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An approach towards achieving net-zero-carbon concrete

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Abstract:
Carbon dioxide is the primary greenhouse gas contributing to climate change. The construction industry is a main contributor to carbon dioxide emissions worldwide and must make conscious efforts towards becoming a green industry by using materials that are better for the environment. This study investigated innovative approaches to reduce carbon dioxide emissions in concrete production by replacing traditional Portland cement and paving the way to achieving net-zero-carbon concrete. Geopolymer mixes were evaluated as cement alternatives. In addition, alternative partial replacements for cement like ground granulated blast furnace slag, alongside the incorporation of various admixtures such as titanium dioxide, zinc oxide, and biochar, were tested. Tests were conducted to evaluate the compressive strength, durability, and carbon dioxide emissions. Comparisons to conventional Portland cement concrete were performed to quantify the environmental benefits of the developed concrete mixtures. The full replacement using a geopolymer was a significant step towards net-zero carbon emissions. It yielded higher strength and durability than the ordinary Portland cement concrete. The geopolymer concrete showed promising results with no curing and better results after 1 day of heat curing, which set this material steps ahead of other alternatives. Alongside the admixtures, the studied concrete model maintained the characteristics needed for structural concrete while reducing the contribution to the carbon dioxide present in the atmosphere and providing greater strength. The use of admixtures like titanium dioxide and biochar effectively enhanced geopolymer concrete while increasing its carbon dioxide absorption.

Keywords:
portland cement concrete; sustainability; net-zero-carbon concrete; geopolymer; GGBS; titanium dioxide; zinc oxide; biochar

1 Introduction

There has been a rising demand for building materials, especially Portland cement, which makes up approximately 10-15 % of a concrete mix and is essential in the construction industry [1]. The cement industry accounts for a staggering 8 % of carbon dioxide emissions [2]. This alarming statistic underscores the urgent need for a paradigm shift towards net-zero-carbon concrete because the repercussions of unmitigated carbon emissions extend far beyond construction sites. The production of cement releases a substantial amount of carbon dioxide into the atmosphere. For every kilogram of cement produced, 0,9 kg of carbon dioxide is emitted [3]. Considering that cement is the second most utilised substance globally after water, the environmental impact of its production cannot be overstated [3]. The consequences of this excessive carbon footprint extend beyond the construction sector and touch upon critical global challenges, the most notable of which is climate change. Alarming increases in global temperatures, sea levels, and the frequency and intensity of wildfires are undeniable indicators of the effects produced by the widespread use of lime and conventional Portland cement, demonstrating the need for third-generation binders, which are often referred to as amorphous alkali aluminosilicates [4].

The Earth's average temperature has risen by a concerning 0,18 °C per decade [5]. This may seem to represent a modest increase, but its implications are profound. Rising temperatures contribute to countless environmental issues, including disruptions to the food supply. Changes in temperature and precipitation patterns directly affect crop yield, thereby posing a threat to global food security [4]. Moreover, the melting of the polar ice caps and glaciers, which is a direct consequence of higher temperatures, contributes significantly to rising sea levels. This phenomenon poses a threat to coastal communities and ecosystems, and highlights the urgent need for industries to address carbon emissions [5].

An increase in the frequency and intensity of wildfires is another symptom of human-induced climate change. Higher temperatures, changes in precipitation, and prolonged droughts create conditions that render wildfires more destructive and pose severe threats to both human life and biodiversity [4]. The previously highlighted consequences of climate change have severe impacts on the environment, which are alarming. These consequences demonstrate the need to change to net-zero-carbon concrete. It is important to maintain building structures without worsening climatic conditions. Embracing green and durable materials, particularly net-zero-carbon concrete, will indeed be an efficient strategy for mitigating the effects of the construction industry on climate-change disasters.

The main goal of this research was to examine the impact of incorporating innovative materials and binders in the pursuit of achieving net-zero-carbon concrete. In this study, a comprehensive analysis and assessment was conducted on calcium silicate as an alternative binder to cement. In addition, the effects of zinc oxide, titanium dioxide, and biochar on capturing carbon dioxide were investigated. Regarding the environmental aspects, a comparative feasibility study was conducted to evaluate the differences between the developed mixes and a conventional concrete mix. In addition, an attempt was made to quantify the carbon dioxide emissions and carbon dioxide absorption and compare them with those of conventional concrete.

2 Literature review

2.1 Geopolymer concrete

Geopolymers are recognised as successors to lime and conventional Portland cement, marking them as third-generation binders, which are often referred to as amorphous alkali aluminosilicates [6]. Several factors influence the strength of geopolymer concrete: increasing the molar concentration of the sodium hydroxide solution enhances compressive strength; a higher sodium silicate-to-sodium hydroxide ratio by mass boosts the compressive strength; curing at temperatures between 30 and 90 °C results in greater compressive strength as the temperature rises; and longer curing times of 6-96 h improve the compressive strength, although the gains beyond 48 h are minimal. Adding a high-range water-reducing admixture of up to approximately 2 % fly ash by mass improves the workability of fresh geopolymer concrete with little impact on its compressive strength [7]. Increasing the molarity of the NaOH solution also reduced the slump [8].

Singh et al. examined the environmental impact of Portland cement concrete, noting its high energy consumption and significant CO_2 and greenhouse gas emissions. Their study proposed the use of a geopolymer binder as an alternative, which was created by combining fly ash, metakaolin, or ground granulated blast-furnace slag with an alkaline solution of sodium hydroxide and sodium silicate. The study explored various factors, including the impact of temperature on geopolymer concrete, setting time variations with different sodium hydroxide concentrations, along with the effects of the curing technique (ambient or steam curing) with different alkaline solution ratios on the 28-day compressive strength. The results showed that geopolymer concrete cured at ambient temperature or through steam curing has potential as an alternative to Portland cement concrete, with optimal results obtained with a sodium silicate-to-sodium hydroxide ratio of 2 [9].

A recent review by Verma et al. highlighted several advantages of geopolymer concrete, such as cost-effectiveness, low energy consumption, workability, eco-friendliness, and durability. It also helps reduce the carbon footprint by utilising industrial wastes, such as slag, fly ash, and rice husk ash. Consequently, geopolymer concretes hold significant promise in the construction industry and are suitable for bridges, high-rise buildings, highways, tunnels, dams, and hydraulic structures owing to their excellent performance [10].

2.2 Titanium dioxide

The incorporation of titanium dioxide in concrete offers various environmental and mechanical benefits. Nano-titanium dioxide has been shown to improve the mechanical properties of concrete, including the compressive strength, abrasion resistance, and fire resistance. Studies have demonstrated that incorporating TiO_2 in amounts ranging 0,5-12,0% enhances these properties [11]. The use of titanium dioxide as a 1,5 % cement replacement increased the compressive, split tensile, and flexural strengths of concrete [12]. Concrete containing titanium dioxide exhibits lower water absorption, enhancing its durability. For example, concrete cubes with TiO_2 absorbed 28,4 % less water than conventional concrete cubes [13]. TiO_2 enhances the resistance to freeze-thaw cycles and scaling, improving its durability under harsh environmental conditions. It also accelerates early age hydration and reduces pore connectivity, which is beneficial for structural applications [14]. An optimal compressive strength and minimal corrosion rates in self-cleaning concrete were achieved with 2,5 % nano-titanium dioxide. Additionally, the use of TiO_2 in lightweight foamed concrete (LFC) improves its durability properties, such as its water absorption, porosity, and drying shrinkage [15; 16].

