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GAMMA RAY DOSES PROCEEDING FROM NATURAL OCCURING RADIONUCLIDES IN CLOSED ENVIRONMENTS.

Vitor Angelo P. de Aguiar[1], Nilberto H. Medina[1], Marcilei A. Guazzelli da Silveira[2], and Ramon H. Moreira[2]

[1]Departamento de Física Nuclear – Instituto de Física da USP
Universidade de São Paulo
Rua do Matão, travessa R 187
05508-090 São Paulo, SP
vitor.angelo.aguiar@usp.br
medina@if.usp.br

[2]Departamento de Física
Centro Universitário da FEI
Av. Humberto de Alencar Castelo Branco 3972
09850-901 S. B. do Campo, SP
shila@if.usp.br
rheleno@hotmail.com

ABSTRACT

In this work we report on the application of gamma-ray spectrometry in the study of the effective dose coming from terrestrial natural elements present in building materials such as sand, cement, lime (CaO) and milled granitic stones. The major contribution to annual gamma-ray radiation effective dose is due to the natural occurring radionuclides ^{40}K, ^{232}Th and ^{238}U. Two spectrometry systems were employed to measure the gamma radiation: one with a 60% efficient GeHP detector and the second one with a 2"x2" NaI(Tl) scintillator. The estimated effective dose coming from the three reference rooms assumed is 0.63 mSv/yr, proceeding from terrestrial natural elements. The principal gamma radiation sources are cement, sand and bricks.

1. INTRODUCTION

Human beings are always exposed to ionizing radiations, such as X-rays, gamma rays, charged particles, etc. There are two principal sources of these radiations in natural environment: the cosmic rays, which come as a result of the interactions of high-energy particles, coming from outer space, with our atmosphere; and the natural occurring radionuclides, which have always been present in Earth since its formation [1]. These radionuclides, such as ^{40}K and the ^{238}U and ^{232}Th families, may be found in our bodies, in our food [2], and in the walls of our houses [3].

In fact, since the building materials are made of minerals which are found in nature, we can expect that the same radionuclides present in a sample of soil may be present in a sample of cement or bricks, for example. The interest for quantifying the effects of indoor radiation exposure due to the radionuclides present in the building materials has started a long time ago [4,5], and it was shown by several studies that the higher contribution for the gamma-ray dose is due to ^{40}K and the ^{238}U and ^{232}Th decay series. The gamma radiation from the ^{40}K nuclide comes from the electron capture decay to the stable ^{40}Ar isotope, emitting a gamma-

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ray of 1460 keV, which occurs in 10.7% of the cases; the other 89.3% β^- decays producing ^{40}Ca with a lifetime of 1.3x10^9 yr [6]. The chains of ^{232}Th and ^{238}U are sequences of unstable nuclei that decay emitting gamma-rays, alpha or beta particles until the stable nuclei ^{208}Pb and ^{206}Pb, respectively, are reached [6]. In order to estimate the activity concentration of these nuclei in each sample, we analyzed the gamma-rays emitted by ^{40}K (1460 keV), the uranium daughter ^{214}Bi (1764 keV) and the thorium daughter ^{208}Tl (2613 keV) [6]. On the other hand, the concentration measurement of these radionuclides in building materials is not enough to estimate the absorbed dose in a human being placed inside a closed environment; it is necessary to consider the geometry of the room to correctly calculate the absorbed dose. Several models have been considered in the past years [4,7].

This work focused on the measurement of the activity concentrations of ^{40}K nuclide and ^{232}Th and ^{238}U families in the most common building materials used in Brazil, such as Portland cement, clay bricks and milled granitic stones. The results were obtained taking into account one calculating model for the absorbed dose in three reference rooms and compared with experimental results obtained *in situ* [8].

2. EXPERIMENTAL PROCEDURES

We have collected one sample of each of the most used building materials in Brazil: clay bricks, the compact type and the holed type, Portland cement, milled granitic stones, lime and sand, collected in the region of Jacareí, São Paulo State, Brazil. The samples of stones and bricks were crushed in a mill until its medium sized grains were about 150 µm large. Lime, cement and sand samples were already in a suitable grain size. 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 two experimental systems: a NaI(Tl) scintillator detector, placed inside a 7.0 cm thick Canberra lead shield, with a Genie 2000 software for spectrum analysis and the UniSpec integrated multi-channel analyzer, and a 20% efficient HPGe detector placed inside a 15 cm thick homemade lead shield, with the Upak software for data acquisition and analysis [9]. For energy calibration, a ^{60}Co standard source was used, and the activity calibrations were carried out using standard samples with known concentration of K, ^{232}Th and ^{226}Ra. The data for each sample has been collected for nearly 12 hours, and a background measurement acquired for about 24 hours.

