

MEASUREMENT OF OXYGEN CONSUMPTION AND DIFFUSION IN EXPOSED AND COVERED REACTIVE MINE TAILINGS¹

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Abstract. When sulphidic tailings are exposed to atmospheric conditions, oxygen can flow in the material where it will be consumed by oxidation reactions. This paper presents results from a project studying the in situ oxygen diffusion and consumption in tailings. It provides measured values for the oxygen reaction rate K_r and the effective diffusion D_e coefficients based on oxygen consumption and diffusion tests that are performed under various conditions. Based on the test results, the K_r and D_e values determined in situ are compared to values obtained through simple predictive models. Results indicate that the value of these two parameters is influenced by factors such as the degree of saturation S_r and in situ porosity n . A lower S_r or a higher n may increase the oxygen flux toward the reactive tailings, which can increase the production rate of acid mine drainage (AMD).

Additional Key Words: Acid Mine Drainage, Mine Tailings, Fick's Laws, Oxygen Fluxes, Reaction Rate.

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Introduction

The oxidation of reactive sulfide minerals can cause acidification and release of heavy metals in surface and ground water. To inhibit the formation of such acid mine drainage (AMD, or ARD for acid rock drainage) the interactions between the constitutive components (sulfide, H₂O or O₂) must be limited. Under a humid climate, the control of the O₂ flux is often the best strategy. In this regard, one of the possible ways to prevent AMD generation on a mine site is to cover the acid generating tailings with a moisture retaining cover made of geological materials, also known as cover with capillary barrier effect (CCBE) (Aubertin et al., 2002). Typically, a CCBE acting as an O₂ barrier will be made of at least three layers. The surface layer composed of coarse materials behaves as a protection layer and also limits evaporation from the layers below. The middle layer is usually built with fine-grained materials and serves as a water-retention layer that must stay close to saturation. The bottom layer is made of coarse materials and serves as a support layer for the cover. The bottom layer creates the capillary break that will allow the water retention layer to stay saturated (Aubertin et al., 1995). The efficiency of the cover will then depend on the difference between the O₂ fluxes in the tailings before and after the installation of the cover system. The cover efficiency over a period of time can be assessed using the following equation (Aubertin et al., 1999):

$$E(\%) = \left(1 - \frac{F_C}{F_0}\right) \times 100\% \quad (1)$$

- where E = Efficiency of the cover system (%);
F₀ = Oxygen fluxes in the uncovered tailings [ML⁻²T⁻¹];
F_C = Oxygen fluxes through the CCBE [ML⁻²T⁻¹].

The O₂ flux into exposed tailings or through a CCBE is influenced by the O₂ diffusion and reactivity rate coefficients (D_e and K_r respectively). Different features such as SO₄⁻² release, O₂ gradient, volumetric water content profile and O₂ consumption can be used to estimate the degree to which a CCBE may limit O₂ migration and AMD production (Collin and Rasmuson, 1988; Elberling et al., 1994; Aubertin et al., 1995; Elberling and Nicholson, 1996; Tibble and Nicholson, 1997; Aubertin et al., 1999; Mbonimpa et al., 2003; Dagenais, 2005). This paper presents an approach based on the modified O₂ consumption tests. It gives the means to determine O₂ fluxes into uncovered tailings or through a CCBE. Two sample cases are presented to illustrate how this technique has been applied, for uncovered mine tailings and for a CCBE.

Test procedure

The modified oxygen consumption (MOC) test is based on direct measurement of O₂ flux into covered or uncovered tailings. It is a variation on the testing procedure proposed by Elberling et al. (1994). The objective of the MOC test is to estimate the value of parameters required to calculate the flux through uncovered or covered tailings, i.e. the effective diffusion and reactivity rate coefficients. The test is designed to cause relatively minor disturbance of the tested materials. It consists of driving a stainless steel (or Al) cylinder (with a diameter of approximately 15 cm) into the soil or mine tailings to create a zone isolated from its

