



Effect of SiO_2 , Al_2O_3 and Na_2O content and fly ash fineness on the structure and mechanical properties of fly ash based geopolymer

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ABSTRACT

Nowadays, the geopolymer technologies using secondary raw materials are more and more widespread; however, some of them (for example fly ash) have originally low reactivity which can be tailored by a mechanical or chemical activation, or by the addition of various high reactive materials. This study investigates the effect of silica, alumina, and sodium oxide contents on the structure and mechanical properties of high calcium fly ash based geopolymers. Metakaolin (MK) was used as an additional alumina and silica source. It was added at various dosages (0; 5; 10; 15; 25; 50 and 75 % by weight) as a replacement of the fly ash (FA). The experimental results confirm that the compressive strength of the geopolymer is greatly affected by the $\text{SiO}_2/\text{Al}_2\text{O}_3$, $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$, and $\text{SiO}_2/\text{Na}_2\text{O}$ ratio. The addition of MK improved the compressive strength of geopolymer by 92 %. In addition, the effect of mechanical activation of FA on the structure and strength of the geopolymer was investigated in case of a given MK content. Based on the results it can be stated that the mechanically activated FA resulted in higher compressive strength. The addition of MK and the fineness of FA changed the structure of geopolymers, which was detected using FT-IR spectroscopy method.

1. Introduction

Geopolymers are three-dimensional amorphous-to-semi-crystalline aluminosilicate materials (Davidovits, 1989), which can be produced from natural/synthetic aluminosilicate minerals or industrial aluminosilicate byproducts (for example fly ash, red mud, slag, metakaolin, perlite, glass, rice husk ash, clay, or a combination of them) mixed with an alkaline (potassium/sodium hydroxide, potassium/sodium silicate) or acidic solution (phosphoric acid) (Palomo et al., 1999; Komintzas and Zaharaki, 2007; Vaou and Panias, 2010; Mucsi et al., 2014; Tchakoute et al., 2017; Singh et al., 2018). Geopolymers possess good physical-chemical and mechanical properties like relative high strength, fire and

chemical resistance, and thermal stability. Due to these properties geopolymers give an opportunity to replace conventional structural materials in the fields like road construction and building industry, metallurgy, mining industry, and high-tech industry (Davidovits, 2002). The properties of geopolymers are affected by the composition and reactivity of the material (Kumar et al., 2017; Singh et al., 2018), the composition of the activator solution (Fernandez-Jimenez and Palomo, 2005; Molnár et al., 2017; Tchakoute et al., 2017; Cheng et al., 2018; Singh et al., 2018), the curing condition (treatment temperature and time) (Palomo et al., 1999; Molnár et al., 2017), and the compression method (especially vibrating compaction or high-pressure compaction) (Wallah and Rangan, 2006; Živica et al., 2011). The alkaline solution

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plays an important role in dissolving Si and Al atoms to form geopolymer precursors and aluminosilicate material. The mechanical properties and microstructure of geopolymer are affected by the molar ratio of $\text{SiO}_2/\text{Al}_2\text{O}_3$.

Chindaprasirt et al. (2012) studied the effect of silica and alumina contents on setting, phase development, and physical properties of high calcium fly ash based geopolymers. The control of setting and hardening properties were investigated by changing $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of the starting mix. $\text{SiO}_2/\text{Al}_2\text{O}_3$ content of the mixtures were changed in the range 2.87-4.79. They found that the increases in either silica or alumina content shortened the setting time of high calcium-based systems.

Chen et al. (2018) investigated the effect of sodium polyacrylate (as organic polymer) on the mechanical properties and microstructure of metakaolin-based geopolymer with different $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio (2.0-4.0). They found that the toughening effect of organic polymer on geopolymer depended on the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of geopolymer, and the geopolymer with lower $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio (no more than 2.5) can be significantly toughening modified by organic polymer.

He et al. (2016) investigated the effect of Si/Al ratio on the structure, mechanical properties and chemical stability of metakaolin based geopolymers. Geopolymers with Si/Al ratios of 2 and 2.5 showed similar structure and property and geopolymers with Si/Al ratios of 3, 3.5 and 4 were similar. Geopolymers with Si/Al of 4 showed much higher mechanical properties than geopolymers with Si/Al of 2, which was due to the increased Si-O-Si bonds and residual silica as reinforcement. However, geopolymers with $\text{Si/Al} \geq 3$ showed worse chemical stability than those with $\text{Si/Al} \leq 2.5$, with the presence of efflorescence on the surface, which was attributed to their higher residual free K^+ .

