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A Comprehensive Review of Polypropylene-Based Hybrid Composites: Structure, Properties, and Applications

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Abstract - The increasing demand for lightweight, high-strength, and cost-effective materials in engineering and industrial applications has driven the development of reinforced and hybrid polymer composites. This review comprehensively examines the evolution and current advancements in polypropylene (PP)-based composites, particularly focusing on hybrid systems incorporating both short fibers and inorganic fillers such as wollastonite, talc, and glass fibers. The article highlights the mechanical, thermal, morphological, and crystallization behavior of these composites, drawing from both historical and recent studies. Key strategies, such as the use of compatibilizers (e.g., PP-g-MA), nanofiller hybridization, and bio-based reinforcements, are explored for enhancing interfacial adhesion, impact resistance, and thermal stability. The role of wollastonite as a multifunctional filler—improving dimensional stability, heat deflection temperature, and processability—is emphasized. Additionally, the review discusses rheological behavior and processing considerations critical to optimizing composite performance. The findings underscore the potential of PP-based hybrid composites to meet diverse application requirements in automotive, construction, packaging, and electronics industries, while also offering pathways for sustainable and multifunctional material solutions.

Key Words: *Thermoplastics, Hybrid composites, Mechanical properties, rheological properties*

1.INTRODUCTION

This article shows the suggested setup and attendance of a manuscript organized for ISJEM monthlies. Established papers will be for a living typeset. This template is intended to be a tool to improve manuscript clarity meant for the referees. The final layout of the typeset paper will not match this template layout. The growing technological advancements and industrial demands have necessitated the development of materials offering high strength-to-weight ratio, enhanced stiffness, and cost-effectiveness. Conventional plastics, limited by low strength and rigidity, are being reinforced to expand their utility in structural applications. Reinforced plastics exhibit superior specific strength, modulus, and corrosion resistance, making polymer composites a promising solution. These composites comprise strong reinforcement fibers within a polymer matrix, significantly improving mechanical performance.

The 1980s marked a shift with the advent of advanced thermoplastic matrices, enabling faster processing and superior damage tolerance compared to traditional thermosetting polymers. Among thermoplastics, polypropylene (PP) stands out due to its affordability, ease of processing, solvent resistance, and balanced mechanical properties. Its broad application base includes automotive, appliances, and construction sectors [1]. The use of mineral fillers in PP enhances properties such as stiffness, shrinkage control, and flammability resistance while lowering costs [2,3].

PP composites now rival engineering plastics like ABS and nylon in performance and cost-efficiency. Common fillers include talc, $CaCO_3$, kaolin, mica, carbon black, and glass fibers. The hybridization of multiple reinforcements allows for property optimization—achieving strength from one filler and dimensional stability or cost reduction from another [4–6]. Hybrid composites are particularly relevant in applications prioritizing high strength-to-weight ratios, such as automotive and mechanical engineering [7–12].

Hybrid composites not only offer tailored mechanical properties but also bring economic benefits and synergistic enhancements [13]. Commercially available hybrid PP composites, like glass/wollastonite systems, combine the strength of glass fibers with the dimensional stability of wollastonite. Varying filler levels enables customized material performance. Short fiber reinforced polymers (SFRPs) are gaining popularity due to their favorable mechanical properties and compatibility with rapid, low-cost injection molding processes [14–16]. However, limited literature addresses hybrid systems incorporating both short fibers and inorganic fillers [17–19].

Although glass fibers are well-established, mineral fibers like wollastonite are gaining attention for their superior dispersion, thermal and chemical stability, and processing advantages. High-volume wollastonite inclusion enhances tensile and impact properties, offering a cost-effective reinforcement strategy [20–25]. It also replaces natural fibers where dimensional stability is prioritized [26,27]. Compared to glass, wollastonite improves flow properties, heat deflection temperature (HDT), and scratch resistance, while supporting better interfacial adhesion and tensile properties. Other mineral fillers like talc and $CaCO_3$ are widely used in semicrystalline polymers, with mica and wollastonite applied to a lesser extent [28].

