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Performance of bauxite refinery residues for treating acid mine drainage

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ABSTRACT

Novel application of alkaline waste materials from the alumina industry (Bayer precipitates) to both neutralize and remove dissolved metals from acid mine drainage (AMD) has been demonstrated. The hypothesis was that Bayer precipitates could not only economically treat a wide range of AMD compositions but also do this in an environmentally acceptable manner. Therefore, the aim was to synthesise a range of Bayer precipitates and test their remediation effectiveness with a variety of real AMD samples. Thermal activation of Bayer precipitate at 380 °C produced a material which required the least mass to neutralize AMD and removed greater amounts of problematic species such as manganese (*e.g.* 74 to 82.8% reduction for Bayer precipitate and 83.7 to 91.6% reduction for thermally activated Bayer precipitate). The presence of an amorphous mixed magnesium/aluminium oxide and lack of carbonate species in the thermally activated Bayer precipitate was proposed to explain the improved performance. The resultant sludge was composed of hydrotalcite and possibly calcium sulphate and calcium carbonate, with precise quantities dependent upon the AMD tested. Exposure of sludge to water promoted the dissolution of a fraction of the hydrotalcite which enhanced concentrations of Mg and Ca in solution; albeit, contact of the sludge with AMD resulted in further removal of dissolved metals and raised the pH of the AMD. In summary, this study has developed a new process using two industrial waste products to solve a critical environmental problem. Future research should investigate the commercial feasibility.

1. Introduction

Acid mine drainage (AMD) is an accelerated form of the naturally occurring acid rock drainage (ARD) at mine sites and is one of the most critical problems caused by mining [1,2]. Acid rock drainage is a natural process caused by oxidation of sulphides, which normally occurs slowly as the surface of the earth erodes. In the presence of oxygen, water or oxidising bacteria sulphide minerals oxidise and form sulphuric acid and can cause liberation of metals from affected soils and rocks [3,4]. The Department of Natural Resources and Mines (Queensland Government) states that there are over 15,000 abandoned mine sites across Queensland [5]. As mining activities increase, the amount of waste material produced concomitantly increases and in turn the formation of AMD [6,7]. Pollution caused by AMD can in some cases impact plant and animal life in streams [8,9].

Treatment of acid mine drainage can be classified in terms of active/physical or passive/biological systems, based on their requirements for chemical addition, infrastructure, maintenance and monitoring. In an active chemical treatment system, acid mine drainage is treated by the addition of alkaline materials such as lime, magnesia or sodium hydroxide [3]. This procedure encourages the precipitation of iron oxyhydroxides and heavy metals that can then be removed through co-precipitation [10,11]. Incomplete removal, high-energy requirements, high cost and production of toxic sludge are the main shortcomings associated with these methods [12,13]. To overcome the outlined disadvantages various approaches have been made for the development of economical and more effective technologies [14,15]. As the chemical treatment of AMD is an inherently expensive process, a number of studies have used industrial residues such as by-products from coal industry [16–18], alumina industry [19–21] and calcined eggshells [22] to treat acid mine drainage. The high density sludge (HDS) process for treating AMD has also received attention recently [23], which recycles lime sludge produced by the treated effluent in the initial lime mixing tanks. A key benefits of using the HDS process includes the formation of a dense and stable sludge with minimal release of metals back into the environment [24].

A substantial source of highly alkaline waste material can be found in the alumina refining industry. Notably, alumina refiners are often in the same geographical location as mines which produce acid mine drainage (such as Queensland). The waste residue produced from the extraction of alumina from bauxite ore is comprised of sodium aluminate ($NaAl(OH)_4$), sodium hydroxide ($NaOH$) and sodium carbonate

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Table 1
Water compositions found at Mt Morgan mine site.

		Concentration (mg/L)												
Site	**Containment type**	**pH**	**SO_4**	**Al**	**Fe**	**Mn**	**Cu**	**Zn**	**Co**	**Ni**	**Mg**	**Ca**	**Na**	**Si**
Airfields	Tailings	2.70	38,000	1703	194	186	101	73	5.84	1.23	3545	431	157	52.3
Frog Hollow	Slag / Waste rock	3.21	16500	1094	1045	150	87	49	3.65	0.93	1929	490	274	65
Mundic Creek East	Waste rock	2.77	19500	1532	500	134	81	45	3.59	0.88	1976	445	221	45
Mundic Creek West	Tailings / Waste rock	2.85	19780	1516	241	186	83	55	4.42	1.49	2597	495	536	62
No. 2 Mill	Tailings	3.00	22500	1364	1370	161	60	39	2.88	0.9	2723	523	202	46
No. 2 South	Tailings	2.86	22500	1500	169	156	61	29	3.66	0.67	2874	455	251	39
Open Pit	Waste rock	3.74	17,430	1233	17	162	77	49	4.06	1.54	2265	535	648	46
Shepherd's Holding	Waste rock	3.14	15000	474	153	81	16	15	1.09	0.24	2496	494	219	45
Shepherd's Spring	Waste rock	3.11	18000	1105	63	135	48	24	2.85	0.56	2526	445	260	38

(Na_2CO_3) [25]. Typically, this wastewater stream has a pH greater than 13, and as such is normally required to be neutralised prior to disposal. Several methods of bauxite refinery residue neutralisation have been reported including: infiltration of rainwater and atmospheric CO_2, treatment with strong acids, gypsum addition, and seawater neutralisation [26–28]. The neutralisation of bauxite refinery residues with seawater forms a Mg/Al layered double hydroxide structure [Eq.1] as reported by Palmer *et al.* [29].

