Exploring the Suitability of Fly Ash and Rice Husk Ash in One-Part Geopolymer: A Case Study for Sustainable, Low-Carbon Construction.

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Abstract—**The high carbon footprint and energy-intensive production processes associated with clinker-based cement necessitate developing sustainable and environmentally friendly alternatives urgently. This study focuses on the properties of a one-part geopolymer utilizing industrial and agricultural waste materials: Fly Ash (FA)and Rice Husk Ash (RHA). Fly ash was used as the primary aluminosilicate source, and rice husk ash was used as a silicate supplementary material. Solid NaOH was utilized as the alkaline activator. The physical and chemical properties of one-part geopolymer were changed with varying water-to-solid (W/S) ratios (i.e., 0.5, 0.6, 0.7), Si/Al ratios (i.e., 2.5, 3.0, 3.5), and NaOH concentrations (i.e., 6M, 9M, 12M), and they were systematically evaluated to optimize the mix design. Furthermore, microstructural and chemical bonding also depend on W/S, Si/Al, and NaOH concentrations, and the optimised geopolymer revealed well-developed bonding mechanisms and structural integrity. Furthermore, this experiment tested that One-Part Geopolymer (OPG) has lower embedded Carbon (0.4 kg CO_2e), which is more than half of the embedded Carbon (0.85 kg CO_2e) of Ordinary Portland Cement (OPC). It is an excellent sign for the sustainability of OPG over OPC. Therefore, this study underscores the suitability of coal power plant fly ash and industrial waste rice husk ash for synthesizing one-part geopolymers as a transformative solution for advancing environmentally sustainable construction practices that support the UN Sustainable Development Goals 9 and 12.**

Keywords—aluminosilicate, supplementary, activator, geopolymer, cement, strength, microstructure

I. INTRODUCTION

The construction industry has experienced significant growth in recent years, driven by the increasing global population and the need for infrastructure development. Concrete, primarily composed of Ordinary Portland Cement (OPC), is one of the most widely used construction materials due to its availability, versatility, strong mechanical properties, durability, and affordability [1]. The global cement market is projected to reach USD 493 billion by 2028, with a compound annual growth rate of 5.4% between 2023 and 2028 [2]. However, OPC production imposes a substantial environmental burden due to its energy-intensive processes, reliance on non-renewable raw materials, and significant greenhouse gas emissions [3]. Each 1 ton of OPC production consumes approximately 6.1 GJ of energy and 1.5 tons of raw materials, releasing 0.8–1.0 tons of carbon dioxide (CO_2) [4]. Consequently, researchers are exploring alternative cementitious binders to partially or fully replace OPC, aligning with the Sustainable Development Goals of SDG 9 and SDG 12[5].

Geopolymers have emerged as a promising alternative due to their lower carbon embodied energy compared to OPC [6]. Discovered in the mid-1970s [7], geopolymers have found applications in soil stabilisation and construction materials[8]. Among that, geopolymer gives superior properties for weak soft soil as a stabilizer [9] which has minimum environmental impact, owing to its superior compressive strength, shear strength, minimal shrinkage, and enhanced durability [10,11]. According to [9], the unconfined compressive strength(UCS) of the stabilised weak soil by geopolymer ranges from a minimum of 45 kPa to a maximum of 971 kPa. Furthermore, stabilised soil with a 1:3 ratio of fly ash to soil and an alkali activator with a 50:50 ratio of NaOH to Sodium silicate exhibits the highest UCS.

Although the activator in a two-part geopolymer creates barriers for field application as a soft soil stabilizer, the One-Part geopolymer plays a more vital role in soil stabilization than the two-part geopolymer, which has many disadvantages (TABLE I) compared with OPGs [9].