Several studies (Criado et al., 2007; Silva and Crenstil, 2008; Vargas et al., 2011; Tchakoute et al., 2013; Bignozzi et al., 2014; Gao et al., 2014; Khedmati et al., 2018) have reported that, in addition to $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio, the $\text{Na}_2\text{O}/\text{SiO}_2$ and $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ ratios also play an important role in the morphological, microstructural, and mechanical properties of fly ash based geopolymer.

Vargas et al. (2011) investigated the influence of different $\text{Na}_2\text{O}/\text{SiO}_2$ molar ratio, curing temperature and age on the mechanical and microstructural properties of fly ash based geopolymers. The results showed that the $\text{Na}_2\text{O}/\text{SiO}_2$ ratio played an important role in the mechanical and morphological characteristics of geopolymers. Geopolymer with higher $\text{Na}_2\text{O}/\text{SiO}_2$ ratio showed higher compressive strength and more compact structure.

Khedmati et al. (2018) received similar results. They found that the higher $\text{Na}_2\text{O}/\text{SiO}_2$ molar ratio showed more N-A-S-H (sodium aluminosilicate hydrate) gel formation, which increased the bonding strength between the geopolymer binder and the aggregate.

Tchakoute et al. (2013) investigated the effect of $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ molar ratio of soda-volcanic ash. Based on the results, they found that the optimal $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ molar ratio of volcanic ash to produce geopolymer mortars ranged between 0.13 and 0.18.

The aim of the present research reported in this paper is primarily to study the effect of SiO_2 , Al_2O_3 and Na_2O contents on the structure of geopolymer and to examine the relation among silica, alumina and sodium oxide content and geopolymer compressive strength. The additional goal was to investigate the dependence of strength on the fly ash fineness.

2. Material and methods

For the experiments FA from the lignite burned power plant (Visonta, Hungary) and MK (IMERYS Ltd.) were used as solid. FA was replaced with MK in various amounts (0; 5; 10; 15; 25, 50 and 75 wt %) in the solid part. The mixture of sodium-hydroxide (8 M) and sodium-silicate (waterglass) solutions was applied as alkaline activator. NaOH solution was prepared by dissolving sodium hydroxide pellets with a purity of 99 % in distilled water. The chemical composition of the sodium silicate solution was as following: 25.3 % SiO_2 , 13.7 % Na_2O , 2.7 % K_2O of and 58.3 % H_2O .

The particle size distribution of the raw materials and the ground FAs was measured by HORIBA LA-950V2 laser diffraction particle size analyzer in wet mode using distilled water as dispersing media and sodium-pyrophosphate as dispersing agent, applying the Mie-theory as evaluation method. The geometric (outer) specific surface area (SSA) was calculated using PSD data by the laser sizer software, the shape factor was 1. The particle density (ρ_p) was determined by pycnometer method using alcohol as media. The chemical composition of FA and MK was determined using X-ray fluorescence spectroscopy analysis (XRF). The structure of raw materials and geopolymers was detected by Fourier Transform Infrared Spectroscopy (FT-IR) in reflection mode with diamond ATR. The main physical properties and chemical composition of FA and MK are found in Table 1 and Table 2.

Based on Table 2 it can be stated that the SiO_2 content of FA and MK was similar, but the Al_2O_3 content was rather different. The Al_2O_3 content in MK was three times higher than in FA. The Na_2O content was low in both materials (under 0.4 wt %), but in FA was higher than in MK.

Table 1
Physical properties of raw materials

	Fly ash	Metakaolin
particle density (g/cm^3)	1.93	2.73
x_{10} (μm)	10.8	1.8
x_{50} (μm)	52	5.2
x_{80} (μm)	119.3	8.5
SSA (cm^2/g)	1,152	11,307

Table 2
Chemical composition of starting materials

Component (wt %)	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	MgO	Na ₂ O	K ₂ O	TiO ₂	P ₂ O ₅	MnO	SO ₃	L.o.I.*
Fly ash	48.1	14.42	11.76	10.97	3.34	0.37	1.66	0.492	0.264	0.171	0.575	2.2
Metakaolin	51.4	45.9	0.4	1.12	0.26	0.05	1.34	0.05	0.064	0.033	< 0.013	-

