

From Waste to Strength: A Comprehensive Review on Using Fly Ash in Composites with Enhanced Mechanical Properties

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ABSTRACT

This article explores the diverse applications of fly ash (FA), a by-product generated during the combustion of coal. The introductory segment thoroughly comprehends the origins, composition, and widespread occurrence of FA. FA, which comprises an estimated 38% of worldwide power generation, frequently encounters disposal and storage obstacles on account of its classification as non-hazardous waste in the majority of countries. The environmental issues linked to the dispersal of FA are underscored in the problem statement, which further emphasizes the urgency for sustainable alternatives. Due to the fugitive emissions and potential health hazards associated with metal melting in FA, it is critical to investigate novel applications and disposal techniques immediately. Environmental sustainability is a primary focus of research, with the development of synthetic FA composites being one such alternative. The analysis presents significant findings that underscore the wide-ranging applications of FA. These applications include its utilization as a filler in composites, as well as its incorporation into cement and geo-polymerization processes. Notably, (10-20) wt. % Nano-FA enhances epoxy-based composites, showcasing remarkable improvements in tensile strength, flexural strength, and impact resistance. In thermoplastic composites, substantial enhancements occur within the (5–10) wt. % FA range, but exceeding optimal ranges weakens matrix-fiber interaction, leading to diminishing returns. The article emphasizes the criticality of FA in improving the mechanical and thermodynamic characteristics of substances, specifically within the domain of composites. The investigation into FA nanoparticles, including their processing techniques and surface treatments, unveils encouraging prospects for enhancing material characteristics.

Keywords: Fly Ash, Composites, Mechanical Properties.



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1 Introduction

Fly ash (FA) is a residual substance that is generated by the combustion of coal for the purpose of energy generation. The substance in question is the ultimate byproduct resulting from the burning process of crushed bituminous or sub-bituminous coal, specifically lignite, within the combustion tanks of thermal power plants. FA is composed of the un-combusted mineral constituents of coal. Upon the introduction of pulverized coal into the combustion chamber of a boiler, a rapid ignition process occurs, resulting in the generation of molten mineral residue. Following the extraction of heat from the firebox, the molten residue undergoes a cooling process, resulting in its solidification and subsequent formation of ash.

The larger fraction of this residual material, referred to as bottom ash, settles at the lower part of the chamber, whereas the smaller particles of ash stay suspended within the flue gas [1]. Typical raw FA is depicted in

Fig. 1.



Fig. 1 Raw Fly ash [2].

Coals are responsible for generating approximately 38% of global power. FA is a waste product generated from the combustion of coal. It is the main component of coal ash and contains silica, aluminum, iron, calcium, and oxygen. FA also contains trace levels of heavy metals such as arsenic and lead. Coal combustion FA contains valuable elements, including rare earth elements (REEs). It is a readily available residue from thermal power plants [3]. FA has low density, good thermal resistance, and wide availability. FA as fillers is used to enhance the mechanical and thermal properties of composites. Most nations do not consider FA a hazardous waste, leading to a lack of regulations on its disposal and storage. FA that is not reused is stored in landfills and surface impoundments.

Fugitive emissions and the melting of metals from FA may pose health risks. Limited epidemiological studies have been conducted on the health effects of FA exposure [4]. Dumping FA is a serious challenge to the environment. Researchers are focusing on artificial FA aggregates as an alternative. FA mixed with cement can be used as a binder in concrete. Type-C FA is recommended as a partial replacement for cement [5]. Coal FA has been successfully used in Portland cement concrete (PCC) as a mineral admixture. Moreover, in recent times, it has been utilized as a component in the production of blended cement. FA is used as an admixture in Portland cement, either as a partial substitute or an additional component. The process involves the direct integration of the material into the ready-mix concrete at the batching plant. Blended cement can be created through two methods: inter-grinding FA with cement clinker or blending it with Portland cement [6]. ASTM C595 categorizes blended cement products into two types, which involve the inclusion of FA as an additive. Two categories of cement include pozzolan:

Portland-pozzolan cement (Type IP) with a pozzolan content ranging from 15 to 40 percent, and Pozzolan modified Portland cement (Type I-PM) with a pozzolan content below 15 percent [7]. The physical properties of FA are listed in Table 1.

Table 1 Physical Properties of FA [8]

Properties	Values
pH	7.20 - 7.80
Bulk density [gm.cm ⁻³]	0.98 - 1.32
Particle density	2.10 - 2.31
Porosity (%)	46.5 - 49.00
Electrical conductivity [ds.cm ⁻¹]	0.522 - 0.614
Water holding capacity	48.6 - 64.75
Specific surface area [cm ² .gm ⁻¹]	7117.67

Other names for FA include coal ash, pulverized flue ash, and pozzolana. FA exhibits several distinctive properties, including its low mass, glass-like silicate appearance, spherical morphology, grey hue, polymeric composition, alkaline nature, and refractory behavior. Furthermore, it should be noted that FA possesses a pozzolanic characteristic [9].

Table 2 ASTM C618 FA Specification [10].

Item (%)	Purpose of item	Class F	Class C
SiO ₂ + Al ₂ O ₃ + Fe ₂ O ₃	The sum of silica, alumina, and iron oxides controls quality and may limit other oxides.	>50	>50
CaO	Sulfate attack and alkali-silica reaction mitigation depends on CaO. Latent hydraulic phases in higher CaO fly ashes	≤18	≥18
SO ₃	Excess sulfate can cause issues with hydration, setting, and strength gain.	<5	<5
Moisture content	Many suppliers transfer fly ash into storage silos pneumatically, but high moisture content prevents this.	<3	<3
LOI	Controlling LOI prevents fly ashes from absorbing large amounts of air.	<6	<6
Fineness – amount retained on 45µm sieve	Fineness affects fly ash reactivity. Limiting very coarse particles reduces contamination.	<34	<34
SAI, 7 days, % of control	SAI indirectly indicates fly ash reactivity and distinguishes it from unreactive materials.	>75	>75
SAI, 28 days or 56 days, % of control		>75	>75
Water requirement, % of control	Water requirements reveal fly ash's general workability.	<105	<105
Soundness – autoclave expansion/contraction	To prevent fly ashes from expanding due to MgO content, soundness is controlled.	<0.8	<0.8

The objective of this article is to provide a comprehensive overview of recent research conducted on the utilization of FA in diverse composite material sectors. In this analysis, the various applications of the technology in the domains of material science, construction, and environmental remediation are examined. Furthermore, emphasis is placed on the amalgamation of FA with various substances, such as nano-fillers, to enhance the quality of the resulting goods and their mechanical characteristics. The development of materials that are both desirable in property and environmentally sustainable is frequently facilitated through collaboration. In this discourse, an overview of the several applications of FA and its synergistic use with other materials to enhance their performance is intended to be presented.

