Biomass Fly Ash-Based Geopolymers

Subjects: Materials Science, Ceramics

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The production of conventional cement involves high energy consumption and the release of substantial amounts of carbon dioxide (CO_2), exacerbating climate change. Additionally, the extraction of raw materials, such as limestone and clay, leads to habitat destruction and biodiversity loss. Geopolymer technology offers a promising alternative to conventional cement by utilizing industrial byproducts and significantly reducing carbon emissions.

Keywords: biomass ; fly ash ; geopolymer ; circular economy

1. Introduction

Based on a recent report published by the World Meteorological Organization, the year 2021 has been characterized as a period of unprecedented achievement. The sea level has recently attained a new record high, posing significant challenges for coastal populations and small islands. The levels of ocean heat and acidification are currently unparalleled. As anticipated, the concentration of greenhouse gases continues to increase [1]. Between 2000 and 2010, greenhouse gas emissions had grown by 24%, 3 times as much as the increase in the previous decade [2]. Consumption of fossil fuels, particularly coal in power plants and the iron/steel production industries, significantly impacts CO_2 emissions [3]. Moreover, the cement industry also emits a non-negligible and increasing amount of greenhouse gases. In 2015, cement production accounted for approximately 2.8 billion tons of CO_2, about 8% of global emissions and roughly 4 times more than air transport [4].

Concrete is a composite material composed of sand, water, and cement, which undergoes a process of hardening and bonding the individual sand grains together. It consists of calcium oxide (CaO), silica (Si_2O), and additional binding agents. The process involves subjecting a mixture of pulverized limestone and clay to high temperatures of approximately 1450 °C in kilns. The thermal process causes a chemical transformation of limestone into calcium oxide, contributing to approximately 50% of the carbon dioxide emissions associated with cement production [5]. The emissions resulting from the heating process, which accounts for the remaining 50% of the cement production's emissions, are primarily caused by the combustion of coal or gas (**Figure 1**) [6][7].

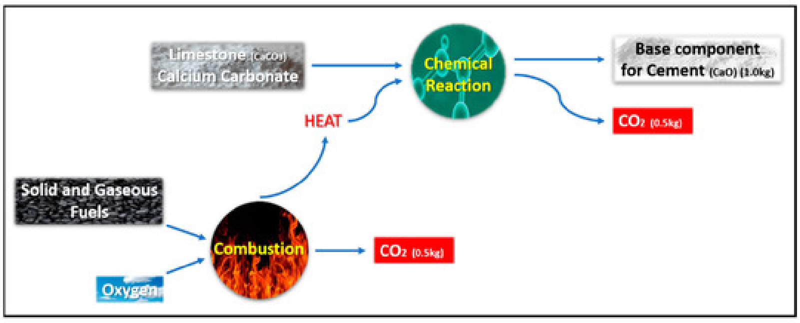

Figure 1. Simple recipe for cement and CO_2 emissions.

Due to the increase in the share of renewable energy sources in total energy production, there has been an increasing interest in biomass energy use. Burning biomass, mainly wood (bark, sawdust, leaves, wood chips, cellulose, sludge, etc.), is a way to achieve a higher percentage of coal-free energy sources. However, with more energy to produce, burning biomass causes more ash to accumulate in landfills as a waste product of the combustion process (sending ash to landfills adds to the cost of energy production). Therefore, finding new ways of recycling fly ash is an essential and timely issue. One of the most economical, efficient, and modern ways to eliminate accumulated fly ash is to process it with alkalinized materials known as geopolymer (inorganic polymers) composites [8][9][10][11][12]. Due to their long-term, low-cost, low-CO_2 emissions during production [13], extraordinary thermal and chemical resistance [14], and highly porous structure [15], geopolymers have gained rapid interest and experienced rapid growth over the last 20 years.

The energy and minerals industry produces valuable resources, including fly ash and other byproducts, with significant potential for various applications [16][17][18]. These byproducts have captured significant attention due to their sustainable use options across various sectors. One of the prominent and widely recognized applications of fly ash lies in the realm of cement production [19][20] and geopolymer concrete [9][21][22]. By replacing a portion of cement with fly ash, the resulting concrete exhibits enhanced workability, improved long-term strength, and reduced permeability. Geopolymer concrete not only offers a sustainable alternative to conventional cement materials but also provides an avenue for utilizing large quantities of fly ash that would otherwise be disposed of in landfills [23][24]. Several studies have been conducted to investigate the greenhouse gas emissions caused by concrete and cement, as well as the effect that the addition of fly ash has on this overall amount [6][25][26]. Comparisons between cement and geopolymer were first made in the literature, and the majority of those comparisons were based on the production stage of each material [27]. According to the findings of these studies, geopolymer manufacturing results in greenhouse gas emissions that are anywhere from five to six times lower than those of cement production [28]. This is accomplished by avoiding the significant direct emissions of CO_2 that are produced during cement production and cutting back on part of the energy used in processing [28][29][30].

