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Evaluating waste concrete for the treatment of acid sulphate soil groundwater from coastal floodplains

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ABSTRACT

The treatment of acidic groundwater generated from acid sulphate soil (ASS) terrain is a challenging environmental issue in coastal floodplains of Australia. In this study, a laboratory column experiment was conducted to assess the performance of waste concrete for treating the acidic groundwater leachate from ASS terrain of the Shoalhaven region of NSW. The groundwater was highly acidic (pH of 2.5–3.5) and contained elevated concentrations of iron (10–90 mg/L) and aluminium (30–45 mg/L). Passage of the acidic groundwater through the column filled with waste concrete resulted in a significant improvement in water quality. Reduction in the concentration of iron and aluminium to below detection limits and improvement of the pH from acidic to near-neutral (pH 6–8) were observed, along with a significant release of alkalinity over a six month period under controlled laboratory conditions. The results show that the working lifetime of waste concrete as the reactive media was governed primarily by the precipitation of secondary minerals despite the high acid neutralisation capacity of the waste concrete material.

Keywords: Acid sulphate soil; Contaminated groundwater; Column test; Groundwater remediation; Waste concrete

1. Introduction

Acid sulphate soils (ASS) have been identified since the early 1960s as a considerable environmental, economic and social problem that affects over three million hectares of coastal floodplains and estuaries in Australia [1,2]. Under reducing conditions, ASS remains chemically inert. When the water table decreases exposing sulphidic minerals such as pyrite to air, oxidation of these mineral occurs generating acidic drainage that is high in potentially harmful metals such as iron (Fe) and aluminium (Al) [3]. Large-scale artificial drainage in low-lying coastal areas of Australia has increased the distribution, magnitude and frequency of acid generation, which in turn has increased the rate of estuarine acidification by many orders of magnitude greater than that which might have occurred under natural drought/flood cycles [4]. Severe and periodic discharges of acidic water corrode steel and concrete infrastructure, clog waterways with Fe flocculates, kill aquatic ecology and produce large acid scalds that render land unsuitable for agricultural purposes [5].

Engineering solutions such as weirs and modified two-way floodgates have been installed near Broughton Ck, southeastern New South Wales, Australia to reduce the amount of acid generated through abiotic and biotic oxidation of pyrite by elevating the water table and to

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buffer acidity before it is released into the estuarine environment [6,7]. However, these mitigation measures are not feasible in very low-lying areas due to the risk of flooding during significant rainfall events. Biological oxidation of pyrite under submerged conditions can also still prevail if the organic content and sulphidic constituents of the soils are high. Thus, permeable reactive barriers (PRBs) have been identified as a potential remediation technique in these low-lying areas. A PRB is an engineered treatment zone consisting of a trench filled with reactive materials placed in the subsurface in order to remediate contaminated fluids as they flow through it. To date, PRBs have been proved as an efficient remedial method for groundwater contaminated with chlorinated volatile organic compounds [8], radionuclides [9], heavy metals [10] and acid mine drainage [11]. The use of PRBs to treat ASS acidic groundwater has been recently demonstrated in Australia [12]. A pilot subsurface alkaline PRB using waste concrete aggregate to neutralise acidic groundwater due to ASS was installed at a farm in Bomaderry, southeastern NSW in 2006. Although a slow decrease in performance was observed over three years of operation [13], the PRB maintained near neutral pH with complete removal of Fe and Al from the groundwater [14].

The objective of this study was to assess the performance of waste concrete obtained from a construction waste depot in neutralising acidity due to ASS groundwater and removing dissolved metal cations such as Fe and Al under varying flow conditions. In order to achieve this goal the performance of the waste concrete as reactive media was evaluated in a laboratory column experiment using real ASS groundwater garnered from the pilot PRB field site. The influence of chemical armouring and physical clogging of the waste concrete due to the precipitation of Fe and Al was studied.

