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EXHALATION OF RADON FROM PHOSPHOGYPSUM PLATES OF DIFFERENT ORIGINS

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ABSTRACT

Radon is formed as a product of the radioactive series of uranium from the alpha decay of radio and has the characteristic to spread through soil and building materials, which may come to internal environments. Materials classified as NORM or TENORM are potential sources of radon isotopes, since they concentrate in significant quantities of natural radionuclides precursor to isotopes. The phosphogypsum is within the group of materials classified as TENORM and exhalation of radon by this material is a major environmental concern associated with the storage of phosphogypsum stacks in the open air and to their use as building material. In places with little air exchange, such as closed homes, the concentration of radon and its progeny may reach alarming levels in terms of radiological protection. The objective of this work is to determine the rate of exhalation of ^{222}Rn using the UNSCEAR model, determining the concentration of ^{226}Ra in phosphogypsum boards by using gamma spectrometry. The radon exhalation rate varied from 0.30±0.01 to 7.00±0.01 (Bq.m^{-2}.h^{-1}). From these measurements, the doses were calculated for an individual living in a house built with phosphogypsum based materials.

1. INTRODUCTION

Natural sources of radiation are known to be the main cause of exposure to the population. They can be classified as external sources such as cosmic rays and radioactive elements in the soil, building materials and internal sources that result from inhalation and ingestion of naturally occurring radioactive elements in air and in the diet. A peculiar feature of radiation arising from natural sources is that it affects the entire world population to a relatively constant rate over a long period of time [1].

Among the natural radioactive series, those of ^{232}Th and ^{238}U are the most interesting in terms of radiological protection, since the first isotope represents 100% of natural thorium

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and the second is present with 99.27% percent of the natural uranium isotope. The ^{235}U is only 0.7% of natural uranium and its contribution to population exposure is negligible [2].

With the development and technological advancement, some practices or human activities have concentrated natural radionuclides, causing a dose increase in certain population groups. This situation has attracted the attention of the scientific community and detailed studies have been conducted on the influence of such activities on exposure. Materials containing natural radionuclides can be classified as NORM (Naturally occurring Radioactive Materials) or TENORM (Technologically Enhanced Naturally occurring Radioactive Material). The materials including the so-called NORM radioactive elements are found in nature and although the concentration of NORM in most natural substances is relatively low, high concentrations can occur due to human activities. When the processing industrial material leads to a concentration of natural radionuclides and the increased level of radioactivity in this material, it is called TENORM.

Among the products from natural raw materials, for which there is human intervention, there is the phosphogypsum, a byproduct of phosphate fertilizer industries obtained from raw materials apatite and phosphorite in the production of wet phosphoric acid. The process consists in the attack of phosphate rock by concentrated sulfuric acid, producing phosphoric acid (H_3PO_4) in the liquid phase and phosphogypsum ($CaSO_4.2H_20$) in the solid phase [3].

The Brazilian production of phosphogypsum reaches 5.5 million tons per year and the major generating industries are located in Rio Grande, southern Rio Grande do Sul, Cajati Cubatão in the state of Sao Paulo and Uberaba, in Minas Gerais state [4].

Most of the phosphogypsum production has been stored in piles outdoors and studies show that only 14% is reprocessed [5, 6]. Thus, this generation is inevitable, making necessary studies on the feasibility of the reuse of phosphogypsum produced by the phosphate fertilizer industries. This practice has, at first glance, the advantage of minimizing environmental and economic problems arising from the disposal of this waste on the environment.

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In Brazil, the phosphogypsum is considered an environmental liability owing to the lack of specific rules governing its use. This implies a utilization of only 10% of the total amount available, using it concentrated in cement, paper and agriculture [7, 8].

The series of natural radionuclides ^{238}U, ^{232}Th and ^{40}K are usually present in phosphogypsum. Radionuclides, among them the ^{226}Ra, a member of the ^{238}U series is one of the most important in terms of radiological protection, both for the external exposure caused by gamma radiation and the internal exposure of the respiratory system caused by inhalation of its direct descendant, the ^{222}Rn. The ^{222}Rn is an inert gas that spreads very easily in the materials and when inhaled, can damage people health because it produces radioactive solids that children retain in the respiratory system [9]. Being an inert gas, radon can move freely through the pores of construction material, although only a fraction of the produced gas reaches the surface of the material and exhale into the air. It is important to distinguish between the emanation of ^{222}Rn, which is a process that controls the movement of radon atoms from the interior of the solid particles into the pore space that contains it and the exhalation of ^{222}Rn, which is a process of transfer of radon atoms from the pore space into the atmosphere. The main source of ^{222}Rn inside a residence is the soil beneath the house, but in some cases the building materials can contribute significantly. Lately, in the construction industry, the use of industrial waste from ore processing has been growing, including chemical gypsum or phosphogypsum. This practice can increase exposure to certain population groups, because this material concentrates the natural radionuclides at significant levels from the standpoint of radiation protection.

