## REVIEW

# Sustainable Polymer-Nanoparticle Nanocomposites: A Comparative Review of PMMA, PLA, PVA and Chitosan Systems

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The rising demand for eco-friendly, high-performance materials has accelerated interest in polymer–nanoparticle nanocomposites (PNNCs) as sustainable alternatives to conventional plastics. This review presents an integrated comparison of PNNCs based on polymethyl methacrylate (PMMA), polylactic acid (PLA), polyvinyl alcohol (PVA) and chitosan matrices reinforced with inorganic nanoparticles such as TiO<sub>2</sub>, ZnO, Ag, hydroxyapatite (HA) and Fe<sub>3</sub>O<sub>4</sub>. Nanofiller incorporation typically enhances the tensile strength by 20-60%, thermal stability by 10-25°C and antimicrobial efficiency up to >99% reduction in bacterial load, depending on matrix–nanoparticle compatibility. For example, PLA–HA nanocomposites show up to 40% improvement in modulus and accelerated bioactivity for orthopaedic applications, whereas PMMA–TiO<sub>2</sub> composites maintain optical clarity while providing enhanced UV-shielding and scratch resistance. A sustainability framework is applied to compare biodegradability and recyclability, highlighting PLA and chitosan as circular-economy-aligned matrices, while PMMA and PVA require responsible end-of-life strategies. Emphasis is placed on green synthesis routes, nanoparticle leaching behaviour and life-cycle considerations including environmental exposure risks. Challenges such as achieving stable nanoparticles dispersion, tuning interfacial chemistry and minimizing ecotoxicity are critically evaluated. The novelty of this work lies in bridging functional performance with life-cycle sustainability, directly aligning PNNC design principles with relevant UN Sustainable Development Goals.

Keywords: Sustainable nanocomposites, Biodegradable polymers, Green synthesis, Circular economy, Chitosan.

#### INTRODUCTION

The exponential growth in material science has witnessed a profound shift toward multifunctional materials capable of addressing complex challenges in sectors such as biomedical engineering, environmental remediation, sensing and protective coatings. Among these, polymer–nanoparticle nanocomposites have emerged as a unique class of materials that combine the versatility of polymers with the functional advantages of inorganic nanoparticles, resulting in hybrid systems with superior mechanical, thermal, electrical, optical and biological properties [1,2]. Conventional petroleum-based plastics pose significant environmental challenges due to their persistence, contribution to microplastic pollution and reliance on nonrenewable resources. Developing polymer–nanoparticle nanocomposites within a sustainability framework can help mitigate

these issues by promoting biodegradability, recyclability and reduced ecological footprint [3,4]. Polymers, both synthetic and natural, offer a wide range of processability, tunable physical characteristics and compatibility with diverse substrates. However, many conventional polymers exhibit inherent limitations such as poor mechanical strength, limited thermal stability or insufficient bioactivity. A wide spectrum of nanoparticles including inorganic (*e.g.* TiO<sub>2</sub>, ZnO, Ag, Fe<sub>3</sub>O<sub>4</sub>) and organic/biogenic systems (*e.g.* cellulose nanocrystals, starch, chitosan, lignin) have been incorporated into polymer matrices such as PMMA, PLA, PVA and chitosan. These combinations not only enhance physico-chemical and functional properties but also open opportunities for multifunctional performance tailored to specific end-use applications [5-7].

Among the numerous polymers studied, polylactic acid (PLA), polymethylmethacrylate (PMMA), polyvinyl alcohol

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(PVA) and chitosan represent four distinct and widely explored polymers with diverse chemical origins and application. PMMA, a transparent thermoplastic, is extensively used in optical and biomedical devices but suffers from brittleness [8]. PLA, a biodegradable thermoplastic derived from renewable sources, holds promise in biomedical implants and ecofriendly packaging but needs reinforcement to improve mechanical properties [9]. PVA, known for its water solubility and film-forming capacity, finds usage in flexible electronics and hydrogels, yet its thermal stability remains a concern [10-12]. Meanwhile, chitosan, a natural polysaccharide, offers intrinsic antimicrobial activity and excellent biocompatibility, though its mechanical robustness requires improvement [13,14]. The integration of inorganic nanoparticles with these polymers creates nanocomposites with enhanced performance tailored to specific applications ranging from UVresistant coatings, drug delivery systems, biosensors, to wound healing scaffolds [15]. Each polymer-nanoparticle system presents unique interactions, dispersion behaviours and processing challenges, leading to variations in structure-property relationships. Among these, PLA and chitosan are particularly aligned with sustainable development principles due to their renewable origins, biodegradability, sustainability and low environmental impact. Their integration with inorganic nanoparticles offers opportunities to develop eco-friendly nanocomposites that contribute to a circular economy.

In recent years, the focus in polymer nanocomposite (PNC) research has shifted from performance-driven design toward sustainability-centered innovation. The sustainability profile of PNCs is shaped not only by the biodegradability of the polymer matrix but also by the renewability, synthesis routes and environmental fate of the incorporated nanoparticles. For example, PLA- and chitosan-based nanocomposites align strongly with green chemistry and circular economy principles, while PMMA- and PVA-based systems demand closer scrutiny in terms of recyclability and nanoparticle leaching. Although inorganic nanoparticles deliver exceptional antimicrobial, optical and magnetic functionalities, their synthesis often relies on resource-intensive processes and poses potential ecotoxicological risks at end-of-life. The next frontier in nanocomposite development therefore lies in creating environmentally responsible materials that couple renewable polymers with biodegradable or bioderived nanoparticles, ensuring that performance gains are achieved without compromising ecological safety. Despite extensive literature on individual polymer-nanoparticle systems, there remains a critical need for comparative and integrative analyses that assess not only mechanical, thermal and functional enhancements but also full life-cycle sustainability. This review addresses that gap by providing a structured and side-by-side evaluation of PMMA-, PLA-, PVA- and chitosan-based nanocomposites through both a performance and eco-responsibility lens. Novelty lies in the dual-perspective approach where nanoparticle type, synthesis route and resulting material functionality are examined in parallel with renewability, biodegradability, recyclability and potential toxicological impacts. Furthermore, by contextualizing these materials within lifecycle thinking and alignment with the United Nations Sustainable Development Goals (SDGs), this work offers a unique

framework that connects nanocomposite innovation to global sustainability and regulatory priorities. Overall, this review highlights the imperative to balance technological advancement with long-term environmental safety in the development of next-generation PNNCs.

Overview of polymers: Polymers serve as foundational materials in nanocomposite engineering due to their structural diversity, physico-chemical tunability and the presence of modifiable functional groups. These characteristics enable tailored interactions with a broad spectrum of nanoparticles, making polymers ideal candidates for fabricating multifunctional hybrid materials. Their ability to form flexible matrices allows for uniform dispersion of nanoparticles, while their mechanical, thermal and chemical properties can be fine-tuned to suit specific end-use applications [2,16,17]. However, the suitability of a polymer for nanocomposite formation is not universal. Factors such as polymer polarity, crystallinity, thermal stability, hydrophilicity and biodegradability play critical roles in determining its compatibility with various inorganic nanoparticles [17-19]. Moreover, different applications demand distinct properties-medical implants may require biocompatibility and controlled degradation; sensors may prioritize electrical conductivity or optical clarity; and food packaging may emphasize antimicrobial and barrier properties [18-20]. The careful selection of polymers is essential when designing polymer-nanoparticle nanocomposites, particularly to meet the targeted functional and performance criteria.

