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Potential for use of industrial waste materials as filter media for removal of Al, Mo, As, V and Ga from alkaline drainage in constructed wetlands – Adsorption studies

T. Hua [a,1], R.J. Haynes [a,*], Y.-F. Zhou [a,1], A. Boullemant [b], I. Chandrawana [c]

[a] School of Agriculture and Food Sciences/CRC CARE, The University of Queensland, St Lucia, QLD 4072, Australia
[b] Legacy Management Europe, Rio Tinto, 725 rue Aristide Bergès, Voreppe BP 7, 38341 Cèdex, France
[c] Rio Tinto Alcan, Queensland Research and Development Centre, 1 Technology Court, Pullenvale, Brisbane, QLD 4069, Australia

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ABSTRACT

The potential to remove Al, Mo, V, As and Ga from alkaline (pH 8.0–8.6) drainage originating from seawater neutralized bauxite processing residue storage areas using constructed wetland technology was studied in a laboratory study. Bauxite processing residue sand, bauxite, alum water treatment sludge and blast furnace slag were investigated as potential active filter materials. Al was shown to precipitate as $Al(OH)_3$ in the pH range 7.0–8.0 in aqueous solution and 6.0–8.5 in the presence of silica sand particles that provided a surface for nucleation. For V As Mo and Ga, adsorption to the surfaces of the adsorbents decreased greatly at elevated pH values (>pH 6–9). Water treatment sludge and bauxite had a greater ability to adsorb V, As and Mo at high pH (As and V at pH 7–9 and Mo at pH 5–7) than processing sand and slag. Adsorption isotherm data for As and V onto all four adsorbent than processing sand and slag. Adsorption isotherm data for As and V onto all four adsorbent materials fitted equally well to the Langmuir and Freundlich equations but for Ga, and to a lesser extent Mo, the Freundlich equation gave higher R^2 values. For all four ions, the maximum adsorption capacity (Langmuir value q_{max}) was greatest for water treatment sludge. Bauxite adsorbed more Mo, Ga and V than residue sand or slag. The pseudo-second order equation gave a better fit to the experimental kinetic data than the pseudo-first order model suggesting that chemisorption rather than diffusion/exchange was the rate limiting step to adsorption. It was concluded that water treatment sludge and bauxite were the most effective adsorbents and that for effective removal of the target ions the pH of the drainage water needs to be decreased to 6.0–7.0.

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* *Corresponding author*. Fax: +61 733651177.
E-mail address: r.haynes1@uq.edu.au (R.J. Haynes).
[1] Fax: +61 733651177.
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1. Introduction

Bauxite ore is mined and refined using the Bayer process in which Al-containing minerals are dissolved in hot NaOH. For each tonne of alumina, 1–2 tonnes of insoluble solids (bauxite processing residue) are produced and these are usually deposited in land-based lagoons or impoundments surrounding the refinery. Bauxite residues have a high pH (11–13) and possess high salinity and sodicity (Jones and Haynes, 2011). As a result, drainage from storage areas can pose an environmental risk and must be managed and drainage may need to be treated for many decades after closure. For this reason, an increasing number of refineries are now partially neutralizing their residues before they are deposited in storage areas and seawater neutralization is used by several refineries situated close to the marine environment. By addition of Ca and Mg in seawater, alkaline buffering anions in the residue are neutralized by precipitation of poorly soluble hydroxides and hydroxycarbonates (Kirwin et al., 2013). As a result, pH is reduced to below 9.0. Even so, drainage from seawater neutralized storage areas has been shown to be high in Al, Mo, As, V and Ga (Department of Environment and Heritage Protection, 2012). Treatment of drainage waters by conventional methods is likely to be expensive, especially if it is to be continued for many decades after closure. A low-cost, passive option, with green connotations, such as a constructed wetland, is an attractive alternative.

