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Research Article

Evaluation of Radioactivity In Tailings From Niobium Mining

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ABSTRACT:
Background: This study evaluates the radioactivity of niobium tailings based on barite (Light sample) and magnetite separation (Dark sample) collected from the exploitation of niobium in the city of Araxa (MG), Brazil.
Purpose: The investigation is part of a study aimed at using mineral waste as raw materials for the production of cement mortar in the construction industry.
Methods: First, the samples were monitored with a GM-type detector. Subsequently, gamma spectrometric analysis with a NaI(Tl) detector revealed the presence of ^{214}Bi and ^{208}Tl above background radiation in both samples.
Results: Using an Ge(HP) detector, the concentrations of ^{226}Ra and ^{228}Ra were measured in the Light and Dark samples. The results were 0.24 ± 0.004 and 0.80 ± 0.003 Bq/g and 0.17 ± 0.01 and 0.87 ± 0.01 Bq/g respectively. The results of the nuclear activity analysis show uranium concentration of $51\mu g/g$ and $12 \pm 2\mu g/g$ and a thorium concentration of 70 ± 1 and $137 \pm 2\mu g/g$ for the Light and Dark samples, respectively.
Conclusions: The results showed that the niobium tailings from the Araxa mine do not represent a radioactivity risk of exposure dose to the population according to the reference standard CNEN NN3.01, 2014 of the National Nuclear Energy Commission. Nevertheless, the presence of ^{226}Ra should be investigated, as ^{226}Ra is a source of radon gas exhalation, which may be a risk factor in the use of these mineral tailings as construction materials.

KEYWORDS: Araxa niobium exploitation, Radioactive contamination study, Radioactive waste management, Niobium tailings characterization, Federal University of Minas Gerais, Belo Horizonte

1. Introduction

Like other mining countries, Brazil is struggling with tailing disposal, which requires appropriate technological solutions in order to mitigate the environmental and social problems [1]. Social responsibility and sustainability are constant concerns of all productive sectors. Mineral tailings have potential application in building industry, especially in the production of concrete and mortar [2]-[4]. This alternative results in cost reduction, as well as offers an environmentally sustainable solution in the use of industrial byproducts and reduction in the exploitation of natural resources to produce conventional aggregates and cements [5] [6]. However, when sterile rock sediments or sediments are manipulated by industrial processes, natural radiation can be exposed from radionuclides in explored geological formations. They must be analyzed in the light of normative limits regarding radioprotection [7]. The isotopes that mostly contribute to natural radiation are radio nuclide series of ^{235}U, ^{238}U, ^{232}Th, and ^{40}K [8]. Measuring radiation exposure using appropriate equipment that can detect each type of radiation is essential for precautionary purposes and exposure control, thus reducing risk. The objective of this study is to quantify the radioactivity of niobium tailings collected from an exploration area in the city of Araxa in the Brazilian state of Minas Gerais. Niobium mining plays an expressive role in

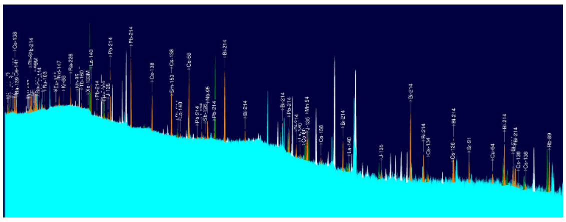

Figure 1: Dark sample resulted obtained by the Ge(HP) detector.

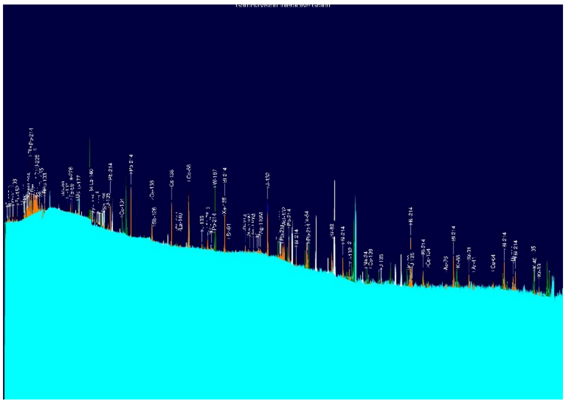

Figure 2: Light sample resulted obtained by the Ge(HP) detector.