3. EXPERIMENTAL RESULTS AND DISCUSSION

The results for the specific activities are shown on Table 1. As it can be seen, the results for both experimental systems are compatible for all the building materials, except for the holed clay brick, in the case of the activity concentration of ^{40}K. The Table also shows that the highest activity concentration is that of ^{40}K isotope. Lime showed just a little specific activity of ^{40}K isotope, and negligible contribution from ^{232}Th and ^{238}U. Due to the higher resolution of the HPGe system, it was possible to detect a contribution of ^{232}Th in milled granitic stone, which was not found with the NaI(Tl) system. Figures 1 and 2 show the gamma-ray spectra obtained for cement and holed clay brick samples, with the two systems.

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Table 1. Activity concentrations of the samples analyzed

Sample	Activity Concentrations (Bq/kg)					
	^{40}K		^{232}Th		^{238}U	
	HPGe	NaI(Tl)	HPGe	NaI(Tl)	HPGe	NaI(Tl)
Compact Clay Brick	76(3)	53(11)	70(3)	64(7)	16.2(7)	31(9)
Holed Clay Brick	99(4)	56(12)	56.2(22)	50(7)	17.2(7)	17(9)
Portland Cement	108(6)	122(11)	19.7(10)	15(8)	93(5)	113(12)
Milled Granitic Stone	54(6)	78(8)	2.0(2)	LLD	50(5)	63(9)
Lime	17.4(9)	22(10)	LLD	LLD	LLD	LLD
Sand	315(12)	264(15)	74(3)	77(10)	35.3(14)	32(10)

*LLD – Lower than the limit of detection

It must be mentioned that these results show good agreement with previous results from other authors for activity concentrations of the isotopes analyzed in those building materials [8].

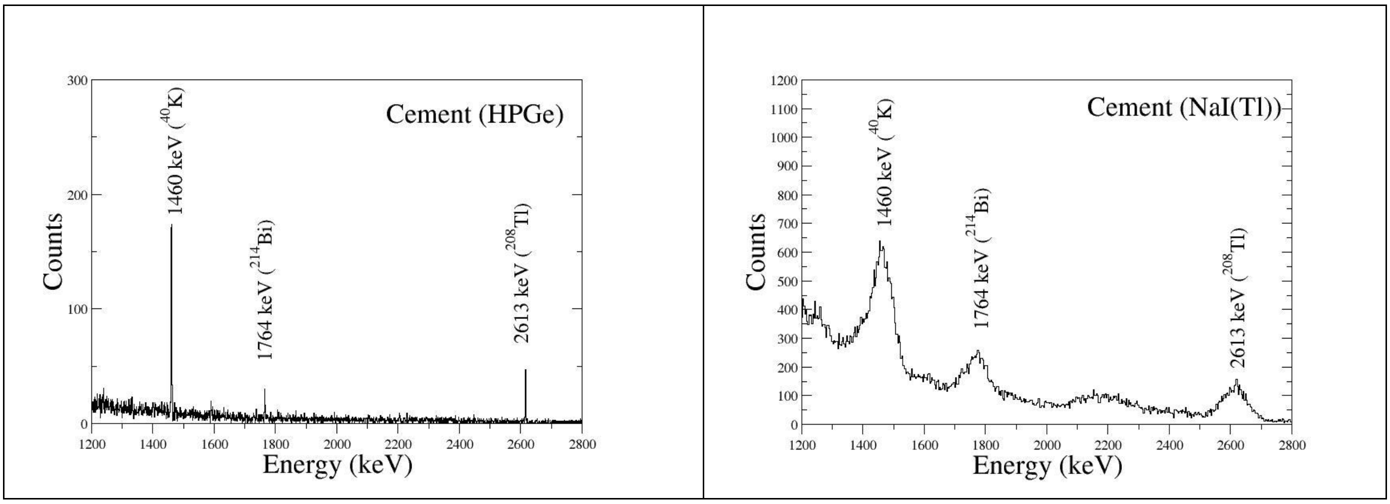

Figure 1. Gamma radiation spectrum obtained with a HPGe detector (left) and NaI(Tl) scintillator (right) for the cement sample.

