

Investigating the Possibility of Geopolymer to Produce Inorganic-Bonded Wood Composites for Multifunctional Construction Material – A Review

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Wood-based composites are widely used in consumer products, either in structural or non-structural applications. One of the basic elements for wood-based composites is the binder itself. Recent years have seen great development and trends in the field of eco-friendly binders in wood-based composite. There have been many concerns on the effects of formaldehyde and volatile organic compounds (VOC) released from wood-based products. Researchers have put lot of effort into developing environmental friendly products with enhanced sustainability. Binder materials with a focus on geopolymers (*i.e.*, alumino-silicates) are discussed in this publication. The development and utilization of geopolymeric binders is relatively new in the field of wood-based composites. Up to the present there has been insufficient information regarding the manufacturing conditions and properties of wood-nonwood composite materials prepared using a geopolymeric binder. This paper considers the background of geopolymer materials and the possibilities of producing inorganic-bonded wood composite using geopolymer.

Keywords: Geopolymer; Wood composite; Inorganic binder; Alkaline activator

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INTRODUCTION

Wood-based composite manufacturing has become a significant industry nowadays. Many countries rely heavily on it for construction applications. The main advantages of wood composite materials are availability, renewability, low processing costs, and simplicity of dismounting and disposal at the end of their services life. Wood-based composites encompass a variety of products with different typical elements including fibers, particles, flakes, veneers, laminates, or lumber. Maloney (1986) proposed four major classification systems of wood-based composites, namely veneer-based material, laminates, composite material, and wood-nonwood composites. Each categorization of elements size and geometry dictate the product manufacture and product performance.

Traditionally, wood-based composites have fully depended on the use of synthetic adhesives as a binder due to its low costs and excellent properties. Synthetic adhesives, produced by the controlled polymerization of various monomeric organic substances, have dominated the application for the last 80 years and still dominate the field of wood panel bonding (Pizzi 2014). However, due to the acute sensitivity of the general public to

anything that has to do with the environment and its protection and the recent development towards reducing formaldehyde emissions from wood composites, there is growing interest in finding substitutes for the currently used binders for wood-based composite materials. Some of the alternative approaches are developments in bio-based wood adhesives (Amini *et al.* 2013; Sulaiman *et al.* 2013), modification of the chemical structure of the adhesives (Liu and Li 2002; Vitale *et al.* 2014), and utilisation of inorganic-bonded composite materials (Al Rim *et al.* 1999; Gouny *et al.* 2014).

Inorganic-bonded wood composites are molded panels that contain between 10 and 70 percent by weight of wood elements and conversely 90 to 30 percent of inorganic binder. Gypsum-bonded, ceramic-bonded, and cement-bonded (Magnesia and Portland) are some of the inorganic-bonded wood composite products that exhibit specific properties that are superior to those of the component materials alone (Simatupang and Geimer 1990). However, the use of cements in wood-based composites involves limitations, in terms of both environmental impact caused by the extraction of raw materials and carbon dioxide emissions during cement manufacture (Kamarudin *et al.* 2011). In order to address these concerns and other environmental problems relating the use of cements, another form of cementitious materials, the geopolymers, was discovered by Glushkovsky in the former Soviet Union in the 1950s and developed by Davidovits in the late 1970s (Kamarudin *et al.* 2011; Shaikh 2013).

Geopolymers are amorphous three-dimensional alumino-silicate binder materials (Shaikh 2013), which have been proposed as an ecologically friendly alternative to concrete, as their production does not involve limestone calcinations (Duxson *et al.* 2007). The need for environmentally friendly construction materials for sustainable development is an important environmental issue at present. Geopolymer, an inorganic material produced by the alkali-activation reaction, is well known as a potential replacement to ordinary cement. This paper summarizes some important research findings on the geopolymer composites and describes the geopolymerization process. Moreover, the paper emphasizes the potential to produce inorganic-bonded wood composites using geopolymer as an alternative to ordinary inorganic binders.

INORGANIC-BONDED WOOD COMPOSITES

Inorganic-bonded wood composites had been established commercially in the early 1900s. In 1914 the first low-density inorganic-bonded wood composite panels were developed in Austria using magnesite binder. They are mainly used for interior applications in many countries in the world, mostly in panel form. Soon after, gypsum-bonded, Portland cement-bonded and ceramic-bonded wood composites became popular, and these had superior properties, with further improvements made subsequently. Typical inorganic-bonded wood boards applications vary widely between the different board types due to the variable properties of each, but common applications include tile substrates, flooring and underlay, kitchen counters, backsplashes, roofing, shingles and shales, weatherboards, façade, cladding, prefabricated houses, exterior and partition walls, acoustic and thermal insulation, soffits, ceilings, and architraves.

