## TECHNICAL ARTICLE

# Treatment of acidic coal mine drainage: design and operational challenges of successive alkalinity producing systems

Jayanta Bhattacharya · Sang Woo Ji · Hyeon Seok Lee · Young Wook Cheong · Gil Jae Yim · Jeong Sik Min · Yong Suk Choi

Received: 21 September 2006/Accepted: 6 July 2007/Published online: 7 November 2007 © Springer-Verlag 2007

**Abstract** Successive alkalinity producing systems (SAPS) provide a popular bioremediation alternative to anoxic limestone drains for acid mine drainage treatment. This paper reviews the designing of SAPS and then discusses the performance of an existing SAPS-based system in Hanchang coal mine of Kangwon province of Korea. The results indicate that sulfate reduction ceased in the VFR within the first year of operation, implying that metal hydroxide precipitation and adsorption prevailed over metal sulfide formation and precipitation.

**Keywords** Design · Iron removal · South Korea · Successive alkalinity producing systems (SAPS) · Sulfate removal · Performance

## Introduction

In many areas prone to acid mine drainage (AMD), passive treatment systems have proven to be more cost effective than conventional treatment plants. This is especially true when potentially responsible parties cannot be identified, or when they have insufficient funds to pay for conventional water treatment.

In South Korea, abandoned coal mines contribute as much as 12,000 m<sup>3</sup>/day of mine drainage to surface

J. Bhattacharya (🖂)

S. W. Ji · H. S. Lee · Y. W. Cheong · G. J. Yim · J. S. Min · Y. S. Choi Geological and Environmental Hazards Divison, KIGAM, 30 Gajeong-Dong, Yuseong-Gu, Daejeon 305-350, Korea streams (Cheong et al. 2001). Successive alkalinity producing systems (SAPS) have been moderately successful in such applications. SAPS are the most commonly used passive treatment approach in South Korea because the variation in iron removal rates is relatively small, compared to other passive treatment processes. However, the investment in design and construction of SAPS is based, in South Korea at least, on the assumption that a SAPS will efficiently lower acidity and sulfate concentrations for 5 years. This study was initiated at the Hanchang coal mine to understand the challenges of effective SAPS design and performance.

The mine is located in the southern part of the Jeongseon coalfield of South Korea, and was closed in 1993. A passive treatment system was constructed there by the Coal Industry Promotion Board (CIPB, 2004) in June 2001. The treatment system consists of an ALD inside the mine adit, two SAPS units, an oxidation pond, an aerobic wetland, and a small pond (Fig. 1). The treatment system was designed to treat 300 m<sup>3</sup>/day. A waste rock pile is 70–80 m away from one side of the treatment system, and the drainage from that pile does affect the system, as explained later. A spring at a farmhouse, which had been used for drinking water but is not used nowadays due to the contamination, is about 200 m away from the mine. Drinking water is now supplied from further upstream in the valley.

### Method

Water samples were collected from February 2002 to January 2004 from each stage of the passive treatment system (Fig. 2), along with spring water (GW) and drinking water (DW) at the farmhouse. The pH, ORP, temperature, DO, conductivity, and TDS were measured

Department of Mining Engineering, Indian Institute of Technology, Kharagpur 721302, India e-mail: jayantaism@gmail.com



Fig. 2 Map showing treatment

locations in the Hanchang coal

mine



in-situ. Water samples for cation determination were filtered and acidified to pH <2.0 by adding conc. HNO<sub>3</sub>. Acid was added to lower the pH to near 4.5, and BaCl<sub>2</sub> was added to precipitate dissolved sulfate as BaSO<sub>4</sub>, which was recovered by centrifuging. Dissolved cations were determined using an inductively coupled plasma atomic emission spectrophotometer (ICP-AES; Jobin Yvon Co. 138 Ultrace) at the Seoul branch of the Korea Basic Science Institute (KBSI). Anions were determined using the

ion chromatograph (IC; Dionex series 500DX) at the Busan branch of the KBSI, and alkalinity was determined by titration. Acidity was calculated by equation, as suggested by Hedin et al. (1994)

A sulfur isotope study was carried out to identify the sources of sulfur. Since sulfate reducing bacteria need less energy to break the <sup>32</sup>S–O bond than the <sup>34</sup>S–O bond, the reduction of <sup>32</sup>S is faster than that of <sup>34</sup>S. As reduction proceeds, <sup>34</sup>S accumulates in the residual sulfate. The  $\delta^{34}$ S

value of residual sulfate can be enriched by up to 25‰ (Krouse et al. 1970). Böttcher et al. (2001) reported that  $\delta^{34}$ S values increased by 11–18.4‰ under biotic anoxic conditions. Habicht et al. (1998) reported that the sulfur isotope fractionation by sulfate reducing bacteria in fresh water ranges between 5 and 46‰, and that the decrease of  $\delta^{34}$ S in marine sediments by sulfate reduction ranges up to 70‰.

