

Rapid fabrication of highly dense geopolymers using a warm press method and their ability to absorb neutron irradiation



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HIGHLIGHTS

- Geopolymers with appropriate shape inside the mold could be fabricated for a few hours.
- The hardening mechanism by a warm press method was discussed.
- This highly dense geopolymer had an excellent ability to absorb neutron irradiation.

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ABSTRACT

A highly dense pure phase geopolymer was fabricated in only a few hours using a warm press technique. The resultant geopolymer had a high compressive strength of 149 MPa after pressing at 200 MPa and 130 °C for 1 h. The heat and pressure applied during the processing accelerated the geopolymerization and solidification of the starting materials, which consisted of fly ash, a sodium hydroxide solution, and water glass. This highly dense geopolymer had an excellent ability to absorb neutron irradiation, compared to geopolymers fabricated by a conventional curing process.

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1. Introduction

Large quantities of fly ash are discharged worldwide from coal-fired power plants every year [1]. A portion of this fly ash is used in cements and asphalts as a filler material, but additional opportunities for reuse and new applications of fly ash should be developed in order to further reduce waste. Generally, fly ash is primarily composed of an amorphous aluminosilicate, quartz, and mullite ($3\text{Al}_2\text{O}_3 \cdot 2\text{SiO}_2$). In addition, fly ash can contain small amounts of iron oxide, calcium oxide, magnesium oxide, sodium oxide, and potassium oxide. Recently, geopolymer materials have been investigated as an attractive method of fly ash reuse [2–8]. These geopolymers have good mechanical properties and excellent chemical stability [2]. However, acceptable mechanical properties only emerged after curing for 3 days to one week between room temperature and 50 °C. In contrast, increasing the curing temperature to approximately 100 °C decreased the hardening period of the geopolymers to less than one day. However, the mechanical strength drastically decreased due to the formation of many cracks

within the hardened body. Therefore, in order to fabricate geopolymers without cracks in several hours, the authors considered a warm press method, in which the starting materials are pressed at high pressure while being heated to between 100 and 150 °C. Using this method, highly dense geopolymer bodies without cracks can be obtained.

To date, acceleration of geopolymerization by the warm press technique has been studied as a lay-up technique for composites consisting of geopolymer and alumina fibers [9]. However, highly condensed single-phase geopolymers have not been fabricated using the warm pressing technique. In the present study, the effects of the heating temperature and pressing time on the density and compressive strength of the resultant geopolymers were investigated. In order to clarify the densification mechanism, X-ray diffraction (XRD), scanning electron microscopy (SEM), Fourier transform infrared (FT-IR) spectroscopy, magic-angle-spinning nuclear magnetic resonance (MAS-NMR), and pore size distribution measurements were performed on the geopolymers. Geopolymers may be useful as encapsulation materials for nuclear waste due to their high chemical corrosion resistance, resistance to high temperatures, water content that is comparable to that of zeolites [10,11]. Therefore, their ability to absorb neutron irradiation was

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evaluated and compared to that of conventionally cured geopolymer cements and cements.

2. Experimental procedure

Coal fly ash (JIS Type II) discharged from a Japanese power plant was used as a starting material. The chemical composition of the fly ash used is shown in Table 1. A slurry was formed by well mixing 100 g of the fly ash, 28 g of 10 mol/L sodium hydroxide solution, and 50 g of water glass, and this slurry was heated to 130 °C for 2 h to form a dry starting material. The starting material was ground to a particle size finer than 500 μm , passed through a sieve, and then placed in a stainless steel mold. The steel mold was inserted into a warm press, and pressed at 200 MPa while being heated to between 75 and 140 °C for various lengths of time to form a cylindrical sample, 15 mm in diameter and 30 mm in height. As a reference, a conventional geopolymer sample was also fabricated by curing at 50 °C and 80% relative humidity for 3 days using the same starting slurry and a mold. The compressive strengths of the samples were measured using a universal testing machine (5582, Instron). The crosshead speed was 1 mm/min. The crystal phases in the samples were analyzed by XRD (XD-D1). In order to confirm the formation of a geopolymer, KBr pellets containing the sample were examined using FT-IR spectroscopy (Spectrum 100). In addition, the hardened samples and the original fly ash were investigated using solid-state ^{27}Al and ^{29}Si MAS-NMR spectroscopy (Unity Inova 400 Plus) at 104.2 and 79.6 MHz, respectively. The chemical shifts were calibrated with respect to the zero reference of $\text{AlCl}_3 \cdot 6\text{H}_2\text{O}$ for ^{27}Al and tetramethylsilane for ^{29}Si . The pore volume and pore size distribution in the samples were evaluated using N_2 sorption isotherm analysis (Autosorb-1). In particular, the porosities of the bulk samples were characterized using the Barrett–Joyner–Halenda (BJH) method [12]. The water content per unit volume of each sample was calculated from the weight loss after heating at 500 °C and the apparent density of the samples.