*L.o.I. = Loss on ignition (at 950 °C)

3. Experimental

The mechanical activation experiments of FA under dry condition were carried out in a conventional tumbling laboratory ball mill with the size of Ø305×305 mm (smooth walled), with steel balls (minimum and maximum ball size was 12 mm and 50 mm, respectively) as grinding media. The mill filling ratio of the grinding media was 30 volume %, the material filling ratio (compared to the pore volume among the grinding media) was 110 volume %. The operational revolution number to the critical revolution number was 80 %. The residence time of mechanical activation was 5, 10, 20, 30, 60, and 120 minutes.

As a first step, the raw or ground FA (and MK) and alkaline activator (mixture of 8 M NaOH and waterglass) were mixed together using 0.82 liquid/solid ratio (L/S ratio). Then the paste was placed to pre-oiled moulds and compacted by vibration for 1 minute. The compacted mixture was kept in moulds for 24 hours in sealed condition in a climate chamber at 23 °C, before removing the specimens. It was followed by heat curing at 30 °C for 6 hours. After the heat treatment, the specimens were stored in a climate chamber at 23 °C and 90 % relative humidity until the measurement of compressive strength. Five specimens of geopolymer were prepared in all cases for the strength investigations. The mechanical test was carried out by Compression Testing Machine for 7 days.

4. Results

4.1. Mechanical activation of FA

The effect of the grinding on the particle size and SSA of the raw and ground FA is summarized in Table 3. The 50 percent particle size (median) of the raw FA was 48.4 µm. During the grinding the particle size was significantly decreased and SSA increased. After 120 min grinding in a ball mill 8.4 µm median particle size was achieved from the initial value of 48.4 µm, indicating a 5.76 size reduction rate. Particles with smaller size than

1 µm also appeared in a significant amount (more than 10 %). Additionally, the “outer” specific surface area increase was significant; from 1,140.9 cm²/g it reached 12,228.8 cm²/g due to ball milling.

4.2. Effect of SiO₂/Al₂O₃, SiO₂/Na₂O and Al₂O₃/Na₂O molar ratios

The use of MK had a beneficial effect on the compressive strength of geopolymers. Based on the Figure 1, it can be seen that the increasing amount of MK in the solid part of mixture increased the geopolymer strength and density. Geopolymer with the highest strength and density was produced by 50 % MK content in the solid part. These values were 22.1 MPa and 1.61 g/cm³. However, it is important to note that the increasing amount of MK reduced the workability of the mixture. It was not workable by 75 % MK content. In this case the fresh paste had a short setting time resulting in the paste quickly bound forming granules during the mixing.

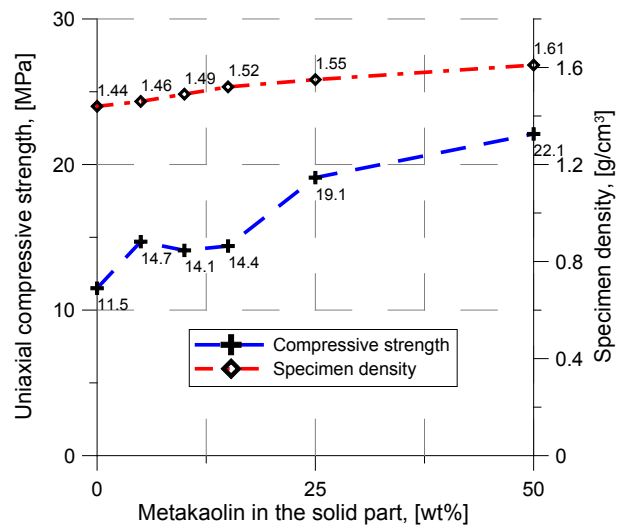


Figure 1. Effect of metakaolin dosage

The relation between the SiO₂/Al₂O₃ molar ratio of

Table 3
Characteristic particle size and SSA of FA after various grinding time

Material properties	Grinding time (min)						
	0	5	10	20	30	60	120
x ₁₀ (µm)	10.3	9.7	8.4	6.5	5.8	4	0.5
x ₅₀ (µm)	48.4	41.1	29.6	19.5	16.4	12.3	8.4
x ₈₀ (µm)	112.6	89.5	68.1	45.9	33.7	22.6	16.1
SSA (cm ² /g)	1,140.9	1,219.3	1,417.9	1,807.4	2,056.5	2,937.9	12,228.8