Research on PP/wollastonite/silicone rubber composites shows that adding wollastonite enhances HDT and notched impact strength but lowers tensile strength and unnotched

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impact strength. Silicone rubber, used to improve flexibility, helps achieve balance between stiffness and flexibility, with maleic anhydride acting as a compatibilizer [29–33].

The push for multifunctional materials has accelerated the development of PP-based hybrid composites with capabilities like self-healing, EMI shielding, and recyclability. The global PP composites market is expected to grow at a CAGR of 6.8% from 2023 to 2030, driven by increasing use in electric vehicles, renewable energy, and sustainable packaging [34,35].

Research by Putfak and Larpkasemsuk demonstrated that varying the ratios of wollastonite and talc in PP composites can tailor properties like impact strength, tensile modulus, and heat distortion temperature, catering to specific application requirements [36].

2. MECHANICAL PROPERTIES OF HYBRID COMPOSITES

At the first manifestation of an contraction, spell it out followed by the acronym in additions, e.g., charge-coupled diode (CCD). Over the past four decades, considerable research has been dedicated to fiber-reinforced composites across diverse applications. The growing demand for materials with higher strength-to-weight ratios, improved stiffness, and safety features has driven advancements in structural design and economic material development [37].

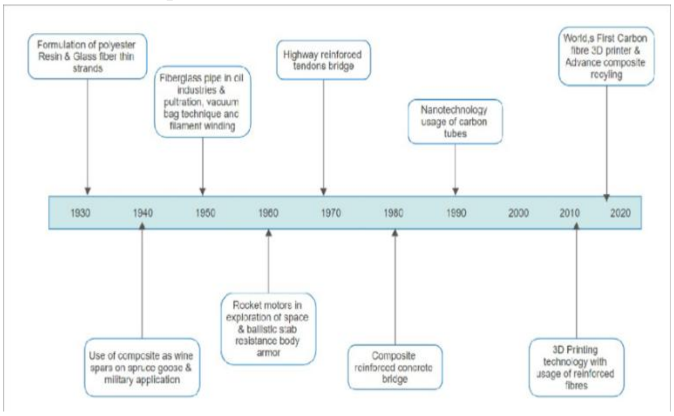

Fig -1: Timeline diagram of advancement in composites over a different period [38,39,40]

Plastic materials, although lightweight, face limitations due to their lower strength and rigidity, making their use in structural parts challenging. A broad range of fibers has been utilized to enhance the mechanical performance and cost-effectiveness of composites [41-46].

Table -1: Comparison of mechanical properties of common reinforcement fibers

Fiber Type	Tensile Strength (MPa)	Young's Modulus (GPa)	Density (g/cm³)
Glass Fiber	2000–3500	70–85	2.5–2.6
Carbon Fiber	3000–7000	230–400	1.7–1.9
Wollastonite	300–500	80–550	1.44–2.7

G. Kretsis described the hybrid effect as the enhancement of failure strain in lower elongation reinforcing materials. Due to the diversity in glass fibers, carbon fibers, and epoxy resins used until 1987, he compared composite performance data qualitatively [47].

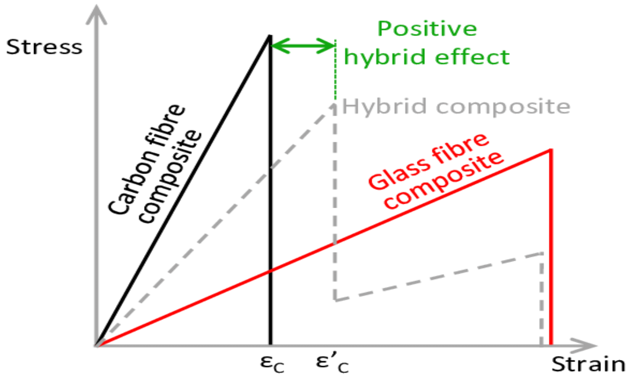

Fig -2: Schematic stress-strain diagram of a hybrid composite and its two reference composites. The hybrid shows two peaks, which are linked to failure of the carbon and glass fibre composite respectively [48]

Manders employed interferometry to assess strain distribution under tensile loads, correlating composite behavior with fiber-resin bonding. Avest and Sillwood, along with Bunsell and Harris, observed crack spacing in hybrid composites, attributing delamination prevention to minimum crack spacing of 1–1.5 mm [49]. Cantwell et al. emphasized the influence of molding conditions on GF/PP and GMT composites and reported a 15% increase in tensile strength of PP/GMT with coupling agents, although Charpy impact resistance decreased by 50%. He later introduced a low molecular weight PP coupling agent, which enhanced multiple mechanical properties [50].