2 Classification

ASTM C618 establishes two categories of FA that are suitable for utilization in concrete applications. There are two main classes of coal ash: Class F, which is typically produced from burning anthracite or bituminous coal, and Class C, which is typically produced from burning lignite or sub-bituminous coal. ASTM C618 also specifies the physical, chemical, and mechanical properties required for these two categories of FA, and are mentioned in Table 2.

Class F FA is a type of ash that exhibits pozzolanic properties and has limited or no ability to act as a cementing agent on its own. Class C FA possesses both self-cementing properties and pozzolanic properties. The difference in pozzolanic reactions between these types of FA is pointed out in Table 3.

FA undergoes pozzolanic reactions over extended timeframes, typically spanning months to years. The process involves the transport of calcium hydroxide through water within the soil, interacting with aluminate and/or silicate clay minerals [11]. The high surface area of these minerals, acting as pozzolan phases, reacts with water and alkali, yielding cementitious materials like calcium silicates and aluminate hydrates. Dissolved Ca²⁺ ions further react with SiO₂ and Al₂O₃ on clay particles, forming hydrated gels (C-S-H and C-A-H), enhancing soil cohesion and stiffness [12] [13]. This consumption of water content results in a more robust, less prone to shrinkage/swelling soil matrix, commonly referred to as a geo-polymer.

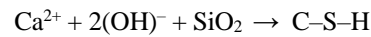


Table 3 Difference in pozzolanic reactions of class F and C FA:

Properties	Class F	Class C
Chemical Composition	Low CaO content (<18%) allows for a higher proportion of pozzolanic phases (SiO ₂ and Al ₂ O ₃) that react with Ca(OH) ₂ to form CSH gel.	High CaO content (≥18%) contributes to self-cementing properties but not directly to pozzolanic reactivity.
Pozzolanic Reaction	Pozzolanic: Alumino-silicates react with Ca(OH) ₂ released during cement hydration to form additional CSH gel, contributing to long-term strength development.	Self-cementing: Free lime readily reacts with water to form calcium hydroxide (Ca(OH) ₂), which then reacts with pozzolanic phases to form calcium silicate hydrates (CSH) gel and contribute to early strength development.
Reaction Rates	Shows slower pozzolanic reactivity, leading to slower early strength development but potentially higher long-term strength due to a more complete formation of CSH gel.	Exhibits faster pozzolanic reactivity due to the presence of free lime, leading to rapid early strength development.
Effects on Concrete	Slightly increases early strength but to a lesser extent than Class C FA.	Increases early strength development due to the self-cementing reaction.

When class F and C fly ashes are added to clay, the liquid limit (LL) and plasticity index (PI) go down. The LL and PI trends stay the same for 25% FA and then get stronger as more FA is added. Class C FA has a bigger effect on the Atterberg limits (a basic measure of the critical water contents of fine-grained soil [14]) than class F does because it has more lime in it. When FA is added, the highest dry density of the mixture goes down because FA is less dense than soil particles. By adding FA, the ideal moisture level goes up as well [15].

When there is 25% FA, the unconfined compression strength is at its highest. Adding FA until the right amount may cause pozzolanic and cementation interactions that make the unconfined compressive strength go up. The curing period makes the results better. For example, in a 25% FA class F case, the unconfined compressive strengths were 1.7 times higher after seven days of curing and 2.5 times higher after 28 days of curing than they were after one day of curing. The specimen stabilized with class C had a higher unconfined compression strength than the one stabilized with class F. This is because class C has a high level of self-cementation. Research from the past has shown that materials that don't stick well can be mixed with a small amount of cement and lime or cement kiln dust. Because of this, examples stabilized with class F FA and a small amount of Portland cement, lime, or cement kiln dust are likely to show the same results [16]-[18].

3 FA Particle Processing

3.1 Ball Milling

The main constituents of FA nanoparticles include silicon dioxide, magnesium oxide, sulfur trioxide, calcium oxide, aluminum oxide, and ferric oxide [19]. FA nanoparticles have surface polarity due to the presence of OH groups. FA nanoparticles can be used as reinforcement in polymer composites [20]. Converting FA into nano-scale particles typically involves advanced milling techniques like high-energy ball milling [21] and attrition milling. These methods apply mechanical forces to reduce particle size, necessitating specialized equipment and precise control to achieve the desired dimensions [22]. FA nanoparticles have particle sizes of less than 45 microns. They consist of glassy spheres with some crystalline matter and unburnt carbon [23]. Attrition milling reduces particle size from 29 μm to 6 μm . Vibration milling reduces particle size to 7 μm . Density increases from 2.44 g/cm^3 to 2.84 g/cm^3 (attrition milling) and 2.79 g/cm^3 (vibration milling). The compressive strength of the binder using attrition-milled FA is 61 MPa. The compressive strength of binder using vibration-milled FA is 49 MPa [24]. The surface and bulk properties of milled FA were enhanced. The specific surface area of milled FA increased during grinding. The crystallite size of quartz in FA decreased during grinding [25].

FA nanoparticles have diameters ranging from 0.1 μm to >100 μm . FA nanoparticles contain quartz as the major crystalline element. The size of banana FA nanoparticles is approximately 28.23 μm . The size of pineapple FA nanoparticles is approximately 19.73 μm . The size of coir FA nanoparticles is approximately 37.14 μm [26]. They have low density, wide availability, good filler factor, good thermal resistance, and glassy nature. FA particle size depends on factors such as the combustion process, collection methods, coal type, and mechanical processing. FA nanoparticles are composed of silica, aluminum, iron, calcium, and oxygen. FA nanoparticles may contain trace levels of heavy metals such as arsenic and lead [27]. They have a spherical shape, facilitating easy blending and flow. FA nanoparticles exhibit both amorphous and crystalline mineral characteristics. They have advantageous properties such as capillarity and pozzolanic activity. The properties of FA nanoparticles can vary depending on their source [28]. FA nanoparticles can have different levels of fineness and spherical particle shape. Some FA nanoparticles have high CaO content and high free lime content [29]. FA nanoparticles contribute to a more compact and homogeneous structure. FA particles have a denser structure and stronger bonding. They contribute to the

hardness and resistance of the composite material. The smaller particles increase the density and hardness of the material. The distribution of FA particles affects the porosity of the composite [30].

