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# Red mud carbonation using carbon dioxide: Effects of carbonate and calcium ions on goethite surface properties and settling



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#### G R A P H I C A L A B S T R A C T



#### A R T I C L E I N F O

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#### ABSTRACT

Carbonation using CO<sub>2</sub> appears as an attractive solution for disposing of red mud suspensions, an aluminum industry hazardous waste since it also offers an option for CO<sub>2</sub> sequestration. Here we report the novel findings that  $CO_3^{2-}$  together with  $Ca^{2+}$  can significantly affect the surface properties and settling of goethite, a major component of red mud. Specifically, their effects on the goethite surface chemistry, colloidal interaction forces and settling in alkaline solutions are investigated. The surface potential becomes more negative by the formation of carbonate inner-sphere complexes on goethite surface. It is consistent with the strong repulsion, decreased particle size and settling velocity with increased carbonate concentrations as measured by atomic force microscopy, particle size analysis, and particle settling. Adding Ca<sup>2+</sup> that forms outer-sphere complexes with pre-adsorbed carbonate changes goethite surface charge negligibly. Changing repulsion to the attraction between goethite surfaces by increasing calcium dosage indicates the surface bridging, in accordance with the increased settling velocity. The adverse effect of carbonate on goethite flocculation is probably due to its specific chemisorption and competition with flocculants. By forming outer-sphere complexes together with the flocculant-calcium bridging effect, calcium ions can eliminate the negative influence of carbonate and improve the flocculation of goethite particles. These findings contribute to a better understanding of goethite particle interaction with salt ions and flocculants in controlling the particle behavior in the handling processes, including the red mud carbonation.

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#### 1. Introduction

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https://doi.org/10.1016/j.jcis.2018.02.006 0021-9797/© 2018 Elsevier Inc. All rights reserved. In the Bayer process used in the aluminum industry, the crushed bauxite ore is digested by concentrated caustic solution

at high temperature and pressure, which leaves behind large amounts of insoluble residue of fine particles, called "red mud" [1]. Red muds from different bauxite ores have a wide range of composition and generally composed by goethite, hematite, boehmite, quartz, sodalite [2,3]. Due to the high alkalinity (pH > 13) and large quantity, the treatment and disposal of red mud have posed serious environmental challenges in soil and groundwater pollution [4,5]. Although many techniques have been investigated to dispose of red mud, the carbonation neutralization with CO<sub>2</sub> shows the greatest potential to provide sustainable outcomes. It is regarded as an inexpensive and safe process that leads to the formation of thermodynamically stable products [6,7]. The innovative nature of carbonation neutralization is that while decreasing the main hazardous alkalinity and overcoming problems associated with red mud storage, the consumption of CO<sub>2</sub> to reduce the greenhouse gas emissions via this sequestration technique can provide additional benefits [8]. In addition, some extra Ca sources can be available or added in a semi-soluble mineral and/or salt form (gypsum or CaCl<sub>2</sub>) to accelerate and enlarge the CO<sub>2</sub> sequestration in neutralization [9]. These studies of neutralization are mainly focused on the conversions of mineral components and decrease of alkalinity in red mud. However, while CO<sub>2</sub> and extra Ca sources are added into the red mud slurry, the soluble carbonate ions  $(CO_3^{2-})$  should be the dominant products of the carbonation process, along with a soluble excess of calcium  $(Ca^{2+})$  [6]. The introduction of these soluble ions may also strongly affect the surface properties and interactions of red mud particles, and subsequently, change the performance of these colloidal particle suspensions in the further storing or handling processes. It is, therefore, worth to investigate the behaviors of  $CO_3^{2-}$  and  $Ca^{2+}$  to understand their roles played in the solid particle suspensions.

For the pervasive distribution and behavior of carbonate in nature, there are particular interests and focus on the carbonate adsorption.  $CO_3^{2-}$  was reported to have some adverse impacts on the red mud flocculation, which decrease the settling velocity and deteriorate the clarity of supernatant [10–12]. In addition, these studies show that  $CO_3^{2-}$  reduce anion adsorption by directly competing for binding sites on the mineral oxide surfaces, and reduce cationic species adsorption by the formation of aqueous carbonate complexes with different sorbing abilities [13–17]. They uptake enhanced proton sorption by direct adsorption or ligand exchange of surface hydroxides, and sorb strongly to minerals with high specific surface areas and have the potential to cover a large fraction of active sites, particularly of metal oxides [18,19]. These macroscopic observations suggest a strong surface interaction between  $CO_3^{2-}$  and minerals in the aqueous systems.

