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Permeable Reactive Barrier (PRB) Technology: An Innovative Solution for the Remediation of Acidic Groundwater from Acid Sulphate Soil (ASS) Terrain

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ABSTRACT

The remediation of acidic groundwater contaminated with potentially toxic metals such as aluminium (Al) and iron (Fe) resulting from the oxidation of sulphidic materials in acid sulphate soils (ASSs) is a challenging geo-environmental problem that requires innovative engineering solutions. In low-lying coastal floodplains, the remediation strategies of groundwater manipulation (e.g. fixed-level weirs) and tidal buffering (e.g. two-way modified floodgates) are not feasible due to the risk of flooding during heavy rainfall events and their inability to prevent pyritic oxidation. In view of this in 2006, the first pilot subsurface permeable reactive barrier (PRB) using recycled concrete for the remediation of acidic groundwater (~ pH 3) was employed in ASS terrain in southeast New South Wales, Australia. While monitoring has confirmed the PRB has successfully neutralized the acidic groundwater to ~ pH 7.3 and removed ~ 95% of Al and Fe, this technology is not without its challenges. These have included the: (1) selection of the appropriate reactive material; (2) elucidation of the mechanisms involved in the neutralization of the acidic groundwater; (3) chemical armouring and possible clogging of the recycled concrete by Al and Fe oxy/hydroxide precipitates; and (4) thus, uncertainty regarding the longevity of the PRB. This paper will present details on the screening process of reactive materials, the installation of the PRB, the column experiments simulating the flow of acidic groundwater through the PRB for the determination of the predominant neutralization reactions occurring within the PRB, the long-term performance of the PRB and the current research strategy for determining its longevity.

INTRODUCTION

The acidification of coastal waterways because of acid sulphate soil (ASS) is a severe environmental, economic and social problem within Australia and worldwide. ASS have been estimated to affect over 3 million hectares of coastal Australia, with up to 0.6 million hectares in New South Wales (NSW) alone (White et al., 1997). ASSs are soils that contain appreciable amounts of sulphidic materials such as cubic iron pyrite (FeS_2), iron monosulphide (FeS), greigite (Fe_3S_4) and

various organic sulphides in lower concentrations (Bush and Sullivan, 1997). If undisturbed and under reducing conditions, pyritic material is relatively chemically inert. Increased population since the 1960s and the installation of deep flood mitigation drains in southeast NSW for the establishment of agricultural land has resulted in the environmental degradation of estuarine ecosystems (Lin et al., 1995). This is due to pyrite oxidation and the generation of acidic drainage with abnormally high trace metals such as aluminium (Al) and iron (Fe), which leaches from the ASS into nearby waterways. This acidic drainage has the capacity to directly impact upon the basic natural assets of soil, water and biota, and may affect agriculture, fishing, aquaculture, recreation, tourism, as well as human health and visual amenity (Sammut et al., 1996).

Low-cost remediation strategies of fixed-level weirs (Indraratna et al., 2001) and modified two-way floodgates (Glamore and Indraratna, 2004) were installed in the Shoalhaven Floodplain, southeast NSW in order to raise the groundwater table above the pyrite layer and allow for tidal buffering of acidic drain water, respectively. While the weirs offered a substantial improvement in the management of acid generation and transport to nearby drains with respect to the existing flood mitigation drainage system, they did not improve long-term groundwater or soil quality due to their inability to manage the leaching of stored acid previously generated within the ASS profile. The groundwater pH was highly acidic (pH 3.5-4.0) and the concentration of Al remained high (30-100 mg/L) due to dissolution of acidic minerals (Indraratna et al., 2001). The modified floodgates decreased the hydraulic gradient between the drain and groundwater and, thus, reduced the 'acid reservoir' effect and decreased drain water Al and Fe by 50%. However, these remediation strategies are unsuitable in low-lying floodplains due to the risk of flooding during significant rainfall events. A sub-surface horizontal lime-fly ash barrier installed in 2004 to decrease pyritic oxidation and improve ground and surface water quality had only a localized impact on groundwater chemistry and its longevity was uncertain (Banasiak, 2004; Indraratna et al., 2006). Therefore, a more suitable remediation technique is needed and the focus of current research is to examine the application of a permeable reactive barrier (PRB) for the treatment of acidic groundwater from ASS.

