

## Article

# Effect of Calcium Hydroxide on Compressive Strength and Microstructure of Geopolymer Containing Admixture of Kaolin, Fly Ash, and Red Mud

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**Abstract:** The current study aims to investigate the effect of calcium hydroxide on the geopolymer derived from an admixture of the natural mineral (kaolin) and industrial by-products (fly ash, red mud). The compressive strength and microstructure were studied using compressive strength tests, X-ray diffraction, infrared spectroscopy, BET method, and scanning electron microscopy. For the investigated NaOH activator concentrations ranging from 4 M to 10 M, the compressive strength of the geopolymer first increases, then decreases with the increase of calcium hydroxide content. The optimal content of calcium hydroxide, which can give the highest compressive strength of the geopolymer prepared, is about 13% wt. of solid raw materials. The geopolymer materials produced at the 8 M NaOH activator have higher compressive strength than those prepared at 4 M, 6 M, and 10 M NaOH. There is a coexistence of geopolymerization gel and C-S-H/C-A-S-H gel in the materials prepared. Both porosity and the formation of N-A-S-H/C-S-H/C-A-S-H during the polymerization process are important for the mechanical properties of materials.

**Keywords:** geopolymer; calcium hydroxide; red mud; kaolin; fly ash

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## 1. Introduction

Geopolymer, which was introduced by Joseph Davidovits in 1976 [1], is an inorganic polymeric material of great interest in the last few decades because of its environmentally friendly, cement-free properties and wide potential applications such as construction materials, adsorbents, and so on [2–4]. Geopolymer is a type of alkali-activated material that possesses a three-dimensional network structure composed of SiO<sub>4</sub> and AlO<sub>4</sub> tetrahedra sharing an oxygen atom as a linking bridge. In general, the formula of geopolymer is M<sub>n</sub>[-(SiO<sub>2</sub>)<sub>z</sub>-AlO<sub>2</sub>]<sub>n</sub>.mH<sub>2</sub>O, where M is the ion used to balance the charge such as Na<sup>+</sup>, K<sup>+</sup>, etc., z is the Si/Al molar ratio; n is the degree of polymerization, and m is the amount of water. Geopolymer material is generally produced by the polymerization of aluminosilicate sources exposed to an alkaline activator solution. The alkaline activators are normally alkaline hydroxides such as NaOH, KOH, and water glass solution. The source materials could be natural minerals such as clays [5,6] and kaolin [7] or waste/by-product materials such as ashes [8,9], slag [10,11], rice hush [12,13], red mud [14–17], etc. One source material or an admixture of more than one source material could be used to prepare geopolymers. In the particular case of red mud, which is the major waste of alumina production from bauxite via the Bayer process [18] and has a quite low Si/Al ratio in composition, the admixture of red mud and other source materials such as fly ash [16,19], metakaolin [20,21], slag [22–24], biomass fly ash, and metakaolin [14] should be used to prepare geopolymers with sufficient qualities.

For application purposes, a lot of effort has been made by scientists to improve the properties of geopolymers. Recently, researchers have found that the calcium component ( $\text{Ca}^{2+}$ ) could have a distinct influence on the properties of geopolymer products. Hu et al. [2] fabricated a micropore-foamed geopolymer composed of fly ash, ZSM-5 zeolite, and calcium hydroxide. According to the authors, the addition of calcium hydroxide brought a positive effect on enhancing the mechanical properties of geopolymer materials obtained. The formation of N-A-S-H and C-A-S-H, which were crystalline phases as unreacted filler derived from materials, was observed. Zhao et al. [25] investigated the effect of calcium on the existing form of the gel product of fly ash-based geopolymers and found that the appropriate amount of added calcium hydroxide could enhance the mechanical properties of geopolymers. Nnaemeka et al. [26] found out that fly ash-based geopolymer blended with kaolin had higher resistance to chemical attack than geopolymer material with 100% fly ash. The authors supposed that the development of N-A-S-H and C-A-S-H in geopolymer could create strong resistance to acid attack. Madani et al. [27] produced the geopolymer brick using wastes of aggregate industries, sand, and calcium hydroxide. They found that the geopolymer brick prepared at 8 M NaOH concentration had higher compressive strength than those prepared at 4 M and 12 M NaOH. Curing geopolymer specimens at temperatures from 70 °C to 105 °C would also enhance the compressive strength of geopolymer brick specimens. Matalkah et al. [28] found that the addition of 10% wt. of calcium oxide dramatically increased the compressive strength of metakaolin geopolymer by 105%. It has been demonstrated that adding a calcium component during the preparation of geopolymer can form more complex calcium-containing sodium aluminosilicate (C,N-A-S-H), hydrated calcium silicate (C-S-H), and hydrated calcium aluminosilicate (C-A-S-H) [29]. The positive effect of the calcium component on the properties of geopolymer materials has mainly been reported based on the formation of C-S-H and/or C-A-S-H, which act as fillers and help bridge voids and gaps in the material [30]. Different types or amounts of calcium components play an important role in the existence or coexistence of gel forms in the products, such as sodium aluminosilicate geopolymer (N-A-S-H), hydrated calcium silicate (C-S-H), and hydrated calcium aluminosilicate (C-A-S-H), which could result in different properties of geopolymer materials. To date, there is still a gap in investigating the effect of calcium hydroxide on geopolymer derived from an admixture containing red mud.

In this study, we effectively optimized the influence of the calcium component ( $\text{Ca}(\text{OH})_2$ ) on the properties of geopolymer. The geopolymer was prepared using an admixture of the natural mineral (kaolin) and industrial by-products (fly ash and red mud). We demonstrated the compressive strength and microstructure of the geopolymer prepared at different activator concentrations and amounts of  $\text{Ca}(\text{OH})_2$ . The results we obtained have the potential to be used in near-future applications of geopolymer as a method for the solidification treatment of waste/by-products and as unsintered materials in the construction field.

