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A Review of Geopolymers-Based Artificial Aggregates Technology Developed Using Waste Materials

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Abstract Scholars around the world are concerned with the continuous reduction of aggregates-related natural resources globally. This led to recent studies on the importance of reusing and recycling waste generated from biological materials and industry by-products. The interest in the exploration of waste was due to the increase in the demand for aggregates normally used as a major component in producing concrete. With the continuous development of technology, the stock of natural aggregates on Earth is declining, hence, an alternative is required to replace natural aggregates. Geopolymer artificial aggregates are aggregates made by several methods such as sintering, autoclaving, and cold bonding with alumino-silicate precursors obtained from waste materials such as metakaolin, slag, red mud, fly ash, and calcined kaolin sludge which are activated using an activator. The activated precursor using this activator causes a polycondensation process called geopolymerization. Geopolymers system was discovered to have remarkable attributes such as exceptional force, enhanced endurance, and heightened fire tolerance, which can be a compelling substitute for aggregates in Ordinary Portland Cement (OPC) concrete. Therefore, this study explores and collects various studies related to the methods used to produce geopolymer-based aggregates and their characterization. The focus is on the production of these produced aggregates, and appropriate approaches are established to improve the quality of the aggregates produced, accompanied by insightful suggestions for future investigations.

Keywords Artificial Aggregates, Waste Materials, Sintering, Cold Bonding, Geopolymers

1. Introduction

Aggregates such as gravels, crushed stones, and sands are one of the basic materials in producing concrete and asphalt through their mixture with a binder [1]. Aggregates can be classified into coarse types such as gravel as well as fine types such as sand. The coarse aggregates are further categorized into two including artificial and natural. The continuous development of technology was observed to be depleting natural aggregates stock on earth, thereby leading to the demand for alternative materials to be used as replacements [2, 3]. This led to the development of artificial aggregates such as fly ash from waste. Fly ash, one of the disposable wastes from combustion, can be mixed and reacted with an activator to form aggregates [4].

The need to handle these problems led to the proposition of three technological methods including recycling aggregates from waste concrete [3], directly utilizing coarse waste particles, for example, steel slag and bottom ash as aggregates [5], as well as producing manufactured aggregates obtained from industrial waste or by-products [6]. Recycling aggregates encompasses the process of reusing waste materials derived from construction and demolition (C&D) activities such as glass, steel, and potentially hazardous substances such as mercury and

asbestos. However, these materials require additional treatment procedures. Direct use of coarse waste materials like bottom ash [7] as well as steel slag [5] also demands improved processing methods in the form of steam pressure and wet treatment. This is the reason artificial aggregates are continuously being recognized as the viable option to generate coarse and fine aggregates needed to fulfill mass production and waste recycling to confirm environmental sustainability.

Using waste materials is also one of the means to reduce the problem of waste processing, significantly conserve irrecoverable natural resources, reduce environmental pollution, and save the production of recycled energy [8]. However, there is a need to consider industrial wastes and by-products as the materials having high potential to be used for this purpose. This is due to the fact that the application of the right treatment and methods to these materials can produce environmentally friendly products. Some of these industrial wastes that can be used as raw materials for artificial aggregates comprise coal fly ash, palm oil fly ash, bottom ash, agricultural and plastic wastes, as well as crumb rubber as shown in Figure 1.

The features of artificial aggregates need to match those of natural aggregates. Therefore, there is a need to focus on the processing and production methods. The two popular methods normally used to produce artificial aggregates are sintering [10] and cold bonding [11]. The sintering relies heavily on high temperatures, typically exceeding 1000 ℃ to crystallize raw materials. Meanwhile, the conventional cold-bonding procedure typically requires using cementitious pastes to bind the raw components. The products from these two methods are usually more lightweight, water-absorbent, and weaker than natural rock aggregates. Despite having higher water absorption, lower strength, and higher bulk density compared to sintered aggregates that require more energy for the same raw materials, cold-bonded aggregates are regarded as more cost-effective and eco-safe, especially when produced on a large scale [12].

