Permeable reactive barrier (PRB) technology for the remediation of acidic groundwater from acid sulphate soil floodplain in south east New South Wales

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PERMEABLE REACTIVE BARRIER (PRB) TECHNOLOGY FOR THE REMEDIATION OF ACIDIC GROUNDWATER FROM ACID SULPHATE SOIL FLOODPLAIN IN SOUTH EAST NEW SOUTH WALES

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ABSTRACT

The release of acidic water contaminated with toxic metals such as aluminium (Al) and iron (Fe) resulting from pyrite oxidation within acid sulphate soils (ASSs) is a major environmental issue in Australia. Groundwater manipulation using weirs and modified floodgates is not effective in low-lying floodplains due to the high risk of flooding and their inability to prevent pyrite oxidation. Permeable reactive barrier (PRB) technology has been looked at as an alternative remediation technique in ASS terrain. Batch and short-term column tests were used to choose a suitable reactive material for the remediation of acidic water and the removal of Al and Fe, with recycled concrete deemed the most effective. Following these tests, a pilot-scale PRB was installed in ASS terrain on the Shoalhaven Floodplain, southeast New South Wales, Australia in October 2006. Despite a slow decrease in the performance of the PRB due to armouring effects caused by the precipitation of Al and Fe on the surface of the recycled concrete, continuous monitoring of the performance of the PRB over the last five years has shown an increase in pH to neutral (~ ph 7.3) and ~ 95% removal of Al and Fe from the groundwater. A long-term (30 months) column test conducted under controlled laboratory conditions for simulating the flow of acidic groundwater through the PRB determined that the predominant neutralisation reactions occurring within the PRB were the dissolution of carbonate/bicarbonate alkalinity, the re-dissolution of Al hydroxide precipitates and the re-dissolution of Fe oxyhydroxide minerals.

Keywords: Environmental geotechnics

1 INTRODUCTION

The acidification of coastal waterways because of acid sulphate soil (ASS) is an intractable environmental, economic and social problem within Australia. ASS are soils that contain appreciable amounts of sulphidic materials such as pyrite (FeS2), iron monosulphide (FeS), greigite (Fe3S4) and various organic sulphides in lower concentrations (Bush and Sullivan, 1997). If undisturbed and under reducing conditions, this pyritic material is relatively chemically inert. However, increased population since the 1960s and the installation of deep flood mitigation drains in southeast New South Wales (NSW) to enhance the usability of agricultural land has resulted in the environmental degradation of estuarine ecosystems (Lin et al., 1995) due to the oxidation of pyritic material and the generation of sulphuric acid (H2SO4). The acid liberates toxic metals such as aluminium (Al) and iron (Fe) from the clay minerals (Dent, 1986), which in turn leaches into nearby drains and waterbodies. Acidic drainage from ASS has severe environmental, economic and social problems. ASS have been estimated to affect over 3 million hectares of coastal Australia, with up to 0.6 million hectares in NSW alone (White et al., 1997). Acidic drainage water enriched with metals ions can be highly toxic to plants and gilled organisms and can corrode concrete and steel infrastructures (Sammut et al., 1996).

Low cost engineering strategies such as fixed-level weirs (Indraratna et al., 2001) and modified two-way floodgates (Glamore and Indraratna, 2004) have been installed in the Shoalhaven Floodplain, southeast NSW. While the weirs raised the groundwater table above the pyritic layer, they were unable to manage the leaching of stored acid already generated within the soil. These remediation techniques are not feasible in very low-lying areas, due to the risk of flooding during significant rainfall events. In addition, biological oxidation of pyrite under submerged conditions can still prevail if the organic content and sulphide constituents of the soils are high. A sub-surface horizontal lime fly ash barrier was installed in 2004 to decrease the oxidation of pyrite and to improve the groundwater and surface water quality (Indraratna et al., 2006). However, this barrier had only a localised impact on groundwater chemistry and its longevity was uncertain. Thus, another more suitable remediation technique is needed and the current research strategy is to examine the application of permeable reactive barriers (PRBs) for the treatment of acidic groundwater from ASS.