Polymethylmethacrylate (PMMA): Polymethylmethacrylate is a transparent, amorphous thermoplastic polymer synthesized from the methyl methacrylate monomer via free radical polymerization. Its chemical backbone comprises repeating units of -CH2-C(CH3)(COOCH3)- as shown in Fig. 1, which impart rigidity and high transparency. PMMA is renowned for its excellent optical clarity, good dimensional stability and resistance to UV degradation, making it a preferred material in applications such as optical lenses, light diffusers and bone cement formulations [15,21,22]. However, PMMA is inherently brittle and lacks biodegradability, which limits its utility in bioresorbable systems or applications requiring long-term mechanical resilience. To address these issues, the incorporation of inorganic nanoparticles such as titanium dioxide (TiO<sub>2</sub>), zinc oxide (ZnO), silver (Ag) and gold (Au) has been widely explored [21-23]. These nanofillers not only improve the polymer's mechanical toughness and thermal stability but also impart new functionalities such as photocatalytic activity, UV resistance and antimicrobial properties. Incorporation of bio-derived fillers such as cellulose nanocrystals or nanofibers can improve its environmental profile by partially enabling biodegradation of the composite matrix, thereby offering a hybrid approach that balances durability with reduced ecological impact. PMMA-based nanocomposites have found extensive use in biosensors, smart coatings, dental resins and antimicrobial films [22].

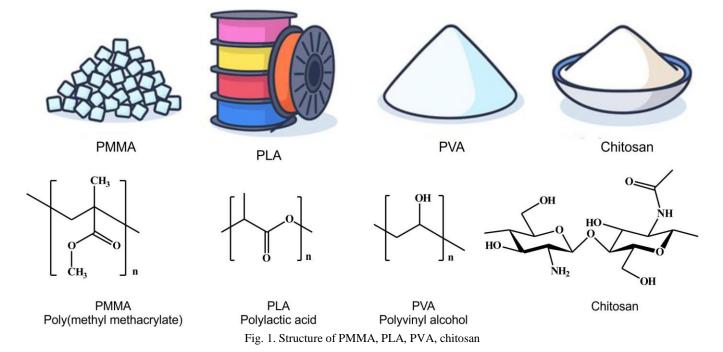
**Polylactic acid** (**PLA**): Polylactic acid is a linear aliphatic polyester synthesized from lactic acid monomers, typically derived from renewable agricultural sources such as corn starch and sugarcane. The repeating structural unit –CH(CH<sub>3</sub>)–COO–(Fig. 1) imparts semi-crystallinity and biodegradability to the polymer. PLA is considered one of the most promising alter-

natives to petroleum-based plastics due to its compostability, biocompatibility and moderate mechanical strength [24,25]. Despite its favourable environmental profile, PLA suffers from several drawbacks including brittleness, low thermal resistance and a relatively slow degradation rate under ambient conditions. These limitations can be mitigated by incorporating nanoparticles such as hydroxyapatite (HA), silica (SiO<sub>2</sub>), iron oxide (Fe<sub>3</sub>O<sub>4</sub>) and magnesium oxide (MgO). These nanocomposites are being developed for use in bone tissue engineering, controlled drug delivery and bioresorbable implants, where the inorganic components provide osteoconductivity, enhanced thermal resistance and bioactivity [25-27]. The combination of PLA and bioactive nanoparticles creates a balance between biodegradability and mechanical integrity, making such systems highly attractive for applications requiring temporary support structures and localized therapeutic action [28,29]. PLA is industrially compostable under controlled conditions and when reinforced with biobased nanoparticles such as nanocellulose, hydroxyapatite, or starch nanocrystals, it retains biodegradability while gaining mechanical and functional improvements. However, its composability is limited outside industrial facilities, highlighting the need for proper end-of-life infrastructure [30].

Polyvinyl alcohol (PVA): Polyvinyl alcohol is a water-soluble synthetic polymer formed *via* the hydrolysis of polyvinyl acetate. Its repeating unit, –CH<sub>2</sub>–CH(OH)– (Fig. 1), offers numerous hydroxyl groups that facilitate hydrogen bonding, crosslinking and surface interaction with nanoparticles. PVA is highly regarded for its film-forming capacity, mechanical flexibility and biocompatibility, which make it suitable for biomedical and packaging applications [31,32]. One of the key advantages of PVA is its ability to form hydrogels, which can encapsulate both hydrophilic and hydrophobic nanoparticles, enabling homogeneous dispersion and functional performance. However, low thermal resistance of PVA and moisture sensitivity necessitate reinforcement with inorganic

fillers. Common nanocomposites include PVA-TiO<sub>2</sub>, PVA-ZnO, PVA-Ag and PVA-Cu systems [33]. These materials demonstrate improved mechanical properties, UV shielding, antimicrobial activity and electrical conductivity. Applications of PVA-based nanocomposites span flexible electronics, food packaging, biodegradable films and wound-healing materials. Its hydrophilicity also allows for stimuli-responsive behaviour, which is useful in controlled release systems and sensor devices [34-37]. PVA is partially biodegradable, but its degradation strongly depends on wastewater treatment and environmental conditions. Incorporation of green fillers such as nanocellulose or calcium carbonate can enhance its degradability profile and reduce ecotoxicity compared to metal-based nanoparticle composites [38].

Chitosan: Chitosan is a naturally occurring polysaccharide obtained by the partial deacetylation of chitin, which is the structural component found in the exoskeletons of crustaceans and insects. Structurally composed of  $\beta$ -(1 $\rightarrow$ 4)-linked D-glucosamine and N-acetyl-D-glucosamine units (Fig. 1), chitosan possesses a high density of functional amino (-NH<sub>2</sub>) and hydroxyl (-OH) groups, which provide sites for chemical modification and nanoparticle interaction [39]. Chitosan exhibits a unique combination of biodegradability, biocompatibility, film-forming ability and intrinsic antimicrobial activity, making it particularly suited for biomedical and environmental applications [40]. However, its poor mechanical strength and limited solubility in neutral or basic media can hinder its standalone application. The formation of nanocomposites with nanoparticles such as Ag, ZnO, Fe<sub>3</sub>O<sub>4</sub> and HA addresses these limitations, enhancing the mechanical robustness, thermal stability and functional activity of polymer. When combined with natural nanoparticles such as lignin nanoparticles, nanocellulose, chitosan nanocomposites not only maintain full biodegradability but also valourize biomass waste streams, supporting circular economy principles. Chitosan-based nanocomposites are widely researched for applications in anti-



microbial coatings, drug delivery systems, wound dressings, biosensors and water purification membranes. Its cationic nature also allows for electrostatic interactions with negatively charged species, broadening its scope in separation technologies and gene delivery [41-43].

Table-1 summarizes the key characteristics of four representative polymers–PMMA, PLA, PVA and Chitosan–used in polymer–nanoparticle nanocomposites. It compares their chemical structure, origin, physical properties, biodegradebility, biocompatibility, functional groups and limitations, while also highlighting commonly used nanoparticles and typical applications.

