

External dose assessment and radon monitoring in an experimental house built with phosphogypsum-based materials

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Abstract. Phosphogypsum, a fertilizer industry by-product, is being worldwide stockpiled, posing environmental concerns. Since this material contains natural radionuclides in significant concentrations, its use as a building material has radiological implications. In order to confirm the feasibility of the use of a new material mainly composed by phosphogypsum, an experimental house was recently built, having one of its rooms entirely lined with this new material. Measurements of samples of this material resulted in a value of 1.8 for the external radiation index, thus justifying a more detailed investigation. In this paper, the application of a previously developed computational model to forecast external doses indoors is described. A comprehensive radiological evaluation is being performed, including measurement of the external gamma exposure and radon concentrations in one of the rooms of the house. The results show that the annual increment in the equivalent dose to a hypothetical inhabitant of the house will remain below the 1 mSv limit for every reasonable scenario.

1. INTRODUCTION

Phosphogypsum, a fertilizer industry by-product, is being worldwide stockpiled, posing environmental concerns. Since this material contains natural radionuclides in significant concentrations, its use as a building material has radiological implications. The limit adopted by the International Atomic Energy Agency [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 [2,3] have shown evidences that the radioactive level of most of the Brazilian phosphogypsum is below 1 Bq g^{-1} . Phosphogypsum in Brazil is unofficially used in cement and paper industries, as well as a soil amendment [4]. However, there is not yet a Brazilian guideline establishing criteria for exemption and clearance recommendations specifically applied to phosphogypsum.

In this paper, results from a radiological evaluation in an experimental house built with gypsum and phosphogypsum-based material are presented.

2. THE EXPERIMENTAL HOUSE

A new building material, constituted mainly by gypsum or phosphogypsum processed by using a novel patent-applied technique consisting of sequential humidification, drying and compaction was recently developed. In order to confirm the feasibility of the use of this new material, an experimental house was recently built, having one of its rooms entirely lined with this material [5].

The house was constructed as a pilot standard dwelling, thus it has all the requirements to be actually inhabitable, including doors, windows, piping and electrical wiring. On the other hand, it has some features to allow the implementation of technological studies, such as the modularity and ease of reconfiguration of its constitutive structural elements, when needed. The floor plan of the house is shown in Figure 1.

The room 2 of the house was 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 walls of that room are composed by double sets of joined panels. The walls' panels are sheathed with blankets in between, as well as between the roof and the ceiling. The internal dimensions of the room are 3.36 m and 3.298 m. The phosphogypsum panels' thickness is 1.5 cm for the walls and 1 cm for the ceiling. The double sets composing the walls have a 15 cm gap between the panels.

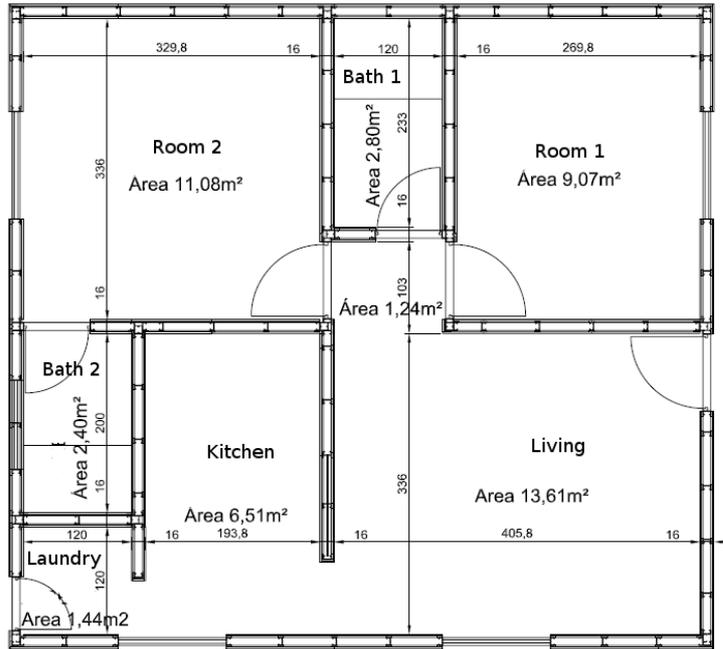


Figure 1. Floor plan of the experimental house

3. MATERIAL AND METHODS

3.1 Gamma measurements

Phosphogypsum panels were analyzed for the gamma emitters' determination. Panels' samples were crushed and sealed in polyethylene flasks with 0.1 L volume. Samples were measured after a minimum 30-day delay period in order to obtain $^{226}\text{Ra}/^{214}\text{Pb}/^{214}\text{Bi}$ radioactive equilibrium. The counting system was composed by a Canberra model GX2518 HPGc detector associated to a DSA1000 electronic module, comprising high voltage supplier and multichannel analyzer. The spectra were submitted to computer assisted analysis with Genie 2000 software, using previously determined calibration parameters and background corrections.

3.2 Radon concentrations determination

Radon concentration measurements indoors are being currently performed. Radon is being monitored by using CR-39 solid state nuclear track detectors, to evaluate the alpha activity in air.

