

RADIOLOGICAL CHARACTERIZATION OF PELOIDS MATURATED WITH ÁGUAS DE LINDÓIA, POÇOS DE CALDAS AND PERUÍBE WATERS

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ABSTRACT

One of the concerns about using mineral clay for therapy treatments is its radioactivity content due to natural radionuclides, normally, associated with the clays. This work proposes to characterize the natural radionuclides of the peloids obtained by the maturation process of mixing bentonite and montmorillonite with different mineral medicinal waters from Águas de Lindóia (SP), Peruíbe (SP) and Poços de Caldas (MG). For this procedure, gray end green bentonite samples were left in contact with running water for three, six and nine months, after this, they were collected, dried, transferred to a mortar, crushed and placed in approximately 40 cm³ polyethylene flasks, sealed and set apart for about four weeks, prior to the measurements. The concentration of ²¹⁰Pb was determined by measuring the activity of its low energy peak (47 keV). Comparing the gray and green bentonite peloids, all the activity concentration of radionuclides are higher in gray ones, except ⁴⁰K. The activity concentration varied from 84 to 156 Bq kg⁻¹ (²²⁸Ra), 25 to 156 Bq kg⁻¹ (²²⁸Th), 9 to 161 Bq kg⁻¹ (²²⁶Ra), 39 to 256 Bq kg⁻¹ (²¹⁰Pb) and 162 to 1070 Bq kg⁻¹ (⁴⁰K).

1. INTRODUCTION

Peloids have been used as thermal therapeutic agents in many spas and thermal centers since ancient times. The term ‘peloid’ is used to refer to different types of sediments or deposits whose compositions include mainly silicates (micas, clays, feldspars, etc.) but also carbonates, sulphates, sulphides and a variable amount of organic substances. When mixed with sea or mineral-medical waters these elements form pastes or poultices for thermal uses [1]. For peloid obtainment, it passes by a maturation process, during which its characteristic greasiness is acquired, due to components mixing and the growth of organic constituents that arise from biological activity [2]–[4]. Several authors have studied some properties related to the suitability for therapy in clays from various parts of the world, such as Italy [5]–[8], Portugal [9]–[12] and Spain [1], [13]–[15]. In addition, some properties of peloids prepared

with clays, different water types and different maturation conditions were also studied [4], [16]–[23].

Treatment with peloids is called “pelotherapy” and it is famous as an extremely effective therapy worldwide and is a commonly used method for the cure of numerous syndromes, for example, skin, rheumatic and joint diseases [24]. A concern about using mineral clay for therapy treatments is its radioactivity content due to natural radionuclides, normally, associated with the clays and during the treatments, natural radioactivity can be beneficial to patients, but negative side effects may occur at greater doses, e.g. chronic lung diseases, leukaemia and bones, kidney and pancreas cancers, nevertheless, there are very few studies evaluating the radioactivity of thermal muds or clay minerals [24], [25], [26]. Hence, measurement of gamma radiation levels provides valuable information about dose levels [24].

Exposure to radioactivity is commonly considered objectionable at all exposure levels, even though no harmful effects have been reported at very low levels of radioactivity [27]. Natural radioactivity in soils is caused mainly by the $^{238}\text{U}/^{232}\text{Th}$ decay series and natural ^{40}K [28].

This study proposes to determine the radiological characterization of the peloids artificially obtained by the maturation process of mixing two types of bentonites with mineralo-medicinal water from Águas de Lindóia (SP) and Poços de Caldas (MG), and sea water from Peruíbe (SP).

Because of their various properties, bentonites are being used in an increasing number of applications, especially in pelotherapy and medical applications [4]–[6], [29]–[34]. In this research were used two types of bentonites to the maturation process, both acquired in formal market. Macroscopic observation showed that the two bentonites are fine-grained, homogeneous and generally light-colored: gray and green.

Few studies about natural radioactivity have been found in the waters used for the peloid maturation used in the present work. A study made by Negrão, 2012 showed that the activity concentration of ^{226}Ra and ^{228}Ra in Águas de Lindóia water ranged from 4.6 - 19 mBq L⁻¹ and 38 - 54 mBq L⁻¹, respectively, concluding that these values are not harmful to the humans [35]. This water is also known as hypo-saline water that emerges at 37 °C and is commonly used for rheumatic and skin affections treatment.

