Review Article

Geopolymer Binders: A Need for Future Concrete Construction

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Applications of polymer based binder material can be an ideal choice in civil infrastructural applications since the conventional cement production is highly energy intensive. Moreover, it also consumes significant amount of natural resources for the large-scale production in order to meet the global infrastructure developments. On the other hand the usage of cement concrete is on the increase and necessitates looking for an alternative binder to make concrete. Geopolymer based cementitious binder was one of the recent research findings in the emerging technologies. The present study is aimed at providing a comprehensive review on the various production processes involved in the development of a geopolymer binder. More studies in the recent past showed a major thrust for wider applications of geopolymer binder towards a cost economic construction practice. This also envisages the reduction of global warming due to carbon dioxide emissions from cement plants.

1. Introduction

Research studies in the past had shown that fly ash-based geopolymer has emerged as a promising new cement alternative in the field of construction materials. The term geopolymer was first coined and invented by Davidovits [1] which was obtained from fly ash as a result of geopolymerization reaction. This was produced by the chemical reaction of aluminosilicate oxides (Si_2O_5 , Al_2O_3) with alkali polysilicates yielding polymeric Si–O–Al bonds. Hardjito and Rangan [2] demonstrated in their extensive studies that geopolymer based concrete showed good mechanical properties as compared to conventional cement concrete. A comprehensive analysis on the various works done in geopolymer concrete is listed in Table 1.

Geopolymer can be produced with the basic raw materials containing silica and alumina rich mineral composition. Several studies have reported the use of the beneficial utilization of these materials in concrete. Most of the studies investigated the use of alkali activators containing sodium hydroxide and sodium silicate or a potassium hydroxide and potassium silicate. Cheng and Chiu [3] reported the production of geopolymer concrete using slag and metakaolin with potassium hydroxide and sodium silicate as alkaline

medium. Palomo et al. [4] produced geopolymers using fly ash with sodium hydroxide and sodium silicate as well as with potassium hydroxide with potassium silicate combinations. The results from the studies exhibited an excellent formation of geopolymer with rapid setting properties. It can be noted that the presence of calcium content in fly ash played a significant role in compressive strength development [5]. The presence of calcium ions provides a faster reactivity and thus yields good hardening of geopolymer in shorter curing time.

2. Background of Geopolymerization Process

Polymerization reaction is best observed in the presence of alkaline medium such as sodium hydroxide, or potassium hydroxide and the addition of silicates can be additional ionic composition with good bonding effects. The reactants in the chain reaction can be accelerated due to higher molar concentration of alkali ions; however, the increase in the concentration leads to rapid loss in consistency during mixing attributed to faster polymer reaction. The inclusion of sodium silicate in sodium hydroxide solution provides higher silicate content and due to which the gel formation is likely to provide faster polymerization. A similar reaction is observed in the case of potassium silicate added to potassium

TABLE 1: Summary of various works done on geopolymers concrete.

