International Journal of Recent Technology and Engineering (IJRTE)
ISSN: 2277-3878 (Online), Volume-14 Issue-3, September 2025

Mechanical Properties of Hardened Geopolymer Concrete Mixes Including Nano Particles: A Comprehensive Review

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***Abstract:** The increase in global warming due to CO_2 emissions from cement production is a critical problem, considering climate change these days. The alternative binder, instead of cement in the production of concrete, is to find a material that has the same chemical properties, is cost-effective, and is more sustainable than ordinary Portland cement, to mitigate the harmful CO_2 impact on the environment and human beings. The primary objective of this research is to investigate the mechanical properties of various sustainable precursor-based geopolymer concrete (GPC) mixes, including those incorporating nanoparticles, in comparison to GPC without nanoparticles. This study aims to demonstrate a significant enhancement in the mechanical properties and durability of GPC, based on research conducted in this field. Different precursors were used in the production of GPC, including fly ash, metakaolin (MK), ground granulated blast furnace slag, and silica fume, which are the primary sources of alumina and silica. The nanoparticles used in GPC mixes are silica nanoparticles, carbon nanotubes, clay nanoparticles, alumina nanoparticles, and graphene oxide nanoparticles, each separately, to enhance the mechanical properties in different precursor-based GPC mixes. Moreover, the mechanical properties of hardened GPC, including nanoparticles, will provide compressive strength, tensile strength, and splitting tensile strength, among others. An optimum percentage of 0.35% nano graphene oxide and 2% carbon nanotubes, separately added to FA-based GPC, enhances the mechanical properties of GPC. The maximum limit for nano alumina is up to 3% for FA-based GPC, after which the mechanical strength will decline significantly. Furthermore, the maximum limit of carbon nanotubes is 2% in FA-based GPC, and then the strength will be reduced.*

***Keywords:** Nano Silica, Carbon Nanotube, Nano Graphene Oxide, Nano Alumina, Geopolymer Concrete.*

Nomenclature:
AAs: Alkali Activators
CNT: Carbon Nanotubes
FA: Fly Ash
GPC: Geopolymer Concrete
GGBFS: Ground Granulated Blast Furnace Slag
GO: Graphene Oxide
MK: Metakaolin
NA: Nano Alumina
NC: Nano Clay
NS: Nano Silica
NT: Nano Titanium
OPC: Ordinary Portland Cement
RC: Reinforced Concrete
ASz: Alkali Activators
RGA: Recycled Glass Aggregates
MOE: Modulus of Elasticity

Manuscript received on 01 September 2025 | Revised Manuscript received on 09 September 2025 | Manuscript Accepted on 15 September 2025 | Manuscript published on 30 September 2025.

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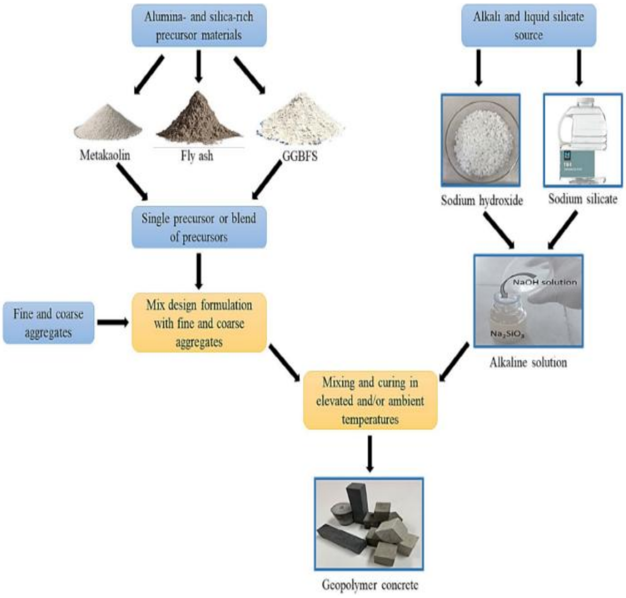
I. INTRODUCTION

The impact of climate change on the probability of corrosion initiation due to CO_2 or chloride ion penetration into concrete microstructure has significantly increased the risk of corrosion to the vital reinforced concrete (RC) structure members, especially bridges, based on the results obtained by Hassan et al. [1], Hassan and Amleh, [2], and Hassan et al. [3]. Additionally, the presence of cracks in the RC structures reduces their service life and durability according to Hassan and Amleh [4], Hassaan et al. [5], Hassaan et al. [6], and Hassaan et al. [7]. Davidovits, who is the scientist for geopolymer, invented the term 'geopolymer' to identify the unique artificial aluminosilicates with similar microstructures to the zeolites, a material widely found in nature [8], to limit the corrosion of RC structures.

