



Article Microstructure and Physical-Mechanical Characteristics of Treated Kaolin-Bentonite Mixture for Application in Compacted Liner Systems

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Abstract: Treated bentonite-rich soils used as liner materials in landfills may provide an effective solution to the problems of increased void ratios upon swelling at reduced suction as well as desiccation cracking when suction is increased during desaturation. Accordingly, this study provides an understanding of the evolution of void ratio of the mixed materials during swelling at three different suction levels upon saturation as well as the soil water retention (SWR) during desaturation. For the treatment process, low quantity of cement binder whose production leverages raw material resources with efficient dry-process kilns and the benefit of lower energy consumption were used. Results indicated increased mixed soils' strength irrespective of increased fines content due to thixotropy. The mixed soils exhibited almost equal values of void ratios at different hydration stages, suggesting that slightly reduced expansion mostly affects the subsequent phases of moisture ingress at full saturation compared to the natural soils. Lower values of void ratio obtained at full saturation also suggests possible reduced infiltration of water into landfills. The observed increased moisture retention within the osmotic suction zone and a decrease in the same as the fines content increased in the mixed soils can aid contaminant encapsulation while also reducing desiccation cracking. The findings of this research are intended to serve as a benchmark for further studies using other sustainable materials for treatment of mixed soils.

Keywords: cement; sodium bentonite; montmorillonite; stabilisation; clay liners; kaolinite; landfills

1. Introduction

Engineered hydraulic barriers can function optimally when designed and constructed to meet the requirements set out in regulatory guidelines [1,2]. Bentonite clays are some of the most important elements of compacted clay liners (CCL). They consist mainly of montmorillonite mineral and therefore belong to the family of smectites. They are geologically formed mainly due to activities of chemical weathering of volcanic ashes. Sodium bentonite has a large specific surface area and is chemically monovalent (Na⁺) which means that it has a greater potential of being readily absorptive and expansive, especially when in contact with water compared to calcium (Ca²⁺) bentonite. Since natural sodium bentonite deposits are rarely found in abundance in some parts of the globe, clays containing kaolinite mineral (chemically weathered feldspar) of relatively lower swelling capacity are also sometimes used as sealants to contain wastes. Still, a mixture of bentonite with kaolinite and or without sand in different proportions have been used in the past as a probable compromise to satisfy cost and performance [3–6].



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Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). To effectively safeguard ground water and the environment from pollutant leakages emanating from hazardous municipal and industrial landfilled wastes, a well-designed and engineered lining system is required. Compacted liners used as top covers in sanitary landfills can prevent water infiltration into the contained wastes beneath the system. However, this water does eventually percolate into the solid waste products and supplements the leachate that is generated.

Sodium bentonite clays are most frequently chosen for the construction of compacted lining systems mainly due to their high adsorptive, retention and swelling capacities. However, bentonite materials may undergo significant volumetric changes upon wetting and depending on the operational circumstance, their void ratios may increase considerably with a reduction in suction, particularly at higher degrees of saturation [7,8]. The change in void ratio due to saturation can have an effect on the performance of the lining system in terms of its hydraulic conductivity and diffusion property.

On the other hand, increased suction levels under extreme desaturation can result in low absorption rates and possible desiccation cracking, leading ultimately to increased permeability [9,10]. Reasonable treatment of the bentonite-rich engineered landfill system with binders can offer sustainable solutions to these challenges.

Treated or stabilised as-compacted clays utilised especially in cover liner systems are constructed close to the ground surface (active zone) and, as such, exist mostly in an unsaturated condition. Hence, their hydraulic characteristics described as a relationship between pore water suction and moisture content can be understood and interpreted using the soil water retention curve (SWRC). The SWRC defines an inverse but unique relationship between the mass of moisture present in a soil and the corresponding energy state or suction within the pore water. This study therefore aims to further an understanding of the evolution of void ratio during swelling at reduced suction as well as the water retention characteristics of five cement-treated clays having different proportions of sodium bentonite.

