2015 International Nuclear Atlantic Conference - INAC 2015
São Paulo, SP, Brazil, October 4-9, 2015
ASSOCIAÇÃO BRASILEIRA DE ENERGIA NUCLEAR - ABEN
ISBN: **978-85-99141-06-9**

EVALUATION OF Ra-226, Th-232 AND K-40 ACTIVITIES CONCENTRATIONS AND RADIUM EQUIVALENT INDEX IN SEVERAL BRAZILIAN ECONOMIC WALL PAINTS

Leandro M. Fonseca and Brigitte R. S. Pecequilo

Instituto de Pesquisas Energéticas e Nucleares (IPEN / CNEN - SP)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
lmfonseca@ipen.br
brigitte@ipen.br

ABSTRACT

The titanium dioxide used as the white pigment in paints is produced from the processing of ilmenite minerals. As monazite, the main ilmenite radioactive contaminant, contains 1 to 20% thorium dioxide and also some uranium traces, so, eventually, wall paints can contain radioactivity. Activity concentrations of the naturally occurring radionuclides ^{226}Ra, ^{232}Th and ^{40}K were determined in 15 brazilian economic wall paints samples, by high resolution gamma-ray spectrometry. The activities concentrations in the studied samples ranged from 1.3 ± 0.2 Bq/kg to 23.4 ± 0.7 Bq/kg for ^{226}Ra; from 2.5 ± 0.4 Bq/kg to 45.8 ± 1.5 Bq/kg for ^{232}Th and from 5.8 ± 2.1 Bq/kg to 157 ± 22 Bq/kg for ^{40}K. The radium equivalent index, calculated from the ^{226}Ra, ^{232}Th and ^{40}K concentrations, varied from 1.30 Bq/kg up to 95.9 Bq/kg, below the value of 370 Bq/kg recommended by OECD for a safety use in residential building applications.

1. INTRODUCTION

Naturally occurring radionuclides with half-lives of the same order that the age of the earth, like ^{40}K and the radionuclides from the ^{238}U and ^{232}Th series, are the major source of exposure for mankind [1,2]. Buildings and houses are very important in human life, as quite a lot of the lifetime is spent at home and/or office. Virtually, every building material contains the natural radionuclides ^{226}Ra, ^{232}Th and ^{40}K, so, knowledge of the levels of natural radioactivity in building materials is important for the assessment of population exposure to natural radioactivity.

Several studies have been conducted to evaluate natural radioactivity in building materials such rocks, granites, marbles, sand, etc. [3-5], however, to date, there are no studies concerning the natural radioactivity that eventually the wall paints used internal or externally in buildings and houses used could contain.

The basic raw materials for the production of almost all kinds of paints are made up of resins, pigments, solvents and additives. The pigments give the paint color, solvents make it easier to apply, resins help it dry and additives serve as everything from fillers to antifungal agents. The basic pigment is titanium dioxide, providing the whiteness and opacity of paints. Titanium dioxide is a simple inorganic compound, processed from ilmenite minerals, representing 92% of the world demand of titanium minerals [6]. As monazite, the main ilmenite radioactive contaminant [7], contains 1 to 20% thorium dioxide and also some uranium traces, it is easy to assume that wall paints, as other construction materials, could contain natural radioactivity [8].

Several studies have been conducted to evaluate the occupational exposure of titanium production industry workers [9-11], but the natural radioactivity effect to the humans from the wall paints used in the buildings and houses remains still unknown.

The use of titanium dioxide in Brazil is divided between 85.5% for paints, 8.6% for steel industry, 6.4% for iron alloys, 1.6% for electrodes and 0.8% for floors and tiles [12].

Brazil is one of the world top five markets for coatings, manufacturing paints in a variety of applications. There are hundreds of large, medium and small manufacturers spread throughout the country. The top ten manufacturers account for 75% of total sales [13].

2. EXPERIMENTAL

Fifteen different brands of economic wall paints were kindly provided by SENAI (National Industrial Training Service) and ABRAFATI (Brazilian Coatings Manufacturers Association). Each sample was tightly sealed in a 100-mL HDPE flat-bottom cylindrical flask with screw cap and bubble spigot and stored for approximately 4 weeks, in order to ensure radioactive secular equilibrium [14].

All samples were measured during 150000 seconds with a coaxial extended range high-purity germanium detector (Canberra XtRa GX2520 detector), with conventional electronics and an EG&G ORTEC Spectrum Master 919 4-k multichannel analyzer. The spectra were analyzed with the WinnerGamma software [15]. 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 as the samples.