3.2 Surface Treatment

Alumina $\text{Al}_2(\text{SO}_4)_3$ treatment, combined with NaOH, has led to substantial improvements in tensile and compressive strength in bamboo fiber composites. This chemical treatment removes impurities from FA and enhances interfacial bonding with natural fibers [27], [28], [31]. The NaOH concentration also plays a significant role in the strength development of the geo-polymer. A higher NaOH concentration increases the reactivity of the FA, resulting in a faster and more complete polymerization reaction, which in turn leads to higher strength properties [32]. However, an excessive NaOH concentration can lead to an over-alkalization of the mix, resulting in a decrease in strength [33]. Chemical solutions (NaOH, HF, and H_2SO_4) enhance fly ash surface conditions, resulting in increased interfacial composites. Increased tensile bonding with unsaturated polyester matrix increases fly ash/UP tensile strength by 91%, 82%, and 55% for NaOH, HF, and H_2SO_4 at 30% fly ash content (weight) [34].

The amount of Si and Al released from FA during geo-polymerization is highly dependent on the alkali activator used and its concentration. There is an optimum concentration range for NaOH solutions generally between 10-15 M, although using higher molarity solutions leads to more dissolving and higher compressive strengths in the finished geo-polymer. By encouraging the development of geo-polymer gel, increasing the sodium silicate to NaOH ratio also increases compressive strength [35].

4 FA in Polymer Composite

Composites are a combination of two or more materials with unique properties that together provide better performance and functionality than individual components [36]. Thermoplastic and thermosetting polymers are two types of materials widely used in various industries due to their excellent mechanical, thermal, and chemical properties [37]. Thermoplastic composites are formed by combining a thermoplastic matrix with a reinforcing material, such as carbon or glass fibers, and can be melted and reformed multiple times without undergoing any significant chemical change [38]. Thermosetting composites, on the other hand, are created by combining a thermosetting matrix with a reinforcing material, such as carbon or glass fibers, and cannot be melted or reformed once cured [39]. The utilization of FA as a filler substance in thermoplastic along with thermosetting composites has received considerable interest in recent times owing to its cost-effectiveness, ample accessibility, and prospective environmental advantages [40]. The presence of FA particles plays a significant role in influencing crack initiation and impact strength. However, manually mixing FA with epoxy resin can pose challenges due to its nature. To overcome this, surface modification techniques are employed for reinforcing FA in bio-composites. These techniques encompass the sol-gel process, in-situ polymerization, solution mixing process, melt mixing process, and in-situ intercalative polymerization [20].

The addition of polyamide (PA) to the binding agent SEBS (styrene-ethylene-butylene-styrene) loaded with maleic anhydride (SEBS-g-MA) resulted in an observed increase in the intensity of the distinctive bands associated with N-H and C=O. The explanation for the interfacial adhesion between the

components involves the potential reactions between the maleic anhydride groups of S-g-M and the amine-containing end groups of PA, as well as between the maleic anhydride groups and the hydroxyl molecules of FA [41]. Surface modification can greatly enhance the composites' mechanical characteristics. The surface tension of FA is drastically decreased if a simple alkali such as NaOH, KOH, Ca(OH)₂ treatment [42] is conducted before mechanical activation. The tensile characteristics of rigid PVC composites with FA particles are significantly impacted. A 10 wt. % FA addition increases Young's modulus by 30%. The

50µm FA's high hardness may explain the stiffer composites of bigger particles. Due to the consistency of particle size and the dispersion and homogeneity of the polymer matrix, 25µm and 50µm FA composites show higher modulus. PVC/FA composite containing FA particles with a diameter of 25µm exhibits the best tensile strength among all other composites. This observation implies that there is a significant transfer of load from the matrix polymer to the FA particles [43]. A summary of the discussion is mentioned in Table 4 .

Table 4 Enhancement of Mechanical Properties in Polymer-Based Composites with FA Fillers and Coupling Agents

Matrix material	Coupling Agent	Wt. %	Highlighted Findings	Ref.
polyamide 6/acrylonitrile-butadiene-styrene (PA/ABS)	----	9	Tensile strength - 11.9% ↑ Young's modulus - 35.5% ↑	[41]
Polyvinyl chloride (PVC)	----	5	Tensile strength - 12.67% ↑ Impact strength - 86.8% ↑ Flexural strength - 21.25% ↑	[44]
(Waste + High density) Polypropylene	----	5	Tensile strength - 14.67% ↓ Flexural strength - 6.40% ↑ Impact strength - 7.16% ↓	[45]
Lapox L12 epoxy Resin	Hardener K6	20	Tensile strength 42% ↑ Compressive strength 27.27% ↑ Tensile modulus 20% ↓	[46]
Lapox L-12 epoxy resin	Hardener K6	10	Tensile strength 19% ↑ Compressive strength 22.5% ↑ Hardness 29% ↑	[47]
Bisphenol A diglycidyl ether epoxy resin	Amine Containing Silicone (ACS)	5	Tensile strength 9.3% ↑ Tensile modulus 30.77% ↓	[48]
Bisphenol-A epoxy resin	Isophorone diamine (IPD)	4	Tensile strength 7% ↑ Compressive strength 24.34% ↑ Flexural strength 21.35% ↑	[49]
Bisphenol A diglycidyl ether epoxy resin	----	10	Tensile strength 93.48% ↑ Flexural strength 81.51% ↑ Impact strength 71% ↑	[50]
Bisphenol A diglycidyl ether epoxy resin	Triethylene-tetra-amine	30	Tensile strength 12.5% ↑ Compressive strength 42% ↑ Tensile strength 25% ↑ Compressive strength 51% ↑	[51]
Epoxy resin	----	20	Tensile strength 77.5% ↑ Impact strength 43.75% ↑	[52]
Epoxy resin	----	10	Tensile strength 22.53% ↓ Flexural strength 20% ↓	[53]
Epoxy resin	Ethylene-di-amine	5	Flexural strength 32.5% ↓ Compressive strength 7% ↑ Elasticity modulus 24% ↑	[54]

The superior performance of Nano FA in the composite can be attributed to its nano-sized particles. Nanoparticles disperse more uniformly within the matrix due to their reduced size, minimizing the chances of agglomeration [55]. Moreover, their increased surface area facilitates stronger bonding with the matrix material, leading to improved tensile strength and modulus. In epoxy composites, FA nanoparticles enhance the compressive strength. Nanoparticles of FA enhance the impact resistance of epoxy composites. FA nanoparticles enhance the thermal stability of composite materials [56]. The thermal behavior of recycled polyethylene terephthalate (rPET) improved with the addition of FA. Microstructural development

showed even filler distribution within the polymer matrix. However, increased FA loading led to voids and agglomeration, affecting thermal behavior [57].

