



# A Critical Appraisal of Soil Stabilization Using Geopolymers: The Past, Present and Future

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## Abstract

Soft and highly compressible soils beneath civil engineering structures are often responsible for severe problems such as excessive settlement, cracking, sinking, etc. Over the years, these soft soils have been stabilized with Ordinary Portland Cement (OPC). Unfortunately, a known demerit of OPC as a traditional stabilizer is its energy-intensive nature and emission of large quantities of CO<sub>2</sub>, which has immediate- and long-term detrimental effects to the environment. This shortcoming has led to the emergence of geopolymers as a viable alternative to OPC with desirable properties such as high strength, stiffness, reduced shrinkage, and low energy consumption. This paper presents a critical appraisal (including a statistical approach) of geopolymer-stabilized soils, from relevant published literature sources covering various soil types and industrial by-products as activators for geopolymers. A review of 50 selected references on geopolymer-stabilized soil revealed that a maximum Unconfined Compressive Strength (UCS) of 82.5 MPa at an elevated temperature of 150 °C and a minimum UCS of 0.38 MPa at room temperature (23 °C) were obtained. The molarity of the alkali precursors (NaOH, KOH, Na<sub>2</sub>SiO<sub>3</sub>, Ca(OH)<sub>2</sub>) used by different researchers ranged from 1.7 to 23.0 M, while the stabilized soil types ranged predominantly from silty to lateritic clays. From the statistical analysis performed on 50 selected references, a statistically significant difference between molarity, curing temperature, and UCS was observed; while, there was a statistical correlation of 0.539 obtained between UCS and curing temperature. Furthermore, a regression equation was developed to predict the UCS of geopolymer-stabilized soil. However, a major limitation affecting the wide utilization of geopolymers in soil stabilization is the absence of design templates compared to the well-established OPC or lime stabilization parameters. Future research should focus more on long-term durability of geopolymer-stabilized soils, especially wet–dry, freeze–thaw durability, and sustainability assessment.

**Keywords** Geopolymers · UCS · Molarity · Curing temperature · Long-term durability

## Introduction

Most civil engineering infrastructures are supported by different soil types which vary mainly due to particle size and mineralogy. With an underlying soil, classified as soft to highly compressible clays, the super-structure has a high tendency to experience significant damages (such as cracking, sinking, collapse) due to weak microstructural bonds inherent in the clay particles. Since excavating and hauling a large

depth of these problematic soils before construction may not be economically viable, improving the engineering properties (such as shear strength, compressibility, and stiffness or ductility) of these soft soils through the process of soil stabilization has proven to be a step in the right direction, and has been widely practiced and documented in various literature sources. The two major techniques of soil stabilization are mechanical and chemical stabilization, which are sometimes used in hybrid form based on the construction complexity. Mechanical stabilization involves reduction of the air voids in the soil while chemical stabilization requires the introduction of additives such as lime and OPC blended with industrial by-products into the weak soil to achieve increased strength, stiffness, reduced permeability, etc.

Historically, the act of chemical stabilization was first recorded when the Mesopotamians and Romans improved the load-carrying capacities of pathways by mixing the soil

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beneath with pulverized limestone [1]. As industrialization increased, lime and Ordinary Portland Cement (OPC) gained more adoption for usage in construction works, particularly for road subgrades. However, a major problem with these conventional soil stabilizers (i.e., OPC and lime) is that their production processes are energy intensive and they emit a large amount of carbon dioxide [2]. This was buttressed by [3, 4] who noted that the global cement industry contributes around 1.35 billion tons of the greenhouse gas emissions annually, or about 7% of the total man-made greenhouse gas emissions to the earth's atmosphere. Furthermore, according to Khedari et al. [5], approximately one ton of CO<sub>2</sub> is emitted into the atmosphere to produce one ton of cement. Zhang et al. [2] noted that the readily available raw materials to produce cement are being over-consumed.

In cement-stabilized soils, the stabilization mechanisms are associated with hydration and pozzolanic reactions [6, 7]. When lime is mixed with clayey soils, the clay particles become closer and the soil is stabilized through flocculation and pozzolanic reactions [7, 8]. More importantly, one of the most promising alternatives to the conventional stabilization methods with OPC and lime [6, 7, 9] is the use of geopolymers (alkali-activated binders blended with industrial wastes such as fly ash, blast furnace slag, etc.). Past literature sources on geopolymers over the last decade have focused on creating eco-friendly and sustainable concrete, with very little attention given to soil stabilization.

This paper presents an evaluation of geopolymer-stabilized soils within a large framework of the past, present, and future to have a clear grasp of the overall effectiveness of this stabilization technique. The past and present give a synopsis of geopolymer-stabilization methods used since inception till date with different industrial by-products such as blast furnace slag, fly ash, etc. A review of different pozzolans (fly ash, blast furnace slag, etc.) composition used to form the geopolymer is also conducted as this has a significant effect on the compressive strength of the treated soil. Other properties such as shear strength, deformability are also discussed. The future gives a likely trend of the research direction, particularly transition from liquid to solid state regarding the usage of alkali-activators.

## Geopolymers

As the drive for sustainable infrastructure increases, geopolymers have become the center of attention. In the early 1970s, a French material scientist, Davidovits developed geopolymer as an inorganic aluminosilicate material, being projected as an alternative to cement. Formation of geopolymer occurs through polycondensation of tetrahedral silica (SiO<sub>4</sub>) and alumina (AlO<sub>4</sub>), which are linked alternately by sharing all the oxygen atoms [10, 11]. According to Zhang

et al. [11] the chemical structure of geopolymer can generally be expressed as:

$$M_n \left\{ -(\text{SiO}_2)_z - \text{AlO}_n - \right\}_n, \quad (1)$$

where M is an alkali cation such as potassium (K<sup>+</sup>) or sodium (Na<sup>+</sup>) that balances the negative charge for Al, n is the degree of polycondensation, and z is the Si/Al molar ratio, ranging from 1 to 15, and may go up to 300 [10, 11]. Depending on the value of z from Eq. (1), geopolymers can assume one of the several basic systems [12]; z > 3 produces a rubbery geopolymer of a linear linked two-dimensional network and z < 3 produces a brittle cementitious product of a cross-linked three-dimensional network suitable for soil stabilization [2, 13–17]. Three typical structures of geopolymer are highlighted in Table 1.

Geopolymers exhibit different physicochemical properties with varying Si/Al molar ratios [18]: Si/Al ratio < 3 result in three-dimensional and cross-linked networks with stiff and brittle properties, and hence can be used as cementitious and ceramic materials; and higher ratios of Si/Al > 3 result in two-dimensional and linearly linked networks with adhesive and rubbery properties [18]. Geo-polymerization requires Al and Si to be dissolved in the alkaline solution before the dissolved species is transported. As shown in Fig. 1, the geopolymerization process can be simplified into two major steps that interact with each other along the reaction: dissolution of amorphous aluminosilicate materials by alkali hydroxide solution and/or alkaline silicate solution to form reactive silica and alumina and polycondensation of the dissolved species into amorphous or semi-crystalline oligomers which further polymerizes and hardens into synthetic aluminosilicate materials [19]. Figure 2 shows a schematic diagram of a typical geopolymer mix design which involves three main components, soil, pozzolans and alkali precursor with the curing at either ambient or elevated temperature. The final product is a 'geological polymer', also known as geopolymer.

