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Mineral Precipitation and the Associated Reduction of Hydraulic Conductivity in a PRB

U. Pathirage and B. Indraratna

Centre for Geomechanics and Railway Engineering, University of Wollongong, Australia

ABSTRACT: Permeable reactive barriers (PRB) have been used worldwide for the in-situ treatment of contaminated ground water. One such contamination found in Australia is acidic groundwater enriched with high concentrations of dissolved aluminium and iron produced in acid sulfate soil (ASS) terrains. To treat this acidity in groundwater, a PRB was installed in the Shoalhaven Floodplain and then monitored over a seven year period. This remediation process was satisfactory but there was a small decrease in efficiency due to a secondary mineral precipitation (aluminium and iron oxy/hydroxides) which reduced the hydraulic conductivity of the PRB. Numerical modelling carried out through MODFLOW and RT3D software showed that this reduction in hydraulic conductivity due to secondary mineral precipitation was 3% at the entrance of the PRB after seven years of installation. This result was satisfactory considering that the predicted longevity of the barrier was 19.5 years, assuming a mean groundwater velocity of 0.05 m/day.

1 INTRODUCTION

For the first time in Australia, this study mainly focuses on combining geochemistry with geohydraulics to capture time-dependent modelling and performance analysis with respect to the remediation of acidic groundwater using a PR Binstalled in the Shoalhaven Floodplain, South of Sydney, where acidic groundwater generated from acid sulfate soils create severe environmental, social, and economic problems. Recycled concrete aggregates were used as the reactive material in this PRB to remediate the acidic groundwater. This acidic groundwater consists of low pH and high concentrations of toxic heavy metals such as dissolved aluminium and iron, so it was important to neutralise the groundwater and remove these toxic metals to an acceptable limit.

The alkalinity generated from recycled concrete can neutralise the acidity and also precipitate the dissolved Al and Fe in their forms of oxy/hydroxides. This secondary mineral precipitation would result in chemical armouring (encrustation on reactive surface) and clogging (accumulation on pores), which would reduce the efficiency of the reactive material and the porosity and hydraulic conductivity of the reactive medium. Chemical armouring/clogging of PRB due to mineral precipitates has not studied in depth, sothis paper presents an original modelling and field verification for a PRB in acid sulfate soil terrains.

2 METHODOLOGY

2.1 *Pilot-scale PRB*

The PRB installed at the Manildra Environmental farm was 17.7m long, 2.1m wide and 3m deep. A barrier trench was excavated and lined with geotextile material, and then filled with crushed recycled particles of concrete (d_{50} = 40mm). The geotextile was used to prevent the recycled concrete from being clogged by sand, clay and other debris in the groundwater. There are 36 observation wells, 15 piezometers and 3 data loggers to monitor the water quality parameters and hydraulic head at the up-gradient, inside and down-gradient of the PRB. Samples of groundwater were collected on a monthly basis and analysed for Al^{3+}, total Fe (Fe^{2+} and Fe^{3+}), primary cations (Ca^{2+}, Mg^{2+}, Na^+, K^+), anions (Cl^- and SO_4^{2-}), alkalinity and acidity.

2.2 *Model development*

This study developed an innovative model that combined the geochemical reaction kinetics with transient groundwater flows, which was then incorporated into the commercial finite different codes, MODFLOW and RT3D. A novel algorithm was also developed for RT3D to compute the geochemical reactions taking place in the PRB.

The development of this detailed mathematical model is given in Indraratna et al., (2014a and b), but for clarity in this paper, the most important equations are given below. The reaction kinetics for generating alkalinity from recycled concrete and secondary mineral precipitation was captured in the model through the Transition State Theory (Eq. 1).

$$r = -k_r \left(1 - \frac{IAP}{k_{eq}} \right) \qquad (1)$$

where r is the reaction rate, k_r is the effective rate coefficient, IAP is the ion activity product, k_{eq} is the equilibrium solubility constant. Saturation index (SI) (Eq. 2) is used to calculate the IAP/k_{eq} values using PHREEQC software.

$$SI = \log(IAP) - \log(k_{eq}) \qquad (2)$$

The changes in porosity due to secondary mineral precipitation on recycled concrete particles were calculated using Eqs. 3 and 4.

