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Assessment of the use of tailings based on the legal requirements for radiation protection, from niobium mining in Minas Gerais – Brazil

Soares[3] E. J. D.; Santos[1] T. O.; Mazzaro[2] F. S.; Almeida[2] F. C. R.; Mendes[4] B. M.; Gomes[4] R. P.; Fonseca[3] T. C. F.

[1] Department of Anatomy and Medical Imaging, Federal University of Minas Gerais, 31270-901, BH MG Brazil

[2] Department. of Materials and Construction Engineering, Federal University of Minas Gerais, 31270-901, BH MG Brazil [3]Department of Nuclear Engineering Federal University of Minas Gerais, 31270-901, BH MG Brazil

[4] Nuclear Technology Development Center / CNEN, 31270-901, BH MG Brazil

ejdsoares@hotmail.com

ABSTRACT

Brazil is the world's largest supplier of niobium to industry, accounting for 98% of world production, with Minas Gerais supplying 80% of total production. The mineral exploration industry generates millions of tons of waste annually. In several mining industries, waste is considered a burden for companies. Based on the radiation protection exemptions for the disposal of mining waste, the study analyses the use of waste as a raw material for the construction industry. The minimum dose rate found for gamma radiation in the waste was 0.24 µSv/h and a maximum dose of 0.33 µSv/h, which corresponds to an annual dose above the population exposure limit. The radio concentrations from gamma spectrometric analyses with the Ge(HP) detector for the two samples are a maximum of 240 Bq/kg for Ra-226 and a maximum of 840 Bq/kg for Ra-228. Despite the dose values determined for gamma radiation, CNEN Resolution 179 of 2014 considers materials with natural radioactive concentrations of radium 226 and 228 of up to 1000 Bq/kg suitable for use in the cement industry. Nevertheless, further analysis must be carried out. Since the tailings contain a concentration of Ra-226 and the radio is a source of radon gas, new analyses need to be carried out targeting the exhalation of radon.

Keywords: **Natural radioactivity; Mining tailings; Niobium.**

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1. INTRODUCTION

Social responsibility and sustainability are constant concerns of managers in all productive sectors. The products derived from mining are present in the daily life and are considered the basis of the industrial production. However, the mining sector is also considered one of that change the environment the most [1].

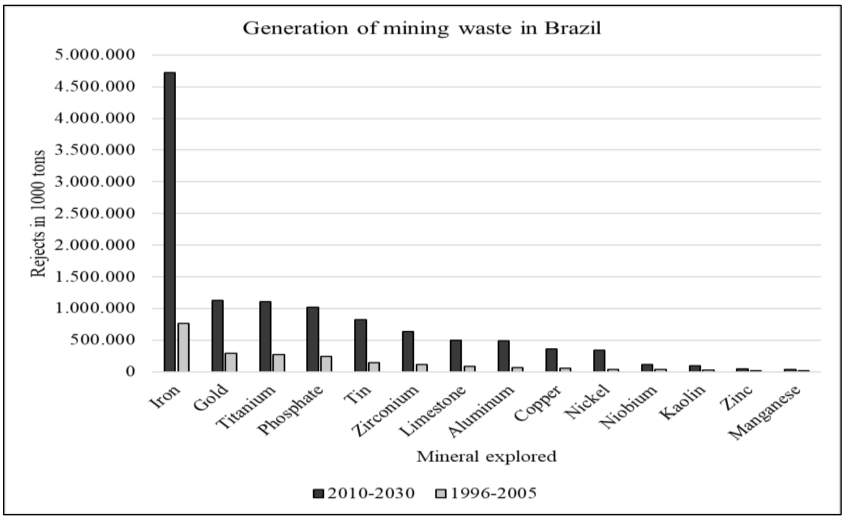
The use of tailing and discarded materials from the mineral extraction has been seen as a possible way in the discovery of new material resources [2]. These residues can be widely used for construction materials, especially for the production of concrete and mortar [3–6]. This alternative results in cost reduction and offers an environmentally sustainable solution in the use of industrial by-products and reduction in the exploration of natural resources to obtain conventional aggregates and cements [7,8]. However, the use of sterile rock sediments or those manipulated by industrial processes are evaluated for use, exposures to natural radiation resulting from radionuclides present in explored rock formations must be analyzed with respect to the normative limits for radiation protection [9].

