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Using one waste to tackle another: Preparation of a CO_2 capture material zeolite X from laterite residue and bauxite

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HIGHLIGHTS

- Valuable zeolite X has been synthesized from laterite residue and bauxite.
- High product purity has been achieved by optimizing the process conditions.
- Prepared zeolite X shows comparable gas adsorption properties to commercial ones.
- Prepared zeolite X can be used for carbon capture by vacuum swing adsorption.

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ABSTRACT

In this work, zeolite X, a benchmark adsorbent for carbon capture, has been successfully prepared from low cost waste minerals namely laterite residue and bauxite using alkali fusion process followed by hydrothermal treatment. The structure and morphology of the as-synthesized zeolite X were verified and characterized with a range of experimental techniques such as X-ray diffraction, scanning electronic microscopy and infrared spectroscopy. The surface area and (N_2 and CO_2) gas adsorption isotherms of this product were found comparable to that of commercial ones, demonstrating the effectiveness of synthesizing zeolite X from laterite and bauxite. Further improvement of the product purity was also accomplished by optimizing the process conditions.

1. Introduction

Nickel is a strategic metal and widely used in stainless steel, batteries, fuel cells, electroplating, catalyst, and etc. [1–3]. Currently, the world's recoverable nickel resources can be divided into two main categories, respectively sulfide ores and laterite ores, depending on the geological compositions [4]. The global nickel supply has been predominantly from sulfide deposits, although laterite ores comprise approximately 70% of the continental nickel resources [5,6]. The reason is that the recovery process of nickel from the laterite ores is quite costly, because of the complex mineralogy and low nickel content of laterite [7]. Recently, however, the laterite exploitation has received extensive attentions, due to the reduction of nickel sulfide reserves as well as significant increase in nickel demands [8,9]. Therefore, there is an increasing interest in the utilization of laterite.

In general, nickel can be recovered from the laterite by using pyrometallurgical, hydrometallurgical or combined (pyro-hydrometallurgical) techniques [10,11]. Pyrometallurgical process comprises of reduction roasting, electric furnace for ferronickel production, and rotary kiln carbon reduction, etc. [12,13]. Nevertheless, the hydrometallurgical processes consist of ammonia leaching, acid leaching, high-pressure acid leaching and bacteria leaching, etc. [14,15]. However, these processes can generate large amount of solid waste, which causes serious environmental pollution. Large volumes, approximately 40 million tons, of laterite residue are generated annually in China alone [16]. Current metallurgical processes usually focus on the recovery of Ni and/or Co, whereas laterite residue containing other waste minerals are all discarded, which brings enormous burden to the environment [17]. Therefore, converting laterite residue into useful commodity could offer a number of benefits from both economic and environmental aspects. Nevertheless, to our best knowledge, little information

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is available in literature on the effective utilization of the laterite residue. Laterite residue consists essentially of SiO_2, Al_2O_3 and Fe_2O_3 [3], presenting a similar composition to that of natural zeolites, which makes it possible to use laterite residue as source materials in zeolites production. Zeolite X is one of the zeolites that have been widely used in adsorption, separation, ion exchange and catalysis, due to its large surface areas, molecular sieving ability and acidity [18,19]. Particularly, zeolite X is also a well-known adsorbent for CO_2 capture from flue gas streams owing to its high selectivity of CO_2 against N_2 [20,21]. In this paper, we report the synthesis of zeolite X using laterite residue as raw materials, in which another common mineral bauxite was added to the starting material to maintain an appropriate Si/Al ratio during the hydrothermal treatment, because otherwise the large ratio of Si/Al in the laterite residue would inhibit the formation of zeolite X during the hydrothermal reactions.

The ultimate aim of this work was to develop a new route to utilize the laterite residue for the production of zeolites that can be used for CO_2 capture from flue gas streams. To obtain high purity of zeolite X, the molar ratios of the components in the starting materials and during the hydrothermal treatment should be carefully controlled. The optimum synthesis procedure was established in this study, and the carbon capture potential of our as synthesized products was also investigated.

2. Experimental

2.1. Synthesis

Laterite residue and bauxite was obtained from Sichuan Province (China) and Indonesia, respectively. The chemical compositions of laterite residue, bauxite and prepared zeolite X were analyzed by XRF (ZSX100e, Rigaku) and described in Table 1 showing 39.1 wt.% Si and 1.2 wt.% Al in laterite residue and 8.7 wt.% Si and 31.5 wt.% Al in bauxite. For comparison purposes, commercial zeolite X beads were also purchased from Sinopharm Chemical Reagent Co., Ltd.