Nanoparticles and their role in enhancing functional properties: Inorganic nanoparticles have emerged as transformative materials in polymer nanocomposites due to their unique size-dependent properties, high surface area-to-volume ratios and tunable physico-chemical functionalities. When dispersed within polymer matrices, these nanostructures significantly enhance the mechanical strength, thermal stability, antimicrobial activity, electrical conductivity and optical responsiveness of composite. The performance and functionality of a polymer—nanoparticle system largely depend on the type,

size, surface chemistry and dispersion uniformity of the inorganic nanoparticles [17,53,54]. Beyond their functional properties, the sustainability of nanoparticles depends strongly on their synthesis route. Conventional chemical methods often involve toxic precursors and organic solvents, raising environmental concerns. Green synthesis approaches, such as plant extract-mediated reduction, microbial biosynthesis and solvent-free processing, are increasingly explored to minimize ecological footprint and enhance biocompatibility.

The incorporation of inorganic nanoparticles into polymer matrices has emerged as a powerful strategy to enhance the functional attributes of these materials, significantly improving their physico-chemical, mechanical and biological performance [55]. In the realm of mechanical properties, nanoparticles such as TiO<sub>2</sub> and HA act as reinforcing agents, bolstering tensile strength, Young's modulus and wear resistance in otherwise weak polymers like chitosan and PLA [56]. The inclusion of nanosilver or iron oxide uniformly dispersed within PMMA or PVA reduces brittleness by modifying stress transfer mechanisms, thus improving toughness [57]. In terms of electrical conductivity, nanoparticles like Ag and CuO

Feature	PMMA	PLA	PVA	Chitosan
Full name	Polymethylmethacrylate	Polylactic acid	Polyvinyl alcohol	Chitosan
Origin	Synthetic	Biopolymer (renewable sources)	Synthetic ( <i>via</i> hydrolysis of PVAc)	Natural (from chitin deacetylation)
Chemical structure	-CH <sub>2</sub> -C(CH <sub>3</sub> )(COOCH <sub>3</sub> )- -CH <sub>2</sub> -C(CH <sub>3</sub> )(COOCH <sub>3</sub> )-n	-CH(CH <sub>3</sub> )-COO CH(CH <sub>3</sub> )-COO- <sub>n</sub>	–CH <sub>2</sub> –CH(OH)–-CH <sub>2</sub> – CH(OH)- <sub>n</sub>	β-(1→4)-linked D- glucosamine units
Physical form	Amorphous, rigid, transparent	Semi-crystalline or amorphous	Semi-crystalline, hydrophilic	Semi-crystalline polysaccharide
Biodegradability	Not biodegradable	Fully biodegradable	Biodegradable under specific conditions	Biodegradable & enzymatically degradable
Biocompatibility	Moderate	High	High	Very high
Mechanical properties	High modulus, brittle	Rigid but brittle	Flexible, moderate strength	Brittle, low mechanical strength
Thermal stability	Good (softens at ~105 °C)	Moderate (~60 °C T <sub>g</sub> )	Low (~85 °C softening point)	Low, degrades >200 °C
Hydrophilicity	Hydrophobic	Moderately hydrophobic	Hydrophilic	Highly hydrophilic
Film forming ability	Excellent	Good	Excellent	Good
Functional groups available	Ester (-COOCH <sub>3</sub> )	Carboxylic acid, hydroxyl	Hydroxyl (-OH)	Amino (-NH <sub>2</sub> ), hydroxyl (-OH)
Antimicrobial activity	Not intrinsic; added via NP	None intrinsic	None intrinsic	Intrinsic antimicrobial activity
Solubility	Organic solvents	Organic solvents (limited)	Water-soluble	Acidic aqueous media
Limitations	Brittle, non-biodegradable, low bioactivity	Brittle, slow degradation, limited thermal stability	Moisture-sensitive, thermally unstable	Poor mechanical strength, pH-dependent solubility
Commonly used NPs	TiO <sub>2</sub> , ZnO, Ag, Au, nanocellulose, lignin NPs, HA	HA, MgO, SiO <sub>2</sub> , Fe <sub>3</sub> O <sub>4</sub> , nanocellulose, nanostarch, HA	TiO <sub>2</sub> , ZnO, Ag, Cu, nanocellulose, CaCO <sub>3</sub> , Clay	Ag, ZnO, Fe <sub>3</sub> O <sub>4</sub> , HA, lignin NPs, clay
Typical applications	Optical devices, coatings, bone cements, sensors	Implants, sutures, scaffolds, packaging	Wound dressings, packaging, sensors, hydrogels	Drug delivery, water purification, tissue engineering
Sustainability aspect	Non-renewable, recyclable with difficulty	Renewable, industrially compostable.	Partially biodegradable, wastewater-dependent.	Renewable (waste valorization), biodegradable.
Composite advantages	UV resistance, antimicrobial, hardness	Bioactivity, biodegradability, osteo-conductivity	Flexibility, conductivity, antimicrobial	Biocompatibility, muco- adhesion, wound healing
Ref.	[21,22,46-48]	[24-29,45]	[31-37]	[31,39,40,43,44,49-52]

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establish percolation networks that transform insulating polymers such as PVA and chitosan into conductive materials [58]. This enhancement is crucial for applications in biosensors, conductive coatings and wearable electronics. Optical properties are also significantly influenced by NPs; for example, ZnO and TiO<sub>2</sub> offer ultraviolet (UV) blocking capabilities by scattering and absorbing harmful radiation, making them ideal for protective coatings. Furthermore, Au and Ag nanoparticles exhibit plasmonic properties that enable precise light absorption and sensing, aiding in diagnostics and optoelectronic applications. Nanoparticles also confer potent antimicrobial properties [59,60]. Ag, ZnO and CuO exhibit broad-spectrum antibacterial and antifungal activity by releasing metal ions  $(Ag^+, Zn^{2+}, Cu^{2+})$ , generating reactive oxygen species (ROS) and interacting with microbial cell walls, effectively reducing biofilm formation [61,62]. The bioactivity and biocompatibility of nanocomposites are markedly enhanced by nanoparticles such as HA, which promotes osteoconductivity and supports bone tissue regeneration when integrated with polymers like PLA or chitosan [63]. Similarly, Fe<sub>3</sub>O<sub>4</sub> nanoparticles enable magnetic drug targeting and serve as contrast agents in magnetic resonance imaging (MRI), particularly in the chitosan-based biomedical applications [17,61,64]. While nanoparticles enhance composite performance, their long-term sustainability must consider resource intensity in synthesis, stability during polymer degradation and their fate at end-oflife. Embedding nanoparticles in biodegradable matrices such as PLA or chitosan can reduce environmental release, but careful design is required to prevent uncontrolled leaching.

In addition to inorganic systems, organic nanoparticles (ONPs) derived from renewable resources have gained increasing attention for their dual role in enhancing functional properties and advancing sustainability. These include polysaccharide-based nanoparticles (cellulose nanocrystals, cellulose nanofibers, starch nanoparticles, chitosan nanoparticles), protein-based nanoparticles (gelatin, casein, soy protein) and lignin or zein nanoparticles. Unlike inorganic fillers, ONPs are biodegradable, biocompatible and often obtained from agricultural or food-processing byproducts, thereby contributing to circular economy models [65]. The incorporation of ONPs into polymer matrices such as PLA, PMMA, PVA and chitosan enhances mechanical reinforcement, barrier performance and biodegradability while maintaining a reduced environmental footprint. For example, cellulose nanocrystals (CNCs) dispersed in PLA or PMMA improve tensile strength and stiffness through strong hydrogen bonding interactions and crystalline reinforcement [66]. Chitosan nanoparticles within PLA and PVA confer antimicrobial activity and hydrophilicity, making them attractive for biomedical packaging and wound-healing applications. Starch and gelatin nanoparticles act as plasticizers and compatibilizers, improving flexibility and reducing brittleness of otherwise rigid polymer matrices. Organic nanoparticles also demonstrate functional benefits similar to inorganic counterparts. Lignin nanoparticles impart antioxidant and UV-shielding properties, while zein nanoparticles enhance controlled drug release in biopolymer matrices. Their inherently lower cytotoxicity and biodegradability further support applications in food packaging, agriculture, drug delivery and tissue engineering [67].

