

Alkali Fusion Synthesis of Zeolitic Materials from Waste Dehydrated Cake Discharged from Recycling of Construction Waste Soil

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Abstract

An inorganic cation exchanger, zeolitic material, was synthesized from dehydrated cake, which was discharged from recycling of construction waste soil, using the alkali fusion method. The waste clay was mixed with NaOH powder (the weight ratio of NaOH/waste clay = 1.0) and then heated at 300°C for 1 h to make a fused material. This fused material was then added to distilled water, and then heated at 90°C, 120°C, 150°C and 180°C for 12 h in reaction bombs under autogenous pressure in order to synthesize the cation exchanger. As a result, waste cake can be converted into fused material with high solubility, and zeolitic materials can be synthesized from the fused material. A mixture of zeolite-X and hydroxysodalite was synthesized at 90°C and 120°C, hydroxysodalite alone was synthesized at 150°C and 180°C. By increasing the synthesis temperature, the cation exchange capacity (CEC) of the product decreased, and the highest CEC of the product at 90°C, including zeolite-X, was 2.06 mmol/g, which is 64.3% of commercial zeolite-13X (3.2 mmol/g).

Keywords

Waste Clay, Alkali Fusion, Zeolite-X, Hydroxysodalite, Cation Exchange Capacity

1. Introduction

The construction industry is an important economic sector that has a large environmental impact in terms of natural resources extraction, energy consumption, pollutants release, greenhouse gases emissions and amount of waste generated. Hence, promoting and practicing sustainability in construction can help preserve the planet's ecosystems, conserve natural resources and improve the environmental conditions of all living organisms on earth. Recycling and reuse

of construction and demolition wastes (CDW) is one such attempt to achieve this goal.

Construction waste soil is one of the CDW, and desired to be recycled. Construction waste soil was sorted into three materials, crushed stone, sand and dehydrated cake, in the recycle plant. Recently, crushed stone and sand are major materials of construction to be sufficient demand. However, dehydrated cake has insufficient demand for construction and other industries, and new utilization using this cake is desired.

In a previous study, we converted waste sandstone cake, which was discharged from one of the quarry in Japan, into a zeolitic material by the alkali fusion method and succeeded in synthesizing crystalline zeolite-13X with a high cation exchange capacity (CEC) [1] [2] [3]. By alkali fusion, most of the crystalline phases could be converted into soluble phases and transformed into zeolite crystals. This is a new way of converting the inorganic waste into functional materials.

In this study, we attempted to convert waste dehydrated cake discharged from construction waste soil into inorganic materials with CEC using alkali fusion. The crystalline phases in the cake were converted into the fused material with soluble phases by alkali fusion, and were then used to synthesize an inorganic cation exchanger, zeolitic material.

2. Materials and Methods

2.1. Waste Cake

The waste cake was obtained from one of the companies in Akita Prefecture, Japan. Before the experiment, the waste cake was crushed and sieved under 500 μm , and then dried in an oven at 60 °C overnight.

The chemical compositions of the waste cake determined by X-ray fluorescent spectrometry (XRF) (ZSX101e, Rigaku) are listed in **Table 1**. The waste cake was mainly composed of SiO_2 and Al_2O_3 , and some minor contents.

2.2. Zeolite Synthesis

The experimental procedure is shown in **Figure 1**. First, 10 g of waste cake was mixed with 10 g of NaOH and ground to obtain a homogeneous mixture. This

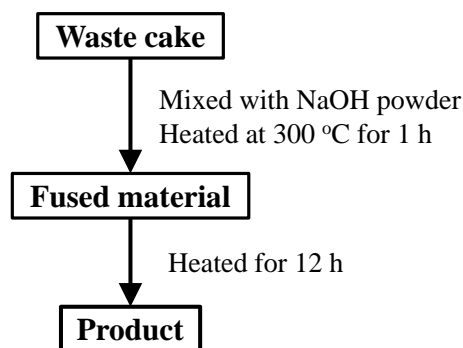


Figure 1. Experimental procedure of this experiment.

Table 1. Chemical composition of waste cake..

