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Investigation into coal-based magnetizing roasting of an iron-rich rare earth ore and the associated mineralogical transformations



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ABSTRACT

The factors influencing goethite to magnetite conversion during magnetizing roasting of an iron-rich rare earth ore using coal as the reductant were explored in the present paper. The effect of temperature, time and reductant addition on magnetite conversion; and the optimal conditions which resulted in complete conversion of goethite to magnetite were determined through the use of satmagan and quantitative X-ray diffraction. For this particular ore, optimal roasting conditions were determined to be a roasting temperature of 600-650 °C with the addition of 10-20 wt% coal in the mixture and a roasting time of 90 min. The mineralogical changes occurring during magnetizing roasting were also investigated using X-ray diffraction. Magnetite formation was found to proceed by the reduction of hematite formed from the dehydroxylation of goethite. X-ray diffraction showed that further reaction of magnetite to form wüstite and fayalite had taken place under highly reducing conditions and temperatures exceeding 650 °C. The primary rare earth mineral in this ore, monazite was stable at all roasting conditions whilst florencite underwent thermal decomposition resulting in an increase in the monazite content of the roasted ore.

1. Introduction

The rare earth elements (REEs) are a group consisting of the lanthanides (lanthanum to lutetium) plus scandium and yttrium which are of strategic importance due to their use in high-end technologies (Hoatson et al., 2011). The largest end uses of REEs are in permanent magnets (neodymium-iron-boron and samarium-cobalt) which find use in wind turbines and electric vehicles, rechargeable batteries (nickelmetal hydride batteries), catalysts (petroleum cracking catalysts, catalytic converters) and metal alloys (Krishnamurthy and Gupta, 2004; Lucas et al., 2014; Xie et al., 2014). Rare earths do not occur naturally as metallic elements but are present in a wide range of minerals (Faris et al., 2017; Jordens et al., 2013; Jun et al., 2011; Zhu et al., 2015). Around 200 rare earth (RE) bearing minerals have been identified to date however only a handful of minerals are of commercial importance. In terms of mineral resources 95% of the total rare earths are contained in three minerals; bastnaesite [(Ce, La)CO₃F], monazite [(Ce,La)PO₄] and xenotime (YPO₄), with other less common minerals and ion adsorption clays making up the balance (Krishnamurthy and Gupta, 2004). Total RE reserves in the world are approximated at 130 million metric tonnes (oxide basis) with 50% of the reserves located in China and the remainder being located in the CIS, Brazil, Vietnam, India and

Australia (USGS, 2016). The current supply base of REEs is dominated by China (Jaireth et al., 2014), which controls over 90% of world RE production (Haque et al., 2014; Kanazawa and Kamitani, 2006).

Rare earths are produced mainly from the Bayan Obo deposit of Inner Mongolia, an iron-rare earth-niobium ore deposit in which the rare earths are recovered as a by-product of iron ore mining and processing operations (Humphries, 2012; Kanazawa and Kamitani, 2006). The chief RE minerals in Bayan Obo are bastnaesite and monazite disseminated with hematite, magnetite and gangue minerals such as fluorite, barite and dolomite (Li and Yang, 2014). Overall resource utilisation is poor with respect to RE recovery (16.8% recovery - Qiu et al. (2016)) and is due to the complex mineralogy and textural characteristics of the ore, fine dissemination of mineral values and poor separability from gangue (Li and Yang, 2014). Analogous challenges have also been encountered in the recovery of rare earths from similar ores outside of China in which iron oxides are present as major gangue (Bisaka et al., 2016). Potential alternative avenues to beneficiate these ores involve the application of pyrometallurgical pre-treatment processes typically used in the beneficiation of refractory iron ores which are unresponsive to conventional mineral separation processes (Iwasaki and Prasad, 1989; Kwauk, 1979; Uwadiale, 1992).

Magnetizing roasting is a method for the beneficiation of refractory

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iron ores not amenable to conventional mineral beneficiation due to fine grain size, complex mineralogy and textures. The method involves roasting the ore in a reducing environment at 500-700 °C resulting in partial reduction of hematite to magnetite followed by application of low intensity magnetic separation (LIMS) to produce an iron-rich magnetic product and tailings (Iwasaki and Prasad, 1989). The reducing atmosphere is generated via reforming of hydrocarbons or by solid carbonaceous reductants such as coal (Iwasaki and Prasad, 1989; Uwadiale, 1992). Despite the energy penalty associated with roasting, a magnetizing roast can be advantageous as magnetic separation is simpler and more selective than other separation techniques (e.g. flotation), has reduced grinding costs due to the increased friability of the roasted ore, and generally results in improved settling and filtration characteristics of the ore due to dehydroxylation of clays during roasting (Iwasaki and Prasad, 1989; Uwadiale, 1992). A review on the magnetizing reduction of iron ores by Uwadiale (1992), cites many examples where this method has been successfully applied in the beneficiation of fine-grained iron ores.

