



The effect of sodium sulphate on the hydrogen reduction process of nickel laterite ore



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ABSTRACT

A series of nickel laterite ores with different calculated amounts of anhydrous sodium sulphate were prepared by physical blending or sodium sulphate solution impregnation. The reduction of the prepared nickel laterite ore by H₂ was carried out in a fluidised-bed reactor with provisions for temperature and agitation control, and the magnetic separation of the reduced ore was performed using a Davis tube magnetic separator. The mineralogical properties of the raw laterite ore, reduced ore and magnetic concentrate were characterised using ICP, TG–DSC, N₂ adsorption, X-ray diffraction and optical microscopy. The catalytic activity of sodium sulphate was also studied by using Hydrogen temperature-programmed reduction. The experimental results indicate that Na₂SO₄ could overcome the kinetic problems faced by the laterite ore and that it exhibited noticeable catalytic activity only if the temperature reached at least 750 °C. This high temperature accelerated the crystal phase transition of the silicate minerals and increased the utilisation of H₂. In comparing the results from the two different methods for adding Na₂SO₄, the nickel content and recovery of the magnetic concentrate were increased by using the impregnation method rather than the physical blending method and the increasing amount of sodium sulphate assisted in the further beneficiation of nickel. The partial pressure of H₂ and the reducing time also affected the reduction process of the iron oxides. The results of the microscopic study indicated that the formation of a Fe–S solid solution, which was derived from the SO₂ sulphide reduction of FeO, was conducive to mass transfer and accelerated the coalescence of metallic ferronickel particles. For the nickel laterite ore, under the synergistic effect of sodium sulphate and hydrogen, a nickel content and nickel recovery of 6.38% and 91.07% were obtained, respectively, with high product selectivity.

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1. Introduction

Nickel is the main alloying element of stainless steel alloys, and the rapid growth (an average of 9% over the past few years) in stainless steel demand has led to a dramatic increase in the production of nickel metal in recent years (Moskalyk and Alfantazi, 2002; Johnson et al., 2008). Nickel sulphide ores, which currently account for over 60% of nickel production (Harris et al., 2011), are an easily-processed ore resource, which can be treated using mechanical beneficiation methods such as stage-grinding, gravity separation and flotation. However, with the continued exploitation of nickel sulphide deposits, miners of nickel sulphide ore have been confronted with increasing challenges due to deeper drilling requirements and reserve depletion (Zhu et al., 2012a,b). Taking full advantage of nickeliferous laterite ore, which is calculated to

account for over 70% of the world's land-based nickel resources, has attracted greater attention in recent years (Norgate and Jahanshahi, 2011).

Nickel laterite ores are difficult to concentrate due to their low nickel content and isomorphous goethite or serpentine crystalline structure (Mudd, 2010; Eckelman, 2010). The extensive and complex structure has prevented the nickel grade from being pre-concentrated by fine grinding or other methods of physical beneficiation (Norgate and Jahanshahi, 2011; Wills and Napier-Munn, 2006). Thus, Pyrometallurgical or Hydrometallurgical production methods are exploited for manufacturing nickel metal from low-grade ores. Hydrometallurgical processes, including high pressure acid leaching (HPAL), atmospheric acid leaching (AL) and ammonia-ammonium carbonate leaching (Caron process) (Georgiou and Parangelakis, 1998; Wang et al., 2013; Loveday, 2008), are more appropriate for treating limonite ore that has a high iron content but a low garnierite content. Nevertheless, the disadvantages of these processes need to be resolved, especially the limited extraction of nickel and substantial energy and reagent

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requirements (Li et al., 2010; Guo et al., 2009). Pyrometallurgical techniques are well suited to treating saprolite, where the garnierite content is high and the iron content is low. The primary disadvantage of these processes is that they require substantial energy inputs Norgate and Jahanshahi (2010). As a result, pre-reduction is used during the pyrometallurgical treatment of nickel laterite ores to reduce the load on the nickel smelting furnace.

Because the pre-reduction of nickel laterite ore is also an energy-intensive metal extraction process, several investigations have been performed to study the effect of different reductants such as carbon or CO, on the reduction of laterite ore. From an economic perspective, the continuing increase in the market price of steam coal, as well as its importance as a primary energy source in other fields, disadvantages carbon thermal reduction processes with high costs and relatively low energy efficiency. One option for improving the energy efficiency of ferronickel production is to utilise renewable energy sources, such as waste heat emitted from a smelter or from the carbonisation of coal Kim et al. (2010). China Shanxi is a significant producer of coal and coke, and has a large amount of surplus coke oven gas that contains 55% hydrogen gas Du et al. (2012). To take advantage of this resource, the comprehensive utilisation of waste heat and reducing gas from coke oven gas used in ferronickel production is proposed by our laboratory. As a preliminary study of this innovative process, hydrogen gas was employed as the preferred reducing agent for the processing of nickel laterite ore.

