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Mineralogical, microstructural and porosimetry analysis in three different clayey soils

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Abstract. Diffraction, scanning electron microscope and helium porosimetry techniques have been shown to be suitable for obtaining physical and mineralogical properties in soil samples. The X-ray fluorescence spectroscopy allows obtaining the elemental composition of the sample, while the X-ray diffraction technique permits obtaining a qualitative and quantitative composition of the soil minerals. On the other hand, scanning electron microscope shows the microstructure of the soil and the helium porosimetry measures the pore size, volume, distribution, density and other porosity-related characteristics of the material. This document presents the characterisation of three different clayey soils by an experimental plan that included mineralogical, microstructural and porosimetric tests. The soils used were bentonite, kaolin and a natural clay retrieved near to Bogotá city, Colombia. Results obtained from this study allow providing a better understanding of soil behaviour, which traditional mechanical tests do not explain.

1. Introduction

Clays are building materials used widely around the world. These materials are confirmed of solid particles and voids filled, with liquid or gas, and have high variability due to geological, environmental, physical and chemical process [1]. Chemically, clays are constituted of minerals of phyllosilicates and alumina sheets. Budhu [2] defines the silicates as a group of minerals with a structural unit called the silica tetrahedron. A central silica cation (positively charged ion) is surrounded by four oxygen anions (negatively charged ions). In some cases, the aluminium (Al^{3+}) can be replaced others elements, such as iron (Fe^{2+}) or magnesium (Mg^{2+}), to polarize the system with a negative charge. This configuration generates an expansion effect when the clay is submerged into an electrolytic media since a cationic covering covers the particles and increases the space between them.

Mineralogy is the main factor controlling size, shape and some properties of clay particles [3]. The main groups of materials that compose clays are kaolinite, illite, and montmorillonite. However, such minerals present different configuration; for instance, the montmorillonite is a mineral with superposition of multiple thin laminae. Figure 1 displays the arrangement of different clay minerals. In addition, in such Figure are detailed the spaces between the layers, as well as why the clays can collect water molecules and change the volume due to the expansion and contraction of such spaces.