2. Materials and method

2.1. Materials

The waste concrete used for the column experiment was collected from a construction waste depot, and was the same batch of concrete used in the pilot PRB installed in ASS terrain [13,14]. Large pieces of the waste concrete were crushed to a smaller size suitable for the column. The particle size distribution of heterogeneous crushed concrete ranged from 1.18 mm to 9 mm with an effective mean diameter (d_{50}) of 5.2 mm similar in character to medium-coarse gravel. Accurate identification of the composition of hydration products of the concrete was difficult due to the physical, chemical and mechanical changes in solidified materials in cementitious systems. In addition, accurate quantification of the minerals was a challenging task due to the heterogeneity of the concrete particles. Therefore, small samples of the concrete were ground in a mill ball to a fine powder for further mineralogical analysis. X-ray diffraction (XRD) was carried out to identify the major mineral phases present in the waste concrete before studying its acid neutralisation behaviour. The detailed elementary composition of the waste concrete was presented in Regmi et al. [13].

2.2. Groundwater sampling

Groundwater was collected on a monthly basis from sampling wells at an ASS affected farm in Bomaderry, southeastern NSW, Australia. Chemical composition of the acidic groundwater passed through the column at different time intervals is shown in Table 1. The groundwater was acidic (pH in range of 2.5–3.5) with high concentrations of Fe and Al.

2.3. Experimental procedure

A laboratory column test was conducted to assess the performance of waste concrete for the neutralisation of and the removal of metals from the acidic groundwater. A 5 cm internal diameter, 65 cm long acrylic column was packed with the waste concrete. The concrete was placed within a zone approximately 50 cm in length in the column bounded by sand layers approximately 10 cm and 5 cm in thickness at the base and the top of the column, respectively following the procedures in [13] (see Regmi et al. [13] for further details of the column configuration). The average total porosity was 0.52 (total void volume was 515 cm^3). The porosity was determined by dividing the total void volume by the volume of the column while the total void volume was determined by weighing the column dry and fully saturated.

A Masterflux peristaltic pump was used to inject the influent continuously through the column at a constant flow rate of 1.15 mL/min for 260 days at room temperature (23–25°C). The column experiment was conducted at a higher flow rate than the groundwater flow rate in the field to illustrate the acid neutralisation behaviour of the material in a short time period in the laboratory. Effluent samples were collected from five sampling ports at intervals of 10, 20, 30, 50 and 55 cm (outlet) along the length of the column. Measurements of pH, oxidation reduction potential (ORP), acidity and alkalinity were determined immediately after sampling. Samples were filtered through 0.45 μm cellulose acetate filter paper, and both acidified and non-acidified samples were collected and stored in a refrigerator at 4°C prior to analysis. Major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}, total Fe and Al^{3+}) were determined using inductively coupled plasma optical emission spectroscopy (ICP-OES) and atomic absorbance spectroscopy (AAS). Anions (SO_4^{2-} and Cl^-) were measured using ion chromatography. All chemical analyses were performed according to the standard method for water and wastewater examination [15]. Acid Neutralization Capacity (ANC) of the concrete was determined

Table 1
Composition of feed water quality used in column experiment

Input solution	Duration (d)	pH	EC (μS/cm)	Eh (mV)	Acidity mg/L as $CaCO_3$	Na^+	Ca^{2+}	K^+	Mg^{2+}	Total Fe (mg/L)	Al^{3+}	Cl^-	SO_4^{2-}
Feed 1	0–35	3.4	4100	520	360	364	155	39	86	6.9	44.2	606	1092
Feed 2	35–70	3.5	4100	520	315	358	155	38	84	7.2	44.0	606	1041
Feed 3	70–133	3.2	3950	525	315	352	136	39	83	7.0	45.0	559	993
Feed 4	133–165	3.01	4080	525	525	375	157	37	157	89.4	30.4	604	1291
Feed 5	165–176	3.13	4080	527	360	375	157	40	97	16.4	28.6	762	1318
Feed 6	176–196	2.57	4220	505	530	514	147	40	160	78.3	32.9	781	1318
Feed 7	196–230	2.97	4220	500	405	293	143	40	129	20.0	42.7	609	1134
Feed 8	230–253	2.76	4060	526	405	248	143	40	129	52.0	42.7	491	1134
Feed 9	253–260	2.76	4060	526	405	384	143	40	129	52.0	42.7	801	1134

Acidity was calculated corresponding to pH 7

following the Acid Sulphate Soils Laboratory Method Guidelines [16].

