RADIOISOTOPIC CHARACTERIZATION OF A CUBAN PELOID BY GAMMA SPECTROMETRY

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ABSTRACT

Peloids or thermal muds are complex colloidal systems with known therapeutic properties. In Cuba, peloids have been used for more than two centuries in an empiric way. The chemical characterization of these matrixes will help to obtain scientific bases to understand the behavior of bioactive and toxic elements in different human pathologies, as well as, spreading their use in medical treatments. The use of nuclear analytical techniques is suitable to characterize mainly the inorganic (including radioactive isotopes) phase of peloids. The objective of this work was to determine the natural and artificial radionuclides (gamma emitters) in the San Diego de los Baños peloid (Pinar del Rio, Cuba) by means of gamma spectrometry. The spectra show that the most significant radionuclides are ⁴⁰K, ¹³⁷Cs, and those from ²³⁸U and ²³²Th decay series. The activity concentrations of these radionuclides are in good agreement with those from regions of normal radioactive background. These results are consistent with references for similar sediments; nevertheless, ¹³⁷Cs has been reported for the first time in Cuban peloids.

Keywords: Peloid, sediment, spa, gamma spectrometry.

INTRODUCTION

Peloids or thermal muds are sediments that constitute complex colloidal systems formed by two (organic and inorganic) phases with known therapeutic properties [1, 2]. Thermal muds are hydrothermal or hydrothermalized pastes produced by primary or secondary mixing of clay materials with salty thermo-mineral waters, accompanied by organic materials produced by the biological-metabolic activity of micro-organisms growing during the so-called "maturation" process [3].

In Cuba, peloids have been used for more than two centuries in an empiric way, in the topical treatment of different human pathologies. The chemical characterization and quality control of these sediments are needed to evaluate their therapeutic properties. This characterization helps to correlate the components in the peloid to their biological action, as well as, spreading their medical use. In addition, the radioisotopic characterization facilitates the assessment of the radiological doses to patients and occupational workers.

The use of nuclear analytical techniques is suitable to perform the radioisotopic characterization of sediments. The objective of this work was to determine the natural and artificial radionuclides (gamma emitters) in San Diego de los Baños peloid (Pinar del Río, Cuba) by means of gamma-spectrometry.

SAN DIEGO DE LOS BAÑOS

San Diego de los Baños is a Thermal Center located in Pinar del Río, Cuba (Figure 1). The mud used at this Thermal Center, is an estuarine sediment, originated in the southern coast of Pinar del Río province. Sediment samples from San Diego River's mouth (22°19'49" N, 83°16'28" W), depth interval of 0-20 cm, were matured for 30 days with thermal waters (calcic sulfated, sulfhidric and radonic waters) to fulfill the requirements for therapeutic muds [1]. These peloids are used in therapeutic treatment of ostiomioarticular, immune and dermatologic diseases.

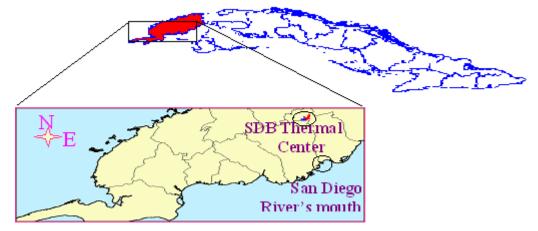


Figure 1. San Diego de los Baños Thermal Center.

EXPERIMENTAL

<u>Samples preparation</u>: The peloid was collected from the maduration pool in San Diego de los Baños Thermal Center. Afterwards, it was centrifuged, dried at room temperature to constant weight, homogenized, and sifted at $125 \,\mu$ m.

For gamma analysis, pretreated samples (50g) were packed in a cylindrical container (60 mm diameter and 20 mm height), sealed and stored for 28 days in order to attain secular equilibrium between the long-lived radioisotopes ²³⁸U, ²³²Th, ²²⁶Ra and their respective progeny.

Gamma spectrometry

Measurements were performed in a DSG Detector Systems GmbH gamma spectrometer, equipped with a HPGe Coaxial Detector (Type: NGC 3019) 30% relative efficiency and 1.9 keV resolution for the 1332 keV photopeak of ⁶⁰Co, coupled to a multi-channel analyzer. The detector has a beryllium entrance window (6 cm) that enables detection of gamma rays below 100 keV with excellent efficiency. In order to reduce laboratory background, the detector is shielded with iron 21 cm, cadmium 2 mm and very old lead 4,5 cm thick, and internally lined with 1 mm thick electrolytic Cu.

The certified material for soil IAEA-375 (soil from Chernobyl region (1990)) [4] was used for measurement calibration, combined with a point source of ²²⁶Ra. The sample counting time was 48 hours. The spectra were analyzed with Winner 6.0 software.

⁴⁰K, ²¹⁰Pb and ¹³⁷Cs activities were obtained from their corresponding gamma rays at 1460.83 keV, 46.54 keV and 661.66 keV, respectively. ²²⁶Ra determination was based on measurement of its decay chain descendant ²¹⁴Pb (351.93 keV) and the assumption of secular equilibrium. The activity concentrations of ²³⁸U and ²³²Th were determined by means of ²³⁴Th (63.29 keV) and ²²⁸Ac (991.07 keV) peaks, respectively. The errors on the activity concentrations arise from the statistical uncertainty in the peak areas. The background activity was properly subtracted.

