# Assessment of Alkali Activated Geopolymer Binders as an Alternative of Portlant Cement

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This study summarizes the recent achievements in the development of alternative cement free binder materials (cementless) called alkali activated binder or more popularly known as geopolymers. The history of the development of alkali activated binder and recent terminology are discussed. The influence of the type of the base materials on the properties of goepolymers are thoroughly reviewed. The effect of composition and concentration of alkali and curing temperature on the properties and mechanical behavior of the resulting alkali activated material are discussed. Different type of base materials used in the formation of alkali activated material are blast furnace slag, metakaolin, fly ash, bottom ash, volcanic ashes, and red mud. The state-of-the art in this field demonstrated that this type of materials are ecologically friendly alternative of the Ordinary Portland cement.

Keywords: OPC, concrete, cement free, alkali activated material, geopolymer, terminology, concentration of alkali, curing temperature.

The Ordinary Portland Cement (OPC) has been used over the centuries for material construction especially as a main material in production of concrete. However, there are a few shortages with the using of OPC that have been found especially in terms of properties and greenhouse effect

OPC has become the most widely used concrete base material - since it was invented in the early of 1800s and has been recorded as a second widely consumed material in the world (behind water) [1]. However, the cement industry is claimed to be one of the major contributors to the global warming. Generally, 1 ton of carbon dioxide ( $CO_2$ ) is released with the production of 1 ton of OPC [2]. The emission of  $CO_2$  gas occurred at the first stage of Portland cement clinker production which is at equation (1). In overall, there have 3 stages of Portland cement clinker production which is decarbonation, rapid neutralization, and formation of major component of Portland cement such alite and details is summarized as below:

Stage 1: Decarbonation of calcium carbonate by calcination at 900°C (endothermic) where CO<sub>2</sub> released:

$$CaCO_3 \rightarrow CaO + CO_2$$
 (1)

Stage 2 : Rapid neutralization of free lime (exothermic):

$$3CaO + Al_2O_3 \rightarrow C_3A \tag{2}$$

$$2CaO + Fe2O3 \rightarrow C2F$$
 (3)

$$2CaO+Fe2O3 \rightarrow C2F$$
 (4)

In details, a total global carbon dioxide  $(CO_2)$  emission over the world in 1990 reported was approximately 22.7 billion tones. The trending of  $CO_2$  emissions then increasing into 33.9 billion tones in 2012 and recently, the  $CO_2$  emission reported is over than 34.5 billion tons [3]. The development of modernization in line with the increase of population over the world leads to the increasing of building construction. The dependent of people to the OPC cannot be avoided and getting higher from time to time. In this respect, a new material have been developed for the OPC replacement where a new kind of inorganic cementatious binder called *geopolymeric binder* have been discovered [3]. This kind of new material improves the greenness of normal concrete and also have a better properties compared to the OPC [4].

The history of geopolymer began with the first patented by a famous chemist and engineer Kühl from Germany in 1908 involving a reaction of a solid precursor which have alumina and silica content (vitreous slag) with alkali (alkali sulfate or carbonate) source lead to the production of solid material comparable to the OPC [5]. The development of this binder was enhanced by Purdon where he published an important journal regarding to the achievement rates of strength and the final strength that is comparable to the OPC activated by solution of sodium hydroxide (NaOH) or calcium hydroxide (Ca(OH)<sub>2</sub>) combined with different

(5)

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sodium salts tested under 30 different types of blast furnace slags [6, 7]. Purdon also stated his method of concrete production is appropriate to be used in precast as well as ready mixed where the ratio of activator can be controlled accurately. Unfortunately, Purdon face a problem with the handling of high concentrated solution and also sensitivity of the total water added from the activation conditions. Researches of alkali activated binder then broaden to eastward for a few decades including of Soviet Union and China where a new alternative materials as a replacement of OPC were needed [8]. Glukhovsky [9] from Soviet Union issued a theoretical basis and development of alkaline cements that leads to the using of material called as alkaline cement. Then, investigation towards ancient constructions repaired with OPC by Malinowsky concluded that the repairing material was disintegrated about at least 10 years proved a low durability when compared to the structure that have been repaired [10].