surroundings. In covered tailings, the cylinder will be pushed through the moisture retaining layer into the tailings underneath. A longer cylinder is preferred, in order to prevent lateral gas movement in the unsaturated materials (Aubertin et al., 2000b; Bussière et al., 2002; Mbonimpa et al., 2002a). The diameter of the cylinder should be sufficient large with respect to the particle size to minimise boundary effects. At the surface, a 2 to 5 cm headspace is left to create an O₂ chamber (see Fig. 1). The cylinder is then sealed hermetically by a cap instrumented with an O₂ sensor to measure the decrease of O₂ concentration in the reservoir. With the MOC test, the O₂ concentration measurements last long enough to measure a significant variation in the O₂ concentration in the reservoir (between 3% and 5%). In exposed reactive tailings, the test usually takes between 2 and 6 hours. In covered tailings, this period can be up to 5 days. A control O₂ sensor is placed in an isolated box and simultaneous measurements are taken with the two sensors during the test. With this control sensor, a correction can be applied to the measurement taken in the reservoir to account for the fluctuations during the test caused by factors like ambient temperature and atmospheric pressure variations (Tibble, 1997; Bussière et al., 2002; Mbonimpa et al., 2002a). Once a test is completed, samples of the tested materials (i.e. the cover material or the tailings) are taken and analysed to obtain some properties necessary and useful for interpretation of the test (particle size distribution curves, specific gravity of solid grains G_s , in-situ porosity n and in situ volumetric water content θ_w).

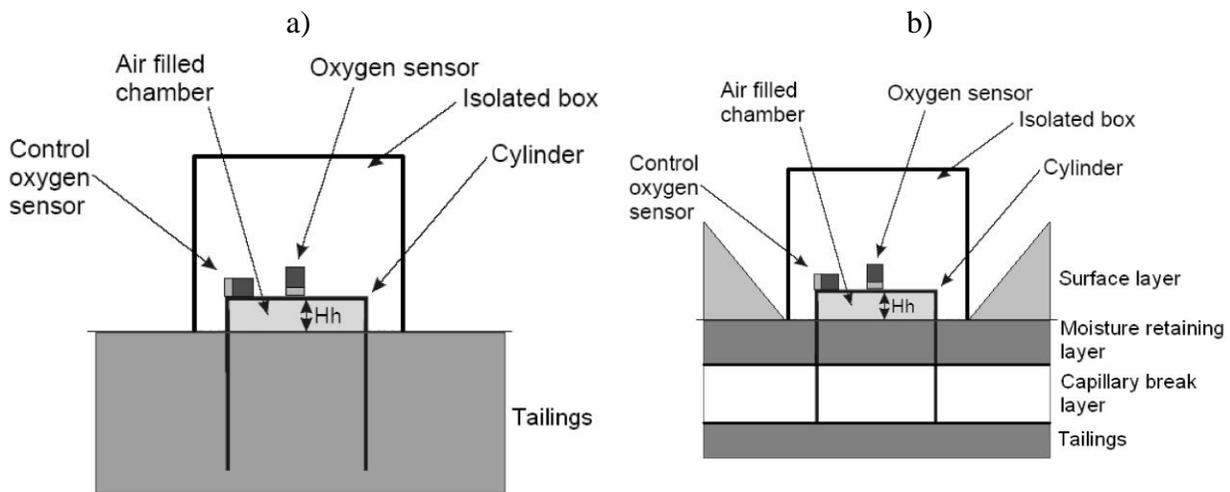


Figure 1. Schematic representation of the modified oxygen consumption test in a) exposed tailings; b) tailings covered with a CCBE (adapted from Bussière et al., 2002).

Theory and Interpretation

When performed on exposed tailings, the modified oxygen consumption (MOC) test can be used to determine both the O₂ diffusion and reactivity rate coefficients for a given type of tailings. This information can be used to help determine the type of rehabilitation method that will be put in place (in combination with other information such as the geochemistry of the tailings and the hydraulic regime of the impoundment). MOC tests can also be conducted to evaluate the amount of O₂ flowing to reactive tailings.

