

Investigations on mineralogical characteristics of rare earth minerals in Bayan Obo tailings during the roasting process

ZHENG Qiang (郑强), WU Wenyuan (吴文远)*, BIAN Xue (边雪)

(School of Metallurgy, Northeastern University, Shenyang 110819, China)

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Abstract: This paper focused on the investigation of the mineralogical characteristics of rare earth minerals from Bayan Obo tailings during the roasting process in the presence of coal, $\text{Ca}(\text{OH})_2$ and NaOH . Roasting experiments and leaching experiments were carried out to study the decomposition of rare earth minerals. The results indicated that bastnaesite and monazite could be completely decomposed at $650\text{ }^\circ\text{C}$ and the leaching ratio of rare earths could reach 89.78%. The reaction mechanisms of bastnaesite and monazite at $650\text{ }^\circ\text{C}$ were analyzed. For bastnaesite, both the outer layer decomposition and inner core decomposition occurred simultaneously during the roasting. However, monazite was decomposed in a spatial sequence starting from the outer layer and proceeding to the inner core.

Keywords: mineralogical characteristics; rare earths; Bayan Obo tailings; roasting; reaction mechanism

China is the global leader in the rare earth (RE) industry and exploits abundant RE minerals, providing over 95% of the world's RE output^[1]. However, with the large demand and rapid development of RE elements, the issues of resource management and environmental protection have become more prominent^[2,3]. It is generally known that resource management and environmental protection are the basis for the sustainable development of human society. Therefore, some new technologies and mineral resources have been developed to effectively and comprehensively recover the RE resources^[4-6]. The Bayan Obo tailings are an important secondary resource containing various valuable resources such as iron, RE and fluorine and produced using the processes of low-intensity magnetic separation, high-intensity magnetic separation and flotation^[7]. However, a total of 160 million tons of rare earth tailings have been discarded into the tailing dam, resulting in a low utilization ratio of valuable resources^[8-11]. Many complicated factors such as the low grade, fine-grained dissemination and complex mineral composition make it difficult to recycle these resources^[12].

Aiming at comprehensive recovery of these valuable resources, many researchers have devoted their efforts to the investigations of novel technologies for tailing sutilization. Prior to 2000, physical dressing processing was the primary technology applied to the tailings, for example, inverse flotation-direct flotation^[13] and combined process of gravity separation and flotation^[14]. These technologies could result in the enrichment of the rare earths and iron, but the recovery rate of rare earths was

low. Then, coal-based reduction was applied to smelt the tailings. The basic theory is that the hematite in the tailings could be transformed into magnetite or iron by reduction roasting, and then magnetite or iron was separated by low-intensity magnetic separation^[15-17]. However, the direct reduction roasting process only focused on the recovery of iron, ignoring the decomposition of rare earth minerals composed of bastnaesite and monazite. During reduction roasting, the decomposition of bastnaesite would release fluorine-containing gas that can pollute the environment. Subsequently, the carbochlorination roasting^[18-20] methods were investigated for the recycling of the rare earths from the tailings. The fluoride-containing gas could be controlled to a certain degree by adding the defluorination agents. However, these techniques were not widely used due to the large equipment requirement and high cost. Recently, the Chinese government has begun to pay more attention to the comprehensive utilization of the tailings. Reduction roasting with CaO ^[21-23] was investigated for the recovery of rare earths and iron. In the temperature range of $600\text{--}1200\text{ }^\circ\text{C}$, rare earth minerals could be effectively decomposed and the extraction ratio of rare earths was high. Considering the above mentioned results and previous study on the roasting decomposition of mixed rare earth concentrate^[24-30], the improved roasting method was investigated by adding coal, $\text{Ca}(\text{OH})_2$ and NaOH . Carbon as a reducing agent was used to reduce hematite to magnetite. The use of $\text{Ca}(\text{OH})_2$ and NaOH as additives aids in the decomposition process of rare earths, as well as in the incorporation of fluorine into the products. Moreover,

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* **Corresponding author:** WU Wenyuan (E-mail: wuw_y_neu@163.com; Tel.: +86-24-83680527)

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magnetite can be separated by low-intensity magnetic separation, and rare earths can be enriched in the magnetic tailings.

To date, few studies have focused on the solid-state roasting mechanism of rare earth minerals. Therefore, elucidating the mineralogical characteristics during the roasting process is necessary for the comprehensive utilization of the tailings. The emphasis of this paper was on the phase transitions and reaction mechanism of rare earth minerals mixed with coal, $\text{Ca}(\text{OH})_2$ and NaOH . Together with a forthcoming study, the results of this would provide a basis for the novel approach for the comprehensive utilization of Bayan Obo tailings.