mixture and the compressive strength is shown in Figure 2. The $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio was changed in the range 3.67 and 7.49, which was modified with MK content in the solid part of mixture. The $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio decreased with the content of MK in solid part (due to higher Al_2O_3 content of MK). Based on Figure 2 it can be stated that increasing the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio decreased the geopolymer strength. The figure shows that the maximum compressive strength (22.1 MPa) was obtained when $\text{SiO}_2/\text{Al}_2\text{O}_3$ was 3.67. Furthermore, it can be observed that the geopolymers had almost the same strength (14.1 - 14.7 MPa) between $\text{SiO}_2/\text{Al}_2\text{O}_3$ of 5.68 and 6.77.

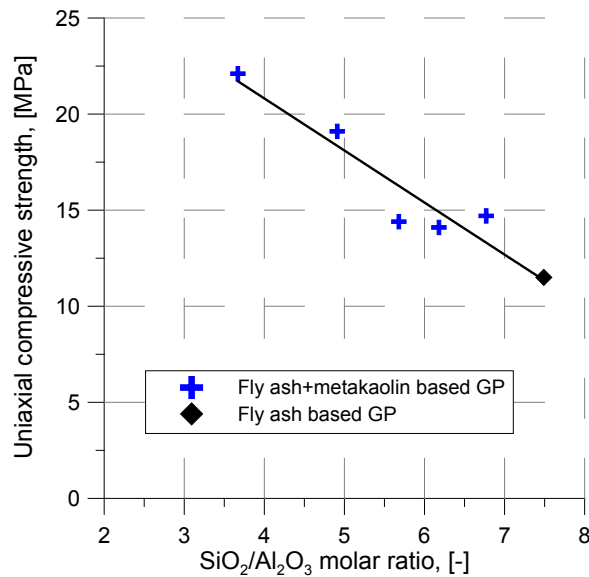


Figure 2. Effect of $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio

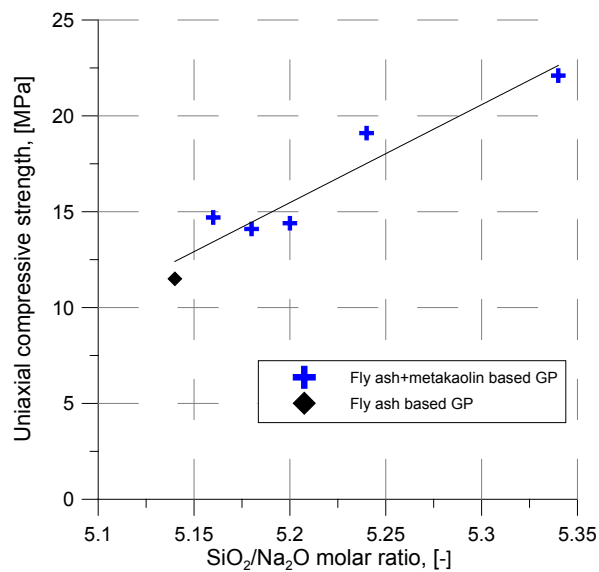


Figure 3. Effect of $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio

Figure 3 shows the compressive strength of geopolymers at various $\text{SiO}_2/\text{Na}_2\text{O}$ ratios. The ratio

was changed in the range of 5.14 and 5.34. The results show that the strength of geopolymers increased with $\text{SiO}_2/\text{Na}_2\text{O}$. The highest compressive strength (22.1 MPa) was obtained when $\text{SiO}_2/\text{Na}_2\text{O}$ was 5.34.

A similar conclusion can be drawn from Figure 4 as shown in Figure 3. Increasing $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ ratio increased the geopolymer strength. The ratio was changed in the range 0.69 - 1.45. Based on the result it can be stated that not only the SiO_2 content plays an important role in geopolymerization, but also the Al_2O_3 content.