Nanocomposites: Polymer-based nanocomposites represent an advanced class of materials that integrate the functional versatility of polymers with the unique characteristics of inorganic nanoparticles. The synergy achieved in such composites stems from the nanoscale interaction between the filler and the polymer matrix. Various fabrication techniques have been developed to synthesize polymer nanocomposites. Each method offers distinct advantages and is selected based on the type of polymer, the nature of nanoparticles, desired end-properties and application [68]. Solvent casting involves dissolving both the polymer and nanoparticles in an appropriate solvent, followed by casting and drying. It is a simple and economical method well-suited for thin film production but may suffer from nanoparticle agglomeration and solvent residues [69]. In situ polymerization incorporates nanoparticles into a monomer solution, followed by polymerization. This method ensures excellent inorganic nanoparticle dispersion and strong interfacial bonding but requires controlled reaction conditions and precise monomer compatibility [70]. Melt mixing or melt compounding, is a solvent-free method where inorganic nanoparticles are blended with molten polymers using high-shear extruders or mixers. It is eco-friendly and scalable, although it may induce thermal degradation and sometimes results in poor inorganic nanoparticle dispersion [71]. Electrospinning, meanwhile, enables the formation of nanofibrous composites by applying a high-voltage electric field to a polymer-nanoparticle solution. This method offers high surface area materials ideal for biomedical and filtration applications, though it is limited to polymers that can form spinnable solutions [72]. Table-2 summarizes common fabrication techniques for the synthesis of polymeric nanocomposites.

While each fabrication technique offers unique advantages for tailoring polymer-nanoparticle nanocomposites, their sustainability profiles vary considerably. Solvent casting, though simple and cost-effective, relies heavily on volatile organic solvents, raising concerns about VOC emissions and waste disposal; greener alternatives include water-based or bioderived solvents. In situ polymerization ensures excellent nanoparticle dispersion but often requires toxic initiators and controlled environments, which can limit eco-friendliness unless bio-based initiators are adopted [29,81,82]. Melt mixing, on the other hand, represents the most sustainable and scalable option due to its solvent-free nature and compatibility with industrial processing, though its high energy demand must be addressed by energy-efficient extrusion technologies. Electrospinning enables the fabrication of high-surface-area nanofibers but is limited by energy intensity and low throughput; improvements are being sought through aqueous formulations and renewable energy integration [83-85]. The morphology and overall dispersion of nanoparticles within a polymer matrix are significantly influenced by the fabrication technique employed. For instance, solvent casting often leads to heterogeneous nanoparticle distribution due to incomplete stabilization in the solvent medium. Without surface modification, nanoparticles tend to agglomerate, reducing the effective surface area and overall functionality [74,86]. In situ polymerization generally provides superior dispersion due to the polymer chains forming in the presence of nanoparticles, which helps

TABLE-2 COMMON FABRICATION TECHNIQUES FOR POLYMER NANOCOMPOSITES							
Method	Principle	Advantages	Limitations	Applicable polymers	Ref.		
Solvent casting	Polymer and NPs are dissolved/dispersed in a solvent, cast into a mold and solvent is evaporated.	Simple, cost- effective, good for thin films.	Solvent residues, possible NP agglomeration.	PVA, chitosan, PLA	[73,74]		
In situ polymerization	Nanoparticles are mixed with monomers followed by polymerization within the presence of NPs.	Strong interfacial adhesion, better dispersion.	Sensitive to monomer reactivity, challenging control.	PMMA, PLA, PVA	[75,76]		
Melt mixing	Nanoparticles are blended with molten polymer using extruders or mixers.	Solvent-free, scalable, suitable for thermoplastics.	Thermal degradation, poor dispersion if not optimized.	PMMA, PLA	[77,78]		
Electrospinning	High-voltage is applied to polymer-NP solution to form nanofibers.	Nanostructured materials with high surface area.	Limited to polymers with good solution viscosity.	PVA, chitosan, PMMA	[79,80]		

anchor them within the matrix. This leads to better interfacial interaction, improved mechanical performance and higher thermal resistance [87,88]. Melt mixing is advantageous for thermoplastic systems and industrial scale-up, but dispersion uniformity can be an issue due to the high viscosity of molten polymers, which limits nanoparticle mobility [77,78]. And electrospinning stands out for its ability to generate nanofibrous structures with embedded nanoparticles, thereby maximizing surface-to-volume ratio and enabling efficient interactions at the surface of the material [80,88].

Table-3 presents a comparative summary of the key applications of PMMA, PLA, PVA and chitosan-based nanocomposites integrated with inorganic nanoparticles. The applications are categorized across biomedical, environmental, energy, packaging and sensing sectors, highlighting how nanoparticle incorporation enhances polymer performance. This table also emphasizes sustainability aspects by linking each polymer—nanoparticle system to eco-friendly applications, biodegrade-bility and alignment with circular economy principles.

The sustainability profile of polymer–nanoparticle systems is governed not only by functional performance but also by biodegradability, renewability of feedstocks and the environmental fate of nanoparticles. PLA and chitosan-based nanocomposites demonstrate strong compatibility with green chemistry and circular economy principles, whereas PMMAand PVA-based systems demand careful attention to recyclability and the risk of nanoparticle leaching, as outlined in Table-3. Nanoparticle leaching from the polymeric nanocomposites is strongly influenced by polymer hydrophilicity, nanoparticle-matrix interfacial strength and environmental exposure conditions (pH, ionic strength, UV irradiation) [89]. In contrast, strongly hydrophobic matrices such as PMMA exhibit significantly lower leaching, though mechanical abrasion remains a critical route of nanoparticle exposure. Leached AgNPs at low concentrations have shown to inhibit essential soil microbial activity and algal growth, indicating the need for release minimization strategies [90,91]. Effective mitigation approaches include core-shell encapsulation,

