Gamma-ray spectrometry to recognize mineralized reliquary zones potentially associated with acid mine drainage generation in an uranium mining waste rock pile (Caldas, Brazil)

Anna Carolina Gastmaier Marques, César Augusto Moreira, São Paulo State University (UNESP)

Abstract

Metal mining deeply modifies local environmental dynamics and produces large volumes of waste rock and residues, consequently impacting the soil, water and the ecosystem. In this sense, understanding the complexity of the geological structures is key to preventing degradation. The Osamu Utsumi mine was a pioneer uranium ore exploitation site in Brazil, located in the State of Minas Gerais. The open pit and the high concentration of sulfide in waste rock piles are nowadays a dispendious concern to the Nuclear Industries of Brazil, due to sulfide oxidation and the resultant acid mine drainage (AMD). The objective of this research is to recognize radioactive sources in the waste rock pile, BF-04, of the Osamu Utsumi mine. The processing data mapped uranium, thorium and potassium concentrations in the study area. Potassium concentrations distinguishes zones of rock depletion, lowest concentrations, and radioelement sedimentation. The mineralized uranium ore includes sulfides that are rapidly destroyed by surface hydration conditions, responsible for generating acid mine drainage. In this sense, superficial zones in the waste rock pile with high levels of eU and eTh are areas exposed to oxidation and hydration of sulfides, and, therefore, their recognition and neutralization can contribute to decrease AMD generation in the study site and areas with similar characteristics.

Introduction

The Brazilian Mining Institute (IBRAM) indicates that mineral substances correspond to 14.5% of the country total exportation, represents 52% of exportation trade balance and points to the participation of the mineral extractive sector in 3.7% of Brazil's Gross Domestic Product (GDP) (IBRAM, 2019).

Brazil possesses the world's sixth largest uranium ore reserves, which amounts approximately 310,000t of triuranium octoxide (U_3O_8), recoverable at low cost (Brazil, 2017). All processes concerning uranium and other radioactive elements, such as mining, are national monopoly activities.

Social and environmental impacts of mining must be an integral part of all mine planning and mineral development from the discovery phase through to closure process, regarding the complexity that involves the mined ore. Physically landscape disturbance is a result of mining, including waste rock and tailings disposal areas and facility development. Operations and waste products associated with metal extraction and processing are one of the main causes of environmental concerns about metal mining, which may increase the acidity of soils and consequent degradation of both surface and groundwater quality, as well as increase air-borne dust and other emissions (Hudson, Fox and Plumlee, 1999).

The exposure of natural reactive minerals to superficial environmental conditions can trigger key processes for the generation of highly complex contaminants. Studied across the globe, metal sulfide minerals are present in host rock associated with most types of metal mining activity. In the presence of moisture and atmospheric oxygen, sulfide minerals are oxidized and generates acid mine drainage (AMD). Although the oxidation of these minerals and the formation of sulfuric acid is a function of natural weathering processes, mining activity, such as extraction and beneficiation operations, increase the chemical reactions by exposing large volumes of sulfide rock material with increased surface area. Among others, pyrite, arsenopyrite, chalcopyrite and chalcocite oxidation can occur for several years following mine closure (USEPA, 1994; Akcil and Koldas, 2006).

Followed by the generation of AMD, secondary factors act to either neutralize the acid produced by oxidation of sulfides or may change the effluent character by adding metals ions mobilized by residual acid. The generation of AMD and the potential to release contaminants are dependent on many factors, including specific characteristics found at different locations, such as climate and physical properties of the waste material. In the geological environment, the occurrence of radioelements associated with sulfide rocks under chemically reducing conditions create an environment where uranium and thorium exist in a tetravalent state, a configuration relatively insoluble. However, under oxidizing conditions uranium occurs in hexavalent uranyl ion (UO_2^{2+}), soluble in water and mobilized by AMD (USEPA, 1994; Dentith and Mudge, 2014).

The radiometric method measures naturally occurring radioactivity in the form of gamma-rays, most originated from mineral species containing radioactive isotopes of potassium, uranium and thorium (^{40}K, ^{238}U, ^{235}U, and ^{232}Th). This study consists in the recognition of shallow mineralized reliquary zones in a waste rock pile of Osamu

Utsumi mine, through the geophysical method of gamma-ray spectrometry.

Method

Gamma-ray spectrometry

Gamma-ray spectrometry is a radiometric geophysical passive method which quantifies occurrence of radioactivity through emission of gamma rays of several naturally occurring or artificial radionuclides. Among the group of natural occurrences are those of cosmogenic origin (^{14}C, ^{3}H, ^{36}Cl, etc.) and the ones associated to the geological environment (^{40}K, ^{235}U, ^{238}U, e ^{232}Th), both groups widely known by the acronym NORM (Naturally Occurring Radioactive Materials).

Spectrometry has a distinctive advantage which is the capacity to provide information about the geochemical nature of the radioactive source (Dentith and Mudge, 2014). Spectrometers are sensible to the energies of the radioelements of geological interest, once it can be chosen K, U and Th energy windows to detect energetic ɣ-rays emitted from relevant elements in parts of the energy spectrum where emissions from other elements are weak or rare.

