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RESEARCH ARTICLE

Some Geotechnical Properties of Clay Nanocomposites

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Abstract

Clay soils have various areas of use in geotechnical engineering. The common use of clayey soils are for landfills, dams, nuclear plants, etc. However, when the clay soils interact with water, the geotechnical properties such as the swelling, the shear strength, and the compressibility properties of clay change gradually. Therefore, in this research, using a hydrophobic organo-clay, polymers (locust bean gum, latex, glycerine, and vinyl acrylic copolymer), and rubber powder, clay-nanocomposites were developed to solve the problems due to the clay-water interaction encountered in the clay liners. This study focuses on the geotechnical properties of the clay-nanocomposites researched experimentally in laboratory conditions. The test results found that, the specific gravities, consistency limits, compaction parameters, and unconfined compressive strengths of clay-nanocomposites change significantly, when compared to those of natural clay and hydrophobic organo-clay.

Keywords

Biopolymer, clay-nanocomposite, compaction parameters, consistency limits, contact angle, hydrophobic organo-clay, geotechnical properties, specific gravity, unconfined compressive strength, vinyl acrylic copolymer

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1 Introduction

Clay minerals are very small particles which are very active electrochemically. The presence of even a small amount of clay minerals in a soil mass can markedly affect the engineering properties of that mass [1]. The common use of clayey soils are for landfills, dams, nuclear plants etc. However, clay minerals, especially smectites are strongly influenced by presence of water. The variation of water content gives rise to plasticity, and the Atterberg limits are an indication of this influence. Clay minerals, being relatively small particles, have large specific surfaces, and they have high ion exchange capacity [1, 2]. Natural clay minerals are low-cost materials with high sorption properties [3]. In this context, smectites (especially montmorillonites) are a valuable mineral class for industrial applications [4, 5, 6]. Due to these properties of clay minerals, the geotechnical properties such as the swelling, the shear strength, the compressibility properties of clay change gradually. Commonly the clay has been gained negative features because of the change of clay properties due to the clay-water interaction. With the increasing of swelling or settlement the volume of the clay has been changed and it could affect negatively the stability of clay. To remove the negative features that could effect the stability of clay soil stabilization by chemicals have been studied by a number of researchers [7, 8, 9, 10]. Additionally, with the increasing of interest on polymer technology, the researchers are interested in surfactants and polymers to modify clays for improving their engineering properties [11, 12, 13, 14, 15]. The organically modified silicate nanolayers are called ‘nanoclays’ or ‘organosilicates’ [16]. The hydrophilic surface of the aluminosilicate surface of smectites can be rendered hydrophobic by exchanging the naturally occuring inorganic cations with organic cations [17]. Alkylammonium ions, in industrial applications mainly quaternary alkylammonium ions, are widely used in modifying bentonites [18].

Polymer-clay nanocomposites are a new class of materials in which the polymer matrix is reinforced by uniformly dispersed inorganic particles, having at least one dimension in the nanometer scale [19]. The clay-nanocomposites are prepared by incorporating finely-dispersed layered silicate materials in a polymer matrix. In recent years, polymer/layered silicate nanocomposites have attracted great interest from researchers [4, 5, 12, 13, 16, 17, 20, 21, 22, 23, 24, 25]. Hackman and Hollaway (2006) mentioned the advantages of using nanocomposite materials in civil engineering over those of the more conventional construction materials [26].

Biopolymers are naturally occurring polymers that are found in all living organisms. Biopolymer-clay nanocomposites are a new class of materials with potentially improved mechanical properties [27]. They are renewable source-based materials that would not involve the use of toxic or noxious components in their manufacture and could allow degradation via a natural composting process [28].

Contact angle measurements are often used to indicate clay wettability and interfacial tension [29] and provide an insight into surfactant behaviour. Rogers et al. (2005) examined the contact angles of some compatibilisers for polymer-silicate layer nanocomposites [29]. While native montmorillonite surface is hydrophilic, adsorption of a small amount of surfactant on the surface can render it hydrophobic [25]. In this study, hydrophobic organo-clay is used to eliminate the negative effects of clayey soils like swelling, settlement and strength when interacted with water.

