RADIOLOGICAL ASSESSMENT OF USING PHOSPHOGYPSUM AS BUILDING MATERIAL

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Abstract

In order to assess the feasibility of using phosphogypsum as a building material, an experimental house was built with phosphogypsum panels of different origins. The aim of this study is to assess the external and internal exposure of residents. Phosphogypsum samples were analysed by high resolution gamma spectrometry for their ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K content. The activity concentrations were 0.016–0.393 Bq/g for ^{226}Ra, 0.026–0.253 Bq/g for ^{232}Th and 0.027–0.852 Bq/g for ^{210}Pb. The results for ^{40}K were lower than 0.081 Bq/g. The effective dose was evaluated for each type of phosphogypsum panel. The effective doses due to external exposure were always below 1 mSv/a, the dose limit for the general public. Radon measurements in the house were carried out using the passive method with solid state nuclear track detectors (CR-39) over a period of 15 months. The detectors were changed every three months, in order to determine the long term average levels of the indoor radon concentrations with varying seasons. The radon concentration varied from 45 to 119 Bq/m^3. These results are below 200 Bq/m^3, the recommended investigation level for radon in dwellings.

1. INTRODUCTION

It is well known that natural radioactivity in building materials and radiation from the ground constitute the most important sources of radiation exposure [1]. Concerns about health hazards and environmental pollution have heightened the interest in radon levels in building materials. Building materials that may be of radiological significance include marl, blast furnace slag, flyash, phosphogypsum, Portland cement clinker, anhydrite, clay, and granites rich in radium or thorium [2].

Chronic exposure of humans to low levels of radiation can cause health effects which may appear several years after the exposure. The main contributors to external exposure from gamma radiation are the radionuclides of the ^{238}U and ^{232}Th series together with ^{40}K that are present in small quantities in the earth's crust and in building materials. The most important contributors to the internal exposure of the population to natural sources are the short lived decay products of radon (^{222}Rn) and thoron (^{220}Rn).

In recent years, various surveys of the radionuclide content of building materials were published [3–6], in order to estimate the indoor radiation exposure of the occupants. Indoor exposure to gamma radiation and radon inhalation can be enhanced if industrial by-products such as phosphogypsum are used to replace some of the natural components of building materials.

Phosphogypsum is a by-product obtained from the wet acid processing of phosphate rock to produce phosphoric acid. For every tonne of phosphoric acid produced in the reaction of phosphate rock with sulphuric acid, about 4–5 t of phosphogypsum are produced. The Brazilian annual production of phosphogypsum is up to 5.4 million t [7]. All the countries that produce phosphate fertilizer by wet acid 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 the disposal of large amounts of this material. Phosphogypsum can be classified as NORM, and its safe application should comply with specific regulations.

At the international and regional levels, the IAEA and the European Commission (EC), respectively, have recently published recommendations on the application of the concepts of exclusion, exemption and clearance, the scope of application of which includes NORM activities [8–10]. In IAEA Safety Reports Series No. 44 [9], the concepts of exemption and clearance have been applied to bulk amounts of material, by taking into account exposure scenarios that included those described above. These scenarios were applied only to artificial radionuclides, however. For radionuclides of natural origin, the recommended criteria for regulation, 1 Bq/g for ^{238}U and ^{232}Th series radionuclides and 10 Bq/g for ^{40}K, were based on the global distribution of radionuclide activity concentrations in rocks and soil. The rounded exemption and clearance levels established by the EC [11] are 0.5 Bq/g for ^{226}Ra and ^{228}Th; 1 Bq/g for ^{228}Ra; 5 Bq/g for U_{nat}, ^{210}Pb, ^{210}Po and ^{232}Th; and 10 Bq/g for ^{230}Th.[1]

The activity concentrations in Brazilian phosphogypsum for all sources of production [7] are below the criteria for regulation recommended by the IAEA and the exemption levels recommended by the EC. In Brazil, the regulatory agency (Comissão Nacional de Energia Nuclear (CNEN)) has recently published a radiation protection guideline on the mining and processing of NORM that may generate enhanced concentrations of radionuclides [12], according to which the phosphate industry activity is classified as category III on account of the activity concentration in phosphogypsum. In terms of this guideline, the facility should

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[1] The values for ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{228}Th include the progeny of these radionuclides.

evaluate the environmental impact of the disposal of phosphogypsum. However, there is not yet a Brazilian guideline giving criteria for exemption and clearance that apply specifically to the use of phosphogypsum. The use of phosphogypsum is very important from the social and economic point of view and also regarding environmental protection. Phosphogypsum can be used as a base for roads, as a building material and in agriculture as a soil amendment.