An investigation into new concepts regarding the infiltration rates and fluid transport in natural bentonite-rich expansive clays used in engineered landfill systems have received considerable attention in recent studies [11–13]. However, not many research studies have considered the water retention properties of bentonite and/or kaolinite materials in terms of the relationship between pore water in the soil and suction for application in engineered barriers [14–16]. Seiphoori et al. [7] suggested that the expansion of bentonite-based liners at reduced suction levels, and at particularly high degrees of saturation, can affect moisture diffusion and hydraulic conductivity. Changes in the hydrated void ratio relate to the pore structure evolution on the hydration path as a consequence of the formation of new pore levels. Hence, void ratio modifications during the swelling process can be fully understood by a consideration of the unsaturated hydraulic property through the moisture retention curve [7]. Ghavam-Nasiri et al. [8] stressed further, the importance and the effect void ratio changes can have on the retention capacity of soils as a porous media. It was reported that air-entry value (AEV) (the suction value that marks a transition to a desaturated state) on a drying SWRC and water-entry value (WEV) (the values of suction at which transition to saturated condition begins) on a wetting SWRC can change with void ratio.

Conversely, CCL can be subjected to desiccation cracking that may lead to an increase in hydraulic conductivity as a result of changes in climatic conditions [17,18]. He et al. [10] noted in their studies that the self-healing properties of an expansive clay such as bentonite, used as final liner covers, cannot be relied on to completely eliminate cracking due to desiccation. Nonetheless, an earlier research had suggested that the onset and subsequent crack occurrence relates to some soil-specific "critical" level of suction. Hence, the SWRC was used to provide input to the design of a compacted liner cover based on a study of their saturation rates and desiccation process [9].

It is proposed in this study that minimal treatment of highly expansive clays can aid a reduction in hydrated void ratio at low suction as well as enhance the moisture retention capacity of these clays, hence minimising cracking during desaturation. Although, the

general mechanical and hydraulic performances of chemically treated expansive clays used in lining systems have been investigated in several research works [19–27], studies devoted to the moisture retention behaviour of these stabilised systems are rare. Consequently, this research aims to study the mechanical behaviour as well as provide an understanding of moisture retention properties of medium-to-highly expansive clays treated with cement. The cement used, depends mostly for its manufacture, on the use of raw material resources with efficient dry-process kilns with the benefit of lower consumption of energy.

2. Materials and Methods

Most researchers have focused on the use of relatively less proportions of bentonite (ranging from 2% to 20%), but it is pertinent to note that higher proportions have also been reported in literature [28,29]; hence, this study intends to experiment on expansive clays with proportions of sodium bentonite of up to 75% by weight of the entire soil mass. Since sodium bentonite is essentially composed of a high percentage of aluminates and silicates but a very low amount of calcium, 8% of cement was utilized to stabilise the soils based on established practices, and in order to prevent subsequent drying upon compaction [6,30]. Compared to the highest amount of the bentonite used for initial stabilisation of the mixture, this quantity of cement could be considered as being minimal.

2.1. Test Materials

In order to meet the objective of this study, clays with potential to exhibit varying absorptive capacities were selected. Kaolinite with a medium swelling capacity, and the very highly absorptive sodium bentonite generally utilised for lining systems, were adopted in this study. Both materials were sourced from Mistral Industrial chemical in Northern Ireland, UK. Table 1 gives the chemical compositions of these soil minerals. The studied clays were simulated by adding varying proportions of the bentonite to the kaolinite (considered here as the parent soil) and thoroughly mixing the powders to produce five soils of wide-ranging swelling capacities in the following ratios: 0:100, 10:90, 25:75, 50:50 and 75:25 by dry weight of the total mass [31]. The five investigated kaolin-bentonite mixtures in the present study are labelled as soil 1, soil 2, soil 3, up to soil 5 in order of increasing bentonite content.

Material	Oxide Content (% by Mass)												
	SiO ₂	Al_2O_3	Fe ₂ O ₃	CaO	MgO	K ₂ O	TiO ₂	Na ₂ O	SO_3	Mn_2O_3	LOI		
Kaolinite	49	36	0.75	0.06	0.3	1.85	0.02	0.1	-	-	12		
Na-Bentonite	57.1	17.79	4.64	3.98	3.68	0.9	0.77	3.27	-	0.06	7.85		
CEM I, (52.5N)	20.7	4.6	2.6	65	1.7	0.4	0.3	0.1	2.9	0.1	2.9		

Table 1. Chemical composition of materials.