1 Experimental

1.1 Raw materials

The tailings used in this work were produced from Bayan Obo during a low intensity magnetic separation process. The results of the chemical analysis of the tailings showed that the REO is 9.45 wt.% as shown in Table 1. The X-ray diffraction analysis of the tailings found that REs occur in the form of bastnaesite and monazite, as shown in Fig. 1. The electron photomicrograph of tailings is shown in Fig. 2. Bastnaesite and monazite disperse as single particles in the tailings, and the grain size of the rare earth minerals varies in the range of 10–50 μm .

The coal used as reductant in this study was obtained from Shanxi Province in China. The coal was crushed to 100% passing 2 mm using a laboratory roller crusher, and more than 80% of coal was in the size range of $>74 \mu\text{m}$. The coal was composed of 85.10 wt.% fixed carbon, 14.42 wt.% volatiles, 0.48 wt.% ash and 3.28 mass% moisture. The $\text{Ca}(\text{OH})_2$ and NaOH used in the experiments were of analytical reagent grade.

Table 1 Main chemical composition of tailings (wt.%)

Ca	F	Fe	REO	Si	Ba	Mn
18.63	15.03	14.10	9.45	5.87	3.24	2.15
Mg	Al	S	P	Na	K	Ti
1.81	1.75	1.70	1.46	0.96	0.39	0.35

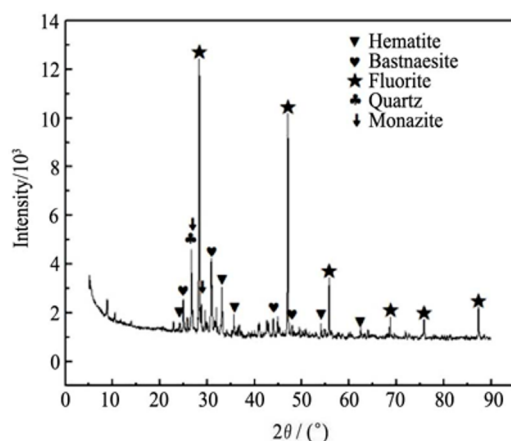


Fig. 1 XRD pattern of tailings

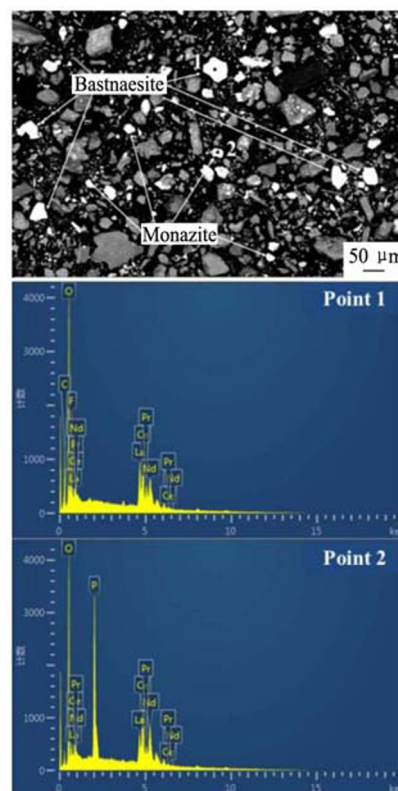


Fig. 2 SEM and EDS images of tailings

1.2 Experimental procedure

Before submitting to roasting experiments, the mixtures of tailings, coal, $\text{Ca}(\text{OH})_2$ and NaOH were fully mixed at a proper ratio. The coal dosages were calculated based on the mole ratio of the fixed carbon to removable oxygen or the iron oxides.

Roasting experiments were performed in a muffle furnace. The mixtures of tailings, coal, $\text{Ca}(\text{OH})_2$ and NaOH (200 g) were put in a graphite crucible of 500 mm in diameter and 600 mm in height, then the graphite crucible was put into muffle furnace with an automatic temperature control system and held for scheduled time when the temperature arrived at the predetermined temperature. In order to cool the roasted samples quickly and avoid the re-oxidization of magnetite, the roasted samples were put in cold water immediately after roasting and cooled down to room temperature. After dried for 4 h in an electric vacuum drying oven at 60 $^{\circ}\text{C}$, the roasted samples and raw tailings were ground and wet separated in a magnetic tube with magnetic field intensity of 0.15 T.

The obtained tailings of magnetic separation were used for leaching experiments. The leaching experiments were operated at room temperature, with the liquid-to-solid ratio of 15 mL/g, hydrochloric acid of 1.50 mol/L and stirring speed of 300 r/min for 60 min.

1.3 Analysis and characterization

The phase changes in the mixtures were identified by X-ray diffraction analysis (XRD, X'Pert Pro, MRDDY2105, PANalytical, Netherlands). A scanning

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