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Characterization of calcium bentonite to use as a natural barrier in a surface repository

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1. Introduction

The nuclear technology used in Brazil in different areas generates radioactive waste that is mainly classified as low and intermediate level, which requires efficient management in order to maintain the safety of the environment, human beings and future generations [1]. CNEN has standards that establish how to properly manage these wastes and their disposal in a repository and coordinates the CENTENA Project (RBMN Project), which will implement in Brazil the near-surface repository using multi-barriers, considering that which one is self-sufficient to avoid and to delay the output of radionuclides [2].

Regarding natural barriers, the International Atomic Energy Agency (IAEA) reported that clays have good applicability in the base, filling and covering layers for repositories [3]. Among the clays, bentonite was chosen for this work, for being internationally studied for use in natural barrier of different repositories, for having good sorption capacity for several radionuclides and low hydraulic conductivity, continuing a research, at the CDTN, started with a clay characterization protocol [4], in order to use this material as a natural barrier at CENTENA, the first Brazilian repository for low- and intermediate-level radioactive waste.

Generally, clays are incorporated into the soil of the repository site to improve the sorption and retention of radionuclides present in the radioactive waste. Mixtures of bentonite and soil was studied, since the CENTENA site is in the final selection phase, the CDTN site soil was chosen for the study. Barroso performed its characterization and also of the mixture containing this soil with 30% sodium bentonite, in order to determine the influence of this mineral clay on the properties: particle size distribution, moisture content, specific surface, particle density, exchange capacity cationic, compaction curve and hydraulic conductivity. The soil evaluated properties were improved with the bentonite addition [5].

Based in these results, another research was carried out to characterize calcium bentonite mineralogical and physiochemically. This paper summarizes the results of this study, and presents a proposition of a work to study different addition of bentonite to the soil, in order to optimize economically and operational feasibility the use of bentonite in the natural barrier system of the repository.

2. Methodology

The methodology used for both studies, with calcium bentonite and with different mixtures of soil and sodium bentonite, follows the Protocol for characterization of clays, which was also efficient for soils and mixtures [4] and [5]. Then, mineralogical analyzes and physicochemical characterization were carried out with calcium bentonite and they will be the same used to study two mixtures of soil, with 8% and 15% of sodium bentonite, respectively.

The mineralogical analysis tests are X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM). The first one is to identify the phyllosilicates and iron oxides and determine the type of clay, using the Rigaku D/Max diffractometer from the SETEM X-ray Diffraction Laboratory - Mineral Technology Service at CDTN. The morphology and qualitative chemical composition of the surface of are analyzed using a scanning electron microscope with field effect emission - FEG-MEV - of the Sigma VP model, from the Nanotechnology and Nuclear Materials Service - SENAN at the CDTN.

The physical-chemical characterization tests are: the moisture content, according the TR 0417 [6]; the cation exchange capacity (CTC) by the methylene blue adsorption method, following NI-SEGRE-01 [7]; the particle size analysis, by laser diffraction with Cylas equipment; particle density, using the Ultrapycnometer Quantachrome, Multipycnometer model, and the specific surface, by the BET multiple point method with Nova-2200 Quantachrome, these tree ones performed at LABCON/SENAN/CDTN.

3. Results and Discussion

In this paper the results of the characterization of calcium bentonite are presented. The results from the proposed work using the mixtures of soil and 8% and 15% of sodium bentonite, respectively, will be compared with those presented by Barroso [5], who studied soil with addition of 0% and 30% of Na-bentonite, and then to verify the effect of each addition in the properties.

3.1 X-Ray Diffraction Analysis (XRD) and Scanning Electron Microscopy (SEM)

In Fig. 1, the diffractogram shows that the sample composition is 66.4% nontronite, 16.5% quartz, and 8.1% montmorillonite, and 9% related to the amorphous part of the material. Nontronite and montmorillonite have more intense peaks that are very coincident, so the defined content values are estimates without accuracy for these species in the sample. As it is bentonite, the presence of montmorillonite was expected and nontronite belongs also to this group of clays. Scanning electron microscopy analysis has not yet been performed, but it is expected that the result will evidence the presence of characteristic particles of the structure of nontronite and montmorillonite, which morphology is lamellar.

