

POTENTIAL OF GEOPOLYMER COATING FOR LIGHTWEIGHT AGGREGATE VIA MILLING AND DIPPING METHOD: A REVIEW

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Abstract

The high demand for construction materials and building products, particularly aggregates, necessitates the development of innovative as the natural aggregate resource depletes. Lightweight aggregates are one of the alternatives has meet industry demand with low apparent density, excellent thermal insulation property and fire protection. Unfortunately, lightweight aggregate has a high-water absorption rate, which results in low strength and high porosity. These conditions had a negative impact on the qualities of fresh concrete, such as strength, slump, and cracking. Surface coating is thought to be a good way to prevent water absorption by lightweight particles, which lowers the concrete's performance. To reinforce the concrete, inorganic polymer composite material and cement were commonly employed to coat lightweight particles. The article presents a review on current issues facing by lightweight aggregate, mix proportion of geopolymer coating (aluminosilicate raw materials, alkaline activator ratio) and coating method (milling and dipping). This review has highlighted performance of lightweight aggregate for concrete application can be enhanced by approaching geopolymer coating as surface coating.

Keywords: *lightweight aggregate, geopolymer coating, aluminosilicate raw materials, alkaline activator, milling coating, dipping coating.*

Introduction

Rapid urbanization has accompanied economic progress, resulting in massive new construction projects, particularly in emerging economies. An aggregate is a material that is useful in the construction industries. The most basic and widely used way for classifying aggregates is by specific gravity which is divided into three categories which normal weight, lightweight, and heavyweight [1]. Due to environmental purposes, lightweight aggregate has replaced natural aggregate as a construction material in the construction industries. Lightweight aggregates are divided into two categories which natural and artificial. Pumice, diatomite, volcanic cinders, and tuff are the most common natural lightweight aggregate [2]. The artificial aggregates are further divided into two categories which are industrial by-products and modified naturally occurring materials [2-3]. Sintered slate, sintered pulverized fuel ash, expanded, or foamed blast-furnace slag and colliery refuse are examples of industrial by-product materials used as lightweight aggregate [3]. Natural materials that require further processing such as shale,

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expanded clay, slate, vermiculite, and perlite, are also employed in the building industry as lightweight aggregate [3-4].

Lightweight aggregates are known as granular and porous materials used in architecture, engineering, landscaping, and geotechnics. Lightweight aggregates have low apparent density, excellent thermal insulation property and fire protection. Prefabricated structural units and structural lightweight concretes, particularly in high-rise structures, as well as track ballasts and road coatings, all contain lightweight aggregate [5]. Unfortunately, the inner structure of lightweight aggregate can have many porous which contribute the porosity problem. Besides that, it also has a high absorption ratio therefore the lightweight aggregate makes it difficult to keep track of how light aggregate-concrete is mixed during construction [6]. On account of the disadvantages, infrastructure maintenance and repair have become a major issue, and the protection of structures and elements has become a top priority. Therefore, the coating is the option to maintain and protect the surface of the structure [7]. Enhancing the mechanical properties of lightweight aggregate by coating is one of the solutions nowadays which is inorganic coating and organic coating. In recent years, a new type of inorganic coatings based mostly on geopolymers has been researched and used to protect the surfaces of concrete, steel, and wood structural elements [8].

Geopolymers are gaining popularity as a coating material due to their inert silicate and aluminate connections, which environmentally safe, provide significant resistance to water, corrosion, and fire [9]. In terms of raw ingredients, production techniques, and final qualities, geopolymer coatings have proven to be superior to current inorganic cement coating and organic coating systems [10]. Geopolymer is known as a mixture of aluminosilicate raw material and alkaline activator and crated geopolymerization at elevated or ambient temperatures [11], [12]. The geopolymer technology offers a new beneficial and green alternative for the exploitation of fly ash, minimizing its negative environmental and ecological impact [7]. The alkali activation is carried out by incorporating NaOH, KOH, Na₂SiO₃, or K₂SiO₃ into fly ash, either combined or individually [13]. Furthermore, the NaOH molarity, Na₂SiO₃/NaOH ratio, geopolymer/alkaline activator ratio, and curing temperature have a significant impact on the final properties of the geopolymers [8]. The raw material that is used as a geopolymer is fly ash, metakaolin, blast furnace slag, natural zeolite, and granite [14], [15]. There are several types of geopolymer coating methods such as dipping and milling methods based on the coating material.

