Synthesis of Environmentally Friendly Activated Alkali Concrete (Geopolymer) Based on Bentonite

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Abstract

Geopolymers can be mentioned as a new class of green cement adhesives. Geopolymers have recently been introduced as a greener alternative to conventional Portland cement (OPC) with the potential to minimize the negative environmental consequences of OPC such as carbon footprint and energy consumption. In this experimental study, the effects of different alkaline activating solutions on compressive, tensile, and indirect flexural strength, water absorption, and acid resistance of bentonite-based geopolymer concrete (GPC) were investigated. Also in this study, a new type of alkaline activator was used. In this study, GPC samples made from bentonite were produced and baked at 90 ° C. The results showed that the addition of NaOH to the mixture after 3 minutes of mixing KOH and Na₂SiO₃ with dry components (1/3 of the total mixing time) increases the compressive, tensile, and flexural strength and we also see that with this new method, water absorption capacity and the amount of weight loss of geopolymer concrete samples are reduced in acidic conditions.

Keywords: Eco-Friendly Cement, Geopolymer Concrete, Bentonite, Mechanical Strength, Alkaline Solution, Compressive Strength, Acid Resistance.

1. Introduction

The most widely employed cementitious material in the construction industry is ordinary Portland cement (OPC) [1]. It is the most important component of ordinary concrete. OPC manufacturing has significant environmental consequences, including significant energy and natural resource use and increasing carbon dioxide (CO_2) emissions [2], with around 0.73-0.99 tons of CO₂ emissions for every ton of OPC manufactured [3]. Climate change, on the other hand, is among the most serious environmental issues that have received a lot of attention in current history. The principal cause of global warming is greenhouse gas emissions, especially CO₂ [4]. The production of OPC was accountable for 7 to 10% of global CO₂ emissions [4]. Geopolymers have recently been introduced as environmentally friendly cementitious materials that can aid in mitigating the negative environmental effects of OPC. In 1978. Davidovits introduced geopolymers as a new class of inorganic polymer binders. Geopolymers are inorganic alumino-silicate compounds composed of silica (SiO₂) and alumina (Al₂O₃)-rich raw materials combined with an alkaline activator solution [5]. Geopolymer Concrete (GPC) offers superior chemical and mechanical qualities to Portland Concrete (PC), notably exceptional mechanical strengths [6, 7], exceptional corrosion resistance [8], lower permeability, improved resistance to solvents, significantly acids [9], and decreased creep effects [10].

*Corresponding author Email address: alireza.esparham@ut.ac.ir Natural resources such as bentonite, synthetic materials such as metakaolin, or waste industrial materials such as fly ash and Granulated Ground Blast Furnace Slag (GGBFS) may be used in the synthesis of geopolymers, depending on desired characteristics, cost, and resource availability [11]. The use of industrial by-products in GPC minimizes not only pollution but also the cost of maintaining these materials[12].

Another pillar of the geopolymerization process that plays an important role in the formation of crystalline structures of Si and Al is the alkaline activator solution, which is usually a combination of sodium hydroxide (NaOH) or potassium hydroxide (KOH) and sodium silicate (Na₂SiO₃) or potassium silicate (K₂SiO₃)[8, 13, 14].

However, the most commonly used alkaline solution is a combination of NaOH and Na₂SiO₃ [15]. Xu et al [16] tested NaOH and KOH without using a silicate solution and observed that KOH led to a better compressive strength. The majority of these investigations focused on the role of the alkaline activator solution in the polymerization reaction, and they observed that adding a silicate solution to the NaOH and/or KOH solution boosted the polymerization process. Palomo et al [17] reported that combining NaOH and Na2SiO3 solutions increased compressive strength over KOH and K₂SiO₃. Sharma et al [18] and Parveen et al [19] demonstrated that elevating the NaOH concentration up to 16 M increased compressive strength, but at 18 M there was no significant difference. Patel et al. [20] reported that raising the concentration of sodium hydroxide up to 12 M increased compressive strength but decreased compressive strength beyond that point. Petrus et al

