



## Research article

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

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## ABSTRACT

In Japan, the quantity of incineration fly ash discharged from biomass power plants has increased rapidly due to the feed-in tariff established by the Japanese government. The costs of disposing of this biomass incineration ash are generally high; therefore, a method must be developed to utilize this by-product. In this context, we propose the syntheses of potassium-type zeolites (K-zeolites) from biomass incineration ash and coal fly ash via a hydrothermal route, which represents a novel use for this material. As the aqueous solution extracted from biomass incineration ash contains high concentrations of potassium, it was employed to substitute the KOH solution that is typically used in the synthesis of K-zeolites from coal fly ash. Consequently, we successfully synthesized a K-zeolite, containing phillipsite and chabazite phases, from coal fly ash using the extracted solution obtained from incineration fly ash. The  $\text{NH}_4^+$  adsorption capacity of our synthesized K-zeolite was comparable to that of the K-zeolite synthesized using only KOH. We also confirmed that the prepared K-zeolite exhibited higher cesium adsorption selectivity than sodium-type zeolites, which are commonly produced industrially.

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## 1. Introduction

Following the Great East Japan Earthquake in 2011, the Japanese government began to consolidate the use of renewable energy for the generation of electric power. In addition, it established the Feed-in Tariff Scheme for Renewable Energy in 2012 [1,2], which promotes power generation using woody biomass. As a result, by 2014, the power generation capacity of woody biomass power plants in Japan had risen to 2.5 million kW [3]. To achieve the governmental target of increasing the renewable power generation capacity to 3.8 million kW using 6 million  $\text{m}^3$  of biomass fuel [4], the cost of power generation must be significantly reduced. More specifically, a reduction in the disposal cost of incineration ash is necessary. Currently, the incineration ash eliminated from the biomass combustion process in Japan must be reclaimed in a final disposal site as industrial waste [5]. Since the cost for conveying and landfilling is particularly high, this inhibits the expansion of woody biomass power plants, and so the development of a suitable scheme for the utilization of biomass incineration ash is necessary [6].

A number of uses for the incineration ash produced by the combustion of biomass have been reported, including its application as a soil improving agent [7,8], a fertilizer [9,10], and an adsorbent material [11], among others [12,13]. Moreover, the addition of biomass ash to

soil improves its texture, aeration, water holding capacity, and salinity. However, as the dominant minerals identified in biomass incineration ash are extremely water-soluble and do not persist in soil, the application of ash has only a minor effect on the mineralogy of the soils. In addition, if the ash is used as a balanced fertilizer, its combination with an additional nitrogen source is necessary, as the ash essentially contains no nitrogen [7]. From the viewpoint of environmental impact, the utilization of biomass incineration ash would be prohibited where it does not meet certain environmental regulations, as this ash contains small amount of harmful heavy-metal components, such as Pb, Cd, Cr, and As [6]. Therefore, these applications have yet to be achieved on an industrial scale. It has been also reported that the aqueous solution obtained from the extraction of biomass incineration ash exhibits a pH range of 11.5–13.0 [14,15]. In addition, as biomass incineration ash contains water-soluble potassium as a main component [15], an aqueous alkaline solution can be obtained containing a high concentration of potassium.

One potential route to the effective utilization of biomass incineration ash is in the preparation of potassium-type zeolites (K-zeolites) from coal fly ash using this extracted solution. Although a number of studies have reported zeolite formation from coal fly ash using hydrothermal treatment methods [16–23], a large quantity of highly concentrated alkaline agent is required for the elution of these ions from coal fly ash [24]. Thus, to effectively reduce the quantity of alkaline agent required, an aqueous solution extracted from biomass incineration ash could potentially be employed as the alkali source.

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

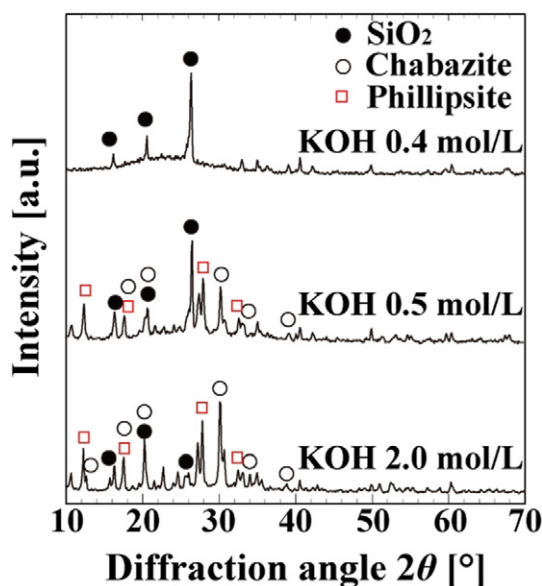
Component [mass%]	Coal fly ash		Biomass incineration ash	
	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>
	52.1	22.3	3.0	2.3
	2.3	20.1	20.1	19.4
	<1	<1	<1	<1
	5.2	55.2	5.2	55.2
	18.1	2.71	18.1	2.71
Median diameter [μm]	3.36	2.71	3.36	2.71