The article presents a review on current issues facing by lightweight aggregate, mix proportion of geopolymer coating (aluminosilicate raw materials, alkaline activator ratio) and coating method (milling and dipping).

Current issues facing by lightweight aggregate

Lightweight aggregate is known as an ingredient to create a concrete. Lightweight aggregate are mineral aggregates with particle densities ranging from 800 kg/m³ to 2000 kg/m³ or loose bulk densities less than 1200 kg/m³ (EN13055-1, 2002) [16]. The inner structure of LWA can be porous to attain the low density required by aggregates used in lightweight concrete [17, 18]. This condition gave adversely affect such as low strength (Aggregate Impact Value (AIV)), high water absorption and high porosity. Table 1 has disclosed the current issues facing by previous study on lightweight aggregate respected to four main properties; density, water absorption, porosity and aggregate impact value.

Kwek and Awang (2018) has studied properties of artificial aggregate containing palm oil fuel ash (POFA) and silt from water treatment sludge [19]. According to this study, the lightweight aggregate made of silt and POFA as based material has a potential in replacing the natural aggregate in concrete production. However, the satisfactory range of water absorption and AIV properties still become an issue. Rehman et al., (2020) has produced of lightweight aggregates through cold bonded pelletization using two different industrial by-products; coal fly

ash (FA) and ground granulated blast furnace slag (GBFS) [20]. This finding can be effectively used as a replacement of natural coarse aggregate in formulation of structural lightweight concrete. Though, the results still have low strength and high-water absorption which need to be overcome in order to increase the efficiency of this lightweight aggregate.

Hanif et al., (2021) presents the production of ultra-lightweight geopolymer foam aggregate utilizing coal fly-ash [21]. The properties obtained would be suitable to design lightweight concrete both for structural and insulation purposes. Satisfactory results of this finding need to be enhanced. Kockal and Ozturan, (2010) has study lightweight aggregates from fly ash which met the requirement for freezing and thawing durability [22]. Unfortunately, the porosity and water absorption still high, and the AIV properties still weak which need to overcome. Aho and Ungwa, (2015) has studied the volcanic scoria to be applied as coarse aggregate [23]. The quality of scoria as an aggregate show that scoria concrete has sufficient strength to be used as a construction material. However, the total porosity is high and AIV is still satisfactory which need to be enhanced to increase the durability of construction materials.

Table 1. current issues facing by previous study on lightweight aggregate [19, 20, 21, 22, 23]

Kwek and Awang (2018) [19]	Water absorption 27.80 – 28.0% AIV 22.86 – 28.28%
Rehman et al., (2020) [20]	Loose bulk density 867 – 872 kg/m ³ Water absorption 14.55 – 13.97% AIV 12.67% – 25.1%
Hanif et al., (2021) [21]	Loose bulk density 443 kg/m ³ Water absorption 22.37% Total Porosity 54.97% AIV 24.62%
Kockal and Ozturan, (2010) [22]	Loose bulk density 789 kg/m ³ Water absorption 25.5% Total Porosity 31% AIV 35.0%
Aho and Ungwa, (2015) [23]	Loose bulk density 679 kg/m ³ Total Porosity 40.0% AIV 33.2%

The aggregate impact value test determines the aggregate's resistance to sudden loading [24], which may differ from the resistance to progressively applied compressive stress [25]. As a result, the aggregates must be strong enough to withstand impact disintegration. The impact value of aggregates is defined as the ratio of the weight of fines produced to the weight of the complete sample taken, represented in percentage terms [25] as presented as Table 2. The performance of aggregate via AIV should be within 20% and below to ensure aggregates are resistance towards the impact load. Unfortunately, lightweight aggregate has faced the issue of low aggregate impact value that influenced the durability of the concrete performance itself.