The development of alkaline activation in Western country was about no progress after the last development by Purdon until in the late of 1979 [11]. The name of aluminosilicate-based material was first given by Davidovits in 1979 with the terms of *geopolymer* [12]. Since 1979 onwards, many kind of formulations related to this material (aluminocilicate-based) was introduced and patented by Davidovits [13]. After that, the geopolymer was developed from time to time and some of the problems related to the geopolymer were solved with the enhancement properties. Some of the important people of developing geopolymer after the year of 1982 were Forss [14] in Finland share experiences in developing F-cement where a binder based on alkali activated blast furnace slag were used. Meanwhile, in Ukraine, alumino-silicate based called as Geocements also used the natural minerals [15, 16]. The interest of researchers towards varieties aluminosilicate based materials with the alkali activator has grown up until presence.

This review summarizes the recent achievements in the development of alternative cement free binder materials (cementless) called alkali activated binder or more popularly known as geopolymers. The influence of type of the base materials, composition and concentration of alkali and curing temperature on the properties of geopolymers is thoroughly discussed.

# Materials in Alkali Activated Production Base Materials

Base material or also known as binder normally has been used from the early history until present can be divided into two categories which are high calcium and low calcium. The base material with high calcium is slag blast furnace. Meanwhile, the base materials that categories under low calcium are fly ash, bottom ash, volcano ash, metakaolin and etc. Basically, terms of geopolymer is applied to widely range of alkali activated material mainly

refer to low calcium content of base materials. Some other people understand the word of *geopolymer* exclusively refer to alkali activated binder only from metakaolin due to direct referring to Davidovits which is first person who introduce *geopolymer* using metakaolin as base material. However, the suitability of geopolymer's term for high calcium content base material such as blast furnace slag is still questionable and under debating.

## Blast furnace slag

There are many different types of precursor also called as binder used in the synthesizing of alkali activated. The first binder used in alkali activated history was from the combustion of steel making called as slag. Many kinds of different slag are formed in the production of steel from scrap metal during the different level of metal processing. The slags is formed in the final stage of the metallurgical process naturally and functioned as a covering layer for the molten metal to avoid from oxidation and also functioned to minimized heat loss [17]. Slag has a high content of silica and alumina with a high composition of calcium. Slags that normally have been used as binder in the alkali activated material were called as blast furnace slag. Blast furnace slag is a non-metallic material formed as a molten stream at a high temperature which is around 1400 – 1600°C during the making of pig iron in the burning chamber called as blast furnace [18]. Blast furnace slag is one of the industrial solid wastes where approximately about 300 kg of blast furnace slag were formed from one tones of pig iron. As a result, there are about 167 million tones waste of blast furnace slag were produced across the world in the year of 1996 [19] and the value was increase annually until a year of 2012, where the production of blast furnace slag increase to approximately 270-300 million tones [20]. Recently, according to U.S. Geological Survey, Mineral Commodity Summaries, the production of global blast furnace slag in 2015 is approximately in the ranges of 300-360 million tones [21]. Productions of blast furnace slag making interest to the researchers to explore and utilize the potential of this waste material especially in the construction field.

There is a very slight deference in the chemical composition of the blast furnace slag depending on the compositions of the available iron ores, fuels, flux stones and the proportion required for efficient furnace operations. Table 1 shows the chemical composition of major elements in the most of blast furnace slag involved of 5 different countries over the world. Lime, magnesia, silica, and alumina are the major oxides and cover about 95 % of the composition [22]. Minor element existed in blast furnace slag are normally sulfur, manganese, alkalis, iron, etc. The cooling process plays important role in forming different types of slags, however the chemical composition is remain constant. Physical properties (glassy or stony, compact or vesicular) of blast furnace slag vary with the changes in the cooling process [18, 23, 24].

