Bioresource Technology 102 (2011) 683-689

Contents lists available at ScienceDirect

Bioresource Technology

journal homepage: www.elsevier.com/locate/biortech

Sulfidogenic fluidized bed treatment of real acid mine drainage water

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ARTICLE INFO

Article history: Received 1 July 2010 Received in revised form 15 August 2010 Accepted 16 August 2010 Available online 24 August 2010

Keywords: Acid mine drainage Fluidized bed reactor Metal precipitation Sulfate reduction

ABSTRACT

The treatment of real acid mine drainage water (pH 2.7–4.3) containing sulfate (1.5–3.34 g/L) and various metals was studied in an ethanol-fed sulfate-reducing fluidized bed reactor at 35 °C. The robustness of the process was tested by increasing stepwise sulfate, ethanol and metal loading rates and decreasing feed pH and hydraulic retention time. Highest sulfate reduction rate (4.6 g/L day) was obtained with feed sulfate concentration of 2.5 g/L, COD/sulfate ratio of 0.85 and HRT of 12 h. The corresponding sulfate and COD removal efficiencies were about 90% and 80%, respectively. The alkalinity produced in sulfidogenic ethanol oxidation neutralized the acidic mine water. Highest metal precipitation efficiencies were observed at HRT of 24 h, the percent metal removal being over 99.9% for Al (initial concentration 55 mg/L), Co (9.0 mg/L), Cu (49 mg/L), Fe (435 mg/L), Ni (3.8 mg/L), Pb (7.5 mg/L) and Zn (6.6 mg/L), and 94% for Mn (7.21 mg/L).

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1. Introduction

Metal-containing acid mine drainage (AMD) is created by the interaction of air and water with sulfides, such as pyrite (FeS₂), found in overburden piles and also in mine shafts consisting of sub-commercial grade mining material left over from the mining operations.

Other sulfide minerals are oxidized in a similar way as pyrite, releasing metals and sulfate in solution and therefore, AMD may contain several metals such as Cu, Fe, Zn, Al, Pb, As and Cd at high concentrations. AMD is acidic in nature and generally has low organic carbon content. If uncontrolled, AMD flows into local streams, lakes and rivers, contaminating soils and destroying plant and animal biota. The formation of acidic wastewaters can continue even tens and hundred years after mine closure if the conditions remain favorable (Szczepanska and Twardowska, 1999).

Sulfate reducing bioreactors are becoming an alternative to conventional chemical processes for the treatment of acidic and metalcontaining wastewaters due to high efficiency (Sahinkaya, 2009). In the presence of organic compounds, sulfate is microbially reduced to hydrogen sulfide under anaerobic conditions and heavy metals (especially, Cu, Pb, Zn, Cd, Ni and Fe) form stable precipitates with the produced sulfide. Moreover, produced bicarbonate increases the pH of the wastewater. This way, metals and sulfate are concomitantly removed and pH can be increased to neutral values in a single reactor (Kaksonen et al., 2003a,b; Nagpal et al., 2000a,b).

Ethanol is a good substrate for sulfate reducing bacteria and suitable for large scale applications (Kaksonen et al., 2003a; Nagpal et al., 2000a). Under sulfidogenic conditions, ethanol is first converted to acetate and in the second step acetate may further be oxidized to CO_2 . In this case, acetate oxidation is obligatory to produce alkalinity (reactions (1) and (2)) as the first step (conversion of ethanol to acetate) does not produce alkalinity (Sahinkaya, 2009). Some SRB oxidize organic substrates completely to CO_2 , while others incompletely to acetate (Sahinkaya, 2009). It is well known that acetate may accumulate in the reactor at high loadings, which may decrease the sulfide and alkalinity production (Kaksonen et al., 2004; Sahinkaya and Yucesoy, in press; Bayrakdar et al., 2009). Hence, in the case of ethanol supplementation, the loading rate should be carefully adjusted not to accumulate acetate and cease alkalinity production:

$$\begin{array}{ll} 2CH_{3}CH_{2}OH+SO_{4}^{2-}\rightarrow 2CH_{3}COO^{-}+HS^{-}+H^{+}+2H_{2}O & (1) \\ CH_{3}COO^{-}+SO_{4}^{2-}\rightarrow 2HCO_{3}^{-}+HS^{-} & (2) \end{array}$$

Most of the studies on the treatment of metal-containing wastewater using bioreactor processes have been conducted with synthetic AMD water containing few metals for the sake of simplicity, whereas, real AMD may contain a number of metals, which makes biotreatment of real AMD more challenging. In this context, this study aims at evaluating the efficiency of an ethanol supplemented sulfidogenic fluidized bed reactor (FBR) for the treatment of real AMD water containing high concentrations of sulfate and various metals, and high acidity.



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2. Methods

2.1. Bioreactor

A laboratory scale FBR (Fig. 1) inoculated with anaerobic digester effluent was used in the study. Concentrated anaerobic digester



Fig. 1. Schematic diagram of fluidized bed reactor.

