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Engineering Geopolymers for Sustainable Construction: From Synthesis to Performance

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ABSTRACT

Because of the large amount of CO_2 emissions associated with its production, ordinary Portland cement (OPC) is gradually replaced by more eco-friendly binders, among which geopolymers are the most promising. This review highlights the most important features of geopolymer cements, the production of which uses aluminosilicate precursors, mainly metakaolin, fly ash, blast furnace slag, and various agricultural wastes, cured at a temperature below 100°C. The review discusses the chemical composition, nanostructure, mechanical performance, and durability of geopolymers, as well as their CO_2 footprint, comparing them to OPC. It was found that geopolymers have a unique amorphous aluminosilicate binder formed by cross-linked SiO_4 and AlO_4 tetrahedra, whereas the OPC binder consists of calcium-silicate-hydrate gel. The formation of a geopolymer's nanostructure allows achieving compressive strengths of 20 MPa after 4 h and 100 MPa after 28 days. Geopolymers are resistant to acid attack, alkali-silica reaction, high temperatures, and permeability. Moreover, the carbon footprint of fly ash- and slag-based geopolymer concrete can be as low as 70% lower than that of OPC concrete. Nevertheless, owing to a production cost that remains roughly twice that of Portland cement, less consistent carbonation resistance, and the continued absence of widely adopted design codes, geopolymers cannot yet be routinely recommended as a default binder for large-scale construction, even though demonstration-scale infrastructure projects show that these barriers are not insurmountable.

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1. Introduction

To facilitate a more sustainable future, sustainable materials must be cheap to produce, of adequate strength, with the appropriate physical/chemical characteristics and must not produce a significant amount of environmental pollution. Cement, in the form of ordinary Portland cement (OPC), is the world's principal building material. But Portland cement manufacturing is not sustainable: roughly one tonne of CO_2 is emitted for every tonne of OPC produced; overall production is responsible for around 5-7% of global CO_2 emissions — about 10% if you count all building materials [1-3].

As the biggest element of the greenhouse effect (accounting for approx. 65% of global warming potential), CO_2 is being scrutinized closely, especially as up to 85%

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of emissions from a cement plant arise from calcination of raw materials into clinker at 1450°C. The remainder results from combusting fuel and plant energy usage. This fact has led to extensive work in developing low-carbon alternatives to conventional cement, such as geopolymers, over the past several decades [4].

2. What is a Geopolymer?

The word "geopolymer" was created in 1978 by the French materials chemist Joseph Davidovits. He discovered for the first time a new type of natural or synthetic inorganic mineral polymer and gave it the name "poly(sialate)" (for "silico-oxo-aluminate chain"). A geopolymer is a 3-dimensional, ceramic-like, amorphous to semi-crystalline, inorganic polymer created by chemical activation of solid, mineral-based, aluminosilicate materials with a high concentration of alkaline solution at lower temperature (typically lower than 100°C and often at room temperature). Examples of such materials are calcined clays, fly ash, and slag [5, 6].

Geopolymers are defined structurally as corner-sharing SiO and AlO tetrahedra joined by common oxygens, yielding a chain or ring structures of formula -(-Si-O-Al-O-)n-, as opposed to oxide representation of traditional cement nomenclature (i.e., 2SiO·AlO). Based on the Si/Al ratio and linkage the major molecular species can be divided into the poly(sialate) family (-Si-O-Al-O-), the poly(sialate-siloxo) family (-Si-O-Al-O-Si-O-), poly(sialate-disiloxo), organo-siloxo (poly-silicone), the poly(alumino-phospho) family, and the poly(ferro-sialate) family [7-9].