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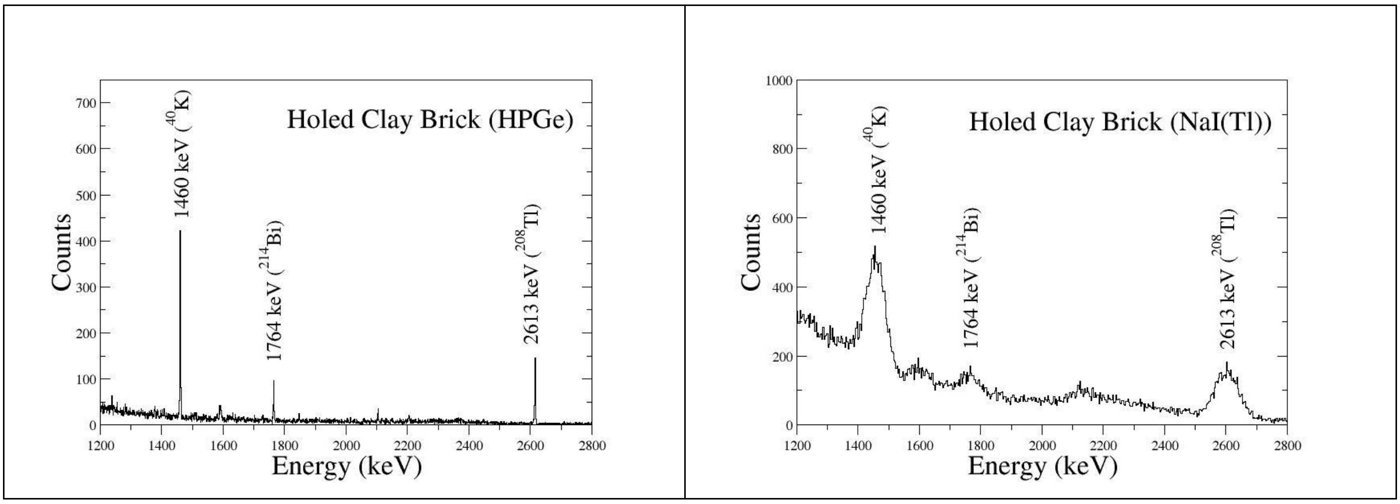

Figure 2. Gamma-ray spectra obtained with a HPGe detector (left) and Nai(Tl) scintillator (right) for the holed brick sample.

In order to calculate the effective dose in a closed environment, it is necessary to consider a reference room, which is an idealized room with pre-determined dimensions and building materials percentages [7]. In this work we considered three types of reference rooms: a compact brick made, a holed brick made and a concrete block made. The reason for these considerations was that, in Brazil, all these types of buildings can be easily found. All these models consider that: 1) the walls are made of the principal building material considered, plus cement and sand, in proportions commonly used by the constructors; 2) the floor was made of concrete (a mixture of sand, cement and milled granitic stones in a appropriate proportion) and the roof was made of the same material of the walls, but with a smaller thickness. The dimensions of the reference rooms used are: 5.0 x 5.0 x 3.0 m^3.

Accordingly to Ref. [1], the dose in a closed environment can be twice as large as the dose contribution in an open environment with the same activity concentration of the isotopes considered. It is suggested by Ref. [7] that the weight factors of the radiations must be altered from those used for the infinite-plane approximation. The weight factors used are then: 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. To transform the absorbed dose in air, calculated using the mentioned weight factors, into effective dose, we have taken into account another conversion factor for external terrestrial gamma radiation which is 0.7 Sv/Gy [1]. It was also taken into account also the time fraction spent indoors by a person, which was assumed to be 0.8 [1,7]. Accordingly to Ref. [7], the effective dose is given by:

$$D_{eff} = pTb \times 10^{-6} \sum_i \left[\left(q_K C_{K,i} + q_{Th} C_{Th,i} + q_U C_{U,i} \right) m_i \right] \quad . \qquad (1)$$

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Where the q symbols are the weight factor for each radiation, the C factors are the activity concentrations of the referred isotope in the i^{th} material, m_i is the mass of the material in the reference room environment, p is the time fraction spent indoors, T is the total hours of the year and b is the conversion coefficient of absorbed dose to effective dose. Table 2 shows the results obtained for effective doses for three different types of reference room.

