Can geopolymer mixture be a solution for utilizing waste glass and basic oxygen furnace slag as aggregates?

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Abstract. This research studies the influence of aggregate proportions, namely Waste Glass Sand (WGS) and Basic Oxygen Furnace Slag (BOFS), on the expansion and compressive strength characteristics of both geopolymer and Ordinary Portland Cement (OPC) mixtures. The research includes an experimental program in which geopolymer mixtures were prepared with different combinations of WGS and BOFS contents (0/100, 25/75, 50/50, 75/25, 100/0, respectively) alongside reference OPC samples containing only WGS or BOFS. The samples prepared for testing expansion behavior were cured under 2 conditions: water submersion and 1M NaOH solution submersion, both maintained at 80°C. Regardless of different combinations of SWS and BOFS, the 28-day compressive strength results for all geopolymer mixtures exceeded the value of 40 MPa. The geopolymer samples experienced different expansion patterns depending on factors such as aggregate content and curing conditions. Specifically, a mixture with 25% WGS and 75% BOFS showed rapid expansion in water, while others maintained expansion within acceptable limits (0.1% threshold value). This was analyzed via literature review, highlighting the role of the alkali-silica reaction. At the same time, the OPC mortar mixture demonstrated significant expansion values, particularly in the NaOH solution, which is connected to the interaction between cement hydration products and NaOH. Based on compressive strength and expansion results, the geopolymer mixture having the 50/50 combination of WGS and BOFS seems to be the optimum mixture in compressive strength and expansion behavior.

1 Introduction

Concrete is one of the most frequently used materials in construction. Thus, ordinary portland cement (OPC) is traditionally used as the primary binder in concrete production. However, the manufacturing of OPC produces a significant amount of carbon dioxide (CO_2),

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contributing to a major environmental issue related to climate change. As the world strives to address problems associated with global CO_2 emissions, it is essential to develop alternative methods for concrete production that do not rely on OPC [1]. Moreover, the issue of managing industrial waste materials and municipal waste materials is rising due to their ecological impacts on the environment.

Geopolymer concrete (GPC) is becoming one of the solutions that can address the issues by making a considerably eco-friendly one that utilizes these waste materials. Geopolymer concrete can be considered a promising alternative to ordinary concrete made of OPC with a significant carbon footprint, offering a sustainable pathway for using such industrial by-products. Characterized by its binder system formed from the reaction of aluminosilicate-rich materials with alkaline solutions, geopolymer concrete provides a viable method to reuse these waste materials. Not only this, but GPC has shown outstanding mechanical properties that are superior to the properties of ordinary concrete, exceeding the expectations of researchers [2].

With increasing economic and ecological significance, the utilization of Waste Glass Sand (WGS) and Basic Oxygen Furnace Slag (BOFS), one of the steelmaking by-products, has become an interest in current GPC studies. Often left in landfills, these waste products represent a lost opportunity for resource reallocation and sustainable materials utilization [3]. However, the presence of free lime (f-CaO) and free magnesia (f-MgO) in BOFS, known for causing expansion in hydration, limits its use as aggregates in construction applications. Moreover, using WGS in regular concrete also causes volumetric expansion problems due to the formation of an alkali-silica reaction (ASR).

Therefore, this study investigates the potential of geopolymer mixtures to stabilize these reactions. In geopolymer concrete, the interaction between f-CaO, f-MgO, and free silicon (f-Si) from Na_2SiO_3 can lead to the formation of stable substances like $CaSiO_3$ and $MgSiO_3$, potentially mitigating expansion issues associated with BOFS [4]. Moreover, Kozhageldi et al. [5] report that major alkalis in pore solution that induce ASR are consumed by the amorphous component in the FA and GGBFS in less geopolymer mixture, leading to converting alkalis to cementitious binders and zeolite forms. As a result, fewer alkalis remain in the geopolymer mixture and minimize the chance of causing the ASR (less expansion) compared to ordinary Portland cement concrete/mortar.

In spite of some existing research, the impact of the combined use of WGS and BOFS on key properties like expansion behavior and compressive strength has not been thoroughly researched. Therefore, this study aims to evaluate the influence of aggregate proportions on the expansion and compressive strength characteristics of geopolymer and OPC mixtures. It is essential to understand the behavior of these materials in combination and within the system of geopolymerization technology, as it can potentially be one of the crucial sustainable construction practices. This paper is structured first to present the experimental program, followed by test results and discussions analyzing the trends shown in the study.