## 2. Materials and Methods

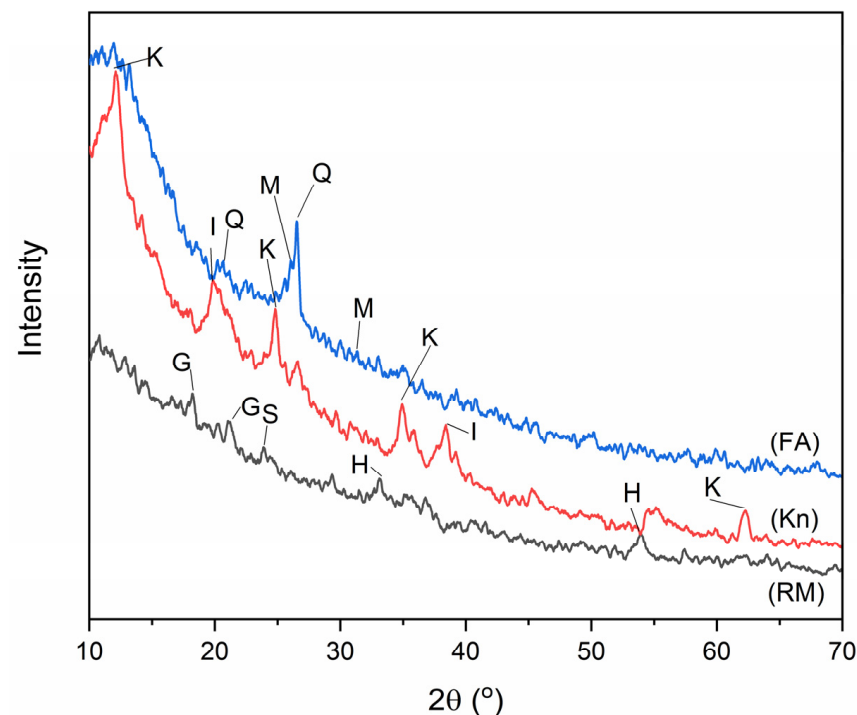
### 2.1. Materials

The red mud used in this study was collected from the waste storage of the Tan Rai alumina plant of Lam Dong Aluminum Company Limited, Lam Dong, Vietnam. Kaolin is commercially available from the Truc Thon clay mine in Hai Duong province, Vietnam. The fly ash was provided by Pha Lai thermal power plant, Hai Duong, Vietnam. The red mud was dried, then ground by a ball mill and sieved to collect the fraction passing a 90  $\mu\text{m}$  mesh screen. Calcium hydroxide (analytical grade,  $\geq 95\%$  purity) was purchased from Xilong Science Co., Ltd., Shantou, China, as a white powder consisting of a small amount of impurities such as sulfate, magnesium salt, and substances precipitated from ammonium hydroxide. Sodium hydroxide was analytical grade ( $\geq 98\%$  purity, China). All the raw materials were dried at 105 °C for 24 h before use.

The chemical compositions of raw materials (kaolin, fly ash, and red mud) were analyzed using X-ray fluorescence (XRF: S4 Pioneer, Bruker, Billerica, MA, USA) and are given in Table 1. Silicon dioxide and aluminum oxide are the main components of fly ash and kaolin, while iron oxide and aluminum oxide are the dominant components of red mud. The X-ray diffractograms of raw materials in Figure 1 demonstrate that kaolin, fly ash, and red mud mostly contain amorphous phases indicated by low-intensity peaks and diffracted patterns in the range of  $2\theta = 10\text{--}17^\circ$  [16,31]. Kaolin consists of crystalline phases of kaolinite, quartz, and illite. The crystalline phases in the red mud are mostly composed of hematite, gibbsite, and sodalite. Fly ash contains crystalline phases of quartz and mullite. Fly ash has the highest Si/Al ratio of 1.9. The Si/Al ratios of red mud and kaolin are 0.3 and 1.2, respectively. The admixing of kaolin with fly ash results in an increase in the Si/Al ratio of materials, which is believed to improve the geopolymerization process and increase the mechanical properties of the materials.

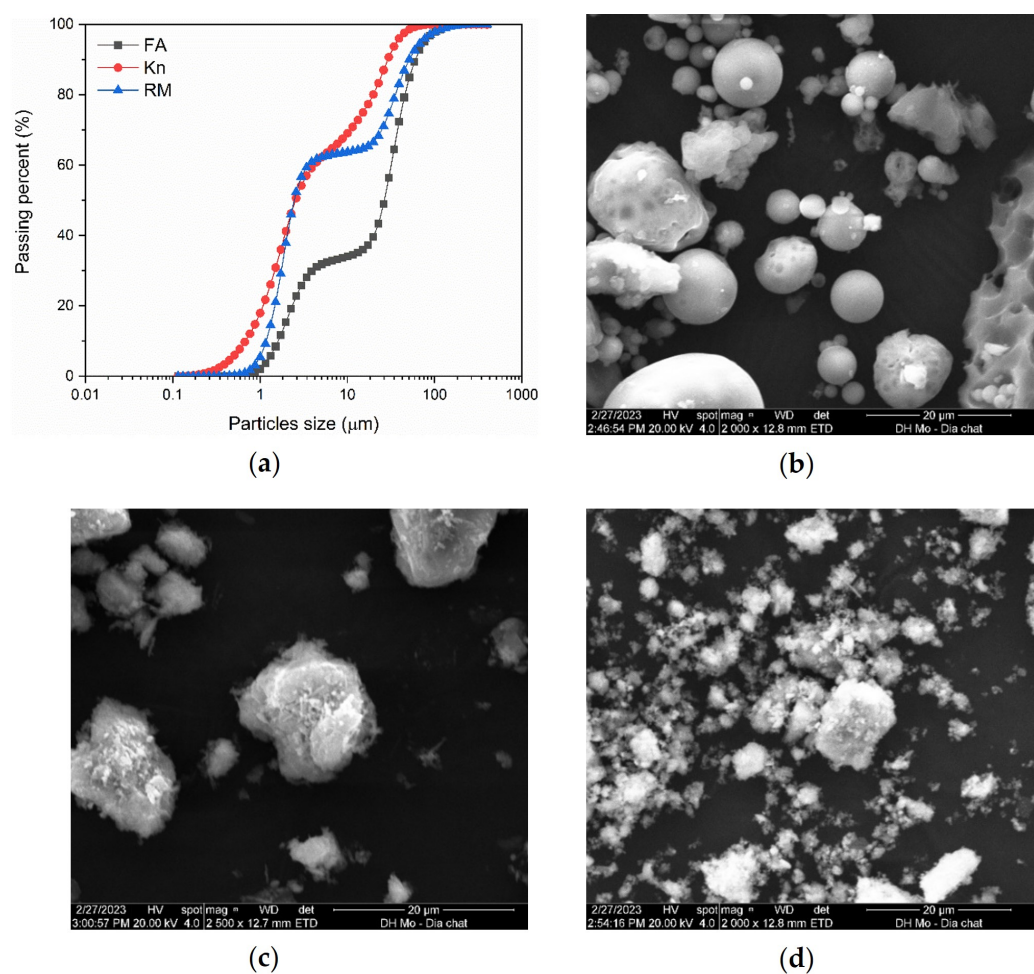
**Table 1.** Chemical composition of red mud, kaolin, and fly ash.

