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RADIOLOGICAL IMPACT OF THE APPLICATION OF PHOSPHOGYPSUM IN CIVIL CONSTRUCTION: AN OVERVIEW OF BRAZILIAN STUDIES

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ABSTRACT

In the last decades considerable attention has been given to naturally occurring radioactive material (NORM). Within this frame, of particular concern is the phosphate fertilizer industry. In Brazil, four main industries (Ultrafertil, Copebrás, Fosfertil and Bunge Fertilizantes) are responsible for the production of $1.2x10^6$ tons per year of P_2O_5. Phosphogypsum is the by-product of the phosphoric acid industry, obtained by reacting phosphate rock with sulphuric acid. This waste is stockpiled in the surrounding environment of the facilities at a rate of $5.5x10^6$ tons per year. The level of radioactivity present in the phosphogypsum, among other impurities, prevents its reuse for a variety of purposes. All the countries that produce phosphate fertilizer by acid wet processing of phosphate rock are facing the same problem of finding solutions for the safe application of phosphogypsum, in order to minimize the impact caused by its disposal. This paper aims to present a review of the research carried out at Instituto de Pesquisas Energéticas e Nucleares (IPEN), São Paulo, Brazil, focusing in the radiological implications of using Brazilian phosphogypsum as building material: bricks and plaster. The results and conclusions achieved can contribute to the development of national standards and guidelines concerning the safe use and management of phosphogypsum as a building material.

1. INTRODUCTION

In the last decades considerable attention has been given to naturally occurring radioactive material (NORM). Within this frame, of particular concern is the phosphate fertilizer industry. In Brazil, four main industries (Ultrafertil, Copebrás, Fosfertil and Bunge Fertilizantes) are responsible for the production of $1.2x10^6$ tons per year of P_2O_5. Phosphogypsum is the by-product of the phosphoric acid industry, obtained by reacting phosphate rock with sulphuric acid. This waste is stockpiled in the surrounding environment of the facilities at a rate of $5.5x10^6$ tons per year. The level of radioactivity present in the phosphogypsum, among other impurities, prevents its reuse for a variety of purposes.

Phosphogypsum is being worldwide stockpiled, posing environmental concerns. Brazil, such as other countries that produce phosphate fertilizers, tries to find solutions for the safe applications of phosphogypsum, in order to minimize the impact caused by its disposal. Phosphogypsum can be classified as TENORM (Technologically Enhanced Naturally Occurring Radioactive Materials) and its safe application should comply with specific regulation. In the international scenario, the EC (European Commission) and the IAEA (International Atomic Energy Agency) have published recommendations on the application of the concepts of exclusion, exemption and clearance that apply to TENORM activities [1, 2]. The limit adopted by the IAEA [1], for the exemption and clearance of materials for general purposes, is 1 Bq g^{-1} for natural radionuclides comprising the decay series of U and Th and 10 Bq g^{-1} for ^{40}K. Previous studies [3, 4] showed that the radioactive level of most of the Brazilian phosphogypsum is below 1 Bq g^{-1}. In Brazil, the regulatory agency (Comissão Nacional de Energia Nuclear – CNEN) has recently published a guideline concerned with mining and milling of natural occurring material, which may generate enhanced concentrations of radionuclides, under the radiological protection point of view [5]. Such activities include the mining and processing of ores as well as storage of raw material, products, by-products, residues and wastes containing radionuclides of the U-238 and Th-232 series, which may incur undue exposures of members of the public and occupationally exposed. According to the Brazilian guideline CNEN-NN-4.01 [5], the phosphate industry activity is classified in category III due to the levels of radioactivity present in the phosphogypsum. To comply with the statements of this guideline, the installation should evaluate the environmental impact of the disposal of phosphogypsum. However, there is not yet a Brazilian guideline concerned with the criteria of exemption and clearance recommendations which apply specifically to the use of phosphogypsum in the civil construction.

This paper aims to present a review of the research carried out at IPEN, focusing in the radiological implications of using phosphogypsum as building material: bricks and plaster. The results and conclusions achieved can contribute to the development of national standards and guidelines concerning the safe use and management of phosphogypsum as a building material.

2. RADIOLOGICAL CHARACTERIZATION OF BRAZILIAN PHOSPHOGYPSUM

A review of the results obtained for phosphogypsum samples from the 3 main Brazilian industries, from Saueia and Mazzilli [3] and Saueia et al [4], are presented in Table 1. All the radionuclides analyzed migrate preferentially to the phosphogypsum, except for the U isotopes, which concentrates in the phosphoric acid. The raw material processed in the installations of Bunge Fertilizantes presented concentrations of the U and Th series below the detection limits of the equipments used.

