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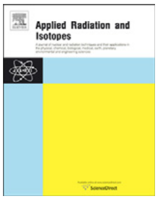

Activity measurements of radon from construction materials ☆

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ABSTRACT

This work presents the results of radon concentration measurements of construction materials used in the Brazilian industry, such as clay (red) bricks and concrete blocks. The measurements focused on the detection of indoor radon activity during different construction stages and the analysis of radionuclides present in the construction materials. For this purpose, sealed chambers with internal dimensions of approximately $60 \times 60 \times 60\ \text{cm}^3$ were built within a protected and isolated laboratory environment, and stable air humidity and temperature levels were maintained. These chambers were also used for radon emanation reduction tests. The chambers were built in four major stages: (1) assembly of the walls using clay (red) bricks, concrete blocks, and mortar; (2) installation of plaster; (3) finishing of wall surface using lime; and (4) insulation of wall surface and finishing using paint. Radon measurements were performed using polycarbonate etched track detectors. By comparing the three layers applied to the masonry walls, it was concluded that only the last step (wall painting using acrylic varnish) reduced the radon emanation, by a factor of approximately 2. Samples of the construction materials (clay bricks and concrete blocks) were ground, homogenized, and subjected to gamma-ray spectrometry analysis to evaluate the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K. The values for the index of the activity concentration (I), radium equivalent activity (Ra_{eq}), and external hazard index (H_{ext}) showed that these construction materials could be used without restrictions or concern about the equivalent dose limit (1 mSv/year).

1. Introduction

Numerous and systematic studies that have been performed in different countries over many decades have resulted in the explicit conclusion that exposure to radon and its progeny is the main cause of lung cancer among non-smokers (NRC, 1988). All three natural isotopes of radon (^{222}Rn, ^{220}Rn and ^{219}Rn) are produced from three natural radioactive decay chains. Specifically, ^{222}Rn is produced by the decay series of ^{238}U and proceeds from the α-decay of ^{226}Ra.

It is well known that more than 50% of the annual effective radiation dose received by a human being is related to the inhalation of radon. Among the main sources of the radon in the air of a dwelling are its diffusion and emanation from soil and building materials, as well as its release from water (ICRP, 1991; UNSCEAR, 1993).

This paper presents the results of correlation studies of the radon concentrations in the air and the construction materials used in the Brazilian industry, such as clay (red) bricks and concrete blocks. The measurements focused on the indoor radon activity detected during different construction stages, and the radionuclides present in the construction materials used in these stages were analyzed. Some preliminary measurement results were reported during the International Nuclear Atlantic Conferences (INAC) in 2007 and 2009 (Fior et al., 2007; Paschuk et al., 2009; Corrêa et al., 2009).

2. Materials and methods

The measurements of the isotope ^{222}Rn were performed within artificial test chambers with internal dimensions of approximately $60 \times 60 \times 60\ \text{cm}^3$. These chambers were built within a protected and isolated environment to maintain stable air humidity and temperature levels. For the radon emanation reduction tests, the chambers were built in four major stages: (1) assembly of the walls using clay (red) bricks, concrete blocks, and mortar; (2) installation of plaster; (3) finishing of wall surface using lime; and (4) insulation of wall

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surface and finishing using paint. The bottom (the floor) and upper (the ceiling) part of each chamber were assembled from Medium-density fiberboards (MDFs) covered (painted) with synthetic resin to prevent possible external humidity and radon entrance into the chamber volume during the measurements (see left photograph of Fig. 1, Fior et al., 2007). The upper part (the cap, the ceiling) was prepared to be removable to give access to the internal volume of the chamber during plaster and finishing materials installation stages. To diminish the moisture content influence at radon emanation from the walls, the test chambers were dried and ventilated after each step of their assembly during approximately two weeks after it was concluded. After that the test chambers were sealed (the caps—the ceiling plates were installed) with diffusion chambers inside for radon measurements. The airtightness of the internal volume was guaranteed by using rubber strips glued at the upper edge of the chamber walls (see right photograph of Fig. 1) and installing some heavy blocks at the ceiling plates. During the second assembly stage each wall was coated with 5 mm layer of cement plaster whose composition is basically a mixture of the Portland cement, sand, lime and water. For this purpose it was used some commercial bagged mixture, typical at the South region of Brazil. The third assembly stage consisted of lime plaster coating when each wall received about 1–2 mm layer of stucco. For this purpose it was used some commercial mixture as well. At the last fourth stage, the internal surface of each test chamber was painted with synthetic polyvinyl acetate latex. As previously, for this purpose it was used some commercial paint typical at the South of Brazil.