Chemical Composition	Kaolin (wt%)	Fly Ash (wt%)	Red Mud (wt%)
SiO <sub>2</sub>	48.85	40.87	6.01
Fe <sub>2</sub> O <sub>3</sub>	0.64	6.44	48.29
Al <sub>2</sub> O <sub>3</sub>	35.45	18.44	16.16
TiO <sub>2</sub>	0.03	0.77	6.44
MnO	0.18	0.06	0.08
Na <sub>2</sub> O	0.05	0.20	4.27
K <sub>2</sub> O	1.47	3.32	0.09
CaO	0.06	1.18	3.35
MgO	0.04	1.22	0.13
P <sub>2</sub> O <sub>5</sub>	0.04	0.11	0.19
SO <sub>3</sub>	<0.01	1.02	0.54
LOI (Loss on ignition)	12.98	26.27	14.30



**Figure 1.** XRD diffractograms of red mud (RM), kaolin (Kn), and fly ash (Fa) (Q: quartz; M: mullite; K: kaolinite; I: illite; H: hematite; S: sodalite; G: gibbsite).

The particle size distributions of kaolin, red mud, and fly ash were determined using a LA-960 laser scattering particle size distribution analyzer (Horiba Scientific, Kyoto, Japan), as shown in Figure 2a. The particle size diameter distribution of RM, Kn, and FA is in the range of 0.45–200  $\mu\text{m}$ , 0.15–89  $\mu\text{m}$ , and 0.67–229  $\mu\text{m}$ , respectively. The median particle size ( $D_{50}$ ) of red mud, kaolin, and fly ash, which indicates that 50% of total particles are smaller than this size, is 2.5  $\mu\text{m}$ , 2.6  $\mu\text{m}$ , and 26.1  $\mu\text{m}$ , respectively. The  $D_{90}$  values of red mud, kaolin, and fly ash are 51.5  $\mu\text{m}$ , 29.9  $\mu\text{m}$ , and 58.9  $\mu\text{m}$ , respectively. The morphology of kaolin, fly ash, and red mud is presented in SEM pictures in Figure 2b–d. Fly ash exhibits a sphere shape, while kaolin has a plate-like structure. Red mud consists of irregularly shaped aggregates formed from smaller particles.



**Figure 2.** Particle size distribution of kaolin (Kn), fly ash (FA), and red mud (RM)—(a) and SEM pictures of fly ash (b), kaolin (c), and red mud (d).

## 2.2. Preparation of Geopolymers

In the preparation process, dry powders of kaolin, fly ash, red mud, and  $\text{Ca}(\text{OH})_2$  were first mixed for 10 min to obtain a well-blended mixture. A geopolymer material containing up to 25% red mud could be feasibly produced with good properties [15]. Therefore, in this study, the weight ratio of kaolin, fly ash, and red mud in the raw material admixture was kept at 50%:25%:25%, respectively, which was 150 g:75 g:75 g in a particular batch. The content of  $\text{Ca}(\text{OH})_2$  was varied from 0 to 75 g, which corresponds to 0 to 20% of the solid materials by weight. The investigation range of  $\text{Ca}(\text{OH})_2$  amount was decided by referring to several existing studies (e.g., Matakah et al. [28] and Zhao et al. [25]). An activator solution of NaOH with different concentrations (4 M, 6 M, 8 M, and 10 M) was added to the dry mixture, followed by blending for 10 min to obtain a well-mixed paste. Table 2 shows

the mixing ratio of raw materials used for geopolymer preparation. The water/solid ratio by weight of the samples was kept constant at 0.37. The water content was kept as low as possible because the lower water content is more favorable for producing geopolymeric materials with higher mechanical strength [17,32]. The paste was then molded in steel molds (40 mm × 40 mm × 40 mm) under vibration to avoid entrapped air bubbles. After drying at 60 °C for 24 h, the samples were demolded and cured at ambient temperature until mechanical testing at 3 days, 14 days, and 28 days of curing time.

