Passive treatment of high-iron acid mine drainage using sulphate reducing bacteria: comparison between eight biofilter mixtures.

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Abstract Eight different biofilter mixtures were compared in batch and column tests for their capacity to treat a high iron acid mine drainage (pH 3.5, $[SO_4^{2-}]$ = 9000 mg/L and [Fe] = 4000 mg/L). For most mixtures in batch test, the pH increased to 6.5, Eh decreased below 0 mV after 10 days, and dissolved metals concentrations were reduced by more than 99%. This series of batch tests showed that (1) mixtures based on chicken, cattle or sheep manures give similar results, (2) SRB inocula coming from sediments are not essential, and (3) mixtures containing 50% of calcite or sand performed well. The second series of tests were performed in columns (11 L, hydraulic retention time of 5 to 7 days) for three selected mixtures based on batch tests results. Neutralization was effective for all columns and metal removal efficiency reached values higher than 90% for Al, Cd, Cr, Ni, Zn, approximately 80% for Pb, and between 30 and 40% for Mn. Nevertheless, iron removal for the tested AMD never exceeded 50% for the three mixtures.

Key Words mine water treatment, sulphate reducing bioreactor, biofilter mixture.

Introduction

Mine water contamination from mine sites comes mainly from a phenomenon called acid mine drainage (AMD). AMD generation consists of a chain of chemical and biochemical reactions due to sulphide minerals oxidation in tailings impoundment, waste rock pile, mine openings, etc. (Aubertin et al. 2002); increasing acidity and dissolved metals concentration. During mining operation, conventional active treatments are usually used to treat AMD in order to respect the existent legislations concerning liquid effluents (Aubertin et al. 2002). Recently, passive systems like biofilters with sulphate-reducing bacteria (SRB) have been proposed to neutralize and remove metal from AMD (e.g. Neculita et al. 2007). This approach is particularly adapted for abandoned mine sites often located in remote areas. Sulphate reducing biofilter contains mainly a mixture of carbon source which allows production of hydrogen sulphide and alkalinity by electron exchanges during the SRB metabolism (Neculita et al. 2007). These reactions lead to precipitation of dissolved metal. However, studies (Potvin 2009, Neculita and Zagury 2008) show that metal sulphides precipitation is not the only metal removal mechanisms. Metals can be also extracted from the contaminated water by sorption, precipitation or coprecipitation of hydroxides and carbonates (Neculita and Zagury 2008). Performances of SRB bioreactors depend mainly on carbon source and on contact time between AMD's sulphates and bacteria. Indeed, many authors worked on mixture optimization (e.g. Cocos et al. 2002, Neculita and Zagury 2008). Recent works of Potvin (2009) and Neculita et al. (2008) demonstrate that an appropriate mixture can treat an AMD with iron concentrations (around 500 mg/L) for a 10 days hydraulic retention time. The present study aims at evaluating the capacity of SRB biofilters to treat a high iron concentrated AMD (between 1000 and 4000 mg/L), typical of those found on numerous hard rock mine sites in Canada (Aubertin et al., 2002).

Materials and methods

Before designing a full scale biofilter, the reactive mixture should be optimized in laboratory. This is done mainly in three steps. The first step is performed in batch tests (approximately 1 L) (Neculita and Zagury 2008, Cocos et al. 2002). This step allows preselecting the most promising mixture. In the second step, column tests (»10 L) are performed to verify the effectiveness of the related mixture (Beaulieu 2001, Neculita et al. 2008). This step is also need to define the hydraulic retention time. Finally, medium size tests (50 L or more) can be used to evaluate the scale effect (Potvin 2009). The full scale passive biofilter is constructed once the mixture selection steps are performed. The first two steps of the proposed design approach are presented in the present study.

Table 1 Biofilter mixtures compositions

Dry weight %	#1	#2	#3	#4	#5	#6	#7	#8
Maple chips	10	10	10	5	10	10	6	10
Maple sawdust	20	20	20	10	20	20	11	20
Chicken manure	10			5		10	8	10
Cattle manure		10						
Sheep manure				10				
Compost	20	20	20	10	20	20	12	20
sand	20	20	20	10	20	35	50	20
Sediment	15	15	15	8	15		8	15
Urea	3	3	3	2	3	3	3	
Calcium carbonate	2	2	2		2	2	2	
Calcite sand				50				5
Municipal sludges					10			

Table 2 AMD compositions

Concentrations	mg/L
Al	1
Cd	0,5
Cr	1
Fe (in AMD)	4000
Fe (in AMD light)	1000
Mg	10
Mn	10
Ni	2
Pb	0,5
SO_4^{2-}	9000
Zn	0,5
pH	3

Eight mixtures were tested on a synthetic high-iron acid mine drainage using batch tests. Mixtures proportions (% dry weight) are described in Table 1. The composition of mixture #1 comes from Neculita and Zagury (2008). Mixtures #2, #3 and #5 were tested to evaluate the possibility of using other sources of organic material (Cocos et al. 2002). Mixture #6 allowed comparing the efficiency of the same mixture with or without a sediment SRB inoculum. Mixture #4 and #7 are tested to investigate the effect of mixture composition on the saturated hydraulic conductivity (sand size particle addition; Rötting et al. 2008). Finally, mixture #8 was not boost by urea (nitrogen nutriment for SRB) and contained a low cost neutralizing agent (calcite sand). Initially, each material used in the biofilter was physically and chemically characterized. More details on the characterization will be presented in further publication (Genty 2011).