The process of polymerization occurs in silicate/aluminate-rich precursor materials in the presence of alkaline solutions. The alkaline solutions used in geopolymer concrete (GPC) are sodium hydroxide, potassium hydroxide, and soluble silicates, which are suitable under curing environments. Moreover, three-dimensional microstructures on the backbone of Al and Si ions are generated (see Fig. 1). The objective of the precursor material is to provide SiO_2 and Al_2O_3, while the alkali activator's role is to break the Si–O and Al-O bonds and accelerate the formation of the tetrahedral framework in the geopolymer products.

GPC is rich in alumina and silica, and exhibits characteristics similar to those of cement. Geopolymer concrete exhibits high durability compared to ordinary Portland cement (OPC) and possesses excellent properties in both acidic and saline environments. Moreover, GPC produces low-carbon footprint concrete. The advantages of GPC include the use of sustainable construction materials, resistance to elevated temperatures, high performance in acidic and sulfate-rich environments, low maintenance costs, and low porosity. All these aspects make the GPC a prospective candidate for use in the field of civil engineering and retrofitting of existing buildings. GPC is used in various civil engineering RC structures. GPC shows improved strength and durability, and is environmentally friendly; however, GPC has some cons.



[Fig.1: Typical Process for Producing GPC Mix [9]]

The primary sources of materials used in the GPC are fly ash, ground granulated blast furnace slag (GGBFS), MK, and silica fume, which are rich in silica and alumina oxides. The sources of aluminosilicate (AS) are FA, GGBFS, MK, and Rice husk ash for producing GPC. Moreover, the use of additive sources, such as Calcium Aluminate Cement, Nano-Silica, Ca(OH)2, OPC, and SF, was also utilised in making GPC. The binding characteristics of a geopolymer depend on generating a 3D amorphous aluminosilicate network. The microstructures and properties of various precursor-based GPC mixes are defined below:

A. **Fly Ash (FA):** FA contains alumina and silica as key constituents that help in the formation of cement as a substitute. FA concrete has high strength and is more durable than traditional concrete. Low calcium FA, such as class F FA-based GPC, produces a dense concrete microstructure with better mechanical strength. Additionally, the microstructure of FA-based geopolymers offers high resistance to severe environments (e.g., acid, sulfate, and chloride) and fire. FA-based geopolymers have a lower environmental impact than MK-based GPC.

B. **Metakaolin (MK):** MK is generated by the calcination of kaolin clay at temperatures ranging between 500°C and 800°C. The resulting MK has a high content of silica and alumina, which makes it suitable for geopolymerization. MK-based geopolymers are characterized by high compressive strength and good durability.

C. **Ground Granulated Blast Furnace Slag (GGBFS):** The chemical properties of GGBFS contain a high amount of calcium oxide. The addition of GGBS to GPC will result in the formation of calcium aluminate hydrate (C-A-H) and other products. Moreover, the presence of GGBFS in GPC enhances the geopolymerization process, resulting in the rapid development of early strength in alkali-activated cement.

D. **Alkali Activators (AAs):** AAs are produced from a combination of alkali hydroxides with alkali silicates. This combination of alkali activators initiates the geopolymerization reaction and facilitates the formation of the polymeric gel matrix.

The primary concern for the current research is that the production of concrete accounts for approximately 30% of global CO2 emissions. Cement is a high-emitting product in terms of greenhouse gases. The cement industry, as one unit, generates high emissions of CO_2.

Various methods can reduce CO2 emissions:

(1) Replacement of cement with secondary cementitious materials that have a low impact on the environment,

(2) Use of environmentally friendly fuel in cement production,

GPC is one of the sustainable means to reduce CO_2 emissions during the utilization of cement in the construction industry. Table I presents the differences between the OPC and GPC in terms of mechanical properties and durability characteristics for both.