A PRB is an in-situ and passive remediation technique that utilizes the natural hydraulic gradient to treat contaminated groundwater through physical, chemical and/or biological processes (Golab et al., 2009). They commonly consist of a trench filled with reactive material selected to treat contaminants of concern. Research has just begun in Australia to investigate the use of PRBs to treat acidic groundwater from ASS (Waite et al., 2002; Indraratna et al., 2010). This paper outlines the findings of current research on the application of a PRB utilizing recycled concrete for the remediation of ASS in the Shoalhaven Floodplain of southeast NSW, Australia. This includes details on the selection process of reactive materials, PRB installation and the evaluation of its long-term performance as well as the determination of the predominant neutralization reactions occurring within the PRB and the current challenges and uncertainties for determining its longevity.

SELECTION OF REACTIVE MATERIAL

Selection of an appropriate reactive material is a major challenge in PRB design that requires thorough analysis of numerous performance parameters essential for PRB operation. In the case of ASS remediation, the reactive material must be able to increase the groundwater pH that allows for the precipitation of Al and Fe. Extensive batch tests (Golab et al., 2006) and short-term column tests (Golab et al., 2009) were performed to screen 25 alkaline materials including recycled concretes, limestone, oyster shells, calcite-bearing zeolitic breccias, air-cooled blast furnace slag (ACBFS), lime and fly ash with drain water collected from the field site (~ pH 3.0-3.2; Al 113 mg/L; Fe 3.98 mg/L) (Figure 1). The recycled concretes, fly ash and ACBFS achieved a pH consistent with the dissolution of lime (pH 11-12). The limestone and zeolitic breccias achieved a pH consistent with the dissolution of calcite (CaCO_3) (~ pH 7.4). Recycled concrete, ACBFS, three zeolitic breccias and limestone successfully removed ~ 98% of the Al and Fe from the drain water and were, thus, shortlisted and further examined for acid neutralization capacity (ANC) by repeatedly replacing the acidic water and monitoring the resultant pH.

Following these batch tests, recycled concrete and oyster shells were chosen for preliminary short-term columns tests for simulating flow of acidic water (pH 3, drain water collected from field site) through the barrier. Three columns of 1.5 L capacity (65 cm in length, 5 cm internal diameter) were used with a water flow of 16 mL/min from the bottom to the top of the column (Figure 2.).

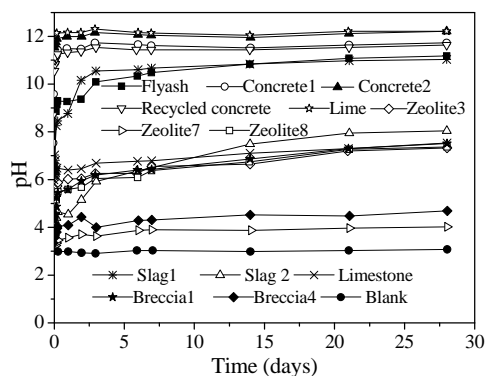


Figure 1. pH vs. time for the selected reactive materials (Adapted from Golab et al. (2006)).

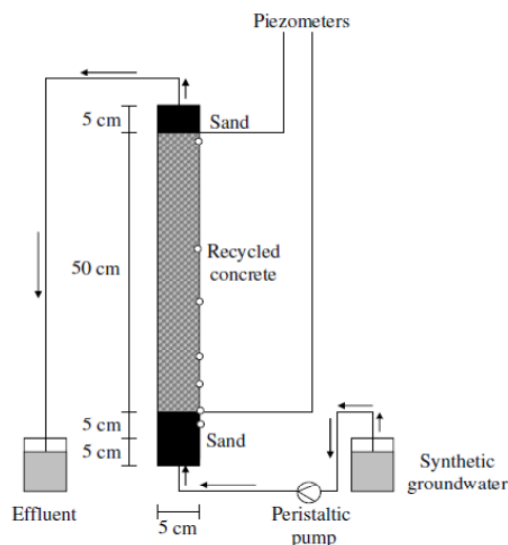


Figure 2. Schematic of preliminary column tests and location of sample ports (7.5, 10.5, 15, 20, 30, 40, 60 cm).

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 simulate anaerobic conditions in the subsurface environment and encourage potential bacteria growth. Samples were collected from sampling ports located along the column and

analyzed for pH, electrical conductivity (EC), oxidation reduction potential (ORP) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) analysis for major anions and cations.

