

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 an HPGe detector for evaluation of²²⁶Ra, ²³²Th and ⁴⁰K 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 granites of thickness 2 cm and considering an annual exposure time of 7000 h. This value was 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 ²²²Rn, determined with CR-39 solid state nuclear track detectors using the sealed can technique, considering a ventilation rate of 0.5 h⁻¹ and an annual exposure time of 7000 h. The results of this study showed that the increase in the annual effective dose ranged from $0.39 \pm 0.04 \ \mu Sv.a^{-1}$ to $70 \pm 4 \ \mu Sv.a^{-1}$ from radon inhalation. All results stayed below 1 mSv.a⁻¹ recommended by the European Commission on Radiological Protection.

1. INTRODUCTION

The natural radionuclides present in building materials can promote people exposure to radiation (homes, schools, shops). This increase may be mainly to: external gamma dose and the 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 ²³⁸U, ²³⁵U and ²³²Th series and the ⁴⁰K isolated radionuclide represent 16.97% of the mean annual effective dose [1].

The internal dose results mainly from inhalation of the radon isotope, ²²²Rn, which is a noble gas caused by the emission of alpha particles during the radioactive disintegration of ²²⁶Ra, a decay product of ²³⁸U series. Inhalation of radon represents 47.6% of the mean annual effective dose due to natural radionuclides [1].

Rocks with high radioactivity can be an important dose source when being used as building materials (structural and 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 are no studies on radiological emission. In this way, the objective of this work is to do this evaluation [2, 3].

2. MATERIALS AND METHODS

2.1. Sample area and Sample 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.



Figure 1: Maps of the study area in: A. Brazil; B. Paraná; and C. Curitiba.

Samples were obtained in the form of plates measuring 15 cm x 15 cm x 2 cm (width, length and thickness, respectively). For the analysis by gamma spectrometry, all samples were pulverized and sealed in standard 100-mL HDPE flat-bottom cylindrical flask with screw cap and bubble spigot and measured in triplicate, after a 4 weeks in growth period for radioactive equilibrium in the ²³⁸U and ²³²Th series. The ²²⁶Ra, ²³²Th and ⁴⁰K activities concentrations were measured by high-resolution gamma-ray spectrometry with a coaxial high-purity germanium detectors (HPGe) with conventional electronics and an a 919 ORTEC EG&G Spectrum Master 4k multichannel analyzer. All spectra were acquired the by software Maestro 3.04 and analyzed with the InterWinner 6.0 [4].

2.2. Annual Effective Dose Due To External Gamma-Rays

To calculate the Annual Effective Dose it is necessary to first calculate the absorbed dose rate. Due to construction materials this rate (indoor) is calculated using dosimetric models that are based on concentrations of gamma activity from the samples and their form and amount of application.

In this study we addopted the model suggested by the European Commission of Radiological Protection [5]. The dose rate absorbed in the air (within a standard model room) was derived from superficial materials (thickness 3 cm density 2.6 g.cm⁻³, disregarding doors and windows). For this configuration of coating rocks the dose rate is given by the equation,

$$\dot{D} = 0.12 A_{Ra} + 0.14 A_{Tb} + 0.0096 A_{k} \tag{1}$$

In this equation \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 ²²⁶Ra, ²³²Th e ⁴⁰K in Bq.kg⁻¹, respectively.

The coefficients of activity in dose conversion, shown in equation 1, were calculated by the mathematical model described by [6]. This model allows the evaluation of several parameters (model room dimensions, wall thickness and material density), thus providing some scenarios. Among these, the one that best applies to the present work corresponds to rocks used for internal coating (equation 1).

The Annual Effective Dose $D_{ef(gamma)}$, considering an occupancy time of 7000 h in the standard room, was calculated by the equation 2.

$$D_{ef(aama)} = \dot{D}(nGy.h^{-1}) x \, 0.7 \, (Sv.Gy^{-1}) x \, 7000(h)$$
⁽²⁾

Where \dot{D} is the dose rate, 0.7 is the dose rate conversion factor for Annual Effective Dose and 7000 h is the exposure time considered in the model [5].

2.3. Annual Effective Dose Due To Radon Inhalation

In this work, the determination of radon and its descendants was performed by the passive detection technique using SSNTD detectors type CR-39, using the Sealed can technique [7], which basically consists of SSNTD detectors fixed internally on top of a chamber (usually cylindrical) sealed.

This technique was performed as an alternative to reproduce the exposure scenarios as the standard room, internally coated with each sample studied. Thus, the alternative was to use the Sealed can technique, which in addition to providing the radon exhalation rate measured in each sample is used as a parameter in dosimetric models to estimate the doses.

For such technique, a cylindrical vessel with dimensions of 26.5 cm x 23.5 cm (height and diameter, respectively) was used where the SSNTD detector, CR-39, was placed inside a diffusion chamber model NRPB/SSI which is attached to the top of the container [7]. The natural sample (a cut of granite slab with dimensions: 15 cm x 15 cm x 2 cm; length, width and thickness, respectively) was placed in the base of the container (Figure 2).



Figure 2: Diagram illustrating the Sealed can technique.

This can was sealed for 30 days, chosen time for integration of traits seeking an optimization between counting statistics and measurement time. After the counting time, the sealed chamber was opened, the diffusion chambers were removed and the CR-39 detectors were subjected to standard chemical etching at the Nuclear Track Detector Laboratory (LRA/GMR) with a 30% KOH solution at 80°C for 5.5 hours in a shaking water bath [7]. The next step was to read the samples in the optical microscope. Therefore, the calculation of radon concentration was determined from the manual counting of the tracks, using ZEISS software "KS100 version 3.0" [8].