Inorganic contaminants are sequestered within the wetland filter matrix by adsorption/surface precipitation and bulk precipitation reactions (Westholm, 2006; Gustafsson et al., 2008; Vymazal and Kropfelova, 2008; Haynes, 2015). Commonly used filter materials in constructed wetlands are sands and gravels but these do not normally have a high adsorption capacity so that the capacity of a wetland to remove inorganic contaminants can decrease greatly over time and/or be highly variable (Haynes, 2015). Nevertheless, the removal efficiency of wetlands for inorganic ions can be improved with the use of active filter materials with reactive Fe/Al hydrous oxide adsorption surfaces (Genc-Fuhrman et al., 2007; Gustafsson et al., 2008). The most commonly used materials are blast furnace slag, steel slag and alum water treatment sludge (Vohla et al., 2011; Babatunde et al., 2009) but other materials such as iron ore, bauxite and bauxite processing residue have also showed promise (Haynes, 2015).

There is very little research on the use of constructed wetlands to treat alkaline wastewater and so far work has been concerned with wastewater streams high in Ca^{2+} where insoluble Ca compounds can precipitate in the wetland (Mayes et al., 2006). However, in alkaline drainage from bauxite residues the main cation present is Na^{+}. This presents practical problems since most Na compounds are highly soluble so that adsorption reactions onto the surfaces of wetland filter media are potentially the major removal mechanism within a wetland. The drainage can have a pH of 8.0–8.6 and contain concentrations of Al of <20 mg L^{-1}, those of Mo, Ga and V < 1.0 mg L^{-1} and As < 0.1 mg L^{-1}. The purpose of this study is to use batch adsorption studies to investigate materials with potential use as filter materials in constructed wetlands for treatment of the drainage. Potential filter media, readily available in the study area (bauxite residue sand, bauxite and alum water treatment sludge), were used as adsorbent materials along with blast furnace slag for comparison (the most commonly used waste adsorbent filter material) and the effects of pH, initial adsorbate ion concentration and contact time on Mo, Ga, V and As adsorption were investigated.

2. Materials and methods

2.1. Sample description

Water treatment sludge was obtained from the Seqwater Mount Crosby Water Treatment Plant (Brisbane), water-quenched BF slag was obtained from the BlueScope Steel Ltd., Port Kembla Steelworks (New South Wales, Australia) and bauxite (from the Weipa deposit, Queensland) and bauxite residue sand were sourced from Rio Tinto Alcan (Yarwun Alumina Refinery). The residue sand was treated with acid (HCl) to give a pH of approximately 9.0 and then washed with distilled water. All samples were dried and ground (<200 μm) prior to use.

Elemental composition of materials was measured by inductively couple plasma atomic emission spectrometry (ICP-AES) after digestion in a microwave system with HNO_3, HCl and HF (CEM, 1993). Specific surface area of samples was determined by the BET/N_2 adsorption method, and zero point of charge (PZC) was determined using a Laser Zeta metre. Materials were subjected to X-ray diffraction analysis to determine their mineralogy. Effective cation exchange capacity, pH and electrical conductivity (EC) (1:5 material/water ratio) were determined as described by Rayment and Higginson (1992). Cu, Zn, Cr, Cd, Pb and As were extracted from wastes by the Toxicity Characteristic Leaching Procedure (TCLP) (USEPA Method 1311; USEPA, 1992), and metals were measured by ICP-AES.

2.2. Aluminium precipitation

To determine the pH range over which Al would precipitate, an Al solution of 10 mg L^{-1} was prepared in 0.01 M NaCl using $AlCl_3$. A concentration of 10 mg Al L^{-1} was used as this is within the range normally found in the drainage water. Aliquots (20 mL) were transferred to 50 mL centrifuge tubes and the pH adjusted from 3 to 12 using HNO_3 and NaOH. Once the pH had stabilized the solutions were shaken for 2 h. During that time, pH was checked and readjusted if necessary every half hour. Resulting solutions were centrifuged (400 rpm for 10 min) and the filtered supernatant was analysed for Al by inductively coupled plasma atomic emission spectrophotometry (ICP-AES). To determine if Al precipitation is affected by the presence of a potential filter material, the above procedure was repeated but centrifuge tubes also contained 200 mg of acid-washed silica sand. Experiments were carried out in triplicate.

To determine the potential cumulative effect of precipitated Al over the surfaces of a filter material on adsorption of the other contaminant ions (Mo, V, As, Ga), solutions of 10 or

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