A PRB is an in-situ and passive remediation technique that utilises the natural hydraulic gradient to treat contaminated groundwater through physical, chemical and/or biological processes (Golab et al., 2009). PRBs commonly consist of a trench filled with reactive material specially selected to treat the contaminants of concern. The majority of PRBs worldwide have been used to treat chlorinated volatile organic compounds (Wilkin and Puls, 2003), acid mine drainage (Blowes et al., 2003), radionuclides and heavy metals (Gu et al., 2002). Research has just begun in Australia to
investigate the use of PRBs to treat acidic groundwater from ASSs (Waite et al., 2002; Indraratna et al., 2010). This paper outlines the findings of current research on the application of a PRB for the remediation of ASS in the Shoalhaven Floodplain of southeast New South Wales, Australia, including reactive material selection, PRB installation and monitoring as well as the identification of the dominant acid neutralisation processes occurring within the PRB through column experiments and field observation.

2 SELECTION OF REACTIVE MATERIAL

Many reactive materials could potentially be used in a PRB to treat acidic groundwater in ASS terrain. The selection of these materials requires a careful analysis of numerous performance parameters essential for PRB operation. Since the contaminants in the groundwater in ASS terrain are acidity as well as soluble [Al\(^{3+}\)] and [Fe\(^{3+}\)], the reactive material must be able to increase the groundwater pH and allow for the precipitation of [Al\(^{3+}\)] and [Fe\(^{3+}\)]. Extensive batch tests (Golab et al., 2006) and short-term column tests (Golab et al., 2009) have been conducted to screen 25 alkaline materials including recycled concrete, limestone, oyster shells, calcite-bearing zeolitic breccia, air-cooled blast furnace slag (ACBFS), lime and fly ash with drain water collected from the field (Figure 1). Recycled concretes and ACBFS achieved a pH consistent with the dissolution of lime (pH 11 to 12). The limestone and zeolitic breccia achieved a pH consistent with the dissolution of calcite (CaCO\(_3\)) (pH ~7.4).

Following the batch tests, recycled concrete and oyster shells were chosen for preliminary column tests for simulating flow of acidic water (pH 3, drain water collected from field site) through the barrier (Golab et al., 2009). Column tests are commonly performed on a wide range of contaminants for the determination of reactive material effectiveness (Waybrant et al., 2002, Komnititsus et al., 2004). Three columns of 1.5 L capacity (65 cm in length, 5 cm in internal diameter) were used with a water flow of 16 mL/min from the bottom to the top of the column (Figure 2). The columns contained: (1) crushed oyster shells (Column A), (2) recycled concrete of size range 1.18-10 mm (Column B) and (3) 50:50 recycled concrete:oyster shells (Column C). They were covered in dark plastic to exclude light to simulate the subsurface environment and to encourage potential growth of bacteria. Samples were collected from sampling ports located along the column and analysed for pH, electrical conductivity (EC), oxidation reduction potential (ORP) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) analysis for major anions and cations. The oyster shells (Column A) successfully maintained a pH above 6.8, due to the dissolution of CaCO\(_3\), even after 300 pore volumes (PVs defined here as the void volume of the column) of acidic water had passed through the column (Golab et al., 2009). However, the oyster shells were rapidly consumed by the acidity, causing the precipitation of Fe and sulphate (SO\(_{4}^{2-}\)) out of solution leading to clogging of the column. The recycled concrete maintained a pH above 10.5, removed Al to below the detection limit even after 90 PVs, and did not show any sign of diminished neutralisation capacity over the 30 day testing period. Column C produced an alkaline effluent of pH 9.8. However, recycled concrete appeared to be the most suitable material when cost, availability, permeability to allow groundwater flow though the PRB, acidic neutralisation capacity (ANC) and Al and Fe removal efficiencies were taken into consideration.

Figure 1: pH as a function of time for the selected reactive materials (Adapted from Golab et al., 2006).

![Figure 1](image1.png)

Figure 2: Schematic of preliminary column tests showing location of sampling ports (7.5, 10.5, 15, 20, 30, 40 and 60 cm).

