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REVIEW

Review of Geopolymer Technology, Barriers and Limitations

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ABSTRACT

Geopolymers are kind of inorganic polymeric green materials, which are considered alternatives to some traditional cementitious materials. Geopolymerisation involves reaction between amount of solid materials rich of high reactive amorphous or semi-amorphous silica and alumina in the existence of alkaline medium (Na_2O or K_2O) to formulate tri-dimensional alumina-silicate polymeric strong network. The present research is targeting to review Geopolymer technology and the major barriers and limitations that impede the implementation and development of Geopolymer technology. The review based on the reports, statistics and the opinions of the construction industry stakeholders, in addition to the past literatures to assess the potential barriers. The results of the research showed number of substantial barriers that still exist. Parameters such as alkaline sources, variety in the Al-Si source materials and the mix design details are mainly the target of this review. These including substantial decrease in the produced amounts of main Geopolymer precursors such as pulverised fly ash (PFA) and slags in the next 10 years due to new regulations regarding the decrease of the greenhouse gas releases. Other barriers including the difficulties in the mix fabrication, efflorescence and the absence of the standards and specifications for this technology. Some novel solutions are suggested for future research interests.

Keywords: Geopolymer, Geopolymer commercialisation, Alkali activated materials

1. Introduction

Manufactured concrete forms one of the most versatile construction materials that has been utilised worldwide. Concrete represents the best choice for a wide range of applications such as housing, bridges, highway pavements, manufacturing, water-containing and retaining structures. The current world production of the Ordinary Portland Cement (OPC) is growing up to 4.1 billion metric tonnes in 2017 [1]. The use of Portland cement in concrete construction is under serious evaluation because of substantial challenges are

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facing the method of production conventional cement. Firstly, the energy consumption through the whole process. Secondly, The high quantity of carbon dioxide gas (CO_2) released to the atmosphere through the production of cement [2]. (OPC) is fundamentally resulting from calcination of limestone (calcium carbonate) and silica rich clays up to 1450 °C which make (OPC) is environmentally not sustainable.

Therefore, two directions of explorations are being developed. The first one includes the use of new technologies and processes but keeping raw materials same. For instant, the utilisation of alternative fuels such as biomass fuels and fuels derived from waste materials instead of the normal fuel, however, not all fuel can reduce CO_2 emissions and can derive the cost and energy up [3].

The second direction comprises of using new methods and raw materials different from the raw materials that are used in producing (OPC). Low carbon cements and binders are encouraging alternatives to (OPC). Low Carbone cements started originally by substituting either fine aggregate with alternatives with less production energy [4–6] or replacing various ratios of supplementary cementitious materials (SCM) with (OPC) to generate what is called blended cements in order to reduce the produced (OPC) and up to 50% to produce a cement binder that has same performance of (OPC) [7–10]. SCM are including natural pozzolanic and industrial wastes and by-product materials that have substantial properties to behave as cementitious materials such as Pulverised Fly Ash (PFA), Ground Granulated Blast Slag (GGBS) and Metakaolin (MK). Large volumes of wastes and by-products that are non-recyclable are generated every year from different locations around the world. Some industrial wastes cannot be disposed or landfilled as it can lead to environmental problems with great damage to the soils, plants and animals. In addition, landfills generating hydrological threat and pollution to the ground water. Incineration treatment has worse effects even with the huge attempts to reduce the pollutants but still pose great polluters compared to other disposing method [11].

Alternatively, there are noteworthy investigations to develop other types of green cements that entirely free of (OPC) and based principally on green raw materials [12, 13]. Mineral Products Association (MPA) in UK listed number of novel (non-Portland) cements with low energy in their fact sheet [14]. For example, CSA (calcium sulfo-aluminate)-belite cements are used in the industrial sector in China for about 35 years. CSA cement is manufactures by heating/sintering wastes such as fly ash, gypsum and limestone at 1200 C-1250 C in kilns. However, when comparing with OPC CEM 1, the energy savings are just 25% as maximum with CO_2 emissions reductions around 20%.

One of these novel green cements is Geopolymer cement. Geopolymer cement synthesised through Geopolymerisation innovative chemical reaction. The resulted product has excellent features as high early strength, high resistance to elevated temperature and aggressive conditions. Geopolymerisation involves reaction between amount of solid materials rich of high reactive amorphous or semi-amorphous silica and alumina in the existence of alkaline medium (Na_2O or K_2O) to formulate tri-dimensional alumina-silicate polymeric strong network [15–19]. Although Geopolymer has substantial advantages but still has not been used widespread in the glob due many problems and limitations. Therefore, the current review is directed to evaluate the main barriers that facing the use of Geopolymer cement and giving suggested solutions.

