East African Journal of Engineering, Volume 6, Issue 1, 2023
Article DOI: https://doi.org/10.37284/eaje.6.1.1361

East African Journal of Engineering
eaje.eanso.org
Volume 6, Issue 1, 2023
Print ISSN: 2707-5389 | Online ISSN: 2707-5397
Title DOI: https://doi.org/10.37284/2707-5397

EAST AFRICAN NATURE & SCIENCE ORGANIZATION

Original Article

Effect of Sodium Silicate to Hydroxide Ratio and Sodium Hydroxide Concentration on the Physico-Mechanical Properties of Geopolymer Binders

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Article DOI : https://doi.org/10.37284/eaje.6.1.1361

Publication Date:

08 August 2023

Keywords:

Geopolymers, Sodium Silicate, Sodium Hydroxide, Metakaolin, Mechanical Tests

ABSTRACT

Geopolymers are emerging materials in the construction industry that are yet to achieve uniform standards in their mode of synthesis. Several factors over the years are said to affect the performance of the binders which vary widely. This paper investigates the effect of silicate-to-hydroxide ratio and sodium hydroxide concentration on the mechanical properties of metakaolin-based geopolymer binders. Four SS/SH ratios of 0.5, 1.0, 1.5, and 2.0 and four SH concentrations of 8M, 10M, 12M, and 14M were studied with the kaolin calcined at 800°C. Physical and mechanical tests were conducted on sixteen (16) samples at testing ages of 7, 14, and 28 days. The mean density of the geopolymer binders was 1.86 g/cm3, and the samples increased with increasing SS/SH ratio and SH molarity with the 2MK14 having the maximum density of 1.95 g/cm^3. The compressive strength and flexural strength of the samples generally increased with testing age (7-28 days). The compressive strength of the samples further showed a direct increase with SS/SH ratio, though, above SS/SH = 1, the compressive strength began to decrease gradually. The maximum compressive strength for these binders 14.6MPa which was recorded by the 1MK10 sample. The Pareto chart for the compressive strength of the samples showed that both the SS/SH ratio and SH concentration contributes significantly (95% significant level) to the compressive test results. However, the SS/SH ratio had a more standardised effect on the compressive strength of the binders. The maximum flexural strength (3.45MPa) was obtained from the 1MK8 geopolymer binder, and similar to the compressive test results, strength values were obtained at the SS/SH ratio of 1.

APA CITATION

Enoh, M. K. E. & Ushie, D. O. (2023). Effect of Sodium Silicate to Hydroxide Ratio and Sodium Hydroxide Concentration on the Physico-Mechanical Properties of Geopolymer Binders *East African Journal of Engineering*, *6*(1), 113-121. https://doi.org/10.37284/eaje.6.1.1361

CHICAGO CITATION

Enoh, Maria Kaka Etete and David Ogar Ushie. 2023. "Effect of Sodium Silicate to Hydroxide Ratio and Sodium Hydroxide Concentration on the Physico-Mechanical Properties of Geopolymer Binders". *East African Journal of Engineering* 6 (1), 113-121. https://doi.org/10.37284/eaje.6.1.1361.

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HARVARD CITATION

Enoh, M. K. E. & Ushie, D. O. (2023) "Effect of Sodium Silicate to Hydroxide Ratio and Sodium Hydroxide Concentration on the Physico-Mechanical Properties of Geopolymer Binders", *East African Journal of Engineering*, 6(1), pp. 113-121. doi: 10.37284/eaje.6.1.1361.

IEEE CITATION

M. K. E. Enoh. & D. O. Ushie "Effect of Sodium Silicate to Hydroxide Ratio and Sodium Hydroxide Concentration on the Physico-Mechanical Properties of Geopolymer Binders" *EAJE*, vol. 6, no. 1, pp 113-121, Aug. 2023.