Chemical composition (wt%)	USA	South Africa	Turkey	Malaysia	Germany
	[22, 25]	[25]	[25]	[26]	[27]
Lime (CaO)	29-50	30-40	34-41	39.32	34.6-42.5
Silica (SiO <sub>2</sub> )	30-40	30–36	34–36	33.87	33.5-39.5
Alumina (Al <sub>2</sub> O <sub>3</sub> )	7–18	9–16	13–19	14.15	10.1-15.3
Iron Oxide (Fe <sub>2</sub> O <sub>3</sub> )	0.1-1.5	<del>-</del>	0.3-2.5	0.52	
Magnesium Oxide (MgO)	0–19	8-21	3.5-7	8.1	9.8-17.3
Manganese Oxide (MnO)	0.2-1.5	<del>-</del>	1-2.5		0.06-0.62
Sulfur (S)	0–2	1–1.6	1.2	<del>-</del>	1.2-1.39

Table 1
CHEMICAL COMPOSITIONS RANGES FOR
THE MOST BLAST FURNACE SLAG
PRODUCED IN 5 DIFFERENT COUNTRIES
[22, 25-27]

Blast furnace slag was proposed for the first time as an alternative of Portland cement by Kühl [28]. The slag is composed mainly of alkali sulfate or carbonate with vitreous phase, with or without added of alkaline earth oxides or hydroxides. The study on blast furnace slag for the formation of cement was continued by Feret [28]. Thirty types of furnace slags as a binder in the formation of alkali activated under various conditions were studied [7]. The material obtained through out activation of the slags by NaOH and combination of different sodium salts with Ca(OH), showed strength comparable to the Ordinary Portland Cement. Moreover, the flexural strength and the tensile were enhanced with low heat evolution and low solubility. However, the handling of high concentrated solution Sand the sensitivity of total water added from the activation conditions posed technological limitations. This problem was settled by Wang et al. in 1995 when a scientific understanding to this problem was proposed [29].

#### Metakaolin

Attention of researchers worldwide towards the production of alkali activated material is not depending just only on blast furnace slags. Beginning in 1979, Davidovits working in France was first introduced a new material of low calcium (including calcium free) replacing blast furnace slag as a binder in alkali activated material. First material that have been developed by Davidovits as binder was calcined kaolin (metakaolin) and the product of alkali activator material was first called as geopolymer in 1979 [12]. Calcined kaolin normally obtained better mechanical properties. Liew et al. [30] stated calcination of kaolin lead to the transformation of crystalline phases to highly amourphous phases and help to enhance the reactivity towards geopolymerization reaction. Term "geopolymer" related to the 'polysialates' nomenclature [31] in which sialates is refer to the aluminosilicate oxide. The network of sialate composed of [SiO<sub>4</sub>]<sup>4</sup> and [AlO<sub>4</sub>]<sup>5</sup> which is tetrahedrally ions that are sharing O<sub>2</sub> and require positive ions such as Na<sup>+</sup>, K<sup>+</sup>, Li<sup>+</sup>, Ca<sup>+</sup>, Ba<sup>+</sup>, NH<sub>4</sub><sup>+</sup>, and H<sub>3</sub>O<sup>+</sup> to generate electric charge of Al<sup>3+</sup> in tetrahedral coordination (the coordination number of aluminum change from 6 (octahedral) to 4 (tetrahedral) after the dehydroxilation process) [2]. The empiric formula for polysialate was discussed briefly by Pacheco-Torgal [2] as follow:

$$M_n \{-(SiO_2)_z - AlO_2\}_n, wH_2O$$
 (6)

where n represent as degree of polymerization, z is value ranging from 1 until 3, and M is alkali cation such as K or Na, forming different kinds of polysialates as in figure 1.

Low calcium binder in alkali activated materials including metakaolin, fly ashes, feldspars, volcanic ashes, low calcium metallurgical slags, and other types of clays. The basic binder structure of low calcium alkaline activated material system is generally to be highly disordered and highly cross linked aluminosilicate. The

Poly(sialate) Si:Al=1 (-Si-O-Al-O-)	SiO <sub>4</sub>	AlO <sub>4</sub>
Poly(sialate-siloxo) Si:Al=2 (-Si-O-Al-O-Si-O-)	Sir	
Poly(sialate-disiloxo) Si:Al=3 (-Si-O-Al-O-Si-O-Si-O)	Sin	
Si:Al>3 Sialate link	O Si O Si	0

Fig. 1. Formation structure of geopolymer proposed by Davidovits [32]

main element in binder which is Si and Al arranged in tetrahedral coordination. Structure of low calcium alkaline activated material was claimed similar to the structure of zeolite [33-35]. For example, the local structure for low calcium binder (metakaolin) were proved very similar to zeolite in terms of length scale up to 5 to 8 Å after heating beyond 1000°C by using X-ray and neutron pair distribuition analysis [36-39].

