

The Effect of NaOH Molarity and Alkali Activator Ratio on The Properties of GGBFS-Based Geopolymer as a fully Cement Replacement

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تأثير تركيز هيدروكسيد الصوديوم ونسبة المنشط القلوي على خصائص الجيوبوليمر المحتوي على خبث الأفران العالية المحبب كبديل تام للإسمنت

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Received: March 13, 2026

Accepted: April 28, 2026

Published: May 12, 2026

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Abstract:

The cement sector is one of the largest CO₂ emitters worldwide, which creates the need for sustainable alternatives. This study explores the use of GGBFS as a 100% replacement for cement to prepare geopolymer binders. The influence of molarity of NaOH (5M, 10M, 15M), ratio of NaOH/Na₂SiO₃ (0.5–3) on setting time, soundness and compressive strength were studied. The results indicate that raising molarity and ratio considerably reduces setting times. Drop in initial set for 16 minutes at 15M. All blends demonstrated satisfied volume stability. The 10M mix gained 43.48 N/mm² at 28 days, 52% improvement compared to OPC. The study validates GGBFS geopolymers as a potential high performance sustainable alternative to cement.

Keywords: Geopolymer, GGBFS; Alkali-activator, Setting time, Compressive strength.

المخلص:

يُعدّ صناعة الإسمنت من أكبر مصادر انبعاثات ثاني أكسيد الكربون عالميًا، مما يُحتم الحاجة إلى بدائل مستدامة. تبحث هذه الدراسة إلى استخدام خبث الأفران العالية المحبب (GGBFS) كبديل تام للإسمنت في تحضير مواد رابطة من الجيوبوليمر. تم دراسة تأثيرات تركيز هيدروكسيد الصوديوم (5 مولار، 10 مولار، 15 مولار) ونسبة هيدروكسيد الصوديوم إلى سيليكات الصوديوم (0.5-3) على زمن التصلب والصلابة ومقاومة الضغط. تتبين النتائج إلى أن زيادة التركيز والنسبة تُقلل بشكل ملحوظ من زمن التصلب، حيث انخفض زمن الشك الابتدائي إلى 16 دقيقة عند تركيز 15 مولار. أظهرت جميع الخلطات استقرارًا حجميًا جيدًا أقل من 2 مم. اكتسبت الخلطة بتركيز 10 مولار مقاومة ضغط بلغت 43.48 نيوتن/مم² بعد 28 يومًا، مع تحسن في الأداء بنسبة 52% مقارنةً بالإسمنت البورتلاندي العادي. تُؤكد هذه الدراسة أن جيوبوليمرات GGBFS تُعدّ بديلًا مستدامًا عالي الأداء للإسمنت.

الكلمات المفتاحية: جيوبوليمر، خبث الأفران العالية المحبب، منشط قلوي، زمن الشك الابتدائي، مقاومة الضغط.

Introduction:

Portland cement (PC) is responsible for 7–8 % of global CO₂ emissions [1]. A sustainable alternative is offered by Geopolymers [2], a term coined by Davidovits, which are alkaline-activated aluminosilicate sources. Ground Granulated Blast Furnace Slag (GGBFS), a by-product of iron industry rich in SiO₂

and Al_2O_3 , can be used for geopolymerization [3]. The key to the performance of geopolymers based on GGBFS is the activator chemistry. Si/Al dissolving rate is dependent on the NaOH molarity [4] and gel structure and strength are influenced by the NaOH/ Na_2SiO_3 ratio [5]. However, the effect of wide-range molarities (5M-15M) and ratios (0.5-3) on fresh and hardened characteristics is lack studied. The objectives of this study are to examine the impacts on setting time and soundness, to evaluate the growth of compressive strength up to 28 days, and to determine optimum mixes that conform to the Libyan criteria.

Material and methods:

Materials:

- **GGBFS:** Libyan Iron and Steel Plant (Misurata) – ground for 5 hours.
- **Alkali activator:** NaOH flakes (98% purity) and Na_2SiO_3 solution ($SiO_2/Na_2O \approx 2.0$). 5M, 10M and 15M solutions prepared 24 hours in advance.
- **Standard Sand:** Fine aggregate substitute was replaced with crushed GGBFS of similar grading to standard sand.
- **Reference:** Ordinary Portland Cement (OPC) for comparison.