In each series of radon activity measurements, 56 polycarbonate etched track detectors (LEXAN, GE) mounted in diffusion cells were used. Of these, six were installed in the internal volume of each chamber (a total of 8 duplicate chambers), for a total of 48 internal detectors. Eight more external detectors were used to measure the natural radiation background of the laboratory environment where the chambers were built.

An exposure time of about 11 days was established based on a previous calibration performed at the Institute of Radiation Protection and Dosimetry (IRD/CNEN) (Melo, 1999), where an efficiency of 70% was obtained for the density of alpha particle tracks of about 13.8 cm^{-2} per exposure day and per 1 kBq/m^{3} of radon activity. The LEXAN etched track detector was chosen for the radon measurements because its 100 μm protection layer creates a Bragg peak of 5.5 MeV α-particles just at the surface of the LEXAN film, which simplifies the chemical track development process. Figs. 2 and 3 show an overview of the diffusion cells, along with the aerosol filter and LEXAN film. The chemical development of the alpha tracks was accomplished using electrochemical etching (see Table 1, Corrêa et al., 2007; Corrêa, 2006). The exposed LEXAN detectors were chemically pre-etched (1000 V at 100 Hz) for 40 min, after which they were etched (800 V at 3000 Hz) at 30 °C in a solution of 6 N KOH + 80% of C_2H_5OH for 3 h. These pre-etching and etching times were chosen to obtain an alpha particle track size of about 100 μm, which could be easily identified and visualized by computer scanning with a 1200 dpi resolution. The obtained images (JPEG) of developed LEXAN detectors, which normally had 24-bit color, were manually counted at the computer screen by a trained person.

Fig. 1. Different views of test chambers built from concrete blocks and clay (red) bricks: 1—removable MDF cap, 2—bottom MDF fiberboard, 3—rubber strips used for airtightness of the internal volume of test chamber. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

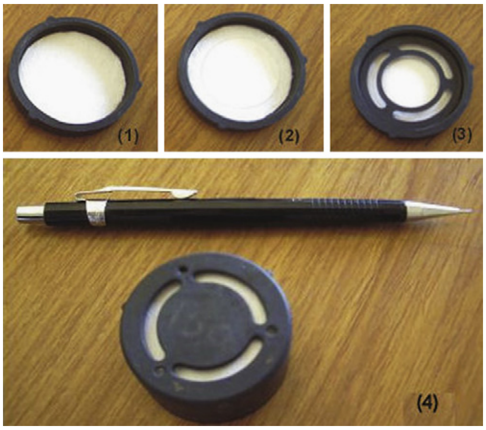

Fig. 2. Detection system: (1) cover and filter; (2) cover, filter, and detector; (3) cover, filter, detector, and ring-support; (4) diffusion chamber ready for exposure.

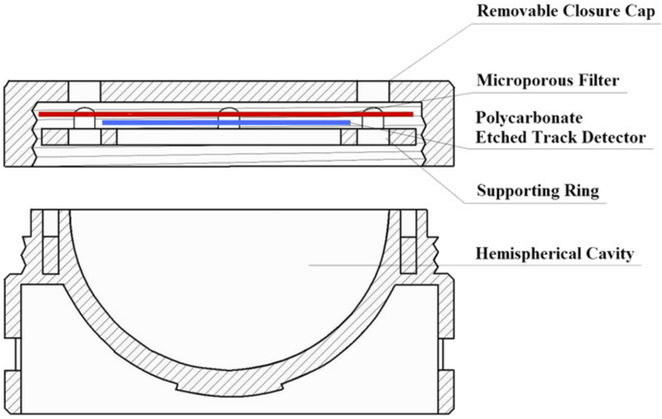

Fig. 3. Schematic drawing of Karlsruhe radon diffusion chamber used in present measurements (Fior et al., 2007).

Table 1
Details of electrochemical development of SSNTD.

