

Utilisation of coal/biomass fly ash in the control and treatment of acid mine drainage

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Abstract

Coal/biomass fly ash (CBFA) was blended with various percentages of mine tailings, bentonite, and glacial till soil to investigate its potential as an additive to control and treat acid mine drainage (AMD). The materials were chosen due to their availability and potential as a cost effective alternative to traditional technologies and/or materials. Mixtures were prepared in kinetic columns and continuously permeated with AMD to monitor changes in hydraulic conductivity, effluent pH, and effluent metal concentrations. Results indicate that the addition of CBFA with mine tailings, bentonite, and glacial till soil has potential applications in the geo-environmental field to prevent, contain, and/or treat AMD. A significant decrease of up to three orders of magnitude in the hydraulic conductivity was observed when AMD was continuously permeated through CBFA and through mixtures with higher percentages of CBFA. Little or no decrease in hydraulic conductivity was observed in the lower CBFA / mine tailing mixtures and the CBFA / glacial till soil mixtures. The addition of 10% bentonite to CBFA was effective in obtaining and maintaining a hydraulic conductivity of 1×10^{-7} cm/s or less. Analysis of the leachate indicate that the addition of CBFA to mine tailings, to bentonite, and to glacial till was effective in neutralising and reducing the concentrations of the heavy metals of AMD flowing through the mixtures. The significant reductions in the hydraulic conductivity of the CBFA and higher percentage CBFA amended columns obtained in the study can lead to a low permeable barrier with potential applications as a liner, cap, and dam core to contain acidic mine tailings and AMD. The results showed the addition of CBFA to bentonite can assist in reducing the volume of bentonite required in traditional containment applications while neutralising and treating the seepage that occurs. The little or no decrease in hydraulic conductivity of the lower percentage CBFA amended mine tailings and glacial till soil along with its neutralising effects and heavy metal reductions can serve as a flow through reactive barrier to treat AMD migration offsite.

1 Introduction

The disposal of sulphide bearing mine tailings in surface facilities is a major environmental concern due to the potential of mine tailings producing acid mine drainage (AMD). Control and treatment of AMD, particularly in decommissioned and inactive mine sites, are major environmental challenges. Often, these mine sites have ongoing AMD formation which impacts the adjacent terrestrial and aquatic ecosystems. With continued infiltration recharge, the AMD effluent may flow in the groundwater regime as well as in adjacent streams and other water bodies down gradient from the disposal site.

Several AMD prevention techniques exist, including water and soil covers. Construction of containment dams and dykes can improve the hydrological properties within the tailings thereby increasing the water table and reducing oxidation of reactive mine tailings. In areas where a water cover is not practical, a low permeable cover may be used as a cap to prevent oxidation of reactive mine tailings. Reactive barriers can be an effective passive technology for treating groundwater contamination associated with AMD (Blowes et al., 2003a). This study investigates the potential of using mixtures of coal biomass fly ash (CBFA) with mine tailings, bentonite, and glacial till soil as either a low permeable or flow-through reactive barrier to treat and/or contain AMD (Penney et al., 2009a, 2009b, and 2010).

The effects of AMD flowing through kinetic column packs containing coal fly ash (CFA) either by itself or amended with other materials, e.g. mine tailings and bentonite, has been studied (Xenidis et al., 2002; Shang et al., 2006; Wang et al., 2006; Yeheyis et al., 2007; Perez-Lopez et al., 2007). These studies showed that CFA can substantially increase the pH of AMD from acidic (pH < 3.8) to alkaline (pH > 7), and reduce its

concentrations of heavy metals. A decrease in hydraulic conductivity was also observed when higher percentages of CFA were used in the kinetic column packs.

2 Materials and methods

2.1 Coal/biomass fly ash

CBFA was obtained from the Atikokan Thermal Generating Station, in Atikokan, Ontario, Canada, when co-firing 16 % (by mass) softwood biomass with 84% western Canadian lignite coal. Major CBFA constituents (reported as oxides) and concentrations of heavy metals are shown in Table 1. Acid based accounting (ABA) by the Modified Sobek Method (BC-MEMPR, 1989) indicated a net neutralising potential (NNP) of 298 tonnes CaCO₃/1,000 tonnes.

