Development of Passive Treatment Systems for Treating Acid Mine Drainage at Stockton Mine

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Abstract

Acid mine drainage (AMD) at Stockton Coal Mine is generated from the oxidation of pyrite in carbonaceous mudstones exposed during surface mining. Acidity production causes metals such as Fe and Al to leach from overburden materials including feldspars. Water chemistry and flow were monitored at numerous AMD seeps at Stockton. Manchester Seep, which daylights at the toe of an overburden embankment, was identified as a suitable research site for trialling passive-treatment systems designed to neutralize acidity and sequester metals in AMD. Median dissolved metal concentrations from the Manchester seep were; 62.9 mg/L Fe, 32.5 mg/L Al, 0.0514 mg/L Cu, 0.175 mg/L Ni, 0.993 mg/L Zn and 0.00109 mg/L Cd.

Treatment of this water is achieved downstream by the Mangatini fine limestone dosing plant, however in the interest of assessing other technologies this work investigated the use of bioreactors to assess the potential of passive treatment technologies to treat the Manchester Seep AMD. Geotechnical parameters, including hydraulic conductivity, were measured for various mixtures of organic and alkaline waste products suitable for use as bioreactor media. Seven mesocosm-scale bioreactors were fed aerated Manchester Seep AMD in a laboratory set-up for nearly four months. Bioreactors incorporating mussel shells performed better than limestone and were capable of sequestering >0.80 mol metals/m^3 substrate/day (or neutralising acidity at rates >66 g $CaCO_3/m^2$/day) while removing >98.2% of all metals. Tracer studies were conducted on two bioreactor systems containing the same substrate composition but different reactor shapes. Results will be applied to reactor models to better ascertain the relationship between reactor hydraulics and treatment performance.

Pilot-scale passive systems incorporating three treatment stages were designed and are currently being installed to treat a portion of Manchester Seep AMD on site. The first stage consists of a sedimentation basin to remove sediment. The second stage includes three bioreactors in parallel to test treatment effectiveness of different substrate mixtures, depths and hydraulic configurations. Data derived from the mesocosm lab study were used to optimise these designs. The final treatment stage consists of three different aerobic wetland configurations, also operated in parallel, to compare their effectiveness at providing oxygenation and tertiary treatment of metals (primarily Fe) from bioreactor effluent.

Keywords:

acid mine drainage (AMD), sulphate-reducing bioreactors (SRBR), Stockton Mine, mine-water treatment, passive treatment systems

Published in the peer-review *proceedings of the New Zealand Minerals Conference,* Australian Institute of Mining and Metallurgy, Auckland, New Zealand, 29 Aug.-4th Sept. 2008, pp. 329-338.

Introduction

Acid mine drainage (AMD) has impacted an estimated 125 km of freshwater streams on the West Coast of the South Island, New Zealand (James, 2003). Stream biodiversity and ecological health have been significantly altered in these systems (Harding and Boothryd, 2004; Harding, 2005) and taxonomic richness of invertebrates was significantly lower (Anthony, 1999; Winterbourn et al., 2000). The Brunner Coal Measures, present at Stockton coal mine were geologically formed in a marginal marine setting and consist of carbonaceous mudstones, sandstones, and coal containing abundant sulphide and subsequently high acid-generating capabilities (Black et al., 2005; Trumm et al., 2005; Pope et al., 2006). Acidity generation and Fe release occurs during pyrite (FeS_2) oxidation (Skousen, 1996; Rose and Cravotta, 1998; Watzlaf et al., 2003). Under acidic conditions Al leaches from ubiquitous micaceous and feldspathic-rich rocks (Black et al., 2005), in itself generating additional metal (Lewis) acidity (Younger et al., 2002; Watzlaf et al., 2003). Other (divalent) metals such as Cu, Ni, Zn, Cd, As, Pb and Mn also dissolve from parent bedrock when exposed to acidity so AMD generation causes a compounding metal mobility challenge.