$$\frac{\partial \phi_k}{\partial t} = M_k R_k \qquad (3)$$

$$n_t = n_0 - \sum_{k=1}^{N_m} M_k R_k t \qquad (4)$$

where ϕ_k is the volume fraction of precipitated mineral, M_k is the molar volume of mineral ($m^3 mol^{-1}$) and R_k is the total reaction rate for a particular substance ($mol m^{-3}_{bulk} s^{-1}$), N_m is the number of minerals and n_0 and n_t are the initial porosity and porosity at time t, respectively. The change in hydraulic conductivity was calculated using the normalized Kozeny Carman equation (Eq. 5).

$$K = K_0 \left[\frac{n_0 - \Delta n_t}{n_0} \right]^3 / \left[\frac{1 - n_0 + \Delta n_t}{1 - n_0} \right]^2 \qquad (5)$$

where K_0 is the initial hydraulic conductivity and Δn_t is the difference in porosity at two consecutive time intervals.

The centre line of the PRB was discretised into a 0.1m x 0.1m mesh that was 1.2m wide (Fig. 1).
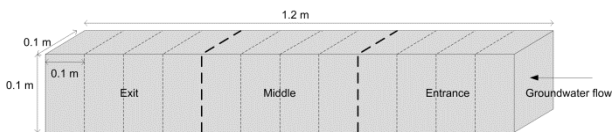

Fig. 1 Finite different mesh (after Indraratna et al., 2014a)

A transient groundwater flow condition was assumed and the pressure head calculation was done through Eq. 6.

$$h = \left(\exp\left[-\frac{\mu^2 BK_0}{S\sum_{k=1}^{N_m} M_k R_k} \frac{(1-n_0)^2}{n_0^3} \left\{ \begin{array}{l} \alpha^2\left(1.5 + 1/\beta\right) \\ -3(\alpha + \ln\beta) \end{array} \right\} \right] \right) \cdot$$
$$(C \sin \mu x + D \cos \mu x)$$

where $\alpha = n_0 + \sum_{k=1}^{N_m} M_k R_k t$ and

$$\beta = 1 - n_0 - \sum_{k=1}^{N_m} M_k R_k t \qquad (6)$$

where B is the thickness of the aquifer, S is the storage co-efficient, μ, C and D are constants which can be evaluated using the initial head conditions at both ends of the PRB. RT3D was used to solve the contaminant transport equation (Eq. 7).

$$R_e \frac{\partial C}{\partial t} = D \frac{\partial^2[C]}{\partial x^2} - u_b \frac{\partial[C]}{\partial x} - R_k M_k C \qquad (7)$$

where C is the concentration of the contaminant, R_e is the retardation coefficient ($R_e=1$, as sorption was ignored), D is the dispersion coefficient with a longitudinal dispersivity of 0.3 m. A mean groundwater flow velocity (u_b) of 0.05 m/day was assumed for an approximated PRB porosity of 50% in the model.

Eq. 6 was used to calculate the starting head for every time step because it has the ability to update the change in porosity and hydraulic conductivity due to secondary mineral precipitation (Indraratna et al., 2014a and b). Therefore, MODFLOW was run to couple the advection, diffusion and dispersion (Eq. 7) with RT3D.

3 RESULTS AND DISCUSSION

3.1 *Performance of PRB*

The PRB was promising in terms neutralizing the acidic groundwater. Fig. 2 shows the pH in the up-gradient, inside, and down-gradient of the PRB. Data loggers inside the PRB have monitored pH above 7 for the past seven years of operation (pH 8 to 7.2 except for an anomaly at the start due to a not fully saturated condition).
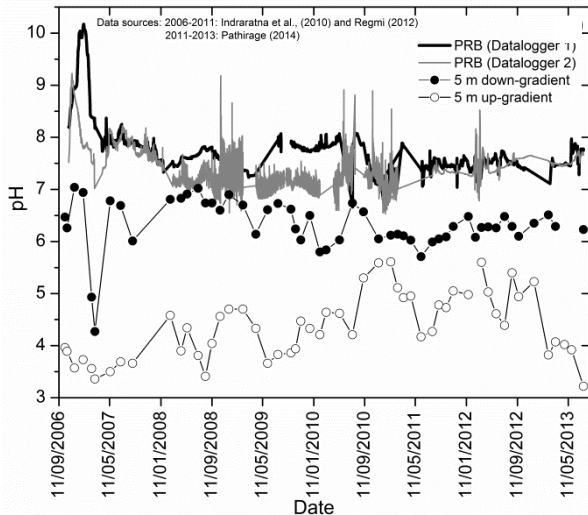
Fig. 2 pH profile in the up-gradient, inside and down-gradient of the PRB (after Indraratna et al., 2014a)