Alves and Coutinho (2019) have demonstrated that the wastes from niobium production resulting from the magnetic separation, flotation and desliming stages may contain some of these radionuclides, especially ^{238}U, ^{226}Ra,^{210}Pb, ^{232}Th and ^{228}Ra [10] .By conducting radiometric analyses of the waste generated by niobium mining in the city of Araxá (MG), the study aims to evaluate the use of this waste as a raw material for the construction industry based on radiation protection exemption standards for waste disposal of mining.

1.1 Mining waste

Brazil, as other countries, is struggling with tailings disposal, which requires appropriate technological solutions [11]. A significant increase in mineral activity is forecast as shown by the Institute of Applied Economic Research (IPEA) on the production of overburden from the fourteen main minerals explored (Figure 1) [16].

Figure 1 - *Forecast of tailings generation from fourteen main ores explored in Brazil*



Source: IPEA 2012 modified

The amount of soil material to be removed and processed in the mining exploration depends on the type of rock formation, the mineral being explored and in the concentration in the rock being explored, beside the depth the rock from the surface. Such solid wastes are divided into sterile wastes and overburden. Sterile wastes are materials generated during excavation, and mining, that have no economic value. Tailings are residues resulting from the industrial processing of the mined ore [12,13]. As a result of management and changes in geological structure caused by mineralogical activities, the resulting residues that exhibit natural radiation are defined as Naturally Occurring Radioactive Material (NORM). The processing of the waste to separate the exploited ore can significantly change the concentration of radionuclides. These materials are referred to as Technologically Enhanced Naturally Occurring Radioactive Material (TENORM) [14].

1.2 Niobium exploration

In Brazil, most niobium ore occur in two geologic settings, with carbonate complexes having the greatest production potential, followed by pegmatites associated with granitic magma. Reserves of carbonate 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 carbonate complexes, which are rocks formed by magmatic crystallization. Table 1 shows mineral origin by ore in the four main producing states of Brazil: Minas Gerais (MG), Goiás (GO), Amazonas (AM) and Rondônia (RO). The highest concentrations of niobium deposits are located in regions of carbonate complexes, in the state of Minas Gerais [1].

Table 1 - Niobium production from main producing states of Brazil, in 2015.

Substance	**Quantity (*ton*)**	**Mineral amount (*ton*)**	**Average Nb_2O_5 content (%)**
MG- Pyrochlorine	132.501	71.453	53.93
GO -Pyrochlorine	20.208	10.578	52.35
AM- Columbite Tantalite	16.815	1.681	10.00
RO - Columbite Tantalite	5.160	477	9.24
Total	174.683	84.189	48.20

Source : modified - Mineral resources of Minas Gerais: Niobium [1]

1.3 Regulations

In Brazil, radioactivity parameters for the use, disposal, transport and storage of naturally radioactive material are set by the National Nuclear Energy Commission (CNEN).

The standard CNEN NN 4.01 aims to establish the safety and radiation protection requirements for mining industrial installations of materials containing radionuclides from the natural series of uranium and/or thorium [15]. CNEN NN 8.01, on radioactive waste management, which in Art. 35 provides for the unconditional exemption of large quantities of materials superficially contaminated with radionuclides from the natural series [16]. CNEN NN 3.01 defines the basic guidelines for radiation protection, with the limit for exposure of the public being an annual radiation dose of 1mSv [17]. Regulatory position 3.01/001:2001 sets parameters for solid waste management based on the radioactive activities of the natural radionuclides present [18].

The Economic Cooperation and Development / Nuclear Energy Agency (OECD/ NEA) has carried out a study on the permissible radioactive limits for building materials in countries member of the group and has determined that for an absorbed dose of no more than 1 mSv/year [19], the material must not have an activity > 370Bq/kg in radio equivalence. In Brazil, according to CNEN Resolution 179 of 2014, materials with natural radioactive concentrations of radium 226 and 228 of up to

1000Bq/kg of material are considered suitable for use in the cement industry [20]. Table 2 summarizes some guidelines and standards that regulate the disposal and use of industrial waste in terms of radioactive activity content.

Table 2 - Standards and guidelines for natural radioactivity limits from building materials.