While the oyster shells maintained a pH > 6.8, due to the dissolution of CaCO₃, even after 300 pore volumes (PVs, defined here as the column void volume) of acidic water had passed through the column (Golab et al., 2009), they were rapidly consumed by the acidity, causing precipitation of Fe and sulphate (SO₄²⁻) leading to clogging of Column A. The recycled concrete maintained a pH > 10.5, removed Al below the detection limit even after 90 PVs and did not show any sign of diminished ANC. Although Column C produced an alkaline effluent of pH 9.8, recycled concrete was deemed the most suitable PRB reactive material when cost, availability, ANC, grain size, Al and Fe removal efficiencies were considered.

PRB INSTALLATION AND LONG-TERM PERFORMANCE

In October 2006, the first pilot PRB using recycled concrete for the remediation of acidic groundwater was installed at Manildra Group's environmental farm in ASS terrain in southeast NSW, Australia (Figure 3A). Baseline monitoring of the study site was carried out for over a year prior to installation of the PRB for the purpose of site characterization. The parameters that were studied include variations in phreatic surface, groundwater chemical composition, soil hydraulic conductivity, porosity and grain size, and hydraulic conductivity. The site was chosen 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) it is low-lying (0-1 m Australian Height Datum (AHD)); (3) there is a nearby drain for treated groundwater to flow into; and (4) there is a zone of preferential groundwater flow for passive interception by the PRB.

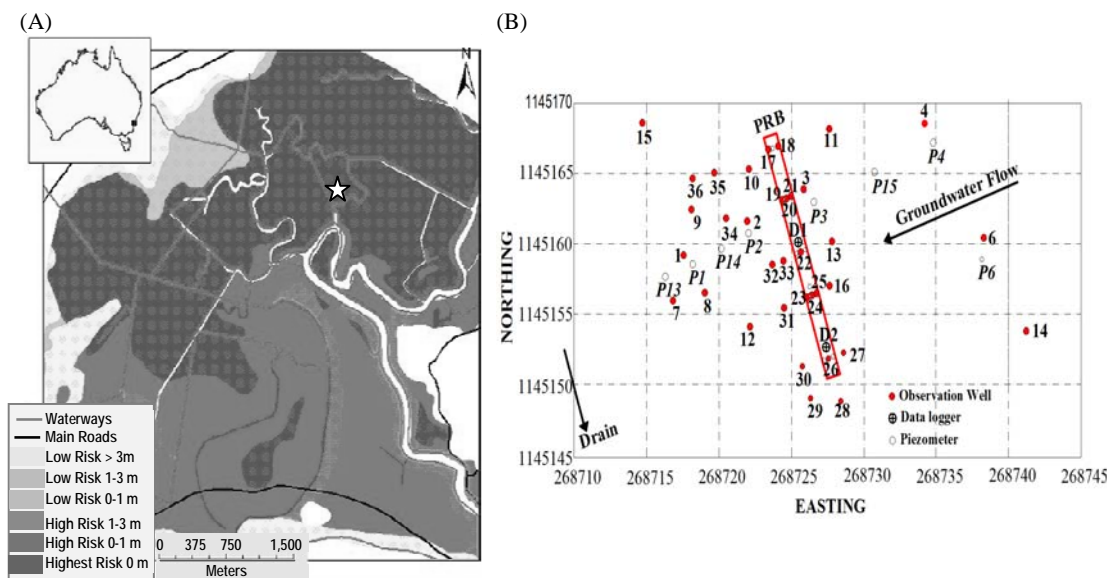


Figure 3. (A) Location of study area, Shoalhaven Floodplain, southeast NSW (PRB is indicated with a star) and ASS risk of soils; (B) Layout of PRB (Adapted from Indraratna et al. (2010)).