To examine neutron transmission, geopolymer samples 45 mm in diameter and 10 mm thick were prepared by the warm press method. Gold filaments were attached to both sides of each sample. After neutron irradiation from one side of the geopolymer sample for 15 min, the radiant intensity of the gamma beam from the gold filaments was evaluated. Finally, the neutron transmission ratio was calculated from the difference between the radiant intensities of the gamma beams from the gold filaments at the top and bottom. The neutron irradiation intensity was $1.9 \times 10^4 \text{ n/cm}^2/\mu\text{A}$.

3. Results and discussion

3.1. Compressive strength

Fig. 1 shows the compressive strength of geopolymer samples heated to various temperatures for 30 min under a uniaxial pressure of 200 MPa. Below 130 °C, the compressive strength of the samples increased with increasing heating temperature, reaching a maximum value of 133 MPa at 130 °C. However, increasing the heating temperature to 140 °C decreased the compressive strength of the samples. Fig. 2 shows the compressive strength and bulk density of geopolymer samples pressed for different lengths of time at 130 °C and 200 MPa. Increasing the pressing time increased the compressive strength of the geopolymer samples. The maximum compressive strength was 149 MPa at 60 min. Furthermore, the bulk density of the geopolymer samples increased with increasing pressing time. In other words, sample densification progressed slowly with pressing time at 130 °C and 200 MPa. According to the true sample density determined using a pycnometer, the relative density (apparent density/true density) of the geopolymer sample pressed for 60 min was 71%, which was relatively large

Table 1

Chemical composition of as-received coal fly ash (mass%).

Composition	Mass%
SiO_2	56.2
Al_2O_3	25.7
Fe_2O_3	7.9
CaO	3.2
TiO_2	2.9
P_2O_5	2.2
K_2O	1.6
Total	99.7

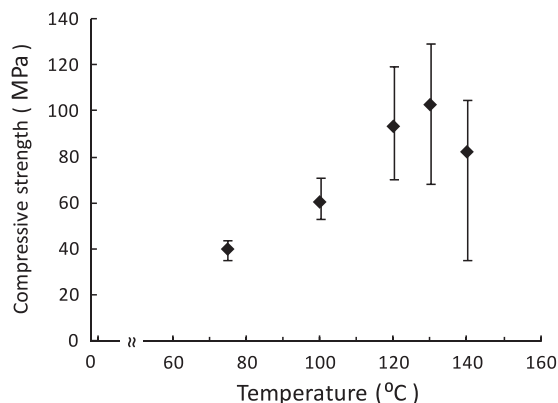


Fig. 1. Compressive strength of geopolymer samples heated to different temperatures for 30 min at 200 MPa.

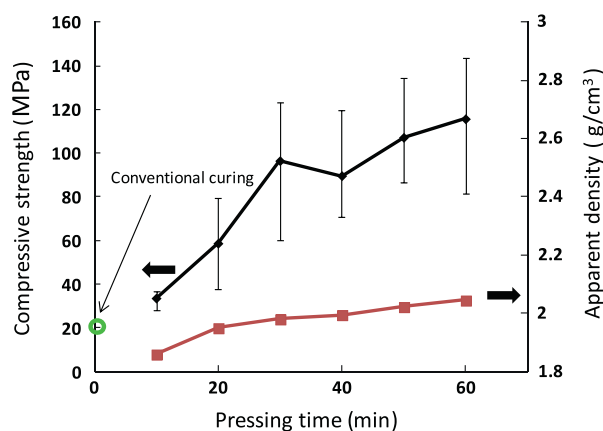


Fig. 2. Compressive strength and bulk density of geopolymer samples pressed for different lengths of time at 130 °C and 200 MPa.

compared to the value of approximately 50% for the conventionally cured sample. Hence, the increase in compressive strength with increasing pressing time was thought to be caused by an increase in the density of the geopolymer samples.

3.2. Effect of warm press on the local structure of the geopolymer

XRD analysis of the conventionally cured geopolymer sample and samples before and after warm pressing was performed. In all samples, XRD identified the crystal phases of quartz and mullite which were contained in the original fly ash. Therefore, warm pressing did not alter the crystal phases present in the geopolymer samples. FT-IR analysis of the conventionally cured geopolymer sample and samples before and after warm pressing was employed. A typical adsorption band around 990 cm^{-1} , which was assigned to the asymmetric stretch vibration of T–O (T: Si, Al) [13], was observed for all samples. Thus, warm pressing did not alter the T–O (T: Si, Al) network structure. Fig. 3 shows (a) ^{27}Al and (b) ^{29}Si MAS-NMR spectra of samples before and after warm pressing. For reference, the spectra of fly ash are also displayed in the figure. In the ^{27}Al MAS-NMR spectra, a resonance peak associated with Al(VI) at 0–20 ppm was present in all processed samples. A peak due to Al(IV): AlO_4 at 50–70 ppm was also detected in the processed samples. It has been reported that upon geopolymerization of fly ash, a transformation from Al(VI) to Al(IV) occurs [14]. That is, the coordination number of Al changes to four, which is the same as that of Si(IV), due to the formation of an Al–O network in the silica network chain. In contrast, in the ^{29}Si MAS NMR spectra of samples before and after warm pressing, the resonance peak

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