

IMPROVED PROPERTIES OF ALTERNATIVE BINDERS AND THEIR POTENTIAL APPLICATION FOR SOIL STABILIZATION

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Abstract

This paper investigates the influence of mechanical activation of different types of fly ash on the properties of alternative binders – geopolymers based on alkali activated fly ash. Fly ash was firstly mechanically and then alkali-activated. Mechanical activation of fly ash was conducted in a planetary ball mill. Alkali activation of fly ash was carried out at room temperature by use of sodium silicate solution as an activator. Structural changes of geopolymers were assessed by SEM and EDS analysis. Mechanical activation of fly ash led to a significant increase in strength of these alternative binders, thereby expanding their potential application, such as soil stabilization.

Keywords: mechanical activation, alkali activation, fly ash.

INTRODUCTION

Fly ash (FA) is generated as an industrial waste material in the process of coal combustion in thermal power plants. There is an ongoing demand worldwide for the use of large quantities of FA. Geopolymers based on alkali activated fly ash (FA) represent quite attractive binding material, known for high compressive strength, good durability in aggressive environments, low shrinkage and good thermal resistivity (Provis and van Deventer, 2009). On the other hand, the lack of a good foundation ground layer that could be used for construction proposes is a common problem all over the world. Geopolymers also emerged as a good alternative to traditional binders (lime and cement) for soil stabilization (Chen, et al. 2020). Portland cement is energy-intensive with high-carbon footprint. Today, more and more researches are focused on finding new solutions to improve the properties of binders in order to obtain a stabilized foundation layer.

The advantage of low carbon emission and energy consumption — which are synonymous with the use of geopolymers — are undoubtedly desirable. Thereby put them ahead of cement and lime stabilization in the face of the current heightened demand for greener methods (Jeremiah, et al. 2021). The limiting factor for wider use of FA in the synthesis of geopolymers is its low reactivity and consequent low strength gain when cured at room temperature. The reactivity of initial FA in the reaction of alkali activation can be improved by the appropriate choice of the alkali reaction conditions or by mechanical activation of FA (Marjanović et al., 2014).

In this work the reactivity of FA was enhanced by mechanical activation. The effects of mechanical activation of FA on the strength and microstructure of the resulting alkali-activated binders – geopolymers were investigated.

MATERIALS AND METHODS

Materials

In the experimental work, four samples of fly ash from thermal power plants (TPP) in Serbia were used as starting material:

1. FA TENT A, TPP “Nikola Tesla”, Unit A, Obrenovac, Serbia
2. FA TENT B, TPP “Nikola Tesla”, Unit B, Obrenovac, Serbia
3. FA Kolubara, TPP “Kolubara”, Veliki Crljani
4. FA Kostolac, TPP “Kostolac” B₁, Kostolac, Serbia

The following materials were used to prepare the alkaline activator solution:

- Sodium silicate solution (“Galenika–Magmasil”, Zemun, Serbia, 13.60% Na₂O, 26.25% SiO₂, 60.15% H₂O, with starting modulus n (SiO₂/Na₂O mass ratio) 1.93).
- Sodium hydroxide pellets (NaOH (p.a. min. 99%, “Lach-Ner”))

Mechanical activation of FA

Mechanical activation of FA (TENT A, TENT B, Kolubara and Kostolac) was carried out in a planetary ball mill (Fritch Pulverisette type 05 102, Germany). FA to ball mass ratio was 1:20 (Marjanović et al., 2014). FA samples were mechanically activated in an air atmosphere for 15 minutes, while the speed of rotation was 380 rpm.

Preparation of geopolymer samples

Starting sodium silicate modulus was adjusted to the value 1.5 by adding NaOH pellets. The concentration of the activator was 10% Na₂O with respect to the FA mass. Geopolymer mortar prisms (40 mm × 40 mm × 160 mm) were prepared by mixing the initial FA (FA) or mechanically activated FA (MFA) with alkali activator and water, and then with sand.