![Figure 2](image2.png)
3 PRB INSTALLATION AND PERFORMANCE

A pilot-scale alkaline PRB (17.7 m x 1.2 m x 3.0 m) using recycled concrete aggregates to neutralise the acidic groundwater was installed in October 2006 at Manilda's environmental farm near Broughton Ck in ASS terrain in the Shoalhaven Floodplain, southeast NSW (Figure 3A). This site was specifically selected because: (1) the groundwater is acidic (~pH 3) with high Al (up to 40 mg/L) and Fe (up to 530 mg/L) levels; (2) the site is low-lying (0-1 m AHD); (3) there is a drain 15 m away for treated groundwater to flow into; and (4) there is a zone of preferential groundwater flow that can be intercepted by the PRB (Golab and Indraratna, 2009). The recycled concrete aggregates (40 mm diameter) used in the PRB were sourced from a refuse depot. The trench containing the aggregates was lined with geotextile fabric to protect the reactive material from physical clogging by soil and other particles. The chemical composition of the major cations in the virgin recycled concrete is predominantly Ca (57.3%), Fe (21.4%), Al (9.85%), Mg (5.27%), Si (3.06%) and others (3.04%) (Regmi et al., 2009). The performance of the PRB has been continuously monitored using 30 observation wells and 12 piezometers installed inside, up-gradient and down-gradient of the PRB (Figure 3B) for variations in the phreatic surface, hydraulic gradients, permeability and groundwater chemistry (pH, EC, ORP, temperature). Also pH, dissolved oxygen (DO), water pressure and temperature data were recorded on an hourly basis using two set of sensors which were installed within the PRB and connected to two dataloggers. Groundwater samples were also frequently collected for the analysis of [Al^{3+}], [Fe^{2+}] and other major trace metals using the standard methods for water and wastewater examination (APHA, 1998).

Figure 3: (A) Location of the study area, Shoalhaven Floodplain, southeast NSW (PRB is indicated with a star) and ASS risk of soils; (B) Layout of the PRB showing transect a-a (Adapted from Indraratna et al. (2010)).

Throughout the monitoring period, the groundwater pH up-gradient of the PRB has always been very acidic (pH 3.2-4.5; average pH 3.7) over the study period, as shown in Figure 4A. However, changes in pH are highly dependent on dilution during heavy rainfall events and the flushing of acid during small rainfall events. The groundwater pH inside the PRB has been consistently alkaline to neutral (pH 10.2 to 7.3). An initial increase in pH within the PRB was due to the dissolution of Ca-bearing cementitious materials within the recycled concrete aggregates and the release of carbonate alkalinity (Regmi et al., 2009). Despite the generation of acidity due to pyrite oxidation down-gradient of the PRB, the groundwater pH is ~6. This is a result of dilution of the existing acidic water by alkaline effluent from the PRB. High oxidation reduction potential (ORP) of up to 500 mV (Figure 4B) up-gradient of the PRB is indicative of pyrite oxidation in the study area. ORP decreased dramatically inside the PRB and has been maintained negative to small positive values indicating that the reducing condition is strongly dominant for the dissolution of limes from the concrete (Regmi et al., 2009). Most of the [Al^{3+}] and [Fe^{2+}] contained in the groundwater precipitated rapidly when alkaline minerals from the recycled concrete started to dissolve, as indicated in Figure 5. The maintenance of [Al^{3+}] and [Fe^{2+}] below average values of 2 and 0.5 mg/L inside the PRB over a 3.5 year monitoring period, respectively. The result indicates exceptional removal efficiency of the recycled concrete for [Al^{3+}] and [Fe^{2+}] (~95%). [Al^{3+}] and [Fe^{2+}] concentrations increased with the down-gradient distance away from the PRB. This is possibly due to the active oxidation of pyrite and the liberation of these metals from the clay minerals in the soil. The high concentrations of [Al^{3+}] (5.5-38 mg/L) and [Fe^{2+}] (44-234 mg/L) up-gradient of the PRB can be attributed to the aerobic (Indraratna et al., 2010) and highly acidic conditions (Figure 4). Slight decreases in the [Al^{3+}] and [Fe^{2+}] removal efficiencies over a 3.5
year monitoring period indicate that some chemical armouring of the surface of the reactive media has occurred and affected the reactivity of the recycled concrete (data not shown). However, this chemical armouring has negligible effects on the permeability of the PRB i.e. does not cause clogging, as reflected by steady piezometric head within the PRB (Indraratna et al., 2010). There were no discernible changes in the concentrations of the other ions (including sodium, chloride, potassium, magnesium and sulphate) within the PRB compared to their concentrations in groundwater up-gradient and down-gradient of the PRB. This indicates that these constituents are not influenced by the neutralisation reactions occurring within the PRB, which will be discussed in Section 4.