2 Experimental program

2. 1 Materials and Mix proportions

Three types of cementitious materials were used in this study. ASTM class F fly ash (FFA) and ground granulated blast furnace slag (GGBFS) were used as binders for the geopolymer mixture. OPC was used for reference mixtures of normal mortar. The specific gravity (SG) of FFA, GGBFS, and OPC are 1.87, 2.99, and 3.15, respectively. The mixture proportion considered each material's specific gravity and absorption capacity.

The SG of WGS and BOFS aggregate is 2.51 and 3.16, respectively. Table 1 presents the chemical compositions of both WGS and BOFS aggregate.

Table 1. Chemical Composition of WGS and BOFS in % by weight.

	SiO_2	Al_2O_3	Fe_2O_3	CaO	SO_3	MgO	TiO_2	Na_2O	K_2O	MnO	ZnO
WGS	66.20	1.78	0.66	8.45	0.44	2.48	0.06	12.27	1.20	0.02	0.10
BOFS	8.73	1.66	24.82	47.95	1.65	11.26	0.38	0	0.11	1.88	0.82

Tables 2 and 3 summarize the materials used for normal OPC mortar and geopolymer mixtures. For clarity, MWGS-0% means the normal mortar with 0% WGS and 100% BOFS aggregates, whereas WGS-25% means geopolymer mixture with 25% WGS and 75% BOFS aggregates.

Table 2. Mixture proportions for geopolymer concrete (kg/m^3).

Mixture	FA	GGBFS	BOFS	WGS	AAS
WGS-0%	579.72	386.48	414.08	0	386.48
WGS-25%	575.12	383.41	308.10	102.70	383.41
WGS-50%	570.59	380.39	203.78	203.78	380.39
WGS-75%	566.14	377.42	101.10	303.29	377.42
WGS-100%	561.75	374.50	0	1401.25	374.50

Table 3. Mixture proportions for regular mortar (kg/m^3).

Mixture	Cement	BOFS	WGS	Water
MWGS-0%	555.00	1777.18	0	260.85
MWGS-100%	555.00	0	1428.49	260.85

For geopolymer mixtures, an alkaline-activating solution (AAS) consisting of solid sodium hydroxide (NaOH), liquid sodium metasilicate (Na_2SiO_3), and water was used to activate the process of geopolymerization. NaOH was dissolved in the water, having a resultant concentration of 10M. Constantly remained proportions are water to binder ratio (w/b) = 0.35, FFA/GGBFS = 1.5, aggregate/b = 0.3, AAS/b = 0.4, and Na_2SiO_3/NaOH (sol) = 1.5. Keeping these parameters constant, for geopolymer mixture, the variables are set to be proportions of WGS and BOFS (100% BOFS, 75% BOFS + 25% WGS, 50% BOFS + 50% WGS, 25% BOFS + 75% WGS, 100% WGS).

Reference OPC mortar mixtures had only 2 different proportions of WGS and BOFS (100% BOFS and 100% WGS). The mixture design of reference was made by fixing water to cement ratio (w/c) = 0.4. Finally, The aggregate gradation was fixed according to ASTM C1260 and presented in table 4.

Table 4. Aggregate gradation.

Sieve number	#8	#16	#30	#50	#100
Proportions (%)	10	25	25	25	15

2.2 Mixing Procedure Test Methods

The mixtures were prepared using a Hobart mixer with 5 L volume. AAS was prepared 24 hours before mixing. First, a dry mix of cementitious materials was performed for 30 seconds at a slow speed. Then, water and AAS were added simultaneously and mixed slowly for 1 minute. The last added materials were aggregates, and mixing continued for 1 minute at a slow rate. Then, the medium mixing mode was switched, and the entire substance was mixed for 1 minute. Afterward, hand scraping lasted for 1.5 minutes, leading to another mix using a fast mixer mode lasting 1 minute.

This procedure, including hand and machine mixing, ensures the homogeneous consistency of the mixture. After mixing, the mixtures were placed into molds of compression cubes and expansion bars, according to each test requirement. Table 5 depicts all the testing methods and the number of samples conducted for each test, sample size, and testing age used in this study. After 2 hours, the samples were demolded and cured according to the testing requirements until the testing day. For the expansion test, the bar samples were submersed in water or at 1M of NaOH solution heated by the oven at an inside temperature of 80°C. For the compression test, the samples were stored in wet paper towels wrapped under plastic sheets to ensure moisturizing and avoid vaporization of water. The paper towels were changed every 3 days to ensure that the paper excluded drying of samples.

Table 5. Testing methods.