Table 1. Conditions of mechanical and alkaline activation of FA and curing conditions

Sample	Duration of mechanical activation (min)	Water/binder ratio	Curing condition	
			Temperature (°C)	Time (days)
FA TENT A	0	0.85	20±2 °C	1, 3, 7, 28
FA TENT B		0.80		
FA Kolubara		0.65		
FA Kostolac		0.69		
MFA TENT A	15	0.40		
MFA TENT B		0.40		
MFA Kolubara		0.40		
MFA Kostolac		0.40		

In the case of geopolymer mortars based on the initial FA (G-FA) water was added in the amount to obtain equal consistency (mortar flow measured on a flow table was 120±5 mm). On the other hand, geopolymer mortars based on mechanically activated FA (G-MFA) were all prepared with the same water/binder ratio. Water in water/binder ratio represents the total amount of water in the system, including water from the activator, while binder represents total fly ash mass and solid part of activator. FA/MFA : sand mass ratio was 1 : 3. Mortar prisms were cured at room temperature in a humid chamber (90±5% rel. humidity) for 1, 3, 7 and 28 days (Table 1). Preparation of the geopolymer pastes was performed in the same manner as the preparation of mortars, only without the sand. After the period of curing, paste samples were crushed and soak in isopropyl alcohol in order to stop further reaction.

Methods

The determination of strength of geopolymer mortars was performed using the CONTROLS ADVANTEST 9 device. Morphological characterization of the geopolymer pastes was done by SEM (VEGA TS 5130 MM, Tescan), while energy dispersive X-ray spectroscopy (EDS) was performed by INCAP-entaFET-x3 (OXFORD Instruments).

RESULTS AND DISCUSSION

Visual appearance of geopolymer mortar prisms based on FA/MFA TENT B is given in Figure 1. There are no visible cracks or deformations on geopolymer mortar prisms. The color of the geopolymer mortar is strikingly transformed by mechanical activation, from light gray (Figure 1a) to dark gray, almost black (Figure 1b). This is most likely the result of a greater dispersion of unburned carbon particles, which are known to be possible supplement for carbon black pigment (Marjanović et al., 2014).

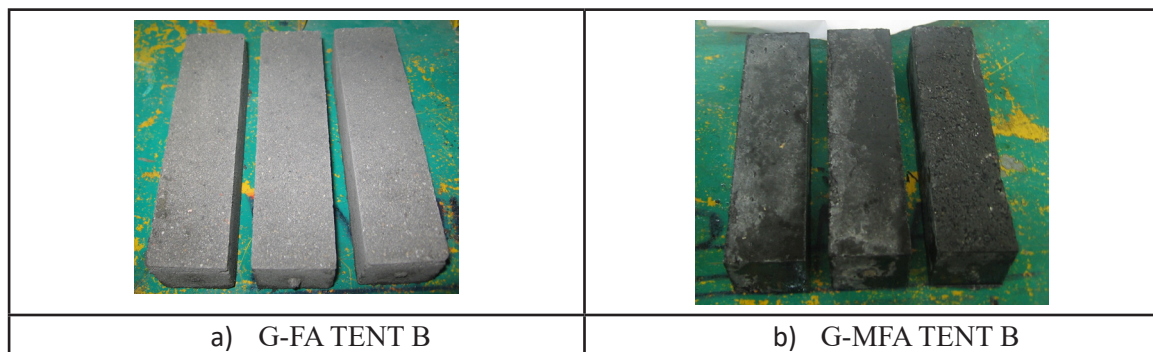


Figure 1. Geopolymer mortars based on initial FA and mechanically activated MFA

The water demand for G-MFA was considerably lower than in the case of G-FA. This can be explained by the destruction of unburned carbon particles in FA samples by mechanical activation. It is known that, when FA is used as a substitution for cement, the presence of porous unburned carbon particles influence the higher water demand for standard consistency of mortars (Marjanović et al., 2014).

The mechanical strength of geopolymers based on FA and MFA after curing at room temperature are given at Figure 2. Geopolymers with very low flexural and compressive strength were obtained by alkaline activation of the initial FA

after curing at room temperature. Geopolymer mortars based on the initial FA were fragile and could easily broke under the hand. In contrast to that, the mechanical strength of geopolymers based on MFA, exceeded the values of 50 N/mm². Based on the presented results, it can be unambiguously concluded that mechanical activation affects the drastic enhancement of FA reactivity in the process of alkali activation.