**Table 2.** Mixing ratios of raw materials for geopolymer preparation.

No.	Kaolin (g)	Fly Ash (g)	Red Mud (g)	Ca(OH) <sub>2</sub> (g)	NaOH (M)	Water/Solid (w/w)
1	150	75	75	0	4	0.37
2	150	75	75	15	4	0.37
3	150	75	75	30	4	0.37
4	150	75	75	45	4	0.37
5	150	75	75	60	4	0.37
6	150	75	75	75	4	0.37
7	150	75	75	0	6	0.37
8	150	75	75	15	6	0.37
9	150	75	75	30	6	0.37
10	150	75	75	45	6	0.37
11	150	75	75	60	6	0.37
12	150	75	75	75	6	0.37
13	150	75	75	0	8	0.37
14	150	75	75	15	8	0.37
15	150	75	75	30	8	0.37
16	150	75	75	45	8	0.37
17	150	75	75	60	8	0.37
18	150	75	75	75	8	0.37
19	150	75	75	0	10	0.37
20	150	75	75	15	10	0.37
21	150	75	75	30	10	0.37
22	150	75	75	45	10	0.37
23	150	75	75	60	10	0.37
24	150	75	75	75	10	0.37

### 2.3. Testing and Measurement

**Compressive test:** The compressive strength of the materials was evaluated using an Advantest 9 Controls Model 50-C9030 apparatus with a constant loading rate of 2.5 kN/s until failure of the cubic specimen. The two parallel ends of each specimen were polished using sandpaper to obtain flat surfaces before testing. Three specimens were tested for each sample investigated. The reported compressive strength was the average of the three specimens tested. The formula used to calculate compressive strength is  $F = P_{\max}/A$ , where  $F$  represents the compressive strength (MPa),  $P_{\max}$  is the maximum load (or load until failure) applied to the material (N), and  $A$  is the area of the material resisting the load (mm<sup>2</sup>).

**Microstructure investigation:** X-ray diffraction (XRD) was employed to determine the crystalline phases present in the materials. XRD patterns of the materials were measured using a D8 Advance-Brucker diffractometer with CuK $\alpha$  radiation ( $\lambda = 1.549 \text{ \AA}$ ). In order to observe the morphology of the materials, scanning electron microscopy (SEM) images were taken using an S4800-Hitachi apparatus. Fourier transformation infrared (FT-IR) spectroscopy of the materials was obtained using an IRAffinity-1S-Shimadzu spectrometer. The porosity properties of the materials were determined using the Brunauer–Emmett–Teller (BET) method with a TriStar II Plus analyzer.



