Absorbed Gamma-Ray Doses due to Natural Radionuclides in Building Materials

Vitor A.P. Aguiar[a], Nilberto H. Medina[a], Ramon H. Moreira[b], Marcilei A.G. Silveira[b] and José R.B. Oliveira[a]

[a]*Instituto de Física,Universidade de São Paulo, SP, Brazil.*
[b]*Departamento de Física,Centro Universitário da FEI, São Bernardo do Campo, SP, Brazil.*

Abstract. This work is devoted to the application of high-resolution gamma-ray spectrometry in the study of the effective dose coming from naturally occurring radionuclides, namely ^{40}K, ^{232}Th and ^{238}U, present in building materials such as sand, cement, bricks and granitic gravel. Four calculation models were applied to estimate the effective dose and the hazard indices. The maximum estimated effective dose coming from the three reference rooms considered is 0.90(22) mSv/yr, and maximum internal hazard index is 0.77(14), both for the compact clay brick reference room. The principal gamma radiation sources are cement, sand and bricks.

Keywords: gamma-spectrometry, building materials, hazard indices, effective dose.
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INTRODUCTION

The most important source of ionizing radiation to which humans are exposed are the naturally occurring radionuclides; the most common are ^{40}K and elements of ^{232}Th and ^{238}U decay families, elements that are present on Earth since its formation [1,2], besides the cosmic rays, which are highly energetic particles coming from space. The radionuclides cited may be found almost everywhere, and there are several studies on this subject [3,4].

Focusing on radiation dosimetry, it is important to measure the activity of these isotopes, in order to establish low dose limits, and, more importantly, check areas/foods/materials potentially dangerous for human health. Since building materials are made of materials found in nature, we find these radioisotopes in many of the building materials used in the civil construction, and many studies were carried out with the aim of quantifying these nuclides [4-7], including in Brazil [8].

The gamma radiation from the ^{40}K isotope (which represents only 0,0117% of natural potassium in nature) comes from the electron capture decay (10,7% of the cases) to the stable ^{40}Ar isotope, emitting a gamma-ray of 1460 keV (the other 89.3% β^- decays producing the stable nucleus ^{40}Ca). The lifetime of ^{40}K is 1.25x10^9 yr [9]. The decay-chains of ^{232}Th and ^{238}U are sequences of unstable nuclei that decay emitting gamma-rays, alpha or beta particles until the series stop at the stable nuclei ^{208}Pb and ^{206}Pb, respectively [9].

When studying the radioactivity hazard in closed environments, it is not sufficient to consider just the activity concentration of each radionuclide: it is important to use a mathematical model that takes into account the geometry of the room. In this work four models were considered and a comparison with *in-situ* values [10, 11] was carried out.

MATERIALS AND METHODS

We have collected 2-4 different samples of each of the most used building materials in Brazil: clay bricks (compact and hollow types), Portland cement, granitic gravel and sand. The samples of stones and bricks were milled to powder and all samples were dried and sealed in 200 cm^3 polyethylene cylindrical recipients. The samples were stored for 30 days, in order to reach secular equilibrium for radon and its daughters.

The gamma-ray activities were measured using gamma spectrometry technique, with a 20% efficient HPGe detector placed inside a 15 cm thick homemade lead shield. Upak software was used for data acquisition and analysis [12]. For energy calibration, a ^{60}Co standard source was used, and the activity calibrations were carried out using standard samples with known concentrations of ^{40}K, ^{232}Th and ^{226}Ra. The data for each sample has been collected for nearly 12 hours. A gamma-ray background measurement of about 24 hours was acquired.