Many other researchers interested in using metakaolin as a binder in synthesizing geopolymer [40-43]. Metakaolin was preferred due to its advantages in terms their properties including high reactivity and normally high purity compared to others, and also give high strength to the geopolymer [43] with low permeability [44]. Besides, metakaolin also have good properties to be used as fire resistance materials after been tested in terms of thermal conductivity, spalling, cracking, compressive strength, flexural strength, residual strength, temperature capability, and non-combustibility [45]. However, Provis et al. proved there have drawbacks regarding to high water needed in the formation of geopolymer in metakaolin-based mix design [46]. Plus, the metakaolin is high coasting give an issue in the production of geopolymer [40].

Coal ash *Fly ash* 

Currently, the most common raw material used as binder in the formation of geopolymer are fly ash [47-52]. Function of fly ash in cementitious binder component was first discovered since in the year of 1937 where fly ashes have been blended with OPC [53]. Fly ash is one of waste materials produced from the combustion of coal industry. Generally, combustion of coal from the industry produces four types of waste which are bottom ash, boiler slag, flue gas desulfurization sludge, and fly ash [54]. Fly ash

Chemical composition	Metakaolin (wt%)	Fly Ash (wt%)
SiO <sub>2</sub>	55.87	55.86
A12O3	42.25	31.47
K <sub>2</sub> O	0.31	1.15
MgO	0.04	0.39
Fe <sub>2</sub> O <sub>3</sub>	0.38	3.28
CaO	0.04	1.67
TiO <sub>2</sub>	0.20	1.24
P2O5	0.14	0.18
Na <sub>2</sub> O	0.26	0.42
Loss of Ignition	0.61	3.42

Table 2CHEMICAL COMPOSITION OFMETAKAOLIN AND FLY ASH [41]

compose of 75-80 % from total mass production and it's comprises of fine powder, a particles of spherical aluminosilicate, and normally produced from the electrical power plant [55]. Production of waste from coal ashes was reported as one of the world's top 5 raw material sources with the production was estimated 600 matrices tons annually [56].

From the previous research, fly ash geopolymer was proved to have better mechanical properties [57-59] and better durability [59-62] compared to OPC. Fly ash was claimed better than slag as binder in geopolymer due the particle size of fly ash is finer than slag. Plus, fly ash has advantage in getting the source of material compare to slag. The using of fly ash as a binder in cementitious also gives advantage in terms of energy consumption. Tempest et al. [63] mentioned the energy consumed for making concrete base on geopolymer (fly ash) was estimated 70 % lower than the energy consumed for making concrete base on OPC with the same strength.

Generally, fly ash are divided by two types which are fly ash Class C and fly ash Class F. Fly ash from class C usually formed from the combustion of younger lignite or subbituminous coal while fly ash from class F produced from the burning of harder bituminous coal and older anthracite [64]. Some other said class C and class F fly ash comes from the burning of brown coal and black coal respectively [65]. Fly ash class C was classified according to ASTM C618 [66] is fly ash that have a higher calcium content than Class F and normally contain more than 20 wt. % calcium oxide (CaO) as in table 3. Meanwhile, fly ash from class F contain CaO less than 20 wt. %. Fly ash from Class F must have a percentage of unburned material (LOI) less than 5 %, Fe<sub>2</sub>O<sub>3</sub> content must not reached 10 %, and low CaO content, the reactive silica should in the range of 40-50 %, and 80-90 % of the particles size should be less than  $45~\mu m$  in order to produce an optimum binding characteristic,. Meanwhile, ASTM C618 requires class C fly ash contains pozzolanic compounds (silica oxide, alumina oxide, and iron oxide) in the ranges of 50-70 wt. %. Class C Fly ashes getting less studied by previous researchers compare to fly ash from Class F [40].

The key advantage of fly ash from class C is in terms of the density. The class C fly ash geopolymer is denser than the class F fly ash geopolymer. This is proved by Lloyd et al. [67], where a comparison between class C fly ash geopolymer gel with class F fly ash geopolymer gel were done with a similar mix designs as in figure 2. Figure 2 (a) represents as class C fly ash geopolymer, while figure 2 (b) represents as class F fly ash geopolymer where high elemental number alloy (Wood's metal) were filled into both binder's pores which was able to penetrate pores larger than size of 11 nm under the experimental conditions [67]. The bright regions in SEM images (fig. 2) represents as pores.