Although Poços de Caldas water is known to be radioactive, it was not possible to find scientific records of studies about determination of radionuclides in this water. The Poços de Caldas water is sulphurous, reach the surface at 45 °C and is commonly used for rheumatism treatments. Sea water, from Peruíbe, was used because of its use in thalassotherapy treatment.

2. METHODOLOGY

2.1. Maturation process

For the maturation process both bentonites were left in contact with running water in the Balneário Municipal de Águas de Lindóia, in Águas de Lindóia (SP), Thermas Antonio Carlos, in Poços de Caldas (MG) and in standing water at the Complexo Thermal da Lama Negra de Peruíbe, in Peruíbe (SP).

One of the clays was bought in a place for clay products, sold as “bentonite”, with gray color (B1) the other clay was bought in a natural products shop, sold as “montmorillonite”, with green color (B2).

The Águas de Lindóia there are four springs and they are protected in a construction of approximately 4 m², and within this construction it is possible to see the budding of water from the rocks. For the maturation process in this place was chosen the spring named “Madame Curie”

In Poços de Caldas there are three springs named “Chiquinha”, “Mariquinha” and “Pedro Botelho”, the three springs are totally piped and the only access to the water is through faucets. The plumbing of springs is inside a building of approximately 100 m². Due to this the samples were matured in two different springs, that is, different waters. The gray bentonite was matured in “Chiquinha” and other one in “Mariquinha” spring.

In Peruíbe, the maturation was performed keeping the mud in contact with standing sea water, taken 2 km far from the coast and no agitation was employed in the process. This procedure was made following Silva et al., 2015 [36].

Therefore, in this study, the procedure of maturation of 2 kg of clay at each site was adopted for nine months and collections were made every three months.

For the identification of the samples, the following criteria were used: the first two characters of the codification of each sample refers to clay color used, thus, B1 (gray bentonite) and B2 (green bentonite), the following two letters refer to water used for maturation, then AL (Águas de Lindóia), PC (Poços de Caldas) and PE (Peruíbe) and finally, the number represents maturation time, 3 for three months, 6 for six months and 9 for nine months. The letter “P” after the number of maturation months means that the clays were matured in standing water with Águas de Lindóia water.

2.2. Gamma spectrometry

Activity concentrations of ²²⁶Ra, ²²⁸Ra, ²¹⁰Pb, ²²⁸Th and ⁴⁰K were measured by gamma spectrometry with a high-pure germanium detector, GX2020, from Canberra. The detector was calibrated with ⁴⁰K, ²³²Th and ²³⁸U standards. Samples were placed in 40 cm³ polyethylene flasks, sealed and set apart for about four weeks, prior to the measurements, to ensure reaching of radioactive equilibrium between ²²⁶Ra and its short-living decay products. The ²²⁶Ra activities were determined by taking the mean activity of three separate photopeaks of its daughter nuclides: ²¹⁴Pb at 352 keV, and ²¹⁴Bi at 609 keV and 1120 keV. The ²²⁸Ra content of the samples was determined by measuring the intensities of the 338 keV and 911 keV gamma-ray peaks from ²²⁸Ac. The ²²⁸Th activities were determined by taking the mean activity of three separate photopeaks of its daughter nuclides: ²¹²Pb at 238 keV and ²¹²Bi at 727 keV. The concentration of ²¹⁰Pb was determined by measuring the activity of its low energy peak (47 keV). The concentration of ⁴⁰K was determined by measuring the activity of its high energy peak (1046 keV) Self-absorption correction was applied because the attenuation for low energy gamma rays is highly dependent upon the sample composition [37].

3. RESULTS AND DISCUSSION

The activity concentrations obtained for ^{228}Ra , ^{228}Th (^{232}Th -series), ^{226}Ra , ^{210}Pb (^{238}U -series) and ^{40}K in non matured clays and the peloids obtained after maturation are presented on Table 1.

Table 1: Activity concentrations of ^{226}Ra , ^{228}Ra , ^{210}Pb , ^{228}Th and ^{40}K (Bq kg^{-1}), in gray and green clays before and after the maturation process as well the worldwide average according to UNSCEAR (2000) [38].

