

NATURAL GAMMA-RAY EMITTERS IN CLAYS USED FOR THERAPEUTIC PURPOSES

Marcelo F. Máduar¹, Ricardo Ponciano² and Paulo S. C. da Silva³

¹Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
mmaduar@ipen.br

²Universidade Nove de Julho (UNINOVE)
Av. Dr. Adolpho Pinto 109
01156-050 São Paulo, SP
ricardopon@bol.com.br

³Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN)
Av. Professor Lineu Prestes 2242
05508-000 São Paulo, SP
pccsilva@ipen.br

ABSTRACT

The use of minerals for therapeutic purposes is an ancient practice, especially clay minerals topically employed in the therapy of skin disorders and rheumatic processes. In Brazil, an example of such application is the use of a clay-based mineral extracted from a deposit in Peruíbe, a resort town in the countrys southeast. Such mineral has been used both in raw form, after a washing process or as an active component of cosmetics. Currently, a comprehensive characterization of samples from this deposit is underway. As a part of this work, this paper presents preliminary results of the determination of natural radionuclides in samples of this clay and its derived preparations, in order to evaluate the radiological implications of such practice. The raw samples, after preparation, were analyzed by gamma-ray spectrometry. Self-attenuation corrections for the ^{210}Pb gamma radiation were applied. The radionuclide concentrations in dried samples varied in the ranges: for ^{226}Ra , 15.3 ± 0.5 to 18.4 ± 0.7 $\text{Bq}\cdot\text{kg}^{-1}$; for ^{228}Ra , 31.2 ± 0.5 to 37.8 ± 0.9 $\text{Bq}\cdot\text{kg}^{-1}$; for ^{210}Pb , 18 ± 5 to 32 ± 6 $\text{Bq}\cdot\text{kg}^{-1}$; for ^{40}K , 373 ± 14 to 454 ± 20 $\text{Bq}\cdot\text{kg}^{-1}$. The concentrations obtained are consistent with previously ones reported for clays. For the cosmetics, none of the target radionuclides were detected above the minimum detectable activities of the counting system, which are 2.5, 2.0, 10 and 20 $\text{Bq}\cdot\text{kg}^{-1}$ respectively for ^{226}Ra , ^{228}Ra , ^{210}Pb , and ^{40}K . An estimation of the effective dose on the skin due to the clay application by a modelling tool, with conservative parameters, led to an effective dose of 0.8 microsievert, indicating that the studied practice causes no significant radiation dose increment to the public.

1. INTRODUCTION

The use of minerals for therapeutic purposes is an ancient practice, especially clay minerals topically employed in the therapy of skin disorders and rheumatic processes [1, 2]. In Brazil, an example of such application is the use of a clay-based mineral extracted from a deposit in Peruíbe, a resort town in the countrys south-east. Such mineral has been used both in raw form, after a washing process or as an active component of cosmetics. Currently, a comprehensive characterization of samples from this deposit is underway.

As a part of this work, this paper presents preliminary results of the determination of natural radionuclides in samples of this clay and its derived preparations, in order to evaluate the radiological implications of such practice.

2. METHODS

2.1. Sample Preparation

The raw samples were dried and crumbled before conditioning, while the cosmetics samples were prepared as they are marketed. Both kinds of samples were conditioned in 100 mL polyethylene flasks and counted after a minimum 30 days delay to allow equilibrium between ^{226}Ra and its short-lived decay products.

2.2. Gamma-ray spectrometry

Determination of gamma radioactivity emitters content in clays and other ores is usually carried out by gamma-ray spectrometry [3].

Activity concentrations of ^{226}Ra , ^{228}Ra , ^{210}Pb , and ^{40}K were determined by counting the samples in a HPGe detector with 25% relative efficiency and Be-layer, extended range close-end coaxial geometry, model GX2518 from Canberra Industries, and associated electronics. Prior to the determinations, the gamma-ray detection system must be calibrated employing reference standards prepared in the same geometry of the samples.

The calibration procedure consists in analyzing multielementar radionuclide sources for the particular counting geometry employed. As a result, a counting efficiency vs. gamma energy fit is obtained. Then the samples are counted and, by application of the efficiency curve previously obtained, the radionuclide activities is determined.

Prior to the measurements, samples were homogenized, sealed and left apart for a 30-days minimum delay to ensure that radioactive equilibrium between ^{226}Ra and its short-lived decay products is obtained.

Table 1 shows the gamma-ray lines used in the determination of the activity concentrations. Self-attenuation by the samples for the ^{210}Pb gamma radiation can occur, due to its low energy. The correction for this effect was performed for each sample according to the procedure described by Cutshall et al. [4] and adapted for the geometry of 100 mL cylindrical polyethylene flasks by Máduar & Miranda [3]. Spectra analysis was carried out using a code for peak analysis by Máduar & Pecequilo [5].