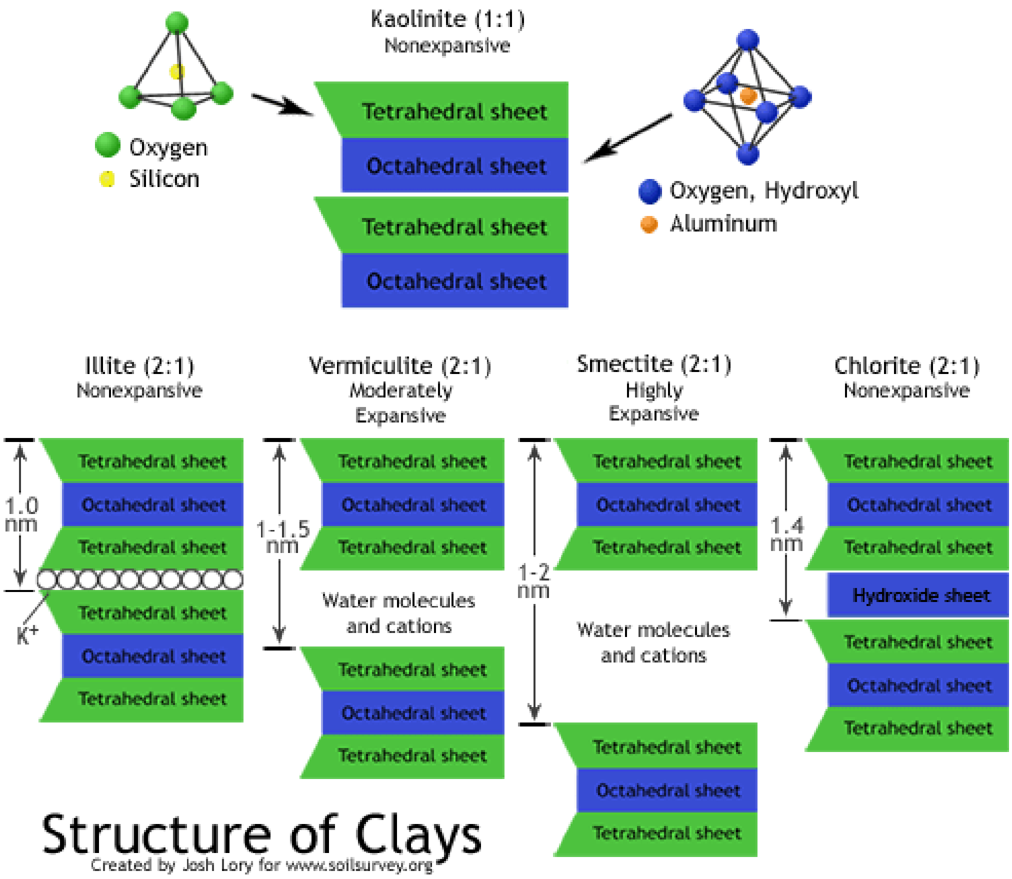


Figure 1. Structure of some clay minerals [4].

On the other hand, the assessment of soil structure by using SEM images allows obtaining the shape and distribution of the particles [5], which are key factors in the soil behaviour. Since the mechanical properties of clays depend directly on the microstructure of their particles; for instance, the angle of friction, cohesion and hydraulic conductivity. In addition, the identification of the pore size and distribution of a soil sample, by HeP tests, provides data about the material capacity to collect fluids (liquid and gas) in its voids [6].

This research work presents an assessment of the characteristics at the micro level of three different clays using X-ray fluorescence (XRF), X-ray diffraction (XRD), scanning electron microscope (SEM) and helium porosimetry (HeP) tests. The results are compared and disused in order to provide some insights about the behaviour at the macro level of these soils.

2. Methodology

2.1. Materials

In this study were used three different clayey soils: bentonite, kaolin and natural clay. Bentonite and kaolin samples were acquired commercially. Natural clay samples were retrieved at 2.0 m - 2.5 m depth in a pilot site at the Nueva Granada Campus of the Nueva Granada Military University, located near in Cajicá near to Bogotá city, Colombia. Torres *et al.* [7] and Caicedo *et al.* [8] describe the geological formation process of clays in this site, as well as Molina-Gómez *et al.* [9] present a experiential study about the compressibility of such soils. Table 1 summarizes the physical properties of the clayey soils.

Table 1. Physical properties of the clayey soils.

Clayey soil	Gs	LL (%)	LP (%)	IP (%)
Bentonite	2.69	315.3	72.2	243.1
Kaolin	2.68	62.4	28.7	33.7
Natural clay	2.70	48.5	31.8	16.8

2.2. Experimental procedures

The preparation of the samples for XRF analysis consists of grinding (if the particle size of the sample is greater than 75 μm), calcination and pearling. The crushing is done in order to obtain a homogeneous sample that prevents the formation of agglomerates of crystals when making the pearl. The calcination process consists in taking the sample to 1000 °C and keeping it at that temperature for 1 hour [10]. In this processes is evaluated the percentage by weight lost during the treatment by weighing the sample before and after test. The loss on calcination (LOI) represents the number of volatile components (H_2O, CO_2, F, Cl and S), as well as the organic matter that is not detected by the equipment. The calcined sample is mixed with a borate and lithium bromide flux (49.75% $Li_2B_4O_7$ - 49.75% $LiBO_2$ - 0.50% LiBr) in sample proportion (flux of 2:6). This mixture is then taken to a percolator in glass discs 33 mm diameter.

For the mineralogical characterization of the clays, an analysis based on the XRD technique was performed. The preparation of the sample begins with a phase of pulverization, in which particle size of less than 75 μm is guaranteed. Then, the sample is poured into a holder, and the excess of material is removed. After creating the flat surface, the test is carried out. On the other hand, XRD test for the particles with size higher than 75 μm includes three phases: (i) dehydration, (ii) solvation with ethylene glycol, and (iii) calcination. Zhang *et al.* [11] introduces this procedure in detail.