## The Past and Present: Soil Stabilization Using Geopolymers.

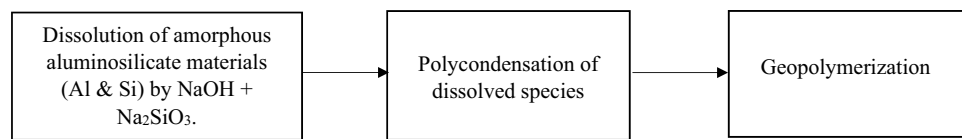
Table 2 shows a summary of past literature sources which assessed the performance of geopolymer-stabilized soils

**Table 1** Three basic typical structures of geopolymers

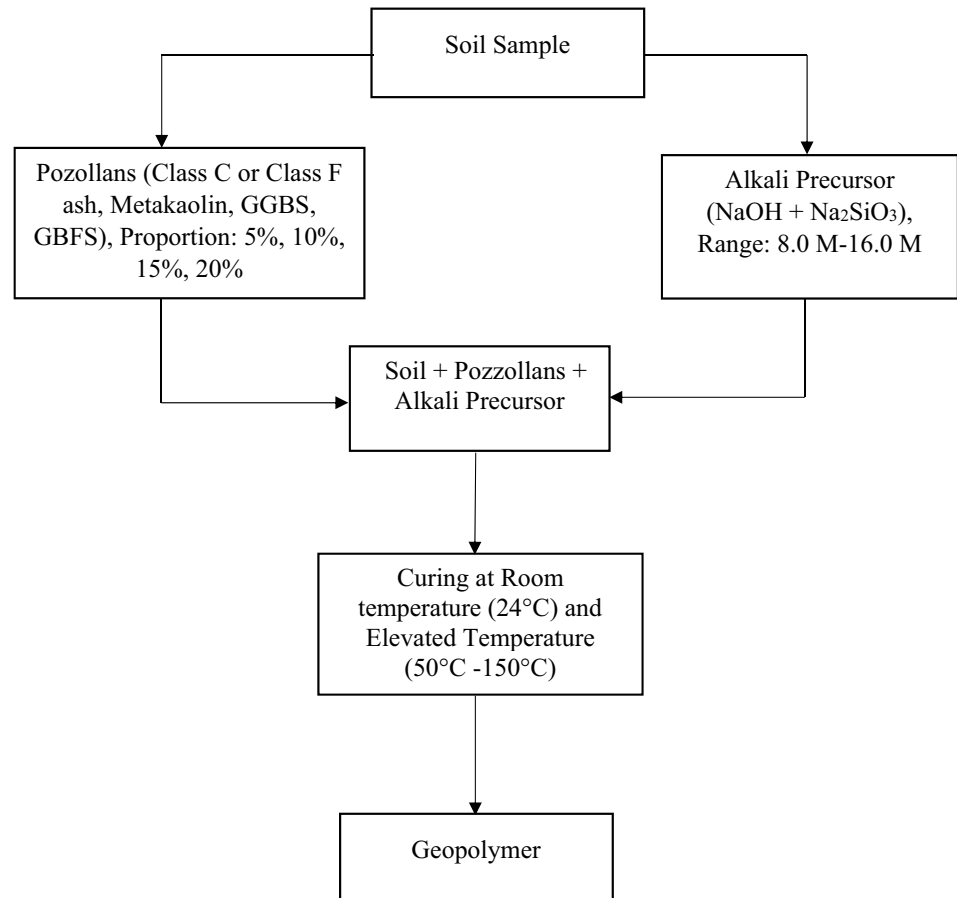
S/N	Geopolymer Structure
1	$M_n \{-\text{Si}-\text{O}-\text{Al}-\text{O}-\}_n$ ,
2	$M_n \{-\text{Si}-\text{O}-\text{Al}-\text{O}-\text{Si}-\text{O}-\}_n$ ,
3	$M_n \{-\text{Si}-\text{O}-\text{Al}-\text{O}-\text{Si}-\text{O}-\text{Si}-\text{Si}-\}_n$

NB: M alkali cation, n degree of polycondensation

**Fig. 1** Flow chart showing a simplified geopolymerization process



**Fig. 2** Typical geopolymer mix design



in terms of compressive strength, compressibility, shear strength, permeability, and shrinkage. For the purpose of this review, two main variables (molarity, curing temperature) will be analyzed in predicting the UCS parameter. Figure 3 shows a summary of molarity, curing temperature and UCS parameters from 50 selected literature sources, which gives graphical representation of the results achieved since the introduction of geopolymers as a sustainable alternative. The number labels on the horizontal axis show the reference of each extracted data. From the data gathered from published information in Fig. 3, it can be observed that the maximum values of UCS, molarity and curing temperature are 82.5 MPa, 23.0 M, and 150 °C, respectively.

Davidovits [12] noted that energy consumption and CO<sub>2</sub> emission could be largely reduced by replacing OPC with geopolymer. According to Sindhunata et al. [80], the common synthesis temperature of geopolymer ranges between 25 and 80 °C. Also, geopolymers can be synthesized from

a wide range of low-cost aluminosilicate materials or even industrial wastes, such as metakaolin, fly ash, furnace slag, red mud, and rice husk ash [11, 81–84]. Furthermore, geopolymers have excellent mechanical properties such as compressive strength, stiffness, effective resistance to heat, organic solvents, and acids.

Zhang et al. [2] investigated the stabilization of low plasticity clay with metakaolin-based geopolymer. The lean clay was prepared at Maximum Dry Density (MDD) of 1800 kg/m<sup>3</sup> and Optimum Moisture Content (OMC) of 15% and cured at an ambient temperature of 23 °C and relative humidity of 40–50%. UCS values of 20.27 MPa and 31.22 MPa were recorded at 7 and 28 days, respectively. Thus, from Fig. 4, it was observed that the geopolymer-stabilized soils achieved increased compressive strength as compared to the traditional OPC. Microstructural analysis also confirmed the formation of geopolymer gels in the soil pores.

