FIELD ASSESSMENT OF SULPHIDE OXIDATION RATES IN COLD ENVIRONMENT: CASE STUDY OF RAGLAN MINE

V. Coulombe[1], P. Eng. Jr., M.Sc., B. Bussière[1], P. Eng. Ph.D, J. Côté[2], P. Eng., Ph.D and M. Paradis[3], P. Eng., M.Sc., PMP

[1] Research Institute on Mines and the Environment, Université du Québec en Abitibi-Témiscamingue (UQAT), *445 boul. de l'Université, J9X 5E4, Rouyn-Noranda (Québec), Canada.*

[2] Civil and Water Engineering Department, Laval University, 1065, avenue de la Médecine, G1V 0A6, Québec (Québec), Canada

[3] SNC Lavalin, Sustainable Mine Development, Global Mining & Metallurgy, Québec (Québec), Canada

ABSTRACT

In this field study conducted on Raglan Mine's tailings storage facility (TSF), tailings oxidation rates were characterized with the oxygen consumption (OC) method. Surface tailings unfrozen volumetric water content and temperature were also measured simultaneously with OC tests. Oxygen fluxes between 30 and 550 $mol{\cdot}m^{-2}{\cdot}yr^{-1}$ were observed during summers 2011 and 2012 when tailings temperature and unfrozen volumetric water content varied between -0.1 and 12.8ºC and 0.09 and 0.23 respectively. These oxygen fluxes are non-negligible, but lower than those measured in the lab for the same tailings, which varied between 390 and 1070 $mol{\cdot}m^{-2}{\cdot}yr^{-1}$ at 21°C. Oxygen flux drops in October 2011 to values below 10 $mol{\cdot}m^{-2}{\cdot}yr^{-1}$ with unfrozen volumetric water contents less than 0.09 and temperatures of around -4°C. Results of this study show that, as expected for dry stack tailings, the degree of saturation at Raglan Mine's TSF (generally between 40% and 60%) is not sufficiently high to control oxidation reactions. Oxygen fluxes decrease with temperature and they are greatly reduced when tailings temperature reaches a value of -6°C, conditions observed for a period of 168 days of the year. This study also showed that Arrhenius' law can be used to approximate the effect of temperature on oxygen fluxes for activation energy values between 60 kJ/mol to 124 kJ/mol.

Key Words: Oxidation, Tailings, Oxygen Consumption, Temperature, Arctic.

INTRODUCTION

Interaction between sulphide minerals contained in tailings, water and oxygen can produce acid mine drainage (AMD), an environmental problem of the mining industry. It is generally accepted that molecular diffusion is the principal mechanism of oxygen migration in fine grained tailings (Collin and Rasmuson 1988). Modified Fick's second law can be used to describe oxygen diffusion through reactive tailings for a first-order reaction. Solving Fick's second law under steady state condition ($\partial C/\partial t = 0$) for homogeneous reactive tailings and specified boundary conditions gives the incoming oxygen flux (F_L) through a tailings surface (eq. 1) (Elberling et al. 1994; Mbonimpa et al. 2011). Oxygen fluxes have been used in field and laboratory studies to assess tailings oxygen consumption, in both temperate and Nordic conditions (Elberling 2001; Meldrum et al. 2001; Tibble and Nicholson 1997).

