

Stepwise carbothermal reduction of bauxite ores

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ABSTRACT

The commercial technologies for aluminium production include production of alumina from bauxite and smelting of alumina to produce aluminium. The current technology is energy intensive, and a major source of greenhouse gas emissions and harmful fluoride emissions. Carbothermal reduction of bauxite is a promising alternative technology for aluminium and aluminoalloy production. Western Australia and Queensland bauxite ores were carbothermally reduced in steps in argon. Experiments were performed in a high temperature vertical tube furnace, and the off-gas composition was monitored using an infra-red gas analyser. The phase composition of reduced samples was characterised by XRD. Oxygen and carbon contents in reduced samples were determined by LECO analysers. The morphology of the surface and intersections was observed by SEM. The chemical compositions of the phases in the reduced samples were detected by EDS. The results of this study have proved the concept of stepwise reduction of bauxite ores in solid state by appropriate control of reduction temperature. Below 1100 °C, only iron oxides were reduced to metallic iron. A ferroalloy phase was formed at 1200 °C and above. The products in the bauxites reduced to 1600 °C include a ferroalloy of silicon and aluminium, carbides of titanium, silicon and aluminium, and unreacted alumina.

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1. Introduction

The commercial technologies for aluminium production include production of alumina from bauxite and smelting of alumina to produce aluminium. The current technology is energy intensive, and a major source of greenhouse gas emissions and harmful fluoride emissions.

Carbothermal reduction of alumina is a promising alternative technology for aluminium and aluminoalloy production. Compared with the Hall–Heroult process, carbothermal reduction of alumina offers advantages of a simpler process, lower cost and a lower requirement for raw materials. Previous assessments showed that carbothermal reduction has the potential to reduce energy consumption by up to 38%, capital costs by more than 60% and decrease CO₂ emissions by up to 30%. It has no fluoride emission, and may decrease overall operating costs by 25–30% (Bruno, 2003; Sayad-Yaghoubi, 2007; Myklebust and Runde, 2005).

In most works on development of carbothermal reduction of alumina, the strategy of two stage reactors was adopted (Gruenert and Mercier, 1963; Cochran and Fitzgerald, 1981; Persson, 1983; Dougan and Southam, 1985; Dmitriev and Karasev, 2000; Johansen et al., 2003). In these works, an alumina–aluminium carbide melt is first formed, then aluminium is produced from the melt. A technical and economic assessment of the process was presented by Choate and Green

(2006). Because of the corrosion/erosion effects of the melt at high temperatures, generally over 2000 °C, there are engineering issues in commercialisation of these processes.

Li et al. (2011) demonstrated that alumina can be carbothermally reduced in solid phase to form aluminium carbide. The latter can be decomposed to produce aluminium. This suggests a new potential technology of aluminium production by carbothermal reduction–decomposition.

Production of alumina from bauxite ores consumes a large amount of caustic soda, and generates a large amount of “red mud” slurry waste. A desirable process for aluminium production is to start from bauxite in which the impurities, oxides of iron, silicon and titanium can be converted into useful products. Investigations on direct reduction of bauxite are limited in literature. Fujishige et al. (1986a,b) investigated the effects of the addition of CaO and other Ca containing compounds on the reduction of bauxite in a blast furnace. CaO and CaCO₃ addition promotes the formation of an aluminium alloy by decreasing the formation of aluminium volatiles. Bauxite with high Fe and low SiO₂ content was considered to be the favourable raw material for aluminium in a blast furnace. Wang et al. (2010) patented a technology to reduce bauxite flotation tailings in an electric arc furnace in order to produce a high aluminium alloy at 2300–2500 °C. They also reported the formation of an Al–Si alloy at temperatures above 1800 °C. They further claimed that the optimum conditions for Al–Si alloy production are: a pressure at 0.1 MPa, a temperature of 1900 °C, a carbon content of 95% of the theoretical amount, and one hour heating (Yang et al., 2010).

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Goldin et al. (1999) proposed vacuum carbothermal processing of low-iron bauxites. Reduction by dry firing produces ceramic products containing oxide, carbide, and oxycarbide of aluminium, silicon and titanium. Kitajima and Kasai (1999) and Mishra et al. (2002) investigated the processing of the red mud waste from the Bayer process to recover value added products. The former investigation tested the recovery of the iron value by carbothermal reduction. They claimed an achievement of 90% iron recovery by adding CaCO_3 as an iron grain coalescence agent. Mishra et al. attempted the recovery of alumina in red mud by soda ash sintering and caustic leaching, and iron recovery by carbothermal reduction.

The present research investigates carbothermal reduction of bauxite ores at solid state. Both temperature programmed reduction (temperature is ramped continuously with time) and stepwise reduction (where temperature is changed in steps) procedures are used in this work.

2. Experimental

The major compositions of Western Australia and Queensland bauxite ores are presented in Table 1. Western Australia bauxite comprised approximately 40% alumina, 19% iron oxides, 17% silica and ~1% titania. Compared with Western Australia bauxite, the most significant differences of Queensland bauxite are the alumina and silica contents. Queensland bauxite consists of more than 50% alumina, 13% iron oxides and 6% silica. Titania content is 2.6% in Queensland bauxite.