**Table 2** Literature review of soil stabilization using geopolymers

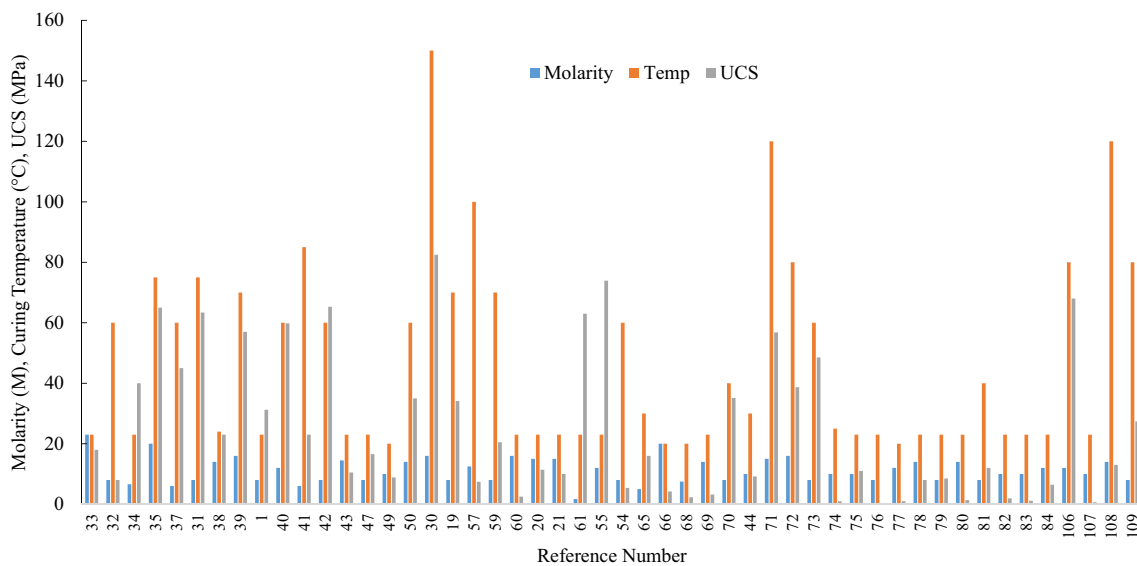
References	Soil type	Alkaline activator	Molarity(M)	Pozzolan	Alkaline/Ash ratio	UCS-7 days (MPa)	UCS-28 days (MPa)	Curing temp (°C)
[20]	Si–Al minerals	NaOH+ KOH	23	–	–	–	18.00	23
[21]	FA paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8.0	Kaolinite	–	–	8.00	60
[22]	GP paste	NaOH	6.6	FA	–	–	> 40.0	23
[23]	FA paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	10.0–20.0	High Calcium FA	–	10	65.0	75
[24]	FA paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	-	FAF	0.40	–	–	70 & 80
[25]	FA + GBFS paste	NaOH	6.0	GBFS + Class F FA	0.35	–	45.00	27, 60
[26]	GP paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8.0	FAC	–	–	63.40	23 & 75
[27]	GP paste	NaOH	4.5–14	Ground FA	–	–	23.00	24
[28]	GP paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	6.0–16.0	FA	–	–	57.00	70
[2]	Lean clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8.0	MKG	0.45	20.27	31.22	Ambient (23)
[29]	FA paste	NaOH	12.0	FA	–	–	59.81	Room Temp. & 60
[30]	GP mortar	NaOH	6.0	FAF	–	–	23.00	65 & 85
[31]	FA paste	NaOH+ KOH	8.0	FAF	–	–	65.28	25, 60
[32]	Clayey soil	NaOH+ KOH	12,14.5	Class C FA + GGBS	0.65	–	10.50	Ambient
[33]	Lateritic soil	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	10	FFA + CCR	–	–	9.20	27 & 30
[34]	RAP	NaOH	2% to 4%	FA	–	–	–	–
[35]	Colombian soil	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	5.0–12.0	FAF	–	–	–	–
[36]	Black cotton soil	KOH+ Ca(OH) <sub>2</sub>	100:0	FA	–	–	16.55 <sup>a</sup>	23
[37]	Silty clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	NR	FA + Slag	–	–	6.00	–
[38]	Lean clay + Sand	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	10.0	Low Calcium FA	0.50	3.41	8.86	Ambient (20)
[39]	GP paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	6.0–14.0	FA	–	–	35.00	29 & 60
[40]	Black cotton soil	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	5.0	FA	–	–	–	–
[41]	FA paste	NaOH	16.0	FA + Elazığ Slag	–	–	82.50	50,100, & 150
[42]	FA paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8.0	FA + Carbon Fibers	–	–	34.10	70
[43]	Pb soil	CaO + Na <sub>2</sub> SiO <sub>3</sub>	-	FA + GGBS	–	–	4.35	22
[44]	Plastic lean clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8.0	Class F FA	0.5	5.38	–	60
[45]	GP mortar	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	12.0	LSS	0.45–0.75	52.0	73.90	Room (23)
[46]	Lateritic clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	12.0	Quarry Dust (QD)	–	–	–	–
[47]	Silty clay	KOH	10.0,12.5	POFA	–	–	7.42	50, 100
[48]	FA paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8.0	FA	–	–	–	–
[49]	GP paste	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	16.0	FA, GBA	0.35	17.56	20.51	70
[50]	BCS	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	16.0	FA	–	–	2.50	Room (23)
[51]	GP mortar	Na <sub>2</sub> SiO <sub>3</sub> + H <sub>2</sub> O	MR (1.7)	GGBFS	–	–	63.00	Room (23)
[15]	Sandy clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	10–15.0	FAF	–	–	11.40	Room (23)
[16]	Sandy clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	10–15.0	FAF	–	–	10.00	Room (19–23)
[52]	Fat clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	10	FAC & FAF	–	–	1.80	23
[53]	Silty sand	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	–	FAF	–	–	0.60	20
[54]	Soft clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	8	GBFS + BOFS	–	–	–	20, 45
[55]	Silty & Clayey sand	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	5	FAC + GGBFS	–	–	16.00	27, 30
[56]	Silty sand	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	5, 12.5	FAC, FAF	–	–	4.20	20
[57]	Silty clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	3, 18	FAF, CCR	–	–	1.20	25, 40
[58]	Silty sand	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	7.5	FAF	–	–	2.30	20
[59]	Clay	NaOH+ Na <sub>2</sub> SiO <sub>3</sub>	14	FAC + GGBFS	–	–	3.20	23

**Table 2** (continued)

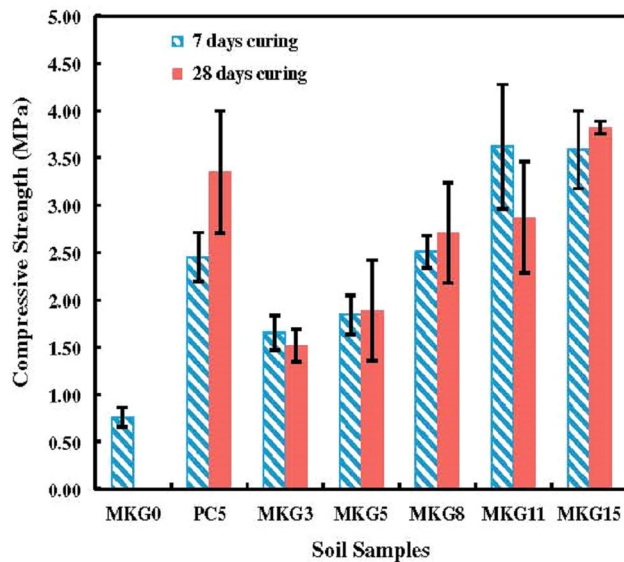
References	Soil type	Alkaline activator	Molarity(M)	Pozzolan	Alkaline/Ash ratio	UCS-7 days (MPa)	UCS-28 days (MPa)	Curing temp (°C)
[60]	FA paste	KOH	8	MK, Dam Sludge	–	–	35.12	40
[61]	FA paste	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	15	BC Fly Ash	–	13.4	56.80	120
[62]	FA paste	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	16	FAF	0.45–0.62	–	38.70	80
[63]	FA paste	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	8	GBS	–	–	48.55	60
[64]	Bangkok clay	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	10–18	BA	–	–	0.973	25
[65]	Red soil	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	10	FAF	0.2	–	11.01	23
[66]	Soft clay	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	8	PJA	–	–	0.37	23
[67]	Silty clay	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	12	FAF	–	–	1.00	20
[68]	Kaolin clay	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	14	FAF + GGBFS	–	–	8.00	23
[69]	Coode island silt	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	8	FAF	–	–	8.50	23
[70]	Silty sand	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	14	FAF	–	–	1.35	23
[71]	Plastic clay	NaOH	4–12	VA	–	–	12.00	40
[72]	Clay	KOH	10	POFA	–	–	1.93	23
[73]	Marine clay	KOH	10	FAC	–	–	1.15	23
[74]	Clay	KOH	12	FAF	–	–	6.45	23
[75]	Clay	KOH	–	MK	–	–	0.85	22
[76]	FA paste	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	12	FAF	2.0	68.00	–	80
[77]	Coarse sand	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	10	FAF	3.0	0.39	0.64	23
[78]	Basaltic tuff	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	10, 12, 14	BT	–	–	30.00	120
[79]	FA paste	NaOH + Na <sub>2</sub> SiO <sub>3</sub>	8	FA	0.4	–	27.36	80
[41]	FA paste	NaOH	8–16	FA + Elaziğ slag	–	–	82.50	50, 100, 150