RESULTS AND DISCUSSION

The activity concentration, under the assumption of secular equilibrium, was calculated by comparison of the areas of the photopeaks of ^{40}K (1460 keV), the uranium daughter ^{214}Bi (1764 keV) and the thorium daughter ^{208}Tl (2613 keV), of each sample with the standard samples. Figure 1 shows a gamma-ray spectrum obtained for a Portland Cement sample, and Table 1 presents mean values for activity concentrations:

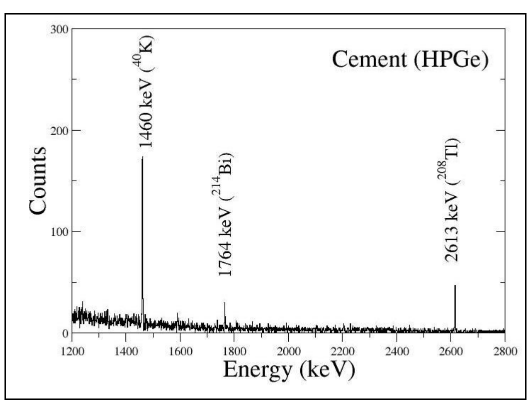

FIGURE 1. A gamma-ray spectrum of a Portland Cement sample analyzed.

TABLE 2. Mean activity concentrations of the samples analyzed.

Activity Concentrations (Bq/kg)			
Sample	**^{40}K**	**^{232}Th**	**^{226}Ra**
Portland Cement	144(19)	43(12)	88(3)
Sand	366(38)	70(4)	33(3)
Hollow Clay Brick	32(22)x10	82(26)	29(12)
Compact Clay Brick	154(52)	138(46)	53(19)
Granitic Gravel	43(19)x10	47(24)	73(22)

Errors in Table 1 are given by standard deviation of the mean. These values may be overestimated, since we have measured only few samples.

As mentioned before, four models were applied for calculation of the radiological hazard. The first model, proposed by [7] is based on a reference room model, which is an idealized room with pre-determined dimensions and building material percentages [7]. In this work, as in a previous article [1], we considered three types of reference rooms: constructed with compact bricks, hollowed bricks or concrete blocks, since all three types of buildings can be easily found in Brazil. More details of this model can be found in Ref. [1,7]. The dimensions of the reference rooms considered are: 5.0 x 5.0 x 3.0 m^3. As suggested by Ref. [6], the weight factors of the radiations for a closed environment dose estimation were 0.054 (nGy/h)/(Bq/kg) for ^{40}K, 0.62 (nGy/h)/(Bq/kg) for ^{232}Th and 0.89 (nGy/h)/(Bq/kg) for ^{238}U, different, therefore, from those used in an infinite-plane approximation.

To transform the absorbed dose in air into effective dose, the conversion factor is 0.7 Sv/Gy [2]. We have also taken into account the fraction of time spent indoors by a person, which was assumed to be 0.8 [2,7]. According to Ref. [7], the effective dose is given by Eq. 1:

$$D_{eff} = pTb \times 10^{-6} \sum_i \left[\left(q_K C_{K,i} + q_{Th} C_{Th,i} + q_U C_{U,i} \right) m_i \right]$$

In Eq. 1 the q symbols are the weight factor for each radiation, the C factors are the activity concentrations of the referred isotope in the ith material, m_i is the mass of the material in the reference room environment, p is the fraction of time spent indoors, T is the total hours of the year and b is the conversion coefficient of absorbed dose to effective dose [1,7]. This dose estimation can be compared with the upper limit established by ICRP [13] of 1.5 mSv/yr.

The other three models are based on the so called Hazard Index [4], which is an index that accounts for the gamma-rays effective dose relative to the limit of ICRP. The first HI model is used for a room with walls of infinite thickness and no windows or doors; the second model is used for finite walls with windows and doors. These two models are called the External Hazard Indices (we may call the second model the "corrected external hazard index"). The third model takes into account the internal exposure to radon and its daughters, through respiration, and is called the Internal Hazard Index. The formulas for calculating these indices, as given in Ref. [4], are presented in Eq. 2, 3 and 4, respectively:

$$H_{ex} = \left(C_{Ra}/370\right) + \left(C_{Th}/259\right) + \left(C_K/4810\right)$$
$$H_{ext}^{corr} = \left(C_{Ra}/740\right) + \left(C_{Th}/520\right) + \left(C_K/9620\right)$$
$$H_{\mathrm{int}} = \left(C_{Ra}/185\right) + \left(C_{Th}/259\right) + \left(C_K/4810\right)$$