Table 1 Compositions of CBFA, mine tailings, bentonite, and soil

Component	CBFA (%)	Mine Tailings (%)	Bentonite (%)	Soil (%)	Element	CBFA (g/t) ²	Mine Tailings (g/t) ²	Bentonite (g/t) ²	Soil (g/t) ²
SiO ₂	45.2	31.7	58.4	74.4	Ag	<2	<2	<2	<2
Al ₂ O ₃	21.5	8.06	15.6	11.3	As	<30	<30	<30	<30
Fe ₂ O ₃	4.04	42.3	5.38	3.41	Ba	3,900	300	430	420
MgO	2.51	2.97	1.80	0.45	Be	4.2	0.64	1.5	1.6
CaO	13.6	3.58	2.11	0.80	Cd	<2	<2	<10	<10
Na ₂ O	7.33	1.31	2.40	3.52	Co	16	190	5	9
K ₂ O	0.73	0.97	0.52	3.11	Cu	41	1,200	26	98
TiO ₂	0.95	0.59	0.14	0.39	Li	16	<5	<10	<10
P ₂ O ₅	0.62	0.11	0.07	0.06	Mo	10	<5	<5	<5
MnO	0.02	0.11	0.32	0.05	Ni	27	4,900	<20	<20
Cr ₂ O ₃	0.01	0.04	<0.01	0.07	Pb	48	<100	<40	<40
V ₂ O ₅	0.03	0.03	<0.01	<0.01	Sr	3,300	130	170	70
SO ₄	0.2	1.22	0.21		S	<100	136,000	<100	na
LOI ¹	0.36	6.53	13.1	1.99	Zn	53	180	64	68

Notes: ¹ Loss on ignition; ² gram/tonne.

2.2 Mine tailings

Fresh mine tailings were obtained from Vale, in Sudbury, Ontario, Canada. Major mine tailings constituents (reported as oxides) and concentrations of heavy metals are shown in Table 1. A large concentration of sulphur (136,000 g/t) is associated with sulphide and sulphate minerals. The most abundant heavy metals are nickel and copper with concentrations in the thousands of grams per tonne. ABA indicated a NNP of 416 tonnes CaCO₃/1,000 tonnes, which corresponds to a strong acid generating capacity.

2.3 Bentonite

Bentonite used in this study was laboratory grade sodium montmorillonite from Wyoming, USA. Major bentonite constituents (reported as oxides) and concentrations of heavy metals are shown in Table 1.

2.4 Glacial till

The soil used in this study was a glacial till obtained from a pit near a decommissioned base metals (zinc, lead, and copper) mine in central Newfoundland, Canada. Major soil constituents (reported as oxides) and concentrations of heavy metals are shown in Table 1. The soil contained trace amounts of heavy metals, e.g. copper and zinc, at concentrations of a few tens of grams per tonne. ABA on this soil indicated a low NNP of -1.2 tonnes CaCO₃/1,000 tonnes.

2.5 Synthetic acid mine drainage

Synthetic AMD solutions were prepared in the laboratory. Two solutions were produced with compositions and pH representative of typical field AMD (Table 2). Columns permeated with solution 1 include the mine tailings, mine tailings amended with CBFA, CBFA, soil, and soil amended with CBFA. The columns of CBFA amended with bentonite were permeated with Solution 2.

Table 2 Composition of synthetic AMD solutions

Element/Parameter	Solution 11	Solution 21	Element/Parameter	Solution 11	Solution 21
pH	2.8	2.7	Magnesium	23	17.8
Aluminum	10	5.6	Nickel	2	20.1
Calcium	170	83.8	Sulphur	300	178
Copper	1.3	12.9	Lead	0.04	4.3
Iron	40	55	Zinc	2.5	21

Note: 1 concentrations in mg/L unless specified otherwise.

2.6 Kinetic column tests

Laboratory kinetic column tests were carried out to study the effects of AMD flow on the hydraulic conductivity and effluent composition of various mixtures of mine tailings, glacial till soil and bentonite amended with CBFA. The columns were constructed of either 50 or 75 mm inside diameter clear acrylic tubing. Inert materials (nylon, vinyl, PVC, and stainless steel) were used as tubing, fasteners, collection containers, etc. The mixtures used as column packs were thoroughly mixed in a bowl to ensure homogeneity. The mixture was prepared at ~2% wet of optimum using distilled water and placed in the column in three equal layers. This moisture content achieved minimal hydraulic conductivity while nearly maximising the density. Each layer was rodded 25 times with a 10 mm diameter plastic rod. After rodding, the sides of the column were tapped to remove air bubbles. Next, the top surface of the pack was trimmed and covered with a porous fabric and acrylic porous plate. Upon final assembly, the pack was confined by applying a confining load to the top plate to prevent uplift under hydrostatic loading.

The columns were first permeated with distilled water to establish the reference hydraulic conductivity. Equilibrium was achieved after 7.6 to 20.5 continuous pore volumes (PV) of distilled water had flowed through the columns. At that time, the permeate solution was switched to synthetic AMD. Column tests were then ran, where practical, for at least 70 PV using AMD as the permeation fluid.

The concept of PV, which is the total of the quantity of leachate flowing through the sample divided by the initial pore volume of the sample, allows comparing results from test columns having different initial pore volumes. During the tests, effluent samples were collected, weighed, time recorded, and the hydraulic head measured. The hydraulic gradient was adjusted throughout the tests to allow for the collection of an acceptable volume of effluent (30 ml or more) in a reasonable time period (1 to 5 days). All effluent samples were analysed for pH. Select effluent samples were acidified to pH < 3 with nitric acid and analysed for elemental composition by inductively coupled plasma – atomic emission spectrometry (ICP-AES). Effluent metal concentrations were compared to Environment Canada Metal Mining Effluent Regulations (EC-MMER, 2003).