Table 2. Classification aggregate impact value (AIV) [25]

Aggregate Impact Value	Classification
< 10%	Exceptionally strong
10 – 20%	Strong
20 – 30%	Satisfactory
>35%	Weak

The water absorption capacity of lightweight aggregate is determined by the degree of connected porosity related to the surface. Normally water absorption of lightweight aggregate is within the ranges of 10% to 20%, depending on the raw material and manufacturing method [26]. However, large lightweight aggregate water absorption ratios are undesirable. Lightweight

aggregate with high water absorption makes controlling the mixing of lightweight aggregate-concrete during construction challenging. High water absorption leads to reduction in free water in fresh concrete, causing workability and compaction issues which decreases the durability of the concrete. The structure of the lightweight aggregate is one of the reasons for the water absorption. The interior structure of lightweight aggregate can be porous to obtain the low density required of aggregates used in lightweight concrete [21].

Total porosity of aggregate is the behavior of material after immersion in fluid and test assists in the evaluation of total pores in aggregate by calculating the true density and particle relative density. Hence, properties of aggregates have a great impact on the volumetric weight and on physical properties of concrete [19 - 24]. The strength qualities of the concrete are typically poor due to the lightweight aggregate's weaker and more porous nature [23]. Furthermore, the porosity of lightweight aggregate was in agreement with water absorption and density results. When porous lightweight aggregate is used, the adhesion of the aggregate materials to the cement paste depends mainly on the pore size characteristic of the aggregate surface. Because lightweight aggregates are porous, their water absorption is higher than that of normal weight concrete where the amount of pores can be up to 67 %.

With the aim to overcome the current issues facing by lightweight aggregate, there are several design and technique on surface coating on lightweight aggregate have been investigated nowadays. These involve coats of cementitious pastes or pozzolanic materials carbonate deposition on aggregate surface and polymers [19 - 23]. Use of such solutions helps in the impregnation of surface pores, reducing the water absorption of the aggregates and improving their mechanical and durability related performance. The potential of geopolymer paste as a coating material with optimum design (aluminosilicate raw materials, alkaline activator ratio) would become a major solution to limit deterioration by protecting the surface of lightweight aggregate.

Potential of geopolymer coating for lightweight aggregate

Geopolymer is an inorganic material that is used for several applications such as coating application [27]. Geopolymers are created by aluminosilicate materials mixed in an alkali activator solution at ambient or high temperatures, resulting in an amorphous phase and three-dimensional silicoaluminate structure. The traditional geopolymer production process consisted of a two-part mixing process that included preparing an alkali activator one day ahead of time and mixing solid aluminosilicates with the prepared alkali activator [28]. Geopolymers are a novel type of inorganic polymer material having excellent strength, temperature tolerance, corrosion resistance, workability, and adhesive strength between diverse materials [29]. Geopolymer coatings are considered sustainable materials because of their low pollution, cost-effectiveness, and environmental friendliness, as well as their superior performance [28, 29]. The geopolymer coating demonstrated excellent durability and bonding strength [28, 29]. The constituent ingredients and manufacturing of geopolymer coatings are intimately linked to their long-term viability. Reusing and recycling solid wastes and industrial byproducts such as natural source materials, waste or by-product can be used to make geopolymer coating [27-29].