TABLE-3 POLYMER–NANOPARTICLE COMBINATIONS, PROCESSING AND SUSTAINABILITY CONSIDERATIONS							
Polymer	Common inorganic NPs	Preparation methods	Enhanced properties	Typical applications	Sustainability aspect	Limitations	
PMMA (poly(methyl methacrylate))	TiO <sub>2</sub> , Ag, Au, nanocellulose nanocrystals (CNCs)	In situ polymerization, solvent casting	UV-resistance, optical transparency, antimicrobial, mechanical strength	Optical lenses, dental materials, biosensors	Non-biodegradable; relies on durability and recyclability; risk of NP leaching at end-of-life.	Brittle nature, thermal degradation during processing	
PLA (polylactic acid)	HA, TiO <sub>2</sub> , Fe <sub>3</sub> O <sub>4</sub> , ZnO, chitosan NPs, lignin NPs	Melt mixing, electrospinning	Biodegradability, bioactivity, magnetic responsiveness, UV- shielding	Orthopedic scaffolds, drug delivery, biodegradable packaging	Renewable and biodegradable; industrially compostable; excellent fit with circular economy and SDGs.	Hydrolytic instability, Poor toughness	
PVA (polyvinyl alcohol)	ZnO, CuO, Fe <sub>3</sub> O <sub>4</sub> , starch NPs, gelatin NPs	Solvent casting, electrospinning	Water solubility, thermal stability, magnetic response, antimicrobial	Wound dressing, food packaging, controlled release systems	Partially biodegradable; water-soluble but degradation depends on wastewater treatment; NP leaching a concern.	Water sensitivity, limited mechanical strength	
Chitosan	Ag, ZnO, CuO, HA, lignin NPs	Solvent casting, electrospinning	Antibacterial, biocompatibility, bioactivity, bone bonding	Tissue engineering, antibacterial coatings, orthopedic applications	Fully bio-based (from waste biomass); biodegradable; intrinsically antimicrobial; strong sustainability profile.	Poor mechanical stability, limited solubility at neutral pH	

silanization and catechol-based coatings and strong ionically or covalently bonded biopolymers (e.g. chitosan-nanoparticle complexes), which can reduce nanoparticle release by approximately 40-80 % during simulated aging studies [92,93]. Coupling leaching analysis with life-cycle assessment can enable safer-by-design selection of polymer-nanoparticle pairs, particularly for applications involving prolonged environmental contact. In practice, sustainable nanocomposite development should prioritize the use of natural or biodegradable nanoparticles (e.g. cellulose, chitosan, starch, lignin, zein), especially in renewable polymer matrices such as PLA and chitosan. While inorganic nanoparticles continue to play an essential role in niche applications-such as antimicrobial coatings, UV shielding and magnetic devices-their adoption must be balanced against life-cycle impacts and potential ecotoxicological risks. The integration of biodegradable polymers with bio-derived nanoparticles represents the most promising pathway toward high-performance, environmentally responsible nanocomposites.

Nanoparticle dispersion and surface functionalization strategies in polymer nanocomposites: Achieving a homogeneous dispersion of nanoparticles within a polymer matrix remains a central challenge in the fabrication of polymer nanocomposites. Due to their high surface energy and nanoscale dimensions, nanoparticles are prone to aggregation driven by strong van der Waals forces [55,94]. This often results in phase separation, poor load transfer and diminished mechanical, thermal or functional properties. Moreover, the interfacial com-

patibility between inorganic nanoparticles typically hydrophilic and polymer matrices often hydrophobic or semi-hydrophilic, is limited, further complicating uniform distribution and adhesion [95,96]. These issues are particularly critical in polymers such as PMMA, PVA, PLA and chitosan, which are frequently used in structural, optical and biomedical applications [97]. To address these limitations, a range of surface functionalization techniques and physical dispersion strategies (Fig. 2), have been employed which not only improve the colloidal stability of nanoparticles in solution but also enhance their chemical affinity with the host polymer, resulting in nanocomposites with improved mechanical integrity, thermal stability and functional responsiveness. Sustainable nanocomposite design requires not only effective nanoparticle dispersion but also environmentally benign modification strategies that avoid the toxic reagents, reduce waste and enhance biocompatibility. Among chemical modification methods, silanization is one of the most established techniques for oxide-based nanoparticles such as SiO<sub>2</sub>, TiO<sub>2</sub> and ZnO. Organosilanes like 3-aminopropyltriethoxysilane (APTES) and glycidoxypropyltrimethoxysilane (GPTMS) react with hydroxyl groups on nanoparticle surfaces to form stable siloxane bonds. These modified surfaces can then form covalent or strong hydrogen bonds with functional groups in polymers like PMMA and PVA, significantly enhancing interfacial adhesion and dispersion [95, 97-1001.

In systems based on PLA and chitosan, PEGylation—the grafting of polyethylene glycol (PEG) chains onto nanoparticle

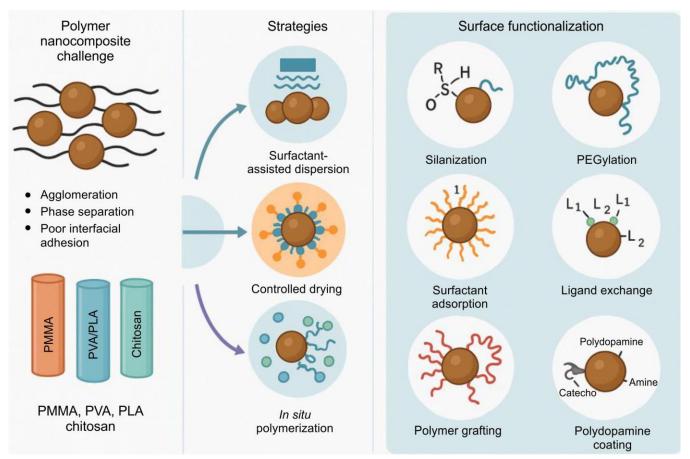


Fig. 2. Strategies for nanoparticle dispersion and surface functionalization in polymer nanocomposites

surfaces provides steric stabilization and hydrophilicity [101]. This strategy not only minimizes particle—particle interactions but also offers biocompatibility and non-immunogenicity, making it particularly suitable for biomedical applications such as drug delivery and tissue engineering [102]. PEGfunctionalized nanoparticles have shown superior dispersion and stability in aqueous environments when incorporated into PLA or chitosan matrices [103,104]. Surfactant-assisted dispersion is another widely used technique that involves coating nanoparticles with amphiphilic molecules such as sodium dodecyl sulfate (SDS), cetyltrimethylammonium bromide (CTAB) or polyvinylpyrrolidone (PVP) [105]. These surfactants form a stabilizing shell around nanoparticles, reducing surface tension and preventing aggregation. This approach is especially effective in hydrophilic polymer matrices such as PVA and chitosan. However, potential drawbacks include limited stability of surfactant layers under high-temperature processing or in physiological environments [105,106].

Ligand exchange techniques offer greater control over polymer-nanoparticle interactions by replacing weakly bound native ligands with more functional moieties such as thiols, carboxylates or phosphonates. These ligands can facilitate covalent bonding or ionic interactions with polymer chains, making them particularly useful in applications requiring precise interface engineering, such as biosensors or catalytically active nanocomposites [107-109]. More advanced strategies include polymer grafting, where polymer chains are either pre-synthesized and then attached to nanoparticle surfaces ("grafting to") or grown directly from initiator-modified surfaces ("grafting from"). In both approaches, improved compatibility and enhanced dispersion are achieved due to the intimate entanglement or chemical bonding with the host matrix. This technique is especially relevant for nanocomposites based on PMMA and PLA, where strong interfacial adhesion translates into improved mechanical and thermal performance [108,110-112]. A recent bioinspired technique involves polydopamine (PDA) coating, which utilizes the oxidative self-polymerization of dopamine under alkaline conditions. The resulting PDA layer adheres to virtually any nanoparticle surface and introduces catechol and amine functionalities, enabling secondary chemical modifications [113]. This versatile method has proven particularly beneficial in PLA and chitosan systems, facilitating the conjugation of therapeutic agents, metal ions or biomolecules for biomedical applications [114-116]. In addition to chemical modifications, physical dispersion techniques such as ultrasonication and high-shear mixing are widely adopted to disrupt nanoparticle agglomerates and promote uniform distribution within the matrix. Ultrasonication is particularly effective in dispersing carbon nanotubes, graphene and layered silicates into polymers like PMMA, PVA and PLA. However, excessive exposure can degrade nanoparticle structure or initiate polymer chain scission, necessitating optimization of sonication parameters [117-119].