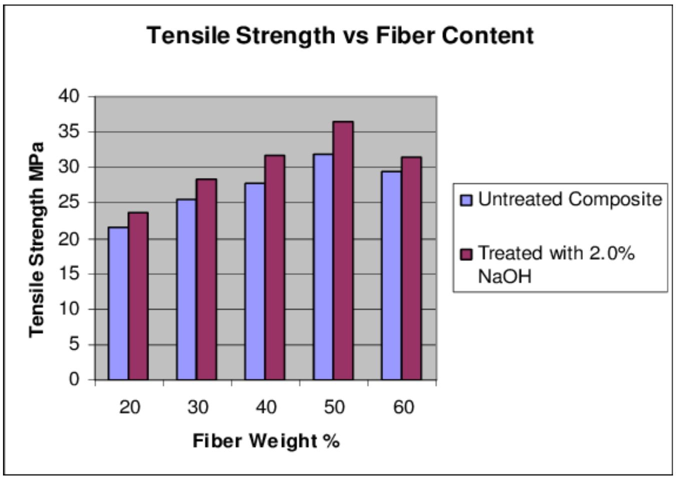

Fig -3

Fig -3: Improving Mechanical Properties of Eco-Friendly Polymer Hybrid Composites [51]

By 1994, research on glass mat reinforced thermoplastics (GMTs) had expanded to include combinations with matrices like PP, linear polyester, and polyamides. Frejes Kozma showed that fracture toughness in GMT-PP composites remained stable with reinforcement, but initial fracture energy declined as reinforcement increased [52-58].

J. Thomason used single pull-out tests to determine critical fiber length, revealing that laminate stiffness was largely independent of fiber length (above 0.5 mm) and fiber concentration (above 40% w/w). Hybrid composites were found to offer tailored mechanical properties and cost advantages over single-fiber systems [59-67]. Wollastonite, a cost-effective inorganic filler, has been widely used as reinforcement in polymers such as PA6-PP, PMMA, epoxy, and polyamide. It features low density (1.44–2.7 g/cm^3) and high modulus (80–550 GPa), making it suitable for hybrid composites [67,68]. Hybrid composites of glass fiber and wollastonite have demonstrated enhanced strength, dimensional stability, and warp resistance. Traditional PP toughening with rubbers like EPR and SEP improves toughness but compromises strength and hardness, while rigid fillers may aggregate, leading to crack initiation and reduced mechanical performance [69-78].

Resistance to crack propagation in PP is low due to filler aggregation-induced voids. Under external force loading, those voids in the aggregation usually act as crack initiators and have catastrophic effects on mechanical properties since the cracks are more readily initiated inside these loose aggregations of filler prior to the filler particles debonding from matrix [79-83]. In 1984, Pande et al. noted constant flexural strength in SGF/glass-particle/PP hybrids with reduced PP content (80% to 70%) at a fixed SGF/glass ratio (2:1) [81]. Yilmazer (1992) observed that adding small SGF amounts to glass-particle/ABS composites reduced elongation at break and impact resistance. Similarly, Hargarter (1993) found that incorporating talc into SGF/talc/PBT hybrids reduced tensile and flexural strength [84-85].

Wollastonite's acicular shape and high aspect ratio (10-20) make it ideal for reinforcement. Numerous studies have assessed the mechanical performance of PP-wollastonite composites, with and without surface modification [86-93]. The effectiveness of silane coupling agents and PP-g-MA in PP/CaSiO3 composites was examined, with results showing moderate tensile strength changes and substantial impact strength improvement at 5% wollastonite and 10% MAPE. Increased interfacial bonding at higher wollastonite content improved tensile strength.