1.1. Geopolymer background and terminology

Geopolymer technology is a term given to the reaction of the solid Al-Si materials in an alkaline medium. Davidovits [20, 21] coined Geopolymer term on the 3D Al-Si chains synthesized with the existence of (Na_2O, K_2O, etc.). However, the early attempts with these materials started in Ukraine through 1950's by Glukhovsky [22] where it was originally called "soil silicate concretes" or "soil cements" and encompasses calcium silicate hydrate (CSH) and (Al-Si) phases. Provis [23] revealed that Geopolymers are types of the alkali-activated binders that are resulted from the reaction of solid Si-Al with highly concentrated aqueous alkali hydroxide or silicate solution to produce in general alkali alumina-silicate materials and suggested that all these materials preferably to be termed an 'inorganic polymers'. Krivenko [24] named geocements on the structure formation processes in the alkaline alumina-silicate binders corresponded to the zeolites, which are geological transformations of alumina-silicate volcanic rocks in the presence of alkaline solutions at low temperatures (below 200–300 °C, depending on a zeolite type). Davidovits explained in the Geopolymer Camp (2016) that Alkali-Activated Materials (AAM) are not Polymers and they cannot be termed Geopolymer. Geopolymers are not subset of (AAM) because they are not a calcium hydrate alternative and does not have (N-A-S-H) or (K-A-S-H) hydrates [25]. Moreover, Davidovits clarified that alkali activation is different from Geopolymerisation reaction because there is nothing to activate and the start materials of the Geopolymer are already reactive materials and the process preferably to called alkalination as there is no Geopolymer activator but it's called "reagent" or "hardener". With all these different names and terminologies, the system and the internal structure of the product materials is remaining the same with alkali-alumina-silicate products, which are connected with strong bonds to yield good properties.

Geopolymer binders started recently to gain great consideration in the construction sectors. Geopolymer cement (GPC) is one of the favorable choices to be a satisfactory alternative to the Ordinary Portland cement (OPC). It is now already used in the industrial sectors in UK, USA and Australia. However, there still substantial barriers and obstacles towards the real applications of Geopolymer cement as it still used just in the pre-cast elements. Several factors forms significant issues that inhibits the widespread of this innovative technology.

2. Method

The current paper is aiming to evaluate and review the main barriers and the difficulties that exist in the current Geopolymer synthesis process. The study will depend on the past investigations and the technical reports that are published so far of the recent development of Geopolymer technology, as Geopolymer concrete synthesis depending on several variables. Therefore, some of these variables will be evaluated to check which one has the most impact on limiting (GPC) from being wide spread in the industrial sector. Factors such as alkaline sources, variety in the Al-Si source materials and the mix design details will mainly be the target of this review.

3. Geopolymer synthesis process

Geopolymerisation include alkaline source and solid alumina-silicate raw material in order to form the polymeric final chains. The reaction is exothermic process that is carried out through oligomers (dimer, trimer) which provide the actual unit structures for the three-dimensional macromolecular network. Once the Al-Si powder is mixed with the alkaline solution, a paste forms and quickly transforms into a hard Geopolymer. Therefore, there is no sufficient time and space for the gel or paste to grow into well-crystallised structure which is the fundamental difference between zeolite and Geopolymer [16, 26]. Fig. 1 illustrate the major stages of Geopolymerisation reaction. The mechanism of

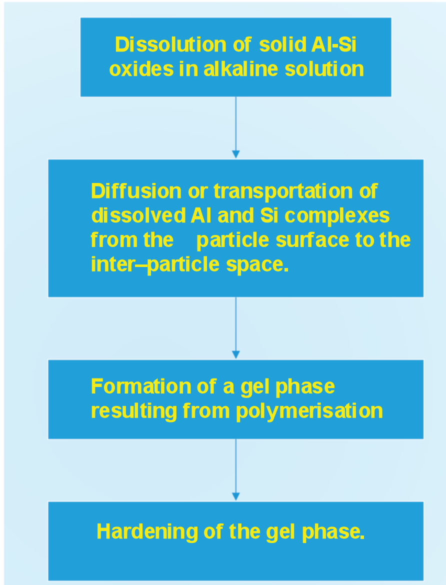

Fig. 1. Geopolymerisation stages.

