Dose rates evaluation of some granitic rocks from the Paraná State

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ABSTRACT

Granitic rocks, widely used as building materials, are known to contain natural radionuclides and can be an important source of radiation for the population. Thirty-four samples of granite rocks from geological occurrences in Paraná state were measured with detector for evaluation of ^{226}Ra, ^{232}Th and ^{40}K activity concentrations. The effective annual external dose was evaluated from these radionuclides activities using a dosimetric room model with dimensions 4 m x 5 m x 2.8 m in which all walls are internally coated with 2 cm thickness granites and considering the annual exposure time of 7000 h, suggested by the European Commission on Radiological Protection for superficial coating materials. The internal exposure was evaluated from radon air concentration of the model room, simulated from an exhalation rate of ^{222}Rn, determined with CR-39 solid state nuclear track detectors by the sealed can technique, considering a ventilation rate of 0.5 h^{-1} and the same annual exposure time of 7000 h. The results for external gamma rays showed an increase in the annual effective dose ranging from 96 ± 4 μSv.a^{-1} to 223 ± 7 μSv.a^{-1} and, for radon inhalation, an increase in the ranging from 0.4 ± 0.04 μSv.a^{-1} to 70 ± 4 μSv.a^{-1}. All results stayed below the recommended value by the European Commission on Radiological Protection, of 1 mSv.a^{-1}.

Keywords: granitic rocks, radon exposure, annual effective dose.

1. INTRODUCTION

Natural radionuclides present in building materials (homes, schools, shops) can promote mankind exposure to radiation, mainly due to external gamma dose and internal dose of radon.

The external gamma dose originates from exposure to natural radionuclides that may occur in isolated form or in radioactive series. The ^{238}U, ^{235}U and ^{232}Th series and the ^{40}K single radionuclide represent 16.97% of the mean annual effective dose. The internal dose results mainly from inhalation of the radon isotope, ^{222}Rn, a noble gas, daughter of ^{226}Ra, a decay product of ^{238}U series. Inhalation of radon represents 47.6% of the mean annual effective dose due to natural radionuclides [1].

Rocks with high radioactivity content can be an important dose source when used as building materials (structural and/or coating).

The geology of the state of Paraná has several types of rocks. Of those, granites have their main application in construction as coating rocks. The Crystalline Shield of Paraná is the main source of this type of rock and there are no studies on radiological emission. In this way, the objective of this work is to evaluate the internal dose due to radon and the external dose due to gamma radiation, caused by granite rocks of geological occurrence in the state of Paraná, used in civil construction, as structural and/or coating materials [2, 3].

2. MATERIALS AND METHODS

2.1. Sample area and collection

A total of 34 samples were obtained in the Metropolitan Region of Curitiba (MRC), Figure 1, representing two factories that are responsible for almost all rocks extraction for internal cladding purposes in the state of Paraná. Samples were obtained as commercialized, in plates, with 15 cm x 15 cm x 2 cm (width, length and thickness, respectively).

Figure 1: *Maps of the study area in: A. Brazil; B. Paraná State; and C. Metropolitan Region of Curitiba (MRC), in red is highlighted the city of Curitiba.*
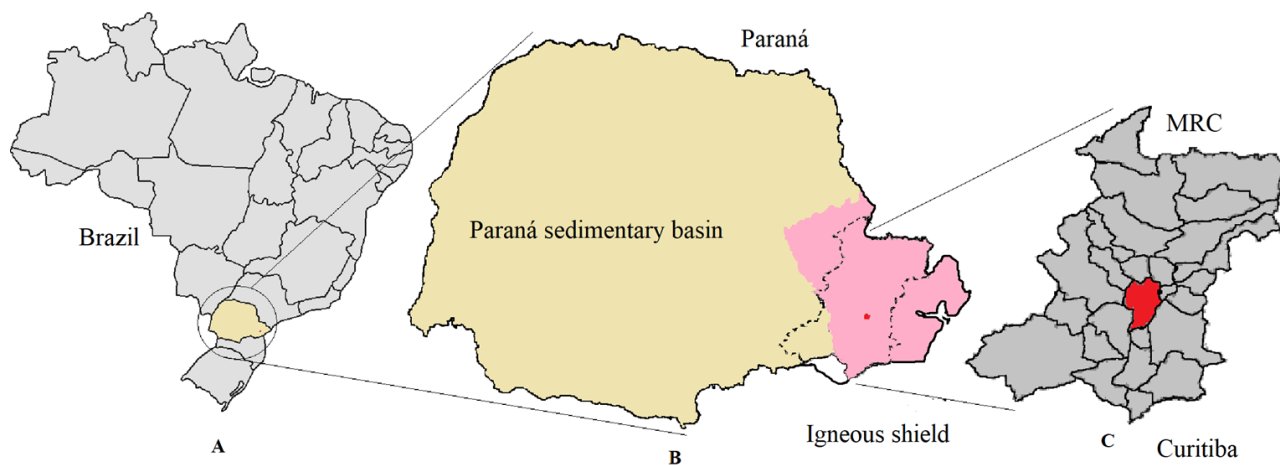
Source: Adapted from [2]