The groundwater analysis data from 2007-2013 revealed that the Al^{3+} concentration at the up-gradient varied from 1.5-51mg/L, while the total Fe concentration varied from 3.3-291mg/L. As Fig. 3a and b shows, the concentrations of Al^{3+} and total Fe were less than 2 and 0.5mg/L, respectively after remediation inside the PRB. There was a small increase in these ion concentrations at the down-gradient that was subjected to mixing with untreated acidic groundwater.
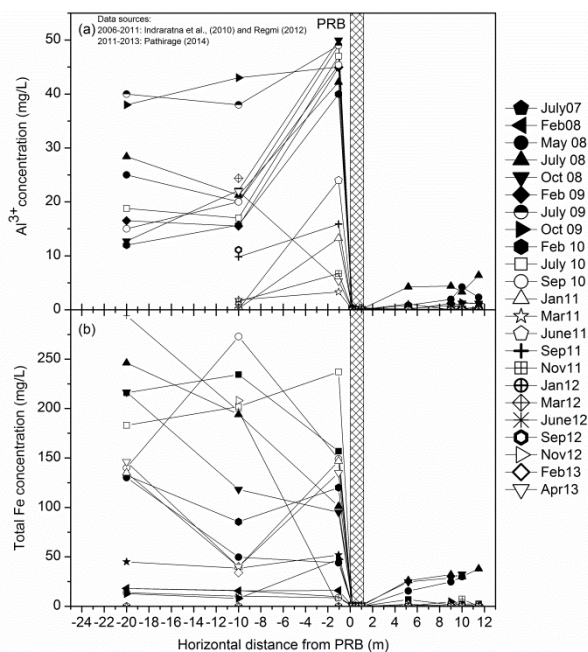

Fig. 3 Concentration of (a) Al^{3+} and (b) total Fe in the up-gradient, inside and down-gradient of the PRB (after Indraratna et al., 2014a)

3.2 Model output

The model predictions and observed data from the field were in agreement, as Fig. 4 shows. The average values of up-gradient groundwater pH, Al^{3+} and total Fe concentrations were 3.6, 27mg/l and 80 mg/L, respectively for 2013. The average values of treated groundwater pH, Al^{3+} and total Fe concentrations inside the PRB were 7 and 1mg/L each, respectively. The values inside the PRB predicted by the model were 7.3, 0.5mg/L and 0mg/L for pH, Al^{3+} and total Fe concentrations, respectively.
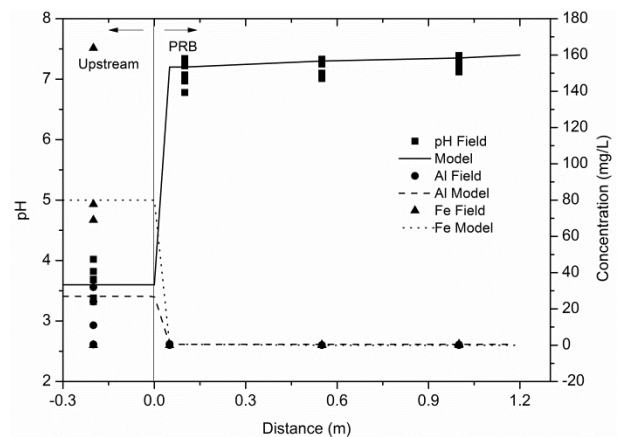

Fig. 4 Model predictions for pH, Al^{3+} and total Fe concentrations (after Pathirage et al., 2014)

The abrupt decrease in these ions confirmed that they had been precipitated inside the PRB in their forms of oxy/hydroxides, a result that would decrease the porosity and hydraulic conductivity.