Therefore, the geopolymer material is suitable to be used as a coating agent. Based on the pervious study, to obtain the best design of geopolymer, there are some parameters that need to be set such as raw material, solid to liquid ratio and sodium hydroxide to sodium silicate ratio [30-35]. The most important part of producing and manufacturing geopolymer coating is ensuring that the essential circumstances for geopolymerization reactions are met, which is fundamentally different from the physicochemical mechanism. Selecting proper raw materials and conducting fair and proper combination proportions of constituent materials are vital and crucial in order to generate geopolymer coating that are worth widely used.

Aluminosilicate source materials

Materials mostly composed of amorphous silica (SiO_2) and alumina (Al_2O_3) can be used to produce geopolymers [30-35]. Minerals and industrial wastes such as kaolin, metakaolin, metahalloysite, volcanic scoria, palm oil fuel ash, red mud, fly ash, and slag have all been investigated as raw materials for geopolymer synthesis [27-35].

Fly ash

Fly ash-based geopolymers have gotten a lot of interest in the last two decades because of their wide range of uses in the building and construction industries [30-36]. Because of its accessible availability, superior workability during processing, and better durability in the final product, fly ash has recently become more popular in geopolymers [36-37]. Fly ash is an industrial by-product generated during the combustion of coal for energy production [33, 36-38]. They are called green materials because of the energy-efficient manufacturing process used to create them, which results in low carbon emissions and the reuse of industrial waste in large quantities [30-38]. Fly ash has spherically shaped particles, which are believed to be more workable compare to other cementitious materials [33, 36-38]. Besides, because of its appropriate silica and alumina composition and low water demand, fly ash is a good material for manufacturing geopolymer binder and has good thermal stability [33].

Based on the main chemical composition of fly ash, fly ash can be classified into two classes which is Class C and Class F according to ASTM C618 [39] as shows in Table 3. When the overall content of these components (SiO_2 , Al_2O_3 , and Fe_2O_3) in fly ash exceeds between 50% to 70% with CaO level exceeding 20%, it is categorized as class C. Class C fly ash has a high calcium concentration and is mostly produced by lignite coal combustion [30, 39]. If the overall components (SiO_2 , Al_2O_3 , and Fe_2O_3) in fly ash exceed 70%, it is classified as class F and a CaO level of less than 20% [39]. The high calcium content of fly ash may cause a direct interaction between calcium, silicate, and aluminium, resulting in the formation of calcium silicate hydrates (C/N-A-S-H), or the hydrates may directly form C-S-H gels, resulting in fast setting and high early strength compared to fly ash with a low calcium content. Calcium (Ca^{2+}) works with sodium (Na^+) to balance the negative charge during the formation of sodium aluminosilicate geopolymer [30-38].

Table 3. Classification of fly ash according to ASTM C618 [39]

Chemical	ASTM C618	
	Class C	Class F
SiO_2 , Al_2O_3 and Fe_2O_3	50% - 70%	>70%
CaO	>20%	<20%

Slag

Slag has a Si/Al ratio of 1.71-3.67, making it one of the most frequent raw materials for geopolymers. Slag is extremely reactive to the synthesis of geopolymers, and a satisfactory reaction rate may be obtained at temperatures as low as 0 °C [30, 35]. Slag is a byproduct of the manufacturing of ferrochrome alloys, and its principal oxide components are SiO_2 , Al_2O_3 , MgO , and CaO . The slag is predominantly crystalline and may be used to make geopolymer binder [40]. Slag can be used to increase the porosity, long-term strength, and resistance to sulphate and alkali silicate reactivity of concrete, as well as to lower its water consumption, permeability, and hydration heat [41]. In that case, slag is chosen as a raw material for geopolymer coating because slag can reduce porosity of material. Because of the cement's good adhesion, slag-based geopolymer coatings could be used to protect marine concrete [42].

