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ORIGINAL RESEARCH
PAPER



Effect of liquid-solid ratio on metakaolin-based geopolymer binder properties

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ABSTRACT

This article aims to achieve the hardening of geopolymer binder-based metakaolin under the same setting time and hardening temperature as traditional cement. It studied the hardening of the geopolymer binder-based metakaolin at two temperatures of 60 °C and room temperature using different liquid-to-solid ratios of 0.8, 0.95, and 1.1. The bulk density, compressive strength, setting times, thermal conductivity, and microstructure were measured for geopolymer binders. Based on the specified range for cement characteristics, the geopolymer binder was solidified at room temperature, utilizing a liquid-to-solid ratio of 0.8 to achieve optimal results stratified according to the cement's ideal characteristics. It had a bulk density of 1.264 g cm⁻³, compressive strength of 19.12 MPa, initial setting time of 288 min, final setting time of 358 min, and thermal conductivity of 0.49 W m⁻¹ K.

KEYWORDS

geopolymer binder, metakaolin, liquid-solid ratio, setting time, thermal conductivity, compressive strength

1. INTRODUCTION

Geopolymer is well known as an alternative to conventional Portland cement, representing a development shift in construction. There are many advantages to opting for geopolymer over cement, foremost among them being the reduction in CO₂ emissions, alongside reduced energy used, and it had lower thermal conductivity. The geopolymer binder needs curing by heat to harden, which hinders use as a cement replacement [1, 2].

Geopolymer binder has a quality performance comparable to conventional cement due to its strength, fire resistance due to low thermal conductivity, and acid and alkali resistance, in addition, it produces less than 20% of the carbon emissions of conventional Portland cement [3]. Geopolymer is formed by activating materials rich in aluminosilicate like fly ash, kaolin, metakaolin, slag, mine, etc., obtained from natural minerals or solid waste materials [1, 4]. The highly alkaline conditions are used to induce the formation of polymeric aluminosilicate compounds from geological materials by using alkali metal silicate solutions [4–6].

Geopolymers are utilized in various applications due to their versatile properties. They are prominent in the production of fire-resistant materials and innovative ceramics. They are effective alternatives to asbestos, offering safe and durable building materials. Geopolymers also play a critical role in stabilizing hazardous waste, providing a secure method for waste containment. Additionally, they are used as adsorbent materials in water treatment processes, enhancing the removal of contaminants and improving water quality [5].

Metakaolin will be expected as a major resource for the geopolymer binder due to its main contents of SiO₂ and Al₂O₃, and excellent chemical stability compared with other waste materials. The obtained by heating kaolin at temperatures higher than 650 °C. Consequently, the mass of the kaolin was reduced by approximately 14 wt% due to the removal of strongly

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attached hydroxyl ions and the disintegration of the kaolin structure, resulting in the contraction and dissolution of the organized arrangement inside the alumina and silica layers [6]. Metakaolin, outstanding in its amorphous structure and unique characteristics, emerges as a compelling alternative to cement in geopolymer production, especially when compared with other aluminum silicate sources. Its inherent pozzolanic and latent hydraulic reactivity, a significant surface area, minimal impurities, and a substantial amorphous composition resulted in solidification without requiring heat curing [7].

Multiple factors affect the geopolymer characteristics: raw materials type, alkali activator molarity, $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio, liquid-to-solid ratio, curing conditions (temperature, and time), etc. All these factors significantly contribute to influencing the properties of the geopolymer binder. It affects density, strength, porosity percent, sitting time, thermal conductivity, and microstructure, depending on these properties geopolymer binder could be used in many fields for example decorative stone, thermal insulation, refractory materials, foundry applications, concrete, aircraft ways, etc. [8, 9].

N A Jaya et al. [10] investigated the impact of various Liquid-to-Solid (L/S) ratios specifically 1.66, 1.42, 1.25, 1.11, and 1 on the properties of metakaolin-based geopolymers. The study used a $\text{NaOH}/\text{Na}_2\text{SiO}_3$ ratio of 1 and used a 10 M NaOH solution. After aging the samples for 28 days, it was found that the optimal compressive strength of 32 MPa was achieved with a L/S ratio of 0.8. Adelino Lopes et al. [11] recommended, preventing a reduction in the strength of metakaolin-based geopolymers, it is recommended to use curing temperatures below 30°C . Samples cured at 50°C show a decrease of over 35% in flexural strength and more than 60% in compressive strength compared to those cured at room temperature.

