

REMEDIATION OF ACID ROCK DRAINAGE AT THE POÇOS DE CALDAS URANIUM MINING SITE – BRAZIL.

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ABSTRACT

Acid Rock Drainage is of great importance to environmental regulators and mine operators in many countries around the world. During the operational of the life the installation, the collect and treat strategy is a commonly employed strategy to reduce pollutant emissions to the environment. Regarding the post-operational scenarios a suite of strategies is available in the literature. Acid drainage is a crucial problem at the uranium mining site of Poços de Caldas. Two waste-rock piles (of about 60 ha each) resulted from mining activities. Presently, acid waters are being collected and neutralized, the solid material being disposed off in the tailings dam. The objective this work was to examine the problem of acid rock drainage generation in one of these piles (Waste-Rock-Pile 4) in order to propose remediation to be implemented based on cost x effectiveness analysis.

Key words: acid mine drainage, uranium mining, remediation, cost x benefit analysis

INTRODUCTION

Open pit mines involve the mobilisation of large amounts of overburden that need to be disposed in waste rock piles. If pyrite is present, acid drainage may result. The overall equation of pyrite oxidation is shown below. The disposal site for the waste rock piles is determined primarily by the convenience of the site to the mining operation, and is not usually based on considerations that will allow a convenient management and remediation. If the mine operator has a fair understanding about how the design of the waste pile may help in the mitigation of acid drainage generation, improved layout and design of waste rock piles may be achieved.



The chemical composition of the acid drainage is controlled by the pyrite oxidation rate, which is a function of temperature, pH, oxygen concentration, composition and amount of infiltrating water and the population of micro-organisms Ritchie (1994) The dimensions of different oxidation and reaction zones in a waste rock pile will depend on the rate that the pile is being oxidised and also on the rate and pathways of water transportation into the pile. The region under oxidation may encompass the whole dump or present a thickness of only few meters. Moreover, due to the heterogeneity of the material forming the pile, these regions may present different sizes in different places in the dump.

Oxygen transport into the dump will be related to three major processes:

1. Transport as dissolved O_2 in the infiltrating water;
2. Diffusion of O_2 gas through rock pore spaces and;
3. Advective transport of O_2 gas through rock pore spaces

Because leach solutions cannot carry significant oxidant with them as they move through the dump, air is the main source of oxidant. Diffusive transport is the major process of oxygen transport in the vast majority of cases. Oxygen removed from the dump pore space in the process of pyrite oxidation will establish a concentration gradient in the pore space. Despite the fact that gas removal will set up a pressure gradient, the gradient will be rapidly removed by mass transport of air.

IOR (Intrinsic Oxidation Rate) is a key parameter for understanding how oxidation is taking place in a dump. It is merely the rate of oxygen consumption by the material deposited in the dump under the conditions applicable to the particular case and is a function of a large number of variables including pore-gas oxygen concentration, particle-size distribution, mineral surface area, bacterial population, temperature, pH, and ferric ion concentration. If one considers the possibility of including the functional dependence of the IOR on these variables, numerically intensive calculations will be achieved.

The IOR in pyritic waste rock is a critical parameter for estimating the potential for acid drainage and its probable duration. It should be of a form that is physically reasonable and which allows the following goals to be achieved:

- i. identification of the mechanisms controlling the global oxidation of the pyritic material in the pile;
- ii. planing the appropriate remedial actions;
- iii. assessing the rehabilitation schemes to be applied;
- iv. assessing the building construction techniques

- v. quantification of pollutant loads, with emphasis on the peak values and the related time for its occurrence.

Values of IOR in the range of 10^{-8} to 10^{-9} $\text{kg (O}_2\text{)}\text{m}^{-3}\cdot\text{s}^{-1}$ are typical for a large number of waste rock piles Ritchie (1995). Values greater than 10^{-7} can be considered extremely high whereas values around 10^{-10} are related to marginal acid drainage environmental problems.

Temperature and oxygen profiles can be analysed both to extract the oxidation rate in a region of pyritic material and to determine the dominant gas-transport mechanisms. With this information and data on bulk physical properties of the dumped material, the primary pollution-production rate can be calculated; from this result an estimate can be made of the time dependence of pollution load in drainage from the base of the pyritic material.

Pore gas oxygen concentration and temperature profiles can be achieved by means of drilling probe holes into the dumped material and installation of liners. There should be enough probe holes to obtain profiles through enough different types of material, to represent the pile. Recognising that *in situ* measurements of oxygen concentration and temperature profiles in a waste pile is the best way to assess the IOR and consequently predict the generation potential of acid drainage and duration, it should be pointed out that other techniques are available to deal with the problem.