The cement used was supplied by the Hanson Heidelberg group, UK, and was produced to comply with the requirements of BS EN 197-1. The oxide compositions of the cement are given in Table 1.

2.2. Experimental Procedure and Testing

2.2.1. Particle Grading

Analysis of the particle sizes of the soils in their dry state was done by the Malvern Mastersizer 2000, which uses the technology of laser diffractometry. The actual percentage size distribution was subsequently determined from the granulometry bins by using a visual basic grain size distribution and statistics (GRADISTAT) version 4 spreadsheet package developed by Blott and Pye [32]. According to Figure 1, Soil I (Kaolinite) is uniformly graded, and the particle size distribution achieved for the other four soils would remain within the delineated boundary and progress towards the more gap-graded bentonite as the fines content increases in the mixture. The precise fitting curve for particle-



Figure 1. Particle size analysis of the soils.

Geotechnical Parameter	Range	Source
Particle grading	% gravel \leq 30; fines content \geq 20%	[34]
	% gravel \leq 20–50; clay fraction \geq 30%	[35]
	Clay fraction $\geq 10\%$	[36]
Consistency limits	$20\% \leq LL \leq 90\%$; $7\% \leq PI \leq 65\%$	[34,37–39]
Unconfined compressive strength	\geq 200 kPa	[40]
Hydraulic conductivity	$\leq 10^{-9} \text{ m/s}$	[1]

2.2.2. Index Property Testing

Tests to determine the index properties of the samples were conducted based on the procedures stated in the ASTM standard technical documents. Table 3 enumerates the basic geotechnical properties of the natural soils.

Clay Sample Notation	K:B	LL	PL	PI	Specific Gravity	Silt Content	Clay Content	MDD	ОМС	USCS Classification
	ASTM D 4318-1				ASTM D 854-10	ASTM D 422-63		ASTM D 1557		
	(%)	(%)	(%)	(%)		(%)	(%)	(kN/m ³)	(%)	
Soil 1	100:0	58	30	28	2.60	74	26	14	15	CL
Soil 2	90:10	85	37	48	2.65	70	30	13.9	21	CH
Soil 3	75:25	130	48	82	2.69	65	35	13.5	23	CH
Soil 4	50:50	222	58	164	2.7	58	42	13.2	25	CH
Soil 5	25:75	285	72	213	2.76	48	52	12.9	30	CH

Table 3. Properties of the investigated clays.

Where: K = Kaolinite; B = Bentonite, LL = Liquid limit; PL = Plastic limit; PI = Plasticity index; MDD = Maximum dry density; OMC = Optimum moisture content; USCS = Unified soil classification system.

2.2.3. Compaction

The moisture content of the soils required for subsequent engineering tests were determined based on their optimum compaction conditions using the standard proctor testing according to ASTM D1557-12e1 (2012). The soils treated with the cement binder were compacted at the optimum moisture content (OMC) and maximum dry density (MDD) of the natural soils but with the addition of not more than 2% water to each of the mixes [41]. For the performance of engineering tests, the samples were carefully extracted from the compaction mould and trimmed accordingly. The treated samples were subjected to a temperature-controlled (± 22 °C) curing for 7 days to promote hydration.