In this work, two factors effects were studied the liquid-to-solid ratio and curing temperature. The objective was to prepare a metakaolin-based geopolymer binder solidifying at room temperature within an acceptable setting time.

2. MATERIALS AND METHODS

2.1. Raw material

The solid raw material for preparing the geopolymer binder was kaolin and the materials to prepare the activator solution were water glass, sodium hydroxide flakes, and distilled water. Table 1 shows the water glass properties and Table 2 sodium hydroxide properties, identified by the manufacturer company.

2.2. Characterization techniques of the kaolin and metakaolin

The transformation phases of kaolin into metakaolin were analyzed through X-ray diffraction using the Rigaku Mini-flex II diffractometer model. The oxide composition of metakaolin was determined using X-ray fluorescence with the Rigaku SuperMini 200 spectrometry.

Table 1. Water glass properties

Name	Silicate sodium
Formula	Na_2SiO_3
Case	Liquid
Density	1.37 g cm^{-3}
Silicate modulus	3.80
Chemical composition	H_2O : 61.06 wt% SiO_2 : 30.95 wt% Na_2O : 8.00 wt%

Table 2. Properties sodium hydroxide flakes

Name	Sodium hydroxide
Formula	NaOH
Case	Solid (flakes)
Purity	~98%

Placeholder granulometry 715 measured partial size in a range of 1–192 μm .

2.3. Geopolymer binder preparation

Metakaolin was prepared by calcinating the kaolin at 750°C for 3 h. The metakaolin was milled to fine powder by using a ball mill model Retsch PM 400, milling conditions speed of 180 rpm and milling time of 20 min. Five silica balls were used to mill with different sizes (the diameter of three balls was 150 cm and for the other two balls was 30 cm). The sodium hydroxide solution was made to a concentration of 10 mol L^{-1} by dissolving 800 g of sodium hydroxide flakes in 2 L of distilled water. It was mixing an equal weight percentage of water glass and sodium hydroxide solution to prepare the activator solution [12]. The geopolymer binder samples were prepared by using a silicon mold cubic with a length of 5 cm, the samples were cured at two different temperatures: 60°C and room temperature, and formulated with different liquid-to-solid ratios of 0.8, 0.95, and 1.1. The geopolymer binder mix proportion design is shown in Table 3 for a cubic length of 5 cm.

2.4. Characterization techniques of the geopolymer binder

The geopolymer binder's bulk density and compressive strength were assessed using cubic samples measuring 5 cm long after a 28-day curing process at 60°C for 24 h and at ambient room temperature. Bulk density was determined for

Table 3. Composition of geopolymer binder with a $\text{NaOH}/\text{Na}_2\text{SiO}_3$ ratio of 1 and NaOH with (10 mol L^{-1})

Samples Cods	L/S	Liquid (g)		Solid (g) Metakaolin
		Na_2SiO_3	NaOH	
GP 0.8	1.1	50	50	125.0
GP 0.9	0.9	50	50	111.1
GP 1.1	0.8	50	50	90.9



an average of five samples according to ASTM C642-06:2006 [13] by dividing the mass of the sample by its volume. The compressive strength of the samples was evaluated using an automated Smart Power and Control System specifically designed for standard failure testing, the Zwick/Roell Z100. Three cubic samples were subjected to testing at a speed of 2 mm min^{-1} , following the ASTM C109/C109M-02:2002 [14] standard. Compressive strength was calculated by dividing the maximum force applied by the cross-sectional area.

The C-Therm TCi thermal conductivity analyzer instrument has been used to measure the thermal conductivity of the geopolymer binder. The sample cubic shape with a side length of 2.5 cm conductive materials was applied to the sample's surface before it was placed on the sensor. The test was performed at room temperature.