Passive treatment systems have successfully treated AMD at abandoned coal and metal mine sites throughout North America and Europe (Younger et al., 2002; Watzlaf et al., 2003; Ziemkiewicz et al., 2003; Wildeman et al., 2006). However, there has been little research on their implementation in New Zealand. Trumm et al. (2005 and 2006) summarized AMD chemical signatures and mesocosm field studies for remediating AMD from Sullivan Mine, an abandoned underground coal mine within the Brunner Coal Measures. Results indicated that a vertical-flow wetland (VFW) comprised of a 150 mm limestone layer overlain by a 130 mm mushroom compost layer was successful at generating alkalinity and removing metals. Influent Fe and Al concentrations in this system were approximately 38-62 mg/L and 13-16 mg/L, respectively. Removal efficiencies at day 22 of system operation and five-hour calculated hydraulic residence time were 100% acidity, 97% Fe, 100% Al and 66% Ni. A small-scale VFW was also installed at Pike River on the West Coast of the South Island, yielding successful acidity and metal removal (Trumm et al., 2006). Removal efficiencies by day 58 (of 151 operational days) in this system with an average calculated hydraulic residence time of 20 hours were 100% acidity, 99% Fe, 96% Al, 95% Ni and 99% Zn.

Trumm et. al. (2007) performed mesocosm-scale treatability tests comparing the effectiveness of a limestone-leaching bed (LLB), an open-limestone channel (OLC) and a VFW for passively treating AMD emanating from Herbert Stream on the Stockton Plateau in the Waimangaroa Catchment. The AMD influent concentrations for these systems were reported as; Al (2.9-9.4 mg/L), Fe (0.33-3.45 mg/L) and Mn (0.39-0.92 mg/L) in Trumm et al. (2006) and Trumm (2007). The LLB effluent (effluent pH 7.3-7.9) performed slightly better than the VFW effluent (effluent pH 6.4-7.4), while the OLC was least effective (pH<5.6). All systems removed up to 99% Al, but the LLB also removed 99% Fe while the VFW and OLC removed 97% and 94%, respectively. Similar removal trends were reported for Mn and Zn. An LLB system was chosen for future full-scale AMD treatment at this site based on its effectiveness, simplicity, practicality, site constraints and successful removal of Fe and Al.

The primary objective of this study was to develop appropriate passive treatment designs in New Zealand for treating AMD dominant in acidity, Fe and Al. Initially, numerous AMD seeps at Stockton Opencast Coal Mine were monitored to ascertain the seasonal signatures (chemistry and flow) of the AMD. Manchester Seep, which daylights at the toe of an overburden embankment, was identified as a suitable research site for trialing passive-

Published in the peer-review *proceedings of the New Zealand Minerals Conference,* Australian Institute of Mining and Metallurgy, Auckland, New Zealand, 29 Aug.-4th Sept. 2008, pp. 329-338.

treatment systems designed to neutralize acidity and sequester metals from AMD. From the Manchester AMD signature, bioreactors were chosen as the most feasible and efficient passive treatment technology. Mesocosm-scale laboratory experiments were established to measure hydraulic and contaminant treatment efficiencies for these systems (McCauley et al., 2008). These hydraulic and treatment efficiencies were used to finalise designs of the Manchester seep pilot-scale passive treatment systems planned to treat 0.4 L/sec of AMD flow.

Methods, Results and Discussion

Acid mine drainage seep monitoring

Results from six AMD seeps monitored at Stockton Mine during this study indicated variability in flow and water chemistry at the mine site (Table 1). The primary contaminants at all seeps were typically Fe, Al, and acidity. Concentrations ranged from 0.59 to 1430 mg/L dissolved Fe and 7.43 to 627 mg/L dissolved Al. The pH values ranged from 2.15 to 3.75 and acidity (to pH 8.3) ranged from 80.5 to 7724 mg/L as $CaCO_3$. Other metals contributing to mineral acidity (<2 wt%) included Cu, Ni, Zn, Cd, Pb and Mn.

