

A DETAILED REVIEW OF THE CHEMISTRY OF GEOPOLYMER MATERIALS: SPECIAL FOCUS ON CIVIL ENGINEERING APPLICATION

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ABSTRACT

Geopolymer is an innovative and eco-friendly inorganic building material that is becoming increasingly popular as a sustainable alternative material for Portland cement concrete in the construction sector. Unlike Ordinary Portland Cement (OPC), which primarily comprises limestone and clay extracted in large quantities from the earth, its production minimizes environmental harm. Geopolymer concrete, in contrast, is formed using industrial waste materials like fly ash and ground granulated blast furnace slag (GGBS), combined with appropriate aggregates and an activating solution. Geopolymer-based concrete offers an eco-friendly substitute for traditional concrete, helping reduce the environmental damage caused by cement production while utilizing industrial waste materials, improving durability, and offering significant energy savings. The physicochemical characteristics of Geopolymer concrete (GPC), such as heat of hydration, shrinkage, chemical resistance, and fire resistance, are found to be excellent compared to OPC-based concrete. Another important advantage of GPC is its ability to easily adsorb radioactive substances present in the environment. GPC is considered a sustainable alternative to OPC-based concrete, offering significant environmental benefits and contributing to a more sustainable future. This review article focuses on the methodology for preparing GPC, its main ingredients, and its properties. The prospects of geopolymer concrete are also discussed.

Keywords: Geopolymer Concrete (GPC), Environment Friendly, Construction, Physico-Chemical Properties, Sustainable Future

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INTRODUCTION

Portland cement is indeed the predominantly utilized product in the construction industry worldwide. It is the main ingredient in concrete, which is essential for the construction of buildings, infrastructure, roads, bridges, and many other civil engineering projects.¹ Portland cement can indeed cause severe damage to the skin and respiratory systems due to its chemical composition.² Portland cement particles, especially in extreme cases, can pose significant health risks due to the chemical configuration and physical nature of the material.³ OPC is primarily manufactured from limestone and clay-based minerals, which serve as the essential raw materials. Calcium carbonate (CaCO_3) is commonly found in nature as the mineral limestone. It is one of the primary key components used in the manufacture of OPC. Clay-based minerals, which are commonly used in the manufacture of Ordinary Portland Cement (OPC), consist of a variety of oxides, including MgO , Al_2O_3 , and SiO_2 , among others. OPC's construction activities worldwide have led to a significant extraction of its main raw materials—calcite (limestone) and clay minerals. This over-reliance on these natural resources poses challenges for their long-term availability and environmental sustainability.^{4,5} Alternatives to OPC are critical to reducing environmental harm and ensuring a sustainable future. By adopting innovative materials and methods, we can minimize the negative impacts on both humans and the environment while maintaining construction quality and durability. Geopolymers are emerging as a promising alternative to OPC worldwide due to their excellent durability and mechanical properties. These materials address the environmental challenges associated with OPC and