The waste rock pile (WRP-4) of Poços de Caldas mining site, object of the present study, was chosen because most of the infiltrating water is collected in a single holding pond. This situation represents a particularly unique opportunity that is not generally available and has not been previously exploited. This allows a better assessment of the cover performance.

The results from this work are expected to contribute to the study of problems associated with acid mine drainage prediction and management and should also support the decisions concerning the remedial actions to be implemented by the mining operator.

DESCRIPTION OF THE STUDY SITE

The uranium mine and mill site is located on the Poços de Caldas plateau, in the Southeast region of Brazil. The alkaline complex corresponds to a circular volcanic structure which formation began in the upper Cretaceous (87 ma) and evolved in successive steps until 60 ma. This intrusion is rounded by the levelling of bed rocks, consisting of granites and gneisses. These rocks are frequently cut by diabase dykes, amphibolites and gneisses.

These igneous-polycyclic activities, of alkaline nature, associated to intense metassomatic processes and a strong weathering, gave rise to a variety of rock types belonging to the Nepheline-Syenite family and to uranium mineralisation.

The uranium enrichment in Poços de Caldas mine is related to hydrothermal events (primary mineralisation) and to latter weathering processes (secondary mineralisation).

The mine covers an area of about 2.5 km^2 and is divided into three mineralised units designated as ore bodies A, B and E for mining purposes.

The mining and milling facilities began commercial operation in 1982. However, the original intended production of 500 ton of U_3O_8 per year was never reached. As of 1995, 1,172 tons of U_3O_8 were produced. In the development of the mine $44.8 \times 10^6 \text{ m}^3$ of rock were removed. From this amount, 10 million ton were used as building material (roads, ponds, etc). The rest was

disposed into two major rock piles, waste rock pile 8 (WRP-8) and 4 (WRP-4). In contrast to WRP-8, all the drainage from WRP-4 is collected into a single holding pond.

ASSESSMENT OF GENERATION OF ACID ROCK DRAINAGE

The IOR was determined from available monitoring program data carried out by the mining operator. The monitoring program of the operator is conducted since the beginning of the mining activities in 1982 and represents a robust data base for the purpose of this work.

The chemical equation of pyrite oxidation (equation 1) shows that the reaction is supported by the supply of water and oxygen. In most situations, there will be plenty of water contained within the pore spaces of a dump resulting from rainfall. Initially, the only effective mechanism for oxygen transport into a dump is the diffusion through the pore spaces caused by the oxygen gradient generated by oxygen consumption within the dump. Diffusion is rather a slow transport mechanism and, as consequence, in waste rock dumps where diffusion dominates, oxygen concentration decreases with distance from the surface of the dump. Table 1 shows the physical characteristic of the WRP-4. It was assumed that the main mechanism of oxygen transport into the system is by diffusion of O₂ gas through the rock pore spaces.

Table 1
General Characteristics of WRP-4

Volume	12.4 x 10 ⁶ m ³
Total Mass	29.8 x 10 ⁶ ton
Area	50.7 x 10 ⁴ m ²
Bulk Density of Dumped Material	2,020 – 2,260 kg.m ⁻³

It was also assumed that all sulphate in the drainage results from pyrite oxidation and that no major precipitation reactions involving sulphate take place. Local precipitation averages 1.7 m.a⁻¹. It can be estimated that the average volume of precipitation collected by the dump area in one year is about 9.67x10⁵ m³. The infiltration rate for the undisturbed rock at the mining area is 0.1 mm³ m⁻¹ year⁻¹ according to Cross et al. (1991). This figure is 5% of the local precipitation rate. However, the dump surface is completely altered in relation to the original surface. It is suggested that 40 to 60% of the precipitation will infiltrate into the dump (Morin and Hutt, 1994). It is reported by Nordstrom (1990) that infiltration in different dumps averages 50% of the total precipitation. Stromberg & Banwart (1994) had reported an infiltration rate of 70% for a waste dump in Sweden. Since the infiltration rate for the current site is not known, we elected to use the value reported by these authors since the amount of water collected in the holding pond suggests the occurrence of high infiltration rates. The mine operator reports that 1.5x10⁵ m³ and 1.98x10⁵ m³ have been pumped from the holding pond in the years 1996 and 1997 respectively. These figures correspond to 16% and 20% of the incident rain over the dump. The average of these two values, i.e., 1.74x10⁵ m³ will be used in the further calculations.

If the average sulphate concentration in the drainage water (1,010 mg.L⁻¹) is multiplied by the average amount of water pumped from the holding pond in a year (1.74x10⁵ m³) an amount of, 1.76x10⁸ g sulphate will be recovered.

