

Article

Study on Mechanical Properties of Heap Deposited Fly Ash Based Geopolymers with Different Alkaline Activator Properties

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Abstract: Geopolymers can improve environmental protection and are therefore considered a 21st-century material that requires special attention. The main objective of this study was to examine the utilization of fly ash deposited on a heap as a potential raw material for the geopolymerization process. In this work, flexural and compressive strengths, as well as water absorption of geopolymers, were evaluated based on alkaline activator properties. Na₂O content, SiO₂/Na₂O molar ratio and fly ash-to-water ratio were key observed factors. Mechanical strengths tests were conducted after 7, 28, 90, 180, and 360 days. The research findings showed that geopolymer cured at appropriate conditions and with a suitable alkaline activator can reach a compressive strength of more than 55 MPa. The presented study highlighted the possibility of the efficient use of fly ash to produce useful materials, i.e., geopolymers, which contribute to environmentally friendly solutions. This approach is fully in line with the principles of sustainable development.

Keywords: geopolymer; fly ash; mechanical properties; compressive strength; flexural strength; environmentally friendly materials; sustainable development; green alternative; environmental solutions



Citation: Sisol, M.; Marcin, M.; Dvořák, K.; Sudřová, M.; Ivanková, V. Study on Mechanical Properties of Heap Deposited Fly Ash Based Geopolymers with Different Alkaline Activator Properties. *Buildings* **2022**, *12*, 1780. <https://doi.org/10.3390/buildings12111780>

Academic Editor: Tomáš Dvorský

Received: 21 September 2022

Accepted: 20 October 2022

Published: 24 October 2022

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1. Introduction

In line with the objectives set by the European Union, which also include achieving an economy with net-zero greenhouse gas emissions, it is desirable to reduce the amount of carbon dioxide released into the atmosphere and to avoid loss of the protection of the ozone layer [1,2]. In addition, climate action, responsible production, and sustainable industry are among the 17 Sustainable Development Goals (SDGs) [3]. However, the huge increase in carbon dioxide and waste production in recent decades is a crucial obstacle to sustainable development and sustainability [4]. For these reasons, solutions must be sought in various areas, and manufacturing and processing are no exception. In this context, a significant amount of carbon dioxide is emitted during the calcination process in the process of obtaining ordinary Portland cement (OPC) in which limestone is transformed to lime. Simultaneously, burning fossil fuels to deliver thermal energy for calcination leads to global warming.

In recent years, this issue has become increasingly prevalent. As a result, even a modest decrease in Portland cement usage will significantly reduce carbon dioxide emissions and lead to major environmental benefits. On this basis, it is possible to emphasize the huge potential of geopolymers with respect to environment protection. Alternative binders are required to minimize carbon dioxide emissions and to reduce greenhouse gas emissions [5–7].

This has prompted studies into finding new environmentally friendly, cementitious products that have high strength and longevity while using a reasonable amount of energy resources in the manufacturing process. Many studies have investigated geopolymer and

alkali-activated materials (AAM), which are synthesized using fly ash, metakaolin, or slag, as an alternative to cement with alkaline activator over the last two decades [8–11].

In more detail, geopolymers are semi-crystalline alkali-activated aluminosilicates formed when aluminosilicates react in alkaline media [9]. Geopolymers have a similar chemical composition to zeolites, but their microstructure is amorphous [12]. The development of geopolymer also encourages the use of aluminosilicate materials as the key binding material in geopolymer, such as fly ash, a natural industrial by-product from coal combustion in power plants [13]. The structure of fly ash also entitles it to be included in the manufacture of construction materials [14]. A geopolymerization process is an effective approach for converting a wide variety of aluminosilicate waste materials into usable binders that can be used for building materials. A significant number of naturally occurring and industrially generated aluminosilicates are used for geopolymer synthesis [15,16].

Fly ash is an undesirable byproduct formed by the burning of pulverized coal. Industrialization has contributed to the production and deposition of fly ash in vast amounts over the years [5]. According to statistics, about 780 million tons of fly ash are generated in the world each year, but only about 17–20% are used [17–19]. The primary use of fly ash is in the manufacture of OPC. Furthermore, a recent study shows that fly ash can be used to make carbon nanotubes or as a source of rare elements [20]. The use of fly ash in geopolymers helps to alleviate the growing environmental impact of aluminosilicate minerals including fly ash and slag [21].

The structure of the fly ash along with the alkaline activator have a major impact on the final properties of the geopolymer, according to References [13,22]. There are several types of alkaline activators that can be used to activate fly ash. Sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3), as well as potassium hydroxide (KOH) and potassium silicate, are widely used alkaline activators in the geopolymerization procedure (K_2SO_3) [23]. The molarity of NaOH and Na_2SiO_3 in the alkaline activator solution is one of the parameters that affect the chemical reactions of the geopolymer paste. Numerous scientific studies have observed the impact of the molarity ratios of NaOH and $\text{Na}_2\text{SiO}_3/\text{NaOH}$ on the compressive and flexural strengths of geopolymers [24–28]. Oxide composition of precursors has a direct influence on the compressive and flexural strength [29] as well as the structural and rheological characteristics of raw material [30]. The alkaline solution represents an essential role in the production of geopolymers with high mechanical strength. Its choice is primarily based on its reactivity and the cost of the materials used.

