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Incorporating Rice Husk Ash in Metakaolin-Based Geopolymer Binder

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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ABSTRACT

Geopolymers are interesting materials synthesized by activating aluminosilicate source(s) with alkaline activator(s). The activators often used for this purpose are alkali metals of silicates and hydroxides. Environmental impact assessment carried out on geopolymers however indicates adverse contribution of this process to several other environmental indices despite the reduction in CO_2 emission. Several biomass sources are known to possess a high silica content. This research investigates the effect of incorporating rice husk ash (RHA) in the production of geopolymers. 2, 4, 5, 6, 8 and 10 g of RHA was introduced as a source of silica into the geopolymer binders. Sodium silicate to sodium hydroxide ratio of 0, 1 and 1.5 was used for the synthesis. From the compressive and flexural strength tests, increasing the RHA content improved the performance of the binders.

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Optimum compressive and flexural strength was obtained at 10 g (15.25 MPa) and 4 g (3.45 MPa) RHA, content respectively. Binders produced with only sodium hydroxide (SH) solution and RHA showed a significant increase in strength as the RHA content increased.

Keywords: Geopolymer; metakaolin; sodium hydroxide; rice husk ash; sustainable materials.

1. INTRODUCTION

The synthesis of geopolymers basically requires a precursor and an activator, and the sources of these constituent materials vary widely [1,2]. While the precursors may come from industrial/agricultural residues and rock-based materials, the activators are commonly sourced from alkali silicates and hydroxides of alkali metals of Na and K and sparsely sourced from alkali aluminates, carbonates and sulphates and even acid-based activators like phosphoric acid and aluminum phosphate-based activators [3,4].

Geopolymers have shown to be good alternatives to ordinary Portland cement due to its lower CO_2 emission [5] but one set back with this product is its consumption of a large quantity of hydroxides and silicates during synthesis which has a negative effect on other environmental indices such as abiotic depletion, ozone layer depletion, human toxicity, fresh water ecotoxicity, marine ecotoxicity, acidification, eutrophication and terrestrial ecotoxicity [6]. There is thus a need to investigate methods of reducing or substituting the source of the alkaline silicate solution used given that it is reportedly a major contributor to the environmental indices of geopolymers. This will go a long way in making them more sustainable, renewable and acceptable.

With utilising alkaline activators, apart from problems with handling, recent concerns have been raised, attributing a high environmental impact value to them especially with the silicates [5,6]. Based on these reports, current research is directed towards investigating methods to reduce the negative effects this binder has on the environment with more focus on reducing the quantity of synthetic activators and replacing them with more sustainable sources [2]. The contributing environmental index of the silicate is reputed to be more than that of the any of the other activators, thus it has received more attention in terms of seeking alternative, environmentally friendly sources. Some researchers have attempted to incorporate silica in the mix by introducing materials such as silica fume, RHA, glass powder and sugar cane bagasse ash that have a high silica content of silica [7–9]. This research, however, goes further to compare the effect of increasing the silica content in the ash while reducing the amount of silica in the activator.

2. EXPERIMENTAL

2.1 Materials

The kaolin samples used for this research was extracted manually from a natural deposit in Markarfi, Kaduna State, Nigeria. The samples were homogenized, dried and filtered through a 200µm sieve in order to obtain a more uniform size, remove moisture and filter some large impurities. Rice husk ash was used primarily due to the high content of amorphous silica in it and to utilize waste materials generated during rice production The rice husk was also sourced locally from a rice processing mill in Ogoja, Cross River State, Nigeria. Impurities such as stones and straws were removed manually from the husk after which it was pulverized and burned in a muffle furnace at 600^{o}C. Liquid sodium silicate with 2.5 module (SiO_2/Na_2O) and analytical grade sodium hydroxide with 98% purity was procured at Ibra Hadad Nigeria Ltd, an authorized distributor of Honeywell Research Chemicals in Nigeria.

