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Characterization of expanded vermiculite to use as natural barrier in repository

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ABSTRACT

Brazilian National Nuclear Energy Commission – CNEN – is responsible to build a repository for the low and intermediate--level radioactive wastes generated in Brazil. In accordance of Brazilian legislation it will be a near surface one. The treated wastes will be disposed of at the repository, which will be surrounded by engineering and natural barriers, in order to prevent the release of radionuclides, during the required time to their decay to the limit levels. The focus of this study is to characterize the expanded vermiculite, which can be used as a natural barriers, over and under the engineering barriers and also as a backfill. They have the function of limiting water infiltration and stabilizing the disposal system, filling the empty spaces, controlling the gases and delaying the release of radionuclides. In this paper is presented the characterization of an expanded vermiculite. The tests to determine its physical chemical properties were based on a clay characterization Protocol. The granulometric analysis was performed using a sieving method. The moisture content was established by the gravimetric method of weight loss, and the specific surface area by the BET method. The helium ultrapicnometer method was used to determine the density. The microstructure was analyzed by X-ray diffraction and scanning electron microscopy. The results indicate that the studied vermiculite would be efficient as a natural barrier of the Brazilian surface repository.

Keywords: **Expanded vermiculite, clay characterization, repository, protocol.**

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1. INTRODUCTION

In Nuclear energy, there is a challenge regarding the management of its radioactive wastes, in order to avoid environmental impacts [1]. Brazilian National Nuclear Energy Commission – CNEN – is responsible to build a repository for the low and intermediate-level radioactive wastes generated in Brazil [2]. Focusing on their goals, R&D activities are being developed to support this implementation.

Clays are used as waterproofing barriers, mainly due to their good sorption capacity for several radionuclides, and their low hydraulic conductivity. A Protocol was prepared by Santos [3] with the purpose of standardizing tests to characterize clays, using the bentonite as reference. This characterization aims to know how the properties of materials are to select those that can be used as natural barriers in the repository. The role of these barriers is to slow down and prevent the output of radionuclides to the environment. They also have the function of limiting water infiltration and stabilizing the disposal system.

Expanded vermiculite is a clay, which expands when subjected to temperatures close to 1,000 ^{o}C and has low acoustic conductivity, low thermal conductivity, high cation exchange capacity, and it retains water up to five times its weight. Such characteristics are important in natural barriers of the repository [4]. In surface repositories, the expanded vermiculite would be applied in the barriers over and under the engineering barriers, as well as in the backfill, filling the voids, controlling the gases and delaying the output of radionuclides.

This paper presents the study that were performed to characterize the expanded vermiculite. The following physicochemical properties were determined: granulometry, moisture content, specific surface area and density. In addition, the microstructure was analyzed by X-ray diffraction and scanning electron microscopy.

2. MATERIALS AND METHODS

The material used for the tests was a vermiculite in its expanded form, available in Brazilian territory.

2.1. XRD and SEM

The samples were prepared for X-ray diffraction analysis (XRD) and scanning electron microscopy (SEM) [5]. Both analyses were performed at Nuclear Technology Development Center (CDTN). For the first one it was used a Rigaku D / Max Plus diffractometer of the X-ray Diffraction Laboratory of SETEM (Mineral Technology Service), and for the second one a scanning electron microscope with field effect emission – FEG-MEV – of the Sigma VP model, manufactured by Carl Zeiss Microscopy, of the Nanotechnology and Nuclear Materials Service – SENAN.

2.2. Granulometric analysis

For the analysis, it was followed the procedure described in the Technical Routine CDTN-338 [6], using 50 g of super thin vermiculite and sieves of Tyler series, varying between 9 # and 400 #. After cleaning and weighting them, they were stacked from the smallest to the largest opening. The sieves are agitated for 20 minutes. The 9 # to 28 # sieves were manually shaken to ensure that the grains, which were concentrated in the middle of the sieve, were not forcing the passage of others. The other sieves were put in the stirrer, as shown in Figure 1.

Figure 1: *The stirrer with the sieves*

Source: Author

2.3. Moisture content

For the experiment, the Technical Routine CDTN-417 was followed [7]. The samples were prepared in triplicate (AM1, AM2, AM3), and for each one 20 g of superfine vermiculite were used

(Figure 2 and 3). They were placed in a lab oven at a constant temperature of 110 ± 5 ^{o}C to dry for 16 hours. After that, they were remained in a desiccator for at least one hour, until they have constant mass, indicating that all the water was evaporated.