From the images (fig. 2), Wood's metal was absorbed into the class F geopolymer gel extensively compare to class C geopolymer gel where only a small amount of

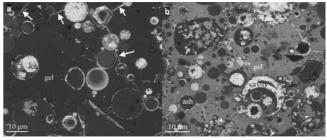


Fig. 2. Back-scattered SEM images for (a) class C fly ash geopolymer and (b) class F fly ash geopolymer. In overall, gel sample in (a) shows brighter than in (b) indicates the gel from class F contains higher porosity than gel from class C. The arrow in (a) indicate cross linking voids due to the particles of fly ash unreact and create a pore volume, where the wooden metal were blocked from entering these spaces due to the fractured of the gel sample during the intrusion process [67]

Wood's metal absorbed inside geopolymer gel [67]. Plus, in theoretical, van Jaarsveld & van Deventer [68] conclude that a higher amount of CaO resulting in higher compressive strength due to the production of calcium aluminate hydrate and other calcium compound as well, especially in the early stage. This is proved by Uysal & Akyuncu [69] where they made a comparison between fly ash class C and class F and the test result indicated that fly ash class C showed higher compressive strength than fly ash class F. The more details the findings made by Diaz Loya et al. studying a concrete made by 25 fly ashes taken from different North American coal fired power stations conclude that the highest strength achieved were from class C fly ash. Nevertheless, this is not the final conclusion cause in separate studies, Oh et al. [70] and Winnefeld at al. [71] proved that various of class C fly ash showed a lower properties in terms of strength development and binder phase evolution compare to class F fly ash.

Meanwhile, some researchers claimed class F fly ash is preferable source material than class C fly ash. The presence higher elements CaO in class C fly ash causing disruption in the geopolymerization process and have potential to change the existing microstructure [72]. Class F fly ash normally has better capability in the resistance against sulphate attack (possibility of concrete to be exposed to sulphate ion and water from ground) and also lower the heat hydration and heat generation of concrete's rate [65]. The main disadvantage of class C fly ash that have been discussed among the expert was the variability of different ashes within class C is much higher compared to class F fly ashes. Hence, there are many detail mix design needed for each source of fly ash in class C fly ash, leading a probability to produce a less robust system than class F fly ash. Another problem that have been discussed from Diaz-Loya et al. [73] is higher water demand or rapid setting (less than 5 min) of some class C fly ashes indicates there is a challenge in making concrete made from class C fly ash in a big scale production.

Chemical compound		OPC		
	Class C	Class F	Class N	1
SiO	39.90	54.90	58.20	22.60
A12O3	16.70	25.80	18.40	4.30
Fe <sub>2</sub> O <sub>3</sub>	5.80	6.90	9.30	2.40
CaO	24.30	8.70	3.30	64.60
MgO	4.60	1.80	3.90	2.10
SO <sub>3</sub>	3.30	0.60	1.10	2.30
Na2O & K2O	1.30	0.60	1.10	0.60

**Table 3**COMPARISON BETWEEN CHEMICAL
COMPOSITION OF FLY ASHES
CLASS C, CLASS F AND OPC [65]

Bottom ash is another product from combustion of coal which is collected from the bottom of furnace. Bottom ash is getting low attention among researchers in geopolymer field compared to fly ash and other popular rich alumino-silica source materials such metakaolin, blast furnace slag, and volcanic ashes. Large and irregularly shape particles with pores and cavities resulting in lower reactivity when used as a geopolymer concrete is the major reasons reported in previous researches of why bottom ash is less popular compare to fly ash [47, 74-80]. Besides, J. Provis et al. [81] state the less reactivity of bottom ash is contributed by very small amount of semi-spherical particles and less glass resulting to a lower strength compared to fly ash.