2.2 Characterization of Raw Materials

The determination of the particle size distribution after preparation of the kaolin was done by sieve analysis with nine different sizes of sieves; 5 µm, 10 µm, 20 µm, 30 µm, 50 µm, 75 µm, 100 µm, 150 µm and 200 µm. The kaolin had an average particle size of 33.8µm while the MK (kaolin calcined at 700^{o}C) had an average particle size of 22.1µm. The finer particle size of the MK7 depicts an increase in the surface area of the particles which accelerates geopolymerisation reaction [10-12]. RHA on the other hand had an average particle size of 8.2µm. Chemical composition of the kaolin and rice husk samples were obtained by carrying out x-ray fluorescence while the mineralogical composition was carried out by x-ray diffraction analysis through an Empyrean diffractometer system. XRD was performed with a CuKα radiation varying from 5^{o}

to 75°, 0.05° 2θ step-scan and 1.0 s/step. Finer particle size distribution is a catalyst for geopolymerization and the particle size distribution of the materials is shown in Fig. 1. It can be observed from this result that the calcination aided in producing a finer particle size which can also improve the chemistry of reaction.

2.3 Synthesis, Testing and Characterization of Geopolymer Binders

A small quantity of the kaolin was taken from the extracted source for chemical analysis by means of x-ray spectroscopy and x-ray diffraction. Metakaolin was prepared by calcination of the kaolin in a muffle furnace at a heating rate of 20°C/min from 25°C to 700°C for 2 hours and the sample was also sent for x-ray spectroscopy analysis. A representative sample of the RHA was collected for chemical characterization. The 10M concentrated NaOH solution was prepared by dissolving the right quantity of pellets with water and this was done at least 24 hours prior to usage. With the aim of determining the effect of reducing the silica content from the activator and increasing the RHA content, different Na_2SiO_3/NaOH (SS/SH) ratio of 0, 0.5 and 1.0 was investigated.

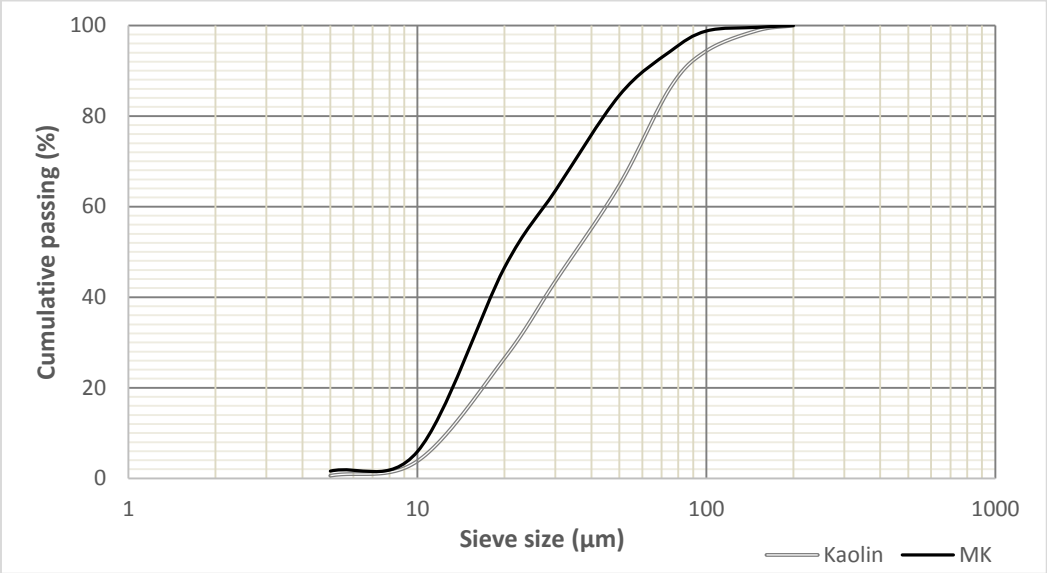

Fig. 1. Particle size distribution of kaolin and metakaolin

Table 1. Mix proportion of samples

	RHA Content	SS/SH ratio	Density (g/cm^3)
1	0	0	2.08
2	0	0.5	2.12
3	0	1	2.18
4	2	0	1.98
5	2	0.5	2.05
6	2	1	2.07
7	4	0	1.87
8	4	0.5	1.92
9	4	1	1.93
10	6	0	1.80
11	6	0.5	1.83
12	6	1	1.87
13	8	0	1.84
14	8	0.5	1.84
15	8	1	1.85
16	10	0	1.81
17	10	0.5	1.83
18	10	1	1.85

Minitab software was used to develop factorial design of experiments for two quantitative factors – RHA content and SS/SH ratio – with six and three levels, respectively. The experimental design was completely randomized, and eighteen (18) mix proportions were developed with three replicates (Table 1).