offer significant advantages in construction and other applications.⁶ Geopolymers have been reported to possess the ability to adsorb atmospheric CO₂, making them a potentially carbon-negative material in construction. This characteristic enhances their sustainability and sets them apart from conventional OPC, which is a major contributor to carbon emissions during its manufacturing process.⁷ This property adds to their sustainability benefits and distinguishes them from traditional OPC, which plays a major role in carbon emissions during its manufacturing process.⁷ Aluminosilicates and basic minerals are indeed the major ingredients of geopolymers. These materials play an important role in the formation of the geopolymer matrix and are responsible for its excellent mechanical and durable properties. Geopolymers are produced through the alkali activation of aluminosilicate materials.⁸ GPC uses industrial byproducts, like fly ash (sourced from thermal power plants) and GGBFS (obtained from iron manufacturing), as a complete alternative to cement in concrete.⁹ In thermal power plants, the ash produced from coal combustion is typically classified into fly ash (80%) and bottom ash (20%) based on its behavior in the furnace.¹⁰ Fly ash particles are considered major pollutants due to their fine size, chemical composition, and potential for dispersal into the air and water. Their management poses significant environmental challenges, especially in areas surrounding thermal power plants.¹¹ Fly ash particles generally contain a mix of major oxides, like, silicon, aluminum, magnesium, calcium, and iron oxides, and trace amounts of heavy metal oxides, like copper, cobalt, arsenic, and lead oxides, making them both versatile and potentially hazardous.¹² GGBFS primarily consists of aluminosilicates and calcium silicates, which are formed as waste material during the production of iron in a blast furnace.¹³ GGBFS, or its precursor, blast furnace slag, can have environmental impacts if not managed properly.¹⁴ GPC, developed using source materials like fly ash, GGBFS, and other aluminosilicate-rich industrial byproducts, exhibits superior characteristics compared to OPC-based concrete.¹⁵ Strength under compression of GPC is often reported to be greater than that of OPC concrete, especially after 28 days of curing. This is due to the unique chemical composition and the geopolymerization process, which results in the creation of a compact and durable microstructure.¹⁶ Research has indicated that the bending stiffness of GPC in saltwater conditions is 3–6% higher than that of OPC concrete under constant pressure.¹⁷ GPC generally exhibits greater bond strength than OPC concrete. The strength of the bond signifies the adhesion between the concrete matrix and reinforcing steel, or between layers of concrete. Higher bond strength in GPC is attributed to its dense microstructure and chemical properties.¹⁸ It was found that the manufacture of OPC is responsible for releasing approximately seven percent of global carbon dioxide (CO₂) emissions annually. This is indeed true, as the cement industry, particularly the manufacture of OPC, is a significant supplier of carbon-emitting gas emissions.¹⁹⁻²⁰ However, Geopolymer-based concrete can significantly decrease carbon-emitting gas emissions compared to OPC.²¹ Apart from the above, GPC is indeed highly versatile and can be more cost-effective than OPC in several respects, aside from its environmental benefits.²² Further, GPC has shown promise in absorbing and immobilizing radioactive materials, making it a potentially valuable material for environmental remediation, especially in the context of nuclear waste management and radioactive contamination.²³ Therefore, GPC plays a significant role in minimizing radioactive pollution, especially in the context of atomic power plants and other nuclear facilities.²⁴ GPC is gaining recognition as an eco-friendly substitute for conventional OPC concrete in terms of environmental sustainability. For the sustainability of the environment, GPC is reported to be an alternative to ordinary concrete in the present scenario. Hence, this review article focuses on the production of GPC using environmentally friendly techniques, its characteristics, advantages, and its application towards a sustainable future.

Manufacture of Geopolymer Concrete

The production of GPC requires careful control over the proportions of different ingredients to ensure the desired performance and sustainability of the material. The main constituents of GPC include GGBS, fly ash, fine aggregates, coarse aggregates, and an alkaline accelerator solution (also known as the alkaline activator solution). These components work together to form the geopolymer matrix that provides the necessary strength, durability, and sustainability of the concrete.²⁵⁻²⁶ The key ingredients of Geopolymer are (1) ingredients more in silicon and aluminum, such as fly ash and GGBS, and (2) a solution of alkaline activators. The basic accelerator solution is typically a mixture of K₂SiO₃ or Na₂SiO₃, along with KOH or NaOH dissolved in water.²⁷ The Geopolymer paste is formed through the chemical reaction between the alkaline activator (which typically consists of alkaline hydroxides and alkaline silicates) and

of SH solution ($M = 10$), SS/SH ratio of 2.0, and AAS content of 40%, as well as slag content at 35%, the same as in Group C. The impact of extra water on the compressive strength of GPC is presented in Fig.-2.

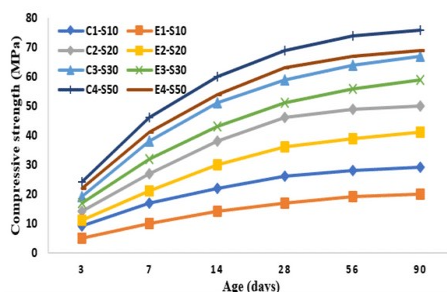


Fig.-2: Effect of Additional Water on Compression Strength of GPC mixes (Group C vs. Group E)³⁹