Metakaolin

Metakaolin is made up of 52% SiO₂, 40% Al₂O₃, and 8% additional elements that are considered impurities, such as feldspar, mica, gypsum, and organic matter [43]. Based on the studies of Kamseu et al the result of calcining kaolin and kaolinitic clays between 650°C and 850°C., depending on the inherent properties of the parent clay. Metakaolin fabrication for geopolymer binders should take into account the mineralogy and thermal behaviour of the source kaolin or clay. Metakaolin is made up of pillared silicate and aluminate layers that have been altered to have the Si atom in a 4-fold coordination and the Al atom in a combination of 4-, 5-, and 6-fold coordination. The product of fine geopolymer paste yields excellent results with this type of calcined clays. It was effectively glazed, and the appearance of shrinkage, deformation, or microcracks was readily controlled by the silica sand residue [44].

Dolomite

Dolomite is a frequent carbonate mineral in the geologic record. CaMg(CO₃)₂ is an anhydrous carbonate mineral comprised of calcium, magnesium, and carbonate. Dolomite is also used to define a sedimentary carbonate rock that is mostly formed of the mineral dolomite, commonly known as dolostone. Dolomite is a mineral resource that may be utilized as a raw material in the production of geopolymer composites [26, 45]. In alkali-activated composites and geopolymer composites, dolomite was solely employed as a filler and substitute for other raw materials [45].

Solid to liquid ratio

The solid-to-liquid ratio (S/L ratio) is one of the key characteristics influencing geopolymer qualities such as compressive strength, water absorption, workability, and geopolymer setting [30-45]. The mechanical strength of geopolymers is significantly affected by the solid-to-liquid (S/L) ratios and sodium silicate-to-sodium hydroxide (Na₂SiO₃/NaOH) ratios [28-32]. In past studies have been conducted to determine the effect of the solid-to-liquid (S/L) mass ratio on mechanical strength, where this factor corresponds to the aluminosilicate precursor-to-activator solution ratio [28-35].

Based on previous research [46], solid/liquid ratios of 0.5, 1.0, 1.5, 2.0, 2.5, and 3.0 were chosen. However, only three ratios were used: 1.5, 2.0, and 2.5. The ratios of 0.5 and 1.0 could not be used because of the high workability of geopolymer paste, which makes it difficult to handle, and the ratio of 3.0 could not be used due to a lack of workability. The ratio of solid to liquid at 2.0 contributed the most compressive strength (73.86 MPa) of the three ratios [47]. Increasing the S/L ratio make the compressive strength of the geopolymer paste increased. However, the compressive strength decreased when the ratio was increased to 2.5 [47]. This could be due to an excess of fly ash, there may not have been enough liquid (alkaline activator) to mix the geopolymers uniformly [46, 47].

Sodium silicate to sodium hydroxide ratio

For the geopolymerization process, alkali metal silicates and/or alkali metal hydroxides have been utilized as alkaline activators [30-47]. Sodium silicate and sodium hydroxide were combined to make an alkali solution. As the alkaline activator to be added to fly ash to form geopolymer coating, a combination of sodium silicate (Na₂SiO₃) and sodium hydroxide (NaOH) has been widely used. The Na₂SiO₃/NaOH ratio is one of the most critical elements influencing the mechanical strength of geopolymer coating materials [27, 48]. The breakdown of aluminosilicate sources requires alkali hydroxide, whereas sodium silicate (Na₂SiO₃) solution works as a binder, alkali activator, dispersant, or plasticizer [30-48].

Furthermore, it will supply extra soluble silicate (SiO₂ content) and alkali (Na₂O content) required for the geopolymerization reaction to continue [42-48]. The SiO₂ content is critical to provide the silicate species required for the fast exchange and oligomerization process between

the aluminate and silicate species from the aluminosilicate-based material and the silicate species from the waterglass [33-48]. In a previous study, six different $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratios were used (0.5, 1.0, 1.5, 2.0, 2.5 and 3.0), the ratio of $\text{Na}_2\text{SiO}_3/\text{NaOH}$ at 2.5 produced the highest compressive strength (61.43 MPa), while greater than 2.5 produced a decrease in strength [42-48] which could be due to the high concentration of OH^- in the mixtures [46, 47].

