



Enhancement of bauxite residue as a low-cost adsorbent for phosphorus in aqueous solution, using seawater and gypsum treatments

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ABSTRACT

Bauxite residue (red mud), the by-product produced in the alumina industry, is being produced at an estimated global rate of approximately 150 Mt per annum. Due to its highly alkaline nature, many refineries use neutralisation techniques such as mud farming (atmospheric carbonation), direct carbonation using carbon dioxide or reactions with seawater, to treat the bauxite residue and reduce its alkalinity prior to disposal in the BRDA (bauxite residue disposal area). Applying a treatment can render the bauxite residue non-hazardous and may also prepare the bauxite residue for reuse, particularly as an adsorbent. In this study, gypsum and seawater treatments were applied to the various bauxite residue samples obtained and the effects on its mineral, elemental and physiochemical properties were examined, as well as the effect on its phosphorus (P) adsorption capacity. It was found that in addition to reducing the alkalinity of all bauxite residue samples used, the P adsorption capacity was also enhanced following amendment with seawater or gypsum, particularly with gypsum. A positive correlation was detected between P adsorption and both Ca and CaO. A negative correlation was detected between the P adsorption and pH of the media. Fitting the data obtained from a batch adsorption experiment to the Langmuir adsorption isotherm, the maximum adsorption capacity was estimated to range from 0.345 to 2.73 mg P per g bauxite residue, highlighting the re-use potential for bauxite residue as an adsorbent for P.

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1. Introduction

During the extraction of alumina from bauxite ore using the Bayer process, a by-product called bauxite residue (red mud) (Kirwan et al., 2013; Liu et al., 2014) is produced. The global inventory for bauxite residue is approximately 3 billion tonnes, with an estimated annual production rate of 150 million tonnes (Evans, 2016; Mayes et al., 2016). Bauxite residue is highly alkaline

(pH > 10) (Goloran et al., 2013), with a high salinity and sodicity (Gräfe et al., 2009). Current best practice within this industry includes careful planning and management of highly engineered bauxite residue disposal areas (BRDAs), avoiding contamination of the surrounding environment (Prajapati et al., 2016). In addition, some refineries use neutralisation techniques for the bauxite residue before disposal into the BRDAs (Klauber et al., 2011; IAI, 2015; Evans, 2016). These techniques include (1) direct carbonation, whereby the residue slurry is treated with either carbon dioxide, sulfur dioxide gas, or undergoes intensive mud farming using amphirollers (atmospheric carbonation) (Cooling, 2007; Fois et al., 2007; Dilmore et al., 2009; Evans, 2016) (2) addition of spent acids

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and/or gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) (Kirwan et al., 2013), or (3) reaction of residues with seawater (Hanahan et al., 2004; Palmer and Frost, 2009; Couperthwaite et al., 2014).

Bauxite residues typically comprise very fine particles, ranging from 0.01 μm to 200 μm (Pradhan et al., 1996). Depending on the type of bauxite ore used, in some refineries the bauxite residue undergoes a separation technique during processing (Evans, 2016), which allows it to be separated into two main fractions: a fine fraction with a particle size <100 μm and a coarse fraction with a particle size >150 μm (Eastham et al., 2006; Jones et al., 2012). The coarse fraction mainly consists of quartz (SiO_2), whereas the fine fraction is dominated by iron (Fe) oxides (Snars and Gilkes, 2009). The ratio of the fine to coarse fraction produced is dependent on the bauxite ore used and the Bayer process employed (Li, 2001). Refineries which carry out the separation technique, have found use for the coarse fraction to create roadways to the BRDA and/or storage embankments (Evans, 2016). However, finding appropriate options for the re-use of the fine fraction bauxite residue remains elusive (Power et al., 2011; IAI, 2015).

Fine fraction bauxite residue comprises Fe oxides (20–45%) and aluminium (Al) oxides (10–22%) (IAI, 2015), which make it suitable as a medium to adsorb phosphorus (P). The European Commission (EC) has identified waste management as an important aspect of the “circular economy” (European Commission, 2015), so in recent years, emphasis has been placed on investigating alternative methods of P recovery from wastewater (Grace et al., 2015, 2016). A move from the more conventional methods of P recovery such as biological removal and chemical precipitation (Wang et al., 2008), to the use of low-cost adsorbents from industrial solid wastes, such as bauxite residue, have been examined. In comparison to standard P removal by sand, bauxite residue has a high P retention capacity (Vohla et al., 2007). However, its P removal potential is enhanced following treatment by heat, acid or gypsum (Table 1). Of the methods employed, acid and heat treatment have proved most successful in increasing the P adsorption capacity of the bauxite residue, with maximum adsorption capacities of up to 203 mg P g^{-1} bauxite being achieved (Liu et al., 2007) compared to untreated residue (0.20 mg P g^{-1} ; Grace et al., 2015) (Table 1). However, whilst acid and heat treatments have proven to be very successful in increasing the adsorption capacity of bauxite residue, they are expensive, energy consuming (using high temperatures up to 700 °C) (Xue et al., 2016), and, without further treatment, do not allow for the easy reuse of the bauxite residue (e.g. as a possible media for plant growth) (Xue et al., 2016).