Standards	Description	Defined Limits
NEA-OECD [21] [19]	building material	Radioactive concentration in radio equivalence ≤ 370Bq/kg
CNEN 8.01 [16]	Solid waste dispensation	Annex VI, Dispense Levels for Solid Materials for Various Nuclides. Surface radioactivity ≤ 3kBq/m^2
CNEN 3.01 [17]	Public exposure	Annual dose ≤ 1mSv
CNEN 3.01 PR 001:2011 [18]	Disposal of mining tailings, without restriction	$\frac{\sum activities\ of\ radionuclides}{\sum reference\ values\ per\ element} \leq 1$
CNEN Resolution 179/14 [20]	Material for use in the cement industry	Ra-226 e Ra-228 ≤ 1000Bq/kg
CNEN 3.01 PR 3.01/007 [22][23]	Radon gas in poorly ventilated areas	Dose<10mSv/year, or concentration ≤ 300Bq/m^3

Source: CNEN and NEA/OECD

2. MATERIAL AND METHODS

The Department of Nuclear Engineering (DEN), the Centre for Development and Nuclear Technology/ National Nuclear Energy Commission (CDTN/CNEN) and the Civil Engineering Department (DEC) collaborated on this study. The samples were provided by DEC. The samples are the subject of feasibility studies on the use of waste from niobium mining as a raw material for mortar production as part of research carried out at DEC. The place where the waste examined by DEC was collected was chosen by the niobium mining industry, which provided the samples. The DEC investigates the physical and chemical properties of mortars produced by adding waste to the composition. This study, carried out in collaboration with DEC, aims to investigate the radiological properties of the wastes used in the production of mortars. The wastes studied come from flotation separation rich in barite, called “Light sample", and magnetic separation rich in iron, called “Dark sample", from industrial processing in the extraction of iron-niobium alloys. The mining industry is located in the city of

Araxá, in the state of Minas Gerais, BR. The site of origin of samples "Light" and "Dark", here assessed, is shown in Figure 2.

Figure 2 - *The site of origin of samples*

First, the samples were analyzed with gas detectors, (Figure 3) to check for the presence of natural radioactivity in the wastes. The analysis was carried out using surface radioactivity detectors, CE, model RDS 80 (A), with a sensitivity of 1 to 100 000cps / 0.01 to 100 kBq/cm2 and the exposure rate meter, CE brand model RDS 30 (B) with a sensitivity of 0.01 µSv/h to 100 mSv/h for photons from 48keV to 1.3MeV. The measurements were made at a fixed distance of 10 cm from the surface of the samples, Figure 4.

Figure 3 - *Geiger-Muller detectors used to measure natural radioactivity*

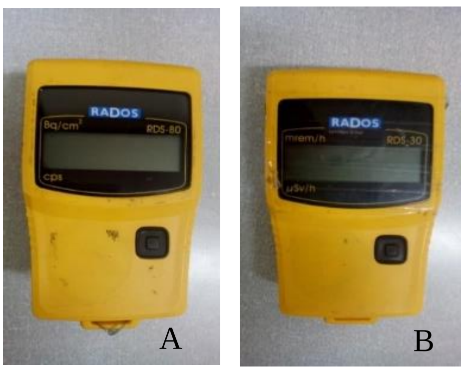
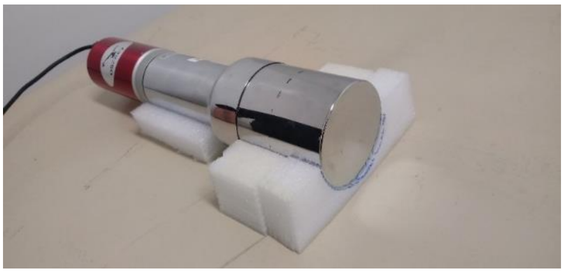


Figure 4 - *Detection of surface activities and measurement of dose rate*

Since the analyses with Geiger-Muller type detectors showed the presence of activity in the sample, it was decided to investigate the gamma energies emitted by the sample. The samples were analyzed by gamma spectrometry using a sodium iodide crystal detector with gamma ray absorption in the internal dosimetry laboratory (LDI) at the CDTN/CNEN. Samples were placed in plastic bags (Figure 2) and left in direct contact with the detector for 10 hours. Was used a Canberra detector model 802, 3"x3" crystal (Figure 5), with an OSPREY multichannel analyzer and the Genie 2000 program.

Figure 5 - *Gamma Spectrometer – NaI(Tl) Detector*



After the results with the NaI(Tl) detector, due to the occurrence of energies above the background energy for some radionuclides, the samples were analyzed again by gamma spectrometry in a detector with higher resolution to quantify the isotopes of radium 226 and 228 [24]. The gamma detector used was Ge(HP) from the Nuclear Spectrometry Laboratory / CDTN / CNEN. The samples were exposed to Ge(HP) for about 24 hours. The system used was the Canberra gamma spectrometry system with a coaxial Ge(HP) detector model 5019 (HP) with 50% nominal efficiency, DSA-2000 (Digital Signal

Analyzer) coupled to a microcomputer with a Multichannel Spectrum Acquisition Board and the Genie2000 program. The Ge (HP) detector system as in the Figure 6.