3 Results and discussion

3.1 Hydraulic conductivity

Hydraulic conductivity results when permeated with AMD for the various mixtures are presented in Figures 1 to 4. Two trends were observed from the results. The packs either observed a minimal change in hydraulic conductivity over the duration of the tests or became less and less permeable with continued AMD permeation. A minimal change in the hydraulic conductivity was observed in mine tailings amended with 5%, 10% and 20% CBFA and soil amended with 5, 10, 20 and 33% CBFA. Tests using 2.5, 5, 7.5 and 10% bentonite amended to CBFA showed an immediate increase in hydraulic conductivity upon switching the permeate from distilled water to AMD (Penney et al., 2009b). Hydraulic conductivity for all but the 2.5% remained consistent when permeated with AMD. This increase in hydraulic conductivity after switching the permeate from distilled water to AMD was also observed by Yeheyis et al. (2007) and Kashir and Yanful (2000 and 2001), who attributed this increase to the collapse of the bentonite’s double layer structure.

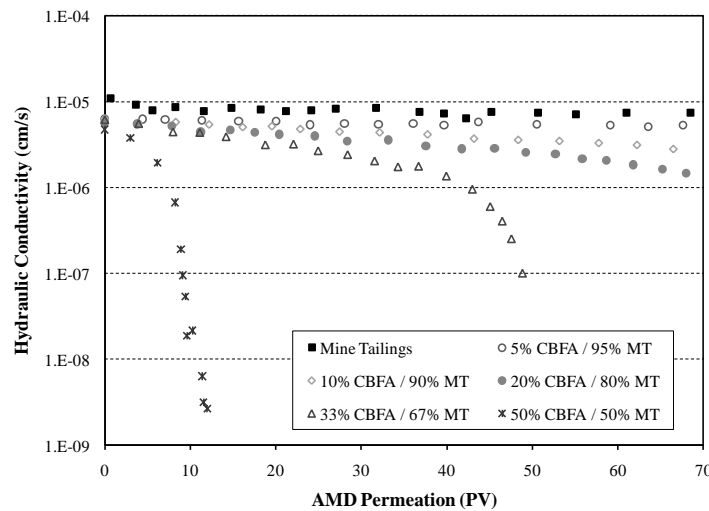


Figure 1 Hydraulic conductivity of mine tailings / CBFA mixtures versus AMD permeate