Treatments such as seawater or gypsum provide relatively

inexpensive, alternative treatments, which may not only enhance the P adsorption capacity of the bauxite residue media, but may also help to improve its physicochemical characteristics. Seawater treatment improves bauxite's physical structure, due to the addition of magnesium (Mg) and calcium (Ca) which behave as flocculating agents, allowing many of the fine particles in bauxite residue to form more stable aggregates (Jones and Haynes, 2011), and a partial decrease in sodium (Na) due to ion exchange with Mg, Ca and potassium (K) (Hanahan et al., 2004). Seawater-treated bauxite residues also allow adsorbed P to become bio-available, unlike the metal cations which are unavailable, highlighting the P and metal retention capabilities (Fergusson, 2009). Revegetation of bauxite residue using gypsum has also improved plant growth by reducing its alkalinity and salinity, and improving the structure of the residue (Courtney et al., 2009; Courtney and Kirwan, 2012). In addition to this, modern alumina refineries are often located close to deep water ports, to allow for the bulk shipment of incoming bauxite (sometimes from multiple sources) to the refinery and/or for bulk shipment of alumina to aluminium smelters situated elsewhere. Therefore, there is ample scope for the increasing use of seawater neutralization technology for pre-treatment of residues in refineries not already employing treatments previously mentioned, prior to their deposition in the BRDA.

To the best of the authors' knowledge, no study has previously compared the use of raw seawater or gypsum treatments on the separate fractions of bauxite residue as a method of neutralisation and preparation for the re-use of bauxite residue in its separated and unseparated fractions as low-cost adsorbents and a potential source of P. The objectives of this study were to (1) characterise bauxite residue from two different sources, before and after treatment with seawater and gypsum, and to investigate their potential to release trace elements (2) investigate the effect of the treated bauxite residue on P adsorption (3) assess the impact of particle size, mineral and elemental (particularly Ca and Mg) composition of the bauxite residue on the adsorption of P.

2. Materials and methods

2.1. Sample preparation

A 1 kg sample of fresh bauxite residue was obtained from Alteo Gardanne [Gardanne, France (4327'9"N, 527'41" E)], who operate a co-disposal method for fine and coarse fractions of bauxite. This sample will be referred to hereafter as UFR. One kilogram of mud-farmed bauxite residue samples (treated by atmospheric

Table 1
Phosphorus (P) adsorption studies that have been carried out using bauxite residues, untreated and treated residues, and their recovery efficiencies.

	P recovery technique	Factors investigated	Type of water	Initial P concentration of the water	P recovered	Reference
Untreated bauxite residue	Batch adsorption experiment	Kinetics, pH and temperature	Synthetic water	5–100 mg P L^{-1}	0.20 mg P g^{-1}	Grace et al., 2015
Gypsum Treated	Batch adsorption experiment	Contact time (3, 6, 24, 48hr)	Synthetic water	20–400 mg P L^{-1}	7.03 mg P g^{-1}	Lopez et al., 1998
Brine treated bauxite residue (Bauxsol™ ^a)	Batch adsorption experiment	pH, ionic strength, time	Synthetic water	0.5–2 mg P L^{-1}	6.5–14.9 mg P g^{-1}	Akhurst et al., 2006
Acid and brine treated bauxite residue (Bauxsol™ ^a)	Batch adsorption experiment	Kinetics and isotherms	Synthetic water	200 mg P L^{-1}	55.72 mg P g^{-1}	Ye et al., 2014
Heat treated bauxite residue	Batch adsorption experiment	Time, pH and initial concentration	Synthetic water	155 mg P L^{-1}	155.2 mg P g^{-1}	Liu et al., 2007
Acid and heat treated bauxite residue	Batch adsorption experiment	Time, pH and initial concentration	Synthetic water	155 mg P L^{-1}	202.9 mg P g^{-1}	Liu et al., 2007
Acid treated bauxite residue	Batch adsorption experiment	Acid type, pH	Synthetic water	1 mg P L^{-1}	1.1 mg P g^{-1}	Huang et al., 2008

^a Bauxsol™ = neutralised bauxite residue produced using the Basecon™ procedure, which uses brines high in Ca^{2+} and Mg^{2+} (McConchie et al., 2001).

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