Table 1. Water Chemistry Results from AMD Seeps at Stockton Mine. Values are given as medians (with ranges indicated in parenthesis beneath). Flow was not monitored for the AMD seeps at C or A Drive or Whirlwind Tributary A.

	Manchester Seep	Collis Seep 1	Collis Seep 3	Whirlwind Tributary A	C Drive	A Drive
N	11-12	2-3	2-3	2-3	1	1
pH	2.81 (2.49-3.34)	2.15 (2.04-2.23)	2.17 (2.07-2.21)	3.13 (2.86-3.16)	3.75	3.13
Diss Fe (mg/L)	62.9 (4.31-143)	(1390-1430)	(1140-1255)	(4.91-6.26)	0.59	NA
Diss Al (mg/L)	32.5 (7.43-56.7)	(586-627)	(429-558)	17.9	13.3	NA
Acidity – pH 3.7 (mg/L as $CaCO_3$)	158 (21.1-373)	3163 (3133-3561)	3071 (2873-3122)	85 (43-111)	14.9	33.0
Acidity - pH 8.3 (mg/L as $CaCO_3$)	363 (78.5-626)	7724 (7352-7851)	6193 (6036-6757)	165 (145-186)	80.5	258
Flow (L/s)	1.84 (0.35-10.5)	0.074 (0.052-0.12)	0.16 (0.12-0.26)	NA	NA	NA

Manchester Seep was identified as the most suitable research site for trialling passive-treatment systems at Stockton Mine given the available land area, AMD signature and potential to demonstrate proof of concept within the prescribed research timeframe. Therefore, monthly chemistry and flow monitoring was conducted for ten months at this site following quality assured/quality controlled (QA/QC) procedures (McCauley et al., 2008). Median flow rate was calculated at 1.84 L/s with a range of 0.35-10.5 L/s. Median dissolved metal concentrations were measured at 62.9 mg/L Fe, 32.5 mg/L Al, 0.0514 mg/L Cu, 0.175 mg/L Ni, 0.993 mg/L Zn and 0.00109 mg/L Cd. The AMD pH ranged from 2.49 to 3.34 and median acidity to (pH 8.3) was 363 mg/L (78.5-626 mg/L) as $CaCO_3$. Fe, Al and H comprised >99% of the acidity from Manchester Seep AMD so removal of these contaminants were prioritised in the treatment designs.

Mesocosm-scale treatability tests

Mesocosm-scale treatability tests were performed in a laboratory set-up measuring the effectiveness of seven continuous flow VFWs (referred to as bioreactors during this study) for treating Manchester Seep AMD, which was shipped from Stockton mine. Bioreactor substrates included a mixture of industrial waste products (Table 2) derived from forestry/timber including organic bark (*Pinus radiata)*, post peel (untreated by-product from fence post manufacture) and bark compost as well as alkaline materials including limestone, mussel shells (from mussel farming) and nodulated stack dust (NSD) (from cement manufacturing). Two different bioreactor dimensions (337 L trapezoidal containers (440 mm substrate depth) and 138 L cylindrical drums (562 mm substrate depth)) were established. Bioreactors containing the same substrate mixtures (P-2: drum and S-4: trapezoidal) were subjected to hydraulic tracer tests to ascertain how reactor shape and substrate depth influence treatment performance. Further detail about the experimental set-up and treatment efficiencies of these mesocosm-scale systems, which were operated for nearly four months has been discussed (McCauley et al., 2008).

Table 2. Bioreactor substrate compositions (volumetric percent).