2.2.4. Microstructure and X-ray Spectroscopy Observation

Scanning electron micrographs (SEMs) obtained from JEOL JSM-5900LV and ZEISS EVO apparatus and the corresponding Energy dispersive X-ray spectroscopy (EDS) were also employed to study the microstructure, chemical and elemental compositions on the sample surfaces responsible for possible fabric changes. Figure 2 shows the micrographs of the compacted samples revealing their aggregate structure. These micrographs confirm that the microfabric of Na-montmorillonite rich clays are likely to be characterised by dispersed and undulating filmy particles as compared to the low swelling kaolinite with more of a leaf-like arrangement [42]. As the montmorillonite content increases, the compacted mixtures tend to exhibit more aggregated and concentrated clusters of clay particles. This behaviour could result in impervious layers that gives rise to an initial low rate of swelling upon moisture ingress but with the ultimate free swelling taking a longer time to be completed. X-ray diffraction (XRD) analyses of representative samples are shown in Figure 3. Since only two pure minerals were blended to achieve the natural soils, it was only necessary to take the XRD on Soil I (kaolinite) and Soil V (containing 75% of the bentonite). Apart from the presence of a larger amount of kaolinite in Soil I, the XRD also confirms traces of other minerals like quartz, muscovite (mica) and microcline (feldspar) present in the clay (Figure 3a). XRD for Soil V seems slightly complex. Even though the mineral montmorillonite is present in abundance, another mineral called the Loughlinite seems to be also available in larger quantities (Figure 3b). The Loughlinite is basically hydrous sodium magnesium silicate that is found mostly in oil shales [43].

2.2.5. Swell-Consolidation and Strength Tests

The swell characteristics of the natural soils and treated soils upon the completion of curing were determined through a series of conventional one-dimensional oedometer testing under saturation according to ASTM D4546-14e1 (2014).

The UCS test was conducted as per ASTM D2166-00 (2000) on the natural and treated samples after the stipulated curing period. Loading was carried out at a rate of loading of 1 mm per minute.



Figure 2. Microstructure of natural soils.



Figure 3. XRD of natural soils: (a) XRD of Soil I, (b) XRD of Soil IV.

2.2.6. Water Retention Property

The composite nature of most lining systems and the potential of capillary breakage due to the presence of cover may significantly reduce the applicability of direct suction measurement methods [8]. Hence, an indirect method using the non-contact filter paper (N-CFP) provides a successful solution for measuring a wide range of suction (10 kPa-100 MPa) as applied by Acikel et al. [44] and Risken et al. [45] in engineered liners. The procedure set out in ASTM D5298-16 (2016) utilizing Whatman Grade No. 42 qualitative type N-CFP of 55 mm diameter was applied in this research to measure total suctions of the samples. The as-compacted natural and treated samples were first brought to full saturation using distilled water in a syringe (with 1 hour allowed to ensure adequate penetration and absorption of the moisture) and then allowed to desaturate and the filter paper employed to measure suctions when approximately 2 g of water was observed to have been lost from each sample. Void ratios were measured at corresponding saturation levels. Calibration equations proposed by Leong et al. [46], Equations (1) and (2), were applied to obtain suction from the measures N-CFP moisture contents.

$$\varphi = 10^{2.909 - 0.0229_{w_f}} \quad w_f \ge 47 \tag{1}$$

$$\varphi = 10^{4.945 - 0.0673_{\rm W_f}} \quad w_f < 47 \tag{2}$$

where:

 φ = suction

 w_f = filter paper water content

2.2.7. Mathematical Models for Soil Water Retention Curve (SWRC)

Laboratory suction data were subjected to a nonlinear regression fitting process to obtain the SWRC by using the model proposed by van Genuchten [47], Equation (3). The soil module function of the SoilVision program (version 5.4.08) was utilized to enable an effective non-linear fit of suction data using the in-built fitting model.

$$\frac{s}{s_0} = \left[\frac{1}{\left(1 + \left(\frac{\psi}{a}\right)^n\right)^m}\right] \tag{3}$$

where:

s = degree of saturation

 s_0 = degree of saturation at suction $\psi = 0$

 φ = soil suction (kPa)

e = exp(1), base of natural logarithm

- a = fitting parameter, which relates to the air entry value of the soil (kPa)
- n = fitting parameter, being a function of the slope of the SWRC
- m = fitting parameter, being a function of the residual water content

2.2.8. Test Sample Size

The laboratory tests in this research require a good number of samples to be prepared. In order to minimise errors due to the sample size used, for the unconfined compression test, three samples (at most) were used for each test and the average value derived for each data points. The oedometer and suction tests utilised two samples each. Hence, by also considering that the specified curing duration had to be completed before the respective testing, a total of 40 compacted (treated and untreated) samples were prepared and tested.