Table 2. Effective doses for three different types of reference room

Effective absorbed dose in the reference room (mSv/yr)			
Reference room type	**HPGe**	**NaI(Tl)**	**Mean**
Compact clay brick	0.493(23)	0.54(9)	0.52(9)
Holed clay brick	0.513(21)	0.51(9)	0.51(9)
Concret block	0.617(27)	0.64(11)	0.63(11)

As it can be seen in Table 2, the reference room type which most contributes to the total gamma-ray effective dose is that made of concrete block. This result could be expected, since cement, sand and stone have the highest activities concentrations for ^{40}K and ^{238}U. In the uncertainty estimation it was not taken into account the features of the reference rooms.

A comparison with experimental data obtained in São Paulo city [8] in various types of commercial establishments, such as banks, shopping centers and street stores, was carried out. The results of Ref. [8] showed that the effective dose for the banks and the other commercial establishments is 1.6(3) mSv/yr, which is about 1 mSv/yr higher than the greatest value obtained by this work. It is important to mention two assumptions on this comparison: 1) in this work there was no dose contribution from cosmic rays, since we considered only the three gamma-rays from natural occurring radionuclides. In Ref. [8], there was an unknown contribution from cosmic rays, but it should be less than 0.22 mSv/yr; 2) the reference room considered in this work may be much smaller than the environments analyzed in Ref. [8], which would decrease the dose value obtained. Despite these discrepancies, the comparison of the values encountered in this work and the mean of all the results published in the literature [10] shows a good agreement among them: the mean of all the results are between 0.24 and 2.29 mSv/yr.

In this study only the effective dose due to gamma-rays coming from the walls was considered – we have not studied the effects of radon and radon daughters. Nevertheless, there are many studies focused specifically on this subject.

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4. CONCLUSIONS

This work showed that the major contribution to the total effective dose is due to the radionuclides ^{40}K and those of ^{232}Th decay chain. It was also showed that the type of building material is of great influence. The buildings made of concrete blocks, concrete, cement and sand are those under which the effective dose is the highest, namely 0.63 mSv/yr for the reference room adopted. The values encountered are inside the range of variation of the data obtained in many countries, but are smaller than the ones collected *in situ* in São Paulo city. This must be due the fact that real constructions may have different building materials and/or dimensions than the reference room assumed in this work.

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REFERENCES

1. UNSCEAR, 2000. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York.
2. V.A.B. Zagatto, N.H. Medina, N.K. Umisedo, E. Okuno, "Natural Radioactivity in Bananas". In: *8th International Symposium on Natural Radiation Environment, 2008, Búzios, RJ. AIP Conference Proceedings.* Melville, NY, USA. American Institute of Physics, 2008. **v. 1034**. p. 264-268.
3. X. Lu, F. Wang, X. Jia, and L. Wang, "Radioactive Analysis and Radiological Hazards of Lime and Cement Fabricated in China", *IEEE Transactions on Nuclear Science*, v. **54**, p. 327-332 (2007).
4. E. Stranden, "Radioactivity of building materials and the gamma radiation in dwellings", *Phys. Med. Biol*. **v. 24**, p. 921-930 (1979).
5. K.D. Cliff, B.M.R. Green and J.C.H. Miles, "The levels of radioactive materials in some common UK building materials", *The Science of the Total Environment*, **v. 45**, p. 181-186 (1985).
6. http://www.bnl.nndc.gov . last access 05/07/2009
7. J.G. Ackers, B.F.M. Bosnjakovic, L. Strackee, "Limitation of radioactivity concentrations in building materials based on a practical calculation model", *Radiation Protection Dosimetry*, **v. 7**, p. 413-416 (1984).
8. S.M. Otsubo, 2001. Dissertação de Mestrado. IFUSP
9. W. Milner, Upak Notes, Oak Ridge National Lab. (1986). Private communication
10. UNSCEAR, 1993. Sources and effects of ionizing radiation. United Nations Scientific Committee on the Effects of Atomic Radiation, United Nations, New York.