The sitting times of the geopolymer binder were measured with the used Vicat apparatus according to standard EN 196-3:2005 [15]. The Vicat mold was filled with a geopolymer binder, and the filled mold with a base plate was put in the container, adjusting the distance zero between the end of the needle and the base, the test was made at room temperature 23°C . A thin metal needle with a diameter of 1 mm was used to measure the initial time. It checked the needle penetration every 15 min. The onset of the initial setting time was determined by observing the penetration of the thin metal needle into the mixture until it reached a depth of 5 mm. To measure the final setting time, invert the mold for the other face and use the hollow circular needle with a diameter of 5 mm. The determination of the final setting time was by observing the penetration of the hollow needle of 0.5 mm. Figure 1 shows the Vicat apparatus for measuring geopolymer binder sitting time.

Scanning Electron Microscope (SEM) was employed to examine the microstructure and analyze the predominant elemental composition of the geopolymer binder. The Thermo Helios G4-PFIB CXe Dual scanning electron microscope, equipped with a Bruker microprobe and operated at a voltage of 20 kV, was utilized for this purpose. Before SEM imaging, the fractured samples underwent a coating process with a layer of gold to enhance conductivity.

3. RESULTS AND DISCUSSION

3.1. Metakaolin characteristics

The effect of the calcination process on the phase transformation of kaolin as a crystal phase to metakaolin as an amorphous phase is shown in Fig. 2. Following calcination at 750°C for 3 h, the kaolin initially contained 16 wt% of amorphous phases, which then increased to 74 wt%, resulting in the formation of metakaolin as it is shown in Table 4 detected by X-ray diffraction results. The findings of the ray fluorescence study, which was used to determine the composition of metakaolin, are presented in Table 5. Depicts the particle size distribution of metakaolin particles in Fig. 3, with an average size of $5.4 \mu\text{m}$.



Fig. 1. Vicat apparatus for the measurement of geopolymer binder test (Source: Authors' photo)

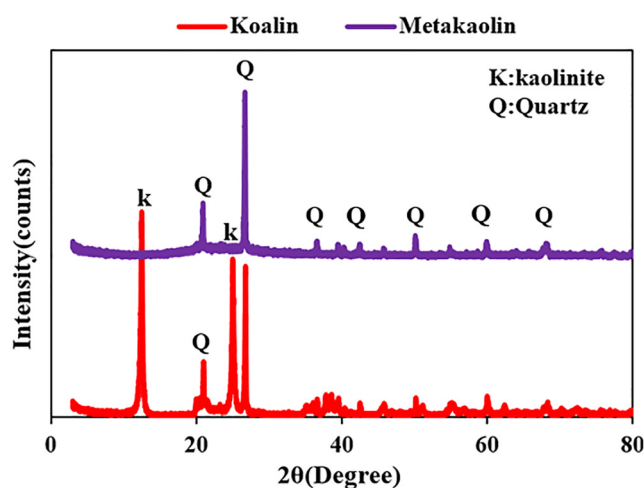


Fig. 2. The X-ray pattern for the kaolin and transformation into metakaolin through the calcination process

3.2. Geopolymer binder characteristics

The bulk density of the geopolymer binders in samples that were cured at two different temperatures and had different

Table 4. X-ray results show the quantity of kaolin and metakaolin's crystalline and amorphous phases

Phases	Kaolin (wt%)	Metakaolin (wt%)
Quartz	22.68	26.00
Kaolinite	61.32	0.00
Amorphous	16.00	74.00
SUM	100.00	100.00

Table 5. Metakaolin composition

Oxide composition	(wt%)	Oxide composition	(wt%)
SiO ₂	58.30	CaO	0.180
Al ₂ O ₃	39.40	Fe ₂ O ₃	0.470
Na ₂ O	0.06	P ₂ O ₃	0.013
MgO	0.33	Other	0.987
K ₂ O	0.26	SUM	100