3. Raw Materials and Synthesis Route

Any material having significant amounts of reactive silica (SiO_2) and alumina (Al_2 O_3) capable of dissolving under alkaline conditions qualifies to be used as geopolymer precursor materials. Geopolymers may be synthesized using two principal chemical reaction mechanisms. Under the alkaline activation method, aluminosilicate precursors are reacted with alkali hydroxide (NaOH/KOH) and soluble alkali silicate leading to the creation of polymeric structures like poly(silicates), poly(siloxo), poly(silicoaluminates) and poly(sialate) [11]. The second method involves acid activation method where phosphoric acid is used as an activating medium giving rise to poly(alumino phosphate) [12, 13]. There have been a number of natural and industrial aluminosilicate materials which have been effectively used as precursors in geopolymer preparation. Some examples are metakaolin (produced by calcination of kaolin clay at about 750 °C), fly ash, granulated blast-furnace slag and silica fume. Furthermore, there have been a number of agricultural byproducts such as rice husk ash, corn cob ash, bagasse ash, palm bark ash, bamboo leaf ash, and sugarcane bagasse ash which have great potential in replacing cementitious and pozzolanic materials in geopolymer systems [14-16].

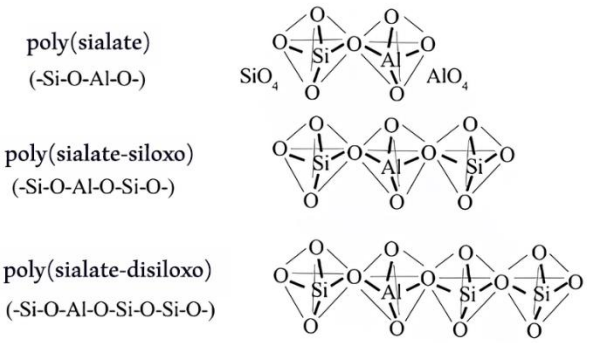

Figure 1. Chemical structure of the poly(sialate) family: mono-, di- and tri-silicate linkages between SiO_4 and AlO_4 tetrahedra (after Davidovits) [10].

3.1. Setting and Hardening Mechanism

Reaction is a two-stage process of geopolymerization, first dissolution of the aluminosilicate precursor in an alkaline activating solution and second polycondensation of the liberated Al and Si complexes to form a solid, three-dimensional network.

Rate of dissolution is a function of alkali content of solution, nature of alkali-metal cation, agitation rate, duration of dissolution, composition and structure of aluminosilicate source. Nucleation of the gel is initiated at the surface of the particles. Diffusion of Al and Si complexes into the growing gel phase occurs and subsequent sequence of events is that of dissolution-diffusion and subsequent condensation. +Hardening occurs by continued polycondensation [17, 18].

Temperature, pH, size of alkali cation have effect on condensation rate; elevated temperature, elevated pH, large cation size enhance the hardening. Unlike ordinary Portland cement, water is not part of the structural framework in geopolymer; rather it serves to provide workable mix during mixing and then escapes during heating and does not appear in the resulting sodium–alumino–silicate–hydrate (N-A-S-H) or potassium–alumino–silicate–hydrate (K-A-S-H) gels [19].

Standard procedure is to combine powder aluminosilicate precursor with NaOH/KOH, sodium-silicate solution to make a homogeneous paste, pour into a form, leave to stand at ambient temperature for a few hours,

after which curing can be performed at ~80°C for 1-2 days and followed by 7-28 days of further development [20, 21].

4. Nanostructure

Geopolymers are defined macromolecules in terms of size and molecular weight that have had their micro-structure and molecular structure elucidated in the solid state using electron microscopy (TEM/SEM), light scattering, and Si/Al MAS-NMR spectroscopy [22]. Zhang et al. applied TEM to a fully cured potassium poly(sialate-siloxo) geopolymer. This work showed a microstructure that consisted of a 5-15 nm particle network with interstitial nanospaces and evidence of order on a 3-10 nm scale (i.e., nanoporous and spongelike). The elemental composition from EDS data was approximately 2:1 Si:Al and suggested short range order and repetitive sequences [23]. Škvára et al. reported a very similar microstructure (5-20 nm) for fly ash-based geopolymer where the nanoparticles appeared similar in structure to surfactant micelles arranged into ordered assemblies; the aggregates are occasionally referred to as precipitated particles. The nanoparticle network structure persists when geopolymer is heated to 1000°C and remains amorphous to X-ray scattering, hence behaving like a polymer [24, 25].