[21] investigated the effect of the Na₂SiO₃/NaOH weight ratio on the compressive strength of GPC in another investigation. They came to the conclusion that raising the weight ratio of Na₂SiO₃/NaOH from 1 to 1.5 will enhance the compressive strength of GPC. The weight ratio of 1.5 had the best compressive strength while increasing the ratio from 1.5 to 2.5 resulted in a considerable loss in compressive strength. Sanni et al. [22] been shown raising the weight ratio of Na₂SiO₃/NaOH to 2.5 resulted in an improvement in compressive strength for fly-ash-based GPC.

Tensile and flexural strengths of various GPC forms, as well as associated features, have received less attention than compressive strength. Rajiwala et al. [23] discovered that a KOH active alkali solution yielded higher tensile and flexural strengths than NaOH at various ages in fly ashbased GPC.

According to Wang et al [24], raising the NaOH content from 4 to 12 M enhanced the flexural strength of Metakaolin-based GPC. According to Mishra et al [25], raising the NaOH concentration enhances the tensile strength of fly ash-based GPC. Morsy et al [26] discovered that raising the Na₂SiO₃/NaOH weight ratio from 0.5 to 1 and 1 to 2.5 increased and lowered flexural strength in fly ash-based GPC, respectively. In contrast, Sanni et discovered al [22] that increasing the Na₂SiO₃/NaOH weight ratio to 2.5 enhanced tensile and flexural strength. Because the effects of alkaline activating solutions on compressive, tensile, and flexural strengths, as well as water absorption and acid resistance of bentonite-based GPC, have not been reported consistently, the purpose of this study was to investigate the effects of alkaline activating solutions on mechanical properties and acid resistance. Chemical properties of bentonite-based GPCs (such as compressive strength, tensile strength, flexural strength, water absorption, and acid resistance) were also investigated provide insight to into the

development of practical applications. The previous study [13] studied the impacts of using KOH and NaOH solutions concurrently, and the results showed that using KOH and NaOH solutions concurrently lowered GPC compressive strength after 3, 7, and 28 days due to interference in Na+ and K+ chemical reactivity. This article focused on a new way to solve this problem.

2. Materials and Methods

The alumino-silicate used in this investigation was bentonite, which came from the Qara Naz mine in Iran's Zanjan district. Table. 1. given the chemical analysis of bentonite employed in this research using X-ray fluorescence (XRF) and in Fig 1, it also displays X-ray diffraction (XRD) spectroscopy ,(Includes the main phases of cristobalite, montmorillonite, calcite, and sub-phases of muscovite and albite). The alkaline activator solution was made with 98 percent pure NaOH, 90 percent pure KOH, and liquid Na2SiO3 with a SiO₂/Na₂O molar ratio of 2. The chemical analysis of the Na₂SiO₃, NaOH, and KOH compounds can be seen in Table. 2. Aggregates with granular sizes ranging from 7 to 10 mm were utilized as coarse aggregate (sand), and aggregates with granular sizes less than 4 mm were used as fine aggregate. ASTM C33 [27] was used to sift fine and coarse aggregates. As indicated in Table. 3., the coarse and fine aggregates were examined for SSD specific gravity and water absorption using the ASTM C127 [28] and ASTM C128 [29] methods. The fine aggregates' fineness modulus (as measured by ASTM C136 [30]) and sand equivalent (as determined by ASTM D2419 [31]) were 3.01 and 73, respectively. To reduce water content and improve concrete workability, a polycarboxylate-based Super Plasticizer (SP) was utilized.

Table. 1. The XRF analysis of bentonite used.

Chemical substance	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	Na ₂ O	K ₂ O	MgO	TiO ₂	LOI
Weight %	68.930	11.140	2.680	1.080	0.410	0.590	2.540	0.120	15.060

Table. 2. Chemical analysis of NaOH, KOH, and Na ₂ SiO ₃	solutions(wt %).