3. Results and discussion

3.1. X-ray diffraction/mineral characterisation

The results of the XRD analysis of the concrete are presented in Fig. 1. Significant peaks of quartz and Ca-bearing minerals were identified, indicating quartz, feldspar (anorthite, albite, etrringite etc.) and calcite as the predominant mineral phases in the waste concrete. The peak of Ca-bearing minerals are attributed to the cement whereas the peak of quartz is attributed to the sand and aggregates. Table 2 shows the XRD quantitative analysis, confirming that different phases of feldspar were high compared with portlandite. Although a significant amount of quartz was observed in the concrete, it was chemically inert in the acid neutralisation reaction.

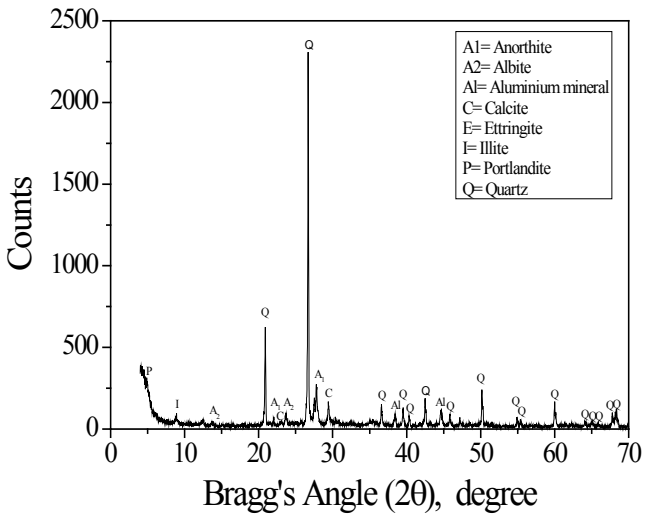

Fig. 1. X-ray diffraction (XRD) analysis of the crushed concrete powder.

Table 2
Mineralogical analysis (XRD) of the waste concrete used in the column experiment

Minerals	Formula	Percentage by weight (%)
Quartz	SiO_2	64.4
Portlandite	$Ca(OH)_2$	0.3
Anorthite*	$CaAl_2Si_2O_8$	16.8
Albite (low)*	NaAlSi3O8	8.4
Illite 1	$(K,H_3O)(Al,Mg,Fe)_2(Si,Al)_4O_{10}[(OH)_2,(H_2O)]$	0.3
Ettringite*	$(CaO)_6(Al_2O_3)(SO_3)_3.32\ H_2O$	4.8
Calcite	$CaCO_3$	4.4
Muscovite	$(KF)_2(Al_2O_3)_3(SiO_2)_6(H_2O)$	5.0
Total		100

* Feldspar mineral

3.2. Water quality parameters

The high initial effluent pH (10.8–8.0), as shown in Fig. 2, was due to the dissolution of soluble minerals portlandite and ettringite from the concrete and the subsequent release of hydroxyl and carbonate alkalinity. Initial pH of 10.8 could not be maintained for a long time period due to the negligible amount of portlandite in the concrete (Table 2). Maintenance of the pH above 8 for a few days was attributed to ettringite, which undergoes rapid dissolution between pH 10.7 and 9.5. Álvarez-Ayuso and Nugteren [17] and Golab et al. [18] suggested that the theoretical equilibrium pH for portlandite and ettringite are 12.1 and 10.7, respectively. The effluent pH decreased rapidly from 10.8 to 8.0 within 50 days, after which two long plateaus were observed. A near-neutral plateau (pH 8.0 to 7.5) continued until day 135 followed by a slow decrease to 6.0 at day 190. The pH then dropped abruptly reaching the next plateau at pH 4.2–4.0.