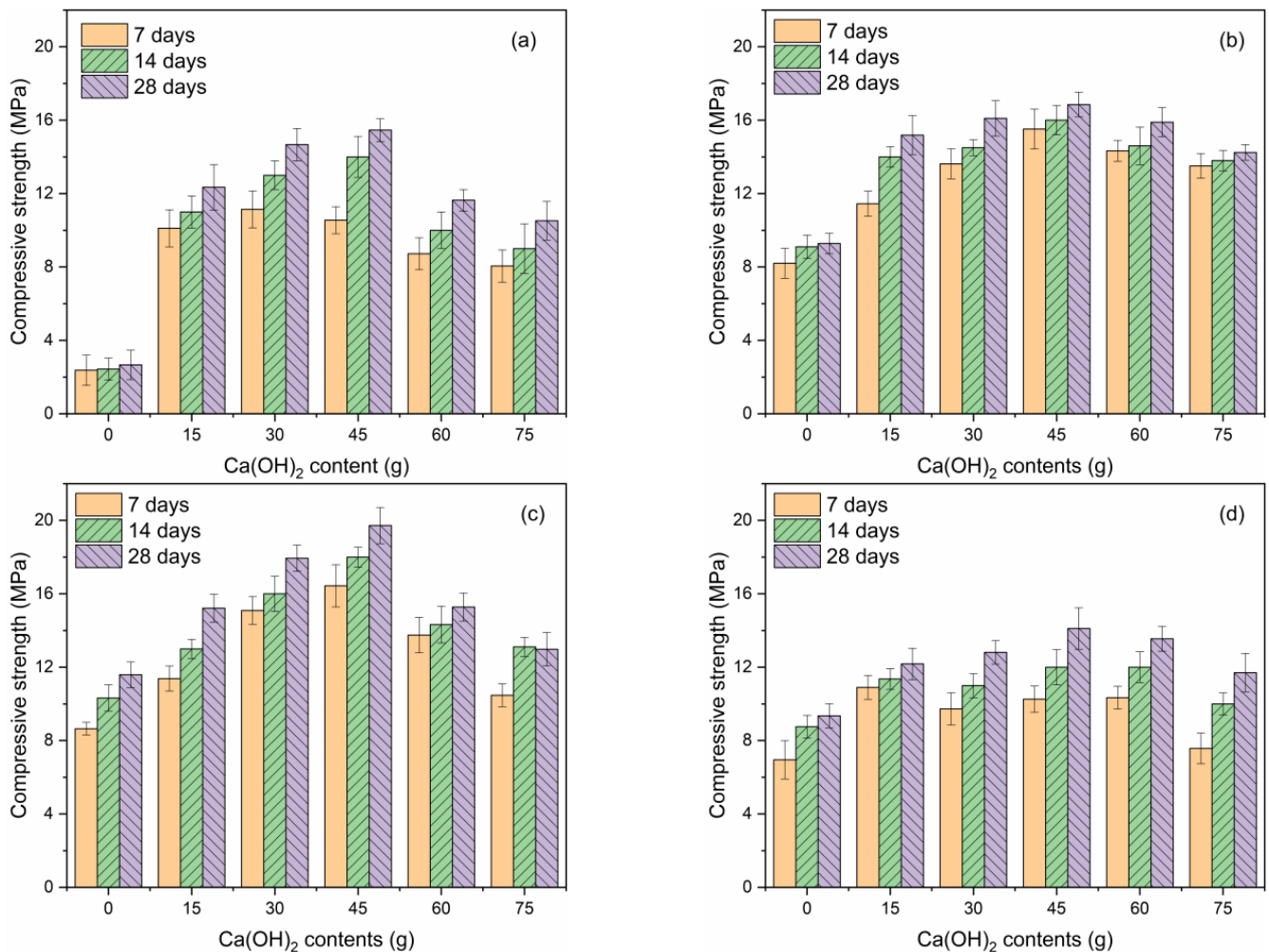
### 3. Results and Discussion

#### 3.1. Compressive Strength of Geopolymers

The results in Figure 3 show the compressive strength of materials prepared with different amounts of NaOH activator and calcium hydroxide. It can be seen that the compressive strength of materials increased with increasing calcium hydroxide content up to 45 g (about 13% by weight of the solid raw materials) in the mixture. Higher content of  $\text{Ca}(\text{OH})_2$ , which is over 45 g, resulted in a decrease in compressive strength. This behavior was observed for all NaOH concentrations investigated in this study. It is in agreement with the behavior of having an optimal amount of  $\text{Ca}(\text{OH})_2$  for the mechanical enhancement of geopolymer materials which has been found by Huang et al. [33] and Zhao et al. [25]. In the study of Huang et al. [33], the optimal  $\text{Ca}(\text{OH})_2$  content in the geopolymer material containing municipal solid waste incineration bottom ash and granulated blast furnace slag was 5%. The research of Zhao et al. [25] showed that the optimal  $\text{Ca}(\text{OH})_2$  amounts were 5.3% and 8.2% in the fly ash-based geopolymer prepared by using 8 M NaOH and 2 M  $\text{Na}_2\text{SiO}_3$  activators, respectively. However, Hu et al. [2] found that the compressive strength of micropore-foamed geopolymer containing fly ash and ZSM-5 zeolite waste continuously increased. This might be because the amount of calcium hydroxide investigated, which was increased up to 6 g (about 2% of solid raw materials), was still small enough for the compressive strength to lay on the increasing part of the curve. When the amount of calcium hydroxide was added in excess, the reactions forming geopolymeric gel and C-S-H gel could compete with each other for soluble silicates and available space. As a result, the geopolymer materials would be more disordered, with more residual voids and holes, leading to the reduction of compressive strength [30]. Another possibility is that the crystal strength of calcium hydroxide is low, and excessive calcium hydroxide surely has a serious influence on the strength of geopolymer materials [33]. The different raw materials and their ratios in the mixtures could result in the different optimal content of calcium hydroxide added to give the highest mechanical enhancement of geopolymer products. In this study, 45 g of  $\text{Ca}(\text{OH})_2$ , which is about 13% by weight of solid raw materials, was the most appropriate value for materials to obtain the best compressive strength. Compared to the results from the literature mentioned above, the optimal content of calcium hydroxide in this research was higher. It can be explained that the geopolymer materials prepared contain red mud, which has a relatively high amount of non-reactive components such as iron oxide, so the higher optimal content of calcium hydroxide was obviously measured.

Comparing the compressive strength of materials prepared without the presence of calcium hydroxide to those prepared with the presence of 45 g of calcium hydroxide at different concentrations of sodium hydroxide activator, it can be seen that the compressive strength of geopolymer with  $\text{Ca}(\text{OH})_2$  presence was 580%, 173%, 170%, and 150% of that of materials prepared without  $\text{Ca}(\text{OH})_2$  when the NaOH activator concentration was 4 M, 6 M, 8 M, and 10 M, respectively. The significant high increment of compressive strength for the 4 M NaOH activator could be understood mainly because the low activator concentration in this particular case can hardly activate the silica and alumina sources of raw materials to form geopolymeric products, leading to relatively low compressive strength. The presence of calcium hydroxide can make the formation of some products, such as C-S-H, possible at low activator concentrations that can give rigidity to the material making higher compressive strength [34]. At higher concentrations of activator, the raw materials were accelerated to form a geopolymeric network, resulting in the compressive strength of materials prepared with NaOH concentrations of 6 M, 8 M, and 10 M being about 3.5, 4.4, and 3.5 times higher than that of materials prepared with 4 M NaOH, respectively. Therefore, the less increment of compressive strength of materials prepared without  $\text{Ca}(\text{OH})_2$  and with the presence of 45 g of  $\text{Ca}(\text{OH})_2$  was observed at higher than 4 M NaOH activator. The highest compressive strength of 19.7 MPa was obtained for the material prepared with the presence of 45 g of  $\text{Ca}(\text{OH})_2$  and 8 M NaOH. In all ranges of NaOH concentration studied, the highest compressive strength was observed for the

geopolymer materials prepared at an 8 M NaOH activator, so it was taken into consideration for the next investigations.



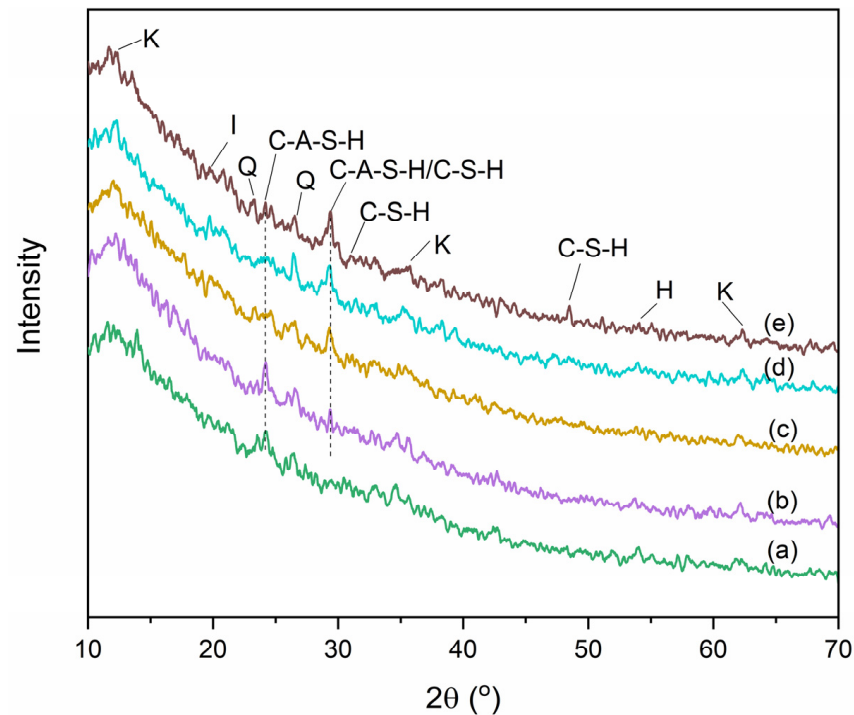
**Figure 3.** Compressive strength of geopolymeric materials vs. the amount of calcium hydroxide at different concentrations of NaOH: 4 M (a), 6 M (b), 8 M (c), 10 M (d).