Kurt and Akbulut (2014), produced clay nanocomposites by means of the sol gel method, using a hydrophobic organo-clay, polymers (locust bean gum, latex, glycerine, vinyl acrylic copolymer), and rubber powder to understand the dynamic properties (secant shear modulus and damping ratio) and swelling of the clay nanocomposites [30]. The objective of this paper is to modify clay-nanocomposites and to investigate the effects of locust bean gum (biopolymer) (LBG), latex (LTX), glycerin (GLC), vinyl acrylic copolymer (VA), and rubber powder (RP) on the geotechnical properties (specific gravity, consistency limits, compaction parameters, and unconfined compressive strength) of different combinations and preparations.

2 Materials

2.1 Natural Clay and Hydrophobic Organo-clay

The clayey soil samples (Table 1), originating from a clay pit in Oltu-Narman, deposits in Erzurum/Turkey and these clays are classified as high plasticity clay (CH), according to the Unified Soil Classification System (USCS), and also based on some engineering properties of natural clay (Table 2). Natural clay was used as the template sample. Additionally, hydrophobic organo-clay was used for preparing nanocomposites and improving clay and hydrophobic organo-clay properties. The hydrophobic organo-clay (Table 2) was prepared using the following procedure, as described by Kurt (2009) [31] and Kurt and Akbulut (2010) [32]. A cationic surfactant, dialkyl ammonium meta sulfate (DAMS), was used in the preparation of hydrophobic organo-clay. First, 40 g of clay was dispersed in 8 L of deionized water and stirred with a magnetic stirrer at 1000 rpm for 2 h. Previously prepared surfactant solution (DAMS and deionized water) was added slowly to the clay suspension at 30ºC. The modified product (hydrophobic organo-clay) was dried at room temperature. The surfactant included 5% by weight of clay [30].

Table 1 The chemical composition of clay

Chemical Composition		Clay
SiO2	%	41.48
Al2O3	%	12.22
CaO	%	11.14
Fe2O3	%	9.88
MgO	%	8.10
K2O	%	1.23
TiO2	%	0.53
Na2O	%	0.2
LOI	%	13

Table 2 Some engineering properties of natural clay and hydrophobic organo-clay [31]

Some Properties		Clay	Hydrophobic organo-clay
Clay content, %	< 0,002mm	56	-
Specific gravity, %	G_s	2.62	2.52
Liquid limit, %	w_L	72	-
Plasticity index, %	I_p	39	-
Contact angle, º	-	37	88
Cation exchange capacity	(meq./100 g dry soil)	26.25	21.62
Optimum moisture content*, %	w_{omc}	16.5	14
Maximum dry density*, kN/m³	γ_{dmax}	17.55	16.67
Unconfined compressive strength*, kPa	q_{uu}	1048	998
BET (N_2) surface area, ²/g	-	10,19	5
Soil classification	(USCS)	CH	-

*The results were investigated from the samples compacted with 2597kJ/m³ energy level

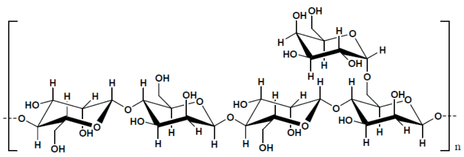
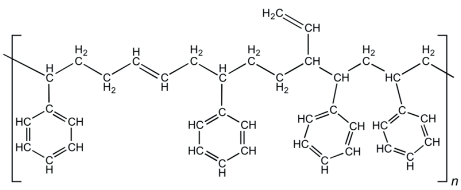
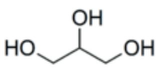
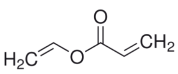
2.2 Polymers and Other Materials

In this study, locust bean gum, latex, glycerine, vinyl acrylic copolymer, and rubber powder (Table 3, 4) were used for preparing nanocomposites and improving clay and hydrophobic organo-clay properties. The conventional use of locust bean gum as an excipient in drug products generally depends on thickening, gel forming, and stabilizing properties [33]. Locust bean gum is a virtually neutral galactomannan polymer extracted from the seeds of the carob tree [34]. Locust bean gum markedly increases gel strength and changes the gel character from brittle to elastic [35].