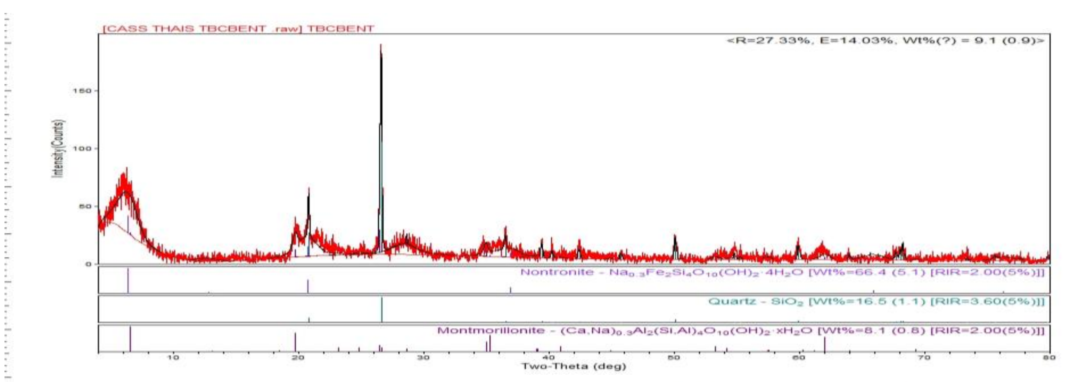

Figure 1: Diffractogram of Ca-bentonite.

3.2 Particle size analysis

The results of particle size distribution analysis by laser diffraction are in Fig. 2. It was identified that

40% of the measured sample presented an average particle size between 8.0 and 10.0 µm. According to the limits of soil fractions by grain size defined by ABNT [8] it is the clay fraction, which was expected since bentonite is predominantly constituted of clay minerals.

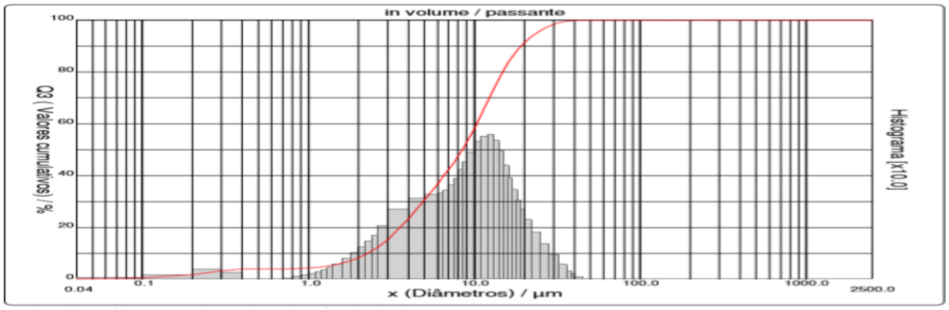

Figure 2: Particle size analysis of bentonite by laser diffraction.

3.3 Moisture content

The global average of bentonite moisture was 14%. Eq. 1 was used to calculate the individual moisture content (A).

$$A = \left[\frac{M_{rau} - M_{ras}}{M_{ras} - M_r}\right] x\ 100 \qquad (1)$$

Where M_{rau} is the mass of the container with the wet sample; M_{ras} is the mass of the container with the dry sample, and M_r is the mass of the empty container.

3.4 Determination of Specific Surface and Particle Density

The results for the specific surface and particle density are presented in Table I.

Table I: Specific surface and particle density results.

Test	Result
Specific surface (m^2/g)	82.741
Particle density (g/cm^3)	2,5287 ± 0,0057

3.5 Cationic Exchange Capacity (CTC)

Fig. 3 shows the appearance of the droplet at the end of the assay for CTC determination. For the calculation of CTC, in $mmol.kg^{-1}$, is used the Eq. 2, in which C_{AM} is the concentration of methylene blue, in $mol.\ L^{-1}$, V_{AM} is the spent volume of methylene blue, in ℓ, and m_a is the dry mass of the sample, in kg. The result was 750 ± 18.71 $mmol.kg^{-1}$.

$$CTC = C_{AM}\ x\ V_{AM}\ x\ \frac{1000}{m_a} \qquad (2)$$

Figure 3: Droplet appearance in the methylene blue adsorption process for CTC determination.

4. Conclusions

The calcium bentonite has good properties as natural barrier for the radioactive waste repository. The techniques and the tests developed will be used in the future work with mixtures of soil and clays to determine the optimum ratio to be used in CENTENA.

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