For the SEM analysis, three representative samples 3.5 cm×2.5 cm×1.0 cm covered with a transparent resin were assembled. Additionally to the resin, it was added graphite to create a conductive film, which helped in scanning under high vacuum mode. The micrographs were taken with a secondary electron detector and the X-ray dispersive energy (EDX) spectra. In the elemental analysis was obtained with a PJT microprobe with a Si-Li detector and cooling with a liquid nitrogen system (LN_2). The x-ray checking time for each spectrum was 60 seconds.

To assess the pore size distribution the HeP technique was implemented. The method involves two phases: (i) the measurement of the total volume of the soil sample, and (ii) the intrusion of helium at high pressure. The direct measurement of the total volume is made by means of a calibrator or the Archimedes principle in the cases of the regular and irregular geometry, respectively. On the other hand, the intrusion of helium is carried out by means of the double cell type Boyle. An inert gas of high diffusivity should be used to avoid the reaction and the adsorption of the same in the sample, which would give measurements of porosity greater than the real ones. This method considers a case of ideal conditions where the process occurs at a constant temperature and the same amount of a single gas is involved. Hence, the volume of a given mass of ideal gas varies inversely with the pressure applied to it. To measure the pore volume, the gas is entered into a cell where the study sample is contained at a given pressure (usually 100 psi) and then is expanded to a connected reference cell that results in a lower equilibrium pressure. Based on the difference of pressures the volume is calculated.

3. Analysis of results

3.1. X-ray fluorescence

Usually, the XRD results are presented by a diffractogram (powder sample) where the x axis corresponds to the intensities of each diffracted mineral peak, and the y axis shows the 2θ angle according to Bragg's law. Bragg (1910) showed that there is a relation between the interplanar distance in the crystalline structure of a mineral or minerals and the wavelength of the X-rays, as shows Equation (1).

$$n \cdot \lambda = 2d \cdot \sin(\theta) \tag{1}$$

where, n is a positive integer, λ is the wavelength of the incident wave, d is the inter planar distance, and θ is the diffraction angle. In this study, it was decided to present the XRF test results through a bars diagram in which the percentage by weight of each mineral is shown (see Figure 2), in order to facilitate the interpretation of the mineralogy composition.

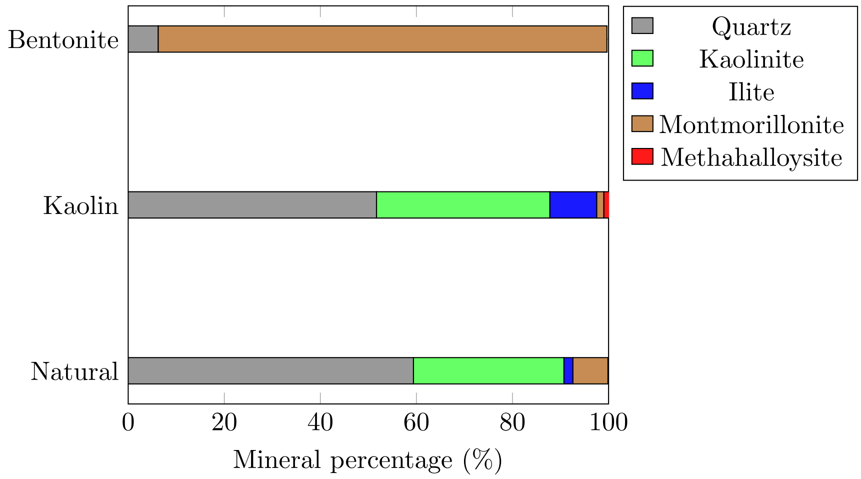


Figure 2. Compositional proportion in percentage of the samples.