NB: FA fly ash, FAF class F fly ash, FAC class C fly ash, MKG Metakaolin, GBFS ground bottom furnace slag, GGBS ground granulated blast slag, RAP recycled asphalt pavement, NR not reported, LSS lead smelter slag, GP geopolymer, GGBFS ground granulated blast furnace slag, CCR calcium carbide residue, MK metakaolin, BC brown coal, GBS ground bottom ash, BA bagasse ash, PJA Prosopis Juliflora ash, VA volcanic ash, POFA palm oil fuel ash

<sup>a</sup>UCS after 90 days.



**Fig. 3** Summary of Molarity, Curing temperature and UCS for geopolymer-stabilized soils compiled from 50 literature sources



**Fig. 4** UCS of MKG stabilized soils and Portland cement-stabilized soil samples after 7- and 28-day curing (Zhang et al. [2], with permission from ASCE)

Türkmen et al. [41] utilized fly ash and Elazığ slag to investigate the mechanical properties of geopolymer paste. NaOH was used as the alkali precursor at various molarities of 8, 10, 12, 14 and 16 M and cured at 50 °C, 100 °C and 150 °C for 72 h. Results revealed a high compressive strength of 82.5 MPa at 28 days.

Xiaolu et al. [26] studied the compressive strength and microstructure characteristics of Class C Fly Ash geopolymer paste at curing temperatures of 75 °C for 8 h followed

by ambient (23 °C) for 28 days. The Alkali precursor used was a mixture of NaOH and  $\text{Na}_2\text{SiO}_3$  in a ratio of 1.5. A UCS value of 63.40 MPa was noted at 28 days. The main geopolymeric gel and calcium silicate hydrate (C–S–H) was observed to co-exist from the XRD test.

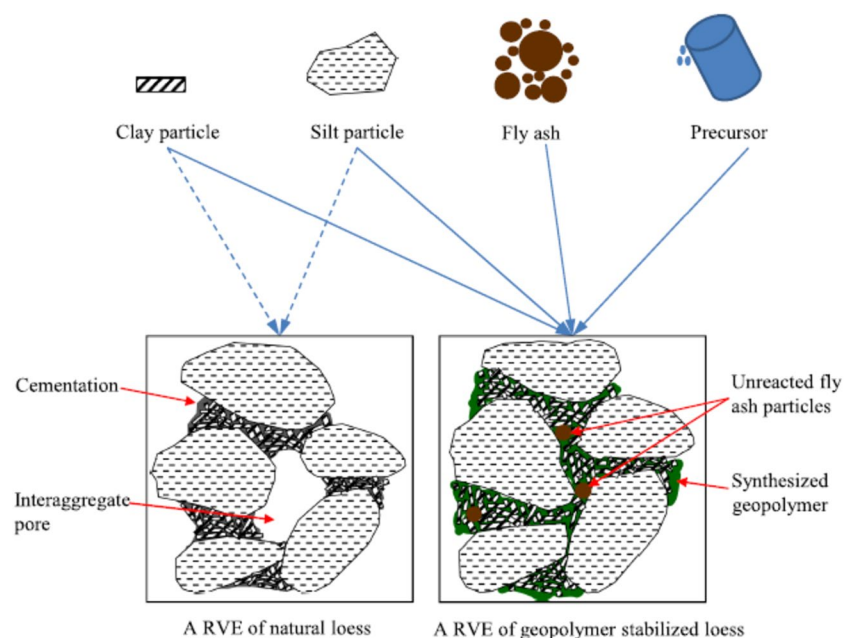
Swanpoel and Strydom [21] used fly ash as a pozzolan with kaolinite and the alkali precursor (NaOH and  $\text{Na}_2\text{SiO}_3$ ) at an elevated temperature of 60 °C (See Table 2). The samples were cured at 40 °C, 50 °C, 60 °C and 70 °C for different time intervals (6, 24, 48, and 72 h). The optimum condition was found to be at 60 °C for a period of 48 h. Compressive strength measurements show a maximum strength of almost 8 MPa after 28 days. Infrared spectroscopic measurements were obtained of the samples after 7 and 28 days.

Liu et al. [17] investigated the stabilization of loess soil with fly ash-based geopolymer using two different precursors, NaOH and KOH. It was found that KOH offers a higher compressive strength than NaOH. More importantly, sealing up the inter-aggregate pores of the stabilized loess soil was observed as shown in Fig. 5.

Conversely, Abdul Rahim et al. [31] compared the use of KOH and NaOH (8 M each) activated fly ash on the mechanical properties of geopolymer paste. It was observed that the highest compressive strength up 65.28 MPa was obtained using NaOH. Meanwhile, geopolymer synthesis using KOH only recorded 28.73 MPa compressive strength. The compressive strength was higher when cured at elevated temperature (60 °C) than room temperature (25 °C).

Kumar et al. [25] investigated the influence of Granulated Blast Furnace Slag (GBFS) on the reaction, structure and properties of fly ash-based geopolymer. It was observed that the reaction at 27 °C is dominated by the GBFS activation,

**Fig. 5** A conceptual micro-structural model of geopolymer-stabilized loess (Liu et al. [17], with permission from Elsevier)



whereas the reaction at 60 °C is due to combined interaction of fly ash and GBFS.

Recently, Cristelo et al. [15, 52] investigated the effectiveness of both low-calcium and high-calcium fly ash-based geopolymers in deep soft soil improvement-grouting process. These studies were conducted by thoroughly mixing alkali-activated fly ash slurry, the geopolymer precursor, with soft soils, and their results indicated that fly ash-based geopolymers were comparable to cement and lime in the stabilization of deep soft soils. According to Pacheco-Torgal et al. [85], geopolymers have been proven to be an effective alternative to OPC in providing civil infrastructures. Furthermore, geopolymers have low shrinkage potential and excellent adhesion to aggregates, suggesting that it can be used as an effective soil stabilizer [86, 87].

Abdullah and Shahin [88] studied the strength characteristics of low and high plastic clay stabilized with Class F fly ash-based geopolymer. The liquid to ash ratio was maintained at 0.4; while for the alkaline precursors, a 14 M of NaOH and Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio of 2.33 was used. Curing of the specimens was executed between 20 and 25 °C with curing periods at 7, 28, and 90 days. Results show that, for both low and high plastic clay, the peak UCS values of untreated clay and geopolymers treated clay were 370 kPa, and 1304 kPa, respectively. When the geopolymer content increased to 20%, the UCS value of geopolymer-stabilized clay increased to 1680 kPa.