### 3.2. Microstructure of Geopolymers

#### 3.2.1. Phase Analysis

Figure 4 shows the XRD diffractograms of the geopolymer materials prepared using an 8 M NaOH activator solution, different amounts of calcium hydroxide, and a curing time of 28 days. The typical diffraction peaks representing the phases of quartz ( $2\theta = 20.56^\circ, 26.6^\circ$ ), mullite ( $2\theta = 26.1^\circ, 31.2^\circ$ ), kaolinite ( $2\theta = 12.2^\circ, 24.8^\circ$ ), illite ( $2\theta = 38.5^\circ$ ), hematite ( $2\theta = 33.1^\circ, 53.8^\circ$ ), gibbsite ( $2\theta = 18.2^\circ, 20.2^\circ$ ), and sodalite ( $2\theta = 23.9^\circ$ ) in raw materials were also observed with relatively low intensity in the prepared geopolymer materials [15,16,31]. It is noticeable that the typical peaks at  $2\theta = 24.2^\circ, 29.4^\circ$  and  $2\theta = 29.4^\circ, 31.1^\circ, 48.5^\circ$ , which are the characteristics of C-A-S-H [2,35] and C-S-H [36–38], respectively, were found for the obtained geopolymers. The formation of C-A-S-H, which is the most stable gel type in geopolymer, is believed to play a crucial role in enhancing the compressive strength of the materials investigated in this study [35]. Moreover, the rise in intensity of the peak at  $2\theta = 29.4^\circ$ , which was clearly observed, could confirm that a greater amount of C-S-H/C-A-S-H was formed in the geopolymer materials with an increasing amount of added calcium hydroxide. Combining the results of intensity increase at  $2\theta = 48.5^\circ$ , which was clearly observed for the material prepared with 75 g of calcium hydroxide, it could be concluded that more C-S-H was formed with a higher amount of added calcium hydroxide. The

presence of C-S-H and C-A-S-H phases confirmed by XRD analysis highly supports the results of compressive strength discussed in the previous section.

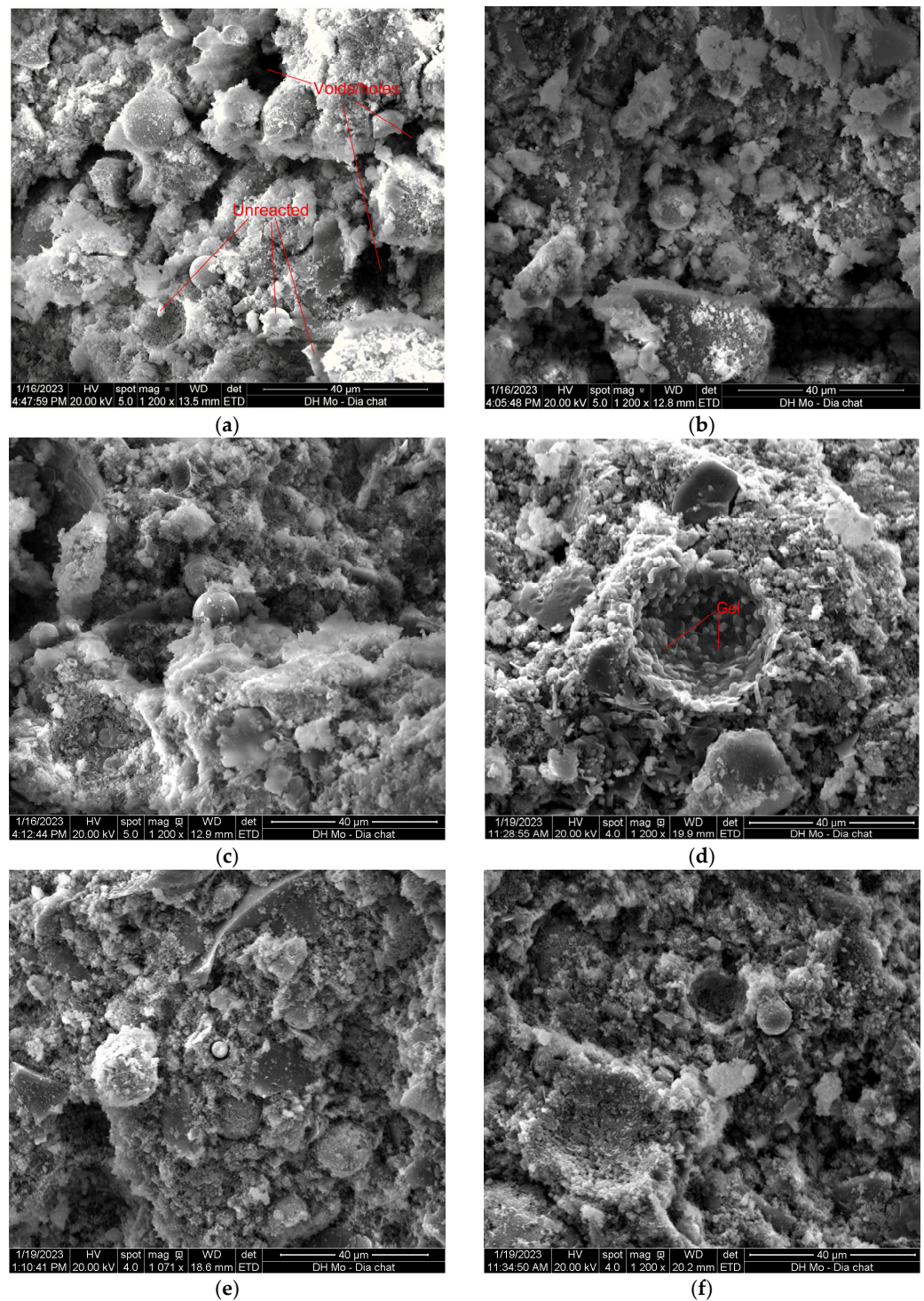


**Figure 4.** XRD diffractograms of geopolymer materials prepared using 8 M NaOH activator and  $\text{Ca}(\text{OH})_2$  content of 15 g (a), 30 g (b), 45 g (c), 60 g (d), 75 g (e) at 28 days of curing time (Q: quartz; K: kaolinite; I: illite; H: hematite; C-A-S-H: calcium aluminosilicate hydrate; C-S-H: calcium silicate hydrate).