Concentration of sodium hydroxide (NaOH)

The concentration of NaOH had a significant impact on the dissolution of silica and alumina from fly ash, with increasing molarity increasing the dissociation of the active species of raw material and resulting in the creation of more geopolymer gel networks [36-47]. However, due to an overabundance of OH ions, a high NaOH concentration can disturb the geopolymerization process, resulting in an inefficient reaction [46, 47]. According to previous research, the 12M NaOH solution produced the highest compressive strength of geopolymer paste when the tests were conducted seven days after the samples were prepared, measuring 68.48 MPa but compressive strength decreases after the 12M NaOH solution [46, 47]. This is due to an increase in the concentration of Na ions in the system, which was critical for geopolymerization because Na ions were used to balance the charges and form the alumino-silicate networks as the binder in the mixture [46, 47]. The dissolution of Si^{4+} and Al^{3+} ions from fly ash also increased when the NaOH concentration was high, and the creation of sodium aluminosilicate was boosted, resulting in an increase in strength [46, 47]. The setting time and compressive strength improved as the NaOH content increased, resulting in a robust matrix with less unreacted fly ash in the microstructure [36-47]. The S/L ratio and the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio are two other factors that influence the compressive strength of geopolymer coating material [46, 47].

As a conclusion, the ratios of fly ash to alkaline activator and sodium silicate to NaOH influenced the strength of the geopolymer. The specific surface composition of initial raw materials, the kind, composition, and relative amount of alkali activator, and the situation during the beginning period of the geopolymerization process all have an impact on the attributes of geopolymers. The effect of NaOH molarity may also be significant in the production of high-strength geopolymer. Knowing how much alkali to use not only helps to acquire optimal strength but may also help to handle the efflorescence problem with geopolymers, and other alkali activated materials.

Coating method for lightweight aggregate

The coating method is also an important parameter in finding the best surface coating for lightweight aggregate or any substrate. This is thought to be heavily influenced by the geopolymer coating formulation, which is calculated by taking into account the content and composition of the amorphous part of the geopolymer used which contributed to the viscosity of the coating paste [27, 34, 48]. Coating is a process that improves the surface attributes of an object or substrate, such as appearance, adhesion, wet ability, corrosion resistance, wear resistance, scratch resistance, and thermal resistance. The application of the coating also extends the life of existing infrastructure or products while lowering maintenance costs [27, 34, 48]. One of the enhancements of the properties of aggregate is coating. Coating significantly reduces aggregate porosity and density, therefore enhancing aggregate microstructure and mechanical qualities [27, 34, 48]. One coating material that been use is geopolymer that is frequently used to create and is coated on a ceramic or a metal substrate [27, 29, 34, 48]. Due to the obvious wide range of applications and needs in various industries, coating processes are available in a large variety. Many alternative on-line or off-line parameters are used in these processes, which result in a wide range of material microstructure, efficacy, appropriateness, and durability outcomes. Coating methods, on the other hand, are useful in specific applications based on the intended functionality. There are various influencing parameters for successful coating deposition on a substrate,

including deposition materials, substrate materials, feedstock shape, and deposition processes [27, 29, 34, 48].

Milling coating method

Milling is a widely used technique for dispersing particles on typically metal powders: it is a non-equilibrium process that operates at low temperatures, with essentially linear processes and a cheap total cost [49]. In order to have optimal control of the results, the ball milling is difficult; several variables such as ball size, rotation speed, duration, atmosphere, and process control agents (PCA) need to be considered [49]. Yazdani, and Zake, (2015) found out that the discovery clearly supports and verifies the chaotic ball dynamics notion in a planetary ball mill with a huge number of balls [50]. It was discovered that mechanical and metallurgical bonding mechanisms have a role in the adherence of Ni coatings to the substrate. Furthermore, the substrate location at the top of the vial was shown to give higher mi- and tribological characteristics for the coating as compared to the bottom position as illustrated in Fig. 1 [50]. Milling coating method would be good method for surface coating lightweight aggregate with the aim to uniform and fully cover the surface of the lightweight aggregate.