TABLE I. ADVANTAGES OF OPG'S AND DISADVANTAGES OF TWO-PART GEOPOLYMER[9]

Advantages of One-Part Geopolymer(OPG)	**Disadvantages of Two-Part Geopolymer**
✓ Solid form (like OPC) ✓ Active by just adding water (like OPC) ✓ High safety ✓ Easy to manage and transport	✓ High viscosity liquid ✓ Challenging to manage ✓ Highly corrosive ✓ Less safety ✓ Difficult to transport

Recent advancements in OPG production emphasize the utilization of waste alumino-silicate (precursor) materials(TABLE II) to address waste disposal challenges, reduce by-product management costs, and minimize landfill usage [10,11].

TABLE II. Commonly Used Industrial Waste Precursors With Activators [6,7,8]

Alumino-silicate(precursor)	Solid Activators
Fly ash + Ground Granulated Blast Furnace Slag(GGBS)	Na_2SiO_3+ NaOH
Fly ash	NaOH+Na_2SiO_3
Metakaolin	NaOH+$(Na_2SiO_2)_nO$

RHA, a silica-rich agricultural by-product, exhibits high pozzolanic reactivity and minimal contaminants, making it a promising precursor for OPGs. The chemical and physical properties of RHA depend on combustion conditions, with temperatures below 700 °C yielding amorphous ash with enhanced pozzolanic activity [12,13].

However, since geopolymer is an inorganic product with polymeric Si-O-Al-O-Si bonds [14] and RHA lacks sufficient alumina content, it should be associated with an alumina source during the production process. Other than the precursors, alkaline activators also play an important role in geopolymer production as they raise the pH of the reaction mixture and facilitate dissolution [15]. Sodium hydroxide (NaOH), sodium silicate (Na_2SiO_3), potassium hydroxide (KOH), and potassium silicate (K_2SiO_3) are a few of the most widely used activators, either alone or in combination [16]. Among them, Na_2SiO_3 and NaOH have the potential to gradually activate aluminosilicate materials, forming geopolymer binders[17]. However, the commercial production of $Na_2SiO_3((Na_2SiO_2)_nO)$ emits a substantial amount of CO_2 and consumes a higher amount of energy, indicating that it is non-cost-effective and environmentally unfriendly as an activator [16,18].

This study addresses critical environmental concerns, such as CO_2 emissions and waste disposal, by utilizing FA and RHA as precursors and NaOH as an activator in OPG production. The optimal ratios of active silicate and aluminate were experimentally assessed by evaluating the mechanical, microstructural, and durability properties of OPGs, paving the way for sustainable soil stabilizer and building materials. The findings promote the 3R concept (Reduce, Reuse, Recycle) and propose a viable solution for soft soil stabilization in an environmentally friendly and efficient manner.

II. METHODOLOGY

A. *Material and Mix Proportions*

1) *Fly Ash*

The FA used in this investigation is a by-product of coal-fired power plants, with a bulk density of 836.12 kg/m^3(TABLE IV), measured according to ASTM C311 [19] guidelines. Given that the chemical composition of FA varies depending on its source, a chemical analysis was conducted following BS EN 450-1:2012 to determine the oxide composition (TABLE III). The FA was classified as Class F (low calcium oxide content) based on ASTM C618, with its CaO content below 10% by mass, indicating low self-cementitious properties [20]. The analysis revealed that the reactive silica (amorphous) content was 11%, which is less than the 25% recommended in BS EN 450-1:2012 [20], further confirming the minimal pozzolanic reactivity of the FA. As a result, a supplementary reactive silicate source is required to enhance the material's reactivity and make it suitable for use as a precursor in OPG synthesis.

2) *Rice Husk Ash*

To determine the most suitable type of RHA for geopolymer production, samples were collected from two sources: 1) RHA from a brick manufacturing industry, and 2) RHA from a boiler in the paper recycling industry. Both samples underwent a thorough investigation of their physical and chemical properties. The RHA obtained from the brick industry was burned at a temperature of 700 ˚C and had a bulk density of 283.93 kg/m^3, according to ASTM C311 guidelines [19], while the RHA collected from the boiler in the paper recycling industry was burned at a temperature of 400 ˚C, with a bulk density of 316.28 kg/m^3 (TABLE IV). Additionally, the Loss on Ignition (LOI) test [19] indicated that the boiler RHA contained a higher amount of unburnt carbon (LOI = 39.4%) compared to the brick industry RHA (LOI = 3.48%) (TABLE IV). Previous studies have shown that a high amount of unburnt carbon in RHA may reduce the resulting OPG's compressive strength [21].