Concrete composition:

For paste specimens, eleven geopolymer pastes were produced with activator/GGBFS=0.3. Three molarity series (5M, 10M, 15M) with different NaOH/ Na_2SiO_3 ratios (0.5, 1, 1.5, 2, 3). Compressive strength: mortar (GGBFS: sand=1:3) mixed with 10M-1 (ratio=1, activator/GGBFS=0.4) was employed. OPC control: water-cement ratio 0.4, cement-sand ratio 1:3.

Test Procedures:

- **Setting Time:** Vicat apparatus (ASTM C191).
- **Soundness:** Le Châtelier molds (ASTM C151), expansion after boiling.
- **Compressive Strength:** 50x50x50 mm cubes evaluated on 3, 14, 28 days.

Results and discussion:

Setting Time:

A comparative analysis across molarities reveals that Figures 1-a, 1-b, 1-C clearly demonstrates a systematic reduction in setting time with increasing both NaOH molarity and NaOH/ Na_2SiO_3 ratio, and three distinct behaviors emerge. **First**, low molarity (5M) shows a gradual as illustrates in Figure (1-a), controlled reduction from 212 min (ratio 0.5) to 110 min (ratio 1), with a slight anomaly at ratio 1.5 (133 min) before dropping to 28 minutes at ratio 3; this anomaly suggests that at lower molarities, the high silicate content from Na_2SiO_3 temporarily stabilizes the system, slowing reaction mechanism [6].

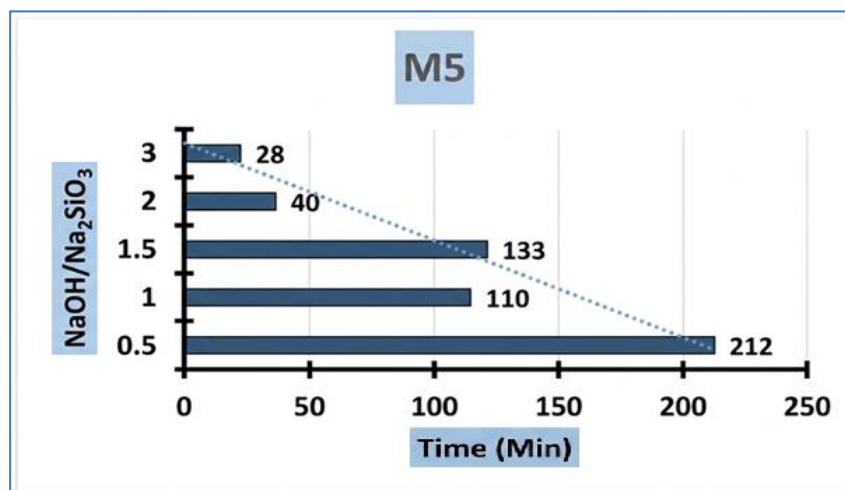


Figure (1-a): Comparative study of initial setting time for 5% of molarity

Second, medium molarity (10M) as shown in Figure (1-b) exhibits a dramatic drop between ratio 0.5 (182 min) and ratio 1 (43 min). A 76% reduction and beyond ratio 1, further increases to 2 results in minimal additional reduction (36 min), indicating that a saturation point is reached where additional NaOH no longer accelerates setting [7].

