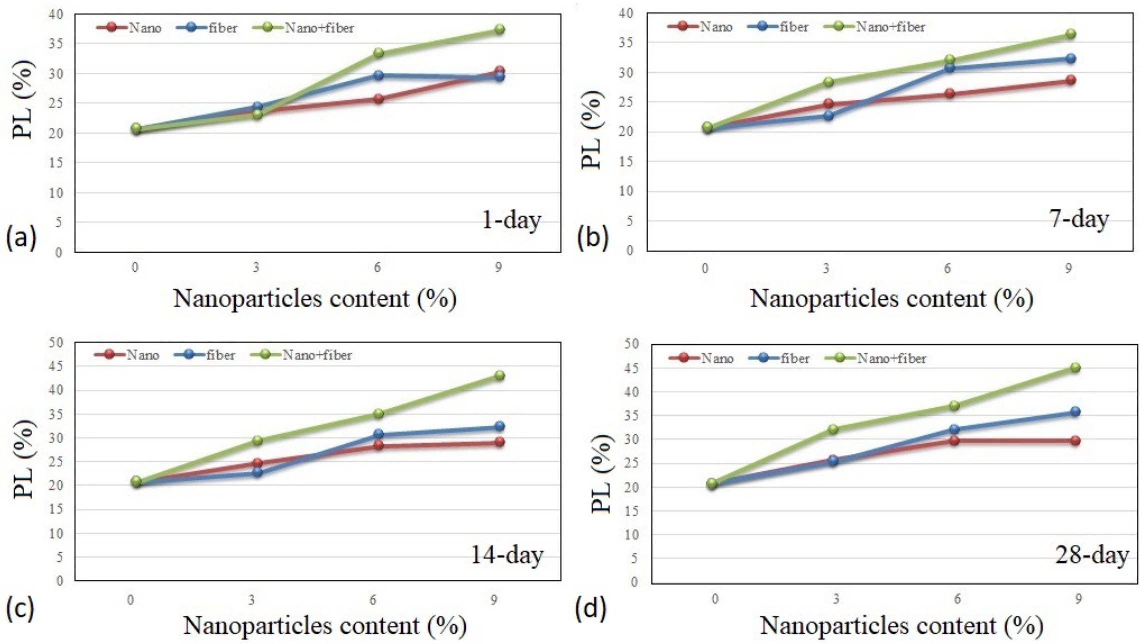

Figure 7. Plastic limits results were measured for different groups of the samples: (**a**) 1-day, (**b**) 7-day, (**c**) 14-day, (**d**) 28-day.

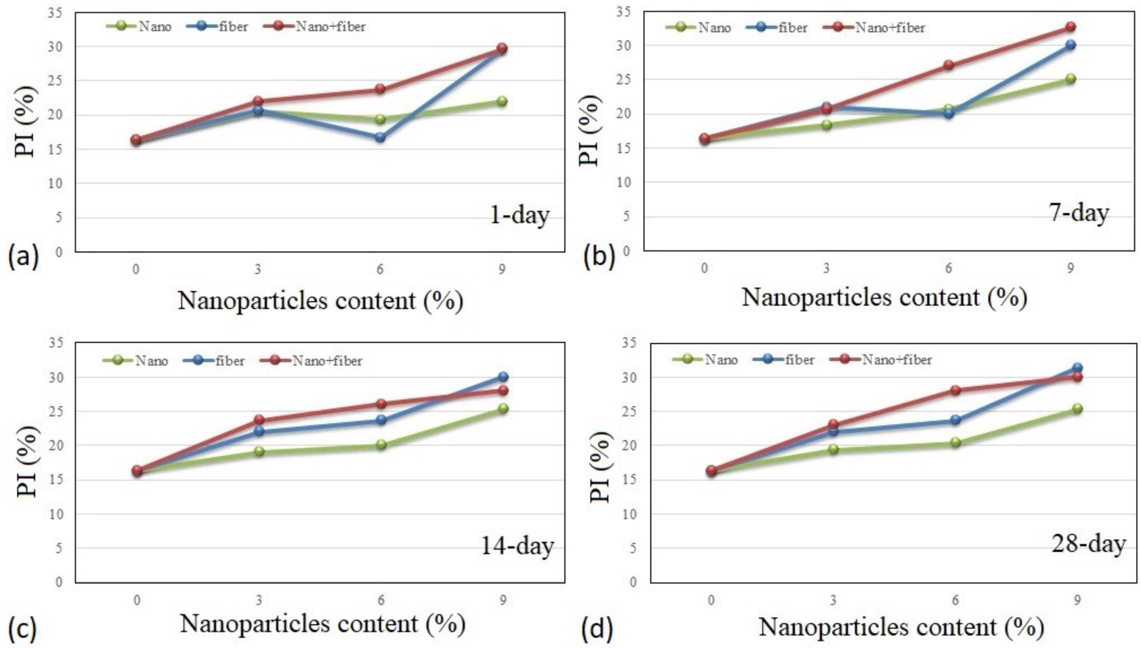

Figure 8. Plasticity index results were measured for different groups of the samples: (**a**) 1-day, (**b**) 7-day, (**c**) 14-day, (**d**) 28-day.