Sample	^{232}Th -series		^{238}U -series		^{40}K
	^{228}Ra	^{228}Th	^{226}Ra	^{210}Pb	
GRAY BENTONITE	142 ± 65	148 ± 49	114 ± 17	256 ± 7	162 ± 11
B1AL3	156 ± 71	149 ± 50	110 ± 17	159 ± 14	203 ± 14
B1AL6	140 ± 63	133 ± 43	93 ± 15	72 ± 25	154 ± 9
B1AL9	157 ± 71	136 ± 45	115 ± 17	150 ± 10	212 ± 14
B1AL6P	140 ± 64	152 ± 49	104 ± 16	ND	189 ± 12
B1AL9P	150 ± 68	147 ± 49	105 ± 16	56 ± 34	139 ± 10
B1PC3	156 ± 71	156 ± 53	110 ± 17	147 ± 6	191 ± 13
B1PC6	155 ± 71	146 ± 47	115 ± 17	284 ± 8	328 ± 22
B1PC9	156 ± 73	133 ± 44	172 ± 27	304 ± 12	987 ± 55
B1PE3	119 ± 54	120 ± 39	96 ± 15	276 ± 5	198 ± 13
B1PE6	126 ± 57	123 ± 39	102 ± 15	198 ± 6	160 ± 11
B1PE9	131 ± 60	122 ± 42	103 ± 16	388 ± 8	192 ± 12
GREEN BENTONITE	54 ± 25	73 ± 25	53 ± 8	63 ± 4	676 ± 41
B2AL3	69 ± 31	68 ± 23	53 ± 8	51 ± 10	760 ± 44
B2AL6	68 ± 31	75 ± 27	52 ± 8	140 ± 6	800 ± 48
B2AL9	74 ± 34	68 ± 24	61 ± 10	236 ± 5	793 ± 47
B2PC3	73 ± 35	73 ± 28	9 ± 2	109 ± 26	1070 ± 66
B2PC6	73 ± 33	73 ± 25	56 ± 8	76 ± 14	934 ± 56
B2PC9	72 ± 34	65 ± 24	60 ± 10	53 ± 12	802 ± 45
B2PE3	59 ± 27	57 ± 20	50 ± 8	39 ± 8	782 ± 45
B2PE6	55 ± 25	59 ± 19	52 ± 8	54 ± 5	795 ± 46
B2PE9	73 ± 34	89 ± 31	52 ± 8	53 ± 5	843 ± 50
UNSCEAR (2000)	30		35		400

The gray bentonite non matured showed activity concentration for radionuclides from ^{232}Th -series about three times greater than green bentonite and the ^{238}U -series about two times. However, the activity concentration of ^{40}K is about four times lower.

After the maturation process the gray bentonite showed a change in the activity concentrations only for the radionuclides ^{226}Ra , ^{210}Pb and ^{40}K matured with water from Poços de Caldas (Chiquinha spring), and was noted that the activity concentration increase according to the maturation time. The increase of ^{210}Pb may be related to the increase of organic matter after maturation with this water. For the other radionuclides no difference in activity concentrations with maturation in any of the waters was observed.

Although the activity concentration of the ^{40}K radionuclides increased in green bentonite after the first three months of maturation with Poços de Caldas water, it was noticed a decrease of these concentrations after nine months, however, in gray bentonite was noticed a increased of ^{40}K according to maturation time.

In gray bentonite was possible to noted a increased of activity concentration of ^{210}Pb in maturation with Poços de Caldas and Peruíbe waters after nine mounths and slightly difference was noted in maturation with Águas de Lindóia water. However, for this radionuclideo, in gray bentonite, was noted a increased of activity concentration according to maturation time with Águas de Lindóia water, and a decreased with Poços de Caldas water. It is important to remember that the clays were matured at different springs in Poços de Caldas, and this may have occasioned the different behavior of these radionuclides after maturation. In peloids obtained in Águas de Lindóia water, the difference in ^{210}Pb behavior should be related only to the difference between the clays used.

The maturation process with Peruíbe water have caused a slightly increased in activity concentration of ^{228}Ra in gray bentonite and ^{228}Ra and ^{228}Th in green one.

Comparing the results obtained in these samples with the worldwide average values of radionuclides ^{226}Ra , ^{232}Th and ^{40}K , according to UNSCEAR (2010) [39], it was verified that gray and green clays presented activity concentration slightly higher for the radionuclides of the ^{232}Th -series and the gray peloids presented higher values (about four times). For the ^{40}K , the green peloids presented values above the worldwide average, while those of gray peloids were below.