According to A.D. Drojov, higher fiber content leads to more microdomains with plastic deformation [94,95]. Fu et al. found that adding particles to SGF/ABS composites reduced tensile strength in high adhesion cases but had minimal effect in low adhesion cases [96,97]. In short fiber composites, stress transfer is crucial and differs between single and multi-fiber systems, with neighboring fibers influencing matrix stress transfer. Shao Yun Fu and others developed models to describe these differences [96-103]. Investigations into PP/CaSiO3 composites showed improved stiffness and tensile strength with enhanced interfacial adhesion.

Upinder and Bidyut observed that adding silicon rubber and maleic anhydride to wollastonite-filled PP slightly affected mechanical properties but significantly enhanced HDT and notched impact strength, while reducing unnotched impact strength and tensile modulus [104].

Efforts to develop PE-wollastonite-sisal fiber hybrids via rotational molding highlighted advantages like minimal residual stress and low material waste, though with limitations in processing time and mold design. Wollastonite's needle-like structure enhances mechanical and thermal properties, dimensional stability, and processing efficiency due to increased crystallization temperatures. High-volume wollastonite use boosts tensile and impact properties while reducing costs. Wollastonite's contribution to dimensional stability complements natural fibers' ability to reduce composite density [25-27, 105-107]. Hybrid composites using both glass fibers and inorganic fillers have outperformed single-filler systems. For example, PBT/wollastonite composites in 70/30 and 50/50 ratios showed significant tensile modulus improvements.

Himani and Purnima (2010) developed PP/GF/wollastonite hybrids via extrusion and injection molding. Their study reported a positive hybrid effect on ultimate strength and a negative effect on tensile modulus [108]. Advanced Mechanical Properties

Dynamic Mechanical Analysis (2023 Findings)

Recent DMA studies reveal that:

• 3D-printed PP/wollastonite/short carbon fiber hybrids exhibit 25% higher storage modulus at 100°C compared to injection-molded counterparts [109]

• Self-reinforced PP/wollastonite composites demonstrate unusual toughness enhancement (150% increase in impact strength) through transcrystallization mechanisms [110].

3. Morphology of Composites

Wetting and adhesion between fillers and the matrix are fundamentally governed by the surface energy properties of each component. Measurements of surface properties in pure materials enable prediction of interfacial adhesion within the composite structure [111–114]. The characteristics of the components, particularly those influencing interfacial adhesion, directly affect the final performance of the composite material [115-117].

Fiber end geometry plays a crucial role in the fiber/matrix stress transfer mechanism, with failure typically initiating at the fiber-end/matrix interface due to stress concentrations from geometric and material discontinuities [118-121]. Fiber efficiency has been shown to depend on several factors [124–129]. M. Shuster et al. used numerical and experimental approaches—including photoelastic methods—to study stresses in the polymer matrix around short fibers, demonstrating that spherical enlarged fiber ends reduce the risk of interfacial debonding and thus improve modulus [123,126,130–134]. Despite this, it was reported that fiber/matrix adhesion does not significantly affect fracture resistance due to variations in fiber length distribution [135].

Mechanical properties in short-fiber/inorganic filler/polymer composites are influenced by the amount of matrix, hybrid ratio of short fiber to filler, fiber orientation, and fiber length distribution [136,137]. Fiber efficiency factors linked to strength and rupture energy indicated that wollastonite incorporation affects fiber pull-out force and energy. Weak interfacial adhesion may arise from multiple causes and can be optimized through surface treatment and suitable adhesion promoters [138]. Turcsanyi et al. noted that with isotropic filler geometry and no anisotropy during processing, composites behave isotropically. In the absence of adhesion, the matrix bears the entire load, matching the yield stress of unfilled polymer [139]. Key factors influencing particulate composite properties include filler size, concentration, matrix properties, interfacial adhesion, and surface characteristics. Lange, Radford, Evans, Newaz, Maxwell et al., and Kinlock et al. investigated fracture mechanisms in particulate filler composites [140–144].