![Figure 4: Groundwater (A) pH and (B) ORP up-gradient (left), inside and down-gradient (right) of the PRB along transect a-a (Adapted from Regmi et al. (2009)).](image)

![Figure 5: Concentration of (A) Al and (B) Fe within the groundwater up-gradient (left), inside and down-gradient (right) of the PRB along transect a-a (Adapted from Regmi et al. (2009)).](image)

4 IDENTIFICATION OF NEUTRALISATION PROCESSES OCCURRING WITHIN PRB

A further column experiment was undertaken to determine the neutralisation reactions occurring within the PRB using a similar column set-up as described in Section 2. However, a synthetic acidic influent solution was used for simulating groundwater at the PRB field site using the average value of contaminants measured of a 6 month monitoring period. A constant flow rate of 2.4 mL/min was applied. The average total porosity of the reactive material was 0.52 (total PV was 534 mL). The recycled concrete effectively treated the acidic groundwater, resulting in near-neutral effluent pH and complete removal of Al and Fe until the depletion of alkalinity (Figure 6A). Three pH plateaus were observed: (1) pH at ~7.9-7.7 for 40 < PVs < 155 followed by a gradual decrease to pH ~6.5 at 235 PVs, (2) pH at ~4 for 300 < PVs < 500, and (3) pH at ~2.7 after 500 PVs. These plateaus were attributed to three distinct pH-buffering reactions: (1) the dissolution of carbonate/bicarbonate alkalinity from the concrete at near neutral pH, (2) the re-dissolution of Al hydroxide precipitates at ~ pH 4, as was similarly reported by Blowes et al. (2003), and (3) the re-dissolution of Fe
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oxyhydroxide minerals at pH < 3 (Regmi et al., 2010). The initial effluent pH was high (~11.2) due to the dissolution of a minor amount of portlandite (Ca(OH)₂) from the concrete and the generation of hydroxyl/carbonate alkalinity. The dissolution of calcium aluminate hydrated compounds (C₃A·H₂) and other Ca-bearing minerals, generated bicarbonate alkalinity for maintaining a near neutral effluent pH (~7.9-7.6) until 250 PVs. [Al³⁺] removal was >99% until the depletion of bicarbonate alkalinity. [Fe³⁺] removal was 100%, due to ferric oxyhydroxide buffering and the presence of goethite (FeOOH), until the depletion of Al oxyhydroxide buffering at 480 PVs. On completion of the experiment, concrete armoured with precipitates was removed from the column and examined using scanning electron microscopy-energy dispersive x-ray spectroscopy (SEM-EDS) in order to characterise the precipitates. EDS spectra for the virgin and armoured concrete (Figure 5B) confirmed the presence of Al and Fe-bearing minerals on the armoured concrete. Armouring on the surface of Ca-based reactive materials by precipitated minerals can influence the long-term performance of PRBs (Waybrant et al., 2002; Komnitsas et al., 2004). Armouring of the recycled concrete, as evidenced by white and orange precipitates within the column, could result in a decrease in the acid neutralising capacity (ANC) of the concrete. The accumulation of secondary minerals and chemical armouring of the recycled concrete by the precipitates (Figure 6B) decreased the actual ANC (71 mg/g as CaCO₃, 250 PV) of the concrete by approximately 50% compared to its theoretical ANC (145 mg/g as CaCO₃, 510 PV) (Regmi et al., 2010). Examination of the nature of geochemical clogging through chemical and microbiological analysis of barrier specimens is being planned.

![Diagram](attachment:Diagram.png)

Figure 6: (A) pH and the removal of Al and Fe as a function of pore volume; (B) SEM-EDS analysis of (inset a) virgin and (inset b) armoured recycled concrete (Adapted from Regmi et al. (2010)).

5 CONCLUSIONS

The performance of the pilot-scale PRB using recycled concrete for the remediation of acidic groundwater from ASS in the Shoalhaven Floodplain, southeast Australia has been monitored for the last five years. Despite monitoring revealing a slow decrease in performance due to ageing effects caused by the precipitation of Al and Fe on the recycled concrete aggregates, the PRB has been successful in maintaining a neutral pH (~pH 7.3) and removing ~95% dissolved [Al³⁺] and [Fe³⁺] from the groundwater. While the PRB cannot prevent the oxidation of pyrite in the soil, it can dramatically improve the down-gradient groundwater quality. The column experiment confirmed that the treatment mechanisms occurring within the PRB was mainly due to the release of carbonate/bicarbonate alkalinity and the precipitation of [Al³⁺] and [Fe³⁺] as hydroxide/oxyhydroxide minerals. Continuous precipitation within the PRB may, over time, decrease the surface area of the reactive material available for neutralising acidity, thus decreasing the longevity of the PRB. Research is currently underway to determine the longevity of the PRB and further study clogging within the PRB and the armouring of the surface of the recycled concrete by Al and Fe-bearing mineral precipitates.

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7 REFERENCES