Brazil, as the country holds around 95% of all deposits in operation in the world [9]. The carbonatitic complexes are the greatest production potential, followed by pegmatites associated with granitic magma. Reserves of carbonatitic complexes are mined in pyrochlore rocks $[(Na, Ca)2Nb_2O_6(OH, F)]$, and reserves of pegmatite complexes are mined in columbite-tantalite rocks $[(Fe, Mn)(Nb, Ta)_2O_6]$ and rapakivi granites. In addition, niobium also occurs in alkaline carbonatitic complexes, which are rocks formed by magmatic crystallization [9]. Alves and Coutinho [10] verified that the niobium production wastes contain radionuclides, especially ^{238}U, ^{226}Ra, ^{210}Pb, ^{232}Th and ^{228}Ra. The analysis aims to evaluate the potential radioactivity risk for occupationally exposed person or the general public using niobium tailings as raw material for the construction industry compared to radiation protection standards [11].

2. Method & Materials

Two samples were provided by the niobium mining industry located in the town of Araxa (MG, Brazil). The geological formation from where the samples were obtained has a diameter of 5 km [11]. The two tailings analyzed were named "Light Sample" (obtained from flotation stages), and "Dark Sample" (from the magnetic separation stages) [12]. The main mineral compositions of both samples are barite and magnetite/hematite, respectively. They were collected by a private company and transported in sealed containers to Laboratory of Characterization of Construction and Mechanical Materials at the Federal University of Minas Gerais-DEMC/UFMG, in Belo Horizonte, MG, Brazil.

The two samples (Light and Dark) were analyzed with gas detectors type Geiger-Muller to check for the pres-ence of natural radioactivity in the Tailings. The analysis was carried out using surface radioactivity detectors, CE model RDS 80, with a sensitivity of 1 to 100000cps / 0.01 to $100kBq/cm^2$ and the exposure rate meter, CE brand model RDS 30 with a sensitivity of $0.01\mu Sv/h$ to $100mSv/h$ for photons from $48keV$ to $1.3MeV$. The measurements were made at a fixed distance of 10 cm from the surface of the two samples (Table 1). The analyses with Geiger-Muller type detectors showed the presence of radioactivity in the two samples. To investigate the gamma energies emitted from the radionuclides found in the samples, the samples were placed in small plastic bags separately as Light and Dark samples and taken to the Internal Dosimetry Laboratory (LDI) in the CDTN/CNEN. The samples were analyzed using the gamma spectrometry system using the model 802. 3"x 3" sodium iodide crystal detector (NaI(Tl)) and an OSPREY multichannel analyzer connected to a computer. Each sample was placed close to the main entrance window of the NaI(Tl) detector and left there for 10 hours of monitoring. The Genie 2000 software of Canberra was used to obtain the two gamma spectra result from the Light and Dark samples. The results were published at BJRS [11]. Using the Neutron Activity Analyze (NAA) method, $1g$ of each sample passing in a #150 μm mesh was placed in the equipment. The samples were exposed to a neutron flux for 3 hours at the TRIGA Reactor at $UQNR - LAN$ Laboratory from $CDTN/CNEN$. The activated samples were analyzed

Table 1: Results obtained with a GM detector [11]

Dark sample		Light sample	
Pulse (cps)	**Dose (μ Sv/h)**	**Pulse (cps)**	**Dose ($\mu Sv/h$)**
4.00	0.42	3.00	0.22
3.00	0.28	3.00	0.27
3.00	0.3	3.00	0.22
3.33±0.47	0.33±0.06	3.00±0.00	0.24±0.02

Table 2: Results from Ge (HP) analyzes.

Radio-226 (Bq/g)		Radio-228 (Bq/g)	
Light sample	**Dark sample**	**Light sample**	**Dark sample**
0,24	0,18	0,84	0,85
0,23	0,17	0,64	0,92
0,23	0,16	0,82	0,79
0,27	0,19	0,92	0,90
0,24	0,17	0,78	0,86
0.24±0.004	0.17±0.004	0.80±0.008	0.87±0.008

with a Canberra gamma spectrometry system using a coaxial Ge(HP) detector model 5019 (HP) with 50% nominal efficiency, $DSA-2000$ coupled to a micro-computer with a Multichannel Spectrum Acquisition Board and the $Genie2000$ program at the Nuclear Spectrometry $Laboratory/CDTN/CNEN$ (Table 3). The spectrum resulted obtained with the NaI(Tl) detec-tor showed photopeaks, above the background, of energies characteristics of some radionuclides, although the low resolution of the NaI(Tl) detectors, the samples were taken and analyzed using the Ge(HP) from the Nuclear Spectrometry Laboratory (NSL) of CDTN/CNEN which has a higher resolution and allows to identify and quantify the isotopes of radium 226 and 228 [12]. The 14.65 g of Light sample and 16.64 g of Dark sample were analyzed for approximately 24 hours. The chemical and mineralogical analysis were carried out by the supplier, using X-Ray Fluorescence (FRX) [13].