Figure 2. TEM image of a nanoparticle (“geopolymeric micelle”), 5–15 nm in size, marked by the arrow [10].

5. Setting Time and Mechanical Properties

Geopolymer cement develops rapid setting strength at room temperature: a reported compressive strength in excess of 20 MPa is obtained within four hours, and up to 70-100 MPa (or even more) at 28 days at 20°C. The low porosity of geopolymer compares favourably with that of Portland cement pastes and mortars, which gives rise to better mechanical strength [26, 27].

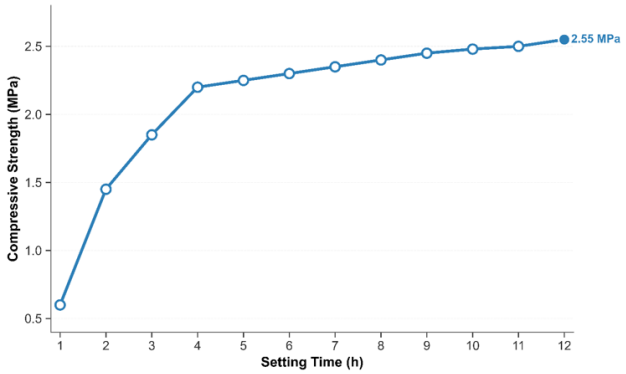

Figure 3. Setting behavior of a potassium poly(sialate-siloxo) cement at ambient temperature: compressive strength vs. time (hours) [10].

Comparative testing against Type I and Type III Portland cement shows that geopolymer concrete both sets faster and ultimately develops significantly higher strength.

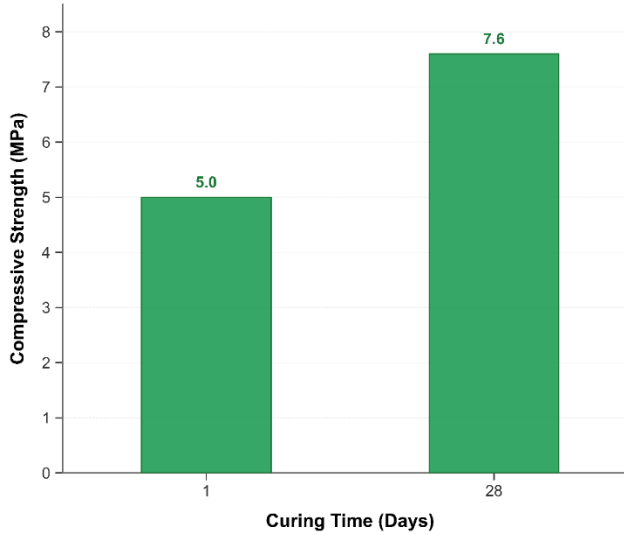

Figure 4. Geopolymer strength gain at 1 and 28 days.

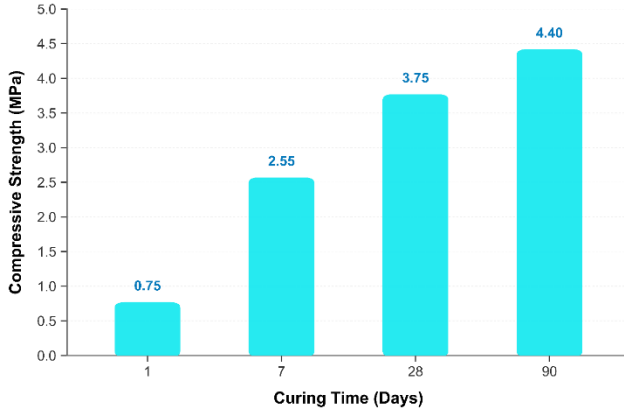

Figure 5. Portland cement strength gain at 1, 7, 28 and 90 days, for comparison [10].