The Si/Al ratio, hydroxide concentration, efficiency of the geopolymerization reaction, and curing conditions all have a significant impact on the mechanical properties of geopolymers. Based on the materials used or the molar ratio, geopolymers may have a variety of mechanical, chemical, and thermal properties [31–34]. Some of these properties are high initial strength, resistance to chemical attacks from substances like acids or sulfates, thermal and fire stability at high temperature, as well as resistance to freezing [35]. As a result of all of these outstanding qualities, they are interesting renewable structural materials for the future. Since they are a non-flammable material with reasonably good insulation, geopolymers are an ideal alternative to materials such as polystyrene and glass [36].

At this point, it is also possible to speak of a circular economy with great opportunity for sustainable development. The circular economy is an economic model that bears in its name the policy of managing resources in one round, in circulation, continuously round and round, until the maximum possible use and closure of material flows. Accordingly, fly ash and other waste materials generated by the industrial and manufacturing sectors can be included in this circle and thus be used to make new products, which is in line with the circular economy. In fact, construction is a highly energy-intensive sector and one of the main sources of environmental degradation. On this basis, there is a great need for the research and development of new and sustainable approaches [37]. Evidence shows that new building materials based on alkaline activation technology are very promising and provide environmental and ecological benefits, especially in relation to products that need significant technological properties and durability [38]. In this context, geopolymers are a

greener substitute for cement to significantly reduce environmental degradation [37,39]. Their use leads to large savings in energy and primary materials, which corresponds to sustainable production. Also, it reduces the need for OPC consumption and the consequent production of carbon dioxide [4].

Theoretically, this study contributes to the knowledge of environmentally friendly materials, i.e., geopolymers, in the conditions of Slovakia. It also helps policy makers to better understand the issue and develop more effective strategies and mechanisms to improve environmental protection, remove obstacles, and achieve sustainable development. Practical use is obvious in the construction industry. The experimental evidence of this study may encourage civil engineers in Slovakia to use green alternatives, which is currently cautious. This issue has long been neglected and underestimated in practice in Slovakia, and therefore this study can be a valuable platform for proactive and evidence-based solutions.

In Slovakia, almost all of the heating plant waste, especially fly ash, is not utilized. It can be concluded that little attention is paid to this issue in Slovakia, which is reflected in the lack of evidence-based practices [40]. In this paper, the utilization of fly ash from Slovakia, mainly deposited in a heap, which creates an environmental problem, was used as a potential aluminosilicate material for the geopolymerization process. In this study, the created alkali-activated materials are regarded the same as geopolymers. Examined mechanical properties, such as compressive strength, flexural strength, and water absorption, were observed over the course of a year. The influence of different parameters of alkaline activators, such as Na_2O amount, SiO_2 -to- Na_2O molar ratio, and water-to-fly ash ratio, was the main observation point in terms of their effect on the above-mentioned mechanical properties of geopolymers.

2. Materials and Methods

The main objective of this study was to examine the utilization of fly ash deposited in a heap as a potential raw material for the geopolymerization process. The raw material for this experiment was a heap of deposited fly ash (Class F) (HDFA) from the District Heating Plant in Košice, Slovakia using melting boilers with a temperature of 1400–1550 °C. HDFA was collected from an upper layer sludge bed. Chemical and mineralogical analyses are shown in Tables 1 and 2. Based on its chemical composition provided by analysis, the $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio is 2.99. The particle distribution of HDFA is shown in Figure 1.

Table 1. Chemical analysis of a raw material.

wt.%	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Other	LOI
HDFA	45.63	15.28	8.74	3.62	1.68	2.68	23.87

Table 2. Mineralogical analysis of a raw material.

wt.%	Amorphous	Hematite	Quartz	Mullite
HDFA	83.52	1.57	6.42	8.46

Chemical analysis was performed by atomic absorption spectroscopy (AAS) (Perkin Elmer instrument, model 1100B). For mineralogical analysis, an X-ray diffractometer URD-6/ID 3003 (Rich. Seifert-FPM, Germany) was used, under the following conditions: X-ray radiation $\text{Co K}\alpha$, high voltage of 40 kV, current 35 mA, step scan mode with the step of $0.05^\circ 2\theta$, time per step 3 s.

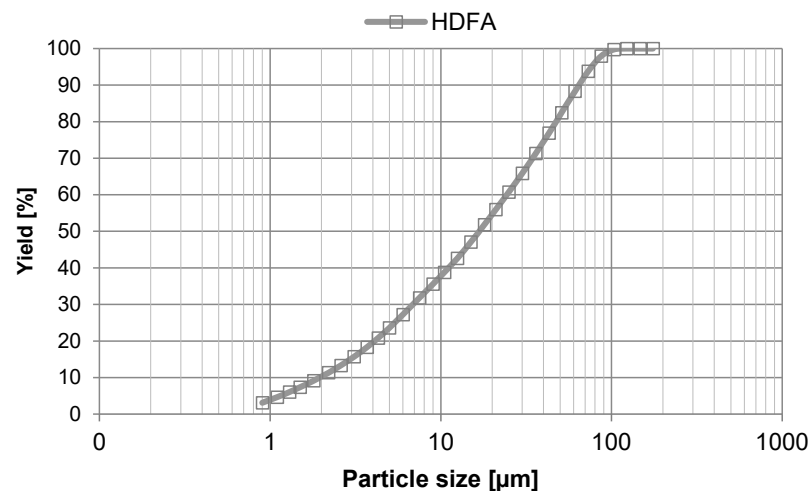


Figure 1. Particle size distribution of HDFA.