Material	LEXAN, polycarbonate
Manufacture	General Electric Co.
Thickness	250 μm
Filter	Fiberglass
Chemical pre-etching	
Time	3 h
Solution	PEW solution
Temperature	27 °C
Electrochemical etching	
Time	(1st)1 h, (2nd) 2 h
Solution	PEW solution
Voltage	(1st) 1000 V, (2nd) 800 V
Frequency	(1st) 100 Hz, (2nd) 3000 Hz
Sensibility	0.58 Tr cm^{-2}/Bq h m^{-3}

After the equilibrium between the radioactive series of ^{238}U and ^{232}Th was achieved, three samples of each construction material (red clay bricks and concrete blocks) were ground, homogenized, and subjected to gamma-ray spectrometry measurements using a

Table 2
Gamma transitions used in gamma spectrometry analysis.

Element	Gamma transition (keV)
^{214}Pb	295.21 and 351.92
^{214}Bi	609.3
^{212}Pb	238.6 and 300.1
^{212}Bi	727.3
^{228}Ac	911.1 and 968.9
^{40}K	1460.7

high-resolution EG&G ORTEC detector with conventional electronics from EG&G ORTEC, along with an analytical multichannel EG&G ORTEC Spectrum Master 919. The detector had a relative efficiency of 15% and its resolution for 1332 keV (^{60}Co) was 2.7 keV. The gamma-ray spectrometry analysis was performed using the commercial software package WinnerGamma (ORTEC InterWinner Spectroscopy Program Family, version 4.1). Table 2 shows the gamma transitions of ^{214}Pb, ^{214}Bi, ^{212}Pb, ^{212}Bi, ^{228}Ac, and ^{40}K, which were used in the gamma spectrometry analysis for determining the activity of ^{226}Ra, ^{232}Th, and ^{40}K in the studied building material samples.

Once the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in the material samples were measured, the radium equivalent activity, Ra$_{eq}$, was evaluated following the recommendations of OECD (1979). It should be mentioned that this parameter is commonly used to evaluate the radiological risk of building materials. For this purpose, the following equation was used (Beretka and Matthew, 1985; Malanca et al., 1993):

$$Ra_{eq}=C_{^{226}Ra}+(1.43\cdot C_{^{232}Th})+(0.077\cdot C_{^{40}K}),$$

where $C_{^{226}Ra}$, $C_{^{232}Th}$, and $C_{^{40}K}$ are the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K in units of Bq/kg, respectively. According to this equation, 1 Bq/kg of ^{226}Ra, 0.7 Bq/kg of ^{232}Th and 13 Bq/kg of ^{40}K provide the same γ-ray dose (OECD, 1979; Malanca et al., 1993).

Following the recommendations of the European Commission on radiological protection principles concerning the natural radioactivity of building materials (European Commission, 1999) and assuming that the permissible equivalent dose limit is equal to 1 mSv/year (ICRP, 1990), the results obtained for the activity of ^{232}Th, ^{226}Ra, and ^{40}K were converted into activity concentration indices (I) (Beretka and Matthew, 1985)

$$I=(C_{^{232}Th}/200)+(C_{^{226}Ra}/300)+(C_{^{40}K}/3000),$$

where $C_{^{226}Ra}$, $C_{^{232}Th}$, and $C_{^{40}K}$ are the activity concentrations of ^{232}Th, ^{226}Ra and ^{40}K in Bq/kg, respectively. The evaluation of the activity concentration index is used to guarantee that its value does not exceed 1, indicating that the annual effective dose for a person occupying a dwelling built from materials with a certain specific radioactivity is less than 1 mSv.

Another way to evaluate the safety of construction materials is the calculation of the external hazard index (H_{ext}) and the internal hazard index (H_{in}), which are defined (Beretka and Matthew, 1985; Xinwei, 2004) as follows:

$$H_{ext}=(C_{^{226}Ra}/370)+(C_{^{232}Th}/260)+(C_{^{40}K}/4810)\leq 1$$

and

$$H_{in}=(C_{^{226}Ra}/185)+(C_{^{232}Th}/260)+(C_{^{40}K}/4810)\leq 1.$$

The internal hazard index (H_{in}) is usually used to evaluate the internal exposure to radon and its progeny and to reduce the acceptable maximum concentration of ^{226}Ra to half the normal limit. It should be noted that for the building materials to be used in dwelling construction, both indices (H_{ext} and H_{in}) should be less than 1 to limit the radiation dose to the permissible amount of 1 mSv/year and ensure that the effects of radon and its short-lived progeny on the respiratory organs of individuals occupying the dwelling will be negligible.