Geopolymer is an eco-safe option to replace Portland cement. This material is normally created through the chemical procedure or polymerization of an alkaline activator and an aluminosilicate substance obtained as a byproduct of industry, such as blast furnace slag or fly ash [13]. Fly ash is usually obtained through combustion or it can be referred to as a waste component [14]. It can be further mixed with alkaline activator to produce artificial aggregates [14]. Geo-polymerization entails a rapid chemical process between Si-Al minerals in alkaline states. This reaction results in the formation of a three-dimensional polymer chain and ring structure that comprises Si-O-Al-O bonds [15].

Geo-polymerization process aims to obtain more environment-safe artificial aggregates. The process involves the manufacture of new materials in the form of a tetrahedral bonded SiO_4 (Silicate) and AlO_4 (Aluminate) polymer framework from the geosynthesis of alkali-silicates and polymeric aluminosilicates. This tetrahedral bonding of SiO_2 (Silicon Dioxide) and Al_2O_3 (Aluminum Oxide) by sharing oxygen atoms requires the presence of positive ions (K+, Na+, Ca++, Mg++, NH++) in the skeleton holes to balance the negative charge of Al_3+. The product usually contains a high volume of silica and alumina.

Figure 1. Waste feasible for use as aggregates [3, 9]

To attain the desired characteristics of cold-bonded aggregates, geopolymers can be utilized. Previous research demonstrated that geopolymer lightweight aggregates (GLAs) can achieve strength without the need for high-temperature sintering. Instead, they acquire strength through the alkali activation of pozzolanic materials [16]. Meanwhile, standard cold-bonding with ordinary Portland cement binders requires adjusting the geo-polymerization process through certain factors such as the precursor kinds activator contents, activator kinds, and curing methods, to reach the preferred properties [17].

The features of concrete are significantly determined by aggregates, which constitute 70% of its total volume. The continuous expansion of the worldwide population and construction requirements has led to a substantial decline in these aggregates. This further increased the need to produce artificial aggregates to suit global concrete industry needs [18].

Pelletization is popular globally despite the application being limited to steel and iron making, as well as agricultural industries due to the resources available in the construction field. However, its benefit is presently earning attention to solve the aggregate deficiency. This was observed to have led to the conduct of research on artificial aggregate manufacture involving industrial by-products, for instance, bottom ash, fly ash, and others through the agglomeration technique [19]. Synthetic aggregate manufacture can also be accomplished by agglomerating fly ash into lumps with a pelletizer and employing various hardening approaches like cold bonding, sintering, water curing, steam curing, and autoclaving for further processing [20]. Sintering was observed to have been used extensively when the fly ash has a carbon range of 2-6%. Meanwhile, the alternative methods, autoclaving and cold bonding, rely on the inclusion of binders like CaO, regardless of the carbon [20]. Cold bonding requires significantly less energy. It was noted that the manufacturing efficiency of these pellets is normally determined by certain aspects such as binder type, raw components, angle of the drum, the speed of revolution, duration, and dosage of the pelletization [21]. Furthermore, different types of pelletizers such as drum, disc, mixer, and cone types can also be used. The disc type is considered preferential because of the ease of controlling the dispersal of the pellet sizes [22]. Previous studies also showed that the addition of chemical substances is essential to enhance both strength and durability [23].

2. Geopolymers as Artificial Aggregates

Aggregates are responsible for more than 50% of the elements used in producing concrete in the construction industry. Nevertheless, many countries and cities have implemented restrictions on the use and extraction of natural aggregates like sand, rock, and gravel. These measures aim to protect nature from the detrimental effects of excessive exploitation of these valuable resources [24,25]. This led to the introduction of several alternatives to minimize the instantaneous usage of natural aggregates in reducing the pollution connected with the increase in waste and limited availability of landfills. To promote reusing waste materials, various initiatives have been implemented, including the introduction of recycled and man-made aggregates. These efforts aim to encourage the utilization of these alternative materials in place of natural aggregates [26].

Artificial aggregates were introduced to reduce the concern for the growth of waste products in the environment. The process involved using wastes as precursors to produce environmentally friendly products. Recycling solid wastes into artificial aggregates was believed to be contributing to endurable growth due to the reduction in the deficit of natural aggregates caused by infrastructure expansion [27, 28]. Moreover, the inclusion of geopolymers in the manufacture of artificial aggregates was focused on producing a material with a strong surface coating and qualified structural accomplishment to be ideally used in lightweight concrete and others such as lightweight geotechnical filling and insulation of goods. A summary of some of the literature can be seen in Table 1 and Table 2.