Test method	Test specification code	Testing age	No. of the test specimen	Dimensions
Compressive Strength	ASTM C109	7/14/28 days	4	50x50x50 mm
Expansion	ASTM C1260	3 and 4-day interval	4	25x25x285 mm

The Compressive strength test and expansion test were mainly conducted. The compressive strength of both geopolymer mixture and OPC mortar mixture was tested according to ASTM C109/C109M Standard Test Method for Compressive Strength of Hydraulic Cement Mortars [6]. There were 4 cubic specimens tested with dimensions 50 x 50 x 50 mm with 0.350 MPa/s rate for each mixture. The testing was performed for 7, 14, and 28 days of strength development. Therefore, a total of 84 cubic samples were tested.

The expansion behavior of the mixtures was examined according to the ASTM C1260 Standard Test Method for Potential Alkali Reactivity of Aggregates (Mortar-Bar Method) [7]. Eight bar specimens with 25 x 25 x 285 mm dimensions were prepared for each mixture type. Half of them were submerged in water and put in an oven at 80 °C. The other group was submerged in the solution of 1M NaOH and put in the oven at the same temperature. The measurements were taken periodically until 28 days.

3 Results and Discussion

3.1 Compressive strength

The compressive strength results for geopolymer and normal mortar mixtures are presented in Fig. 1. For all normal mortar and geopolymer mortar samples, except for geopolymer mixtures with 50% and 100% WGS replacement, the compressive strength increased gradually with days passed. Moreover, all geopolymer mixtures exceeded 40 MPa for 28-day compressive strength, while regular mortar samples did not reach such indicators. This shows that geopolymer concrete is more promising in terms of having high-strength mechanical properties. In addition, the maximum strength gain for regular mortar mixture is indicated for the mixture of MWGS-0% and is equal to 4.1 MPa from 7-day to 28-day compressive strength. At the same time, the minimum strength gain value for the geopolymer mixture is 11.1 MPa for WGS-100%, and the maximum strength gain is 19.7 MPa for WGS-50%.

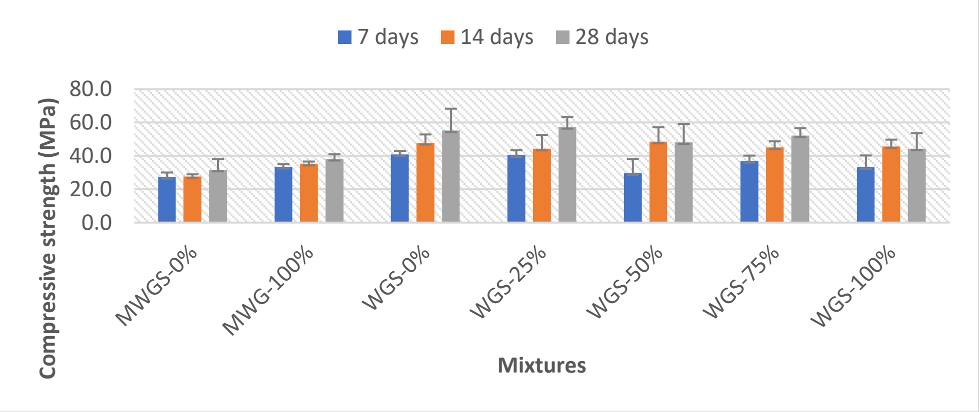

Fig. 1. Compressive strength test results of geopolymer and regular mortar mixtures.

Interestingly, WGS-50% and WGS-100% mixtures also showed the least strength among geopolymer mixtures in the first 7 days. This result may be due to the compaction non-uniformity. Geopolymer mixtures of WGS-0% and WGS-25% had the highest average compressive strength values. This may also be attributed to the high proportions of BOFS in these mixtures. It should be noted that BOFS has higher specific gravity than WGS, and well-crystallized BOFS (less porous structure) provides higher strength compared to WGS. Tukaziban et al. [8] have a similar finding. They explain that BOFS is rich in calcium, silica, and alumina. The calcium content in BOFS can react with silica and alumina to form additional C-A-S-H (calcium aluminate silicate hydrate) gel, a binder phase that significantly contributes to strength [9]. Also, the silica (SiO_2) component in WGS may participate in the pozzolanic reaction, resulting in more C-A-S-H production. That is the reason why the WGS-25% mixture has the highest compressive strength.

