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Comparison of concrete decalcification tests

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1. Introduction

Currently, nuclear energy corresponds to 10.4% of the world's electricity production [1] and 2.5% of Brazil's domestic supply [2]. Nuclear fuels are a promising energy source for being efficient, safe, profitable and practically free of greenhouse emissions. Due to these advantages, the 2030 National Energy Plan aims to expand the installed capacity of the country's nuclear power plants to 4,000 MW [3], a challenge that demands projects focused on radioactive waste management (RWM). A material is defined as radioactive waste when it has radionuclide concentrations above the exemption limits and have unforeseen or improper reuse [4]. The administrative and technical strategies from the collection to the final and safe disposal of these wastes make up the RWM [5]. Thus, the Centro Tecnológico Nuclear e Ambiental (CENTENA) will act as the licensed institution and responsible for the disposal of low- and intermediate-level radioactive waste, as well as for RD&I activities in this area.

For the safe disposal of radioactive waste for the environment and the public, the principles of multi-barriers are adopted using different permeable and impermeable confinement materials, such as concrete [6,7,9], as shown in Fig. 1. Thus, concrete engineered barriers minimize and prevent the dispersion of radionuclides, as in containers for packaged waste [6,8], which are of particular interest in the construction of CENTENA.

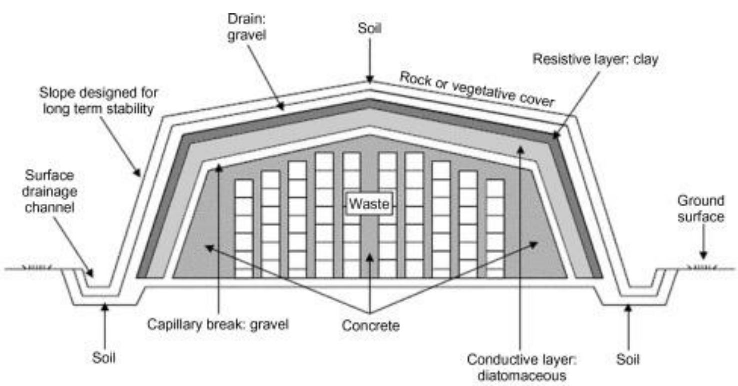

Figure 1: Schematic of a near-surface disposal facility for radioactive waste similar to the structure of the CENTENA repository [9].

However, for use in engineered barriers, concrete must be qualified for integrity, durability and shielding capacity. For this, different mechanisms that affect its properties must be studied, one of them being the leaching, which consists in the extraction of ions by dissolution [7]. For concrete, calcium leaching – decalcification – has a great impact on the service life required for radioactive nuclear facilities, being permeability, porosity and mass transport properties standing out as the most affected by it [7,10]. Thus, over the last few years, several authors have used leaching tests to measure the quality parameters of concrete used in radioactive waste repositories [5,6,8]. Therefore, this work aims to compare two studies seeking to have a reference for tests for the concrete under development for use at CENTENA.

2. Methodology

It was searched for papers of the last decade focused on concrete decalcification, which could be applied to the study of the repository. Using the keywords *calcium leaching*, *concrete* and *repository* and the period 2011-2021, many articles were obtained from which 10 were selected for study. Among these, two were chosen for this work: one focused on mathematical modeling and the other for its experimental part in the laboratory. From the study of these articles, three criteria were qualitatively compared:

- Scope (characteristics of the analyzed concrete and considerations imposed for the study);
- Experimental methodology and main results related to leaching;
- Potential contributions to the development of the concrete used in the CENTENA repository.

3. Results and Discussion

The chosen articles are "*Modeling the effect of temperature gradient on moisture and ionic transport in concrete*", by Bai et.al., published on the Cement and Concrete Composites journal (2020) [11], and "*Decalcification of cement paste in NH_4NO_3 solution: microstructural alterations and its influence on the transport properties*", by Phung et.al., published on the 10th fib International PhD Symposium in Civil Engineering (2014) [12]. The three compared criteria for each of them will be discussed below.