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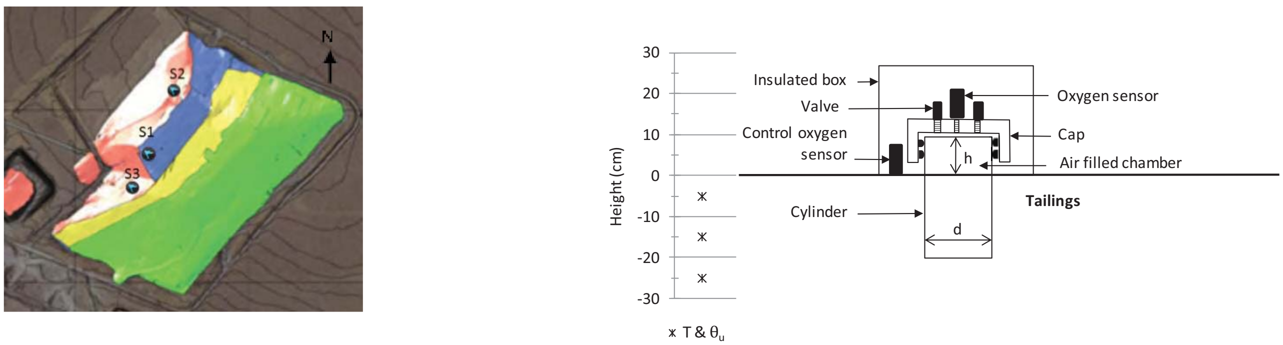
The flux of oxygen is controlled by the oxygen diffusion coefficient (D_e) and by the reaction rate coefficient (K_r). The value of the diffusion coefficient depends on the tailing's degree of saturation; maximal oxygen concentration through water-filled pores ($C_w = 9.2$ mg/L) is 30 times less than the oxygen concentration in air ($C_a = 276.7$ mg/L). A study by Ouangrawa (2007) shows that at saturation greater than 85%, sulphide oxidation rate is significantly reduced. Since oxidation reaction kinetics is affected by temperature, Arrhenius' law can be used to describe effect of temperature on tailings oxygen fluxes (Elberling 2001). Studies on tailings oxidation rates in Nordic environments have shown that oxygen consumption can take place when tailings temperature reaches values of -2°C (Meldrum et al. 2001) and -4°C (Elberling 2001). In this study, oxygen consumption (OC) tests were performed for a one year period at different locations on the Raglan Mine tailings storage facility (TSF) to characterize the *in situ* tailings oxidation rates. Measurements of tailings unfrozen volumetric water content and temperature were also performed simultaneously with the OC tests. The main objective of this study is to better understand the evolution of tailings oxygen consumption rate during a typical year and to relate it with two important factors of influence: temperature and volumetric water content.