	S-1	**S-2**	**S-3**	**S-4**	**P-1**	**P-2**	**P-3**
	Trapezoidal Containers - 337 L (Substrate Depth – 440 mm)				*Cylindrical Drums* - 138 L (Substrate Depth – 562 mm)		
Limestone	12.5	0.0	0.0	5.0	0.0	5.0	2.5
Mussel Shells	0.0	20	20	12	30	12	12
NSD	0.0	0.0	0.0	0.0	0.0	0.0	5.0
Bark	35	40	30	30	30	30	30
Post Peel	37.5	25	35	38	25	38	35
Compost	15	15	15	15	15	15	15

Influent and effluent Fe and Al concentrations from the mesocosm-scale experiments are shown in Fig. 1. The x-axis illustrates Manchester Seep AMD and effluent from each bioreactor. The y-axis shows Fe (orange bars) and Al (grey bars) concentration ranges (on a logarithmic scale). Horizontal black lines represent median Fe and Al concentrations. Data shown in Fig. 1 is representative of metal loading rates ranging from 0.23 to 0.83 mol/m^3 substrate/day and acidity loading rates ranging from 25 to 80 g (as $CaCO_3$)/m^2/day. Bioreactors containing 20-30 vol.% mussel shells (P-1, S-2 and S-3) showed the best metal removal of the feasible options evaluated in this study. Metal removal was also good for P-3 (containing NSD), but effluent pH was 9.0-10.5, and therefore considered too caustic for discharge to a freshwater ecosystem so was eliminated from further study. Bioreactors containing limestone (especially S-1) showed the least effective metal removal. Bioreactor P-2 outperformed S-4 (duplicate reactors) indicating cylindrical drum reactors (562 mm substrate depth) perform better than trapezoidal prism reactors (400 mm substrate depth) for this bioreactor design. Metal removal from S-2 and S-3 (both contained 20 vol.% mussel shells) were similar throughout this study except at the highest loading rates tested at 1.4 mol metals/m^3 substrate/day and 135 g as $CaCO_3$/m^2/day (data not shown) (McCauley et al., 2008). Bioreactor S-2 had lower metal removal than S-3 at the highest metal and acidity loading rates tested, possibly due to increased hydraulic short circuiting caused by the higher percentage of flat, plate-like bark.

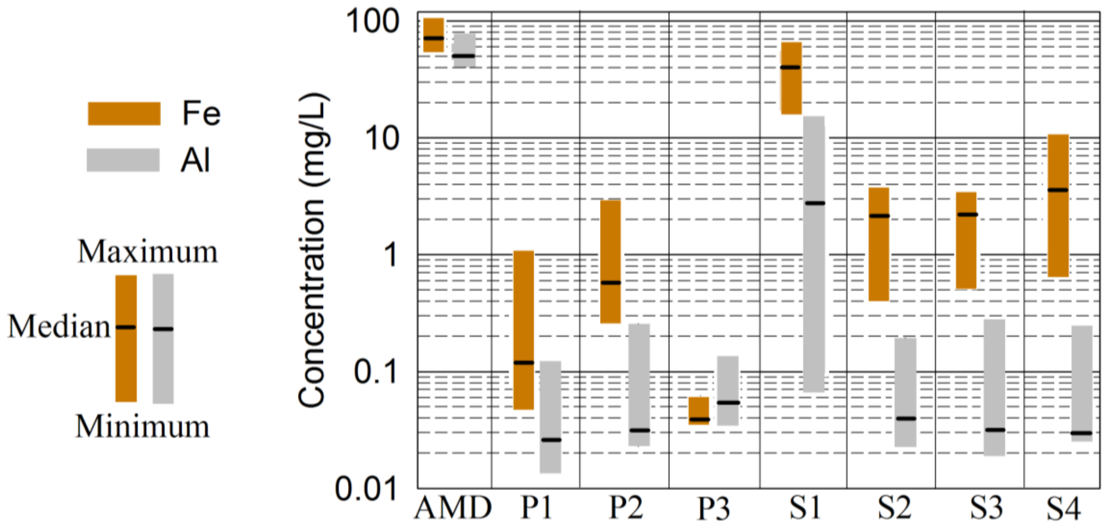

Figure 1. Influent (AMD) and effluent (P-1, P-2, P-3, S-1, S-2, S-3 and S-4) dissolved Fe and Al concentrations from mesocosm experiments during metal loading rates from 0.23 to 0.83 mol/m^3 substrate/day and acidity loading rates from 25 to 80 g (as $CaCO_3$)/m^2/day.