3.3 Modeling of the external dose in air and gamma direct measurements

Provided that the concentration activities of the radionuclides in the building material are known, the absorbed dose in air at any point of interest inside a room can be calculated, for practical purposes, by using a simple linear function given by:

$$\dot{D} = \sum_i q_i C_i \quad (1)$$

where \dot{D} is the absorbed dose rate in air (Gy s^{-1}), q_i is the dose conversion factor for the radionuclide precursor of the decay series i (Gy s^{-1} per Bq kg^{-1}), and C_i is the activity concentration of the radionuclide precursor of the decay series i (Bq kg^{-1}).

The factors q_i were calculated by applying a previously developed computational model to forecast external doses indoors [6], arising from the gamma transitions of ^{40}K and of ^{226}Ra and ^{232}Th respective decay series.

To apply this model, the room was geometrically defined as a set of several rectangular slabs of finite thickness, in a parallel and/or perpendicular disposition, as needed. For each slab, the dose was calculated at a point O , considering a uniform radionuclide distribution in a finite rectangular slab with thickness t and parameters x_1, x_2, y_1, y_2 . The point O is at the origin of the Cartesian coordinates system, at a distance h from the slab face toward it. This construction is shown in Fig. 2.

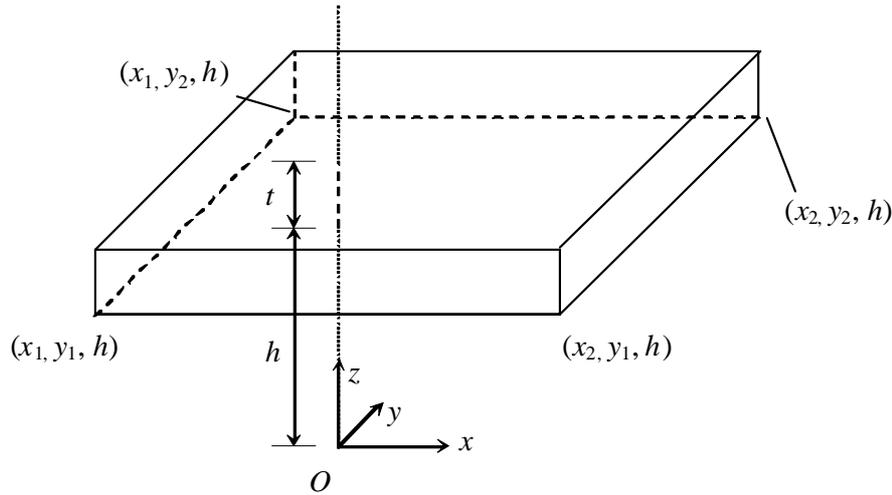


Figure 2. Geometric model for each slab

As the dose rate function is additive, the final dose inside a compartment will be the sum of the doses due to each slab considered separately.

The phosphogypsum slabs have density $1.8 \text{ g}\cdot\text{cm}^{-3}$. Table 1 summarizes the geometric parameters for all the phosphogypsum slabs composing the room. The minus signs in some house elements indicate that doors and windows were modeled as slabs and then the resulting terms were subtracted from the total dose.

The radiation sources were assumed to be uniformly distributed in the volume of the slabs. The point of calculation is the center of the room, 1.1 m above the floor, to coincide with the location of dosimeters and nuclear track detectors.

Table 1. Dimensions of the room elements

House element	Parameters (x_1, x_2, y_1, y_2) of the slab (cm)	Distance h from the calculation point to the slab face toward it (cm)	Slab thickness t (cm)
Inner slabs:			
2 lateral walls	-165, +165, -110, +150	168	1.5
2 frontal walls	-168, +168, -110, +150	165	1.5
Ceiling	-165, +165, -168, +168	150	1
Entrance door (-)	-168, -86, -110, +100	165	1.5
Bathroom door (-)	-165, -93, -110, +100	168	1.5
Window (-)	-63, +57, -10, +90	165	1.5
Outer slabs:			
2 lateral walls	-165, +165, -110, +150	182.5	1.5
2 frontal walls	-168, +168, -110, +150	179.5	1.5
Entrance door (-)	-168, -86, -110, +100	179.5	1.5
Bathroom door (-)	-165, -93, -110, +100	182.5	1.5
Window (-)	-63, +57, -10, +90	179.5	1.5

External dose is being directly assessed using thermoluminescent dosimeters (TLD). Both detectors (TLD and track detectors) are placed in field and substituted each 3 months.

4. RESULTS AND CONCLUSIONS

4.1 External doses in air

Forecast doses, in the form of dose conversion factors separated for ^{40}K , ^{226}Ra +progeny and ^{232}Th +progeny, were assessed. The activity concentrations applied were the mean values obtained for each determined radionuclide in the phosphogypsum samples. These data and the resulting annual absorbed doses in air are shown in Table 2, resulting in a 0.34 mGy sum for all the radionuclides.