3. Results and Discussion

3.1. Consistency Limits

Consistency limits of the treated soils along with the natural clays are presented in Figure 4. Before addition of cement to the natural soil, it was important to allow a minimum

of 24 h for sufficient initial hydration of the natural clay samples with water due to the presence of the heavier bentonite fines, which have a tendency for slow absorption of water. There is a continuous rise in LL of the natural soils from 58% for Soil I up to 285% for Soil V as the percentage of bentonite fines increases from 0 to 75% (Figure 4a). This is due to the greater affinity or demands for moisture in the soils as clay fines increase in an attempt to give a more workable product. Nevertheless, the modification effect of cement seems to cause a slight increase in LL of Soil I but a consistent reduction for the rest of the soils as a result of initial hydration reactions and reduced diffuse double-layer thickness. On the other hand, Figure 4b indicates an overall decrease in PI when the soils are treated with cement. The percentage decrease in PI (54%, 23%, 4%, 13% and 1% for treated Soil I-Soil V, respectively) corresponds to an increase in the amount of bentonite in the soil-cement mix. This phenomenon signifies the reduced influence of treatment on soils as the amount of clay fines increases. Hence, more cement may be required to cause an obvious decrease in the highly rich bentonite mixtures. Within the range of LL and PI required for clay liner applications as suggested in Table 2 and also depicted in Figure 4, both the natural and treated Soils I and II (having 0 to 10% bentonite content) would meet these conditions.



Figure 4. Consistency limits of natural and treated soils: (a) Liquid limit, (b) Plasticity index.

3.2. Unconfined Compressive Strength

The UCS of the natural soils do not show much variation (fluctuating between 190 and 270 kPa) despite the different quantities of the added bentonite (Figure 5). This lack of sufficient difference is noted in literature [48,49]. Bentonite quantity in a soil beyond a certain limit may not result in any appreciable effect on the UCS of the compacted clay. Muntohar and Yogyakarta [50] have indicated that soil strength is not directly proportional to the amount of bentonite present. A threshold limit of 30% by mass of bentonite fines in a bentonite-rich mix has been suggested as that which can generate similar mechanical behaviour to that of pure bentonite used alone. As noticed in Figure 5, all five soils can only marginally meet the minimum requirement for strength (≥ 200 kPa) for lining systems. However, with the soils stabilised by cement, UCS appear to increase up to Soil III (having 25% bentonite content) and then decreases with further increase in the bentonite fines, but not below the lowest acceptable limit for lining systems. A more rational explanation of this phenomenon is that the cement may have had a relatively reduced effect on both Soil IV and V, which possess 50% and 75% of the bentonite proportion, respectively. Stabilisation by cement resulted in the formation of cementitious gels and products of hydration, hence enhancing strength. However, it is believed also that the presence of bentonite might have also aided strength increment in spite of its very high plasticity. This can be explained by a process called "thixotropy", as confirmed in Mitchell and Soga [51] and Zhang et al. [52]. Thixotropy in this case refers to a reversible, isothermal and time-dependent process that

1200 1000 1000 400 200 0 Soil I Soil II Soil II Soil II Soil II Soil IV Soil V Soil V



Treated soil

Figure 5. Unconfined compressive strength of natural and treated soils.

Natural soil

3.3. Microstructure

Morphological complexities of the stabilised soils are immediately apparent compared to the natural soils (Figure 6). The formation of cementitious gels (calcium aluminosilicate hydrates, C-S-H and C-A-H) and ettringite (calcium-sulpho-aluminates and Monosulfoalumino-ferrite) which are strength-enhancing components are evident. Moreover, it could also be observed that the spaces between the particles are much narrower, with dense packing resulting from the binding of the soil particles by cement. EDS for stabilised Soils I and IV shown in (Figure $6c_{e}$) indicate the principal peaks consisting of calcium, oxygen, aluminium and silicon, and traces of sulphur, magnesium and sodium, all of which confirm the compounds of hydration and pozzolanic reaction product CSH formed in the stabilised soils. However, on average, the EDS showing the stabilised Soil IV containing 50% bentonite gives much-reduced quantities of the elements which comprise the compounds of the products of hydration. This further explains that little quantity of cement may have been available to be used up by soil to enable reasonable cationic exchange and pozzolanic reactions. This was experienced previously by the small effect the cement addition had on LL and PI, as stated previously. Hence, an increase in the amount of the binder may be needed to cause an obvious decrease in the highly rich bentonite mixtures, given their very high plastic characteristics brought about by high bentonite content.