The addition of FA increased the flexural strength. The inclusion of FA increased the impact strength. The FA in the bio-composites showed a moderate level of porosity. In the bio-composites, FA strengthened the adhesive bonding surface. Bio-composites were created with the use of a basic compression molding method. The epoxy matrix was strengthened with FA at weight percentages of 5, 10, and 15%. A blend consisting of 25% Bimal fiber and 75% epoxy matrix was determined to be ideal. The filler material was heated to 110°C in an oven throughout

the production process. With filler added, tensile strength of 23.78–33.79 MPa was measured. Elevated impact and flexural characteristics by 21.77% and 22.11%, respectively. Fillers increase mechanical qualities and enhance the adhesion between the fiber and matrix. The SEM data demonstrate the filler powders' good bonding nature. Silica and other inorganic materials are present in polymer composites. The composite's tensile and flexural moduli are improved by the FA component [56]. CaCO₃ nanoparticles have a high surface area, strong interfacial adhesion, and great dispersion [58] within the matrix, which is why they increase their content up to 5 wt. % enhances tensile strength. Finding the appropriate nanoparticle loading for a given composite application is crucial since, beyond this concentration, undesirable effects or declining returns may be detected [52]. The tensile strength of micro-filled composites has a negative correlation with the FA filler content ratios, as the insufficient adhesion between the matrix and fillers results in a reduction in the strength of the epoxy composite [53] and the decrease in elasticity at break values seen in the composite as the filler content increases may be explained to the dependence of the composite's elastic characteristics on the polymer matrix. The presence of fillers causes the polymer matrix to show brittle behavior, leading to a reduction in elongation at break values [54]. Nano-FA is also being used to reinforce epoxy composite specimens with Lapox L12 acting as the matrix. FA's hollow particle structure and high interfacial energy between the matrix and the nano-filler serve as advantages in this reinforcement strategy, leading to a linear increase in tensile and compression strengths as the amount of FA is increased. At the concentration

5 FA in Natural Fiber-Based Composite

The incorporation of FA into natural composite materials has the potential to improve their inherent characteristics. The incorporation of FA into natural materials such as clay [66], abaca [67], jute [68], [69], sisal [70], coconut [71], bamboo [72], banana [73], pineapple [74], coir fibers [75], or rice husk [76] results in the development of composites that exhibit enhanced mechanical strength, durability, and thermal resistance. Its applications include reinforcement in thermoplastics, thermosetting polymers, and elastomers. In rubber formulations, FA acts as a reinforcing agent, reinforcing the polymer matrix and improving tensile strength, modulus, and abrasion resistance. FA can also be used as an inexpensive and highly effective adsorbent for the removal of environmental pollutants because of its broad specific surface area, high porosity, and other distinctive properties [72]. It also aids in reducing heat build-up during mechanical deformation, thereby extending the material's lifespan [77]. These composite materials have shown to be highly advantageous in construction due to their ability to minimize the reliance on conventional building materials such as cement and steel, which are known for their high energy consumption. FA is utilized as a reinforcing filler in natural composites. The fine particles effectively occupy the interstices among the natural fibers, augmenting the material's overall structural stability. This application utilizes a by-product and fosters sustainability within the building sector [78].

Natural fibers are eco-friendly alternatives to synthetic fibers in various industries. Natural plant-based fibers are studied as alternatives to man-made fibers in composites. Natural fibers have advantages like environmental friendliness and cost-effectiveness. The hydrophilic nature of natural fibers limits their compatibility with hydrophobic matrices [79]. Pineapple and sisal fibers were used with epoxy as a matrix. FA from coconut,

of 20% FA, the tensile strength is observed to be at its maximum. However, it is important to note that concentrations beyond this threshold have been shown to complicate the curing process [46]. The water absorption behavior of various composites is influenced by the immersion time and the amount of FA content. The decrease in water absorption is attributed to the hydrophobic nature of FA [59]. The higher reinforcement percentage results in lower water absorption, while the composite with a constant filler percentage shows lower water absorption [60]. FA improves the water stability of composites. Hybrid interfaces formed by FA greatly enhance composite properties. Optimal water stability and tensile strength are achieved at specific conditions [61]. The addition of lignite FA (LFA) nano-filler results in increased values for tensile strength, flexural strength, and impact resistance. Moreover, the incorporation of LFA nano-filler leads to a significant reduction in moisture absorption, with a noteworthy decrease of 55% [62]. FA is being used as an inorganic filler in plastic products. FA alters the properties of composites and improves their performance. FA has high dielectric and thermal properties. FA composites show improved mechanical, thermal, and electrical properties [63], [64]. The addition of 5% Cashew Nut Shell Liquid and 15% FA in the Vinyl Ester polymer matrix enhanced the ultimate tensile strength of the composite to 37 MPa, which brought the Young's modulus up to 3.84 GPa. The yield strength increased to 10 MPa. However, the addition of Cashew Nut Shell Liquid caused these parameters to decrease, while the addition of FA caused them to take on higher values [65].

pineapple, and banana plants was used as fillers. Incorporating fillers improved the mechanical properties of the composites [26]. Natural fiber-based composites have biodegradable, low-cost, and lower-density properties. Properties depend on fiber treatment, matrix combination, and fabrication process. Natural fibers like hemp, jute, sisal, flax, rice husk, etc. are used. Chemical treatments improve adhesion between fibers and polymers [80].

5.1 Surface Treatment of Fibers

The alkali pre-treatment eliminates lignin, pectin, and hemicellulose and roughens the surface of the fiber. This enhances geo-polymer matrix-fiber bonding. Geo-polymer prevents the thermal breakdown of treated fiber [81]. When hemp fibers are treated with alkali, the porosity of the strengthened mortar seems to go down even after the wet/dry cycles. Compared to fiber-reinforced mortar that hasn't been treated, plain mortar absorbs 19% less water. Since the alkali treatment of the fibers lowers the ability of the fibers to absorb water and improves the pore structure of mortars, it also decreases the ability of the fiber-reinforced mortar to absorb water by an average of 6% for all sodium hydroxide concentrations. The alkali process gets rid of impurities on the surface of the fibers and causes the fibers to bond together [82].