	NaOH			KOH Na ₂ SiO ₃				
Chemical substance	Result	Unit	Chemical substance	Result	Unit	Chemical substance	Result	Unit
NaOH	98	%	КОН	90.7	%	SiO ₂	30	%
Na ₂ CO ₃	1	%	K ₂ CO ₃	0.3	%	Na ₂ O	14.5	%
NaCl	200	ppm	KC1	0.006	%	Water	55.5	%
Fe	6	ppm	Fe	2	ppm			
SiO ₂	15.7	ppm	NaOH	1.2	%			



Fig. 1. X-ray diffraction pattern of the studied bentonite sample and identified phases: Cr; Cristobalite, M; Montmorillonite, C; calcite, Mo; muscovite, A; albite.

Table. 5. Specific gravity and water absorption of aggregates	avity and water absorption of aggregates.
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Material	SSD Specific gravity (gr/cm3)	Water absorption (%)
Coarse Aggregates	2.62	1.3
Fine Aggregates	2.59	3.2

Table. 4. Composition of the alkaline solutions.

Alkaline solution ID	NaOH 12M (%)	KOH 12M (%)	Addition Time delay (min)
Ν	100	0	-
K	0	100	-
T-K50N50	50	50	0
3-K50N50	50	50	3
6-K50N50	50	50	6

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Mix design ID	Bentonite	NaOH 12M	KOH 12M	Na ₂ SiO ₃	Coarse aggregate	Fine aggregate	Extra water	SP
Ň	400	144	0	216	850	850	10	8
K	400	0	144	216	850	850	10	8
T-K50N50	400	72	72	216	850	850	10	8
3-K50N50	400	72	72	216	850	850	10	8
6-K50N50	400	72	72	216	850	850	10	8

Table. 5. Mix design of specimens (kg/m3).

2.2. Experimental Program 2.2.1. Mix Designs

As given in Table. 4., the initial step of the inquiry involved the creation of 5 alkaline solutions to investigate the influence of various alkali solutions on the compressive, tensile, and flexural strengths of GPC. The NaOH and KOH solutions were all 12 M in concentration, and the weight ratios of Na₂SiO₃/KOH, Na₂SiO₃/NaOH, and Na₂SiO₃/KOH+NaOH were all adjusted to 1.5. Furthermore, when the first series of specimens were created, the weight ratios of the alkaline solution/bentonite and fine/coarse aggregate ratios were 0.9 and 1, respectively. Table. 5. depicts the mixed sample design for the first portion of the investigation.

To commence, 12 M NaOH and KOH solutions were made. Following that, the mix designs N, K, and T-K50N50 were prepared by adding these solutions to the Na₂SiO₃ solution 24 hours before the trials. The "T" prefix denotes the inclusion of KOH and NaOH solutions to the mixing procedure at the same time. Dry components, including bentonite and coarse and fine aggregates, were combined for 3 minutes to produce the GPC specimens. The dry mix was then mixed for 10 minutes with an alkaline activator solution containing NaOH (mix design N) or KOH (mix design K) or NaOH+KOH (mix design T-K50N50), Na₂SiO₃, and SP. The KOH and Na₂SiO₃ solutions with SP and dry components were mixed for the 3-K50N50 and 6-K50N50 combinations. Thereafter, after 3 and 6 minutes of mixing, NaOH solution was added to the mixes to test the effect of

time delay in adding the NaOH solution on the compressive strength of GPC. The prefixes "3" and "6" in the mix design IDs denote 3- and 6-minute latencies in the NaOH addition to the mixing process, accordingly. The overall mixing time for the 3-K50N50 and 6-K50N50 mixes was 10 minutes, similarly to the other mixtures.



Fig. 2. Samples a. made of geopolymer concrete based on bentonite.