Table 2. Resulting absorbed doses in air from the modeling

Radionuclide	Dose conversion factor (nGy/h per Bq/kg)	Mean activity concentration (Bq/kg)	Annual absorbed dose in air (mGy)
^{226}Ra	0.0800	250	0.18
^{232}Th	0.103	180	0.16
^{40}K	0.00776	20	0.0014

The annual radiation exposure increment in the room was evaluated with a TLD set placed in it and another set placed outdoors, in the backyard of the house, to enable the assessment of the increment in the gamma radiation due to the building materials employed. The external radiation exposure increment was periodically evaluated and normalized to 90-days periods, resulting in an average value from May to November/2007, normalized to one year, of 0.32 mGy per year.

4.2 Measurements of radon concentrations and external gamma doses

Radon concentrations in the room under study were $98 \pm 18 \text{ Bq.m}^{-3}$ in the May to Aug/2007 period and $105 \pm 18 \text{ Bq.m}^{-3}$ in the Aug to Nov/2007 period. The annual dose increment in air obtained with thermoluminescent dosimeters, 0.32 mGy, was consistent with the corresponding figure of 0.34 mGy forecasted by the present model from gamma emissions from ^{40}K , ^{226}Ra and ^{232}Th , suggesting that the model could be applied to different scenarios as well.

4.3 Radiological evaluation

For radiological studies involving the use of building materials, European regulations [7] recommends the application of the index derived from the activity concentrations C_{Ra} , C_{Th} and C_K present in the building material. This index I is defined as

$$I = \frac{C_{Ra}}{300 \text{ Bq kg}^{-1}} + \frac{C_{Th}}{200 \text{ Bq kg}^{-1}} + \frac{C_K}{3000 \text{ Bq kg}^{-1}} \quad (2)$$

This criterion is ultimately derived from specific dose rates values, given in nGy h⁻¹ per Bq kg⁻¹, of 0.92 for ^{226}Ra , 1.1 for ^{232}Th and 0.080 for ^{40}K . These values were obtained from a well established model of a room defined as a 4 m × 5 m × 2.8 m compartment delimited by 20 cm-thick concrete structures with density 2350 kg m⁻³. Application of expression (2) to the mean values of activity concentration in the material under study results in $I = 1.8$, thus surpassing the $I \leq 1$ criterion for bulk usage.

Nonetheless, the same regulations [7] state that the use of such index should be limited to screening purposes, in order to identify building materials of concern, hence justifying the use of a more realistic dose forecast model in the present situation. The application of this model to the experimental house leads to specific

dose rates that are 8.7% for ^{226}Ra , 9.4% for ^{232}Th and 9.7% for ^{40}K of the corresponding values for that standard room. Such lower figures were expected as the building material is used in the form of thin plates and have lower density than of the standard room, thus the overall mass of the material that constitutes the radiation source is also significantly lower.

Furthermore, with the increment of the external absorbed dose in air of 0.34 mGy obtained in this work, it is expected that the equivalent dose increment will remain far below 1 mSv per year, for every reasonable scenario of human occupation of the house.

Radon concentrations in the experimental house were found to be comparable with the average radon concentrations in Nordic dwellings, built with conventional building materials [8]. The results obtained show that radon concentrations are below the investigation level of $200 \text{ Bq}\cdot\text{m}^{-3}$ [9].

The present work aims to contribute to the regulatory authority in establishing standards, from the radiological point of view, for application of phosphogypsum in civil construction.

Acknowledgments

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References

- [1] International Atomic Energy Agency (IAEA), 2005. Derivation of activity concentration values for exclusion, exemption and clearance. Safety Reports Series No. 44. Vienna, 2005.
- [2] B.P. Mazzilli, V. Palmiro, C.H.R. Saueia and M.B. Nisti. *J. Environ. Radioactivity*, 49: 113-122 (2000).
- [3] A.J.G. Santos, P.S.C. Silva, B.P. Mazzilli and D.I.T. Fávoro. *Radiat. Prot. Dosimetry*, 121(2): 179-185 (2006).
- [4] B.P. Mazzilli and C.H.R. Saueia. *Radiat. Prot. Dosimetry*, 86(1): 63-67 (1999).
- [5] W.M. Kanno, H.L. Rossetto, M.F. Souza, M.F. Máduar, M.P. Campos and B.P. Mazzilli, in: *Book of Abstracts of the 8th International Symposium on the Natural Radiation Environment (NRE VIII), Búzios, Brazil, 2007*.
- [6] M.F. Máduar and G. Hiromoto, *Radiat. Prot. Dosimetry*, 111(2): 221-228 (2004).
- [7] European Commission on Radiation Protection (ECRP 112), Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials (ECRP, Brussels, 1999).
- [8] Radiation Protection Authorities in the Nordic Countries (RPANC), Naturally Occurring Radioactivity in the Nordic Countries – Recommendations (RPANC, 2000).
- [9] International Commission on Radiological Protection (ICRP Publication 65), Protection against Radon-222 at home and at work. (Oxford, Pergamon Press, 1993).