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$^{232}Th/^{238}U$ IN A URANIUM MOBILITY ESTIMATE IN AN AGRICULTURAL AREA IN THE MUNICIPALITY OF PEDRA – PERNAMBUCO- BRAZIL

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ABSTRACT

The mobility of the radionuclides in soil depends primarily on the physic-chemical parameters. The uranium is easily oxidized in aqueous environment, which allows its characterization with higher mobility. The Thorium is practically insoluble, mainly if the environment has organic matter and sulfates. The geochemical characteristics of the rocks, associated with the weather and metamorphism produce alterations in the concentration diagrams of the natural radionuclides in different types of soil. The ratio $^{232}Th/^{238}U$ has been used as an indicator of oxidizing and reducing conditions. Th/U less than 2 suggests that the uranium is in its concentrated form abundantly when compared to the thorium. In reducing conditions, the value Th/U higher than 7 indicates a removal of the uranium. In this work it was possible to analyze the agricultural soil in the municipality of Pedra, Pernambuco, Brazil where there are uranium anomaly and thorium in rocky outcrops. Sixty-two samples of the horizon C soil were collected, in an area of 2 km², where the main uranium occurrences are located. The analyses were done by High-Resolution Gamma-Spectroscopy. In the analyses the secular equilibrium was assumed and the ^{238}U and the ^{232}Th specific activities were used to estimate the oxidizing and reducing conditions defining the uranium mobility in the soil. The obtained findings show that the ratio Th/U varied from 0.3 to 13.4, with average of 4.6. The biggest ^{238}U fraction was fix (80.3%), with low mobility; the smallest fraction concentrated (6.6%) and a lixiviated intermediate fraction (13.1%).

1. INTRODUCTION

The natural radionuclides mobility in the environment is essentially differentiated by the physic-chemical conditions available. The uranium is easily oxidized in aqueous solutions, which allows its characterization with higher mobility. In acidic rocks, the uranium concentrations may be 100 times high to the ones seen in the majority of the rocks [1]. The

Thorium is practically insoluble, mainly if the environment has organic matter and sulfates [2].

In dry weather regions with short rainy periods, the soil pH may vary from 7 to values higher than 7, and the predominant uranium ions in a period of one year will be $UO_2(OH)_3^-$, $UO_2(OH)^+$ and UO_2^{2+}. The UO_2^{2+} mobility in acidic environment is very high. It is worth noticing that the excess of carbonate ions reduces the uranium mobility [3,4].

According to Anjos et al. (2005) [5], the abundances of thorium and uranium normally increase from the basic to the acidic rocks. The majority of the radioactive occurrences in rocks is found in granites and is associated with the presence of pegmatites. According to these authors, the ratio thorium-uranium (Th/U) has been used as an indicator of oxidizing and reducing conditions: values Th/U below than 2 suggest that the uranium is disposed in the concentrated form (in abundance when compared to the thorium). In reducing conditions, Th/U values higher than 7 suggest a uranium removal, possibly due to the lixiviation.

According Mckee et al. (1987) [4], the U^{6+}, in oxidizing environments, is associated to the ferric ion Fe^{3+}, while the U^{4+} is found in the same conditions in which the iron is reduced (ferric ion). It might be possible that the ferric ion reduces the uranyl ion [4]. Reducing environment, with pH higher than 4.5, propitiate the uranium precipitation. The UO_2^{2+} is a strong oxidant and therefore the joined occurrence of uraninite and hematite (Fe_2O_3) are derived, due to the Fe^{2+} to Fe^{3+}oxidation when it goes through an aqueous solution rich in UO_2^{2+}.

Considering the possibility of determining the oxidizing and reducing conditions for the uranium by the ratio Th/U, this work had as its objective to determine the uranium mobility in the uraniferous occurrence in the municipality of Pedra, Pernambuco, Brazil [6,7], utilizing the analyses of the ^{238}U and ^{232}Th by High Resolution Gamma Spectrometry in soil samples collected in the adjacencies.

2. MATERIAL AND METHODS

2.1. Study area, sample collection and preparation

The study area is located in the municipality of Pedra, Pernambuco, Brazil. The municipality is located in the realms of the hydrographic bay of Ipanema river, with water from the ponds of Arcoverde, Mossoró, besides the lagoons of Bicheiro, Jacu, of the Grande, Algodão and of Anzol. The average temperature is of 22,9°C and its vegetation is of the hypoxerophytic caatinga. Cattle farming represents 65.3% of the economic activities in the municipality, being the dairy cattle the main income source of the municipality, with a monthly production of over 2,000,000 liters of milk.

The collecting area has the coordinates: 08º37.693'S e 36º55.220'W with an extension of approximately 2 km^2. This area was chosen for its rocky outcrops with uraniferous anomalies. Moreover, the anomalous rocks in uranium are located in dairy farms.