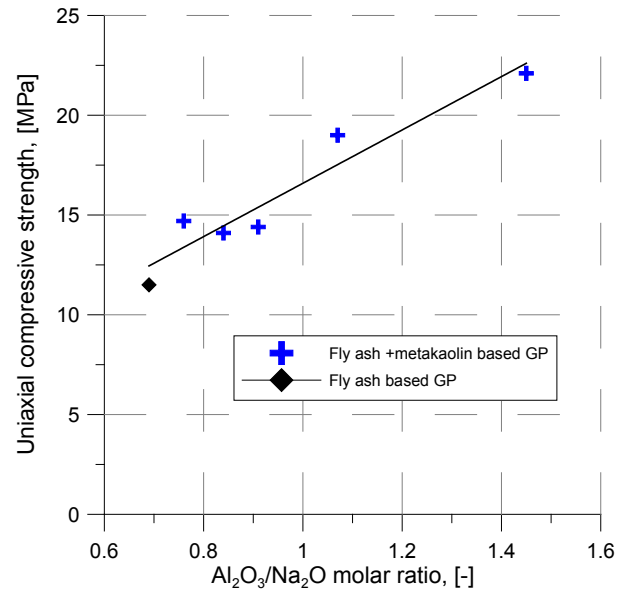


Figure 4. Effect of $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ molar ratio

4.3 Effect of grinding fineness of FA

The effect of grinding fineness of FA was investigated by metakaolin content of 25 % taking into the workability of mixture. Based on Figure 5 it can be stated that increasing the grinding time (by changing of FA fineness) increased the density and the compressive strength of the geopolymers. While the raw FA based geopolymers ($\text{SSAFA}=1,141 \text{ cm}^2/\text{g}$) had compressive strength of 19.1 MPa and specimen density of $1.54 \text{ g}/\text{cm}^3$, the geopolymers which were made using SSAFA of $12,229 \text{ cm}^2/\text{g}$ (grinding time of 120 min) had compressive strength of 25.2 MPa and specimen density of $1.64 \text{ g}/\text{cm}^3$. As a result of the milling, the specific surface of the material was increased as well, resulting in more Al and Si solved by the alkaline solution from the FA, and it was advantageous for the emergence of the geopolymer gel. Another explanation of the increasing compressive strength (and specimen density) can be that the finer particles resulted in more compact microstructure.

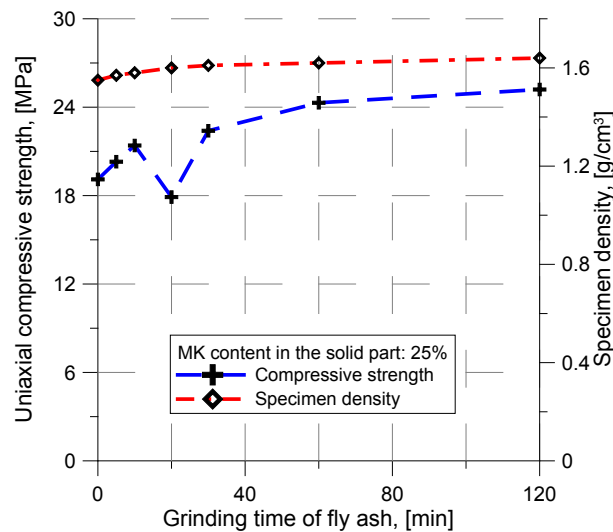


Figure 5. Effect of fineness of FA on the compressive strength and specimen density

4.4. FT-IR results

The FT-IR spectra of raw materials (FA and MK) can be seen in Figure 6. The IR spectrum of MK contains one very intense band characteristic of the internal vibrations in TO_4 tetrahedral (T=Al, Si). This peaks at around $1,064\text{ cm}^{-1}$, and is associated with T-O-Si bond (T= Al, Si) asymmetric stretching vibrations. In case of FA this peak appeared at around $1,100\text{-}1,009\text{ cm}^{-1}$ in the form of a double band. At 800 cm^{-1} , a band appeared corresponding to Al-O bending of tetrahedral Al (Granizo et al., 2000). The peak at $1,457\text{ cm}^{-1}$ corresponds to O-C-O stretching vibration, symmetrical stretching vibrations of Si-O-Si and Al-O-Si bonds are observed at 677 and 598 cm^{-1} (Swanepoel and Strydom, 2002; Panias et al., 2007).