Table I: Comparison Between Geopolymer and Ordinary Portland Cement Concrete Properties

Properties	OPC	GPC
Compressive Strength	Lower	Higher
Tensile Strength	Lower	Higher
Durability	Lower	Higher
Acid Attack	Lower Resistance	Higher Resistance
Fire Resistance	Limited	Higher Resistance
Freeze and thaw cycling	More sensitive	Less sensitive
CO_2 Emission	High	Low

GPC offers improved strength and durability for its microstructure according to Singh and Middendorf [10], Kakria et al. [11], Shah et al. [12], Amran et al. [13], Xu, and Shi [14]. GPC exhibits superior resistance to high temperatures compared to OPC concrete, which can withstand temperatures ranging from 1000°C to 1200°C. It was deduced that when the GPC elements are subjected to maximum temperatures ranging from 150°C to 350°C, the residual compressive strength increases significantly; however, the compressive strength of RC sections made of GPC decreases at a temperature of 400°C or above, according to Manzoor et al. [15]. Enhancements in GPC properties to resist high temperatures can be achieved by incorporating materials such as fibres, waste glass, and rubber particles [16]. The utilization of a GPC member composed of 50% FA and 50% GGBFS significantly inhibited the probability of chloride-induced corrosion initiation compared to that of a standard concrete member subjected to various maximum temperatures ranging from 25°C to 45°C [17]. Carbon fibres are the most used fibres in GPC due to their high tensile modulus and thermal conductivity [18]. GPC exhibits excellent resistance to chloride penetration within the concrete microstructure [19]. Oyebisi et al. [20] found that GGBFS-corncob ash-based GPC exhibited a lower environmental impact and higher sustainability compared to ordinary Portland cement concrete. These findings can lead to a cleaner environment achieved through sustainable construction materials. Shehata et al. [21] showed the contributions of GPC to sustainable development goals and the implementation of various wastes for the creation of a GPC-based circular economy. Optimising the elements for the GPC, especially the alkaline activators, would lead to a greener circular economy.

FA-based GPC is the most common type of concrete used in construction projects due to its low carbon emissions, reduced environmental impact from global warming, and extended service life of RC structures [22]. Özkılıç et al. [23] recommended using 10% glass aggregate with a sodium hydroxide molarity of 16 to achieve the optimum GPC mix at different stages. However, when utilizing recycled glass aggregates (RGA) in GPC with a ratio of 30% concerning the total amount of coarse aggregate, the compressive strength is decreased significantly by 21.6%, compared to the reference sample, including 0% of RGA, according to Özkılıç et al. [23]. Çelik et al. [24] deduced that combining an optimum percentage of micro silica and lathe scraps in a GPC mix will improve the compressive, flexural, and splitting tensile strengths of GPC, compared to the reference sample (see Table II). The potential of GPC mixes in various engineering applications highlights their relevance in modern sustainable construction materials.

Table II: Mechanical Properties of GPCs, Including Various Percentages of Micro Silica and Lathe Scraps Replaced with the Amount of FA in GPC

Percentages of Micro Silica and Lathe Scrap Utilized in GPC Mixes	Mechanical Properties of GPC		
	Compressive Strength	Flexural Strength	Splitting Tensile Strength
5% Micro silica	↑ 14.4%	↑ 7.45%	↑ 6.18%
20% Micro silica	↓ 17%	↓ 35%	↓ 25%
1% lathe scraps	↑ 11.4%	↑ 6.35%	↑ 8.23%
3% lathe scraps	↓ 12%	↓ 11%	↓ 10%
Combining 1% lathe scraps and 5% micro silica	↑ 25.7%	↑ 14.4%	↑ 12%

The maximum compressive strength, at the age of 28 days, is produced by the GGBFS-based GPC with 3% dosage of superplasticizer according to Gupta et al. [25]. Ramujeea and PothaRajub [26] found that the GPC reached its target strength much faster under heat-cured conditions than under ambient conditions. Moreover, the relationship between split tensile strength and compressive strength of GPC can be expressed using a regression model analysis that resembles that given by ACI-318-99 for OPC concrete. The increase in geopolymer recycled aggregate concrete (GPRAC) content in GPC will lead to an increase in flexural strength according to Zhang et al. [27]. Moreover, the porosity of GPRAC decreased after the addition of nano-silica and rice husk ash.

The modulus of elasticity for ordinary Portland cement concrete is investigated using (1). A few researchers studied the modulus of elasticity (MOE) for GPC mixes. The MOE generally ranges between 23 GPa and 31 GPa and between 11.2 GPa and 41.2 GPa, respectively. Geopolymers differ only in ductility compared to ordinary Portland cement.

$$E_C = 0.043\rho^{1.5}\sqrt{f_{cm}} \pm 20\% \quad \ldots \quad (1)$$

where: f_{cm}: average compressive strength (MPa) and ρ: density.