Table 3 Properties of locust bean gum, latex, glycerine, vinyl acrylic copolymer and rubber powder [30]

Properties	Locust bean gum	Latex	Glycerine	Vinyl acrylic copolymer	Rubber
Chemical formula	(1-4) linked beta-D mannose residues and the side chain of (1-6) linked alpha-D galactose.	C_3H_3N	$C_3H_8O_3$	-	-
Chemical composition	Galacto-mannan (a group of hydrocol-loids)	Styrene Butadiene Emulsion	Glycerol	Vinyl Acrylic Copolymer Emulsion	Styrene Butadiene Copolymer
pH	5-7	8 -12	7	5 ± 1	-
Viscosity (cps)	2000-3500	-	1200	1000-5000	-
Density (g/cm^3)	-	1.015	1.261	1.03	1.15-1.198

Table 4 Chemical structures of locust bean gum, latex, glycerine and vinyl acrylic copolymer [30]

	Chemical structure
Locust bean gum	
Latex	
Glycerine	
Vinyl acrylic copolymer	

Latex, which was the other polymer used in the experiments, is an elastomer. Natural rubber is produced from the latex of the Hevea brasiliensis tree [36]. The presence of glycerine is essential for preparing nanocomposites [37]. In some studies for preparing nanocomposites, glycerine was used as plasticizer [38,39,40].

Vinyl acrylic copolymer emulsion is a water-based resin [41]. Acrylic resins such as vinyl acrylic copolymers may be used as the adhesive component of the composition [42]. The pigment binding and film forming of vinyl acrylic copolymer are significant, and its water stability is also notable. Vinyl acrylic copolymer was also used for its adhesive properties [30].

The rubber powder used in the present study was purchased from Kahya Rubber, Sakarya, Turkey. Due to its light-weight nature and its capacity for damping energy, the rubber powder can be used to mitigate seismic forces and to absorb earthquake vibrations [43]. The use of waste fiber materials in geotechnical applications was investigated by Akbulut et al. (2007) to evaluate the effects of scrap tire rubber and synthetic fibers on the unconfined compressive strength parameters, and on the dynamic behavior, of clayey soils [44].

3 Characterization of Composites

3.1 Sample Preparation and Tests

The clay nanocomposites were obtained by the sol-gel method [45]. The clay-nanocomposite samples were prepared using the following procedure, as described by Kurt and Akbulut (2014) and Kurt (2014) [30,46]. First, LBG (0.5%) was added to 2 L of water and mixed using a mechanical stirrer at 4000 rpm until dissolved. During stirring, latex (10%) was added and mixed for 20 min. Then, glycerine (10%) was added and the solution was mixed for 10 min. Next, 2500 g of hydrophobic organo-clay and 1 L of water were added and mixed for 1 h. Finally, vinyl acrylic copolymer was added in different proportions (0%, 5%, 10%) and mixed with a mechanical stirrer. (These products were clay nanocomposite samples without rubber powder additive CNC). Rubber powder (5%) was also added and mixed for 10 min to produce clay nanocomposite samples with rubber powder additive (CNCR). The leach products (clay nanocomposites) were dried at room temperature for 48 h. All of the percentages of polymers and additives were used as the percentages of dry hydrophobic clay weight (Table 5).

Table 5 Clay-nanocomposite contents

	Content
C	Natural clay
HOC	Hydrophobic clay
CNC0	HO-%0,5LBG-%10LTX-%10GLC
CNC5	HO-%0,5LBG -%10LTX-%10GLC-%5VA
CNC10	HO-%0,5LBG -%10LTX-%10GLC-%10VA
CNCR0	HO-%0,5 LBG -%10LTX-%10GLC-%5RP
CNCR5	HO-%0,5LBG-%10LTX-%10GLC-%5VA-%5RP
CNCR10	HO-%0,5LBG-%10LTX-%10GLC-%10VA-%5RP

For measuring contact angles, a goniometer, which allows the user to measure the contact angle visually, was used. The droplet is deposited by a syringe pointing down vertically onto the sample surface, and a high-resolution camera captures the image, which can then be analysed either by eye (with a protractor) or using image analysis software [25]. In this study, the contact angles were measured with a goniometer (CAM 101, KSV Instruments, Finland), using hydrophobic organo-clay and clay-nanocomposites.