As one of the dominant multivalent ions in soil solutions and mineral systems, the presence of Ca<sup>2+</sup> has been shown to affect the adsorption of naturally dissolved matters or synthetic polyelectrolytes onto iron oxides at high pH. It may change the surface charge of iron oxides and accordingly colloidal stability and sorption properties [20]. The study of competitive interactions of ions with charged goethite particles shows that a monodentate calcium outer-sphere surface complex dominates its adsorption at acid pH range and a monodentate inner-sphere complex at basic pH range (FeOHCa) [21]. In the study of the influence of calcium ion on the adsorption of polyelectrolytes onto oxide surfaces [22], it is found that the polymer adsorption is markedly increased by Ca<sup>2+</sup> ions. Due to the possibility of forming intermolecular bridges between the carboxylic groups (RCOO<sup>-</sup>–Ca<sup>2+</sup>–<sup>-</sup>OOCR), divalent cations screen the negative charges of the polyelectrolytes better than univalent ions.

As presented above, both  $CO_3^{2-}$  and  $Ca^{2+}$  exist in the original and carbonated red mud. But there is few research on their synergetic effects and direct interactions with particles, especially in the alkaline conditions. In this work, goethite is chosen as the typical

iron-containing monomineral in the red mud under the digestion condition of gibbsite bauxite [23,24], which can be regarded as a main stable and insoluble component with the treatment of carbonation neutralization. The effects of  $CO_3^{2-}$  and  $Ca^{2+}$  on the goethite surface properties and aggregation with or without polymeric flocculants are investigated by surface potential analysis, particles free settling test, and interaction force measurement using atomic force microscopy (AFM). This study is necessary to predict the effect of  $CO_3^{2-}$  and  $Ca^{2+}$  ions on the particle properties and settling of red mud, which can help to improve the disposal of red mud ultimately.

#### 2. Experimental

#### 2.1. Materials and reagents

#### (1) Goethite particles

Goethite is an  $\alpha$ -iron (III) hydroxy-oxide mineral with the chemical formula FeOOH. A goethite powder (99% purity, Bioscientific, Sydney, Australia, A16267.30 T) with a mean size (50% volume percentage of passing size), D(v, 0.5) of 3.3  $\mu$ m was used, as obtained, for the surface (zeta) potential measurement and settling tests. The BET surface area of the goethite particle was measured to be 17.45 m<sup>2</sup>/g by N<sub>2</sub> adsorption (Micromeritics Tristar 3020, USA).

#### (2) Goethite substrate wafer and AFM colloidal probe

The goethite substrate wafer for the AFM measurements was prepared from a piece of goethite crystal (Crystal Peak Mining district, USA). The crystal was embedded into an epoxy resin and cut to give a thickness of 3 mm. Then the substrate wafer was manually polished with 3  $\mu$ m and 1  $\mu$ m diamond suspensions (Struers, Milton, Australia), thoroughly washed with acetone, ethanol and copious amounts of deionized water, dried in a stream of high purity nitrogen and then UV-treated for 10 min [25]. The colloid probe was prepared by attaching the goethite particle (10–30  $\mu$ m from crushed goethite crystals selected under a microscope using a 3D micro-manipulator) to the apex of a micro-fabricated AFM cantilever with an epoxy resin [26,27] as shown in Fig. 2.

#### (3) Other materials

The commercial anionic polymer flocculant (Industrial reagent, Nalco 9779, Nalco Water, Australia), primarily consisting of ammonium polyacrylate, was used in the flocculation tests [28,29]. According to the manufacturer's descriptions, the flocculant has high molecular weights with hydrocarbon backbones, as shown in Fig. 1. Stock flocculant solutions were prepared in alkaline solutions (0.25 M NaOH and 1 mM NaCl) and left to equilibrate for 24 h.

Sodium hydroxide (AR grade, Chemsupply, Australia) and hydrochloric acid (RCI Premium grade, RCI Labscan, Australia) were used for the pH adjustments. Sodium carbonate, calcium chloride, and sodium chloride (AR grade, Sigma-Aldrich, Australia)



Fig. 1. Schematic of the functional groups present in the polymer flocculant.

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