The dimensions of the PRB (17.7 m × 1.2 m × 3.0 m) were determined based on the results of the column tests and soil conditions at the field site. These dimensions were used to optimize residence time within the barrier, to ensure that acidic groundwater did not pass under the PRB and to capture the zone of maximum groundwater flow. Excavation and backfilling techniques utilized in the installation of the PRB were determined based on geotechnical testing. Steel boxes were placed along the walls of the trenches to ensure that the walls did not collapse due to the low undrained shear strength of the soil (< 10 kPa) (Golab and Indraratna, 2009). The trench containing the recycled concrete was lined with geotextile fabric to prevent physical clogging of the PRB by soil and other particles. The recycled concrete aggregates (40 mm diameter) used in the PRB were sourced from a refuse depot, after the demolition of old concrete structures. The chemical composition of the virgin recycled concrete was primarily Ca (57.3%), Fe (21.4%), Al (9.85%), Mg (5.27%), Si (3.06%) and others (3.04%) (Regmi et al., 2011). The performance of the PRB has been continuously monitored for the last 4.5 years using 36 observation wells, 15 piezometers and 2 data loggers 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, DO, ORP, major anions and cations).

Up-gradient of the PRB, the groundwater is acidic (~ pH 3.4-5.6; average pH 4.2; Figure 4), with changes in pH depending on dilution during large rainfall events and flushing of acid during small rainfall events following dry periods (e.g. Nov 2006 and Nov 2008). The average pH down-gradient of the PRB is > 6.0 due to dilution of existing acidic water by alkaline effluent from the PRB. Groundwater inside the PRB is consistently alkaline to neutral (pH 10.2-7.3; average 7.6); thus, confirming that the recycled concrete is an effective neutralising material for acidic water despite the variable environmental field conditions. The initial increase in pH was attributed to the dissolution of Ca-bearing minerals within the recycled concrete and the release of carbonate alkalinity (Indraratna et al., 2011), which will be further discussed in the following section.

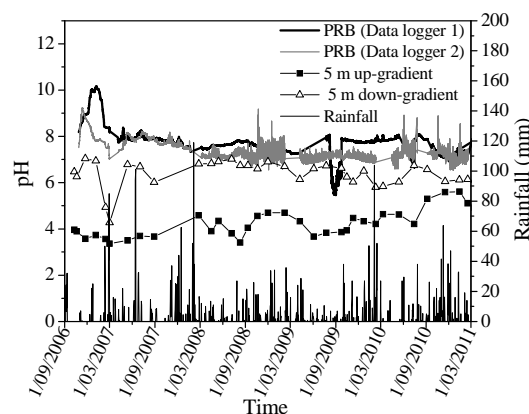


Figure 4. Temporal and spatial variation of groundwater pH up-gradient, inside and down-gradient of the PRB (Adapted from Indraratna et al. (2011)).

Monitoring of Al and Fe in the groundwater shows that most of the Al and Fe precipitated rapidly within the PRB (Figure 5). The maintenance of Al and Fe below average values of 0.5 and 2 mg/L, respectively, inside the PRB over the 4.5-year monitoring period indicates the excellent removal efficiency of recycled concrete for Al and Fe. The concentration of Fe and Al increased with distance down-gradient of the PRB, due to active and on-going pyrite oxidation generating fresh acid, releasing Al and Fe into the groundwater. However, slight decreases in groundwater pH, Al and Fe removal over the monitoring period has occurred due to exhaustion of alkalinity generating minerals as well as assumed armouring by precipitates over the surface of the recycled concrete. However, this armouring has had negligible effects on PRB permeability (i.e. has not caused clogging), as indicated by a steady piezometric head within the PRB (Indraratna et al., 2010).

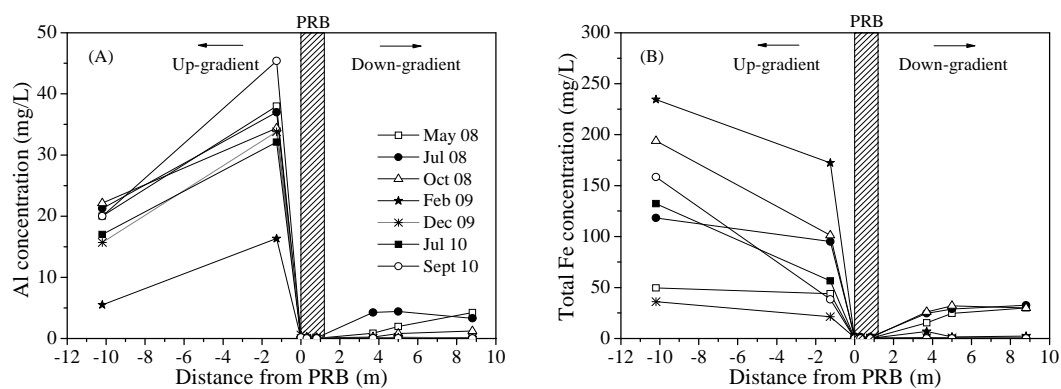


Figure 5. (A) Al and (B) Total Fe concentration in groundwater up-gradient, inside and down-gradient of the PRB along transect a-a (Adapted from Indraratna et al. (2011)).