Samples for sulfur isotope analysis were collected in 500 mL polyethylene bottles. Sulfur isotope composition was analyzed using a PRISM II mass spectrometer (model VG Isotech, UK) at Daejeon headquarter of the KBSI, and was expressed in CDT (Canyon Diablo Troilite). The precision of analysis was  $\pm 0.2\%$ .

#### **Design of SAPS: considerations and review**

#### AMD entrainment

The design of SAPS is not an exact science. Typically, the water contains dissolved oxygen and ferric iron, making it unsuitable for treatment using an anoxic limestone drain (ALD). In such applications, a sedimentation pond is often placed in front of the SAPS vertical flow pond, to capture as much of the ferric iron as possible and thereby reduce the amount that has to be reduced. However, at our site, the water was only partially oxygenated (typically 2-4 mg/L, ranging up to 6 mg/L), and had been partially neutralized by an ALD that had been constructed inside the mine. The ferric iron concentrations were low (25-50 mg/L), so rather than aerate it and then attempt to deoxygenate it, it was entrained using full cross-sectional flow directly into a vertical flow reactor (VFR) column containing spent mushroom compost substrate (SMS, Beyer 1999), where the bacterial activity would consume the oxygen and reduce the ferric iron(Fe<sup>+3</sup>), rendering limestone neutralization possible.

#### Spent mushroom compost substrate

Spent mushroom compost substrate (SMS) is an organic waste produced during commercial mushroom production. The major ingredients of SMS are straw, hay, peat, horse manure, poultry manure, and gypsum. In addition to these bulk components, SMS may contain some nutrient additives, such as potassium salts, ammonium nitrate, super phosphate, etc. On average, fresh SMS is approximately 60% water (by mass) (Levanon and Danai 1995), whereas  $\approx 65\%$  of the dry matter is organic (Gerritis 1994). The major inorganic cations in SMS leachate are K<sup>+</sup>, Na<sup>+</sup>, Ca<sup>2+</sup>,

and Mg<sup>2+</sup>. With increased weathering, NH<sub>4</sub><sup>-</sup> decreases and becomes undetectable. Dominant inorganic anions like Cl<sup>-</sup>, SO<sub>4</sub><sup>-2</sup>, and NO<sub>3</sub><sup>-</sup> in SMS leachate range from 27 to 196, 79 to 324, and 0.03 to 118 mmole L<sup>-1</sup>, respectively (Guo et al. 2001). The dissolved organic content (DOC) and the dissolved organic nitrogen (DON) concentrations range from 800 to 11,100 mg L<sup>-1</sup> and from 70 to 1,900 mg L<sup>-1</sup>, respectively, depending on the substrate column length and composition (Guo et al. 2001).

#### Design aspects

Sulfate reducing bacteria can acclimate to temperatures of 40°C to as low as 1°C, with the sulfate reduction rate only decreasing up to 20% over this range (Gusek et al. 2002). Typical sulfate reduction rates range from 200 to 600 mmole/m<sup>3</sup>/day but an average rate of 300 mmole/m<sup>3</sup>/ day has been recommended for the design of sulfate reduction systems (Wildeman et al. 1994) The above equation means that a daily supply of 150-400 (average 300) mmole/m<sup>3</sup> of organic matter needs to be available (molar ratio being 2:3). Carbon production in natural wetlands has been reported to be about 1 kg/m<sup>2</sup>/year (Whittakar and Likens 1972). A 1 m thick substrate can provide 23 mmole of carbon/m<sup>3</sup>/day. If one assumes that only 50% of the carbon molecules in the column react, a VFR with an approximate cross-section of 30 m<sup>2</sup> will be required  $(300 \times 2/23)$ .