Figure 6: *Gamma Spectrometer – Ge (HP)*

Source: https://www.gov.br/cdtn/pt-br/laboratorios/laboratorios-de-analitica-e-radioquimica/laboratorio-de-espectrometria-nuclear

3. RESULTS AND DISCUSSION

The results found with the gas detectors are described in Table 3. The values correspond to the arithmetic mean of three measurements for each analysis, pulse number (cps) and dose rate (µSv/h) per sample and the standard deviations calculated around each mean value.

Table 3 - Results of surface radiation monitoring and dose rate.

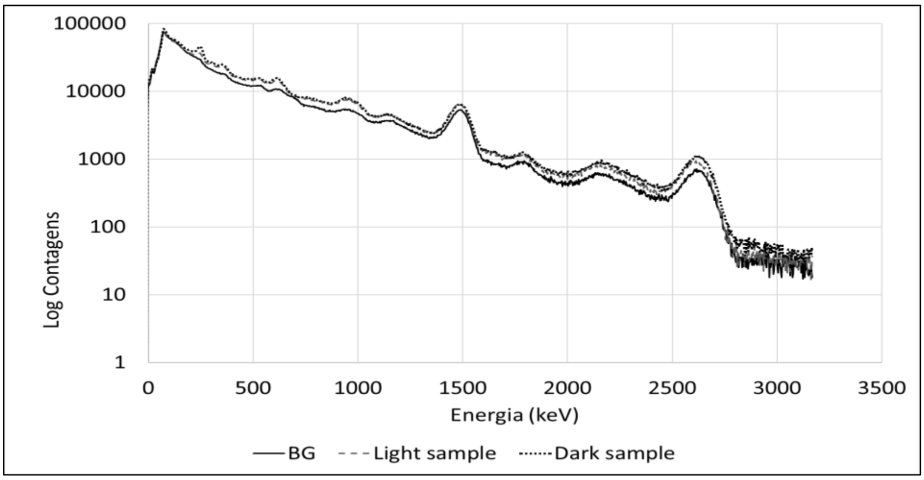
Sample	Surface radiation (**cps**)	Dose rate **(µSv/h)**
Dark	3.33 ±0.47	0.33±0.06
Light	3.00±0.01	0.24±0.02
Background (BG)	1	0.09

Source: author

Palácio and Napolitano (1979), who performed radiometric analysis in a niobium exploration plant in the city of Catalão in state of Goias, BR, found the minimum value of dose rate of 5 μSv/h in the exploitation area and the maximum value of 15 μSv /h in the barite tailings basin and slag deposit [25]. The tailings from niobium exploration in the city of Araxá have values that are less than one tenth of the minimum value found in the industrial mining site in Catalão. Niobium mining in Catalão (GO) takes place in the same rock formations explored in the city of Araxá (MG), as shown in Table 1, where pyrochlore is mined to extract niobium [1].

Using the IAEA and ICRU established residence time of 7000 h/year [23,26] as the parameter to calculate the dose/year for the results in Table 3. The annual exposure dose for the dark sample is 2.33 mSv/year and 1.68 mSv/year for the light. The values obtained are less than 2.4 mSv/year when taking into account the average radiation dose rate of the world population as reported by UNSCEAR in their report [27]. However, the values would be above 1 mSv/year if the annual dose limit for the population defined in the standard CNEN NN 3.01 is taken into account [17]. Figure 7 shows the diagram created from the results of the Na(Tl) gamma spectrometer analysis of the counts per energy channel.

Figure 7 - *Gamma ray emission from samples, obtained by NaI(Tl) detector*



The graph shows the difference between the detection of background radiation and the radiation emitted by the samples, in terms of counts per channel of radiant energy. Based on the gamma-ray energies found, it is possible to detect activities in the samples in the gamma bands characteristic of

^{214}Bi (609 keV), ^{40}K (1460 keV) and ^{208}Tl (2614 keV), which are above the gamma-ray energies of the surrounding background radiation.

The concentrations of Ra-226 and Ra-228 from gamma spectrometric analyses with the Ge(HP) detector for the two samples are shown in Table 4. The diagram in Figure 8 and Figure 9 shows the quality analysis of the samples in terms of gamma emission energy per second. The analyses of the samples with gamma radiation show that the presence of Ra-226 in the light sample, 0.236±0.012 Bq/g has a greater concentration than in the dark sample, 0.172±0.010 Bq/g.