A dose is considered insignificant if the values of these indices are lower than unity. It must be noted that these models do not take into account the type of construction, but only the use of each material in a construction. In order to correct this assumption and make these models more realistic and comparable with the first model, we propose a modification on these models, where we calculate the hazard index by a weighted fraction of each material, as used in the calculations for the reference rooms. Table 2 shows the results for all the models considered.

TABLE 2. Effective absorbed doses and hazard indices in the reference rooms.

Effective Absorbed Dose and Hazardous Indices in the Reference Room				
Reference room type	**Effective dose (mSv/yr) (model nº1)**	**H_{EX} (model nº2)**	**H_{EX}^{CORR} (model nº3)**	**H_{INT} (model nº4)**
Compact clay brick	0.90(22)	0.62(12)	0.31(6)	0.77(14)
Hollow clay brick	0.68(18)	0.45(6)	0.225(28)	0.56(6)
Concrete block	0.71(10)	0.44(3)	0.221(12)	0.58(3)

As can be seen from Table 2, all four models considered for the three types of construction that can be encountered in Brazil agree in the fact that the radiation dose is under the limits of ICRP [13], that is, 1.5 mSv/yr for the effective dose and a value below unity for the hazard indices. Therefore, uncertainties reveal that the rooms made of compact bricks show a value of effective dose compatible with 1.5 mSv/yr, and internal hazard index compatible with unit. From Table 2 we also note that the room type with the highest dose contribution is the one made of compact clay brick, which also has higher values for the hazard indices. This is due to the higher activity concentration of sand and gravel for ^{40}K and cement and granitic gravel for ^{226}Ra, besides the higher activity of ^{232}Th in the bricks, as can be seen on Table 1. We must also mention that our modified model for the hazard indices only takes into account the composition of the walls, but not its geometry. Other calculation models must take this into consideration, just like the reference room model.

The last analysis carried out was a comparison of the order of magnitude with some *in-situ* measurements in São Paulo City [10,11], performed by the Dosimetry Laboratory of the Physics Institute, São Paulo University. In Ref. [10] the authors analyzed the gamma-ray dose in closed commercial establishments, like shopping centers, banks and commerce in general. On the other hand, Ref [11] shows the results of the gamma-ray dose in open environments and also the contribution of the cosmic rays. Subtracting the median value of Ref. [11] (0.710 mSv/yr) and 80% of the cosmic radiation contribution (0.096 mSv/yr), from the mean values for commerce in general (street stores) (1.547 mSv/yr) [10], we find the value of 0.741 mSv/yr, which has the same order of magnitude of the values encountered in our analysis. We must emphasize that the values of Refs. [10] and [11] are affected by a high variability on the construction materials, sizes of the rooms and soil composition.

Values of Ref. [8] are also comparable with the ones encountered in this work. Differences arise mainly in the construction geometry, percentages of each material used and in the composition of each material, which can vary significantly.

CONCLUSIONS

This work showed that the type of building material is of great influence on the effective dosis, since its content of ^{40}K, ^{232}Th and ^{226}Ra is variable. The buildings made of compact clay bricks, concrete, cement and sand are those for which the effective dose is the highest, namely 0.90(22) mSv/yr for the reference room model adopted. The values of the calculated hazard indices are all below unity, but in the case of the compact clay brick room the internal hazard index is compatible with unity, and, in principle, there may be a risk for inhabitants in using the materials analyzed in this work. The values encountered are inside the range of variation of the data obtained in many countries [2].

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