2.2 Samples analysis

2.2.1 ^{226}Ra, ^{232}Th and ^{40}K activities concentrations

The ^{226}Ra, ^{232}Th and ^{40}K activities concentrations were determined by high resolution gamma ray spectrometry [2]. All samples were first pulverized to 60 Mesh, tightly sealed in standard 100-mL HDPE flat-bottom cylindrical flasks with screw cap and bubble spigot and stored for approximately 4 weeks, in order to ensure radioactive secular equilibrium. The samples were measured during 86000 seconds, with an ORTEC GEM coaxial high-purity germanium detector (HPGe) of 15% relative efficiency and 2.8 keV effective resolution for the 1.33 MeV transition of ^{60}Co, with conventional electronics and a 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer and afterwards analyzed with the InterWinner 6.0 software [4]. The background radiation was determined by measuring an ultra-pure water sample and the detector efficiency curve was determined with a multi-element standard aqueous radioactive solution sample, both in the same geometry of the samples [2, 3]. As samples densities ranged from 1,57 g.cm^{-3} to 2.02 g.cm^{-3} and the standard solution density is 1 g.cm^{-3}, measurements were made to determine the self-attenuation factors for all samples [5, 6].

The activity concentration of ^{226}Ra was determined by the weighted mean of the ^{214}Pb and ^{214}Bi gamma transitions and the activity concentration of ^{232}Th by the weighted mean of the ^{212}Pb, ^{212}Bi and ^{228}Ac gamma-ray transitions. The activity concentration of ^{40}K was calculated through its single gamma transition of 1461 keV. All uncertainties were calculated by error propagation [2, 3].

1.1. 2.2.2 ^{222}Rn activities concentrations

The determination of radon was performed by the Sealed-can passive detection technique with SSNTD CR-39 detectors [2, 7]. The CR-39 detectors were placed inside a diffusion chamber model NRPB/SSI, attached to the top of a cylindrical vessel of 26.5 cm height and 23.5 cm diameter. The sample, as purchased (a granite slab of 15 cm length x 15 cm width x 2 cm thickness was placed at the bottom of the container.

Seeking an optimization between counting statistics and measurement time, the can was kept sealed for 30 days [2]. After, the CR-39 detectors were subjected to standard chemical etching with a 30% KOH solution at 80°C for 5.5 hours in a shaking water bath [7]. The tracks were manually counted with a Zeiss optical microscope and the KS100 version 3.0 software [8].

2.3   Annual effective doses

1.2. 2.3.1 Annual effective dose due to external gamma-rays

For assessing the Annual Effective Dose, it is necessary the knowledge of the absorbed dose rate. For construction materials, this indoor rate can be calculated with dosimetric models based on gamma activities concentrations.

In this study, it was used the European Commission of Radiological Protection suggested standard model room (4m x 5m x 2.8m) [9]. The dose rate absorbed in the air was derived from superficial materials (thickness 3 cm and density 2.6 g.cm^{-3}, disregarding doors and windows). For this configuration of coating rocks the dose rate is given by equation 1, as

$$\dot{D} = 0.12A_{Ra} + 0.14A_{Th} + 0.0096A_k \tag{1}$$

Where $\dot{D}$ is the absorbed dose rate taxa in the air in nGy.h^{-1} and A$_{Ra}$. A$_{th}$ and A$_K$ are the activity concentrations of ^{226}Ra, ^{232}Th e ^{40}K in Bq.kg^{-1}, respectively.