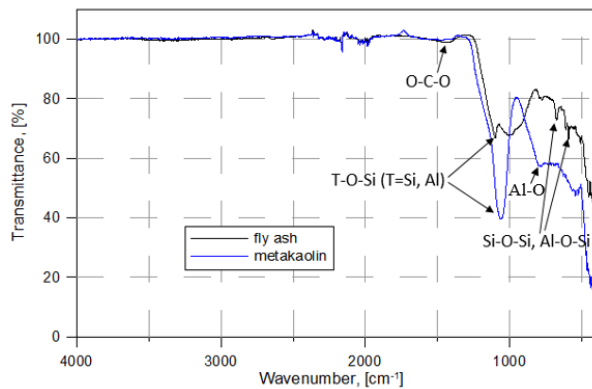


Figure 6. FT-IR spectra of raw materials

FT-IR spectra of geopolymers with various metakaolin content can be seen in Figure 7. The most characteristic difference was observed between the IR spectra of geopolymers and FA at the band of T-O-Si bonds. This band that appeared as a broad band between $1,100$ and $1,009\text{ cm}^{-1}$ in the FT-IR spectrum of FA became sharper

and shifted to lower wavenumber ($\sim 943\text{ cm}^{-1}$) in the FT-IR spectrum of FA based geopolymer, indicating the formation of a new product (the amorphous aluminosilicate gel phase), which is associated with the dissolution of FA amorphous phase in the strong alkaline solution (Swanepoel and Strydom, 2002; Panias et al., 2007). This bond is often used to determine the degree of polymerization. This peak observed between 951 and 973 cm^{-1} in the FT-IR spectra of MK containing geopolymers. The peaks that were observed in IR spectra of FA and MK between 800 and 598 cm^{-1} disappeared after geopolymerization, which is associated with the dissolution of Al and Si in the NaOH solution.

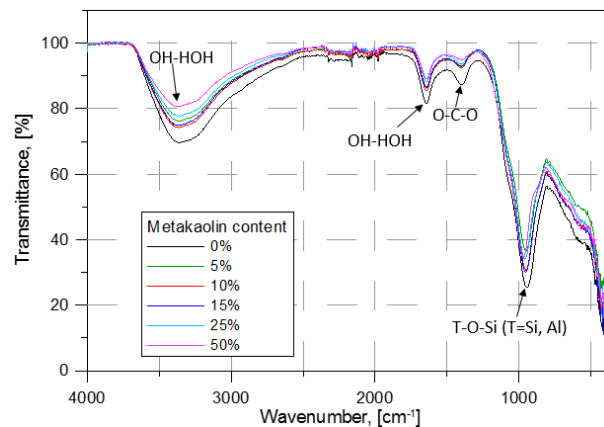


Figure 7. FT-IR spectra of geopolymers with different MK content

The broad bands which appeared in all FTIR spectra in the region of $3,370$ and $1,640\text{ cm}^{-1}$ are assigned to stretching (-OH) and bending (H-O-H) vibrations of bound water molecules, that were either surface absorbed or caught in the structure cavities (Swanepoel and Strydom, 2002; Panias et al., 2007; Ozer and Soyer-Uzun, 2015). The intensity of these bands decreased by higher MK content, which could be correlated with higher mechanical strengths.

The peak observed at $1,457\text{ cm}^{-1}$ in case of FA shifted a lower wavenumber (approximately $1,400\text{ cm}^{-1}$) in all the FTIR spectra of geopolymer, which are attributed to stretching vibrations of O-C-O bond. This band is related to carbonate formation because of alkali sodium hydroxide reacting with the atmospheric CO_2 , which is diffused on the geopolymeric materials surface (Swanepoel and Strydom, 2002). The intensity of these bands decreased by higher metakaolin content, which could be correlated with increased mechanical strengths. It is associated with higher proportion of Na^+ ion built into the geopolymer structure.

Figure 8 shows FT-IR spectra of geopolymers with different fineness of FA and MK content of 25%. Based on Figure 8 it can be stated that the spectrum of raw FA based geopolymer is separated from the other FT-IR spectra. In case of the ground FA based geopolymers, the peak intensities were higher than by the raw FA based

geopolymer. It can be also observed that the different FA fineness did not cause a significant change in the structure of the geopolymers. The position and intensity of the absorption bands were almost the same.