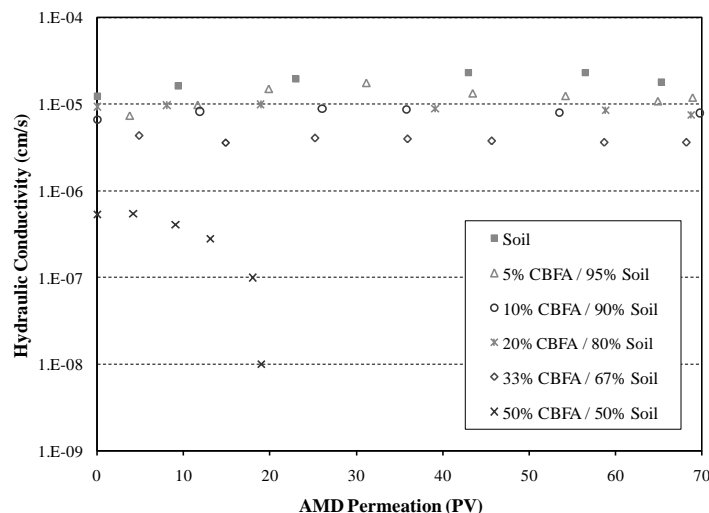


Figure 2 Hydraulic conductivity of soil / CBFA mixtures versus AMD permeate

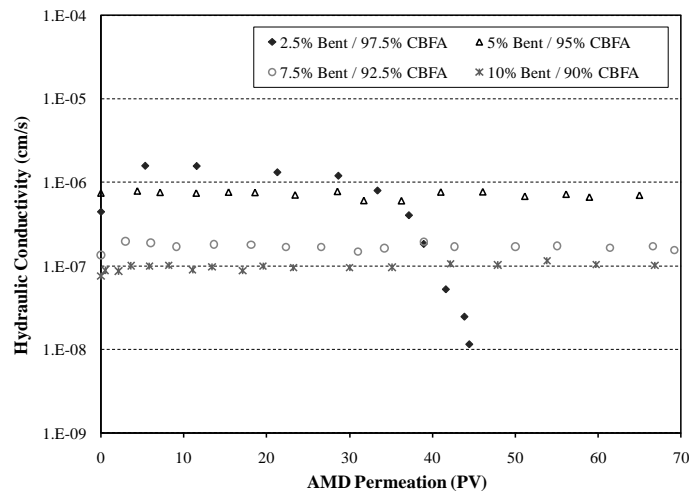


Figure 3 Hydraulic conductivity of bentonite/CBFA mixtures versus AMD permeate

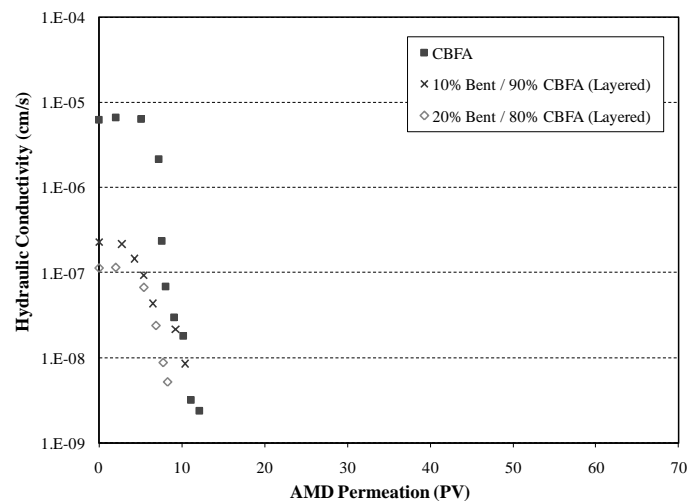


Figure 4 Hydraulic conductivity of layered CBFA and bentonite mixtures versus AMD permeate

A substantial decrease in hydraulic conductivity with continued AMD permeation was observed in the tests using mine tailings amended with 33 and 50% CBFA; soil amended with 50% CBFA; 2.5% bentonite amended to CBFA; layered CBFA with 10% and 20% bentonite amended to CBFA and CBFA. Similar trends in hydraulic conductivity were observed by Shang et al. (2006), Perez-Lopez et al. (2007) and Yeheyis et al. (2007). The decrease in hydraulic conductivity is attributed to the formation of secondary minerals and precipitation of heavy metals from the reaction of the AMD permeating through the CBFA (Yeheyis et al., 2007; Penney et al., 2009b).

3.2 Effluent pH

The kinetic column tests effluent pH results are shown in Figures 5 to 8, when permeated with AMD. The effluent pH of all tests with the exception of the mine tailings and soil controls remained above the EC-MMER lower regulatory pH limit of 6. With the exception of the CBFA amended soil columns, all columns that containing CBFA observed exceedances of the EC-MMER upper regulatory pH limit of 9.5. Typically pH was found to be greatest at the start of the test and decreased with continued AMD permeation. These trends were also observed by Shang et al. (2006), Perez-Lopez et al. (2007) and Yeheyis et al. (2007).