Only a small proportion of the phosphogypsum produced worldwide (14%) is used as a building material. However, phosphogypsum often contains significant amounts of radioactivity originating mainly from the ^{238}U and ^{232}Th decay series, which, according to Ref. [13], can create a health hazard. Radium, which decays to ^{222}Rn through alpha particle emission, is one the most important radionuclides from the point of view of radiation protection. Exposure to radium and radon originating from phosphogypsum is an important health concern associated with the use or disposal of this material. The main health concern associated with ^{222}Rn arisies from its short lived alpha emitting progeny, which can cause damage to the lungs after chronic exposure. Radon is an inert, noble gas and may become airborne by diffusing into the air. One of the most important sources of indoor radon is the underlying soil; however the contribution from building materials should also be taken into account.

In order to assess the feasibility of using phosphogypsum as a building material, an experimental house was built with phosphogypsum panels (manufactured with phosphogypsum of various origins) in São Carlos, São Paulo State, Brazil. The panels were developed with a new process (patent applied for), denominated UCOS (humidification, compaction and drying), which works with plasters made from both mineral gypsum and phosphogypsum [14]. The plaster prepared through this process may be used inside and outside the building and is sturdy enough to be used in the construction of buildings up to eight storeys high. The house walls are light, easily mounted and have thermal and acoustic insulation. Other advantages of these panels compared to the conventional process are the short construction time (a seven room house such as the experimental house takes only one month to build) and cost, which is approximately 30% lower than that for conventional construction methods.

In the experimental house, two bedrooms and one bathroom were lined entirely with phosphogypsum and designed so as to enable a comprehensive radiological assessment to be performed, including the modelling of the indoor external dose rate and measurement of the external gamma exposure and radon concentration. The walls consisted of double sets of joined panels. The internal cavities between wall panels, as well as between the roof and ceiling, were lined with blanketing material. The floor areas were 9.07 m^2 for the first bedroom, 11.08 m^2 for the second bedroom and 2.4 m^2 for the bathroom. The phosphogypsum panel thickness was 1.5 cm for the walls and 1 cm for the ceiling. The wall cavities were 15 cm wide.

The aim of the study was to assess indoor external exposure and radon concentrations in a house constructed with phosphogypsum panels of various origins. Samples of the material were analysed by high resolution gamma spectrometry for their ^{226}Ra, ^{210}Pb, ^{232}Th, and ^{40}K content. The radium equivalent activity and effective dose from external exposure were also calculated. Radon measurements were carried out using passive solid state nuclear track detectors (CR-39) over a period of 15 months. The detectors were changed every three months, in order to determine the long term average levels of the indoor radon concentrations with varying seasons. The results obtained from this study can contribute to the development of national standards and guidelines concerning the safe use and management of this material as a building material.

2. MATERIALS AND METHODS

An experimental house was constructed, in which the walls and ceilings were built of phosphogypsum panels. These panels were manufactured with phosphogypsum from different producers, namely Ultrafertil, located in Cubatão (CT), Bunge, in Cajati (CA) and Fosfertil, in Uberaba (UB). The bathroom and one bedroom were built with phosphogypsum panels from Cubatão and the other bedroom was built with Cajati phosphogypsum panels.