MLA CITATION

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INTRODUCTION

Geopolymers are inorganic polymers formed by dissolving an aluminosilicate (AS) material in a highly alkaline liquid at a relatively low temperature (Askarian et al., 2019; Provis, 2014; Singh & Middendorf, 2020). Raw rock-based materials such as clay, feldspar, natural pozzolans, and industrial/agricultural residues such as fly ash, ground blast furnace slag, silica fume, rice husk ash, and crushed glass are suitable precursors for the making of GPs due to an appropriate proportion of alumina and silica in their composition (Duxson et al., 2007). Apart from these precursors, a strongly alkaline solution made from hydroxides, silicates, carbonates, and sulphates of alkali metals is the necessary activator required to complete the geopolymerisation process (Ishwarya et al., 2019; Ma et al., 2022).

The research interest in GPs has increased over the past years with several potential applications for use as resins, binders, cement, composites, and waste encapsulation. Their ability to use various by-products during synthesis while reducing or diversifying the depleting rate of natural resources and minimising the stash of by-products, and reduction in the emission of carbon dioxide are what make them sustainable for these applications (Nodehi & Taghvaee, 2022; Singh & Middendorf, 2020; Zhao et al., 2021). High strength, rapid setting, high resistance to acids and alkalis, high thermal resistance and high endurance to thermal shock are some of the other desirable properties of GPs, though these properties are not inherent in all compositions because they rely on the chemical makeup, mineralogical composition, morphology and glassy phase content (Davidovits, 2002; Lahoti et al., 2019).

The discovery of GPs has therefore created new materials for coating and adhesives, binders for fibre composite, encapsulation of wastes and cement for concrete. Their areas of application are on a constant increase, and this includes; fire-resistant materials, decorative stone artefacts, waste encapsulation, 3D printing, low-energy ceramic tiles, refractory items, biotechnology, foundry industry, cement and concretes, composite for infrastructure repair and strengthening, arts and decoration, composites for aircraft interior design, thermal insulation and archaeology (Davidovits, 2002).

As with every new technology, GPs have encountered several challenges in their commercialisation and acceptability (Danish et al., 2022). The inability to tailor similar properties over a wide variety of sources is one of them. Several factors are known to have contributing effects on the final properties of the binder. These properties may include the ratio and nature of the activator, the source material and their composition, the Si/Al ratio in the mix proportion of the geopolymer binder, the solid to liquid ratio, SS/SH ratio, sodium hydroxide molarity, curing time, curing medium, time of mixing etc. (Jan et al., 2022; Komljenović et al., 2010; Lin et al., 2021). There are several research related to the effect of SS/SH ratio and SH molarity on the mechanical properties of geopolymer binders. Improving the molarity of SH reportedly increases the solubility of alumina and silicate in the mix, thus leading to higher strength (Hardjito et al., 2008). Increasing the SS/SH ratio is also reported to improve the strength of the binder, but

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the limit of this increase varies among researchers (Jan et al., 2022; Malkawi et al., 2016).

Therefore, to carefully understand each distinct source, the characterisation and synthesis of the identified source are essential. A few research have examined the effect of several factors in the synthesis of geopolymers, but this source of MK has not been investigated yet; thus, the reason for this research is to set the foundation for further research. The ratios and concentrations investigated fall within a range that has reportedly produced exceptional properties.

MATERIALS AND METHODS

Materials

Kaolin was used as the aluminosilicates source or precursor and was collected from a deposit around Markarfi in Kaduna State, Northern area of Nigeria. Sodium silicate (SS) mixed with sodium hydroxide (SH) was used as the alkaline activators in this study due to their performance, availability, and lower cost. The SH with 97% purity was purchased in pellet form and dissolved in distilled water to produce the required concentrations. Sodium silicate with the composition of SiO_2; 30%, Na_2O; 12 % and H_2O; 58% were purchased in liquid form.

