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Review on Recent Advances in Filler-Reinforced Polymer Composites: Natural, Mineral, and Hybrid Systems

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Abstract

Recent advances in polymer composite technology have focused on incorporating sustainable natural fillers and performance-enhancing inorganic additives to achieve balanced mechanical, thermal, and environmental properties. The collection of studies presents a comprehensive overview of these innovations across thermoset and thermoplastic systems, including vinyl ester, epoxy, polypropylene (PP), high-density polyethylene (HDPE), and polylactic acid (PLA). Each investigation explored unique filler-matrix interactions, fabrication methods, and structure-property relationships. Natural fillers such as groundnut shell powder, pecan nutshell, coffee husk, coconut shell, and agricultural residues demonstrated notable improvements in tensile, flexural, and impact properties at optimized filler ratios, while inorganic and nano-fillers like SiC, TiO_2, and red mud provided significant stiffness, wear resistance, and hardness enhancement. Hybrid composites and bio-nano systems introduced synergistic effects that improved interfacial adhesion, durability, and multifunctional performance, such as flame retardancy, thermal conductivity, and environmental remediation capabilities. A notable trend across these works is the shift toward waste-to-wealth material engineering, emphasizing green processing, circular economy benefits, and scalability for industrial applications. Collectively, these findings indicate that optimizing filler surface treatment, dispersion, and hybridization can yield composites combining sustainability with high mechanical performance. This review consolidates the state-of-the-art developments, compares fabrication and performance parameters, and identifies future opportunities for bio-based hybrid composites as eco-efficient alternatives to conventional materials.

Keywords: Polymer Composites, Natural Fillers, Mineral fillers, Hybrid Systems, Mechanical Properties, Sustainable Materials.

1. Introduction

The field of filler-reinforced polymer composites has expanded rapidly over the past two decades, driven by the demand for materials that combine lightweight performance, mechanical strength, and environmental sustainability. A series of recent investigations [1-5] demonstrated the transition from traditional single-filler composites to hybrid formulations integrating natural, mineral, and biodegradable fillers. For instance, vinyl ester composites reinforced with groundnut shell powder and calcium carbonate exhibited enhanced tensile and flexural properties [1], while epoxy composites containing micro-sized wood flour improved strength and toughness [2]. Similarly, argan nut shell particles in HDPE matrices [3], lignocellulosic fillers in PP [4], and pecan nutshell biofillers in PLA [5] showed distinct performance gains, confirming the versatility of agricultural waste materials.

Inorganic and nano-reinforcements have also contributed to property enhancement. The use of TiO_2 nanoparticles in vinyl ester [6], thyme waste and compatibilizers in HDPE [7], mate-tea and eucalyptus particles in PP [8], coconut shell powder in epoxy [9], and coconut shell in polyethylene [10] established how filler dispersion and interfacial bonding affect mechanical integrity.

Subsequent works [11-15] extended these principles to functional composites using coffee husk fibers, SiC-filled glass fiber epoxy, and nano-red-mud polyester systems, achieving improved modulus, wear resistance, and interlaminar shear strength. Later studies [16-20] diversified into environmental and hybrid applications, such as adsorbent bio-waste for Cd(II) removal [16], honeycomb and Tectona Grandis fiber composites [17,18], and hybrid tamarind-palm polyester systems [19].

The most recent advancements introduced nano-hybrid composites integrating rice husk ash, Al_2O_3, SiC, graphene, cellulose nanocrystals, and basalt–graphite combinations, achieving significant improvements in mechanical strength, conductivity, flame retardancy, and durability [21-30]. Together, these high-quality studies demonstrate a consistent methodological rigor through ASTM-standard testing, microstructural (SEM) analysis, and thermomechanical evaluations (DMA, TGA, FTIR). Overall, the collective research reflects a progressive evolution in composite engineering - transitioning from natural and agricultural fillers to multi-functional hybrid systems, advancing the dual goals of sustainability and high performance in modern material science.

2. Methodological Overview

Across the studies, fabrication techniques such as hand lay-up, compression molding, extrusion-injection molding, and ultrasonication were frequently employed. Characterization tools included tensile, flexural, and impact testing, SEM for morphology, and thermal analyses (TGA, DSC, DMA). Coupling agents such as maleic anhydride grafted polypropylene (MAPP) or MAPE were often applied to improve filler–matrix compatibility. Optimal filler loadings typically ranged between 5-20 wt% for microscale fillers and 1-5 wt% for nanofillers, reflecting the balance between mechanical reinforcement and dispersion quality.

3. Discussion of Individual Studies

3.1. Natural and Agricultural Fillers in Thermoset Composites

Groundnut shell powder and calcium carbonate in vinyl ester matrices improved both tensile and flexural strength at 20 % GNP + 15 % CC [1]. Wood flour-filled epoxy [2] and argan nut shell-HDPE systems [3] demonstrated how alkaline treatment and small particle size enhance interfacial adhesion and strength. Coconut shell powder in epoxy [9] and polyethylene [10] produced notable stiffness and hardness improvements, although water absorption increased. Tectona Grandis flour [18] raised tensile and compressive strengths with increasing fiber volume. Honeycomb natural fibers [17] and tamarind-palm hybrids [19] further confirmed that optimal loadings (~9-12 wt%) maximize mechanical performance before agglomeration reduces bonding.

3.2. Lignocellulosic Fillers in Thermoplastics

Polypropylene and HDPE matrices reinforced with wood, rice husk, walnut shell, and thyme waste [4,7,8,11,13] highlighted sustainable biocomposite trends. The addition of compatibilizers (MAPP, MAPE, PEGM) markedly improved tensile and flexural strength while limiting water absorption. For instance, coffee husk fiber with MAPE increased tensile strength by 127 % and reduced water absorption by 70 % [11]. Studies on mate-tea and eucalyptus fillers [8] confirmed that mixed agro-waste can yield cost-effective yet mechanically balanced composites.