Twelve samples of phosphogypsum panels (five from Cubatão, four from Cajati and three from Uberaba) were crushed and packed in a 100 mL polyethylene flask and sealed for about four weeks prior to the measurement, to ensure that equilibrium was reached between ^{226}Ra and its short lived progeny. The activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the samples of phosphogypsum panels were measured by gamma spectrometry with a Eurisys EGNC 15-190-R hyperpure germanium detector. The relative efficiency of the detector was 15% and the effective energy resolution for the 1.33 MeV ^{60}Co gamma transition was 1.9 keV. The detection efficiency curve was calculated for aqueous solutions containing certified activity concentrations of gamma emitters covering a wide range of energies, encompassing the radionuclide energies determined in the samples. Background measurements were taken and subtracted in order to get net counts for the sample. Samples were measured during a period of 40 000–200 000 s, depending on the radioactivity levels in the samples. All spectra were analysed with Eurisys Interwinner 4.1 software for personal computer analysis of gamma spectra from HPGe detectors [15].

The activity concentration of ^{40}K was determined directly by its own gamma peak at 1460.8 keV. The activity concentration of ^{226}Ra was determined using the 295.2 and 351.9 keV gamma emissions from ^{214}Pb and the 609.3 and 1120.3 keV gamma emissions from ^{214}Bi. The activity concentration of ^{232}Th was determined using the 338.5, 911.1 and 968.9 keV gamma emissions from ^{228}Ac and the 727.2 and 1620.6 keV gamma emissions from ^{212}Bi. In a previous paper [7], it was shown that the measurements of ^{232}Th in phosphogypsum samples, analysed by alpha and gamma spectrometry, gave results which are representative of the radionuclide activity. In this paper, the ^{232}Th activity concentration was performed by gamma spectrometry on its progeny gamma emitters. The activity concentration of ^{210}Pb was determined by its 46.5 keV photopeak. A self absorption correction was applied owing to the low energy gamma ray attenuation within the sample. The approach used was the same as that described in Ref. [7]. The minimum detectable activity concentrations in the phosphogypsum panel samples were, for a counting time of 200 000 s, 0.0028 Bq/g for ^{226}Ra, 0.0051 Bq/g for ^{232}Th, 0.039 Bq/g for ^{40}K and 0.018 Bq/g for ^{210}Pb.

Various well established techniques are available for measuring the concentrations of radon and its progeny. The passive method with solid state nuclear track detectors (SSNTDs) has been used widely, since it is regarded as being inexpensive, reliable and unaffected by widely varying climatic conditions [16]. In this study, the ^{222}Rn concentration was obtained by the passive detection method with SSNTDs placed in diffusion chambers. CR-39 in 2.5 cm squares was used as the detection medium. As a diffusion chamber, a hemispherical ‘closed-can’ of 4 cm radius was selected. This chamber consists of a polypropylene holder made of an upper and lower half which snaps together during assembly. The fit of the two halves is quite tight, in order to exclude moisture, dust and radon progeny. Thoron is excluded from the diffusion chamber because of its short half-life and diffusion time. The detectors were suspended in the dwellings from the ceiling at a height of 1.7 m from the ground and placed away from any surface to avoid any plate-out effects. After exposure for approximately 3 months, the detectors were collected and replaced by fresh ones. The detectors were etched in a KOH solution (30% mass concentration) at 80ºC for 5.5 h in a constant temperature bath. After etching, the detectors were washed, dried and scanned under a microscope for track density measurements. The background was 220 ± 16 tracks/cm^2. Using a calibration factor of 0.0216 ± 0.0015 tracks/cm^2 per Bq·d·m^{-3}, obtained with a Pylon model RN-150 calibrated radon gas source; the track density was converted to radon concentration in the environment.

The radon concentration was calculated from the following equation [17]:

$$C = \frac{D}{k \cdot t} \tag{1}$$

where C is the radon concentration (Bq/m^3), D is the track density (tracks/cm^2), k is the calibration factor (tracks/cm^2 per Bq·d·m^{-3}) and t is the exposure time (d). Radon measurements were carried out over a period of 15 months, changing the detectors every three months, in order to determine the long term average levels of the indoor radon concentrations with varying seasons. The detectors were placed in the two bedrooms and the bathroom.

