

Geo-polymer Concrete as a New Type of Sustainable Construction Materials

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ABSTRACT

The development of alkali activated binders with superior engineering properties and longer durability has emerged as an alternative to OPC. In the near future, geopolymers or alkali-activated cementitious materials as a new construction material will be to use high performance materials of low environmental impact that are produced at reasonable cost. These materials can be replaced to the binder in concrete as a major construction material. Geo-polymer materials are inorganic polymers based on alumina and silica units, they are synthesized from a wide range of de-hydroxylated alumina-silicate powders condensed with alkaline silicate in a highly alkaline environment. Using lesser amounts of calcium-based raw materials, lower manufacturing temperature and lower amounts of fuel, result in reduced carbon emissions for geopolymer cement manufacture up to 22%-72% in comparison with Portland cement. Geopolymer concretes, its specifications, benefits, applications and evaluation of carbon footprint and cost have been studied in this paper.

Keywords: geopolymer concrete, alkali activated binders, alumina-silicate, and sustainable construction material

1. INTRODUCTION

Concrete is a durable construction material produced by mixing Portland cement, water, aggregates and additives with special proportion. Revising the ingredients and production method of conventional concrete is important with respect to high consumption of concrete as a construction material. High consumption of concrete causes vast requirements of cement production. Portland cement is used as a binder in the production of concrete and is produced by mixing selected raw materials (clay and lime) with a given proportion, grinding and heating it at 1500°C. In the process of producing 1 tone cement, 125 lit fossil fuel and 118 KWH electricity is consumed while cement transportation is 11.9 percent of the country roads' transportation (Vazinram and Khodaparast, 2009). Unfortunately, the production of Portland cement as a major construction material worldwide releases large amounts of CO₂ in to the atmosphere (production of 1 tone OPC releases 1 tone CO₂), and this gas is a major contributor to the greenhouse effect and the global warming planet (Bilodeau and Malhotra, 2000). The main benefit of geopolymeric cement is reduction in environmental impacts to move toward sustainable development which is defined as the optimum usage with correct and efficient operation of basic and natural resources

for providing the requirements of the future generation. This includes less emission of carbon dioxide; a more efficient use of mineral and metal resources; increase the use of recycled materials and issue the long-term durability by the mechanical properties of these novel materials studied in relation archeology analogues, namely ancient Roman cement. More than 65% of the crust of the earth consists of Al-Si minerals and use of lesser amounts of calcium-based cement do not release large quantities of CO₂, so it is most useful to understand how these minerals convert to cement (Davidovits, 1994). For geo-polymeric cement production, no temperature higher than 750 degree centigrade is ever needed. The above item means only one third of the fuel requirement is needed for this kind of cement production. Therefore geo-polymeric cements don't rely on the calcinations of calcium carbonate, not require high temperature kilns, with large expenditure of fuel, so the chemical process and fuel much less reduce the carbon dioxide production for geo-polymeric cement manufacture. Thus a good solution for producing an environmental friendly type of concrete is to reduce the amount of Portland cement and replacing it with material which has lower manufacturing temperature such as geopolymer cement. This paper presents a review on the geopolymer structure, setting and hardening mechanism, engineering properties, evaluation of carbon footprint and cost of geopolymer concrete while mentions some of its applications.

2. GEOPOLYMERS AND MINERAL RESOURCES FOR PRODUCING GEOPOLYMER CEMENT

Geopolymer was invented by Davidovits in 1979 as a 3 dimensional alumina silicates while the idea came out of the Great Pyramid Mystery. Davidovits states that supplementary cementing materials which are coal and lignite fly ash, rice husk ash, palm oil fuel ash, other ashes, blast furnace and steel slag, silica fume, limestone, metakaolin, natural pozzolan can produce geopolymer with mineral and metal resources. Although the tendency in world electricity production is the opposite of implementing more and more coal-fuelled power plants, until year 2015 a maximum amount of 290 million tones of fly ash about 8% of the worlds cement market would be available as a mineral resource for geopolymer cement applications (Davidovits, 1994). Quenched vitreous slag as a metal resource could represent the availability of respectively 290 to 560 million tones of blast slag in the year 2015 for cement production (Davidovits, 1994). Metakaolin is a highly pozzolanic material produced by calcinic China clay at a temperature of 700-900c (EN 206/BS 5328). Metakaolin is a product, which is manufactured for use rather than by-product, and the process of producing this material seems similar to geopolymers.