### 3.2.2. Morphology Analysis

The changes in the amount of calcium hydroxide should influence the microstructure of the materials prepared. The morphology of geopolymer materials containing different calcium hydroxide content was observed through SEM investigation. SEM pictures of materials prepared using an 8 M NaOH activator and calcium hydroxide of 0–75 g at 28 days of curing time are shown in Figure 5. It can be seen that raw materials are embedded together in geopolymers. However, some particles or fragments with relatively weak adhesion on the surface were also observed for all samples. More unreacted and nonactive particles were detected for the geopolymer sample prepared without calcium hydroxide addition (Figure 5a). It is clearly seen that as more calcium hydroxide was added, the geopolymer materials became denser, with fewer voids or holes present. This result is due to the formation of geopolymeric products and C-S-H, C-A-S-H, which can help bridge the gaps between the unreacted or nonactive particles. Significantly, the gel form, which could result from a geopolymerization reaction under an alkaline activator, was obviously seen in the sample prepared with 45 g  $\text{Ca}(\text{OH})_2$ . This finding is greatly consistent with the enhancement of its compressive strength analyzed above.



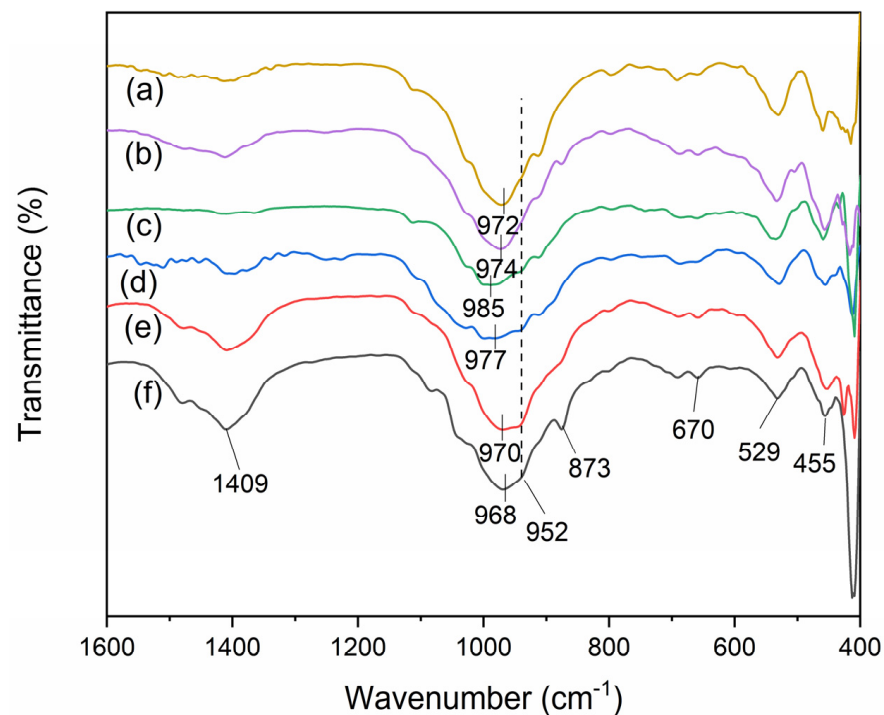


**Figure 5.** Scanning electron micrographs of geopolymer materials prepared using 8 M NaOH activator and  $\text{Ca}(\text{OH})_2$  content of 0 g (a), 15 g (b), 30 g (c), 45 g (d), 60 g (e), 75 g (f) at 28 days of curing time.

### 3.2.3. FT-IR Spectroscopy Analysis

Figure 6 shows the FT-IR spectra of materials prepared with an 8 M NaOH activator and different amounts of calcium hydroxide. The spectral results focused on the range from  $400\text{--}1600\text{ cm}^{-1}$ , where some important broad bands characterizing T-O bonds (T: Si, Al) are located [2]. The band in the range of  $800\text{--}1200\text{ cm}^{-1}$  corresponds to the asymmetric and symmetric stretching vibration of Si-O bonds. The characteristic broad band located in the range of  $900\text{--}1200\text{ cm}^{-1}$ , which is associated with the asymmetric stretching vibration

of the Si-O-T, is the key focus of geopolymer specimens [2]. It can be seen that the peak position of the wavenumber shifted from  $972\text{ cm}^{-1}$  to  $985\text{ cm}^{-1}$  as the calcium hydroxide content was increased from 0 to 30 g. Then, a higher amount of calcium hydroxide than 30 g would make the peak position shift to a lower wavenumber. Zhao et al. [25] also found a shift in the Si-O-T asymmetric stretching vibration when the amount of  $\text{Ca}(\text{OH})_2$  was increased in the fly ash-based geopolymers. Particularly, the peak shifted first to a lower wavenumber and then shifted to a higher wavenumber as the amount of  $\text{Ca}(\text{OH})_2$  was increased. The authors believed that the polymerization of Si-O-Al should take place when  $\text{Ca}(\text{OH})_2$  is less than a critical value of content, while the C-S-H gel is generated when higher than this critical value of  $\text{Ca}(\text{OH})_2$  amount is added. In this study, the shifting of the asymmetric stretching vibration to a higher wavenumber should indicate that C-S-H gel coexists with N-A-S-H gel in the geopolymer materials prepared with up to 30 g of  $\text{Ca}(\text{OH})_2$ . The shifting to a lower wavenumber (from  $985\text{ cm}^{-1}$  to  $968\text{ cm}^{-1}$ ), as higher than 30 g of  $\text{Ca}(\text{OH})_2$  was added, might correspond to the coexistence of C-A-S-H gel formed by ion exchange with N-A-S-H gel [37]. Moreover, the band shoulder at  $952\text{ cm}^{-1}$ , which is assigned for C-A-S-H gel, was more clearly observed for materials prepared with 60 g and 75 g of  $\text{Ca}(\text{OH})_2$ . Hence, C-A-S-H gel could coexist with N-A-S-H and C-S-H gels in materials prepared using higher than 30 g of calcium hydroxide.

