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Recovery of gallium from Bayer red mud through acidic-leaching-ion-exchange process under normal atmospheric pressure

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ABSTRACT

The Bayer red mud (BRM), generally containing 0.002 wt%–0.008 wt% of gallium (Ga), is an overlooked resource of Ga. In this work, an efficient method, called acidic-leaching-ion-exchange process (ALIEP), was developed to extract Ga from BRM. The ALIEP method involved three main steps: the BRM sample was firstly dissolved by mineral acid and the obtained leaching solution was further purified to remove the Fe^{3+}. Consequently, the purified solution, pre-concentrated by re-circulation process, was efficiently treated by ion-exchange process for the Ga recovery. The main influencing factors of the acidic-leaching process were systematically investigated. The optimal Ga leaching conditions were determined as HCl 159 g/L, liquid-to-solid ratio of 8 mL/g, 55 °C, and 5 h, attaining Ga leaching rate of 94.77% and the corresponding Ga^{3+} concentration of 3.91 mg/L in leachate. A nearly complete iron removal from the leachate was achieved by employing LSD-396 resin under conditions of 45 ± 2 °C, resin dosage of 0.6 g/mL and 2 h. Subsequently, the recovery efficiency of Ga from re-circulation leaching solution using the ion-exchange technology was evaluated. The results indicated that an adsorption rate of 59.84% and desorption rate of 95.32% for Ga were obtained. The concentrated solution contained 97.54 mg/L of Ga, which was enriched 24.95 and 4.75 times compared to the initial leaching solution of 3.91 mg/L and re-circulation solution of 20.52 mg/L, respectively.

1. Introduction

The Bayer red mud (BRM), a highly alkaline solid residue, is produced during the Bayer process for extraction of alumina from bauxite ores (Zhu et al., 2012). Approximately 0.6–2.5 t of BRM is generated per ton of alumina from the process, depending on the original properties of bauxite and operating conditions (Li et al., 2016). With the increasing alumina demand worldwide, the accumulated red mud was estimated to be 4 billion tons in 2015 (Zhu et al., 2015).

The disposal of BRM poses a serious environmental problem due to the corresponding highly pH nature (10–12.5) and complex chemical composition. Currently, most of raw red mud is directly stored in high-sized holding ponds without treatment (Sahu et al., 2010; Gomes et al., 2016). In contrast, the residue is regarded as a “polymetallic raw material” or an “artificial ore” because it is enriched in alumina, soda, silica, iron, calcium oxide and rare metals (Qu et al., 2013, 2015; Liu and Naidu, 2014). To alleviate the storage pressure increase of BRM, extensive research efforts have been devoted to the development of processes for utilization of the waste in the past several decades. Particularly, the extraction of valuable elements from the residue has received much interest (Balomenos et al., 2011; Lindsay, 2011; Samouhos et al., 2013; Gladyshev et al., 2015). The recovery techniques of the base metals from the residue, such as alumina, soda, ferric and titanium oxides, have been studied by many researchers (Agatzini-Leonardou et al., 2008; Bruckard et al., 2013; Ghosh et al., 2011; Zhang et al., 2011; Liu et al., 2012a,b; Huang et al., 2016); however, only a few techniques have been applied in commercial scale production (Balomenos et al., 2011). Also, certain special metals have been recycled as secondary raw materials, including boron dioxide (Cengeloglu et al., 2007), scandium (Tsakanika et al., 2004; Wang et al., 2013), and other rare-earth elements (Borra et al., 2015; Borra et al., 2016). Apart from the aforementioned valuable metals, Ga has also been discovered in BRM with its content ranging from 0.002 wt% to 0.008 wt% (Liu and Li, 2015). In the Bayer process, approximately 70% of Ga occurring as Ga_2O_3 in bauxite is dissolved out and accumulates in the Bayer liquor, which produces approximately 90% of the worldwide Ga quantities.

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The remaining 30% of undissolved Ga in bauxite is left in the BRM (Figueiredo et al., 2002). Ga metal is extensively employed in high-tech industries as it provides the benefits of low energy consumption and high computational speeds (Swain et al., 2015). Accordingly, Ga production has been continuously increasing and is estimated to increase 20-fold by the year 2030 compared to the 273 metric tons during 2012 (Alonso et al., 2012; Frenzel et al., 2016). It is worth mentioning that the Ga-bearing host minerals are quite scarce, consequently a large amount of research has been done on the potential Ga resources. Unfortunately, few efficient investigations have been conducted to extract Ga from the BRM, except a recent report from Abdulvaliyev et al. (2015). The researchers treated red mud by using Bayer-hydrogarnet process. In this process, the red mud sample contained 0.0025 wt% Ga was leached using a 240 g/L Na_2O solution in the presence of lime for 1.5 h at 240 °C under 20 atm with a liquid-to-solid ratio of 6 (abbreviated as L/S ratio hereafter). The leaching efficiency of Ga by this method was approximately 58%. The leaching solution was then treated by precipitation, followed by carbonation process to generate a concentrate (0.32 wt% Ga), which needed further treatment to recover Ga. In this work, an efficient method, called acidic-leaching-ion-exchange process (ALIEP), was developed by our group to recover Ga from the residue. The method mainly consisted of three steps: the BRM sample was firstly dissolved by a mineral acid under normal atmospheric pressure and the Ga was transformed into the liquid phase, followed by the removal of iron from the filtered leached solution. Subsequently, the purified solution, pre-enriched by circulation process, was efficiently treated by the ion-exchange process to concentrate Ga. The main benefits of this process are high extraction efficiency, simple procedure, and excellent Ga selectivity of resin. The aim of this work was to evaluate the Ga recovery from the BRM using the ALIEP method. The main factors influencing acidic-leaching of Ga from the BRM were investigated in detail. Also, Ga concentrations from the leaching solution by the ion-exchange method were performed.