Preparation of Samples

The kaolinite was ground and sieved through a 200 µm sieve to get finer and more uniform particle sizes. The clay sample was then calcined in a muffle furnace at three different temperatures, 800 ^oC, to produce metakaolin (MK) with a mean particle size of 23.5 µm obtained from sieve analysis. In preparing the SH concentrated solutions of 8 M, 10 M, 12 M and 14 M, SH pellets were dissolved in distilled water. For instance, to prepare a 6 M concentrated sample of SH solution, 240 g (40g x 6) of SH pellets were dissolved in 1000 ml of water, and the reaction was exothermic.

Characterisation of Materials and Binders

X-ray diffraction (XRD) and X-ray fluorescence (XRF) were used to determine the mineralogical and chemical composition of the kaolin. For the XRD graph (Figure 1) of the calcined samples, it is observed that the sharp kaolinite peaks diminish with an increase in calcination temperature and become less distinct. This disappearance of the peaks is an indication of the presence of an amorphous phase (metakaolinite) due to the dehydroxylation of the raw clay sample (Elimbi et al., 2011). Studies of phase transformations of kaolin to MK from different regions have also produced similar results (Eliche-Quesada et al., 2020; Mark et al., 2019). The peaks of the kaolin diminish with an increase in calcination temperature, gradually transcending from a crystalline to an amorphous material.

From the XRF results in *Table 1*, the combined composition of SiO_2 (51.2%) and Al_2O_3 (32.4%) was about 85% which makes it a suitable aluminosilicate precursor.

Figure 1: XRD result for kaolin and metakaolin calcined at 800oC

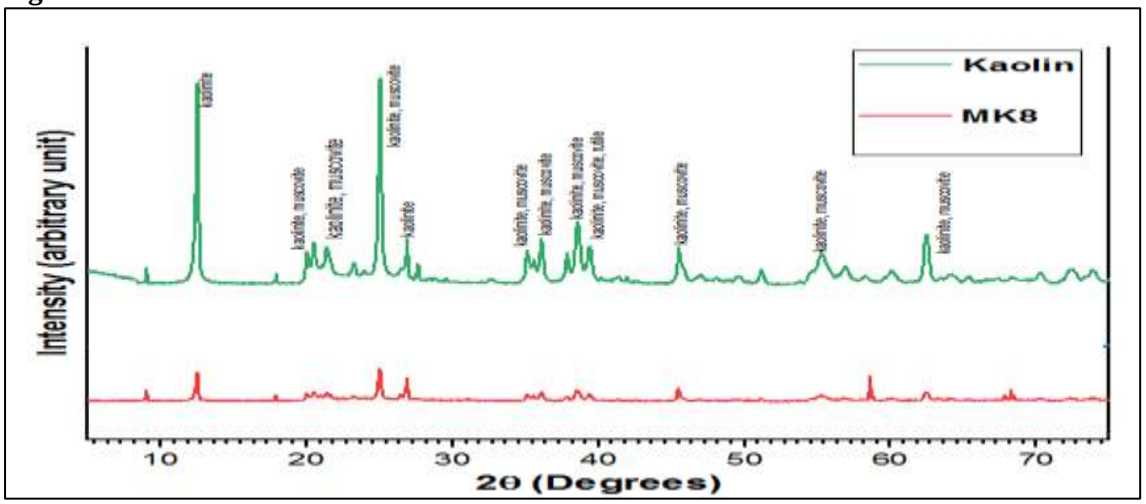

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Table 1: XRF results for kaolin

Raw Material	Chemical Composition										
	SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	K_2O	MgO	Na_2O	CuO	Others	LOI
Kaolin	51.18	32.35	0.34	0.88	0.02	1.87	0.01	0.01	0.25	1.03	12.06

Synthesis and Testing of Materials

The geopolymer binders were produced using the two-part mixing process. The SH and SS solution were mixed in ratios of 0.5, 1, 1.5 and 2 for 1 minute before pouring immediately into a measured quantity of solid metakaolin sample with a solid-to-liquid (S/L) ratio of 1.2. The solid and liquid constituents were mixed thoroughly for 10 minutes with the aid of a hand mixer and poured into a silicone mould of size 50 mm x 50 mm x 250 mm. It was observed that the viscosity of the slurry increased directly with SS/SH ratio and SH molarity content, thus making the paste difficult to mix at higher concentrations and ratios. The samples in the mould were vibrated for 3 mins to remove trapped air in the paste and allowed to cure at room temperature. Samples were investigated at testing ages of 7, 14 and 28 days. The mix proportion is presented in *Table 2*.