It was reported that GPC with 8 molar alkaline solutions, viz., NaOH and Na₂SiO₃, can significantly enhance compressive strength. The strength of the alkaline activator is crucial in the geopolymerization process, as it governs the dissolution of the precursor materials and the subsequent formation of the aluminosilicate gel, which is the binding phase important for imparting the mechanical properties to GPC.⁴⁰ Temperature curing significantly enhances the mechanical characteristics of GPC, including compression resistance, elastic modulus, and other key behaviors, compared to ambient curing. The increased strength and improved performance can be ascribed to the acceleration and improving the geopolymerization process at high temperatures.⁴¹ The elastic modulus of GPC is normally reported to be lower than that of OPC concrete at comparable compressive strengths (30–35 MPa). This difference arises due to variations in the binder composition and the resultant microstructure of the two types of concrete.⁴² Replacing slag with quarry rock dust (QRD) in GPC up to 15% enhances its stretchability, compressive strength, and bending strength. However, mechanical characteristics tend to decline beyond this percentage due to changes in the composition and microstructure of the concrete.⁴³ The Si/Al ratio (silicon to aluminum ratio) is a critical factor influencing the mechanical properties of GPC, particularly its compressive strength. The 1.5 to 2.0 range for the Si/Al ratio is considered ideal for achieving higher compressive strength. This range creates a balanced amount of silica and alumina in the geopolymer matrix, promoting strong chemical bonding, and a well-structured gel.⁴⁴ Nano-silica as a reinforcing material in GPC has been reported to considerably enhance the mechanical properties and the material's durability. Nano-silica, because of its tiny particle size and large surface area, can improve the structure of the geopolymer matrix at the nanoscale level, contributing to stronger bonds, reduced porosity, and enhanced performance under various environmental conditions. The split tensile strength of nano-silica (NS) incorporated GPC is presented in Fig.-3.

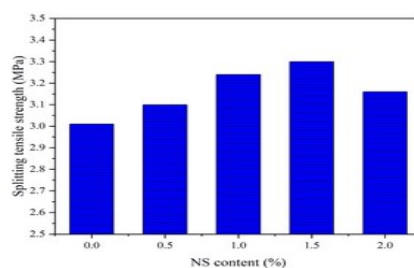


Fig.-3: Split Tensile Strength of NS-GPC⁴⁵

GPC with nano-silica enhanced the elastic modulus by 17.2%, split tensile strength by 9.6%, and compressive strength by 25.8% compared to control concrete.⁴⁵ As shown in Fig.-3, with the enhancement in the amount of nano-SiO₂ in GPC, the split pulling strength improved by 9.6%.

Chemical Resistive Properties of GPC

GPC has been shown to exhibit excellent resistance to a variety of chemicals, making it a highly durable material for applications where exposure to harsh environments is a concern. After 28 days of exposure to concentrated sulfuric acid, GPC samples showed no sign of degradation, highlighting the material's

superior acid resistance.⁴⁶ The oxy-aluminum bond (-Al-Si-O) is indeed a key contributor to the strength characteristics of GPC. This bond is essential in facilitating the geopolymerization process, which is the formation of a strong, durable, and cohesive material. Sulfuric acid can indeed attack the oxy-aluminum bridges (-Al-Si-O) in GPC, which could result in a decrease in its compression resistance over time, although GPC is generally more resistant to acid attack than conventional concrete.⁴⁷ Blended ash-based GPC performs better than OPC concrete subjected to a 2% sulfuric acid solution, highlighting the superior acid resistance of GPC. The aluminosilicate polymer structure in GPC is a key factor that contributes to its superior performance, especially in harsh environments like exposure to sulfuric acid⁴⁸. GPC has been shown to exhibit enhanced compressive strength (up to 15%) when exposed to seawater, while its porosity can still be affected by sulfate attack over time.⁴⁹ GPC generally exhibits excellent resistance to chemical attacks, including exposure to 5% hydrochloric acid (HCl) and 5% sulfuric acid (H₂SO₄) solutions, particularly when immersed for prolonged periods (15, 45, 75, and 105 days) at room temperature.⁵⁰ It was found that GPC exhibited superior characteristics when exposed to sulfuric acid (pH = 0.8) and acetic acid (pH = 2.4) solutions for 1, 2, 3, 4, and 5 months, respectively. The strength of the acid solutions was determined based on the actual exposure of concrete in food processing, mining, and sewage treatment plants.⁵¹ The GPC and conventional concrete were immersed in hydrochloric acid solution according to the procedure prescribed in ASTM G20-08 for 60 days. The acid test resulted in greater weight loss for conventional concrete compared to GPC.⁵² When a metakaolin-based geopolymer composite mixed with silica fume, colemanite waste, and slag was immersed in 10% (v/v) sulfuric acid and hydrochloric acid solutions for up to one year, its oxy-aluminum bond (-Al-Si-O) weakened.⁵¹ The results of the residual flexural strength and compressive strength noted in GPC based on metakaolin after exposure to sulfuric acid and hydrochloric acid are presented in Fig.-4 and Fig.-5, respectively.⁵³ When GPC samples were treated with hydrochloric acid solutions of varying concentrations (3%, 5%, and 7%) for 3 and 6 months, no apparent damage was observed in the outer portions of the GPC.⁵⁴ The Geopolymer (GP) mortars, made from the residuals of kaolin and bamboo, along with ordinary Portland (OPC) mortars, were exposed to a 5% H₂SO₄ solution for fourteen, twenty-eight, and forty-two days. The results showed that the GP mortars exhibited better characteristics than the OPC mortars, with lower loss in weight and compression resistance.⁵⁵ The compression resistance and mass of the GPC specimens were slightly reduced after being immersed in H₂SO₄ solutions with varying concentrations (0.5%, 1%, and 2%) for 3 months.⁵⁶ After exposing the GPC and OPC specimens to 5% sodium sulfate and 5% sulfuric acid solutions, the GPC specimens exhibited superior durability characteristics against both acid and sulfate attacks.⁵⁷