STUDY SITE AND FIELD TESTS DESCRIPTION

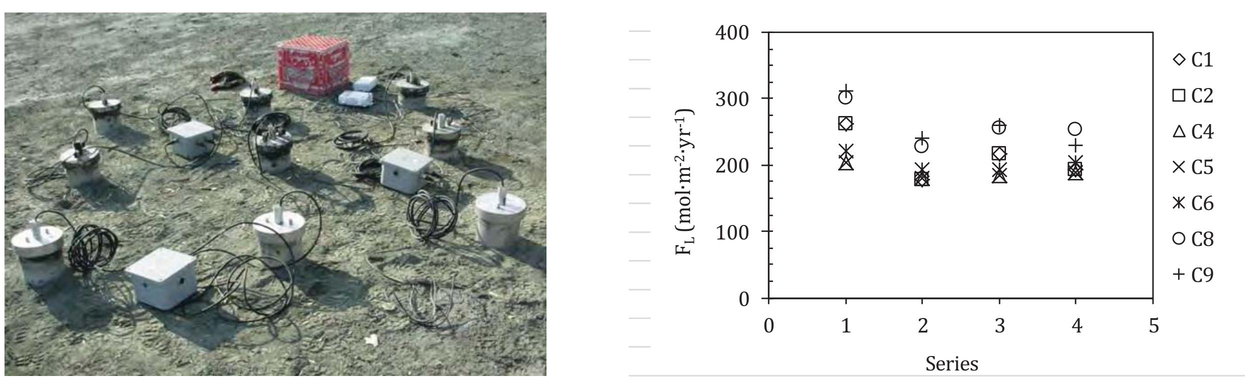
Raglan Mine is located in Northern Quebec, Canada and has operated since 1997 to produce nickel and copper concentrate. The site is located in subarctic desert climate in a region of continuous permafrost. The climate is severe for both wind and weather conditions (mean annual air temperature of -10.3°C). The tailings deposition method used at the site is referred as dry stack tailings, which consists of filtering tailings coming from the milling process to a solid percentage around 85%, so tailings have a "cake" like consistency. Solid tailings are then transported by truck and stored at the tailings storage facility. In this study, tailings temperature, unfrozen volumetric water content and oxygen fluxes were measured at three different locations (S1, S2 and S3) on different configuration of the TSF surface (see Fig. 1-a). Temperature and unfrozen volumetric water content were recorded simultaneously with the 5TM sensor from Decagon Devices. A calibration curve was established in the laboratory to increase the accuracy in volumetric water content measurements in tailings (Coulombe 2012). Sensors were installed at a depth of 5, 15, and 25 cm under the tailings surface and measurements were taken every hour from July 5, 2011 to July 18, 2012. Tailings oxidation rate is characterized with the oxygen consumption test, which gives the oxygen flux (F_L) consumed by the tailings oxidation reaction. The OC test measured the decrease of oxygen concentration over time in a sealed chamber over tailings during a period of three to five hours. The test is performed in a 30 cm stainless steel cylinder of 15 cm diameter which is inserted in the tailings leaving an empty space "h" at the top of the cylinder ("h" in this study ranged between 10.2 cm to 13.8 cm). Oxygen concentration decrease was measured with the SO-110 sensor (Apogee Instruments) having an accuracy of ± 0.02% O_2. A schematic view of a typical monitoring station is show in Figure 1-b. Three OC setups were installed at each monitoring station and between 6 and 8 OC tests were performed in each cylinder from July 2011 to July 2012 at different times of the year. Tests are interpreted with the method described by Elberling et al. (1994), for a maximum decrease in oxygen concentration of 3%. With this method, slope of the plot of the logarithm relative change in oxygen concentration with time gives the parameter $(K_rD_e)^{0.5}$, that is inserted in eq. 1 to obtain the oxygen fluxes consumed by tailings.



a) b)

Figure 1 a) Station locations on the Raglan Mine TSF; b) Schematic view of the oxygen consumption test setup and instrumentation localization (5TM sensors measure temperature (T) and unfrozen volumetric water content (θ_u) simultaneously).

Variability of the OC test was studied on the Raglan Mine TSF. Four series of seven OC tests were performed in a three days period (from July 19 to 21, 2012) on a surface of 4 m^2 between stations S1 and S2 on the flat tailings surface. Tests were interpreted with the approach and the conditions described above (see Coulombe (2012) for details).



a) b)

Figure 2. a) Setup used on TSF to assess the variability of OC tests; b) variability of oxygen fluxes measured with the OC test.

Results presented in Figure 2-b vary from 204 to 312 mol·m^{-2}·yr^{-1}, 178 to 241 mol·m^{-2}·yr^{-1}, 184 to 268 mol·m^{-2}·yr^{-1} and 190 to 254 mol·m^{-2}·yr^{-1} for the four test series (Figure 2-b), representing an oxygen flux variability between 25% and 35%. This variability is mainly due to tailings geochemical and unsaturated hydrogeological heterogeneities. Despite the variability observed with the oxygen consumption test, it is considered here as an appropriate tool to study the *in situ* oxidation rates of sulphide tailings.

MATERIAL CHARACTERIZATION

Tailings were sampled on the Raglan Mine's TSF at stations S1 to S3 and characterized in the laboratory for their physical, chemical and mineralogical properties (Table 1). Specific gravity measured with a helium pycnometer varied between 2.895 and 2.931. Tailings have a typical grain size distribution with a D_{10} (grain size at 10% passing) ranging from 0.0015 to 0.002 mm and 79% to 83% of the particles passing 80 µm. Carbon and sulfur analyses conducted with an induction furnace show that tailings sulfur content varies between 4.38 ± 0.5% and 4.95 ± 0.5%. The main sulphide mineral found by X-Ray diffraction (DRX) in these tailings samples is pyrrothite with traces of pentlandite. The main gangue minerals are lizardite, chlorite, magnetite and hornblende. Tailings samples are considered acid generating because of their sulfide content between 10% and 20%wt and their low neutralization potential (absence of carbonate minerals).

