Soil gas compositions as a tool for understanding acid mine drainage formation and air flow in a uranium waste rock pile

Carolina B. Abreu[1], Peter M. Fleming[2], Virginia S. T. Ciminelli[1], Dina L. Lopez[3], Paulo C. H. Rodrigues[2], Otávio E. A. Branco[4]

[1]Department of Metallurgical and Mining Engineering (CPGEM), Fed. University of Minas Gerais, Belo Horizonte, Brazil
[2]Nuclear Technology Development Center (CDTN), National Nuclear Energy Commission (CNEN), Belo Horizonte, Brazil
[3]Department of Geological Science, Ohio University, Athens, USA
[4]Department of Sanitation and Environmental Engineering, Fed. University of Juiz de Fora, Juiz de Fora, Brazil

Abstract The first site of exploitation and processing of uranium ores in Brazil is located at Plateau of Poços de Caldas, in Caldas City, Minas Gerais, which is the largest alkaline Complex in South America. The activities of mining and processing of uranium occurred between 1982 to 1995. Fractures and faults with high permeability act as channels to conduct the underground flow of water and gases. They also determine the main features of the surface drainage network. In geological systems, gases can migrate preferentially through fracture zones. These gases can provide information on the conditions that allow its formation, accumulation and migration. During the mine operation, tailing dams and waste rock piles (WRP) were constructed for the disposal of uranium waste. The Waste Rock Pile 4 (WRP4) was built on the valley of Consulta Stream, an area adjacent to the pit mine. An artificial diversion channel for this stream, about 500m long, was built. Currently, the WRP4 generates acid mine drainage (AMD) with pH of approximately 3.5 and uranium concentration of 8.7mg/L. Despite mitigation actions, the discharge of AMD has increased 60% over the past ten years. It has been shown that waste piles may present regions of recharge and discharge of gases, which influence the oxidation pattern within the pile. In this context, soil gas sampling was carried out, at a depth of 40cm, to establish the concentrations of O_2, N_2, H_2, and CO_2. The results helped to identify the regions of the pile close to the surface where atmospheric air is likely to be entering the pile and regions where air seems to be discharged, after reacting with the sulfides. Understanding the spatial consumption of O_2 due to sulfide oxidation and patterns of gas concentrations and distribution will help the development of remediation approaches.

Key Words Acid mine drainage, fluid flow, waste rock pile, uranium

Introduction

During the oxidation of sulfides present in waste rock piles (WRP), oxygen is consumed and other gases (CO_2, N_2 and H_2) may be enriched and released to the interstitial air. These variations in gas composition within the pile can be used to understand the chemical and physical processes, especially the transport processes. The formation of acid waters is directly affected by the transport of oxygen within the pile. The main objective of this paper is to understand the acid mine drainage formation by means of an approach based on the gas distribution in the superficial layer of a uranium waste rock pile. The pile under investigation, waste rock pile 4 (WRP4), is located in a former mining site, at Caldas, Minas Gerais, Brazil. The site is located on the premises of Nuclear Industries of Brazil (INB) (Figure 1). The uranium deposit at Poços de Caldas Plateau was discovered in 1970. The mining and treatment of uranium ores started in 1982 and operated until the depletion of the deposit in 1995. The industrial facilities cover an area of approximately 15km^2 and consist of an open pit mine, named Osamu Utsumi, waste rock piles, tailing dams and deactivated plants for physical and chemical treatment of uranium ores, and sulfuric acid production.

The Uranium Mining and Milling Facilities of Poços de Caldas (UMMF) exploited a uranium mine located at a large alkaline intrusion, a 35km-diameter round caldera along its northwest-southeast axis, extending over an area of about 800km^2. The altitude lies between 1300 and 1600m. The Poços de Caldas Plateau is characterized by fractured aquifers, whose recharge areas occur mainly at the points of convergence of coincidence between the drainage and surface fractures. It is believed that neotectonic movements have had great influence and produced significant changes in surface drainage, generating faults that facilitate the movement of water and form channels of rivers (Franklin, 2007). The average annual temperature in the region is approximately 19 °C. The maximum temperature is about 36 °C and minimum around 1 °C. Dry and rainy seasons alternate throughout the year, and 80% of annual rainfall occurs between the months of October and March. Annual precipitation corresponds to about

Figure 1 *Location of Caldas City at Minas Gerais, Brazil (left). Partial view of the Uranium Exploration Site with emphasis on Osamu Utsumi Mine and Waste Rock Pile 4 (WRP4) (right).*

1700mm, with over 120 rainy days each year (Holmes *et al.* 1990; Schroscher & Shea, 1991).