A significant drawback of using titanium dioxide is the observed reduction in compressive strength, both at early and later ages, particularly when TiO_2 is incorporated at higher concentrations. Studies indicate that dosages exceeding 5 % of the cement by weight can lead to a noticeable decrease in the 28-day compressive strength, with some research reporting reductions of up to 15-20 % with a 10 % TiO_2 addition. This strength reduction is often attributed to the high specific surface area of nano-TiO_2 particles, which can increase the water demand and lead to incomplete hydration or agglomeration, creating weak zones within the cement matrix [17-19]. Different crystalline forms and surface treatments of TiO have been shown to influence its mechanical and photocatalytic behaviours, with anatase-rich composites exhibiting higher reactivity, often leading to the more aggressive degradation of admixtures. For instance, Słosarczyk et al. [17] demonstrated compressive strength decreases exceeding 30 % in mixes containing 5-10 % TiO_2. Similar outcomes were observed by Shafaei [20], who analysed hundreds of NT-based composites.

Furthermore, the fine particulate nature of TiO_2 can adversely affect the workability of fresh concrete, leading to a decrease in slump and an increase in viscosity. This requires a high water-to-cement ratio to maintain the desired consistency, which can compromise the strength and durability of the hardened concrete [21; 22]. The tendency of nano-TiO_2 particles to agglomerate because of their high surface energy poses another challenge, because these clusters can act as defects within the matrix, hindering the formation of a homogeneous and dense microstructure [23].

Photocatalytic activity, which is beneficial for environmental applications, can potentially degrade organic admixtures such as superplasticisers, leading to a loss of workability over time or reduction in the efficiency of other chemical additives. This degradation can complicate the mix design and quality control, requiring careful selection of compatible admixtures or dosage adjustments [24].

2.3 Zinc oxide

The addition of ZnO to concrete improves its compressive and flexural strengths. Studies have shown that ZnO nanoparticles can increase the compressive strength by up to 20 % and improve the flexural strength when added at small percentages [25; 26]. ZnO enhances the durability of concrete by counteracting the deterioration caused by pollutants. Experimental tests have demonstrated that this can significantly improve the longevity of concrete structures [27]. ZnO is an effective corrosion inhibitor for the steel reinforcements in concrete. It has a significant inhibition efficiency, especially at certain concentrations, protecting steel from corrosive environments [28]. ZnO microcapsules have been used to improve the compressive and flexural strengths of mass concrete and enhance its crack-sealing capabilities [29]. ZnO increases the skin friction and adhesion capacity of concrete, making it more resistant to environmental factors, such as water absorption and freeze–thaw cycles. This improvement in durability has been attributed to the pore-filling capability of the ZnO nanoparticles, which also accelerate cement hydration [30]. The use of highly amorphous metakaolin eliminated chemical retardation, while maintaining this acceleration. In addition, this increased the 28-day compressive strength [31]. Mortars with 1 % by weight ZnO additions to kaolinite calcined at 650 °C showed slightly higher 7- and 28-day strengths compared to non-ZnO samples. Chemical retardation was almost eliminated for the highly dehydroxylated metakaolin when ZnO was used. This allows the use of various ZnO-containing waste materials as concrete additives [32].

The primary disadvantage of adding ZnO to concrete is its pronounced retardation effect on the cement setting time and subsequent reduction in the early age compressive strength. This delay is attributed to the formation of a zinc hydroxide ($Zn(OH)_2$) layer around the cement particles, which acts as a barrier impeding the dissolution of calcium ions and formation of calcium silicate hydrate (C-S-H), which is the primary phase responsible for the strength of concrete. Specifically, Tamashiro et al. [33] demonstrated that ZnO nanoparticles induce a continuous $Zn(OH)_2$ layer that delays cement hydration. This barrier effect is particularly problematic at higher ZnO dosages, where the setting time can be extended by several hours or even days, rendering the concrete impractical for typical construction applications requiring rapid strength development [26; 31].

Beyond the delayed setting, ZnO incorporation typically leads to a reduction in both the early- and later-age compressive strengths. For instance, Liu et al. [34] observed that even small amounts of ZnO reduced the early strength owing to inhibited hydration. ZnO delayed setting and reduced the compressive strength of alkali-activated cements. A 1 % addition of ZnO can decrease the 28-day compressive strength by 10-20 %, whereas higher additions, such as 3 %, resulted in a more substantial reduction of up to 40 %, as corroborated by Li et al. [35]. This strength reduction is not merely a consequence of delayed hydration, but also arises from the disruption of the microstructure, potentially leading to increased porosity and a less dense C-S-H gel structure [36]. The impact on strength is often more severe at early ages (e.g., 3 and 7 days) but can persist for 28 days and beyond, indicating a fundamental alteration in the hydration process rather than a temporary delay [31].

2.4 Biochar

Biochar has the potential to improve the properties of concrete and contribute to sustainability. Biochar, which is a carbon-rich product derived from organic materials via pyrolysis, offers several benefits when incorporated into concrete mixtures. Biochar can improve the mechanical strength and the thermal and physical properties of concrete. Research indicates that biochar enhances the durability of concrete by improving its resistance to various attacks, shrinkage, and permeability, while also increasing its carbon sequestration capacity [37]. Biochar not only enhances the durability of concrete but also provides environmental benefits by reducing CO_2 emissions from cement production. This improves the sustainability of construction materials and aligns with the goals of eco-friendly construction practices [38]. Studies have shown that biochar can increase the compressive strength and improve the flexural performance of reinforced concrete beams. Thus, it is a viable option for enhancing the mechanical properties of reinforced concrete structures [39]. The optimal amount of biochar for enhancing concrete properties is generally small, with studies suggesting that replacing up to 1-3 wt% of the cement with biochar can significantly improve the compressive strength and other mechanical properties [40]. The ability of biochar to sequester carbon and improve its environmental profile is a significant advantage. It captures and stores carbon, thereby reducing the overall carbon footprint of construction projects [41].

The most prominent drawback of using biochar in concrete is the significant reduction in the compressive strength that results, especially at higher dosages. This strength loss is largely a result of the high porosity and low density of the biochar particles, which act as voids within the cement matrix, increasing the overall porosity of the hardened concrete and reducing its load-bearing capacity. For example, Yang and Wang [42] observed a 20-30 % decrease in the 28-day compressive strength with a replacement dosage of 5 %. A critical review by Winters et al. [43] reinforced this finding, highlighting that even though biochar enhances the carbon sequestration and environmental value of concrete, its integration into cement systems often leads to mechanical compromises owing to its porosity, heterogeneity, and disruption of the hydration process. Similar findings were reported by Huang et al. [44], who confirmed that the highly porous structure of biochar-modified concrete is a key factor in its strength reduction.