The first series of batch tests was performed in 1L glass reaction flasks, at room temperature (21 °C) under nitrogen atmosphere. The ratio between mixture and AMD was 200 g : 600 mL. AMD composition is given in table 2 and is typical of an AMD coming from hard rock mines (Aubertin et al. 2002). Parameters such as pH, oxydo-reduction potential called Eh (values are corrected relative to the standard hydrogen electrode), sulphates and metals were measured during 40 days.

The second series of tests was performed in columns (14 cm diameter and 70 cm height) for optimal mixtures #1, #4 and #7. Two columns (#4 and #7) were duplicated. Columns were fed from the bottom with AMD (see table 2) to allow constant anoxic conditions. A perforated plastic plate covered with a geotextile was placed at the bottom to uniformly feed the column. During the first 173 days and after 291 days, the upflow rate was set to have a hydraulic residence time (HRT) of approximately 5 days; between 173 and 291 days, the hydraulic residence time was increased to 7 days. After 55 days, AMD iron concentration was decreased to 1000 ppm (called AMD light in Table 2) and after 229 days increased to the initial value of 4000 ppm. The change in iron concentration allowed evaluating the effect of iron on the treatment efficiency. The pH, Eh, sulphates, acidity, alkalinity, metals and saturated hydraulic conductivity were monitored during the experiment. SRB enumeration was performed on the effluent of the two duplicates #4 and #7 using the Most Probable Number technique (Beaulieu, 2001).

Results and discussion
Batch tests results
Some results obtained during batch tests are discussed in this section. The pH in each flask reached 7 after 10 days, except for mixture #8 which needed 30 days (figure 1a). The oxydo-reduction potential of each flask decreased below 0 mV (reducing conditions), except for mixtures #4 and #8 which stayed around 200 mV (figure 1b). All flasks removed more than 99% of iron before 20 days (figure 1c). As regard to sulphates concentration, there was no significant decrease before 20 days, which corresponded to the lag period for SRBs (Cocos et al. 2002). This last result shows that during the first days of biofilter operation, sorption, hydroxides and carbonates precipitation are probably the main mechanisms of metal removal. After 40 days, the sulphates concentration decreased by a proportion between 25 and 75% depending on the mixture (figure 1d). Since mixture #1, #2, #3, #5 gave similar results, different organic material sources could be used in a SRB biofilter. Mixture #6 demonstrated that a SRB inoculum coming from sediments was not essential. A bacterial enumeration performed on manure and compost showed also the presence of SRB. Finally, mixtures #4 and #7 also performed well. Consequently, three mixtures (#1, #4 and #7) were selected based on the results of batch tests and were investigated further in column tests.
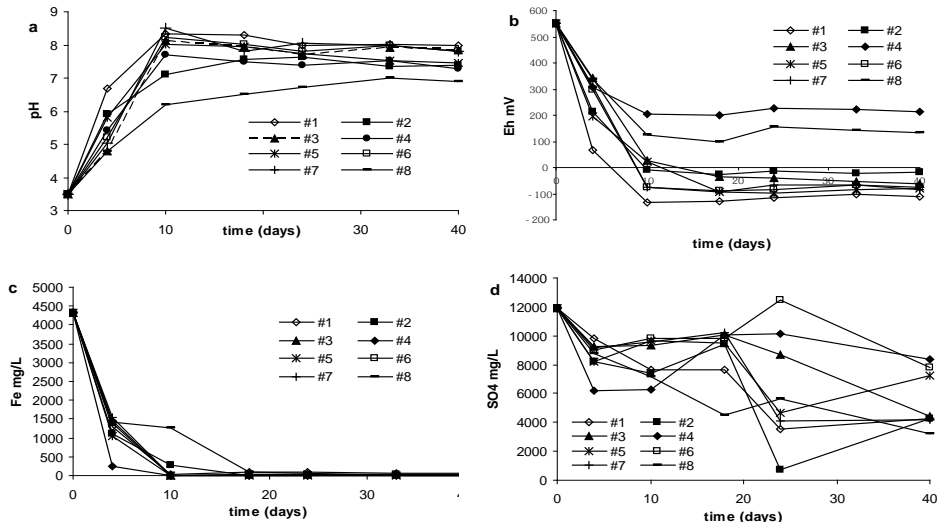
Figure 1 (a) pH, (b) Eh, (c) iron removal % and (d) sulphates in batch tests

Column tests

Figures 2a to 2d show the main columns results for pH, Eh, sulphates and iron removal.