The specific gravities and consistency limits (liquid limit and plastic limit) were determined in the clay-nanocomposite samples in accordance with ASTM D 4892 and BS 1377, Part 2, 1990, respectively. The cone penetrometer (fall cone) method was used to determine liquid limit. Additionally, plastic limit tests were performed on the material prepared for the liquid limit test. The maximum dry unit weight (γ_{dmax}) and moisture content were determined in the clay-nanocomposite samples in accordance with ASTM D 1557. The compaction effort of the modified proctor tests were 2597kJ/m^3 (E1).

The unconfined compressive strength tests were carried out in accordance with ASTM D 2166. The tests were conducted on the compacted samples with moisture contents as in the above-mentioned compaction tests. The unconfined compressive strength tests were conducted on the natural clay, hydrophobic organo-clay and clay-nanocomposite samples that compacted with E1 and 5192kJ/m^3 (E2) energy levels.

3.2 Contact Angles

The contact angle measurements of the clay-nanocomposite samples (Figure 1, 2) revealed that the contact angle values of clay-nanocomposites were increased. The contact angle measurements indicated that, the clay water affinity was decreased by polymers and a hydrophobic surface produced. With increasing VA content, the contact angles of the CNC and CNCR samples increased and became more hydrophobic when compared with the hydrophobic organo-clay.

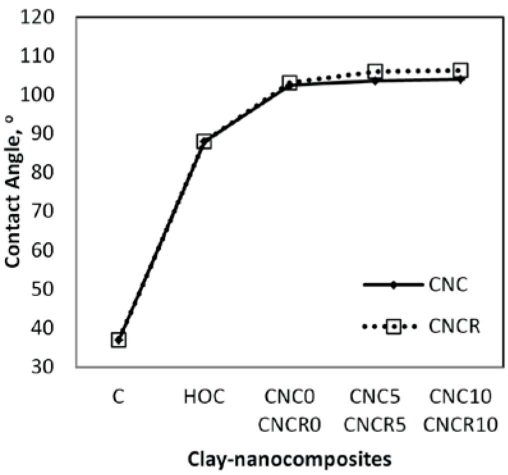


Fig. 1 Contact angle measurements of the clay-nanocomposite samples

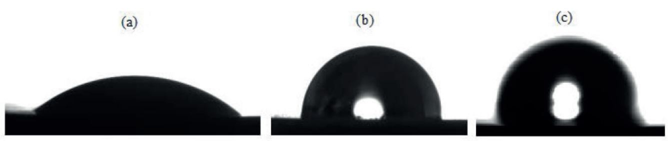


Fig. 2 The contact angles of some clay-nanocomposite composite samples. a) Contact angle image of the natural clay (37°); b) Contact angle image of the HOC (88°); c) Contact angle image of the CNCR10 sample (106.3°)

3.3 Specific Gravities

The specific gravity test results (Figure 3) revealed that, the specific gravities of CNC and CNCR samples were decreased when compared with natural clay and hydrophobic organo-clay. The decrease in the specific gravities of CNC and CNCR samples are due to the increasing of pore ratios [13, 47]. Denham (1999) indicated that, the lower specific gravity of the organo-clay could be attributed to a change in the soil fabric. Conversely, it could be thought that, the decreasing of specific gravity values are due to the increasing of pore sizes and basal spacings of the clay-nanocomposites.