Column studies by Thomas et al. (2006) indicated that Fe, Al, Cu, Zn, and Ni can all accumulate in the substrate over time, although the predominant phase of accumulation changes with loading rate and time for these metals. Aluminum solubility is governed largely by water pH and is unaffected by the oxidation and reduction processes that affect other metals (Stumm and Morgan 1996). Dissolved Al concentrations remain high at pH 3.5 or less, but decrease to less than 1 mg/L at a pH between 5 and 8 (Hedin et al. 1994). The Al compounds in the column are generally hydroxides and sulfates (Thomas et al. 2006). In reality, Al(OH)<sub>3</sub> is Al(H<sub>2</sub>O)<sub>3</sub>(OH)<sub>3</sub>; this neutral substance has a very low solubility.

In all SAPS, the deoxygenated water flows from the VFR into a limestone bed, which is typically 0.5-1.5 m thick. The limestone should be at least 90% CaCO<sub>3</sub> and should have a maximum size of 1.3-1.9 cm in diameter. There should be enough limestone present to generate at least 100 mg/L of alkalinity for a period of 10 years. The most rapid gain in alkalinity occurs within the first several hours of AMD—limestone contact; after that, the rate of limestone dissolution slows with time as the water reaches saturation with respect to calcite (Jage et al. 2001; Rose et al. 1979).

#### **Results and discussion**

## Physicochemical properties

Discharge of mine drainage (HC1) ranged between 250 and 450 m<sup>3</sup>/day. When the flow rate increased very dramatically after a severe rainfall in September 2003, the mine adit plug was damaged, so that some mine drainage now flows along the side of the waste rock pile and joins the seepage water from the waste rock pile. The pH of water from the mine adit was generally >6.5 (Fig. 3), which could have been due to the existence of a marine-origin lenticular limestone layer in coal seams, as well as the ALD inside the adit (Ji 2004). A similar pH was earlier reported for AMD in the same area (Jung 2003).

The GW and seepage water from the retaining wall (RSW) had low pHs in all seasons, despite flow variations. This may indicate that the confined aquifer was continuously influenced by AMD generated inside the mines. In the case of seepage water from the retaining wall, pH values from summer to fall in 2003 were lower than those of 2002, which could be due to increased pyrite oxidation in the waste rock pile. During dry seasons (winter–spring), the quality of the stream water was most affected by the seepage from the retaining wall, the sediment bed, and the effluent of the treatment system. Therefore, the pH of the stream was depressed during the dry season, and elevated during the wet season by dilution.

Figure 4 shows the variation of acidity and concentrations of Fe, Al, and  $SO_4^{2-}$  for mine drainage (HC1), waste rock pile seepage water (PSW), spring water (GW), retaining wall seepage water (RSW), and stream water (SW). It should be noted that some AMD might have flowed directly into the oxidation pond, without going through the SAPS because of overflow (OF) taking place in SAPS II. Water was found seeping through cracks in the retaining wall located between the SAPS II and the oxidation pond. This seepage water (RSW) ran off on the surface and finally mixed with the water from the oxidation pond at the boundary of the stream (GW).

Sulfate concentrations initially increased after February 2002, six months after construction, and then gradually decreased until becoming almost constant at slightly more than 500 mg/L. Iron concentrations of the mine drainage were between 18 and 27 mg/L in March-June, 2002 but increased to 84 mg/L in September 2002 and 43 mg/L in October 2003. Acidity and concentrations of Fe and  $SO_4^{2-}$  in the retaining wall seepage water showed the highest concentration during the spring (March-June), decreased during the wet season (July-October), apparently due to dilution, and showed the lowest level in winter, which might have been due to other reasons, such as run-off from old AMD generation sites inside the mine. Iron concentrations were between 640 and 313 mg/L, and acidity was between 1,624 and 1,027 mg/L of CaCO<sub>3</sub>, in spring. In autumn, their values decreased from 313 to 19 mg/L (Fe), and from 402 to 198 mg/L CaCO<sub>3</sub> (acidity), respectively. The high concentration of iron and acidity in spring might have been caused by the flushing of iron sulfate salts that accumulated on the mine surfaces during the winter; similar instances have been reported elsewhere (Rose et al. 1979). Since the mine drainage had a neutral pH in the spring, metals were found precipitated around the mine adit at that time. Water quality downstream was also dependent on the composition of the seepage water flowing along the hydraulic gradient from the retaining wall. The extent of its effect also depended on the seasonal variation of flux of the stream. However, seasonal variations of other contaminants were largely limited to slightly higher overall concentrations in the winter. Iron concentrations were lower than other waters, but Al concentrations were much higher, except for retaining wall seepage water. This might have been caused by the fact that Fe hydroxides precipitate at a lower pH range than Al. Such phenomena have been reported elsewhere (Ji et al. 1997).