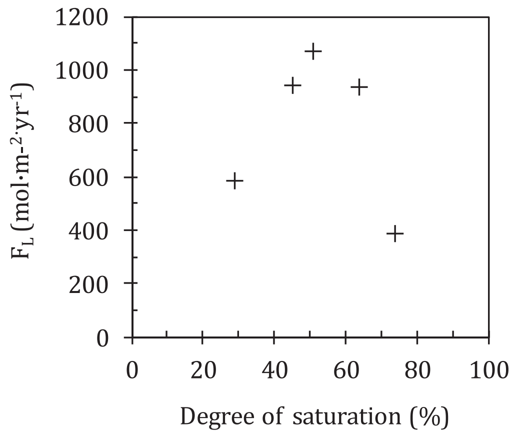
Physical	S1	S2	S3
Specific gravity	2.931	2.930	2.895
Grain size D_{10} (mm)	0.0015	0.0017	0.002
Grain size D_{60} (mm)	0.0229	0.0287	0.0323
C_U	14.8	16.4	16.5
Particles passing 80 µm (%)	83	79	80
S (wt%)	4.38	4.95	4.69
C (wt%)	0.294	0.264	0.185
Mineral content (%)			
Chlorite $(Mg, Al, Fe)_6(Si,Al)_4O_{10}(OH)_8$	12.8	9.2	12.2
Hornblende $Ca_2[Mg_4(Al,Fe)]Si_7AlO_{22}(OH)_2$	9.7	9.2	10.8
Lizardite $Mg_3Si_2O_5(OH)_4$	40.9	38.4	51.5
Magnetite Fe_3O_4	7.7	8.5	12.4
Pentlandite $(Fe,Ni)_9S_8$	0.9	1.4	0.4
Pyrrhotite $Fe(1\text{-}x)S$	10.4	13.2	12.1
Other	17.6	20.1	0.6

Table 1. Main physical, chemical and mineralogical properties of the tailings studied

Oxygen consumption tests on Raglan Mine's tailings were performed in the laboratory to determine oxygen consumption rates at ambient temperature (21°C) for different degrees of saturation. Five OC tests were performed on tailings in a column having a diameter of 14 cm and a height of 29 cm (see Figure 3-a) at porosity between 0.37 and 0.43. Tests were interpreted with the previously described approach of Elberling et al. (1994). Results of the oxygen consumption tests are shown in Figure 3-b. The maximum oxygen consumption rates obtained is 1070 $mol\cdot m^{-2}\cdot yr^{-1}$ at 51% saturation. When the degree of saturation reaches values close to 20% and to 80%, oxygen consumption decreased to values less than 580 $mol\cdot m^{-2}\cdot yr^{-1}$. These laboratory results are consistent with those presented in the literature that show an optimal sulphide oxidation for degrees of saturation between 40% and 60% (Bouzahzah 2013; Godbout

2012). This is explained mainly because gas diffusion in a porous media is negligible at high saturation degree (Aachib et al. 2004) and because not enough water is available to feed the oxidation reaction at low saturation (Godbout 2012).

a)



b)

Figure 3. Laboratory evaluation of tailings oxygen consumption a) column used for OC tests in laboratory; b) Oxygen fluxes measured in laboratory at different degree of saturation and at ambient temperature

