



Application of ferrous pyrometallurgy to the beneficiation of rare earth bearing iron ores – A review



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ABSTRACT

This review is concerned with the application of ferrous pyrometallurgy to the beneficiation and comprehensive utilization of rare earth ores enriched in iron and other valuable metals such as niobium of which a large number of such deposits exist and are located in Australia, Brazil, China, Russia and USA. Due to the complex mineralogical and textural characteristics and very fine grain size of the minerals, these ores are refractory to conventional mineral beneficiation processes. Pyrometallurgical processes typically used in the beneficiation of fine grained iron ores may solve these problems. The largest deposit of this kind and the supplier to over 90% of the world's rare earths is the Bayan Obo Fe-REE-Nb deposit of Inner Mongolia, China. This deposit has had the most research conducted regarding pyrometallurgical processing as a means to beneficiate the contained iron and rare earth metal values. A detailed case study is presented of the research carried out to date on the application ferrous pyrometallurgy for the comprehensive utilization of Bayan Obo ore and the applicability of these processes to the beneficiation of similar ore deposits outside of China is considered.

1. Introduction

The rare earth elements (REEs) comprise a group of seventeen elements; fifteen lanthanides (lanthanum to lutetium) plus scandium and yttrium. Scandium and yttrium are included in the lanthanides group owing to their similar chemical and physical properties resulting in their common co-occurrence with the lanthanides in many ore deposits (Jaireth et al., 2014). 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 (nickel-metal hydride batteries), catalysts (petroleum cracking catalysts, catalytic converters), ceramics, phosphors, metal alloys (Krishnamurthy and Gupta, 2004; Lucas et al., 2014; Xie et al., 2014) and polishing powders (Humphries, 2012; Lucas et al., 2014). Over the past two decades, the global demand for REEs has increased significantly in line with their expansion into high-end technology, environment and economic areas (Hoatson et al., 2011).

Rare earths do not occur naturally as metallic elements but are present in a wide range of mineral types including oxides, carbonates, halides, phosphates and silicates (Table 1). Around 200 minerals are known to contain REEs although only a relatively small number are

commercially significant. In terms of mineral resources 95% of the total rare earths are contained in the three minerals; bastnasite [(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 reserves of rare earths in the world are approximately 130 million metric tonnes (oxide basis) with 50% of the reserves located in China (USGS, 2016). The balance of global rare earth resources are located in the CIS, Brazil, Vietnam, India and Australia. The current supply base of REEs is dominated by China (Jaireth et al., 2014), which controls over 90% of world rare earth production (Haque et al., 2014; Kanazawa and Kamitani, 2006).

Rare earth elements occur in a broad range of deposit types (Table 2) yet rarely are these deposits mined exclusively for their rare earth inventory; exceptions include Mountain Pass, Mount Weld and ion adsorption clay deposits in Jiangxi Province, China. In general, rare earths are usually recovered as concentrates produced as a by-product of other mineral processing operations. For example the Bayan Obo Fe-REE-Nb deposit is China's largest iron ore mine with iron ore reserves in excess of 1 billion metric tonnes (Krishnamurthy and Gupta, 2004). The deposit is also the world's largest resource of rare earths having an average rare earth oxide content of 6.19 wt% (Krishnamurthy and

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Table 1
A selection of commonly occurring REE-bearing minerals listed in order of estimated REO content.

Mineral ^a	Formula	Approximate REO _T wt%	Major deposit type
Bastnasite	REE(CO ₃)F	53–79	Carbonatite, Hydrothermal metasomatic
Monazite	(REE, Ca, Sr, Th)(P, Si)O ₄	38–71	Carbonatite, Hydrothermal metasomatic, Laterite, Placer, Fe-oxide Phosphate
Xenotime	(REE, Zr)(P, Si)O ₄	43–65	Granite pegmatite, Hydrothermal metasomatic, Laterite, Placer
Parisite	Ca(REE) ₂ (CO ₃) ₃ (F, OH) ₂	58–63	Carbonatite, Hydrothermal metasomatic
Fergusonite	REENbO ₄	43–52	Granite pegmatite, Hydrothermal metasomatic
Synchysite	CaREE(CO ₃) ₂ F	48–52	Carbonatite, Hydrothermal metasomatic of carbonatite and granite
Ba-REE fluorocarbonates ^b	Ba _x REE _y (CO ₃) _{x+y} F _y	22–40	Hydrothermal metasomatic, Carbonatite
Churchite	REEPO ₄ ·2H ₂ O	43–56	Laterite

^a Other RE minerals referred to in the text include: aeschynite, REE(Ti, Nb)₂O₆; fersmite, (Ca, REE, Na)(Nb, Ta, Ti)₂(O, OH, F)₆; allanite, (Ca, REE)₂(Al₂Fe²⁺)₃(SiO₄)₃(OH).

^b Includes huanghoite (x = y = 1), zhonghuacerite (x = 2, y = 1), and cebaitite (x = 3, y = 2).