2.3. Modelling of doses

The clay mineral under study is been employed, in the context of a complementary therapy for the treatment of rheumatic diseases [6]. Since such mineral contains natural radionuclides, a radiation dose assessment to estimate the radiological implications of such practice to the public was applied.

Table 1: Gamma-ray lines for the activity concentrations determination

Radionuclide	Gamma-ray Energy (keV)	I%
²²⁶ Ra	295.2	18.4
	351.9	35.5
	609.3	44.1
²²⁸ Ra	911.1	28.2
	968.9	17.0
²¹⁰ Pb	46.5	4.1
⁴⁰ K	1460.8	10.7

To perform the modelling, a deterministic code to calculate the absorbed dose to the skin from the activity concentrations and exposure geometry, VARSKIN 3 [7], was employed.

Very conservative assumptions were adopted. A hypothetical mud, containing simultaneously radionuclide concentrations above the maximum experimental concentrations of ²²⁶Ra, ²²⁸Ra, ²¹⁰Pb and ⁴⁰K determined in the samples, was adopted. In the model, the mud application is modelled as a 2-dimensional source disk directly on the skin. Table 2 shows the input parameters for VARSKIN 3.

The parameters simulate 1 kg of the hypothetical mud distributed in a 2-D disk with area 2000 cm², which is 10% of the total body surface area of a male adult [8] and assuming 200 h of mud application on the skin.

According to the procedure described in the ICRP Publication 103 [8], the equivalent dose H_T was assessed by

$$H_T = \sum_R D_{T,R} \cdot w_R \quad (1)$$

where H_T is the equivalent dose absorbed by tissue T , $D_{T,R}$ is the absorbed dose in tissue T by radiation type R and w_R is the radiation weighting factor, equal to 1 for both beta and gamma radiation.

As the application involves exclusively the topical application on the skin, the effective dose E was assessed as

$$E = \sum_T w_T \cdot H_T \quad (2)$$

Table 2: Input parameters for application of VARSKIN 3 code

Parameter	Value
Source geometry	2-D disk
^{226}Ra	$0.01 \text{ Bq}\cdot\text{cm}^{-2}$
^{228}Ra	$0.02 \text{ Bq}\cdot\text{cm}^{-2}$
^{210}Pb	$0.025 \text{ Bq}\cdot\text{cm}^{-2}$
^{40}K	$0.25 \text{ Bq}\cdot\text{cm}^{-2}$
Skin density thickness	$7 \text{ mg}\cdot\text{cm}^{-2}$
Air gap thickness	0 mm
Protective clothing thickness	0 mm
Protective clothing density	$0 \text{ g}\cdot\text{cm}^{-3}$
Source diameter	50.463 cm
Source area	2000 cm^2
Irradiation time	200 h
Irradiation area	2000 cm^2

Where w_T is the tissue weighting factor, equal to 0.01 for the skin.

3. RESULTS AND DISCUSSION

3.1. Radionuclide concentrations

Table 3 shows the results of activity concentrations for each sample and radionuclide.

Samples description are as follows: LN1, LN2 and LN3 are raw black mud and LNM1 is black mud submitted, at the SPA where it is applied, to a washing process with sea water collected at the site. Samples from "hair conditioner" to "shampoo" are of cosmetic products having various black mud contents on their formulations. The dried samples were collected from the same batches of the four first samples, as described.

The highest radionuclide concentrations were found in the dried samples, and varied in the ranges: for ^{226}Ra , 15.3 ± 0.5 to $18.4 \pm 0.7 \text{ Bq}\cdot\text{kg}^{-1}$; for ^{228}Ra , 31.2 ± 0.5 to $37.8 \pm 0.9 \text{ Bq}\cdot\text{kg}^{-1}$; for ^{210}Pb , 18 ± 5 to $32 \pm 6 \text{ Bq}\cdot\text{kg}^{-1}$; for ^{40}K , 373 ± 14 to $454 \pm 20 \text{ Bq}\cdot\text{kg}^{-1}$. The concentrations obtained are consistent with previously ones reported for clays. For the cosmetic products, none of the target radionuclides were detected above the minimum detectable activities of the counting system, which are 2.5, 2.0, 10 and $20 \text{ Bq}\cdot\text{kg}^{-1}$ respectively for ^{226}Ra , ^{228}Ra , ^{210}Pb , and ^{40}K

Table 3: Activity concentrations ($\text{Bq}\cdot\text{kg}^{-1}$) determined in samples of black mud and derived cosmetic products

