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Research paper

The synthesis and application of zeolitic material from fly ash by one-pot method at low temperature

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Abstract

A new route to prepare zeolitic material was introduced in this work. Compared with traditional methods, the new route showed lower energy consume. The effect of pre-treatment conditions on structure and crystalline phase was investigated, revealing that the mullite crystalline phase in fly ash could be converted to amorphous phase by alkali at low temperature. The removal performance of heavy metal ions on designed material was also investigated, and we found that the intermediate product showed higher adsorption capacity on Ni^{2+} than zeolite A.

Keywords: Fly ash; Zeolitic material; Removal of heavy metal; Intermediate

1. Introduction

Thermal electric power plants are responsible for generating a substantial amount of pollutants including solid waste, such as fly ash. World production of fly ash is over 750 million tons/year, whereas the fly ash from power plant has become the first solid waste in China. The total amount fly ash has reached 578 million in 2014 and the amount is increasing with the increase of coal-fired power plant scales. The large amount of fly ash are stored for piles instead of comprehensive utilization, which brings about severe environmental pollutions. Presently, the main use of fly ash is building materials and related application. For example, fly ash is used in the production of cement and concrete due to its pozzolanic properties. Obviously, such low value product is not an ideal product from the economic view. It is necessary to develop various economical viable technology routes to consume such pollution source.

The major components in fly ash are Al, Fe and Si, with smaller concentrations of Ca, K, Na, Ti and S [1,2], which is available source for zeolite synthesis. It has been found that zeolite has widespread applications in ion exchange, molecular sieve and adsorption due to their structural characteristics and valuable properties. Many synthesis methods of fly ash zeolite have been reported, such as hydrothermal process [3–6], salt-thermal [7], alkali fusion [8,9], microwave-assisted synthesis [10,11] and two-step process method [12,13]. For example, W. Franus et al. synthesized several types of zeolites with different channel system sizes, such as zeolite P, zeolite X and sodalite, according to different synthesis conditions (i.e., NaOH concentration and reaction temperature) [4]. M. Wdowin et al. synthesized the high-purity Na–P1 (zeolite content 81 wt%) from the coal fly ash in a very effective and efficient way, and the obtained material is mainly mesoporous (c.a. 61%) [14]. Each method has its advantages and limitations, for example, the alkali fusion method can prepare high purity zeolite with higher energy consume, while the hydrothermal

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process leads to lower energy consume but low purity. Anyway, the core mechanism of each method is to convert amorphous silica and aluminum in fly ash to zeolite crystalline structure, so releasing the amorphous silica and aluminum from fly ash is a crucial step.

The zeolite from fly ash has a higher value added and it has been used for a wide range of purposes, including agricultural, environmental and industrial application [12,15–17]. Among these application fields, using zeolitic material from fly ash as inexpensive adsorbents for wastewater treatment containing heavy metal has been proved to be an economically viable solution [18,19]. Hui et al. investigated the removal performance and the selectivity sequence of mixed heavy metal ions in aqueous solution on pure-form zeolite A prepared from fly ash, commercial grade zeolite A. They found that the equilibrium data matched with the Langmuir model and showed the affinity order: $Cu^{2+} > Cr^{3+} > Zn^{2+} > Co^{2+} > Ni^{2+}$ [18]. Wang et al. explored the possibility of utilizing the synthetic pure-form zeolites from fly ash to remove and recover heavy metal ions. Copper and zinc ions were chosen as target metal ions for the adsorption studies. The removal performance of heavy metal ions on both synthetic pure-form zeolites (A and X) were investigated and the influencing factors were studied [19]. L. Bandura et al. investigated the sorption performance towards BTX of zeolite Na–P1 and Na-X synthesized from fly ash, and the Na-X exhibited the greatest sorption capacity due to the major contribution of micropores in the mineral structure of faujasite [20]. Another work by L. Bandura found the synthetic zeolites obtained from fly ash were promising alternatives for natural mineral sorbents for land-based petroleum spills cleanup [21]. S. Chalupnik et al. suggested the method to remove radium isotopes and some other stable pollutants by mixture of zeolite with fly ash and sand [22], which proposed new ideas for the application of zeolite from fly ash.

The ultimate aim of this work is to develop a new route to synthesize zeolitic material with low energy consume, which can be used to remove heavy metal ions. Nickel was chosen as target metal ion for the adsorption studies. The removal performance of heavy metal ions on zeolitic material was investigated, meanwhile commerce zeolite A and fly ash were used as comparison. Moreover, an attempt was made to investigate the removal mechanism in order to develop industrial application.

2. Experiment

2.1. Preparation

Before any treatment, the sodium hydroxide was carefully ground into powder to ensure the fly ash contact with sodium hydroxide as complete as possible. Then, sodium hydroxide and fly ash were mixed together with pre-determined different ratios and transferred to autoclave with teflon bottle as liner. After 24–48 h at different temperatures from 50 to 110 °C for pretreatment process, deionized water was added to the mixture. The detailed pretreatment parameters were listed in Table 1. The slurry was stirred at room temperature for better mixing, a portion of slurry was filtered and washed as intermediate samples marked as II-x-Z (x = 1, 2, 3 …). After pretreatment and the other portion was transferred to autoclave and kept at 110 °C for 24 h for hydrothermal reaction. Afterwards the resultant products were washed by deionized water until being neutral, filtrated and dried in an oven at 110 °C overnight, the synthesized zeolite can be obtained, labeled as II-x (x = 1, 2, 3 …).

2.2. Characterization

X-ray diffraction (XRD) patterns were measured with a Rigaku Ultima X-ray diffractometer using Cu Kα radiation at 40 kV and 40 mA. The fourier transform infrared (FT-IR) spectra were collected on a Bruker Spectrometer using the KBr wafer technique. The Scanning electron microscope (SEM) was obtained using Zeiss 1555 VP-FESEM instrument operating at 15 KV. The X-ray fluorescence (XRF) was measured with RIGAKU ZSX Primus II to detect the sample composition. The inductively coupled plasma (ICP) was used to measure the Ni concentration on Shimadazu ICPE-9000 spectrometer. Thermo gravimetric (TG) curve was obtained by TA Q50. The Magic-angle spinning nuclear magnetic resonance (MAS NMR) spectra were acquired on an Infinityplus 300 MHz spectrometer. The Langmuir surface areas were obtained on ASAP Micromeritics 2040.

2.3. The evaluation and application

To investigate the adsorption potential of synthesized zeolites in the removal of heavy metal ions from aqueous solution, batch adsorption experiment was conducted at 20 °C. 1 g of each synthesized samples was mixed with 100 ml Ni^{2+} solution (1500 ppm, pH = 3) in a conical flask for 24 h. During the adsorption process, the test conical flasks were agitated in a shaking bath at 200 rpm. In the end of the experiment, the syringe equipped with syringe filter was used to collect resulting Ni^{2+} solution. The residual nickel ion concentration was measured with the help of Inductively Coupled Plasma-atomic Emission Spectroscopy (ICPE-9000). The equilibrium sorption capacity was calculated from Eq. (1):

$$q_e = (C_i - C_e)\frac{v}{m} \quad (1)$$

Table 1
The experiment conditions for pretreatment process.

Factor	T/°C	Ash: Alkali	t/h
II-1	110	1.25	24
II-2	110	1.25	48
II-3	110	0.625	24
II-4	110	0.625	48
II-5	50	1.25	24
II-6	50	1.25	48
II-7	50	0.625	24
II-8	50	0.625	48

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