The use of inorganic-bonded wood composites in the construction industry is increasing due to its advantages over traditional building materials. The properties are significantly influenced by the amount and type of the inorganic binder and the wood element as well as the density of the composites. The largest portion of the inorganic-

bonded wood composites board market is filled by Portland cement-bonded medium-high density boards, which is mainly used as wall and roof materials in the construction of houses and in such diverse applications as sound barrier walls bordering highways. Production of gypsum-bonded particleboards and fiberboards is steadily increasing while magnesia-bonded medium-high density boards have been available for a relatively long period of time but have faced a declining market due to relatively high costs (Simatupang and Geimer 1990).

Inorganic-bonded composites are made by blending wood elements with inorganic materials in the presence of water and allowing the inorganic material to cure or set-up to make a rigid composite. Wood and other plant fiber in combination with inorganic binder has been practiced intensively. Successful combination of these disparate materials depends upon an understanding of their chemical interactions and resultant performances (Frybort *et al.* 2008). According to Simatupang and Geimer 1990, certain woods and their extractives retard the cure of inorganic binders and alter the form and dimension of the crystalline structures. The manufacturing of inorganic-bonded composites is adaptable to either end of the cost and technology spectrum. This is facilitated by the fact that no heat is required to cure the inorganic material. A very small capital investment may be sufficient to produce satisfactory inorganic-bonded wood composites on a small scale using mostly unskilled labor. Besides savings in cost, space, time and convenience, it is a highly-durable construction material with high impact-resistance. Most of the inorganic-bonded wood composites are very resistant to deterioration by decay fungi, insects, and vermin, and most importantly it is fire and water resistant.

GEOPOLYMER

Raw Materials Involved in Geopolymer

Geopolymer materials contain aluminium (Al) and silicon (Si) species that are soluble in highly alkaline solutions (Davidovits 2008). Any material that is rich in Si and Al in amorphous form can be a possible source material for geopolymer binder (Shaikh 2013). Ikeda (1988) stated in his study that two sources are needed for the synthesis of geopolymers; (i) raw materials where it could be natural (alumino-silicate) minerals or industrial wastes, (ii) geopolymer liquor, which is an alkali hydroxide solution required for the dissolution of the raw materials. Natural minerals or industrial wastes that have been used as a raw material are kaolinite, metakaolin, calcium- and silica-based geopolymer, fly ash, slag, silica fume, and natural pozzolans.

Kaolinite and metakaolin are primarily used as sources of alumino-silicate oxides in order to synthesize geopolymers. Many other researchers have also focused on the manufacture of geopolymeric products by using either kaolinite or metakaolin (Xu and Van Daventer 2000; Shaikh 2013; Barbosa and Mackenzie 2003). Both materials have been preferred due to their high rates of dissolution in the reactant solution (Xu and Van Daventer 2000), improved mechanical strength (Ambroise *et al.* 1994), and reduced transport of water and salts in the final product (Kostuch *et al.* 1993). However, due to certain disadvantages, kaolinite and metakaolin have been ruled out with respect to mass use in geopolymer applications. Disadvantages include, for example, the high cost of metakaolin (Shaikh 2013), the fact that a large amount of added kaolinite does not take part in the synthesis reaction, affecting the properties of the final product (Van Jaarsveld

et al. 2002; Komnitsas *et al.* 2009), and the requirement for too much water due to porosity and consequently becoming too soft for construction application (Kostuch *et al.* 1993).

The utilization of industrial wastes or by-product materials in geopolymerisation has been investigated extensively in recent years. Fly ash and slag are among the industrial wastes that have high potential for being used as source materials for geopolymers. According to Shaikh (2013), fly ash is considered to be advantageous due to its high reactivity that comes from its finer particle size than slag ash. Ahmaruzzaman (2010) mentioned that by reacting fly ash with calcium hydroxide, the cementitious compound becomes stable, which increases its resistance to corrosion. This is due to the reaction products tending to fill the capillary voids in the concrete mixture and reducing permeability of the concrete. In 1999, Van Jaarsveld and Deventer found that fly ash with a higher amount of CaO produced higher compressive strength, due to the formation of calcium-aluminate-hydrate and other calcium compounds. High calcium content slags also have been investigated extensively and have been used in large scale construction applications. Cheng and Chiu (2003) conducted research on the use of granulated blast furnace slag as active filler for production of geopolymer. They found that the highest compressive strength was 79 MPa. It was also found that geopolymer setting time correlates well with temperature, potassium hydroxide concentration, metakaolinite, and sodium silicate addition.