Table 1

Operational conditions of FBR.

effluent was injected to the reactor to have an initial volatile suspended solids content of 1 g in the FBR. After the injection of the anaerobic consortia, the reactor was run in recirculation mode for 2 days without feeding to let the biomass to attach on the carrier material. The FBR was maintained in a temperature controlled room and operated at about 35 °C. The active bed volume of the FBR was 300 mL. Activated carbon (particle size range of 0.5-1 mm) was used as biomass carrier and bed fluidization was 15-20%. During the reactor operation of around 220 days, we did not observe significant abrasion of the media due to inter-particle collision caused by liquid flow. For both reactors, the recirculation flow rate was between 60 and 70 mL/min, giving the recirculation ratio of 150–300 depending on the feed flow rate. The reactor can be considered as completely mixed due to high recirculation rate. Similarly, Chung et al. (2006) stated that a recirculation ratio of 150 promoted completely mixed conditions.

The FBR was fed with AMD water collected from a copper mine area near Elazığ, Turkey. The AMD water was collected three times (between August 2009 and May 2010) during around one year to determine the variation in the composition and to study the robustness of the FBR.

Ethanol was supplemented as a carbon and electron source for sulfate reducing bacteria (SRB). For the first 64 days (period 1), as a start-up strategy, the FBR was fed with a metal deficient synthetic wastewater containing 2000 mg/L sulfate to enrich sulfate reducing bacteria (SRB) (Table 1). After start-up (period 1), in period 2, the FBR was fed with AMD1 to adapt SRB to the real AMD as AMD1 was the most dilute AMD. The pH and sulfate concentrations and metal concentrations of the AMD1, 2 and 3 were presented in Tables 1 and 2, respectively. In periods 3 and 4, the FBR was fed with AMD2, which was the most concentrated AMD. In order to see the impact of COD/sulfate ratio on the reactor performance, COD/sulfate ratio was kept at 0.67 and 0.85 during

Parameter	Periods										
	1	2	3	4	5	6	7				
Days	0-64	64-74	74-100	100-134	134-163	163-188	188-220 ^a				
Feed	Synthetic	AMD1	AMD2	AMD2	AMD3	AMD3	Simulated AMD2				
Feed sulfate (mg/L)	2000	1500	3360	3360	2500	2500	3500				
Feed ethanol (mg COD/L)	1340	1005	2180	2900	1950	2150	2975				
COD/sulfate	0.67	0.67	0.67	0.85	0.78	0.85	0.85				
HRT (h)	24	24	24	24	16	12	24				
Feed pH	6.5–7	4.3	3.04	3.04	2.7	2.7	3				

^a Except for days 191–196 when metal deficient water was used to recover the FBR.

Table 2

Average influent metal of	concentrations (mg	(L) for each	period.
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Periods	Days		Al	Ca	Cd	Со	Cr	Cu	Fe	К	Mg	Mn	Na	Ni	Pb	Zn
2 (AMD 1)	64-74	Conc. Stdv.	0.16 0.01	236 3	0.002 0.00	2.19 0.30	0.03 0.01	3.98 0.30	2.27 0.20	1.89 0.12	368 7	6.48 1.10	10.69 1.20	0.77 0.05	0.99 0.02	3.01 0.30
3 (AMD 2)	74–100	Conc. Stdv.	55.36 0.17	120 2	0.01 0.01	9.05 0.48	0.09 0.05	45.48 1.03	435 49	19.54 0.37	366 0.10	7.21 2.61	13.11 1.04	3.79 0.13	7.48 3.37	6.64 0.83
4 (AMD 2)	100–134	Conc. Stdv.	53.05 3.13	111 4	0.01 0.01	8.26 0.66	0.08 0.04	49.17 2.90	354 14	18.35 1.11	368 4	5.23 0.37	9.91 0.73	3.66 0.17	4.93 ND	5.27 0.31
5 (AMD 3)	134–163	Conc. Stdv.	39.40 2.26	122 7	0.00 0.00	5.34 0.08	0.07 0.01	38.47 1.06	164 19	16.58 1.32	248 16	3.72 0.03	9.28 0.01	2.50 0.03	ND ND	3.90 0.17
6 (AMD 3)	163-188	Conc. Stdv.	44.98 1.70	116 5	0.00 0.01	6.01 0.17	0.18 0.08	39.69 0.90	139 5	15.85 0.41	245 25	2.97 1.68	13.73 2.20	1.69 1.43	5.39 4.63	2.57 2.20
7 (AMD 2)	188-220*	Conc. Stdv.	17.61 28.37	48 72	0.00 0.01	7.11 0.88	0.10 0.08	45.65 3.16	260 88	26.23 9.70	253 9	6.86 2.24	13.04 ND	3.52 0.70	9.01 0.54	7.43 2.88

Except for days 191-196 when metal deficient water was used to recover the FBR.

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