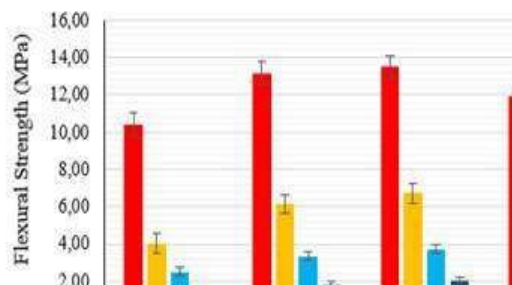


Fig.-4: Bending Strength of Metakaolin-based GPC after Sulphuric Acid Effect⁵³

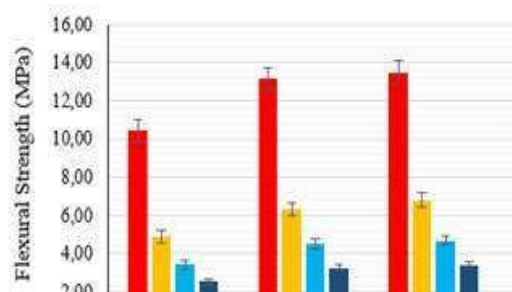


Fig.-5: Bending Strength of Metakaolin-Based GPC after Hydrochloric Acid Effect⁵³

The GPC and OPC specimens were subjected to durability studies under different aggressive environments (distilled H_2O , 0.005 M H_2SO_4 , 0.05 M H_2SO_4 , 10% NaCl, and 10% Na_2SO_4) for thirty, sixty, and ninety days. The results exhibited that GPC specimens exhibited good resistance to chemical attacks. However, the compressive strength of both OPC and GPC samples was reduced with an increasing exposure period.⁵⁸ The Geopolymer mortars were prepared by mixing agro-industrial wastes, such as ash derived from palm oil fuel and crushed fuel ash, with the appropriate alkaline activators instead of cement. When these Geopolymer samples were treated with 5% Na_2SO_4 solution and 2% H_2SO_4 solution for 28, 56, 90, 180, and 365 days, they demonstrated better resistance to these chemical solutions.⁵⁹ The physical presence of the geopolymer and OPC samples after 365 days of immersion in 5% Na_2SO_4 solution is presented in Fig.-6. The GPC exhibited smaller adsorption and Cl^- infiltration depth compared to OPC when the specimens were exposed to saltwater containing 3.5% NaCl.⁶⁰ The GPC and OPC specimens were treated with MgSO_4 , HCl, and H_2SO_4 solutions. The GPC specimens exhibited better resistance to these chemical solutions than the OPC specimens.⁶¹

Fire resistive properties of GPC

A special case of GPC specimens was found to exhibit better fire-resistant characteristics than OPC. When exposed to high temperatures, they demonstrated better durability behavior.⁶² Industrial products such as bottom ash, iron-making slag, cement kiln dust, metakaolin, rice husk dust, and others were very useful in the formulation of GPC. Three different raw materials—viz., calcined clay, GGBS, and fly ash—along with SiO_2 fume were utilized to prepare GPC. These specimens exhibited better fire resistance characteristics at 600°C, 800°C, and 1000°C.⁶³ It was reported that GPC exhibits better fire resistance properties than OPC when exposed to 500°C for 2 hours. It was also found that the residual compression resistance of GPC increased from 13% to 45% after subjection to 500°C, while OPC was unable to sustain such high-temperature exposure.⁶⁴ The effect of replacing nano-silica (nS) with rice husk ash (RHA) in recycled aggregate geopolymer concrete (RAGC) was studied. RAGC with RHA exhibited optimum post-fire residual strength compared to RAGC containing Ns.⁶⁵ It was reported that GPC coated with a fire-retardant coating resulted in the highest compression resistance strength of 54.7% after burning the samples at 1000°C.⁶⁶ The muffle furnace box used for fire testing is shown in Fig.-7.