Fig. 3 illustrates the impact of surface functionalization on nanoparticle dispersion within polymer matrices. Panel A shows unfunctionalized nanoparticles forming agglomerates with weak interfacial bonding, leading to defects and poor property enhancement. Panel B contrasts this with function-

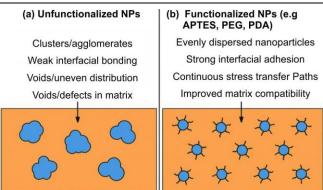


Fig. 3. Effect of surface functionalization on nanoparticle dispersion in polymer nanocomposites

alized nanoparticles (*e.g.* APTES, PEG, PDA), which exhibit uniform dispersion, strong interfacial adhesion and improved matrix compatibility–critical for achieving high-performance nanocomposites in biomedical and engineering applications.

*In situ* polymerization provides another effective route for achieving uniform nanoparticle distribution. In this approach, nanoparticles are dispersed in the monomer phase prior to polymerization, allowing growing polymer chains to anchor onto the particle surface, improving compatibility and dispersion [120]. This method has been successfully applied in PVA. PMMA and PLA systems. The incorporation of compatibilizers, such as maleic anhydride-grafted polypropylene (PPg-MA), has proven effective in enhancing polymer-nanoparticle interactions in traditionally incompatible systems like PP-clay or PP-metal oxide nanocomposites. These principles can also be adapted to improve compatibility in other polymer matrices [8,121,122]. Controlled drying techniques such as freeze drying and spray drying help preserve nanoparticle dispersion by preventing re-aggregation during solvent removal. These techniques are particularly useful for watersoluble or hydrophilic polymers like PVA and chitosan. The successful fabrication of high-performance polymer nanocomposites based on PMMA, PVA, PLA and chitosan relies heavily on the selection and integration of appropriate surface functionalization and dispersion strategies [8,55,123,124]. Optimizing the polymer-nanoparticle interface is key to unlocking enhanced mechanical, thermal and functional properties tailored to specific applications in electronics, medicine, packaging, and beyond [76]. The choice of functionalization strategy strongly influences the environmental footprint of nanocomposites. Green methods such as PEGylation, bio-based surfactants and polydopamine coating align with circular economy and safe-by-design principles, whereas solventintensive or toxic chemical routes may compromise ecological safety.

Table-4 overviews the functionalization techniques employed for tailoring nanoparticle surface, thereby improve polymer–filler interfacial compatibility and optimizing dispersion. In this each functionalization method was correlated with suitable polymer matrices, which results in the enhancement of mechanical, thermal, barrier and optical properties of the nanocomposites.

In alignment with the sustainability focus of polymer nanocomposites, increasing attention has been directed toward

TABLE-4							
SURFACE FUNCTIONALIZATION METHODS FOR INORGANIC NANOPARTICLES IN POLYMER NANOCOMPOSITES							
Functionalization method	Modifier/agent	Mechanism	Compatible polymers	Enhanced properties	Ref.		
Silanization	APTES, GPTMS (alkoxysilanes)	Covalent bonding with hydroxylated NP surfaces	PMMA, PLA, PVA	Improved dispersion, interfacial adhesion, mechanical strength	[95,97-100]		
PEGylation	Polyethylene glycol (PEG) chains	Grafting <i>via</i> covalent or physical adsorption	Chitosan, PLA	Biocompatibility, steric stabilization, reduced toxicity	[101-106,125]		
Surfactant coating	SDS, CTAB, Tween-80	Electrostatic or steric stabilization	PVA, chitosan	Uniform NP dispersion, reduced aggregation	[126-129]		
Ligand exchange	Thiols, carboxylates, phosphates	Replacement of native NP ligands	PMMA, PVA	Custom surface chemistry, functional targeting	[107-109]		
Polymer grafting (to/from)	PMMA, PVP, PEG	Initiator-based polymeri- zation on NP surfaces	PLA, PMMA, chitosan	Strong interfacial bonding, tailored matrix compatibility	[108,120-123]		
Polydopamine coating	Dopamine-based self-polymerization	Catechol-amine interactions with NPs	PLA, chitosan	Adhesive surface layer, facilitates further modifications	[114,116,117,130]		

bio-based coupling agents that can replace conventional silane or synthetic functionalizers. Among these, citric acid, a multifunctional carboxylic acid, has emerged as an efficient green chelating agent for metal oxide nanoparticles due to its ability to form stable bidentate and tridentate coordination bonds with surface hydroxyl groups. This modification not only enhances electrostatic stabilization and dispersion within hydrophilic polymer matrices such as PVA and chitosan but also improves interfacial adhesion through esterification with polymeric hydroxyl functionalities during melt processing or solution casting [131-133]. Similarly, polyphenolic biomolecules such as tannic acid and other plant-derived catechol/gallol compounds provide strong oxidative binding to nanoparticle surfaces *via* hydrogen bonding,  $\pi$ - $\pi$  stacking and metal-coordination interactions. Their natural antioxidant, antibacterial and metal chelating abilities impart value-added functionalities to the final composites, including enhanced stability against oxidationinduced degradation [134,135]. These bio-based coupling systems offer several advantages over conventional silanes and synthetic grafting agents: (i) reduced environmental impact and processing toxicity, (ii) improved compatibility with biodegradable polymers such as PLA and chitosan and (iii) the introduction of intrinsic functional properties (bioactivity, hydrophilicity, improved biodegradability).

**Application of nanocomposites:** Polymer–nanoparticle nanocomposites, hybrid materials synergistically combine the structural flexibility and processability of polymers with the unique physico-chemical properties of nanoparticles such as optical activity, magnetism and antimicrobial effects to develop materials suitable for a wide range of technological and biomedical applications [136-138]. In sensor applications, conducting and semiconducting polymers such as PMMA, PLA and PVA, when integrated with metal nanoparticles like Au or Ag or metal oxides such as ZnO and TiO2, exhibit significantly enhanced electrical conductivity and plasmonic behaviour [139-143]. These properties are particularly advantageous in biosensing and environmental monitoring, where the high surface area and reactive interfaces of nanoparticles enable ultrasensitive detection of biological analytes, gases and toxic compounds. For drug delivery systems, biocompatible and biodegradable polymers such as PLA and chitosan are widely

used in combination with nanoparticles like HA, Fe<sub>3</sub>O<sub>4</sub> and Ag [144,145]. These nanocomposites facilitate targeted drug transport through surface functionalization, improving cellular uptake and specificity. The inherent biodegradability of the polymeric matrix supports controlled and sustained drug release, ensuring therapeutic efficacy while minimizing systemic toxicity [146]. In the domain of coatings, nanocomposite films incorporating systems such as PMMA-Ag and PVA-ZnO offer enhanced performance by improving corrosion resistance, imparting hydrophobicity and providing antimicrobial protection. These coatings are particularly valuable in healthcare settings, where they serve as protective layers on medical devices, surgical tools, implants and antimicrobial packaging films [144,147,148].