The particle size distribution of raw material is one of the most important characteristics that affect its reactivity [41,42]. The size distribution of HDFA was obtained using a Diffraction Spectrometer Helos 12 LA (Sympatec GmbH, Clausthal-Zellerfeld, Germany) with a dispersing Feeder Rodos 12 SR for dry, pulverulent solids. Particle size analysis was in the range of 0.9–170 µm. HDFA 50% comprehends particles smaller in size than 17 microns.

Scanning electron microscopy (SEM) was used to investigate the morphology of the HDFA (Figure 2) using a Jeol JSM 7000F device.

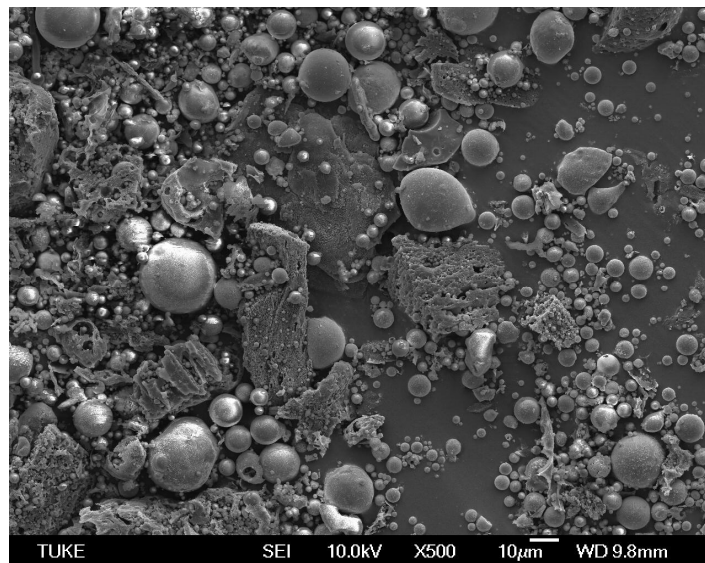


Figure 2. SEM picture of HDFA.

The mechanical properties of geopolymers were examined depending on the alkaline activator used. The effect of Na_2O , the effect of $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio, and the water-to-HDFA ratio were observed. Samples were prepared by mixing HDFA with an alkaline activator. The $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio (Ms modulus) in alkaline activator was adjusted by NaOH addition to Na–water glass. The alkaline activator was made by combining NaOH solid pellets with Na–water glass and distilled water. Sodium–water glass (Kittfort Praha Co.) with a density of 1.328–1.378 g/cm^3 was used. The sodium–water glass contains 36–38% Na_2SiO_3 and the molar ratio of $\text{SiO}_2/\text{Na}_2\text{O}$ is 3.2–3.5. Solid NaOH with a density of 2.13 g/cm^3 (Kittfort Praha Co., Neratovice, Czech Republic) containing at least 97–99.5% of NaOH.

The design of the experimental work was divided into 3 parts. Each part of the experiment was conducted with the same conditions, but the alkaline activator varied. Table 3 describes the composition of each tested sample with highlighted varied parameter.

Table 3. Design of experiment.

Material	Na ₂ O	SiO ₂ /Na ₂ O	W
HDFFA	6	1.25	30
HDFFA	7	1.25	30
HDFFA	8	1.25	30
HDFFA	9	1.25	30
HDFFA	8	0.75	30
HDFFA	8	1	30
HDFFA	8	1.25	30
HDFFA	8	1.5	30
HDFFA	8	1.65	30
HDFFA	8	1.25	30
HDFFA	8	1.25	32
HDFFA	8	1.25	35