Figure 6. SEM and EDS of treated soils: (a) SEM of treated Soil I, (b) SEM of treated Soil II, (c) EDS of treated Soil I, (d) SEM of treated Soil IV, (e) EDS of treated Soil IV.

3.4. Volume Change

An evaluation of the swelling potential of bentonite-based lining systems at different levels of suction in terms of saturated void ratios is very critical. Figure 7 indicates that hydration from the initial suction (as-compacted state) results in a progressive increase

in the volumetric swelling of the natural samples up to saturation and beyond, until the final equilibrium state (i.e., when no further swelling is observed (ese). Swelling increased only marginally from the initial state (e_{SI}) to that which corresponds to an almost zero suction under full saturation, (e_{50}) for the samples (approximately 10%, 14%, 7%, 4% and 5% for soils I, II, III, IV and V, respectively). This outcome suggests that soils having less percentage of the heavier bentonite clay seem to exhibit higher expansion at the initial stages of moisture ingress due to their leaf-like or flaky characteristics, giving rise to a relatively more porous structure, as confirmed previously in the micrographs of Figure 6. However, in the course of time, much higher volumetric expansion at equilibrium (under full saturation or very low suction) would be accorded Soils III, IV and V, which possess so much of the clay fines. The increased void ratio at equilibrium swell is ascribed to the modification of the montmorillonite particles in the path of hydration, as more water is drawn into the spacing between particles due to osmotic forces, which then subsequently results in an emergence of new pore levels in the structure of the bentonite. This phenomenon is expected to bear an influence on the hydraulic performance of lining systems in terms of moisture diffusion and permeability, as confirmed by Seiphoori et al. [7]. Treatment with cement slightly reduces volumetric expansion, as shown in Figure 7b. The formation of crystalline calcium alumino-silicate compounds in the process of time caused by cement treatment, as alluded to in Figure 6, prevents further moisture ingress and expansion of the soils. Figure 7b also indicates a slight incremental trend in swelling corresponding to an increase in bentonite content in the soil. Compared to the natural soils, there is approximately only a 10% reduction in the swelling when treatment by cement is applied. Interestingly though, for the treated soils, almost equal values of void ratios at the different stages of hydration (saturation states corresponding to initial, zero suction and equilibrium swelling) in each of the soils are noted. Compared to the natural soil, this phenomenon indicates that, although swelling is only slightly reduced by cement treatment, this reduction mostly affects the subsequent stages of moisture ingress at full saturation given that the void ratios at these stages do not increase more than those at the initial as-compacted state before moisture application. Lower void ratio obtained under saturation enhances the performance of lining systems because this ensures limited hydraulic conductivity and diffusion coefficient [53]. Hence, treatment with the cement in this research is very promising for compacted liner or top cover systems because, while the desired swell properties are preserved, infiltration or percolation of water into the landfill wastes underneath is minimised (when the system is fully saturated). Subsequent investigation of the retention characteristics of the natural and treated systems are given below.



Figure 7. Void ratio at different saturation levels: (a) Natural soils (b) Treated soils.