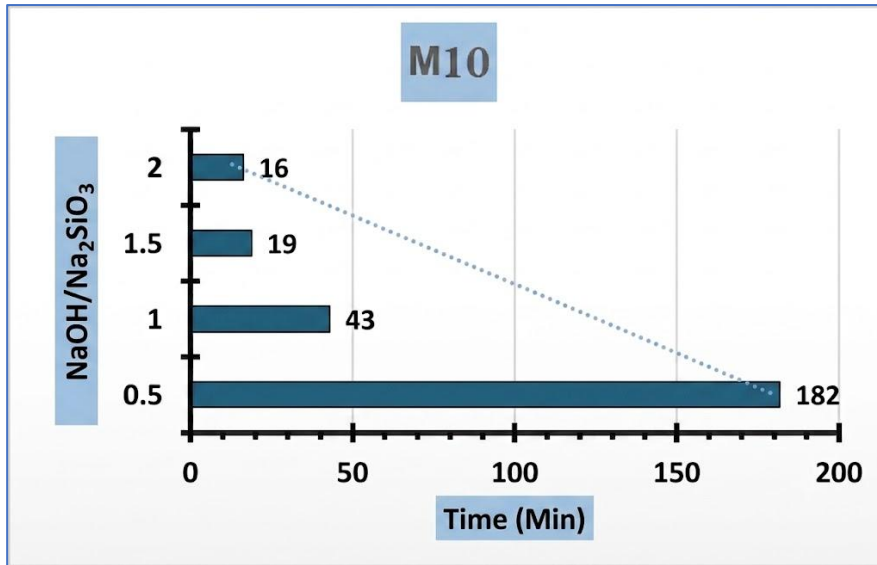


Figure (1-b): Comparative study of initial setting time for 10% of molarity

Third, high molarity (15M) in Figure (1-C) shows the most extreme behavior, with ratio 0.5 giving 213 minutes (similar to 5M-0.5) except ratio 1 dropping to 16 minutes.

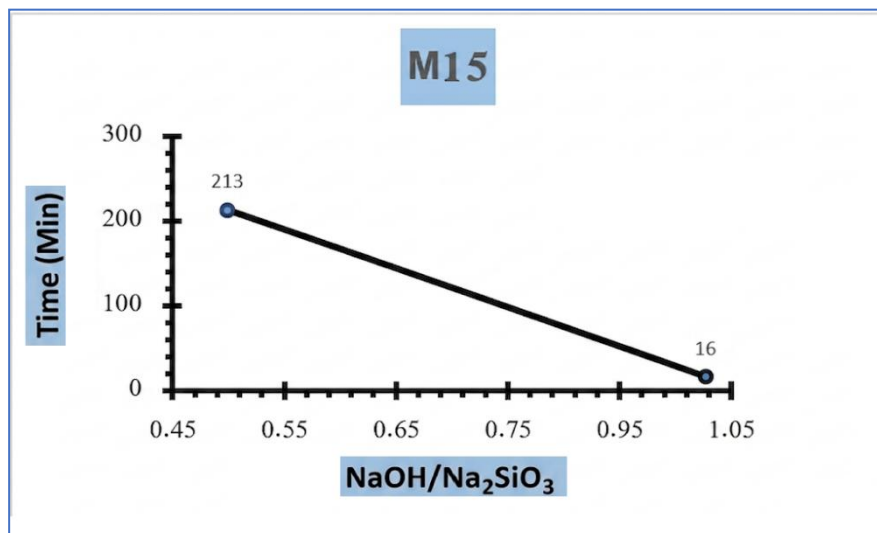


Figure (1-C): Comparative study of initial setting time for 5% of molarity

The fastest setting among all mixes; this paradoxical behavior at ratio 0.5 (high Na₂SiO₃) suggests that at very high molarities, the viscous silicate solution impedes initial dissolution before rapid acceleration. Turning to mechanistic interpretation, the acceleration mechanism involves OH⁻ ions attacking Si-O-Si and Si-O-Al bonds on GGBFS surfaces, releasing silicate and aluminate species [8]; higher molarity provides more OH⁻ ions, accelerating bond breakage. However, at ratio 0.5, excess Na₂SiO₃ creates a viscous medium that limits OH⁻ mobility, explaining the delayed setting, whereas at ratio ≥1, sufficient OH⁻ is available to overcome this limitation. Regarding standard compliance, Libyan Standard 340/2009 requires initial setting time ≥45 minutes. Therefore, the only mixes with initial set >45 min is suitable for conventional construction. The acceptable mixes include all 5M mixes except ratio 3, 10M-0.5 (182 min), and 15M-0.5 (213 min); mixes like 10M-1 (43 min) and 15M-1 (16 min) set rapidly for most applications but could benefit specialized uses requiring rapid setting as shown in Table 1.