The total alkalinity of the effluent is governed by the release of alkaline materials from the recycled concrete and the initial chemistry of the groundwater. At the beginning of the column experiment, the total alkalinity decreased from 70 to 40 mg/L within the first 40 days as the hydroxyl and carbonate alkalinity generated by trace amounts of alkaline portlandite and ettringite was depleted (Fig. 3). Correspondingly, the effluent pH decreased rapidly until the generation of bicarbonate alkalinity (a strong buffer) in the column by anorthite and calcite, whereby a long plateau (pH 7.9–7.5) was observed until day 135 (Fig. 2). In fact, from day 40 onward, as bicarbonate alkalinity was released, the total alkalinity increased and reached a peak value of 140 mg/L (as $CaCO_3$) at approximately 85 days, followed by a gradual decrease. Fast depletion of alkalinity after 180 days indicates that a decrease in efficiency of the reactive material occurred from this point. Complete depletion of alkalinity at day 190 was accompanied by a sharp decline in pH to about pH 4 and an increase in ORP (Figs. 2–4) indicating that the column system was not buffered. The pH remained stable until equilibrium with the most soluble Al hydroxide mineral was attained. In good agreement, Jurjovec et al. [19] and Regmi et al. [20] observed a similar stable pH behaviour during the generation of carbonate/bicarbonate alkalinity followed by a rapid drop in pH after the total depletion of these carbonate minerals in acid mine drainage and ASS, respectively. In addition, Jurjovec et al. [19] reported the plateau of pH due to equilibrium of the effluent water with respect to gibbsite ($Al(OH)_3$) at pH 4.0 in the remediation of acid mine drainage.

The initial ORP of the effluent was low (100 mV) and remained between 200–250 mV until a near neutral pH was maintained (Fig. 4); indicating poor oxidation conditions. Drops in pH corresponded to rapid increases in ORP in the bottom part of the column (Fig. 2 and Fig. 4) at 27, 37, 61, 141 and 190 days for sampling ports at distances

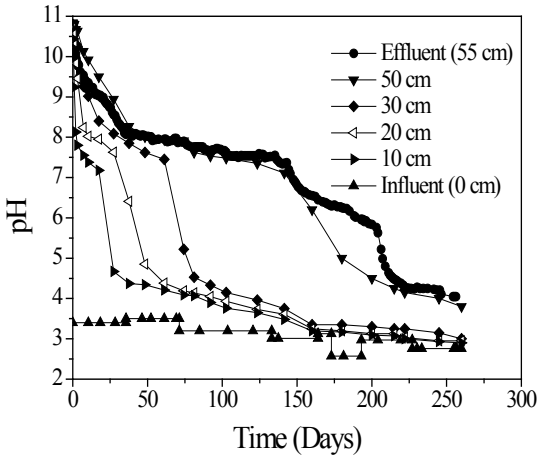

Fig. 2. pH as a function of time at different sampling ports from the interface of the bottom sand and concrete layer.

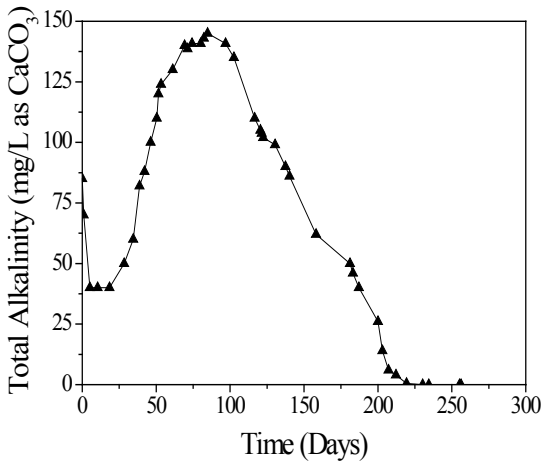

Fig. 3. Effluent total alkalinity (mg/L as $CaCO_3$) as a function of time.