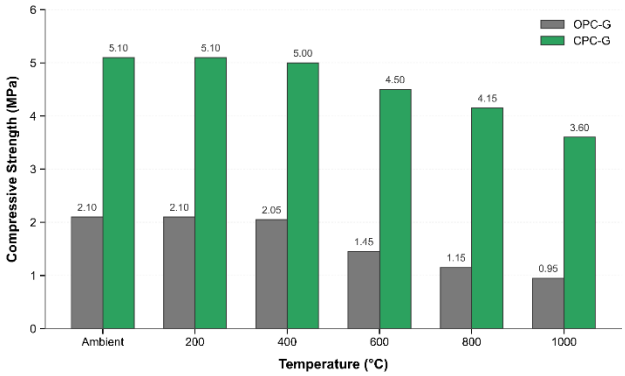

Figure 6. Strength development of ordinary Portland cement (Types I and III) compared with a geopolymer concrete over the first 12 hours [10].

Nascent geopolymers from sodium have been found to retain their amorphous structure up to 500°C, but potassium-based geopolymers can maintain their amorphous character even up to 1000°C. Both have been found to preserve greater strength when compared with Portland cement pastes that undergo continuous degradation by dehydrating C-S-H followed by decomposition of $Ca(OH)_2$ and $CaCO_3$ [28, 29].

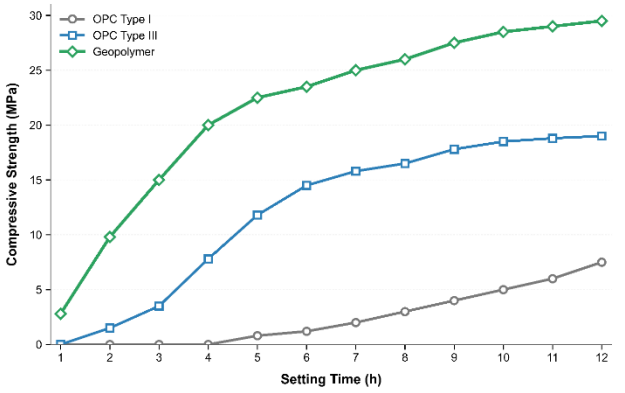

Figure 7. Compressive strength of Portland concrete (OPC-G) versus a geopolymer concrete (CPC-G) after exposure to increasing temperatures (40–500°C) [30].

6. Chemical Durability

There are two major processes through which concrete becomes susceptible to deterioration: sulfate attack and the corrosion of reinforcement caused by chlorides. Since geopolymers have very low calcium content in comparison to Portland cement, the calcium-based expansive products that cause the sulfate attack process in OPC concrete are unlikely to form, thus ensuring high resistance of geopolymer concrete against sulfate attack. In addition, the geopolymer binder matrix is not as permeable as the hydrated Portland cement paste [31].

The geopolymer cements are also quite resistant to alkali-silica reaction, which may cause damage to OPC-based concrete made with some reactive aggregates, although the alkalinity of geopolymer cement can be very high (up to 9.2% vs. low alkali requirement for OPC). The geopolymer concretes have been found to resist well against acid attack, to have low permeability (10^{-9} cm/s, like OPC) and good freeze-thaw durability [32].

Carbonation resistance, however, is one aspect of geopolymer durability that does not uniformly favor the new binder. Because atmospheric CO_2 reacts with the alkaline pore solution and lowers pH through a mechanism distinct from the decalcification of $Ca(OH)_2$ and C-S-H in OPC, the degree of protection depends strongly on precursor chemistry and activator dosage. Fly ash- and slag-based geopolymers with limited free alkalinity have shown carbonation depths comparable to those of ordinary Portland concrete of similar strength grade, whereas mixes activated with a high dose of soluble sodium silicate tend to carbonate faster and lose surface alkalinity more readily, which can threaten the passivation of embedded steel reinforcement earlier than in OPC systems [39]. This sensitivity to mix design is not yet captured by a single predictive model, and it tempers the otherwise favorable durability profile described above.