Alkali treatment serves to enhance the surface topology of fibers and promotes better interlocking with the matrix. Mercerization is employed to eliminate impurities and heighten fiber density. Sodium hydroxide (NaOH) is a frequently utilized substance for alkali treatment in the context of natural fibers. This treatment contributes to heightened fiber strength and improved overall properties. Additionally, alkali treatment induces fibrillation and increases the aspect ratio of fibers, further optimizing their characteristics [83]. The tensile strength of

composites made from Kenaf fibers experiences improvement through alkali treatment. Furthermore, the incorporation of FA fillers contributes to enhanced mechanical properties in these composites. Specifically, composites with 10 wt. % FA reinforcement exhibits higher tensile strength. However, beyond this threshold, a slight decrease in tensile strength is observed. It is noteworthy that alkali treatment plays a crucial role in enhancing interfacial adhesion and bonding strength in these composites [84].

The treatment process involved the application of linseed oil and turpentine oil to the coir fibers. Notably, the treated fibers demonstrated a substantial enhancement in the mechanical properties of the soil. The results revealed that the treated fibers exhibited improved peak stress and displayed enhanced post-peak behavior. This suggests that the combination of FA and treated coir fibers has a positive impact on reinforcing soil [85]. Brief bamboo fibers underwent surface treatment with a 10% aluminum sulfate solution, resulting in a surface with increased roughness compared to untreated fibers. This enhanced rough surface is crucial for promoting effective bonding between the fibers and the matrix. The introduction of these treated bamboo fibers led to notable improvements in both compressive and split tensile strengths. Optimal performance was achieved with a bamboo fiber loading of 1.4% and a length of 20 mm. The addition of bamboo fibers contributed to a remarkable enhancement in compressive strength, demonstrating an increase of up to 292.41%. Similarly, the split tensile strength experienced a significant improvement, reaching up to 355.82% compared to control samples, underscoring the effectiveness of the specific surface treatment employed [86].

The application of sodium hydroxide treatment to hemp fibers has a positive impact on the properties of mortars. This treatment results in reduced porosity and decreased water absorption capacity in the mortars. Additionally, the compressive and flexural strengths of the mortars exhibit improvement following the treatment. Moreover, there is an increase in the energy absorption capacity of the mortars. Surface treatment of the fibers plays a crucial role in enhancing their distribution within the matrix, further contributing to the overall improvement in the performance and characteristics of the mortars [87].

5.2 Performance of Fiber-Reinforced FA Composites

Nano-FA added to laminated composites of epoxy, basalt, jute, banana, and carbon significantly improves their mechanical properties. The Nano-FA and BJBC-enforced composite has much greater tensile strength than the epoxy + BJBC counterpart. The inclusion of Nano-FA also improves performance concerning flexural strength. Epoxy + BJBC + 4 wt. % Nano-FA composite also shows better results in terms of tensile modulus than epoxy + BJBC composite [88]. The composites reinforced with FA demonstrated a tensile strength of 40 MPa, while those without FA reinforcement exhibited a tensile strength of 23.90 MPa. These findings endorse the efficient use of natural composites in automotive applications. The fabrication involved a hand-layup process, where Kenaf fiber mats were placed over a wooden mold. Subsequently, a mixture of epoxy resin and FA was poured onto the reinforcement. Four layers of woven Kenaf fiber mats were employed in the process. After applying a nominal load for 2 hours, the composites were cured at room temperature for 24 hours [84]. Key findings of such research are listed in Table 5.

Table 5 Natural fiber composite studies with FA as filler

Matrix material	Fillers	Wt. % / Vol %	Reinforced Fiber	Findings	Reference
Epoxy resin	FA	4 wt.%	Basalt (B), Jute (J), Banana (B), and Carbon (C) layers	Tensile strength 107% ↑ Tensile modulus 25% ↑ Flexure strength 141% ↑ Flexural modulus 227% ↑	[88]
Epoxy resin	FA 20nm	2 wt.%	Hemp	Tensile strength 38.33% ↑ Tensile modulus 17% ↑ Flexure strength 70.5% ↑ Impact strength 51.25% ↑ Water absorption 16.67% ↑	[89]
Epoxy Resin	FA	5 wt.%	Rice husk 10 wt.%	Impact strength 8.91 J/m Tensile strength 35 MPa Water absorption 5%	[90]
Epoxy Resin	FA	10 wt.%	Jute layer (JJJJ) 20 wt.%	Tensile strength 34.61% ↑ Flexural strength 25% ↑ Flexural modulus 20% ↑	[91]
Epoxy resin	FA	10 wt.%	Kenaf fabrics 20 wt.%	Tensile strength 30% ↑	[92]
Epoxy resin	Banana FA	3 wt.%	30 wt.% of Sisal (S)/Pineapple (P) hybrid fiber	Tensile strength 29 MPa Flexural strength 78 MPa Impact strength 61 J/m	[93]
	Coir FA			Tensile strength 33MPa Flexural strength 70MPa Impact strength 60 J/m	
	Pineapple FA			Tensile strength 28 MPa Flexural strength 71 MPa Impact strength 64 J/m	

Incorporating sawdust enhances flexural strength in the FA-based matrix. Additionally, plant waste materials such as corn

husk, rice husk, and coffee grounds offer potential for reinforcement in the matrix. Moreover, there are untapped

possibilities with unexplored natural fibers like henequen, ramie, sunn, and Kenaf for further reinforcement applications [94].

Sisal fiber has poor thermal conductivity, lowering composite thermal conductivity. This can help thermal insulation applications [95]. FA composites can improve tensile and flexural strength with sisal fiber. The addition of FA in sisal fiber has increased the composite material's tensile strength from 33.02MPa to 35.02MPa [96]. Because of their high strength-to-weight ratio, sisal fiber can reinforce the composite. Sustainable composites can be used because they are non-toxic and eco-friendly. The rough surface of sisal fiber can link the polymer matrix. Tensile strength in Kenaf fiber-reinforced composites was increased by 29% after alkali treatment as a result of increased fiber-matrix adhesion. Including small particles of FA (5%-15 wt. %) enhanced the tensile strength, with the maximum being reached at 10%.

Fig. 2 shows that FA particles were added to the composite, which enhanced the tensile characteristics to some extent by replacing voids with filler FA. When the filler content was

increased beyond 10%, the matrix-fiber bond was weakened, and a minor drop occurred.

Coir FA (CFA) composite demonstrated the highest tensile strength at 33 MPa, while Banana FA (BFA) composite exhibited the highest flexural strength at 78 MPa among the various FAs used in natural composites. The tensile strength of the Pineapple FA (PFA) composite was 28 MPa, and its flexural strength was 71 MPa. CFA and the BFA composite each had an impact strength of 60 J/m, while the PFA composite was slightly higher at 64 J/m. These differences illustrate the unique mechanical properties conferred by various FA in natural composites [93].