In general, bottom ash normally not accepted as a blend material with OPC binder due to poor reactivity and mostly content of heavy metals. Although bottom ash is not suitable used in OPC binder, this kind of source material has potential used in alkali activated binder, at least on laboratory scale. The ability of bottom ash as alkali activated binder could be enhanced if the particles size, morphology, amourphous phase, and surface properties was studied and understand. As an example, strength up to 58 MPa was achieved by Santhonsaowaphak et al. [82] when bottom ash was milled to a sufficient fineness and activated by mixing with appropriate molarity of NaOH, liquid of alkaline to bottom ash ratio, and sodium silicate to NaOH ratio. From a limited study of bottom ash in geopolymer, there are majority focusing on the properties of geopolymer pastes [62, 76, 83, 84] and geopolymer mortars [74, 75, 77, 80, 85-87]. Apart from that, there is

only a single study regarding to the geopolymer concretes [88] formed by using bottom ash as raw material.

#### Alkali activators

Alkali activator used in alkali activated binder is to activate the binder in the geopolymerisation process. The alkali activator normally used comes from combinations of alkaline solution and water glass. The most common alkaline solution used in the formation of alkali activated is NaOH or KOH, while the most common water glass used in geopolymerisation process is sodium silicate (Na\_2SiO\_3) or potassium silicate (K\_2SiO\_3). Types of alkali activator play significant role in geopolymerisation process. Apart from prime material (binder), the product properties depend also on the alkali activator. In overall, the alkali activator is divided into 4 categories which are alkali hydroxides, alkali silicates, alkali carbonates, and also alkali sulfates as summarized in table 6.

#### Alkali Hydroxides

Alkali hydroxide is a strong base that composed of metal alkali cation and hydroxide anion. Alkali hydroxide generally called as metal hydroxide (MOH) is very corrosive. The metal alkali cation may come from sodium (Na), potassium (K), litium (Li), rubidium (Rb), caesium (Cs). The most common metal hydroxide than have been used as alkali activator is NaOH and KOH. This is due to the cost is cheaper and suitability to activate the binder in the geopolymerisation process. NaOH and KOH are cheap and used commercially as a product such as drain cleaner (made from NaOH) and cleaning solution (made from

 Table 6

 SUMMARY OF THE INDICATION OF THE QUALITY OF DIFFERENT COMBINATIONS BETWEEN BINDERS WITH ALKALI ACTIVATORS [8]

	MOH	M <sub>2</sub> O· rSiO <sub>2</sub>	$M_2CO_3$	M <sub>2</sub> SO <sub>4</sub>	Others
Blast furnace slag	Acceptable	Excellent	Good	Acceptable	
Fly ash	Excellent	Excellent	Poor *becomes acceptable with cement /clinker addition	Poor-only with cement/clinker addition	NaA1O2- acceptable
Metakaolin	Acceptable	Excellent	Poor	Only with cement/clinker addition	
Natural pozzolans and volcanic ashes	Acceptable/Excellent	Excellent			
Framework alumino silicate	Acceptable	Acceptable	Only with cement/clinker addition	Only with cement/clinker addition	
Synthetic glassy precursors	Acceptable/Excellent *depending on glass composition	Excellent			
Steel slag		Excellent			
Phosphorus slag		Excellent		 	[
Ferronickel slag		Excellent			
Copper slag		Acceptable *grinding of slag is problematic			
Red mud		Acceptable *better with slag addition			
Bottom ash and municipal solid waste incineration ash		Acceptable			3

KOH) for cleaning terrace and all kinds of buildings structure made by wood that easily to buy at most of hardware stores. Another potential for NaOH and KOH is both metal hydroxide have high solubility in water which is reach 20 mol/kg H<sub>2</sub>O at 25°C [89, 90]. Both metal hydroxides is very popular used as activator in alkali activated binder especially when using with class F fly ash binder. Meanwhile, LiOH, RbOH, and CsOH is not suitable used as activator due to the cost is expensive, stability, and also low of solubility in water especially for Li which is only under 5.4 mol/kg H<sub>2</sub>O at 25°C) [91].