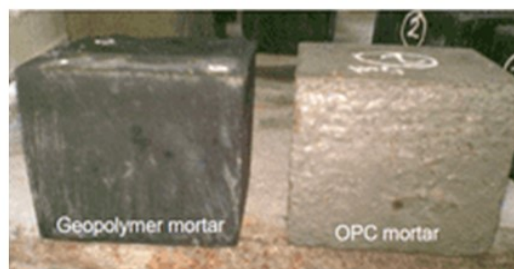


Fig.-6: Physical Appearance of Geopolymer and OPC Specimens after 365 Days Immersion in 5% Sodium Sulphate Solution⁵⁹

It was illustrated that lightweight aggregate geopolymer concrete (LWAGC) exhibited enhanced compression resistance up to 300°C, after which its strength deteriorated up to 800°C due to the variation in thermal elongation amongst the lightweight aggregate and the geopolymeric paste.⁶⁷ GPC and OPC panels, each comprising a layer of steel wire, were subjected to fire for about 2 hours. After fire exposure, cracking was less pronounced in the GPC panels compared to the OPC panels, and the residual capacity was higher in the GPC panels (61% to 71%) than in the OPC panels (50% to 53%). Hence, the fire endurance of GPC was greater than that of OPC.⁶⁸ The geopolymer-coated concrete exhibited cracking at the surface because of the removal of geopolymeric water at certain high temperatures during fire exposure.

However, the structural integrity of the GPC was fully preserved. These results indicate the potential for successful passive fire protection in the construction of underground tunnels.⁶⁹ Class C fly ash-based GPCs containing potassium-based activators exhibit better thermal and fire resistance characteristics⁷⁰. It was found that GPC with a ratio of 95% class F fly ash to 5% class C fly ash exhibited better fire resistance characteristics than ordinary cement mortar.⁷¹ The surface integrity of GPC was maintained

even after exposing it to fire at temperatures between 500°C and 1200°C sustained for 2 hours, with only crazing cracks and no spalling.⁷² The weight-holding capacity of MK-based GPC increased to about 12% of its initial strength under low-level fire exposure.



Fig.-7: Furnace Box for Fire Testing⁶⁶

However, its strength decreased to about 28% of its initial strength with high-level fire exposure for a longer duration. The loss in strength was primarily due to spalling.⁷³ The moisture uptake behavior of GPC samples increased after subjection to 700°C fire, in comparison with the untreated samples.⁷⁴ The fire-resistant properties of GPC containing basalt fiber (BF), polypropylene fiber (PF), and crimped steel fiber (SF) were evaluated at high temperatures (>900°C). The results showed that GPC with BF exhibited reduced compressive strength after exposure to 925°C for 60 minutes.⁷⁵

Radioactive Resistive Properties of GPC

GPC structures have several advantages compared to conventional concrete structures. It was found that GPC-based buildings offer better resistance to gamma and neutron radiation. GPC with higher GGBS content is more effective at absorbing gamma photons. Similarly, GPC with a higher concentration of quartz sand is effective in reducing the impact of fast neutrons.⁷⁶ The presence of heavyweight coarse aggregates (HWCAs) in GPC provides better radiation protection, even after exposing the building to high temperatures (i.e., 900°C for 3 hours).⁷⁷ Geopolymer is recognized as an outstanding cementitious material, offering reduced embodied energy and a lower carbon footprint in comparison with the materials based on OPC. It has also been found that GPC is a suitable material for the encapsulation and immobilization of nuclear waste.⁷⁸ Fly Ash Geopolymer (FAGP) combined with 15% BaSO₄ results in the highest Z_{eff} (Effective Atomic Number) and the lowest radiation dosage (34.68 µGy), making this combination a promising radiation shielding material.⁷⁹ It has been reported that Radiation Shielding Concrete (RSC), a type of GPC containing magnetite aggregates with a density of 3.2 – 4 t/m³, can be used in healthcare facilities, nuclear power plants, storage casks for radioactive waste, and nuclear research facilities.⁸⁰ It was found that RSC can be an effective alternative to conventional shields in controlling neutron and gamma radiation.⁸¹ To control the simulated radionuclide ¹³³Cs⁺, fly ash-based GPC was prepared by adding cesium nitrate. It was found that Cs⁺ compounds exhibit superior physico-chemical properties, making them suitable as stimulants for ¹³⁷Cs.⁸² GPC has two advantages: (i) quick dewatering, which makes the material stable at high temperatures, and (ii) the ability to trap radioactive elements within the zeolite framework, significantly controlling their effects.⁸³ The classification of RSC is shown in Fig.-8.⁸⁴ A high content of heavyweight coarse aggregates is required in the formulation of RSC.