Utigard et al. (2005) studied the reduction kinetics of Goro nickel oxide using hydrogen and provided theoretical guidance for this research. However, the nickel in nickel laterite ore is finely disseminated in the molecular lattice of goethite or hydroxyl silicate (Li et al., 2010). This structural difference makes the reduction unlike chemical grade NiO which has faster reduction kinetics. To address this problem, catalysts are necessary to overcome the insufficient kinetics conditions. There have been previous reports that indicated that alkali metal sulphates are the best-known group of activators (Li et al., 2012). Thus, in this study, the author conducted hydrogen reduction of low-grade nickel laterite ore with the addition of sodium sulphate followed by magnetic separation to determine the effects of temperature, time, partial pressure of H₂ and content of sodium sulphate on the nickel reduction process. Moreover, the effects of different sodium sulphate addition methods, such as physical mixing and the sodium sulphate solution impregnation method, were also examined. The phase transformation accompanying reduction and the catalytic mechanism of sodium sulphate were analysed using instrumental analysis techniques such as ICP, XRD and DTG–DSC, and thermodynamic calculations.

2. Experimental

2.1. Characterisation

The main chemical composition of low-grade nickel laterite ore was analysed by inductively coupled plasma emission spectroscopy (ICP) on an ICP-9000 (N + M) spectrometer made by Thermo Jarrell-Ash Corp., USA. The distribution of nickel in the laterite ore was analysed using the chemical phase analysis methodology.

The X-ray diffraction (XRD) analysis was performed on a Rigaku D/max2500 diffractometer, using Cu K α radiation (40 kV, 100 mA) at a scanning rate of 4°/min from 5° to 85°.

DTG–DSC analysis was conducted on a NETZSCH STA409 PC/PG unit in a nitrogen atmosphere (flow rate: 50 mL/min and heating rate: 10 °C/min).

The reducibility of laterite ore with the addition of sodium sulphate was measured by H₂-TPR. The experiments were conducted under a mixture of 40% H₂ in argon and a total flow rate of 40 mL/min flowing through the sample at a heating rate of 5 °C/min. For

comparison, H₂-TPR measurements of raw nickel laterite ore were also carried out.

Optical microscopy analysis was performed on an Axio scope A1 microscope made by Carl Zeiss AG, Germany. The powder samples were treated with a vacuum inlaying machine and polished using sand paper. The polished sections were prepared to observe morphological changes.

2.2. Materials

The low-grade nickel laterite ore used in this study was collected from Indonesia. Hydrogen was employed as a gaseous reductant and reagent grade sodium sulphate was selected as a catalyst.

2.3. Sample preparation

Nickel laterite ore that contained a large amount of absorbed and crystallised water was first dried at 60 °C for 24 h in a dryer oven and then crushed using a jaw crusher. The crushed nickel laterite ore was then screened using a sieve to a particle size of less than 2 mm and the particles larger than 2 mm were ground to the required size using a laboratory-scale rod mill.

Reduced samples containing different amounts of Na₂SO₄ were prepared using two distinct methods:

(a) Physical blending method

The ground laterite ore was uniformly mixed with sodium sulphate by mechanical stirring. The nominal content of Na₂SO₄ was 5, 10, 15 and 20 wt%, and the corresponding nickel laterite ore was denoted as PM ore.

(a) Sodium sulphate solution impregnation

In a typical synthesis procedure, the ground nickel laterite ore was considered as a carrier of sodium sulphate and prepared using an immersion method. Six samples of the ground nickel laterite ore, each 10 g, were calcined at 300, 400, 500, 600, 700 and 800 °C for 1 h in a corundum crucible within a muffle furnace before being analysed with a sorption analyser. The specific surface areas and pore diameters for ores calcined at various temperatures are presented in Table 1, which shows that the surface areas decreased with increases in the calcination temperature (due to the interparticle sintering) and that the pore sizes fall into the range of microporous and mesoporous. Then, different amounts of sodium sulphate (the dosages of sodium sulphate equalled the masses used in the physical blending method) were dissolved into 500 mL of distilled water at 40 °C, followed by magnetic stirring. After the sodium sulphate was completely dissolved, 1 kg of the calcined ore which with largest specific surface area was immersed in different concentrations of sodium sulphate solution and air-dried naturally (coded as IM ore). Finally, the air-dried sample was milled in a rod mill for 3 min.

Table 1

The specific surface areas and pore size distributions of the laterite ore calcined at different temperatures.

Calcination temperature (°C)	300	400	500	600	700	800
Surface area ^a (m ² /g)	81.05	77.52	72.19	50.68	44.48	18.17
Pore size (nm)	1.583	1.223	2.727	3.046	2.724	2.292

^a Multi-point BET surface area.

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