Table 1. Previois studies on Geopolymers-based Artificial Aggregates with diverse mixing parameter ratios and processing techniques

Researchers	Precursors		Mixing Parameter Ratio	Processing
	Raw Materials	Alkali Activator		
[2]	Red mud, coal fly ash	Na_2SiO_3	Solid-to-liquid ratio (0.34–0.39) Modulus ratio, Ms (1.29–3.84)	The shaping process used a disc pelletizer. Subsequently, curing was performed at 20 ◦C and relative humidity of 50%
[8]	Coal fly ash and slag	NaOH, Na_2SiO_3	Various fly ash levels (80–90%), various slag levels (10–20%)	Cold pelletization bonding was implemented, utilizing dry curing with a raised temperature of 70 ◦C to cure the aggregates
[22]	Class F fly ash and slag	NaOH, Na_2SiO_3	Diverse alkali equivalent, AE (5–9%) and alkali modulus, Ms (0.6–1.0)	The hardened paste of alkali-activated fly ash-slag was shaped by crushing and then split into coarse and fine sizes through sieving
[25]	Class F fly ash, silica fume	NaOH, Na_2SiO_3	Alkali activator percentage (NaOH-Na_2SiO_3) 20–80, 30–70, 40–60	Hand shaping and then microwave curing
[26]	Class C fly ash, Class F fly ash	NaOH, Na_2SiO_3	The influence of various raw materials, specifically Classes C and F fly ash, on the performance of fine aggregates was examined.	The samples were subjected to heating at 100 ◦C for 1 hour and subsequently left at ambient temperature for one day.

Table 2. Aggregates size, curing, water abs, and crushing strength

Researchers	Raw Materials	Size of Aggregates (mm)	Curing (℃)	Water Abs (%)	Crushing Strength (MPa)
[29]	FA, Cement, Lime	4,75 - 19	21	30	-
[30]	FA (F Class)	10,25	1100	25,125	1,4
[31]	FA (F Class)	10 - 20	100	21,56 - 52,65	2,04 - 8,725
[32]	FA (F Class)	2,36 - 19,1	70	6,08 - 10,24	-
[33]	FA (F Class)	10 - 12,5	80	4,3 - 17,8	1,8 - 4,1
[34]	FA (F Class)	2 - 10	20	22 - 23	-
[35]	FA (F Class)	6 - 20	80	10 - 100	26 - 75
[36]	FA, Cement	0,25 - 8	21	57,8	0,96
[37]	FA (F Class), Bentonite and GP	10	110, 1100, 1150, 1200	0,7 - 18,4	5,1 - 23,1
[38]	FA (C Class), Cement	10 - 12,5	21	1,23	-
[39]	FA, Bentonite, Kaolinite	4,75 - 19	>1000	-	-
[40]	FA (F Class)	10	1100	21 - 22	-
[41]	FA+GGBS+RHA	18 - 20	23	9,8 - 10,1	8,1 - 8,8
[42]	FA	2 - 8	In water	0,2 – 3,55	30 - 52

Geopolymers are binder materials with zero Portland cement content, created through polycondensation of alumino-silicate sources obtained from waste materials, in alkaline solutions [43-45]. Geo-polymerization normally leads to the production of inorganic polymer 3D networks with cross-linked polysialate chains [46]. It is important to note that regular Portland cement usually uses calcium silicate hydrates (C-S-H) gel as the primary binding compound. Meanwhile, geopolymers are formed through a polycondensation process called geo-polymerization, which involves the reaction of silica (SiO_2) and alumina (Al_2O_3) sources in a highly alkaline environment, typically using NaOH or KOH solutions [43]. During geo-polymerization, oxygen, aluminum, and silicon atoms combine to form a chain consisting of SiO_4 and AlO_4 units, where they are linked alternately by sharing oxygen atoms [46]. However, the term poly-sialate is proposed as the chemical designation for geopolymers that are primarily composed of silica-aluminate compounds [47]. The simplified representation of the involved chemical processes in the geo-polymerization is presented in the following Figure 2 [43].

