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Original Research Paper

Utilization of incineration fly ash from biomass power plants for zeolite synthesis from coal fly ash by microwave hydrothermal treatment

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ABSTRACT

Potassium-type zeolite (K-zeolite) was synthesized from coal fly ash and woody biomass incineration ash by microwave hydrothermal treatment. The woody biomass incineration ash was discharged from a biomass power plant, and extracted solutions of the material had a pH range of 11.5–13.0 and a high potassium concentration; therefore, it could be employed to replace the KOH solution that is typically used in the synthesis of K-zeolites. Consequently, we successfully synthesized a K-zeolite containing phillipsite phases from coal fly ash using extracted solutions obtained from biomass incineration fly ash. The ammonium adsorption capacity of the K-zeolite synthesized by microwave hydrothermal treatment was comparable to that of K-zeolite synthesized by oil-bath hydrothermal treatment (external heating). We also confirmed that the microwave heating method could shorten the K-zeolite synthesis time compared to that required when using oil-bath heating.

1. Introduction

The use of renewable energy sources for the generation of electric power in Japan is being promoted by the Feed-in Tariff Scheme for Renewable Energy, which was established in 2012 [1–3]. Using woody biomass for electricity generation can give a stable supply of electricity that is not influenced by the weather while also contributing to the suppression of carbon dioxide emissions and the conservation of forests. Therefore, the idea of a boiler power plant using woody biomass as a fuel has been attracted attention in recent years [4]. In Japan, the ash generated during biomass incineration is discharged from power plants and transferred to a final disposal site as industrial waste without any further use [5]. Since the costs of conveying and landfilling is particularly high (about 20,000 JPY/t), this appears to be sufficient to inhibit the spread of boiler power plants using woody biomass as a fuel, and so the development of a suitable scheme for the utilization of biomass incineration ash is necessary [6].

It has been reported that the incineration ash discharged after the combustion of biomass could be utilized for a variety of purposes, for example as a soil improving agent [7,8], a fertilizer [9,10], or an adsorbent material [11]. One potential way of utilizing biomass incineration ash is in the preparation of potassium-type zeolites (K-zeolites) from coal fly ash using extracted solutions of the biomass incineration ash, which exhibit pH values ranging from 11.5 to 13.0 and contain high concentrations of potassium [12,13]. Although a number of studies have reported zeolite formation from coal fly ash using hydrothermal treatment methods [14–21], a potential problem with these systems is that they require a large quantity of highly concentrated alkaline agent for the dissolution of the ions necessary for zeolite synthesis. Thus, aqueous solutions extracted from biomass incineration ash could potentially be employed as alkali sources to reduce the quantity of alkaline agent required. We have previously reported the synthesis of K-zeolites from coal fly ash via an external heating method using woody biomass incineration ash as an alkali source [22]. The zeolites were successfully prepared at a KOH concentration that was much lower than the concentration required to synthesize K-zeolite using a KOH solution alone as the alkaline source. However, long thermal treatment times were necessary to synthesize the zeolite. Therefore, shortening the thermal treatment time is necessary for practical use.

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Microwaves are capable of heating matter rapidly, directly, and selectively. They generate heat by rotating the dipole of an object. In our previous studies, we have investigated the effect of microwave irradiation on zeolite synthesis from coal fly ash using pure chemicals as alkali agents, and have reported that microwave heating reduced the treatment time necessary for the reaction to go to completion [23–25].

In this study, we investigated the effect of microwave heating on the rate of zeolite synthesis from coal fly ash in an extracted solution of biomass incineration ash. Furthermore, the zeolite crystal synthesis rates achieved using the microwave and external heating methods were compared based on the investigation and an empirical equation.

2. Experimental methods

Table 1 lists the properties of the coal fly ash and biomass incineration ash employed herein. Coal fly ash (JIS Z8910, No. 10) with a median diameter of 3.4 μm was used for preparation of the zeolite framework as a source of Si and Al. Biomass incineration ash, which had a potassium concentration of 57.6 mass%, was also used. The ash was collected from the bag filter dust collector following combustion of dry sawdust, bark, and wood chips of Japanese cedar using a fluidized bed furnace in the Chugoku Mokuzai woody biomass power plant in Hiroshima, Japan. The X-ray diffraction (XRD) patterns of the biomass incineration ash before and after extraction with deionized water are shown in Fig. 1. The crystalline phases of the biomass incineration ash were KCl and K_2CO_3, both of which are readily soluble in water. The residue contained almost no potassium, rendering it suitable for use as a raw material for cement (N.B., potassium degrades the quality of cement). In addition, the mass of the biomass incineration ash decreased by ~40% following the extraction process. Therefore, our reported method of using biomass incineration ash halped to both reduce the mass of biomass incineration ash and produce useful byproducts while reducing the quantities of waste materials produced. Prior to zeolite synthesis, the coal fly ash and biomass incineration ash (before extraction) were dried at 100 °C for 24 h.