Figure-2: *Materials for moisture determination*

Figure-3: *Crucibles with clay*

Source: Author

2.4. Determination of specific surface

The BET method was used to determine the specific surface area [8]. Samples containing 2 g of superfine vermiculite were subjected to the degassing process, so that their surfaces were free of volatile substances, the process took place inside the equipment for 2 hours at 100 ^{o}C. Then the sample was kept at -196 ^{o}C while N_2 gas was added to be adsorbed on the sample surface and pores. The multiple point technique was used and Figure 4 shows the equipment used, Nova-2200 Quantachrome at the Nuclear Fuels Laboratory of the nanotechnology service – LABCON / SENAN / CDTN.

Figure 4: *Specific surface area analysis*

Source: Author

2.5. Determination of particle density

The experiment started with the preparation of the sample, which was previously dried in the oven at 100 ^{o}C and was cooled in a desiccator. Then, the sample was directed to the electronic densimeter, Ultrapycnometer 1000 from Quantachrome Instruments, available at the Nuclear Fuel Laboratory (LABCON / SENAN / CDTN) [9], as shown in Figure 5. The equipment accurately measures the actual density of solid materials by the principle of displacement of an Archimedes fluid and the expansion of gases in Boyle's Law. Helium gas penetrated the finest pores and eliminated the influence of chemistry on the surface [10]. The equipment recorded the density readings and ended when presents three consecutive values with an error less than 0.005 $kg.m^{-3}$. In this test, 20 readings were taken.

Figure 5: *Ultrapicnometer Quantachrome*

Source: Author

2.6. Compaction Curve

For this test, the standard NBR 7182 [11] was used, which describes the method for the relationship between the moisture content and the apparent density of the soils when compacted. The method consists of using five samples with different humidity to draw the compaction curve, which has a maximum point that indicates the ideal humidity. First, the samples were moistened, and then the compaction was performed with 26 strokes of the Proctor Normal compaction cylinder. The amount of water for each sample was calculated using equation 1. M_s, M_T and w represent the mass of the solid, total mass and moisture, respectively.

$$M_s = \frac{M_T}{(1 + w)} \tag{1}$$

3. RESULTS AND DISCUSSION

3.1. XRD and SEM

From XRD analysis, biotite and muscovite were the main peaks identified, both minerals from the micas group. The figure 6 presents this result: 62.5% biotite, 37.5% muscovite, 0.8% vermiculite, showing that the expanded vermiculite studied is different of the vermiculite in natura, since when the expansion occurs, some characteristics and even minerals are changed due to high temperature [5]. SEM analysis confirms that expanded vermiculite has lamellar morphology (Figure 7).

Figure 6: XRD result

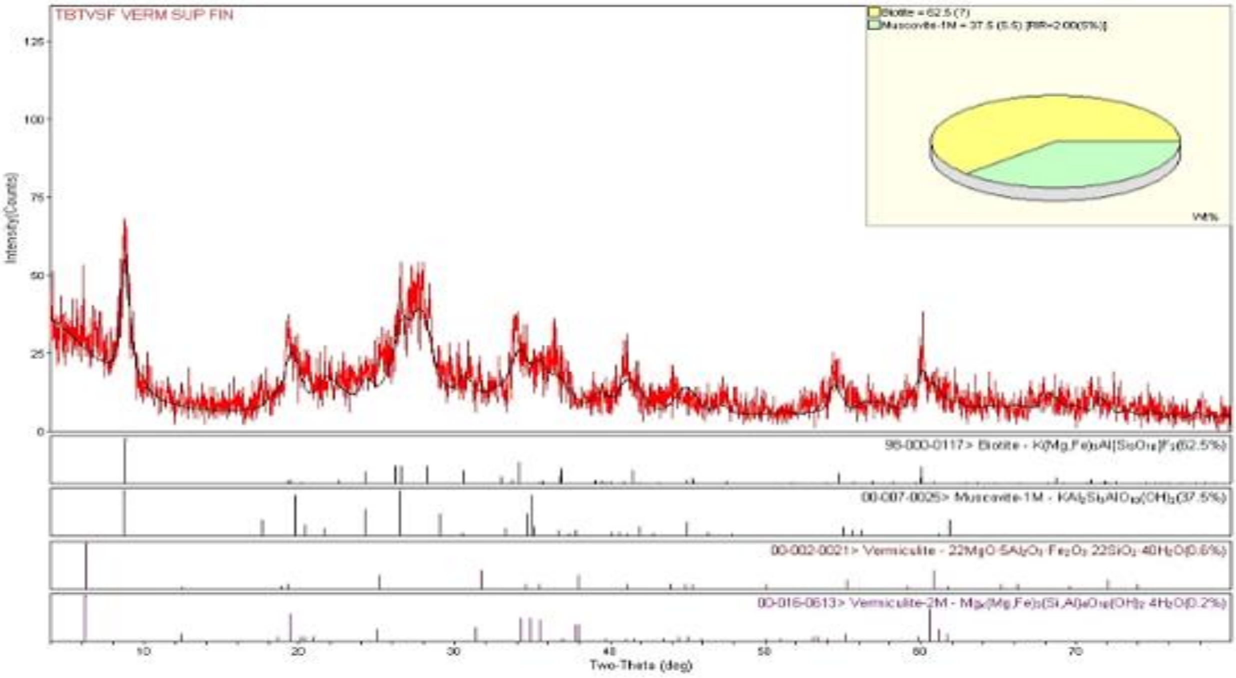
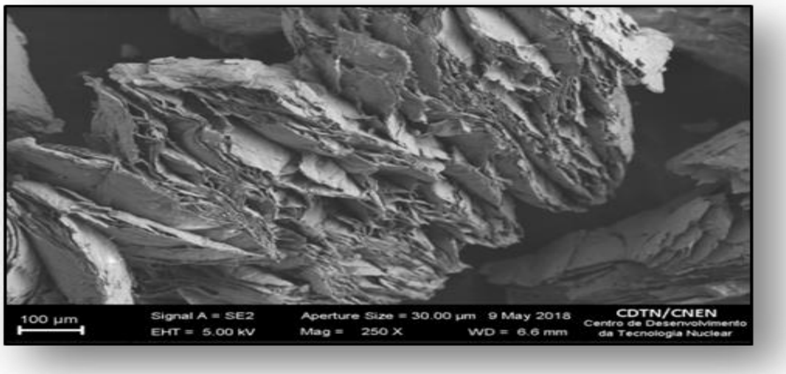