Combining 15 wt. % FA with 15 wt. % Bhimal-fiber bio-composite yielded the highest flexural strength at 159.96 MPa. For impact strength, the optimum was achieved with 10 wt. % FA and 15 wt. % Bhimal fiber, reaching 94.25 Jm. The resulting bio-composite exhibited a moderate porosity level ranging from 16.07% to 19.1% [56].

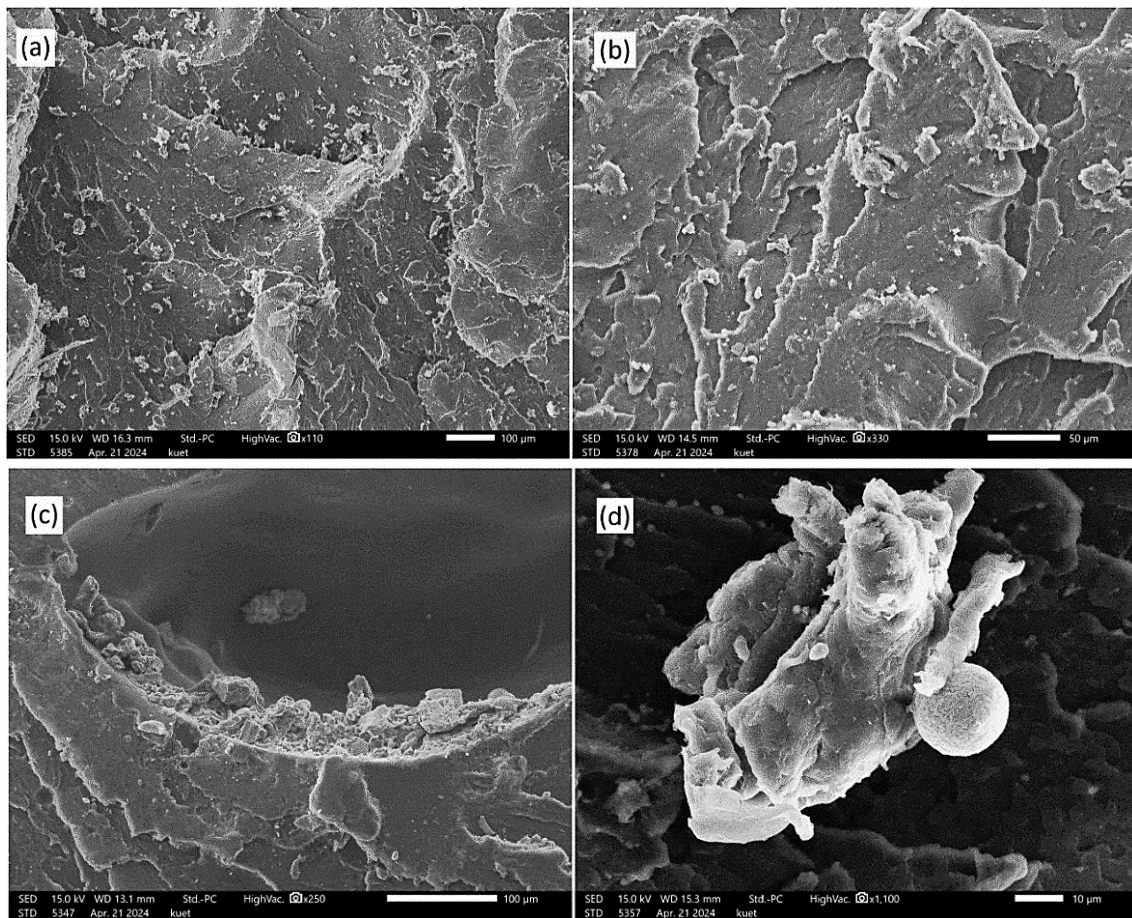


Fig. 2 SEM image of (a) RAW, (b) Water treated-FA, (c) Alkali treated-FA, (d) Acid treated-FA incorporated composites [92].

6 FA in Geopolymer

Adding natural fibers and applying pre-compression to lime-FA-stabilized soil can improve the soil's compressive strength, ductility, and stiffness. Pre-compression reduces the strength of the composite by redistributing the bond strength and causing matrix cracking and fiber-matrix interfacial de-bonding. Pre-compression can improve the strength of the composite at the low fiber content. When the amount of fiber is more than 0.25%, the compression strength of uncompressed specimens is higher than that of pre-compressed specimens. For pre-compression

stress levels, the loss of strength is more noticeable at 0.75% fiber content [97]. Sisal and coconut fibers significantly boost tensile and flexural strengths in FA geo-polymer mortars. While their inclusion improves strength, higher fiber content leads to decreased workability in the mortars [98].

Table 6 contains the highlighted outcome of the incorporation of FA as reinforcement in geo-polymer. The incorporation of 0.10% graphene oxide (GO) and 2% polyvinyl alcohol (PVA) fiber significantly bolstered the 28-day

compressive and flexural strengths of the HVFA system by 244.1% and 46.4%, respectively. These improvements stem from enhanced stress transfer through a robust interfacial bond

between PVA fibers and the GO-reinforced matrix, alongside increased damping sources for superior energy dissipation and vibration reduction [99].

Table 6 Impact of FA in Natural Fiber Reinforced Geo-polymer Composites

Matrix material	Chemical treatment	Fiber/Filler	Wt.%	Finding	Ref.
FA	16 M NaOH	Pineapple leaf	0.50	Compressive strength 272% ↑ Flexural strength 25% ↑	[33]
FA	NaOH +Ultrasonic treatment	Flax tow	1	Flexural strength 35% ↑	[32]
FA	5% NaOH + 10% Al ₂ (SO ₄) ₃	Bamboo fiber	1	Tensile strength 355.82% ↑ Compressive strength 292.4% ↑	[31]
Portland cement + FA	----	Polyvinyl alcohol fiber (PVA) + graphene oxide (GO)	2	Compressive strength 46.4% ↑ Flexural strength 244% ↑	[99]
FA	Na ₂ SiO ₃ + NaOH 12M	Polypropylene	1.2 vol. %	Compressive strength 1.15% ↓ Flexural strength 14% ↑	[100]
		Polyvinyl alcohol		Compressive strength 4.3% ↑ Flexural strength 40% ↑	
FA	----	Polyvinyl alcohol fiber	1.2	Compressive strength 50.5% ↑ Flexural strength 66.3% ↑	[101]
Sulfoalminate cement 80 wt.%+ FA 20 wt.%	----	Polyvinyl alcohol fiber	0.50	Compressive strength 8% ↑ Flexural strength 32.5% ↑ Bending strength 102% ↑	[102]
			1.5	Compressive strength 2.27% ↑ Flexural strength 56.25% ↑ Bending strength 125% ↑	
FA + River sand	Na ₂ SiO ₃ + NaOH 10M	Sisal fiber	1.0 vol.%	Flexural strength 106.25% ↑ Tensile strength 73.7% ↑ Compressive strength 9.83% ↑	[103]
		Coir fiber		Flexural strength 110% ↑ Tensile strength 16% ↑ Compressive strength 18% ↓	

