



Recovery of magnesium and potassium from biotite by sulfuric acid leaching and alkali precipitation with ammonia



Zheng Luo^{a,c}, Jing Yang^{a,b,*}, Hongwen Ma^{a,b,*}, Meitang Liu^a, Xi Ma^a

^a School of Materials Science and Technology, China University of Geosciences, Beijing 100083, China

^b Beijing Key Laboratory of Materials Utilization of Nonmetallic Minerals and Solid Wastes, China University of Geosciences, Beijing 100083, China

^c Bluesky Technology Corporation, Beijing 100083, China

ARTICLE INFO

Article history:

Received 24 February 2015

Received in revised form 19 August 2015

Accepted 22 August 2015

Available online 29 August 2015

Keywords:

Biotite

Magnesium hydroxide

Ammonium–potassium sulfate

Recovery

ABSTRACT

A process for the recovery of magnesium (Mg) and potassium (K) from biotite was developed using sulfuric acid leaching and precipitation with ammonia. The optimum conditions for dissolution of biotite were found to be sulfuric acid solution of 2.5 mol/L, sulfuric acid/biotite ratio of 4 mL/g, leaching temperature of 90 °C and leaching time of 2 h. Under these conditions, the leaching efficiency of Mg and K was found to be 95.8% and 94.9% respectively. By using the solubility differences of corresponding elements, a two-step precipitation process was applied to remove Fe and Al and recovery of Mg. Subsequently, magnesium hydroxide was prepared by precipitation from the leach solution with ammonia as the precipitant. X-ray diffraction (XRD) and scanning electron microscope (SEM) analyses showed that the high purity Mg(OH)₂ existed in the form of flower-like spherical aggregation. Furthermore, the final solution of ammonium–potassium sulfate can be used for agricultural compound fertilizer.

© 2015 Elsevier B.V. All rights reserved.

1. Introduction

Magnesium hydroxide is a popular inorganic compound because of its wide range of applications. It plays an important role in many fields, such as flame retardant in polymers, special ceramics, fillers in bleaching agent and a raw material for preparation of magnesium oxide (Sonawane et al., 2010; Pavlović et al., 2010; Liu et al., 2011). Magnesium hydroxide can be prepared by several methods, such as hydration of magnesium oxide, electrolysis of magnesium salt aqueous solutions, sol–gel techniques and precipitation (Yu et al., 2004; Utamapanya et al., 1991). Magnesium is found in many minerals such as magnesite, dolomite, brucite, serpentinite (Tran et al., 2013). Elsner and Rathon (1998) extracted magnesium from a magnesite ore by leaching with HNO₃. This was followed by purification at pH 5–6 using NH₄OH and H₂O₂ and then precipitation to produce Mg(OH)₂. Nduagu et al. (2012) processed a serpentinite ore containing 21.8% Mg to prepare high purity Mg(OH)₂. Mixtures of the ore with ammonium sulfate at a mass ratio of 0.5–0.7 were roasted at 400–550 °C and Mg(OH)₂ was recovered at pH 10–11 using NH₃ gas after purification. Xiong et al. (2014) extracted magnesium from dolomitic phosphate ore (7.67% MgO) with dilute waste acid. Operating under the optimum leaching conditions, a 98.31% recovery of Mg with 0.02% P₂O₅ loss was obtained. Magnesium hydroxide was prepared by precipitation from the refined

solution with ammonia as the precipitant and sodium dodecyl sulfate as the modifier.

In many studies, magnesium could be recovered from tailings and waste residues. Ma et al. (2014a) treated boron mud with about 40% magnesia which is a potential magnesium source for synthesis of Mg(OH)₂ and proposed the formation mechanism of the flower-like Mg(OH)₂ via anti-drop precipitation method. In another study, Özdemir et al. (2009) used hydrochloric acid to recovery magnesium from magnesite tailings. MgCl₂ · 6H₂O with a purity of 91% was produced by evaporation of leach solution obtained at 40 °C for 60 min, 1.0 M acid, solid-to-liquid ratio of 10 g/L. Karidakis et al. (2005) investigated the recovery of magnesium from nickel laterite with sulphuric acid leaching followed by precipitation with Ca(OH)₂ and the precipitate was a mixture of Mg(OH)₂ and CaSO₄ · 2H₂O. The magnesium recovery rate of 90–99% could be achieved at only 20 °C for the precipitation.