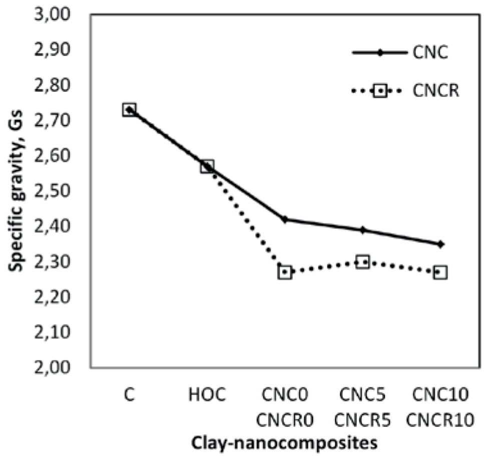


Fig. 3 Specific gravities of the nanoclay-composites

3.4 Consistency Limits

The clay-nanocomposites gained non-plastic (NP) behaviour [31,32] (Figure 4, Table 6). It should be pointed out that, there has been no general consensus regarding the effect of surfactants and chemicals on clays' consistency limits [25]. Similarly, there is no study on the consistency limits of clay-nanocomposites. The consistency limit tests that focused on the natural clay showed that, the classification of natural clay is CH (high plasticity clay). However, the hydrophobic organo-clay and clay-nanocomposites showed non-plastic feature.

Fig. 4 Water drops on the hydrophobic clay [31]

Table 6 Consistency limit test results of the clay-nanocomposites

	w_L, %	w_P, %	IP, %	Soil classification
C	70	33	37	CH
HOC	NP	NP	NP	NP
CNC0	NP	NP	NP	NP
CNC5	NP	NPw	NP	NP
CNC10	NP	NP	NP	NP
CNCR0	NP	NP	NP	NP
CNCR5	NP	NP	NP	NP
CNCR10	NP	NP	NP	NP

3.5 Compaction Results

Modified proctor tests were conducted on the clay-nanocomposite samples to determine their moisture content and maximum dry unit weight (γ_{dmax}) relationships (Figure 5). Because of the hydrophobic property of clay-nanocomposites, the compaction tests were conducted on the clay-nanocomposites during the drying process. The compaction test results of the clay-nanocomposites (Figure 6, 7) indicated that by increasing the VA content, the maximum dry densities of the CNC and CNCR samples are decreased. Akbulut et al., (2013) reported that, the maximum dry unit weights decrease with the increasing percentage of the cationic surfactant, due to the low specific gravity of organo-clays [13]. In this sense, it could be said that, the decreasing of the maximum dry unit weights of the CNC and CNCR samples are related with the lower specific gravities of the clay-nanocomposites (Figure 3).

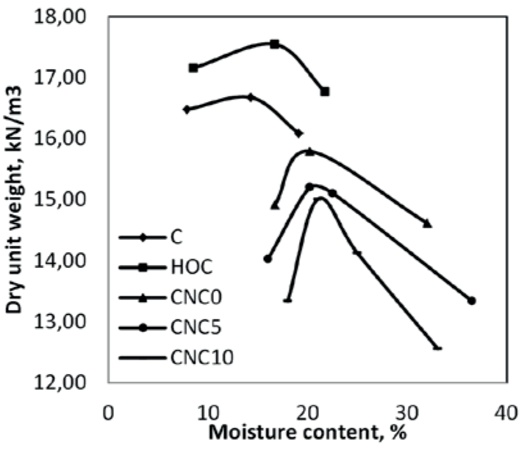


Fig. 5 Compaction curves of the clay-nanocomposite samples

The void ratios and moist unit weights

The minimum void ratios (e_{min}) are determinated from the maximum dry densities (ρ_{dmax}) and specific gravities (Gs) of the clay-nanocomposites with Eq. (1). In this equlation, ρ_{water} means the density of the water. The minimum void ratios of the CNC samples are increased when compared with the hydrophobic clay (Figure 8). Also, the minimum void ratios of the CNCR samples are not change significantly. It could be said that, at first, the minimum void ratios of the CNC and CNCR samples drop to a lower void ratio value and then, increase gradually with increasing VA content.

$$e_{\min} = \left(\left(Gs^{*} \rho_{water} \right) / \rho_{d\max} \right) - 1 \quad (1)$$