The results of ^{226}Ra and ^{228}Ra obtained in this work for the gray and green bentonite peloids, are according to the values studied by Silva et al., 2011 [26] in kaolins mainly composed of well-crystallized kaolinite and bentonites containing smectites commercial clays for pharmaceuticals and cosmetics propouses. All the peloids studied here presented activity concentration of ^{226}Ra and ^{40}K in agreement to the values measured by Karakaya., 2015 [24] (8.91 to 401.64 Bq kg⁻¹ for ^{226}Ra and 64.82 to 1698.60 Bq kg⁻¹ for ^{40}K) in peloids used for musculoskeletal disorders and aesthetic treatments from Turkey spas and these values are not significant.

Compared with mud used in a spa in Serbia, the activity concentration of ^{226}Ra in the present samples were lower for gray and green peloids (259 Bq kg⁻¹) and the activity concentration of ^{40}K were of the same order (219 Bq kg⁻¹) for gray peloids (except in samples matured in Poços de Caldas waters) and higher in green ones [25].

One of the concerns regarding the use of clays in pelotherapy, generally, is the dose of radiation that can be received by patients because their content of natural radionuclides.

The absorbed dose rate (D) is related to the risk due to the amount of radiation deposited in a body per unit of time that arises from terrestrial gamma emitters. D can be derived (nGy h⁻¹) from the measured activity concentrations and the following conversion factors, as given by UNSCEAR (2010) [39] and shown in Eq 1:

$$D = 0.462C_U + 0.604C_{Th} + 0.0417C_K \quad (1)$$

The results presented here are about two to three times greater than the worldwide average, which is 58 nGy h⁻¹ [40]. Table 2 shows the results of absorbed dose rate.

Table 2: Absorbed dose rate (Gy h⁻¹) in gray and green clays before and after the maturation process.

Sample	Dose rate	Sample	Dose rate
GRAY BENTONITE	145 x 10 ⁻⁹	GREEN BENTONITE	85 x 10 ⁻⁹
B1AL3	154 x 10 ⁻⁹	B2AL3	98 x 10 ⁻⁹
B1AL6	134 x 10 ⁻⁹	B2AL6	98 x 10 ⁻⁹
B1AL9	157 x 10 ⁻⁹	B2AL9	106 x 10 ⁻⁹
B1AL3P	139 x 10 ⁻⁹	B2PC3	93 x 10 ⁻⁹
B1AL6P	140 x 10 ⁻⁹	B2PC6	109 x 10 ⁻⁹
B1AL9P	145 x 10 ⁻⁹	B2PC9	105 x 10 ⁻⁹
B1PC3	153 x 10 ⁻⁹	B2PE3	91 x 10 ⁻⁹
B1PC6	160 x 10 ⁻⁹	B2PE6	90 x 10 ⁻⁹
B1PC9	215 x 10 ⁻⁹	B2PE9	103 x 10 ⁻⁹
B1PE3	124 x 10 ⁻⁹		
B1PE6	130 x 10 ⁻⁹		
B1PE9	135 x 10 ⁻⁹		

The results of the absorbed dose rate of the non-matured gray clay and its peloids varied from 124x10⁻⁹ to 215x10⁻⁹ Gy h⁻¹, while those of green clay presented slightly lower values varying from 85x10⁻⁹ to 106x10⁻⁹ Gy h⁻¹. Most of the calculated absorbed dose rates are varying from 28x10⁻⁹ to 120x10⁻⁹ Gy h⁻¹ recommended by UNSCEAR (2008) [38].

The maturation process caused a significant increase in the absorbed dose rate only in gray clay matured with Poços de Caldas water for nine months, varying from 153x10⁻⁹ to 215x10⁻⁹ Gy h⁻¹, which represents, in the worst case, 1.3 μSv for 100 h of application, which is 1000 times less than the allowable dose increase of 1 mSv per year. For comparison in Europe the annual effective dose from all sources of radiation in the environment is estimated to be about 3.3 mSv [25].

4. CONCLUSION

The aim of this study was evaluated the maturation process of two types of bentonites in different waters from Águas de Lindóia, Poços de Caldas and Peruíbe. The activity concentration of ²¹⁰Pb, ²²⁸Ra and ²²⁸Th is greater than the worldwide average for all clays and peloids studied in this work but it agrees when compared with peloids from different spas. However, by calculating the absorbed dose rate in the worst scenario, this may not be a problem for topical application. Although the waters from Águas de Lindóia and Poços de Caldas are considered radioactive they have not caused a significant increase in radionuclide concentrations absorbed dose rate.

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