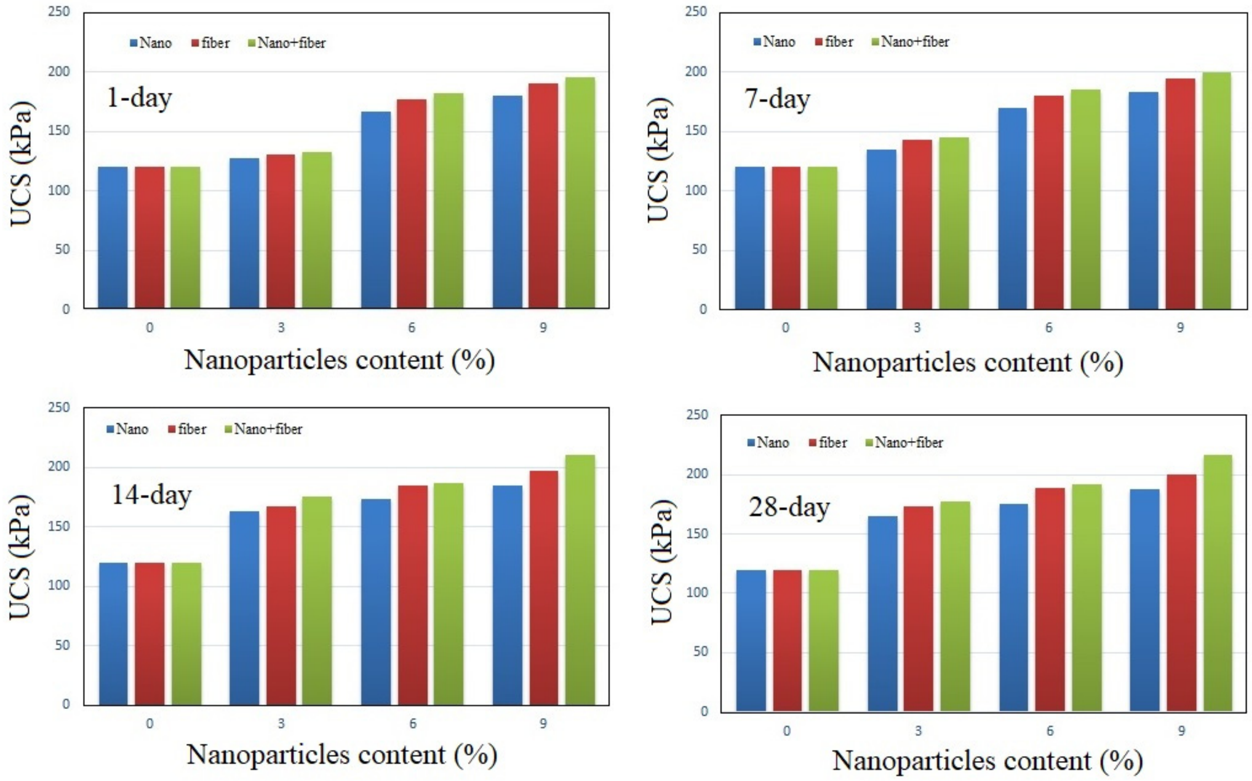

Figure 9. UCS results for a different group of the samples.