The dose conversion coefficients 0.12, 0.14 and 0.0096, were calculated with the Markkanen mathematical model [10] for the adopted scenario of a standard room of 4m x 5m x 2.8m and coating

walls with rocks of thickness 3 cm and density 2.6 g.cm^{-3}. Considering that the studied samples have thickness of 2 cm and density ranging from 2.55 ± 0.01 g.cm^{-3} to 2.86 ± 0.01 g.cm^{-3}, with an average density value of 2.63 ± 0.03 g.cm^{-3}, the addopted model will lead to over-estimated dose results, regarding radiological protection and safety. If any of the dose results exceed the established limits, other models must be used to calculate doses for the samples specific parameters [2, 11, 12].

1.3. 2.3.2 Annual effective dose due to radon inhalation

The Annual Effective Dose due to the increase of the radon concentration caused by the building materials in a residence, $D_{ef(Rn)}$, is calculated by equation 3 [2, 9, 10].

$$D_{ef(Rn)} = C_{Rn} x 20 \frac{\mu Sv}{Bq.m^{-3}} \tag{3}$$

Where C_{Rn} is the radon concentration in the standard model room considered (4m x 5m x 2.8m), given in Bq.m^{-3}, and 20 is the derived conversion factor considering an annual exposure time of 7000 h inside the standard room and an equilibrium factor of 0.5 [9].

The radon concentration C_{Rn} was calculated from the surface exallation rate of radon of the samples, measured by the sealed can technique. [2, 10].

The equilibrium factor is dimensionless and refers to the radioactive balance between the radon and its progeny in the environment (considered). In a sealed environment this value is 1, indicating total equilibrium. In real conditions this does not occur because radon behaves differently from its daughters.

3. RESULTS AND DISCUSSION

Figure 3 presents a typical gamma-ray spectrum of the granitic rock sample # 34, with some transitions of all radionuclides used in the calculations marked.

Table 1 shows the results of ^{226}Ra, ^{228}Ra and ^{40}K gamma activities concentrations together with the radon concentrations for all 34 granitic rocks samples.

Figure 3: *Gamma ray spectrum of granitic rock sample # 34, counting time 86 ks.*
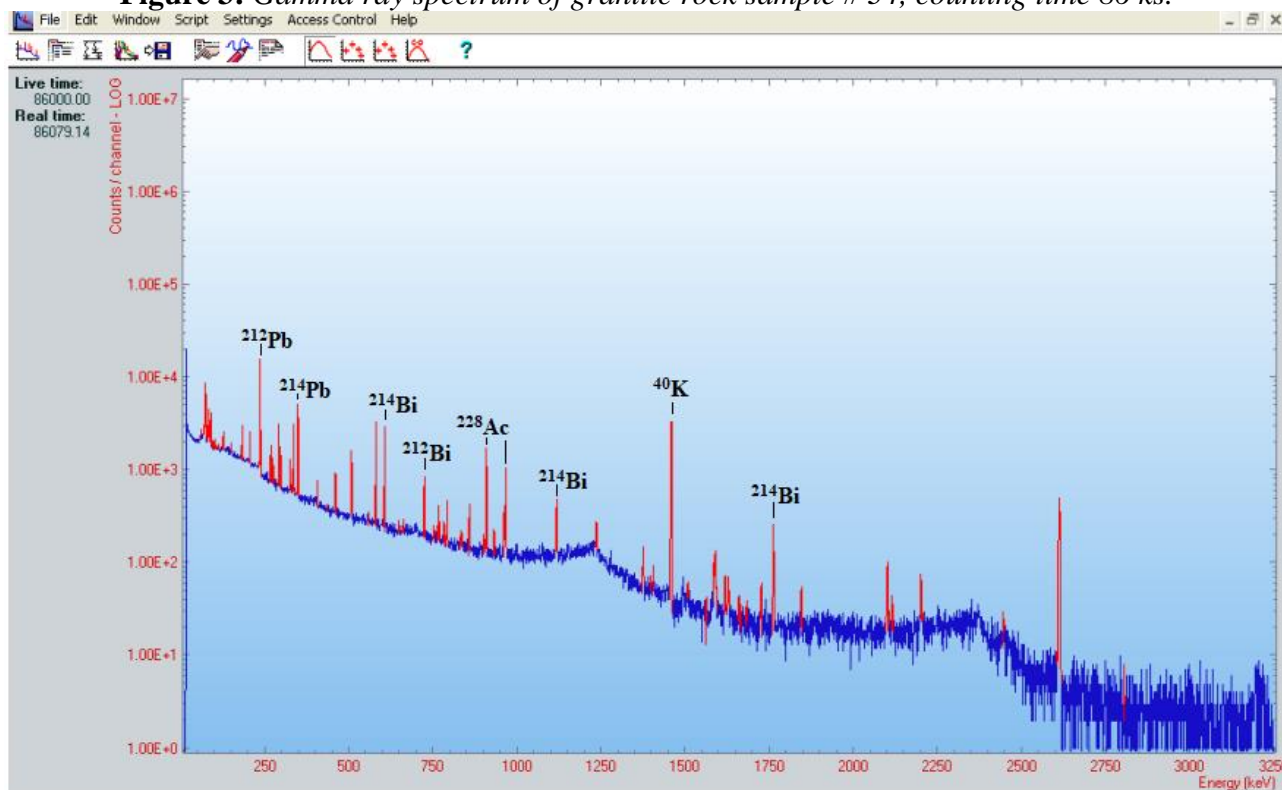
Table 1: Gamma activity concentration and radon concentration in granitic rocks samples from Curitiba Metropolitan Region [2].