	SiO ₂	Al ₂ O ₃	Na ₂ O	K ₂ O	MgO	CaO	Fe ₂ O ₃	SO ₃	P ₂ O ₅	TiO ₂
Oxide (wt.%)	57.1	22.1	1.8	1.9	2.7	3.6	8.7	0.5	0.4	0.9

mixture was then heated in a nickel crucible, which is stable for alkali fusion in this experiment, in air at 300°C for 1 h. The resultant fused mixture was cooled to room temperature and ground again. To determine the solubility of Si and Al in raw cake and fused material, 0.1 g of the solid was added into 10 mL of 1 M HCl solution, and shaken for 6 h. This mixture was filtered, and the concentrations of Si and Al in the filtrate were determined by atomic absorption spectrometer (AAS) (AAna list 200, Perkin Elmer Japan) to calculate the solubility of the solid (mg/g). Next, 2 g of the mixture was added to 10 mL of distilled water in reaction bombs and heated at 90°C, 120°C, 150°C and 160°C for 12 h in an electric oven under autogenous pressure. After heating, the solid product was filtered, washed with distilled water, and dried in a drying oven at 60°C overnight. The phases of the product were analyzed by X-ray diffraction spectroscopy (XRD) (XRD-DSC-XII, Rigaku) and the CEC of the product was measured by the modified Schörrenberg's method [4] as follows. The exchangeable cations in the product were replaced by NH₄⁺ using 1 M ammonium acetate solution. This process was repeated three times at 20 min per exchange. The sample was then washed with 80% EtOH solution to remove excess salt. The NH₄⁺ was then replaced by K⁺ using 10% KCl solution for 20 min. This process was also repeated three times. Finally, the NH₄⁺ in the KCl solution was analyzed by the method described by Koyama *et al.* [5] to determine the CEC of the sample.

3. Results and Discussion

Figure 2 shows the XRD patterns obtained for (a) raw cake and (b) fused material. The raw cake comprised silicate and aluminosilicate minerals, such as quartz, albite and clinocllore (**Figure 2(a)**). After fusion, most of the phases in the raw cake were initially converted into soluble sodium silicate and sodium silicate hydrate (**Figure 2(b)**).

Table 2 shows the solubilities of Si and Al from raw waste cake and fused material. The solubility of raw cake is low, Si: 4.45 mg/g and Al: 5.77 mg/g. After alkali fusion of waste cake, the solubilities of Si and Al from fused material are 74.1 mg/g and 26.51 mg/g, respectively, which are higher than those from raw cake.

From these results, waste cake can be converted into the fused material with high solubility, due to the formation of sodium salts, such as sodium silicate, by alkali fusion.

Figure 3 shows the X-ray diffraction patterns of the product synthesized at (a) 90°C, (b) 120°C, (c) 150°C and (d) 180°C. Zeolite-X and hydroxysodalite were synthesized from fused material, and different phases in the product were confirmed at each reaction temperature. The mixture of zeolite-X and hydroxysodalite was synthesized at 90°C and 120°C, and hydroxysodalite alone was

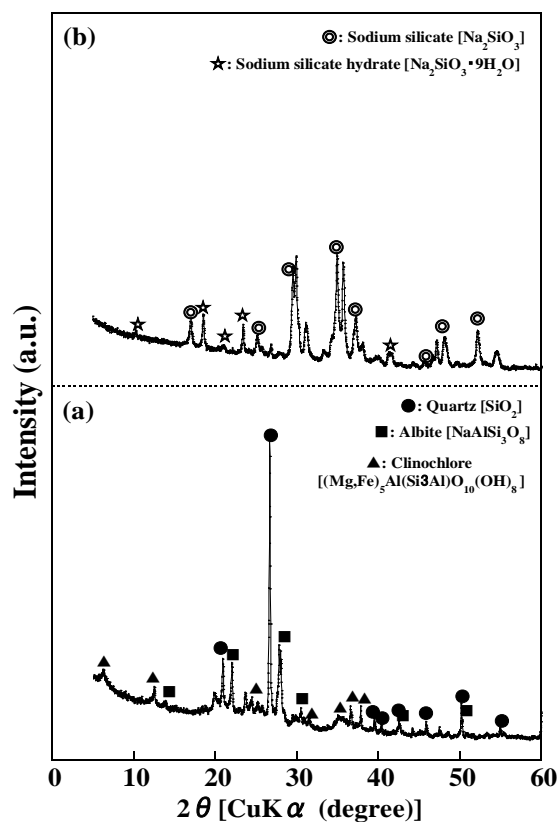


Figure 2. XRD patterns of (a) raw waste cake and (b) fused material.