However, as WGS content increases, the compressive strength of the geopolymer mixture decreases. WGS primarily consists of silica (SiO_2). While silica can participate in the geopolymerization process, it does not react as a material with higher alumina content. High amounts of WGS may not contribute significantly to the chemical reaction that forms the geopolymer matrix, thus not adding much to the compressive strength. Additionally, the smooth and non-porous surface of glass particles might lead to a weaker bond in the geopolymer matrix, as Ahmed et al. suggested [8].

3.2 Expansion behavior

As stated earlier, the volumetric expansion behavior was examined under 2 test conditions. While the specimens submersed in water monitored the expansion due to $Ca(OH)_2$ formation, the specimens submersed in 1M NaOH solution examined the expansion due to ASR gel formation. The results of the expansion characteristics in water are plotted in Fig. 2. As presented in Fig. 2 (a), the expansion values for all geopolymer mixtures, except for the WGS-25% mixture, are below 0.1%, and expansion behaviors of these geopolymer mixtures fluctuated between -0.08% and 0.02% expansion. The fluctuation of the expansion in the geopolymer samples submersed in water can be explained by chemical heterogeneity within the mix, such as variations in the reactivity of the alkaline activators. These chemical variations might not be visually apparent but can significantly affect the geopolymerization process, as indicated by Thaarrini and Ramasamy's research on using different industrial by-products in geopolymers [10].

Interestingly, the WGS-25% geopolymer mixture reached 0.1% in 2 weeks and was broken before 28 days. The expansion behavior of the WGS-25% mixture can be explained by the localized expansion that happens when the curing space is limited, and water is not enveloping the samples properly. Tukaziban et al. [8] indicate that BOFS inherently leads to volumetric expansion behavior due to the chemical reaction between f-CaO and water. The WGS-25% mixture samples stayed between other samples, which might have caused this issue. Fig. 3 (a) supports the localized expansion due to $Ca(OH)_2$ formation for the WGS-25% mixture.

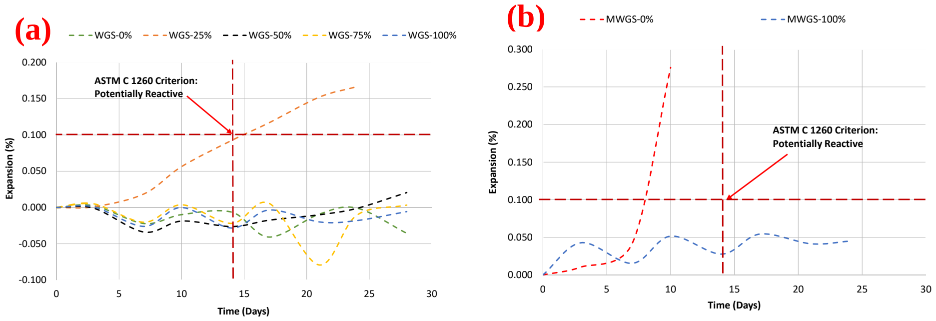

Fig. 2. Expansion behavior of specimens submersed in water: (a) geopolymer mixture and (b) normal mortar mixture.

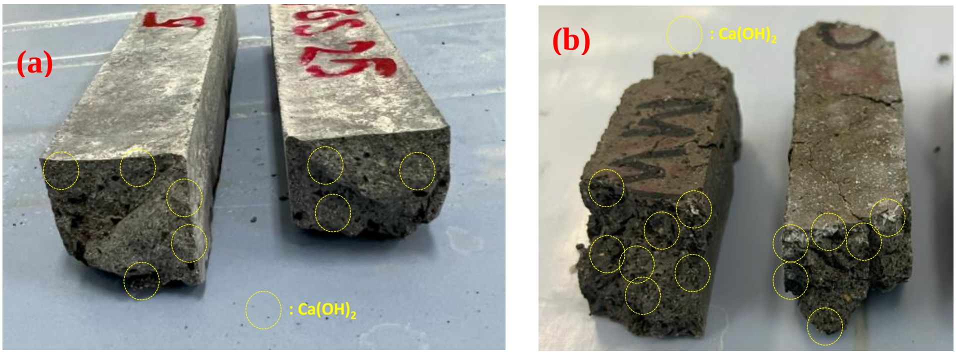

Fig. 3. Broken samples due to the localized expansion caused by $Ca(OH)_2$ formation: (a) WGS-25% geopolymer mixture and (b) MWGS-0% normal mortar mixture (100% BOFS).