Dissolved metal influent (AMD) and summarised effluent concentrations and calculated removal efficiencies from bioreactors containing 20-30 vol.% mussel shells (P-1, S-2 and S-3) during metal loading rates of 0.23 to 0.83 mol/m^3 substrate/day and acidity loading rates of 25 to 80 g (as $CaCO_3$)/m^2/day, are shown in Table 3. Metal removal was most effective for Al, Cu, Ni, Zn, Cd and Pb (Table 3), but a substantial amount of Fe (96.5-99.8 %) was also removed. McCauley et al. (2008) recommended a conservative design criteria of 0.8 moles of metals removed/m^3 substrate/day and acidity removal of 66 g $CaCO_3$/m^2/day for bioreactors used in this study containing 20-30 vol.% mussel shells.

Table 3. Dissolved metal influent (AMD) and summarised effluent concentrations and removal efficiencies from bioreactors containing 20-30 vol.% mussel shells (P-1, S-2 and S-3) during metal loading rates from 0.23 to 0.83 mol/m^3 substrate/day and acidity loading rates from 25 to 80 g $CaCO_3$)/m^2/day. Median concentrations were computed assuming sample concentrations detected below laboratory practical quantitation limits (PQLs) were equal to one-half the PQL values.

	AMD Conc. (mg/L)	Effluent Conc. (mg/L)			Removal Efficiency (%)
	Median	*Median*	*Min*	*Max*	*Range*
Fe	78.4	1.04	0.120	3.46	96.5-99.8
Al	53.6	0.031	0.0170	0.277	99.5-99.9
Cu	0.209	0.00025	<0.0005	<0.001	<99.7->99.9
Ni	0.230	0.001	<0.0005	0.0020	99.3->99.7
Zn	1.27	0.002	<0.001	0.005	99.7->99.9
Cd	0.00186	0.000025	<0.00005	<0.00005	>98.3-98.9
Pb	0.0152	0.00005	<0.0001	0.0001	99.5->99.7

Geotechnical parameters of substrate mixtures

Hydraulic conductivity (constant head) and air porosity were measured on substrate mixtures in bioreactors P-1, P-2/S-4, S-2 and S-3 (Table 4) prior to conducting the treatability tests. Hydraulic conductivity was of the order 1E-3 m/s for all substrate mixtures. The porous properties of most substrates employed in these systems (except compost) make them less prone to hydraulic plugging from sediment, immobilised metals and biofilms than systems utilised overseas, which incorporate only compost or a mixture of compost and limestone (Watzlaf et al., 2003; Ziemkiewicz et al., 2003). The hydraulic conductivity of compost used in this study was one to two orders of magnitude less (1E-1 to 1E-2 m/s) than substrate mixtures indicating that system clogging is more likely if only compost was used. Air porosity is important because it represents the fraction of voids not occupied by substrate media. It also plays an important role in reactor hydraulics including calculation of theoretical hydraulic residence time, which was calculated at 2.09-8.50 days during this study (when metal loading rates ranged from 0.23 to 0.83 mol/m^3 substrate/day and acidity loading rates from 25 to 80 g (as $CaCO_3$)/m^2/day).

Table 4. Hydraulic conductivity and air porosity of substrate mixtures.