3.3. Biopolymer Composites and Renewable Fillers

PLA-based systems [5,14] reinforced with pecan nutshell, hemp hurd, alfalfa, and grape stem revealed that treated biofillers improve modulus and maintain biodegradability. Hemp hurd exhibited the best adhesion and stiffness (modulus ≈ 10.5 GPa). These works underscore the potential for biodegradable, rigid packaging and consumer goods applications.

3.4. Inorganic and Nano-Filler Reinforcements

Significant advances in mechanical and tribological performance were observed with inorganic additions. TiO_2 nanoparticles in vinyl ester [6] enhanced tensile and flexural modulus at 2.5 wt% before agglomeration effects appeared. SiC-filled glass-fiber epoxy composites [12] achieved optimal tensile and impact strengths at 10-15 wt% SiC. Nano-red-mud-reinforced polyester [15] improved wear resistance by nearly 48 % at 2.5 wt% of 110 nm particles, demonstrating the advantages of nano-scale dispersion.

3.5. Functional and Environmental Applications

Studies extended beyond structural enhancement to environmental remediation and resource recovery. Polyalthia longifolia seeds effectively adsorbed Cd(II) ions with 97.6 % removal efficiency [16], proving bio-waste's potential in water purification. Hybrid epoxy composites with cellulose micro-filler from peanut oil cake [20] offered improved tensile and thermal behavior, linking agricultural residues to high-performance material development. Subsequent works explored hybrid nano-biofillers, surface modification, and multifunctional uses such as flame retardancy and electrical conductivity enhancement, reinforcing the trend toward sustainable and multifunctional composites [21-30].

4. Comparative Analysis

The comparative assessment of studies reveals the progressive enhancement in composite technology through the incorporation of natural, mineral, and hybrid fillers. Across all systems, the results confirm that the type, particle size, surface treatment, and concentration of fillers critically influence the mechanical, thermal, and tribological performance of polymer composites [1-30].

Natural fillers such as groundnut shell powder, wood flour, argan nut shell, and coconut shell powder [1-10] exhibited clear mechanical improvements, particularly in tensile and flexural strength at filler loadings between 10-25 wt%. The positive effect was attributed to uniform filler dispersion and good interfacial adhesion within epoxy, vinyl ester, and polyethylene matrices. However, higher filler content often led to particle agglomeration, resulting in slight reductions in elongation and impact strength.

Incorporation of inorganic and nano-fillers, including TiO_2 nanoparticles, SiC particles, and nano-red mud [6,12,15], provided a significant increase in hardness, wear resistance, and dimensional stability even at very low filler levels (1-3 wt%). These systems showed improved load transfer and microstructural compactness, indicating that nanoscale reinforcement is highly effective for enhancing matrix rigidity and durability.

Hybrid composites further extended these advantages by combining bio-based and inorganic components [20-30]. Systems such as cellulose microfiller–flax/epoxy [20], rice husk ash–Al_2O_3/epoxy [21], banana fiber-SiC/PP [22], and graphene-basalt/polyester [30] achieved simultaneous improvements in tensile modulus, flexural strength, thermal conductivity, and flame retardancy. The synergistic interaction between fillers minimized interfacial voids and enhanced load transfer efficiency.

A distinct pattern was also observed in biopolymer-based matrices, such as PLA and HDPE composites [5,11,14,24,29], where chemical coupling agents like MAPP and MAPE improved filler-matrix compatibility and reduced water absorption. This demonstrates the importance of surface modification for optimizing the performance of natural filler composites.

Comparing the overall data, the hybrid and nano-filler systems [21-30] outperformed single-filler composites in almost all mechanical and functional categories, offering superior stiffness, wear resistance, and moisture durability. The combination of low-density natural fibers with high-modulus inorganic nanoparticles effectively balanced strength, cost, and environmental sustainability.

The collective findings emphasize that filler hybridization and surface modification are key strategies for developing next-generation, eco-efficient polymer composites with tailored mechanical and thermal properties suitable for engineering and environmental applications.

5. Applications

The reviewed works collectively demonstrate that natural and hybrid fillers can serve in diverse sectors:

- Automotive & Transportation: Lightweight interior panels, dashboards, and semi-structural parts [1,2,7,12,19].
- Construction & Furniture: Ceiling boards, decking, and partition materials utilizing lignocellulosic fillers [4,8,13,18].
- Packaging & Consumer Goods: PLA- and HDPE-based biocomposites offering biodegradability [5,14].
- Environmental Applications: Adsorbent materials for wastewater purification [16].
- Aerospace & Industrial Components: Nano-TiO_2, SiC, and red-mud-reinforced thermosets for wear-resistant or high-temperature performance [6,12,15].

6. Conclusions

The collective research demonstrates a decisive trend toward eco-efficient and multifunctional composites. By strategically combining natural, mineral, and nano-fillers, these systems achieve enhanced stiffness, strength, and environmental sustainability. The inclusion of hybrid filler systems marks a significant evolution, offering improvements in thermal conductivity, wear resistance, flame retardancy, and moisture durability. Future studies should emphasize large-scale manufacturing, advanced interfacial chemistry, and performance modeling to accelerate the industrial adoption of sustainable composites. Additionally, incorporating advanced surface modification and nanotechnology-based treatments could further improve interfacial bonding and long-term durability of hybrid composites. Future research should also consider large-scale manufacturability, cost evaluation, and environmental life-cycle assessment to ensure practical and sustainable implementation of these materials.

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Ethical considerations

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Conflict of Interest

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