3. RESULTS AND DISCUSSION

3.1. Activity concentration and radium equivalent activity

Phosphogypsum panels from three production locations, Cubatão (CT), Cajati (CA) and Uberaba (UB) were analysed for ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K activity concentrations. The radium equivalent activity (Ra_{eq}) is a common index that compares the activity concentration of ^{226}Ra, ^{232}Th and ^{40}K in building materials, taking into account the radiation hazards associated with them. This index is based on the assumption that 1 Bq/g of ^{226}Ra, 0.7 Bq/q of ^{232}Th and 13 Bq/g of ^{40}K produce the same gamma dose rate. According to UNSCEAR recommendations [1, 18], the maximum value of Ra_{eq} in building materials must be less than 0.37 Bq/g in order to keep the dose from external gamma exposure below 1.5 mSv/a. The radium equivalent activity provides a useful guideline on radiation protection for the general public in dwellings. It was calculated as follows:

$$Ra_{eq} = C_{Ra} + 1.43\ C_{Th} + 0.007\ C_K \tag{2}$$

where C_{Ra}, C_{Th} and C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively (Bq/g). Table 1 presents the average activity concentrations and standard deviation for ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K in the phosphogypsum panels used in the house construction. The radium equivalent activity is also presented.

The activity concentrations varied widely for the three phosphogypsum origins. Panels from Cubatão (CT) showed the highest activity concentrations for all the radionuclides analysed, while the lowest values were observed in phosphogypsum from Cajati (CA). The activity concentrations in the products

TABLE 1. ACTIVITY CONCENTRATIONS IN PHOSPHOGYPSUM PANELS

Origin of panel	Average activity concentration (Bq/g)				
	^{226}Ra	^{232}Th	^{40}K	^{210}Pb	Radium equivalent
CA	0.016 ± 0.001	0.026 ± 0.003	<0.039	0.026 ± 0.007	0.056
CT	0.392 ± 0.10	0.253 ± 0.003	<0.081	0.852 ± 0.138	0.759
UB	0.294 ± 0.003	0.151 ± 0.006	<0.056	0.295 ± 0.016	0.513

from Cajati are lower than the worldwide average values for building materials of 0.05 Bq/g for ^{226}Ra, 0.05 Bq/g for ^{232}Th and 0.5 Bq/g for ^{40}K [18]. All the phosphogypsum panels exhibited ^{40}K concentrations lower than the world averages for building materials. The results obtained in this work are in agreement with national and international values published in the literature for phosphogypsum [7, 19].

There is a great variation in the radium equivalent activity results, depending on the origin of the phosphogypsum. The highest values were reported for Cubatão (CT) and Uberaba (UB) panels, both of which exceeded the value recommended by UNSCEAR (0.37 Bq/g), while the panels from Cajati (CA) are lower than this value. However, it should be noted that the radium equivalent activities obtained from the maximum activity concentrations in industrial by-products used for building materials in the European Union reaches 1.351 Bq/g [10], a figure significantly higher than the maximum value of 0.759 Bq/g obtained in the present work for Brazilian phosphogypsum.

3.2. Effective dose for external exposure

The effective dose received indoors from external exposure to phosphogypsum panels was assessed according to UNSCEAR procedures through the standard room concept, according to the equation:

$$E = T f b \times 10^{-6} (q_{Ra}C_{Ra} + q_{Th}C_{Th} + q_K C_K)\, m \tag{3}$$

where E is the effective dose from external exposure; T is the exposure time (8760 h/a); f is the fraction of time spent indoors (0.8); b is the conversion factor from absorbed dose in air to effective dose (0.7 Sv/Gy); q_{Ra}, q_{Th} and q_K are the conversion factors from ^{226}Ra, ^{232}Th and ^{40}K activity concentrations to absorbed dose in indoor air, respectively (nGy/h per Bq/g); C_{Ra}, C_{Th} and C_K are the ^{226}Ra,