	P-1	**P-2/S-4**	**S-2**	**S-3**
		Hydraulic Conductivity		
(m/s)	1.78E-3	1.18E-3	2.16E-3	1.96E-3
(m/day)	154	102	186	169
		Air Porosity		
fraction	0.51	0.47	0.48	0.48

Concurrent and Future Research

Tracer studies

System hydraulics are important parameters in contaminant reactor design and operation as short-circuiting can impair treatment efficiency in these systems. Tracer studies are commonly conducted to develop residence-time distribution curves that provide information on wastewater flow characteristics and actual hydraulic residence time (Levenspiel, 1999). Data can also be applied to commonly employed reactor models such as the tanks-in-series and plug flow (dispersion) models. Tracer tests for this study were performed on two occasions (data from the first of these events is presented below). Each bioreactor was "instantaneously" spiked with a sodium bromide tracer solution. The AMD collected from Manchester Seep was continuously fed into the reactors at controlled flow rates equivalent to about 0.9 mol metals/m^3 substrate/day. Effluent samples from both bioreactors were collected regularly and analysed for dissolved Br concentration by ICP-MS (Method APHA 3125 B) (APHA, 2005). Maximum recovered Br concentrations corresponded well with theoretical hydraulic residence times (3.0 days for P-2 and 3.5 days for S-4) as shown in the residence-time distribution curves illustrated in Fig. 2 during the first sodium bromide spike. Ideally, effluent samples should have been collected for a longer time duration until nearly all Br was flushed out of the reactors in order to understand the complete operational hydraulics of contaminant transport in these systems. Consequently, during the second tracer study, effluent samples were collected for up to 11 days following a complete flushing period between tests. A representative plug-flow trend is illustrated by the black data points in Fig. 2 (representing bioreactor P-2). The development of a second Br spike is shown for bioreactor S-4 (grey data points after about 4.5 days) signifying potential flow channelling and/or internal recirculation confirming that deeper, cylindrical shaped bioreactors may be preferable to trapezoidal shaped ones for optimum treatment performance. Data from both tracer tests is currently being analysed and modelled to further compare system hydraulics in duplicate substrate reactors of different dimensions (P-2 and S-4).

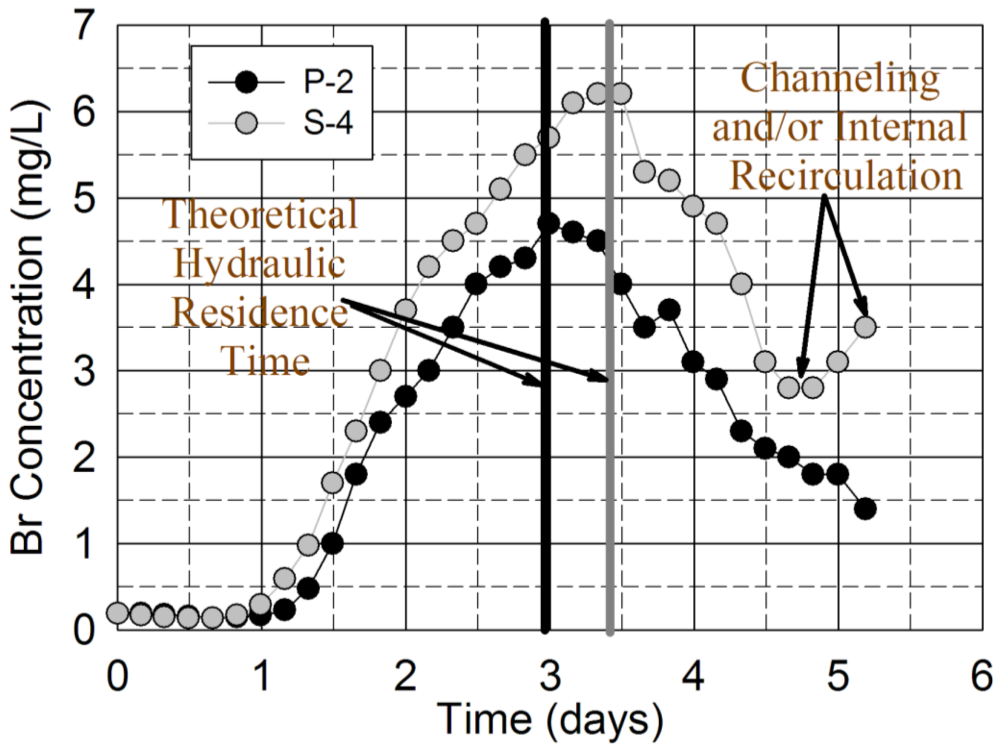

Figure 2. Residence time-distribution curve during the first Br spike showing Br concentration versus time. Bromide concentrations peaked at the theoretical hydraulic-residence time for both bioreactors (P-2 and S-4).