The equation used to calculate the MOE for GPC as a function of maximum temperature values is shown in Equation (2), based on the Australian Standard (AS 3600).

$$E_{GPC} = 0.9713 + 0.0009\,T - 5 \times 10^{-7} \times T^2 \qquad 100°\mathrm{C} \le T \le 1000°\mathrm{C} \quad \ldots \quad (2)$$

where: E_{GPC}: elasticity modulus of GPC at room temperature, and T: temperature (℃).

The constitutive stress-strain model of ordinary Portland cement concrete is shown in (3). However, the stress-strain relationship of GPC at elevated temperature is shown in (4) and (5) [28].

$$\frac{f_c}{f_{cc}} = \frac{n\left(\frac{\varepsilon_c}{\varepsilon_{cc}}\right)}{n - 1 + \left(\frac{\varepsilon_c}{\varepsilon_{cc}}\right)^n} \quad \ldots \quad (3)$$

$$\varepsilon'_{GPC} = \left(\frac{f'_{GPC}}{E_{GPC}}\right)\left(\frac{\varphi}{\varphi - 1}\right) \quad \ldots \quad (4)$$

$$\varphi = \left(\mathrm{f'}\frac{\mathrm{GPC}}{17}\right) + 0.8 \quad \ldots \quad (5)$$

Where: f_c, f_{cc}, ε_c, ε_{cc}: the stress at any point, the maximum stress, the strain, and the strain corresponding to the maximum stress, respectively; and n is the curve adjustment coefficient. ε'_{GPC}, f'_{GPC}, and $EGPC$ represent the strain, stress, and elastic modulus at elevated temperatures, respectively; φ is a fixed parameter.

This paper is mainly concerned with the mechanical properties of various precursor-based GPC mixes, including compressive strength, tensile strength, MOE, etc. In addition, the behaviour of nanoparticles in GPC mixes will be discussed to determine the optimum percentage of nano silica, carbon nanotubes, nano clay, nano alumina, and nano graphene oxide in enhancing the mechanical properties, including tensile strength, compressive strength, and splitting tensile strength, for different precursor-based GPC mixes.

II. BASIC MECHANICAL PROPERTIES OF GPC

A. Compressive Strength

The most common precursor materials used in GPC are FA, MK, and GGBFS. The raw materials of GPC exhibit significant differences in chemical composition due to their diverse origins. In most cases, raw geopolymer materials are typically rich in silicon and aluminium, with a low calcium content. The content of GGBFS is generally controlled between 20% and 50%. When the calcium ion content reaches a specific value, the calcium ions undergo a hydration reaction, resulting in a product that forms an inhibition effect in the geopolymer and reduces its properties. Researchers studied the impact of the proportion of FA and GGBFS on the GPC and found that when the two accounted for 50% of the mixed preparation, the highest compressive strength of GPC was obtained. Incorporating FA or GGBFS increases the compressive strength of GPC under high temperatures. GGBS is also used to produce GPC mixtures because of its high aluminosilicate content.

GGBS-based GPC improved the compressive strength of hardened GPC. In contrast, MK-based GPC suffers a decrease in compressive strength and the generation of several cracks at high temperatures. According to Meesala et al. [29], a GPC made of class F FA has high durability and strength.

B. Tensile Strength

The optimum dosage of FA and GGBFS had an impact on the physical and mechanical properties of GPC. The splitting tensile strength of GPC increases up to 400°C, and a downward trend is observed between 400°C and 800°C. The GPC achieved a higher tensile strength compared to ordinary Portland cement concrete [30]. GGBFS-based GPC has a significant impact on tensile strength. The tensile strength of cleavage was linearly positively correlated with the mass ratio of the alkali solution. The value of the tensile strength can be estimated by its relationship with compressive strength. Many researchers have proposed a corresponding empirical formula as shown in (6) to (9) [30].