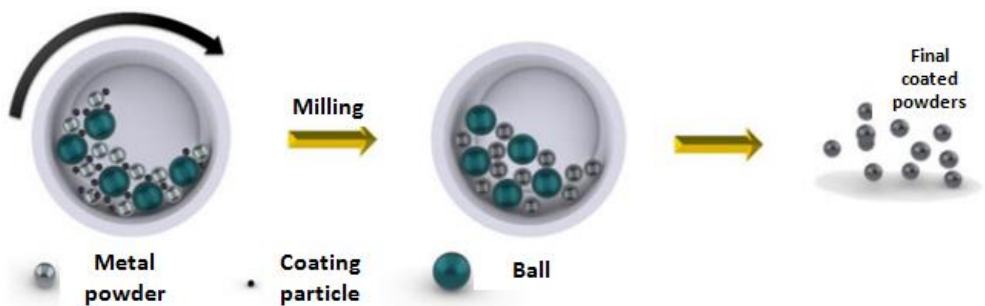


Fig. 1. Scheme of milling coating process [50]

Dipping coating method

Dipping coating is a simple and cost-effective process for depositing onto a variety of substrates, including metallic, ceramic, polymer films, and fibrous materials [51, 52]. Dipping coating may be applied to practically any substrate, even those with a lot of surface area [51-53].

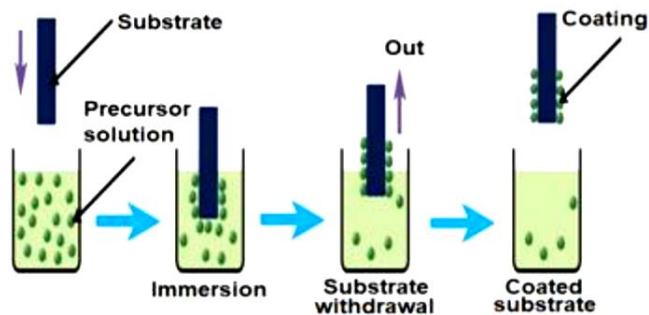


Fig. 2. Scheme of dipping coating process [54]

The overall process can be described as in Fig. 2. The substrate is immersed at a constant speed in the precursor solution, followed by a certain dwell time, to allow for sufficient interaction

time of the substrate with the coating solution for complete wetting [51-54]. It was discovered that extending the time for sol synthesis prevents the formation of a crack-free separation layer on membrane support due to the formation of thick active layers that would otherwise crack during the drying and carbonization steps [51-53]. Similarly, a very short synthesis time necessitates the need for multiple coating cycles to obtain a defect-free active layer. Controlling the microstructure of the substrate and the dip coating slip characteristics, uniform, and dense coating layers with a wide range of thickness can be generated would also be good for surface coating lightweight aggregate.

Conclusions

As a conclusion the coating of lightweight aggregate using geopolymer coating is an excellent idea. The coating of geopolymer has been proven to enhance the properties of lightweight aggregate. The coating of aggregate can decrease the water absorption of the lightweight aggregate. In the geopolymer it is important to determine the rate of the alumina and silica releases during the reaction. This is because alumina and silica releases influence the formation of the geopolymer network. Beside that the parameters of solid to liquid ratio influence the viscosity of the geopolymer material. These parameters also influence the setting time of the geopolymer coating. One of the results of the viscous nature of the water glass liquid as the $\text{Na}_2\text{SiO}_3/\text{NaOH}$ ratio increased and the high quantity of water glass liquid caused the geopolymer slurry to become exceedingly sticky. Therefore, the parameter of the geopolymer is really important to determine the design of geopolymer coating. On the other hand, the method of coating is one of the important mechanisms to be determined to show the thickness of the material is evenly.

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