A two-part mixing method was used to prepare the geopolymer binders. The solid samples (RHA and metakaolin) were measured in the required quantity and mixed while the liquid samples (SS and SH) were also measured based on their ratios. A precursor (solid) to activator (liquid) (S/L) ratio of 1.2 was used to prepare all samples. This was the most suitable ratio obtained after conducting various test mixtures because it had the best consistency and flow. In producing the GP binder, the alkaline solution was poured into a measured quantity of the MK and mixed at a low velocity (190rpm) for 2 minutes and at a higher velocity (210rpm) for 3 minutes before finally mixing at low velocity for another 3 minutes. The final binder was then poured in a silicone mould of size 50mm x 50mm x 250mm, vibrated for 5 mins to remove air bubbles and allowed to cure at ambient temperature for 28 days. Subsequently, the samples were subjected to compressive and flexural testing when they attained their curing time according to the ASTM C109 and ASTM C293, respectively. Samples for compressive test were cut to cubic size of 50 mm x 50 mm x 50 mm. Once the specimen was fractured, the pieces were preserved in an airtight zipper bag.

Minitab software was used to statistically analyze the results after imputing the response (compressive and flexural strength). The aim of this analysis was to get a correlation between the factors and the responses received and to determine the optimal levels of each one. Pareto charts were also presented to provide a better understanding from the ANOVA analysis. This chart shows the contributing effect of each factor to the response, in this instance the responses are flexural and compressive strength. This chart has a reference line that indicates the statistical significance of each term with a t-level associated to the null hypothesis for $\alpha = 0.05$ (5% significance level)

3. RESULTS AND DISCUSSION

3.1 Kaolinite and RHA Characterization

Table 2 shows the chemical composition of kaolin and RHA. The kaolin is composed of silica (51.18) and alumina (32.35) with a silica to alumina ratio of 2.69. The total content of the SiO_2 and Al_2O_3 in the kaolinite is approximately 84% which is typical of most aluminosilicate sources [13], [14]. The diffractogram of the raw clay shows kaolinite with little content of muscovite. Comparing the XRD patterns of the raw and calcined kaolin, diminishing patterns of the peaks of the kaolin are visible and this is because the calcination produces a more amorphous structure which will make the precursor more reactive (Fig. 2).

RHA had a larger content of silica (76.49) and smaller amount of CaO (2.87), Al_2O_3 (4.75) and Fe_2O_3 (3.11). Several other research have recorded an even higher content of silica in RHA above 90% [15] and this significant content of silica is what makes it suitable for being used as an alternative to the silicate activator.

3.2 Effect of RHA Content on the Compressive Strength of the Geopolymer Binder

Fig. 3 depicts the compressive test result of the metakaolin-based geopolymer binder reinforced with RHA at different weight percent and at different SS/SH ratio. Generally, binders produced with only SH solution (SS/SH at 0) produced lower compressive strength but the strength increased from 5.2 MPa to 9.5 MPa with the introduction of the RHA to 10 g. Geopolymer binders produced with SS/SH ratio of 1 had the best performance of 15.25 MPa at 8 MPa while those with SS/SH ratio of 0.5 had a comparable strength of 14.75 MPa.

Table 2. XRF analysis of kaolin and RHA

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
RHA	76.49	4.75	3.11	0.22	2.87	1.41	2.37	0.01	0.02	2.25	6.5