Palomo *et al.* (1999) and Xu and Van Deventer (2000) state that calcined materials such as slag, fly ash, and metakaolinite, which are mostly amorphous, usually display a higher reactivity compared to non-calcined materials, which is regarded as beneficial during geopolymerisation. Xu and Van Deventer (2000) added that this can be explained by the fact that calcination activates materials by changing their crystalline into amorphous structure with subsequent storage of the extra energy.

Alkaline Activator

A highly alkaline solution of some type is one of the important 'ingredients' in geopolymerisation. Such solutions are used to induce the silicon and aluminum atoms in alumino-silicates minerals to dissolve and form the geopolymer mixture. Studies by Hardjito *et al.* (2004) and Palomo *et al.* (1999) have shown that the curing temperature, the curing time, and the type of activator affect the compressive strength. Shaikh (2013) mentioned that the most common alkali activator used in geopolymerisation process is the combination of sodium silicate (Na_2SiO_3) or potassium silicate (K_2SiO_3) with sodium hydroxide (NaOH) or potassium hydroxide (KOH).

The types of alkaline activators used play an important role with respect to the polycondensation process. According to Van Jaarsveld *et al.* (2002), alkali metals cations contribute to the structures formation of geopolymers. They affect almost all stages of geopolymerisations, particularly during gel hardening and crystal formation. Cheng and Chiu (2003) found that the activation reaction rate depends on several factors such as particle size distribution and the mineral composition of the starting material as well as the type and concentration of the activator.

Reaction Mechanism of Geopolymerization

The reaction mechanism of geopolymerisation is a challenging field to understand. The basic steps of geopolymerisation involve mixing of alumino-silicates oxides (solid powder form) with strong alkaline solutions such as example NaOH or

KOH solutions, to form a sodium silicate solution or potassium silicate solution (Zuhua *et al.* 2009). The reaction mechanism shown in Fig. 1 demonstrates the basic process occurring in the transformation of a solid aluminosilicate source into a synthetic alkali aluminosilicate.

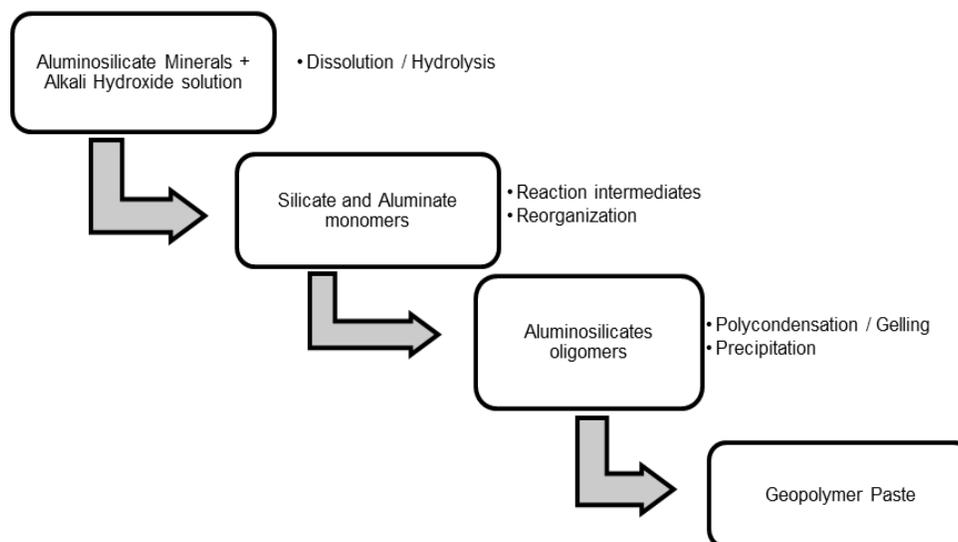
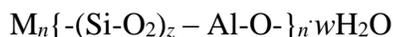