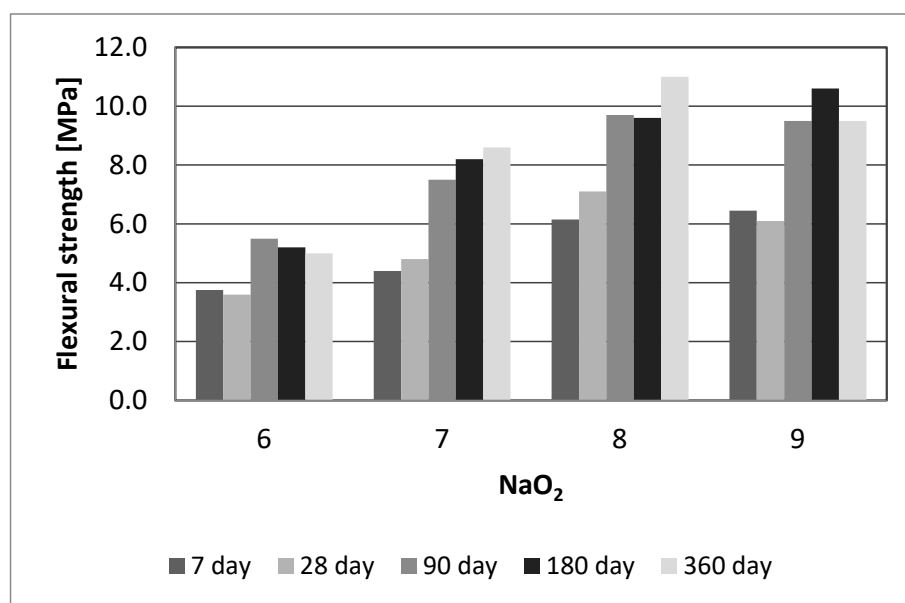
Altogether, 12 samples were prepared with a different alkaline activator. The amount of Na₂O ranged from 6 to 9 (%), the SiO₂/Na₂O molar ratio (mol/mol) ranged from 0.75 to 1.65, and water to HDFFA ratio (W) varied from 30 to 35 (%). HDFFA mixture with alkaline activator was stirred for 10 min before the homogenous mixture was formed. The mixture was poured into molds with dimensions of 40 × 40 × 160 mm and compacted on a vibration table. Samples were then cured in a hot air-drying chamber for 6 h at 80 °C. Following that, the samples were taken out of the forms, labeled, and deposited in laboratory conditions with room temperature (20 °C) before the mechanical properties' tests. Mechanical strength measurements were made following the Slovak standard STN EN 12390-3, which indicates that compressive and flexural strengths were tested after 7, 28, 90, 180, and 360 days. The same three samples were split in half to assess flexural strength, and six halves were tested for compressive strength. The mechanical strengths of the cured samples were analyzed using the Form + Test MEGA 100-200-10D hydraulic device. A part of the samples was stored at laboratory temperature for 28 days before undergoing a water absorption test following Slovak Standard STN 73 1316.

3. Results and Discussion

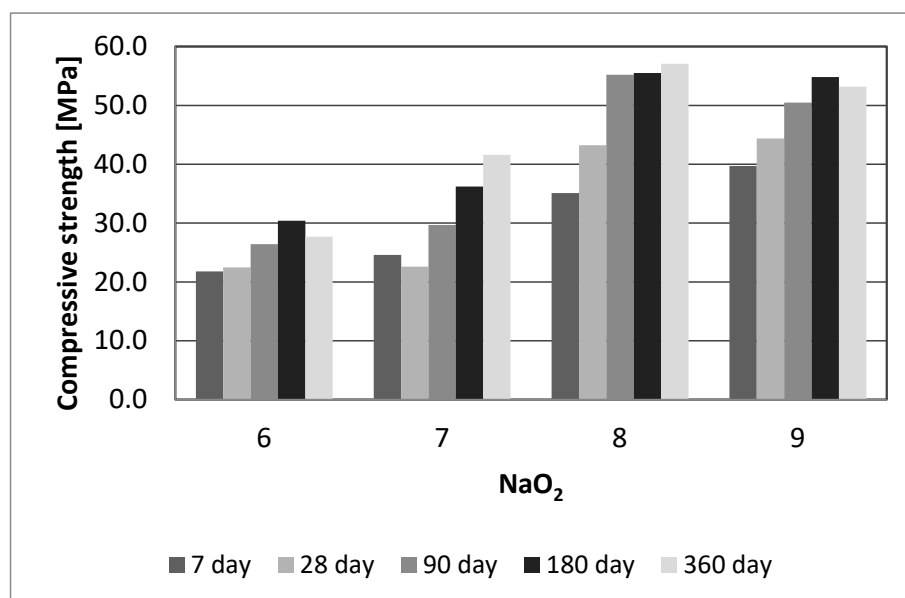
3.1. Effect of Na₂O Amount

The first series of strengths and water absorption tests were focused on Na₂O concentration changing in alkaline activator solution. This concentration was in the range of 6–9%. Figure 3 shows all flexural and compressive test results.

The geopolymer with the lowest strength was obtained with 6% Na₂O in the mix. After 360 days, the flexural strength was 5.0 MPa, and the compressive strength was 27.7 MPa. This is a factor that noticeably affects the alkaline solution's content is the concentration of an alkaline solution. Higher strength is acquired by applying an alkaline solution of a higher concentration, which indicates that with lower concentrations, the strengths were lower [43,44]. A sample with 8% Na₂O in the mixture had the highest strength, with a flexural strength of 11.0 MPa after 360 days and a compression strength of 57.1 MPa. The resulting strength difference was approximately twice as high. Intense increases in mechanical strength occurred during the first 90 days when the samples reached their maximum strength. Their strengths did not change significantly, except for a sample containing 7% Na₂O, for which strength increased throughout the testing.



(a)



(b)

Figure 3. Flexural strengths (a) and compressive strengths (b) results with different Na₂O amounts in the alkaline activator.

Water absorption results with different Na₂O amounts in alkaline activator are shown in Figure 4. The increased amount of Na₂O in the mixture had a positive effect on the water absorption of the cured geopolymer samples. Samples with 8 and 9% Na₂O had a very similar trend of water absorption over time and reached the lowest values. The water absorption of the sample was 6.68% with 8% Na₂O in the mixture and 6.71% with 9% Na₂O. The sample with 6% Na₂O in the mixture (12.98%) had the highest water absorption. Again, samples with higher strength were shown to have lower water absorption.

