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Coupled hydro-geochemical modelling of a permeable reactive barrier for treating acidic groundwater

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ABSTRACT

This study focuses on coupling geochemistry with geo-hydraulics to enable time-dependent modelling of the remediation of acidic groundwater using an alkaline permeable reactive barrier (PRB). Chemical clogging due to secondary mineral precipitates reduces the porosity and hydraulic conductivity of the reactive medium. The governing equations are incorporated into commercial numerical codes, MODFLOW and RT3D. An original algorithm was developed for RT3D to simulate geochemical reactions occurring in the PRB. The results and the model predictions are in agreement, confirming that the hydraulic conductivity reduction due to mineral precipitation occurs at the start of permeation and continues until halfway through the testing phase.

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1. Introduction

Acidic groundwater generated from acid sulphate soil (ASS), which occupies over 200,000 km^2 of the Australian land is a major environmental and socio-economic problem in Australia. Changes in land use pattern (e.g., construction of deep flood mitigation drains) and hydrological systems (e.g., rainy and drought seasons) can promote the oxidation of ASS (pyrite) in shallow zones, with the associated generation of sulphuric acid in soil, which results in mobilizing toxic metals (aluminium (Al) and iron (Fe)) from the soil [1–3]. Therefore, the transportation of acidic water along with high concentrations of dissolved Al and Fe towards the water bodies has significantly degraded the coastal environment of Australia.

A permeable reactive barrier (PRB) offers an in situ technology for passive treatment of contaminated groundwater [4–7]. Recycled concrete has been recommended as one of the suitable reactive media for the PRB based on the batch test analysis among 24 different types of alkaline materials [8] for its ability to remove Al and Fe effectively out of solution, and most importantly to maintain near neutral pH for a considerable time. A pilot-scale PRB (17.7 m × 1.2 m × 3.0 m) was installed in the acid sulphate soil terrain located in Lower Shoalhaven floodplains area near the town of Bombaderry, (about 100 km South of Sydney) in October 2006 (Fig. 1). The PRB was filled with crushed recycled concrete (d_{50} = 40 mm) and the trench was lined with geotextile fabric to protect the reactive media from physical clogging by soil and other fine particles entering the barrier. A total of 30 observation wells and 15 piezometers were installed inside, up-gradient and down-gradient of the PRB to monitor phreatic surface variations, hydraulic gradients, permeability and groundwater chemistry. Groundwater elevation and water quality parameters such as pH, oxidation reduction potential (ORP) and temperature were directly measured in the field every month from October 2006 using water level metre and multi-parameter field electrode probes. In addition, pH, DO (dissolved oxygen), water pressure, and temperature were measured hourly by two multi-parameter automated data loggers installed within the barrier. Groundwater samples were collected frequently for analysis of iron, aluminium, major cations, anions and other trace metals. To the knowledge of the authors, this is only the second pilot scaled PRB under reducing conditions that has been installed for treating acidic water from acid sulphate soil after a natural limestone PRB reported by [9].

Generally the performance of PRBs has been satisfactory [4,10–14]. On the other hand, questions remain about the

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Nomenclature

r	reaction rate ($molm^{-3}_{bulk}s^{-1}$)
k_{eff}	effective rate coefficient ($molm^{-3}_{bulk}s^{-1}$)
IAP	ion activity product (depend on the reaction)
K_{eq}	solubility constant (depend on the reaction)
S_{Fe^0}	current reactive surface area of zero-valent iron (m^2)
$S^0_{Fe^0}$	initial reactive surface area of zero-valent iron (m^2)
φ_{Fe^0}	current volume fraction of zero-valent iron
$\varphi^0_{Fe^0}$	initial volume fraction of zero-valent iron
SI	saturation index
Φ_k	volume fraction of mineral
M_k	mineral molar volume ($m^3\ mol^{-1}$)
R_k	overall reaction rate for the mineral ($molm^{-3}_{bulk}s^{-1}$)
t	time (s)
n_o	initial porosity
n_t	porosity at time t
N_m	number of minerals
K_0	initial hydraulic conductivity (ms^{-1})
K	hydraulic conductivity at time t (ms^{-1})
S	storage co-efficient
T	transmissivity ($m^2\ s^{-1}$)
h	head (m)
b	aquifer thickness (m)
μ, C, D	constants
l	length of the column (m)
C	aqueous species concentration ($mg\ L^{-1}$)
D	hydrodynamic dispersion coefficient (m)
v	seepage velocity (ms^{-1})
R	retardation coefficient
λ	first-order decay constant
$\tilde{C}$	solid species concentration ($mg\ m^{-3}$)
$\tilde{r}$	kinetic reaction expression for the solid phase ($molm^{-3}_{bulk}s^{-1}$)