By 1993, limited work addressed the effects of filler size and surface treatments in PP composites. Jian Shen found that fillers reduce mechanical strength and melt flow rate due to voids and weak filler-matrix interactions. This weakness arises from the incompatibility of nonpolar PP with polar inorganic fillers, necessitating surface modification for improved adhesion [145–147. Wollastonite ($CaSiO3$), with its needle-like acicular form and high Mohs hardness, is suitable for polymer reinforcement due to its high aspect ratio and unique silica tetrahedron structure [31,32,148,149]. Enhancing PP/$CaSiO3$ interfacial adhesion requires suitable coatings based on anticipated physical and chemical interactions. PP-g-MA is a commonly used compatibilizer, and KH-550 (g-aminopropyltriethoxysilane) can be used as a filler coating, where the amine group is expected to react with the MA group.

Shao Yun Fu studied fiber length distributions in SGF/ABS composites. At low stress, they deform linearly; at high stress, nonlinearly, due to interfacial microfailures around fibers. Unmodified glass fibers provided the highest interfacial adhesion, followed by aminosilane/polyurethane dispersion-treated fibers, with the lowest adhesion observed in aminosilane/epoxy dispersion-treated composites. Stress-strain analysis of ABS/calcite/SGF hybrids showed brittle fracture characteristics, affirming the superior adhesion of untreated glass fibers.

4. Crystallization Behaviour in Composites

Polypropylene (PP) is one of the most widely used thermoplastics in injection molding applications due to its simple chemical structure and cost-effectiveness [150,151]. However, it is rarely used in its pure form; instead, it is typically compounded with mineral fillers to enhance mechanical, thermal, electrical, and dimensional stability properties [152–157].

Liu Jingjuang investigated the crystallization behaviour of PP-wollastonite composites using Differential Scanning Calorimetry (DSC) and observed that the β-phase melts first and then recrystallize into the α-form (peak Tm4). The melting temperature was only slightly influenced by wollastonite content ranging from 3.2 to 17.7 vol% [158]. Similarly, other researchers have also

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examined thermal conductivity and the influence of fiber treatments on thermal stability [159–160]. While the incorporation of fillers like $CaSiO_3$ may not significantly affect the crystallization temperature or degree of crystallinity of PP [161], they do impact the nucleation and crystallization mechanisms of the polymer matrix.

Hadal (2004) performed DSC analysis on PE–wollastonite composites and found that wollastonite reinforcement raised the onset of crystallization and reduced processing time in injection molding by accelerating crystallization [162]. In another study, Makarand Rishbul et al. examined PBT/wollastonite composites (70/30 and 50/50 ratios), reporting enhanced crystallization and marginal changes in melting behavior [163].

The addition of fillers has also been found to increase the crystallization temperature of PP during cooling, promoting heterogeneous nucleation and faster crystallization rates [164]. Research on nanoscale reinforcements has shown that smaller particles, such as single-walled carbon nanotubes (SWNTs), alter polymer crystallization by acting as nucleating agents, thereby reducing spherulite size and narrowing melting/crystallization peaks in composites compared to neat PP [165–173].

Polypropylene's β-crystals melt at lower temperatures than α-crystals, and DSC analyses indicate narrower exotherm peaks (ΔW) in SHSD-reinforced PP, suggesting a more uniform crystallite size distribution. While fillers decrease the degree of supercooling required for crystallization, the crystallization peak temperature remains largely unchanged [174–175].

Thermal conductivity of composites has also been studied, and recent research by Meng and Dou revealed that pimelic acid-modified wollastonite can influence β-iPP formation during isothermal crystallization, though under non-isothermal conditions, the filler reduced the crystallization temperature [176,177]. Similarly, Akinci reported increased crystallization with the addition of graphite [178].

Wollastonite has emerged as a significant nucleating agent in PP matrices, increasing crystallinity, thermal stability, and mechanical performance, which makes it suitable for high-performance applications [179,180]. Karagöz et al. (2024) evaluated PP hybrid composites with hazelnut shell and wollastonite fillers. While wollastonite enhanced mechanical strength, hazelnut shell inclusion impacted thermal behavior. Compatibilizers such as SEBS and SEBS-g-MA improved impact resistance but slightly reduced other mechanical properties [181].