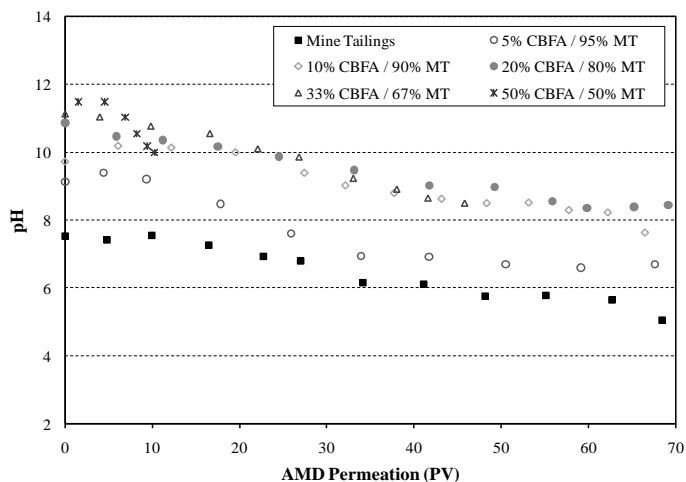


Figure 5 Effluent pH of mine tailings / CBFA mixtures versus AMD permeate

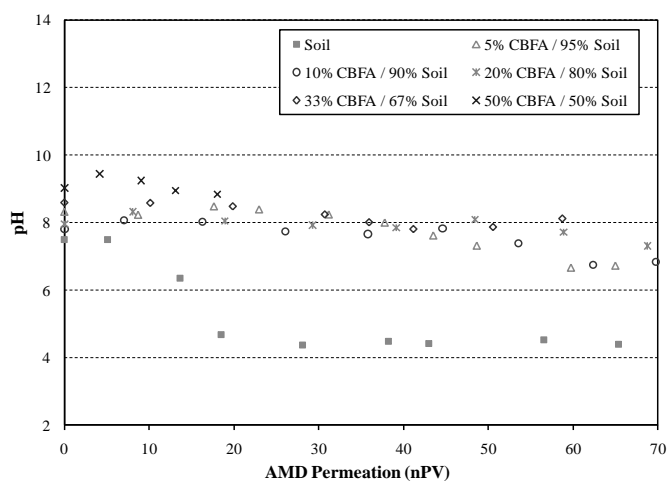


Figure 6 Effluent pH of soil / CBFA mixtures versus AMD permeate

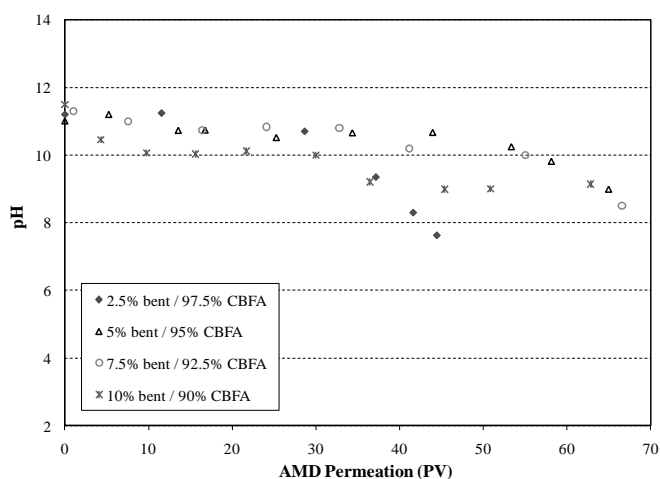


Figure 7 Effluent pH of bentonite / CBFA mixtures versus AMD permeate

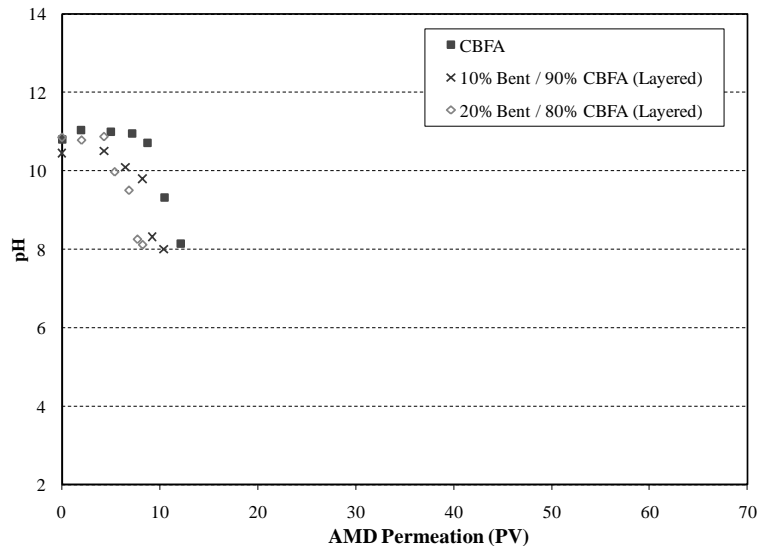


Figure 8 Effluent pH of layered CBFA and bentonite mixtures versus AMD permeate

3.3 Effluent composition

The dissolved concentrations of major elements in the effluents of the column packs were analysed by ICP-AES. Concentrations of elements in the effluent were compared to EC-MMER and the original AMD concentrations. All regulatory limits (EC-MMER, 2003) for metals (arsenic, copper, nickel, lead, and zinc) were met with the exception of the tests using mine tailings and soil only, i.e. control tests. Nickel was found to exceed EC-MMER in the effluent from the mine tailings control during the initial 15 PV of AMD permeations. Nickel and zinc were found to exceed EC-MMER in the effluent from the soil control in 10 PV and 5 PV, respectively, and remained above the regulatory limit for the remainder of the test. While only AMD permeation of 70 PV is reported in this paper, EC-MMER exceedances in nickel and zinc were observed in the 10% CBFA amended soil after 80 PV and 90 PV, respectively (Penney et al., 2010).

Other elements of interest include aluminium, iron, calcium, and sulphur. Although aluminium concentrations are not regulated under EC-MMER, other agencies around the world do regulate them. For example, the United States Environmental Protection Agency (USEPA) sets the total concentration of aluminium at 1 mg/L as the average daily limit value for 30 consecutive days, with a maximum daily value of 2 mg/L (USEPA, 2003). In addition, the toxicity of aluminium to fish and other aquatic organisms is well established at levels as low as 5 µg/L in a range of pH values from acidic to alkaline (WHO, 1997; CCME, 2007).

Aluminium concentrations in the effluent from column packs containing varying amounts of CBFA exceeded 1 mg/L. Comparing the aluminium results with the pH for column tests containing mine tailings and soil amended with CBFA, it is observed that the effluent concentration is greater than the AMD concentration at $\text{pH} \geq 11$ and less than the AMD concentration when $\text{pH} < 11$. Elevated aluminium concentrations were observed in the higher percentages of CBFA, and the CBFA layered bentonite mixtures. These columns appear to be less dependent on pH but did decrease as the hydraulic conductivity decreased. The increased time between sampling intervals, as a result of the decreased hydraulic conductivity, may have resulted in precipitation of aluminium and other elements in the effluent, thereby resulting in a decrease in the effluent pH and aluminium concentrations over time.