Wood geo-polymer composites were analyzed at varying wood/FA ratios to examine geo-polymer formation through XRD and FTIR. The incorporation of wood particles significantly impacts the properties of these composites. The water absorption and thickness swelling characteristics meet standard specifications. Notably, the composite with a 23% wood particle ratio and 77% FA exhibits the highest mechanical properties. Despite these advancements, further enhancements are necessary to attain mechanical properties comparable to cement-bonded particle boards [104].

7 Mechanical Property Enhancement Factors

7.1 FA Weight Percentage

The incorporation of 4 wt.% FA into epoxy composites, which were reinforced with various fibers, including basalt, jute, banana, and carbon fiber, yielded significant improvements in both tensile and flexural properties. Specifically, the addition of FA resulted in an impressive 107% increase in tensile strength, showcasing the material's enhanced ability to withstand tension. Moreover, this positive influence extended to flexural properties, where the same study revealed a substantial 141% increase in flexural strength, indicating improved resistance to bending forces, and an even more impressive 227% increase in flexural modulus, reflecting enhanced stiffness and load-bearing

capacity. These findings underscore the beneficial impact of FA weight percentage on the overall mechanical performance of composite materials, making it a promising avenue for enhancing their structural integrity and utility in various applications.

However, the influence of FA content on mechanical properties can be complex. Some studies have found that increasing FA content in aluminum composites can lead to increased brittleness and porosity in casted samples, which can affect the overall material properties negatively [105].

Another study underscores the time-dependent influence of FA on the strength of mortars. While early-age strength may be reduced with FA additions, extended curing periods demonstrate significant strength improvements, particularly up to a 15% FA replacement rate. 10% FA was determined to be the optimal amount for maximal strength. This finding provides valuable insights for optimizing FA content in mortar mixes to achieve enhanced long-term strength performance. [106].

This significant enhancement can be attributed to several factors:

Reinforcement Effect: The presence of SiO₂ particles in FA, typically ranging from 40% to 50% in FA, plays a crucial role in enhancing the mechanical properties of composites. These particles enable FA to strengthen Metal Matrix Composites (MMCs) by significantly improving their load-bearing capacity [107].

Interfacial Bonding: FA particles possess the ability to form strong bonds with the matrix material in composites. This results in improved interfacial bonding, facilitating the transfer of stresses between the reinforcing fibers and the matrix. Enhanced interfacial bonding reduces the likelihood of crack initiation and propagation within the composite, thereby increasing its overall mechanical integrity [108].

Stiffening Effect: The addition of FA to composites leads to a stiffening effect. This means that the composite becomes stiffer and less flexible, which is reflected in the higher flexural modulus. The increased stiffness can be advantageous in applications where rigidity and dimensional stability are essential [109].

Crack Resistance: FA also serves as a crack inhibitor within composites. It can hinder the propagation of cracks, making the composite more resistant to fracture. This crack resistance enhances the overall flexural strength and durability of the composite material [110].

7.2 Particle Size

Particle size plays a significant role in determining the mechanical properties of composites containing FA. Smaller FA particles, typically in the range of 20-90 μm or even finer at 20 nm, have been observed to exhibit a greater enhancement in tensile strength as the FA content increases. This phenomenon is attributed to several key factors:

Interfacial Area: Smaller particles have a higher surface area per unit volume. As particle size decreases, there is a corresponding increase in the interfacial area available for interaction within the composite. This enhanced interfacial area facilitates a stronger bond between the FA particles and the composite matrix. This improved bonding results in an augmentation of tensile strength. Research has shown that finer 20 nm FA particles, when used in epoxy composites with hemp fiber, led to a significant 51.25% increase in impact strength [89].

Homogeneous Dispersion: Finer particles tend to disperse more uniformly within the composite matrix. This uniform dispersion reduces stress concentration points and enhances the material's resistance to impact. Homogeneous dispersion is particularly advantageous because it minimizes the formation of agglomerations, which can compromise material strength [111].

7.3 Surface Modification

Surface modification of FA can significantly improve adhesion between the matrix material and the filler. This enhanced adhesion leads to better interfacial bonding, resulting in higher compressive and tensile strength in composites [112].

Micro-Bead Surface Coating: Surface modification techniques, including micro-bead surface coating, have been explored to alter the surface characteristics of FA particles. These modifications can influence the interfacial bonding between FA and the matrix material, potentially leading to improved mechanical properties [113].

Ultrasonic Treatment: Ultrasonic treatment of FA can enhance its dispersion within a composite, reducing voids and improving load-bearing capacity. This has been observed in flax tow composites, resulting in a notable 35% increase in flexural strength [32].

7.4 Fiber Effects

Fibers are known for their high tensile strength and stiffness, which make them excellent candidates for reinforcing FA composites. The inclusion of fibers significantly improved the

tensile strength of FA composites. The aligned and rigid structure of fibers reinforces the matrix, reducing crack propagation and enhancing tensile properties. Fibers also contribute to improved thermal properties of FA composites. The natural insulating properties of jute reduce thermal conductivity, making these composites suitable for applications requiring thermal insulation.

Fibers play a crucial role in enhancing the mechanical and thermal properties of composites. Here's how:

Reinforcement and Stress Distribution: Various fibers, including carbon, glass, and natural options like jute, serve as a vital reinforcement component embedded within the composite matrix. Their primary role is to enhance the structural integrity of the composite, substantially boosting its load-bearing capacity and, consequently, elevating its mechanical strength. In practical terms, when external forces act upon the composite, these fibers excel at evenly dispersing the stress across the material. This uniform stress distribution not only averts localized failures but also substantially enhances the composite's overall toughness and resistance to fracture [114].

Improved Modulus: Fibers significantly increase the stiffness and modulus of composites. This leads to higher rigidity and resistance to deformation under load, making them suitable for structural applications [115].

Weight Reduction: Fibers, including carbon, glass, and aramid, enhance mechanical properties while reducing weight in composites. Carbon fiber composites offer an exceptional strength-to-weight ratio, ideal for aerospace components. Glass fibers provide strength with lower weight and are used in boats, wind turbine blades, and autos [116].