Geopolymerisation may be varied depending on the source materials that the Geopolymer synthesised.

By the end of the reaction, the final Geopolymerisation chemical equation [16].

$$\begin{aligned} & n\left(Si_2O_5, Al_2O_2\right) + 2nSiO_2 + 4nH_2O + NaOH \text{ or } (KOH) \\ & \quad \rightarrow Na^+,\ K^+ + n(OH)_3 - Si - O - \underset{\displaystyle (OH)_2}{\underset{|}{Al^-}} - O - Si - (OH)_3 \\ & \qquad \text{(Geopolymer Precursor)} \end{aligned} \tag{1}$$

The final geopolymeric product could be one of the following depending on the Si:Al atomic ratio as shown in Table 1 [27, 28].

Past studies listed the main factors and parameters that can affect the Geopolymerisation [29, 30]:

1. The weight ratio of alumino-silicate powder to alkaline solution and preferably in range (3.0–5.5).
2. The alkaline material type and amount.
3. The molar ratio Si/M controls the nature and quantity of the siliceous species.
4. When the alkaline material is notably reactive, this enables a fast oligomer formation and, consequently govern the Geopolymerisation reaction and the final performances of the materials.

Table 1. Geopolymer products.

Atomic ratio Si:Al	Chemical group	Chemical formation
1	sialate, poly(sialate)	-Si-O-Al-O-
2	sialate-siloxo, poly(sialate-siloxo)	-Si-O-Al-O-Si-O
3	sialate-disiloxo, poly(sialate-disiloxo)	-Si-O-Al-O-Si-O-Si-O-

5. It is observed that the workability as well as compressive strength of Geopolymer mortar increases with increase in concentration of alkaline medium in terms of molarity.
6. The SiO_2/M_2O molar ratio of the activating solution.

3.1. Geopolymer system

The conventional method of producing Geopolymer involves two main parts, which are the raw materials that form the source of Al-Si. The second part forms the source of alkali, which normally is an alkali (Na, K, Ca…) or silicate hydroxide. Table 2 shows the categories of the start raw materials that usually been examined by past studies. The most widely known alumina-silicate source that has been used as a raw material in Geopolymer cement is ground granulated blast furnace slag (GGBS) which is a by-product waste material produced from iron manufacture. Extensive laboratory studies have used (GGBS) in their Geopolymer cement formation [19, 31–35]. Despite the advantages of GGBFS, it is reported that the increase of GGBS percentage results in reducing the initial setting time and the workability of the mixture, which makes the Geopolymer cement inefficient for mixing, transporting and finishing [19, 36]. GGBS supplies a clear source of alumina-silicate, but carries a high price in many markets and so drives up cost [17].

Among others, fly ash has been used in formulating Geopolymer cement [37–41] and these studies proved that fly ash offered similar characters and properties of GGBS in terms of the mechanical and some durability properties but increased the setting time. However, using fly ash can have disadvantages such as more air-entraining agent requirements to give the mix the desired air content when finer fly ash with high carbon content is used. This is in addition to high permeability, cracking and shrinkage, alkali aggregate resistance issues and low carbonation resistance [42, 43]. GGBFS and PFA are the most promising raw materials that have been used in GPC manufacturing due to their high content of (Al and Si) which can be easily activated. However, in reality, there are many obstacles related to the long-term usage of these precursors in GPC production. The most significant issue is the availability in terms of the location and the amount of these materials available in local areas. Recent implications are that there is a high risk related to the production and availability of these main raw materials, which forms the main barrier in implementing GPC [17, 44–47]. Latest work on Geopolymer cement concentrated on using metakaolin (calcined kaolin) which contains pure kaolin or what is called (China clay) [48, 49]. These studies showed that metakaolin based Geopolymer concrete has high compression strength and low permeability. Many Recent studies argued that the use of metakaolin as a binder has many drawbacks such as the difficulty and the cost of extraction and production of metakaolin [50]. In addition, the unfeasibly relating to high water demand due to the high porosity of GPC based metakaolin mix design is a significant negative aspect as well [17, 51]. Other sources of alumina-silicate materials have been examined by several studies to check their suitability as Geopolymer precursors. Biomass ashes are new promising sources of pozzolanic materials rich of alumina and silicate. Some of these materials need to be treated and processed to increase their reactivity. A study [52] investigated the effect of sugar cane straw ash (SCSA) as solid precursor on the alkali-activated binders based on

Table 2. Geopolymer precursor's categories.