WRP4 contains waste material with estimated uranium grades of 680g/t (Wiikimann, 1998). Having a $12.4x10^6m^3$ volume and a $56.9x10^4m^2$ surface area, the pile was built on the Consulta Stream Valley, which was diverted from its original bed to an artificial channel, about 500 meters long, along the valley. Upstream of the diversion channel, a dam, called Carlaile Basin, was built to regulate the Consulta Stream discharge on the diverted stretch. The original Consulta Valley was previously prepared to receive the pile. Nestor Figueiredo Basin (BNF) was constructed to collect the water discharged from WRP4. The wastewater is treated by adding lime (CaO) or calcium hydroxide ($Ca(OH)_2$) to raise the pH to a range of 8 to 11. In this pH range, most metals and radionuclides are precipitated, promoting significant decrease in the ^{238}U and ^{226}Ra concentration in the effluent. Fernandes & Franklin (2001) estimated that the production of acid drainage could last approximately 600 years.

The end-dumping method used to build the pile promoted the classification of the solid particles; coarser material is located at the toe of the benches while the finer material occupies the regions near the surface. The areas where the coarser material is located may work as areas of gases recharge, while the surface of the pile, where the finer material is typically found, works as discharge areas (Lefebvre, 1995; Kuo & Ritche, 1999; Fala *et al.,* 2005; Doe *et al.,* 2008). The heterogeneity caused by the wide variation in the solid particle size may promote areas with different water content, thus enabling the formation of preferential flow inside the pile. The development of preferential flow is a result of changes in hydraulic conductivity of the material that makes a particular area of the pile be more conductive than the surrounding area. The preferential flow along WRP channels has been documented by several authors (Lopez & Smith, 1995, Smith *et al.,* 1995).

The role played by gases in the formation of acid mine drainage

Studies of concentration profiles of gases and temperature in WRP have revealed the importance of convective transport of air in the unsaturated zone of piles containing reactive materials. At the surface of the pile, barometric pressure, oxygen concentration and temperature are established according to local ambient and atmospheric conditions. Oxygen concentrations are expected to be near the atmosphere (21%) at the surface (Kuo & Ritche, 1999; Lefebvre *et al.,* 2001). The active processes of oxygen transport in pile include barometric pumping, diffusive and thermal convection (Smolensky *et al.,* 1999).

Pressure gradients can be induced by wind flow over the pile. Seasonal variations in atmospheric pressure can also affect the air flow rate into the pile (Kuo & Ritche, 1999). Seasonal variations on temperature stimulate convective process as great as the temperature range between ambient and the inner part of the pile. Smolensky *et al.* (1999) observed that during the winter months thermal convection was favored. Modifications in the rate of gas flow and oxygen concentration were observed by Wels *et al.* (2003) as a response to the pressure changes. Diffusive and thermal convection transports are also directly related to pyrite oxidation, where oxygen is consumed and heat is released. Partial reduction of oxygen concentration due to the consumption of the gas in the oxidation reaction promotes a concentration gradient that drives the gas into the pile through diffusive process. Oxygen concentrations are gradually reduced as the air penetrates the pile. Heat released from pyrite oxidation (exothermic reaction) generates temperature gradients that reduce gas density and form convection cells which promote the heat and fluid flow within the pile. However, the permeability must be sufficiently high to enable the convection cell formation (Lefebvre *et al.,* 2001).

The construction method affects the characteristics of the pile regarding heterogeneity, anisotropy of the medium, as well as particle size segregation that influence the transport of air. Heterogeneity due to the wide variation in particle size can promote areas with different water content within the pile, enabling the existence of preferential path flow. The end-dumping method also creates a steep slope with high vertical permeability, when compared with horizontal permeability, facilitating the air flux in inner regions of the pile (Lahmira *et al.,* 2007). Gas and water vapor flows occur preferentially in regions of coarse grain size. These regions are normally saturated with air and therefore the flow of gases and water vapor is facilitated.

During dry periods, the sulfide minerals will react with the gas phase occupying the pores and interstices. But in view of reduced amounts of water within the pile, precipitation reactions will occur, thus producing a variety of minerals as secondary phases. The precipitates are stored in the pores to be leached out during wet periods. Soil gas distribution can be affected by climatic factors, soil moisture, mobility and solubility of the gas itself. In WRP the gas efflux is dramatically reduced after heavy rainfall due to down flow infiltration front that interferes with gas movement upward. However the impact has a relatively short duration (Hinkle, 1994; Kabwe *et al.,* 2006).