Sample Code	(Bq.kg^{-1})			(Bq.m^{-3})
	^{226}Ra (A$_{Ra)}$	^{232}Th (A$_{Th)}$	^{40}K (A$_K$)	^{222}Rn
1	26 ± 1	58 ± 3	1335 ± 58	452 ± 21
2	59 ± 3	37 ± 2	1227 ± 63	348 ± 21
3	29 ± 2	72 ± 4	1422 ± 83	619 ± 35
4	31 ± 1	84 ± 3	1317 ± 53	773 ± 28
5	50 ± 3	123 ± 6	1194 ± 51	501 ± 35
6	20 ± 2	30 ± 2	1356 ± 72	264 ± 14
7	31 ± 3	47 ± 4	1560 ± 109	536 ± 28
8	38 ± 2	55 ± 3	1562 ± 78	675 ± 35
9	59 ± 4	59 ± 4	1265 ± 79	668 ± 35

10	25 ± 2	63 ± 3	1328 ± 71	181 ± 14
11	32 ± 2	46 ± 3	1228 ± 68	369 ± 21
12	35 ± 3	64 ± 5	1560 ± 117	473 ± 28
13	40 ± 2	80 ± 5	1331 ± 62	522 ± 28
14	50 ± 3	83 ± 6	1504 ± 104	612 ± 35
15	19 ± 1	30 ± 3	1507 ± 87	139 ± 7
16	32 ± 2	53 ± 4	1377 ± 67	132 ± 7
17	50 ± 5	90 ± 8	1308 ± 133	223 ± 14
18	29 ± 3	49 ± 3	1457 ± 78	376 ± 21
19	28 ± 2	46 ± 2	1218 ± 62	70 ± 7
20	34 ± 2	59 ± 3	1347 ± 55	84 ± 7
21	46 ± 2	68 ± 4	1230 ± 67	1274 ± 63
22	50 ± 3	90 ± 5	1327 ± 79	995 ± 49
23	54 ± 3	88 ± 5	1191 ± 67	35 ± 7
24	25 ± 1	49 ± 3	0935 ± 46	14 ± 7
25	24 ± 2	44 ± 4	1400 ± 78	571 ± 28
26	27 ± 2	43 ± 3	1351 ± 63	606 ± 28
27	77 ± 4	66 ± 4	1336 ± 77	1030 ± 49
28	51 ± 3	44 ± 3	1279 ± 75	696 ± 35
29	45 ± 3	39 ± 3	1309 ± 93	689 ± 35
30	35 ± 2	65 ± 4	1308 ± 64	383 ± 21
31	29 ± 2	35 ± 2	1162 ± 66	7 ± 1
32	26 ± 2	71 ± 6	1355 ± 61	362 ± 21
33	16 ± 2	28 ± 2	1604 ± 94	35 ± 7
34	91 ± 5	146 ± 8	1474 ± 87	1332 ± 69

The values of the absorbed dose rate were calculated by equation 1, using the activities concentrations values of A_{Ra}, A_{Th} and A_K from Table 1.

Figure 4 shows the results of the Annual Effective Dose assessment, considering a standard room, due to the external exposure to gamma radiation as described by equation 2 and the internal exposure by the inhalation of radon as described by equation 3.