Sl. no.	Authors/ref.	Year	Test conducted	Types of binder and alkali activator used	Curing regime	Observations
(1)	Goretta et al. [6]	2004	Compressive strength.	Class C fly ash and granulated blast-furnace slag, sodium silicate.	Hot air oven curing at 80°C to 120°C and ambient temperature.	The response was attributed to material loss by propagation of both lateral and radial cracks and presence of microcracks and aggregates in the matrix.
(2)	Bakharev [7]	2005	Compressive strength, FTIR, XRD, and SEM.	Class F fly ash, Sodium silicate and sodium hydroxide. Potassium hydroxide	Hot air oven at 75°C to 150°C.	An increase of temperature of heat treatment caused a decrease of Si/Al ratios in aluminosilicate gel, and long curing at room temperature narrowed the range of distribution of the Si/Al ratios.
(3)	Bakharev [7]	2005	Compressive strength.	Class F fly ash, Sodium silicate and sodium hydroxide.	Hot air oven curing at 75°C and 95°C.	Fly ash activated by sodium silicate, 6 h heat curing is more beneficial than 24 h heat. Fly ash activated by sodium hydroxide had more stable strength properties.
(4)	Fernández-Jiménez et al. [8]	2005	Compressive strength.	Class F fly ash and sodium hydroxide solution.	Hot air oven curing at 80°C.	The particle size distribution and the mineral composition of the starting fly ash, the type and concentration of the activator, and so forth.
(5)	Duxson et al. [9]	2005	Compressive strength.	Metakaolin. Sodium silicate and sodium hydroxide solution.	Hot air oven curing at 80°C.	This demonstrates that the characteristics of geopolymers can be tailored for applications with requirements for specific microstructural, chemical, mechanical, and thermal properties.
(6)	Bakharev [10]	2006	Compressive strength, shrinkage measurements, XRD, and SEM.	Fly ash. Sodium silicate and sodium hydroxide. Potassium hydroxide	Hot air oven at 100°C.	Geopolymer materials prepared using class F fly ash and sodium and potassium silicate show high shrinkage as well as large changes in compressive strength with increasing fired temperature in the range 800–1200°C.
(7)	Škvára et al. [11]	2006	Compressive strength.	Fly ash and ground blast-furnace granulated slag. Sodium hydroxide.	Hot air oven curing at 100°C–120°C.	The hardness of geopolymer is approximately twice higher than for OPC that could indicate less deformability and higher brittleness.
(8)	Chindaprasirt et al. [12]	2007	Compressive strength.	Lignite fly ash (FA) Sodium silicate and sodium hydroxide solution as alkali activators.	Hot air oven curing at 120°C.	The samples with a high strength were obtained using the delay time after molding and before subjecting the sample to heat of 1 h with heat curing in the oven at 75°C of not less than two days.
(9)	Kong et al. [13]	2007	Compressive strength.	Metakaolin and low-calcium fly ash. Grade D sodium silicate solution and potassium hydroxide.	Hot air oven curing at 100°C.	Fly ash pores contain higher proportion of microspores than metakaolin geopolymer. Fly ash-based geopolymer gives better strength than metakaolin.
(10)	Temuujin et al. [14]	2009	Compressive strength.	Fly ash. Sodium silicate and sodium hydroxide solution.	Hot air oven curing at 75°C and 100°C.	Addition of the calcium compounds CaO and Ca(OH) ₂ improves mechanical properties and cured at ambient temperature. Calcium compound addition reduces mechanical properties cured at elevated temperatures.

TABLE I: Continued.

Sl. no.	Authors/ref.	Year	Test conducted	Types of binder and alkali activator used	Curing regime	Observations
(11)	Kong and Sanjayan [15]	2008	Compressive strength.	Low-calcium (class F) fly ash. Sodium silicate solution and potassium hydroxide.	Hot air oven curing at 80°C.	The strength declined with inclusion of geopolymer/aggregate composites. While aggregates undergo expansion at elevated temperatures, the geopolymer matrix experienced contraction.
(12)	Diaz et al. [16]	2010	Compressive strength.	Class F fly ash. Sodium silicate and sodium hydroxide solution.	Hot air oven curing at 80°C.	Higher amount of fine particles will result in higher surface area, higher reactivity resulting in higher compressive strength.
(13)	Kong and Sanjayan [17]	2010	Compressive strength.	Class F fly ash. Sodium silicate and sodium hydroxide.	Hot air oven curing at 100°C.	The rate of expansion of the aggregate with temperature is an influential factor in the performance of geopolymer concrete under elevated temperatures.
(14)	Kumar et al. [18]	2010	Compressive strength. FTIR, XRD, and SEM.	Fly Ash. Sodium hydroxide.	Hot air oven at 100°C to 250°C.	Combined effect of particle size and change in reactivity due to mechanical activation altered the geopolymerisation reaction. The improvement in physical properties is related to the intrinsic structure developed due to enhanced geopolymerisation.
(15)	Wongpa et al. [19]	2010	Compressive strength.	Fly ash and rice husk bark ash. Sodium silicate and sodium hydroxide solution.	Hot air oven curing at 75°C to 125°C.	Paste content and the aggregate content P/Aggregate of 0.34 and Si/Al of 0.63 showed the highest compressive strength.
(16)	Jämstorp et al. [20]	2010	Compressive strength.	Kaolin ($\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$), fumed silica. Metakaolin and sodium hydroxide (NaOH). Fentanyl base and Zolpidem tartrate.	Hot air oven curing at 100°C to 150°C.	Samples with pore sizes of about 40 nm, exhibited a satisfying initial release of 60–80% of the API content within 10 h and nearly all within 24 h, as well as fairly high compression strengths of 50–60 MPa.
(17)	Elimbi et al. [21]	2011	Setting time, linear shrinkage, compressive strength, XRD, and SEM.	Metakaolin, kaolinite, and sodium hydroxide and sodium silicate.	Calcined at 450°C and ambient temperature.	Above 700°C, there is an increase of setting time. The compressive strength increases when the calcination temperature of kaolinite clays is between 500 and 700°C but drops above 700°C.
(18)	Natali et al. [22]	2011	Flexural strength and fracture toughness.	Metakaolin, ladle slag, and sodium hydroxide and sodium silicate.	Calcined at 700°C for 5 hours.	Geopolymer matrix is able to determine a flexural strength increment, ranging from 30% up to 70% depending on the fiber type, compared to the unreinforced material.
(19)	Nazari et al. [23]	2011	Compressive strength.	Seeded fly ash and rice husk bark ash. Sodium silicate and sodium hydroxide.	Hot air oven at 80°C.	The highest strength was achieved using a 12 M NaOH solution. Oven curing of the specimens at 80°C was found to be the optimum temperature.