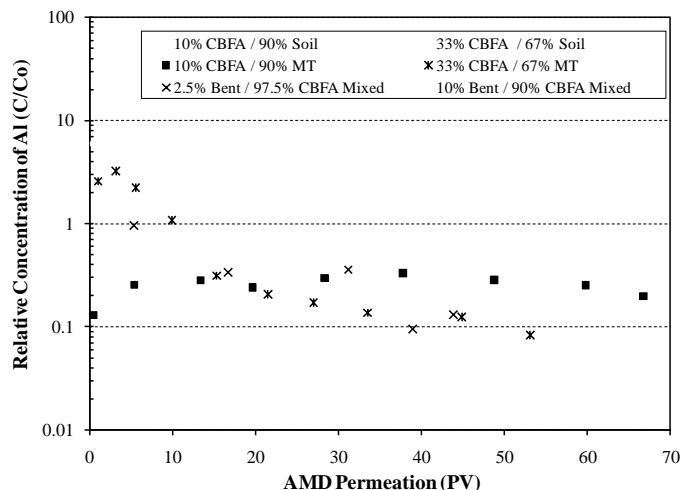


Figure 9 Relevant concentration of aluminium for columns with application as a flow through reactive barrier versus AMD permeate

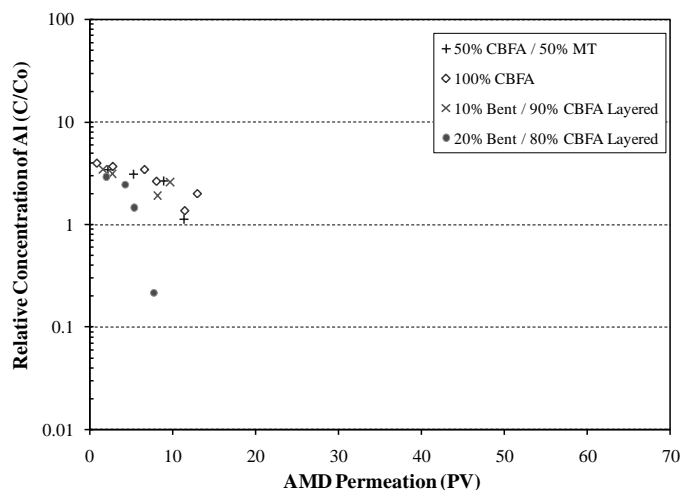


Figure 10 Relevant concentration of aluminium for columns with application as a low permeable barrier versus AMD permeate

The evolution of dissolved aluminium in the effluent of select column packs is shown in Figures 9 and 10. The graphs show the relative concentration, defined as the ratio of the effluent concentration (C) to the concentration of the AMD solution (Co). Relative concentration less than 1 indicates accumulation in the column (e.g. by precipitating). By contrast, relative concentration larger than 1 indicates depletion from the column (e.g. by dissolution).

Iron concentrations are also not regulated under EC-MMER. USEPA sets the total concentration of iron at 1 mg/L as the average daily limit value for 30 consecutive days, with a maximum daily value of 2 mg/L (USEPA, 2003). Only the mine tailings and soil controls exceeded 1 mg/L. All columns containing varying amounts of CBFA show a decrease in iron concentration of at least two orders of magnitude. Since iron is a major component in the formation of AMD particularly at low pH (Blowes et al., 2003b), a substantial reduction in iron can suppress the redevelopment of AMD down gradient of the reactive barrier. The evolution of dissolved iron in the effluent of select column packs is shown in Figures 11 and 12.

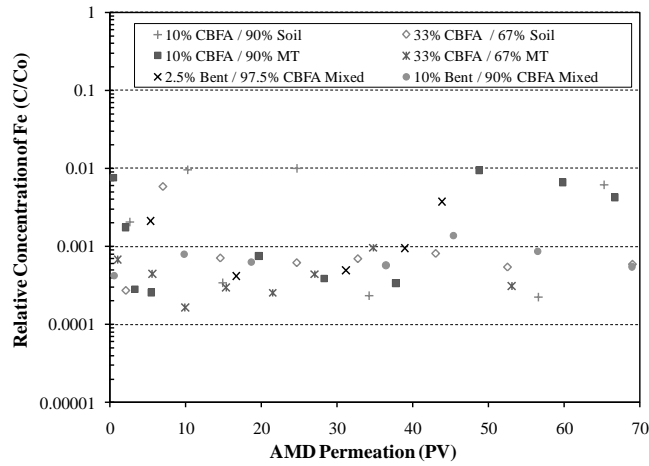


Figure 11 Relevant concentration of iron for columns with application as a flow through reactive barrier versus AMD permeate

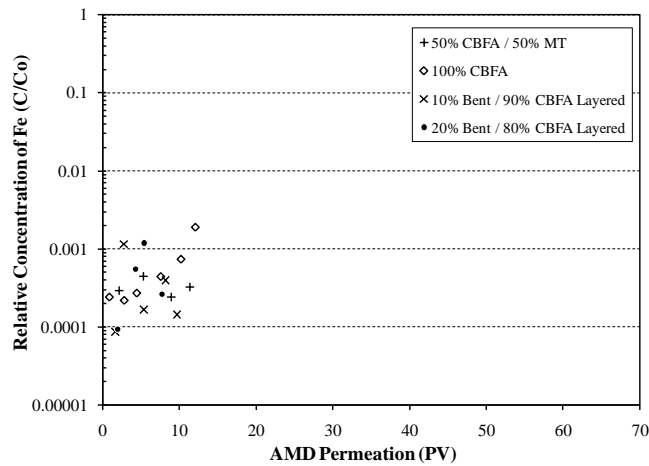


Figure 12 Relevant concentration of iron for columns with application as a low permeable barrier versus AMD permeate