Important in several areas in Geosciences as petrology, hydrochemistry and environment geology (Dentith and Mudge, 2014; Gilmore, 2008; Milson, 2003), ^{235}U, ^{238}U, e ^{232}Th are the first terms of long decay chains, developing several radionuclides until a stable isotope of Pb. On the opposite direction, ^{40}K decays directly to one of his two stable isotopes: ^{40}Ca and ^{40}Ar, this last one through electronic capture with emission of associated gamma-ray of 1461 keV (Chu et al., 1999).

The use of photopics generated by daughter nuclides is a very common approach in gamma-ray spectrometry to determinate the respective primordial parent radionuclide. Thus, the photopeaks of 1765 keV (^{214}Bi) and 2615 keV (^{208}Tl) accounted to estimate concentrations of U and Th, respectively (Erdi-Krausz et al., 2003). When measurement infers indirectly the concentration of the parent, by relating the emission products from daughter element, the concentration of the ɣ-emitting element must be in proportion to the concentration of its radioactive source, meaning the decay series must be in equilibrium (Dentith and Mudge, 2014).

The method can be used through portable or airbone detection systems or profiling probes (Telford, 1990; Ferronsky, 2015). Regarding portable systems, gamma-ray spectrometry, internal memories can store a large quantity of data acquired, which are generally restricted to measurements in the K, U and Th energy windows, and the total (Dentith and Mudge, 2014). Nevertheless, it has some limitations associated with the equipment and environmental conditions: spectral of the detection crystal (scintillometer), topographical irregularities, soil moisture and soil/rock density (Musset and Khan, 2000; Beamish, 2015). Thereby, depth of acquisition is an important factor for both planning and analysis procedures because, according to Taylor et al. (2002), approximately 90% of gamma rays observed on surface are generated only in the superficial 30 cm depth in a relatively dry soil of 1.6 g/cm3.

Gamma-ray spectrometry presents a high applicability in determination of environmental gamma radiation levels, especially for its potential correlation with anthropogenic contamination sources (Dentith and Mudge, 2014; Gilmore, 2008).

Data acquisition and processing

Geophysical acquisition was performed through 281 individual points in 120 seconds each covering the BF-04 most affected area, approximately 28.16 ha. Individual gamma-ray readings were made in direct contact with the surface of the waste rock exposed, without any vegetal cover or soil. The acquisition was concentrated on the average altitude of 1378m, comprising many portions of BF-04, such as the margins of the Consulta Creek, north border and central region of the waste rock pile. Collecting points were disposed approximately along parallel lines according to NE-SW direction, with average spacing of 30m.

The system used was the portable RS-332 Multipurpose Gamma-Ray Spectormeter System from Radiation Solutions INC, presenting high sensibility, thermic protection, easy handling, internal GPS, and a collimator associated to the BGO crystal. This equipment enables registration of energetic intermissions from 30 keV to 3,000 keV, an appropriate energy interval for the recognition of daughters radionuclides of series of ^{232}Th, ^{238}U, and the decay of ^{40}K. Finally, the system automatically converts the unit counting-per-second (cps) in concentrations expressed in parts per million (ppm) for U e Th and percentage (%) for K. The generated data for uranium and thorium are given by the term “equivalent” (eU and eTh) due to indirect estimates base on daughter radionuclides (IAEA, 2003).

After acquiring radiometric data, a processing procedure was performed for generation of maps that reflect radiometric distribution and signature of BF-04 regarding eU, eTh and K.

The 281 collected data points were submitted to an interpolation routine at Oasis Montai platform (Geosoft) in order to generate a bidimensional section of thorium, potassium and uranium concentration values in terms of distance. Due to natural mitigation of gamma rays and receptor capability to recognize energy emitted next to surface, the radiometric data were not deeply processed.

After processing, maps with color-scale or grey-scale are created, representing quantitative contents of elements K (in %), eU and eTh (both in ppm) (Ulbrich et al., 2007). With the processed geophysical data, maps for analysis of radioisotopes are individually prepared.

Results

Geophysical data interpolation allows the acknowledgement of radiometric anomalies on the BF-04 surface. The radioelements uranium, thorium and potassium mobility were described, as well as the

radiometric characteristics of the waste rock pile BF-04 from the Osamu Utsumi mine.

In distinct environment conditions, potassium has a high mobility and geochemistry solubility. In tropical and subtropical climate zones the decomposition of many potassium minerals due to weathering can be clearly observed (Dentith and Mudge, 2014). The elevated annual pluviosity at Poços de Caldas plateau is the main responsible factor for weathering processes, mainly in superficial and exposed potassic rocks, as identified in the BF-04.

Regarding potassium radiometric response, high concentrations can be associated with the following factors: unchanged potassic rocks, potassium precipitation in wetlands and/or adsorption by clay minerals. On the other hand, low concentrations suggest depletion and element mobility due to weathering (Dentith and Mudge, 2014).

Potassium anomaly zones (Figure 1) are observed in north and southeast areas of BF-04, with values between 5.68% and 13.39% (warm colors), while in the central portion is characterized by lower concentrations varying between 1.002% and 4.72% (cold colors).