TABLE I: Continued.

Sl. no.	Authors/ref.	Year	Test conducted	Types of binder and alkali activator used	Curing regime	Observations
(20)	McLellan et al. [24]	2011	Compressive strength.	Comparative study of OPC and fly ash. Sodium silicate and sodium hydroxide solution.	Hot air oven at 100°C.	There is an estimated 44–64% improvement in greenhouse gas emissions over OPC. Emissions from geopolymer concrete can be 97% lower up to 14% higher.
(21)	Somna et al. [25]	2011	Compressive strength.	Fly ash. Sodium silicate and sodium hydroxide solution.	Hot air oven at 100°C.	Sodium hydroxide-activated ground fly ash cured at room temperature can be produced with reasonable strength. Ground fine fly ash can be used as a source material for making geopolymer cured at ambient temperature.

FIGURE 1: Polymerization reaction [1].

hydroxide solution. It is known that the conventional organic polymerization involves the formation of monomers in a given solution in which the reaction can be made faster to polymerize and form a solid polymer. The geopolymerization process involves three separate processes and during initial mixing, the alkaline solution dissolves silicon and aluminium ions in the raw material (fly ash, slag, silica fume, bentonite, etc.). It is also understood that the silicon or aluminium

hydroxide molecules undergo a condensation reaction where adjacent hydroxyl ions from these near neighbors condense to form an oxygen bond linking the water molecule, and it is seen that each oxygen bond is formed as a result of a condensation reaction and thereby bonds the neighboring Si or Al tetrahedra. A clear representation of the chain reaction involved during the polymerization is explained in Figure 1 with a fundamental understanding from the literature.

Polymers are sensitive towards heat and can form a stronger chain due to polycondensation. It is noted from the basic chemical reaction when subjected to heat causes silicon and aluminium hydroxide molecules to polycondense or polymerize, to form rigid chains or nets of oxygen bonded tetrahedra. Also, at higher elevated temperatures it produces stronger geopolymers. Aluminium ions require a metallic Na^+ ions for charge balance. Davidovits and Davidovics [26] reported that geopolymers can harden rapidly at room temperature and can gain the compressive strength up to 20 MPa in 1 day. Comrie et al. [27] conducted tests on geopolymer mortars and reported that most of the 28-day strength was gained during the first 2 days of curing. Geopolymer cement is found out to be acid resistant, because, unlike the Portland cement, geopolymer cements do not depend on lime and are not dissolved by acidic solutions. Most of the studies concluded that the concentration of NaOH solution plays the most important role on the strength of the fly ash-based geopolymers. The addition of calcium oxide along with sodium hydroxide accelerates the geopolymerisation in fly ash. Guo et al. [28] conducted experimental studies in class C fly ash-based geopolymers using a mixed alkali activator of sodium hydroxide and sodium silicate solution. It was reported that a high compressive strength can be obtained when the molar ratio of silicate to sodium is 1.5, and the mass proportion of Na_2O to class F fly ash was 10%. The compressive strength of these samples was around 63 MPa when it was cured at 75°C for 8 h followed by curing at 23°C for 28 d.