The synthesis procedure for zeolite X is summarized schematically in the flow diagram (Fig. 1) which involved two steps *viz.* fusion and hydrothermal treatment. In a typical procedure, 5 g of laterite residue was mixed with bauxite using different molar ratios of SiO_2/Al_2O_3 mole ratios such as 3, 3.2, 3.5, and 3.8. Then, NaOH was added into the resulting mixture to adjust the Na_2O/SiO_2 mole ratio to a desired value in the range of 1.4–2.6. The mixture was fused in a tube furnace at 600 °C for 2 h. Then, the resulting product was dissolved in water with various H_2O/Na_2O ratio (35, 40, 43, 48, 55, and 60 mol mol^{-1}), followed by aging of different time (0, 8, 12, 24, and 48 h). The mixture was transferred into an autoclave and heated at 100 °C for different hydrothermal reaction time (0.5, 3, 6, 12, and 24 h) under static conditions. After cooling to the room temperature, the resultant solid was filtered, washed three times with deionized water, and dried at 100 °C overnight. The synthesis conditions used with different samples are presented in Table 2 (Group A–E). Note that the optimal synthesis conditions determined from the experiments of Group A was carried on to the experiments of Group B, likewise finally to group E.

2.2. Measurement and characterization

The crystalline properties of the raw materials and the synthesized samples were examined by X-ray diffraction (XRD) using a Shimadzu X-ray diffractometer, with a scanning rate of 2° min^{-1} from 4° to 50°. Field Emission Scanning Electron Microscopy (FE-SEM) analysis was conducted by employing a Hitachi S-3400 N scanning electron microscope operated at 15 kV. All samples were platinum coated prior to measurement. BET surface areas were measured on an ASAP2020 analyzer (Micromeritics, USA) using N_2 as probe gases at 77 K in the relative pressure range of 0.05–0.25. CO_2 and N_2 adsorption isotherms were measured on ASAP2020. Prior to analysis, all the samples were degassed under vacuum at 330 °C for 8 h. Infrared spectroscopy was performed using an Agilent Cary 600 FTIR instrument, and samples were mixed with KBr using a mortar before the measurement.

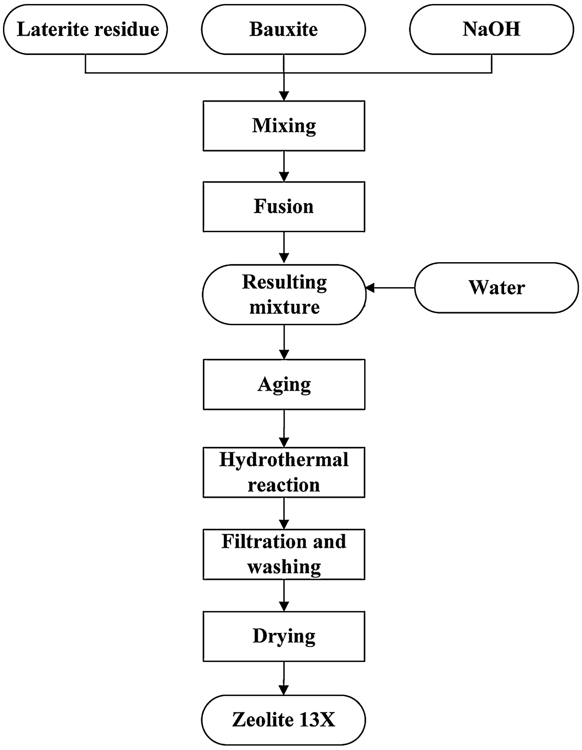

Fig. 1. Flowchart for synthesizing zeolite X from laterite residue and bauxite.

3. Results and discussion

3.1. Raw materials characterization

The X-ray diffraction pattern of laterite residue (Fig. 2) indicates that the crystalline phases are mainly SiO_2, $Mg_3[Si_2O_5(OH)_4]$, $Mg_3Si_4O_{10}(OH)_2$, and $Mg_{1.8}Fe_{0.2}[SiO_4]$, identified by their characteristic sharp peaks at the corresponding positions. The X-ray diffraction pattern for bauxite (Fig. 3) indicates the major crystalline phase is gibbsite, with coexistence of anatase, α-quartz, kaolinite, and hematite [22].

The chemical compositions of the raw materials (Table 2) by XRF show that they are impure in nature. The Si/Al ratio of the laterite residue and bauxite are about 29.034 and 0.248, respectively, indicating that laterite residue and bauxite are rich in silica and alumina, respectively, a favorable combination as raw materials for synthesizing zeolite X. Furthermore, XRF data reveals that the raw materials contain a very small amount of impurity, which suggests high purity of zeolites X could be obtained using laterite residue and bauxite.

3.2. Zeolite X synthesis and characterizations

3.2.1. Effect of Na_2O/SiO_2 mole ratios

The effect of Na_2O/SiO_2 mole ratios on zeolite X formation was determined by mixing NaOH, laterite residue and bauxite at the Na_2O/SiO_2 ratio range of 1.4–2.6 at 600 °C for 2 h. The XRD spectra