The first number in the mixed code signifies the SS/SH ratio, the letters MK represents the precursor (metakaolin), and the succeeding number signifies the SH molarity. For example, 0.5MK8 represents a geopolymer binder prepared with metakaolin with an SS/SH ratio of 0.5 and SH molarity of 8.

The samples were demoulded and left to cure for the stipulated period of time. The prismatic samples were used for the flexural tests. For the compressive testing, however, the prismatic samples were reduced to cubes of size 50 mm, and the compressive and flexural strength were tested in accordance with ASTM C109 and ASTM C293, respectively. At the various testing age, three (3) cubes and prismatic samples were tested for each strength test, and the average values were recorded at each testing age. *Plate 1* shows some of the samples being prepared for testing.

Table 2: Mix the proportion of geopolymer binders

Mix codes	M	MK (g)	SH soln (g)	SS soln (g)	SS/SH ratio	S/L ratio	Curing time (days)
0.5MK8	8	140	77.8	38.9	0.5	1.2	28
0.5MK10	10	140	77.8	38.9	0.5	1.2	28
0.5MK12	12	140	77.8	38.9	0.5	1.2	28
0.5MK14	14	140	77.8	38.9	0.5	1.2	28
1MK8	8	140	58.4	58.4	1.0	1.2	28
1MK10	10	140	58.4	58.4	1.0	1.2	28
1MK12	12	140	58.4	58.4	1.0	1.2	28
1MK14	14	140	58.4	58.4	1.0	1.2	28
1.5MK8	8	140	46.7	70.0	1.5	1.2	28
1.5MK10	10	140	46.7	70.0	1.5	1.2	28
1.5MK12	12	140	46.7	70.0	1.5	1.2	28
1.5MK14	14	140	467	70.0	1.5	1.2	28
2MK8	8	140	38.9	77.8	2.0	1.2	28
2MK10	10	140	38.9	77.8	2.0	1.2	28
2MK12	12	140	38.9	77.8	2.0	1.2	28
2MK14	14	140	38.9	77.8	2.0	1.2	28

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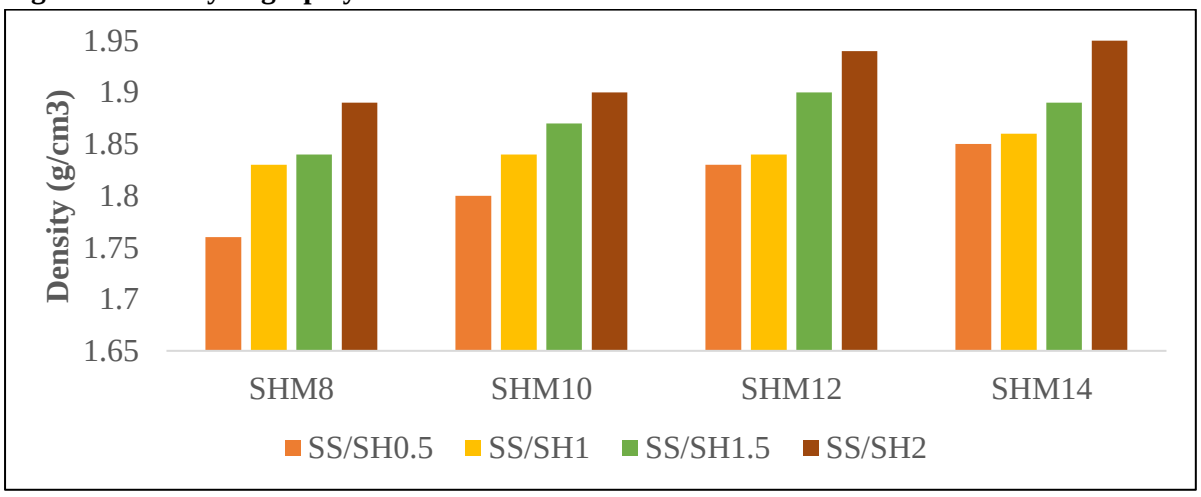
Plate 1: Samples of prepared geopolymer binder