Pilot-scale treatability tests

Construction of pilot-scale treatment systems to treat 0.4 L/s of Manchester Seep AMD is currently being completed at Stockton Mine (Fig. 3). Manchester Seep flows into a sedimentation basin to settle and store sediment that could otherwise clog substrate of the subsequent treatment stage consisting of bioreactors. Outflow from the sedimentation basin will be conveyed into a v-notch weir to measure volumetric flow rates. A portion of the AMD (0.4 L/s total) is then conveyed into the second treatment stage consisting of three bioreactors operated in parallel to neutralise acidity and biogeochemically immobilise metals (metal removal rate criteria of 0.77 mol/m^3 substrate/day used for design). Bioreactor design criteria are given in Table 5, all of which contain 30 vol.% mussel shells. Different substrate depths (1.0 and 2.0 metres) and compost percentages (15 vol.% and 30 vol.%) are incorporated into the designs to test treatment performance. Effluent from each of the bioreactors will be treated by a subsequent aerobic "polishing" stage consisting of one or two cascades followed by either a pond, a rock filter followed by a vegetated aerobic wetland or a pond followed by a vegetated aerobic wetland (Table 5). This final polishing stage is necessary to remove biochemical oxygen demand (BOD) by oxygenating the reduced bioreactor effluent and remove residual metals such as Fe as iron hydroxides. These final aerobic wetland cells are sized to remove 5 mg/L Fe based on an areal design rate of 10 g Fe/m^2/day.

Once the pilot-scale systems are fully operational, chemistry and flow data will be measured biweekly to monthly to ascertain treatment performance so that contaminant mass balances for acidity and metals of concern can be calculated for each treatment stage. Additional water quality parameters will be measured and compared with compliance and discharge consents.

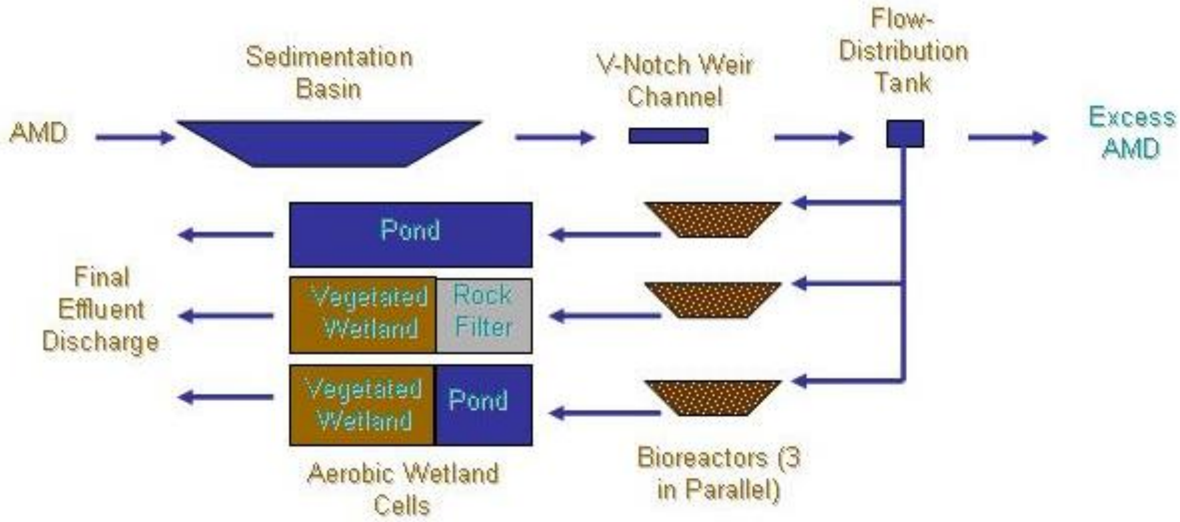

Figure 3. Pilot-scale treatment system design for treating 0.4 L/s of Manchester Seep AMD.