Figure 7: *Morphology showed by SEM*



3.2. Granulometric analysis

The result of the particle size test is shown in the Table 1 with the percentage of grain passed through certain openings of the sieve in mm. Between the sieves of 0,500 and 0,297mm there is greater variation of the percentage passed, which indicates a predominance of these grain size of the studied clay.

Table 1- Vermiculite particle size

Vermiculite

Opening (mm)	*% Passed*
2.00	99.61
1.19	97.66
0.850	91.24
0.590	79.96
0.500	73.35
0.297	47.09
0.210	33.86
0.150	23.74
0.106	16.54
0.075	12.65
0.053	10.51
0.045	6.43
0.038	3.12

3.3. Moisture content

After the tests, the moisture content was calculated resulting in a value of 1.54% ± 0.16.

3.4. Determination of specific surface

The specific surface determined by Nova-2200 Quantachrome was 6.2195 $m^2.\ g^{-1}$.

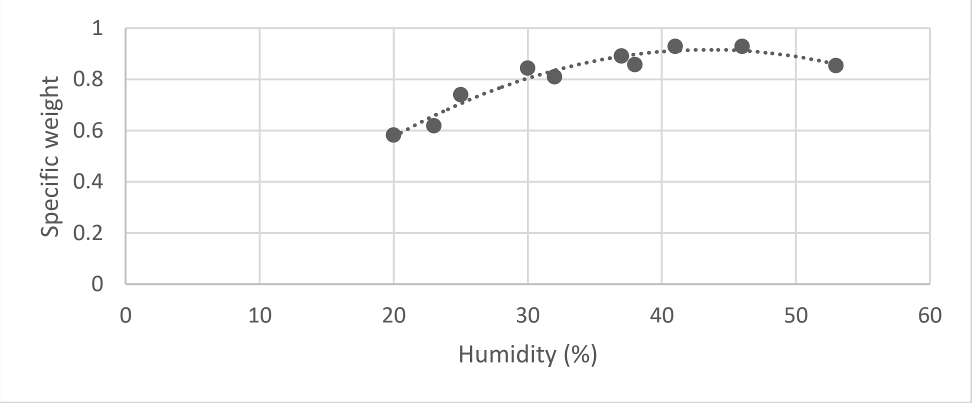
3.5. Determination of particle density

The particle density determined by Quantachrome Instrument was 1.9260 ± 0.0070 $kg.m^{-3}$.

3.6. Compaction Curve

The compaction curve is presented in Figure 8, and the optimum humidity was 44.7% represented by its maximum point. This value was calculated using equation 2 through the polynomial regression (equation 3), evidenced by the trend line.

Figure 8: *Compaction curve*



$$Optimum\ humidity = \frac{-b}{2a} \tag{2}$$

$$\textit{Linear regression equation} = -0.0006x^2 + 0.0536x - 0.2494 \tag{3}$$

The type of absorption curve was different from that in the protocol [3] and in the literature [12], in which only five humidity points are enough to obtain this curve. For the studied clay, it was necessary ten points to achieve this result, because the vermiculite was an expanded one and not *in natura*.

