



Neutralisation of bauxite residue by carbon dioxide prior to acidic leaching for metal recovery



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ABSTRACT

The present work considers the neutralisation of bauxite residue (red mud) with CO₂ as a potential technology for reducing the acid consumption in the acidic leaching step for metal recovery. The pH of bauxite residue was reduced during neutralisation by the transformation of hydroxide ions to (hydrogen) carbonate ions. Neutralisation at high CO₂ partial pressures and high temperatures reduces the alkalinity of the bauxite residue, but it leads to the stabilisation of silicate compounds such as cancrinite and grossular. After acidic leaching of the neutralised product with sulfuric acid, a decrease by 20% in the dissolution yield of Al, Fe, and Ti was observed, due to an insufficient amount of acid devoted to leaching as the transformation of calcite into bassanite and the high concentration of silicate compounds consumed part of the acid. Sc recovery by leaching of the highly neutralised bauxite residue was about 35 wt%, which depends on Fe and Ti recovery. A positive correlation between La/Nd with Sc indicates that the recovery of La/Nd indirectly depends on the extraction of Fe/Ti, as Sc is chemically associated to these major metals.

1. Introduction

Bauxite residue (red mud) is the waste product generated during alumina production by the Bayer process, and it is composed essentially of non-soluble elements such as iron and titanium minerals, un-digested alumina minerals, sodium aluminium hydrosilicates, calcium minerals and significant amounts of minor and trace elements such as scandium, yttrium and lanthanides, i.e. the so-called rare earth elements (REEs) (Evans, 2016). Bauxite residue is an interesting source of REEs and especially scandium, but also of iron and titanium, while the residue after metal recovery can be used for low-carbon building materials (Binnemans et al., 2015). Scandium concentrations in Greek bauxite residue are about 120 mg/kg (Borra et al., 2015b), but it may reach up to 260 mg/kg in some bauxite residues (Borra et al., 2016) depending on the origin and mix of bauxite minerals (raw material). The scandium concentration in bauxite residue is much higher than its average abundance in the Earth's crust (22 mg/kg), and almost twice that in the original bauxite ores (Ochsenkühn-Petropulu et al., 1996). Although, scandium can be found in concentration over 100 mg/kg in natural minerals such as thortveitite ((Sc, Y)₂Si₂O₇) and kolbeckite (ScPO₄·2H₂O), the abundance of these minerals is very scarce (Binnemans et al., 2015; Wang et al., 2011). Therefore, bauxite residue

represents an interesting source for the recovery of this metal. In fact, scandium represents more than 90% of the value of the REEs present in bauxite residue.

Several methods for recovering valuable elements from bauxite residue have been reported (Borra et al., 2016). They are mainly based on hydrometallurgical and pyrometallurgical processes, or combinations of both. Pyrometallurgical processes allow a high recovery of scandium and other REEs (Borra et al., 2015a; Tanutrov et al., 2013), but the energy consumption is high. Minor elements can be recovered by acidic leaching with HCl, HNO₃, or H₂SO₄ (Abhilash et al., 2014; Borra et al., 2015a; Ochsenkühn-Petropulu et al., 2002, 1996), but co-dissolution of a significant amount of iron is a serious drawback for REE recovery (Borra et al., 2015b).

The alkalinity of bauxite residue is an important issue for metal recovery by acidic leaching, because part of the acid must be used for the neutralisation of the alkaline products left behind after the Bayer process. This leads to a large acid consumption and makes the recovery of metals from bauxite residue often not economical. In order to reduce the amount of acid needed for neutralisation of bauxite residue and, consequently, to develop a more sustainable process for metal recovery, alternative neutralisation routes must be considered. The use of carbonic acid (H₂CO₃), formed during the dissolution of CO₂ in water,

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represents an inexpensive and safe technology for bauxite residue neutralisation. The use of this reagent has shown promising results in terms of bauxite residue stabilisation for further safe disposal (Bonenfant et al., 2008; Hanahan et al., 2004; Lutpi and Zhu, 2010; Rai et al., 2013; Yadav et al., 2010). Alkaline leaching of bauxite residue with addition of CO₂ for scandium recovery has also been reported for scandium recovery (Petrankova et al., 2015). The process allows the recovery of scandium due to the high solubility of Sc(OH)₃ in a solution of NaHCO₃ (Pasechnik et al., 2004). However, the process is limited for the recovery of other metals such as iron, aluminium, titanium and the lanthanides. The use of CO₂ as a neutralisation reagent for bauxite residue as part of a flow sheet to reduce acid consumption during the metal recovery by acidic leaching has not been studied yet.

The objective of this paper is to evaluate the recovery of valuable metals after the reduction of bauxite residue's alkalinity by transformation of hydroxides into soluble carbonate compounds during the neutralisation by CO₂, at different conditions. Neutralisation with a mineral acid at similar conditions is compared to the neutralisation with CO₂. The neutralised solid fraction is leached with sulfuric acid at different concentrations to dissolve major and minor elements, and the metal extraction efficiency is correlated with the neutralisation conditions. The acid consumption is also evaluated.