There were 3%, 0.01%, and 0.003% reductions in the entrance, middle, and exit zones, respectively for the PRB hydraulic conductivity after seven years of operation (Indraratna et al., 2014b).

3.3 Longevity of PRB

The longevity of the PRB depends on the exhaustion rate of reactive material and the precipitation rate of secondary minerals (Pathirage and Indraratna, 2014), although continuous secondary mineral precipitation would decrease its effectiveness over time because it clogs the reactive surfaces of recycled concrete particles and reduces the acid neutralisation capacity (ANC).

The column experiments carried out by Pathirage (2014)revealed that the reduction in ANC due to secondary mineral precipitation was 54%. This implies that the long term performance of the PRB would be threatened by the exhaustion of reactive material due to acid neutralisation and armouring of the reactive surfaces by secondary minerals. This pilot-scale PRB contained 80 tonnes of recycled concrete (ANC of 146 g/kg), from

which a capacity of at least 11.7 tonnes of acid neutralisation was expected to be available in this PRB. With a mean groundwater flow velocity of 0.05 m/day and with an initial PRB porosity of 50%, acid transportation through the PRB was about 4.85 x 10^5 L/year. The averaged acidity at the study site from September 2010 to July 2012 was 565 mg/L (equivalent to $CaCO_3$), with a corresponding consumption of reactive material of 0.274 t/year. To consume all these acid neutralising materials would take 42.7 years without considering the effect of armouring caused by the precipitation of secondary minerals. When this effect on ANC was considered, (i.e. 54%), the assessed longevity of the PRB would be 19.5 years for an averaged groundwater flow of 0.05 m/day. The computed longevity of 19.5 years would differ permitting the velocity of groundwater flow and corresponding consumption of reactive material shown in Fig. 5 (Pathirage and Indraratna, 2014).Consumption of reactive materials depends on the groundwater flow as the amount of alkalinity consumed, depends on the amount of acidity brought into PRB. Clogging influences the consumption of reactive minerals because of the adherence of secondary minerals onto reactive surfaces.
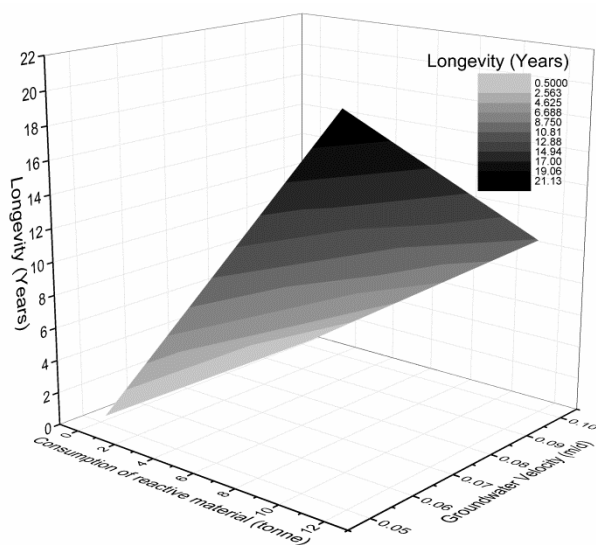

Fig. 5 Longevity of the PRB (after Pathirage and Indraratna 2015)

4 CONCLUSIONS

This paper shows that recycled concrete aggregates can be utilized to treat acidic groundwater in acid sulfate soil terrains. The pH inside the PRB was almost neutral from the time of installation until now, and the removal of dissolved aluminium and iron from groundwater looks promising. The results predicted by the MODFLOW and RT3D models were in good agreement with the field data, and thus confirmed the suitability of the developed mathematical model. The reduction in hydraulic conductivity at the entrance of the PRB was only 3% and it was almost negligible at the middle and exit zones after seven years of operation. The estimated longevity of the PRB was 19.5 years for a mean groundwater velocity of 0.05m/day. This means the developed mathematical and numerical model can be used by geotechnical engineers and environmental scientists for the remediation of acidic groundwater via PRB in acid sulfate soil terrains.

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