2.2.2. Mix Testing

The GPC specimens were molded during the specimen preparation procedure after the material mixing was completed. Each GPC mixture was batched to yield three cube specimens (100x100x100mm) for compressive testing (Fig. 2). According to prior research [13], the produced GPCs were dry-cured at 90 C for 24 hours before being allowed to sit at room temperature. Following that, compressive testing on 7- and 28day specimens were performed in accordance with BS1881: Part116 [31]. Tensile strength tests were cylindrical performed on 3 specimens (300x150mm) for each mix design and performed at 7- and 28-day intervals using the ASTM C496 indirect tensile strength testing method [33]. In terms of flexural tests, three beam specimens (500x100x100mm) were used for each mix design and evaluated using the ASTM C293 [34] 3-point bending test methodology. The water absorption capacity of GPC specimens was investigated using ASTM C642 [35] techniques.

For this objective, three compressive cube specimens for N, T-K50N50, 3-K50N50, and K mix designs were evaluated. The 28-day specimens had first been placed in a 105 C oven to achieve a consistent dry weight before being weighted for water absorption capability (m0). After that, the specimens were placed in a water tank for three

days. They were then removed and weighed again after drying the surface water (m). Eq. (1). is being used to compute the 3-day water absorption capacity (W):

$$W = (m-mo)/mo \times 100$$
 (Eq. 1.)

In addition, to test the chemical resistance of GPC specimens, 28-day specimens of N, T-K50N50, 3-K50N50, and K mix designs were immersed in a solution of water and sulfuric acid with a pH of one for 28 days. The specimens were then subjected to a weight-loss test.

3. Results and discussion

The compressive, tensile, and flexural strengths of specimens at 7 and 28 days are reported in Table 6 (together with the appropriate coefficients of variation) and plotted in Figs 3-7,(As opposed to Fig. 3. to Fig. 5., Fig. 6. and Fig. 7. demonstrate the consequences of adding potassium hydroxide solution in different method (based in time delay). As can be seen, the mix 3-K50N50 with the NaOH solution added after 3 minutes of adding KOH and Na₂SiO₃ to the dry components had the highest initial (7-day) and lateral (28-day) compressive strengths of 23.3 and 25.6 MPa, respectively. The mix T-K50N50 had the lowest 7- and 28-day compressive strengths, measuring 14.3 and 15.7 MPa, respectively. According to the results, the strength gain of the mix N after 7 days of curing was more than that of the other mixes (93 percent). After 7 days of curing, the mix K had 79 percent of its 28-day compressive strength. The rate of strength gain for the mix K from 7 to 28 days, on the other hand, was the most significant of all, at roughly 26 percent. Tensile strength and flexural strength test results follow a pattern similar to compressive strengths. The simultaneous and equal integration of KOH and NaOH solutions (mix T-K50N50) produced the lowest tensile strength values, which were roughly 10% and 12% lower than the N and K single solution mix designs, correspondingly. Mix 3-K50N50, on the other hand, demonstrated the highest tensile strength (about 9, 6, and 21% higher than the N, K, and T-K50N50 mix designs), showing that a 3-minute time delay in KOH addition to the mix had a considerable favorable effect. The flexural strengths follow the same basic tendencies as the aforementioned broad trends.

Mix ID	Compressive strength (MPa)		Tensile str	rength (MPa)	Flexural strength (MPa)		
	7-days	28-days	7-days	28-days	7-days	28-days	
Ν	19.4±0.5	20.9±0.2	3.55±0.1	3.7±0.3	4.46±02	4.69±0.2	
T-K50N50	14.3±0.3	15.7±0.3	3.25±0.1	3.34±0.4	3.84±0.3	4.04±0.4	
3-K50N50	23.3±0.4	25.6±0.1	3.83±0.3	4.04±0.2	4.88±0.1	5.17±0.2	
6-K50N50	20.9±0.1	24.3±0.4	3.69±0.2	3.97±0.2	4.67±0.2	5.02±0.3	
K	18.1±0.2	22.8±0.4	3.48±0.3	3.8±0.3	4.18±0.3	4.81±0.1	

Table. 6. Compressive, tensile, and flexural strength values, as well as their coefficients of variation.