Low-calcium fly ash is preferred than high calcium (ASTM class C) fly ash for the formation of geopolymers, since the presence of calcium in high amount may affect the polymerization process [29]. The suitability of different types of fly ash can be a potential source for studying the type and efficiency of geopolymerization reaction. It was also reported that geopolymerisation reaction can be effective in low-calcium fly ash depending on if it contains unburnt carbon less than 5% and 10% CaO content, reactive silica about 40–50%, and particles finer than 45 microns [30]. However, it was reported by Van Jaarsveld et al. [5] that fly ash with higher amount of CaO produced higher compressive strength, due to the formation of calcium-aluminate hydrate and other calcium compounds, especially in the early ages. The most preferred alkaline solution used in geopolymerisation is a combination of sodium hydroxide (NaOH) or potassium hydroxide (KOH) and sodium silicate or potassium silicate [4, 31–35].

Palomo et al. [4] reported that reactions occur at a high rate when the alkaline liquid contains soluble silicate, either sodium or potassium silicate, compared to the use of only alkaline hydroxides. Xu and van Deventer [33] confirmed that the addition of sodium silicate solution to the sodium hydroxide solution as the alkaline liquid enhanced the reaction with fly ash. Furthermore, geopolymerisation with the NaOH solution resulted in higher dissolution of minerals than KOH solution. A combination of sodium hydroxide and sodium silicate solution, after curing the specimens for 24 hours at 65°C , provided higher strength [33]. It was reported that the proportion of alkaline solution to aluminosilicate

powder by mass should be approximately 0.33 to allow the geopolymeric reactions to occur. Alkaline solutions formed a thick gel instantaneously upon mixing with the aluminosilicate powder. The previous studies also reported that mixtures with high water content, that is, $\text{H}_2\text{O}/\text{Na}_2\text{O} = 25$, developed very low compressive strengths. Palomo et al. [4] reported that curing temperature is an important indicator for strength gain in fly ash-based geopolymers and improves the mechanical strength. Higher curing temperature and optimum curing time were found to influence the compressive strength gain in geopolymer concrete. Alkaline liquid that contained soluble silicates was proved to increase the rate of reaction compared to alkaline solutions that contained only hydroxide.

3. Long Term Durability Properties of Geopolymer Concrete

Durability aspects of geopolymer products have good sustainability to weathering effects; however, they are not resistant towards high temperature beyond 400°C . Several experimental studies showed that geopolymer concrete specimens immersed in sulfuric acid and chloric acid were found to be resistant to acid attack. While the Portland based cement showed deleterious reaction and results in surface deterioration followed by weight loss (Davidovits, 1994). Extensive studies also demonstrated that heat-cured fly ash-based geopolymer concrete has an excellent resistance to sulfate attack due the formation of stronger polymer chain due to polycondensation reaction. The effects of acid attack also cause reduction in compressive strength of heat-cured geopolymer concrete; the extent of degradation depends on the concentration of the acid solution and the period of exposure. However, the sulfuric acid resistance of heat-cured geopolymer concrete is significantly better than that of Portland cement concrete as reported in earlier studies.

Several studies have shown that fiber addition is an effective method to improve the mechanical characteristics of brittle material such as concrete by providing crack arresting mechanism [36]. Limited studies have been carried out to analyze the effect of fibre reinforcement in geopolymer concrete. Future studies are needed to study the effect of steel and glass fibres in geopolymer concrete to be investigated systematically. Also, it is well known that increase in fracture toughness is provided essentially by fiber bridging near the crack opening prior to crack propagation. The linear elastic behavior of the matrix could not be affected significantly for low volumetric fiber fractions. However, postcracking behavior can be substantially modified, with increases of strength, toughness, and durability of the material. The future study has to be focussed on the effect of fibre addition on the postcrack performance of geopolymer concrete.

4. Summary

It is understood from the earlier studies that good scientific information is available on the evaluation of chemical and physical properties of geopolymer concrete. Also, very few works has been reported on the effect of fibre reinforcement

in geopolymer concrete. Further studies are needed to investigate the fracture resistance of this brittle composite. The addition of glass fibres can exhibit a reasonable improvement on the strength properties of geopolymer concrete due to strain hardening properties at failure. The concentration and type of alkali need to be investigated extensively to choose the combination and dosage of alkali for fly ash. The effect of alkali activators on the rate of hardening of geopolymers at different curing regimes needs to be well documented. Curing regime on the hardening properties of geopolymeric concrete needs special attention to improve the strength properties. The rate of strength gain in different curing regimes needs to be explored using ultrasonic pulse velocity measurements. The mechanical characteristics of geopolymer concrete specimens at elevated temperature (600–800°C) need to be assessed for checking its potential applications as heat resisting construction material.

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