3. CLASSIFICATION OF GEOPOLYMERS STRUCTURES

Geopolymers include three classifications of inorganic polymers which depend on the ratio of Si/Al in their structures:

- a) Poly (sialite) (-Si-O-AL-O-)
- b) Poly (sialate-siloxo) (-Si-O-Al-O-Si-O-)
- c) Poly (sialate-disiloxo) (-Si-O-Al-O-Si-O-Si-O-)

The distribution and relative amounts of each of the different Al and Si building blocks affect the chemical and physical properties of the final product.

4. GEOPOLYMERIZATION OF PRIMERY MONOMERS

The three main steps in the process of geopolymerisation are:

- Dissolution of any pozzolanic compound or source of silica and alumina that is readily dissolved in alkaline solution, with the formation of mobile precursors of alumino-silicate oxides through the complex action of hydroxide ions.
- Partial orientation of mobile precursors as well as the partial internal restructuring of the alkali poly-sialates.
- Re-precipitation of the particles from the initial solid phase where the whole system hardens to form an inorganic polymeric structure.

Poly (sialates) as the resulted material has the empirical formula as below: $M_n \{-(SiO_2)_z-AlO_2\}_n, wH_2O$

Wherein M is a cation such as potassium, sodium or calcium, and «n» is a degree of poly-condensation; «z» is 1, 2, and 3. Poly (sialates) is chain and ring polymers with Si^{4+} and Al^{3+} in IV-fold coordination with oxygen (Davidovits, 1991).

5. SETTING AND HARDENING MECHANISM FOR GEOPOLYMERIZATION

Geopolymeric materials are synthesized by activation of alumina silicate materials with alkaline silicates or hydroxides at ambient to maximum 120°C temperatures. Geopolymers have amorphous to semi crystalline structures with nano particle size depend on curing temperatures. Alumino silicates are used in geopolymer synthesis are kaolin, metakaolin, fly ash and metallurgic slag such as blast furnace slag and activators solutions are sodium and potassium silicates which can be used with sodium and potassium hydroxides. When alumina silicates resources are adjacent to alkali solutions, dissolution and precipitation of Al and Si complex begin. The dissolution is affected by alkaline solution consistency, type of alkaline cation, the speed of stirring, and the structures and chemical compositions of alumina silicate. Among these factors the specification of alumina silicate source material and the condensation of alkaline solution are the most important ones. After dissolving alumina silicate particles from their surfaces, precipitation of gel begins and Al and Si complex penetrates in to gel phase. Thus the density of Al and Si complex reduces in the surface of alumina silicate particles and therefore dissolving of Al and Si increases. During the penetration process, the duration and intensity of stirring are the governing factors. Whereas more activation energy is needed for formation of Si-O-Si than Al-O-Si, polymerization of Al and Si complex occurs first. Therefore condensation of Al and Si complex from alumina silicate sources, dissolution and penetration of it will occur at the same time. The factors affect on condensation are temperature, pH and cation size. Higher temperature, higher pH (higher condensation of alkaline solution) and alkaline cation with bigger atomic size make condensation mechanism faster. Geopolymer compositions are developed crystalline and amorphous in this way are used for industrial applications. Crystalline PS and PSS are produced due to exothermal reaction while geopolymer setting at ambient temperature causes an amorphous structure.

6. GEOPOLYMER CONCRETE MIX DESIGN

In geopolymer concrete, aggregates are bound by binder which is composed from two parts

including aluminosilicates and alkali solution and named geopolymer binder. Mix proportioning is based on determining the quantities of the ingredients, when mixed together and cured properly will produce workable concrete that achieves the desired strength and durability when hardened. Therefore different variables including desired workability measured by slump, water to binder ratio, binder content and aggregate proportions should be considered in the mix design procedure. In geopolymer concrete based on alkali activated fly ash the weight ratio of alkali solution to fly ash is suggested in the range of 0.3 to 0.45 while Table 2 shows a good guideline for selecting target workability and compressive strength (Costa et al., 2007).

Table1. Guideline of selecting geopolymer mix design

Compressive Strength (Mpa)	Workability	Mass ratio of water to polymeric material in solid form
60	Very low	0.16
50	Low	0.18
40	Normal	0.70
35	Fluid	0.77
30	Fluid	0.74

7. GEOPOLYMER CONCRETE CASTING AND CURING

The equipments needed for geopolymer concrete production are the same as OPC concrete. Usually for casting this type of concrete, fundamental materials such as fly ash and aggregates are mixed and alkaline solution with additives are added to it. Curing at elevated temperature helps the reaction of the paste in geopolymer concrete. Curing time and temperature have an effective role on compressive strength of geopolymer concrete. Two curing conditions can be considered which are fog and sealed conditions. It is shown that sealed condition shows around 15% more compressive strength than samples are cured at steam condition (Bondar et al., 2009 and 2011).

