Reactions and Phase Transformations During the Low-Temperature Reduction of Bauxite Residue by H_2 in the Presence of NaOH, and Recovery Rates Downstream

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Abstract

The scope of this work is to investigate the reactions and phase transformations that occur during the low-temperature reduction of bauxite residue (BR) by hydrogen in the presence of NaOH, and how that affects the recovery downstream of the different elements. Different processing conditions were studied, such as temperature, time, and NaOH dosage under a pure hydrogen atmosphere. XRD results revealed that hematite (Fe_2O_3) and goethite (FeOOH) were completely converted to magnetite (Fe_3O_4) when a blend of BR and 20 wt % NaOH was treated at 500 °C for 30 min. The iron recovery and grade of magnetic rich fraction after water leaching -magnetic separation of reduced BR (at 500 °C, 20 wt % NaOH, 30 min reduction time) is 87.7 % and 44.5 % respectively, whereas the aluminum and sodium recovery under these conditions is 75 % and 87 %. The formation of $NaAlO_2$ occurs at 400 °C, and the amount formed increases till 700 °C. When NaOH addition exceeds 25 wt % and the temperature is higher than 600 °C, the formation of water-soluble sodium iron oxide ($NaFeO_2$) and sodium-calcium silicate ($Ca_2Na_2Si_3O_9$) along with wüstite (FeO) takes place. This led to lower Fe and Al recovery and further dissolution of Fe, Si, and Ca in the leach liquor during water leaching and wet-magnetic separation. For thermal processing at 500°C for 30 min with 20 wt % NaOH, the amount of Fe_3O_4 forming is the highest, and via magnetic separation, a Fe-rich fraction is recovered. In the non-magnetic fraction, perovskite ($CaTiO_3$), calcite ($CaCO_3$), and quartz (SiO_2) are enriched, whereas the leach liquor is Na and Al-rich, as confirmed by ICP-OES measurements. The partitioning of rare earth elements in the different phases, as well as what is overall the preferable recovery process, are also discussed.

Keywords: Bauxite residue, Hydrogen, Circular economy, Alkaline roasting, Phase transformations.

1. Introduction

The Bayer processing of bauxite for alumina extraction generates a substantial amount of strong alkaline solid waste known as bauxite residue (BR) or red mud. On average, 1.35 tonnes of alumina produces 0.8-2.5 tonnes of bauxite residue, with global stockpiling expected to reach 5 billion tonnes by 2020 [1]. Currently, most efforts on BR utilization focus on recovering key metals, like iron, aluminum, titanium, and REEs, and on manufacturing building and construction materials. However, the utilization rate is less than 3 %. It is worth noting that the composition of BR can vary depending on the source of the bauxite and the conditions under which it is processed. BR is typically made up of 5-65 % iron oxides, 5-40 % aluminum oxides, 5-40 % silicon oxides, and 3 percent–20 % titanium oxides in addition to a sizable amount of strategic metals like scandium (Sc) and lanthanides as REEs [2]. BR may therefore be regarded as supplementary raw material for the recovery of Fe, Al, Ti, Ca, and REEs. Bauxite, which makes up 70 % to 80 % of the deposits of scandium, has not been employed as a raw material to extract scandium. BR is enriched with more than 98 % Sc (50–120 mg/kg) [13, 14] during the alumina production from the Bayer process. This has resulted in the recovery of Fe, Al, Ti, and REEs from BR being explored at the laboratory scale with the objective of achieving near-zero waste valorization and carbon footprint. Metal recycling technologies like physical separation by direct magnetic separation, pyrometallurgical procedures including smelting and reduction roasting-magnetic separation, and hydrometallurgical acid leaching techniques, are the broad categories for the recovery of metals.

One technique for recovering iron fractions while leaving behind the non-magnetic fractions (Al, Si, Ti, Ca, Si) of BR is direct magnetic separation [1]. The bulk of hematite and goethite particles are fine, weakly magnetic, and agglomerate with fine non-magnetic minerals of desilication product (DSP), making them difficult to separate from BR. Additionally, the distribution of minerals phases in BR is complicated and the extraction of iron is intertwined with that of other elements. As a result, using physical separation techniques, it is challenging to extract metals from BR. On the other hand, in the hydrometallurgical route, various acids (HCl, H_2SO_4, HNO_3) are used to dissolve elements (including Fe, Al, Ti, Ca, and REEs) in BR by varying the acid dosage, leaching duration, and temperature. However, multielement dissolution complicates downstream separation processes due to complicated flowsheets, huge volume generation of liquid byproducts, high impurity levels, hazardous/corrosive acids, and/or uneconomic process conditions [3,4].

As a result, BR is an intriguing potential resource, with the techniques investigated generally falling within the categories of reduction smelting and reduction roasting. Slag and a product of pig-iron grade are produced when BR is smelted using a reducing agent (carbon) at higher temperatures such as 1400-1800 °C through the smelting reduction process. High energy consumption and operating temperatures, and high sodium content in the raw materials in the smelting process limit commercial manufacturing on an industrial scale [5]. Researchers have extensively explored the popular technique of reductive roasting of BR and other residues at temperatures between 300 to 1000 °C using coke, CO, or hydrogen to separate iron minerals such as Fe_3O_4, and pure Fe [6-12]. A recovery rate of 88 % Fe and grade of 90 % were observed when BR was reduced with carbon at 1000 °C, with the resulting iron possible to use as an ingredient in iron making directly. It has been suggested from previous research methods [6,7,11] that the sodium-based flux (sodium carbonate, sodium hydroxide, and sodium sulfate) during reductive roasting substantially increases the reduction and recovery of Al, Si, and Na-bearing non-magnetic phases via the formation of water-soluble sodium silicate and sodium aluminate phases from reduced BR after magnetic separation and water leaching [6-13]. The low-temperature reduction roasting of BR at 600 °C with hydrogen and addition of NaOH is investigated to recover the pure iron and aluminum simultaneously but lower recovery rates of Al and Fe were observed. Also, it should be noted that the above-mentioned studies do not provide a sufficient explanation of the non-magnetic, magnetic, and REEs phase assemblage and the ongoing transformations in reduction roasting. Additionally, none of the processes showed proof-of-concept for industrial upscaling, confirming that all the above-mentioned technologies are in their infancy. For the alumina industry to grow sustainably and environmentally friendly, an efficient bauxite residue valorization is a key technology.

In the present study, the reduction roasting of BR by adding NaOH at lower temperatures ranging from 400-700 °C under pure hydrogen is investigated by adjusting time (30-120 min), temperature, and NaOH addition (10-25 wt %). Then, the reduced products were processed to combine water leaching and wet magnetic separation method for the recovery and separation of metals such as Fe, Al, Na, Ca, Ti, and Sc. X-ray powder diffractometer (XRD), Rietveld QXRD analysis, scanning electron, microscope-energy dispersive spectroscopy (SEM-EDS), and electron probe micro-analyzer (EPMA) were utilized to investigate the phase transformation, the authors' opinions, absolving the community of all responsibility. The Harare Project webpage is https://h2020harare.eu/.

5. References

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