Debanath et al. [89] used a fly ash-based geopolymer to treat an expansive soil. The maximum UCS value after treatment with geopolymer was found to be 4.2 MPa at 28 days curing period compared to the UCS of 0.88 MPa for the untreated clay soil. The experimental results also show the decrement of normalized UCS after 20% of geopolymer content. Appraising the past of geopolymer-stabilized soils, it can be confirmed that geopolymers have proven to be effective in treating soft or high compressible soils using a wide variety of pozzolans such as fly ash, blast furnace slag, metakaolin, etc. and alkali precursors. Also, the most commonly used pozzolan is fly ash, mainly Class C due to its high calcium content needed for geopolymerization.

In terms of unconfined compressive strength values from Table 2, a lot of variability can be observed due to different soil types and pozzolan compositions. For example, geopolymer treatment of a Pb-contaminated soil recorded a maximum UCS value of 4.35 MPa as compared to a UCS value of 1.2 MPa after treatment of silty clay [43, 56]. On the other hand, a trend of high UCS values (65.28 MPa, 35.12 MPa, 48.55 MPa) was noted for geopolymer pastes when tested [31, 60, 63]. This shows that the geopolymer paste (alkali + pozzolan) on its own has the tendency to resist a lot more compressive load. However, the variation comes into play when it begins to interact with the microstructure of different soil types and compositions. Another important

factor that affects the production of geopolymers is the ratio of Na<sub>2</sub>SiO<sub>3</sub>/NaOH which varies from 1.0 to about a maximum of 3.0. Experimentally, the most adopted Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios from published information gathered are 1.5, 2.0, and 2.5.

## Engineering Properties of Geopolymer-Stabilized Soils.

Various soil properties have been improved through alkali-activated binders. Two main engineering properties presented here are compressive strength and shear strength of geopolymer-stabilized soils. Other areas of interest also discussed are microstructure, durability, effect of curing and pozzolan composition of geopolymer-stabilized soils.

### Compressive Strength

The compressive strength of a soil is a very important property in supporting the super-structure. One of the most commonly used tests to evaluate the strength of geopolymer-stabilized soils is the Unconfined Compressive Strength (UCS). Past literature sources have observed that the introduction of geopolymer within the soil microstructure improved its unconfined compressive strength [17, 52, 55, 59]. The addition of geopolymer increases the treated soil peak strength, and decreases the corresponding axial failure strain, both contribute towards a stiff response similar to that of OPC-stabilized soils [59, 90].

Generally, the strength improvement of geopolymer-treated soils is a product of the inter-particle bonds induced by geopolymerization of soil particles. However, from analysis of the literature summary presented in Table 2, a high variability in UCS values can be observed from one researcher to another, and it appears that the effect of geopolymer on UCS values varies according to the soil type, alkali activator, and the pozzolans composition (fly ash, blast furnace slag, etc.). According to Cristelo et al. [15], curing conditions (i.e., temperature, and curing age) also affects the volume of reaction products and the level of strength improvement of the stabilized soils. Clay mineralogy also plays a vital role in the level of strength improvement using geopolymer as a result of the interaction between the minerals like (kaolinite, illite or montmorillonite) and the chemicals [91].

### Shear Strength

Shear resistance of a soil is a function of friction and interlocking of particles, which helps to resist imposed load from the super-structure. Shear behavior of geopolymer-stabilized soils is a field that has not been given much attention beyond the routine UCS testing, which has limited utility in the realistic prediction of load–deformation responses due to lack

of control on the drainage conditions or ability to examine the impact of confining pressure [91].

Triaxial test simulates the effects of confining pressure and pore water pressure, which is very pivotal in achieving critical strength evaluation. Published information on triaxial shearing behavior of geopolymer-treated soils considering drained/undrained conditions are very scarce, and most existing studies are mainly focused on stabilizing sands rather than clays. Among the limited research, Rios et al. [56] carried out anisotropic triaxial drained tests on silty sand treated with geopolymer comprised of fly ash (Class F) and a chemical activator based on a 50% weight ratio of sodium silicate to sodium hydroxide. Spectrum of deviator stress was documented for different stabilized mixtures. In all tested samples, a general stress–strain behavior was observed with a brittle response, similar to those of OPC-stabilized soils, in which high peak deviator stresses for stabilized specimens were recorded at low strains, followed by strain-softening [92].

Abdullah et al. [91] investigated the Consolidated Undrained (CU) triaxial behavior of a fly ash-based geopolymer-stabilized clay at ambient temperature incorporating granulated slag. A 14 M NaOH solution was synthesized with the blended pozzolans (fly ash and slag) and used to stabilize the untreated clay. The samples were cured at 20–25 °C for 7, 28, and 90 days, respectively. Results from the CU tests demonstrated that the addition of geopolymer changed the initial characteristics of remolded clays from quasi-over-consolidated to heavily over-consolidated, rendering high yield surface and more effective shear strength parameters (i.e., cohesion and friction angle). Moreover, although the overall qualitative stress–strain and stress path responses of the clays were similar, significant quantitative differences were observed, particularly in terms of the attainable yield strength, stiffness, and shear strength [91].

Thiha et al. [93] studied the shear strength enhancement of compacted clay soils using high-calcium fly ash-based geopolymer. The result of three-ring direct shear tests gives higher shearing strengths for geopolymer-stabilized soils and those strengths increase more in all soil types through curing state. Furthermore, it was noted that fly ash-based geopolymer enhances the shear strength of soils by increasing the cohesion and friction angle.

The shear strength of sand-GGBFS-based geopolymer composite material was also investigated by Hussein and Al-Rkaby [94]. It was found that the structured aluminosilicate-polymerized gel provides significant cohesion, better bonding between the discrete particles of sand, producing a bonded, stable composite. Overall, the drained triaxial strength increased by almost 21.1–53.5% (depending on GGBFS) as the activator ratio changed from 0.2 to 0.4, 6.2, 7.5, and 9.9 times as the GGBFS increased from 0 to

10, 20, 30, 40 and 50%, respectively. Therefore, the inclusion of the GGBFS resulted in a significant increase in the unconfined compression strength, cohesion, and friction angle for all activator contents [94].

Shear strength characteristics of clay stabilized with fly ash-based geopolymer were also studied by Abdullah and Shahin [88]. Based on the results obtained, geopolymer-treated clay specimens exhibited and Consolidated Undrained (CU) peak/residual behavior with higher strength and stiffness compared to untreated clay specimens. The alkali activator solution comprised of fly ash, 14 M NaOH with  $\text{Na}_2\text{SiO}_3$  and cured between 20 and 25 °C for 7, 28, and 90 days. From the triaxial undrained compression results, the effect of the confining pressure was evident as a result of increases in the stiffness and the undrained peak strength of geopolymer-treated clay. In summary, based on the limited published information on shear strength behavior of geopolymer-stabilized soils, it can be inferred that geopolymer enhances the shear strength of coarse and fine-grained soils.