8. PROPERTIES OF FRESH GEOPOLYMER CONCRETE

A geopolymer mix can be timed to set either fast or slow, by adjusting the mixture components. Depending on the synthesis conditions, structural integrity and reasonable strength were attained in a short time, sometimes in as little as sixty minutes (Van Jaarsveld et al., 1997). Using granulated blast furnace slag as the source material with the addition of metakaolinite, Cheng and Chiu (2003) found that the setting time of the geopolymer paste was affected by curing temperature, type of alkaline activator and the composition of source material. They stated that the setting time of above geopolymer paste was between 15 to 45 minutes at 60°C. The time available between the end of mixing and the start of casting of fresh geopolymer concrete may be more relevant in practical applications and measuring the setting time at elevated temperatures may not be appropriate. The laboratory experience by Hardjito et al. (2003a) showed that the fresh geopolymer concrete could be handled for at least 120 minutes after mixing, without any sign of setting and degradation in compressive strength. These results depended on the

composition of the source material, higher CaO contents giving faster setting. The presence of components other than Al_2O_3 and SiO_2 in the source material may also delay the setting. In materials of pure geological origin (say calcined kaolin), the dominant chemical contents are only Al_2O_3 and SiO_2 , whereas byproduct materials such as fly ash may contain other compounds e.g. Fe_2O_3 . Therefore, it appears that pure geological materials may be more reactive with alkaline activators and induce a reduction in initial setting time (in Hardjito et al, 2004). Fly ash based geopolymers showed faster initial setting time at higher temperatures with the final setting of these mortars occurring 15 to 25 minutes after the initial setting (Costa et al., 2007). In natural pozzolans, the higher the amount of silicate concentration the longer the setting time, unless the silicate source consists of minerals such as opal which reacts rapidly with an aqueous alkaline solution (Bondar, 2009).

9. PROPERTIES OF HARDENED GEOPOLYMER CONCRETE

There are many different views as to which are the main parameters that affect the compressive strength and other mechanical properties of geopolymer concrete. Palomo et al. (1999) stated that the significant factors affecting the compressive strength are the type of alkaline activator, the curing temperature and the curing time (in Hardjito et al, 2004a, 2004c). However, other researchers have reported that the important parameters for satisfactory polymerization are the relative amounts of Si, Al, K, Na, molar ratio of Si to Al present in solution, the ratio of alumina silicate mineral to kaolinite (when kaolinite is added), the type of alkaline activator, the water content, and the curing temperature (Xu and Deventer, 2000, Barbosa et al., 2000, Rowles et al., 2003). The presence of silicate ions in the alkaline solution substantially improves the mechanical strength and modulus of elasticity values but has a slightly adverse effect on the otherwise very strong matrix/aggregate and matrix/steel bond (Fernandez-Jimenez et al, 2006). Experimental results show that the $\text{H}_2\text{O}/\text{M}_2\text{O}$ molar ratio in the mixture composition significantly affects the compressive strength of fly ash based geopolymer concrete, whereas the influence of the $\text{Na}_2\text{O}/\text{SiO}_2$ molar ratio is less significant (Hardjito et al, 2004a, 2004c). An increase of the $\text{H}_2\text{O}/\text{M}_2\text{O}$ molar ratio and water to geopolymer solids ratio decrease the compressive strength of geopolymer (Hardjito et al, 2004a, 2004c).

In addition, Van Jaarsveld et al. (2002) found that curing at elevated temperatures for long periods of time may weaken the structure of hardened material. The research on fly ash-based geopolymer binder, Palomo et al. (1999b) has confirmed that curing temperature and curing time significantly influence the compressive strength but this does not seem to be same for different aluminosilicates. Longer curing time and higher curing temperature increased the compressive strength in fly ash based geopolymer concrete, although the increase in strength may not be as significant for curing at more than 60°C and for periods longer than 48 hours (in Hardjito et al, 2004a, 2004c). In most cases, 70% of the final compressive strength is developed in the first 4 hours of setting. Because the chemical reaction of the geopolymer paste is a fast polymerization process, the compressive strength does not vary greatly with the age of concrete, after it has been cured for 24h. This observation is in contrast to the well-known behaviour of OPC concrete, where the hydration process extends over a long time period and hence strength increases over time (in Hardjito et al, 2004a, 2004c). Another kinetic difference between Portland cement and alkaline activated systems is the existence of a relatively low threshold temperature in the former, above which thermal curing can have an adverse effect on the mechanical development and even on material durability.