# Alkali silicates

Alkali silicates are another important chemical compound which has an ability to be a good activator to a binder in alkali activated with a formula of M2OrSiO2. Similar to alkali hydroxides, alkali silicates comprise with metal cation with the most popular used as activator in alkali activated binder is sodium and potassium. Currently, Na2SiO3 and K2SiO3 is widely produced in industrial as alkali activation [92] due to the same factor which is in terms of costing, suitability, and solubility. Other metal silicates are not suitable to be used as activator due to the limited sources, cost, drawback, as well as low solubility especially when used in a large scale. Equation 7 shows the example of the alkali silicates (sodium silicate) formation which is from the reaction between sodium carbonate and silicon dioxide:

$$Na_2CO_3 + SiO_2 \rightarrow Na_2SiO_3 + CO_2$$
 (7)

From equation 7, the formation of sodium silicates creates an issue to the greenhouse where the formation of alkali silicates will release  $CO_2$  to the environment. Plus, high energy consumption needed in order to produce sodium silicate which is by calcinations, dissolving, and filtration process. However, due to the very lower consumption of activator used in alkali activated process compare to the production process of OPC [93], making this material still relevance to be used in construction area.

Na,SiO, and K,SiO, normally suitable to be used as an activator with almost existence binders. This kind of activator sometimes suitable to be used alone and sometimes suitable to be combined with alkali silicates group in order to produce a better activator. Puertas et al. [94] in the study of blast furnace slag has reported combination of NaOH with alkali silicates as activator leads to the lower reaction products of Al/Si molar ratio than the one obtained by using a single NaoH. Meanwhile, Granzo [95] found the effect of different reactions when only NaOH was added as activator compare to mix of NaOH with waterglass on the study of metakaolin. The first reaction, the destroyed products start to accumulate after the phase was dissolute an induction period follows. The second reaction, a quick polycondensation reaction follows right away after a quick phase of dissolution

Many researchers produce alkali activated binder by combining an activator between alkali silicates and alkali hydroxides with appropriate ratios [96-99]. Ratio between alkali silicate and alkali hydroxide plays important role to the mechanical properties of alkali activated binder. For example, Figure 4 shows the effect of alkali silicate to the alkali hydroxide ratios on the compressive strength of the fly ash based alkali activated binder. Palomo et al. found a compressive strength of ranges 35-40 MPa will produced from a reaction of different fly ash with alkali activator (NaOH) of ranges 8-12 M cured at 85°C in 24 h, while the

compressive strength (with the same conditions) will increase up to 90 MPa when alkali silicate ( $Na_aSiO_3$ ) was added with the ratio of  $SiO_a/Na_aO = 1.23$  [100].

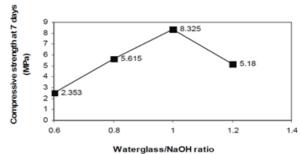


Fig. 4. Effect of Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios to the compressive strength (at 7 days) of the class F fly ash based alkali activated binder [96]

#### Alkali carbonates

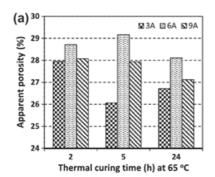
Unlike alkali hydroxides and alkali silicates, alkali carbonates or generally known as M<sub>2</sub>CO<sub>3</sub> (where M is referred as alkali metals) give less attention among researchers in the alkali activated binder's area. The study of using alkali carbonates as alkali activator was started since 1959 by Gluchovskij where he wrote a book regarding to the possibility of reaction alumino-silicate raw materials with alkaline compounds [101]. The alkali carbonate can be produce either by directly mining of carbonate salts, or by the process called as Solvay [102]. The key advantage of using alkali carbonates as an activator is lower impact to the Greenhouse but the alkalinity of this activator is lower than other types of alkali activators making the suitability alkali carbonates getting narrow to the alkali activated material binders. The most familiar of alkali metal carbonate is sodium carbonate. Potassium and other metal carbonate were usually not used as an activator in alkali activated material.

# Alkali sulfate

Generally alkali sulfate chemically known as M<sub>s</sub>SO where M may consist of any alkali metals. Similar to alkali carbonate, only sodium sulfate is suitable to be used as activator to the some of alumino-silicate binders. The production of sodium sulfates consumed less energy than other types of alkali activator where the production process used lower temperature resulting many industrial chemicals manufacture this activator as by product. Plus, this kind of activator can be obtained directly from mining (natural resources of over 109 tonnes are estimated to exist across the world [103]). This kind of alkali sulfate gives good effects to the Green house where the energy consumption for production is lower with a less of CO, emission. The suitability of sodium sulfate is limited to the types of binders and gives a less performance to the alkali activator material product. Almost all the possible binders' needs to be added with cement or clinker to be combined with Na, SiO, activator in forming of alkali activated material.