$$f_t = (f'c)^n \quad \ldots \quad (6)$$

$$f_t = 0.858 \times (f'c)^{0.41} \quad \ldots \quad (7)$$

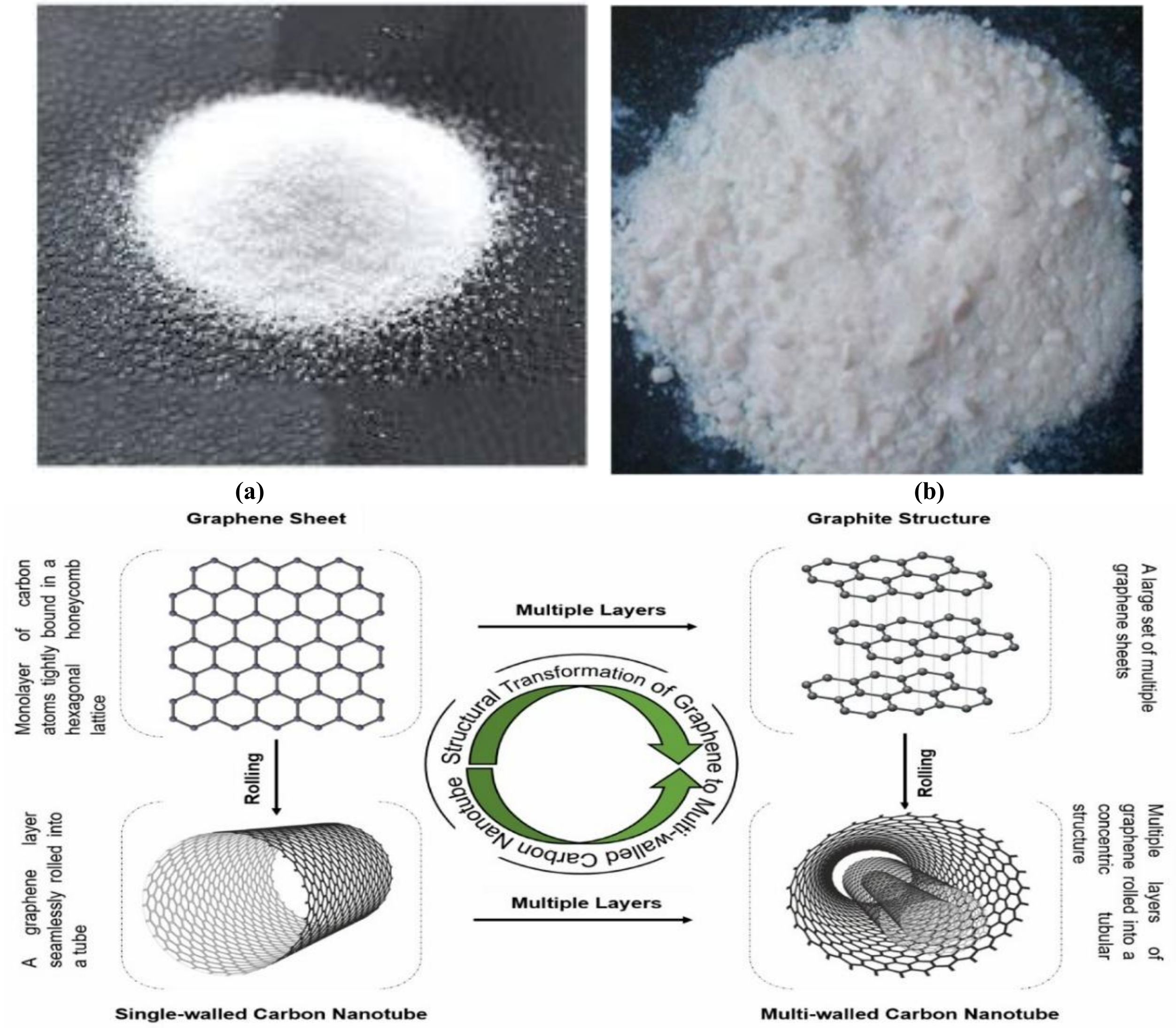
$$f_t = 0.70 \times (f'c)^{\frac{1}{2}} \quad \ldots \quad (8)$$

$$f_t = 0.40 \times (f'c)^{\frac{7}{9}} \quad \ldots \quad (9)$$

Where: f_t: splitting tensile strength of concrete (MPa), f'_c: compressive strength of concrete (MPa).