Potassium (K) is an essential element for plant nutrition and potassium salts are mainly produced by water soluble potassium resources. However, China only possesses 210 million tons of the global potash reserves which is totally 6 billion tons around the world (US Geological Survey, 2014) and imports several million tons of potash fertilizer each year. Under such circumstances, the recovery of K from different indigenous sources is highly essential in China. The N–K nutrient balance was highly significant for dry matter yield and tiller development indicating that optimum yield was obtained only when both elements were adequately supplied (MacLeod, 1965; Grant and MacLean, 1966). Later studies by Opuwaribo and Odu (1978); Lumbanraja and

* Corresponding authors at: School of Materials Science and Technology, China University of Geosciences, Beijing 100083, China.

E-mail addresses: yjcl@cugb.edu.cn (J. Yang), mahw@cugb.edu.cn (H. Ma).

Evangelou (1990, 1992, 1994), and Shen et al. (1997) reported that K^+ , added simultaneously with NH_4^+ , stimulated N_2 -fixation in soils containing illite, vermiculite and mica.

In this context, some efforts have been made to recover potassium for obtaining soluble K from K-feldspar and biotite. Earlier in 1997, Varadachari developed a process for the production of potassium chloride from biotite. The kinetics of reaction of biotite with HCl was revealed that leaching of K^+ from biotite followed first-order kinetics and the key process is the replacement of K^+ by H^+ in the interlayer space. After purification, KCl was recovered using ethanol (Varadachari, 1992, 1997). Jena et al. (2014) observed that the potassium values present in nepheline syenite (5.4% K_2O) were unlocked through roasting with calcium chloride followed by water leaching. In this report, it is possible to recover ~99.6% K_2O value by calcination at 900 °C for 30 min. Ma et al. (2014b) reported that K-feldspar was easily transformed into kalsilite by removing 2/3 SiO_2 from the feldspar in the KOH- H_2O solution. Kalsilite was then decomposed with sulfuric acid and the nearly pure solution of potassium sulfate was obtained. During the process, the recovery ratio of K_2O was up to 94.0%.

Biotite ($K(Mg,Fe)_3AlSi_3O_{10}(OH)_2$) is a kind of layered aluminosilicate mineral with some valuable elements of magnesium and potassium. Compared with other kinds of mica, like phlogopite and lepidolite, which are of practical importance in industry, biotite, containing iron inside, is greatly restrained from practical use due to its poor insulation performance. As a result, biotite slice could only be used as cheap fillings of building materials. In Qinghai province of China, 45,000 t of biotite as a by-product of phosphate ore, containing ~20% MgO and 9–10% K_2O were produced while processing 300,000 t of phosphate ore per year. Therefore, it attaches a great practical importance on researching how to recover Mg and K by using the biotite. Preliminary investigations indicated that metal cations in biotite can be easily dissolved by inorganic acid (HCl and H_2SO_4), and hydrochloric acid was found to be the most effective in terms of rate of leaching efficiency (Varadachari, 1992, 1997). However, it is evident from the literature that Cl can be toxic to some plants and higher than adequate rates of K as KCl caused a yield reduction due to damage caused from an excess of Cl (Rominger et al., 1976). Therefore sulfuric acid was used to destroy biotite in this research.

In this paper, an innovative process to recover magnesium and potassium from the Qinghai biotite is proposed, which is suitable to deal with magnesium-rich biotite. It aims to optimize the conditions for magnesium and potassium recovery from biotite by leaching with sulfuric acid solution and to determine factors affecting its leaching efficiency. Various process parameters like the concentration of sulfuric acid, leaching time and temperature are investigated in terms of metal leaching efficiency. Then, a two-step precipitation process was applied to prepare $Mg(OH)_2$ and recovery of potassium from the leach solution using ammonia. Finally, the production of $Mg(OH)_2$ was also determined by XRD and SEM methods.

2. Materials and methods

2.1. Materials

The biotite mineral sample, which was produced from a beneficiation of phosphate ore, was from Shangzhuang county of Qinghai province (China). Sulfuric acid and ammonia used were of analytical reagent grade and all the solutions with specified concentrations were prepared with distilled water.