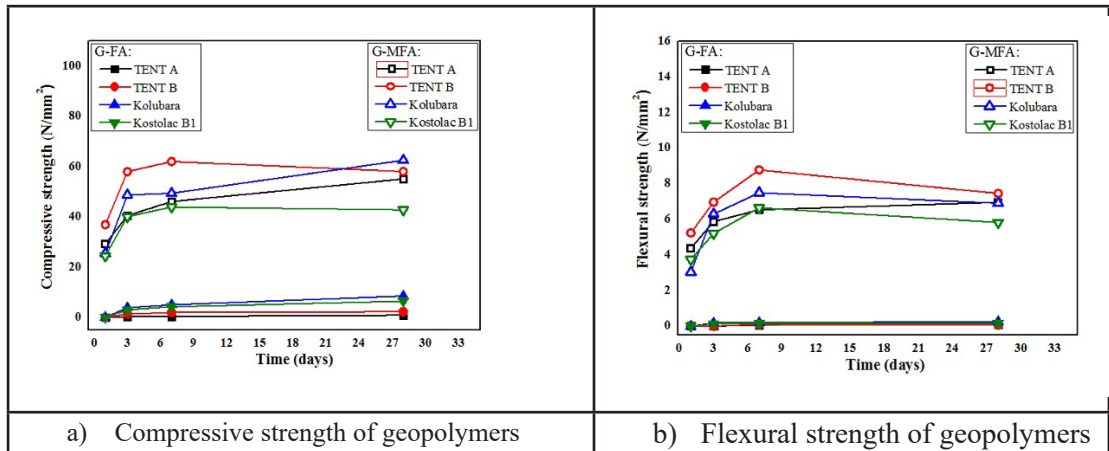


Figure 2. Compressive and flexural strength of geopolymers based on alkali activated FA/MFA

The curing time does not significantly affect the development of the strength of the mortar based on the initial FA, because even after 28 days there is no increase in strength. From this, it can be clearly concluded that the alkali activation of the initial FA at room temperature is extremely slow, i.e. the reactivity of FA at room temperature is very low. In contrast, alkali activation of mechanically activated (MFA) resulted in geopolymers which compressive strength after 1 day of curing was in all cases higher than 20 N/mm², and after 3 days of curing higher than 40 N/mm². According to the quality requirements defined by the SRPS EN 197-1:2013 standard, the compressive strength of Portland cement-based mortar after 28 days of curing must be in the range of 30-50 N/mm², depending on the cement class. Therefore, geopolymer mortars based on mechanically activated MFA after 3 days of curing show strengths comparable to the strengths that Portland cement-based mortars develop after 28 days. With the extension of the curing time up to 7 and 28 days, the compressive strength values slightly increase for geopolymers based on MFA. The geopolymer based on MFA TENT B achieved the highest flexural and compressive strengths in relation to all tested

samples. The compressive strength of this mortar after 7 days (62.08 N/mm²) is more than 30 times higher than the initial FA TENT B mortar (1.99 N/mm²) cured under the same conditions. The presented results clearly show the influence of the mechanical activation of FA on the achieved geopolymer strengths and improved properties.

The differences in the microstructure of geopolymers derived from FA/MFA can be clearly distinguished (Figure 3). The geopolymers based on FA appear highly heterogeneous, with loosely structured precipitates and cavities in the structure (Figure 3 a,c,e,g), originating most probably from the evaporation of water. In contrast to this, the geopolymers derived from MFA showed very dense and compact microstructure, implying the formation of aluminosilicate gel, as the main reaction product, to a greater extent (Figure 3 b,d,f,h).