2. Material and methods

The bauxite residue studied in this paper was kindly provided by Aluminium of Greece (Agios Nikolaos, Greece). It originates from a mixture of karst and lateritic bauxites. It was received from the alumina refinery after dewatering by filter presses and room temperature drying. Upon arrival in the lab, the sample was further dried at 105 °C for 24 h (moisture content 20–30 wt%). Chemical analysis of the major elements in bauxite residue was performed using wavelength dispersive X-ray fluorescence spectroscopy (WDXRF, Panalytical PW2400). Chemical analysis of minor elements was performed after complete dissolution of the bauxite residue by alkali fusion and acid digestion in 3% HNO₃ solution, followed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS, Thermo Electron X Series) analysis. The alkali fusion was carried out by mixing 0.5 g of bauxite residue with 1.5 g of sodium carbonate and 1.5 g of sodium tetraborate decahydrate, followed by heating the mixture in a platinum crucible at 1100 °C for 30 min. The mineralogy of the samples was studied by X-ray powder diffraction (XRD, Bruker D2 Phaser). The obtained data were evaluated with EVA V.3.1 (Bruker AXS) and quantified with Topas-Academic V.5, using the Rietveld method. Thermo-gravimetric analysis (TGA, Netzsch STA 409) were carried out in a nitrogen atmosphere from room temperature to 1000 °C at a heating rate of 10 °C/min in order to identify phase transformation and thermal decompositions in the bauxite residue.

Neutralisation at ambient conditions of bauxite residue with CO₂ gas was performed in a three-neck flask reactor (400 mL) with continuous agitation (agitation at 478 rpm). The agitation was performed by a magnetic stirrer (4 cm long, 7.5 mm diameter, 6 g). Within the reactor, solid particles were mixed with water at a liquid-to-solid ratio, L/S, of 5:1. CO₂ gas (99.9925%, Air Liquide) was introduced continuously at defined flow rate, which was controlled with a flow meter. A diffuser (PTFE, pore size of 10 μm) was used to spread out the gas into the formed slurry.

High-pressure neutralisation experiments were carried out in a titanium autoclave (Parr Company, series 4560) varying the partial pressure of CO₂ and temperature. Bauxite residue and Milli-Q water were mixed during a period of 5 min in a beaker at a L/S of 5:1 at ambient conditions. The slurry was poured into the titanium vessel (200 mL) and mounted in the autoclave. Nitrogen gas at a pressure of 5 bar was used to remove the excess of oxygen gas inside the reactor, so that potential oxidative reactions could be prevented.

Neutralisation of bauxite residue with HCl (37%, Fisher Scientific)

and H₂SO₄ (95–97%, Sigma-Aldrich) was performed with a L/S ratio of 5:1 in sealed polyethylene bottles by constant agitation using a laboratory shaker (Gerhardt Laboshake) at 200 rpm and 25 °C. The slurries after neutralisation were filtered using filter paper (pore size 0.45 μm) and diluted with 2% HNO₃ for Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES, PerkinElmer Optima 8300) analysis for major elements (e.g., Al, Fe, Ca, Na, Si, Ti). Leaching of the non-neutralised and the neutralised bauxite residue was carried out with sulfuric acid (95–97%, Sigma-Aldrich) with a L/S ratio of 10:1. The experiments were performed in the same laboratory shaker at 200 rpm and 25 °C. The leach solution (pregnant leach solution, PLS) was filtered using a syringe filter (pore size of 0.45 μm) and diluted with 2% HNO₃ for ICP-OES analysis for major and minor (e.g., Sc, Y, La, Nd) elements. Lutetium was used as internal standard during the analysis. A synthetic solution with a pre-defined concentration was used as reference. The distribution of major and minor elements was detected using an electron microprobe for microanalysis (EPMA, type JXA-8530 F from Jeol Ltd.).

All the experiments, i.e. neutralisation and leaching, were carried out in triplicate to ensure reproducibility of the results. The errors were determined as the standard deviations on the results.

3. Results and discussion

3.1. Characterisation of the bauxite residue

The concentrations of major elements in the form of oxides in our Greek bauxite residue sample are given in Table 1. Iron(III) oxide is the major oxide in the bauxite residue followed by alumina, calcium oxide, silica, titanium oxide and sodium oxide. Other oxides may also be present in bauxite residue at low concentration (e.g. oxides of arsenic, beryllium, cadmium, chromium, copper, gallium, lead, manganese, mercury, nickel, potassium, thorium, uranium, vanadium, zinc, zirconium, and rare earth elements), as well as non-metallic elements such as phosphorus, carbon and sulfur. In Table 2, the concentrations of selected REEs are shown. The concentrations of Sc, Y, La and Nd are shown because their concentrations in this bauxite residue are relatively high.

Mineralogical analysis allowed the identification of several mineral phases, based on iron (hematite, goethite), aluminium (gibbsite, diaspor, bayerite), calcium (calcite, calcium silicates and calcium aluminosilicates), sodium (sodalite, cancrinite) and titanium (rutile). Perovskite (CaTiO₃) was detected only in very low concentration (below 1 wt%). Approximately 95% of the mineral phases were quantified using the Rietveld method (see Tables S1 and S2 in the Supplementary Information). Table 3 describes the normalised (to 100% total) mineralogical composition of the bauxite residue sample. The quantification of mineral phases was analysed by taking into account the chemical composition of major elements obtained by XRF analysis (Table 1) with an estimated deviation error of approximately 10%.

Table 1
Major chemical components, expressed as oxides, in the bauxite residue.

Compound	wt%
Fe ₂ O ₃	46.7
Al ₂ O ₃	18.1
CaO	8.5
SiO ₂	7.3
TiO ₂	5.8
Na ₂ O	2.8
Loss on ignition	8.5

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