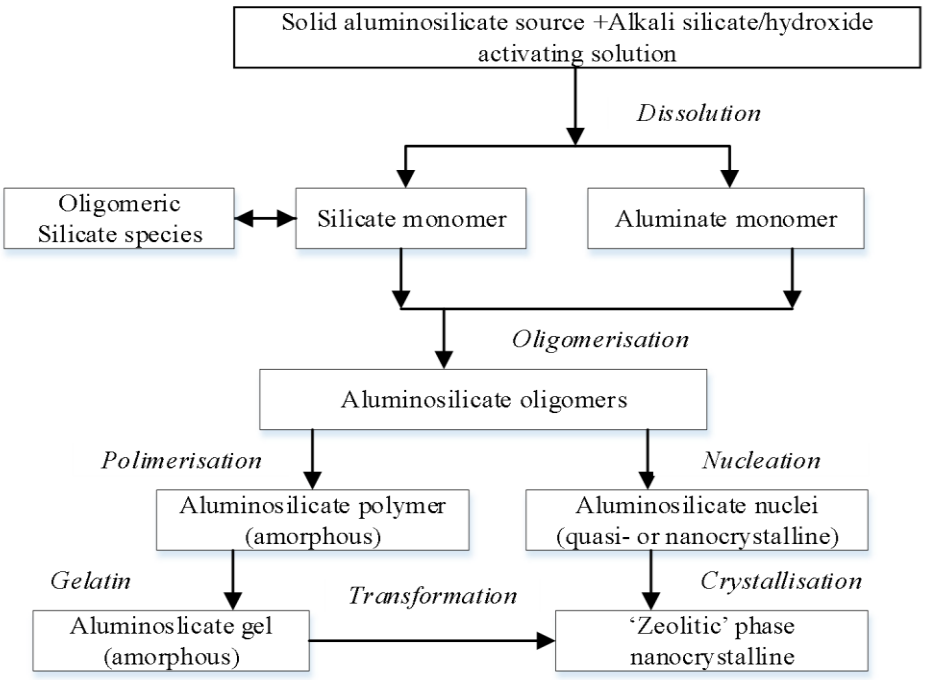

Figure 2. The chemical processes in geopolymerization [43]

An amorphous alumino-silicate cementitious material is geopolymer. The technique of geopolymerization, which involves polycondensation of a geopolymeric precursor and alkali polysilicates, can be used to create geopolymers. Amorphous to semi-crystalline polymeric structures with Si-O-Al and Si-O-Si links are produced via geopolymerization, which is a heterogeneous chemical reaction between solid aluminosilicate oxides and alkali metal silicate solutions at extremely alkaline conditions and mild temperatures [15]. The ability of the aluminium ion (6-fold or 4-fold coordination) to generate chemical and structural changes in a silica backbone is necessary for geopolymerization, a type of geosynthesis (reaction that chemically integrates materials).

Alkali activation of materials made of aluminium silicates is a complicated process that hasn't been fully explained yet. Si-O-Si bonds are first broken when aluminosilicate minerals react in a highly alkaline environment. Later, new phases develop, and the mechanism for their production appears to involve a solution ("synthesis via solution"). A key aspect of this reaction is the penetration of Al atoms into the initial Si-O-Si structure. The majority of generated geopolymer precursors are alumino-silicate gels. Their composition can be characterized by the formula:

$$Mn[-(Si\text{-}O)z - Al\text{–}O]n.\ wH_2O. \quad (1)$$

Where: M = the alkaline element or cation such as potassium, sodium or calcium

n = the degree of polycondensation or polymerisation

z = 1,2,3 or higher

Depending on the composition of the starting components and the circumstances of the reaction, the C-S-H and C-A-H phases may also form. These (poly-condensation) reactions may even result in the formation of secondary H_2O. Depending on the nature of the initial raw materials and the circumstances of the reaction, amorphous (gel-like), partially amorphous, or crystalline substances may be produced. The amount of solid matter present has a significant impact on the alkali activation process [15].

Most proposed mechanism consisting of the chemical reaction of geopolymer may comprise the following steps:

1) Dissolution of Si and Al atoms from the source material through the action of hydroxide ions.
2) Transportation or orientation or condensation of precursor ions into monomers.
3) Setting or polycondensation/polymerisation of monomers into polymeric structures

The term "polysialates" refers to these frameworks, while "sialate" refers to the silicon-oxo-aluminate building block. The SiO_4 and AlO_4 tetrahedra that make up the sialate network are joined by exchanging all of their oxygen atoms. Positive ions (Na+, K^+, Ca^{2+}, etc.) must be present to balance the negative charge of Al in 4-fold coordination. Chains and rings may be formed and cross-linked together, always through a sialate Si-O-Al bridge. Polysialates are chain and ring polymers with Si^{4+} and Al^{3+} in 4-fold coordination with oxygen and range from amorphous to semi-crystalline. The amorphous to semi-crystalline three dimensional silico-aluminate structures were classified as 'geopolymer' of the types [48].