When magnetizing roasting is applied to ores containing other valuable minerals in addition to iron, the result is a tailings enriched in the other valuables which can be concentrated further by conventional methods such as froth flotation. For example, the issues encountered in the beneficiation of refractory iron ores such as fine grain size, complex mineralogy and textural characteristics are also pertinent to RE bearing ores such as RE-enriched lateritic deposits and iron oxide-copper-gold-RE style deposits where the iron oxides are present as major gangue (Faris et al., 2017). Magnetizing roasting has been successfully applied for the treatment of Bayan Obo iron-RE ore resulting in the recovery of an iron enriched magnetic product and tailings enriched in rare earths after magnetic separation. Kwauk (1979) studied the fluidized bed reduction roasting of an iron-RE ore from the Bayan Obo deposit at 550 °C using a mixture of CO and H₂ gas. Application of magnetic separation after roasting produced an iron concentrate grading 62.2 wt% Fe_T starting from a feed grading 36 wt% Fe_T at a recovery of 94%. The rare earths present in the feed reported to the non-magnetic product at a recovery of 80%. Yang et al. (2013) studied the magnetizing roasting of iron-rich rare earth bearing tailings using coal as the reductant. Ore roasted under optimal roasting conditions of 650 °C, roasting time of 2.5 h and a C/O molar ratio of 3.85 was subjected to low intensity magnetic separation resulting in a Fe_T grade of 45 wt% and a recovery of 68% starting from a feed containing 13.1 wt% Fe_T. The rare earths were recovered in the tailings (80% recovery) with a slight increase in grade to 7.5 wt% REO_T relative to the feed which had a grade of 7.1 wt % REO_T.

The removal of iron through reduction roasting and magnetic

separation ahead of froth flotation for recovery of RE minerals offers a number of potential downstream advantages. The typical collectors used in the flotation of RE minerals such as fatty acids and hydroxamates (Jordens et al., 2013; Pradip and Fuerstenau, 1991) also tend to float iron oxide minerals (Fuerstenau et al., 1970) which would lead to poor recovery and lower grades of rare earth concentrates; with high collector and depressant consumption. The removal of iron oxides ahead of RE mineral flotation could result in improved REO grade and recovery to the concentrates, reduced consumption of flotation reagents, and simplified reagent addition schemes.

In this paper, the factors influencing magnetite formation during the reduction roasting of a refractory goethite-RE ore sourced from a lateritic deposit was explored. The ore feed was characterized by chemical analysis (XRF and ICP-OES), quantitative X-ray diffraction (QXRD) and thermogravimetric analysis (TGA). Characterization of the reduced products was carried out by powder X-ray diffraction, QXRD and scanning electron microscopy (SEM) to investigate the mineralogical changes taking place after reduction roasting. This study also looked at the effects of magnetizing roasting on the stability of the RE and gangue minerals and their potential implications.

2. Materials and methods

2.1. Materials

The material used in this study was an iron-rich RE containing ore sourced from a lateritic deposit. The as-received ore was crushed to a P_{80} of 1.18 mm and was blended and riffled to produce representative sub-samples for chemical analysis, QXRD and for roasting experiments.

The reductant used in this study was a sub-bituminous coal of noncoking nature from the Collie Basin, Western Australia. The moisture, ash and volatile matter content of the coal were determined according to ASTM methods D3173, D3174 and D3175 respectively (ASTM D3173/D3173M-17, 2017; D3174-12, 2012; D3175-17, 2017). The fixed carbon content of the coal was calculated by difference and the proximate analysis of the coal used in this study gave a moisture content of 14 wt%, 31 wt% volatile matter, 50 wt% fixed carbon and 5 wt% ash. The coal was screened and the -2.36 mm +150 µm size fraction was used in most experiments. In latter experiments looking at the influence of coal particle size, the bottom size was varied from +150 µm up to +600 µm. The coal particle size was controlled to enable char separation after roasting when required.



Fig. 1. Experimental set-up for reduction roasting experiments.

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