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Study of a mixture of soil and brazilian bentonite using a protocol for clay characterization for near-surface radioactive waste repository

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ABSTRACT

Aiming to implement the Brazilian repository for disposal of low and intermediate level of radioactive wastes, the "Project RBMN" is being coordinated by CNEN. The repository is made of multiple barriers in order to prevent or retain the migration of radionuclides throughout these paths to the environment. Clays are used as backfill and coverage layers, which are the repository natural barriers. Recently, in Nuclear Technology Development Center (CDTN) a protocol for clay characterization was elaborated, and it was used the Brazilian bentonite as a reference. In this protocol are established some physical and chemical tests and their reference values that are essential for the characterization of materials to be used as backfill and coverage layers in the surface repository. This research intended to evaluate the applicability of this protocol to characterize soil without and with bentonite. The bentonite content added to the soil was 30%. The results were compared with the reference values of the protocol and literature for materials with similar mineralogical features. The results showed an increase of the specific surface and of the cation exchange capacity, as the result of the bentonite addition. The hydraulic conductivity is significant decreased by the addition of bentonite.

Keywords: bentonite, soil, repository, radioactive waste, RBMN Project.

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1. INTRODUCTION

Nuclear energy is used in medical, industrial and environmental activities. The radioactive materials generated in these applications, which are no longer useful, are radioactive wastes and they need to be managed properly, in order to avoid possible damages to humans and the environment. The wastes can be classified according to their levels and nature of radiation and half-lives as exempt, very short lived, low and intermediate level and high-level wastes [1,2].

According to the Nuclear Sector and Brazilian Radiological Glossary [3], "radioactive waste is any material resulting from human activities containing quantities of radionuclides higher than the exemption limits established by Brazilian National Nuclear Energy Commission (CNEN – Comissão Nacional de Energia Nuclear), whose reutilization is not appropriate or foreseen." In addition, "repository is a licensed deposit for storage of radioactive wastes, without the intention of removal, in accordance with the criteria of the competent authorities."

In Brazil the radioactive wastes are divided in four classes: 0, 1, 2 and 3. The class 2 is subdivided in other four. Class 2.1 - low and intermediate level radioactive waste of short half-life - includes wastes containing beta or gamma emitters, with half-life of approximately 30 years or less [2]. The greatest volume of wastes generated in Brazil comes from the operation of nuclear power plants, the use of radionuclides in medicine, industry, agriculture and R&D activities, and it is classified as 2.1, being the first ones the biggest generators. In accordance of to CNEN NN 8.02 the disposal for this type of wastes should be in a near-surface repository [4].

The "Project RBMN" is conducted by CNEN, with the objective to construct and to commission the Brazilian Repository for disposing these wastes. Others objectives are to store safely and centrally the radioactive waste generated in the Brazilian territory and to comply a legal requirement from the Brazilian environmental regulator for the operation of the Nuclear Power Plant Angra 3. The life cycle of the repository is expected to be about 360 years, 60 years of operation and 300 years after the closure, called institutional period control [5].

For the Brazilian Repository it was previewed a near-surface disposal with multiple barriers, natural and engineered. This multiple barrier concept is to ensure that there is no contamination of the environment or living beings and that the levels of exposure are below the limits of the radioprotection standards. Their role is to act as physical and chemical barriers to the release of radionuclides [6].

As barrier of a repository, backfill and coverage layers can be used. Backfill is used for a number of purposes: void filling, limitation of water infiltration, sorption of radionuclides, gas control and to facilitate waste retrieval. The backfill and coverage layers may consist of clays or the mixture of clays with cement grout, rock or soil [7].

Bentonite is the term used to designate clays composed mainly of montmorillonite. Bentonite is studied nationally and internationally and has a well established data set. This clay is characterized by high absorption capacity and it shows effectiveness in the retention of contaminants from the product [8]. Brazilian reserves in 2018 were 54.1 Mt, with the following state holdings: 33.6% São Paulo, 49.8% Paraíba and 15.3% Bahia [9].