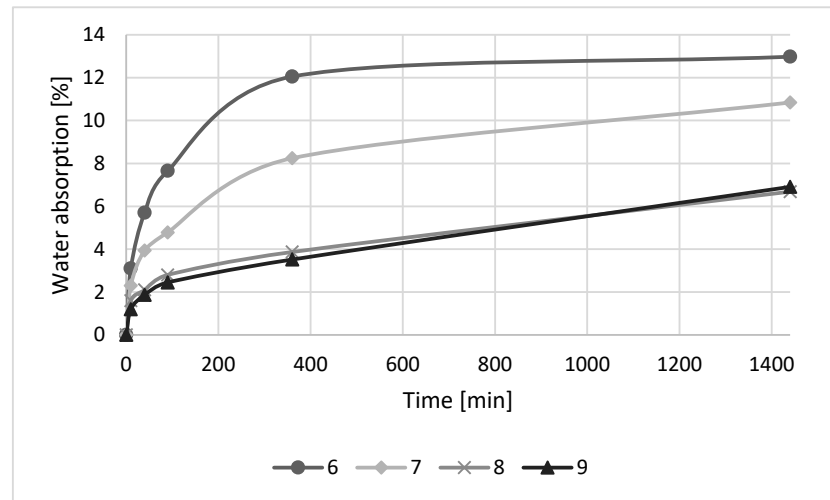
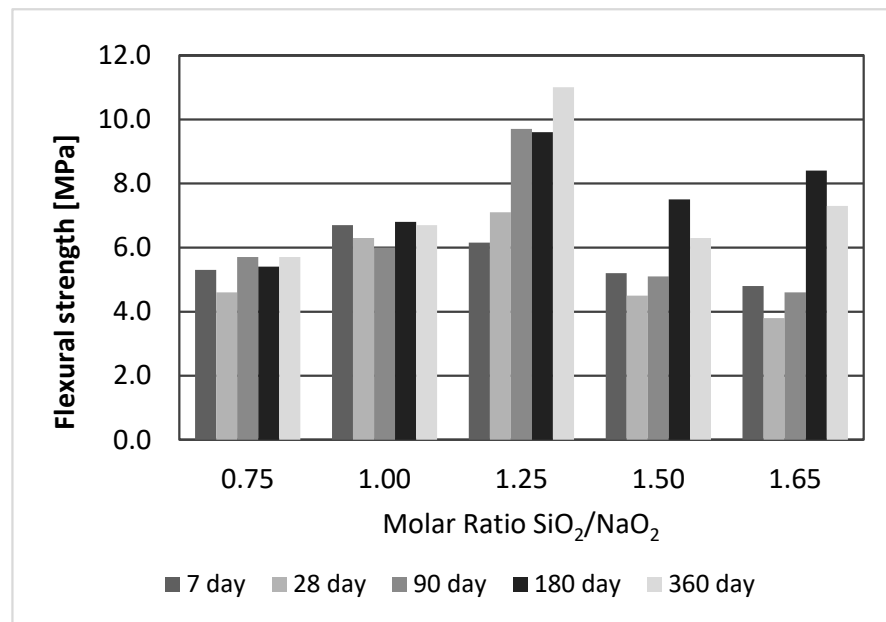


Figure 4. Water absorption results with different Na₂O amounts in the alkaline activator.

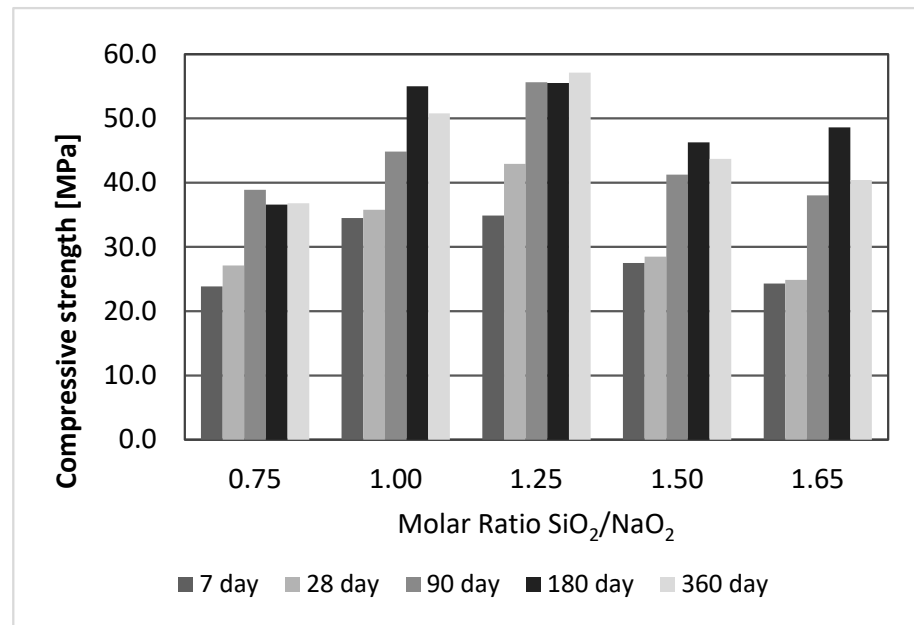
3.2. Effect of SiO₂/Na₂O Molar Ratio

The effects of different alkaline activators on the compressive and flexural strengths of samples with the different SiO₂/Na₂O molar ratios are shown in Figure 5. The values of flexural strength of HDFA geopolymers increase with SiO₂/Na₂O molar ratio from 0.75 to 1.25. However, increasing the SiO₂/Na₂O molar ratio further reduces the flexural strengths achieved. Higher amounts of SiO₂ in the alkaline activator impacted the flexural strengths, hence the strengths were decreased. The highest value was observed after 360 days and is equal to 11.0 MPa (1.25 sample).