Table (1): Initial and Final Setting Times

SN.	Mix	M	NaOH/Na ₂ SiO ₃	Initial (min)	Final (min)
1	OPC	-	-	173	273
2	GPM-5-0.5	5	0.5	212	252
3	GPM-5-1	5	1	110	170
4	GPM-5-1.5	5	1.5	133	175
5	GPM-5-2	5	2	94	118
6	GPM-5-3	5	3	28	133
7	GPM-10-0.5	10	0.5	182	242
8	GPM-10-1	10	1	43	57
9	GPM-10-1.5	10	1.5	40	54
10	GPM-10-2	10	2	36	48
11	GPM-15-0.5	15	0.5	213	262
12	GPM-15-1	15	1	16	32

Soundness:

The Libyan standard specification 340/2009 requires that the measured expansion in the distance between the tips of the two needles of the Luchatelli mold should not be exceeding 10 mm. The findings of the volume stability test revealed satisfied and consistent performance across the categories and at NaOH/Na₂SiO₃ ratios indicating stability of the volume of the geopolymer across diverse mixes. The results are presented in Table 2 and this stability supports the notion that the geopolymer utilized can be a sustainable substitute of Portland cement as it satisfies the required volume stability criteria for engineering and building applications.

Regular Portland cement also exhibited high stability which are likewise within the specification limits. The results of the volume stability test for the geopolymer mixtures are all within the limitations defined by Libyan standard (340/2009) related to standard specifications as indicated in Table 2. This shows that the mixtures developed are of quality and suitability that meet the needed standards.

Table (2): Soundness Test Results of Geopolymer and OPC

SN.	Samples	Groups	Molarity (M)	NaOH/Na ₂ SiO ₃ Ratio	Volume Stability (mm)		
1	1	5	0.5	0.3	1	5	-
2			1		2	2	-
3			1.5		2	1	0
4			2		1	2	3
5			3		1	3	1
6	2	10	0.5		2	3	-
7			1		4	0	2
8			1.5		0	1	1
9			2		1	0	1
10	3	15	0.5		1	2	-
11			1		1	1	0

Geopolymers mixtures showed satisfied volume stability with expansions of 0.5 to 2 mm, which is far below the 10 mm limit of the Libyan standard. Importantly, the geopolymer expansion was 67-92% lower than OPC (6 mm). The improved performance is explained by variations in reaction chemistry:

In OPC delayed expansion is caused by hydration of free lime (CaO) and periclase (MgO) after hardening [9]. However, geopolymerization leads to a three-dimensional aluminosilicate network without free expanding oxides. The early-age reaction consumes all the calcium and magnesium available inside the gel structure, removing sources of delayed expansion.

This has important practical implications: Geopolymers based on GGBFS are more dimensionally stable than traditional cement, thus lowering chances of cracking due to volume fluctuations in constrained constructions.

Compressive Strength: Compressive strength development mechanism: Different strength development procedures exist between geopolymer and OPC as shown in Figure 2. The geopolymer exhibits an early age (3-14 days): Average increase 70% to 17.99 N/mm² Later age (14-28 days): 110% average increase, 37.76 N/mm².