To confirm the effectiveness of using biomass incineration ash as an alternative to KOH, the following starting suspensions for hydrothermal treatment were prepared:

(i). Initial suspension using only KOH as a source of alkali and potassium (conventional suspension): an aqueous solution of potassium hydroxide (1.5 mol/L) was prepared by dissolving KOH (4.21 g, 86.0%, Kanto Chemical Co., Inc., Japan) in deionized water (50.0 mL, SWG203, SANSYO, Japan). Coal fly ash (10.0 g) was dispersed in this KOH solution and used as the starting suspension.

(ii). Initial suspension using the extracted solution obtained from the biomass incineration ash as a source of alkali and potassium: biomass incineration ash (3.0–15.0 g) was dispersed in deionized water (50 mL), and the resulting mixture was stirred for 10 min. A solution containing alkali and potassium was obtained by filtration. Table 2 shows the potassium ion concentration and pH of the obtained solution. The potassium ion concentration and pH increased upon increasing the mass of biomass incineration ash. In our previous work, we have found that pH of more than 13.5 is necessary for stable zeolite synthesis. Therefore, to adjust the pH of this solution to be more than 13.5, KOH (0.842 g, final concentration 0.3 mol/L) was dissolved in the extracted solution. Finally, coal fly ash (10.0 g) was added as a source of Si and Al.

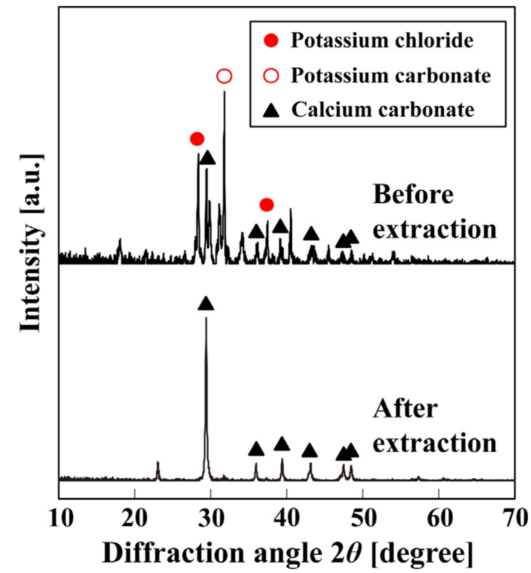

Fig. 1. XRD patterns of biomass incineration ash before and after extraction.

A schematic diagram of the microwave heating equipment is shown in Fig. 2a. Microwaves (2.45 GHz) generated by a magnetron reach the vessel via a waveguide. The prepared starting suspension was irradiated and heated by the microwaves. The temperature of the suspension was held at 413 K, and was controlled by varying the output of the microwaves with a propor tional-integral-derivative (PID) controller. The thermal treatment time was 2–32 h. The treatment pressure was maintained at atmospheric pressure (1.013×10^5 Pa), and the rotational speed of the stirrer was set to 100 rpm. After the hydrothermal treatment, the desired product was isolated by filtration (Büchner funnel), and washed with deionized water thoroughly prior to drying at 373 K for 24 h to give the powdered product.

As a control experiment, the prepared starting suspension was treated hydrothermally under the same conditions using a conventional electric heater, shown in Fig. 2b. The prepared starting suspensions were then loaded into a 100 mL Teflon reaction vessel, and the suspensions were heated in an oil bath (OHB-1000G,

Table 1
Properties of coal fly ash and biomass incineration ash.

	Coal fly ash		Biomass incineration ash	
Component [mass%]	SiO_2	52.1	SiO_2	1.8
	Al_2O_3	22.3	Al_2O_3	1.5
	CaO	2.3	CaO	32.4
	K_2O	<1	K_2O	57.6
	Fe_2O_3	5.2	Fe_2O_3	<1
	Others	18.1	Others	6.7
Median diameter [μm]	3.36		4.40	