3.5. Moisture Retention Capacity

A key element in the analysis of the hydro-mechanical characteristics of liner systems is the establishment of a constitutive relationship between suction and the degree of saturation through the moisture retention curve [54]. The SoilVision package (version 5.4.08) adopting the nonlinear regression fitting van Genuchten function was utilized to enable the soil water retention curve (SWRC) to be obtained from the raw laboratory suction data. Table 4 shows the fitting parameter obtained from the van Genuchten model. This model provided very good fits to the suction data as the regression coefficients (R²) all seem to exceed 0.90 for both natural and treated soil samples. However, no obvious trend between the soils (natural and treated) is noticed for the rest of the other van Genuchten fitting parameters. Pedarla et al. [55] attempted a correlation of the fitting parameters with montmorillonite fractions in an expansive clay and reported a non-consistent trend.

	a	vg	n	vg	m	vg	R ²		
Soil	(1/k	(Pa)							
	Natural	Treated	Natural	Treated	Natural	Treated	Natural	Treated	
Soil I	0.0008	0.0006	1.777	3.000	0.511	0.001	0.992	0.974	
Soil II	0.0021	0.0005	3.000	1.092	0.139	0.284	0.986	0.989	
Soil III	0.0026	0.0008	3.000	1.3889	0.0999	0.198	0.996	0.989	
Soil IV	0.002	0.0015	3.000	3.000	0.090	0.102	0.992	0.997	
Soil V	0.0029	0.0004	2.999	3.000	0.102	0.353	0.966	0.908	

Table 4. Van Genuchten fitting parameter.

From Figure 8a, the SWRC appear to be slightly congruent one to another, particularly at suction values up to about 1000 kPa. Beyond this point, a separation occurs with an obvious "flattening" of the desorption branches of the curves from Soil I to Soil V. The increasing quantity of montmorillonite from Soil I to Soil V corresponds to high moisture retention capacity given that the heavier clays are reluctant to give up water from their pores during desaturation at increased values of suction. This invariably confirms the notion that a relatively higher amount of water is needed to hydrate clays that possess high montmorillonite proportions. Much of this retention or flattening seems to occur from suction values of approximately 1000-1500 kPa. Matric suction, or the effects of capillarity, governs the portion on the SWRC between 0 and 1500 kPa, while osmotic suction (differential salt concentration gradient in the pore water) occupies the high suction values from about 1500 kPa and beyond [56]. In the same vein, matric suction tends to relate more to the energy state at the air-water interface (i.e., the contractile skin), whereas the osmotic suction is more closely associated with the diffused double layer surrounding the clay particles. The interaction in the double layer (combination of clay particles, the interparticle fluid and the bulk pore fluid) and the corresponding repulsions existing between particles are a reflection of osmotic pressure [57]. Thus, for similar moisture content at a certain point of consideration on the curves, a lot of suction energy may be required to extract the "osmotic pore water" remaining in the soils having higher fines content (and by extension higher diffused double layer due to hydration) than those with a lower amount of the clay fines. Treatment with cement increases the retention capacity of the soils, as indicated in Figure 8b–f. This could be partly due to the higher quantity of water available and necessary for the soil-cement hydration to occur. Increased retention aids contaminant encapsulation as well as reduced shrinkage or desiccation cracking upon drying in the landfill top and bottom covers. Miller et al. [9] predicted and confirmed the susceptibility of lining systems to desiccation cracking corresponding to certain compaction conditions inferred from the SWRC for an untreated cover liner. For instance, considering the five soils used in this study, a "critical" section can be located that relates to the onset of cracking corresponding to each degree of saturation. Consequently, a numerical procedure is then applied to determine the minimum degree of saturation or water content for varying

climatic conditions, hence establishing the vulnerability to desiccation and cracking. The amount of retention seems to gradually decrease in the treated soils with an increase in the bentonite fines fractions from Soil I to Soil V. This further confirms a seemingly less influence of the cement as the bentonite content increases as mentioned earlier. For instance, the desorption branch of the SWRC of cement-treated Soil IV almost appears to coincide with that of the natural soil (Figure 8e), while an obvious less retention is observed for treated Soil V for suction values beyond the osmotic range (Figure 8f).

Figure 8. SWRC of natural and treated soils (a) Natural soils (b) Soil I (c) Soil II (d) Soil III (e) Soil IV (f) Soil V.