Natural pozzolans	All alumina-silicate minerals such as (kaolinite, Bentonite and mining tailings)
Industrial waste (by-products)	Fly ash, bottom as, slags and metakaolin
Municipal waste	Glass waste, paper waste construction waste and air pollution residues (APC)

Table 3. Geopolymer synthesis using different precursors.

Study	Type of precursor	Findings
Chindaprasirt and Rattanasak [53]	fluidized bed combustion (FBC) ash pulverized coal combustion (PCC) fly ash Sodium silicate (Na_2SiO_3) and 10 M sodium hydroxide (NaOH) solutions	high strength Geopolymer mortars of 35.0–44.0 MPa.
Detphan and Chindaprasirt [54]	fly ash (FA) and rice husk ash (RHA) sodium hydroxide (NaOH), sodium silicate	compressive strengths are between 12.5–56.0 MPa and are dependent on the Ratio of FA/RHA, the RHA fineness, and the ratio of sodium silicate to NaOH.
López, Sugita [55]	metakaolin (MK) and rice husk ash (RHA) NaOH and Na_2SiO_3 molar ratio, $Na_2O/SiO_2 = 0.25$	silica obtained from rice husk combustion at 700 °C became to be raw material for geopolymers and significantly contributed to the formation of geopolymer matrix with MK.
Ahmari and Zhang [56]	copper mine tailings and cement kiln dust (CKD) sodium hydroxide (NaOH) (10 and 15 M)	significant improvement of unconfined compressive strength UCS of bricks and durability when CKD is used
Bashar, Alengaram [57]	Palm oil fuel ash (POFA), fly-ash (FA), blast-furnace-slag (BFS) sodium hydroxide (NaOH) solution and liquid sodium silicate (Na_2SiO_3)	The calcium rich-BFS increased compressive strength and 50% POFA with BFS produced 28 day compressive strength of 58 MPa.
Sun, Cui [58]	Waste ceramic and alkali hydroxides and/or sodium/potassium silicate solutions	higher compressive strength of 75.6 MPa after heat treatment of 1000 °C
Vafaei and Allahverdi [59]	Waste-glass powder and calcium aluminate Cements CACs, solutions of sodium hydroxide and sodium silicate with different Na_2O contents as alkali activators	Hydrothermally cured mortars of optimum composition exhibited strengths up to 87 MPa.

blast furnace slag (BFS) and NaOH and a mixture of NaOH + sodium silicate solutions, SiO_2/Na_2O ratios (ε) of 0 and 0.75 respectively, were assessed as activating solutions. They found that (SCSA) worked well with (BFS) in H_2O/Na_2O ratio between 22 and 37. It was noticed that the best compressive strength with solid precursor proportion for BFS/SCSA was 75/25. Table 3 shows number of the studies that used different types of Geopolymer precursors and their findings.

4. Geopolymer strength

The compressive strength of geopolymer cement is a key attribute that varies widely based on raw materials, mix design, and curing conditions. With proper optimization, geopolymer cement can achieve high compressive strengths, making it a viable and sustainable alternative to traditional Portland cement in many construction applications. For specific applications, it is essential to tailor the mix design and curing process to meet the required performance criteria.

In a study by Al-Azzawi et al. [90] on Fly Ash-Geopolymer Concrete (FA-GPC), different fly ashes with varying Si/Al ratios (1.58, 1.66, 2.44, 2.57, and 5.08) were examined. The fly ash with a Si/Al ratio of 1.66 showed the highest compressive strength at 34 MPa compared to other fly ashes, indicating that the chemical composition of fly ash influences the geopolymerisation process and ultimately affects the strength of the concrete mixtures. Furthermore, Singhal et al. [91] found that while the concrete's molarity remained unchanged, adding more fly ash to the mixture enhanced its compressive strength in FA-GPC. For instance, at a molarity of 16 M, the fly ash content increased from 350 kg/m^3 to 375 and 400 kg/m^3, accordingly, at the ambient curing age of 7 days, and at the age of 28 days, the compressive strength improved by 11% and 32%.