Fig. 6. The effect of NaOH solution adding duration on the compressive strengths of T-K50N50, 3-K50N50, and 6-K50N50 mixtures.



Fig. 7. The effect of NaOH solution adding duration on the flexural and tensile strengths of T-K50N50, 3-K50N50, and 6-K50N50 mixtures.



Fig. 8. Water absorption and weight loss under acidic conditions of N, T-K50N50, 3-K50N50, and K mixes.

To characterize and comprehend trends, the performance mechanism of alkaline solutions should indeed be investigated. KOH creates more geopolymers than NaOH, resulting in a stronger and more compact microstructure with low 7-day compressive, tensile, and flexural strengths, slow hardening, and high 28-day compressive, tensile, and flexural strengths. However, at the same concentration, NaOH may dissolve more inorganic components than KOH, resulting in a faster reaction rate for Na+ than for K+. Since Na+ has a faster reaction rate, the use of NaOH results in higher initial compressive strength and faster hardening. On the other hand, the combination of NaOH and KOH greatly reduces the compressive, tensile, and bending strength of GPC. This could be because NaOH and KOH behave differently in geopolymerization. The tendency of K+ to condense cannot compensate for the high reactivity of Na+. On the other hand, due to the higher activation energy of potassium compared to sodium, it enters the polymerization reaction later, in addition, adding potassium hydroxide solution to sodium hydroxide solution (taking into account the time delay or simultaneously), increases the workability of mortar (or concrete). However, as the mixtures of 3-K50N50 and 6- K50N50 were shown, the addition of NaOH at 3 and 6 min intervals gave both K+ and Na+ enough time to crosslink in different ways, resulting in more geopolymer gel and a denser geopolymer cement matrix. [14, 36].

Fig. 8. illustrates the outcomes of water absorption and weight loss trials on GPC specimens under acidic conditions. The N, T-K50N50, 3-K50N50, and K mix designs, respectively, had water absorption capacities of 6.8, 7.2, 5.8, and 6.4 percent. The N, T-K50N50, and K mix designs, respectively, lost 4.2, 4.7, 3.8, and 4% of their weight in acidic circumstances. Because of the higher density of the geopolymeric matrix structure in this mix design, the 3-K50N50 mix design had a lower water absorption capacity and weight loss under acidic circumstances than the other specimens.

4. Conclusion

In this investigation, a new type of alkaline activator for GPC based on bentonite was thoroughly studied. Based on the findings of the current experimental experiments, the following conclusions can be drawn:

1. After 28 days, utilizing the KOH alkaline solution in combination with Na_2SiO_3 produced higher compressive, tensile, and flexural strengths than using NaOH alone.

2. Simultaneous application of NaOH and KOH with Na_2SiO_3 as the alkaline solution lowered the compressive, tensile, and flexural strengths of bentonite-based GPC due to interference in the Na+ and K+ reactivity. However, over a period of time, adding the NaOH solution to the new mix resulted in higher compressive strengths.

3. Adding NaOH to the mix after combining KOH and Na_2SiO_3 with dry components for 3 minutes (1/3 of total mixing time) produced the maximum compressive, tensile, and flexural strengths, among other things, in this study.

4. Under acidic circumstances, the addition of NaOH to the mix after 3 minutes of mixing KOH and Na_2SiO_3 with dry components (1/3 of the total mixing time) resulted in the lowest water absorption and weight loss.

5. It is proposed that the novel alkali solution composition technique reported in this study be applied to improve the mechanical and chemical properties of geopolymer concretes manufactured from various alumina silicate sources.

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