### Microstructure

X-Ray Diffraction (XRD) and Scanning Electron Microscopy (SEM) techniques have been used to provide insights into the microstructure of geopolymer-stabilized soils. Beyond the UCS test, these microstructure tests go deeper to reveal inner dynamics of the soil pores. This involves tracking the cementitious growth induced by the geopolymer to explain the improvement mechanism (formation of gels) of the treated soils at the micro-scale. According to Sargent [95], fly ash-based geopolymers have been reported to increase the density of treated soils, in a manner similar to lime- or OPC-stabilized soils. For example, Liu et al. [80] and Cristelo et al. [52] observed that the homogeneity of clay fabric was improved with the addition of the fly ash-based geopolymer, resulting in more closely linked clay particles and fewer voids. This improvement was mainly traced to the precipitation of artificial cementation products, and the subsequent development of inter-particle bonds within soil particles during curing. This finding was supported by Phummiphan et al. [55] who observed, through SEM analysis conducted on marginal lateritic soil, etched holes on the surface of partially reacted fly ash particles within treated soil. It was claimed that these holes were formed by leaching silica and alumina from the surface of the activated fly ash. The partially reacted fly ash particles and cementitious products within the treated soil are believed to serve as nucleation sites that bond clay plates into clusters, thus modifying the structure of the soil and enhancing its mechanical response [91].



## Durability

Durability is measured in terms of two main properties, i.e., moisture and temperature. These are examples of the field-related conditions that are representatives of a durable performance of binder–soil mixtures [96]. Durability is simulated in the laboratory by wetting–drying and freezing–thawing which are found in ASTM D559-03 [97] and ASTM D560-15 [98], respectively, and are used to evaluate the durability behavior of stabilized soils [99]. In both durability tests, cylindrical specimens are cured for 7 days, and then subjected to 12 successive cycles of temperature and moisture variations (48 h each in duration), simulating potential extreme field conditions, with changes in the measured volume and residual strength [92].

Past literatures on durability of geopolymer-stabilized soils are very scarce and often focused on wetting–drying tests. Rios et al. [58] confirmed a stable performance, i.e., low volumetric change and reasonable residual strength, for Class (F) fly ash geopolymer-treated sand against wetting–drying durability cycles, promoting geopolymer as a viable competitor binder for soil stabilization as compared to OPC. For clay treatment, Sargent et al. [53] similarly reported low volumetric changes in geopolymer-treated clay; however, low residual UCS performance was detected. Sargent et al. [53] reported that the low performance of the activated fly ash binder was attributed to the lack of clay content in the stabilized soil, which limited the cation exchange capacity and chemical reaction. However, the effects of clay mineralogy and plasticity which are known to have significant impact on the performance of clay soil stabilization were not considered [100].

Furthermore, Sargent et al. [53] have not discussed the impact of Class C fly ash used in their work, which is not recommended for alkali activation due to its low silica–alumina content and is likely to negatively affect the expected improvement of geopolymer-treated soils. In terms of the freezing–thawing of geopolymer-treated soils, Abdullah et al. [59] observed high volumetric changes and low residual strength for Class F fly ash geopolymer-stabilized kaolin clay, suggesting that the treated clay exhibits a less stable performance in a freezing climate than tropical climate, and confirming the retardation of the geopolymerization reaction at very low temperatures.

## Effect of Curing

Literature shows that curing conditions have a notable effect on the geopolymerization process. Various researchers used different curing methods ranging from ambient to elevated temperature and water-submerged curing. Higher curing temperature enhances the mechanical properties by increasing the rate of dissolution at early ages but does not have

much effect on final strength [101–103]. However, higher temperature over a consistent period of time also causes fast evaporation of water and propagation of shrinkage cracks along the soil-stabilized soil surface; therefore, extended curing should be avoided [104–106]. Also, wet curing causes efflorescence in the samples [106].

## Effect of Pozzolan Composition

Apart from molarity and curing temperature, the pozzolan composition of industrial by-products such as fly ash, blast furnace slag, plays a key role in achieving effective geopolymer synthesis and ultimately the strength of the stabilized soil. The raw materials for geopolymer synthesis could be natural pozzolanic materials such as volcanic ash (tuff), diatomaceous earth, opaline cherts, shales, pumicites, etc. or naturally tempered pozzolanic materials like zeolite, kaolinite, phonolite, etc. Artificial raw materials for geopolymers include pozzolanic materials from industrial or agricultural waste such as low-calcium fly ash, silica fume, brick powder, granulated blast furnace slag, sugarcane bagasse ash, rice husk ash, etc. [93, 94, 107–110].

Pozzolans used in geopolymers should have the required aluminosilicate materials to have a great enhancement effect on the strength of geopolymer-stabilized soils. For example, Class C fly ash has a different chemical composition from Class F fly ash, which affects the geopolymerization process. The  $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}$  content should be more than 75% and CaO should be less than 15% for a fly ash material to be classified as Class F, while Class C fly ash should be between 50 and 70% of the  $\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}$ . [111]. Typically, Class C fly ash contains more 20% CaO. Even though the alkaline activator is suitable for all types of fly ash, Cristelo et al. [52] noted that Class F is more effective for strength improvement than Class C due to a higher content of silica and alumina in Class F compared to Class C, which contributes to producing a higher number of cementations products, and thereby enhances the soil fabric [92]. Apart from the type of aluminosilicate content, the type of activator has a significant impact on soil improvement using geopolymers. Liu et al. [17], endorsed the use of a KOH-based activator over NaOH to achieve greater compaction and higher artificial bonding on the microstructural level within treated soils, and ultimately resulting in considerable strength improvement. Although the KOH-based activator enhances soil structure more than the NaOH-based activator through the geopolymer, it has some cost limitations [111]. According to (Rios et al. [58], Cristelo et al. [15], Cristelo et al. [16], Phummiphan et al. [33], and Phetchuay et al. [57]), the NaOH activator is commonly suggested for geopolymer soil stabilization. The most commonly used pozzolans (fly ash, blast furnace slag) are measured in percentage of the total weight of the soil to be stabilized. Fly ash content adopted varies from 5% to about 30%. Liu et al. [17] utilized Class F fly ash in increments of 10%, 20% and 30% in stabilizing loess soil. Sitarz et al.

[51] used fly ash and Granulated Ground Blast furnace slag (FA-GGBFS) in increments of 10%, 30% and 50%. It was observed that an increase of the GGBFS content generated an increase of compressive strength. The 50 N composition (50% FA and 50% GGBFS) demonstrated the highest mechanical performance with a UCS value of 68 MPa. Also, FA content of 5%, 10%, 20% was used by Li et al. [43] to stabilized lead contaminated soil. When the FA content increased from 5 to 20%, the Pb concentration decreased from 16.6 mg/L to 0.09 mg/L. In summary, increase in pozzolan content has been proven to effectively enhance the UCS and other soil properties. However, from the review of various literature sources, the pozzolan (fly ash, GGBFS, etc.) content ranges from 5 to 50%.

### Statistical Analysis and Results

Interpreting the results of the engineering data from 50 literature sources, statistical analyses were carried out through one-way analysis of variance (ANOVA) method with the aid of IBM SPSS v.27. The ANOVA was used to determine whether more than two population means are equal. The three (3) population groups here are Molarity, Curing Temperature, and UCS. The hypotheses given in the following equation:

$$H_0 : \mu_1 = \mu_2 = \mu_3 \tag{2}$$

$$H_a : \mu_1 \neq \mu_2 \neq \mu_3 \text{ (at least one of the population means is different from the others),} \tag{3}$$

where  $H_0$  is the null hypothesis,  $H_a$  is the alternative hypothesis, and  $\mu_1, \mu_2,$  and  $\mu_3$  are the population means of Groups 1, 2, and 3, respectively. To be consistent, the one-way ANOVA analysis was also used to test the equality of two population means.