A clay characterization protocol was elaborated by Santos and Tello [10] at Nuclear Technology Development Center (CDTN – Centro de Desenvolvimento da Tecnologia Nuclear), in which the Brazilian bentonite was used as a reference to standardize the tests and to specify the value range of the parameters. The protocol is a guideline, which presents the main physical-chemical tests to be carried out in order to characterize clays, or mixtures of clays and soils. This characterization is essential to select the materials to use as natural barriers in the near-surface repository.

This present study intends to characterize a soil and its mixture with bentonite according to the protocol elaborated by Santos and Tello [10], verifying its applicability and the effectiveness of these materials for use as backfill and coverage layers in the surface repository for radioactive waste. The applicability of the protocol was assessed by repeating and refining the described physical and chemical tests and considering the need for the inclusion of new assays.

2. MATERIALS AND METHODS

A soil (Sample 0) and a 70%:30% (soil/bentonite, dry mass) mixture (Sample 1) were studied in order to analyze the influence on mineralogical, physical and chemical properties of the soil with bentonite as additive. The soil/bentonite ratio of the mixture was arbitrarily defined, since the definition of an optimal value for this ratio was not important at the moment [11].

2.1. Soil sampling

As the local of the Brazilian Repository is not defined and due to the impossibility of studying all variety of Brazilian soils it was decided to use CDTN as the site for soil collecting (Figure 1). For this decision it was considered the technical and logistic needs for sampling the material and its reproduction [11].

Soil samples were collected at the geographical coordinate points 19°52'36.7" S and 43°58'16.4" W. The procedure consisted of a central point and four other points around it, at a distance of about three or five meters in the direction of the cardinal points (North, South, East, West). Mini-trenches of approximately 20 cm depth and 50 cm diameter were opened at each of the five points.

Once collected, the sample was dried at room temperature, sieved to 2 mm and the fraction < 2 mm was physically disaggregated. Subsequently, for the preparation of sample 0, the material was homogenized in a longitudinal pile and separated in 1 kg fractions. For the preparation of the sample 1, the soil after disaggregated was mixed with the bentonite, homogenized in a longitudinal pile and separated in 1 kg fractions.

Figure 1: *Sample collection locations in the Nuclear Technology Development Center (CDTN).*

2.2. X-ray diffraction

X-ray diffraction is one of the most used structural and microstructural analysis techniques for characterizing crystalline materials and is too most used in phase quantification, determination of unit cell parameters and size of crystallites [12].

The samples for the mineralogical analyzes by X-ray diffraction were prepared following the procedures proposed by Albers et al. [13], in order to identify the filossilicates and iron oxides to identify the type of clay mineral present in the samples. For the X-ray diffraction analysis, it was used the Rigaku D/Max diffractometer of the X-ray Diffraction Laboratory of the Mineral Technology Service (SETEM – Serviço de Tecnologia Mineral) at the CDTN.

2.3. Particle size distribution

Consists of particles of different sizes and this is the first characteristic to differentiate it. To determine the particle size of a soil a granulometric analysis is performed. The soil granulometric analysis consists of determining the size of the particles that it is and its distribution at determined intervals [14].

For the granulometric analysis the method described in procedure CDTN-RT-338 was used [15]. To sequences of sieves of different opening sizes connected to a vibrating shaker were used. The separation of granulometric fractions is performed during 20 minutes. Granulometric distribution curves are represented in percentage by weight of grains of each of the different size fractions.

2.4. Moisture content

Soils are constituted by sets of particles that can have variable diameters and contain water in their empty spaces [14]. Soil moisture or water content is defined as the ratio of the mass of water to the dry soil mass.

To determine the moisture content the method described in procedure CDTN-RT-417 was used [16]. A sample is dried at a specific temperature until reading the constant dry mass, which means that there is no moisture in the sample. The mass of the water is equal to the difference between the weight of the original sample and the weight of the dried sample.