The main underlying assumption at the core of these tests is that molecular diffusion is the controlling O₂ transport mechanism. The one-dimensional diffusive flux of O₂ F(z,t) in an unsaturated porous medium can then be defined using Fick's first law (equation 2; Mbonimpa and Aubertin, 2003):

$$F(z, t) = -D_e \frac{\partial C(z, t)}{\partial z} = -\theta_{eq} D^* \frac{\partial C(z, t)}{\partial z} \quad (2)$$

where $C(z, t)$ = Concentration of oxygen at time t and depth from surface z [ML³];

D_e = Effective diffusion coefficient [L²T⁻¹];

θ_{eq} = Equivalent (diffusion) porosity [dimensionless];

D^* = Bulk diffusion coefficient [L²T⁻¹] expressed as: $D^* = D_e / \theta_{eq}$.

In this equation, the equivalent porosity θ_{eq} is used to take into account the flux of O₂ in the air phase and in the water phase (Aubertin et al., 1999, 2000a); it is defined in equation 3:

$$\theta_{eq} = \theta_a + H\theta_w \quad (3)$$

where θ_w = Volumetric water content [dimensionless];

θ_a = Volumetric air content [dimensionless] defined as: $\theta_a = n - \theta_w$, where n is the soil total porosity [dimensionless];

H = Henry's equilibrium constant (H ≈ 0.03 at 20° C for oxygen).

The flow of O₂ may be influenced by the oxidation reaction when the tailings contain sulfides (e.g. pyrite). This reaction is usually controlled by 1st order kinetics (Nicholson et al., 1989; Mbonimpa and Aubertin, 2003), which means that the O₂ consumption rate is linearly related to its concentration. The O₂ consumption can be introduced into Fick's second law as follows (Mbonimpa et al., 2002a; 2002b):

$$\frac{\partial}{\partial t} (\theta_{eq} C) = \frac{\partial}{\partial z} \left(D_e \frac{\partial C}{\partial z} \right) - K_r C \quad (4)$$

This equation can be simplified if θ_{eq} and D_e are to be considered time and depth independent (Mbonimpa et al., 2002a; 2002b):

$$\frac{\partial C}{\partial t} = D^* \frac{\partial^2 C}{\partial z^2} - K^* C \quad (5)$$

In these equations, K_r is the O₂ reactivity rate coefficient [L³L⁻³T⁻¹ or T⁻¹]. Similarly to the D^* , there is also a bulk O₂ reactivity rate coefficient ($K^* = K_r / \theta_{eq}$).

Equations 4 and 5 are used with equation 2 for the interpretation of the MOC test (Mbonimpa et al., 2002b).

Because of its relatively long duration, the MOC test has to be interpreted under a transient analysis. It is usually necessary to use a numerical method since no analytical solution exists for such a test under transient conditions. The authors have used the software POLLUTE (Rowe et al., 1998) to interpret these tests. The MOC test interpretation is based on a comparison between the measured and the simulated evolution of the O₂ concentration in the reservoir. An iterative process is used, where values of D* and K* are changed. The actual value of the bulk diffusion D* and O₂ reactivity rate K* coefficients are those that allow a “best fit” to the experimental data. These parameters are associated with the effective diffusion coefficient D_e and the reactivity rate coefficient K_r (using the equivalent porosity θ_{eq}). This iterative comparison method therefore gives D* and K* at a given location. These can be used subsequently to calculate the flux of O₂ into the exposed tailings (or through a CCBE, to evaluate its performance, for example).

During the best fit analysis, the starting D* value can be estimated using basic material properties measured in the field, including the in situ porosity n and volumetric water content θ_w. Aachib et al. (2002, 2004) have developed a simple predictive equation that can be used to evaluate the starting value for D_e (equation 6):

$$D_e = \frac{1}{n^2} \left[D_a^0 \theta_a^{3.4} + H D_w^0 \theta_w^{3.4} \right] \quad (6)$$

where D_a^0 = Diffusion coefficient of oxygen in air ($\approx 1.8 \times 10^{-5}$ m²/s);

D_w^0 = Diffusion coefficient of oxygen in water ($\approx 2.5 \times 10^{-9}$ m²/s).

This equation is based on the Collin and Rasmuson (1988) formulation, which has been shown to provide good results when compared to D_e values measured in the laboratory (Aubertin et al. 1999; Aachib et al., 2004).