Samples Identification	^{226}Ra	^{228}Ra	^{210}Pb	^{40}K
Raw samples				
LN1	8.5 ± 0.2	14.7 ± 0.3	14.9 ± 2.7	179 ± 7
LN2	7.4 ± 0.3	13.7 ± 0.3	7.4 ± 3.1	169 ± 8
LN3	7.7 ± 0.2	15.1 ± 0.3	8.4 ± 1.5	178 ± 8
LN M1	6.5 ± 0.3	13.1 ± 0.3	12.4 ± 3.2	155 ± 7
Hair conditioner	< AMD	< AMD	< AMD	< AMD
Moisturizer	< AMD	< AMD	< AMD	< AMD
LNNE	5.8 ± 0.6	13.6 ± 0.6	42 ± 8	154 ± 11
Natural mask	5.9 ± 0.4	13.1 ± 0.5	41 ± 6	161 ± 9
Bar soap	< AMD	< AMD	< AMD	< AMD
Liquid soad	< AMD	< AMD	< AMD	< AMD
Shampoo	< AMD	< AMD	< AMD	< AMD
Dried samples				
LN1	15.3 ± 0.6	36.1 ± 0.8	18 ± 5	424 ± 19
LN2	16.8 ± 0.4	31.2 ± 0.5	18.3 ± 2.2	373 ± 14
LN3	15.7 ± 0.8	36.0 ± 1.0	15 ± 7	407 ± 20
LN M1	18.4 ± 0.7	37.8 ± 0.9	32 ± 6	454 ± 20

3.2. Effective dose

The absorbed dose rate and total dose are shown in Table 4. The effective dose due to both beta and gamma radiation on the skin, during the treatment time span of 200 h, was assessed as $E = 0.8 \mu\text{Sv}$.

4. CONCLUSIONS

The final result for the effective dose to the member of the public was of the order of $1 \mu\text{Sv}$, figure that is three orders of magnitude lower than the reference level of 1 mSv per year for the public members. Therefore, the radiation dose to the public arising from the practice studied may be considered negligible, even assuming a very conservative scenario.

This study will continue, with the collection of additional samples to improve the statis-

Table 4: Absorbed dose results from VARSKIN 3 code

Radiation type	Absorbed dose rate	Absorbed dose
Beta	3.93×10^{-7} Gy/h	7.86×10^{-5} Gy
Gamma	3.11×10^{-9} Gy/h	6.22×10^{-7} Gy
Total	3.96×10^{-7} Gy/h	7.92×10^{-5} Gy

tical representativeness and to detect possible seasonal variations.

ACKNOWLEDGMENTS

This work has the support of Fundação de Amparo à Pesquisa do Estado de São Paulo, under research project no. 2012/16642-9.

REFERENCES

1. E. B. Quitete, P. R. M. Leal, “As propriedades das lamias na utilização terapêutica”, *Congresso Brasileiro de Geologia*, Araxá (2004).
2. M. Carretero, C. Gomes, F. Tateo, *Developments in Clay Science*, vol. 1, chap. 5, pp. 717–741, Elsevier (2006).
3. M. F. Máduar, P. Miranda Jr, “Efficiency calibration of a gamma-ray spectrometry system and self-absorption measurements of the 46.5 keV gamma-ray emission from ^{210}Pb for the quantitative determination of radionuclides in samples”, *Proceedings of the International Nuclear Atlantic Conference*, Belo Horizonte, 24-28 Oct, 2011 (2011).
4. N. H. Cutshall, I. L. Larsen, C. R. Olsen, “Direct analysis of ^{210}Pb in sediment samples: Self-absorption corrections”, *Nuclear Instruments & Methods in Physics Research*, **206**(1), pp. 309–312 (1983).
5. M. Máduar, B. Pecequilo, “Open-source implementation of an algorithm for photopeaks search and analysis in gamma-ray spectrometry with semiconductor detectors”, *Proceedings of the International Nuclear Atlantic Conference*, pp. 978–85, Rio de Janeiro, Sep 27 - Oct 2, 2009 (2009).
6. Z. M. N. Britschka, W. R. Teodoro, A. P. P. Velosa, S. B. V. de Mello, “The efficacy of Brazilian black mud treatment in chronic experimental arthritis”, *Rheumatology International*, **28**(1), pp. 39–45 (2007).
7. J. S. Durham, “VARSKIN 3: a computer code for assessing skin dose from skin contamination”, *Washington, DC: US Nuclear Regulatory Commission* (2006).
8. ICRP, “The 2007 Recommendations of the International Commission on Radiological Protection”, *ICRP publication*, **103** (2007).