3. Results and conclusions

The measured radon activity concentrations inside the chambers were found to be compatible with the maximum recommended by ICRP 60 (1990) of 600 Bq/m^3 in construction stages (1), (2), and (4): (615 ± 65), (690 ± 74) and (399 ± 51) Bq/m^3 for clay (red) bricks and (594 ± 135), (356 ± 97) and (354 ± 121) Bq/m^3 for concrete blocks, respectively. The results obtained in construction stage (3) for both materials were found to be higher than the recommended value: (1031 ± 137) Bq/m^3 and (874 ± 120) Bq/m^3 for clay (red) bricks and concrete blocks, respectively.

Clearly, the results obtained for the ^{222}Rn concentration in air during the four construction stages were related to the rather small volume of air inside the chamber (0.21 ± 0.01) m^3, as well as the small area of the walls. In other words, considering a cubic chamber of side length h, it can be concluded that the radon activity in air is related to the internal volume of the chamber according to $1/h^3$. At the same time, the total amount of radon in air is proportional to the surface area of the walls ($\sim h^2$) because of the emanation process. This means that, as an approximation, the radon activity in air within used building materials is proportional to the size of the chamber according to $1/h$.

Following this, if we extrapolate the results for the internal volume of a chamber 0.60 m in size to a reasonable room size (3.0 m) within a dwelling built with the same concrete blocks or clay bricks, we should have a decreased radon concentration of about 3.0/0.6=5. In this case, at all construction stages, the concentration of ^{222}Rn inside the room of the dwelling will be below 200 Bq/m^3 (indoor level of action).

Considering that the difference between the obtained experimental results for clay (red) bricks and concrete blocks is not bigger than three standard deviations, it is possible to consider both types of data simultaneously, calculating the average radon activity for both materials taking into account the experimental errors. The results are presented in Fig. 4.

It was observed that during the first two building stages, the radon activity inside the chambers remained almost stable. The radon concentration almost doubled after the lime layer was installed. This fact requires special attention and further

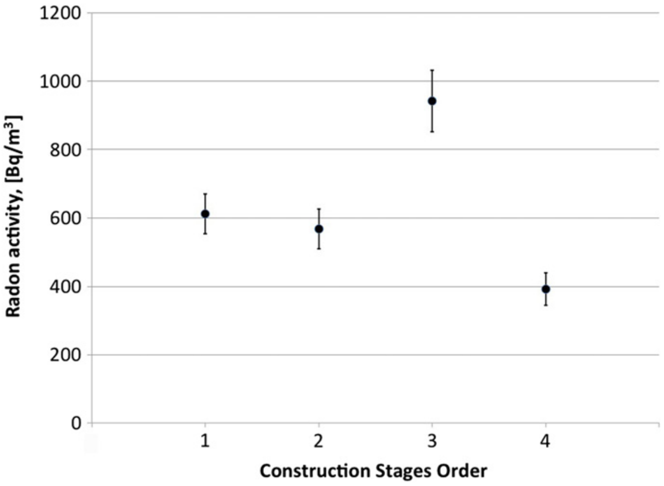

Fig. 4. Average radon activity in air within internal volume of chamber in four main stages (see text).

Table 3
Results of gamma-ray spectrometry analysis for red clay bricks and concrete blocks.

Measured and evaluated activity parameters	Clay (red) bricks	Concrete blocks
Activity concentration (Bq/kg)		
^{226}Ra	38.9 ± 1.7	21.1 ± 0.9
^{232}Th	46.1 ± 1.8	19.7 ± 0.9
^{40}K	188 ± 12	737 ± 44
Index of activity concentration, I	0.423 ± 0.019	0.414 ± 0.022
Radium equivalent activity, Ra_{eq}, (Bq/kg)	119.3 ± 5.2	106.1 ± 5.6
External hazard index, H_{ext}	0.321 ± 0.014	0.286 ± 0.015

measurements because this finishing material is usually considered to be a sealant in radon mitigation techniques. One possible explanation is that the brand of lime used was mixed with gypsum phosphate, which could be contaminated by radioactive decay products of the uranium series, such as ^{226}Ra. Comparing the radon reduction between the three layers applied to the walls, one can see that only the wall painted with acrylic varnish reduced the radon emanation flow by a factor of approximately 2.