Experimental procedure

Two field campaigns were conducted in May and July 2010 for the sampling of the gases in the pile. A grid, where each cell was 50x25m, was designed to collect gas samples, a total of 170 sampling points. Samples were collected using a glass syringe 20mL, aided by an aluminum tube 50cm long, 8mm in diameter and porous tip. Sampling was done by extracting gas at 40cm depth. The sampling bore holes were made using a drill widia 50cm long and 22mm in diameter. Soil was used to close the opening to reduce atmospheric air contamination. After purging the soil three times, the samples were inserted into a 10mL sterile vacutainer. The vacutainers were sealed with silicon and properly labeled. The samples were kept out of direct sunlight and transported in thermal boxes. From each bore hole two samples were taken. The determination of gas concentrations was carried out via a Quadrupole Mass Spectrometer Omnistar 422.

Results and Discussion

Soil gas concentrations for O_2, CO_2, N_2 and H_2 were obtained and mapped using Geographic Information System (GIS) tools. Maps were plotted using the Natural Neighbor Interpolation Method. This method is based on a spatial interpolation which provides a smoother approximation set of values. The maps for the concentrations of O_2 (Figure 2) and CO_2 (Figure 3) show that O_2 is higher to the south-southeast (SSE) side of the pile and lower towards the west (W), in the direction of the pit lake, suggesting that air is entering the pile throughout the old drainage channels. The concentration of

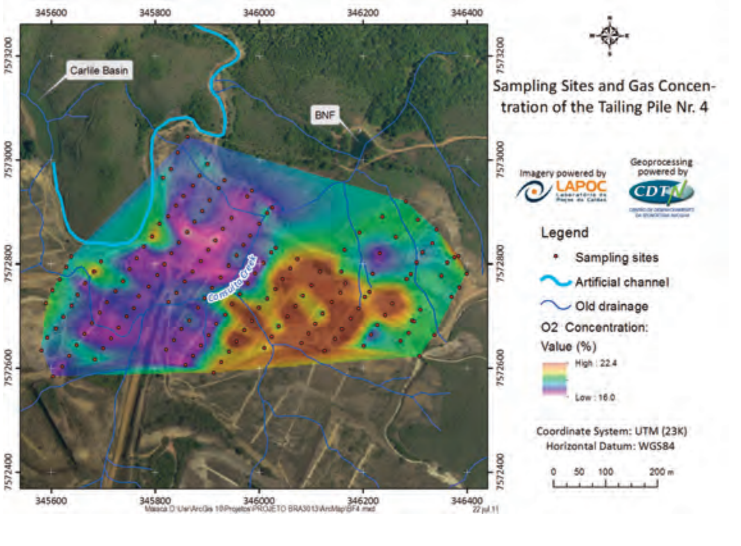

Figure 2 *Gas concentration map for O_2 at WRP4.*

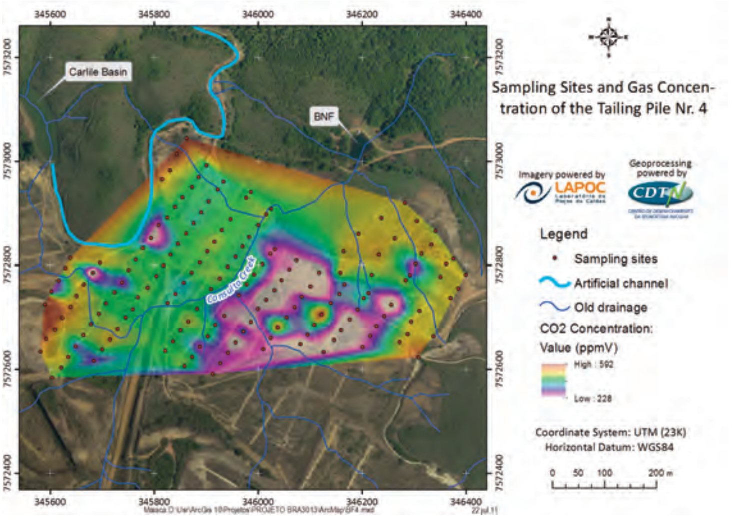

Figure 3 Gas concentration map for CO_2 at WRP4.