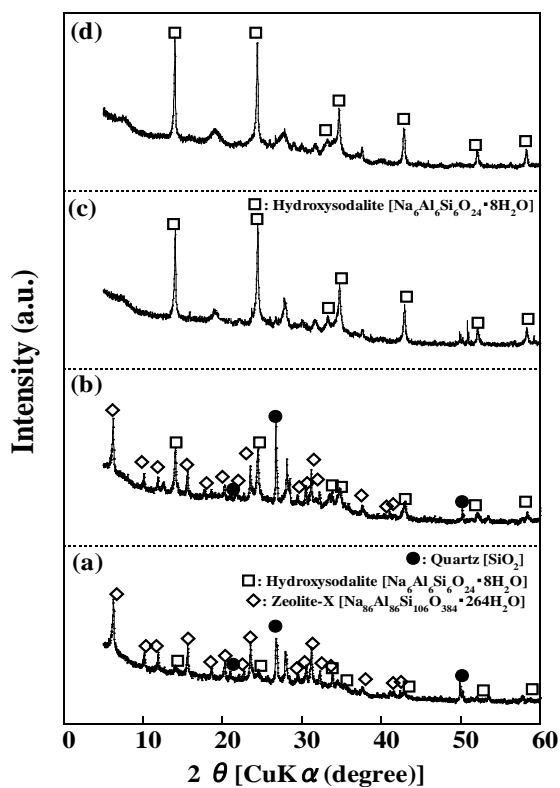


Figure 3. XRD patterns of the products synthesized at (a) 90°C; (b) 120°C; (c) 150°C and (d) 180°C.

synthesized at 150°C and 180°C. It was confirmed that quartz including the raw cake was present in the products obtained at 90°C and 120°C, and diminished in the product at 150°C and 180°C. This means that a part of the mineral phases in the cake, quartz, remained after alkali fusion due to the insufficient fusion condition, and all mineral phases in the cake can be converted into the zeolite phases at 150°C and 180°C through alkali fusion treatment.

Figure 4 shows the SEM photographs of (a) raw cake (b) fused material, and the products synthesized at (c) 90°C and (d) 180°C. Different morphologies of each sample were observed. The raw cake was an aggregate of some minerals (Figure 4(a)), while fused material was a large particle with a melting surface resulting from the formation of sodium salts by alkali fusion (Figure 4(b)). In the product synthesized at 90°C, octahedral crystals of zeolite-X, can be observed in the product, as shown in Figure 4(c), while in the product synthesized at 180°C, ball-like aggregate of hydroxysodalite can be observed (Figure 4(d)).

Table 3 shows the cation exchange capacities of raw waste cake, fused material, and the product synthesized at 90°C - 180°C. The CEC of the product decreased with the increase in heating temperature. Hydroxysodalite has a lower CEC than zeolite-X and is easily synthesized due to the increase of heating temperature. The product with the highest CEC was synthesized at 90°C, and the CEC was 2.06 mmol/g. It is noted that the CEC of commercial zeolite-13X (Wako) is 3.2 mmol/g, which is 1.55 times greater than that of the obtained product.

4. Conclusion

An inorganic cation exchanger can be synthesized from waste cake using the alkali fusion method. Waste cake can be converted into the fused material with

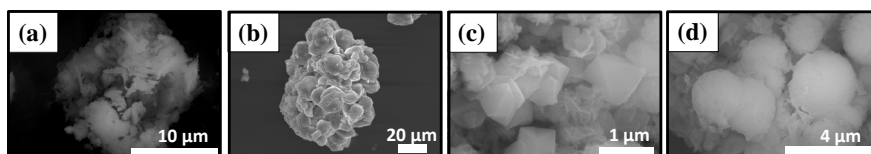


Figure 4. SEM photos of (a) raw cake; (b) fused material, and the products synthesized at (c) 90°C and (d) 180°C.

Table 2. Solubilities of Si and Al from raw waste cake and fused material.

	Raw waste cake		Fused material	
	Si	Al	Si	Al
Solubility (mg/l/g)	4.45	5.77	74.10	26.51

Table 3. CECs of raw waste cake, fused material, and the products.

	Raw waste cake	Fused material	The product			
			90°C	120°C	150°C	180°C
CEC (mmol/g)	0.17	1.16	2.06	2.04	1.84	1.14

high solubility by alkali fusion, and zeolite-X and hydroxysodalite were synthesized. The mixture of zeolite-X and hydroxysodalite was synthesized at 90°C and 120°C, and hydroxysodalite alone was synthesized at 150°C and 180°C. The CEC of the product decreased with an increase in heating temperature, because hydroxysodalite has a lower CEC than zeolite-X and is easily synthesized due to the increase of heating temperature. The product with the highest CEC (2.06 mmol/g) was the product synthesized at 90°C, which is 64.3% of CEC of commercial zeolite-13X.

Acknowledgements

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