MAIN RESULTS

Figure 4 shows the mean (from the three cylinders installed at each station), minimum and maximum oxygen fluxes (4-a), the hourly tailings temperature distribution at 5 cm under the tailings surface (4-b), and the hourly unfrozen volumetric water content distribution at 5 cm under the tailings surface (4-c) for stations S1 to S3. The focus in this study is mainly on sensors located at 5 cm under the tailings surface because tailings oxidation reaction takes place in the first centimetres (Godbout 2012). Oxygen fluxes measured on Raglan Mine's TSF from July 2011 to July 2012 vary between 0 and 550 mol·m^{-2}·yr^{-1}. Tailings' oxygen consumption annual tendency is similar for the three stations but at different intensities. Maximum oxygen fluxes are reached during the months of July (2011 and 2012) with maximum values of 240, 390 and 550 mol·m^{-2}·yr^{-1} for S1, S2 and S3, respectively, when tailings temperature is higher. A maximum tailings temperature of 17.5°C, 12.5°C (incomplete data set) and 25.2°C for S1, S2 and S3 are reached during the months of June or July.

During the period between June and August 2011, the tailings unfrozen volumetric water content remained fairly stable at values ranging between 0.17 and 0.26, which corresponds to value of unfrozen degree of saturation between 40% and 60% (assuming a porosity of 0.43). It is important to remember that a degree of saturation greater than 85% is required to control oxygen diffusion in tailings and sulphide oxidation (Ouangrawa 2007). Tailings temperature reaches 0°C for the first time at mid-September 2011 and remains constant slightly above 0°C until the end of September 2011. Oxygen fluxes are still measurable the 26th of September 2011 when tailings temperature is close to 0°C, with values of approximately 30, 250 and 30 for S1, S2 and S3 respectively. Unfrozen water content varies between values of 0.17 to 0.21 (S1), 0.03 to 0.21 (S2) and 0.09 to 0.18 (S3) from mid-September 2011 to the end of September 2011, during the period where tailings temperature is close to 0°C. Oxygen fluxes reach values lower than 10 mol·m^{-2}·yr^{-1} by the end of October 2011, when tailings temperature and volumetric unfrozen water content values are -4.1, -4.2 and -4.5ºC and 0.01, 0.03 and 0.08 for S1, S2 and S3, respectively. During winter months, unfrozen water content values ranging between 0.01 and 0.05 at 5 cm were measured under the tailings surface. No OC tests were performed during the winter period because snow and ice accumulation was blocking access to the OC test cylinders. Unfrozen volumetric water content slowly started increasing at the end of April 2012, when tailings temperature was close to -8°C, and then increased rapidly when tailing temperature was close to 0°C (in June 2012) to reach values between 0.16 and 0.24 (40% < S_r < 56%), which is similar to data observed during summer 2011.