Table 1. Radionuclides activity concentration (mean ± standard deviation) and range in Bq kg^{-1} for Brazilian phosphogypsum of the 3 main Brazilian industries from [3, 4]

	Copebrás		Fosfertil		Ultrafertil	
	Mean±SD	Range	Mean±SD	Range	Mean±SD	Range
U-238	48±5	42-53	37±19	50-23	49±13	40-58
U-234	59±4	54-63	65±35	40-89	50±3	48-52
Th-230	782±144	631-978	50±25	32-68	293±59	251-334
Ra-226	744±160	550-940	186±50	122-236	344±65	280-434
Pb-210	1061±132	834-1163	182±65	136-228	347±44	316-378
Po-210	742 ± 47	541-777	279 ± 20	255-344	174 ± 18	134-203
Th-232	226±31	189-257	86±21	60-103	204±33	172-243
Ra-228	242±43	210-273	152±39	124-179	219±40	191-247

3. USE OF PHOSPHOGYPSUM AS PLASTER

The use of phosphogypsum in the civil construction as plaster or bricks can cause an increase in the exposure of dwellers since it concentrates natural radionuclides to significant levels from the radiological protection point of view. Therefore, its safe utilization requires an evaluation of the radiological impact in dwellers.

In order to study the feasibility of the use of phosphogypsum as plaster, an experimental house was built in São Carlos, State of São Paulo, having some of its rooms entirely lined with this material. At the experimental house two bedrooms and one bathroom were entirely lined with phosphogypsum and designed to perform a comprehensive radiological evaluation, including the modeling of the external dose indoors, measurement of the external gamma exposure and of radon concentrations.

The radiological evaluation of this experimental house built with phosphogypsum-based material is presented in Maduar et al [6]. The results of the radionuclides activity concentration in phosphogypsum plasters varied from 16 ± 1 to 392 ± 10 Bq kg^{-1}, for ^{226}Ra, from 26 ± 3 to 253 ± 3 Bq kg^{-1} for ^{232}Th and less than 81 Bq kg^{-1} for ^{40}K. Phosphogypsum plasters from Ultrafertil presented the highest activity concentration for all the radionuclides analyzed, while the lowest values were observed in phosphogypsum from Bunge Fertilizantes. The results for phosphogypsum from Bunge Fertilizantes are lower than the world average activity concentrations for conventional building materials of 50 Bq kg^{-1} for ^{226}Ra, 50 Bq kg^{-1} for ^{232}Th and 500 Bq kg^{-1} for ^{40}K [7].

Measurements of samples of phosphogypsum plasters from different origins resulted in values of 0.2 to 2.6 for the external radiation index, thus justifying a more detailed investigation. The values of activity concentration index (*I*) for Ultrafertil and Fosfertil phosphogypsum plasters surpassed the recommended criterion for materials used in bulk amounts. In this case, the external exposure was assessed by applying the gamma-ray transport theoretical model in the house, in order to evaluate a more realistic scenario. By using the actual data related to the dimensions of the experimental house and gamma measurements in the materials employed, a previously developed gamma-ray transport theoretical model [8] provided the dose conversion factors for evaluation of the dose from external exposure from such building material. All phosphogypsum plasters evaluated in this study presented effective dose for external exposure lower than the recommended value of 1 mSv y^{-1}, adopted as a reference level for protection of the public against radiological impact of naturally occurring radioactive materials.

Radon measurements were carried out through the passive method with solid-state nuclear track detectors (CR-39) over a period of 18 months. The detectors were changed every three months, in order to determine the long-term average levels of the indoor radon concentrations. The radon concentration results obtained in this study are below 200 $Bq\cdot m^{-3}$ [9], the investigation level recommended by ICRP in dwellings. It should be observed that the radon concentration results took into account also the radon from soil under the construction.

4. USE OF PHOSPHOGYPSUM AS BRICKS

In order to study the feasibility of the use of phosphogypsum as brick another study was undertaken to evaluate the radon exhalation from phosphogypsum bricks [10]. In this study, a hermetically sealed glass was used as accumulator. The phosphogypsum brick and a diffusion chamber with CR-39 were enclosed inside the accumulator. The radon exhalation rates from bricks manufactured with phosphogypsum from different origins were evaluated by measuring the radon concentration inside the accumulator.

The radon exhalation rates varied from 0.11 ± 0.01 Bq m^{-2} h^{-1}, for the phosphogypsum bricks from Bunge Fertilizantes, to 1.2 ± 0.3 Bq m^{-2} h^{-1}, for the phosphogypsum bricks from Ultrafertil. The results obtained are of the same order of magnitude of literature values for conventional building materials [11, 12]. So, it can be concluded that the bricks manufactured with phosphogypsum from these producers may be used for houses construction, posing no additional health risk due to exhalation of radon.

5. CONCLUSIONS

The use of building materials with elevated or technologically enhanced levels of natural radioactivity could be a reason of higher external and internal doses for residents. However, the results obtained in this study for activity concentrations, radiation doses and radon exhalation rates gave evidence that the use of the Brazilian phosphogypsum as plaster and brick poses no additional health risk to dwellers for the studied scenario.

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