2. Experimental

2.1. Materials and analysis

2.1.1. Major chemical compositions of BRM sample

The BRM utilized in this study was collected from a local alumina refinery in Guizhou Province of China. The sample was first dried in an oven at 105 °C for 48 h, then crushed into a fine powder and stored in a desiccator. From the inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7500a, US) analysis, the Ga of 0.0033 wt% was detected in the sample. The results are presented in Table 1.

2.1.2. Morphological characteristics and particle size distribution of BRM sample

The morphological characteristics of the BRM sample were observed by a scanning electron microscope (SEM, JSM-6700F, Japan) (Fig. 1), and the corresponding particle size distribution was analyzed by a laser particle size distribution analyzer (SEISHIN LMS-30, Japan) (Table 2). As presented in Fig. 1 and Table 2, the fines appeared as crystalline aggregates and some large-sized crystals of various shapes and sizes; the particle sizes of the sample ranged from 0.315 μm to 1.991 μm. The mean particle size was only 0.889 μm. The fine particles contribute to the increase in leaching efficiency increase for all metals.

Table 1
Major chemical compositions of BRM sample (wt%).

Al_2O_3	SiO_2	Fe_2O_3	CaO	Na_2O	TiO_2	Ga	V	LOI
20.73	17.19	20.74	15.85	6.39	5.29	0.0033	0.03387	10.74

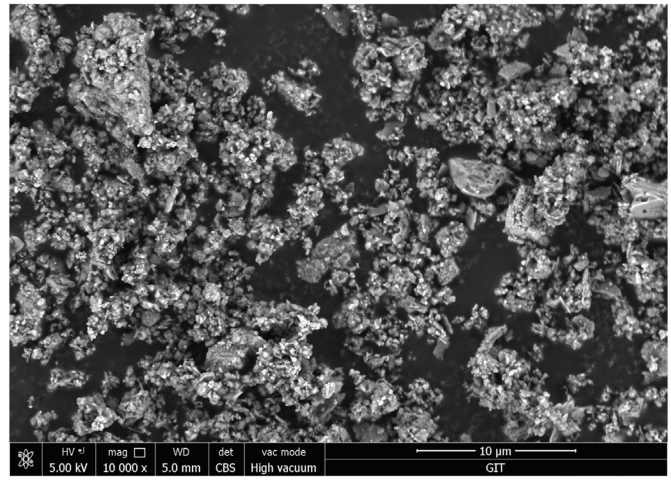

Fig. 1. SEM image of raw BRM.

2.1.3. X-ray diffraction measurement of BRM sample

The X-ray diffraction (XRD) pattern of the BRM sample was determined using a diffractometer (XRD, XRD-6100, Japan) and the result is presented in Fig. 2. The residue mainly consisted of hematite (Fe_2O_3, PDF #89-0596), katoite ($Ca_3(Fe_{0.87}Al_{0.13})_2(SiO_4)_{1.65}(OH)_{5.4}$, PDF #87-1971), cancrinite [$Na_8(Si_6Al_6O_{24})(H_{0.88}(CO_3)_{1.44})(H_2O_2)$], PDF #77-1145) and kaolinite ($Al_2Si_2O_5(OH)_4$, PDF #72-2300). Aluminosilicates were found to be the primary phases in the sample. The Ga-bearing mineral was not observed, likely because it was below the detection limit.

2.2. Methods and procedure

2.2.1. Leaching experiments

The acid leaching experiments were performed in a 1000 mL glass reactor equipped with a mechanical stirrer. The stirrer was equipped with a digital controller unit and a thermostat for controlling the reaction temperature with an accuracy of ± 0.5 °C. In addition, the reactor was installed with a reflux cooler to prevent the mass from evaporation. A specific amount of acid was initially slowly added into distilled water in the reactor according to a predetermined L/S ratio. When the dilute acid solution was heated up to the required temperature, a 50 g BRM sample (dry weight, precision 0.1 mg) was introduced into the reactor and the mixture was agitated well at 300 r/min for a specific duration. The leaching slurry was filtered, and the produced leached residue and solution were chemically analyzed. The extracted amounts of Ga and Al were calculated from chemical analysis of the leached residue.

The effects of acid concentration, leaching temperature, leaching duration and L/S ratio on the leaching efficiency of Ga were studied to determine the optimum conditions for Ga extraction. The leaching behavior of Al was also analyzed due to the geochemical affinity between the two elements (Zhao et al., 2012). From this analysis, the percentage extraction of Ga (or Al) was calculated by Eq. (1) as follows:

$$\eta(\%) = 1\text{-}\frac{(\mathrm{Me})_{\mathrm{RE}} \times \mathrm{M}_{\mathrm{RE}}}{(\mathrm{Me})_{\mathrm{RM}} \times \mathrm{M}_{\mathrm{RM}}} \times 100\% \tag{1}$$

where, η (%) is the leaching rate, $(Me)_{RE}$ and $(Me)_{RM}$ are the contents of metal in the leached residue and the BRM sample (wt%); M_{RE} and M_{RM} are the masses of the dried final leached residue and the BRM sample (g), respectively.

2.2.2. Iron removal experiments

For eliminating the interference of Fe^{3+} with Ga extraction by resin adsorption, the leaching solution was treated to remove iron prior to Ga recovery. LSD-396 is a chlorinated polystyrene macroporous resin,

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