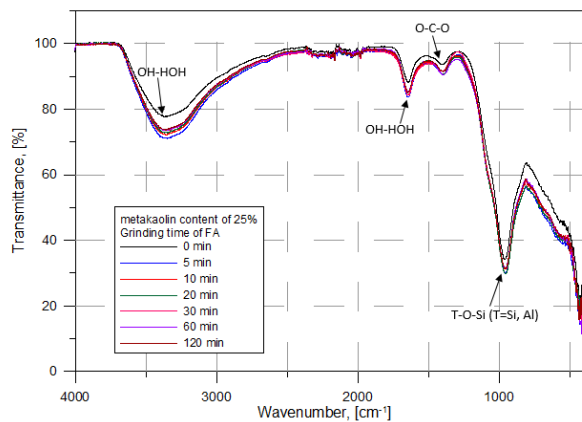


Figure 8. FT-IR spectra of geopolymers with different fineness of FA

5. Conclusions

In this work, the effects of $\text{SiO}_2/\text{Al}_2\text{O}_3$, $\text{SiO}_2/\text{Na}_2\text{O}$ and $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ molar ratios and FA fineness on the structural and mechanical properties of geopolymers are studied and the following results are drawn:

- MK had a positive effect on the compressive strength of FA based geopolymers. Increasing the MK content improved the strength. Geopolymer with the highest strength (22.1 MPa) and density (1.61 g/cm^3) was made by 50 % MK content in the solid part.
- Higher MK content resulted in lower $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of the mixture. There is a relationship between $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio and compressive strength. The lower $\text{SiO}_2/\text{Al}_2\text{O}_3$ molar ratio resulted in a higher compressive strength.
- The strength of geopolymers increased with $\text{SiO}_2/\text{Na}_2\text{O}$ and $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ ratios. The highest compressive strength was obtained when $\text{SiO}_2/\text{Na}_2\text{O}$ was 5.34, and $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ was 1.45.
- Not only did the MK content significantly influence the strength of geopolymer, but also the FA fineness. The mechanical activation of FA enhanced the reactivity and consequently the properties of the resulted geopolymer, i.e. the compressive strength was improved and specimen density increased as well. Geopolymer with the highest compressive strength (25.2 MPa) was made using SSAFA of $12,229 \text{ cm}^2/\text{g}$ (grinding time of 120 min).
- Based on the FT-IR results, it can be stated that the intensity of O-C-O bonds in IR spectra of geopolymers decreased by higher metakaolin content, which could be correlated with the

increased mechanical strengths. The higher alumina content resulted in a higher degree of Na^+ incorporation into the geopolymer structure.

- FA fineness did not cause a significant change in the structure of the geopolymers. The position and intensity of the absorption bands were almost the same.

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Uticaj sadržaja SiO_2 , Al_2O_3 i Na_2O i finoće letećeg pepela na strukturu i mehaničke osobine geopolimera na bazi lebdećeg pepela

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I Z V O D

U današnje vreme, tehnologije geopolimera na bazi sekundarnih sirovina se sve više koriste. Međutim, neke od njih (na primer na bazi lebdećeg pepela) imaju na početku nisku reaktivnost koja se može promeniti mehaničkom ili hemijskom aktivacijom, kao i dodavanjem različitih visoko reaktivnih materijala. U ovom radu je ispitivan uticaj sadržaja silicijum, aluminjum i natrijum oksida na strukturu i mehaničke osobine geopolimera na bazi lebdećeg pepela sa visokim sadržajem kalcijuma. Metakaolin (MK) je korišćen kao dodatni izvor aluminijuma i silicijuma. Dodavan je u različitim dozama (0; 5; 10; 15; 25; 50 i 75 % mase) kao zamena za lebdeći pepeo. Rezultati eksperimenta potvrđuju da na pritisnu čvrstoću geopolimera značajno utiče odnos $\text{SiO}_2/\text{Al}_2\text{O}_3$, $\text{Al}_2\text{O}_3/\text{Na}_2\text{O}$ i $\text{SiO}_2/\text{Na}_2\text{O}$. Dodavanje metakaolina je povećalo pritisnu čvrstoću geopolimera za 92 %. Osim toga, ispitivan je i uticaj mehaničke aktivacije lebdećeg pepela na strukturu i čvrstoću geopolimera u zavisnosti od sadržaja metakaolina. Na osnovu dobijenih rezultata, može se zaključiti da se mehaničkom aktivacijom lebdećeg pepela postigla veća pritisna čvrstoća. Dodavanje metakaolina i finoća lebdećeg pepela su promenili strukturu geopolimera, što je utvrđeno metodom FT-IR spektroskopije.