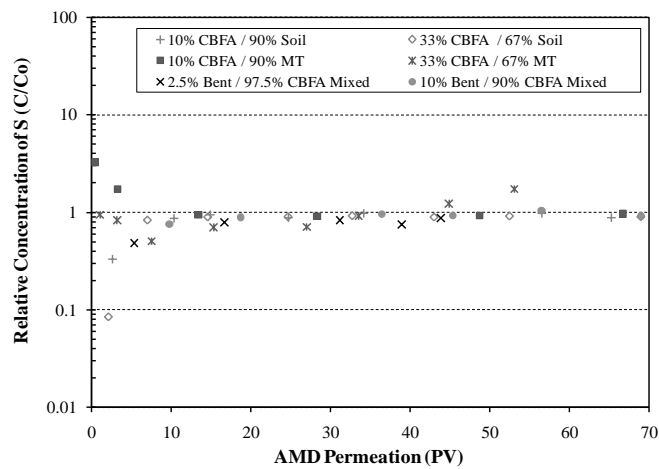


Figure 13 Relevant concentration of sulphur for columns with application as a flow through reactive barrier versus AMD permeate

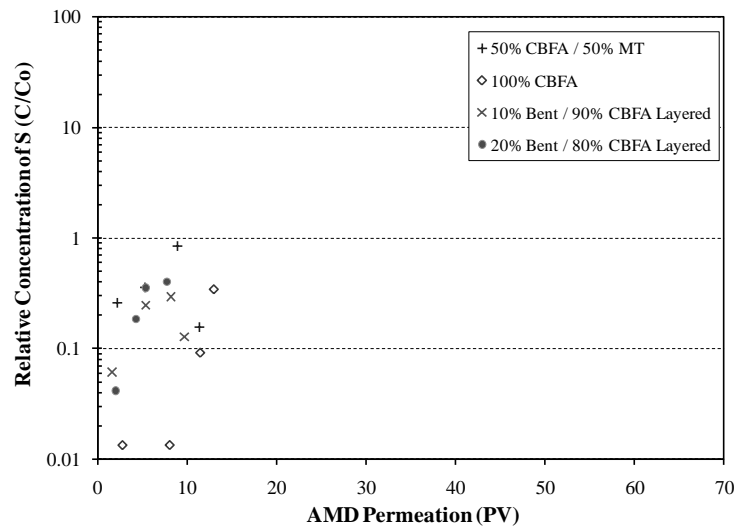


Figure 14 Relevant concentration of sulphur for columns with application as a low permeable barrier versus AMD permeate

Calcium and sulphur concentrations in the effluent were monitored to determine any changes over their initial AMD concentration. Yaheyis et al. (2007) and Penney et al. (2009b) has suggested that the observed decrease in hydraulic conductivity is the result of the precipitation of secondary minerals, particularly gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) and ettringite $((\text{CaO})_6(\text{Al}_2\text{O}_3)(\text{SO}_3)_3 \cdot 32\text{H}_2\text{O})$. Columns observing little change in hydraulic conductivity (potential as a flow through barrier) show break through behaviour for the sulphur, with little change from the original AMD concentration (see Figure 13). Columns observing decreasing hydraulic conductivity (potential as a low permeable barrier) show a decrease in the sulphur from the original AMD concentration (see Figure 14). This reduction in sulphur indicates that it is precipitating within the packs. Comparisons of X-Ray diffraction (XRD) patterns obtained from samples of CBFA before and after column testing reveal the formation of gypsum and ettringite during AMD permeation (Penney et al., 2009b). Gypsum and ettringite were also identified by Yaheyis et al. (2007) on XRD patterns of coal fly ash permeated with AMD. Calcium results are not plotted as they were typically found to exceed the AMD concentrations, most likely due to the dissolution of the calcium oxide minerals (Blowes et al., 2003b).

3.4 Applications of findings

Kinetic column tests show that amending mine tailings and glacial till soil with 33% and less CBFA is effective in buffering pH and reducing the concentrations of heavy metals, while causing little change in hydraulic conductivity. These findings indicate their potential as a flow-through reactive barrier to treat AMD. The experimental setup simulates a flow-through reactive barrier installed in a trench or cavity down gradient of a reactive mine tailings spoil (Figure 15A). The objective of the reactive barrier is to intercept and passively treat plumes of AMD contaminated groundwater, prior to discharging into the environment.