The results of the gamma spectrometry analysis of the activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K; the index of activity concentration (I); radium equivalent activity (Ra_{eq}); and external hazard index (H_{ext}) for the studied clay bricks and concrete blocks are presented in Table 3.

The measured levels of activity concentration for ^{226}Ra, ^{232}Th, and ^{40}K, with non-uniform distribution for the studied materials, are in good agreement with the experimental data from other researches (Xinwei, 2004; Kumar et al., 2003; Fathivand et al., 2007; Somlai et al., 1998), where these values vary from 12 to 109 Bq/kg for ^{226}Ra, from 41 to 89 Bq/kg for ^{232}Th, and from 24 to 1200 Bq/kg for ^{40}K in the case of similar building materials.

It should be noted that the values obtained for the activity concentration index, I, and the external hazard index, H_{ext}, are inferior to 1. This means that the studied construction materials (clay (red) bricks and concrete blocks) can be used without restrictions or any concern about the equivalent dose limit (1 mSv/year) that might be received by people inside a dwelling built with such materials. The same conclusion can be made considering the results for Ra_{eq}, which were below the indoor reference limit (370 Bq/kg) established by international norms and the recommendations of other publications (OECD, 1979; Beretka and Matthew, 1985; Malanca et al., 1993; UNSCEAR, 1982; Venturini and Nisti, 1997).

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References

Beretka, J., Matthew, P.J., 1985. Health Phys. 48, 87–95.
Corrêa, J.N., 2006. Avaliação de Concentração de Radônio em Ambientes de Convívio Humano na Região Metropolitana de Curitiba (in Portuguese). UTFPR, Master Degree Thesis.
Corrêa, J.N., et al., 2007. AIP Conf. Proc. 884, 501–503.
Corrêa, J.N., et al., 2009. In: Proceedings of Fourth International Nuclear Atlantic Conference—INAC 2009 (CD-ROM). Brazilian Association for Nuclear Energy, ABEN, Rio de Janeiro.
European Commission, 1999. Radiological Protection Principles Concerning the Natural Radioactivity of Building Materials, Radiation Protection Report RP-112, EC, European Commission, Luxembourg.
Fathivand, A.A., Amidi, J., Najafi, A., 2007. Radiat. Prot. Dosim. 123, 391–393.
Fior, L., et al., 2007. In: Proceedings of International Nuclear Atlantic Conference, INAC 2007 (CD-ROM). Brazilian Association for Nuclear Energy, ABEN, Rio de Janeiro, RJ.
ICRP 60, 1990. Recommendations of the International Commission on Radiological Protection. Annals of the ICRP, vol. 21, 1–3. Pergamon Press, Oxford.
ICRP 60, 1991. International Commission on Radiological Protection. Recommendations of the International Commission on Radiological Protection. Pergamon Press, Oxford.
Kumar, A., Kumar, M., Sengupta, D., Prasad, R., 2003. Radiat. Meas. 36, 465–469.
Malanca, A., Passina, V., Dallara, G., 1993. Radiat. Prot. Dosim. 48, 199–203.
Melo, V.P., 1999. Measures of the Concentration of Radon in the Alegre Mount Region—PA (in Portuguese). UFRJ, Master Degree Thesis.
NRC, 1988. National Research Council, Health Risks of Radon and other Internally Deposited Alpha-Emitters: BEIR IV. National Academy Press, Washington, DC.
OECD, 1979. Exposure to radiation from natural radioactivity in building materials. Report by a Group of Experts of the OECD Nuclear Energy Agency, Paris, France.
Paschuk, S.A., et al., 2009. In: Proceedings of Fourth International Nuclear Atlantic Conference—INAC 2009 (CD-ROM). Brazilian Association for Nuclear Energy, ABEN, Rio de Janeiro.
Somlai, J., et al., 1998. Health Phys. 75, 648–651.
UNSCEAR, 1982. Ionising radiation sources and biological effects. Report to the United Nations General Assembly, with Scientific Annexes. United Nations, New York.
UNSCEAR, 1993. Sources and effects of ionizing radiation. Report to the General Assembly, with Scientific Annexes. United Nations, New York.
Venturini, L., Nisti, M.B., 1997. Radiat. Prot. Dosim. 71, 227–229.
Xinwei, L., 2004. J. Radioanal. Nucl. Chem. 262, 775–777.