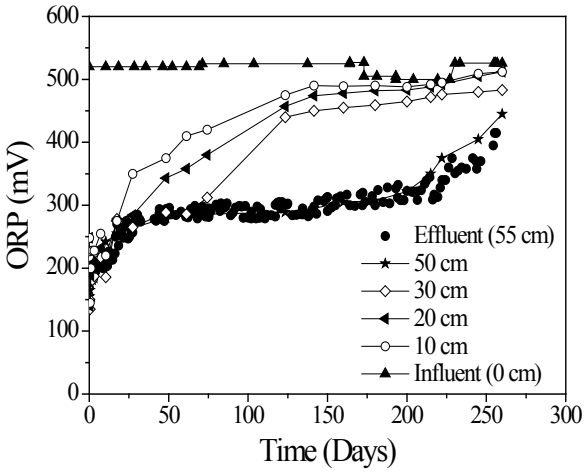

Fig. 4. Oxidation reduction potential (ORP) as a function of time at different sampling ports from the interface of the bottom sand and concrete layer.

of 10, 20, 30, 50 and 55 cm, respectively. This indicates fast depletion of alkalinity at the advancing acid front, which was supported by the growth of visible white and orange precipitates upwards from the bottom of the column. However, the alkalinity generated in the upper part of the column (Fig. 3) maintained the effluent pH plateau from the outlet port at near neutral with continuous release of Ca (Fig. 5) for a longer period. These results illustrate that the waste concrete neutralised the acidic water for a long period under a high flow rate despite the considerable amount of acidic groundwater passed through the column, thus proving its high potential for neutralising contaminated groundwater from ASS under variable environmental conditions.

3.3. Groundwater chemistry

Acidic groundwater was regularly collected from the PRB site to run through the column and as a result ion concentrations in the influent were heterogeneous over time and varied depending on the field conditions and sampling period. Peaks in Fe and Al concentrations (Figs. 6–7) resulted from pyrite oxidation during drought periods and subsequent mobilization from soil during rain events [7]. Observed sudden changes in effluent ions concentrations in short interval of time correspond to changes in the influent ions concentrations caused by the stored groundwater collected at different times.

Despite variations in influent Fe and Al concentrations, the high pH maintained in the column until the presence of bicarbonate alkalinity favoured Fe and Al precipitation (Fig. 6 and Fig. 7). The Fe and Al precipitates that formed on the waste concrete were amorphous and produced broad diffraction peaks in XRD analysis which were difficult to identify by XRD diffractogram curves alone. However, quantitative XRD analysis confirmed the precipitates as Al hydroxides (gibbsite and boehmite) and Fe oxyhydroxides (goethite), respectively, and quantified the ratio of $Al(OH)_3$ to FEOOH as 40–60. The abrupt increase in the Al concentration after day 190 coincided with the depletion of bicarbonate alkalinity and an abrupt decrease in pH from pH 6 to pH 4. A negligible amount of total Fe was observed in the effluent compared with Al until the end of the experiment. This is because Fe continued to precipitate until pH 3.5, whereas Al precipitated until the pH was maintained above pH 4. The column experiment was ceased at 260 days when the effluent reached pH 4. Therefore, it is noted that the further trend of effluent Fe concentration under an acidic condition of below pH 4 was not investigated in this study.

The concentration of the remaining major ions (Mg^{2+}, Na^+, K^+, Cl^- and SO_4^{2-}) in the effluent and influent was relatively constant throughout the entire experiment (Fig. 8) indicating that they were not involved in the acid neutralisation process. However, slightly higher concentration of these ions in the effluent compared to the