Fig. 2 (b) shows the expansion behavior of normal mortar mixtures submerged in water. As expected, the MWGS-0% mixture (OPC mortar mixture with 100% BOFS aggregate) was broken after 10 days, while the MWGS-100% mixture (OPC mortar mixture with 100% WGS aggregate) did not exceed 0.06% expansion, though it showed the fluctuated expansion behavior. The chemical reaction that converts f-CaO in BOFS to $Ca(OH)_2$ in a normal mortar mixture causes the expansion and pressure to exceed the tensile strength of mortar bar samples, resulting in cracks and fractures (Fig. 3 (b)). Since the mortar mixture containing 100% WGS had no ASR in the water, the mixture showed no cracks.

The expansion characteristics of geopolymer and normal mortar mixtures submersed in 1M NaOH solution are shown in Fig. 4. All the geopolymer mixtures had gradual expansion under acceptable limits of up to 14 days. However, after 21 days, the WGS-0% mixture expanded rapidly and reached 0.39% at 28 days. Moreover, WGS-75% and WGS-100% mixtures slightly passed over 0.1% at 28 days. This result is different from Tukaziban et al.'s findings [8]. They explained the mitigation mechanism of ASR for the mixtures having BOFS and siliceous reactive sand: Since alkalis in pore solution in the mixture and 1M NaOH solution and reactive silica in aggregate participate in the geopolymerization process (the engagement of alkalis in binder and pore solution and silica compound), fewer amounts of alkalis and silica are available in the geopolymer system. As a result, ASR expansion can be minimized. In our case, abundant amounts of alkalis and silica components that do not participate in the geopolymeriztion process, especially for the localized areas, are available in the mixture, consequently leading to ASR expansion. Nurruddin et al. [11] highlighted that even subtle variations in test/curing, such as those caused by partial immersion, can lead to differences in properties like expansion. It should be noted that WGS-75 and WGS-100% mixtures contain large amounts of reactive silica, as shown in Table 1. To verify this hypothesis, additional investigation is in progress.

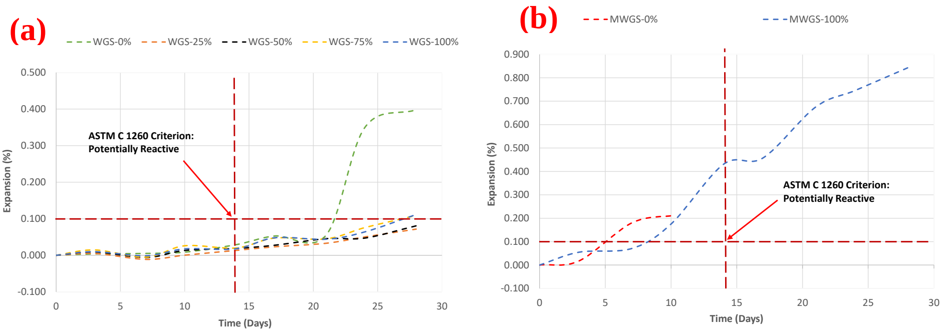

Fig. 4. Expansion behavior of specimens submersed in 1M NaOH solution: (a) geopolymer mixture and (b) normal mortar mixture.

As expected, both MWGS-0% and MWGS-100% mixtures submersed in 1M NaOH solution experienced huge expansion (Fig. 4 (b)). The MWG-0% mixture was broken after 10 days, while the MWGS-100% mixture had significant expansion until 0.85% at 28 days. This aligns with the study of Bocullo et al. [12], in which they suggest the OPC samples' reaction with the NaOH solution can lead to changes in the microstructure and increased expansion due to the formation of expansive compounds. Notably, normal OPC mixtures submerged in NaOH get exposed to additional ASR, which affects their microstructure. Micro and macro cracks in the mortar matrix are built up in the long term.

4 Conclusions

This research investigated the effect of the WGS and BOFS aggregate combination on compressive strength and expansion behavior for geopolymer and normal OPC mortar mixtures. The results are summarized as follows:

- Geopolymer mixtures showed better compressive strength results than normal OPC mortar mixtures, both in average strength gain (15.2 MPa for GPC and 4.5 MPa for OPC) and maximum strength (51.4 MPa for GPC and 34.9 MPa for OPC).
- The highest compressive strength was obtained in the WGS-25% mixture, which has low WGS (25%) and high BOFS (75%) contents.
- Expansion characteristics of geopolymer mixtures were promising, although several mixtures having the 0/100, 75/25, or 100/0 combination of WGS and BOFS exceeded the threshold value (0.1% expansion) under condition submersed1M NaOH solution at 28-day.
- The geopolymer mixture with the 50/50 combination of WGS and BOFS seems to be the optimum mixture in compressive strength and expansion behavior.

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