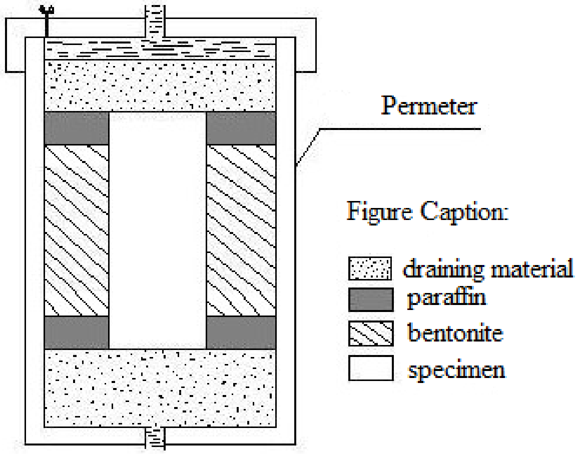
2.5. Compaction curve

The compaction of a soil is its densification by a mechanical equipment [14]. The tests for determination of the compaction curve of the samples were performed according to the procedure of NBR 7.182 of ABNT [17] at the Geotechnical Laboratory of the Federal University of Minas Gerais (UFMG – Universidade Federal de Minas Gerais). The compaction energy adopted was the Proctor Normal, with a 2.5 kg socket, number of layers equal to 3, number of strokes equal to 26 and a drop height equal to 30 cm.

2.6. Hydraulic conductivity

The hydraulic conductivity of a soil is its ability to flow water when submitted to a hydraulic gradient [14]. The tests to determine the hydraulic conductivity of the samples were performed according to the procedures of standards NBR 7.182 and NBR 14.545 of ABNT [17,18] at the Geotechnical Laboratory of the UFMG. For the accomplishment of the tests, the specimen was compacted with the optimum moisture obtained by the compaction curve and the tests were performed using a variable charge permeter (Figure 2).

Figure 2: *Schematic of the hydraulic conductivity test using the variable charge permeter.*



Source: adapted from Batista, 2006 [19]

2.7. Specific surface area

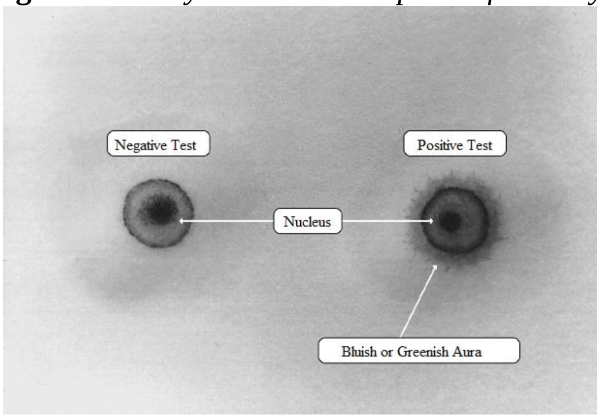
The specific surface is defined as the area of the external surface of the particle, in m^2, per unit of mass, in grams [11]. To determine the specific surface area of the samples, the BET method was used, multiple point technique, Nova-2200 Quantachrome equipment of the Nuclear Fuel Laboratory of the Nanotechnology and Nuclear Materials Service (SENAN – Serviço de Nanotecnologia e Materiais Nucleares) at the CDTN.

2.8. Cation exchange capacity

The cation exchange capacity is a measure of the distribution of electrical charges available on the surface of soil particles for the retention of water and cations dispersed in the soil solution. This parameter indicates the amount of cations that the soil is able to retain under certain conditions and exchange for stoichiometric quantities of other cations [20].

To determine the cation exchange capacity the method described in the internal standard of Cementation Laboratory of the Waste Management Service (SEGRE – Serviço de Gerência de Rejeitos) was used [21]. This test consists of dispersing the material in water and adjusting the pH between 2.5 and 3.8. In sequence a methylene blue aliquot is added into the damp material. Drops of this slurry are put in a filter paper. The appearance of each drop should be observed until emerges a clear halo around the blue drop, indicating the end point, as show in Figure 3.

Figure 3: *Methylene blue adsorption of the clay.*



Source: adapted from Fabri, 1994 [22]

2.9. Particle density

The particle density is an important physical-chemical parameter used in the characterization of materials and is defined as the ratio between mass and volume occupied by a corp or substance [11]. The tests for the determination of the particle density of the samples were performed with the

Ultrapycnometer Quantachrome, Multipycnometer model, of the Nuclear Fuel Laboratory of the SENAN at the CDTN.

3. RESULTS AND DISCUSSION

The diffractograms of samples 0 and 1 and the identification of the minerals found in the respective peaks are shown in Figure 4 and 5, respectively. The minerals identified in sample 0 were kaolinite and quartz, and in sample 1 were kaolinite, montmorillonite, quartz and goethite.