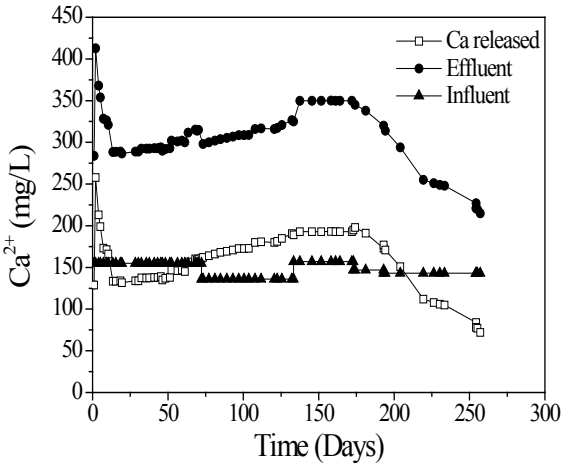

Fig. 5. Aqueous Ca concentration in the effluent and influent and the Ca released from the waste concrete as a function of time.

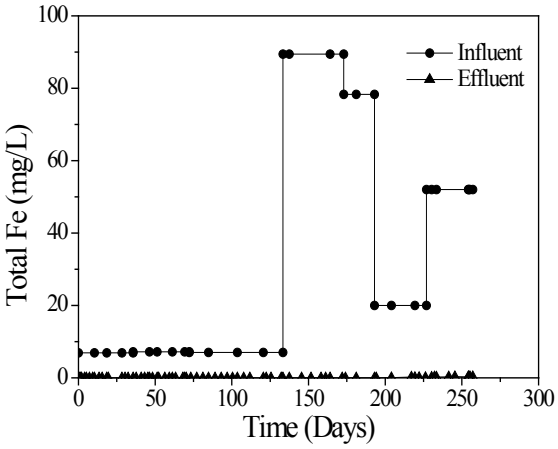

Fig. 6. Aqueous total Fe concentration in the effluent and influent as a function of time.

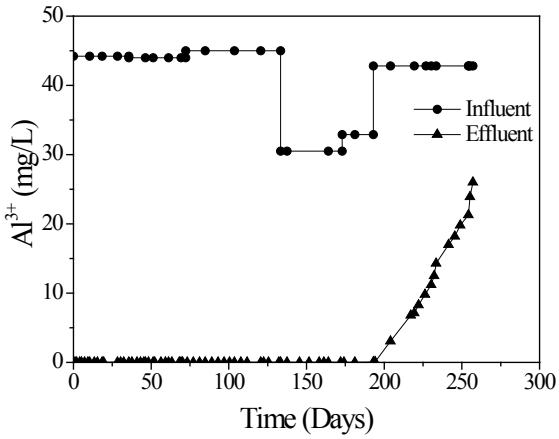

Fig. 7. Aqueous Al concentration in the effluent and influent as a function of time.

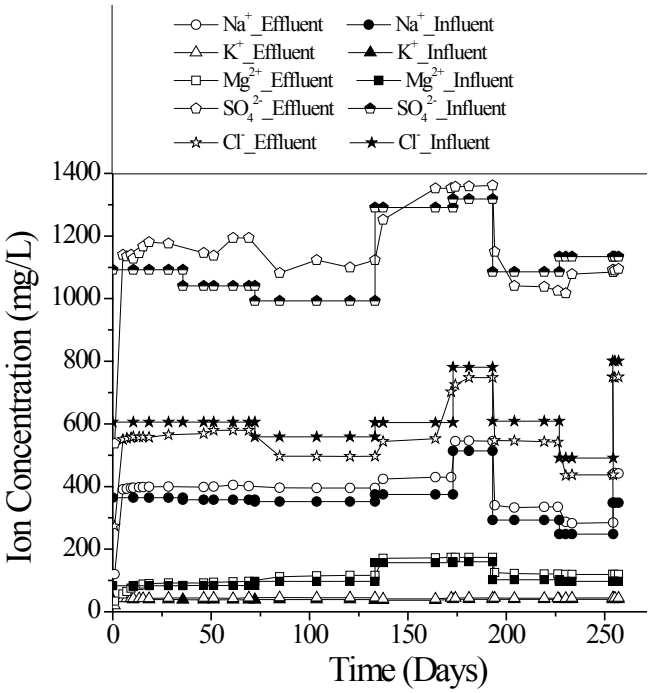

Fig. 8. Concentration of the remaining ions in the effluent and influent as a function of time.