For an activated ash, on the contrary, a suitable choice of reaction time and curing temperature can yield a different reaction products without detracting from material durability, because according to Fernandez et al. (2006) increases in the curing temperature go hand-in-hand with decreases in the amount of Al incorporated into the final product and a concomitant improvement in mechanical properties. Such improvements parallel the formation of a homogeneous aluminaosilicate matrix (Fernandez-Jimenez et al., 2006). The properties of geopolymers based on different alumina silicates are summarized as below:

9.1. Alkali activated slag

Alkali activated slag cements have been known for about four decades and usually for activation 2-7 percent Na₂O or 3-10 percent K₂O of the slag content is necessary. This kind of concrete has shown progressive gain of strength from 21Mpa in 3days to 36Mpa in 1 year and to 40Mpa in 6years (for a concrete mix having binder content 350Kg/m³, binder to aggregate ratio of 1:5.96, water to binder ratio of 0.46 and MSA of 25mm) (Chatterjee, 2003). When alkali-activated slag cement concrete is cured in water, compressive strength of the concrete keeps increasing until 365 days. However, if the concrete is cured in a sealed condition, the strength stopped increasing at about 90 days. This may be attributed to the lack of moisture available for the hydration of slag inside the concrete. The concrete exposed to air exhibits the lowest strength all the time and strength retrogression occurs at ages greater than 28 days. The strength reaches a maximum after 14 to 28 days of hydration, and then starts to decrease (Shi et al., 2006). For Portland cement concrete, the effect of inadequate water on strength is greater at higher water to cement ratio, lower strength development and in the presence of fly ash or slag (Neville, 1995).

9.2. Alkali activated fly ash

Although there are only a few reports regarding the flexural strength and elastic modulus of alkali activated fly ash (AAFA), it seems that both show inferior values to those of Portland cement. Puertas et al. (2003) reported that the flexural strength of alkali activated PFA mortars are 5.79 MPa while OPC based mortars are 7.76 MPa. They also showed that the elastic modulus of OPC mortars was 5.679 GPa, also higher than the values for PFA mortars activated with 8M NaOH (4.441 GPa). Fernandez-Jimenez et al. (2006) found that the addition of soluble silicates in the alkaline solution improved the modulus of elasticity in PFA-based geopolymer concrete. However, this improvement was not sufficient and the alkali activated PFA concrete showed a much lower static modulus of elasticity than expected. The values presented for OPC concrete ranged from 30.3 to 32.3GPa while for geopolymer concrete they ranged from 10.7 (without silicate) to 18.4 GPa (with silicate). Hardjito et al. (2004) observed better elastic modulus results for a concrete samples made in similar conditions: 22.95 to 30.84 GPa. Apart from their short setting times compared to conventional concrete, geopolymers also attain higher unconfined compressive strengths and shrink much less on setting than OPC (for 7 days only 0.2% that of OPC while for 28 days it is 0.5% of OPC) (Van Jaarsveld et al., 1997). One explanation for this behaviour may be found in the microstructural characteristic of the new binder which in alkali activation of fly ash can form a zeolite-type phase. Zeolite properties and microstructure are widely known to be unaffected by the loss of the water incorporated during their synthesis because not only water loss is reversible in most zeolites but also they are able to absorb water from the humidity in atmosphere (Fernandez-Jimenez et al, 2006). The drying

shrinkage strains are extremely small indeed and the ratio of creep strain-to-elastic strain (that is, creep factor) reached a value of 0.30 in approximately 6 weeks after loading on the 7th day with a sustained stress of 40% of the compressive strength. Beyond this time the creep factor increased only marginally (Hardjito et al, 2004). For normal condition the value of 1.6 is suggested in BS 5400-4:1990 for creep factor of OPC concrete, although it depends on environmental conditions, maturity of the concrete at the age of loading and composition of it. Kaewmanee and Okamura (2001) reached to a value of 1.35 in their works on OPC self compacted concrete loading on the 7th day with a sustained stress of 40% of the compressive strength while Tarek and Sanjayan (2008) have shown that the rate of creep factor evolution with time for slag mixes is lower than of OPC mix which is 1.49 at the time of cracking.