Similarly, it is necessary to have an estimate of the bulk oxygen reactivity rate coefficient K* for the initial conditions that are to be included in the POLLUTE file. For that purpose, the O₂ reactivity rate coefficient K_r can be estimated with the Collin (1987, 1998) model based on surface kinetics, in which the rate varies linearly with the proportion of sulfide minerals (i.e. pyrite). This simple model can be written as follows (Collin, 1987; Mbonimpa et al., 2002c):

$$K_r = K' \frac{6}{D_H} (1 - n) C_p \quad (7)$$

where K' = Reactivity of pyrite with oxygen ($K' \approx 5 \times 10^{-10}$ m³ O₂/m² pyrite /s);

C_p = Pyrite content over mass of dry tailings (kg/kg).

Parameter D_H represents an equivalent particle diameter that can be estimated using a relationship with the particle size distribution (PSD) curve (Aubertin et al., 1998; Mbonimpa et al. 2002c):

$$D_H = [1 + 1.17 \log(C_u)]D_{10} \quad (8)$$

where D_{10} = Diameter corresponding to 10% on the cumulative PSD curve [L];

C_u = Coefficient of uniformity [L] expressed as $C_u = D_{60}/D_{10}$;

D_{60} = Diameter corresponding to 60% on the cumulative PSD curve [L].

When using POLLUTE to interpret the data, the value of K^* is related to the half-life time $t_{1/2}^*$ (as an analogy to a radioactive decay parameter). In the case of a first-order reaction, this relationship is expressed as:

$$t_{1/2}^* = \frac{\ln 2}{K^*} \quad (9)$$

Sample Cases

In this section, the authors present two cases where the MOC tests were performed to evaluate O_2 fluxes. The first case (Mine A) consists of an exposed tailings storage facility (TSF) on an operating mine. For the second case (Mine B), the tests were performed on an already rehabilitated TSF where a three layer cover has been installed over acid generating tailings. In both cases, tests were conducted at multiple locations on the TSFs to characterize the variation of the O_2 diffusion and reactivity rate coefficients (D_e and K_r).

Fluxes in exposed (uncovered) tailings (Mine A)

The objective of these tests was to evaluate the oxidation rate of the tailings and to have an idea of the O_2 fluxes in order to prepare a sound management and closure scenario for this facility. To evaluate this, 8 tests were completed at various locations on the TSF. The basic properties of the tailings next to the test locations are presented in Table 1. Because the O_2 flux is influenced by the degree of saturation S_r of the material (as shown by equations 2 and 6), the test locations were chosen to represent a range of water contents, from almost saturated (Tests 3 and 6) to a dry point on the facility (Test 4). Other parameters of interests for the analysis of the tests results are the in situ porosity n , the tailings grain sizes D_{10} and D_{60} (respectively 0.003 mm and 0.038 mm), the specific gravity of the solid grains G_s (of 2.93), and the amount of pyrite in the tailings C_p (of 12%), which were obtained from laboratory samples. These values were assumed to be equal (and constant) for all tests.

Table 1. Basic in-situ properties at the MOC test locations for Mine A.

Test	In situ porosity n	Degree of saturation S_r^*	Volumetric water content θ_w^*
1	0.44	69.5%	0.30
2	0.40	82.3%	0.33
3	0.34	98.3%	0.34
4	0.39	60.1%	0.23
5	0.41	67.6%	0.28
6	0.45	98.0%	0.44
7	0.43	81.7%	0.35
8	0.40	92.2%	0.37

* At the surface of the tailings next to the test location.

The properties in Table 1 are used to interpret the MOC tests. As explained previously, the MOC tests are modeled with various combinations of the effective diffusion coefficient D_e and oxygen reaction rate coefficient K_r (i.e. D^* and K^*) so that computed O_2 concentrations match experimental data as closely as possible. Because POLLUTE is a 1D modeling software, there are only two boundary conditions. The top boundary condition is a specified mass of O_2 fixed by the height of the reservoir (measured at the beginning the test) and the initial O_2 concentration C_0 (9.71 mol O_2/m^3 air or 276.8 mg O_2/m^3 air at 20°C). The bottom boundary condition is a constant concentration of O_2 of 0 at an infinite thickness. The starting values for the two parameters D^* and K^* are obtained using the models presented above (equations 6 and 7). It is also assumed (for simplicity) that the degree of saturation of the tested material is constant over the whole depth of analysis. A sample result following an analysis with POLLUTE (for test #2) is presented in Fig. 2.