Now, if the total area of the dump is taken into account a Global Sulphate Generation Rate (GOR) of 7.9 x 10⁻⁷ kg(SO₄).m⁻².s⁻¹ is calculated. IOR is related to GOR according to equation 8:

$$\text{GOR} = \text{IOR} * L \text{ (equation 2)}$$

Where L is the height of the dump.

Souza (1995) reports that the maximum height of WRP-4 is 90m. As a result, a maximum IOR value of $2.72 \times 10^{-9} \text{ kg (O}_2\text{) m}^{-3}\cdot\text{s}^{-1}$ can be applied to the dump. This result is near the low end of values that are typical of waste rock piles (Ritchie 1995). If it is also assumed that oxygen diffusion is the major mode of oxygen transport into the dump the average depth of the surface

$$X = \sqrt{\frac{2DC_0}{\text{IOR}}}$$

region under oxidation may be calculated according to equation 3 (Ritchie, 1994):

Where C_0 ($\text{mg}\cdot\text{kg}^{-1}$) is the concentration of oxygen in the air ($0.265 \text{ kg}\cdot\text{m}^{-3}$), D ($\text{m}^2\cdot\text{s}^{-1}$) is the diffusion coefficient of oxygen in the pores of the rock. Ritchie (1995) reports that a typical value of D is $5.0 \times 10^{-6} \text{ m}^2\cdot\text{s}^{-1}$. Applying these parameter values to the above equation, the thickness of the oxidising region will be about 31.2 m.

The time needed for a region of a given thickness to be oxidised is given by the equation 4 (Ritchie, 1995):

$$T = \frac{\varepsilon\delta_{rs}}{\text{IOR}} \text{ (equation 4)}$$

Where ε is the mass of oxygen used per unit mass of sulphur oxidised and assumes a value equal to 1.75; δ_{rs} is the sulphur bulk density as pyrite ($30 \text{ kg}\cdot\text{m}^{-3}$). As a result of these calculations the time needed to the region under oxidation to be consumed will be approximately 600 years.

The previous results rely on the assumptions that the dump is homogeneous in respect to the concentration of pyritic material; diffusion is the dominant mechanism of oxygen transport into the dump and no major precipitation reaction (that would cause significant losses of sulphate from the solution) is taking place.

These calculations have also to consider the complexity of a mine rock-pile in respect to the preferential channels for water flow. For large piles only 5 to 20% of the rock surfaces may be regularly flushed by infiltration events (Morin & Hutt, 1994). As a result of the above discussion, the calculations done must be regarded as a general trend and used as an initial guidance for decision making.

REMEDICATION CONTROL OPTIONS

As it has been discussed above, there are three main factors that govern the rate of oxidation of pyritic rock. These are the availability of water, the availability of oxygen and the biomass of catalysing bacteria.

It is easy to show that the water contained in the pore spaces of the rocks is enough to provide the water required to oxidise all of the sulphidic material in the dump. If it is assumed that the production of sulphate measured in the holding pond of the WRP-4 is solely related to pyrite

oxidation (this is a fair assumption since other sulphide minerals are reported as being of less importance), the oxygen consumption associated to sulphate production may be estimated with the aid of equation 1. As a result, 1.03×10^8 g of oxygen would have been necessary to oxide the amount of sulphide to produce the calculated amount of sulphate. If it is considered that average concentration of oxygen in natural waters is 8 mg.L^{-1} , it can be accepted that a total amount of 1.39×10^6 g of oxygen were transported with the total volume of water collected at the holding pond. The obvious conclusion is that the amount of dissolved oxygen in the infiltration water would be not enough to account for the recovered sulphate.

Bacterially catalysed oxidation rates are sensitive to pH from a range of 8 to close to zero. Chemical rates are much less sensitive and both are much higher than the very low intrinsic pyrite oxidation rates that are of significance in pollutant generation rates in waste dumps. Therefore in the case of WRP-4, $1.73 \times 10^{-9} \text{ kg(O}^2\text{)m}^{-3}\cdot\text{s}^{-1}$, this factor (bacterial catalysis) is not a significant factor