The significance level ( $\alpha$ ) was assumed to be 5% (0.05) for all the statistical tests, which means that if the P-value of a statistical test is less than 0.05, at least one of the population means is different from the others; otherwise, the difference among the population means is not statistically significant [112]. The statistical testing groups (molarity, curing temperature and UCS) are summarized in Table 3, with the

means of molarity, curing temperature and UCS being tested individually. For example, if the P-value of each group is less than 0.05, then there is a statistically significant difference between the three population means. Conversely, if the P-value is greater than 0.05, then the three (3) groups would be assumed not to be statistically different. Furthermore, multiple regression analysis was performed using UCS as the dependent variable, while Molarity and Curing temperature as the predictor (independent) variables. The purpose of the regression analysis is to predict the UCS given the Molarity and Temperature values.

### Discussion of Statistical Results

Table 3 shows the descriptive statistics of the three variables (Molarity, Curing Temperature, and UCS). The UCS value ranges from 0.38 to 82.5 MPa, Curing temperature ranges from 20 to 150 °C, and molarity ranges from 1.7 to 23 M.

Table 4 shows the analysis of variance (ANOVA) which analyses the variability between the three (3) main variables (molarity, UCS and curing temperature). From the ANOVA results, it can be observed that the P-value is less than 0.05. Hence, the null hypothesis is rejected, and this means that there is a statistically significant difference between the molarity, UCS, and curing Temperature.

Table 5 shows the Pearson correlations between the analyzed variables. From the output, it can be noticed that there exists a weak positive linear correlation between UCS and

**Table 4** One-Way ANOVA Summary of Geopolymer variables (Molarity, Curing Temperature and UCS)

Testing Variables	Sum of Squares	df	Mean square	F	Sig.
Between groups	31,649.760	2	15,824.880	29.660	0.000*
Within groups	78,429.987	147	533.537		
Total	110,079.746	149			

Sig. Significance

\* The null hypothesis is rejected at the 5% significance level.

**Table 3** Descriptive statistics of geopolymer variables (Molarity, Curing Temperature, & UCS)

	Sample size, N	Mean	Std. deviation	Std. error	95% Confidence interval for mean		Min.	Max.
					Lower bound	Upper bound		
Molarity	50	11.15600	4.197626	0.593634	9.96305	12.34895	1.70	23.00
Curing Temp.	50	46.48000	31.698992	4.482915	37.47125	55.48875	20.00	150.00
UCS	50	25.12286	24.045078	3.400488	18.28932	31.95640	0.375	82.50
Total	150	27.58629	27.180696	2.219295	23.20093	31.97164	0.375	150.00

**Table 5** Pearson Correlation of Molarity, UCS and Curing Temperature

	Molarity	Curing Temp.	UCS
<b>Molarity</b>			
Pearson Correlation	1	0.111	0.000
Sig. (two tailed)		0.442	0.998
N	50	50	50
<b>Curing Temp</b>			
Pearson Correlation	0.111	1	0.539*
Sig. (two tailed)	0.442		0.000
N	50	50	50
<b>UCS</b>			
Pearson Correlation	0.000	0.539*	1
Sig. (two tailed)	0.998	0.000	
N	50	50	50

N Sample Size, Sig. Significance

\*Correlation is significant at the 0.01 level (two tailed)

curing temperature (0.539), while there was no statistical correlation between UCS and molarity which does not tally with the laboratory experimental results. While this weak positive correlation could be attributed to the variability in data as a results of different soil types, it also confirms there are other factors that contribute significantly to the UCS of geopolymer-stabilized soils such as ash/alkaline ration, pozzolan composition.

Table 6 shows a summary of the predictors (molarity and curing temperature).  $R^2 = 0.295$  implies a unique contributor of molarity and curing temperature that explains 29.5% of the variability in the dependent variable (UCS). This

**Table 6** Model summary for molarity, UCS and curing temperature

Model	R	R <sup>2</sup>	Adjusted R <sup>2</sup>	Std. error of the estimate	Change Statistics				
					R <sup>2</sup> change	F Change	df1 <sup>b</sup>	df2 <sup>c</sup>	Sig. F change
1	0.543 <sup>a</sup>	0.295	0.265	20.618495	0.295	9.820	2	47	0.000

<sup>a</sup>Predictors: (Constant), Curing Temp, Molarity. Dependent variables (UCS)

<sup>b</sup>df1 degree of freedom for Curing Temp.

<sup>c</sup>df2 degree of freedom for Molarity.

**Table 7** Regression coefficients between molarity, UCS and curing temperature

Model	Unstandardized Coefficients		Standardized Coefficients	t	Sig	95.0% Confidence Interval for B	
	B	Std. Error				Beta	Lower Bound
1 (Constant)	9.769	9.046		1.080	0.286	- 8.429	27.967
Molarity	- 0.350	0.706	-0.061	- 0.496	0.622	- 1.771	1.070
Curing_Temp	0.414	0.094	0.546	4.432	0.000	0.226	0.602

<sup>a</sup>Dependent Variable: UCS

confirms there are blends of other factors that account for 70.5% of the UCS of geopolymer-stabilized soils such ash/alkaline ratio, NaOH: Na<sub>2</sub>SiO<sub>3</sub>, pozzolan composition, etc.

Table 7 presents the results of the linear regression analysis using UCS as the dependent variable, while Molarity and Curing temperature as the predictors. The Multiple linear regression model from the Table 7 gives the relation presented in Eq. (4):

$$UCS = 0.414 \times \text{Curing Temperature} - 0.350 \times \text{Molarity} + 9.769, \tag{4}$$

which means for known values of curing temperature and molarity, the UCS could be estimated approximately using Eq. 4. However, this regression model would be more accurate for large sample sizes.

From Fig. 6, the Normal P–P Plot shows that the available data are approximately close to the line of fit and hence can be assumed to be approximately normal.

### The Future: Sustainability of Geopolymer-Stabilized Soils

With promising results from geopolymer-stabilized soils, it is important to predict the future of its applications. The flow chart in Fig. 7 shows long-term durability and Life Cycle Cost Assessment (LCCA) as major branches of future geopolymer research. In assessing future relevance of geopolymers, sustainability has been viewed as a major fulcrum. Utilization of geopolymers must transcend the first zone of effectiveness in improving soil properties to the zone of sustainability. This is only possible when it can be fully

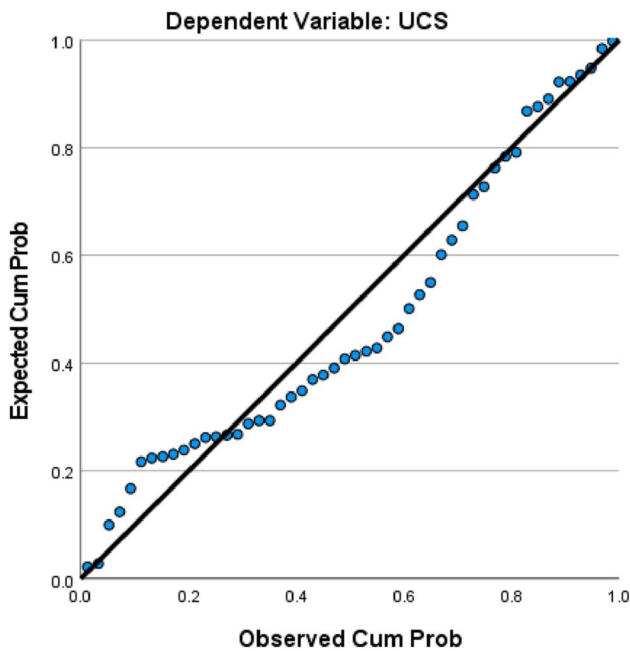


Fig. 6 Normal P-P plot of regression standardized residual

beneficial not only just to infrastructures, but also to the environment at large. Sustainability is relatively an emerging field that cuts across majority of disciplines, though its application seem to have more depth in civil engineering infrastructures. According to Brundtland’s declaration, “sustainable development is development that meets the needs of the present without compromising the ability of future generations to meet their own needs [113, 114]. Published information on sustainability of geopolymer-stabilized soils are very limited in nature, as it appears majority of the research focus is on the present.