2. Biomass Fly Ash-Based Geopolymers

2.1. Description of Geopolymer

Geopolymers are, according to the commonly accepted definition, inorganic, amorphous, synthetic aluminosilicate polymers formed from the synthesis of silicon and aluminum and geologically derived minerals. Their chemical composition is similar to that of zeolite but reveals an amorphous microstructure [31][32][33]. The base material used in this context can be either a natural raw material (kaolin, metakaolin, clay, volcanic tuff, laterite) or a waste material, such as fly ash, slag, or inorganic material with pozzolanic properties [34][35].

Geopolymer materials are mechanically durable with high compressive and flexural strength, elasticity, and chemical and fire resistance. They can exhibit compressive strengths higher or similar compared to Portland cement-based concrete [36]. Bakri et al. developed an experimental plan to assess the impact of different ratios of fly ash and aggregate on the compressive strength of concrete [37]. The study compared the use of fly ash-based geopolymer with ordinary Portland cement (OPC). This study utilized various ratios of FA 50%: aggregate (AGG) 50%, FA 40%: AGG 60%, FA 30%: AGG 70%, and FA 20%: AGG 80% in geopolymer concrete. The identical designs have also been employed as control references for OPC concrete. The strength of the material was assessed through compressive strength testing. The findings indicate that the geopolymer made with 30% fly ash and 70% aggregate exhibits superior compressive strength compared to ordinary Portland cement concrete after 1, 7, and 28 days of testing.

The geopolymer matrix appearance is unchanged at exposed temperatures of 1000–1200 °C [38]. Geopolymers are highly resistant to fire and do not emit harmful vapors or smoke. Geopolymers can also potentially be used for producing fire panels or as fire-resistant coatings on metals. The coatings can be designed to maintain temperatures below 550 °C [39]. The geopolymer material has low thermal conductivity, high mechanical strength, excellent resistance to alkaline and acidic environments due to the low calcium content in its chemical structure [40], and even allows the adsorption of toxic chemical wastes [21]. The ability to add different fillers (particles, fibers) [41][42][43] increases not only its performance parameters (strength, mechanical resistance, thermal conductivity) [44] but also its physical aspects [45][46][47][48][49].

2.2. Biomass Fly Ash in Geopolymer Composites

Current research emphasizes the utilization of geopolymer products derived from biowaste materials. These products exhibit superior durability, strength, and fire resistance compared to conventional building materials [50]. The advantages can be further improved by developing the ability to modify the composition of the geopolymer to achieve specific features [51][52]. **Table 1** summarizes recent research on biomass fly ash based geopolymer production and use.

Table 1. Process of making geopolymers from biomass waste and its areas of application in the latest literature.

Sources of Fly Ash	Geopolymer Preparation Method	Precursor	Application/Goal of Geopolymer	References
Paper waste	A mixture consisting of 15 g of aluminosilicate precursors, comprising 50 wt.% metakaolin and 50 wt.% FA, was subjected to mechanical mixing with 24.38 g of alkaline solution, 4.15 g of water, and 0.75 g of pore-forming agent in order to generate the geopolymer slurry.	Metakaolin	Wastewater treatment	[53]