The activity of ^{40}K was calculated through its single gamma transition of 1461 keV. The activity of ^{226}Ra was determined by the weighted mean of the gamma transitions of ^{214}Pb (295.2 keV and 351.9 keV) and ^{214}Bi (609.3 keV), as seen in Eq. (1), and the activity of ^{232}Th by the weighted mean of the gamma-ray transitions of ^{212}Pb (238.6 keV and 300.1 keV), ^{212}Bi (727.3 keV) and ^{228}Ac (338.4 keV, 911.1 keV and 968.9 keV), as seen in Eq. (2). All activities concentrations were determined without self-attenuation corrections [16].

$$C_{^{226}Ra} = \frac{\dfrac{C_{^{214}Pb}}{\sigma^2_{^{214}Pb}} + \dfrac{C_{^{214}Bi}}{\sigma^2_{^{214}Bi}}}{\dfrac{1}{\sigma^2_{^{214}Pb}} + \dfrac{1}{\sigma^2_{^{214}Bi}}} \quad \textbf{(1)}$$

Where

$C_{^{226}Ra}$: ^{226}Ra activity concentration (Bq/kg);

$C_{^{214}Pb}$: ^{214}Pb activity concentration (Bq/kg);

$\sigma_{^{214}Pb}$: Uncertainty of ^{214}Pb activity concentration (Bq/kg);
$C_{^{214}Bi}$: ^{214}Bi activity concentration (Bq/kg);
$\sigma_{^{214}Bi}$: Uncertainty of ^{214}Bi activity concentration (Bq/kg).

$$C_{^{232}Th} = \frac{\dfrac{C_{^{212}Pb}}{\sigma^2_{^{212}Pb}} + \dfrac{C_{^{212}Bi}}{\sigma^2_{^{212}Bi}} + \dfrac{C_{^{228}Ac}}{\sigma^2_{^{228}Ac}}}{\dfrac{1}{\sigma^2_{^{212}Pb}} + \dfrac{1}{\sigma^2_{^{212}Bi}} + \dfrac{1}{\sigma^2_{^{228}Ac}}} \quad \textbf{(2)}$$

Where
$C_{^{232}Th}$: ^{232}Th activity concentration (Bq/kg);
$C_{^{212}Pb}$: ^{212}Pb activity concentration (Bq/kg);
$\sigma_{^{212}Pb}$: Uncertainty of ^{212}Pb activity concentration (Bq/kg);
$C_{^{212}Bi}$: ^{212}Bi activity concentration (Bq/kg);
$\sigma_{^{212}Bi}$: Uncertainty of ^{212}Bi activity concentration (Bq/kg);
$C_{^{228}Ac}$: ^{228}Ac activity concentration (Bq/kg);
$\sigma_{^{228}Ac}$: Uncertainty of ^{228}Ac activity concentration (Bq/kg).

The Radium Equivalent Index was calculated using the ^{226}Ra, ^{232}Th and ^{40}K activity concentrations, as seen in Eq. (3).

$$Ra_{eq} = 370\left(\frac{C_{^{226}Ra}}{370} + \frac{C_{^{232}Th}}{259} + \frac{C_{^{40}K}}{4810}\right) \quad \textbf{(3)}$$

3. RESULTS AND DISCUSSION

The activity concentration values of the considered natural radionuclides, for each wall paint brand are shown in Fig. 1 and the Radium equivalent index values are shown in Fig. 2.
As seen in Figure 2, the radium equivalent index for all samples is lower than the value of 370 Bq/kg recommended by OECD for a safety use in residential building applications [17].