The absorptive nature of biochar contributes to an increased water demand and reduced workability of fresh concrete. Biochar readily absorbs water from the mix, reducing the amount available for cement hydration, which often necessitates a higher water-to-cement ratio to maintain adequate workability. This increased water demand can further reduce the strength and hinder proper placement and compaction [45]. Gupta et al. [46] highlighted that the unburnt carbon and inorganic impurities present in certain biochars can interfere with cement hydration by limiting the formation of calcium silicate hydrate (C-S-H), which is the primary phase responsible for the strength of cement.

Environmental and consistency concerns also exist. Schreiter et al. [47] documented the potential leaching of contaminants from biochar depending on the feedstock and pyrolysis conditions, raising environmental safety issues in construction applications. This was reinforced by Sargent et al. [48], who reported geochemical contamination risks with certain biochar-based binders. The highly variable nature of biochar, owing to differences in the biomass feedstock and production parameters, further complicates its application to cementitious systems. According to Fleury [49], this variability necessitates extensive material characterisation and batch-to-batch quality control to ensure consistent performance.

2.5 Ground-granulated blast furnace slag

The incorporation of ground-granulated blast furnace slag (GGBS) into concrete improved its mechanical properties and durability. The use of GGBS can enhance the resistance to chemical attacks and improve the overall lifespan of concrete [50]. GGBS can be used as a partial cement replacement in concrete, with a typical substitution range of 30-60 %. This replacement conserves natural resources and improves the strength of concrete over longer hardening periods [51]. Research has shown that replacing up to 30 % of the cement with GGBS can increase the compressive strength of concrete. Beyond this percentage, the strength may start to decline [52]. As a pozzolanic material, GGBS reacts with the calcium hydroxide produced during cement hydration, forming additional calcium silicate hydrate, which enhances the properties of concrete at later ages [53]. Utilising GGBS in concrete not only provides a sustainable way to use an industrial by-product but also reduces the carbon footprint of construction activities. It promotes the use of waste materials and reduces landfill disposal [54].

The most notable disadvantage of GGBS concrete is its slower rate of early-age strength gain compared to ordinary Portland cement (OPC) concrete. This is because GGBS reacts pozzolanically, requiring the liberation of calcium hydroxide (CH) during cement hydration to form additional calcium silicate hydrate (C-S-H). This pozzolanic reaction proceeds more slowly than the direct hydration of cement, resulting in a reduced early-age compressive strength (e.g., at 1, 3, and 7 days). For example, Alkuhly [55] demonstrated that GGBS concrete has a lower strength at early ages, although it gains strength significantly with time owing to continued pozzolanic activity. Tan [56] further emphasised the need for careful early curing to mitigate slow hydration rates and early-age strength deficiencies. For applications requiring early formwork removal such as precast concrete or structures, this delay can lengthen construction timelines and escalate costs.

Another significant concern regarding GGBS concrete, particularly at high dosages, is its increased susceptibility to carbonation, which occurs when atmospheric CO_2 reacts with CH and C-S-H, decreasing the pH of the pore solution. As the GGBS consumes CH during its pozzolanic reaction, its buffering capacity against carbonation decreases. Jia et al. [57] found that concrete with GGBS experienced accelerated carbonation when it was not properly cured. Similarly, Harold [58] confirmed that the carbonation depth is strongly correlated with inadequate early curing and reduced CH content in GGBS mixes. This can compromise the durability of the reinforced concrete by lowering the pH required to passivate the steel.

Additionally, GGBS concrete often requires more water to maintain workability or must rely on superplasticisers because of the finer particle size and glassy texture of slag particles. This increased water content, if uncontrolled, can increase the porosity and decrease the strength, as noted in a binder composition study of Visser [59]. To achieve the desired strength and durability, GGBS concrete requires proper curing, which is critical for facilitating a slower hydration process. Yousuf [60] emphasised the importance of early-age curing for supplemental cementitious materials (SCM)-blended concretes, including those containing GGBS, to prevent poor microstructure-development and carbonation risks. Without adequate curing, the potential benefits of GGBS in terms of durability and sustainability are significantly undermined.

3 Methodology

3.1 Experimental design

The experimental work in this research was divided into two phases. The first phase included material characterisation. In this phase, all the raw materials to be used in the mix-design process were tested to determine their properties. All of the materials met both the ASTM and Egyptian standards. During this phase, nine geopolymer pastes were used to determine the optimal ingredient ratios. The second phase involved the concrete-mix development and testing. In this phase, 16 concrete mixes were developed and tested. One Portland cement mix was used as the control mix, which served as a benchmark for the performance evaluations of the other mixes. Three mixes incorporating different percentages of GGBS as a partial cement replacement were used to study the effect of GGBS, which is a waste that reduces the carbon footprint of the mix. Two other mixes were prepared using geopolymer concrete, in which the binder was a more sustainable alternative than cement. Two mixtures were produced using dioxide injection to study the possibility of carbon dioxide sequestration. In another approach for studying geopolymer concrete for carbon dioxide sequestration, two mixes were poured using carbonated water instead of regular water. One of these two geopolymer mixes was selected to study the effects of three admixtures known to absorb carbon dioxide in the mix. The three admixtures used were titanium dioxide, zinc oxide, and biochar. This procedure and apparatus have been reported previously [61]. The purpose of these 16 mixes was to study the effects of different materials on the carbon footprint of the mix. The detailed phase two plan is shown in Figure 1.

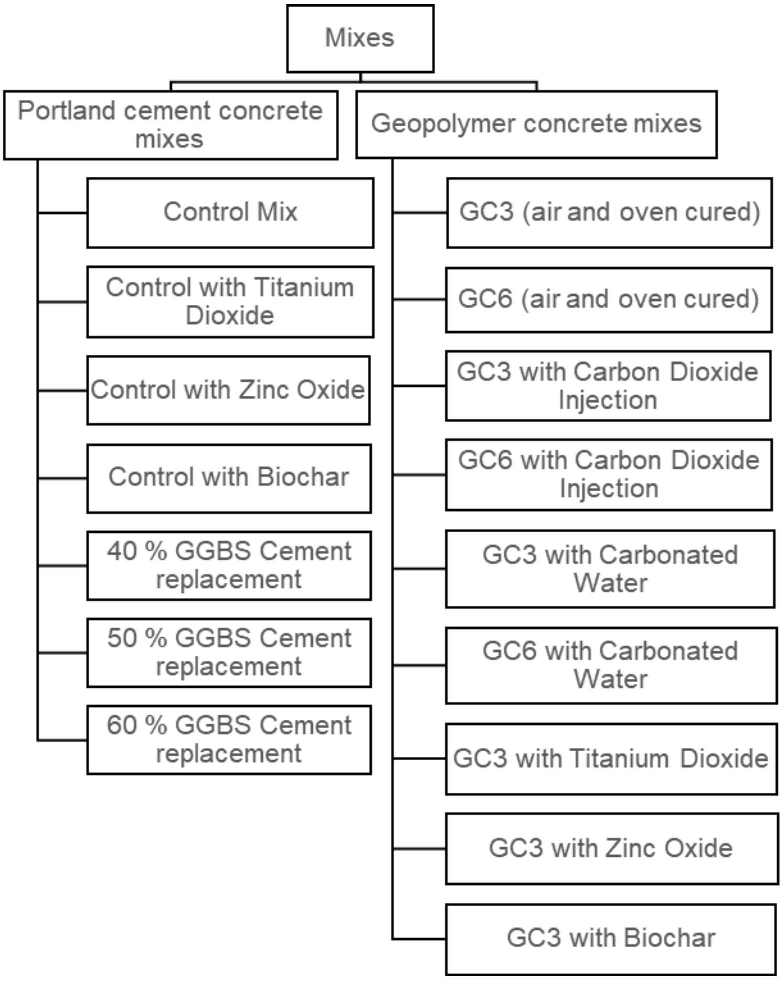

Figure 1. Phase 2 plan

3.2 Materials

Several materials were required to achieve these research goals. The following materials were used in the study after test results showed that they passed all of the ASTM and Egyptian Codes of Practice requirements:

- Type I OPC.
- Well-graded natural sand (passing sieve no. 4 and retained on sieve no. 200).
- Well-graded gravel (passing through a 1,5-inch sieve and retained on sieve no. 4).
- Clean potable water.
- Kaolin hydroxylated to attain metakaolin.
- Sodium hydroxide crystals.
- Calcium silicate powder.
- Titanium dioxide powder.
- Zinc oxide powder.
- Biochar powder.
- GGBS.
- Calcium silicate.
- CO_2 gas.
- Carbonated water.

3.3 Concrete mix design

3.3.1 Portland cement concrete mix

A conventional Portland cement concrete mix was used as a control sample. The target compressive strength of the mixtures was 30 MPa. This compressive strength was selected because it is commonly used in the Egyptian market, while considering durability requirements. To achieve these requirements, a water-to-cement ratio of 0,43 was chosen for these mixes. Three additional Portland cement mixes were prepared to study the effects of waste materials. The Portland cement concrete mixes were thoroughly mixed and cast according to ASTM standards (ASTM C192/C192M). The design of the control mixture is listed in Table 1.

Table 1. Control mix design

Cement (kg/m^3)	**Water** (kg/m^3)	**Coarse aggregate** (kg/m^3)	**Fine aggregates** (kg/m^3)
460	200	1265	475

3.3.2 GGBS partial replacement

Three identical mix designs were tested with different GGBS contents (40, 50, and 60 % of the cementitious material weight). The purpose of this study was to systematically investigate the alterations that occur and interpret the resulting modifications in concrete properties depending on the replacement percentage of GGBS within the mix. The mix designs for the three GGBS mixes are listed in Table 2.

Table 2. GGBS mix designs

Mix number	**w/c ratio**	**Cement** (kg/m^3)	**GGBS** (kg/m^3)	**Water** (kg/m^3)	**Coarse aggregate** (kg/m^3)	**Fine aggregates** (kg/m^3)
GGBS 40 %	0,4	300	200	200	1265	445
GGBS 50 %	0,4	250	250	200	1265	445
GGBS 60 %	0,4	200	300	200	1265	445

3.3.3 Geopolymer paste

In the experimental synthesis process, calcium silicate geopolymer was developed by combining three key components: metakaolin, which was sourced as kaolin and dehydroxylated at 750 °C; calcium silicate, which was subjected to calcination at 1000 °C; and sodium hydroxide, which was diluted with water. The primary objective of this synthesis was to determine the optimal ratio of these three components. This balance is crucial to achieve the desired properties and performance of the end product. Additionally, this study had the goal of determining the most favourable temperature for the reaction to occur. Selecting the optimum temperature is pivotal because it directly influences the kinetics and extent of the chemical reactions between the components, ultimately affecting the structural and functional characteristics of the material.

Nine pastes with different compositions were tested. The mixes were both air- and oven-cured. The percentages of the components for the nine different mix designs are listed in Table 3. It shows the percentages of calcium silicate, metakaolin, sodium hydroxide, water used to dilute the sodium hydroxide, and extra water used for the mix. The label GP is used to designate a geopolymer paste, followed by the mix number, as presented in Table 3.

Table 3. Geopolymer paste mix designs

Geopolymer mix number	**Mix ID**	**Calcium silicate** (%)	**Metakaolin** (%)	**Sodium hydroxide** (%)	**Water with sodium hydroxide** (%)	**Additional water** (%)	**Alkalinity** (%)
Geopolymer paste mix 1	GP1	11,40	57,01	7,64	15,28	8,67	50,00
Geopolymer paste mix 2	GP2	8,69	54,32	9,45	18,90	8,64	50,00
Geopolymer paste mix 3	GP3	8,62	57,47	13,22	17,24	3,45	76,67
Geopolymer paste mix 4	GP4	11,04	54,11	14,29	17,32	3,25	82,50
Geopolymer paste mix 5	GP5	11,98	52,08	15,10	17,71	3,13	85,29
Geopolymer paste mix 6	GP6	11,85	57,80	9,54	12,43	8,38	76,74
Geopolymer paste mix 7	GP7	10,82	58,48	9,94	14,04	6,73	70,83
Geopolymer paste mix 8	GP8	5,11	63,90	9,90	15,34	5,75	64,58
Geopolymer paste mix 9	GP9	0,00	63,29	15,82	20,89	0,00	75,76

3.3.4 Geopolymer concrete

Geopolymer paste mixes GP3 and GP6 listed in Table 3 were selected to produce geopolymer concretes, which were referred to as GC3 and GC6, respectively. The selection of these two pastes for use in concrete mixes is discussed in detail in section 4.2.4. The mix designs for geopolymer concrete mixes GC3 and GC6 are listed in Table 4.

Table 4. Geopolymer concrete mix designs

Mix ID	**Calcium silicate** (%)	**Metakaolin** (%)	**Sodium hydroxide** (%)	**Water** (%)	**Coarse aggregates** (%)	**Fine aggregates** (%)
GC3	3,22	21,49	4,94	7,75	41,01	21,59
GC6	4,34	21,15	3,49	9,42	40,35	21,25

3.3.5 Geopolymer concrete with carbon-capturing admixture

The effects of three carbon-capturing admixtures on concrete were studied. These admixtures were used as partial replacements of the cementing materials (calcium silicate, metakaolin, and sodium hydroxide) by weight. The mix design for GC3 was repeated by replacing 2,5 % of the cementitious material weight with each admixture. For ease of reference, the mixes containing titanium dioxide, zinc oxide, and biochar were labelled GC3TD, GC3ZO, and GC3Bc, respectively. The three mix designs are listed in Table 5.

Table 5. Mix designs of GC3 geopolymer with admixtures

Component	**GC3TD**	**GC3ZO**	**GC3BC**
Calcium silicate (kg/m^3)	79,56	79,56	79,56
Metakaolin (kg/m3)	530,40	530,40	530,40
Sodium hydroxide (kg/m^3)	121,99	121,99	121,99
Water with hydroxide (kg/m^3)	159,12	159,12	159,12
Extra Water (kg/m^3)	36,72	36,72	36,72
Coarse aggregates (kg/m^3)	1038,26	1038,26	1038,26
Fine aggregates (kg/m^3)	546,21	546,21	546,21
Admixture (kg/m^3)	18,77	18,77	18,77

3.4 Experimental testing

3.4.1 Nomenclature of concrete mixes

All the OPC and geopolymer concrete mixes are listed in Table 6, along with their nomenclature.