2.1. Precursors

Industrial by-products, for instance, palm oil waste, fly ash, blast furnace slag, silica fumes, and ferrochrome slag, can pose challenges to the environment in terms of land deficiency for waste dumping, thereby significantly increasing the costs associated with the treatment or disposal [49]. Therefore, the introduction of these resources to produce concrete is directed towards reducing the waste produced by various industries, along with the carbon footprint linked with concrete manufacturing. Recycling and reusing waste materials have increasingly become a subject of research interest in recent years, particularly with a focus on the geopolymer sector [50]. Geopolymers typically possess high silica and alumina contents, often exceeding 50% of the total sum of SiO_2 and Al_2O_3. Consequently, the appropriate selection of aluminosilicate precursors is crucial for their utilization as artificial aggregates. Some of the examples of these aluminosilicate materials that have been extensively explored include slag, kaolin, fly ash, metakaolin, red mud, volcanic ash, and rice husk ash.

Pozzolanic materials such as metakaolin and bentonite were also used in some previous studies as binders in producing geopolymer aggregates [51] instead of the usual alkaline activators [52]. The process involved collecting appropriate sizes of pellets in a disc relying on the pelletizer speed, the disc angle, and the moisture. The fresh pellets were hardened through cold bonding, sintering, or autoclaving methods to improve the aggregate strength.

Any chemically activable by-products or waste materials containing aluminosilicate minerals are generally considered potential raw materials to produce alkali-activated aggregates. These precursors can be employed individually or in combination, encompassing a wide range of materials such as industrial wastes (metakaolin, furnace bottom ash, coal fly ash, ground granulated blast furnace slag, kaolin, red mud, and bentonite), agricultural wastes (peat-wood fly ash, palm oil fuel ash, and rice husk ash), municipal wastes (sewage sludge and dredged sediment), as well as natural resources (volcanic ash and mine tailings). Meanwhile, in recent studies related to alkali-activated aggregates, coal fly ash has emerged as the most frequently utilized material due to its abundant availability and cost-effectiveness [52].

The utilization of geopolymers can be determined by the Si/Al ratio, which plays a crucial role in defining the microstructure of various types of geopolymers. This highlights the significance of silicon (Si) and aluminum (Al) as fundamental elements in the composition of geopolymers. A risen Si/Al ratio can lead to the production of a denser microstructure for geopolymers, thereby enhancing the mechanical property and strength performance. This simply shows that Si/Al, Na/Al, as well as free water, contribute to the micromorphology and macro strength of metakaolin-based geopolymers. Moreover, the ability of SiO_2 and Al_2O_3 to regulate the viscosity of the glass phase has a direct impact on the characteristics of artificial aggregates. It is also important to note that reactions involving aluminate and silicate are faster than those with only silicate. Industrial wastes have been recycled or used to construct artificial aggregates but the application of geopolymers for the same purpose has not been fully explored. This needs to be researched to produce environmentally friendly aggregates for construction purposes. Moreover, the production, shaping, and curing methods need to be studied in addition to the influencing factors.

2.2. Activators

Previous studies employed sodium silicate (Na_2SiO_3) and Sodium hydroxide (NaOH) as alkali activators [25]. Activations were defined as the substances or elements causing other elements to react. The activator used in this study contained NaOH and silica, which is a strong acid with the ability to react with a strong base. Meanwhile, Na_2SiO_3 was used to accelerate the polymerization reaction and NaOH reacted with the Si and Al in the fly ash to produce strong polymer bonds. The alkaline activator was produced by blending the NaOH solution with a Na_2SiO_3 solution, and the mixture was allowed to stay for 1 day to achieve equilibrium before utilization.