RESULTS AND DISCUSSION

Physical Properties of Samples

The density of the prismatic samples was measured, and the results are represented in *2*. Samples with a high SS/SH ratio and high sodium hydroxide molarity (SHM) were denser than others. An increase in SS/SH ratio and sodium hydroxide molarity resulted in an increased density of the sample with a maximum density of 1.94 g/cm^3 for the 2MK14 binder. The high density of the silica and higher density of the hydroxides with increased concentrations can explain the reason for this trend in the density of the samples.

Figure 2: Density of geopolymer binders



Compressive Strength Test Results

The compressive strength development of the geopolymer binders with varying SS/SH ratios and SH molarity are shown in Figure 3. As expected, the strength increased with curing time, but the rate of increase varied because some binders developed early strength than others. Samples with higher SS/SH ratio (1-2) developed most of their strength within the first 14 days, while those with lesser content of silica had a more gradual strength development. This agrees with previous research that suggests that the presence of silica facilitates the setting process (Jan et al., 2022). Despite the early strength development of those with a SS/SH ratio of 2, the optimum compressive strength of 14.6 MPa was obtained when the SS/SH ratio was one, and the

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SH molarity was 10. This decrease in strength at a higher SS/SH ratio may be associated with the saturation of the system with silica which hinders further strength development (Morsy et al., 2014). Increasing the SS/SH from 0.5 to 1.0 improved the compressive strength, but a decline in the strength was, however, noticed from 1.0 to 2.0, despite the increase in SH molarity. The SH molarity had a lower contributing effect than the SS/SH ratio on the strength development of the geopolymer binders. This effect can be seen in the Pareto chart (Figure 4) for the standardised compressive strength effect, which presents the contributing effect of each of the factors (SS/SH ratio and SH molarity) on the response (compressive strength). Both values fall beyond the 95% confidence level, which is represented by the red dotted line, and this signifies that both values contributed significantly to the compressive strength development of the binders.