IDENTIFICATION OF NEUTRALIZATION MECHANISMS OCCURRING WITHIN PRB

A further long-term column experiment using recycled concrete was undertaken to determine the neutralization reactions occurring within the PRB (Regmi et al., 2011). A similar column set-up, as described previously, was used. However in this case, a synthetic acidic influent of constant flow rate 2.4 mL/min was used to simulate groundwater from the field site using the average value of contaminants measured over a 6-month monitoring period. The recycled concrete used in the column experiment was from the same batch of concrete used in the PRB. However, the large recycled concrete fragments were crushed to make smaller aggregates suitable for the column experiment. The particle size distribution of the recycled concrete used in the column was uniformly graded aggregates in the range between 1.18 and 10 mm. The average porosity of the recycled concrete was 0.52 (total pore volume (PV) was 534 mL). The recycled concrete successfully neutralized the acidic influent to a near-neutral pH and completely removed Al and Fe (Figure 6A) until depletion of the available alkalinity at 300 PVs (Figure 6B). Three pH plateaus were observed and attributed to three pH-buffering reactions: (1) dissolution

of carbonate/bicarbonate alkalinity at pH 7.9-7.7 ($40 < \text{PVs} < 155$) followed by a gradual decrease to pH 6.5 (at 235 PVs); (2) the re-dissolution of Al hydroxide precipitates at \sim pH 4 ($300 < \text{PVs} < 500$); and (3) the re-dissolution of Fe oxyhydroxide precipitates at \sim pH 3 (> 500 PVs). The presence of Fe oxyhydroxides can result in the further generation of acidity within the column or PRB (Indraratna et al., 2011). The initial high pH (\sim 11.2, < 40 PVs) was due to the dissolution of a minor amount of portlandite ($\text{Ca}(\text{OH})_2$) from the concrete and the generation of hydroxyl/carbonate alkalinity. The generation of bicarbonate alkalinity after 40 PVs was due to the dissolution of calcium aluminate hydrated (C-A-H) compounds (e.g. anorthite ($\text{CaAl}_2\text{Si}_2\text{O}_8$) from within the recycled concrete (Regmi et al., 2010).

Armouring of the recycled concrete, as evidenced by white and orange precipitates within the column, resulted in a decrease in the actual ANC of the concrete (71 mg/g as CaCO_3 , 250 PVs) by \sim 50% compared to its theoretical ANC (145 mg/g as CaCO_3 , 510 PVs). Scanning electron microscopy-energy dispersive x-ray spectroscopy (SEM-EDS) of the armoured concrete confirmed the presence of Al and Fe-bearing minerals within the precipitates. Although a decrease in hydraulic conductivity within the column was observed (Figure 6B) due to armouring of the concrete by precipitates, this would not be a major problem with the larger concrete particle sizes employed in the PRB compared to the column experiment.