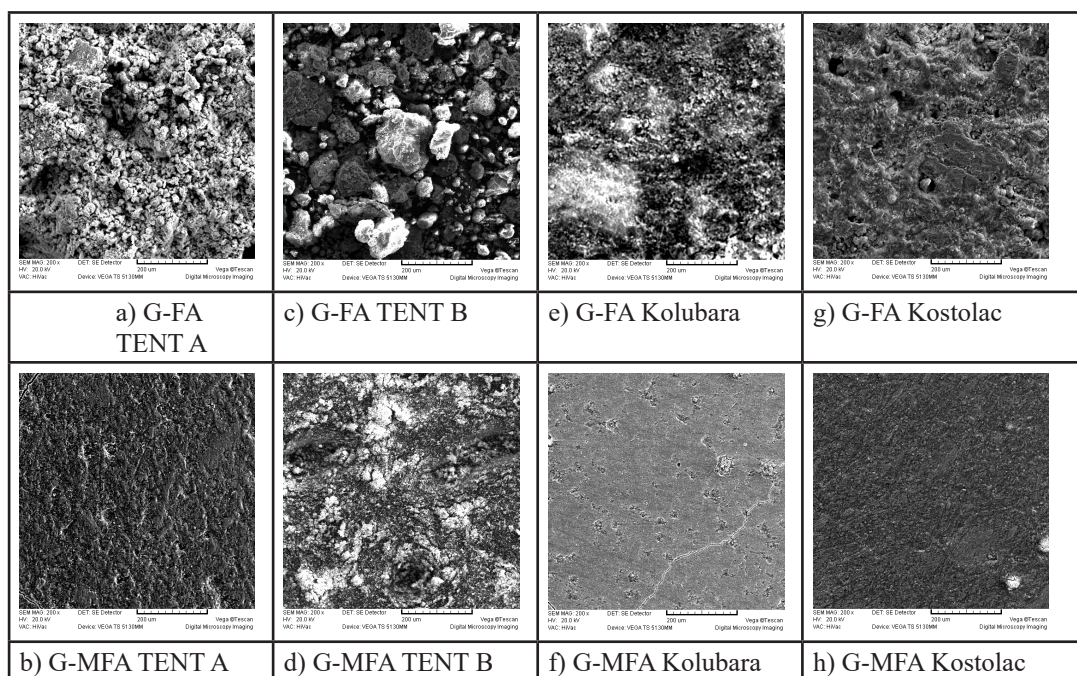


Figure 3. SEM of geopolymers based on initial FA and mechanically activated MFA

The results of EDS analysis (Figure 4) showed that in the geopolymers based on MFA there was a somewhat higher Al/Si ratio, indicating the higher proportion of incorporated aluminum in the aluminosilicate gel phase. Greater extent of aluminum incorporation can be related to the greater availability of this component during the alkali activation. It is known that the amount of available aluminum and the rate of its release during the reaction is very important factor,

since it highly affects the geopolymer gel properties (Provis and van Deventer, 2009). Greater availability (the faster release) of aluminum during the reaction influences the gel homogeneity, contributing to the formation of a more homogeneous gel and better strength development. With greater aluminum availability, faster alkali activation, i.e. formation of aluminosilicate gel occurs and the gel network eventually consists of more aluminum component (Provis and van Deventer, 2009).

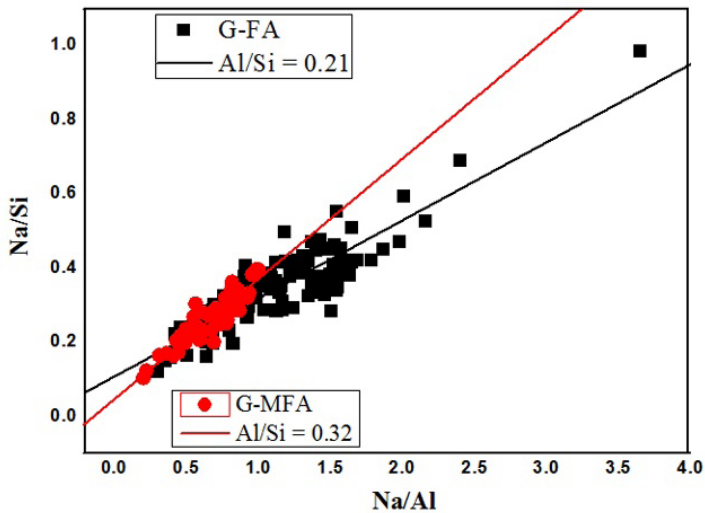


Figure 4. EDS results - ratios of major elements in the geopolimers based on FA/MFA

CONCLUSION

In this paper, the influence of mechanical activation of FA on mechanical properties and structure of geopolymer mortars was investigated. Based on the results presented the following conclusions can be drawn:

- Mechanical activation influences lower water demand in the synthesis of geopolymers
- Mechanical activation seems to affect the greater availability of the aluminum, contributing to the formation of more homogeneous aluminosilicate gel.
- Mechanical activation in a duration of 15 min drastically enhances the FA reactivity in the process of alkaline activation at room temperature, which is confirmed by exceptional increase of mechanical strength of geopolymers based on MFA.

The main advantage of binding materials synthesized by alkaline activation of FA is primarily the significantly reduced emission of carbon dioxide into the atmosphere during their synthesis. Of great importance is the possibility of synthesis at room temperature, which represents a contribution to significant energy savings compared to the synthesis process of Portland cement clinker, which takes place at temperatures of approximately 1500 °C. The use of industrial waste (FA) as a starting material for the synthesis of binders also has positive effects from an environmental point of view, such as the saving of natural mineral raw materials, reduced environmental pollution and the valorization of industrial waste material. An additional advantage and importance of the mechanical activation of FA is the possibility of using the entire amount of this material in the alkali activation.

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