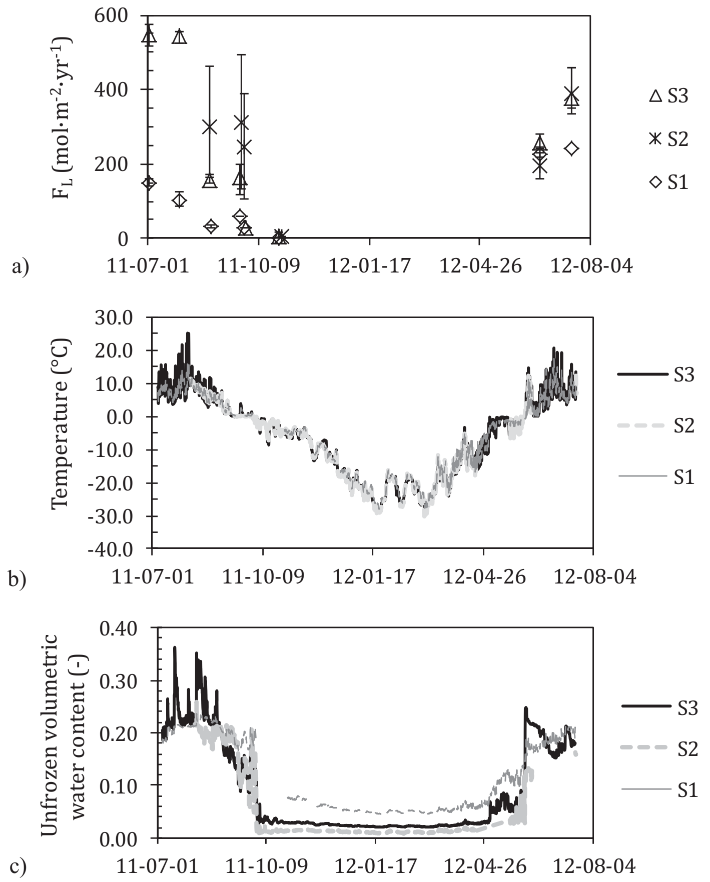


Figure 4. Main results obtained on the Raglan Mine TSF between July 5, 2011 and July 18, 2012; a) Mean, minimum and maximum oxygen fluxes measured; b) hourly tailings temperature distribution at 5 cm under surface; c) hourly tailings unfrozen volumetric water content distribution at 5 cm under the surface

Oxygen fluxes measured after the first winter (in June 2012) were similar to those observed in summer 2011 with values greater than 200 mol·m^{-2}·yr^{-1}. Figure 4 shows that oxygen flux appears to be closely linked with tailings temperature; when tailings temperature decreased, oxygen consumption decreased. Unfrozen volumetric water content (degree of saturation) also influenced tailings oxygen consumption.

The following section further examines the relationships between unfrozen volumetric water content and temperature on tailings oxygen consumption rates.

DISCUSSION

Effect of the Water Content on Oxygen Fluxes

The relationship between oxygen fluxes and tailings unfrozen volumetric water content are shown in Figure 5. Measurements of the oxygen consumption at 21°C performed in UQAT laboratory on tailings sampled on Raglan Mine's TSF are also presented for comparison. *In situ* oxygen fluxes were measured at rates between 30 and 550 mol·m^{-2}·yr^{-1}, for unfrozen volumetric water content values between 0.09 and 0.23, showing that tailings degree of saturation allows the oxidation reactions. Tailings temperature values during these OC tests varied between -0.1°C and 12.8°C. Figure 5 shows that the maximum oxygen flux measured in the field (F_L=550 mol·m^{-2}·yr^{-1}) is almost half the values of the maximum oxygen flux measured in the laboratory at 21°C (F_L=1070 mol·m^{-2}·yr^{-1}). These maximum oxygen flux values in the lab and in the field were observed for unfrozen volumetric water content values between 0.17 and 0.26, which corresponds to degree of saturation between 40% and 60%. Such degree of saturation corresponds to ideal conditions for tailings oxidation reactions to proceed (Bouzahzah 2013; Godbout 2012). At unfrozen volumetric content lower than 0.09, oxygen fluxes drop to values less than 10 mol·m^{-2}·yr^{-1}. Low unfrozen volumetric water content could explain (at least in part) the lower oxygen fluxes of less than 10 mol·m^{-2}·yr^{-1} in October (θ_u = 0.01, 0.03 and 0.08 for S1, S2 and S3). Those tests were performed when tailing temperature was under 0°C (about -4°C). The cold temperature is then another factor that could explain the low reaction rates observed.

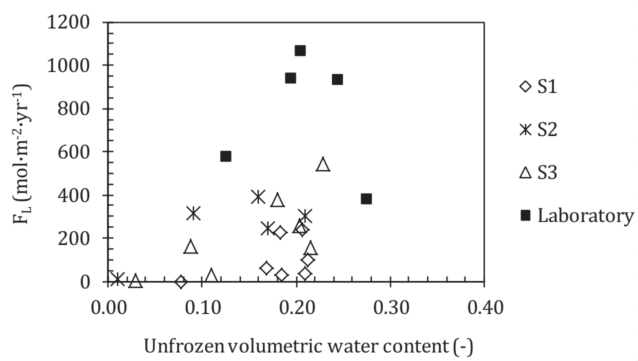


Figure 5. Mean oxygen fluxes measured at different unfrozen volumetric water contents in the fields for tailings temperature between -4.5 and 12.8°C and in the laboratory at 21°C