3. Results and Discussions

The results of the gas detector are shown in Table 1. An average exposure dose of 0.24 $\mu Sv/h \pm 0.02$ and $0.33 \mu Sv/h \pm 0.06$ were estimated for Light and Dark sam-ples, respectively, as described in Table 1. The spectra results showed the gamma-ray photopeak energies identifying the energy characteristic of ^{214}Bi(609 keV), ^{40}K(1460 keV) and ^{208}Tl (2614 keV) [11]. The results indicated the difference of counts per channel of radiation energy between the detection of background radiation and radiation emitted by the samples. The measurements obtained with HPGe detector allowed us to observe in the spectra result, the presence of different radionuclides, Figures 1 and 2. The amount of ^{226}Ra and ^{228}Ra obtained with the Ge(HP) analysis is shown in Table 2 for both samples. The estimated average concentration is shown in Table 2. The results obtained using the NAA techniques showed concentrations of uranium and thorium, in both samples. The results are showed in Table 3. The chemical composition of both samples provided by the niobium mining industry are showing in Table 4.

Using the results of the Geiger-Muller detectors, it is possible to estimate the risk of exposure dose from the samples. In a hypothetical calculation for 2000 working hours per year, radiation dose is about $0.48 mSv$ (for Light Sample) and $0.66 mSv$ (for Dark Sample) per year. According to the radiation protection standard, the radiation dose must be below $1 mSv.y^{-1}$ (for general public) and $20 mSv.y^{-1}$ (for workers) for an environment to be considered radiation-free [14]-[15]. The Ge (HP) gamma spectrometry analysis confirmed the peaks found in the previous analysis with NaI(Tl)

Table 3: Results from NAA analyzes

	Uranium (μg/g)	**Thoron (μg/g)**
Light sample	5±1	70±1
Dark sample	12±2	137±1

Table 4: Chemical composition of the tailing from niobium exploration.

Oxide composition analysis (FRX)		
Oxide/Elements	**Tailing samples**	**Tailing samples**
	Light (%)	**Dark (%)**
BaO	77.24	1.08
Fe_2O_3	2.13	91.83
TiO_2	0.45	3.08
Nd_2O_3	0.4	-
SiO_2	0.39	0.2
Pr_6O_{11}	<0.38	-
La_2O_3	<	-
Al_2O_3	0.32	0.07
Nb_2O_5	0.18	0.66
ThO_2	0.11	-
MnO	0.05	-
U_3O_8	0.05	-
CeO_2	0.04	-
CaO	0.03	-
Ta_2O_5	-	<0.10
PbO	-	0.04

Source: Supplier.

detector, indicating the presence of ^{226}Ra and ^{228}Ra. In the process of geological changes and sedimentation, radium has an affinity for some elements, including barium [16]. Neutron Activity Analysis (NAA) was performed to determine the influence of uranium and thorium in the results of the Ge(HP) detector analyses. According to FRX analyses, has a much higher BaO content in the barite based sample (Light) than in the magnetite-based sample (Dark).

4. Conclusion

In his study the quantification the radioactivity of niobium tailings, collected from an exploration area in Araxa (MG, Brazil) was performed. The niobium mining plays an expressive role in Brazil, as the country holds around 95% of all deposits in operation in the world. The analysis aims to evaluate the potential radioactivity risk for occupationally exposed person or the public using niobium tailings as raw material for the construction industry compared to radiation protection standards. The Geiger-Muller detector was used to indicate the presence of radioactivity from gamma radiation in the waste, the results showed no risk of radioactivity to occupationally exposed persons or the public. The concentration of thorium and uranium detected after the NAA analysis was responsible for ^{226}Ra and ^{228}Ra quantified with the Ge(HP) detector. Although the occurrence of uranium and thorium is greater in the Dark sample (hematite/magnetite) than in the light sample, the activity of ^{226}Ra is greater in the light sample (based on barite) than in the dark sample. This can be explained by the affinity of ^{226}Ra for barium, which is confirmed by the XRF analysis. Assuming that the waste is used as a raw material to produce mortar and concrete, the ^{226}Ra present in the waste may pose a health risk as ^{226}Ra is a source of radon gas. Additional studies using alpha detectors should be conducted to understand the exhalation rate of radon from the waste and the materials produced from this waste.

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Authorship contribution

E. J. D Soares (performed the experiments and measurements, discussion results), F. C. R. Almeida (supplier of tailings and chemical analysis), B. M. Mendes (performed the spectrometric analysis), R. G. Passos (discussion results and review the text) T. C. F. Fonseca (discussion results and review the text).

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Conflict of interest

This article has no conflict of interest and the authors have non-financial interest to disclose.

Declaration

This research has been conducted ethically, reporting of those involved in this article.

Similarity Index

I hereby confirm that there is no similarity index in ab-stract and conclusion while overall is less than 5% where individual source contribution is 2% or less than it.

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