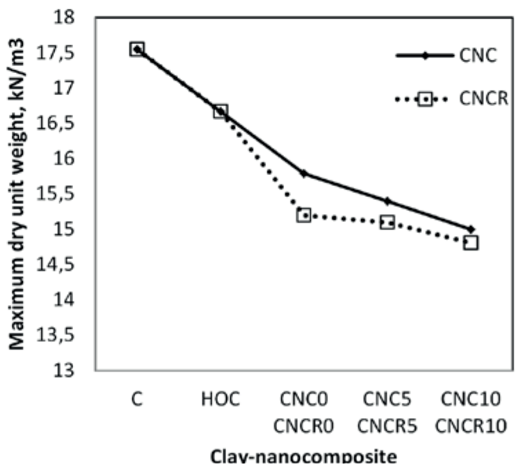


Fig. 6 The maximum dry unit weight values of the clay-nocomposites (compaction effort is 2597 kJ/m³)

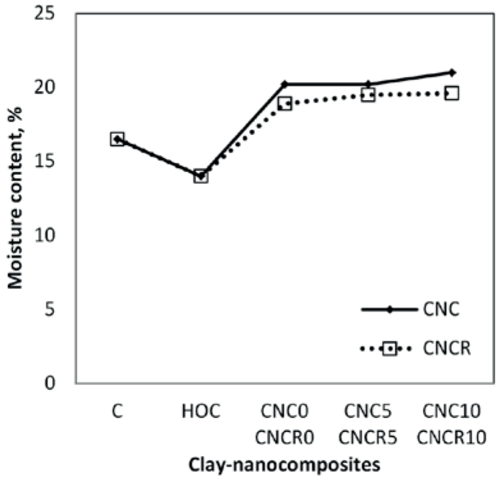


Fig. 7 The moisture values of the clay-nanocomposites

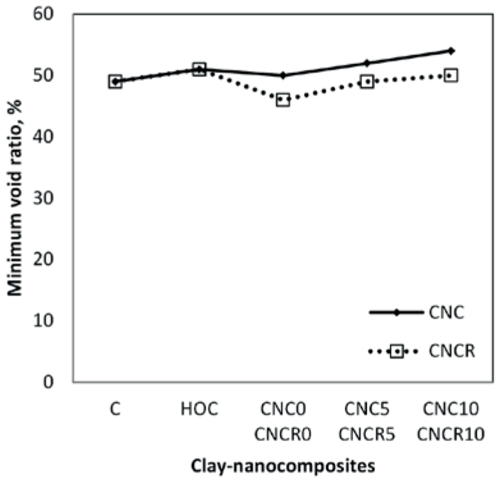


Fig. 8 The minimum void ratios of the clay-nanocomposite samples

The change in the moist unit weights of the CNC and CNCR samples with the VA percentage (Figure 9) indicated that, these values are decreased with the increasing VA content when compared with the hydrophobic organo-clay and natural clay. Furthermore, the moist unit weights of the CNCR samples were decreased more than the CNC samples.