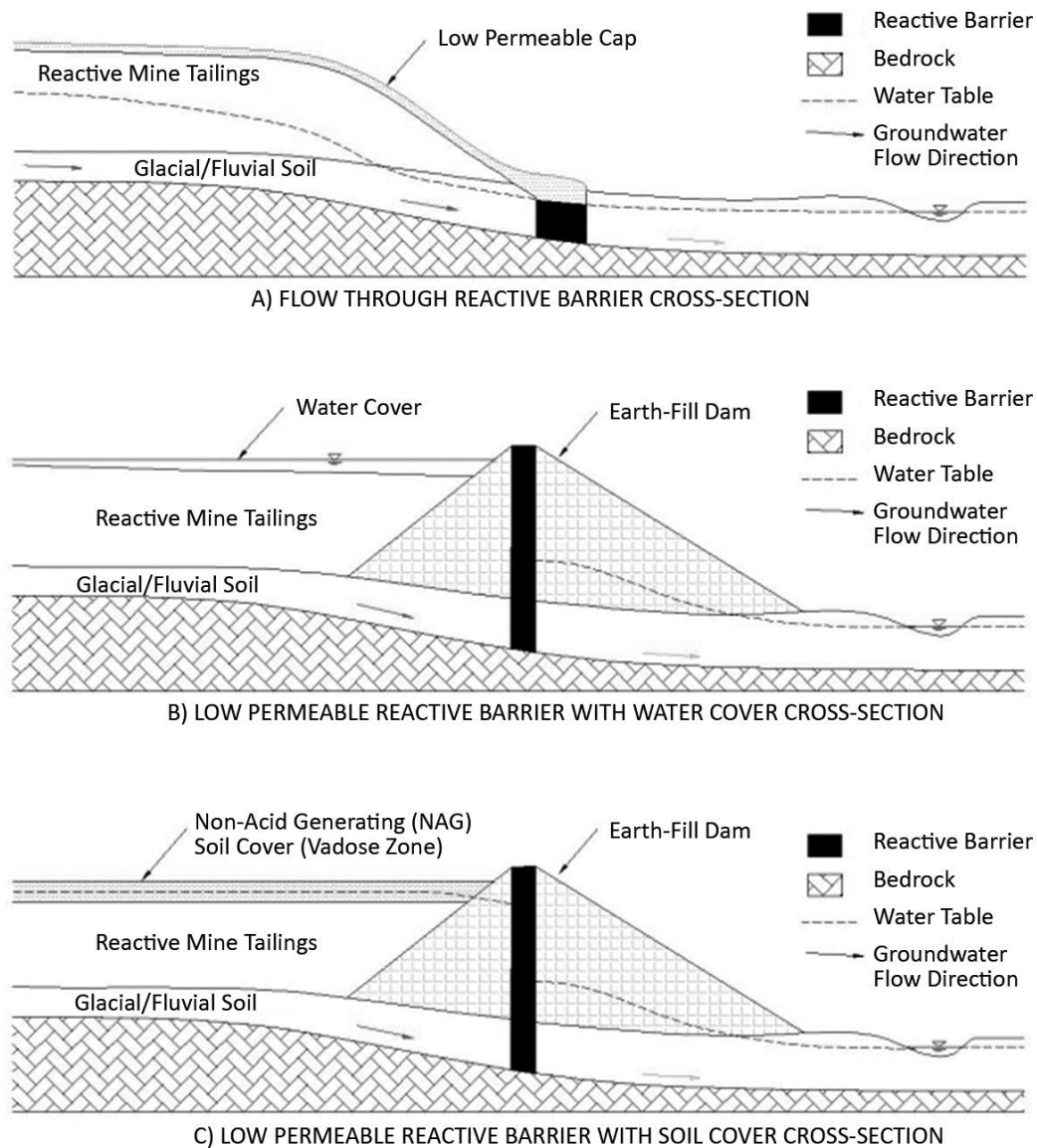


Figure 15 Reactive barrier applications

The 50% CBFA amended column packs and the various combinations of CBFA and bentonite have potential as a low permeable reactive barrier. These packs observe either low hydraulic conductivities throughout the duration of the tests or a substantial decrease shortly after AMD permeation. These tests were also found to be effective in buffering pH and reducing heavy metal concentrations. A practical application for these mixtures is a low permeable core in containment dams. During and following construction, as the AMD flows through the mixtures, the hydraulic conductivity would decrease and the water table elevation would increase. The raising water table would submerge the reactive mine tailings and minimise their oxidation rate (Figure 15B). This application is subject to the surrounding topography, geology and hydrology, but under ideal conditions it could create a containment pond with an adequate water cover. Where the formation of a containment pond is not viable, the water table could be raised to an elevation such that the vadose zone would be in a soil cover having a positive NNP (Figure 15C). The CBFA/bentonite mixtures also have potential as cap or liner applications, particularly when an initial low hydraulic conductivity is required.

4 Conclusions

Kinetic column tests show that amending CBFA with onsite mine tailings and glacial till soil can effectively create a reactive flow through barrier where the pH of the AMD plume is increased and the dissolved concentrations of heavy metals decreased. The formation of secondary minerals (gypsum and ettringite)

within the column packs containing 50% and greater CBFA can effectively decrease the hydraulic conductivity below 1×10^{-7} cm/sec. Mixing bentonite with CBFA can be an effective method to construct a low permeable barrier for applications where an initial low permeability is required. This would be best suited for new mine sites where reliance on the formation of AMD and its reaction with the CBFA are not applicable.

Acknowledgements

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