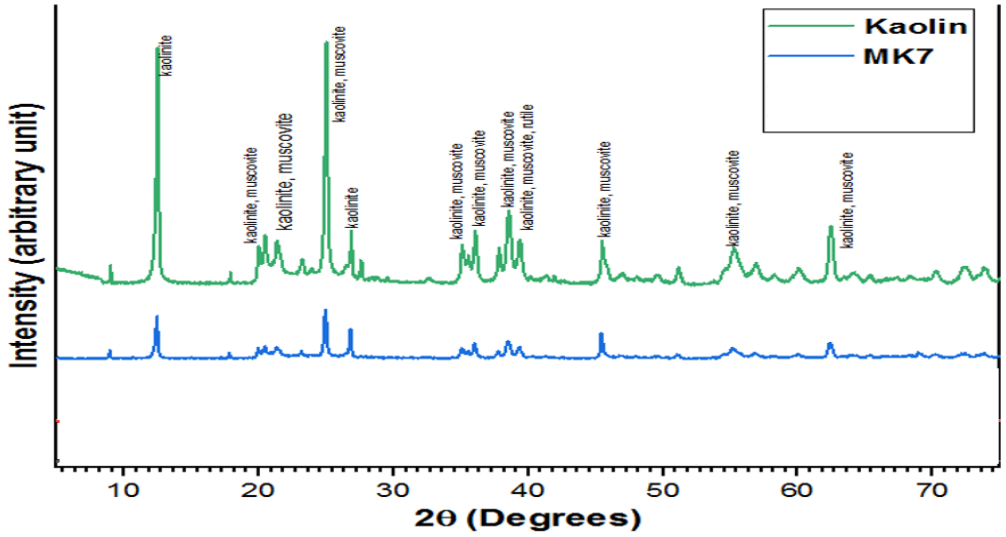

Fig. 2. XRD pattern of raw kaolin and calcined kaolin (700°C)

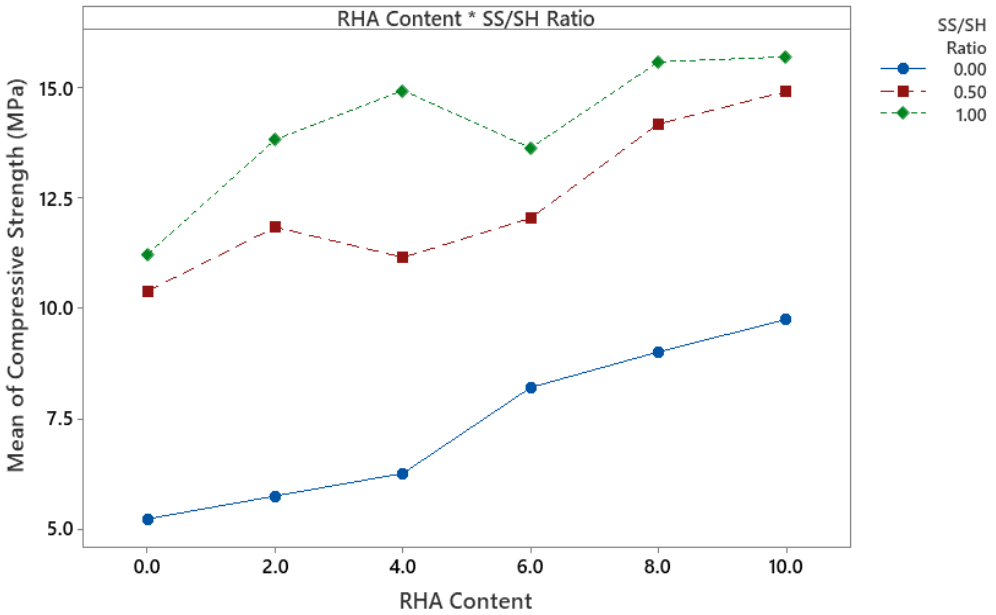

Fig. 3. Interaction plot for compressive strength

The addition of the RHA in the mix in significant quantity (6g, 8g and 10g) increased significantly by about 80%. Lower weights of RHA had no significant contribution to the compressive strength of the geopolymer binder. In some research, it is reported that the increase in fiber/ash content above a particular percentage can lead to a decrease in strength [16]. This however was not observed in this research as the highest strength was obtained at the highest ash content (10 g weight of ash). This may be due to the final particle size of the RHA compared with when fibers are being used. The result obtained suggests that the sodium silicate content can be reduced, and ash introduced to give commensurate properties with those produces with higher synthetic sodium silicate content.