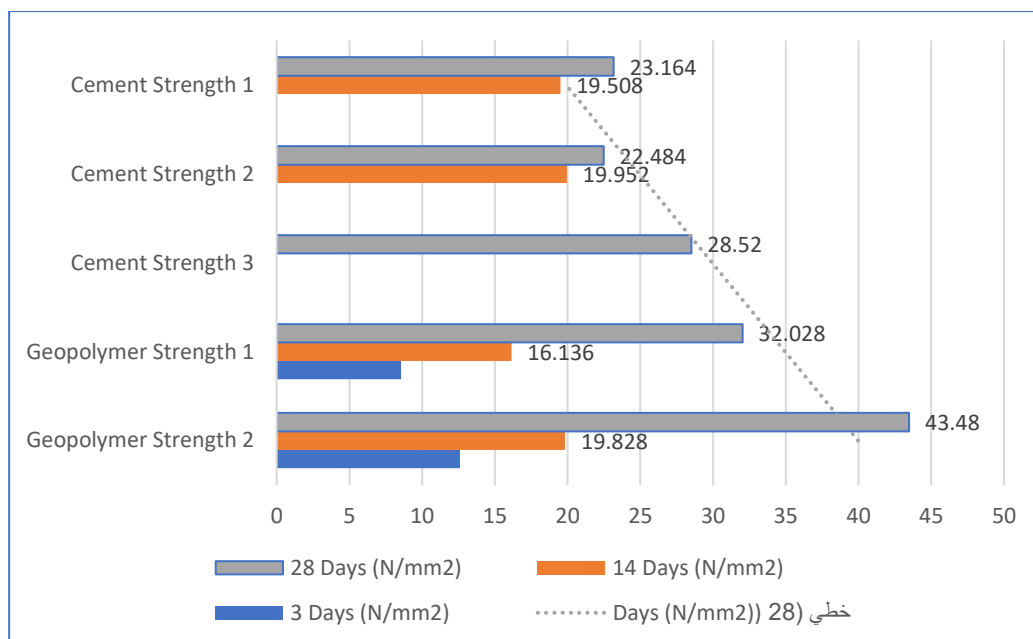


Figure (2): Comparison of compressive strength (Geopolymer & OPC)

This is in direct contrast to OPC where we exhibit 25% growth throughout 14-28 days. The continuing and rapid development in geopolymer shows extended repolymerization, i.e. dissolution of GGBFS particles and production of aluminosilicate gel continues beyond 14 days [10]. However, in OPC, the creation of C-S-H is practically finished in 28 days. The highest strength obtained (43.48 N/mm² at 28 days for geopolymer) is an increase of 52.7% over OPC's top performance (28.52 N/mm²). The performance is caused by the dense, 3D, covalently bound geopolymer gel structure with stronger inter-particle bonding than C-S-H gel [2].

Strength-Setting indicates an inverse relationship between setting time and ultimate strength. This suggests that quicker setting mixtures tend to reach higher compressive strengths. However, relationships are not linear. The optimum balance is at 10M-1: initial set time of 43 min (a little below the 45-min guideline) providing 43.48 N/mm². This blend has practical workability and optimum strength.

The substantial variability in the 28-day results for the geopolymer (32.03 - 43.48 N/mm², 26% variance) compared to OPC (22.48–28.52 N/mm², 21% variation) reflects a significant practical issue.

This variation is presumably the result of:

- Fast setting with fast compaction before increasing in viscosity
- Possible non-uniform distribution of activator in the mix
- Sensitivity of geopolymer to mixing process
- This implies that the quality control measures for geopolymer should be more rigorous than for regular concrete.

Conclusions:

It can be concluded that the increase in alkaline activators molarity and ratio results in a reduction in the setting time, with the most significant modifications at some intermediate concentrations. Nevertheless, only few low ratio mixes match the needed building criteria. All geopolymer combinations exhibit improved dimensional stability than that of regular Portland cement. In addition, the compressive strength of geopolymer is much higher than standard Portland cement and the strength will improve with time due to long-term geopolymerization. For a certain intermediate mixture composition, the optimal compromise between practical workability and compressive strength is attained.

Recommendations:

This can inform future efforts by focusing on the following critical areas:

1. Standardized tests for aggregates. Reproducibility of compressive strength test utilizing standard sand (ISO 679) for international comparison.
2. Develop retarders: Develop chemical admixtures for geopolymer systems to delay setting time while maintaining strength.
3. Microstructural investigation: The 10M-1 mix will be analyzed using SEM-EDS and XRD to relate gel chemistry to the mechanical properties.
4. Durability assessment: Test for the resistance to sulfate attack, acid attack and freeze-thaw cycle for confirmation of long-term performance.

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