^{232}Th and ^{40}K activity concentrations, respectively (Bq/g) and m is the mass fraction of material in the standard room. The choice of conversion factor from activity concentration to absorbed dose is the most important factor for evaluating the external dose from building materials. Gamma radiation from walls is strongly dependent on the wall thickness and material density, so it is useful to adopt the standard room concept to estimate the dose. Values of q_i reported in Ref. [20] were used as the basis for applying a previously developed computational model [21] to forecast external doses indoors using the parameters of the experimental house. The q_i values take into account the gamma transitions of ^{40}K and radionuclides in the ^{226}Ra and ^{232}Th decay series, the wall thickness of the house and the density of the phosphogypsum panels. The estimated effective dose from external exposure for each source of phosphogypsum panel, is presented in Table 2. For all origins, the effective dose from external exposure was lower than the 1 mSv annual dose limit for members of the public.

3.3. Radon concentration

The ^{222}Rn concentrations in the experimental house were calculated from the SSNTD measurements using Eq. (1). The average radon concentrations and standard deviations obtained for the period May 2007 to September 2008 are shown in Table 3. The radon concentrations varied from 45 to 119 Bq/m^3 in the bedrooms and from 83 to 105 Bq/m^3 in the bathroom. The difference in radon concentration between bedrooms 1 and 2 can be explained by the origin of the phosphogypsum. Bedroom 1 and the bathroom were built with phosphogypsum from Cubatão (average ^{226}Ra concentration 0.392 ± 0.010 Bq/g) and bedroom 2 was built with phosphogypsum from Cajati (average ^{226}Ra concentration 0.016 ± 0.001 Bq/g). Reference to Fig. 1 shows that there was no clear seasonal variation of indoor radon concentration.

TABLE 2. EFFECTIVE DOSE FOR EXTERNAL EXPOSURE IN A STANDARD ROOM

Origin of panel	Annual effective dose (mSv)
CA	0.02
CT	0.20
UB	0.14

TABLE 3. RADON CONCENTRATIONS IN THE EXPERIMENTAL HOUSE

	^{222}Rn activity concentration in air (Bq/m^3)				
	May to August 2007	August to November 2007	November 2007 to February 2008	February to June 2008	June to September 2008
Bedroom 1	98 ± 14	105 ± 11	99 ± 5	119 ± 9	106 ± 11
Bedroom 2	—	46 ± 2	50 ± 2	45 ± 2	48 ± 3
Bathroom	88 ± 13	105 ± 8	99 ± 7	83 ± 8	86 ± 9

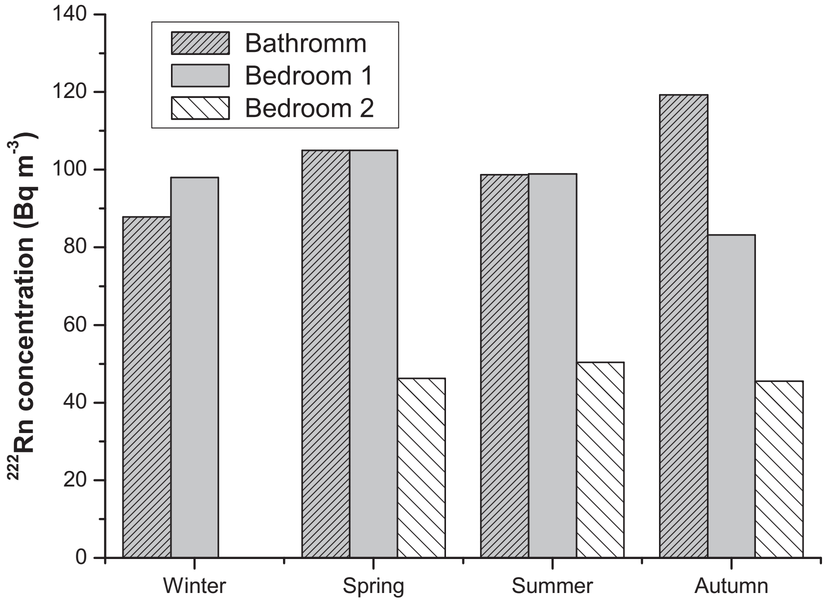

FIG. 1. Seasonal variation of indoor ^{222}Rn concentration in the experimental house.