Gupta, 2004) with reserves in excess of 100 million tonnes (oxide basis) (Smith et al., 2000). Although the deposit is high grade, rare earths are only recovered as a by-product of iron ore mining and processing operations (Humphries, 2012; Kanazawa and Kamitani, 2006). The rare earth beneficiation operation is influenced by the requirement to generate a high grade iron product (Castor and Hedrick, 2006; Li and Yang, 2014) and overall resource utilization is poor with respect to rare earth recovery (16.8% recovery - Qiu et al. (2016)). The rare earth recovery is exacerbated by a mineralogically and texturally complex ore with fine dissemination of mineral values and poor separability from gangue (Gao et al., 2010b; Li and Yang, 2014).

Besides Bayan Obo, a number of iron-rich rare earth deposits of similar complex mineral and textural associations (Table 3) are located in Australia (e.g. Mount Weld, Olympic Dam), Brazil (e.g. Araxá), USA (e.g. Pea Ridge, Mineville) and Russia (e.g. Chuktukon, Tomtor), and analogous challenges have been encountered in the recovery of rare earths from these deposits (Bisaka et al., 2016). One potential way to address the beneficiation of these ores is the application of pyrometallurgical pre-treatment processes as typically used in the upgrading of low grade iron ores (Iwasaki and Prasad, 1989; Kwauk, 1979; Uwadiae, 1992). The thermal pre-treatment step is used to convert iron oxides into a form which facilitates physical separation of iron from the rare earths. The rare earths and other valuables in the concentrated tailings can then be treated for recovery via mineral beneficiation or chemical leaching. Such processes have been extensively researched for the recovery of both iron and rare earths from the Bayan Obo deposit (Gao et al., 2010a,b; Ding et al., 2012) and more recently on a monazite-bearing magnetite ore from Korea (Kim et al., 2014) and iron-rich rare earth ores from South Africa (Bisaka et al., 2016).

The applicability of pyrometallurgical pre-treatment processes for beneficiating rare earth minerals from fine-grained Fe-REO ore types is reviewed. A technical assessment of pyrometallurgical processes and the stability of rare earths in these processes are presented. A critical analysis was conducted of previous research carried out on the application of ferrous pyrometallurgy for the treatment and recovery

of REEs from the world's largest fine-grained Fe-REO deposit, Bayan Obo in China. Finally, the applicability of such processes to the recovery of rare earths and iron from similar deposits outside of China is considered.

2. Pyrometallurgical processing schemes for low grade iron ore

Pyrometallurgical processes previously applied to the beneficiation of low grade iron ores fall into one of three general categories; magnetizing roasting, direct reduction and smelting processes (for the purposes of the current review it is assumed separation of REO is required). Common to all three processing schemes is the application of a high temperature pre-treatment step that converts iron oxides to a form amenable to physical separation from REO contained in the ore, be it via magnetic separation or density difference. The processing schemes are illustrated in Fig. 1.

2.1. Magnetizing roasting

Magnetizing roasting (Fig. 1a) involves roasting the ore in a reducing environment at 600–700 °C resulting in an iron-rich magnetic product (magnetite, Fe₃O₄ or maghemite, γ-Fe₂O₃) and tailings enriched in the other valuables (rare earths for example). The reducing atmosphere is generated via reforming of hydrocarbons or by solid reductants such as coal (Iwasaki and Prasad, 1989; Uwadiae, 1992). For ores in which the principle iron-bearing mineral is siderite (FeCO₃), an oxidizing roast is employed to convert siderite to magnetite due to auto-oxidation of intermediate wüstite (Fe_{1-x}O) by CO₂ (Kwauk, 1979; Uwadiae, 1992). The roasted ore is ground and subjected to low intensity magnetic separation (LIMS) to remove magnetite (Iwasaki and Prasad, 1989). 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

Table 2
Types and examples of rare earth deposits, rare earth mineralogy and associated gangue present. Adapted from (BGS, 2011) and (Hoatson et al., 2011).

Deposit type	Example deposit	REE mineralogy	Gangue mineralogy
Carbonatite	Mountain Pass, USA	Bastnasite	Calcite, dolomite, barite
Associated with alkaline igneous rocks	Thor Lake, Canada ^a	Zircon, fergusonite, allanite, monazite, bastnasite, synchysite	Quartz, K-feldspar, plagioclase, biotite, Fe-oxides
Iron oxide-copper-gold	Olympic Dam, South Australia	Bastnasite, florencite, monazite, xenotime	Hematite, quartz, copper sulphides, fluorite, barite, siderite
Hydrothermal Metasomatic	Nolans Bore, Northern Territory, Australia	Monazite, cheralite, apatite, bastnasite, allanite	Apatite, kaolinite, quartz, calcium silicates
Heavy mineral sands (Placer)	Eneabba, Western Australia	Monazite, xenotime	Zircon, rutile, ilmenite
Laterites – associated with carbonatites	Mt Weld, Western Australia	Monazite, crandallite, florencite, rhabdophane, cerianite, churchite	Goethite, hematite, apatite, dolomite, clays, silicates
Ion adsorption clays	Guangdong, Southern China	REE cations adsorbed onto clays such as kaolinite	–

^a Reference Krishnamurthy and Gupta (2004).

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