The diffusion of oxygen into WRP-4 through the gas-filled pores was assumed to be the main factor governing the rate of oxidation of the sulphidic material. The diffusion coefficient in porous media is closely related to the diffusion coefficient in free air, modified by a factor to account for the restrictions of the pores. To reduce the diffusive flux of oxygen into the WRP-4 we recommend covering the dump with a layer of an appropriate material that has an oxygen diffusion coefficient lower than the rocks forming the dump. This would allow a reduction in the global oxidation rate by a significant factor. The reduction factor will depend on a very large extent on the cover properties than on the waste rock pile properties themselves [The practical problem is to design a cover that remains relatively intact over a long period of time. The Global Oxidation Rate of the entity may be related to the cover thickness X_c and oxygen diffusion coefficient of the cover D_c by equation (5):

$$GOR = \sqrt{2C_0DS^*} (\sqrt{\alpha + n} - \sqrt{\alpha + n - 1}) \text{ (equation 5)}$$

Where

$$\alpha = \sqrt{\frac{X_c}{D_c} \left(\frac{S^* D}{2C_0} \right)}$$

If it is expected that the cover shows some efficiency, α will have to be greater than unity. With the aid of equation 5 the sulfate concentration and load arising from the dump drainage was simulated for different cover thickness and oxygen diffusion coefficient. The infiltration rate was assumed to be equal to 50% of the total precipitation rate. The results are shown in table 2. It can be observed that from the option 4 on, the reductions in the sulphate and uranium concentrations begins to be negligible.

Table 2
Efficiency of the different remediations applied to the dump

Dif. Coef. (m ² .s ⁻¹)	1x10 ⁻⁸ (1)	1x10 ⁻⁸ (2)	1x10 ⁻⁸ (3)	1x10 ⁻⁹ (4)	1x10 ⁻⁹ (5)	1x10 ⁻⁹ (6)	1x10 ⁻¹⁰ (7)	1x10 ⁻¹⁰ (8)	1x10 ⁻¹⁰ (9)
Thick. (m)	0.5	1.0	2.0	0.5	1.0	2.0	0.5	1.0	2.0
GOR kg/m ²	1,8 x10 ⁻⁸	2.2x10 ⁻¹⁰	1.1x10 ⁻¹⁰	2,0x10 ⁻⁹	1x10 ⁻⁹	2,0x10 ⁻¹¹	2,0x10 ⁻¹⁰	1,0x10 ⁻¹⁰	5,0x10 ⁻¹¹
Load t/Year	139	75	38	15	7,7	3,9	1,8	0,76	0,34
[SO ₄] mg/L	287	154	78	32	16	8,0	3,7	1,58	0,79
[²³⁸ U] Bq/L	49	26	13	5,4	2,7	1,3	0,63	0,27	0,13

It has been mentioned that the integrity of the covering system has to be assured in the long term. As a result a three layers cover system is usually employed. The system comprises a lower granular layer, an intermediate one of low gas permeability and an external one of sand and gravel to avoid the erosion of the system by rain and wind. The costs of a system like that, adapted to the dimensions of the WRP-4 is represented in table 3.

Table 3
Covering system associated costs

	US\$/m ²	m ²	Total (US\$)
Lower Layer	3,5	56,5 x 10 ⁴	1,98 x 10 ⁶
Intermediate Layer (low permeability)	11	56,5 x 10 ⁴	6,22 x 10 ⁶
External Layer (gravel and sand)	3,0	56,5 x 10 ⁴	1,70 x 10 ⁶
Total			9,9 x 10 ⁶

Source: Fernandes (1997)

It can be seen that US\$ 10 million approximately would be necessary for the application of a covering system like that to the entity. Other possibilities were considered like the entity covering with a plastic material or the backfilling of the open pit with the material of the waste rock pile. The first option would amount US\$ 12.7 million while the second one would amount about US\$ 70 million. It must be emphasized that these cost refer to only one of the existing waste rock piles at the site.

CONCLUSIONS

Acid drainage generation is a long-term problem at mining sites where sulphidic material is present in the rock. The IOR was determined from monitoring program data through mass balance calculations. This approach depends on the availability of hydrologic and water quality

data and information on the amount, composition and physical properties of the waste rock. It was demonstrated that IOR $1.73 \times 10^{-9} \text{kg(O}^2\text{)m}^{-3}\cdot\text{s}^{-1}$. can be applied for the dump. This rate can be considered as being a low intrinsic oxidation rate. By this reason, remediation should concentrate on the reduction of oxygen diffusion into the dump. This may be achieved by covering the dump with some sort of material (e.g. clay or compacted clay) that has a lower coefficient of diffusion for oxygen and lower permeability to water than that the ones of the rocks at the surface of the dump. Future monitoring of the dump should aim the improvement of the understanding of the actual infiltration rates applicable to the site and of the distribution of oxygen in the dump. These data may be obtained by the installation of lysimeters (work in progress) and installation of oxygen probes to measure oxygen concentration at different depths in the dump (work to be developed).

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