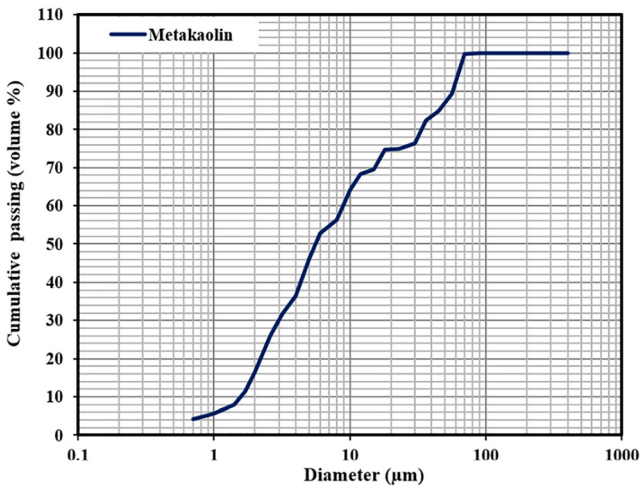


Fig. 3. Metakaolin particle size distribution

liquid-to-solid ratios were measured. The bulk density range of the binders cured at 60 °C for 24 h was 1.172–1.195 g cm⁻³, and that of the binders cured at room temperature was 1.213–1.264 g cm⁻³, as it is shown in Fig. 4. As the curing temperature increases, the binder density decreases due to increased moisture loss early during polymerization. It is necessary to prevent water loss before it is completely cured [16]. Note that the bulk density of the binders reduces with the liquid-to-solid ratio. The higher density was 1.264 g cm⁻³ for the sample with a liquid-to-solid 0.8 ratio and was cured at room temperature. When the ratio of liquid to solid in geopolymers is higher, holes appear, particles are not packed tightly enough, the volume shrinks more, and the porosity increases. All these factors lead to a notable reduction in the binder's density [17].

The compressive strength of the geopolymer binders cured for 28 days is presented in Fig. 5. The selected curing temperature of the geopolymer depended on raw materials for the fly ash-based geopolymer needs a curing temperature of more than 60 °C to get higher compressive strength [18].

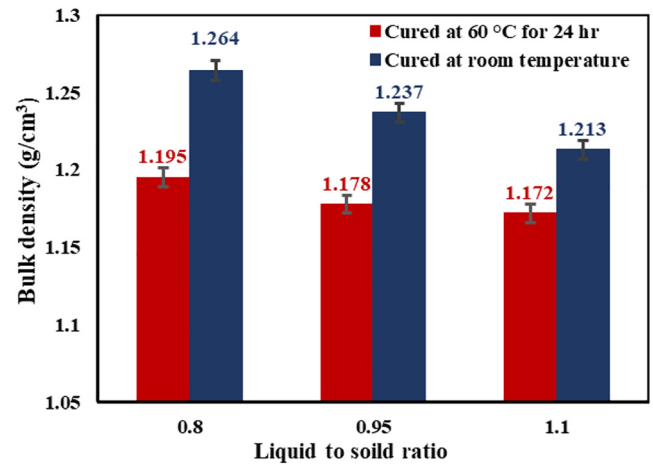


Fig. 4. Bulk density of geopolymer binders with different liquid-to-solid rates cured at two different temperatures

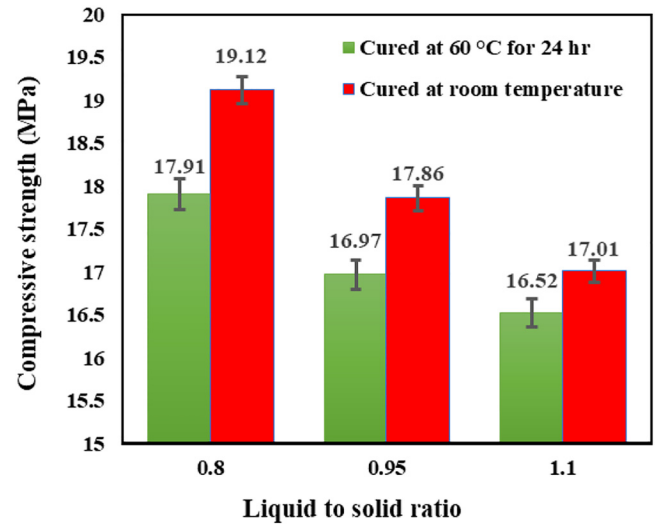


Fig. 5. Compressive strength of geopolymer binders with different liquid-to-solid rates cured at two different temperatures