7. Geopolymer Versus Portland Cement: Chemistry Comparison

Hardening process of ordinary Portland cement occurs due to hydration reactions; C_3 S along with other calcium silicates react with water forming calcium-silicate-hydrates (C-S-H) gel and $Ca(OH)_2$. In geopolymer cement, on the other hand, the hardening process occurs as a result of a polycondensation reaction, also known as geopolymerization, in which Al and Si atoms form covalent bonds and cross-linked molecules, making the geopolymer become rock-like. Geopolymers have a chemical composition closer to natural silicate minerals than to hydrates of the cement paste. This is evident from ^{29}Si NMR spectra of both cases [33, 34].

8. Applications

Geopolymers can be tailored, mainly through the Si:Al ratio, for a wide range of uses, from refractory ceramics to low-carbon binders, heat-resistant composites, and industrial sealants (Table 1).

Apart from these uses, geopolymer cement is under study for use in paving roads, airport runway slabs, precast concrete construction elements, non-combustible construction panels and cladding, hazardous waste

containers, as well as constructions in severe sulfate-chloride environments like culverts and offshore structures. The principal drawbacks associated with it at present include sensitivity to curing and less practical experience compared to normal concrete [35, 36].

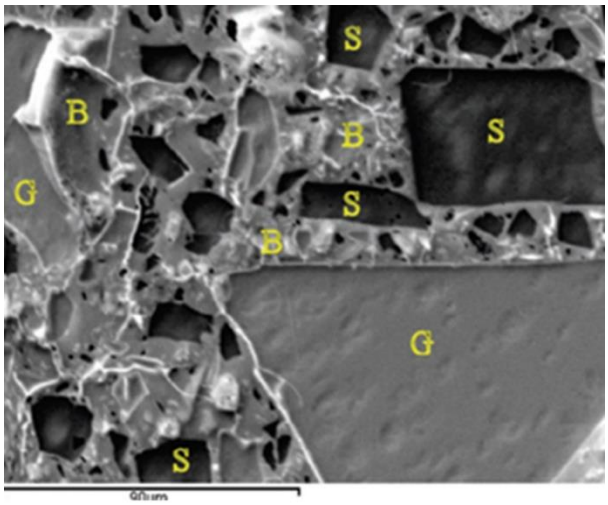

Figure 8. Back-scattered electron (BSE) image of a ferronickel-slag glass geopolymer microstructure (compressive strength > 70 MPa): S = slag, G = glass, B = binder phase [10].

The clearest demonstration that geopolymer concrete can be produced and placed at genuine infrastructure scale is the pavement work carried out at Brisbane West Wellcamp Airport in Queensland, Australia. Roughly 40,000 m³ of a fly ash- and slag-based geopolymer ready-mix concrete, marketed as Earth Friendly Concrete, was used for the airport's apron, taxiway, and turning-node pavements, placed with a conventional slip-form paver in slabs up to 435 mm thick. The project reported approximately 8,640 tonnes of avoided CO_2 emissions relative to an equivalent Portland-cement pavement, alongside flexural strength and workability that the contractor judged suitable for standard paving plant [42]. Cases of this kind remain rare, but they show that the barrier to wider adoption is no longer purely technical.

A large part of that limited practical experience traces back to the regulatory environment rather than to the material itself: dedicated design codes and standardized test methods for geopolymer concrete are still uncommon, and engineers who specify concrete through prescriptive OPC-based codes (defined by cement content and water-cement ratio) have no equivalent framework for an alkali-activated binder with a fundamentally different chemistry. Performance-based specifications, of the kind first adopted in Australia and more recently piloted in the United Kingdom, are widely regarded as the more workable path to standardization, but until such frameworks are adopted more broadly, specifiers and insurers are likely to remain cautious about geopolymer concrete outside of niche or precast applications [40].An emerging application that falls outside the traditional cast-in-formwork model is extrusion-based 3D printing of geopolymer pastes and mortars. Because geopolymer mixes can be formulated with the rapid, controllable stiffening behavior that extrusion printing requires, without relying on cement hydration kinetics, they are being explored as a low-carbon alternative to the OPC-based mortars currently used in most construction 3D-printing trials. The main obstacles are rheological rather than chemical: achieving a mix that is fluid enough to pump and extrude yet stiff enough to retain its shape and bond well to the previously printed layer, across the open time needed for a full print run [43].