Table 6. Concrete mix designations

Designation	Description
Control	Ordinary Portland cement concrete control mix
Control TD	Control mix with titanium dioxide
Control ZO	Control mix with zinc oxide
Control BC	Control Mix with Titanium Biochar
GGBS 40 %	OPC mix with 40 % GGBS by weight of cement
GGBS 50 %	OPC mix with 50 % GGBS by weight of cement
GGBS 60 %	OPC mix with 60 % GGBS by weight of cement
GC3 air	Geopolymer concrete mix 3, air cured
GC3 oven	Geopolymer concrete mix 3, oven cured
GC3CO_2	Geopolymer concrete mix 3 with carbon dioxide injection
GC3TD	Geopolymer concrete mix 3, including titanium dioxide
GC3ZO	Geopolymer concrete mix 3, including zinc oxide
GC3BC	Geopolymer concrete mix 3, including biochar
GC6 air	Geopolymer concrete mix 6, air cured
GC6 oven	Geopolymer concrete mix 6, oven cured
GC6CO_2	Geopolymer concrete mix 6 with carbon dioxide injection

3.4.2 Fresh concrete tests

- Unit weight: This test was performed to obtain the unit weights of the three mixtures according to the ASTM C138 standard.
- Slump: Slump tests were performed according to ASTM C143 to test the workabilities of the different mixes.
- Air content: The air content per mix was tested in accordance with ASTM C231.

3.4.3 Compressive-strength test

The compressive strengths of the 16 concrete mixes were evaluated according to the guidelines outlined in the BS1881 standard. For each mix, three 15 × 15 × 15 cm cubes were cast and subjected to compressive-strength testing at specific time intervals using a universal testing machine until failure occurred. For the Portland cement concrete samples, testing was performed after curing for 3, 7, and 28 days. The GGBS samples were tested after curing for 7, 14, and 28 days. The samples with the titanium dioxide, zinc oxide, and biochar admixtures were tested after curing for 28 days. All of the Portland cement concrete samples were cured in a moist room until tested. The geopolymer concrete samples were tested after curing for 1, 7, and 28 days. The samples were either air cured or oven cured for one day at 60 °C according to the experimental plan. The geopolymer samples with carbon dioxide and admixtures were tested after curing for 28 days. The samples were tested after curing for different lengths of time because of the anticipated strength-development differences between the different mixtures.

3.4.4 Other tests

To assess the durability of the selected mixtures, the samples were tested for abrasion, chemical durability, and permeability. In addition, to study the effectiveness of the carbon-capturing admixtures, mixes that included these admixtures together with the control mix were tested for carbon absorption.

4 Results and discussion

4.1 Fresh concrete tests

4.1.1 Slump tests

The geopolymer mixes posed challenges related to the workability, with an average slump of 2 cm, indicating very low workability. To enhance the workability, mixes were prepared with the addition of a superplasticiser following recommendations from the literature, specifically at a dosage of 1 % of the cementitious material weight. However, the inclusion of a superplasticiser did not yield the desired improvement in workability; moreover, a slight decrease in strength was observed. Considering these findings, an investigation of alternative percentages of superplasticisers is recommended to identify the optimal dosage that balances the workability and strength.

4.1.2 Unit-weight test

The unit-weight test was performed on the two geopolymer mixes and control mix. The results showed that mix GC3 had a unit weight of 2172 kg/m^3, mix GC6 had a unit weight of 2271 kg/m^3, and the control mix had a unit weight of 2400 kg/m^3. Thus, the unit weights of the geopolymer concrete mixes were less than that of the Portland cement mix.

4.2 Compressive-strength testing

4.2.1 Control mix

The control mix was tested after curing for 3, 7, and 28 days. The control mixture achieved the desired 30 MPa threshold. The achieved strengths were 24,5, 27,8 and 30,2 MPa after curing for 3, 7, and 28 days, respectively.

4.2.2 GGBS as a partial replacement

GGBS was used to partially replace the OPC. Three mixes were prepared with 40, 50, and 60 % cement replacements by weight. The compressive strengths were recorded for each mix at 7, 14, and 28 days, and the results are presented in Figure 2. The 40 % replacement achieved the highest compressive strength of 20,24 MPa, which represents a more than 30 % reduction in concrete strength compared with the control mix. Because this was a partial replacement of cement, the reduction in the carbon dioxide cost of the mix was not significant. The combination of a significant reduction in strength and insignificant environmental benefits leads to the conclusion that the use of these mixes is not recommended.

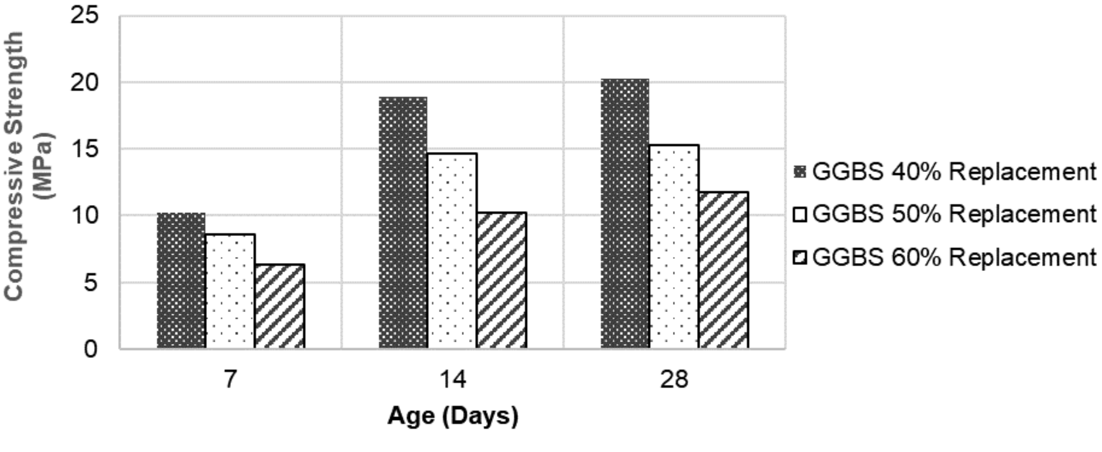

Figure 2. GGBS compressive-strength results

4.2.3 Control mix with carbon-capturing admixtures

The control mix was combined with each of the three carbon-capture admixtures. The three carbon-capturing admixtures used were ZnO, titanium dioxide, and biochar. The compressive strengths of the mixes were tested, and the results are shown in Figure 3. The addition of 2,5 % cement to each admixture significantly increased the compressive strengths of the samples. The increases in compressive strength ranged from 50-65 % depending on the type of admixture used.