III. ANALYSIS OF HARDENED GPC WITH VARIOUS NANOPARTICLES

Nanoparticles are one of the most valuable forms of nanomaterial used as an additive in GPC, but nanomaterials like carbon nanotubes (CNT) were used as nanomaterials in fibre forms [31]. The most useful nanoparticles in GPC are nano silica (NS), nano alumina (NA), and nano titanium (NT) [32], and nanomaterials like nanographene oxide (GO) are available in nano-flake forms according to Szczepanik [33], Ismael et al. [34], Braganca et al. [35] (see Fig.2).



(a)

(b)

(c)

[Fig.2: Different Nanoparticles in GPC. (a) NS, (b) NA, (c) CNTs [36]]

A. Nano Silica (NS)

NS is available in nanoparticle form, with approximately 98% silica content. Naskar et al. [37] found that the impact of the addition of NS with a tiny percentage, approximately equal to 0.75%, 75–80% strength is achieved only in early ages of curing. The effect of the incorporation of NS is shown in Table III.

Table III: Effect of Incorporation of NS in GPC

NS in GPC
The addition of 3% optimum content nano silica significantly improves the compressive strength of GPC [38].
Incorporating 2% NS into class C FA-based GPC increases flexural strength by 82% and compressive strength by 22%. However, tensile strength is decreased by 31% by the incorporation of the same amount of NS [39].
Adding 1% NS to the GPC initial and final setting time reduces them by 31% and 82% respectively.
Porosity is decreased by 8% and flexural strength is increased by 24% after the addition of 1% NS to MK-based GPC.
Compressive strength decreased by 32% after adding 10% NS to FA-based GPC.
The compressive strength increases by 7% after the addition of 1.5% NS to GGBFS-based GPC.

B. Carbon Nanotubes (CNT)

CNTs are fibre forms of nanomaterials that have their benefits for improving the bond strength of concrete. The tensile strength of the GPC mix increases when adding CNTs [40]. It is also observed that carbon nanotubes with multi-walled structures enhance the tensile properties of geopolymer paste [41]. CNT increases the mechanical and durable properties of GPC [42]. Multiwall CNT gives better mechanical properties than single-wall CNT due to the availability of more area for geopolymerization by adding the quantity of C–S–H gel [43]. The effect of the incorporation of CNTs in various precursor-based GPC mixes is shown in Table IV.

Table IV: Impact of The Incorporation of CNT in GPC

CNT in GPC
Compressive strength for MK-based GPC increased by 32%, and flexural strength increased by 66% after adding 0.5% CNT [40].
Compressive strength rises by 33% due to the addition of 2% CNT for FA-based GPC, but it starts decreasing after further addition of CNT [44].
In the case of FA-based GPC, good electrical resistivity and agglomeration are achieved at 0.5% and 1% CNT.
Mechanical properties were increased for MK-based GPC at 0.1% addition of CNT, but there was a reduction in strength after adding 0.2% of CNT [45].
Compressive strength and flexural strength are decreased after the addition of 1% CNT [46].

C. Nano Clay (NC)

NC is produced when kaolin is burned at a temperature of 800 °C [47]. It is used in many civil engineering products. The impact of adding a significant amount of NC to a certain amount increases the mechanical strength of GPC [48] (see Table V).

D. Nano Alumina (NA)

Nano Al_2O_3 is utilized as a nanofiller [52]. This type of nanomaterial can be used with low alumina sources [53]. The highest mechanical strength is achieved after incorporating 1% nano titanium and 1% nano alumina into the GPC mix [37]. The impact of NA incorporation into the GPC mixes is shown in Table VI.

Table VI: Effect of Incorporation of NA in GPC

NA in GPC
The highest mechanical properties are achieved with the optimum addition of 2% nano Al_2O_3 to FA-based GPC [54].
Adding 2% nano alumina to FA-based GPC produces significant mechanical strength. However, when 3% nano Al2O3 is added to FA-based GPC, it leads to a decrease in mechanical strength due to a less dense structure.
Flexural strength is increased by 35% after adding 2% nano alumina to FA-based GPC.
The compressive strength and flexural strength of GPC increased by 90% and 38%, respectively, when 0.5% nano titanium was added to GGBFS-based GPC.

Table V: Effect of Adding Nano Clay in GPC

Nano Clay in Geopolymer Concrete
The porosity for GPC-based FA is reduced by 98% after adding 3–5% NC [49].
Adding 6% NC for FA-based GPC improves flexural strength and tensile strength by 29% and 36%, respectively [50].
NC plays a vital role in increasing the activation reaction of GPC and making the structure dense [48].
The highest flexural modulus and mechanical strength are achieved after incorporating NC at a 2% concentration for FA-based GPC [51].
Compressive strength increases with an increase in the percentage of NC for GGBFS-based GPC.
Compressive strength and flexural strength increased by 23% and 24%, respectively, after the incorporation of NC by 2% especially for FA-based GPC [48].