(a)

Figure 5. Cont.



(b)

Figure 5. Flexural strengths (a) and compressive strengths (b) of samples with different SiO₂/Na₂O molar ratio.

As the SiO₂/Na₂O molar ratio was increased to 1.25, the compressive strength increased, but beyond that value, compressive strength decreased. In this case, only one sample obtained the highest strength after 360 days, and the highest value reached 57.1 MPa. The sample's (1.25) strength did not increase with time, since after 90 days, the strength was 55.6 MPa, meaning the compressive strength remained roughly the same.

The water absorption test was examined over 24 h. These results are presented in Figure 6. Water absorption reflected compressive strength results, with samples with higher compressive strengths achieving better and lower water absorption. The lowest percentage was 6.84% (12.5 sample).

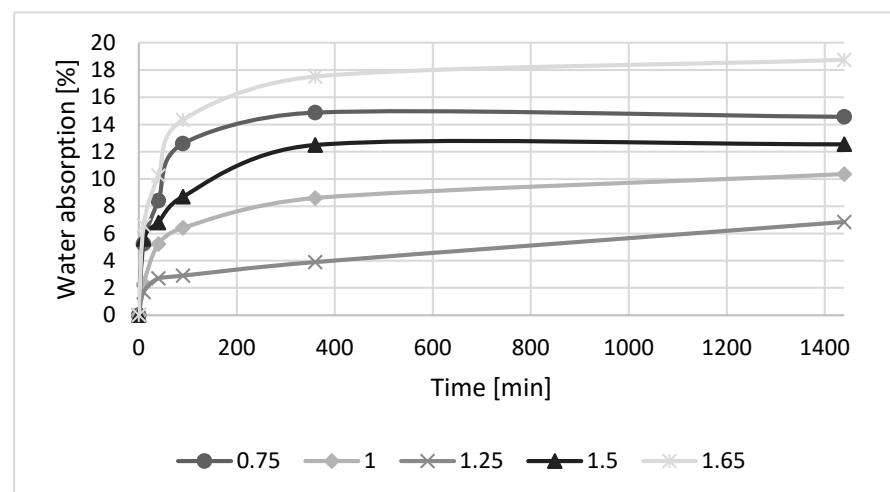
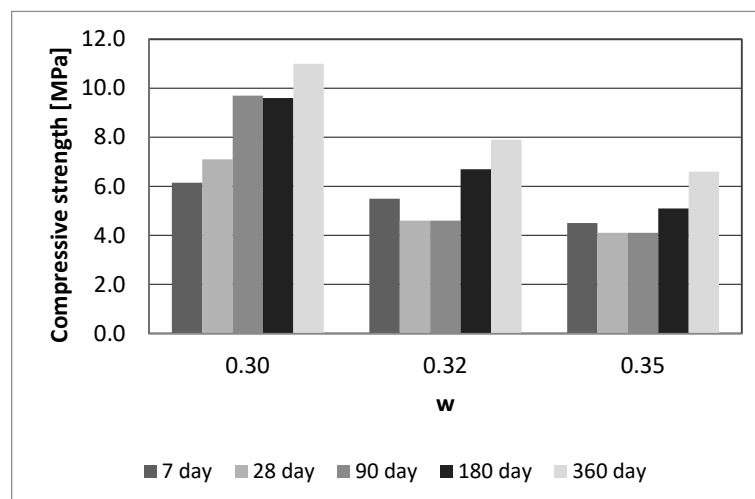


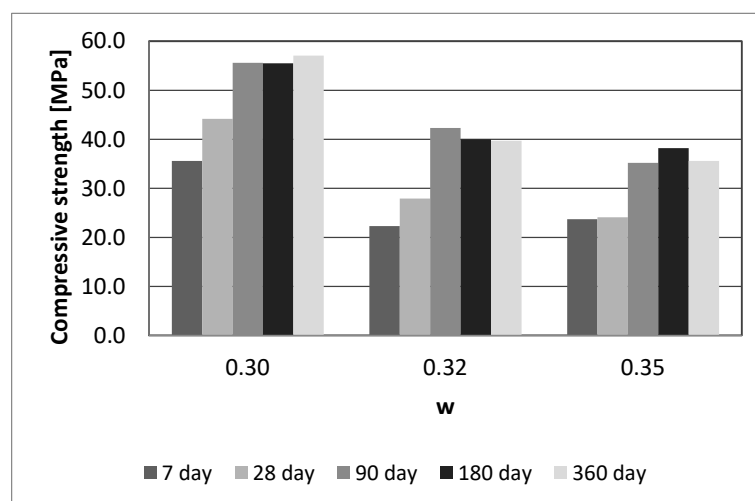
Figure 6. Water absorption results with different SiO₂/Na₂O molar ratio.

3.3. Effect of HDFA/Water Ratio

In this series of experiments, the effect of the HDFS/water ratio in the mixture on the resulting properties of geopolymers was studied. The amount of water (w) in the mixture ranged from 30 to 35%. The results of the flexural and compressive strengths are in Figure 7 and water absorption in Figure 8.