In this work, the metakaolin-based geopolymer gave maximum compressive strength for samples cured at room temperature by adjusting the raw materials ratio and kaolin calcination temperature depending on previous works [12]. The compressive strength range of the binders cured at 60 °C for 24 h (one day) and 27 days at room temperature was 16.52–17.91 MPa, and that of the binders cured at room temperature for 28 days was 17.01–19.12 MPa. The compressive strength of the binders was influenced by the ratios of liquid to solid components. The highest strength recorded was 19.12 MPa, observed in samples with a liquid-to-solid ratio of 0.8, cured at room temperature. Mainly, increasing the solid content in geopolymer binders leads to improving the mixture by good packing, enhanced chemical reaction, and reduced porosity, all of which factor in improving compressive strength. Depending on previous work, it was found to reduce compressive strength and poor workability when the liquid-to-solid ratio was less than

0.4 because the activation solution amount would be insufficient for the completed polymerization process [19].

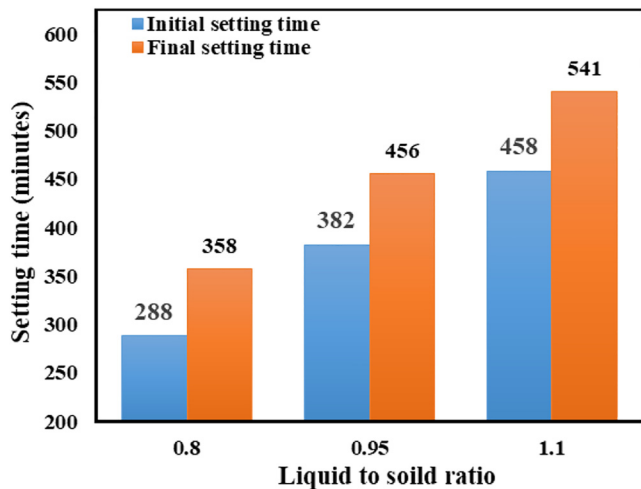


Fig. 6. Setting time of geopolymer binders with different liquid-to-solid rates cured at room temperature

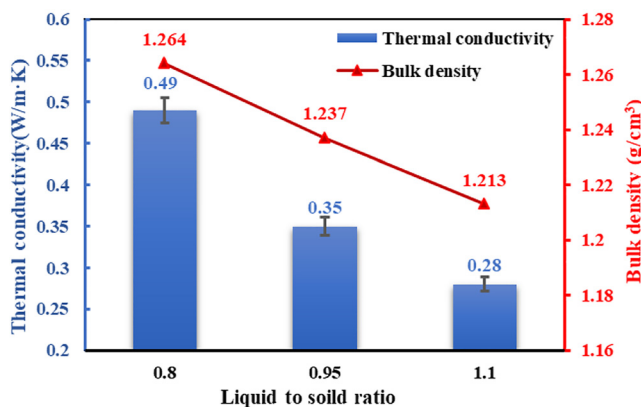


Fig. 7. Thermal conductivity of geopolymer binders with different liquid-to-solid rates cured at room temperature

The setting times both initial and final were measured for geopolymer binder cured at room temperature with different liquid-to-solid ratios as it is shown in Fig. 6. The initial setting denotes the duration required for the binder to transition from a liquid to a solid state following the mixing of the powder with an activator solution. While the final setting time indicates the period needed for the binder to attain hardening. It was found setting time for geopolymer binder decreases as the liquid-solid ratio decreases. The lower setting time was recorded at 288 min for the initial setting time and 358 min for the final setting for the geopolymer binder with a 0.8 liquid-to-solid ratio due to the Increased mixture fluidity leading to delayed formation of geopolymer gel [20]. The standard setting times of cement: the initial setting time should be more than 45 min and the final setting should be less than 390 min.

Regarding this matter, the sample with a liquid-to-solid ratio of 0.8 underwent solidification within the specified standard setting time limit. This is one of the metakaolin-based geopolymers advantages, it solidification at room temperature more rapidly than fly ash-based geopolymers [21]. The ultimate final setting time of fly ash-derived geopolymer solidifying at room temperature is approximately 1800 min [22].