To clarify the source of the contamination, sulfur isotope compositions of various waters were analyzed (Fig. 5). The isotope composition of the spring water was very similar to that of the waste rock pile seepage, while the mine drainage differed by about 4‰. Therefore, it appears that the spring water was mostly affected by waste rock pile seepage, not necessarily AMD.









Waste rock pile seepage

Fig. 5 Sulfur isotope compositions of the Hanchang coal mine waters ( $\delta^{34}$ S unit: ‰)

## Efficiency of each stage

Removal efficiencies of Fe and acidity were calculated per unit area per day at each stage of the passive treatment system, assuming the flux to be  $300 \text{ m}^3/\text{day}$  (Tables 1, 2). Removal efficiencies of acidity in the SAPS and the oxic

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Mine adit

wetland became very poor in January 2004. This might be the result of accumulating contaminants, less activity of sulfate reducing bacteria, and desorption of metals from the surface of the organic material. Aluminum increased from 0.27 to 16.68 mg/L. In the oxic wetland, the decomposition of dead plants during the winter can release absorbed Al

Spring

Retaining wall seepage

Table 1 Iron and acidity removal efficiency for each stage of the Hanchang Coal Mine passive treatment system for an annual average daily flow rate of 300 m<sup>3</sup>/day

Operating process			Area (m <sup>2</sup> )	HRT (day)*	Removal Efficiency per Unit Area (g/m <sup>2</sup> /day)								
					February 2002	June 2002	July 2002	September 2002	April 2003	Juy 2003	October 2003	January 2004	Avg
Iron	SAPS 1 280.	280.3	2.8	5.03	6.85	0.26	31.87	10.47	25.28	15.79	1.10	12.08	
		2	508.5	5.1									
	Oxidation pond		260.8	0.87	0.09	0.55	0.16	0.58	0.25	0.21	1.84	0.00	0.46
	Oxic wetland		316.73	0.5	-0.03	-0.13	0.00	0.00	-0.19	0.00	-0.03	0.00	-0.05
Acidity	SAPS	1	280.3	2.8	9.49	12.86	11.37	59.02	20.37	46.98	29.56	-33.7	16.7
		2	508.5	5.1									
	Oxidation pond		260.8	0.87	-0.06	0.88	38.33	-0.68	-1.32	-1.66	0.95	110.8	18.4
	Oxic wetland		316.73	0.5	0.33	-0.43	1.26	1.07	0.47	1.92	2.51	-609	-75.2

The retaining wall water circuit was in a separately laid channel and is not needed considered in this table

\*HRT Hydraulic retention time; -sign implies addition rather than removal

and Mn. Al and Mn increased in the wetland from 6.8 to 106.5 mg/L and from 1.5 to 29.3 mg/L, respectively.

Sulfate removal by sulfate reduction in the organic substrate is important in many SAPS (Kepler and McCleary 1994; Nairn and Mercer 2004; Zipper and jage 2001), but we observed no such decrease of sulfate concentrations at any stage. Similar findings, and even increases in sulfate concentration (possibly due to external sources or desorption) have been reported by others (Cheong et al. 2001; Ji 2004). This might be due to relatively low levels of biological sulfate reduction and/or leaching of sulfate from organic material, leading to a net increase in sulfate concentration. If sulfate concentrations do not decrease, decreases in metal concentrations may be due to adsorption and/or precipitation rather than sulfate reduction. The sludge cake formed on the surface of the organic layer is suspected to be precipitated metal hydroxides. Iron concentrations in the overflow from SAPS II were 2.4 and 2.8 mg/L, in July and September 2002, and 15, and 13 mg/ L in July and October 2003, respectively. This indicates that Fe passed through the system without being processed.