3.1 – Scope:

Bai et.al. [11] studied Ordinary Type 1 Portland cement, with coarse and fine aggregates with average sizes of 12.7 mm and 2 mm, respectively, water/cement ratio (w/c) of 0.6 and cure time of 28 days, using the experimental set-up shown in Fig. 2. Phung et. al. [12] evaluated two Ordinary Type 1 Portland cement pastes with w/c ratios of 0.325 and 0.425. In both works, the diffusion analysis was one-dimensional

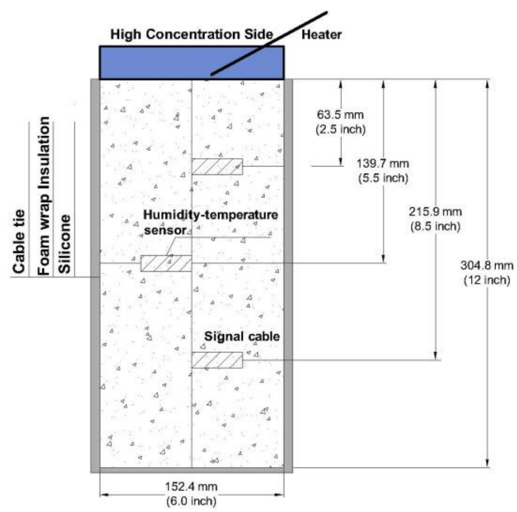

Figure 2: Schematic of the experimental set-up proposed by Bai et.al [11].

3.2 – Experimental methodology:

Bai et.al. [11] methods consisted of modeling an analytical solution to the mass transfer problem starting from the first modified Fick's Law to predict the free chloride concentration over time. The proposed model fitted well with the experimental data and allows to observe that the concentration of free chloride tends to be higher with the temperature gradient, as shown in Fig. 3. On the other hand, Phung et.al. [12] used accelerated concrete leaching tests, in which the samples were immersed in a NH_4NO_3 solution. Among the various analyzes performed in the work, mercury intrusion porosimetry (MIP), nitrogen adsorption (BET) and scanning electron microscopy (SEM) stand out. A linear behavior of the cementitious matrix decalcification with time was observed, and the samples with a lower w/c ratio had a slower degradation speed (0.97 mm/day). An increase of up to 38% in porosity was also observed, which was confirmed by SEM analyzes (Fig. 4), that shows in detail that calcium leaching can create interconnected preferential pathways that increase percolation in the pore system.

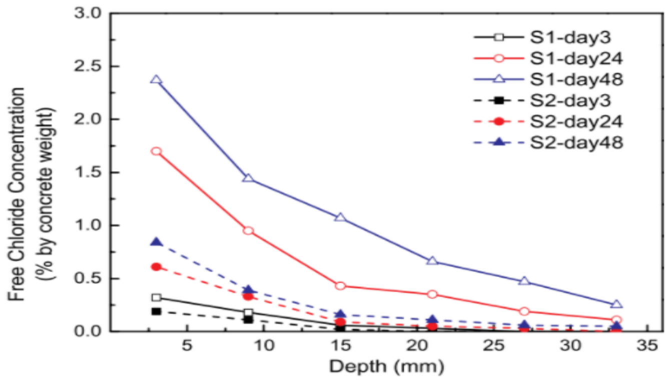

Figure 3: Chloride concentration profile with (S1) and without (S2) a temperature gradient [11].

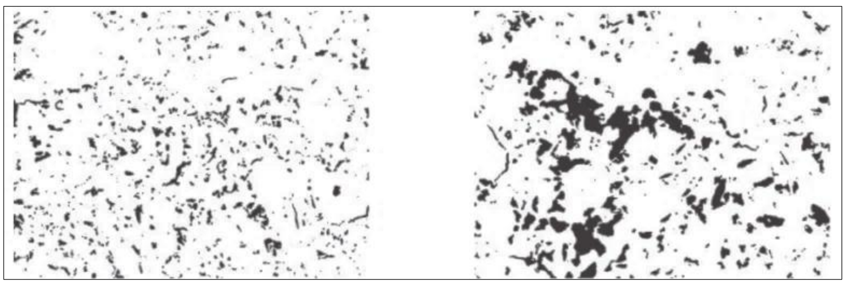

Figure 4: Pores (black) of reference sample (left) and leachate (right) (adapted) [12].

3.3 – Potential contributions:

Although both studies evaluated do not deal specifically with concrete for repositories, the contributions are quite expressive. Bai et.al. [11] successfully validates the use of mathematical models as a reliable means to predict the behavior of concrete under conditions that may be similar to those of the environment in which the CENTENA project repository will be built. These models can be adjusted according to other types of concretes to simulate the leaching, since deviations due to changes in the composition are corrected. Contributions by Phung et.al. [12] have great experimental applicability for the concrete development for CENTENA. There are a number of good practices for leaching tests, such as the preparation of samples with epoxy waterproof resin for one-dimensional analysis and the injection of nitrogen to inhibit carbonization. Another point that stands out is the combined use of instrumental techniques to obtain complementary results that provide rich details about the microstructure. Furthermore, in subsequent studies, the same authors evaluated the adherence of experimental data to mathematical models as a function of porosity and concentration of calcium and nitrate ions, obtaining promising results [13].