5. Geopolymer barriers and limitations

Geopolymer Concrete is generally thought to be an innovative material, which is a sustainable alternative to traditional Portland cement. It is very resistant to several of the aggressive environment conditions such as sulphate and chloride attack [60–62]. It is considered eco-friendly by cutting the greenhouse gases such as the carbon dioxide emissions by 80% when compared to Ordinary Portland cement [18]. A comparison conducted between OPC and GPC has indicated a 70% reduction in CO_2 emissions [63]. It has been reported that Geopolymer Concrete has (44–64)% lower CO_2 emissions than OPC [64]. Geopolymer Concrete strengthens quickly at room temperature and affords compressive strength in the range of 20 MPa after only 4 hours at 20 °C when tested under the conditions applied to hydraulic binder mortars. After 28 days, compressive strength is in the range of 70–100 MPa [65, 66]. Geopolymer Concrete will cure more rapidly than Portland concretes as it may cure at ambient temperature and gains most of its strength within 24 hours [38, 67]. Furthermore, Geopolymer Concrete has very high resistance to elevated temperatures, particularly when incorporating rich (Al and Si) raw materials [68]. Its thermal performance is dependent on the concentration of the alkaline activator (KOH, NaOH, etc.) within Geopolymer Concrete [69, 70].

On the other hand, the activator solutions of alumina-silicate material used to form the geopolymerisation represents a real concern with Geopolymer Concrete. The activator solutions provide the highest single contribution to the embodied carbon dioxide of Geopolymer Concrete [64, 71, 72]. The activator solutions chemicals derive from procedures that make intensive use of resources and energy. The economic analyses of Geopolymer Concrete have shown that the sodium silicate and potassium silicate activators contribute significantly to the total production costs of Geopolymer Concrete [44, 73, 74]. The main difficulty with Geopolymer technology is the use of liquid alkaline activating solutions, which are categorized as extremely corrosive materials. From an operational viewpoint, they are difficult and expensive to handle with significant occupational health and safety concerns [75].

The efflorescence problem is another example of the unwanted issues associated with Geopolymer Concrete. Zheng, Van Deventer [76] realised such issues and the alkaline-silicate solutions cannot be entirely consumed throughout the Geopolymerisation process. This causes severe efflorescence with high permeability and water absorption due to the movement of alkali together with water to the geopolymer surfaces. Despite this, in the several hundred alkaline activated concrete binders-related papers published in Scopus/Elsevier journals, less than ten addressed in some way the efflorescence problem and of those, only three focused directly on the problem [77].

Furthermore, other barriers include the untrusted availability of the solid raw materials of Geopolymer. For instance, in UK there will be substantial decrease in the produced amounts of pulverized fly ash (PFA) and slags in the next 10 years due to the UK's new regulations regarding the decrease of the greenhouse gas releases by at least 80% by 2050 [47].

Past research [78, 79] listed several technical and commercial barriers that are facing Geopolymer technology;

- The complexity of alumina-silicate materials, the final product gel chemistry synthesis and the phase formation compared to the C-S-H gel in OPC.
- Absence of an in-service record of accomplishment equivalent in scale and stability of OPC.
- Lack of research to validate durability-testing methodology and improve Geopolymer cement technology.
- Lack of understanding micro/Nanostructure of Geopolymer gel.
- The difficulty of designing the alkali activators compositions.
- Technical challenges in scaling up-Geopolymer synthesis from laboratory to the real-world and construction industry.
- Geopolymer commercialisation requires vast regulatory, asset management, liability and industry stakeholder engagement process.
- Variability in raw materials properties and characterisations.
- Safety risk associated with alkalinity of activating solution.

A report created by Low Carbon Living CRC [80] in Australia about the barriers and the pathways to overcome Geopolymer commercialsation. The report includes survey about questions for various positions such as (Materials supplier, academic researcher, contractor . . .etc.). The questions were:

1) What barriers for implementation of Geopolymer concrete?
2) What are the suggested pathways to overcome the limitations of Geopolymer?

It was noticed that “not covered in standards” and “lack of standard and specifications” form the highest barriers for Geopolymer widespread with (62.5 and 60%) respectively. Lack of long-term performance data has high responses as well with 60%. Lack of awareness of the mix designs and the details of Geopolymer formed (50%). Suggested pathways were to develop standard specifications (65%) followed by further research on properties and durability (55%). Other significant pathways includes to reduce the cost and increase the availability had (45 and 42.5%) respectively.

The several factors that can affect the properties of the resulted (GPC) is a noteworthy concern that limit the use of this cement. Factors including the dosage of alkaline activators, ratios of the reactive silicate in the raw materials, the mineralogy of the raw materials are the main influent factors on the Geopolymer properties. Therefore, the difficulty in creation and the require of special handling as it has chemicals as essential part in the synthesis process. This fact reveals the reason of only pre-cast utilisation of (GPC).