Sources of Fly Ash	Geopolymer Preparation Method	Precursor	Application/Goal of Geopolymer	References
Paper waste	The SiO_2/Al_2O_3 ratio was 3.1, the Na_2O/Al_2O_3 ratio was 2.0, and the Na_2O/SiO_2 ratio was 0.6. To investigate the influence of the pore-former on porous geopolymer materials, different quantities of H_2O_2 were utilized. Sodium silicate was replaced in these compositions by 0.03, 0.15, 0.30, 0.90, and 1.2 wt.% H_2O_2.	Metakaolin	Board and wall panels	[54]
Co-generation plant (BA)	Here, 75 wt.% BA and 25 wt.% MK were employed in the formulation. The solids were combined for 1 min at 60 rpm in a Kenwood planetary mixer before adding the alkaline activators for 10 min at the same agitation. Stirring was maintained for another 5 min at 95 rpm with the addition of H_2O_2 as needed.	Metakaolin (MK)	Filtration and separation	[55]
Kraft pulp mill (BFA)	The manufacturing process of GP mortars involves several steps. First, MK and BFA were hand mixed for a duration of 1 min to achieve a consistent blend. Second, sodium hydroxide and silicate were homogenized at a speed of 60 rpm for 5 min. Next, the alkaline solution was mixed with the solid precursors (BFA + MK) in a Hobart-type mixer at a speed of 60 rpm for 9 min. Finally, lime slaker grits were added to the mixture and mixed for an additional 1 min at the same speed to ensure uniformity.	Metakaolin(MK)	Construction and masonry	[56]
Wood biomass (BA)	The alkaline activators were added while still being stirred for 10 min after the solids (BA and MK) had been combined for 1 min at 60 rpm in a Kenwood planetary mixer. The mixture was stirred for 5 more min at 95 rpm.	Metakaolin(MK)	Reducing cost of geopolymer	[57]
Mixed waste from Hauts-de-France (BFA)	NaOH (20 wt.% of the activation solution) and Na_2SiO_3 (80 wt.%) are the chemicals used to initiate the geopolymerization process. Na_2SiO_3 was added with the goal of raising the concentration of soluble silicates and the pace of the reaction. A magnetic agitator was used to combine the 2 reagents in a glass container for 6 h before resting the solution in a plastic bottle for 24 h. The alkaline solution was then combined for about 3 min in a mixer with metakaolin and SRS or BFA at a rotating speed of 300 rpm.	Metakaolin (MK) and shooting range soil (SRS)	Immobilization of heavy metal	[58]
Wood biomass (BWA)	Three replacement ratios of FA by BWA were used in the blended biomass wood fly ash–fly ash geopolymer mortars: 10%, 20%, and 30% of the total binder. The activator (Na_2SiO_3 NaOH)/binder and fine aggregate/binder mass ratios for the geopolymer mortars were fixed at 0.5 and 2.0, respectively.	Fly ash	Economic and environmental benefits	[59]
Mix of pine pruning, forest residues	The solid precursors were combined with the activating solution. The concentration of the sodium hydroxide solution was 8 M, and the ratio of sodium silicate to sodium hydroxide was 1.15, which represents the modulus of the activator. The activator was introduced into the precursors that had been previously combined for a duration of 2 min. Subsequently, the mixture was subjected to agitation for an approximate duration of 5 min using a Proeti planetary mixer.	Metakaolin	Building materials, bricks	[60]
Olive and forest pruning (FBA)	The geopolymers were prepared using five different compositions. These compositions included pure MK, as well as four other compositions referred to as GP1, GP2, GP3, and GP4. GP1 consisted of 50% MK, 25% AIS, and 25% FBA. GP2 consisted of 50% MK, 33% AIS, and 17% FBA. GP3 consisted of 40% MK, 35% AIS, and 25% FBA. GP4 consisted of 40% MK, 25% AIS, and 35% FBA.	Metakaolinaluminum industry slags (AIS)	Partial substitutes for metakaolin and Portland cement	[61]

Sources of Fly Ash	Geopolymer Preparation Method	Precursor	Application/Goal of Geopolymer	References
Burned eucalyptus biomass	**The geopolymer mortars were prepared according to a mix design that followed a binder-to-aggregate weight ratio of 1:3. The mixer was supplemented with alkaline activators according to the following procedure: (i) the sodium silicate and NaOH solution were initially homogenized at a rotational speed of 60 rpm for a duration of 5 min; (ii) the alkaline solution was then mixed with the solid materials at the same rotational speed for a period of 10 min; and (iii) the mixture underwent further homogenization and mixing at a rotational speed of 95 rpm for an additional 5 min.**	**Metakaolin andconstruction and demolition waste**	**Applications in building, replacing conventional mortars**	[62]
Wood biomass	**Geopolymers were synthesized by combining a mixture consisting of 2/3 wt.% metakaolin (MK) and 1/3 wt.% biomass FA, which served as an aluminosilicate source. In the present study, various compositions were examined by replacing sodium silicate with different weight percentages (0.03, 0.15, 0.30, 0.60, 0.90, and 1.2 wt.%) of hydrogen peroxide (H_2O_2). The blending of the mixtures was conducted using a mechanical procedure consisting of the following steps: (i) the sodium silicate and NaOH solution were homogenized at a rotational speed of 60 revolutions per min (rpm) for a duration of 5 min; (ii) the alkaline solution was then mixed with biomass FA and MK at the same rotational speed for a period of 10 min; and (iii) H_2O_2 was added to the blend in an amount determined by the formulation, followed by an additional mixing period of 2 min at a rotational speed of 95 rpm.**	**Metakaolin**	**pH regulators for biogas reactors or wastewater treatment**	[63]