Thus, we herein report our investigation into the syntheses of K-zeolites via a hydrothermal method from coal fly ash using the extracted solution from biomass incineration ash. We will also investigate the absorption selectivity of the prepared K-zeolites towards cesium ions. Indeed, the promising ion exchange ability of zeolites has previously been applied to the removal of radioactive cesium [25,26], which has attracted attention in Japan since the above-mentioned nuclear incident. In addition, several studies have reported the effectiveness of zeolites in the removal of radioactive cesium in soil [27]. However, sodium type zeolites (Na-zeolites), which are mainly produced industrially, release sodium ions upon the absorption of cesium ions. This is undesirable because excessive sodium contents in the soil can inhibit the absorption of water and minerals by plants. In contrast, K-zeolites release potassium ions, which actually promote plant growth [28]. As such, the use of K-zeolites for radioactive cesium decontamination would be preferred over Na-zeolites.

## 2. Experimental

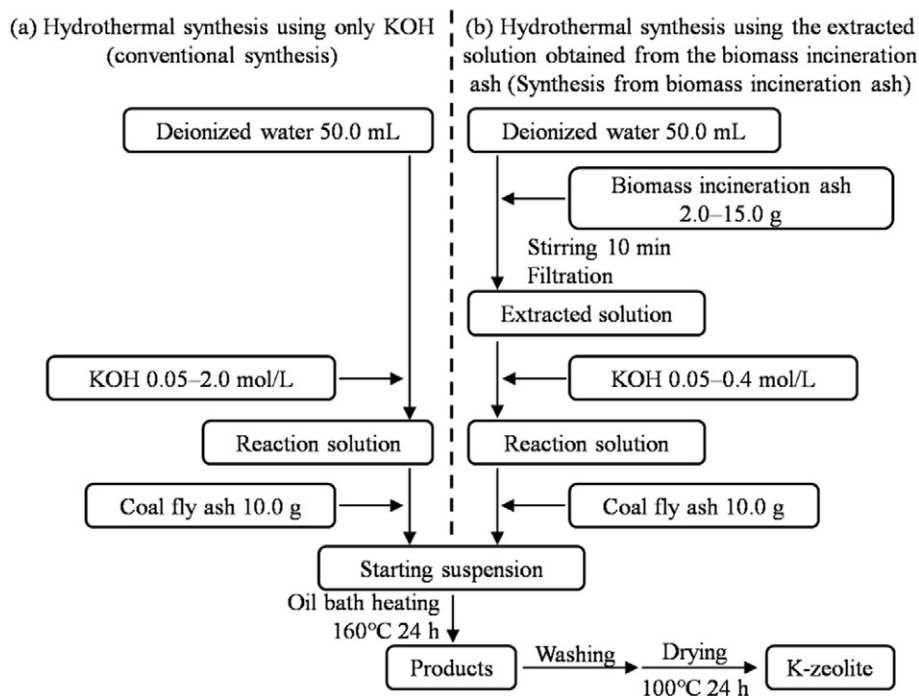
### 2.1. Raw materials

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 as a source of Si and Al for preparation of the zeolite framework. Biomass incineration ash, which had a potassium



**Fig. 2.** XRD diffractograms of the powdered products synthesized using only KOH (conventional synthesis).

concentration of 19.4 mass% (compared to the corresponding metal oxides), was employed as a source of alkali and potassium. The ash was collected at the bag filter dust collector following combustion of the sawdust and bark of Douglas pine using a fluidized bed furnace in the woody biomass power plant of Chugoku Mokuzaï in Hiroshima, Japan. Its crystalline phase is K<sub>2</sub>SO<sub>4</sub>, which is readily soluble in water (see Supplementary data, Fig. S1). It should be noted that the mass of biomass incineration ash decreases by ~34% following the extraction process (see Supplementary data, Table S1). Prior to zeolite synthesis, the coal fly ash and biomass incineration ash (before extraction) were dried at 100 °C for 24 h.



**Fig. 1.** Schematic representation of the synthetic procedures employed in the hydrothermal synthesis of K-zeolites: (a) Hydrothermal synthesis using only KOH (conventional synthesis), and (b) hydrothermal synthesis using the extracted solution obtained from the biomass incineration ash (synthesis from biomass incineration ash).

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