Past research efforts towards improving the sustainability index of concrete have yielded promising results with the use of geopolymers, resulting in early compressive

strength, low permeability, excellent chemical, and fire resistance [86, 115–119]. More importantly, within the context of geopolymer-stabilized soils, sustainability is estimated in terms of a parameter called Sustainability Index ( $I_{Sus}$ ), which is based on a weighted multi-criteria framework recently introduced at University of Texas at Arlington [13, 31]. The  $I_{Sus}$  of a material was proposed to be a function of its resource consumption, environmental impact, and socio-economic impact, as shown in Eq. 5.

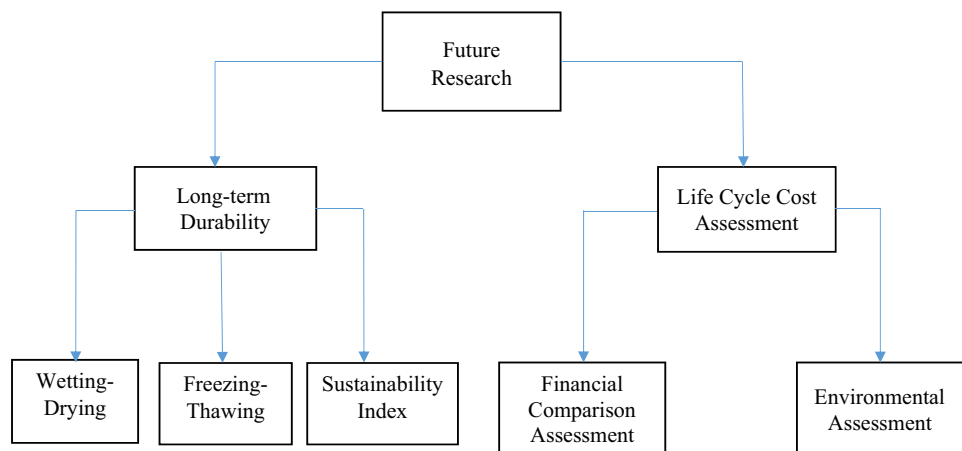
$$I_{Sus} = (W_1 \times I_{Rec}) + (W_2 \times I_{Env}) + (W_3 \times I_{SoEc}), \quad (5)$$

where  $I_{Rec}$  is the resource consumption index,  $I_{Env}$  is the environmental impact index,  $I_{SoEc}$  is the socio-economic impact index, and  $W_1$ ,  $W_2$ , and  $W_3$  are the weighted values of each associated index.

Recent information on this topic was reported by Samuel et al. [75] where the sustainability benefits assessment of metakaolin-based geopolymer treatment of high plastic clay was carried out. Results revealed that the metakaolin-based geopolymer-stabilized soil was found to be a more sustainable alternative with a lower sustainability index,  $I_{Sus}$ . Also, the global warming potential and embodied energy of production of a kilogram of lime were found to be significantly higher than metakaolin, the primary component of the geopolymer. This agrees with the known fact that production of lime and cement has significant contributors to global greenhouse and gas emissions.

The above research on sustainability by Samuel et al. [75], and the framework introduced by the University of Texas at Arlington can be used in carrying out further sustainability benefit assessment of other pozzolans such as fly ash, GGBFS since they are used frequently than other materials. Natural pozzolans like volcanic ash should also be evaluated for sustainability. Furthermore, Life Cycle Cost Assessment (LCCA) should be carried out to compare and contrast the cost-effectiveness of using alkaline activators, especially on road infrastructures.

Fig. 7 Flow chart showing the future of geopolymer-stabilized soils



## Limitations to Full Adoption of Geopolymers for Soil Stabilization

Adoption of geopolymers as a reliable alternative is still being met with a lot of skepticism in the geotechnical engineering community. The reason for this is not far-fetched. Many engineers still prefer the use of traditional binders (OPC and lime) despite their well-documented detrimental effects on the environment in terms of CO<sub>2</sub> emissions [120]. Also, the cost of the alkaline activator chemicals could be counter-productive, especially on a highway project except the cost is balanced out by utilizing a large content of natural or artificial pozzolans from industrial wastes.

Due to the heterogeneous nature of soils, variability of stabilization results is expected because soils responds differently to various alkaline activators. Hence, the absence of practical mix design templates for geopolymers has been a major drawback.

With the risk of handling hazardous chemicals (NaOH and Na<sub>2</sub>SiO<sub>3</sub>) as alkali precursors, the most preferred stabilizing agents would naturally be in powdered form for easy application. Hence, advanced geopolymerization through solid-state chemistry should be endorsed so that the precursors can be produced in powdered form and applied by dry weight of the soil sample to be more eco-friendly. Also, long-term durability under freeze–thaw actions and wet–dry conditions has not been well established for geopolymers compared to the conventional OPC and lime methods.

The use of 150 °C curing temperature as reported by Türkmen et al. [41], even though effective in the laboratory, poses a great difficulty in achieving this in the field. This is a notable limitation of geopolymers in terms of practicability. However, temperature range of 40–60 °C is more practical to simulate in the field. Furthermore, while the use of high temperatures (100–150 °C) could achieve experimental results, implementation of the field should be given more priority to gain more acceptance.

## Conclusions

Geopolymers have been proven over the last decade, to be worthy alternatives to conventional stabilization due to enhanced strength, reduced shrinkage, reduced porosity, etc. This paper provided a critical appraisal of the available literature on using geopolymer as an eco-friendly alternative binder for soil stabilization. From the appraisal, fly ash, GGBFS-based geopolymers can be used successfully as binders to improve soil properties. In addition to the literature review, statistical analysis was carried out using limited data (50 literature references) which showed some correlation between UCS and curing temperature.

The future of geopolymer-stabilized soils revolves mainly around sustainability, and durability. In terms of sustainability, geopolymer-stabilized high plastic clay was found to be more sustainable, with a lower sustainability index,  $I_{Sus}$ , as compared to OPC-stabilized soils. More importantly, the use of geopolymers will need to shift away from curing at elevated temperature to ambient temperature to boost its increased adoption above the conventional OPC.

Published information on long-term durability under freezing–thawing, wetting–drying action was found to be very scarce. Hence, further research should be carried out on long-term durability and prediction models should be developed from the experimental data.

**Author's contribution** 1st author draft paper, run the statistical analysis software, plotted the results; 2nd author contributed discussion and analysis of results, modified the figures, and review the paper.

## Declarations

**Conflict of Interest** There is no conflict of interest. No external funding was received for conducting this study.

**Data availability statement** All data, generated or used during the study may available upon request.

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