3.3 Effect of RHA Content and SS/SH Ratio on the Flexural Strength of the Geopolymer Binder

Fig. 3 shows the flexural test result of the metakaolin-based geopolymer binder reinforced with RHA at different weights and SS/SH ratio. Like the compressive test results, binders produced with only SH solution produced lower

flexural strength, but the strength increased from 1.5 MPa to 2.6 MPa with the introduction of the RHA to 10 g weight. Geopolymer binders produced with SS/SH ratio of 1 had the best performance of 3.45 MPa at the 4 g weight of RHA. A slight decrease in the flexural strength was obtained from the 6 g weight of RHA and the strength remains similar till the 10 g weight RHA. The results obtained here are relatable to those obtained from the compressive strength. The flexural strength increased with increase in SS/SH ratio and RHA content. Reducing the content of sodium silicate and increasing the content of the RHA can give similar strength.

3.4 Standardized Effect of RHA Content and SS/SH ratio on the Compressive and Flexural Strength of the Geopolymers

The pareto chart attempts to describe the effect of the factors (RHA content and SS/SH ratio) on the responses (compressive and flexural strength) for the geopolymer binders. The factor(s) beyond the dotted line means that the factors were significant at the 95% confidence level. Considering Fig. 5, the RHA content and SS/SH ratio contributed significantly to the compressive strength of the geopolymer binders. The