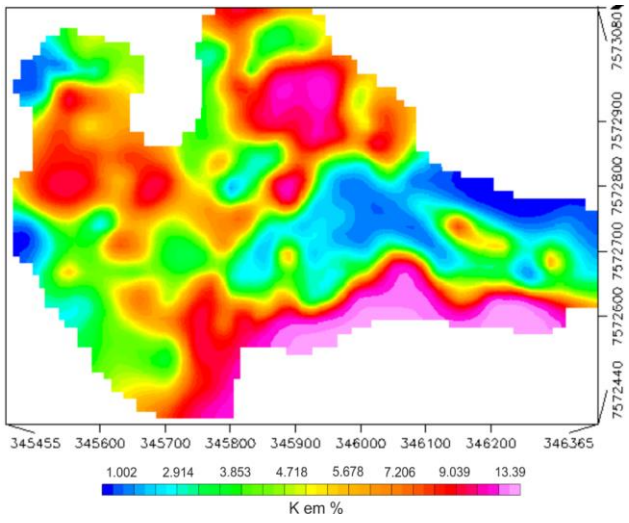

Figure 1 - 2D interpolation map of potassium radiometric response in BF-04

The most important processes associated with uranium and thorium decay are removal, transport, and sedimentation. Uranium (U^{6+}) mobilization occurs in weathering environments and its solubilization is intensified in oxidizing environments due to the formation of the hexavalent uranyl ion (UO_2^{2+}) (Dentith and Mudge, 2014). The main differences between uranium and thorium radioelements mobilization comprise the uranium leaching capacity and thorium detention on organic and inorganic pedogenic substances.

Figure 2 presents a 2D interpolation map of eU and Figure 3 shows the same product for eTh. It is possible to observe similar results in the potassium distribution map, where the highest concentrations of eU and eTh can be found in north and southeast, with eU ranging from 87.76 ppm to 312.61 ppm and eTh between 113.90 ppm and 221.14 ppm.

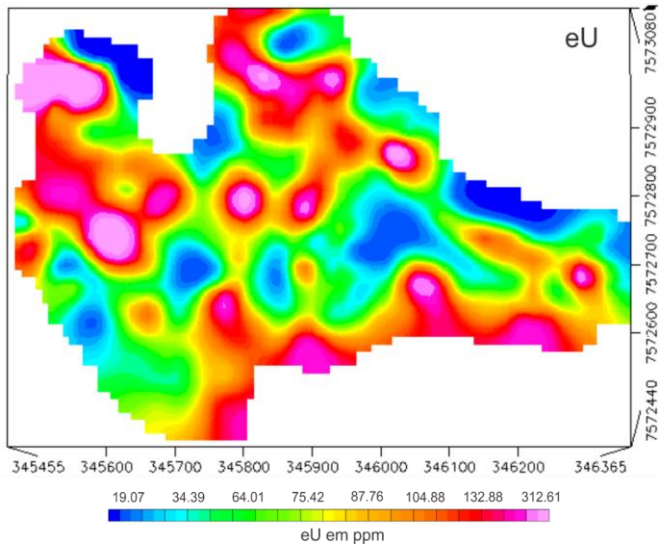

Figure 2 - 2D interpolation map of uranium radiometric response in BF-04

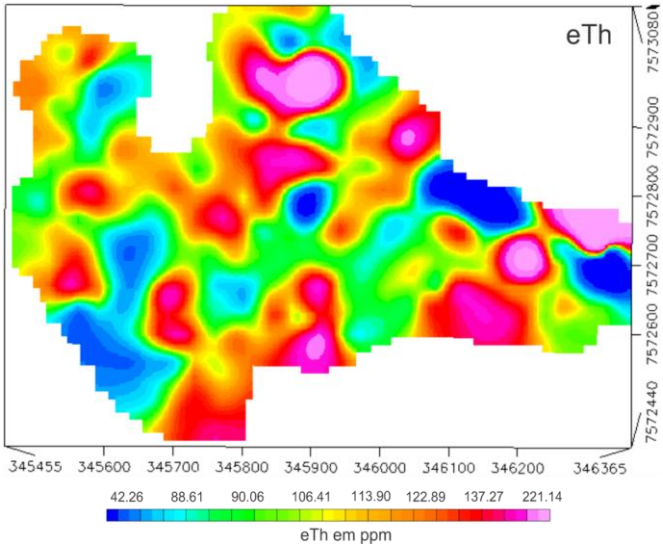

Figure 3 - 2D interpolation map of thorium radiometric response in BF-04

Conclusions

Geophysical investigations have an elevated potential for application in environmental studies, so that the radiometric results provided a very useful shallow analysis of radiometric results in a waster rock pile.

The mineralized uranium ore includes sulfides that are rapidly destroyed by surface hydration conditions and are responsible for generating acid mine drainage. In this sense, superficial zones in the waste rock pile with high levels of eU and eTh are areas exposed to oxidation and hydration of sulfides, and, therefore, their recognition and

neutralization can contribute to decrease AMD generation in the study site and areas with similar characteristics, even indirectly by gamma-ray spectrometry.

Acknowledgments

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