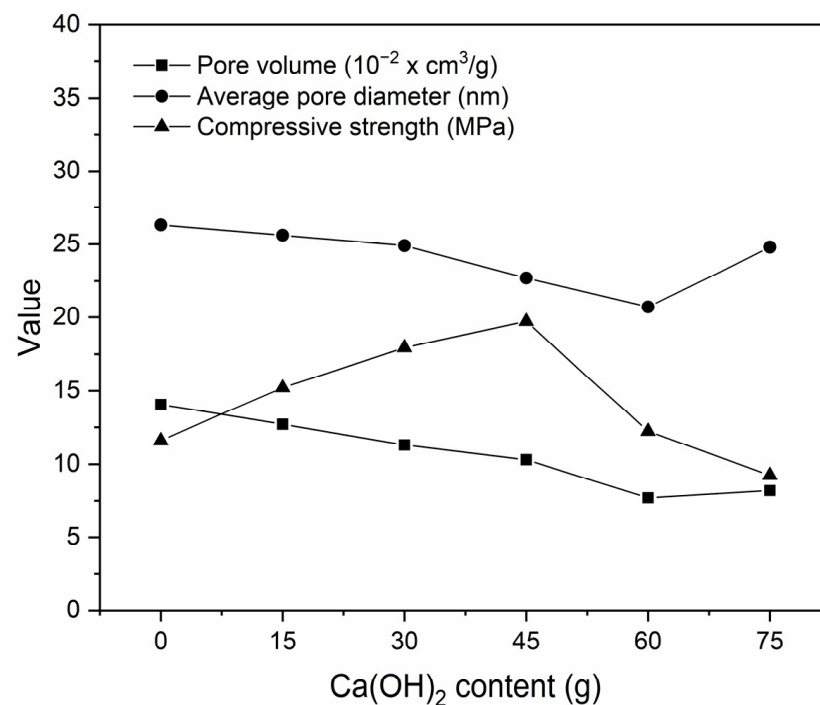


**Figure 6.** FT-IR spectra analysis of materials prepared with 8 M NaOH activator and different amounts of  $\text{Ca}(\text{OH})_2$ : (a) 0 g, (b) 15 g, (c) 30 g, (d) 45 g, (e) 60 g, and (f) 75 g.

### 3.2.4. Porosity

Porosity is an important parameter affecting the mechanical properties of materials. The pore volume and average pore diameter of the geopolymer materials prepared using an 8 M NaOH activator and varying amounts of calcium hydroxide were measured using the BET method and are shown in Figure 7, along with the compressive strength of the corresponding materials. It is evident that the geopolymer prepared without the presence of calcium hydroxide had the highest observed pore volume and average pore diameter. As the content of calcium hydroxide was increased up to 60 g, the pore volume, as well as the average pore diameter of the specimens, continuously decreased, indicating that the presence of calcium hydroxide made the materials more densely structured. This is

because calcium hydroxide not only participates in the geopolymerization process in an alkaline environment but also forms crystalline phases or precipitates that act as fillers, adding particles that fill the observed holes, voids, and gaps in the material [2]. However, when more than 60 g of  $\text{Ca}(\text{OH})_2$  was added, the pore volume and average pore diameter increased. This can be explained by the excessive  $\text{Ca}(\text{OH})_2$  particles forming deposits or precipitates, resulting in a loose internal structure and higher porosity of materials. Although it is well-known that compressive strength should increase when materials become denser, increasing the amount of calcium hydroxide from 45 g to 60 g resulted in a decrease in compressive strength despite the decrease in pore volume and pore diameter. Therefore, it can be concluded that both porosity and the formation of N-A-S-H/C-S-H/C-A-S-H during the polymerization process play a crucial role in the mechanical properties of materials.



**Figure 7.** Pore volume, average pore diameter, and compressive strength of materials prepared by using 8 M NaOH and different  $\text{Ca}(\text{OH})_2$  contents.

#### 4. Conclusions

In this work, the effect of calcium hydroxide on the geopolymer derived from kaolin, fly ash, and red mud was investigated using compressive strength tests, X-ray diffraction, infrared spectroscopy, BET method, and scanning electron microscopy. For all ranges of NaOH activator concentrations investigated from 4 M to 10 M, the compressive strength of the geopolymer first increased, then decreased with the increasing calcium hydroxide content. The geopolymer materials produced at the 8 M NaOH activator had higher compressive strength than those of materials prepared at 4 M, 6 M, and 10 M NaOH. The critical value of calcium hydroxide content, which can give the highest compressive strength of the geopolymer prepared, is about 13% wt. of solid raw materials. There is a coexistence of geopolymerization gel and C-S-H/C-A-S-H gel in the materials prepared. Both porosity and the formation of N-A-S-H/C-S-H/C-A-S-H during the polymerization process are important for the mechanical properties of materials. The highest compressive strength, 19.7 MPa, was obtained for the material prepared with the presence of 45 g  $\text{Ca}(\text{OH})_2$ , 8 M NaOH, and kaolin: fly ash: red mud ratio of 150 g: 75 g: 75 g. The findings of this study can help develop a better understanding of the effect of calcium hydroxide on the properties of geopolymeric products for further research and practical applications.



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