Figure 4: *Diffractogram of sample 0.*

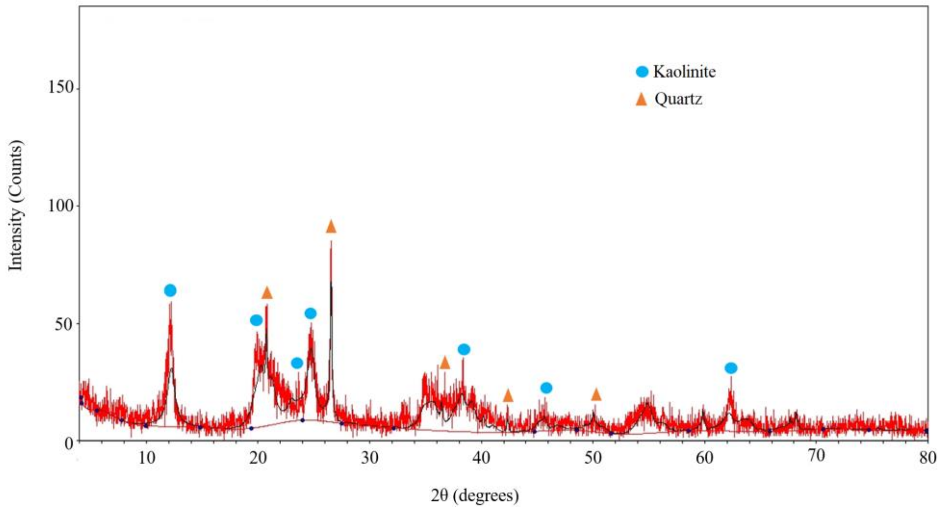
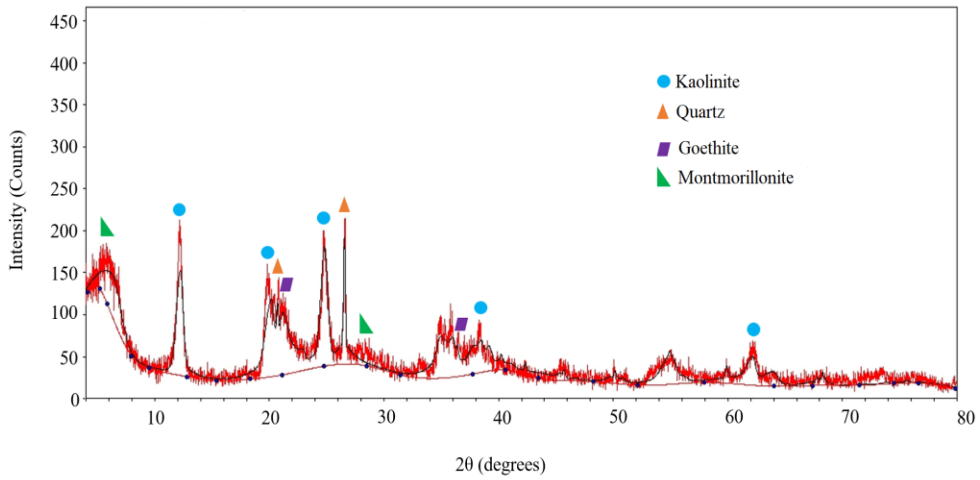