$$6MgCl_{2(aq)} + 2NaAl(OH)_{4(aq)} + 8NaOH_{(aq)} + Na_2CO_{3(aq)} \rightarrow Mg_6Al_2(OH)_{16}(CO)_3.\,xH_2O_{(s)} + 12NaCl_{(s)} \quad (1)$$

Synthetic layered double hydroxides have also been studied extensively for their application in environmental wastewater decontamination [30,31]. Layered double hydroxides (LDHs) consist of positively charged brucite-like octahedral hydroxide layers which are neutralised by the interlayer anion and water molecules ($[M_{1-x}{}^{2+} M_x{}^{3+}(OH)_2]^{x+} A_{x/m}{}^{m-}.nH_2O$, where M^{2+} is a divalent cation, M^{3+} is a trivalent cation and A an inter-lamellar anion) [32,33]. LDHs have been studied for the removal of metals (Cu^{2+}, Zn^{2+}, Pb^{2+}, Cd^{2+}, Ni^{2+}, Co^{2+}, Sc^{3+}, La^{3+}) from aqueous environments by hydroxide formation, adsorption or *via* intercalation in organic modified layered double hydroxide [34–36]. Layered double hydroxides show some interesting properties when calcined: calcination of layered double hydroxides generates mixed metal oxides due to the removal of interlayer water, interlayer anions and hydroxyl groups; thus increasing its reactivity due to higher surface basicity as compared to normal LDHs [37,38]. However, when exposed to water and anions, calcined LDH is able to regenerate the layered structure [39,40]. This latter property of regenerating layered structure has also been used for the removal of different anions and cations [41]. Consequently, the application of waste alkaline residues from the alumina industry to remediate acid mine drainage appears prospective.

Recently, the authors of this study completed a feasibility study involving addition of seawater-neutralised bauxite refinery residues, including thermally activated Bayer precipitate, to an acid mine drainage sample [42]. This study confirmed that Bayer precipitates were indeed effective in neutralising AMD and capable of removing heavy metals, except manganese, to meet acceptable discharge limits. In addition, the performance of Bayer precipitates was shown to be comparable to lime which is the current industry standard material for AMD neutralization. However, previous research regarding the treatment of AMD with Bayer precipitates involved only one AMD water composition. As the composition of AMD varies not only from region to region but also within the same mine site there was a definite need to understand in greater depth the effectiveness of using Bayer waste residue to neutralize a wider range of AMD solutions. In addition, as the neutralisation process itself forms a sludge it was pertinent to examine the stability of this material and whether it was useful for further water treatment application.

Therefore, this study evaluated nine AMD water compositions to establish what impact variations in water quality had upon treatment methods using Bayer precipitates and thermally activated Bayer precipitates. It was hypothesised that Bayer precipitates may be capable of remediating different types of AMD, thus expanding the applicability of this novel approach. Specific research questions addressed included: (1) how does the performance of Bayer precipitates change with variable AMD water composition? (2) what impact does mine water composition have on the quality of treated water and sludge produced? and (3) what impact does mine water composition have on sludge stability? Consequently, AMD samples were collected and analysed from various locations at the Mt Morgan site in Queensland and bench scale tests were conducted to determine the influence of AMD composition upon Bayer precipitate treatment.

2. Materials and methods

2.1. AMD samples and chemicals

2.1.1. Study area and sampling

Mount Morgan mine is a historic site located 32 km south-west of Rockhampton (23°38′S and 150°22′E) and adjacent to the Dee River in central Queensland [43]. From 1882 to 1990 this mine produced a total of 247 t of gold, 37 t of silver, 38,700 t of copper and 100 Mt of sulphide-rich rocks consisting of minerals like pyrite, pyrrhotite, sphalerite, and chalcopyrite. Oxidation of the sulphide bearing minerals in tailings and waste rock deposits has resulted in the production of AMD waters (pH 1.9–3.8) rich in Al, Mn, Cu, and Zn. Acid mine drainage samples were collected from 9 waterbodies across the Mt Morgan mine site in 2014–2015 (Table 1), with “Open Pit” being the largest waterbody with an estimated volume of 9×10^9 L (based on depth profile measurements) [43]. A map with the locations of AMD sites is shown in Fig. SI1 (Supplementary Information). The climate in the area is seasonal with temperatures ranging from 32°C in January to 23°C in July [44]. The mean annual rainfall is 740 mm and the mean annual evaporation is estimated to be about 1840 mm [44]. During overflow events, the adjacent Dee River is the main recipient of acid mine drainage water from this site. A “pump back” system has been introduced across the mine site to pump water collected at sumps to the open pit waterbody. A lime neutralisation plant was commissioned in 2006 to maintain the volume in the open pit to avoid overflow events [43].

2.2. Synthesis of bayer precipitates

2.2.1. Synthesis of Bayer liquor

To simulate 10 g/L Al_2O_3 supernatant liquor at an Australian alumina refinery, saturated evaporative liquor (96 g/L Al_2O_3) was diluted and then had sodium carbonate (Na_2CO_3) and sodium hydroxide (NaOH) added. The 10 g/L Bayer liquor was prepared by half filling a 2 L volumetric flask with deionised (DI) water, to which 21.13 g of Na_2CO_3 was dissolved before the addition of 29.99 g of NaOH. Once the Na_2CO_3 and NaOH were dissolved, 212 mL of saturated evaporative