In recent years, some research groups in Brazil have been studying the possibility of the use of phosphogypsum in construction [9,10,11,12]. This work contributes to these studies and is part of a broader survey in progress at the Laboratory of Radiometry Environmental of Center of Radiation Metrology IPEN, about the radiological implications of the use of phosphogypsum in construction and agriculture. In this work, the rate is being determined by ^{222}Rn exhalation of phosphogypsum when used as building material in the form of prefabricated plates and blocks. These items are made of phosphogypsum coming from the fertilizer industries: Bunge, located in Cajati, state of São Paulo and Valefert, with units in Cubatão, state of São Paulo and Uberaba, in the state of Minas Gerais.

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2. MATERIALS AND METHODS

In this work, the rate of exhalation of ^{222}Rn from plates made with phosphogypsum is studied. The company Inovamat - Inovação em Materiais Ltda., located in Sao Carlos, Sao Paulo State, provided three plates made of phosphogypsum by Bunge and Valefert industries (units Cubatão and Uberaba). Each of the plates was made with one of the origins of phosphogypsum described above. The samples coming from the companies were called Valefert Ultrafértil and Fosfértil, according to the names of the units Cubatão and Uberaba, respectively.

There are several methods to measure the rate of exhalation of radon in building materials, using a scintillation cell, ionization chamber detectors and solid state nuclear features, among others. In this work, the radon exhalation rate was obtained from the model adopted by UNSCEAR [1] for building materials. In this model, the rate of exhalation is calculated from the concentration of ^{226}Ra in the material, among other factors, as shown in equation 1.

$$J_D = C_{Ra} \cdot \lambda_{Rn} \cdot f \cdot \rho \cdot L \cdot \tanh\left(\frac{d}{L}\right) \tag{1}$$

Where: J_D is the radon exhalation rate (Bq m^{-2} h^{-1}); C_{Ra} is the concentration of ^{226}Ra in the sample (Bq kg^{-1}); λ_{Rn} is the decay constant of ^{222}Rn (h^{-1}); f is the emanation fraction, ρ is the density of the sample (kg m^{-3}); d is the half-thickness of building material (m) and L is the diffusion length in the material (m). The diffusion length L was calculated using the equation , where D_e is the effective coefficient of diffusion (m^2/h).

The emanation fraction is related to the size of the grains, since due to the effective diffusion coefficient with moisture present in the sample, this paper adopted the fraction of emanation suggested by UNSCEAR [1]; the effective diffusion coefficient was made using an approximation of the aerated concrete value, obtained in the literature [13,14]. The values of the ^{226}Ra concentration were obtained in the literature [9].

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Table 1. Average concentrations of ^{226}Ra and standard deviations (Bq.kg^{-1}).

Origin	Concentrations of ^{226}Ra (Bq.kg^{-1})
Bunge	15.9 ± 0.5
Ultrafértil	392 ± 10
Fosfertil	294 ± 3

As expected, the phosphogypsum materials from Ultrafértil have the highest concentrations of ^{226}Ra. According to Villaverde, [9], the phosphogypsum produced by the companies Ultrafértil and Fosfertil is obtained from the waste processing and sedimentary rocks; the phosphogypsum from Bunge must come from the processing of igneous rocks, which are predominant in Brazil, known to have lower concentrations of ^{238}U and, consequently, ^{226}Ra. The concentrations of radionuclides in phosphogypsum may vary depending on the source of phosphate rock and the type of procedure chosen to obtain phosphoric acid.

The radon exaltation rate in phosphogypsum plates was determined by means of the model in Annex B of UNSCEAR [1], presented in equation (1) for the concentration of ^{226}Ra in materials, among other factors. Table 2 presents the results of ^{222}Rn exhalation rates for phosphogypsum plates.

Table 2. Radon exhalation rate of theoretical phosphogypsum plates, using the UNSCEAR model.