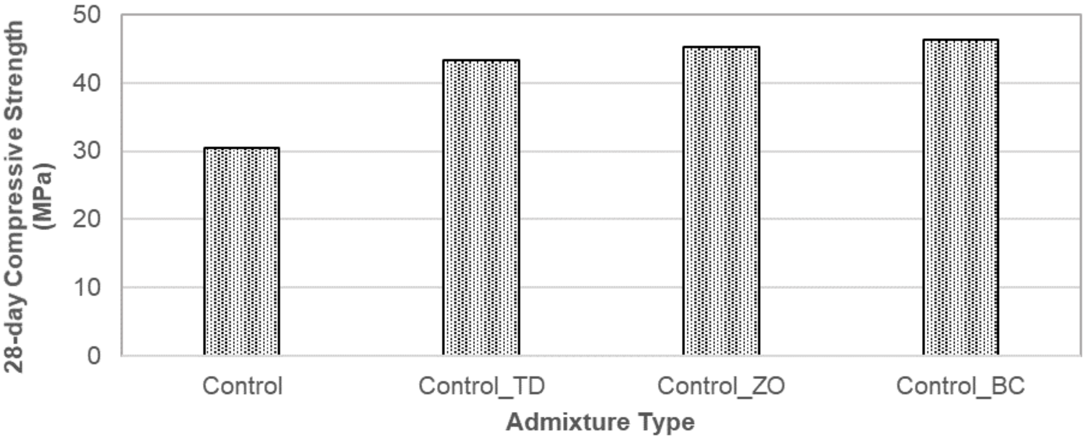

Figure 3. Compressive strengths of control mix and admixtures

4.2.4 Geopolymer paste

The compressive strengths of the geopolymer pastes are listed in Table 3 and shown in Figure 4. Mix GP9 is not shown in the figure because it showed no cohesion as a result of its lack of calcium silicate. This proves that calcium silicate plays an effective role in building the geopolymer bonds and increasing its strength. Oven curing had a positive effect on the strengths of the mixes but not for extended periods. Mixes GP3 and GP6 showed the best and most consistent results; hence, they were selected for use in the development of the geopolymer concrete.

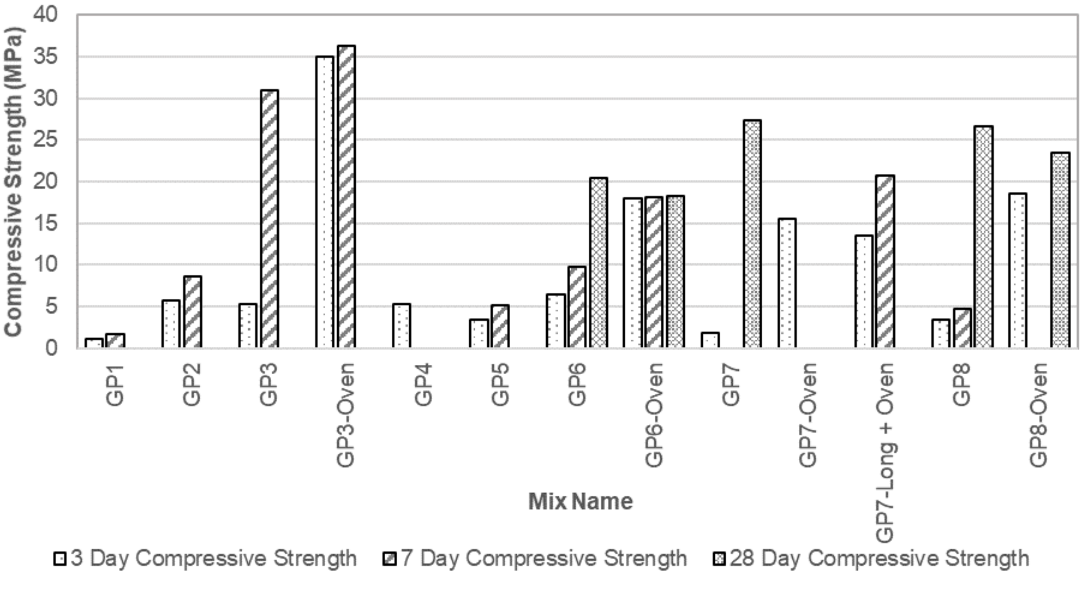

Figure 4. Compressive strengths of geopolymer pastes

4.2.5 *Geopolymer concrete*

Mixes GC3 and GC6 were poured and their compressive strengths were recorded. Each mixture was cured using two methods: air and oven curing. Their compressive strengths, along with those of the control mix, are shown in Figure 5. The inconsistent results for the oven-cured samples of mix GC3 were found to be a result of a problem with the aggregates used in the batch that was tested at 7 and 28 days. Although the maximum strength values of 25,03 and 18,22 MPa were not higher than that of the control mix, they showed that a high compressive strength is attainable.

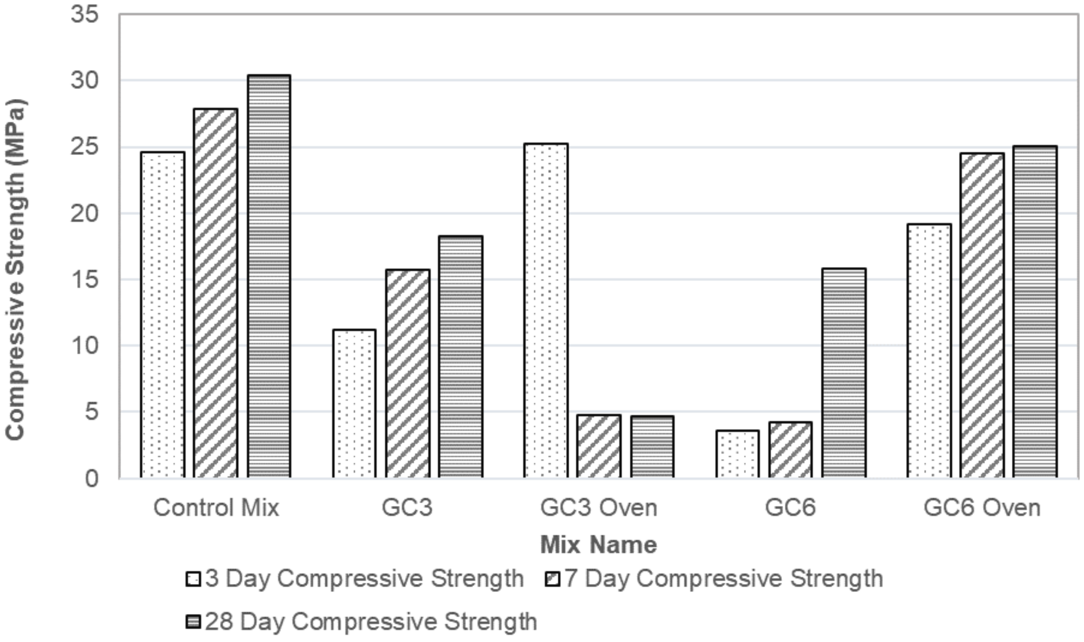

Figure 5. Compressive-strength results for geopolymer concrete

4.2.6 *Geopolymer concrete with carbon dioxide injection*

After carbon dioxide was injected into mixes GC3 and GC6, as outlined in [61], they had no cohesion or strength. The samples that did not fail in the mould exhibited a low strength of 3 kN.

4.2.7 *Geopolymer concrete with carbonated water*

Considering that the problem with the previous method was due to the injection of carbon dioxide as a gas, the use of carbonated water was tested. The same two mixes (GC3 and GC6) were prepared, replacing the normal water with carbonated water as a means of carbon-dioxide sequestration. Both mixes failed inside the mould, and none of the samples showed cohesion.