8 Challenges in FA Addition

While FA offers numerous advantages when added to natural fiber composites, certain challenges must be addressed:

- **Optimizing Filler Content:** Achieving the ideal filler content in composites can be challenging. While the addition of FA enhances mechanical properties, exceeding optimal levels can weaken the matrix-fiber interface and result in a minor drop in tensile strength. The addition of small FA particles (5%-15 wt. %) with alkali-treated fibers enhances tensile strength, with the most significant improvement observed at 10% FA content. However, excessive FA content beyond 10% can weaken the matrix-fiber bond [84], leading to a minor reduction in strength. Therefore, finding the right balance of FA content is crucial for maximizing composite performance.
- **Agglomeration of Nano-FA:** Insufficient dispersion of nanoparticles is a major challenge in nanocomposites. Nanoparticles tend to agglomerate due to their high surface area and volume effect [27], [57]. Beyond 8 wt. % Nano-FA concentration, the increase in tensile strength becomes less noticeable due to particle aggregation. Excessive FA addition can cause agglomeration and stress concentration points [20]. Preventing agglomeration and ensuring even nanoparticle dispersion within the polymer matrix is essential for achieving the desired mechanical improvements. Surface treatments and coupling agents can help mitigate this challenge. If agglomeration occurs, the apparent volume occupied by the filler increases, and agglomeration results in larger particles, resulting in vacuum space, which might be responsible for strain propagation.

9 Future Research Direction

The application of FA as a nanofiller in diverse composite materials has attracted considerable interest recently because of its ability to improve mechanical characteristics, fire resistance, and overall material performance. Future research directions in this field can be classified into many principal areas, including the optimization of processing techniques, the examination of synergistic effects with other nanomaterials, and the assessment of environmental consequences and sustainability.

A potential study field is the mechanical activation of FA to generate nano-sized particles that can function as efficient fillers. Research indicates that mechanical activation can markedly increase the reactivity and surface area of FA, resulting in enhanced bonding and mechanical characteristics in composites. For example, it was shown that nanostructured FA can strengthen acrylonitrile butadiene styrene (ABS) nanocomposites, improving their mechanical properties. It was also observed that high-energy ball milling alters the microstructure of FA, yielding a more reactive and efficient filler. Subsequent research may concentrate on refining milling parameters to attain the requisite particle size and surface properties for particular applications. An important area for future research is to the synergistic effects of FA in conjunction with other nanomaterials, including multiwall carbon nanotubes (MWCNTs) and nano-silica. Investigated the amalgamation of FA with multi-walled carbon nanotubes to improve the fire-retardant characteristics of epoxy composites. Furthermore, highlighted the function of nano-silica in enhancing the characteristics of cement paste containing high calcium FA. The investigation of diverse combinations of FA with various nanofillers may result in the creation of new materials with customized qualities for certain applications, including construction, automotive, and aerospace sectors.

In summary, future research on the utilization of FA as a nanofiller includes optimizing processing methods, investigating synergistic interactions with other nanomaterials, and evaluating environmental sustainability. By focusing on these aspects, researchers may fully realize the potential of fly ash in composite materials, thereby advancing technology and promoting environmental sustainability.

10 Conclusion

This comprehensive review paper delves into the multifaceted applications of FA in various composite materials, with a particular focus on mechanical enhancements, chemical treatments, and particle size considerations. Through a meticulous examination of numerous studies, several key findings emerge, shedding light on the pivotal role of FA in composite material engineering.

First and foremost, the optimal weight percentage of FA within composite materials emerged as a critical factor in achieving improved mechanical properties. It was observed that an optimal range of FA content exists for different types of composites. In cases such as epoxy-based composites, an addition of (10 - 20) wt. % Nano-FA yielded remarkable improvements in tensile strength, flexural strength, and impact resistance. Likewise, in thermoplastic composites, the mechanical properties were substantially enhanced when FA was added within (5–10) wt. % range. Beyond these optimal ranges, the matrix-fiber interaction weakened, resulting in diminishing returns and, in some cases, detrimental effects on the composite's performance.

The size of FA nanoparticles emerged as a critical factor influencing composite properties. Nano-scale FA particles, with sizes typically less than 45 microns, exhibited superior dispersion within the matrix, contributing to improved tensile strength, modulus, and abrasion resistance. Conversely, larger micro-scale FA particles faced challenges in uniform dispersion, leading to weaker interfacial bonding and reduced resistance to abrasion. Achieving proper particle size distribution and dispersion is crucial for optimizing the mechanical benefits of FA in composites. Furthermore, Surface treatment of FA particles was also found to play a pivotal role in enhancing composite properties. Chemical treatments, such as alkali treatments or the use of coupling agents, significantly improved interfacial adhesion between FA and the polymer matrix. An optimum NaOH concentration range, generally between 10-15M, was found to enhance the reactivity of FA, resulting in a faster and more complete polymerization reaction and, subsequently, higher strength properties. However, excessive NaOH concentration could lead to over-alkalization and a decrease in strength. Additionally, the amount of Si and Al released during geo-polymerization was shown to be dependent on the alkali activator used and its concentration. This surface modification mitigated issues like nanoparticle aggregation, ensuring even distribution and preventing agglomeration within the matrix.

The findings of this review paper emphasize that the judicious selection and treatment of FA, in harmony with other components in composite materials, holds the key to unlocking their full mechanical potential and expanding their practical utility in diverse industrial sectors.

11 Key Findings

- For epoxy-based composites, (10 - 20) wt. % Nano-FA yields remarkable improvements in tensile strength, flexural strength, and impact resistance.
- In thermoplastic composites, substantial enhancements in mechanical properties are observed within the (5–10) wt. % FA range.
- Beyond these optimal ranges, matrix-fiber interaction weakens, leading to diminishing returns and, in some cases, detrimental effects on composite performance.
- Nano-scale FA particles (<45 microns) exhibit superior dispersion, contributing to improved tensile strength, modulus, and abrasion resistance.
- Larger micro-scale FA particles face challenges in uniform dispersion, resulting in weaker interfacial bonding and reduced resistance to abrasion.
- Chemical treatments like alkali treatments or coupling agents significantly improve interfacial adhesion between FA and the polymer matrix.
- An optimum NaOH concentration range (10-15M) enhances reactivity, leading to a faster and more complete polymerization reaction and higher strength properties.
- Excessive NaOH concentration can lead to over-alkalization and a decrease in strength.
- The amount of Si and Al released during geo-polymerization is dependent on the alkali activator used and its concentration.

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