

Process optimization of rare earth and aluminum leaching from weathered crust elution-deposited rare earth ore with compound ammonium salts

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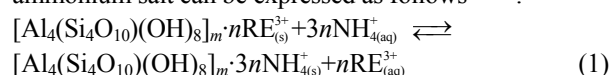
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Abstract: In order to intensify the leaching process of rare earth (RE) and reduce the impurities in the leachate, ammonium chloride (NH₄Cl) and ammonium nitrate (NH₄NO₃) were mixed as a compound leaching agent to treat the weathered crust elution-deposited RE ore. Effects of molar ratio of NH₄Cl and NH₄NO₃, ammonium (NH₄⁺) concentration, leaching agent pH and flow rate on the leaching process of RE were studied and evaluated by the chromatographic plate theory. Leaching process of the main impurity aluminum (Al) was also discussed in detail. Results showed that a higher initial ammonium concentration in a certain range could enhance the mass transfer process of RE and Al by providing a driving force to overcome the resistance of diffusion. pH almost had no effects on the mass transfer efficiency of RE and Al in the range of 4 to 8. The relationship between the flow rate and height equivalent to a theoretical plate (HETP) could fit well with the Van Deemter equation, and the flow rate at the lowest HETP was determined. The optimum conditions of column leaching for RE and Al were 1:1 (molar ratio) of NH₄Cl and NH₄NO₃, 0.2 mol/L of ammonium concentration, pH 4–8 of leaching agent and 0.5 mL/min of flow rate. Under this condition, the mass transfer efficiency of RE was improved, but no change was observed for Al compared with the most widely used ammonium sulfate. Moreover, the significant difference value (around 20 mL) of retention volume at the peak concentration between RE and Al provided a possibility for their separation. It suggested the potential application of the novel compound leaching agent (NH₄Cl/NH₄NO₃). It was found that the relative concentration of RE in the leachate could be easily obtained by monitoring the pH of leachate.

Keywords: weathered crust elution-deposited rare earth ore; aluminum; leaching process; mass transfer; ammonium chloride; ammonium nitrate

With the rapid development of advanced technologies, there is an ever-increasing demand for rare earth (RE) elements in the international markets, especially for the mid-heavy rare earth^[1,2]. The weathered crust elution-deposited RE ores, also named ion-adsorption RE ore, is the main source of mid-heavy RE in the world^[3]. In a warm and humid climate, original rocks contained RE are weathered and converted to clay minerals by biological, chemical and physical effects. During this weathering process, RE minerals are dissociated to be hydrated or hydroxyl hydrated RE ions and further adsorbed by the clay minerals with the migration of natural water^[4,5]. Due to the metallogenic climatic condition, the weathered crust elution-deposited RE ores are mainly distributed in southern China, such as Jiangxi, Guangdong, Fujian, Hunan, Yunnan, Guangxi and Zhejiang^[6]. The specific occurrence state of RE, ion-exchangeable state, decides its unique beneficiation technology^[7]. RE can be easily exchanged from the weathered crust elution-deposited RE ore by monovalent cations and transferred into solution as soluble RE sulfates, chlorides, or

nitrate^[8,9]. The main clay minerals in this RE ore is aluminosilicate, which can be described as [Al₄(Si₄O₁₀)(OH)₈]_m. The ion-exchange reaction with ammonium salt can be expressed as follows^[10,11]:



where *s* and *aq* represent solid phase and aqueous phase, respectively.

It shows that the clay minerals adsorbing RE can be seen as a natural ion exchange resin. The leaching process can be described as a chromatographic column elution process, in which the RE ore is a stationary phase and the leaching agent is a mobile phase^[12]. When the leaching agent flows through the RE ore, the cations (NH₄⁺) in the leaching agent diffuse from solution to the ore particles, and then the RE ions adsorbed on the ore particles are substituted and transferred into the solution^[13]. The ion-exchange process between the RE and NH₄⁺ is a typical mass transfer process, which can be evaluated by chromatographic plate theory^[14].

The study on the intensification of RE leaching proc-

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ess can not only increase the leaching efficiency of RE and reduce the consumption of leaching agent, but also improve the production efficiency and shorten production cycle^[13,14]. During the past years, much effort has been engaged in the development of the leaching techniques^[15] and the improvement of the RE leaching efficiency^[16]. However, limited investigations on the intensification of RE leaching process are reported. Although Tian et al.^[17] introduced sesbania gum as a filter-aid reagent into ammonium sulfate solution and Qiu et al.^[18] applied magnetic field to enhance RE leaching process, the high cost limited the application in the actual production.

