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Synthesis of uniform-sized zeolite from windshield waste

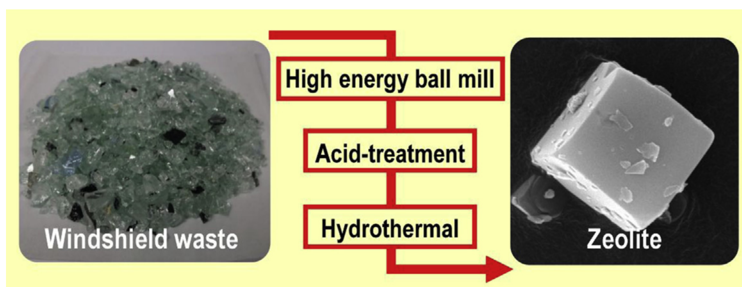
Jae-Chan Kim [a], Mingu Choi [a], Hee Jo Song [b], Jung Eun Park [a], Jin-Ho Yoon [c], Kyung-Soo Park [c], Chan Gi Lee [c], Dong-Wan Kim [a, *]

[a] School of Civil, Environmental and Architectural Engineering, Korea University, Seoul 136-713, South Korea
[b] Department of Materials Science and Engineering, Seoul National University, Seoul 151-744, South Korea
[c] Advanced Materials & Processing Center, Institute for Advanced Engineering (IAE), Yongin-Si 449-863, South Korea

HIGHLIGHTS

- Environmental-friendly recycling of windshield waste for high valuable product of zeolite.
- Synthesis of zeolite form windshield waste via a low-temperature hydrothermal process.
- High-energy milling effect on the uniform cubic shape and high-purity A-type zeolite.

GRAPHICAL ABSTRACT



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ABSTRACT

We demonstrate the synthesis of A-type zeolite from mechanically milled windshield waste via acid treatment and a low-temperature hydrothermal method. As-received windshield cullet was crushed to a fine powder and impurities were removed by HNO_3 treatment. The resulting glass powder was used as the source material for the hydrothermal synthesis of A-type zeolite. Crystal structure, morphology, and elemental composition changes of the windshield waste were evaluated at each step of the process through scanning electron microscopy, X-ray diffraction, X-ray fluorescence spectrometry, etc. After a high-energy milling process, the glass had an average particle size of 520 nm; after acid treatment, its composition was over 94% silica. Zeolite was successfully synthesized in the A-type phase with a uniform cubic shape.

1. Introduction

Every year worldwide scrapped vehicles make a significant contribution to the generation of waste. In recycled vehicle scrap, glass accounts for about 3% by weight. Many solutions have been suggested for recycling waste glass from scrapped vehicles, but a large amount of waste glass still goes to the landfill because of its high recycling cost [1–3]. In particular, the recycling of windshield waste creates a financial burden compared to other waste glass, due to the necessary separation of poly vinyl butyral (PVB) polymer safety film from the glass [4]. To encourage glass recycling, we must develop high-value recycling products to offset the recycling cost.

Many researchers have reported on eco-products that use recycled glass, such as eco-concrete, cement, bricks, and glass fibers

* Corresponding author.
E-mail address: dwkim1@korea.ac.kr (D.-W. Kim).

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[5–9]. Zeolite fabrication is also a possible alternative for recycling waste glass. Much industrial waste is used as source materials for zeolites, including coal fly ash, blast furnace slag, and waste LCD glass [10–12]. Windshield waste is highly suitable for the synthesis of zeolites because its main component is silicate but has not been used in eco-products yet.

Zeolites are microporous structures with three-dimensional cages and channels, made by connecting tetrahedral SiO_4 and AlO_4 units [13]. Zeolites take different forms such as sodalite, calcium aluminosilicate, limonite, zeolite X, and zeolite Y. A-type zeolite (Si/Al = 1) has exceptional capacity for ion-exchange, selective adsorption, and gas separation [14]. Synthetic zeolites are generally produced using a hydrothermal method in an alkaline medium at 200 °C [15]. Various types of zeolite can be synthesized in similar conditions. For producing high-purity A-type zeolite, many researchers suggest the optimization of synthesis variables such as heating rate, heating source, and acid treatment. Recently, M. Kamitani et al. reported the synthesis of A-type zeolite using environmentally friendly flat glass. They optimized acid-treatment and reaction time, but the synthetic zeolite still included impurities and unreacted glass powder. In order to use waste glass as a source material, its characteristics, such as particle size and size distribution, must be optimized to improve the properties of A-type zeolite [10–12,16,17].

Herein, we fabricate A-type zeolite from windshield waste via high-energy ball milling and a low-temperature hydrothermal method. Through high-energy ball milling, the windshield waste was reduced in average particle size for use as a source material for the synthesis of zeolite. Synthesis variables play an important role, but we confirm that the source material's conditions, such as particle size distribution, also affect the properties of synthetic A-type zeolite.