Figure 4: *Increment of Annual Effective Dose caused by the internal coating of a standard room due to external gamma radiation and inhalation of radon.*
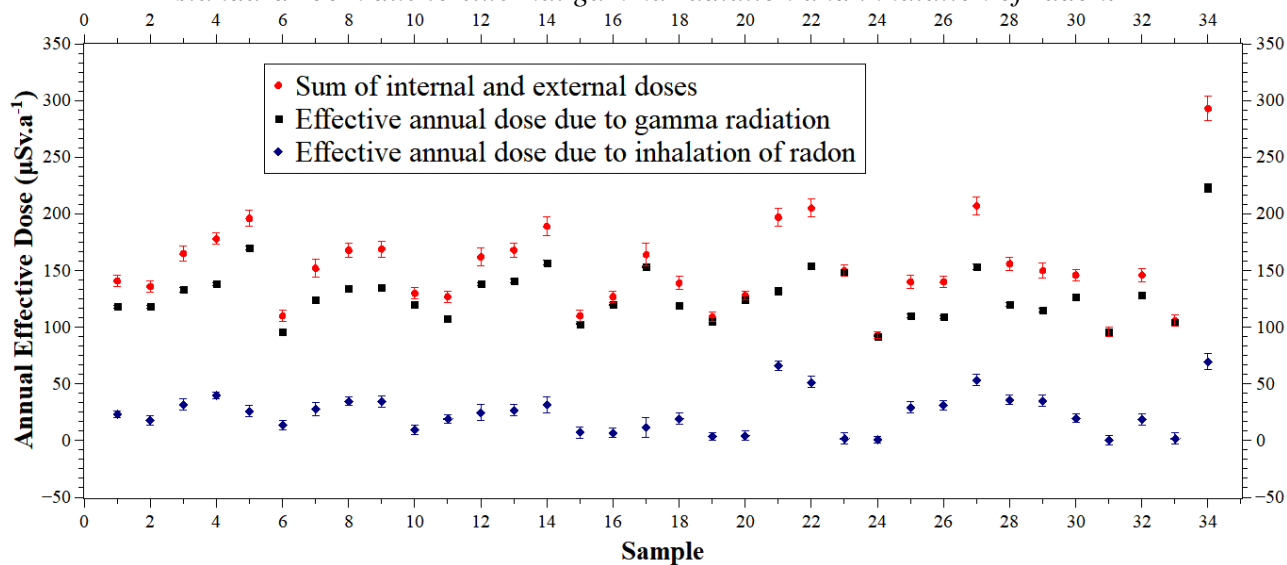
Source: Adapted from [2]

The results showed the total increment of the Annual Effective Dose in a standard room, where all the walls were coated with the studied rocks. The estimated values for the contribution due to radon inhalation ranged from (0.4 ± 0.04) µSv.a^{-1} to (70 ± 4) µSv.a^{-1} and the contribution from gamma external dose ranged from 96 ± 4 µSv.a^{-1} to 223 ± 7 µSv.a^{-1}.

These value ranges are in accordance with literature values [13-17]. In a similar study, Shweikani and Raja assessed the contribution of a combination of marbles and ceramics as finishing materials (coating in a standard room) obtained maximum values of 20 µSv.a^{-1} and 35 µSv.a^{-1} for internal and external doses respectively. [18]

The behavior of the internal and external doses are similar (Figure 4). The sum of these two contributions ranged from 93 ± 3 µSv.a^{-1} to 293 ± 11 µSv.a^{-1}. All values are below the European Commission on Radiation Protection Unit suggested limit of 1 mSv.a^{-1} for the general public [9].

However, the results represent the estimation of increment of the dose due only to the coating materials, without taking into account all other structural materials and other possible sources.

The distribution of the data in Figure 4 shows a good correlation between the external and internal dose, which is expected, since the ^{226}Ra that contributes to the external dose is also the main source of ^{222}Rn (internal dose).

4. CONCLUSION

Comparing the values of the Annual Effective Dose due to granite rocks as inner surface coating, it can be observed that, in most samples, the contribution due to external gamma radiation is greater than the contribution from inhalation of radon.

Regarding the considered scenarios, none of the studied rocks presented radiological risks for the general public, so the granitic rocks of geological occurrence in the Parana State, can be safely used in civil construction, as structural and or coating materials. Also, as these scenarios were defined assuming specific conditions of internal walls coating and whole year exposure, the radiological risks were overestimated, favoring the radiological safety.

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