Figure 5: *Diffractogram of sample 1.*

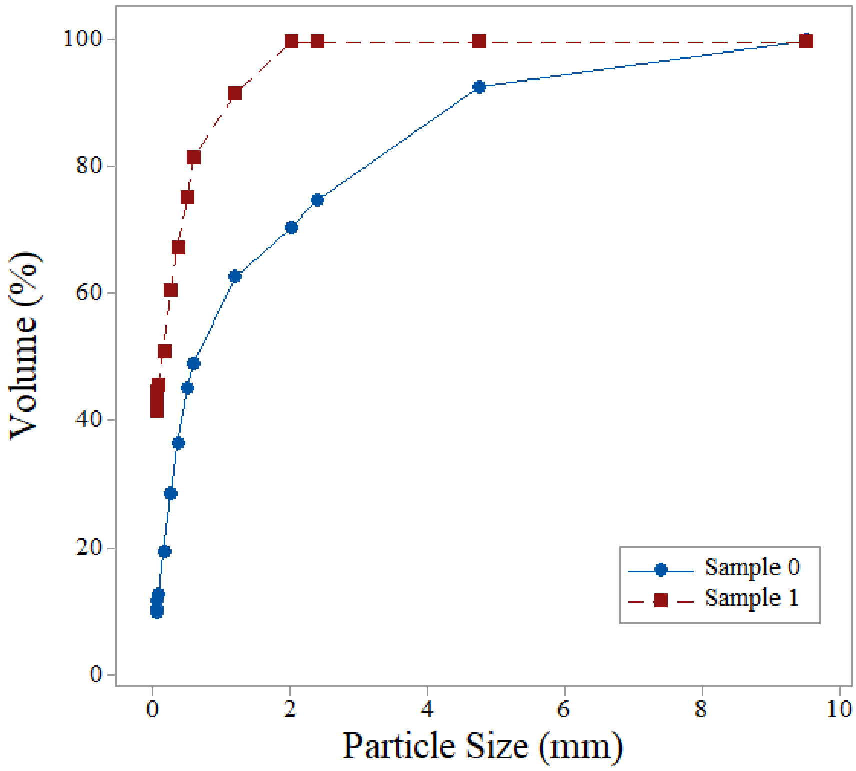


As expected, in the sample 1 the argillomineral montmorillonite was identified, due to adding bentonite to the soil. According to Santos and Tello the bentonite sample is a clay composed predominantly of montmorillonite [10].

Kaolinite is the main clay mineral present in most of tropical and subtropical acid soils, jointly with iron and aluminum oxides [23]. According to IBGE the area of occurrence of tropical soils corresponds to 2.34 billion hectares, of which 790 million are sited in Brazilian territory, corresponding to 87% of Brazil's surface soils [24].

The results of the particle size distribution of samples 0 and 1 are showed in Figure 6. The Table 1 showed the results of the others properties studied in each of the samples.

Figure 6: *Particle size distribution of sample 0 and 1.*



It’s can be observed by particle size distribution curve that in sample 1 the clay + silt content (<0.06 mm) increased due to adding bentonite to the soil. According to Santos and Tello all bentonite sample is in the clay + silt fraction [10]. Thus, in sample 1 the 30% of bentonite added to the soil became of the clay + silt fraction in its granulometric distribution.

Table 1: Physical and chemical properties of samples 0 and 1.

Test	**Results**	
	Sample 0	**Sample 1**
Moisture content	2.2%	5.1%
Compaction curve – Maximum dry density	1.44 $g.cm^{-3}$	1.38 $g.cm^{-3}$
Hydraulic conductivity	$1.1503x10^{-8}$ $cm.s^{-1}$	$4.3556x10^{-9}$ $cm.s^{-1}$
Specific surface area	11.56 $m^2.g^{-1}$	26.19 $m^2.g^{-1}$
Cation exchange capacity	14.89 $mmol.kg^{-1}$	228 $mmol.kg^{-1}$
Particle density	2.58 $g.cm^{-3}$	2.60 $g.cm^{-3}$

Comparing the results of moisture content, specific surface area and cation exchange capacity of sample 0 and of sample 1, an increase in the values is observed. Those results were expected because bentonite has high values for these properties [10]. The increase in the cation exchange capacity value for the sample 1 implies an increase in its ability to react with the surrounding environment, which is desirable for use in engineered barrier systems of repositories.

There was a three-fold reduction in the value of the hydraulic conductivity from sample 0 to sample 1. The hydraulic conductivity is important for choosing the material of the engineering barriers for a radioactive waste repository, because this requires high sealing capacity, consequently a low hydraulic conductivity.

4. CONCLUSION

The clay characterization protocol described in this work can be applicable for evaluation of materials used as backfill and coverage layers in the surface repository for radioactive waste.

The addition of bentonite to the soil was efficient for improve the investigated properties. Increase of the fine fraction soil, specific surface area and cation exchange capacity and reduction of the hydraulic conductivity, making its possible use in engineered barrier systems of repositories.

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