(a)



(b)

Figure 7. Flexural strengths (a) and compressive strengths (b) strength results with different HDFS/water ratios in the mixture.

An increase in the amount of water in the geopolymer mixture has a negative effect on the development of the strength of the geopolymer samples. With a constant amount of Na_2O in the mixture, the molarity of the sodium hydroxide used decreases, and the mechanical strength of the geopolymer samples decreases. Another factor contributing to the weakening of geopolymer samples is porosity, which is caused by a greater number of water-filled pores in a product with higher initial water content. The logical consequence of increasing the porosity of the synthesized geopolymers is their reduced strength. The geopolymer sample with the least amount of water in the alkaline activator solution ($w = 30\%$) had the greatest mechanical strength. Its flexural strength was 11.0 MPa and compressive strength was 57.1 MPa after 360 days of storage in laboratory conditions.

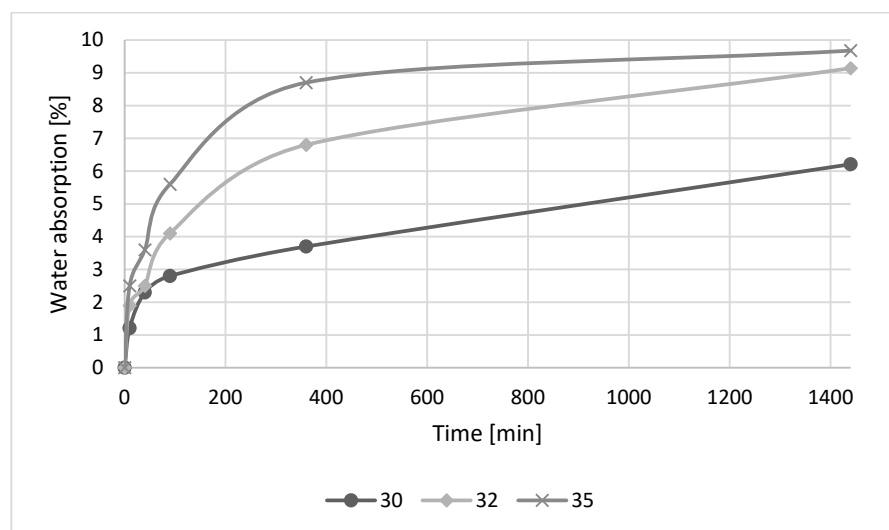


Figure 8. Water absorption results with different HDFS/water ratios in the mixture.

The development of the water absorption of the formed geopolymer samples is also affected by an increase in the amount of water in the geopolymer mixture. The sample with the lowest water absorption, 6.21 percent, had $w = 30\%$. When the amount of water in the geopolymer mixture was increased, the water absorption of the geopolymer samples increased as well. The sample with $w = 35\%$ had the highest absorptency of up to 9.68%.

Although geopolymers are an extensively researched topic, there is a constant need to improve the mechanical performance and properties of geopolymers due to the relentless search for environmentally friendly solutions. In this context, it is generally known that fly ash-based geopolymers have very good properties such as good fire and chemical resistance, high compressive strength development, low thermal contraction, and conductivity [11,24,45,46].

In this study, the composition of the alkaline activator significantly affected the resulting properties of the geopolymers. Increasing the amount of Na_2O in the mixture, at a constant $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio and amount of water, resulted in an increase in strength and a decrease in the water absorption of the geopolymers. Similar results were revealed by [47], who confirmed that higher compressive strength can be obtained with higher Na_2O content, but also with higher curing temperatures and longer pre-curing periods at relatively low temperature. These findings indicate that Na_2O can be considered as the major alkali oxide required for strength development [48]. The amount of sodium hydroxide in the activating solution mainly affects the rate of release of silicon and aluminum from the fly ash into the solution [41], because a partial role of the base is to increase the solubility of fly ash. In the base environment, the acidic hydroxyl groups on the surface of the quartz are neutralized, which allows for the breaking of Si-O-Si bonds [42]. Increasing the Na_2O amount in previous work shows the positive effect on mechanical strengths [48,49]. The mechanical strengths of the prepared geopolymers improved as the amount of sodium water glass in the alkaline activator solution increased while the amount of Na_2O and water in the mixture remained constant. This is consistent with the findings of [50], who revealed that the amount of sodium–water glass had effects on the properties of their samples. At the molar ratio $\text{SiO}_2/\text{Na}_2\text{O} = 1.25$, the maximum compressive strength and lowest water absorption are achieved. The strengths of the geopolymers decreased as the volume of sodium water glass was increased, while the water absorption of the geopolymers increased. Higher $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratios have been shown to reduce the strengths due to sodium silicate diluting the alkalinity, resulting in less precursor dissolution during geopolymerization [34]. The amount of water in the mixture had a huge effect on the mechanical strength of the geopolymer materials that were produced. As the volume of water in the mixture was increased, the mechanical strength of the mixture decreased and the water absorption of the geopolymers increased. A 2% increase in the volume of water

in the mixture resulted in a 30% reduction in mechanical strength and a 30% increase in water absorption. The degradation of geopolymer mechanical properties may be, in part, due to the fact that as the amount of water in the mixture increases while the amount of NaOH remains unchanged, the concentration of NaOH decreases, decreasing the ash particle dissolution rate. Another explanation may be porosity, which is caused by a greater number of water-filled pores in a substance with higher water content. Increasing the porosity of geopolymers has the logical result of lowering their strengths [51].