In this work, a new leaching agent was developed to optimize the leaching process of RE, which demonstrated potential possibility to replace the most widely used leaching agent ammonium sulfate^[19]. Previous study revealed that the exchangeability and permeability of ammonium chloride (NH₄Cl) and ammonium nitrate (NH₄NO₃) both surpassed ammonium sulfate (NH₄)₂SO₄^[20]. However, the practical application of NH₄Cl and NH₄NO₃ were limited by the high content of impurities in the leachate, which would increase the cost of impurities removal, decrease the purity of RE product and even destroy the crystal forming of carbonate RE precipitation^[21,22]. Our latest research found that the mixture of NH₄Cl and NH₄NO₃ could be used as a compound leaching agent, which not only could reduce obviously the leaching of impurities, but also could keep a high leaching efficiency of RE. The effects of molar ratio of NH₄Cl and NH₄NO₃, ammonium (NH₄⁺) concentration, leaching agent pH and flow rate on the mass transfer process of RE were studied and evaluated by chromatographic plate theory. Aluminum (Al) as the main impurity ion in the leachate was also discussed to reveal the leaching performance of impurities.

1 Experimental

1.1 Materials and characterization

The experimental RE ores sample was collected from Dingnan (DN) County in Jiangxi Province of southern China. The main chemical composition of the RE ore was analyzed by X-ray fluorescence (Axios advanced, Panalytical B.V.) and the result is listed in Table 1. It was shown that the RE ore contained 0.10 wt.% RE and 19.53 wt.% Al. Not all the RE and Al, but the exchangeable state RE and Al can be exchanged into solution by the cations. The contents of the exchangeable state RE and Al in the RE ore were 0.09 wt.% and 0.015 wt.%, respectively. The partitioning of the ion-exchangeable state RE determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7700x, Agilent Technologies Inc.) are shown in Fig. 1. 52.06 wt.% of RE was the middle and heavy RE, implying the tremendous

Table 1 Main chemical composition of the RE ores (wt.%)

Component	REO	Al ₂ O ₃	MnO ₂	ZnO	CaO	MgO	K ₂ O	SiO ₂	P ₂ O ₅
Content	0.10	19.53	0.01	0.01	0.02	0.52	3.54	58.09	0.20
Component	SO ₃	TiO ₂	Fe ₂ O ₃	Rb ₂ O	SrO	ZrO ₂	BaO	Loss	
Content	0.04	1.40	8.07	0.02	0.01	0.02	0.04	8.36	

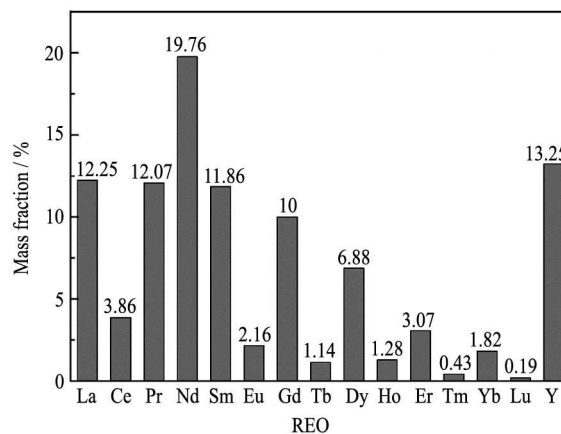


Fig. 1 Partitioning of the ion-exchangeable RE in the RE ores

commercial value.

1.2 Column leaching experiments

The experiments were conducted at ambient temperature (25 °C) in a glass column with 45 mm inner diameter, which can be seen as a chromatographic column. 250 g of dried RE ore sample was uniformly packed in the glass column and the packed ore height was measured. The schematic diagram of experimental apparatus is shown in Fig. 2. The RE ore sample in the glass column was eluted by the leaching agent solution with a certain concentration and pH at a desired flow rate. Leachate was collected from the bottom of the leaching column and the concentrations of RE and Al were analyzed by

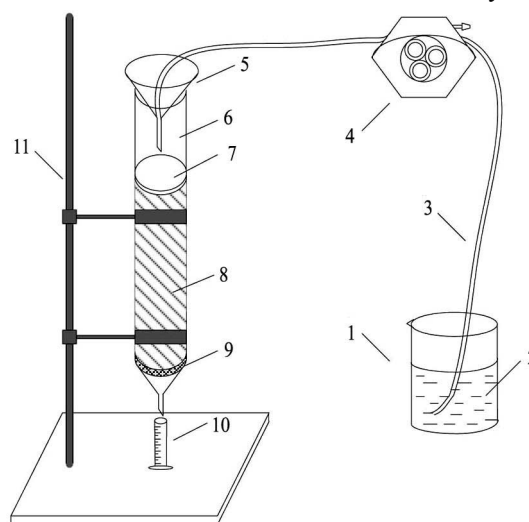


Fig. 2 Schematic diagram of experimental apparatus

1–Beaker; 2–Leaching agent; 3–Silicone tube; 4–Peristaltic pump; 5–Funnel; 6–Glass column; 7–Filter paper; 8–RE ore sample; 9–Sand core filter plate; 10–Precision measuring cylinder; 11–Iron support

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