Figure 3 illustrates the various silica extraction procedures employed to create sodium silicate activators using different silica resources. The activators produced were mainly used to activate fluid catalytic cracking catalysts, metakaolin, slag, red mud, fly ash, calcined kaolin sludge, and water treatment sludge. Moreover, the extraction techniques considered include fusion, hydrothermal, and thermochemical methods. The dissolution of the silica was based on important factors such as processing time and temperature. The stirring and heating method was expected to commonly enhance the dissolution speed of the silica because the kinetics of chemical reactions normally increase with temperature and external stimulus. Therefore, the processing time ranged between 5 min and 72 h, and the temperature from the room value to 120 ◦C to generate alkaline activators employing the hydrothermal procedure [27].

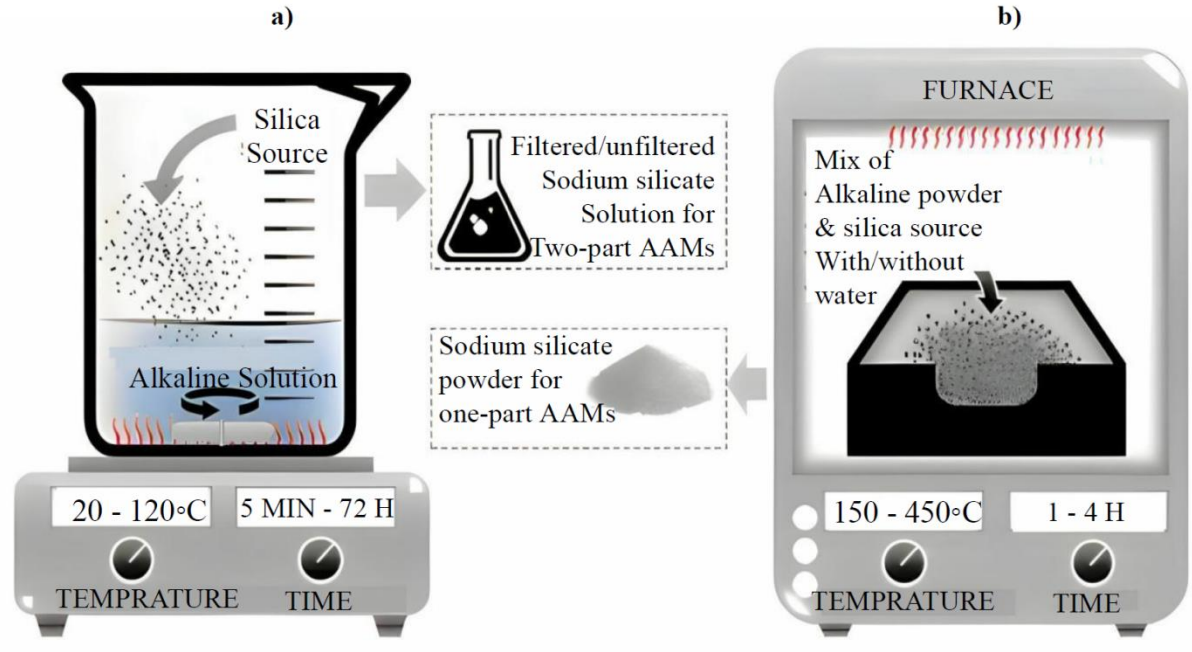

Figure 3. Comparison between (a) hydrothermal and (b) thermochemical procedures with diverse processing parameters (time and temperature) to produce sodium silicate activator [27]

The alkaline solution commonly used in preparing artificial geopolymers aggregates is the solid form of sodium hydroxide (NaOH) which is normally dissolved using distilled water to achieve the desired concentration and then a solution of sodium silicate or water glass (Na_2SiO_3). The inclusion of sodium hydroxide in geopolymers aggregates mixture was to act as an alkaline reactant while the sodium silicate acted as a catalyst to accelerate the polymerization reaction. Furthermore, there was a high level of alkaline solution to the pozzolan used, fly ash, in geopolymers aggregates binder. These levels were important to the water permeability and compressive strength of the concrete produced. The sodium hydroxide applied as the alkali activator reacted Si and Al in fly ash with lime to deliver strong polymer bonds. Meanwhile, the sodium silicate s sped up the polymerization reaction.