The rheological behaviour of polypropylene (PP) filled composites has been the subject of extensive research due to its critical role in determining processability and final product properties. Most polymers, including PP, are viscoelastic solids that exhibit both viscous and elastic characteristics [182]. It is well-established that the addition of fillers generally increases the melt viscosity and reduces melt elasticity. Understanding the melt rheology is thus essential for controlling flow during processing operations such as injection molding and extrusion.

The rheological response of a polymer melt includes its viscous component (characterized by the loss modulus, G") and its elastic component (storage modulus, G') [183]. Oscillatory rheometry is widely used to evaluate these properties [184]. In such tests, specimens are subjected to sinusoidal deformation between parallel plates, and their viscoelastic response is measured. In the Newtonian plateau, G" follows a slope of 1 while G' follows a slope of 2 on a double logarithmic scale. A deviation from this behavior, particularly in the slope of G', often indicates effects such as cross-linking, degradation, or polymer superstructures.

Rotational rheometers using parallel plate (PP) geometry are common for such measurements due to effective gap control [185]. Since polymer processing always involves flow, which is influenced by molecular chain mobility and entanglement, rheological behaviour significantly affects not only processing but also the mechanical performance of the final product. For instance, molecular orientation—largely governed by melt rheology—affects properties of molded products, films, and fibers [186].

Filler addition alters the flow behavior and processability of polymer systems [187]. While the rheological properties of short fibre reinforced thermoplastics differ from those of neat polymers, they are not vastly different. In fact, when aligned properly, short fibre composites can approach the mechanical properties of continuous fibre composites, offering cost and production advantages [188].

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Numerous studies have explored the rheology of short fibre reinforced thermoplastics and elastomers [189]. Gupta and Punvar examined the impact of glass fibres, noting that when fibre aspect ratio nears unity, the behaviour resembles that of particulate-filled systems [190]. Thomas and colleagues also reported rheological behaviour of short sisal, coir, and pineapple fibre-reinforced systems [191–194].

The importance of rheology in diverse polymer processing techniques—including extrusion, blow molding, fiber spinning, and calendaring—has been highlighted by researchers such as Al-Faris, Al-Zahrani, Osswald, Mills, and Birley [195–198]. Rheological methods not only inform on processability but also provide structural insights, including molar mass distribution, crosslinking, branching, and crystallization behavior [199, 200]. These measurements, typically performed using oscillatory techniques with precise temperature control, are most often conducted using rotational rheometers with parallel-plate geometry [201].

Recent studies have extended rheological investigation to nanofilled systems. The rheology and scratch resistance of PP/nano wollastonite composites have been evaluated [203–204]. Improved interfacial adhesion from matrix modification has been shown to enhance storage modulus. Selecting the appropriate rheological technique is crucial; the method should ideally simulate the intended processing application—for example, extrusion-based characterization for extrusion applications.

Himani et al. (2023) performed dynamic rheological testing on PP/GF/WF composites and found that the inclusion of fillers significantly influenced storage modulus, loss modulus, and complex viscosity. These changes directly impacted the composites' processability and mechanical performance [205].

Hybrid Composites: Concept and Advantages

Hybrid composites, which combine two or more types of reinforcements, offer a strategic approach to tailor material properties. The hybridization of glass fibers with wollastonite, for example, provides an optimal balance between strength (from glass fibers) and dimensional stability (from wollastonite) [6]. The benefits of hybrid composites include:

1. Tailored Mechanical Properties: By adjusting the ratio of reinforcements, specific property requirements can be met [13].

2. Cost Efficiency: Partial replacement of expensive fibers (e.g., carbon fibers) with cost-effective mineral fillers reduces material costs.

3. Synergistic Effects: Hybrid systems often exhibit enhanced performance beyond what is achievable with single-fiber composites [13].