On Fig. 2, it appears that the field and modeled results are close. Similar results were obtained with the other tests. On Fig. 3, the results of all 8 tests are presented and compared with the values obtained with models presented in equations 6 and 7 (dashed lines).

As seen on Fig. 3, both the D_e and K_r increase when the degree of saturation of the material decreases. The values of D_e predicted with the equation proposed by Aachib et al. (2002, 2004) are fairly similar to those obtained from the field measurements (Fig. 3a). On the other hand, the values of K_r obtained with the predictive equation are not as close to the measured values (Fig. 3b), in part because this parameter seems to vary with the degree of saturation and this is not taken into account in the Collin (1987) model.

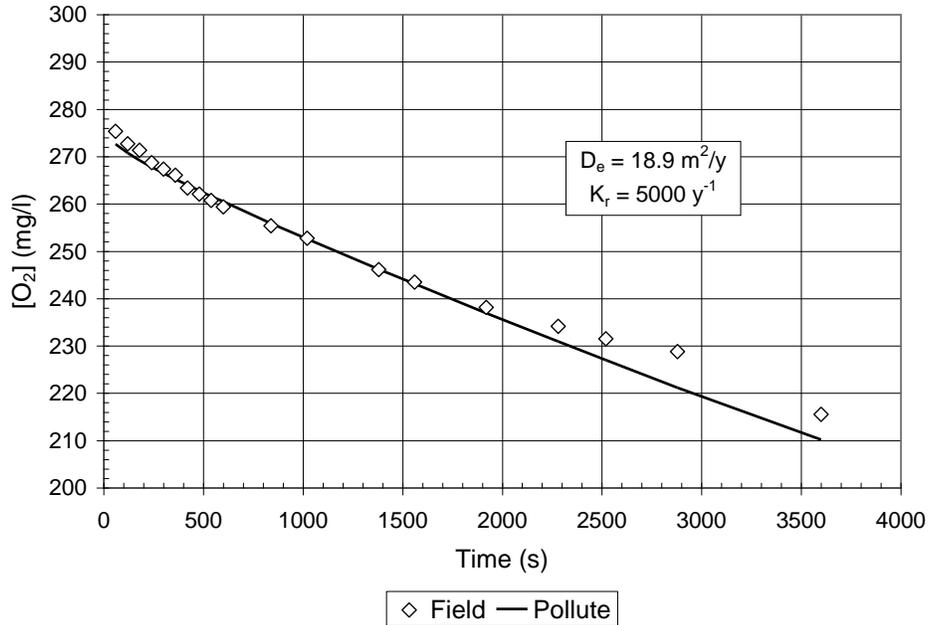


Figure 2. Sample test result for modified oxygen consumption test #2 completed at Mine A; the line corresponds to the model calculations made with the adjusted parameters that provide a best fit to the measured O₂ concentration values in the reservoir.

Following the determination of D_e and K_r , it is then possible to estimate the diffusive flux of O₂ at the surface of uncovered tailings. The diffusive surface flux F_0 entering uncovered mine tailings in a (pseudo) steady-state, can be estimated with a steady state solution to Fick's Law (Elberling et al., 1994; Mbonimpa and Aubertin, 2003):

$$F_0 = C_0 \sqrt{D_e \times K_r} \quad (10)$$

where C_0 is the concentration of oxygen at the surface of the tailings, which is approximately 0.27 kg O₂/m³ air (at STP). This equation supposes that the system attains steady state conditions fairly rapidly and also represents a conservative estimate of the surface fluxes (Mbonimpa et al., 2003). Using equation 10 and the results presented in Fig. 3, the surface flux for the Mine A tailings at various degrees of saturation are shown in Fig. 4.