Fig. 1. Conceptual model of geopolymerisation

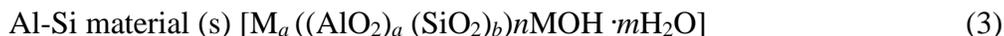
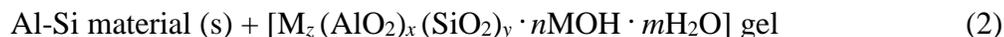
According to Xu and Van Deventer (2000), geopolymerizations involve four basic steps: (1) dissolution of solid aluminosilicates oxides in MOH solution (M: alkali metal), (2) diffusion of dissolved Al and Si complexes from the particles surface to the inter-particle space, (3) formation of a gel phase resulting from the polymerization between added silicate solution and Al and Si complexes, and finally (4) hardening of the gel phase. This is also reported by Alonso and Palomo (2001), Duxson *et al.* (2007), and Komnitsas and Zaharaki (2007).

According to Komnitsas and Zaharaki (2007) the alkali hydroxide solution is required for the dissolution of raw materials, while sodium or potassium acts as binder, alkali activator and dispersant or plasticizer. The dissolved slurry then undergoes polycondensation to produce materials with desired mechanical properties (Sofi *et al.* 2007). They cure and set under ambient conditions into a material with an X-ray amorphous three-dimensional network of aluminate and silicate units with charge-balancing cation (Kamarudin *et al.* 2011). Davidovits (2008) reported that the inorganic polymers developed have the general formula as below,



where M is the alkali metal; z which 1, 2 or 3 and n is the degree of polymerization. Previous study by Xu and Van Deventer (2000) found that the polycondensation taking place during geopolymerisation of minerals were,





The amount of Si-Al material involved in polycondensation depends on the particle size, the extent of dissolution of Al-Si materials, and the concentration during the reaction mechanisms in Eqs. 1 and 2. Furthermore, the geopolymers with amorphous structure are formed during reaction (3), where the time required for Si-Al material to form a continuous gel depends on the raw material processing conditions (Komnitsas and Zaharaki 2007; Kamarudin *et al.* 2011).

Dissolution of the alumino-silicate material and formation of polymeric species depends strongly on thermodynamic and kinetic parameters, which are parallel with the steps show in Fig. 1, as proposed by Duxson *et al.* (2007). It was found that these processes of structural reorganization determine the microstructure and pore distribution of the material, which are critical in determining many physical properties of the geopolymer.

Development and Application of Geopolymer

In the last few years, there has been spectacular technological progress in the development of geopolymeric applications. The new materials design with the help of geopolymerisation reaction processes has opened up new applications and procedures in inorganic and mineral chemistry. According to Davidovits (1994), high-tech geopolymer binders have been developed that already meet the standard applications in all fields of industry, whether used as pure materials, with fillers, or reinforced. These applications are to be found in the automotive and aerospace industries, non-ferrous foundries and metallurgy, civil engineering, cements and concretes, ceramic and plastics industries, waste management, art and decoration, and retrofit of buildings. From the literature, some of the geopolymer applications are still under development, whereas others are already industrialized and commercialized.

Geopolymer cements are manufactured in a different manner than that of Portland cement. They do not require extreme high temperature kilns, with large expenditure of fuel and large capital investment in plant and equipment. Based on Duxson *et al.* (2007), the thermal processing of naturally occurring alkali-silico-aluminates and alumino-silicates provides suitable geopolymeric raw-materials. According to the US Corps of Engineers (Davidovits 1994), alkali-activated alumino-silicate binders (cements) have the potential to become the best and in many cases the most economical binder for routine construction and may evolve into a new generation of building materials. Their unique properties which include high compressive and flexural strength, low shrinkage, freeze-thaw resistance, sulphate and corrosion resistance, heat and electrical resistance make them ideal for various applications of geopolymer technology.

POTENTIAL TO PRODUCE WOOD-GEOPOLYMER COMPOSITE

Woody or lignocellulosic materials play an important role in inorganic-bonded wood composite production, either acting as an aggregate or as a reinforcing element. According to Jorge *et al.* (2004) the properties of inorganic-bonded wood composite are influenced by (i) wood element characteristics; species, size, geometry, chemical

compositions, (ii) binder type, (iii) wood-binder ratio, (iv) environmental temperature and (v) cure time. Simatupang and Geimer (1990) said that, depending on the raw material selection and processing conditions, inorganic-bonded wood composite can exhibit a wide variety of properties and characteristics, including compressive strength, dimensional stability, fire and acid resistance, and thermal conductivity.