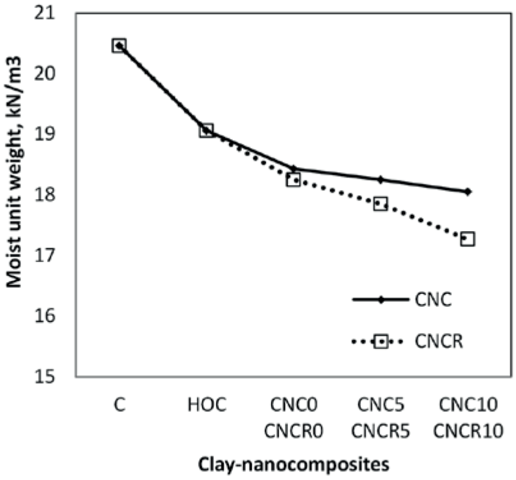


Fig. 9 The moist unit weights of the clay-nanocomposite samples

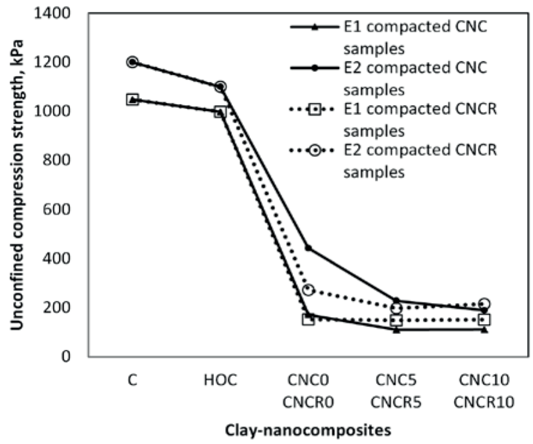


Fig. 10 The unconfined compressive strengths of the clay-nanocomposites

3.6 Unconfined Compressive Strength Tests

The unconfined compressive strength tests were conducted on the clay-nocomposites compacted with E1 energy level and the clay-nanocomposites compacted with E2 energy level. The moisture contents of the samples compacted with E1 energy level was lower than the samples compacted with E2 energy level.

The unconfined compressive strengths of the clay-nanocomposites were decreased with the increasing VA content when compared with the natural clay and hydrophobic organo-clay (Figure 10). Also for %0 VA the decrease is significant. On the other hand, with increasing VA content the decrease is marginal. The unconfined compressive strength values depend on a large number of parameters, which are, the composition of the soil particles, the water content of the compacted soil, the compaction effort for the preparation of the compacted soil, and the shape and size of the soil particles [48]. The decrease of the unconfined compressive strength test values of the CNC and CNCR samples are due to the decrease of the specific gravities and the increase of the void ratios of the clay-nanocomposites [13]. On the other hand, Das (2008) stated the general relation between consistency and unconfined compression strength of clays [49]. According to Das (2008), the clays have 96-192 kN/m^3 uncompression compressive strength called stiff clay [49]. Additionally, damping properties of these clay-nanocomposites have been investigated by Kurt and Akbulut (2014) [30]. They indicated that, the damping ratio values have been increased and the secant shear modulus values have been decreased with the increasing of VA percentage when compared with natural clay and hydrophobic organo-clay. Due to Finegan and Gibson (1999) with the increasing of damping ratio values, the rigidity properties of clay nanocomposites have been decreased [50].

4 Conclusions

This study was undertaken to investigate some geotechnical properties of clay-nanocomposites, and the specific gravity, consistency limits, compaction, unconfined compressive strength values have been compared with natural clay and hydrophobic organo-clay. Based on the test results and the discussion presented in this study, the following conclusions were made:

- The contact angles of the clay-nanocomposites increased when compared with the natural clay and hydrophobic organo-clay.
- The specific gravities of the clay-nanocomposites decreased when compared with the natural clay and hydrophobic organo-clay.
- The consistency limits showed that clay-nanocomposites are non-plastic.
- The compaction parameters of the clay-nanocomposites at modified compaction energy (E1) were determined. The moisture contents of the clay-nanocomposites increased with the increasing VA percentage. Also, the maximum dry unit weights of the clay-nanocomposites decreased with the increasing VA percentage.
- The minimum void ratio values and moist unit weights after the compaction tests showed that the void ratios of the clay-nanocomposites increased in the CNC samples. However, the void ratios of the CNCR samples decreased when compared with the natural clay and organo-clay samples. Additionally, the moist unit weights of the clay-nanocomposites decreased.
- The unconfined compressive strength tests of the clay-nanocomposites, compacted with both E1 and E2 energy levels revealed that, the unconfined compressive strengths of the clay-nanocomposites decreased when compared with natural clay and hydrophobic organo-clay.

Consequently, it is thought that, the clay-nanocomposites are more hydrophobic due to increasing of the contact angles. Kurt and Akbulut (2014) has been investigated the swelling pressures of the same clay-nanocomposite samples [29]. According to Kurt and Akbulut (2014), the swelling pressures of both CNC and CNCR samples have been decreased when compared with natural clay and hydrophobic organo-clay [29]. So, it could be said that, the clay-nanocomposites couldn't have been interacted with water. On the other hand, the void ratio values of clay-nanocomposites are higher and specific gravities are lower than natural clay and hydrophobic organo-clay. According to these statements, it could be said that, the clay-nanocomposites are more porous and lighter than natural clay and hydrophobic organo-clay. In addition to this, unconfined compressive strength tests showed that the consistencies of clay nanocomposites are stiff. Therefore, they can be used as a liner in waste disposal landfills and dams.

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