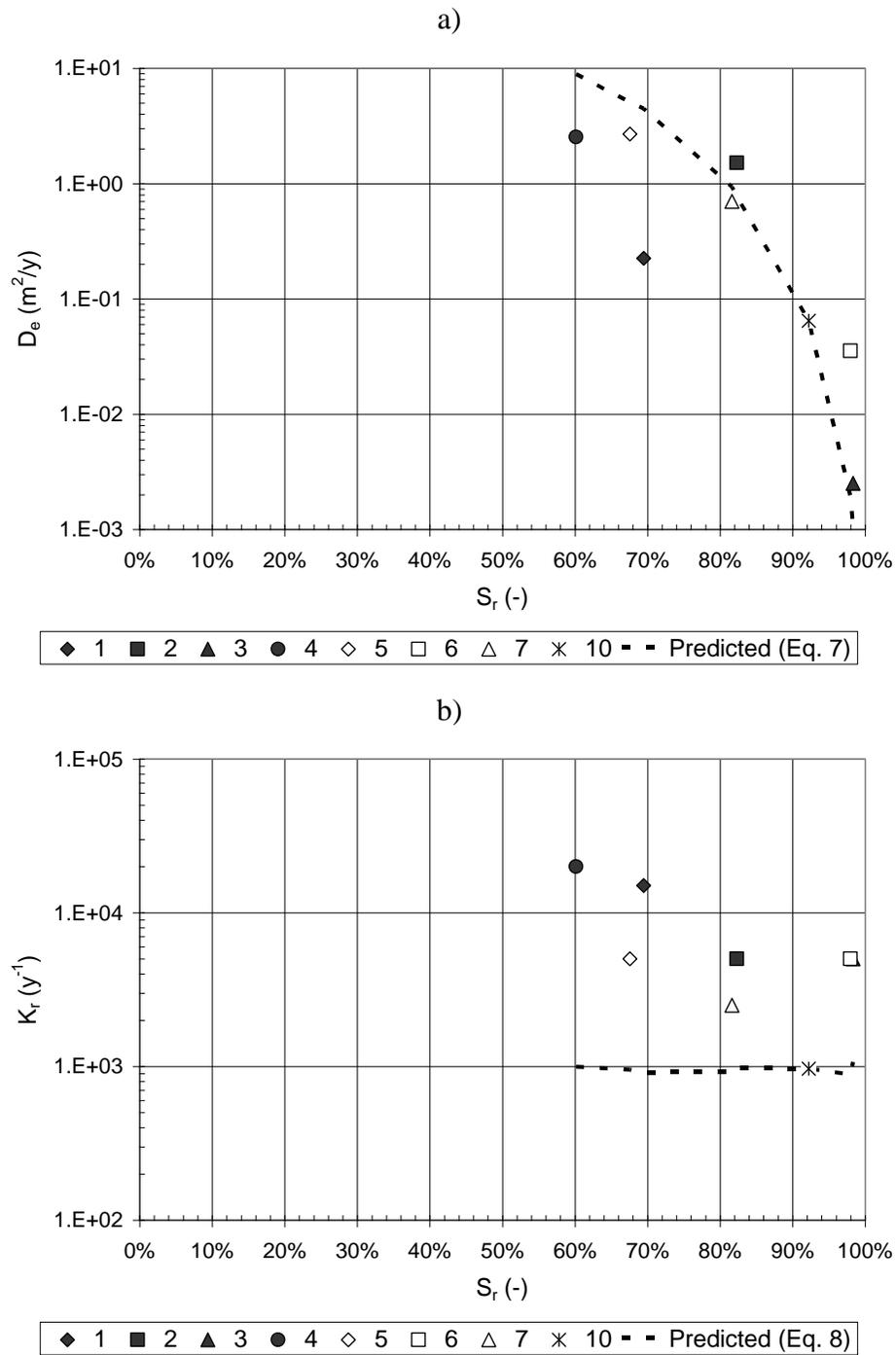


Figure 3. Modified oxygen consumption test results for the tests conducted at Mine A a) Effective diffusion coefficient (D_e), b) Oxygen reactivity rate coefficient (K_r).

The results presented in Fig. 3 and 4 show that the mine tailings studied are fairly reactive. They also emphasise the necessity to keep the materials at a high degree of saturation (an issue

that is explicitly addressed elsewhere in this conference by Dagenais et al., 2006 and Ouangrawa et al., 2006).