One of the geopolymer binders advantages is that it has lower thermal conductivity than cement due to the absence of calcium hydroxide, which is mainly a contributor to the high thermal conductivity value of cementitious materials [23]. Figure 7 presented the thermal conductivity values of geopolymer binders, which were in the range of 0.28–0.49 W m⁻¹ K for samples cured at room temperature with different liquid-to-solid ratios [12]. As the ratio of liquid to solid in a geopolymer binder increases, the thermal conductivity of the material reduces because of the decreased bulk density of the mixture. Denser materials generally have a greater thermal conductivity rating because their atoms and molecules are more closely packed, allowing for active heat transfer.

Figure 8 illustrates the microstructural composition of the geopolymer binder made using two liquid-to-solid ratios:

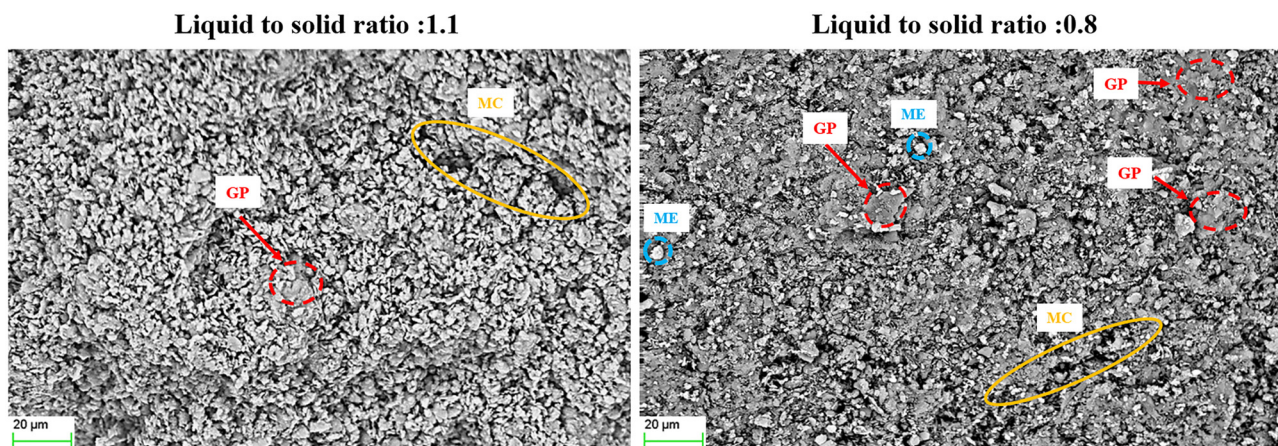


Fig. 8. Electron microscopic images of geopolymer binders with different liquid to solid rates at magnifications 500X harder at room temperature (GP: Geopolymer gel, MC: Micro crack, MK: metakaolin)

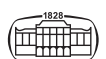
1.1 and 0.8 cured at room temperature the microstructure of the two samples displayed metakaolin unreacted and some microcracks in the gel matrix. In a sample with a lower liquid-to-solid ratio of 0.8, the resultant gel exhibited a comparatively denser and more uniformly structured composition, characterized by reduced pore size. Conversely, in samples with a ratio of 1.1, geopolymer specimens displayed heightened occurrences of cracks and larger pores. Higher liquid-to-solid ratios in geopolymer binder formulations typically result in increased porosity. This phenomenon occurs because a greater amount of water is present, which facilitates the formation of voids within the binder matrix [17]. As a result, the overall porosity of the geopolymer tends to rise, potentially impacting properties such as strength, durability, and permeability.

4. CONCLUSIONS

This article studied the effects of curing temperature and liquid-to-solid ratio on metakaolin-based geopolymer binders. The curing temperature affects the compressive strength and bulk density of the geopolymer binder; the samples cured at room temperature have higher values. The solid and liquid ratio effect increases the bulk density, compressive strength, and thermal conductivity while reducing the liquid-to-solid ratio. As the liquid-to-solid ratio rises, the hardening times increase because of the augmented fluid content, necessitating a longer duration for complete hardening. The sample with the lower value of liquid-to-solid ratio 0.8 cured at room temperature achieved a maximum compressive strength of 19.12 MPa with lower setting times, an initial setting time of 288 min, and a final setting time of 358 min respectively.

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