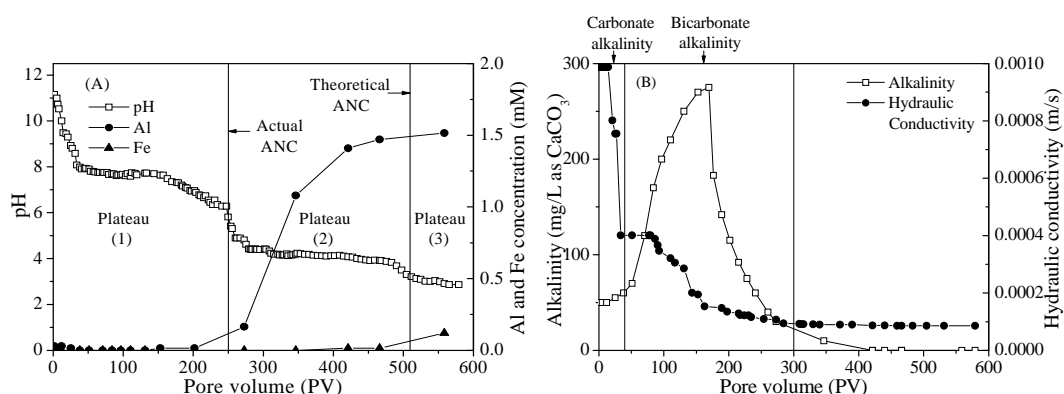


Figure 6. (A) pH and removal of Al and Fe; and (B) Alkalinity and hydraulic conductivity vs. pore volume (Adapted from Regmi et al. (2011)).

DETERMINATION OF PRB LONGEVITY

The precipitates formed in the PRB during neutralization of the acidic groundwater might cause extensive armouring over time, which would in turn decrease the treatment efficiency of the reactive material by decreasing the exposed surface area of the reactive material. Results from long-term column tests confirmed that the loss of ANC caused by chemical armouring is a more significant threat on the performance of recycled concrete than the exhaustion of alkalinity under high Al and Fe feed concentrations. Furthermore, fine precipitates formed inside the PRB and accumulated in the pore space between the reactive media may alter groundwater flow paths through the barrier. Excessive precipitation of Al and Fe metal oxy/hydroxides inside the PRB may threaten its longevity. Thus, it is of utmost

importance to understand the chemical behaviour and model potential armouring/clogging effects in order to evaluate the efficiency and longevity of the PRB.

Extensive studies for physical clogging in granular media have been done to predict PRB longevity (e.g. Indraratna and Vafai (1997)). However, there are limited studies on the long-term performance of PRBs, especially with respect to chemical and/or biological clogging (Liang et al., 2003). Some researchers (e.g. Li et al. (2006)) have suggested numerical modeling for the assessment of clogging by developing a calibrated groundwater flow and reactive transport model. Research is currently being undertaken at the University of Wollongong (UOW) to quantitatively assess changes in flow behaviour (Darcian or non-Darcian) due to armouring/clogging in order to develop a time-dependent porous medium flow model combining particle retention and groundwater flow with chemical precipitation. This will be used to determine the corresponding reduction in void space within the PRB per unit volume, thereby analysing the interrelated effects of acidic flow induced clogging and PRB effectiveness i.e. longevity of the PRB.

CONCLUSIONS

This paper highlights the current research strategy for the remediation of contaminated groundwater from ASS in Australia and demonstrates the effectiveness of the first pilot PRB in ASS terrain. Prior to installation of the PRB, the first challenge was the determination of the optimum reactive material for use in the pilot PRB. Batch tests and short-term column experiments on 25 alkaline materials confirmed that recycled concrete was the most suitable reactive material because of its high neutralization capacity and availability. Despite a gradual lowering of the performance of the PRB during the monitoring period due to armouring effects caused by the precipitation of Al and Fe on the surface of the recycled concrete, the groundwater pH has increased to neutral (~ pH 7.3) and ~ 95% of Al and Fe were removed from the groundwater. A long-term column test under controlled laboratory conditions simulating acidic groundwater flow through the PRB was used to evaluate the predominant neutralization reactions occurring within the PRB, which were the dissolution of carbonate/bicarbonate alkalinity and the re-dissolution of Al hydroxide and Fe oxyhydroxides minerals. Longevity and long-term performance of PRBs are an inherent uncertainty, and in this case, over time, continuous precipitation of Al and Fe oxy/hydroxide minerals may decrease the surface area of the recycled concrete available for neutralizing acidity, thus decreasing the efficiency of the concrete. Indeed, chemical armouring during the long-term column experiment decreased the actual ANC of the recycled concrete by ~50% compared to the theoretical ANC. Thus, research is currently being undertaken to establish a rational approach for quantifying armouring/clogging of the PRB treating acidic groundwater from ASS terrain through geo-hydrological and geochemical modeling. Changes in flow behavior due to armouring/clogging are being extensively studied in column experiments and the PRB in order to develop a time-dependent flow model combining particle retention, groundwater flow and geochemical kinetics. Laboratory testing and trial injections of an alkaline wastewater into an established PRB are

being carried out to investigate how conditioning of the recycled concrete with alkaline wastewater enhances PRB neutralising capacity and longevity.

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