In this area, survival agricultural activities for feeding the cattle or cattle-related (like the cultivation of palm, cut and trampling grass are also developed.

Sixty-two horizon C soil samples were collected, with average depth of 45 cm, which classifies the soil as lit soil, enabling therefore, the collection of autochthon samples.

In the lab, the soil samples were dried in greenhouse at 60ºC, sieved with a 1.0 mm openness sieve, homogenized, quartered and 250 g of each, conditioned in plastic bags hermetically closed for further indirect analysis by gamma-spectrometry.

2.2. Measurements system

The analyses were done in the Departamento de Energia Nuclear of the Universidade Federal de Pernambuco, utilizing an experimental arrangement made by a High Resolution Gamma-Spectrometry HPGe-Be Canberra®, with active volume of 41.1 cm^3. The detector presents resolution of 1,77 keV for the energy of 1,332 keV of ^{60}Co, nominal efficiency of 27.7% and is attached to a model 2002 CSL MCA pre amplifier, with 8 k channels and software Genie®-2000 Canberra®.

2.3 Measurements Standardization and activity calculation

To determine the necessary parameters in the uranium mobility estimate in the environment, gamma transitions with less interference were used, In the analysis of the ^{238}U the gamma transition with approximately 63 keV of ^{234}Th [8] and for the ^{232}Th, the photopics of 338 keV, 911 keV and 969 keV of ^{228}Ac [9], assuming the secular radioactive equilibrium state. The time used for the acquisition was adjusted to 43,200 seconds, where the measures were made at 0 cm of the detector.

For the calculation of the ^{238}U e do ^{232}Th specific activities an efficiency in energy curve was built using the ^{241}Am and ^{252}Eu reference certified by the Instituto de Pesquisas Energéticas e Nucleares (IPEN) of the Comissão Nacional de Energia Nuclear (CNEN), as procedures of the International Atomic Energy Agency, TECDOC 619 [10] and using the equation 1 (IAEA, 1989 apud [11]).

$$A = \frac{N_{Ei}}{\varepsilon \times I_{\gamma} \times t \times m} \quad (1)$$

where: A represents the specific activity determined for the radionuclide considered; the N_{Ei} total liquid area of the interest fotopico; ε the energy counting efficiency ($\varepsilon \leq 1$); t the counting time; I_{γ} the radionuclide gamma abundance ($I_{\gamma} \leq 1$); and m the mass of the analyzed sample .

3. RESULTS

Figure 1 presents the efficiency in energy curve used to determine the efficiency in energy for the gamma transitions used in the quantification of the ^{238}U and of the ^{232}Th, built from standard sources of the ^{241}Am and ^{152}Eu (energetic range varying from59 keV to 1,400 keV).

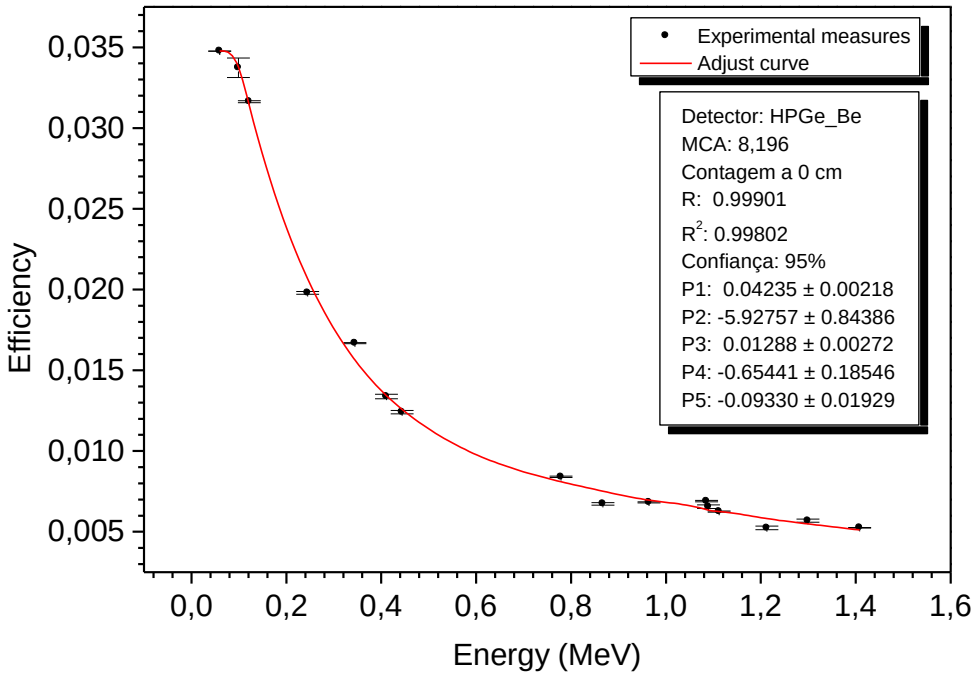

Figure 1. Efficiency in energy curve.