The alkali-activated aggregates were treated with water glass, NaOH solutions, or their combinations, which served as conventional wetting and binding agents. These options were selected because of their favorable performance, widespread availability, and cost-effectiveness. Meanwhile, in a previous study, a combination solution of K-silicate and KOH was employed as an activator for cold-bonded aggregates [53]. There is recently heightened interest in the need to improve sustainability and recycling through the application of waste-derived alkali as activators. A previous study [39] enforced a medley of conventional alkali-activator (NaOH) with waste alkali source ($NaAlO_2$ solution) for cold bonded aggregates production. This method was considered promising for further studies on the lessening of cost and advancement of the environmental performance of these aggregates. The manufacturing of these materials up to date has been through liquid-based activating solutions using the terminology known as two-part mixing [54]. A significant drawback of this method is the high alkalinity of the solutions, which leads to their hostile nature, high viscosity, lack of user-friendliness, and increased costs, particularly during transportation and delivery. One-part (or "just add water") alkali-activated materials developed using solid-based activators were believed the new technology to solve the aforementioned issues. However, the one-part mixing technique has not been thoroughly examined, indicating that the influences of different mixing techniques on the properties and production necessitate further exploration. The higher molecules condensed in a gel form while the alkali attacked the surface of the particle and expanded to a larger hole. It further exposed the hollow or partially filled smaller particles with other yet smaller ashes to bidirectional alkaline attack from the outside in as well as inside out. Consequently, the product of the reaction was generated both inside and outside the shell of the sphere up to the moment the ash particle was completely or almost completely consumed.

3. Processing and Producing Geopolymers-Based Artificial Aggregates

Geopolymers-based artificial aggregates were generally created through a geo-polymerization process involving three steps. The process involves several steps, for instance, the dissolution of Al and Si elements from the raw materials, the orientation or transportation of precursor ions to monomers, and the polycondensation of monomers to form polymeric structures. The mixture of the liquid and solid precursors based on their respective mix marks needed to be followed by the formation of aggregates and the shape can be achieved through different methods including pelletizing, die casting, crushing, and hand forming [55].

The production of artificial aggregates followed a fundamental principle that entailed the mixing of raw materials, agglomeration, particle binding or hardening, and subsequent processing steps such as sintering as well as curing. The desired sizes were determined by agglomerating the raw waste materials during production [56]. Moreover, the known methods available to produce artificial aggregates after the initial steps were cold bonding, sintering, and autoclaving. The common method to obtain lightweight artificial aggregates was found to be sintering process. It was also noted that the application of cold bonded method with alkaline activators for lightweight aggregates led to the making of high-performance concrete [56]. Furthermore, pelletized aggregates were generally produced through the pelletizing method with due consideration for efficiency based on certain factors such as the pellet disc rotation speed, moisture content, inclination angle of the pellet discs, and pelletization duration [55]. The pelletizing process was included in both the agglomeration and granulation stages. The granulation technique influenced aggregates properties linked to the binding aspect such as the interaction forces, binder viscosity or adhesion, binder adhesion in the electric double layer, liquid surface tension, Van der Waals forces, and electric charge attraction between powder [56]. The primary controlling factors in granulation are the first two mentioned. The quality of bond granulation was also observed to be influenced by several factors, including the method applied, the operating conditions of the granulator, the catalyst, and the characteristics of the binder or additives used. Another important observation was that mold casting was commonly preferred for laboratory-scale study due to its suitability. The destruction process has been widely utilized as an effective means to transform natural aggregates into the expected grade. However, hand shaping, being a traditional technique reliant on human labor, was deemed unsuitable for large-scale production. Some studies suggested that the incorporation of waste gas formers into the fresh mixture, shaping it into spherical forms, and subjecting it to high temperatures ranging from 1100-1200 ℃ as indicated in Figure 4 could potentially yield lightweight aggregates.

Figure 4. Particles of geopolymers artificial aggregates at different stages of heating [5]

3.1. Sintering

Sintering was used in previous studies to produce lightweight aggregates in a furnace up to 1000 ℃ - 1200 ℃ in fusing particles at the points of joint contact [56]. The raw materials were pelletized through a mixture with 20 – 30 % water used as a binding agent to obtain the desired and consistent size after which they were dried at 1100 ℃. Various types of pelletizer machines, like disc or pan, drum, cone, and mixer types, can be utilized during agglomeration through granulation [57]. The mechanical and physical characteristics of aggregates were observed to be affected by the revolution speeds during the pelletization process with a rotating disk. The changes were associated with the increase in particle coalescence up to a maximum scale. The sintering method necessitated significant energy input, but it offered high engineering aspects contingent upon the characteristics of the agglomerated materials and procedure efficiency. In some cases, fly ash aggregates underwent a two-step thermal treatment process. The first phase involved applying heat at 750 ℃ for 10-15 minutes, followed by a second expansion phase at 1150-1175 ℃ for the same duration [58]. When compared to products obtained from one-step heat treatment, the materials resulting from the two-step thermal treatment exhibited increased porosity and demonstrated similar properties to commercially available lightweight aggregates.