Overall, fly ash-based geopolymers prove to be a promising material due to the obtained compressive strength, environmental, and economic aspects. It is possible to speak of both environment protection and cost savings [52]. Currently, some geopolymers are already replacing naturally occurring materials due to their unique properties, but the production of carbon dioxide from OPC remains an open and persistent problem, as does the way fly ash is processed [4,53]. Over time, much more stringent emission reduction measures can be expected to slow down global warming, as it is impossible to stop it altogether [52]. Geopolymers help to reduce the carbon footprint and address sustainable growth issues. The role of policy makers is therefore to promote the use of geopolymers in practice and to encourage civil engineers to favor this still little-known alternative in Slovakia. The results of this study provide them with a valuable platform. It should be borne in mind that the sustainable development of new materials can ensure the industry's competitiveness, its stable economic growth, and a healthy environment [39]. Before effectively implementing environmentally friendly approaches, it is first necessary to know the possibilities, and this study offers one of them. The presented findings show the possibility of efficient use of fly ash for the production of useful materials, i.e., geopolymers, which contribute to greener solutions. Based on the results, geopolymers can be better used in practice in Slovakia. This approach is fully in line with the principles of sustainable development.

4. Conclusions

The current state of the environment requires countries to be more proactive in using green approaches, but this cannot be done without experimental evidence. The main objective of this study was to examine the utilization of fly ash deposited in a heap as a potential raw material for the geopolymerization process. An experimental analysis of the mechanical properties of geopolymer prepared using heap-deposited fly ash and various alkaline activators were carried out and described in this study. The following observations can be drawn based on the experimental findings.

The geopolymeric constructions and infrastructure sectors have undergone a revolution as a result of the recent development of unique and ultramodern geopolymer technology and applicable advanced research. As a result, a new age of environmentally friendly construction materials has begun thanks to the practical substitution of geopolymer cement (GPC) for regular Portland cement (OPC). In addition, compared to current OPC manufacturing, this novel, user- and eco-friendly inorganic family of geopolymeric materials has a minimal carbon footprint and can be made using a simple technique at a low temperature and with less energy.

1. The mechanical strengths of the geopolymers improved as the volume of Na₂O in the mixture increased. After 360 days, the compressive strengths have the highest values, but after 90 days, the strength did not improve much.
2. The geopolymer strengths improved as the SiO₂/Na₂O molar ratio increased to 1.25, but a further increase was accompanied by a decline in strength. After 180 days, several samples showed a decrease in strength.
3. The mechanical strength of the geopolymers was decreased as the volume of water in the mixture was increased.
4. A water absorption experiment revealed that an increase in the amount of water in the geopolymer mixture has a negative effect on the development of the strength of

the geopolymer samples. The mechanical strengths of geopolymer samples have an impact on final water absorption.

5. The geopolymer has the greatest mechanical properties when it was prepared with an alkaline activator solution with a $\text{SiO}_2/\text{Na}_2\text{O}$ molar ratio of 1.25, 8% Na_2O in the mixture, and 30% water in the mixture. This alkaline activator solution's formulation was ideal for the HDFFA geopolymers from our investigation. This mixture was used to produce geopolymer, which was cured for 6 h with 80 °C. It had a flexural strength of 11 MPa and a compressive strength of 57.1 MPa after 360 days, and its water absorption was 6.68%.

This study supports the use of fly ash to produce geopolymers in line with reducing environmental degradation and promoting sustainable development. Ultimately, it can be claimed that geopolymers as a material have potential, as they are completely resilient, sustainable, and affordable.

In Slovakia, nearly all the heating plant waste, primarily fly ash, is not utilized and is discarded on in the form of heaps. It can be concluded that little attention is paid to this concern in Slovakia, which is reflected in the lack of evidence-based practices. According to Slovakian legislation, heaps are sources of dangerous wastes, which is the main environmental problem. This issue has long been neglected and underestimated in practice in Slovakia, and therefore this study can be a valuable platform for proactive and evidence-based solutions. In this paper, the utilization of fly ash from Slovakia, deposited on a heap near the city Kosice which creates an environmental problem, was used as a potential aluminosilicate material for the geopolymerization process. As we proved from the results obtained from alkali activation, the heap near Kosice can be utilized, and the environmental problem can be removed by starting to use new methods of discarding fly ash.

Author Contributions: Conceptualization, M.M.; methodology, M.M and M.S. (Martin Sisol); investigation, M.M. and M.S. (Michaela Sud'ová); data curation, M.S. (Martin Sisol) and K.D.; writing—original draft preparation, M.M.; writing—review and editing, M.M. and V.I.; supervision, M.S. (Martin Sisol). All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by the European Institute of Innovation and Technology (EIT), a body of the European Union, under the Horizon 2020, the EU Framework Programme for Research and Innovation (Project BioLeach: Innovative Bio-treatment of RM, grant number: 18259). This research was funded by the European Institute of Innovation and Technology (EIT), a body of the European Union, under the Horizon Europe, the EU Framework Programme for Research and Innovation (Project DYNOSORT Dynamic ore sorting of polymetallic stockpiles, grant number: 21050).

Conflicts of Interest: The authors declare no conflict of interest.

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