TABLE III. Chemical Analysis Of Coal Power Plant FA

Test (% by mass)	Results (% by mass)	The specification given in BS EN 450-1:2012
LOI	3.7	Shall not be greater than 5
SiO_2	45	
Al_2O_3	31.8	
Fe_2O_3	4.4	
MgO	1.1	Shall not be greater than 4
CaO	9	--------
Chloride content	< 0.01	Shall not be greater than 0.10
Total Alkali equivalent as Na_2O	0.4	Shall not exceed 5
SO_3	0.5	Shall not be greater than 3
Reactive Silica	11	Shall not be less than 25

TABLE IV. Bulk Density And Loss Of Ignation(LOI) Of FA, Boiler RHA, Brick Industry RHA Samples

Sample	Bulk density (kg/m³)	LOI (%)
FA	836.12	3.7
Paper industry boiler RHA	316.28	39.4
Brick industry RHA	283.93	3.48

X-Ray Diffraction (XRD) analysis (Regaku Ultima IV; 40 kV, 30 mA), as shown in Fig. 1, revealed that the boiler RHA contained a higher amount of crystalline SiO_2 than the brick industry RHA. However, both RHA samples exhibited low crystallinity, suggesting that they were predominantly amorphous, which is indicative of higher reactivity. Visual observations confirmed that the brick industry RHA was light grey (by comparison, Fig. 2(b) and Fig.3(b)), which aligned with its low LOI value of 3.48%, indicating a low percentage of unburnt carbon. X-Ray Fluorescence (XRF) analysis further revealed that 92.61% of the oxide compounds in the brick industry RHA were silica, while in the boiler RHA, silica accounted for 94.55% (TABLE V). Additionally, the brick industry RHA contained 1.34% Al_2O_3, whereas the RHA from the boiler in the paper industry had no detectable Al_2O_3 content (TABLE V). Consequently, the use of brick industry RHA can result in a stronger binder compared to the boiler RHA from the paper industry. Both types of RHAs exhibited low bulk densities (Table IV), comparable to that of FA. Based on the combined physical and chemical properties of both RHA samples, as revealed through the respective analyses, the brick industry RHA was selected as the supplementary silicate source for OPG production in the current experimental study [22].

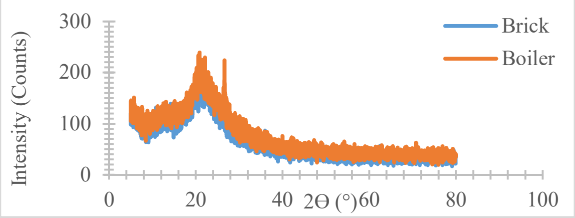


Fig.1. XRD analysis of the Brick industry and the Boiler industry RHA by Regaku Ultima IV (40 kV,30 mA)

(a) (b)

Fig.2. RHA generated from the Boiler in the paper

(a) (b)

Fig.3. RHA generated from the Brick manufacturing industry

TABLE V. XRF ANALYSIS FOR PAPER INDUSTRY BOILER RHA AND BRICK INDUSTRY RHA

Element	Weight (%) in paper industry boiler RHA	Weight (%) in brick industry RHA
MgO	0.73	0.3
Al_2O_3	0	1.34
SiO_2	94.55	92.61
P_2O_5	1.86	1.83
SO_3	0.02	0
K_2O	1.87	2.06
CaO	0.56	0.91
TiO_2	0.02	0.04
MnO_2	0.16	0.21
Fe_2O_3	0.23	0.7
ZnO	0.005	0.002
Re_2O_7	0.004	0

3) Alkaline Activator

An inorganic substance, namely granular sodium hydroxide (NaOH), was utilized as a solid activator to facilitate the dissolution of aluminosilicate in the geopolymer system. The use of granular NaOH in geopolymer production offers a simplified and more time-efficient alternative to traditional alkaline solutions, as it obviates the need for prior preparation of a NaOH solution before mixing [11]. For this study, 98% pure commercial-grade NaOH was employed as the activator.