Table 1.
Typical geopolymer applications as a function of the Si:Al ratio.

Si:Al Ratio	Typical Application
1	Bricks, fire-resistant ceramics, refractory materials
2	Low-CO_2 cements; encapsulation/immobilization of toxic and radioactive waste
3	Heat-resistant composites, foundry tooling/equipment, fiberglass composites
> 3	Industrial sealants / binders

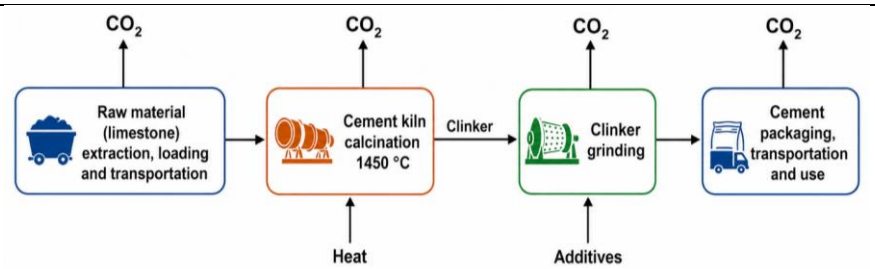

Figure 9. Simplified Portland-cement production flow diagram, showing the three main CO_2 -emission points: raw-material calcination, kiln fuel combustion, and plant energy use.

9. Life-cycle Assessment and Environmental Benefits

Goal and Scope Definition, Life-Cycle Inventory (LCI), Impact Assessment, and Interpretation are the four major steps followed while using Life Cycle Assessment (LCA). Life cycle assessment helps in identifying the input, output, and environmental impacts associated with the entire life cycle of a process or product. Abiotic Resource Depletion Potential (ADP), Global Warming Potential (GWP), Eutrophication Potential (EP), Human Toxicity Potential (HTP), and Ozone Depletion Potential (ODP) are some of the impact indicators [37].

The comparative LCA analysis performed by Weil et al., employing the CML approach, suggests that even if the mass balance of a geopolymer mixture includes a lot of low-impact sand/aggregate, it is still the sodium silicate activator solution and metakaolin that are responsible for the majority of the global warming potential contribution, while fly ash and slag are of relatively low importance (Fig. 10). The optimal ratio between slag and fly ash in the geopolymer concrete mixture (see Table 2), which does not require thermal treatment, had a 70% lower GWP compared to Portland cement concrete, and 21% higher CED for the latter.

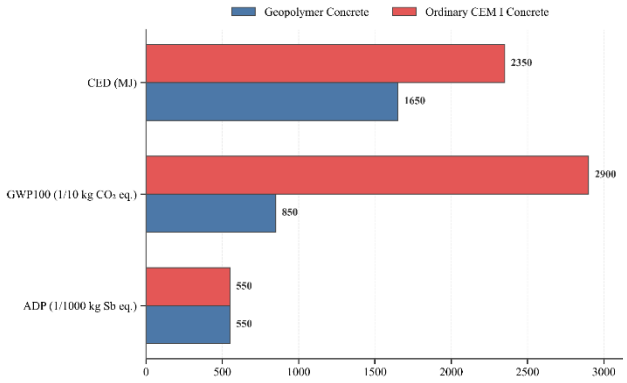

Figure 10. Comparative cumulative energy demand (CED), global warming potential (GWP100) and abiotic resource depletion potential (ADP) of geopolymer concrete versus ordinary CEM I Portland cement concrete.

The findings clearly show that geopolymer concretes formulated using industrial waste products such as slag and fly ash, and which do not need any form of thermal curing, can perform environmentally at par with, or even better than, ordinary Portland cement concrete, especially in terms of GWP, but with the same abiotic resource depletion and energy footprint as ordinary Portland concrete for frost resistance [30, 38].