The immobilization of biomass fly ash is essential for preparing safe concrete for further use [22][64]. Geopolymers play a significant role in environmental protection due to their capacity to immobilize heavy metals. This ability is closely linked to their ion exchange capabilities and extensive surface area development [65]. Metals like cadmium, copper, lead, chromium, zinc, and others can be immobilized within the geopolymer structure. Excessive amounts of zinc or chromium have been found to negatively impact compressive and bending strength [66]. A recent study proved the effective immobilization of toxic heavy metals (such as Cr, Mo, Pb, Sb, Se, and Zn) found in biomass fly ash. This was achieved through the utilization of the geopolymerization/accelerated carbonation technique. The study findings indicate that geopolymerization/carbonation stabilization processes effectively trap various elements, including As, Cr, Mo, Pb, Sb, Se, and Zn, across a broad pH range. The efficiency of this process is significantly influenced by the composition of the metakaolin [58].

Geopolymers can be used for prefabricated building elements, transport structures, and materials that achieve high adhesion to steel, aggregates, and many others [67][68]. Songpiriyakij et al. have demonstrated the bonding strength of geopolymers [69]. Twelve distinct mix proportions of geopolymers were created by adjusting the quantities of the initial binder materials and alkaline concentration. These mixtures were subsequently evaluated for their compressive and bonding strengths. The bonding strengths of the round bar and geopolymer were slightly greater than those of the control concrete, ranging from 1.05 to 1.12 times. The bonding strengths were significantly higher for deformed bars, ranging from 1.03 to 1.60 times. The study also included the presentation of the ratios between bonding strength and compressive strength. The bonding strengths of geopolymer were found to be 1.24–1.81 times higher than those of epoxies when compared to commercial repair materials. Furthermore, geopolymer concrete with the addition of biomass fly ash could be used as an additive for ceramics, chemically resistant exterior and interior cladding, chemically resistant items for industry, a composite item for working with hazardous substances (heavy metals, radioactive substances, etc.), and filler joints for reinforced concrete structures [70][71].

Challenges in utilizing biomass fly ash for geopolymers include the variation in particle sizes, morphology, composition, and reactivity among different fly ash samples [72][73]. The particle differences in biomass fly ash are highly influenced by the production conditions and the composition of the feedstock used in the boiler. It has been observed that the resulting mechanical strengths of geopolymers vary significantly even when using fly ash of apparently similar composition but from different sources, as well as different batches of fly ash from the same source [74][75].

Sharko et al. [76] conducted a recent and comprehensive study on the synthesis of geopolymers utilizing biomass fly ash. Six distinct fly ashes derived from six separate biomass thermal power plants in the Czech Republic were utilized in the study.

Subsequently, these geopolymers were subjected to a mechanical durability test to assess their performance. In addition, a comparison was made between the mechanical test outcomes of geopolymer concrete incorporating metakaolin and conventional concrete. The flexural and compressive strength, as well as impact toughness, exhibit significant variability as a result of variations in chemical composition among different types of biomass fly ash. The experiment demonstrated that the physical properties of geopolymer structures and their durability performances vary depending on the biomass fly ash obtained from different thermal power plants.

To ensure the production of a consistent geopolymer product from a raw material source with varying physicochemical properties, it is necessary to gain a comprehensive understanding of how different synthesis parameters impact the properties of the resulting geopolymer. This understanding will enable precise adjustment of these parameters for the specific product, thereby facilitating potential commercial applications in industries such as construction [77].

The primary determinant of fly ash's chemical composition is the presence of reactive silicon compounds [78][79]. Silicon creates the primary constituent of the internal structure of the geopolymerization products resulting from the alkalinization process of fly ash [80]. The fly ash's reactive silicates dissolve under highly alkaline conditions, resulting in the formation of Si-O-Al polymer bonds (**Figure 2**). Therefore, the presence of abundant reactive silicon compounds leads to the formation of significant quantities of aluminosilicate gel, which contributes to the potential for achieving high strength in the resulting geopolymer material [81][82][83].