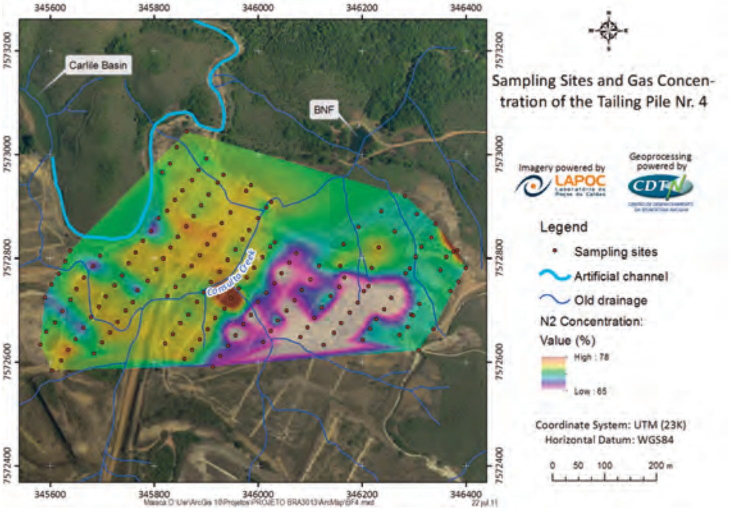

Figure 4 Gas concentration map for N_2 at WRP4.

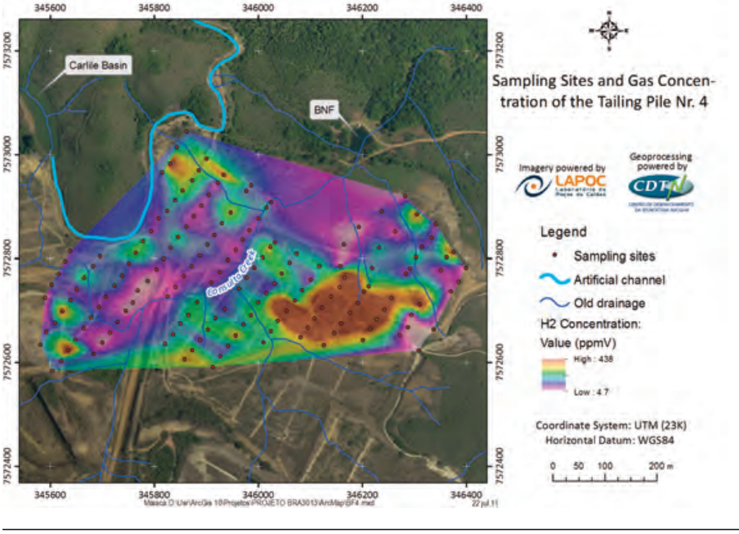

Figure 5 Gas concentration map for H_2 at WRP4.

O_2 is also relatively low at the far end to the east (E) and the concentration of CO_2 is high in that zone. The spatial distribution of N_2 concentration (Figure 4) shows similar results as for CO_2. The slope of the pile towards the north was not sampled due to the high topographic relief. High levels of oxygen may be found there too. The concentration profiles of CO_2 and O_2 can be explained by two phenomena described as follows: i) air is being recharged from the SSE area and possibly along the north slope and it is probably leaving the pile to the W and far E after reacting with the sulfides present in the pile and getting depleted in O_2 and enriched in CO_2; ii) AMD formation at the west side of the pile explains the low O_2 and high CO_2 concentration. O_2 is consumed by sulfide oxidation while CO_2 is produced by microbial respiration, which in turn increases its relative concentration.

The patterns of gas concentrations suggest that air enters the pile along the old drainage from the SSE and throughout the northern slope of the pile, oxidizes the sulfides along the pathway and is discharged from the western section of the pile. The map for H_2 concentrations is similar to O_2 map concentrations; this behavior is due to the fact that H_2 is more stable at reducing conditions (Figure 5). Additional data in the northern slope of the WRP4 pile is needed to reach final conclusions about the way the air phase is moving within the pile.

Conclusion

Gas sampling in waste rock piles can provide very important information about the movement of the gas phase and the sulfide oxidation within the pile. This work has shown that the buried old drainage in waste rock piles continues to be an important source not only for water but also for gases. Fluid flowing along those buried drainages can transport mass throughout open channels. The results helped to identify the regions of the pile close to the surface (40cm), where atmospheric air is likely to be entering the pile, regions with high O_2 and H_2, and low CO_2 and N_2 and regions with low O_2 and H_2, and high CO_2 and N_2, where air seems to be discharged, after reacting with the sulfides. Understanding the spatial consumption of O_2 due to sulfide oxidation and patterns of gas concentrations and distribution will help the development of remediation approaches.

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