Novel Reinforcement Strategies (2020-2024)

Nanofiber Hybridization

Recent studies demonstrate that incorporating cellulose nanofibers (CNFs) with wollastonite in PP matrices improves tensile strength by 40% while maintaining excellent biodegradability [206]. Graphene nanoplatelet (GNP)-modified wollastonite hybrids show 35% higher thermal conductivity compared to conventional fillers, enabling heat dissipation in electronic packaging [207].

Bio-based Hybrids

New developments include:

- Lignin-coated wollastonite/hemp fiber hybrids that enhance UV resistance (85% retention after 500h weathering) for automotive exteriors [208]

- Chitosan-modified clay/wollastonite systems that impart antimicrobial properties (99.9% reduction in E. coli) for medical applications [209]

5. Conclusion

The comprehensive review of polypropylene (PP)-based hybrid composites highlights the significant strides made in enhancing the performance of conventional thermoplastics through reinforcement with both short fibers and inorganic fillers. The incorporation of materials such as glass fibers, wollastonite, talc, and nanofillers has proven instrumental in tailoring mechanical, thermal, and rheological properties to meet specific industrial requirements. Hybridization strategies, particularly those combining mineral fillers with short fibers, offer synergistic benefits including improved strength-to-weight ratios, dimensional stability, and cost efficiency.

Wollastonite has emerged as a key reinforcement due to its acicular shape, thermal stability, and compatibility with PP matrices, especially when used with compatibilizers like PP-g-MA. Additionally, advances in filler surface treatments, bio-based hybrids, and nanofiber integration have expanded the functional scope of PP composites to include self-healing, EMI shielding, and antimicrobial properties. Crystallization and rheological studies further underscore the importance of filler-matrix interactions in determining processability and final performance.

Despite these advancements, challenges such as filler dispersion, interfacial adhesion, and the trade-offs between toughness and stiffness persist. Continued research into novel hybridization methods, surface modifications, and sustainable filler alternatives will be crucial for developing next-generation multifunctional composites. As global demand for lightweight, high-performance, and recyclable materials increases—especially in automotive, construction, and electronics—PP-based hybrid composites are poised to play a central role in future material innovation.

6. Future Perspectives

Future research should focus on:

1. Advanced Surface Treatments: To enhance interfacial adhesion.
2. Sustainable Reinforcements: Natural fibers and bio-based fillers.
3. Computational Modeling: For predictive design of hybrid composites.

4. Cutting-Edge Characterization Techniques

4.1 In-situ Microscopy Advances

- Synchrotron X-ray microtomography now enables real-time visualization of wollastonite fiber debonding during tensile loading at 1000 fps [210]
- Atomic force microscopy-infrared (AFM-IR) reveals nanoscale chemical bonding at PP/wollastonite interfaces modified with novel ionic liquid compatibilizers [211]

5. Sustainable Processing Innovations

5.1 Green Manufacturing (2022-2024)

- Supercritical CO_2-assisted compounding reduces energy consumption by 30% while improving wollastonite dispersion [212]
- Recycled PP/wollastonite composites from post-consumer waste achieve 95% of virgin material properties through reactive compatibilization [213]

6. Emerging Applications

6.1 Energy Sector (Recent Breakthroughs)

- PP/wollastonite/graphene bipolar plates for fuel cells show 80% lower gas permeability than conventional materials [214]
- Phase-change composites with wollastonite/paraffin wax cores demonstrate 5× improved thermal cycling stability for building insulation [215]

6.2 Automotive Electrification

- Crash-resistant PP/wollastonite/carbon fiber hybrids for battery enclosures meet new UL 2596 safety standards [216]
- Triboelectric nanocomposites with surface-modified wollastonite enable self-powered sensors in smart tires [217]

7. Future Perspectives (2024 Onward)

7.1 AI-Driven Material Design

Machine learning algorithms now predict optimal hybrid compositions with 92% accuracy, reducing development time by 70% [218]

7.2 Circular Economy Solutions

- Enzymatic degradation systems for PP/wollastonite composites achieve 80% depolymerization in 48 hours [219]
- Blockchain-enabled tracking ensures 100% traceability of recycled content in automotive parts [220]

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