Sintered aggregates were produced through the fusion of fresh pellet particles at a high temperature which was often above 1000◦C. The production process was gradual and involved the vitrification and expansion of fresh pellets. Sintering is a multifaceted strategy that encompasses both chemical and physical procedures. Therefore, the raw materials and method parameters used have the greatest influence on the features of sintered artificial aggregates. Some of these parameters include temperature and duration similarities with cold bonding and can be divided into three stages: raw material mixing, pelletization or granulation,

and subsequent hardening through sintering at elevated temperatures. Sintering is considered time-saving, even though it is energy-consuming, because the duration for the pre-heating and actual process is estimated at 1 h.

Sintering process is generally perceived as reliable for the making of high-quality and lightweight artificial aggregates. This is because the product's mechanical features are usually comparable to the commercialized standard. Additional heat is normally applied to this technique to enhance the reaction process to generate artificial aggregates, specifically lightweight models. This further expands its applicability to different types of raw materials instead of being limited to only those with cementitious properties.

3.2. Autoclaving

Autoclaving is a process of mixing chemicals including gypsum, lime, or cement with the source materials in the agglomeration step. Subsequently, the sample area is normally autoclaved or cured for several hours in a pressurized environment with saturated steam at 1400C. The procedure can be used to produce artificial aggregates with minimum binding elements and a short curing duration [59].

Commercialized autoclaved construction goods are made by combining calcareous elements like cement and quicklime, siliceous materials like quartz sand or fly ash, and H_2O under pressurized steam at temperatures ranging from 125 to 200 ℃. This showed the possibility of using this method to produce artificial aggregates. It is pertinent to note that the pressure and temperature of the autoclave were influential in reinforcing the pellets produced during the pelletization process. However, to date, there is a scarcity of studies exploring the potential of utilizing autoclaving in the artificial aggregate manufacture, particularly concerning geopolymers.

3.3. Cold Bonding

Cold bonding is the use of conventional water curing at room temperature to connect mixed materials. Products are typically stiffer and have more drying shrinkage. Moreover, its drawbacks can be managed with compaction approaches such as roll pressing, uni-directional pistons, extrusion, and pellet mills during agglomeration. The application of class c fly ash aggregates demonstrated a gain in the span of normal water curing lowered the absorption rate and produced aggregates having similar features with autoclave because of dense microstructure formation.

In this method, the process involved utilizing standard curing water at room temperature where the fly ash (FA) reacted with calcium hydroxide to produce a moisture-proof binder. Compaction agglomeration techniques were used to combat drying shrinkage and bonding material creep. Accordingly, the joining of solid particles to form bigger shapes into larger shapes is achievable through different methods such as agitation, drum, or disc granulation [56]. This method not only assisted in conserving energy compared to other manufacturing methods but also ensured comparable aggregates properties against autoclaving and steam curing, while simultaneously providing maximum strength.

4. Conclusions

In conclusion, geopolymers aggregate were observed to be attaining a reputation because of the deficit of conventional coarse aggregates. The concept gained significant attention in the past decades concerning the production of aggregates despite its long history. This was due to the dual benefit of using precursors in geopolymers aggregates production which was focused on long-term environmental protection and improved properties right from the early stages. The incorporation of geopolymers in the manufacturing process enhanced the characteristics and overall quality. Cold bonding showed fantastic aggregates features, including strength while also preserving energy by eliminating the need for high temperatures during curing and hardening. On the contrary, aggregates produced through autoclave and sintering consistently exhibited prominent quality in terms of lightweight, water absorption, and force characteristics. The inclusion of geopolymers contributed to consistent pore disbandment and enabled the manufacture of lightweight aggregates at lower temperatures compared to those without these materials.

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