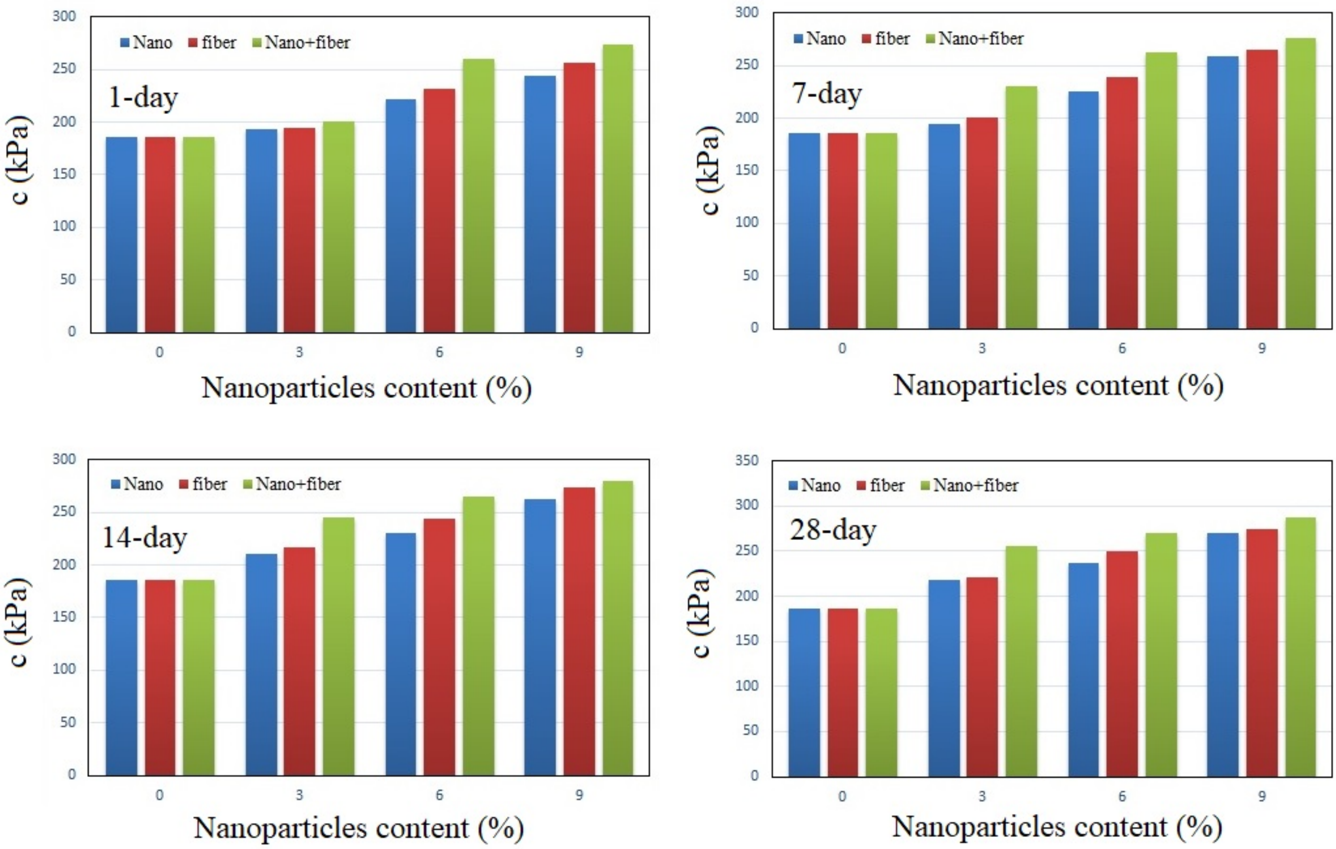

Figure 10. Estimated cohesion for a different group of the samples.