Role of GPC in Reducing the Emission of Greenhouse Gases

The greenhouse effect refers to the rise in environmental temperature caused by the trapping of heat through greenhouse gases such as CO₂, CH₄, and NO_x. This effect can lead to various environmental problems, such as soil erosion, reduced crop production, global warming, climate change, and rising sea levels. Therefore, the concentration of greenhouse gases in the environment must be significantly controlled.⁸⁵⁻⁸⁷ A major component of GPC is fly ash, a premier sustainable construction material due to its ability to replace conventional Portland cement (PC).⁸⁸ GPC has emerged as a potential solution to climate-related issues due to its lower carbon emissions. It demonstrates a 64% lower global warming potential than OPC. In Australia, the use of GPC can cut greenhouse gas emissions by 44-64% in comparison to OPC, although it comes at double the cost of OPC.⁸⁹⁻⁹¹ The CO₂ footprint of GPC is nearly 9% smaller than that of 100% OPC binder.¹⁹ The manufacturing of OPC generates significant greenhouse

gas emissions, making it a key factor in driving global warming.⁹² The CO₂ footprint of GPC is nearly nine percent smaller than that of 100% OPC binder. The production of OPC emits substantial greenhouse gases, which play a significant role in accelerating global warming. It has been reported that the formation of one ton of Portland cement generates about one ton of CO₂ emissions, primarily from the heating of CaCO₃, contributing to about 7% of global CO₂ emissions. However, Geopolymer-based concrete structures are environmentally friendly, as they do not release excessive amounts of CO₂ like OPC.⁹³

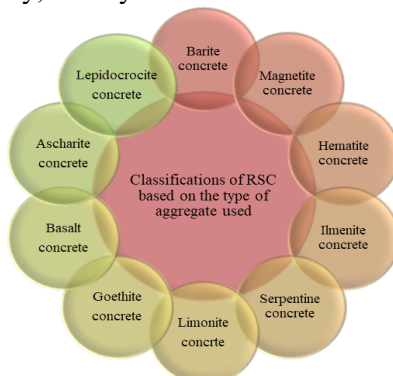


Fig.-8: Classifications of RSC⁸⁴

It has been reported that GGBS-based GPC exhibits superior sustainability, higher economic efficiency, and a lower environmental impact compared to OPC.⁹⁴

CONCLUSION

The production of OPC leads to the release of excessive carbon dioxide emissions, which is a major cause of global environmental issues. Recently, OPC has been replaced by GPC, which is considered more sustainable due to its primary constituent, fly ash, an industrial waste material. GPC also exhibits better durability compared to OPC. The performance of Geopolymer (GP) is primarily based on the reaction between SiO₂ and Al₂O₃ with the basic activator solution. The compression resistance of GPC can be impacted by various factors, such as curing temperature, curing settings, and water content. GPC demonstrates excellent resistance to a range of chemicals, including sulphuric acid, hydrochloric acid, and sodium sulfate. Additionally, GPC exhibits superior fire resistance at temperatures of 600°C, 800°C, and 1000°C. GPC demonstrates better fire resistance than OPC when exposed to 500°C for 2 hours. The inclusion of heavyweight coarse aggregates (HWCAs) in GPC enhances its radiation protection, even when exposed to high temperatures. GPC has emerged as a potential solution to climate-related issues due to its lower carbon emissions. It has been shown to have a 64% reduced global warming impact compared to OPC. Additionally, the CO₂ footprint of GPC is nearly 9% smaller than that of 100% OPC binder.

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CONFLICT OF INTERESTS

The authors declare that there are no conflicts of interest.

AUTHOR CONTRIBUTIONS

All authors made significant contributions to this manuscript, participated in the review and editing process, and approved the final draft for publication. The research profiles of the authors can be verified through their ORCID ids provided below:

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