The air-entry value (AEV) for the natural soils do not appear to show much variation as indicated in Figure 9. For the natural soils, the air entry is occurring within the zone

of matric suction where the curves seem to coincide as noted above. AEV is that value of suction in which air would begin to enter the smallest sized pores in the soil as it desaturates. A relatively faster "draw-down" or desaturation was noticed in Figure 8a for Soil I compared to the bentonite-rich soils. The AEV for the treated soils are much higher than those of their natural counterparts, hence their higher moisture retention capacities. The evolution of the degree of saturation with suction for the treated soils as compared to the natural soils as indicated in Figure 8b–f shows that the denser the soils get due to filling of voids by cement, the higher the AEV would be. Also, a trend of reduction of AEV is noticed as the percentage of fines increases from Soil I to Soil V. Again, this suggests lesser retention for the treated soils with higher proportions of the bentonite fractions.

Figure 9. AEV of natural and treated soils.

4. Volume Change and Suction

Changes in void ratio can affect the moisture retention capacity of deformable soils. More so, void ratio can exert significant influence on the AEV suction, suggesting that the breakthrough of air into the soil pores is more arduous especially for denser soils [54]. Figure 10a–c demonstrates an increase in AEV suction with a corresponding decrease in the void ratio for both the natural and treated soils, which corroborates the trend drawn in research [54,58,59]. This is comparable to an imposed suction procedure whereby, during desaturation, the migration of the liquid phase will be initiated once the AEV suction is overcome. This transition in moisture retention behaviour mirrors a limit capillary tension level that the soil can sustain. This limit tends to vary inversely with pore sizes (and thus with the void ratio) and is the reason why the AEV decreases distinctly as the void ratio increases. The relationship between void ratio and AEV suction, which is essentially a power function, and which holds true for the natural and treated soils, is evident from Figure 10a–c, as suggested by authors in their proposed models, most of which are based on the van Genuchten equation [8,60,61]. Notice also how this relationship applies irrespective of the state or condition of saturation of the soils.

A direct linear relationship exists between the AEV suction value and the initial suction values (i.e., corresponding to dry density and void ratio at the as-compacted state) before saturation, as shown in Figure 10d. Increasing suction at the initial condition prior to saturation results in an increase in the AEV. This phenomenon as could be observed is applicable to both the natural and treated soils.

Figure 10. Void ratio vs. suction relationship of natural and treated soils: (**a**) e_{SI} vs. AEV, (**b**) e_{S0} vs. AEV, (**c**) e_{SE} vs. AEV, (**d**) Initial suction vs. AEV.

5. Conclusions

This research investigated the mechanical responses of treated bentonite-rich clays for potential application in engineered barriers. An understanding of the evolution of void ratio of the treated soils during swelling at different suction levels upon saturation as well as the soil water retention (SWR) characteristics during desaturation were provided. The main conclusions are as follows:

- Plasticity indices of the soil-cement composites reduced with 8% of cement applied but the effect became minimal with increased fines content. Consistency limit properties of both the natural and treated soils with 10% of the sodium bentonite fraction meets the general requirements for most clay liners.
- Increased unconfined strength for the stabilised soils adheres to the requirements for a compacted lining system but may not correspond to the quantity of bentonite in the soils due to thixotropic effects.
- Formation of cementitious gels and ettringite, which are strength enhancing components, were evident from the SEM analyses; however, elemental peaks from the EDS measurements suggested a rather reduced effect of the cement used as the soils became more plastic.
- Almost equal values of void ratios at the different stages of hydration suggested that slightly reduced expansion mostly affects the subsequent stages of moisture ingress at full saturation when compared to the natural soils. The lower values of void ratio obtained at full saturation can mean reduced infiltration or percolation of water into landfill wastes.
- Increased moisture retention occurred with cement treatment but with a decrease in the same as the proportion of bentonite increased in the soils. This outcome can aid

contaminant encapsulation as well as reduce shrinkage or desiccation cracking upon drying in engineered landfill systems.

Finally, it is important to reiterate that most of the discussions, results and conclusions of this research have relied on the usage of a relatively lower quantity of a calcium-based binder, which benefits from efficient dry-kiln process; hence, lower energy consumed from its production, for treatment of the mixed soils. This is intended to serve as a reference for further studies, which the authors are currently embarking on to further an understanding of the concepts proposed herein by incorporating other sustainable binder materials.

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