E. Nano Graphene Oxide (Nano GO)

Graphene oxide is produced via the geopolymerization reaction between alkaline activators and nano GO. The mechanical properties of GPC increased after the incorporation of GO. A low quantity of GO achieves higher compressive strength than various nanomaterials GPC [55]. The compressive strength increased by 77% after the incorporation of 0.04% nano GO [56]. Many researchers observed that the mechanical strength of FA/GGBFS GPC is increased after the incorporation of graphene oxide [57]. The influence of the incorporation of nano GO across various precursor-based GPC mixtures is shown in Table VII.

Table VII: Influence of the Incorporation of Nano GO in GPC

Nano GO in GPC
Flexural strength and modulus of elasticity increased significantly by 134% and 367%, respectively, when adding 0.35% nano GO to FA-based GPC.
For FA-based GPC, including 1% of nano GO, the compressive and flexural strength increased by 144% and 216%, respectively.
The impact of nano GO helps in increasing the flexural strength of GPC by 61.5% after the addition of 0.35% nano GO to MK-based GPC [58].
Compressive strength MK-based GPC is increased with an increase in the percentage of GO up to 3% and then starts decreasing after the addition of more GO. Incorporation of nano GO up to 3% to MK-based GPC results in an increase in compressive strength up to 80% [59].
Compressive, tensile, and flexural strength of 50%FA and 50%GGBS-based GPC is increased by 18%, 61.9% and 60% after incorporation of 0.3% GO [60].

IV. RESULTS AND DISCUSSION

A. The impact of the proportion of 50% FA and 50% GGBFS-based GPC, the highest compressive strength of GPC was achieved.
B. Combining an optimum percentage of 1% lathe scraps and 5% micro silica in a GPC mix improves the compressive, flexural, and splitting

tensile strengths of GPC, compared to the reference sample [24].

C. The increase in geopolymer recycled aggregate concrete content in GPC will lead to an increase in flexural strength [27].
D. The mechanical behaviour of GPC is like that of OPC concrete [26].
E. GGBFS was more resistant to sulfuric acid attack. Moreover, increasing MK content in GPC improves its permeability resistance and compressive strength [27].
F. GPC has excellent resistance against fire.
G. GPC strength is affected by various factors that deal with uncertainty.
H. The addition of nano silica with a tiny percentage, approximately around 0.75% to the GPC mix, 75–80% strength being gained in early ages of curing [37].
I. Adding 2% carbon nanotubes for FA-based GPC significantly increases compressive strength by 33% [44].
J. Adding 0.5% carbon nanotubes, compressive strength for MK-based GPC is increased by 32%, and flexural strength is increased by 66% [40].
K. Adding 6% nano clay for FA-based GPC enhances the flexural strength and tensile strength by 29% and 36%, respectively [50].
L. An optimum percentage of 2% nano alumina to FA-based GPC produces significant mechanical strength.
M. The compressive and flexural strength of GGBFS-based GPC increased by 90% and 38% respectively, after contributing 0.5% nano titanium to the GGBFS-based GPC.
N. Flexural strength and modulus of elasticity increased sharply by 134% and 367%, respectively, when adding 0.35% nano graphene oxide to FA-based GPC.
O. The maximum limit to nano alumina is up to 3% to FA-based GPC, and then the mechanical strength will be reduced [54]. Furthermore, the maximum limit of carbon nanotubes is 2% in FA-based GPC, and then the strength will decrease [44].

DECLARATION STATEMENT

After aggregating input from all authors, I must verify the accuracy of the following information as the article's author.

- **Conflicts of Interest/ Competing Interests:** Based on my understanding, this article has no conflicts of interest.
- **Funding Support:** This article has not been funded by any organizations or agencies. This independence ensures that the research is conducted with objectivity and without any external influence.
- **Ethical Approval and Consent to Participate:** The content of this article does not necessitate ethical approval or consent to participate with supporting documentation.
- **Data Access Statement and Material Availability:** The adequate resources of this article are publicly accessible.
- **Author's Contributions:** The authorship of this article is contributed equally to all participating individuals.

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