Temperature Effect on Oxygen Fluxes

The effect of tailings temperature on oxygen fluxes for the monitoring stations is presented in Figure 6-a. As expected, oxygen fluxes diminished with temperature. The highest oxygen fluxes (from 340 to 540 mol·m^{-2}·yr^{-1}) were measured at temperatures between 7.9 and 12.8°C. Oxygen fluxes that decreased gradually with temperature were still measurable at temperatures close to 0°C with values between 30 and 310 mol·m^{-2}·yr^{-1}. OC tests performed at tailings temperature less than -4°C (-4.1°C, -4.2°C and -4.5°C) yield values less than 10 mol·m^{-2}·yr^{-1}. Results suggest that sulphide oxidation reaction was almost stopped for tailings temperature somewhere between -1°C and -5°C. Assuming that a minimum temperature of -6°C is necessary to control sulphide oxidation reactions, tailings would be below this critical temperature value for about 168 days (46% of the year) at station S3 (similar results are observed for the other stations).

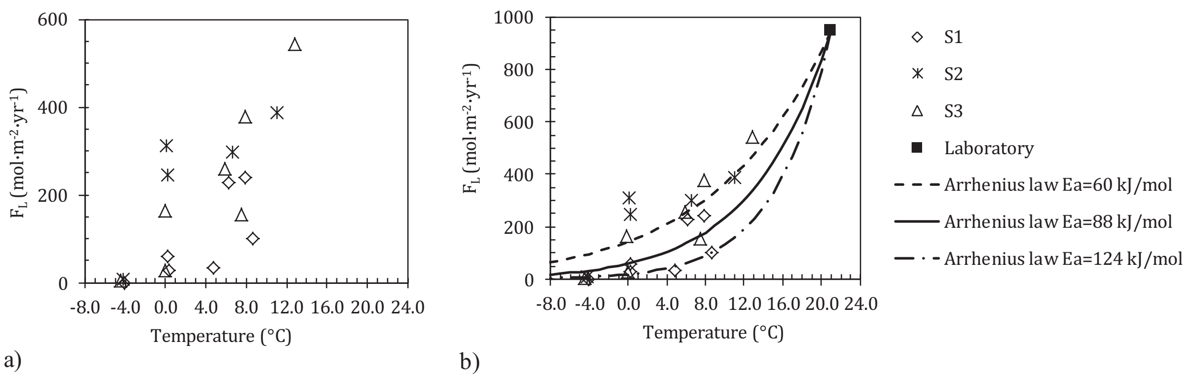


Figure 6. a) Effect of tailings temperature on oxygen fluxes; b) Effect of temperature on oxygen fluxes and prediction with Arrhenius law for different activation energies.

The Arrhenius equation is used to predict the effect of temperature on tailing oxygen fluxes measured at Raglan Mine's TSF (eq. 2), where K_1 and K_2 represent oxygen consumption at temperature T_1 and T_2, E_a is the activation energy and R is the gas constant (Figure 6-b). Three different activation energies have been used (60, 88 and 120 kJ/mol) corresponding to activation energy found in the literature for pyrrhotite (Janzen et al. 2000). A K_1 of 945 mol·m^{-2}·yr^{-1} measured in the laboratory at T_1 of 21ºC is used for the calculation.

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Figure 6-b shows that Arrhenius' law accurately predicts field data for temperatures above 0°C with activation energy of 60 kJ/mol, considering that temperature is not the only parameter affecting oxygen fluxes. Results also show that Arrhenius' law seems to predict oxygen consumption rates at temperatures below 0°C more precisely with higher activation energy (between 80 to 124 kJ/mol). Nevertheless, more work is needed to better understand the fundamental aspects related to the influence of temperature on pyrrhotite oxidation.