Reactive mixture #1, #4 and #7 increased the pH up to an average value of 6.1. The oxydo-reduction potential (figure 2b) decreased from an average of 550 mV for the AMD to 30 mV for the columns effluent whatever the HRT and iron concentration. Although the Eh was not negative, sulphate reduction could happen in these conditions (Neculita et al. 2008). Indeed, SRB enumeration performed on the effluent of duplicated columns #4 and #7 gave $2 \cdot 10^2$ BSR/100 mL after 117
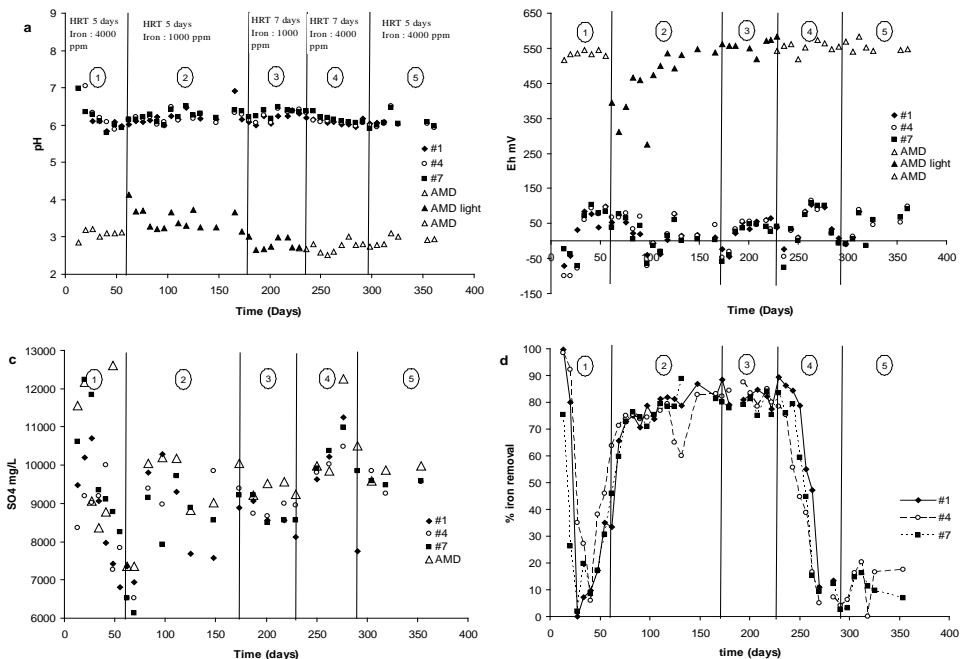

Figure 2 (a) pH, (b) Eh, (c) sulphates and (d) iron removal % in column test with mixture #1, #4 and #7

days. Alkalinity production varied from 200 to 800 mg/L $CaCO_3$ depending of the mixture, with higher values for mixture #1. Figure 2c shows that sulphates removal average percentage was approximately 16% for the column with mixture #1, 17% for mixture #4 and 4% for mixture #7 during the first period (10—55 days). For period 2 to 5, the sulphates removal average percentage never exceeded 5% whatever the columns. During the first period, iron removal dropped quickly and stabilized at values between 10 and 40%. Then, for periods 2 and 3 (AMD light), iron removal rose up to 80%. During these two periods, HRT (5 or 7 days) seems to have no influence on iron removal. During periods 4 and 5, iron removal decreased quickly to stabilize between 0 and 20%. Based on these results, a HRT of 5 days seemed as efficient as 7 days and AMD light was easier to treat than AMD. Although iron removal was not as high as expected, for all the experiments, Al, Cd, Cr, Ni, Zn removal rose up to 90%, Pb removal was between 52—80% and Mn removal varied between 1 and 28%; the last removal percentage is in accordance with the literature which shows that SRB biofilters do not treat efficiently Mn (Neculita et al. 2008). Permeability test results showed that there was no significant change in terms of saturated hydraulic conductivity during tested periods with values of approximately $3 \cdot 10^{-3}$ cm/s for mixture #1, $1 \cdot 10^{-3}$ for mixture #4 and from $1 \cdot 10^{-3}$ to $1 \cdot 10^{-2}$ cm/s for #7. Finally, duplicated columns for mixtures #4 and #7 indicated that experiments were reproducible since the water quality was nearly identical at the exit of the columns during 62 days (the statistic analysis using the paired-different test is not presented in this paper).

Conclusion

Batch tests were used to identify the most promising mixtures among eight for a SRB biofilter. Column tests were performed to evaluate the impact of hydraulic retention time on SRB biofilters capacity to remove dissolved iron. Results showed that biofilters alone cannot treat efficiently high-iron AMD although they can be efficient for low iron contaminated AMD (<500 mg/L; see Neculita et al., 2008). Hence, a combination of passive treatment systems (e.g. Champagne et al. 2005) is probably the best option to treat highly contaminated AMD; SRB biofilter could be an important part of this treatment combination.

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