If sulfate reduction occurred in the SAPS units, the  $\delta^{34}$ S value of the effluent should have been much higher than the  $\delta^{34}$ S values of sedimentary metal sulfides in the SAPS. As shown in Table 2, the change of  $\delta^{34}$ S from inflow to

outflow was relatively small, indicating a low sulfate removal rate of about 20%. Samples collected in September 2002 and July 2003 showed sulfate removal ratios of 17.7 and 20.3%;  $\delta^{34}$ S values of outflows were higher by 2.0 and 2.2‰ than those of the inflows. Samples in October 2002 and May 2003 showed sulfate removal ratios of only 7.9 and 7.8%, respectively;  $\delta^{34}$ S values of both outflows were higher by 0.8‰ than those of the inflows (Fig. 5).

The possible effect of meteoric water was also examined. The reported  $\delta^{34}$ S values of some surface waters from Kangwon Province were in the range of 7.4–8.4‰ (Choi et al. 2000), and that of the drinking water in this area was about 6‰. Even though high  $\delta^{34}$ S values of such waters could affect the  $\delta^{34}$ S values of the outflow, the low concentration of sulfate (about 3 mg/L) should limit the effect. Therefore, the main metal removal mechanism appears to be via adsorption on to the surface of the organic material, and as precipitates of metal hydroxides, rather than due to sulfide formation.

#### Summary

The study identified potential reasons for the operational problems of this SAPS:

Table 2 Variation of sulfate concentration and sulfur isotope composition at the Hanchang coal mine SAPS

Sampling date	Inflow		Outflow		Sulfate removal	Variation of $\delta^{34}$ S (‰)	
	$SO_4^{2-}$ (mg/L)	$\delta^{34}$ S (‰)	$\overline{\mathrm{SO}_4^{2-}}$ (mg/L)	$\delta^{34}$ S (‰)	ratio (%)		
September 2002	669.07	-2.5	550.37	-0.5	17.7	2.0	
October 2002	817.12	-3.0	752.78	-2.2	7.9	0.8	
May 2003	567.42	0.0	522.95	0.8	7.8	0.8	
July 2003	623.54	-1.3	496.80	0.9	20.3	2.2	

- 1. Seasonal flooding caused variable loading to the SAPS that might have caused wear and tear to the compost substrate. Non-reactive channels could have formed in the SMC and/or limestone substrate, enhancing the flow of virtually untreated AMD.
- 2. Heavy flooding could have caused retaining walls to collapse or introduced seepage from other debris/waste nearby. Also the hydraulic gradient from one pond to another could have contributed to the seepage problem.
- 3. One of the highlighted mechanisms of SAPS is bacterial sulfate reduction in the organic substrate. However, our isotope study indicated that biological sulfate reduction was low and/or that leaching of ions from the organic material increased sulfate concentrations in the effluent. Therefore, the decrease in metal concentrations was attributed to pH-dependent precipitation and ionic/non-ionic adsorption, rather than sulfate reduction.
- 4. We also found that within 2–3 years of operation, the DOC source was nearly exhausted in the compost. The substrate composition needs to be improved by adding a higher percentage of lignino-cellulosic material in the compost. This would also likely improve the stabilization of the bio-films in the compost column.
- 5. The SAPS was constructed down-gradient of a confined aquifer; the resulting water fluxes through the sediment were not anticipated. It is also likely that ground water and AMD flow may have upset the capacity and design criteria, particularly during the rainy season.

# Conclusions

SAPS are a popular passive bioremediation choice, finding application in metal and non-metal AMD treatment. However, their efficiency often decreases within two years of construction. This study indicated that a decrease in dissolved organic carbon over time likely contributed to a decrease in biological sulfate reduction. Several other operational and site-specific problems may also have contributed to the decreased performance. Thus reinforces the fact that SAPS require regular monitoring and maintenance, and sometimes even a change of substrate to make them function efficiently. Another area, not addressed in this study, is the disposal of precipitate-laden substrate. The major challenge is not to trigger desorption and re-solubilization of the precipitates. However, at this site, much of the precipitate was associated with organic matter rather than precipitated as metal sulfides; it is likely to be chelated or otherwise sequestrated. If this is confirmed, simply covering the dumped substrate with soil to avoid contact with air is an option.

**Acknowledgments** This study was financially supported by grants from the Sustainable Water Resources Research Center of the 21st Century Frontier Research Program and from the Korea Institute of Geoscience And Mineral Resources (KIGAM).

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