Table 4 - Concentration of Ra-226 e Ra-228 from HPGe detector analyses.

RADIO-226 (Bq/g)		RADIO-228 (Bq/g)	
Light sample	**Dark sample**	**Light sample**	**Dark sample**
0,239	0,176	0,836	0,852
0,232	0,169	0,639	0,922
0,231	0,155	0,819	0,789
0,272	0,194	0,918	0,900
0,236	0,168	0,779	0,864
0.242±0.012	0,172±0.010	0,798±0.071	0,866±0.036

Source: author

Figure 8 - *Counts per second per channel of the photopeak gamma energy diagram*

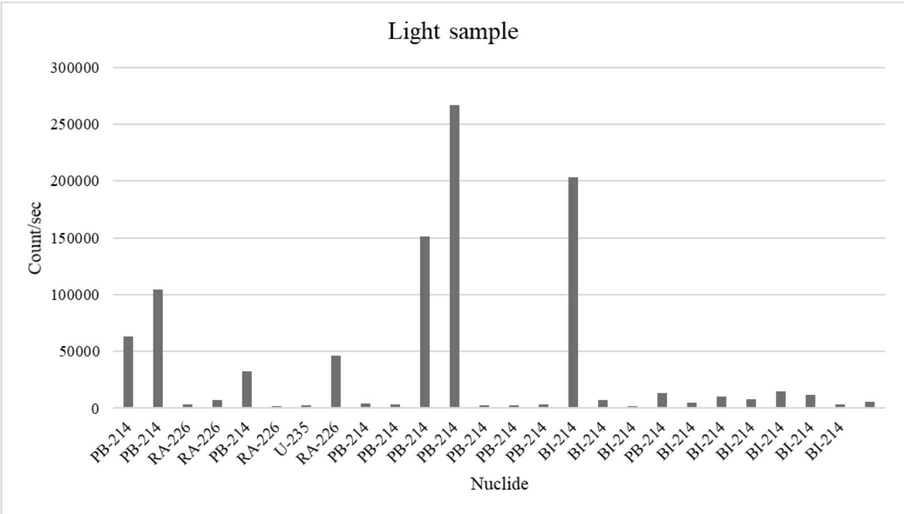
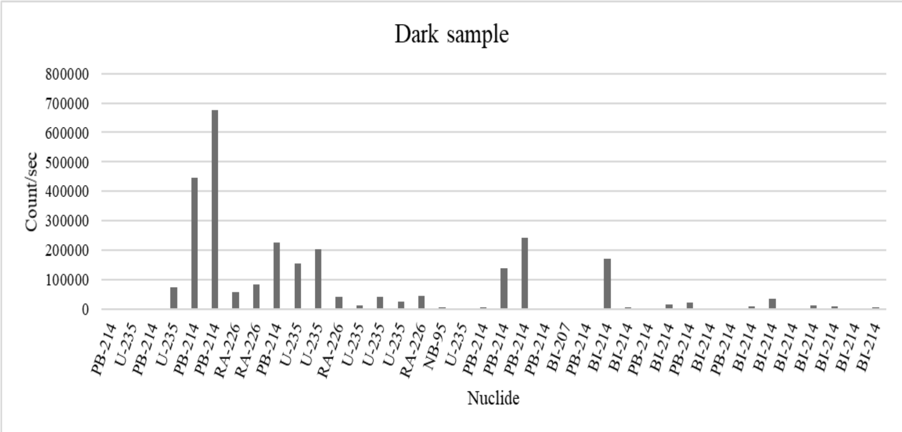


Figure 9 *Counts per second per channel of the photopeak gamma energy diagram*



4. CONCLUSIONS

The use of overburden from niobium mining in Araxá (MG) as a raw material for the construction industry was studied to determine the radioactive concentrations in the overburden and to compare the results with the standards and guidelines that set the limits for natural radioactivity in raw materials and overburden for the construction industry.

Compared to the annual exposure dose of 1mSv/year limited by CNEN NN 3.01, the overburden material will produce a higher exposure dose. Taking into account Resolution 179/14, also from CNEN, which sets a limit of 1000 Bq/kg for radioactivity concentration for raw material for the cement industry, and according to NEA-OECD, which sets a limit of 370 Bq/kg for Ra-226 concentration, the overburden material has a much lower Ra concentration.

The results allow to use the tailing in the context of radiation protection in the construction industry. Nevertheless, further analyses need to be carried out, and the exhalation rate of radon gas from the tailings as a target since there is a considered Ra-226 concentration on the tailings.

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