The first applications involving wood with geopolymer technology were building products developed in the period 1973 to 1976 (Davidovits 2008). These were fire-resistant chip-board panels, comprised of a wooden core faced with two geopolymer nanocomposite coatings, in which the entire panel was manufactured in a one-step process. Gouny *et al.* (2013) studied the mechanical behavior of laboratory assemblies of wood, geomaterial binder and two different types of earth brick. They found that geopolymer binder can give good adhesion between wood and earth brick.

Many methods have been described to increase the compatibility of woody or lignocellulose materials with geopolymer. Chen *et al.* (2014) investigated the mechanical properties of reinforcement fly-ash based geopolymer with alkali-pretreated sweet sorghum fiber. They found the unconfined compressive strength, the splitting tensile, and flexural strengths, as well as the post-peak toughness increased with fiber content up to 2%. Alomayri *et al.* (2013) studied the characterization of cotton fiber-reinforced geopolymer composites and found that the appropriate addition of cotton fibers can improve the mechanical properties of geopolymer composites.

The interaction between the inorganic binder and the woody material is very important. Acceptable properties of an inorganic bonded wood composite can be obtained only when the wood particles are fully encased with the binder (Simatupang and Geimer 1990). As in the case of synthetic resin adhesion, inorganic adhesion of material is thought to result from physical bonding mechanisms as well as mechanical interlocking (Frybort *et al.* 2008). Gouny *et al.* (2013) stated that the adhesion mechanism between geopolymer binder and wood are not well defined, however the mechanical interlocking adhesion and chemical bonding were considered to be involved. By means of SEM and chemical element analysis of the wood interface, the authors showed that the bonding is caused by foam of the geopolymer binder and the alkaline element. Before consolidation, the binder penetrates the pores of the wood, simultaneously reinforcing the wood fiber and inhibiting its hygroscopic behavior at the interface. The alkaline element is essential for wood cell growth and able to move into the wood cells through channels. Duxson *et al.* (2007) mentioned that if the alkaline content is too great in a geopolymer formulation, then the mechanical properties of the geopolymer will deteriorate. According to Davidovits (2008), the pH of an alkali silicate solution is dependent on the molar ratio (MR) of the solutions itself. The pH value is the most important characteristic determining stability of high-modulus silicate solutions, that is, their inclination to the formation of gel or coagulation. The stability of alkali silicate solutions increases steadily as their pH values increases.

The use of inorganic binder for wood-based composites involves limitations and tradeoffs. Simatupang and Geimer (1990) mentioned that hemicellulose, starch, sugar, tannins, and lignin in woody materials might affect the cure rate and ultimate strength of these composites. It has been demonstrated that wood extractives retard the hydration of the inorganic binders and alter crystalline structures. Duxson *et al.* (2007) mentioned that to make strong and durable inorganic-binder wood composites, appropriate measures must be taken to ensure long-term stability of the elements (woody or lignocellulose) in the cement matrix. The assessment of the effect of contaminants present in the raw

materials which might affect the reaction pathways and the properties of the final product as a result of changes in setting time, slump, and strength, need to be considered during the geopolymerisation. In addition, Davidovits (2008) in his book mentioned that geopolymers have three main properties that make them superior to ceramic-matrix composites, plastics and organic composites materials. First, geopolymers are very easy to make, as they handle easily and do not require high heat; second, geopolymeric composites have a higher heat tolerance than organic composites. Tests conducted on geopolymer carbon-composites showed that they will not burn at all, no matter how many times ignition might be attempted; Third, the mechanical properties of geopolymer composites are as good as those of organic composites. Furthermore, geopolymers resist all organic solvents and are only affected by strong hydrochloric acid.

CONCLUSIONS

The stabilization of geopolymer-bonded wood composite is crucial for its applications. Various measures have been carried out in order to meet the required strength properties for certain applications of geopolymer-bonded wood composite. Many studies have been undertaken previously on geopolymer technology; however there is still a lack of information on the properties of geopolymer-bonded wood composites, or even how to produce them. Therefore, further study is needed to investigate the possibility for making geopolymer-bonded wood composites, which have the potential to be used advantageously as environmentally friendly wood products. Using geopolymer type of binder in wood composite manufacturing would result in a potentially important class of formaldehyde-free type of wood composites. Such material would probably be welcomed by the construction material providers, provided that similar properties as wood composites prepared with phenol, melamine, or isocyanate can be achieved. The outcomes of future research will have a large direct impact on the wood based industries, especially to have more global impact for enhanced material greenness. Geopolymer-bonded wood composites have the potential for use in various construction applications.

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