B. Preparation of Samples

In the production of OPG, critical parameters such as the Water/Binder (W/B) ratio, Si/Al ratio, and NaOH concentration play a pivotal role in determining the properties of the resultant binder [17]. Ye et al. [23] explored the influence of the W/B ratio on the compressive strength of OPG derived from red mud, examining ratios of 0.55, 0.60, and 0.65. Their findings indicated that, in the absence of dispersants, compressive strength increased as the W/B ratio decreased. Consequently, this study employed W/B ratios of 0.5, 0.6, and 0.7, aligning with the range identified in their research. Similarly, Hajimohammadi[24] investigated the effect of varying the Si/Al ratio at 1.5, 2.0, and 2.5, concluding that a lower Si/Al ratio accelerated the dissolution of geothermal silica and the formation of the geopolymer matrix. Building on this insight, the present study examined Si/Al ratios of 2.5, 3.0, and 3.5 to determine the optimal value. Furthermore, Memon et al. [25] analysed the impact of NaOH concentration on the fresh properties and compressive strength of self-compacting geopolymer concrete, testing concentrations ranging from 8M to 14M and observing peak compressive strength at 12M. Based on these findings, NaOH concentrations of 6M, 9M, and 12M were selected for this study.

Cylindrical samples, each with a diameter of 38 mm and a length of 76 mm, were prepared for all mix designs and subjected to curing under air for durations of 7, 14, 28, and 90 days(Fig. 4).

C. Testing Procedure

The cylindrical samples, which were cured in air, were subjected to testing after the designated curing periods of 7, 14, 28, and 90 days. The tests conducted included measurements of compressive strength, with the specified curing durations. Compressive strength tests were performed using a computer-controlled universal testing machine, applying a constant strain rate (10mm/min) [26]. The resulting stress-strain curves were utilized to determine the compressive strengths. For the 90days cured samples, additional analyses were carried out by Scanning Electron Microscopy(SEM) to investigate the geopolymer's microstructure and both micro and nano morphological characteristics of the samples(Fig.4).

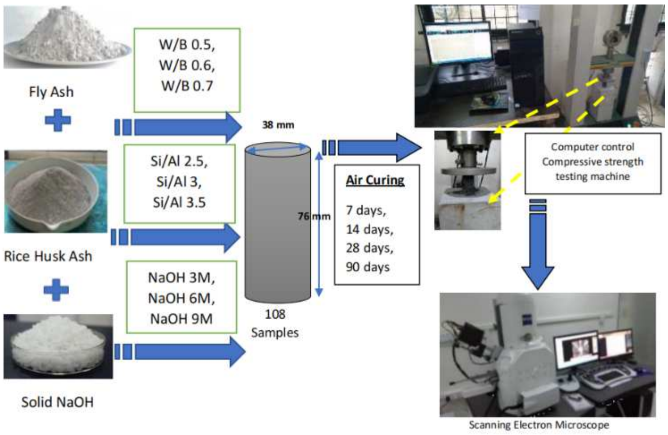


Fig.4. Sample preparation and testing procedure

III. RESULTS AND DISCUSSION

Fig.5 presents the compressive strength results of W/B ratio samples of 0.5, 0.6 and 0.7 at 7, 14, 28, and 90 days. For samples tested at 7 and 14 days, those with a W/B ratio of 0.7 exhibited the highest compressive strength. At 28days, the W/B 0.6 samples achieved the highest compressive strength. After 90days of curing, the W/B 0.6 samples achieved the highest compressive strength, measuring 1.7MPa.