Figure 3: Compressive strength of geopolymer binders

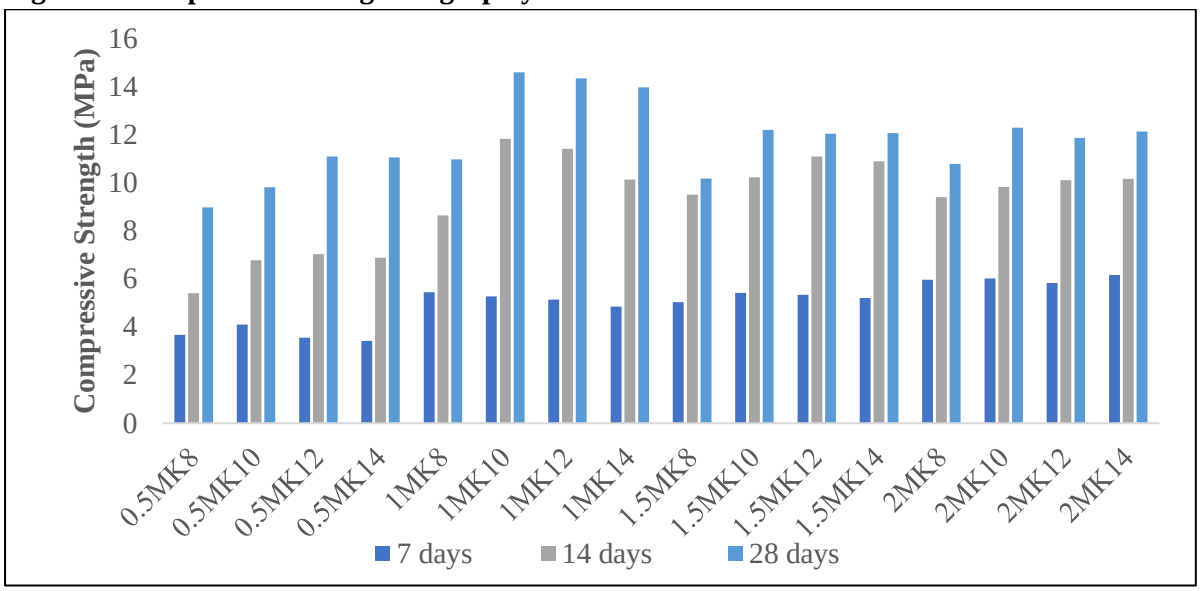
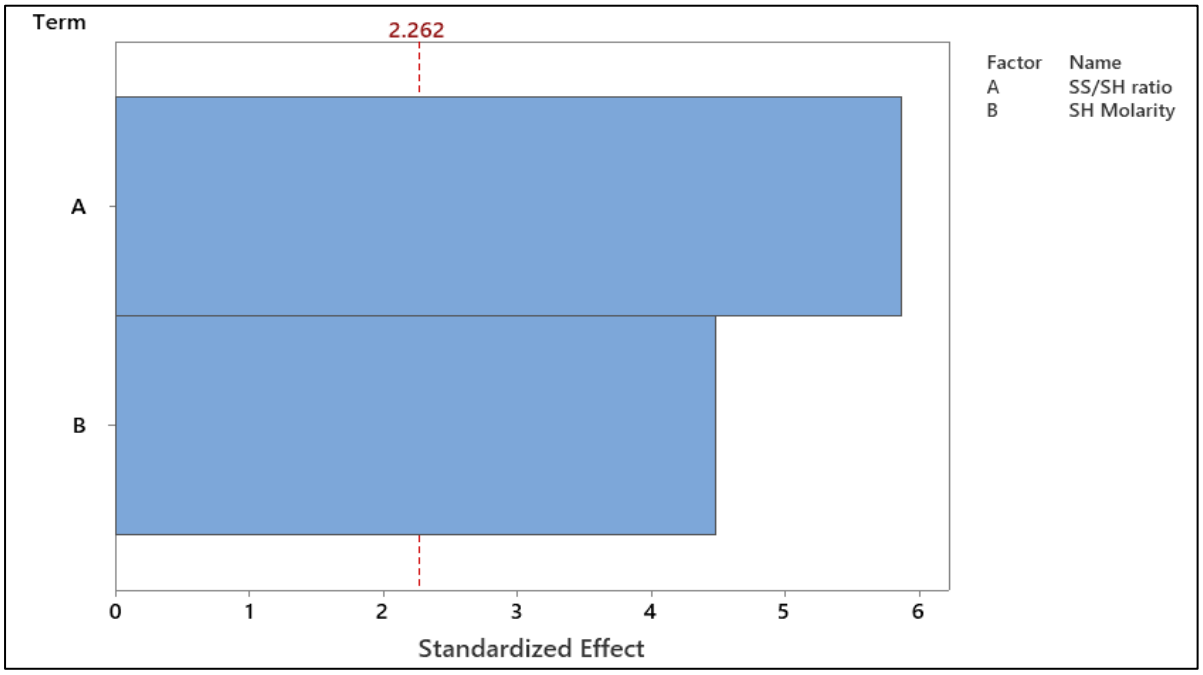