There was a good correlation between the indoor ^{222}Rn concentration and the ^{226}Ra concentration in the phosphogypsum panels (see Fig. 2). Therefore, the ^{222}Rn concentration in a room built with panels from another source can be predicted from the ^{226}Ra concentration in those panels and the slope of regression line in Fig. 2.

4. CONCLUSIONS

The activity concentrations of ^{226}Ra, ^{232}Th, ^{210}Pb and ^{40}K in phosphogypsum panels from Cubatão, Cajati and Uberaba have been measured by gamma spectrometry. The radium equivalent activity and the effective dose for

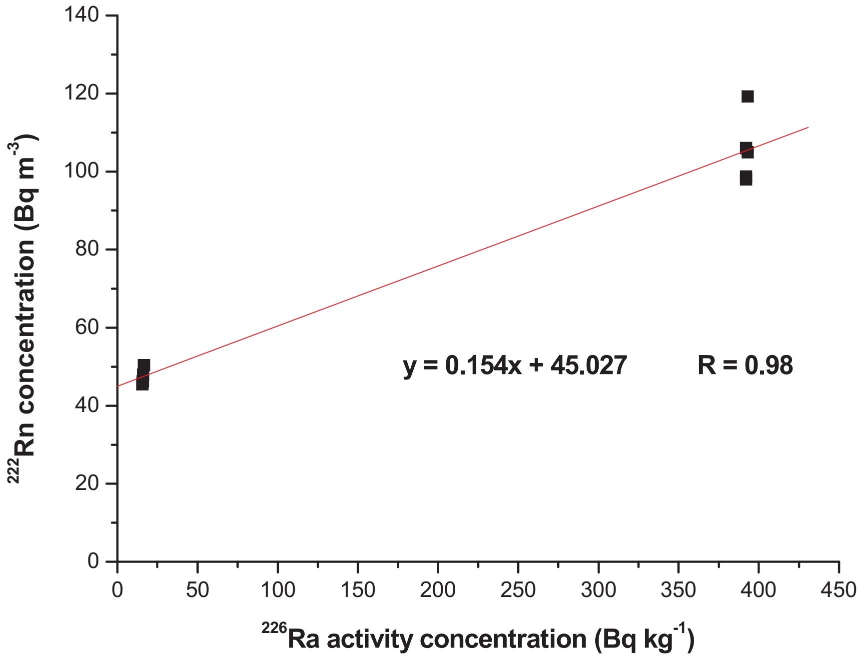

FIG. 2. Relationship between indoor radon concentration and ^{226}Ra activity concentration in the phosphogypsum panels.

external exposure were assessed from the activity concentrations. The results showed that the phosphogypsum panels from Cubatão exhibited the highest radioactivity levels, while those from Cajati exhibited very low activity concentrations and, therefore, negligible risk.

There is no regulatory framework for the use of NORM as a building material in Brazil. Nevertheless, it is possible to adopt 1 mSv/a as a reference level for protection of the public against radiological impact of naturally occurring radioactive materials, as recommended by the International Commission on Radiological Protection (ICRP) [22]. According to EC guidance [10], the annual effective dose due to gamma radiation in dwellings should not exceed 0.3 mSv for the building materials to be exempted from all restrictions concerning their radioactivity. All phosphogypsum panels evaluated in this study gave rise to effective doses for external exposure lower than this recommended value. The present work showed that, according to dose criteria recommended by the ICRP and the EC, the phosphogypsum panels do not pose any significant additional health risk to dwellers and its use can be considered to be safe for inhabitants.

The ICRP recommends that the action level for radon in dwellings should be in the range 200–600 Bq/m^3 [23]. The radon concentrations determined from this study are below 200 Bq/m^3. It should be noted that the radon concentration results took into account the radon from soil under the building.

The results obtained in this study point to the fact that the radon concentrations in dwellings made from phosphogypsum panels are comparable with those in houses built from conventional materials [1, 24]. Therefore, the use of phosphogypsum as a building material poses no additional health risk to occupants.

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