influent was attributed to leaching of the minerals from the concrete. Similar to our study, Indraratna et al. [14] reported that these elements are almost inert in the acid neutralisation of the PRB with an evidence of no significant changes in the remaining ions up-gradient, inside and down-gradient of the pilot PRB in ASS. Although Ca was released from the concrete and there was a high concentration of SO_4^{2-}, gypsum precipitation, as observed by other researchers in acid mine drainage treatment (see for example Komnitsas et al. [21]), was not found in this study. This is because SO_4^{2-} concentrations were less than 2000 mg/L, which is insufficient for gypsum precipitation. Regmi et al. [13] reported that waste concrete contained a negligible amount of heavy metals. Indraratna et al. [20] also demonstrated that heavy metals leached from PRB filled with recycled concrete for treating acidic groundwater in ASS was negligible. Therefore, release of heavy metals from the waste concrete was not considered in this study.

3.4. Efficiency of reactive material

Armouring of the concrete and porosity reduction by accumulation of Fe and Al precipitates in the void spaces was observed throughout the length of the column during the experiment. Significant changes in piezometric head did not occur because of the use of larger concrete particles. This minimised the threat of chemical clogging, one of the expected failure mechanism in reducing the longevity of the PRBs technology, reported by many researchers [22,23]. However, armouring on the surface of the concrete by Fe and Al precipitates could result in a decrease in the rate of mineral dissolution and subsequent exhaustion of the ANC of the concrete. The 147th day was selected as a baseline for evaluating the efficiency of the concrete's ANC based on 240 L treated acidic groundwater over a pH of 7, complete removal of Fe and Al and considerable alkalinity within the column. The average acidity of the field groundwater was 365 mg/L as $CaCO_3$. The average ANC value of the concrete was 145 mg/g equivalent $CaCO_3$ [14]. The concrete used in the column (1206 g) had the capacity to neutralise 483.2 L of acidic groundwater to pH 7. This indicates a nearly 50% loss in the concrete's efficiency by armouring although it had the capacity to neutralise almost the same volume of acidic water treated.

Longevity of the reactive material for treating acidic groundwater from ASS therefore depends on two major factors: (a) fluctuation of the groundwater acidity due to changes in pyrite oxidation rate in shallow depth of soil and (b) the amount of Al and Fe leached to the groundwater. However, continuous neutralisation acidic water collected under variable field conditions and complete removal of the toxic Al and Fe under high flow rate in the laboratory controlled environment illustrates that the waste concrete is a suitable reactive material for treating contaminated groundwater from ASS.

4. Conclusions

This paper evaluates the efficiency of waste concrete for remediating contaminated groundwater generated from acid sulphate soils. A laboratory column test confirmed the suitability of the material in decontaminating acidic leachates loaded with high concentrations of hazardous metal ions such as Fe and Al. The maintenance of near neutral pH and effective removal of Fe and Al was realised when pozzolanic reactions took place between the Ca-bearing minerals of the concrete and acidic groundwater and generated alkalinity within the column. Continuous removal of these dissolved metals was caused by mineral precipitation on the reactive material surface at high pH and led to a longer-term decline in reactivity. Excessive armouring reduced the life of the material by nearly half. However, the possibility of temporal clogging, unlike other fine reactive media used in commercial PRBs, was reduced due to the use of larger concrete particle sizes.

Despite a reduction of material efficiency due to armouring, performance of the waste concrete was high and sustainable for an extended period under controlled laboratory conditions. In conclusion, a long-term column experiment for treating ASS affected groundwater has shown the successful application of waste concrete for the remediation of groundwater in ASS terrain under varying conditions. In addition, it is recommended that the

material used in the PRB should not be left in the ground when the efficiency of the PRB system drops because resolubilisation of previously formed unstable precipitates under acidic conditions will worsen the groundwater quality significantly.

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