4.2.8 *Geopolymer concrete with carbon-capturing admixtures*

The compressive strengths of geopolymer concrete mixes containing carbon-capturing admixtures (GC3TD, GC3ZO, and GC3BC) are shown in Figure 6. Each of the admixtures (zinc oxide, titanium dioxide, and biochar) was added to the control mix and their compressive strengths were compared to that of the original calcium silicate geopolymer concrete, with mixed results. The zinc-oxide mix showed a huge decrease in compressive strength, whereas the titanium-dioxide and biochar mixes showed increases in compressive strength, reaching values of 31,7 and 35,9 MPa, respectively. Calcium silicate geopolymer concrete has a higher compressive strength than OPC concrete and absorbs carbon dioxide, which was experimentally proven in the following tests.

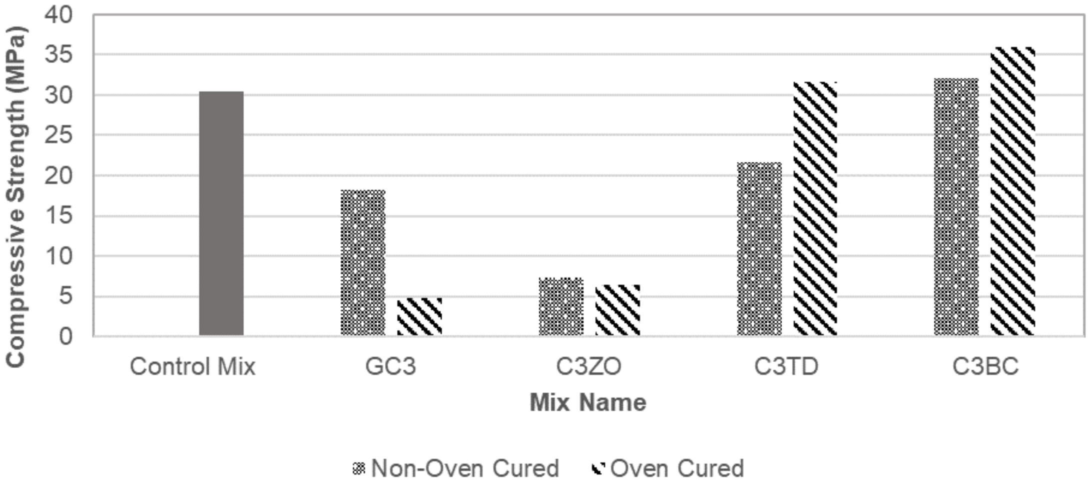

Figure 6. Compressive-strength results for geopolymer concrete with admixtures

4.3 Carbon absorption

The main reason for using the carbon-capturing admixtures was to provide the calcium silicate geopolymer concrete with carbon-capturing properties. Carbon-absorption tests were performed in a control chamber containing no sample and containing samples of each of the calcium silicate geopolymer concrete mixes with the admixtures. Two 150 × 150 × 150 mm cubes were placed inside the sealed chamber. Carbon dioxide was pumped into the chamber, and the carbon-dioxide percentage was measured using a KANE EGA-4 gas exhaust analyser at 1 h intervals over a test period of 7 h to test the rate of carbon-dioxide absorption. As shown in Figure 7, the carbon-dioxide percentage continued to decrease in all cases, yet it decreased to a greater extent when a sample other than the control sample was inside the chamber. The graph shows the behaviours of all the mixes incorporating the carbon-capturing admixtures compared with that of the control. These graphs prove that the calcium silicate geopolymer concrete obtained carbon-capturing properties from each of the three admixtures. Titanium dioxide exhibited the best carbon-capturing behaviour, followed by zinc oxide.

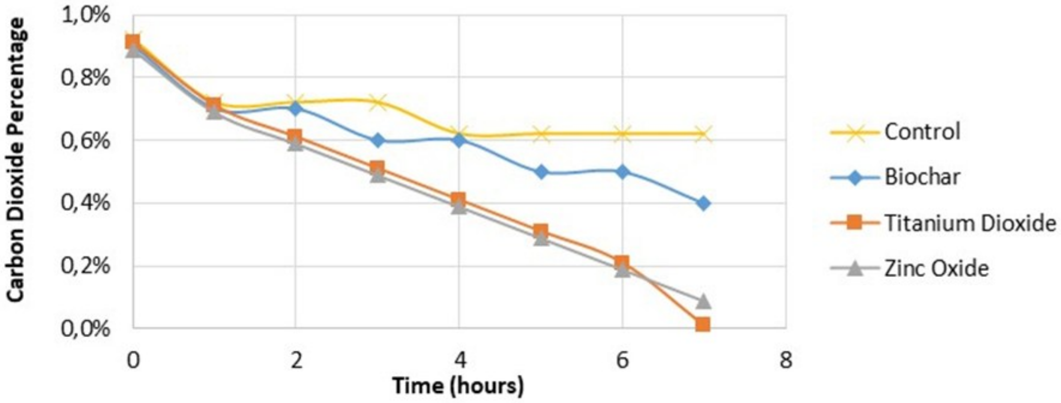

Figure 7. Carbon-absorption results

4.3.1 Carbon-dioxide emissions for calcium silicate geopolymer

The carbon-dioxide emissions were calculated based on the materials used for the calcium silicate geopolymer. The calcination of calcium silicate involves the production of carbon dioxide, with 700 kg of carbon dioxide generated per ton of calcium oxide. The calcium-oxide content of the calcium silicate was 12,57 %. Consequently, the carbon-dioxide emissions were quantified as 88 kg/ton of calcium silicate. Notably, calcium silicate constituted 11 % of the geopolymer mix. As a result, the overall carbon-dioxide emissions attributable to the calcination of calcium silicate in the geopolymer production process amounted to 9,68 kg per ton of geopolymer. However, the dihydroxylation of kaolin and dilution with sodium hydroxide produced zero carbon-dioxide emissions.

Furthermore, the carbon-dioxide emissions were calculated based on the electricity required for the use of ovens. Calcium silicate plays a significant role in the production of geopolymers and emits 190 kg of carbon dioxide per ton when heated. This compound constituted 11 % of the geopolymer mix. Additionally, in the geopolymer mix, kaolin contributed to the carbon footprint with an emission factor of 0,14 tons of CO_2 per ton of metakaolin. Metakaolin, which is a derivative of kaolin, accounted for 72 % of the geopolymer mix. Consequently, the carbon-dioxide emissions associated with the geopolymer production process amounted to 20,9 kg per ton when calcium silicate was used and 100,8 kg per ton when metakaolin was used. The dilution with NaOH produced zero carbon-dioxide emissions.

Figure 8 shows the difference between the total carbon-dioxide emissions for the calcium silicate geopolymer binder and Portland cement. The geopolymer carbon-dioxide emissions were as high as 13 % of its weight compared to 90 % for Portland cement.