The adjustments of the efficiencies were made based on software which enabled the correction parameters establishment, allowing the determination of the equation 2 used for the correction of the interest radionuclides counting efficiencies.

$$\varepsilon = (p_1.e^{E.p_2} + p_3.e^{E.p_4}).e^{-\mu.p_5} \quad (2)$$

In this equation, ε is the counting efficiency ; $p_1 = 0.042$; $p_2 = -5.928$; $p_3 = 0.013$; $p_4 = -0.654$ and $p_5 = -0.093$, are the defined parameters by the software for the adjustment of the curve; E is the energy for which the calculation of the counting efficiency is desirable and μ is the mass attenuation coefficient, defined by equation 3, considering $a_1 = 0.0587$; $a_2 = -0.4160$; $a_3 = 0.0005$ and $a_4 = -2.9430$, the used parameters for the detector characteristics .

$$\mu = a_1.E^{a_2} + a_3.E^{a_4} \quad (3)$$

Figure 2 presents the Th/U ratio diagram for the analyzed soil, as well as the estimate of the uranium availability in function of the gamma activities of the ^{238}U and of the ^{232}Th series elements.

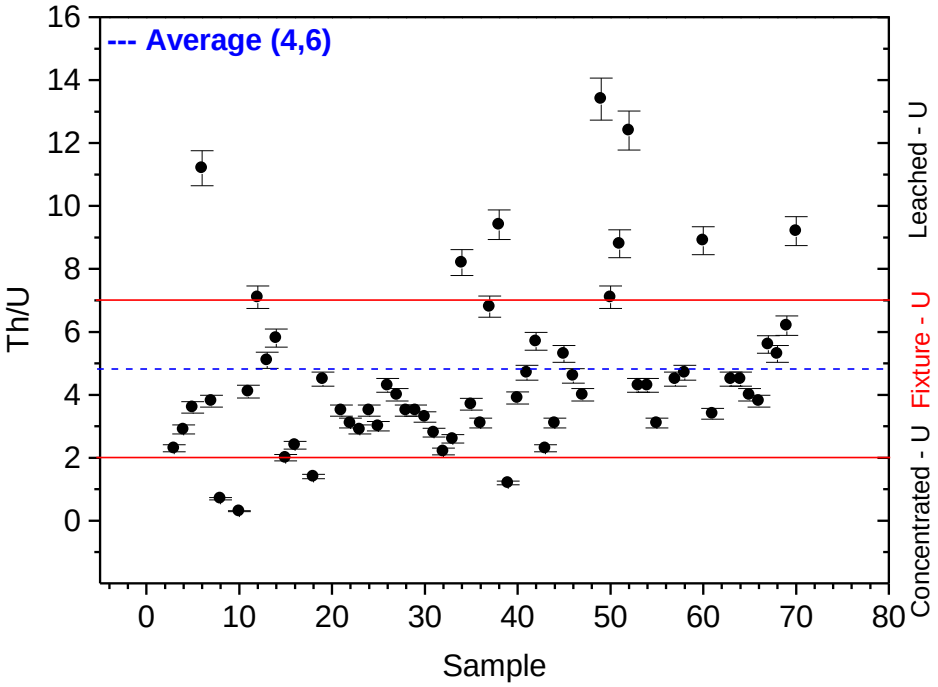

Figure 2. Th/U Ratio Diagram in the studied soil.

In this case, the Th/U ratio has varied from 0.3 to 13.4, with average of 4.6. The bigger ^{238}U fraction in the soil is fixed (approximately 80.3%), characterizing low mobility; the smallest fraction, concentrated (6.6%) and a lixiviated intermediary fraction (13.1%).

The pH measures made for the soil samples have varied from 4.8 to 8.7, with average of 7.2 ± 0.8, with higher frequency predominantly for the values between 6 and 8, which characterizes an oxi-reducer environment.

According to the analyzed tests, there are no direct correlation evidences between the found activities and the pH associated to the elements of the U and of the Th series, however, mineralogical analyses made in anomalous rocks in uranium (cálcio-silicáticas anfibolíticas), have allowed the identification of the presence of maguemite (Fe_2O_3) and of uranium oxids amongst them the UO_2, UO_3 e o U_3O_8 [12], which justifies uranium concentration in the soil with low mobility, due to the presence of maguemite which makes the environment reducer.

4. CONCLUSIONS

According to the Th/U analyzes it was possible to conclude that the major part of uranium in the soil, approximately 80% of the analyzed samples presented low mobility.

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