<u>Spatial Variation of Pyrrhotite Content</u>

Figure 4-a demonstrates that oxygen fluxes can be quite different from one station to the other for an OC test performed the same day in similar conditions. As mentioned in previous sections, tailings degree of saturation is an important parameter controlling the oxygen diffusive flux and temperature affects the kinetics of sulphide oxidation reaction. However, tailings temperature and unfrozen volumetric water content distribution is generally similar from one station to the other (see Figure 4-b and 4-c). Hence, other factors must influence tailings oxygen consumption rates (Coulombe 2012). One of them is

pyrrhotite content. Indeed, a study has shown that the tailings reaction rate coefficient (K_r) varies linearly with relative sulphur content (Collin 1998). To verify this statement, an annual mean oxygen flux is calculated with OC tests results from August 26, 2011 to July 17, 2012 (n = 6) and related to pyrrhotite content (C_p) for station S1 (C_p = 10.4%), S2 (C_p = 13.2%) and S3 (C_p = 12.1%). Annual mean oxygen fluxes of 100, 240 and 170 mol·m^{-2}·yr^{-1} are calculated for stations S1, S2 and S3 respectively. A linear relation with a correlation coefficient of 0.9849 is observed between tailings sulphide content and annual oxygen fluxes (see Coulombe (2012) for more details). Therefore, tailings sulphide content could also be a cause of the variation observed in tailings oxygen consumption rates at Raglan Mine's TSF.

CONCLUSION

In this paper, the influences of temperature and unfrozen volumetric water content on tailings oxygen consumption were studied on Raglan Mine's TSF. Maximum oxygen fluxes of 550 and 1070 mol·m^{-2}·yr^{-1} were measured in the field and laboratory for unfrozen volumetric water content values between 0.17 and 0.26 ($40 < S_r < 60\%$). The study showed that, as expected for dry stack tailings (Bussière 2007), the tailings saturation at Raglan Mine's TSF is not sufficiently high to control oxidation reactions. It is also shown, as expected, that oxygen fluxes decreased with temperature. Tailings oxygen consumption was greatly reduced when tailings temperature reached values below -6°C, conditions observed for approximately 50% of the year. It was confirmed that Arrhenius' law can be used to estimate the effects of temperature on measured oxygen fluxes with activation energy between 60 and 124 kJ/mol. This study also showed that significant differences are observed when assessing field oxygen fluxes at different locations on the same tailings stack due to mineralogical and unsaturated hydrogeological heterogeneities.

Results of this field study demonstrate that cold temperatures can be used to control AMD generation, but appropriate conditions must be reached. This study was completed as part of a larger effort to increase the understanding of Raglan Mine's tailings, their reactivity, and the factors that influence tailings reactivity in the current closure concept. The operation is currently in a pre-feasibility stage study using intermediate scale instrumented experimental cells testing different cover concepts and their effects on mitigating the factors that influence tailings reactivity in an effort to update the current closure concept.

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Overcoming Northern Challenges

Proceedings of the 2013 Northern Latitudes Mining Reclamation Workshop and

38th Annual Meeting of the Canadian Land Reclamation Association

Whitehorse, Yukon September 9 – 12, 2013

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Northern Latitudes Mining Reclamation Workshop

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Polster	Natural Processes: An Effective Model For Mine Reclamation
Dustin	Implementation of contaminated water management system upgrades to allow for dewatering of two open pits at the Vangorda Plateau, Faro Mine Complex, Yukon
Kempenaar, Marques and McClure	Tools for Arctic Revegetation: What's in Your Toolbox?
Smreciu, Gould, and Wood	Establishment of Native Boearl Plant Species On Reclaimed Oil Sands Mining Disturbances
Keefer	Twin Sisters Native Plant Nursery
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Janin	Passive treatment of drainage waters: Promoting metals sorption to enhance metal removal efficiency
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Nadeau and Huggard	Restoration Planning and Application of Ecological Succession Principals
Simpson	Defining Disturbance and Recovery - the influence of landscape specific ecological responses to oil and gas linear disturbances in Yukon

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- To collect information relating to land reclamation and publish periodicals, books and leaflets which the Association may think desirable.
- To encourage education in the field of land reclamation.
- To provide awards for noteworthy achievements in the field of land reclamation.

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