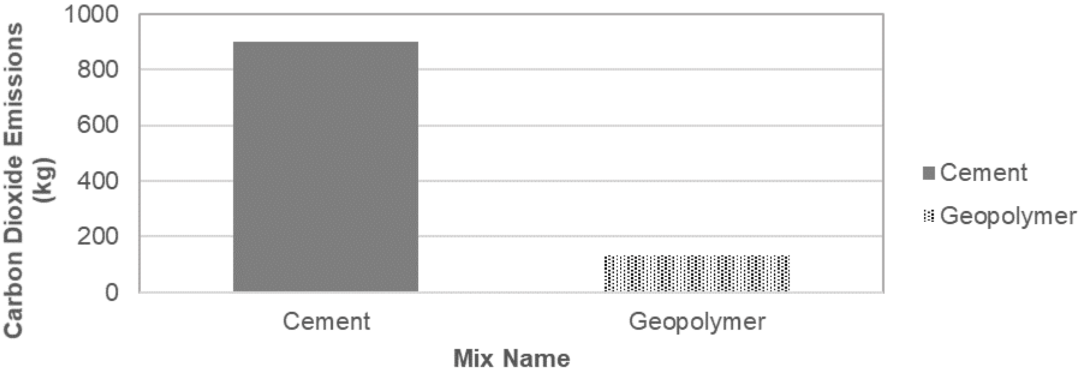

Figure 8. Carbon-dioxide emissions per ton of concrete

4.4 Durability testing

To ensure that the geopolymer concrete had sufficient durability, the following tests were performed to check its durability. These tests included abrasion, permeability, and chemical-durability tests.

4.4.1 Abrasion test

The results of the abrasion test, presented in Table 7, show that geopolymer concrete mix GC3 performed significantly better than the control mix. This result can be observed in the changes in both the mass and dimensions. This proves that not only were the geopolymer mixes better from the perspectives of strength and sustainability, but they were also better in terms of abrasion resistance.

Table 7. Abrasion-test results

Cube #	**Mass reduction** (g)	**Dimension Reduction** (mm)
1	11,5	2,0
2	19,6	2,3
3	11,6	2,0
Average	14,2	2,1
Control Mix	32,2	2,5

4.4.2 Permeability test

The results of the permeability tests showed no water penetration in the geopolymer concrete for either the GC3 or GC6 mix, but a penetration of 2,4 cm for the control mix. These results showed that the geopolymer concrete mixes had full resistance to water penetration.

4.4.3 Chemical-durability test

The test results for the geopolymer concrete against sulfuric acid compared with that for the control mix are presented in Figure 9. The figure shows decrease in the mass and volume percentages. These results confirmed that the calcium silicate geopolymer concrete (GC3) had a higher resistance to sulfuric acid and chemical attacks than OPC concrete.

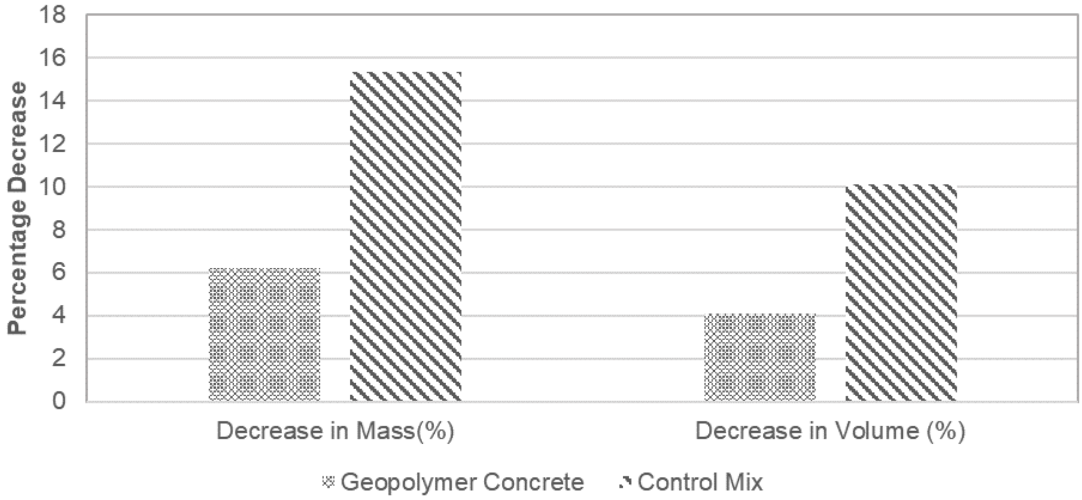

Figure 9. Durability-test results

5 Conclusions and recommendations

Based on the scope of this study and the materials and parameters involved, the following conclusions were drawn:

- This study provided compelling evidence that calcium-silicate geopolymers offer a highly promising route towards achieving net-zero-carbon concrete. With 9 0% CO_2 emissions from conventional concrete compared to a mere 13 % for calcium silicate geopolymers, the environmental benefits are undeniable.
- Calcium-silicate geopolymers not only have excellent sustainability, but also surpass the properties of OPC concrete in several key areas. When enhanced by titanium dioxide and biochar, the compressive strengths of calcium-silicate concrete reached 32 and 36 MPa respectively, exceeding that of OPC concrete. In addition to its strength, the geopolymer concrete mix containing titanium dioxide showed a very high level of carbon-dioxide absorption from the surrounding environment. The zero permeability of calcium-silicate geopolymers compared to the 2,4 cm permeability of OPC concrete highlighted their superior resistance to water penetration, which translated to greater durability over time. This enhanced robustness extended to chemical resistance, with the calcium-silicate geopolymers exhibiting significantly lower mass losses when exposed to sulfuric acid than conventional concrete. The distinctive hardening behaviour of calcium-silicate geopolymers deserves further investigation, because they have a higher hardening rate, reaching their maximum compressive strength in less time than conventional concrete. This rate could be further enhanced by oven curing, offering opportunities for rapid construction. However, additional research is required to address the inconsistencies in compressive-strength development.

- Although carbon-dioxide sequestration during the mixing process did not work when using direct injection or carbonated water, mixes with carbon-capturing admixtures should have good potential. The use of GGBS as a cement replacement at high dosages had a substantial negative effect on the strength of the concrete, outweighing any environmental benefit.

This study demonstrated the remarkable potential of calcium-silicate geopolymers for revolutionising the construction industry by offering a sustainable and superior alternative to conventional concrete. Further research to refine and optimise this technology will pave the way for a greener future and significantly reduce the environmental footprint of the construction sector. As steps on the way, the following are some recommendations for future work:

- Analyses of the tensile strengths and shrinkage tendencies of geopolymers.
- Analyses of the effects of the curing temperature and duration (only attempted at 60 °C for 24 h).
- Analyses of the effects of the absorption and chemical composition of aggregates on geopolymers.
- Long-term analyses of the carbon-capturing potentials of admixtures.
- Analyses of the bonding strengths between geopolymers and steel.
- Analyses of the long-term compressive strengths of geopolymers (after 28 days).
- Determination of superplasticisers that are compatible with geopolymers.
- Determination of the potentials of other mix designs with the guidance of a ternary plot.
- Study of calcium-silicate activation using the CO approach should be conducted with a high CaO content to achieve the required results.
- GGBS approach should be reconsidered with enhancing admixtures.
- Investigation of the carbon-capturing properties of kaolin after acid treatment.

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