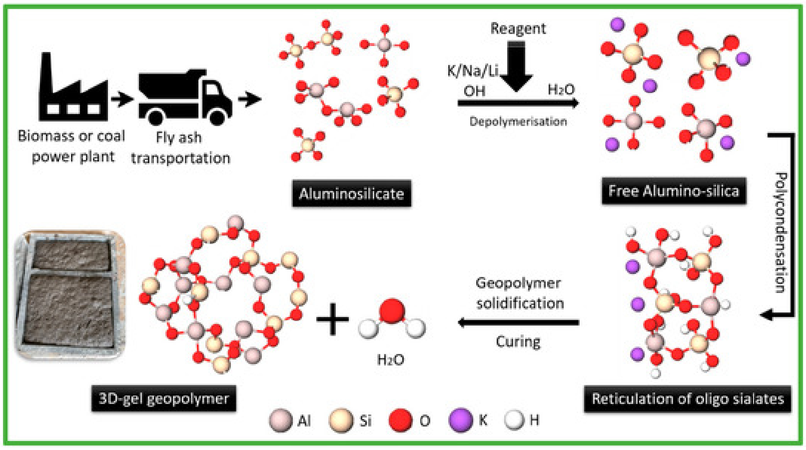

Figure 2. Chemical structure diagram.

The essential characteristics of fly ash that are deemed suitable for the production of geopolymer materials with commendable mechanical properties are as follows: The fly ash should contain a maximum of 5% unburnt material, 10% iron oxide, and 10% CaO [25]. The concentration of reactive silicon should fall within the range of 40 to 50%. The percentage of particles with a size smaller than 45 μm should fall within the range of 80 to 90% [74][84].

2.3. Environmental Impact of Biomass Fly Ash Recovery on Geopolymer Formation

A geopolymer is a replacement for cement that has a significantly lower energy requirement for production and emits a smaller amount of CO_2 greenhouse emissions compared to Portland cement [22][85]. Geopolymer technology offers the benefit of utilizing industrial byproducts, such as kaolin, feldspar, fly ash, slag, palm oil ash, and mining waste, as binders [86]. Geopolymers are a promising area of study in terms of their cost-effectiveness and environmentally friendly nature [87]. The production of geopolymers offers novel technical solutions that enable the utilization of up to 90% of ash, thereby enhancing waste utilization within the circular economy of the country. Furthermore, this procedure has the capability to produce a long-lasting and ecologically sustainable substance [88]. Another significant advantage of porous geopolymer materials is their ability to exhibit low thermal conductivity and high thermal resistance [89][90]. These materials are commonly used in the construction industry as insulation due to their optimal mechanical strength [91][92]. Geopolymers are a viable substitute material in situations where it is crucial to avoid the emission of harmful fumes during combustion due to their non-flammable nature [93][94].

Uses of geopolymers include not just thermal insulation but also pH buffering [95] and wastewater treatment [96]. The feasibility of utilizing biomass fly ash-based geopolymers as lead adsorbents was assessed by Novais et al. [97]. They examined the impact of heavy metal concentration, pH of aqueous solutions, adsorbent quantity, and contact time on the efficiency of lead removal with geopolymers. The results indicate that the novel materials have a lead uptake of up to 35 mg/g, highlighting their potential as effective lead adsorbents. Geopolymer materials have significant potential for enhancing water quality through wastewater treatment. A cost-effective geopolymer was synthesized using solid waste through a process involving acid treatment after geopolymerization [98]. This method effectively removes methylene blue (MB) dye from wastewater. The geopolymer adsorbent demonstrated excellent adsorption performance for a 600 mg/L solution of MB dye (pH = 8) at room temperature. It achieved a maximum adsorption capacity of 115 mg/g and a removal efficiency of 97.8%.

Geopolymer materials do not consistently exhibit promising characteristics. Another study demonstrated their adverse effects. One of the first studies conducted by G. Habert [99] aimed to examine the environmental evaluation of geopolymer-based concrete production through the utilization of the life cycle assessment (LCA) methodology. The researchers substantiated the favorable influence of geopolymer materials on the phenomenon of global warming. However, their investigation also revealed that the utilization of geopolymers is associated with the exacerbation of additional environmental concerns. As an illustration, the researchers documented that the human toxicity of geopolymer-based concrete was 105.4 kg of 1,4-DB eq. (dichlorobenzene equivalent) in contrast to 18.9 kg 1,4-DB eq. for conventional Portland concrete. The ecotoxicity towards freshwater organisms was found to be 2.52 kg 1,4-DB eq. for ordinary Portland concrete, while the corresponding value for geopolymer concrete was more than ten times higher at 27.01 kg 1,4-DB eq. The primary cause of geopolymers' greater toxicity levels in humans can be attributed to the presence of sodium silicate solution, which is necessary for the geopolymerization process of aluminosilicate source materials. The utilization of fly ash and granulated blast furnace slag geopolymer has been determined to possess a diminished environmental footprint due to its activation process involving small quantities of sodium silicate solution. In order to minimize the utilization of sodium silicate solution, it is imperative to take into account the mix design for geopolymer concrete with a focus on optimizing the Si:Al ratio [100].

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