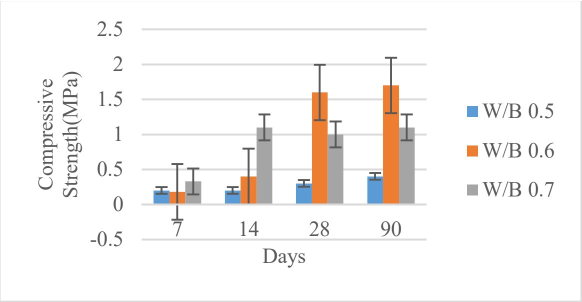
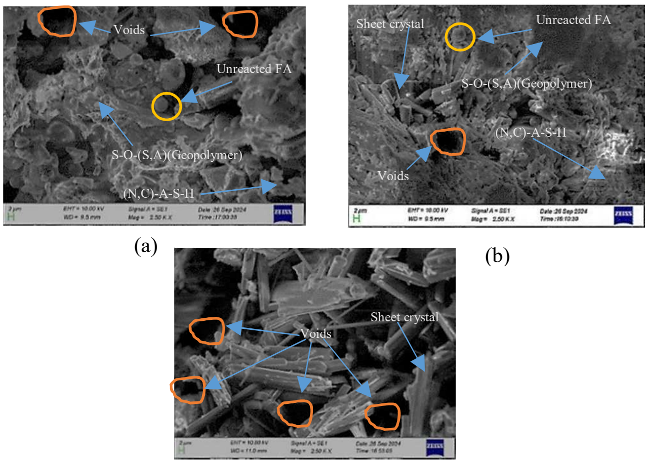


Fig.5. Compressive strength vs. days of W/B 0.5, 0.6 and 0.7 samples

Moreover, SEM images presented in Fig. 6(a), (b) and (c), under identical magnifications, revealed that the OPG with a W/B ratio of 0.6 exhibited a more uniform structure with fewer voids compared to the W/B 0.5 and W/B 0.7 samples.



(a)

(b)

(c)

Fig.6. SEM image of (a)W/B 0.5, (b)W/B 0.6 and (c)W/B 0.7 OPGs in 2.50KX magnification

According to Fig. 7, the Si/Al ratio of 3.5 exhibited the highest compressive strength after 7days of curing. However, after 14 days of curing, the sample with a Si/Al ratio of 2.5 demonstrated the maximum strength enhancement.

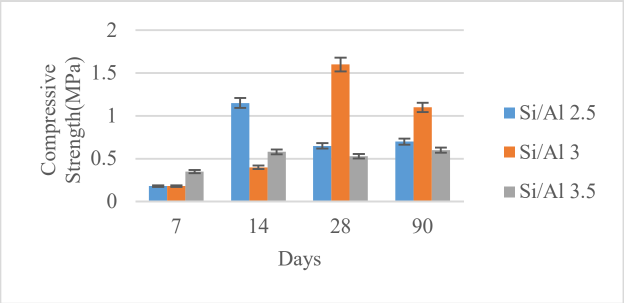
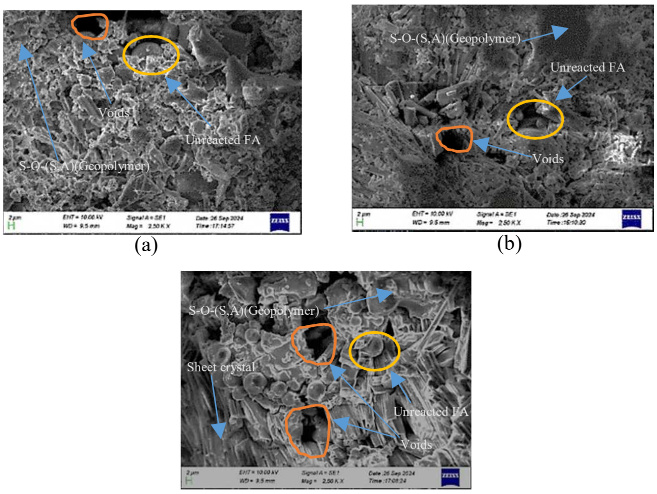