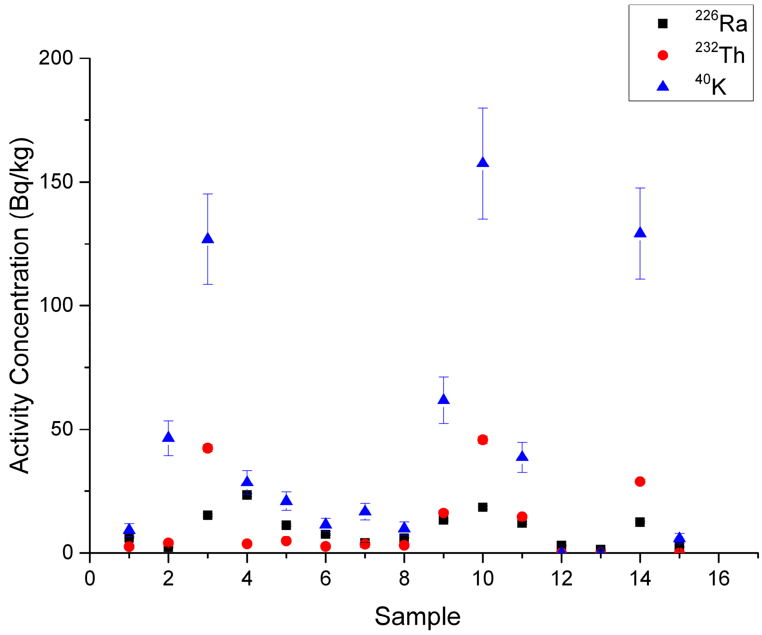

Figure 1: ^{226}Ra, ^{232}Th and ^{40}K activity concentration values assessed for 15 brands of brazilian economic wall paints.

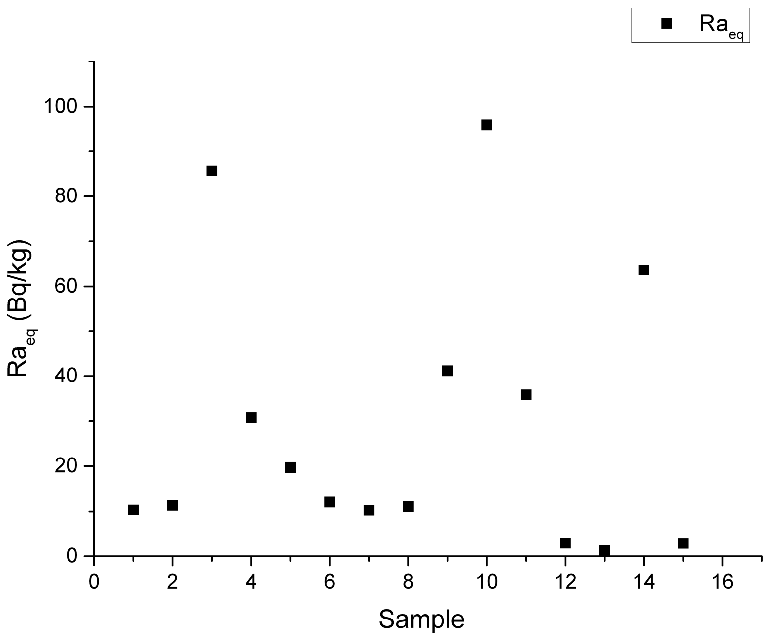

Figure 2: Radium equivalent index values, assessed for 15 brands of brazilian economic wall paints.

In Table 1 the results for the natural radionuclides activities concentrations in brazilian economic wall paints are compared with activities concentration for common building materials.

Table 1: Natural radionuclides activities concentrations of wall paints and common building materials

Material	Activity concentration (Bq/kg)			Reference
	^{226}Ra	^{232}Th	^{40}K	
Wall paints	1.3 – 23.4	2.5 – 45.8	5.8 – 157	Present Work
Concrete	1.0 - 1300	1.0 - 152	7.0 - 1450	[18]
Cement	4.0 - 422	3.0 - 266	4.0 – 846	[18], [19]
Brick	2.0 - 148	2.0 - 164	12 - 1169	[18], [19]
Phosphogypsum	18 - 1406	2.0 - 118	0 - 569	[18]

4. CONCLUSIONS

Activities concentrations of ^{226}Ra, ^{232}Th and ^{40}K in 15 brands of brazilian economic wall paints shows that, as expected because of the titanium dioxide, from the ilmenite, present in the white pigment, there is radiation, but at low levels.

The radium equivalent indexes calculated for the wall paints are below the value of 370 Bq/kg recommended by OECD for a safety use in residential building applications, so these wall paints can be safely used.

As the economic wall paint samples have apparent densities between 1.15 g/cm^3 and 1.40 g/cm^3, higher than the density of the multi-element standard aqueous radioactive solution sample used for the determination of the detector efficiency curve, attenuation factors for all samples will be further determined.

Further, Standard and Premium wall paints will also be analyzed, for an extended evaluation of the radiation levels in brazilian commercial wall paints.

The complete assessment of the natural radioactivity in wall paints will be achieved with the evaluation of the external hazard index (H_{ex}), internal hazard index (H_{in}) and the gamma index ($I_γ$).

ACKNOWLEDGMENTS

Leandro Milhomens da Fonseca would like to thanks CAPES for the scholarship grant. The authors would like to thanks ABRAFATI and SENAI “Mario Amato” for the wall paints samples.