Environmental performance is only part of the adoption calculus, and cost remains a genuine obstacle. A recent meta-analysis pooling production-cost and life-cycle data across a large number of published geopolymer and Portland-cement concrete mixes found that geopolymer concrete is, on average, roughly twice as expensive to produce as conventional concrete of comparable strength, driven mainly by the price of soluble sodium silicate activator. The same analysis confirmed a substantial net reduction in greenhouse-gas emissions for most geopolymer mixes but also found that several other impact categories, including some measures of resource depletion, were not consistently improved relative to Portland cement concrete [41]. Taken together with the LCA results discussed above, this suggests that geopolymer concrete's environmental advantage is real but narrower, and its economic case weaker, than a GWP-only comparison would imply.

10. Conclusions and Recommendations

Geopolymers are inorganic macromolecules with a defined nanoscale architecture (5–20 nm nanoparticles separated by nanopores) formed through a precise chemical mechanism of alkaline dissolution, polycondensation, and hardening of aluminosilicate raw materials or industrial/agricultural wastes. They can be engineered from diverse feedstocks to obtain properties suited to many applications, offering compressive strengths and durability (sulfate, chloride, acid, freeze–thaw, and high-temperature resistance) that generally exceed those of Portland-cement concrete, though with a less consistent advantage in carbonation resistance, together with substantially reduced CO_2 emissions and energy demand.

Based on the findings of the reviewed literature, several strategies can be adopted to further minimize the environmental impacts associated with geopolymer concrete production. First, geopolymer binders should be promoted as a sustainable alternative to conventional Portland cement, particularly in applications requiring high resistance to chemically aggressive environments. Second, incorporating blended aluminosilicate precursors, such as metakaolin and fly ash, can optimize the Al/Si ratio while decreasing the demand for sodium silicate, whose production is a significant contributor to the global warming potential (GWP) of geopolymer systems. Third, developing sodium silicate activators from agricultural waste materials, including rice husk ash, offers a promising approach to reducing the energy consumption and greenhouse gas emissions associated with activator production. Nevertheless, despite the environmental and technical benefits of geopolymers, their widespread industrial implementation remains constrained less by chemistry than by curing sensitivity, an approximately two-fold production cost relative to Portland cement

concrete, and the continued absence of widely adopted design codes and standardized test methods [40, 41]. The Brisbane West Wellcamp Airport pavements demonstrate that these barriers are not insurmountable [42], but they are also not yet routine. Continued research is therefore essential to enhance production robustness, narrow the cost gap, and support the standardization work needed for geopolymers to move from demonstration projects to default specification in large-scale construction.

Table 2.
Example mix design comparison between an optimized geopolymer concrete and an equivalent-performance Portland-cement concrete (kg/m³).

Material	Geopolymer concrete (kg/m³)	Portland cement concrete
CEM I 32.5R Portland cement	—	340
Ground granulated blast-furnace slag	230	—
Fly ash	57	—
Reactive waste material	83	—
Sodium silicate solution (37%)	33	—
NaOH solution (50%)	24	—
Mixing (deionized) water	99	170
Sand – aggregate (0–16 mm)	1878	1878
Total	2404	2388

AUTHOR CONTRIBUTIONS

A.E. conceived the review and defined its scope. A.E. and A.N. jointly conducted the literature analysis. A.N. drafted the initial manuscript, and A.E. critically revised it for intellectual content. Both authors read and approved the final manuscript.

HUMAN AND ANIMAL RIGHTS

Not applicable. This review is based on previously published literature on geopolymer materials science and did not involve human participants, human data, or animal subjects.

CONSENT FOR PUBLICATION

Not applicable.

AVAILABILITY OF DATA AND MATERIALS

No new data were generated or analyzed in the preparation of this review. All information discussed is available within the cited sources listed in the References section.

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CONFLICT OF INTEREST

The authors confirm that there is no conflict of interest related to this manuscript.

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