Overall, the authors of both papers conclude that calcium leaching substantially affects the integrity of the concrete and its durability. Additionally, it was also observed that ion transport is highly dependent on moisture and temperature, which reiterates the importance of evaluating such properties together, and that mathematical models can be reliable tools for studying this phenomenon.

4. Conclusions

The phenomenon of decalcification in the cementitious matrix is a fundamental parameter to determine the integrity and durability of the near-surface repository structure, especially considering the environmental conditions it needs to withstand. For this reason, accelerated leaching tests must be part of the quality parameters validation for the composition chosen for the CENTENA repository. The comparison presented in this work allows to conclude that mathematical models can be used as a complement to these tests, providing a way to measure and qualify the microstructural changes of the material over time and under different environmental conditions. Since decalcification is closely linked to the durability of concrete, it is suggested for future research to study the transport of ions through the cement matrix to assess its shielding capacity, as other authors have proposed in relation to diffusion [14] and adsorption [15] of cesium, for example.

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References

[1] BP. Statistical Review of World Energy. 2020, 69th ed.

[2] MINISTÉRIO DE MINAS E ENERGIA. Resenha energética brasileira: ano base 2020. Secretaria de Planejamento e Desenvolvimento Energético, Brasília, 2021.

[3] ELETROBRAS. Eletronuclear: Nossas atividades - Angra 3. 2019. Disponível em: <http://www.eletronuclear.gov.br/Nossas-Atividades/Paginas/Angra-3.aspx>.

[4] CNEN. Glossário do Setor Nuclear e Radiológico Brasileiro. Diretoria de Radioproteção e Segurança Nuclear, Rio de Janeiro, 2021.

[5] LOPES, I.S.R.F.S. Estudo do critério de resistência à lixiviação para a aceitação de rejeitos radioativos de baixo e médio nível para disposição (thesis). Programa de Pós-Graduação em Ciência e Tecnol. das Radiações, Minerais e Materiais do Centro de Desenv. da Tecnol. Nuclear - CDTN, Belo Horizonte, 2018.

[6] TAVARES, B.L., TELLO, C.C.O. Concrete containers in radioactive waste management: a review. Brazilian Journal of Radiation Sciences, 07-02A, p. 01-17, 2019.

[7] PHUNG, Q.T., MAES, N., JACQUES, D. Current concerns on durability of concrete used in nuclear power plants and radioactive waste repositories. Proceedings of the 4th Congrès International de Géotechnique - Ouvrages -Structures, 2018.

[8] TAVARES, B.L. Contêineres de concreto para acondicionamento de rejeitos radioativos (thesis). Programa de Pós-Graduação em Ciência e Tecnol. das Radiações, Minerais e Materiais do Centro de Desenvolvimento da Tecnol. Nuclear - CDTN, Belo Horizonte, 2019.

[9] OJOVAN, M.I., LEE, W.E. Chapter 19: Nuclear Waste Disposal. An Introduction to Nuclear Waste Immobilisation, p. 269-287, Elsevier, 2005.

[10] TCHERNER, J., VAITHILINGAM, L., HAN, M. Effective aging management of NPP concrete structures. Journal of Advanced Concrete Technology, v. 15, p. 01-09, 2017.

[11] BAI, Y., WANG, Y., XI, Y. Modeling the effect of temperature gradient on moisture and ionic transport in concrete. Cement and Concrete Composites, 106, 2020.

[12] PHUNG, Q.T., MAES, N., JACQUES, D., SCHUTTER, G.D., YE, G. Decalcification of cement paste in NH_4NO_3 solution: microstructural alterations and its influence on the transport properties. 10th fib International PhD Symposium in Civil Engineering, Québec, 2014.

[13] PHUNG, Q.T., MAES, N., JACQUES, D., SCHUTTER, G.D., YE, G. Modelling the evolution of microstructure and transport properties of cement pastes under conditions of accelerated leaching. Construction and Building Materials, 115, p. 179-192, 2016.

[14] KILDA, R., POSKAS, P., RAGAISIS, V. Modelling of nuclide migration for support of the site selection for near surface repository in Lithuania. Nuclear Engineering Laboratory - Lithuanian Energy Institute, Kaunas, 2004.

[15] VOLCHEK, K., MIAH, M.Y., KUANG, W., DEMALEKI, Z., TAZEL, F.H. Adsorption of cesium on cement mortar from aqueous solutions. Journal of Hazardous Materials, 194, p. 331-337, 2011.