long-term efficiency of PRBs that are expected to function for decades or longer [5,15,16]. The performance of PRBs has been hindered by mineral fouling wherein the pore space is reduced by mineral precipitation in the reactive media. Fouling of the pore spaces reduces the porosity and hydraulic conductivity of the reactive medium [16–24], which then directly affects the reorientation of flow paths and changes reactive times. Porosity reductions due to secondary mineral precipitation usually are calculated by the volumes of secondary minerals observed in cores or from stoichiometric calculations using measured changes in aqueous concentrations [5]. Porosity reduction model developed by [25] for an in situ reactive barrier for the treatment of hexavalent chromium and trichloroethylene in groundwater has used the transition state theory (Eq. (1)) to model secondary mineral formation in treatment zone and surface area reduction method to model the depletion of reactive material of zero-valent iron (Eq. (2)). Simulations based on [25], the decrease in volume fraction was approximately 0.5–0.42 after 20 years in the reactive barrier.

$$r = -k_{eff}\left(1 - \frac{IAP}{K_{eq}}\right) \tag{1}$$

$$S_{Fe^0} = S^0_{Fe^0}\left(\frac{\varphi_{Fe^0}}{\varphi^0_{Fe^0}}\right)^{2/3} \tag{2}$$

Moreover, the porosity reductions of nine PRBs are reported by [26], indicating the porosity reductions range from 0.0007 to 0.03 per year. Li [5,26] also used Eq. (1) to develop the geochemical algorithm and Eq. (2) for reactive surface area reduction of zero-valent iron. The current study too adopted the transition state theory (Eq. (1)) to develop the geochemical algorithm for the secondary minerals precipitation and for the dissolution of Ca-bearing minerals in the reactive media. Eq. (2) was not used to model the depletion of recycled concrete aggregates' surface area, as the surface area is not 100% consists of reactive material (Ca-bearing minerals) [27].

Usually the porosity reductions due to secondary mineral precipitation are greater near the entrance face and diminish with distance into the PRB. Wilkin et al. [14] reported the zero-valent iron medium at US Coast Guard Support Centre had a decrease in porosity by 0.032 within the first 25 mm from the entrance face after eight years of operation. Liang [28] used saturation indices of each and every mineral in the system to predict which ones precipitate and which ones dissolve. Liang's [28] model for porosity reduction due to secondary mineral precipitation simulated over 10 years gave 0.188, 0.672, 1.150 and 0.918 reductions in PRBs located at Portsmouth, Moffett field, Monticello and Y-12 site respectively. Although the porosity reduction at entrance face was significant, about 80 mm into the iron medium, the porosity reduction diminished to less than 0.00002. Further studies done by [5], considering the effect of different parameters (geochemical parameters, influent concentration, rate coefficients and aquifer parameters) for porosity reduction in PRBs show that maximum porosity reduction was attained at the entrance face and diminishes with distance going into the PRB.

The intent of this study was to develop a model to understand mineral fouling in PRBs in ASS terrains, incorporating a calibrated flow and a reactive transport model to simulate mineral deposition

Fig. 1. (A) Pilot-scale PRB with piezometers and monitoring wells in ASS terrain on Shoalhaven Floodplain, southeast NSW and (B) installation of the PRB.

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