2. Experimental details

Windshield waste was provided in cullet form through the recycling process from the Seoul Glass Industry (Korea). Windshield cullet was ground using a mortar and pestle, and passed through a 270 mesh sieve. The micron-sized glass cullet was used as a source material for the synthesis of zeolite and is hereafter termed glass powder (GP). Sodium hydroxide (NaOH, 98+%, DAEJUNG, Korea) and sodium aluminate ($NaAlO_2$, anhydrous, Sigma–Aldrich) were used without pretreatment.

A-type zeolite was prepared by a two-step process of high-energy ball milling and a hydrothermal treatment. First, we prepared fine glass powder (FGP) using high-energy ball milling (Laboratory Mill MINICER, NETZSCH, Germany). 2 g GP and 300 mL ethanol was mixed and milled in a rotating chamber with 800-μm spherical ZrO_2 beads at a rate of 3000 rpm for 4 h. After the high-energy ball milling process, the product was washed with ethanol several times using a centrifuge, then freeze-dried. 10 g FGP and 250 mL 5 mol/L of HNO_3 aqueous solution were blended in a sealed bottle and stirred at 70 °C for 4 days. After this acid treatment, the solution was filtered with deionized water and dried at 50 °C overnight, creating acid-treated fine glass powder (FGP-A). 1 g FGP-A and $NaAlO_2$ (Si/Al = 1, based on the XRF results of FGP-A) were mixed with 20 mL 0.5 mol/L NaOH aqueous solution. The mixture was stirred for 10 min and set into a 25 mL-capacity Teflon autoclave chamber, where it was heated at 90 °C for 24 h. After this hydrothermal process, the resulting powder (Zeolite FGP-A) was washed with deionized water until it reached a neutral pH value. We synthesized a second zeolite sample by the same process, but without the high-energy ball milling. This was referred to as Zeolite GP-A and compared with Zeolite FGP-A.

The microstructures of the glass and zeolite samples were characterized by field-emission scanning electron microscopy (FE-SEM, JEOL JSM-4300) and transmission electron microscopy (TEM, TECNAI, FEI). The crystal structures of the glass and zeolite samples were identified by X-ray diffraction (XRD, SmartLab, Rigaku). The size distribution and average particle size were verified using a particle size analyzer (PSA, ELSZ-100, Otsuka Electronics). Component analyses were conducted by X-ray fluorescence spectrometry (XRF, ZSXPrimus, Rigaku) and EDS mapping.

3. Results and discussion

Fig. 1 displays pictures describing the change in windshield waste over the course of treatment. Windshield cullet particles of several millimeters in size (picture 1) became micro-scale GP (picture 2) by grinding and sieving at 270 mesh. GP was reduced in

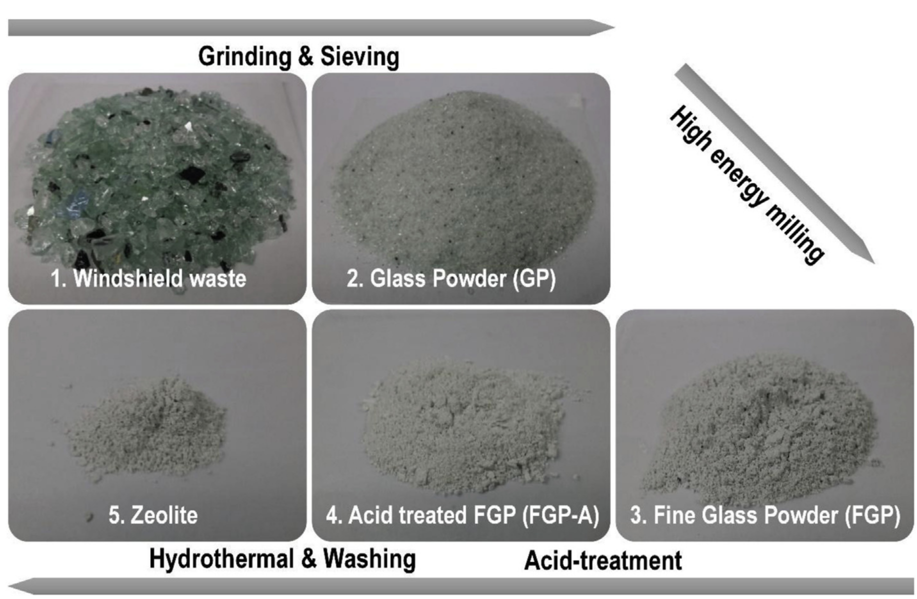

Fig. 1. Flowchart with powder pictures for the synthesis of A-type zeolite.