Origin	J_D (Bq m^{-2} h^{-1})
Bunge	0.3 ± 0.01
Ultrafértil	7 ± 0.1
Fosfertil	4.7 ± 0.1

It is noteworthy that some of the parameters used in the calculation do not correspond to the materials analyzed and the results show the importance of experimental determination of ^{222}Rn exhalation rate, especially if this parameter is chosen in the decision as to the

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assessment of exposure by individuals in a residence. As previously mentioned, one of the parameters determining the rate of exhalation of ^{222}Rn is the concentration of ^{226}Ra present in the material. The other parameters are the grain size, porosity and moisture.

3. CONCLUSIONS

The radon exhalation rate varied from 0.3 $Bq.m^{-2}.h^{-1}$ for the phosphogypsum plates and blocks produced by the company Bunge up to 4.7 $Bq.m^{-2}.h^{-1}$, for the phosphogypsum plates and blocks from Ultrafértil. Many times the radon exhalation rate from models has been over-estimated, hence showing the importance of data confirmation by experimental determination.

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REFERENCES

1. United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR). Sources and effects of ionizing radiation. **Vol. 1** (New York: United Nations Publication) (2000).
2. Paes, V. P., “Caracterização radioquímica do ^{226}Ra, ^{40}K e dos isótopos de urânio e tório no fosfogesso”, MSc thesis, IPEN, São Paulo (2001).
3. Saueia, C. H. R., “Distribuição elementar e de radionuclídeos na produção e uso de fertilizantes fosfatados no Brasil”, PhD thesis, IPEN, São Paulo (2006).
4. Mazzilli, B. P., Palmiro, V., Saueia, C., Nisti, M. B., “Radiochemical characterization of Brazilian phosphogypsum”, *Journal environmental radioactivity*, **Vol. 49,** p. 113-122, 2000.
5. Rutherford, P..M.; Dudas, M.J.; Samek, R.A. “Environmental impacts of phosphogypsum”, *The Science Total Environmental*, **Vol. 149,** pp.1-38 (1994).
6. Lysandrou, M., Charalambides, A., Pashalidis, “Radon emanation from phosphogypsum and related mineral samples in Cyprus”, *Radiation Measurements*, **Vol. 42,** pp. 1583-1585 (2007).
7. Saueia, C. H. R., “Caracterização radioquímica do fosfogesso e implicações radiológicas de sua utilização como material de construção”, MSc thesis, IPEN, São Paulo (1998).
8. Damasceno, E. C., Lima, J. R. B., “Aproveitamento de resíduos da indústria de mineração: o fosfogesso gerado pela solubilização de concentrados fosfáticos”, In: 3º

ISBN: **978-85-99141-04-5**

CONGRESSO ÍTALO BRASILEIRO DI INGEGNERIA MINERARIA, 26-27 settembre, pp. 229-230, Verona, (1994).
9. Villaverde, F. V., "Avaliação da exposição externa em residência construída com fosfogesso", MSc thesis, IPEN, São Paulo (2008).
10. Canut, M.M.C., Jacomino, V.M.F., Bratveit, A., Gomes, A.M., Yoshida, M.I., "Microstructural analyses of phosphogypsum generated by Brazilian fertilizer industries", *Materials Characterization,* **Vol. 59,** pp. 365-373 (2008).
11. Máduar, M. F., Campos, M.P., Mazzilli, B. P., Villaverde, F. L. "Assessment of external gamma exposure and radon levels in a dwelling constructed with phosphogypsum plates", *Journal of Hazardous Materials,* **Vol. 190,** pp. 1063-1067, (2011).
12. Rabi, J. A. da Silva, N. C., "Radon exhalation from phosphogypsum building boards: symmetry constraints, impermeable boundary conditions and numerical simulation of a test case", *Journal of Environmental Radioactivity,* **Vol. 86,** pp. 164-175 (2006).
13. Van der Pal, M., Van der Graaf, E. R., de Meijer, R. J., de Wit, M. H., Hendriks, N. A., "Experimental set-up for measuring diffusive and advective transporto f radon through building materials" The Science of the Total Environment, Vol. 272, pp. 315-321. (2001).
14. de Souza, A. F. C., "Desenvolvimento de uma metodologia para avaliação de doses visando o uso de NORM em materiais de construção", PhD thesis, IME, Rio de Janeiro (2009).