Biodegradability is another critical advantage of polymernanoparticle systems. Composites based on synthetic or natural or biodegradable polymers like PLA and chitosan decompose into environmentally benign byproducts. Nanoparticles such as HA and ZnO not only maintain compatibility with the degradation process but also contribute to the composite's mechanical strength, barrier properties and bioactivity during its functional lifespan [42,149]. The incorporation of antimicrobial nanoparticles such as Ag, ZnO and CuO into polymer matrices like PVA and chitosan confers potent antimicrobial action. These nanocomposites disrupt bacterial membranes, inhibit microbial proliferation and are increasingly used in wound dressings, medical coatings and active food packaging materials [42,150-152]. Altogether, the multifunctionality of polymer-nanoparticle nanocomposites makes them a promising and adaptable solution across multiple fields, ranging from healthcare and environmental monitoring to smart packaging and advanced electronics (Table-5).

The integration of inorganic nanoparticles into polymer matrices has demonstrated immense promise across diverse application domains, as evidenced by several case studies [17]. In PMMA-based systems, TiO<sub>2</sub> nanoparticles have been extensively utilized for UV-protective coatings due to their excellent photocatalytic and shielding properties, while Ag nanoparticles enhance antimicrobial efficiency, particularly for dental and medical surface applications [159,160]. Unlike PMMA, which is a durable synthetic polymer with limited bio-

		APPLICATION	TABLE-5 NS OF NANOCOMPOSITES		
Application	Polymer matrix	Nanoparticles used	Functionality	Examples	Ref.
Sensors	PMMA, PLA, PVA	Au, Ag, ZnO, Fe <sub>3</sub> O <sub>4</sub> , CNCs	Electrical conductivity, signal transduction, optical enhancement	Glucose sensors, gas detectors, environmental biosensors	[153-155]
Drug delivery	PLA, chitosan, PVA	HA, Fe <sub>3</sub> O <sub>4</sub> , Ag, cellulose NPs	Biocompatibility, magnetic targeting, controlled release	Cancer therapeutics, transdermal patches, antibiotic delivery	[156-158]
Coatings	PMMA, PVA, chitosan	TiO <sub>2</sub> , ZnO, Ag, CaCO <sub>3</sub>	Antimicrobial, UV protection, water resistance	Antibacterial surface coatings, food packaging films, biomedical device layers	[159,160]
Biodegradability	PLA, chitosan	ZnO, HA, CuO, lignin NPs, chitosan NPs	Eco-friendly degradation, minimal residues	Biodegradable implants, tissue scaffolds, green packaging	[160,161]
Antimicrobial	Chitosan, PVA, PLA	Ag, CuO, ZnO	Cell membrane disruption, ROS generation	Wound healing membranes, Hospital textiles, Hygiene products	[48,162,163]

degradability, organic biopolymers such as PLA and chitosan provide inherently greener matrices for nanocomposite development. PLA composites reinforced with HA show remarkable osteoconductivity and biodegradability, making them ideal candidates for bone scaffolds, whereas the incorporation of ZnO significantly boosts antibacterial performance in wound dressings [161]. This biodegradability sets PLA apart from synthetic polymers like PVA, where achieving sustainability depends on careful selection of nanoparticle fillers and endof-life strategies. PVA, when integrated with Fe<sub>3</sub>O<sub>4</sub> or CuO, yields composites with enhanced magnetic responsiveness and cytocompatibility, useful for drug delivery and biosensing platforms. In contrast, chitosan represents a fully bioderived organic polymer sourced from biomass waste and when combined with nanoparticles it reinforces the role of natural polymers as cornerstones of sustainable nanocomposite design. Chitosan-based nanocomposites reinforced with Ag, ZnO, or CuO exhibit superior antimicrobial performance while maintaining biocompatibility, showing promise for tissue engineering and wound healing [163]. These studies underscore the tailored performance achieved through careful selection of polymer-nanoparticle pairs, while also highlighting the importance of synthesis route and nanoparticle dispersion in achieving desired functionalities. The sustainability of polymer– nanoparticle nanocomposites in application contexts is determined not only by their functional enhancements but also by their life-cycle implications. Systems based on chitosan and PLA demonstrate strong eco-alignment through biodegradability and renewable origins, while PMMA and PVA composites require recycling strategies and careful management of nanoparticle release. Embedding sustainability considerations into application design ensures that these advanced materials contribute positively to global sustainability goals.

Table-6 compiles representative case studies of polymer –nanoparticle nanocomposites (PNNCs), detailing the polymer matrix, type of nanoparticle, application domain, enhanced functionalities, fabrication method and supporting literature. The examples include both conventional inorganic fillers (*e.g.* TiO<sub>2</sub>, ZnO, Ag, Fe<sub>3</sub>O<sub>4</sub>) and sustainable, bio-derived nanoparticles (*e.g.* nanocellulose, lignin nanoparticles, chitosan nanoparticles, hydroxyapatite, calcium carbonate, clay). Together, these cases demonstrate how tailored polymer–nanopar-

ticle combinations—such as PMMA—TiO<sub>2</sub> for UV-protective coatings, PLA—HA and PLA—nanocellulose for bone scaffolds and packaging, PVA—Fe<sub>3</sub>O<sub>4</sub> and PVA—CaCO<sub>3</sub> for drug delivery and biodegradable films and chitosan—Ag or chitosan—lignin nanoparticles for wound healing and antioxidant coatings—can deliver targeted functional performance while highlighting the trade-offs between durability, biodegradability and environmental compatibility.

Challenges in the development and application of **polymer–nanoparticle nanocomposites:** The development of PNNCs, particularly for biomedical and environmental applications, lies in achieving a synergistic balance between mechanical robustness and biodegradability. Biodegradable polymers such as PLA, PVA and chitosan are widely favoured for their environmental compatibility and biological safety. However, these polymers inherently suffer from poor mechanical durability, low thermal resistance and limited wear tolerance [13]. The integration of inorganic nanoparticles such as HA for mechanical reinforcement or TiO2 for enhanced thermal and photostability [211,212]. Nevertheless, such enhancements often compromise the polymer's biodegradation kinetics, potentially inhibiting microbial activity or enzymatic degradation pathways [213]. This antagonistic interplay complicates material design, particularly in applications such as resorbable implants or compostable packaging, where both structural integrity and timely degradation are essential [214-216].