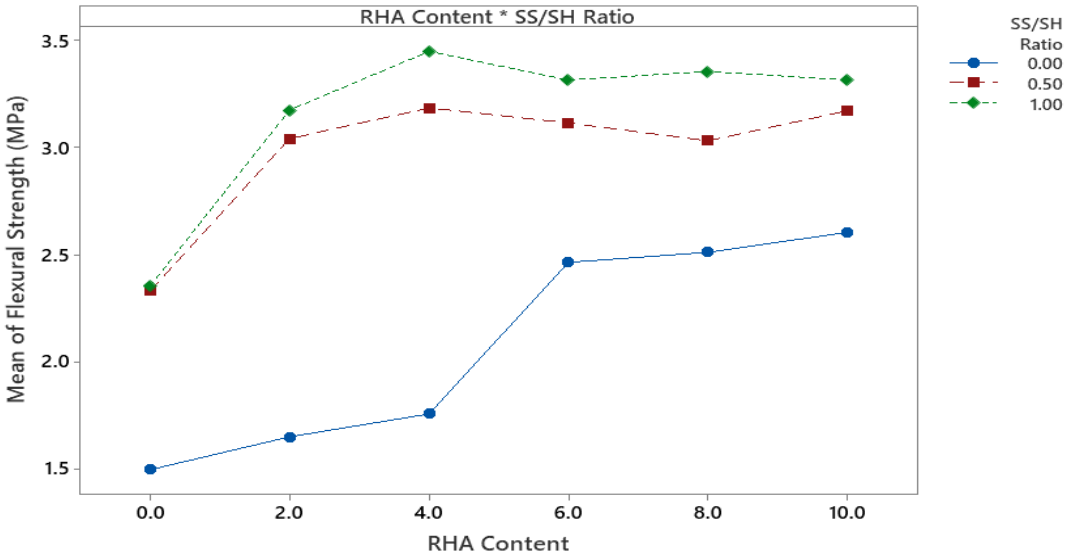

Fig. 4. Interaction plot for flexural strength

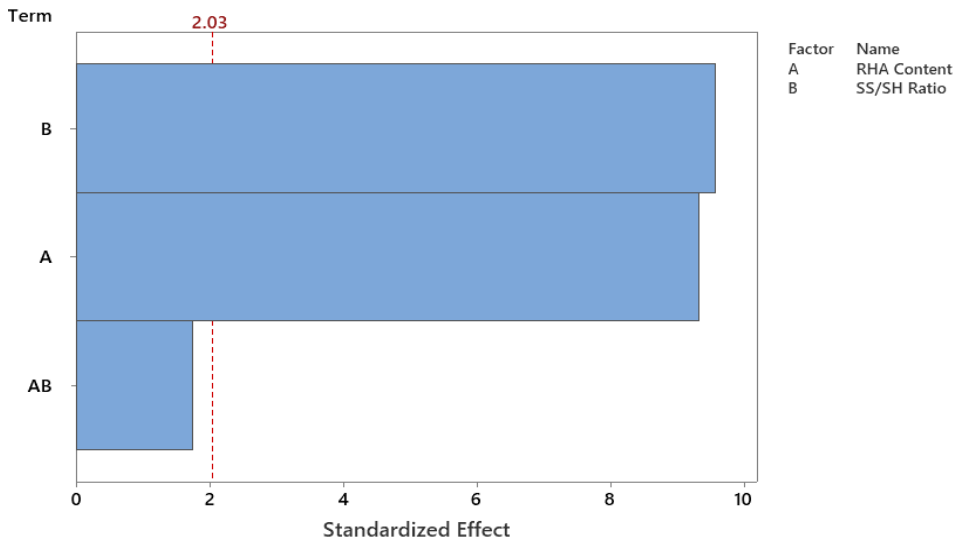

Fig. 5. Pareto chart for compressive strength

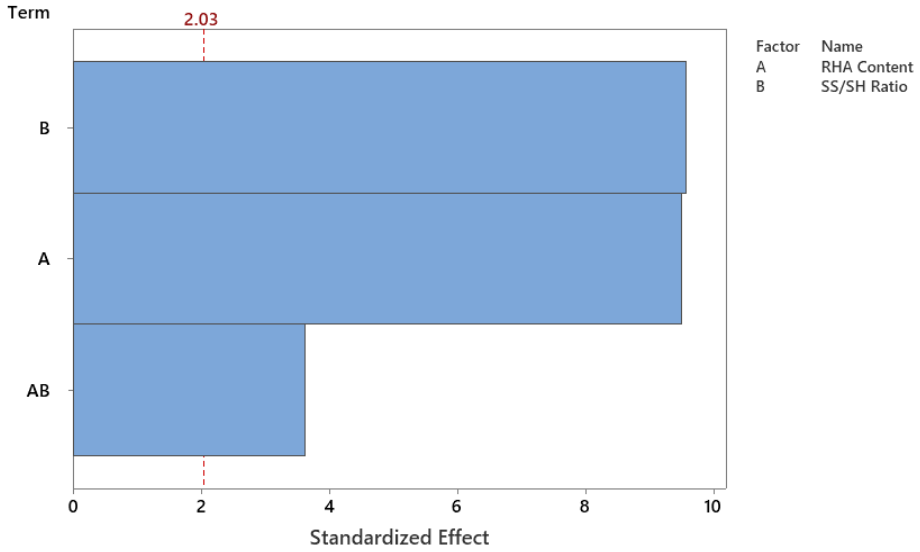

Fig. 6. Pareto chart for flexural strength

combined effect of the factors however did not contribute significantly to the strength of the binders. Similarly, the pareto chart for the flexural strength depicts a significant contribution from both factors; RHA content and SS/SH ratio. Unlike the compressive strength, combined effect of the factors on the flexural strength, got beyond the 95% significant level.

4. CONCLUSION

This research investigated the effect of incorporating RHA (2, 4, 6, 8 and 10g) in the synthesis of geopolymers to reduce the use of synthetic alkali silicate solution and produce more sustainable binders suitable for construction industries. In this study, three SS/SH ratio of 0, 0.5 and 1 were investigated for five different RHA content by weight using the two-part mixing process and cured at ambient temperature for 28 days. The following are the main findings deduced from this study;

1. Increasing the RHA content from 2g to 10g in the geopolymer binders resulted in a constant increase in the compressive strength for all SS/SH ratios (0, 0.5, 1.0) investigated.
2. Increasing the weight of the fibers in the mix resulted in a decrease in the bulk density of the binders from 2.18g/cm^3 to 1.85g/cm^3
3. The Pareto chart shows that both factors (RHA content and SS/SH ratio) have more than 95% level of significance on the mechanical properties of the binders.
4. The optimum RHA content that favoured the compressive strength was 10g (15.25 MPa) while an optimum for flexural strength was obtained at the 4g (3.45 MPa) weight of RHA. It is also worthy to note that reducing the optimum SS/SH ratio by 0.5 and incorporating 10g RHA produced a compressive strength of 14.75 MPa.

DATA AVAILABILITY

The data used to support the findings are available from the corresponding author upon request

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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