The kaolin sample has a quartz percentage of 52.69%, which is a stable primary mineral that does not show expansion. It is also a hard mineral that is mainly found in sands. The second mineral in percentage is kaolinite with 36.48%, which is a soft mineral with a hardness of 2 on the Mohs scale, it does not present expansion. The third mineral is illite with 9.75%, which is a secondary mineral, with hardness from 1 to 2. The fourth mineral present is montmorillonite which is a secondary mineral, soft with a hardness of 1 to 2, another characteristic of this mineral is its high expansion due to the open structure form in which water molecules can be accommodated. Mineralogical values indicates that this soil is a material that does not show expansion and due to its mineral composition presents low Atterberg limits (low plasticity), in relation to other clays, since it contains a high content of quartz.

The bentonite studied has a percentage of 93.43% of montmorillonite, which is a highly expansive mineral. Therefore, this clayey soil experiments high volume changes according to its water content. The second mineral in percentages is quartz with 6.23%, which is very stable and resistant, but in this soil is not representative. The third mineral methahalloysita has no influence in the soil behaviour because of the low proportion. According to the mineralogical characteristics found, this soil is expansive, with high limits of Atterberg, it can not be used for fillings of retaining walls and to place foundations on top of it an expansion potential test should be carried out.

The natural clay studied has a quartz content of 59.38%, which has a hardness of 8 on the Mohs scale and is a stable primary mineral that does not present expansion. The second mineral in percentage is kaolinite with 31.33%, which is a soft mineral with a hardness of 2 and does not present expansion. The third mineral is montmorillonite with 7.43%, which is a secondary mineral, with a hardness of one to two, and presents expansion. The fourth mineral present is the illite with a percentage of 1.86%, which is a secondary mineral and a hardness of 1 to 2. According to described above, this material has a higher hardness than bentonite due to the high quartz content, and a low percentage of expansion is expected in comparison with bentonite due to the lower content of montmorillonite.

3.2. X-ray diffraction

By means of the XRD tests, an excitation was applied to the atoms of the soil samples, in order to emit characteristic radiation called x-ray fluorescence. Table 2 shows the percentages by weight of the minerals that compose each sample.

Table 2. Physical properties of the clayey soils.

Compound	Bentonite	Kaolin	Natural clay
SiO_2	61.16	48.08	64.85
Al_2O_3	18.92	19.08	19.49
Fe_2O_3	3.71	6.37	3.01
CaO	0.12	1.26	0.22
K_2O	1.70	0.76	1.22
MgO	1.67	3.04	1.65
Na_2O	0.91	2.62	0.96
P_2O_3	0.95	0.09	0.09
TiO_2	0.53	0.88	0.86
LOI	10.33	17.82	7.65

A high percentage of silicon (hardness equal 7) with 61.16% and 64.85% in the kaolin and natural clay, respectively, as well as bentonite, has a silicon content of 48.08%. Silicon-containing minerals constitute about 40% of all common minerals and are an important constituent of Portland cement and bricks. The second important component for all the samples is alumina (Al_2O_3) with percentages of 18.92%, 19.08% and 19.49%, respectively. Synchronically with silica, alumina is one of the important element in the constitution of clays. Furthermore, this component provides resistance to the material after being subjected to a heating process [12].

3.3. Helium porosimetry

Table 3 shows the results with an uncertainty of ± 0.47%. The "0" value is the calibration for the correct operation of the equipment a stainless by a steel cylinder. The natural clay was moulded in a rectangular prism. A cylindrical assembly of the powder sample with shrink sleeve around its circumference and metal meshes on the faces was performed. The dimensions were used for the total volume calculation.

Table 3. Porosity results.