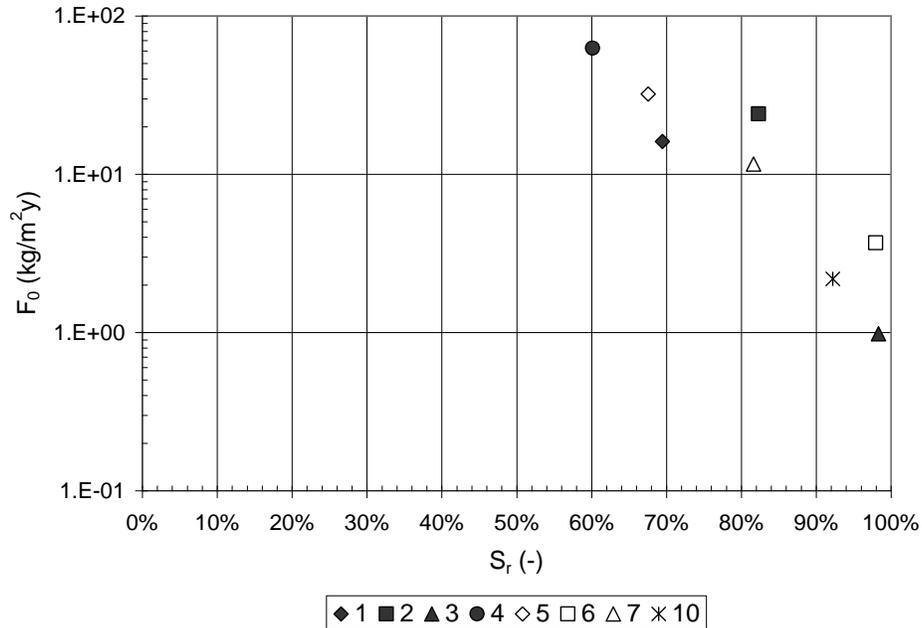


Figure 4. Calculated value of the steady state surface flux with respect to degree of saturation for exposed tailings (Mine A).

Fluxes in tailings covered with an oxygen barrier (Mine B)

The second case studied (Mine B) consists of a base metal mining operation that has been closed for a multitude of years, and whose TSF has been restored with a three layer cover with capillary barrier effects (CCBE): a base layer consisting of 50 cm of sand used as capillary break layer; a moisture-retaining layer made of slightly reactive but non-acid generating tailings material with a thickness of 80 cm; and a 50 cm top layer consisting of sand and gravel to protect the silt layer from evaporation and biointrusion (Aubertin et al., 2000b). Ricard et al. (1999) describe the properties of the cover materials in greater detail. This CCBE was design to act as an O₂ barrier (e.g. Aubertin et al. 1999; Mbonimpa et al. 2003). MOC tests are well suited to evaluate the performance of layered covers placed on reactive tailings (Aubertin et al. 2000b; Mbonimpa et al. 2002b; Dagenais, 2005). The MOC tests proposed here, with the interpretation method, are particularly interesting in this case because the tailings forming the moisture retaining layer contain a small amount of sulphides and are slightly reactive (although they do not cause AMD, contrarily to the tailings of Mine A). Preparation of the test sites was done by stripping the protection sand and gravel layer over an area approximately 1 m by 2 m (as seen on Fig. 1b). The cylinder was then inserted from the water-retention layer to the top of the capillary break layer. As the measurements were made in the water retention layer, the range of degrees of saturation is not as large here as for exposed tailings (Mine A).

Samples were again collected next to test sites to evaluate the basic properties of the materials tested. In general, the degree of water saturation measured in situ is higher than 80%. In the POLLUTE model, the top boundary conditions are similar to those that were used in the previous sample case. The bottom boundary condition is a constant O_2 concentration of 0 kg/m^3 below the water retention layer, dictated by rapid and complete O_2 consumption by the reactive tailings. The capillary break layer, which typically has a low degree of saturation S_r , does not interfere with O_2 diffusion in the media. These boundary conditions then represent a worst case scenario for O_2 fluxes through the cover (Aubertin et al., 1999; Mbonimpa et al., 2003). Figure 5 presents the results for one of the tests.