Table 5. Pilot-scale bioreactor designs. Percent substrate compositions shown are on a volumetric basis.

	BIOREACTOR NAME		
	PS-1	**PS-2**	**PS-3**
Design Metal Removal (mol/m^3 substrate/day)	0.77	0.77	0.77
Flow (L/s)	0.1	0.2	0.1
Substrate Depth (m)	1.0	2.0	1.0
% Mussel Shells	30	30	30
% Pine Bark	20	20	15
% Post Peel	35	35	25
% Compost	15	15	30
Aerobic Treatment Stage Design	2 stage cascade followed by pond	1 stage cascade followed by rock filter and vegetated aerobic wetland	1 stage cascade followed by pond and vegetated aerobic wetland

Conclusions

A systematic process is recommended when considering using passive treatment systems to neutralise acidity and immobilise metals in AMD-impacted water. Robust flow and water chemistry monitoring conducted over a sufficiently long period (accounting for seasonal variability) is required to understand hydrologic variance and chemical loading for a particular AMD source. The most appropriate passive-treatment designs are then considered with a known AMD chemical signature and site limitations. Factors that influence passive treatment feasibility (and success) include land availability, topography, flow, AMD chemistry and operational management. Passive treatment may not be feasible for treating all AMD-impacted waters but can offer a more cost-effective alternative to traditional lime-dosing systems, especially for abandoned and decommissioned mine sites. Mesocosm and pilot-scale treatability tests should be performed prior to design, construction and operation of a full-scale system to ascertain pertinent design parameters such as metal and acidity removal rates. System hydraulics should also be well understood to predict bioreactor optimisation and longevity.

Although Fe and Al were the most prevalent metal contaminants in AMD monitored during this study, Cu, Ni, Zn and Cd were also elevated. Results of mesocosm-scale bioreactors incorporating industrial waste products as alkaline and carbon substrate materials were successful at sequestering metals (Fe, Al, Cu, Ni, Zn, Cd and Pb) and removing acidity from Manchester Seep AMD. Design criteria for bioreactors incorporating 20-30 vol% mussel shells was established at >0.8 mol metals/m^3 substrate/day or >66 g acidity as $CaCO_3/m^2$/day (McCauley et al., 2008). Future analysis on recently conducted tracer tests will be applied to reactor models to better ascertain the relationship between reactor hydraulic residence time distribution and treatment performance. Results of future pilot-scale treatability tests at Stockton Mine will provide performance data of bioreactors and "final polishing" aerobic wetlands for treating Manchester Seep AMD in a field application. Overall, water quality discharging from a passive treatment technology should aim to improve biodiversity and ecological health of the receiving water body.

Acknowledgements

This research was made possible by financial support provided by Solid Energy New Zealand Ltd., a Technology for Industry Fellowship awarded by Technology New Zealand (Foundation for Research, Science and Technology), a grant awarded by the Coal Association of New Zealand and numerous contestable funding research grants awarded by the University of Canterbury Department of Civil and Natural Resources Engineering. Industrial mentoring was provided by Andrew Brough of Pattle Delamore Partners Ltd (PDP). Technical and logistical support was provided by various university technicians including Peter McGuigan and Ian Sheppard and operational staff at Stockton Mine. Nicola McHaffie, a summer undergraduate researcher, helped with the geotechnical analyses. Jenna Hutchinson, a summer undergraduate researcher, helped sample during the tracer studies. Sea Products, Ltd. in Christchurch, New Zealand donated mussel shells.

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