RESULTS AND DISCUSSION

Gamma spectrometry

To our knowledge, it seems to be no information about radioactivity levels in San Diego sediments so far and no data have appeared in the literature concerning radionuclide concentrations in the specific region under study; therefore, no comparison in time is possible.

Gamma spectra of the peloid samples show the peaks corresponding to natural radionuclides from the ²³⁸U (²³⁴Th, ²²⁶Ra, ²¹⁴Pb, ²¹⁴Bi, ²¹⁰Pb), ²³²Th (²²⁸Ac, ²¹²Bi, ²⁰⁸Tl) and ²³⁵U decay series, besides ⁴⁰K. The anthropogenic radioisotope, ¹³⁷Cs, was also found in this matrix. Figure 2 illustrates an example of these spectra.

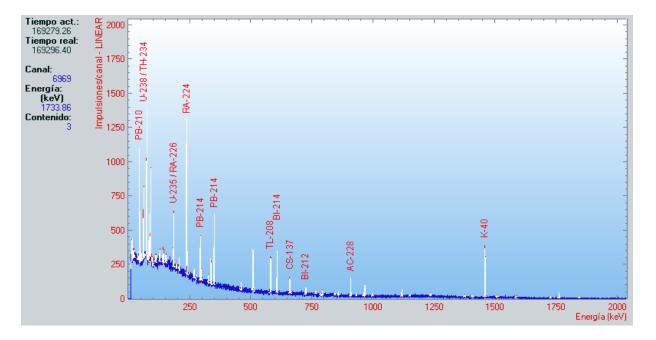


Figure 2. Gamma Spectrum of San Diego de los Baños Peloid.

Table 1 summarizes the dry weight activity concentrations of the radionuclides ⁴⁰K, ²³⁸U, ²²⁶Ra, ²¹⁰Pb, ²³²Th and ¹³⁷Cs in the peloid as well as the world averages for comparison.

Radionuclide	Activity concentration (dry weight) [Bq/kg]	U [Bq/kg]	World average and range (surface sediments) [Bq/kg]	
40 K	237	13	400 (140 - 850) [5]	
²³⁸ U	31	6	35 (16 – 110) [5]	
²²⁶ Ra	29	1	35 (17 – 60) [5]	
²¹⁰ Pb	61	6	40 - 100 [9]	
²³² Th	21	2	30 (11 – 64) [5]	
¹³⁷ Cs	5.0	0.4	6(1-17) (nowadays in northern hemisphere) [10, 11]	

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Table 1. Radionuclides a	ctivity concent	rations in the	peloid and	world averages.

The activity concentrations of natural radionuclides measured in this work are in good agreement with those from regions of normal radiactive background, reported by the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) in its performed gamma-ray surveys in soils and sediments [5]. However, ²²⁶Ra and ²¹⁰Pb contents in San Diego muds are slightly lower than those reported in a radioisotopic characterization of peloids from Elguea Thermal Center, located in the central region of Cuba [6].

The natural radionuclide ²¹⁰Pb ($T_{1/2} = 22.6$ y) has a double origin in the sediments. It is principally produced by ²²⁶Ra decay (via ²²²Rn) in the mineral components of sediment particles and additionally by atmospheric ²²²Rn decay. The atmospheric fraction of the ²¹⁰Pb activity - the so-called excess activity or unsupported fraction - is due to the fact that ²²²Rn partially escapes to

the atmosphere once is generated by ²²⁶Ra disintegration. As a consequence, ²¹⁰Pb is also produced in air and deposited into sediments isolated from its progenies. Therefore, sediments will contain an excess of ²¹⁰Pb with respect to that produced in the own sediment by ²²⁶Ra, which generally decreases with depth according to the half live of ²¹⁰Pb, depending on the sediment deposition rate. In the peloid, ²¹⁰Pb radioactivity reaches about the double of the ²²⁶Ra values. This excess is typical for surface sediments, like the one under study.

By the other hand, ¹³⁷Cs does not exist naturally in soils and sediments; it is a product of fallout radioactivity. ¹³⁷Cs has been deposited in marine sediments from all over the world, mainly in the northern hemisphere, as a result of the atmospheric nuclear weapon tests conducted by several countries in the past century, as well as the nuclear power plant accident at Chernobyl on April 1986. A large number of studies have shown that ¹³⁷Cs is preferentially bound on clay and organic particles and is rapidly and strongly absorbed in cation exchange sites [7, 8, 9].

The content of ¹³⁷Cs in San Diego's peloid (5 Bq/kg dry weight) is in the range for normal background in northern hemisphere (1 - 17) [10, 4, 11, 5]; although, it has not been reported in previous characterization of Cuban peloids [6]. The appreciable content of phylosilicates (39 %) [8], main component of clay particles (<2 µm), in the peloid of San Diego de los Baños contributes to the absorption of ¹³⁷Cs in the surface sediments, where it is virtually non-exchangeable [12, 13, 14, 8, 15].

CONCLUSIONS

According to the radioisotopic characterization by gamma spectrometry, the most significant radionuclides are those of terrestrial origin: ²³⁸U and ²³²Th series, besides ⁴⁰K. There was also found the anthropogenic radionuclide ¹³⁷Cs. The activity concentrations of these radionuclides are in good agreement with those from regions of normal radioactive background. These results are consistent with those reported in the literature for similar sediments; nevertheless, ¹³⁷Cs has been reported for the first time in Cuban peloids. This study will contribute to the assessment of radiological doses to patients and occupational workers in San Diego de los Baños Thermal Center.

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