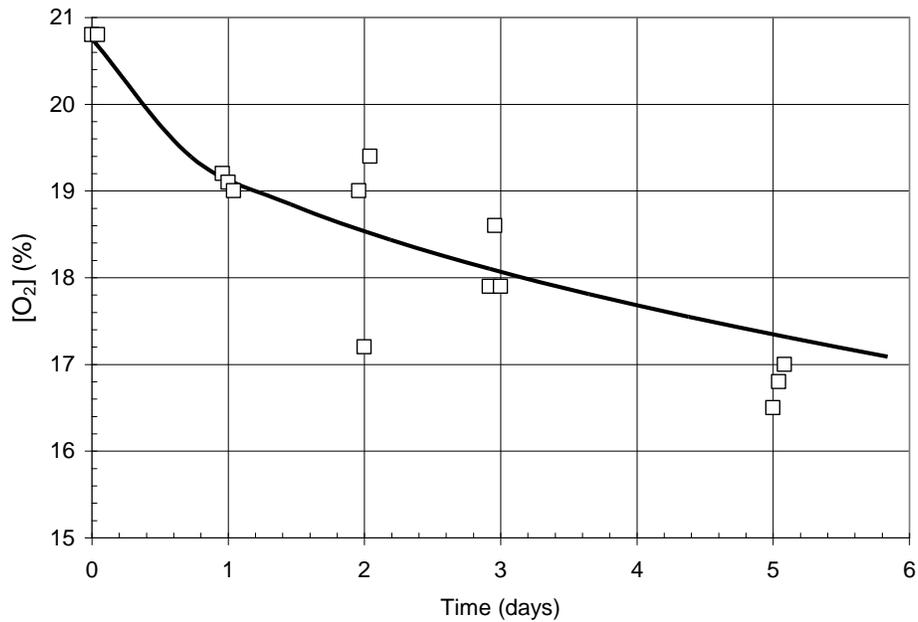


Figure 5. Sample test result for a modified oxygen consumption test completed at Mine B.

For this test, the moisture retaining layer has a degree of saturation S_r of 82 % and a porosity n of 0.45. The interpretation of this test with POLLUTE indicate that the D_e and K_r of the water retaining layer are $0.10 \text{ m}^2/\text{y}$ ($2.7 \times 10^{-4} \text{ m}^2/\text{d}$) and 3.0 y^{-1} ($8.2 \times 10^{-3} \text{ d}^{-1}$) respectively. Using a method presented by Mbonimpa et al. (2002a), the calculated daily flux through the base of the cover (that reaches the reactive tailings underneath) is $1.1 \times 10^{-2} \text{ g/m}^2$. In this case, the small reactivity of the moisture retaining material is beneficial, as it can consume some of the O_2 penetrating the cover, therefore decreasing the amount of O_2 that will actually come in contact with the tailings below (Aubertin et al., 2000b; Bussière et al., 2004). With a moisture retaining layer made of a material that is non-reactive ($K_r = 0$) but with the same D_e , the O_2 flux through the cover would be about $9.6 \times 10^{-2} \text{ g/m}^2/\text{d}$, which is roughly 9 times larger than with the actual materials used to build the cover.

Closing Remarks

The evaluation of the O₂ fluxes through covered and uncovered tailings is a key component to assess the potential for generation of AMD of a tailings storage facility. A technique for the measurement and interpretation of such fluxes has been presented in this paper. Modified O₂ consumption tests combined with the appropriate interpretation methods yield realistic results (compared to predicted values) concerning the fluxes of O₂ through covered or uncovered tailings, using a few simplifying hypotheses. The application requires relatively little equipment and time, and can help those involved in the closure and rehabilitation of mine sites to have a better handle on the properties of the materials they are managing. MOC tests also offer the possibility to evaluate the efficiency of a cover designed to inhibit O₂ fluxes to tailings, in order to minimise generation of AMD.

An equation proposed to predict the effective diffusion coefficient D_e value of the material gives fairly good results for these preliminary analyses. On the other hand, the model used here to predict the reaction rate coefficient K_r is less representative, partly because it does not take into account the influence of the degree of saturation. As shown in the case of Mine A (exposed tailings) this can lead to fairly large discrepancies between the predicted and measured values for K_r. This parameter also varies with other factors such as the state of oxidation, the nature of the sulfides, and bacterial activity. Additional work is underway to study these aspects.

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