Both bauxite ores were reduced carbothermally in flowing argon at 1000 ml/min. The ores were crushed and sieved to $<212 \mu\text{m}$ and then wet mixed with graphite powder ($<20 \mu\text{m}$) in a C/O molar ratio of 1.2:1. After drying, the mixtures were pressed into cylindrical pellets of 8 mm in diameter and about 10 mm in height. Each pellet had a mass of about 1 g. Experiments were performed in a high temperature vertical tube furnace, with a reactor setup presented previously (Zhang and Ostrovski, 2000). The off-gas composition was monitored using an infra-red gas analyser. The phase composition of reduced samples was characterised by XRD. Oxygen and carbon contents in the reduced samples were determined by LECO analysers. The morphology of the surface and intersections was observed by SEM. The chemical compositions of the phases in the reduced samples were detected by EDS.

Temperature programmed reduction experiments were carried out from 850 °C until 1600 °C, and then the temperature was maintained for 30 min. The rate of reduction as presented by CO evolution rate consisted of multiple peaks. In the step by step reduction experiments, reduction temperatures were decided on the basis of the peak temperatures of reduction curves of the temperature programmed reduction. For Western Australia bauxite, 980 °C, 1032 °C, 1360 °C and 1600 °C, and for Queensland bauxite, 1051 °C, 1255 °C and 1600 °C were selected as the step temperatures. The last temperature, 1600 °C, was limited by the maximum operation temperature of the furnace used.

3. Results and discussion

Western Australia and Queensland bauxite ores were reduced using two reduction procedures. Temperature programmed reduction was followed by sample characterisation to identify the stages of reduction of different metal oxides and their temperature ranges of reduction. Further reduction experiments were carried out in different steps with each

Table 1
Major components of bauxites by XRF, wt.%.

| Ore | Al_2O_3 | Fe_2O_3 | SiO_2 | TiO_2 |
|---------------------------|-------------------------|-------------------------|----------------|----------------|
| Western Australia bauxite | 39.9 | 19.3 | 17.3 | 1.3 |
| Queensland bauxite | 52.5 | 13.6 | 6.5 | 2.6 |

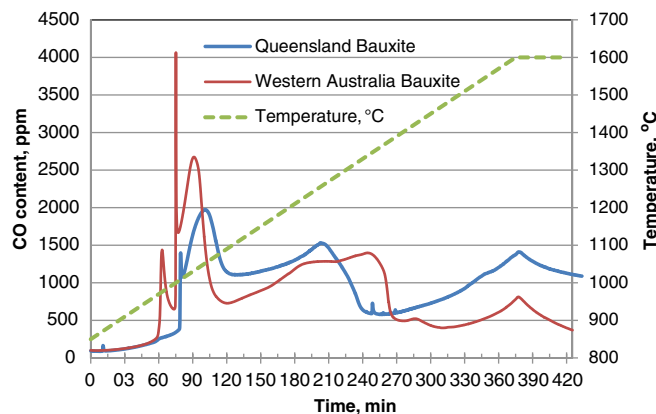


Fig. 1. Change in CO content in the off-gas during temperature programmed reduction of bauxite ores. Temperature was ramped from 850 °C to 1600 °C at 2 °C/min.

step being at constant temperature. This was to further demonstrate the feasibility of stepwise reduction of metal oxides from bauxite ores.

3.1. Progress of carbothermal reduction

Temperature programmed reduction was carried out under a flowing argon gas atmosphere. The rate of reduction was monitored by detection of CO released from the reactions using a $\text{CO}/\text{CO}_2/\text{CH}_4$ infra-red analyser.

Fig. 1 presents the change of CO content in the off gas during experiments for both bauxites. The reduction curves consist of multiple overlapped peaks, showing that the metal oxides in the bauxite ores were reduced in different temperature ranges. The reduction curve of Western Australia bauxite is more complex than that of Queensland bauxite due to more impurities in the former ore.

Based on the major compositions (Table 1) and the reducibility of the metal oxides (Li et al., 2011; Dewan et al., 2009, 2010, 2011) it is expected that the first major reduction stage was reduction of iron oxides which took place in the temperature range of 850 to 1100 °C. The second major stage of reduction corresponding to the reduction of silica and titania, was in the range of 1100 to 1500 °C. Further reduction was mainly attributed to alumina, which was not complete at the end of experiments.

In the step by step reduction experiments, reduction temperatures were decided making reference to the peak temperatures of reduction curves in Fig. 1. The temperature and CO evolution curves during reduction of Queensland bauxite are presented in Fig. 2.

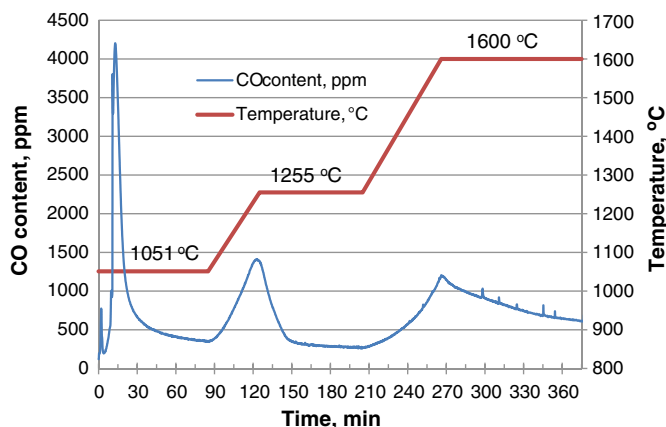


Fig. 2. Changes in experimental temperature and CO content in step by step reduction. The ramping rate was 5 °C/min between two step temperatures.

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