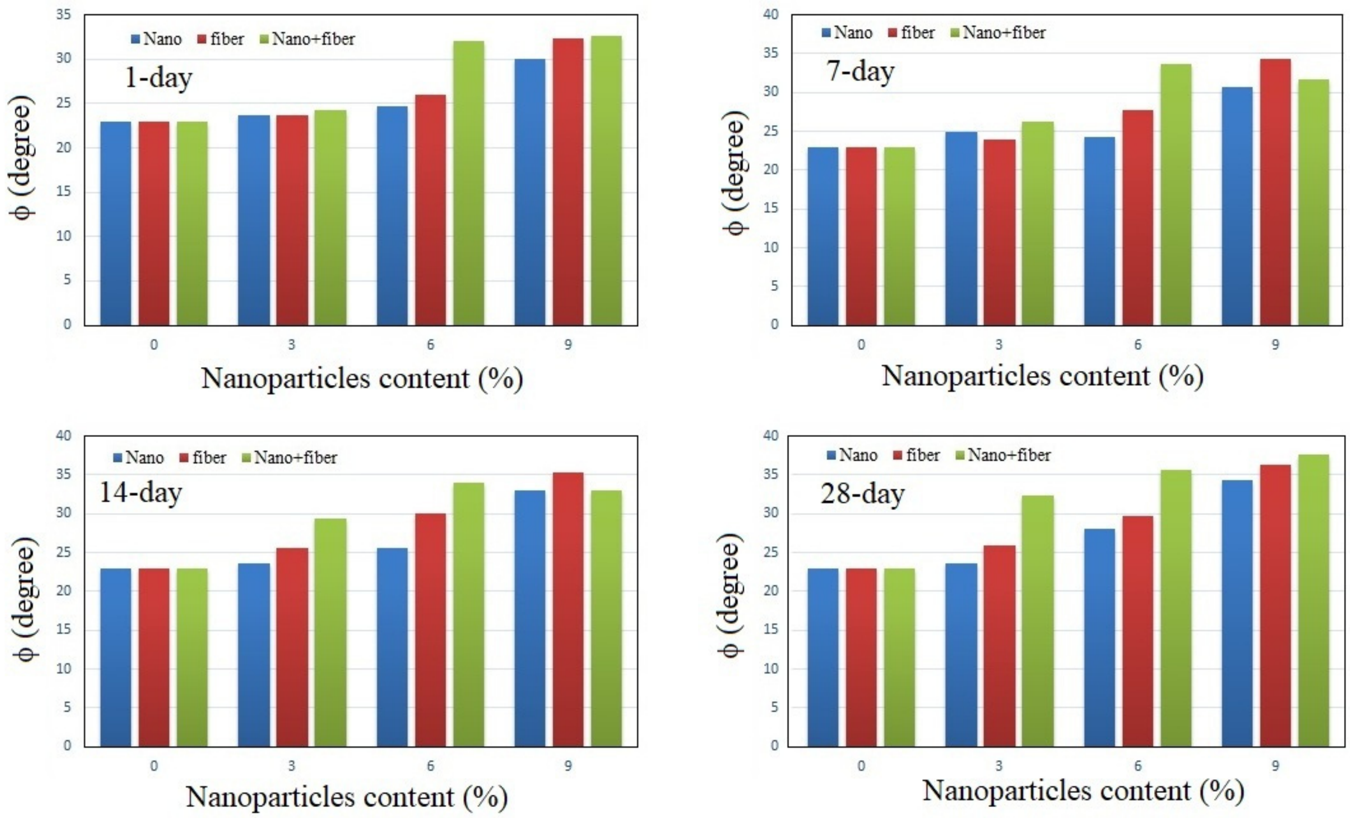

Figure 11. Estimated friction for a different group of samples.

k is the permeability coefficient which decreased, because nanomaterials such as montmorillonite clay is a fine-grained soil with a low permeability capability. Adding such materials with low to good permeability will naturally reduce soil permeability. On the other hand, due to the nano size of these materials, they enter easily into the interparticle spaces in the soil and this action will further reduce the permeability of the soil. On the other hand, with the entry of these particles into the interparticle spaces according to their characteristics, they cause the increase in soil fertility and structural strength. This strength works to increase soil resistance against loading or cutting. Therefore, it is reasonable to expect that by adding nanomaterials, soil resistance will also improve.

4. Conclusions

The presented study attempted to provide extensive experimental research on nanoclay–nanofiber composite application to geotechnical improvements and permeability for landfill liners and its impact on leachate prevention. In this regard, a total of 120 samples were taken from the Tabriz landfill site (northwest of Iran) and prepared for various geotechnical tests to conclude particle size, hydrometry, permeability, Atterberg limits, UCS, and direct-shear. The samples were prepared and we waited for 1, 7, 14, and 28 days before the testing stage. All tests were performed based on ASTM regulations and repeated three times (to reduce human and device error) which represents average values. The taken samples were divided into four groups after preparations: concluded nanoclay additives, nanofiber additives, nanocomposite additives, and control samples (without additives) with 3%w, 6%w, and 9%w mixtures. Based on the results of the study, the following are presented:

1. The studied soil is classified as CL class based on USCS which is estimated using particle size and hydrometry tests. CL stands for clayey soil with low plasticity.
2. Regarding permeability tests, the results indicated that additives such as nanoclay, nanofiber, and nanocomposite (nanoclay–nanofiber) reduce permeability. Nanoclay reduces the permeability from 4.25×10^{-6} cm/s to 5.25×10^{-8} cm/s. Nanofiber reduces

the main value to 3.24×10^{-8} cm/s and nanocomposite reduces it to 6.34×10^{-9} cm/s. Therefore, this estimation application of nanoclay–nanofiber composite shows significant work on permeability reduction.

3. Measured Atterberg limits for different groups showed that the plasticity increased with an increasing in the nanoparticle additives, but it increased more rapidly for LL than PI or PL. Nanoclays were recognized as responsible for the plastic behavior of the soil.
4. UCS and c increase when increasing the type and amount of additives in soil, while φ has not shown the significant action of variations. Results show that the nanoclay–nanofiber composite has provided good improvements in soil durability, with increased UCS from 120 kPa to 220 kPa after a 28-day waiting period. It increased c from 185 kPa to 288 kPa as well.
5. Considering the results of the study it appeared that the nanoclay–nanofiber composite had a good impact on leachate control by reducing permeability and increasing soil durability and mechanical properties which can now be attributed to the optimal design of the landfill liners.

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