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.

$$D_{ef(Rn)} = C_{Rn} \times 20 \frac{\mu S v}{Bq \cdot m^{-3}}$$
(3)

Where, 20 is the derived conversion factor considering an annual exposure time of 7000 h (7000 h per year within the standard room and equilibrium factor of 0.5) [2].

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

Figure 3 shows the results of the estimation of the Annual Effective Dose in 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. The internal annual effective dose was calculated for a room model with the same dimensions as the model used to calculate the external annual effective dose and considering the same annual exposure time (7000 h).



Figure 3: Increment of Annual Effective Dose caused by the internal coating of a standard room due to external gamma radiation and inhalation of radon.

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 $(0.39 \pm 0.04) \mu Sv.a^{-1}$ to $(70 \pm 4) \mu Sv.a^{-1}$ and the contribution from gamma external dose ranged from $62 \pm 3 \mu Sv.a^{-1}$ to $138 \pm 5 \mu Sv.a^{-1}$.

These value ranges are in accordance with the international literature [9, 10, 11, 12, 13]. In a similar study, evaluating the contribution of a combination of marbles and ceramics as finishing materials (coating in a standard room) Shweikani e Raja [14] obtained maximum values of 20 μ Sv and 35 μ Sv for internal and external doses respectively.

The values behavior values obtained for internal and external doses are similar (Figure 3). The sum of these two contributions ranged from $63 \pm 1 \,\mu\text{Sv}$ to $208 \pm 1 \,\mu\text{Sv}$. These values are below the maximum limit allowed for the public, that is 1 mSv, remembering that this is the estimation of increment of the dose due to only the coating materials, without taking into account all other structural materials and other possible sources.

It is also possible to verify a good correlation between external dose and internal dose, which is expected, since the ²²⁶Ra that contributes to the external dose is also the main source of ²²²Rn (internal dose). This correlation is not higher because pretrographic properties of rocks influence radon exhaustion rate.

4. CONCLUSIONS

Considering the scenarios that were described in this study, no studied rock presented radiological risks for the public. However, as these scenarios were defined assuming conditions (internal coating on walls and yearly exposure) that may not reflect the current scenario, overestimating values going towards radiological safety.

Comparing the values obtained to increase the Annual Effective Dose due to rocks application with the inner surface coating of the model, it has been found that most of the time, the contribution due to external gamma radiation is greater than the contribution from inhalation of radon.

A good correlation can be verified between the 226 Ra concentration and the 222 Rn concentration.

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REFERENCES

- 1. Unscear The 2000 "Report to the general assembly with scientic annexes". Technical Report, United Scientic Committee on the Eects of Atomic Radiation, New York, (2000).
- Ferreira, A. O., "Avaliação da radioatividade natural em algumas rochas graníticas do estado do Paraná e sua utilização na construção civil". Tese (Doutorado), Instituto de Pesquisas Energéticas e Nucleares Autarquia associada à Universidade de São Paulo, (2013).
- 3. Ferreira, A.O.; Pecequilo, B.R.S., "Natural radioactivity assessment by gamma spectrometry in some commercially-used granites from Paraná State, Brazil: Preliminary results". *Radioprotection* (Paris. 1966), v. 46, p. S43-S47, 2011.
- 4. InterwinnerTm 6.0 MCA Emulation, Data Acquisition and Analysis software for Gamma and Alpha Spectroscopy IW-B32 2004. ORTEC. Oak Ridge, TN, USA. (2004).
- 5. EC "Radiological protection principles concerning the natural radioactivity of building materials, radiation protection report RP-112". Technical report, European Commission. Radiation Protection Unit, Luxembourg, (1999).
- 6. Markkanen, M. "Radiation Dose Assessments for Materials with Elevated Natural Radioactivity". Report STUK-B-STO 32,. Technical Report, Radiation and Nuclear Safety Authority, STUK, (1995).
- 7. Ferreira, A.O.; Pecequilo, B.R.S.; Aquino, R.R. . "Application of a Sealed Can Technique and CR-39 detectors for measuring radon emanation from undamaged granitic ornamental building materials". *Radioprotection* (Paris. 1966), v. 46, p. S49-S54, 2011.
- 8. ZEISS Ks 100 imaging system release 3.0, (1997).

- 9. Marocchi, M., Righi, S., Bargossi, G. M., Gasparotto, G., "Natural radionuclides content and radiological hazard of commercial ornamental stones: An integrated radiometric and mineralogical-petrographic study". *Radiation Measurements*, v 46,: 538-545, (2011).
- 10. Turhan, S., "Estimation of possible radiological hazards from natural radioactivity in commercially-utilized ornamental and countertops granite tiles", *Annals of Nuclear Energy*, v 44, 34-39, (2012).
- 11. Asaduzzaman, K., Khandaker, M. U., Amin, Y. M., and Bradley, D. A., "Natural radioactivity levels and radiological assessment of decorative building materials in Bangladesh", *Indoor and Built Environment*, v 25: 541-550, (2014).
- 12. Al-Zahrani, J. H., "Estimation of natural radioactivity in local and imported polished granite used as building materials in Saudi Arabia", *Journal of Radiation Research and Applied Sciences*, v 10:3,241-245, (2017).
- 13. Salas, H.T., Nalini, H.A. and Mendes, J.C., "Radioactivity dosage evaluation of Brazilian ornamental granitic rocks based on chemical data, with mineralogical and lithological characterization", *Environ Geol*, **v** 49: 520-526. (2006).
- 14. Shweikani, R. and Raja, G. "Radon exhalation from some finishing materials frequently used in Syria". *Radiation Measurements*, v. 44: 1019-1023, (2009).