

Hydrogen Reduction of High Temperature Sintered and Self-Hardened Pellets of Bauxite Residue produced via the Addition of Limestone and Quicklime

Manish Kumar Kar¹, Casper van der Eijk² and Jafar Safarian³

1. Ph.D. Student

3. Associate Professor

Department of Materials Science and Engineering, NTNU, Trondheim, Norway

2. Senior Research Scientist

SINTEF, Trondheim, Norway

Corresponding author: manish.k.kar@ntnu.no

Abstract

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The reduction of iron oxide by hydrogen and phase transformation in Bauxite Residue (BR) were experimentally studied at 1000 °C. In the present work, two types of oxide pellets were first made from bauxite residue via the addition of CaCO₃ (Limestone) and CaO (quicklime) powder. The Ca was added to form a leachable mayenite phase (12CaO.7Al₂O₃) with the existing alumina in BR. Either only CaCO₃ was added or a mixture of CaO and CaCO₃ was added (Fixed Ca content). The former was sintered at 1150 °C, while the latter could be self-hardened through the cementing effect of CaO in exposure to moisture and air. Both types of pellets were reduced by hydrogen in a thermogravimetry furnace at elevated temperature under similar conditions. The pellets were characterised by X-Ray Diffraction (XRD), Scanning Electron Microscope (SEM) coupled with energy dispersive spectroscopy (EDS), physical and mechanical testing. It was found that the phase formation after reduction is dependent on the characteristics of the used pellets. The high-temperature sintered pellets had higher strength and it was also found that produced oxide pellets were different with respect to the dominant phases present in them. There was a negligible amount of gehlenite (Ca₂Al₂SiO₇) formation in the self-hardened pellets, while this phase was a dominant phase in the sintered pellets. The two pellets showed similar reduction behaviour, while different chemical, physical and mechanical properties were observed. Higher iron recovery was observed in the reduced sintered pellets. On the other hand, the reduced self-hardened pellets had a higher amount of alumina containing mayenite leachable phase.

Keywords: Hydrogen reduction, Bauxite residue, Thermogravimetry, Self-hardened pellet, Sintered pellet, Phase transformation.

1. Introduction

As the modernization of society is growing, the demand for aluminium has increased with time [1]. Bauxite is the major raw material for the production of alumina. Most alumina is produced around the world through the Bayer's process, which generates a waste known as red mud or bauxite residue (BR) in dewatered form [2]. However, the generation of BR has serious environmental challenges due to its properties and challenges to storage [2]. Many researchers have tried to find an efficient way of the utilization of BR because it can be a potential secondary source for the metals like Fe, Al, Ti, Ca, and many more. However, utilization of BR is still impenetrable mainly due to economic reasons and running efficient processes. That is why it is important to find out a different technology for the utilization of BR to get rid of the environmental problem [3]. As the environmental greenhouse gas concern increases with time, green reduction will be a promising technology for the oxide reduction in BR.

Most of the iron production in the world is through the blast furnace and basic oxygen furnace (BF/BOF) route, which needs coking coal both as a reductant and heat source. As the shortage of

coking coal increases and the environmental climate concern increases, hydrogen will most likely be the future reductant for the steel industry [4]. Globalization increases with time, and for future economic growth, industry has to adopt low carbon technologies [5][6]. There is some major difference underlined between hydrogen reduction and carbothermic or carbon monoxide reduction from a metallurgical point of view. Iron oxide reduction with hydrogen is endothermic, however, the reduction kinetics is high therefore the industrial operation must be different compared to conventional carbothermic reduction[7]. The reduction of iron oxide by hydrogen goes through either two or three steps depending upon the applied reduction temperature. Temperature higher than 570 °C, hematite (Fe_2O_3) transforms to magnetite(Fe_3O_4) and wustite (FeO) and finally to metallic iron but at temperatures below 570 °C, hematite(Fe_2O_3) to magnetite(Fe_3O_4) first, and then directly to metallic iron because wustite (FeO) is not stable below 570 °C [7] [8].

Limited research works have been carried out in literature on the hydrogen reduction of BR and bauxite ore. Low temperature Hydrogen reduction of BR has been studied for the conversion of Fe_2O_3 to Fe_3O_4 and formation of leachable sodium aluminate phase [9]. They found the maximum conversion of hematite to magnetite around 96 % with 20 wt % NaOH addition and reduction with 5 vol% H_2 for 120 mins at 500 °C. Hydrogen reduction of Bauxite ore has been studied [3] for the iron separation prior the ore smelting. It was found that the reduction of Hematite to iron starts below 560 °C and the rate and extent of reduction increases with increasing temperature, however, formation of hercynite (FeAl_2O_4) retards the complete reduction above 760 °C. The addition of sufficient Ca to BR avoids the hercynite formation during the reduction step.

This work focuses on hydrogen reduction behaviour of BR pellets, phases formation in the process and mechanical properties of two different types of pellets with similar reduction conditions. CaCO_3 is partly substituted by CaO in order to make self-hardening pellets that do not need to be sintered before a reduction in hydrogen thereby reducing the cost of the process. The effect of CaO addition on the pellets strength and the phase formation have been studied. The mechanical properties are studied through breaking load test, tumbler test, and abrasion test for the sintered and self-hardened pellets, and it is shown that they are related to the type of pellets, in addition to the chemical and physical properties of the pellets

2. Experimental Procedure/ Research Methodology

2.1 Materials

Bauxite residue (BR), limestone (CaCO_3), quick lime (CaO), and water were used for pellet making. BR was supplied from the Mytilineos metallurgy business unit S.A. (previously known as aluminum of Greece). The limestone was provided by VUGIUKLI SA, Greece, and quick lime was from the NorFraKalk. The materials were deagglomerated and sieved below 500 μm and dried in oven for overnight at 80 °C. Before pelletization, the dried materials with the appropriate ratio were mixed well for homogenization.

2.2 Pellets Making

For the self-hardened pellets, 1000g of BR, 497 g of limestone, and 80 g of quicklime were mixed in a rotating drum with clockwise and anti-clockwise rotation for 4 minutes for homogenization, but in sintered pellets only 639 g CaCO_3 used per 1000 g of BR. However, in both mixtures the calcium weight fraction remains fixed ($\text{CaO}/\text{Al}_2\text{O}_3 = 1$) regarding the chemical compositions of BR, Limestone and quick lime presented in Table1.

4. Reduction rate and extent of iron containing oxides are very similar for both pellets
5. There is more formation of alumina leachable phase in reduced self-hardened pellets as compared to reduced sintered pellets
6. Iron particle size of reduced self-hardened pellets is more than the reduced sintered pellets

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