# Effects of conditions to the alkali activated material Alkali concentration

Low performance cementitious material can achieve high compression strength when added with alkaline activator. The alkali activator can increase the performance of alkali activated binder up to 160 MPa after curing at room temperature for 90 days [104]. However, although in an ideal concentration, a loses of mechanical properties may occurred due to the free OH ions in the alkali activated system which resulting in possibility of changing structure of alkali activated binder [105]. Alkali concentration proved



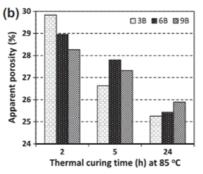


Fig. 5. The apparent porosity of fly ash based alkali activated binder for different concentrations at (a) 65°C and (b) 85°C [105]

to be one of the main parameter in the contribution to the performance of alkali activated binder product.

Other condition factors such curing temperature, humidity, and curing time play less important than activator concentration [106-108]. Alkaline activator plays important role in the dissolution process of silica oxide and alumina oxide in the formation of alkali activated binder. In the study of metakaolin with NaOH as activator, Hong Ling et al. [109] mentioned the increasing concentration of alkaline activator (NaOH) will led to the increasing of amorphous phase. Besides, the effect of alkali concentration to the porosity has been studied by Görhan & Görhan & Kürlü [105] with 3 different concentrations of NaOH which is 3M, 6M, and 9M with 3 different curing temperatures and curing times. A result (fig. 5) doesn't show any specific trending and was not clear to correlate the relationship between concentrations with the degree of porosity.

Curing temperature

The reaction can take place at normal laboratory temperatures or elevated temperatures (typically in the range of 30 - 85°C) [110-115]. Moist curing is typically used to assure a rapid strength gain. The reactants form a series of alumino-silicate chains or networks of oxygen, silicon, and aluminum linked with covalent bonds [116]. This curing temperature will affect the structural and also the mechanical properties of alkali activated binder [117-120].

Curing temperatures are different depending on conditions. A.M. Mustafa Al Bakri et al. [115] stated the optimum curing temperature involving of fly ash (Malaysia power plant) and NaOH 12 M was 60°C. At that temperature, the compressive strength of alkali activated binder was maximum which is 67.04 MPa and the value of compressive strength getting lower when the temperature was raised above 60°C. Meanwhile, Görhan & Kürklü [105] proved the optimum curing temperature for alkali activated binder fly ash (Turkey power plant) with NaOH solution 6 M was 85°C. Some other researchers got different optimal curing which are 60°C [121], 65°C [122], and 80°C [123] depending on the types of fly ash, types of activator, alkali concentration, humidity, curing time and etc. [124].

## **Conclusions**

The review on the alkali activated material have been discussed in this paper including the introduction of alkali activated binder, the base material, the alkali activator, and the effects of conditions to the alkali activated material. The emissions of CO<sub>2</sub> by the OPC product have been presented and the alternative way to overcome this issue toward greener has been proposed by development of alkali activated material. The history of development involving alkali activated have been described in details starting from a famous chemist and engineer Kühl from Germany in 1908 until present and have been grown over the world across from Europe to Asian. The performance of

development of alkali activated always been compared with the OPC and mostly a better properties of alkali activated material have been produced compare to OPC especially in terms of mechanical properties. From this reviews, base material can be divided into 2 categories which is high calcium material and low calcium material. The chemical compositions of stated base material were different among each other and becoming a factor to the contribution toward the properties of product. The structure of poli(sialate) was illustrated to give a clear view to the formation of alkaline activated material by geopolymerisation process. Percentage composition of CaO of fly ashes indicates their categories where high content of CaO considered as Class C while low content of CaO considered as Class F. The CaO content from fly ash is key important to the contribution of product properties especially in terms of compressive strength and density. Alkaline activator play important role to the properties of alkali activator product. The suitability of activator with the base material is very critical in the production of good properties of products. The properties of alkali activated product are very depending on the types of binder, types of activator, concentration of activator, and the curing temperature.

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