9.3. Alkali activated Natural Pozzolan

Geopolymeric concrete mixes based on activated natural pozzolans mostly have shown lower strength and modulus of elasticity than OPC mixes at early ages, but they reach the same and even higher strength and modulus of elasticity than OPC mixes after long-term curing. It is concluded that concrete made with an alkali activated natural pozzolan develops moderate to high mechanical strength and modulus of elasticity and shrinks much less than ordinary OPC. All of the geopolymer concrete mixes show lower ultrasonic pulse velocity than OPC concrete mixes even though they have higher compressive strengths despite lower densities (Bondar, 2009, 2011).

10. EVALUATION OF CARBON FOOTPRINT AND COST FOR GEOPOLYMER CONCRETE

Two potential advantages of concrete made with alkali activated alumina silicate compared with other binders are its carbon footprint and cost. Increased pressure to improve sustainability within the concrete industry makes these factors very important. The relation between CO₂ footprint and cost of geopolymer concrete and its compositions in comparison with Portland-based cements is roughly quantified (Bondar, 2009).

10.1. Environmental benefits

De-carbonation of lime and calcination of cement clinker release CO₂ as a reaction product in OPC concrete while the use of an alkaline hydroxide or silicate activating solution rather than water for cement hydration does reintroduce some CO₂. Production of these activators needs temperature similar to de-carbonation of lime in OPC manufacture. The CO₂ emission of geopolymer concrete can be quantified in terms of its compositions. Referring to the research was done by author, 110kg of activator is needed to be mixed with 400kg pozzolan to produce 1m³ of geopolymer concrete which has the CO₂ emission equal to 27.5% of the same amount of OPC, when pozzolan used in natural state (It is estimated that the production of 1 tone of OPC results in the release of 1 tone of CO₂). If the calcined form is used, the CO₂ emission of AANP concrete would be the summation of CO₂ emission due to producing the required activators and the amount related to calcination procedure. Since the temperature required for calcination these materials, is half of that needed to de-carbonate lime, the CO₂ emission for calcinations of these materials can be considered 50% of equal OPC production. Therefore, in this case the CO₂ emission of geopolymer concrete increases to 77.5% of the amount emitted by the same weight

of OPC. Hence the geopolymer concrete manufacture is liable to reduce CO₂ emission from 22.5% to 72.5% compared to OPC production (Bondar, 2009).

10.2. Supply and cost of activators

Referring to the research was done by author, 66kg of potassium hydroxide and 46kg (considering density equal to 1.35 kg/m³) of water-glass as activator is needed to be mixed with 400kg pozzolan to produce 1m³ of geopolymer concrete. Although, these activators have different prices in different markets, the cost of industrial potassium hydroxide (25kg KOH Flake UNSD made in KOREA costs 81\$) and water-glass [11.35 kg water-glass solution is sold by Sheffield Pottery in U.K. for 12\$] is considered equal to 3.2\$ and 1.1\$ per kg, respectively. Hence calculating as a rule of thumb the activators required for activating 400kg pozzolans cost 266.6\$ and considering 25kg of pozzolan to be comparable to the price of OPC in 25kg bags, the price of required activators would be 17\$ per 25kg (Bondar, 2009).

11. APPLICATION ASPECTS OF GEOPOLYMER CONCRETE

Geopolymer concrete made from alkali activated alumina silicates is a new type of concrete which needs a very good control on site to be used as a comparable alternative to OPC concrete. However its drawbacks, such as loss of workability, quick setting time and the health and safety implications of working with strong alkali solutions can easily be adapted in applications such as pre-cast concrete and mass concretes as in dam construction where roller compacted geopolymer concrete may be a viable construction method. With respect to the description of the specifications of this type of concrete it can also be used as railway traverse, waste water pipe line, hydraulic structures and pre-tension concrete structures. This type of concrete, especially in countries with greater resources of natural pozzolan and alumina silicate by products, can help decrease energy consumption and environmental impacts (Bondar, 2009).

12. CONCLUSION

1. Geopolymer concretes develop moderate to high mechanical strength with a high modulus of elasticity and a shrinkage much lower than with OPC.
2. Geopolymer concrete manufacture is liable to reduce CO₂ emission from 22.5% to 72.5% compared to OPC production.
3. Geo-polymer concrete can be produced with the same cost of OPC concrete and comparable properties.

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