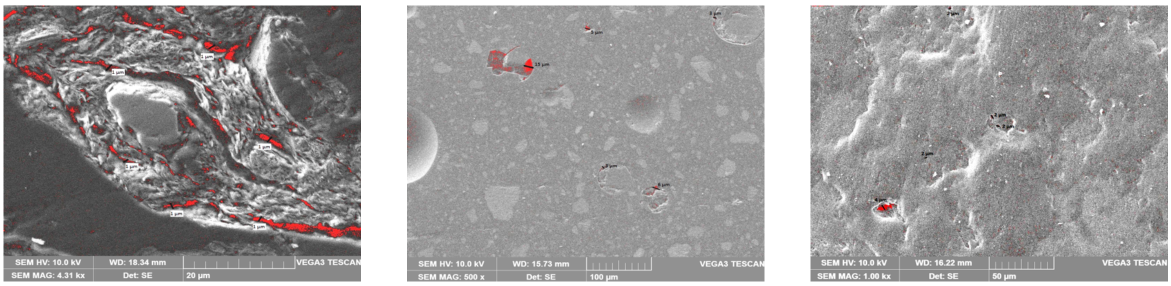
Sample information			Porosity computation	
ID	Type	Name	Porous size (cm^3)	Porosity (%)
0	Steel cylinder	Control	0.12	0
1	Soil	Natural clay	3.37	20
2	Powder	Bentonite	9.17	39
3	Powder	Kaolin	9.28	44

Results of the natural clay sample indicated the porosity in percentage is 20% and this is a fraction of the volume of holes over the total volume which for this material is 3.37 cm^3. According to findings the capacity to absorb water by natural clay is 20% over an estimated volume of 3.37 cm^3, due to the quartz is an important component of its structure with 59.38%. Therefore, this material has low capacity to absorb water since its low liquid and plastic limits.

The capacity to absorb water by kaolin is 44% over an estimated volume of 9.28 cm^3. It can be affirmed that the kaolin has a low capacity to absorb water since this material is composed mainly by quartz. However, the sample was obtained from powder and had to be compacted, due to its level of porosity was induced during the sample preparation.

The capacity to absorb water of the bentonite is 39% over an estimated volume of 9.17 cm^3. It can be inferred that the water adsorption capacity of the material is the highest of this study since montmorillonite is the component of its structure, with 93.43%. The above mentioned is evidenced, clearly, in the values of liquid limit and plasticity index of this soil type.

3.4. Scanning electron microscope
The processing of each micrograph was done using the IMAGE J program, which colors the pores red (interparticle and intraparticle spaces) and microfractures. The program determines those red areas as a percentage of porous area in the rock. Figure 3 presents the final micrographs for each of the samples.



(a) (b) (c)

Figure 3. SEM images: (a) bentonite; (b) kaolin; (c) natural clay.

Figure 3 shows the high porosity of the bentonite sample against the kaolin and natural clay samples. In addition, the structure of each type of soil is observed, highlighting the laminar structure in the bentonite, as well as the high quartz content in the samples of kaolin and natural clay. The laminar structure allows the water cumulation between minerals and quartz content reduces the pore spaces.

On the other hand, it was found that the porosity of the three samples is between 20% and 44%. Nevertheless, SEM results revealed an association of the voids with air bubbles (circular and semicircular figures). Such finding was because of the preparation process, in which the samples manifested small fractures after compaction.Hence, a random distribution of the porous different from the natural voids generation in bentonite and kaolin samples was considered for this analysis.

4. Conclusions
This study proposes an alternative methodology for the characterization of clayey soils. Samples of bentonite, kaolin and natural clay were used. An experimental plan, which included XRF, XRD, SEM and HeP was performed to provide some insights into soil behaviour from the mineralogy and structure of the material. The mineral composition controls the swelling potential of the clayey soils. It was found that for the samples studied, the swelling potential changes according to the proportion of montmorillonite. Besides, such composition has an effect in physical properties such as liquid limit and plastic index. The kaolin and the natural clay present a structure more solid against the bentonite because of mineralogical and

microstructural composition, due to the high quartz content. This composition induces a low porosity and reduces the problems of expansion. Results showed the relevance of evaluating the mineralogical, microstructural and porosimetric characteristics of clays. This test combination allows identifying, simultaneously, the effects in macrolevel of the geotechnical structures caused by the mineralogycal composition and pore size distribution. Therefore, the methodology presented in this document complements the traditional mechanical characterization of this materials.

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