Figure 4: Pareto chart for compressive strength



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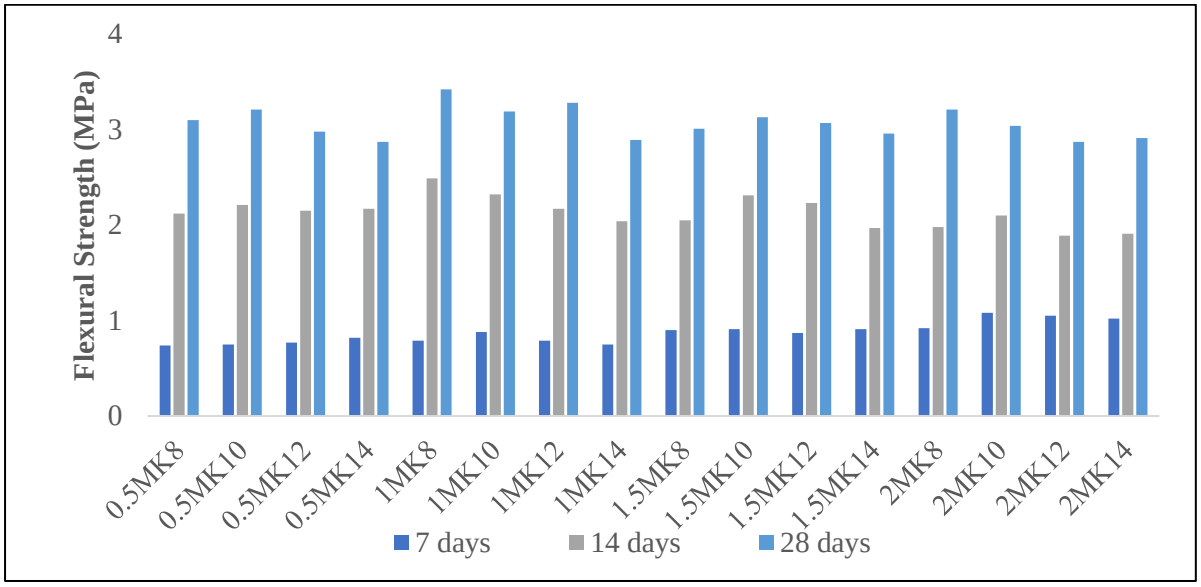
Flexural Strength Test Results

The results of the flexural strength test on the prepared geopolymer binders are reported in the bar graphs in *Figure 5*. Like the results obtained from the compressive strength test, the flexural strength of the samples increased with testing age. Early strength development occurred between the 7- and 14-days testing. The maximum flexural strength of 3.4 MPa was attained at the testing age of 28 days from the 1MK8 sample. Samples with an SS/SH ratio of 1 resulted in high values of flexural strength for all molarities studied apart from the 12M concentration. This is similar to what was reported by Morsy (2014), though a higher value of flexural strength of 6 MPa was achieved at a SS/SH ratio of 1. The reduction in the strength with an increase in the SS/SH ratio can be associated with the presence of excess silicate, which is said to obstruct water evaporation and structure formation in the binders (Jan et al., 2022).

Figure 5: Flexural strength of geopolymer binders



CONCLUSION

This study investigated the effect of the SS/SH ratio and SH molarity on the physical and mechanical properties of metakaolin clay-based geopolymer binder. The results showed that altering the alkaline solution ratios and concentration could affect properties like workability, density, flexural and compressive strength of the binders. From the results obtained, the following conclusions can be drawn:

- The density of the geopolymer binders increased with SS/SH ratio and SH molarity.
- The compressive and flexural strength increased with the testing age from 7 to 28 days.
- The SS/SH ratio had more effect on the compressive strength of the geopolymers than the SH molarity.
- The maximum compressive strength was obtained at 1MK10, and increasing beyond the SS/SH ratio of 1 led to decreased strength.
- The maximum flexural strength was obtained at 1MK8, but samples with 8M and 10 M showed better properties across all SS/SH ratios.
- Using SS/SH ratio above one will not be economical or sustainable given that using lower content of sodium silicate still produces desirable properties.

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