To understand the absorption behavior of the expanded vermiculite, different amounts of water were calculated in order to moisten clay samples. It was verified that the moisture content values obtained after the experiment were below the calculated for all samples. The variation between the actual and the calculated values was between 11% and 24%.

4. CONCLUSIONS

The expanded vermiculite was characterized according to the clay characterization protocol. This protocol was based on a Brazilian sodium bentonite. During the study, it was necessary to improve the Protocol, including experiments to determine the density of particles and the specific surface area by the ultrapicnometer and BET method, respectively.

In addition, it was verified, through the compaction curve experiment, that the expanded vermiculite has a different water absorption behavior in comparison of clays *in natura*. In spite of this, the optimum moisture value indicated that the use of expanded vermiculite as a natural barrier in the surface repository will be beneficial, since the objective of the barrier is to minimize the release of radionuclides.

The other experiments also showed satisfactory results, based on the results of the reference clay Protocol.

Therefore, it was concluded that vermiculite in its expanded form will be efficient for use in the national repository.

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REFERENCES

[1] CNEN – Comissão Nacional de Energia Nuclear. **Gerência de Rejeitos Radioativos de Baixo e Médio Níveis de Radiação.** Resolução 167/14, CNEN NN 8.01. Rio de Janeiro, Rio de Janeiro, Brazil. 2014. Available at: < http://appasp.cnen.gov.br/seguranca/normas/pdf/Nrm801.pdf >. Last accessed: May 2019.

[2] TELLO, C.C.O. “Implementation of the Brazilian national repository”. In: **INTERNATIONAL NUCLEAR ATLANTIC CONFERENCE**, 2013, Recife. Annals… Recife: Comissão Nacional de Energia Nuclear, 2013.

[3] SANTOS, D. M. M. **Elaboração de protocolo para caracterização de argilas como material de recheio e de cobertura para repositório de superfície**. Dissertação de Mestrado em Ciência e Tecnologia das Radiações, Minerais e Materiais, Belo Horizonte: Centro de Desenvolvimento da Tecnologia Nuclear, CDTN, Belo Horizonte, 111p, 2017.

[4] UGARTE, J. F. O.; SAMPAIO, J. A.; FRANÇA, S. C. A. **Vermiculita**. Minerais CETEM. Avaiable at : <:http://mineralis.cetem.gov.br/bitstream/cetem/1142/1/38.%20VERMICULITA%203%20sampaio.pdf>. Last accessed in May 2019.

[5] MARCOS, C; ARANGO, Y. C. C.; RODRIGUEZ, I. X-ray diffraction studies of the thermal behavior of commercial vermiculites. **Applied Clay Science**, 2009. Elsevier Available at: <https://www.sciencedirect.com/science/article/pii/S0169131708000562>. Last accessed: April 2019.

[6] CENTRO DE DESENVOLVIMENTO DA TECNOLOGIA NUCLEAR. **Análise granulométrica de minérios**.. Rotina Técnica – RT 0338 (CT5) LABCIM/CDTN, Belo Horizonte – MG, CDTN, 2001.

[7] CENTRO DE DESENVOLVIMENTO DA TECNOLOGIA NUCLEAR. **Determinação do teor de umidade em argilas**. Rotina Técnica – RT 0417 (SN1) LABCIM/CDTN, Belo Horizonte – MG, CDTN, 2005

[8] TEIXEIRA, V. G.; COUTINHO, F. M. B.; GOMES, A. S. Principais métodos de caracterização da porosidade de resinas à base de divinilbenzeno. **Química Nova**, v. 24, n. 6, pp. 808-818, 2001.

[9] OLIVEIRA, Jr. M. et al. Projeto e construção de um picnômetro a ar para caracterização de insumos e produtos farmacêuticos. **Química Nova**, v. 33, n. 6, p. 1384-1388, 2010.

[10] QUANTACHROME Gas Pycnometers. Available at: <em<https://www.quantachrome.com/pdf_brochures/07171_Gas_Pyc.pdf>. Last accessed: 10 Nov. 2018.

[11] ASSOCIAÇÃO BRASILEIRA DE NORMAS TÉCNICAS. ABNT NBR 7.182: S**olo: ensaio de compactação.** Rio de Janeiro, 9 p. 2016 (New version: 2020).

[12] DIAS Jr, M. S.; ESTANISLAU, W. T. Grau de compactação e retenção de água de latossolos submetidos a diferentes sistemas de manejo. **Rev. Brasileira de Ciência do Solo**, v. 23, n. 1, 1999, pp. 45-51 Sociedade Brasileira de Ciência do Solo Viçosa. 1999.