Fig.7. Compressive strength Vs Days of Si/Al 2.5, Si/Al 3 and Si/Al 3.5

In contrast, after 28 and 90 days of curing, the Si/Al 3.0 samples exhibited the highest compressive strength. Furthermore, according to SEM images of Fig. 8(a), (b) and (c), it revealed that the microstructures of OPG samples with Si/Al ratios of 3.0 and 2.5 were characterised by fewer voids and a more uniform structure compared to the Si/Al ratio of 3.5. This aligns with observations by Hajimohammadi [24], who reported a more homogeneous microstructure with smaller pores in two-part geopolymer systems as the Si/Al ratio increased within the range of 1.5 to 2.5.



(a)

(b)

(c)

Fig.8. SEM image of (a) Si/Al 2.5, (b) Si/Al 3.0 and (c) Si/Al 3.5 OPGs in 2.50KX magnification

In the next stage of the experiment, the concentration of the activator (NaOH) in the OPG matrix was varied to 6M, 9M and 12M to observe the physical and morphological characteristics of OPGs. According to Fig. 9, higher concentration NaOH samples show negligible strength in the early stages. In 7 days and 14 days, 12M has very low strength, and it increases domestically after 28days and 90days. In 14days, 6M and 9M have nearly equal compressive strength. In 90days, 12M samples have higher compressive strength compared with 6M and 9M samples.

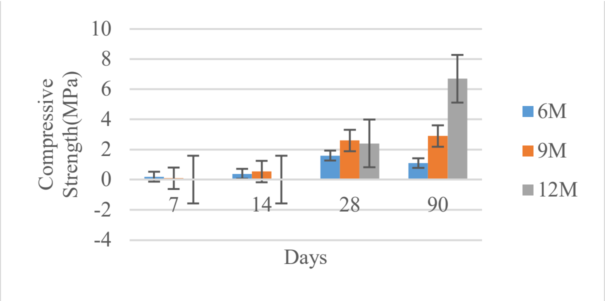
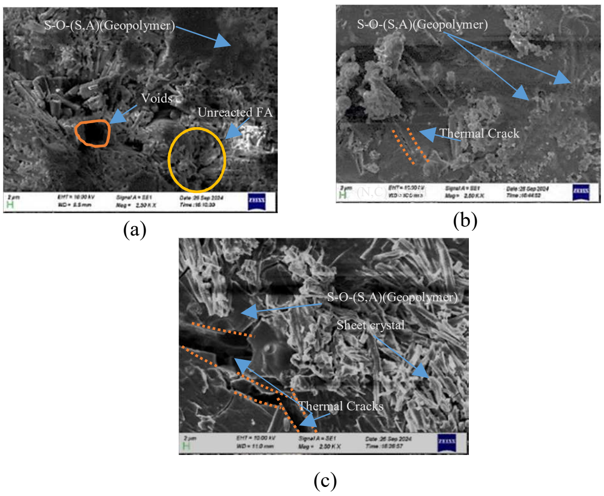


Fig.9. Compressive strength Vs Days of A6M, A9M and A12M

According to SEM analysis of 6M, 9M, and 12M (Fig. 10(a), (b) and (c)), 9M concentration samples have a uniform and less void microstructure than 6M and 12M samples. According to Fig. 10(c), thermal cracks are shown in micro microstructure of 12M samples. Therefore, high activator concentration creates a thermal crack due to the high heat generated in the reaction. Although it has the highest compressive strength of 6.7MPa in all samples.

Furthermore, according to Fig.9, the compressive strength of 12M concentration OPG samples is notably increased with time period than other OPG samples.