6. Future research trends

Novel research are being developed to reduce the obstacles associated with Geopolymer technology. One of the trends involves examining alternatives potentials Geopolymer precursors. Moreover, with the production decline and the unexpected availability of PFA and GGBS in next few years [47], the need for other source of raw materials is substantial issue. Non-ferrous slags such as copper and nickel slags are potential alternatives that recently took great consideration in research field [81–83]. Other trend of research include using alternative activators instead the conventional alkaline solutions. Alternatives could include waste with alkaline content such as industrial glass waste [84–86] or waste from the bauxite residues which is called red mud [87].

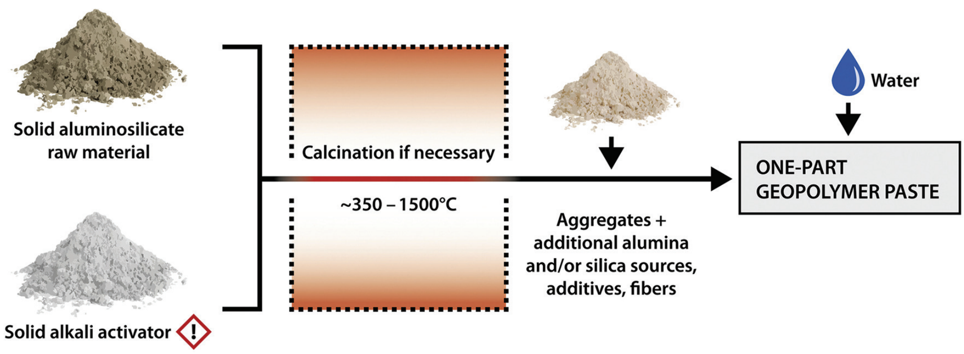

Fig. 2. One part geopolymer synthesis [17].

6.1. One-part Geopolymer system

Conventional (two-part) Geopolymer are shaped by a reaction between a concentrated solution of alkali hydroxide, silicate and solid alumina-silicate precursor, that is, two parts in addition to water [88]. However, the inconveniencies linked to control huge quantities of glutinous, corrosive, and dangerous alkali activator solutions has put force on the growth of one-part or "just add water" Geopolymer that could be used similarly to OPC [17]. In one-part mixtures, only a dry mixture is needed in addition to water. The dry mixture is prepared by mixing a solid alkali-activator with a solid alumina-silicate precursor with or without a calcination step Fig. 2.

One part Geopolymer is a great step in the widespread Geopolymer utilisation especially for in situ applications where handling alkali solutions can be difficult whereas two-part mixtures appear suitable for precast work [89].

7. Conclusions

Based on the recent studies that dealt with the development of Geopolymer cement, the following conclusions can be extracted:

1. Geopolymer cement is different from alkali-activated materials as there is nothing to activate in Geopolymer but there is alkalination as it does not has N-A-S-H and K-A-S-H products.
2. Most of the current Geopolymer use is just in the pre-cast applications such as airports runway and retaining walls.
3. Several factors can affect the kinetic of Geopolymerisation including Si/M, Si/Al, SiO_2/M_2O and Al_2O_3/M_2O.
4. Variability in the properties of Geopolymer precursors forms the biggest concern that limit the use of Geopolymer cement.
5. No warranty in the long-term availability of main Geopolymer raw materials such as (GGBS) and (fly ash) and other materials which in terms form substantial barrier opposite Geopolymer.
6. More research about the properties of alternative precursors such as biomass waste, natural pozzolanic and non-ferrous slags materials is needed to identify their suitability for this technology.
7. The main difficulty with Geopolymer technology is the use of liquid alkaline activating solutions, which are categorized as extremely corrosive materials. From an

operational viewpoint, they are difficult and expensive to handle with significant occupational health and safety concerns.

8. The efflorescence problem is another example of the unwanted issues associated with Geopolymer Concrete.
9. Other barriers include lack of standard and specifications, long-term durability reports and absence of understanding the micro/Nano-structure of Geopolymer gel.
10. Novel research trends involve alternative activators and friendly user instead the conventional hostile alkaline solutions and the development of one-part Geopolymer cement that just need water to create started to take great consideration in the research field.

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Conflict of interest

The authors declare that there is no conflict of interest.

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