2.2. Experimental procedures

A planetary ball mill with a rotation speed of 200 rpm was employed for mechanical activation. Four milling cells were fixed on its platform, and each cell was composed of a 500-mL stainless steel vessel filled with 250 g of 2B mm steel balls, 200 g of $\Phi 10$ mm steel balls and 50 g

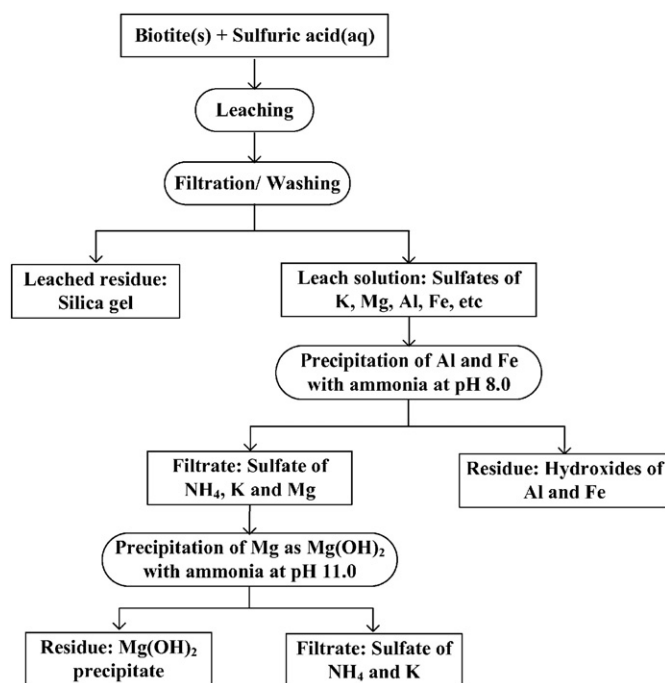


Fig. 1. Flowchart diagram showing process for the recovery of Mg and K from biotite.

of $\Phi 5$ mm steel balls. The milling was operated in the air with 50 g of mineral biotite per cell and a total of 200 g for 2 h. The average particle size (d_{80}) of biotite was 150 μm .

The flowchart for Mg and K recovery from biotite by acid leaching and alkali precipitation is shown in Fig. 1. The leaching experiments were performed in a glass 100-mL flask equipped with a magnetic stirrer with an agitation speed of 200 rpm and a reflux condenser to prevent evaporation loss, which was bathed in a water bath. Biotite (5 g) was reacted with different concentrations of sulfuric acid (1.85–2.7 mol/L), with different sulfuric acid/biotite mass ratios (3:1–7:1 mL/g), different leaching times (0.5–2.5 h) and different leaching temperatures (25, 60, 70, 80 and 90 °C). After the acid leaching, the mixture was filtered through 0.5- μm pore size white filter paper using a pressure filtration unit and washed to separate the leached residue and the solution. The Mg^{2+} , K^+ , Fe^{2+} , Fe^{3+} , and Al^{3+} contents in the solutions were determined by wet chemical analysis. A measured amount of hydrogen peroxide solution containing 30 wt.% H_2O_2 was added to the solution to oxidize Fe^{2+} to Fe^{3+} at room temperature for 30 min under stirring at 200 rpm. The acidic leaching solution was then adjusted to pH 8.0 with ammonia to precipitate Al and Fe, and retain K and Mg in solution.

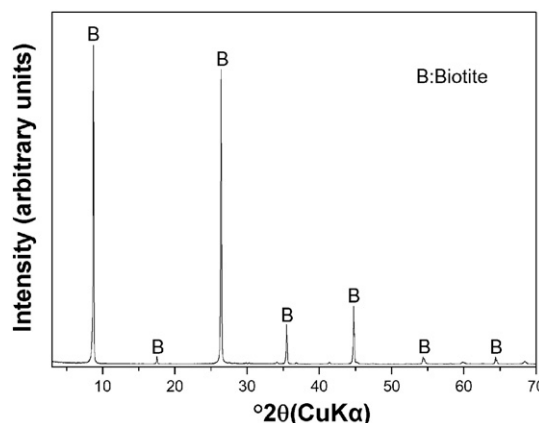


Fig. 2. XRD pattern of biotite mineral powder.

Download English Version:

<https://daneshyari.com/en/article/6659273>

Download Persian Version:

<https://daneshyari.com/article/6659273>

[Daneshyari.com](https://daneshyari.com)