REFERENCES

1. UNSCEAR – United Scientific Committee on the Effects of Atomic Radiation (1993) "Sources, effects and risks of ionizing radiation". New York, United Nations
2. UNSCEAR – United Scientific Committee on the Effects of Atomic Radiation (2000) "The 2000 Report to the General Assembly with scientific Annexes". New York. United Nations.
3. R. Veiga, N. Sanches, R.M. Anjos, K. Macario, J. Bastos, M. Iguatemy, J.G. Aguiar, A.M.A. Santos, Mosquera, C. Carvalho, M. Batista Filho, N.K. Umisedo, "Measurement of natural radioactivity in Brazilian beach sands", *Radiation Measurements*, **41**, pp. 189-196 (2006).
4. A.M. El-Arabi, "^{226}Ra, ^{232}Th and ^{40}K concentrations in igneous rocks from eastern desert, Egypt and its radiological implications", *Radiation Measurements*, **42**, pp. 94-100 (2007).
5. A.O. Ferreira, B.R.S. Pecequilo, "Natural radioactivity assessment by gamma spectrometry in some commercially-used granites from Paraná State, Brazil: Preliminary results", *Radioprotection*, **46**, pp. S43-S47 (2011).
6. G.M. Bedinger, "Titanium Mineral Concentrates", U.S. Geological Survey, Mineral Commodity Summaries (2013).
7. K.R.S. Ferreira, C.H. Sampaio, A. Washington, "Characterization of a concentrate of ilmenite from guaju mine, northeastern Brazil", *XII Balkan Mineral Processing Congress,* Delphi, Greece (2007).
8. J.D. Dana, *Dana's System of Mineralogy,* **V. 2, ed. 7,** reviewed by Charles Palache, Harry Berman and Clifford Frondel, John Wiley & Sons (1951).
9. P.M.B. Harisadan, P.M.B. Pillai, R.M. Tripathi, V.D. Puranik, "Radiological implication due to thorium in titanium mineral separation and chemical processing", *Proceedings of the fifth International Symposium on NORM,* Seville, Spain, 19-22 March 2007 (2008).
10. J.P. Bolívar, R. García-Tenorio, J. Mantero, M.J. Gázquez, R. Lozano, "Occupational exposure assessment at a titanium dioxide plant located in the south-west of Spain", *Proceedings of the sixth International Symposium on NORM,* Marrakesh, Morrocco, 22-26 March 2010 (2011).
11. M.J. Gázquez, J.P. Bolívar, J. Mantero, R. García-Tenorio, F. Vaca, "Physico-chemical and radioactive characterization of co-products from titanium dioxide NORM industry for their use in cement manufacturing", *Proceedings of the sixth International Symposium on NORM,* Marrakesh, Morrocco, 22-26 March 2010 (2011).
12. A.A.A. Neto, "Titânio", *Sumário Mineral Brasileiro,* DNPM (2012).
13. "ABRAFATI – Associação Brasileira de Fabricantes de Tintas" http://www.abrafati.com.br (2014).
14. R.D. Evans, *The Atomic Nucleus*, McGraw-Hill, New York (1972).
15. InterWinner TM 6.0 MCA, 2004. Emulation, Data Acquisition and Analysis software for Gamma and Alpha Spectroscopy IW-B32. ORTEC. Oak Ridge, TN, USA.
16. L.F. Barros, B.R.S. Pecequilo, "Self-attenuation factors in gamma-ray spectrometry of select sand samples from Camburi beach, Vitória, Espírito Santo, Brazil", *Radiation Physics and Chemistry,* **95**, pp. 339-341 (2014).
17. SAAS, Staatliches Amt Für Atomsicherheit Und Strahlenschutz, (SAAS-250), "Grenzwerte für die Konzentration natürlicher Radionuklide in Baumaterialien bei unterschiedlichen Anwendungsfällen". Berlin, pp.205-213 (1979).
18. R. Trevisi, S. Risica, M. D'Alessandro, D. Paradiso, C. Nuccetelli, "Natural radioactivity in building materials in the European Union: a database and an estimate of radiological significance", *Journal of Environmental Radioactivity,* **105**, pp. 11-20 (2012).

19. N. Damla, U. Cevik, A.I. Kobya, B. Ataksor, U. Isik, “Assessment of environmental radioactivity for Batman, Turkey”, *Environmental Monitoring and Assessment,* **160**, pp. 401-412 (2010).