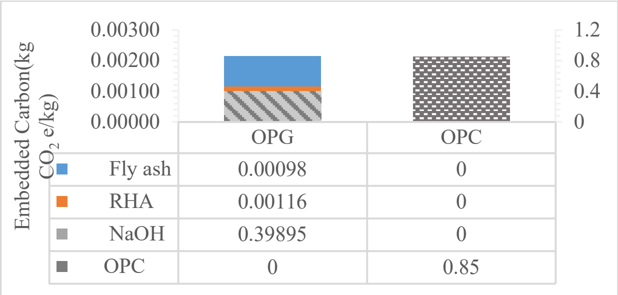


(a) (b) (c)

Fig.10. SEM image of (a) 6M, (b) 9M and (c) 12M OPGs in 2.50KX magnification

IV. EMBODIED CARBON COMPARISON BETWEEN ONE-PART GEOPOLYMER (OPG) AND ORDINARY PORTLAND CEMENT (OPC)

According to Davidovits[27], approximate values of cement CO_2 emissions are in the range of 0.85 to 1.00 kg CO_2e/kg. In Australia, the most recently reported emission factor for cement production is 0.82 kg CO_2e/kg [28]. Fly ash is a waste by-product generated from coal-burning power plants, and CO_2 is emitted due to the results of fly ash capture, milling, drying, and transportation. According to Turner et.al [28], the CO_2 emission factor value for fly ash of 0.027 kg CO_2e/kg. Same as Fly ash, Rice husk ash is also a by-product from the brick industry. Due to that, it can be taken as 0.0027 kg CO_2e/kg by considering the CO_2 emission factor of fly ash. The Sodium Hydroxide (NaOH) (Caustic Soda) is produced from the basic oxide of Sodium Oxide (NaO) dissolved in water. The Caustic Soda manufacturing process is usually conducted by the electrolysis technique. Sodium hydroxide is formed composed of Chlorine (Cl) contained in seawater, by a chlor-alkali method using the electrolysis technique. So that, Davidovits J et.al [27] recommended a CO_2 emission factor for NaOH of 1.915kg CO_2e/kg. According to the above consideration, the embedded Carbon of OPG and OPC is shown in Fig.11. By considering these experimental results, 1000 g of OPG has 361.50 g of Fly ash, 430.17g of RHA and 208.33g of NaOH.



	OPG	OPC
Fly ash	0.00098	0
RHA	0.00116	0
NaOH	0.39895	0
OPC	0	0.85

Fig.11. Embedded carbon in OPG and OPC (by considering 1000 g)

According to Fig.11, OPG has lower embedded Carbon (0.4 kg CO_2e), which is more than half of the embedded Carbon (0.85 kg CO_2e) of OPC.

V. CONCLUSION

The test results identified the optimal water-to-solid ratio and Si/Al ratio for OPG as 0.6 and 3.0, respectively, yielding the highest long-term compressive strength and favourable microstructural development. However, despite varying the W/B and Si/Al ratios, the compressive strength after 90 days of curing remained significantly lower when the NaOH concentration was maintained at a lower level. In contrast, increasing the NaOH concentration (9M and 12M), experiments resulted in simultaneous improvements in both long-term compressive strength. Although it is important to note that elevated NaOH concentrations lead to higher reaction temperatures, which may induce thermal cracking in the geopolymer. This trade-off highlights the need to balance strength improvements with potential durability concerns and increased production costs.

The optimized mix of W/B 0.6, Si/Al 3 and activator concentration 12M achieved a compressive strength of 2.4MPa in 28days under ambient conditions. Furthermore, it has long-term advanced performance in 90 days with a compressive strength of 6.7MPa under ambient conditions. Furthermore, this experimental OPG has lower embedded Carbon (0.4 kgCO_2e), which is more than half of the embedded Carbon (0.85 kgCO_2e) of OPC. It is a great sign for the sustainability of OPG rather than OPC.

These findings demonstrate that utilizing a coal power plant and agricultural by-products such as FA and RHA can impart cement-like properties to OPG without additional energy input. This positions OPG as a sustainable alternative to clinker-based cement, significantly reducing the carbon footprint and energy demand. Its potential applications include soil stabilization, pavement construction, and other infrastructure needs, supporting the development of environmentally friendly construction materials. However, the performance of OPG with different types of soil needs to be studied further.

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