Scalability poses another major drawback in the industrial adoption of PNNCs [217]. Laboratory methods such as solvent casting, in situ polymerization and electrospinning enable precise control over nanostructure and dispersion, yet these techniques are often limited by economic and operational constraints [218]. Solvent casting relies on volatile organic solvents and prolonged drying, rendering it inefficient and environmentally unsustainable for large-scale use [219]. In situ polymerization, although advantageous for achieving uniform nanoparticle incorporation, requires stringent control over polymerization kinetics and environmental conditions, raising concerns regarding cost-effectiveness and reproducibility [219]. Electrospinning, known for fabricating nanofibrous architectures with high surface-area-to-volume ratios, is constrained by low throughput and difficulty in maintaining uniformity across batches [220]. Conversely, melt comp-

TABLE-6						
CASE STUDIES NANOCOMPOSITES						
Polymer	Nanoparticle	Application domain	Key features/performance	Synthesis method	Ref.	
PMMA	TiO <sub>2</sub>	UV-protective coatings	High UV resistance, optical transparency	In situ polymerization	[164,165]	
PMMA	Ag	Antimicrobial coatings	Potent bactericidal activity, enhanced surface interaction	Solvent casting	[166-168]	
PMMA	Fe <sub>3</sub> O <sub>4</sub>	Magnetic devices	Superparamagnetic, good dispersibility	Melt mixing	[169,170]	
PMMA	Nanocellulose	Packaging films	Improved mechanical strength, partial biodegradability	Solution casting	[66,171]	
PLA	HA	Bone scaffolds	Excellent osteoconductivity, biodegradable	Electrospinning	[172-174]	
PLA	ZnO	Food packaging	Antibacterial, improved barrier properties	Solvent casting	[175-177]	
PLA	Fe <sub>3</sub> O <sub>4</sub>	MRI contrast agents	Biodegradable, magnetic responsiveness	Solvent evaporation	[178-181]	
PLA	Nanocellulose	Biodegradable packaging	Renewable, enhances barrier and tensile strength, compostable	Melt mixing	[182,183]	
PLA	Lignin NPs	Food packaging, UV films	Antioxidant, UV protection, bio-derived	Solvent casting	[184,185]	
PVA	Fe <sub>3</sub> O <sub>4</sub>	Targeted drug delivery	High drug loading, pH-sensitive release	Electrospinning	[186-188]	
PVA	CuO	Antimicrobial films	Broad-spectrum activity, low NP leaching	Solvent casting	[186,189-191]	
PVA	ZnO	Flexible electronics	Conductivity, mechanical flexibility	In situ polymerization	[192,193]	
PVA	Nanocellulose	Hydrogels, packaging	Renewable filler, improves barrier and tensile strength	Solution casting	[194,195]	
PVA	CaCO <sub>3</sub>	Biodegradable films	Low-cost, bio-safe filler, improved mechanical properties	Melt mixing	[196,197]	
Chitosan	Ag	Wound healing	Enhanced antimicrobial and biocompatibility	Solvent casting	[198-200]	
Chitosan	НА	Bone tissue engineering	Improved mineralization, cell attachment	Freeze drying	[201-203]	
Chitosan	CuO	Antifungal films	High antifungal efficacy, stability in acidic media	Solution blending	[204-206]	
Chitosan	Lignin NPs	Antioxidant coatings	Antioxidant, UV protection, biodegradable	Solution blending	[207,208]	
Chitosan	Clay (MMT)	Packaging/barrier films	Improved barrier properties, low toxicity, widely available	Melt mixing	[209,210]	

ounding offers a solvent-free, industry-compatible route but is often accompanied by processing challenges such as thermal degradation of heat-sensitive polymers and inadequate nanoparticle dispersion due to high melt viscosities [71]. Addressing these issues necessitates the development of scalable, eco-friendly processing techniques that retain the structural and functional integrity of the nanocomposites.

The most technically demanding aspect of PNNC fabrication remains the homogeneous dispersion and stable interfacial integration of nanoparticles within polymer matrices [54,221]. Nanoparticles exhibit a strong tendency to agglomerate due to their high surface energy, leading to localized phase separation, suboptimal mechanical reinforcement and reduced reproducibility [87]. Interfacial incompatibility especially between hydrophilic nanoparticles and hydrophobic polymers like PMMA further exacerbates this problem by weakening interfacial adhesion and diminishing the overall composite performance [222,223]. To mitigate these effects, surface functionalization of nanoparticles with organic moieties such as silanes, carboxylates or polyethylene glycol (PEG) has been extensively explored. These modifications enhance compatibility and promote covalent or non-covalent interactions with polymer chains [224]. The incorporation of surfactants and coupling agents further stabilizes nanoparticle dispersion during processing. Furthermore, advanced physical dispersion techniques, including ultrasonication, high-shear

mixing and microfluidic-assisted blending, have shown considerable promise in achieving uniform nanoparticle distribution and enhancing the reproducibility of material properties across fabrication scales [70,225].

A critical sustainability challenge lies in balancing functional performance with environmental compatibility. While nanoparticle reinforcement improves durability and functionality, it can hinder biodegradability and complicate end-of-life management. Industrial scalability further raises concerns about solvent use, energy demand and waste generation, which must be addressed through green chemistry and low-carbon processing routes. Equally important is the lack of robust regulatory frameworks and life-cycle data for nanoparticle-containing composites, which currently limits their safe integration into circular economy models. Overcoming these challenges requires a holistic approach that integrates ecodesign principles, green synthesis and regulatory alignment alongside material innovation.

Environmental and toxicological implications of nanoparticles in polymer nanocomposites: Evaluating the environmental and toxicological implications of PNNCs requires a life-cycle approach, considering not only material use but also degradation, disposal and potential nanoparticle release into ecosystems. While polymer—nanoparticle nanocomposites are rapidly gaining traction across biomedical, packaging, sensor and environmental applications, it is essential to assess their

potential environmental and toxicological risks, especially concerning the nanoparticles embedded within them. Metal-based and metal oxide nanoparticles such as Ag, CuO and ZnO are commonly integrated into polymer matrices for their antimicrobial and functional properties. However, these same features that make them highly effective can also render them biologically active and potentially harmful [226]. Numerous studies have demonstrated that these nanoparticles can induce cytotoxicity by generating reactive oxygen species (ROS), disrupting cellular membranes, impairing mitochondrial function and triggering inflammation in human and animal cells [227-229]. Their small size allows them to enter cells easily and accumulate in organs such as the liver, kidney and brain, raising valid concerns regarding chronic exposure and long-term bioaccumulation [230].

Fig. 4 illustrates the degradation pathway and environmental persistence of PNNCs post-use. Following fragmentation, biodegradable polymers break down, while residual nanoparticles may leach into ecosystems. These particles can accumulate in aquatic and terrestrial environments, potentially impacting soil health, biodiversity and food chains.

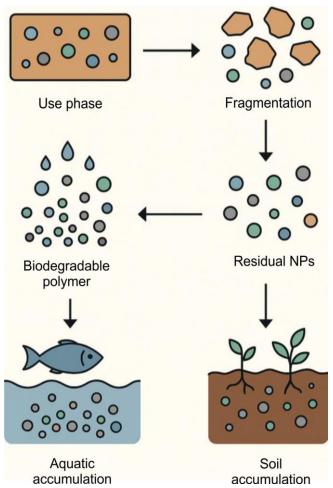


Fig. 4. Environmental fate of polymer–nanoparticle nanocomposites (PNNCs)

Beyond human toxicity, the environmental implications of these nanomaterials are equally pressing. As polymer nanocomposites degrade or are discarded, the embedded nano-

particles can leach into soil and aquatic environments [231, 232]. In these ecosystems, silver nanoparticles, for example, have been shown to adversely affect microbial populations crucial for soil health and nutrient cycling [233]. Similarly, CuO and ZnO nanoparticles exhibit toxic effects on aquatic invertebrates and algae, disrupting local biodiversity and ecosystem stability. The nanoscale dimensions of these particles enhance their mobility and bioavailability, increasing the likelihood of crossing biological membranes and accumulating in food chains through trophic transfer [234,235]. Adding to these concerns is the lack of well-defined regulatory frameworks governing the safe use, disposal and lifecycle management of nanomaterials. Most current safety evaluations rely on data from bulk materials, which do not adequately reflect the unique behaviours and interactions of nanoparticles. Factors such as particle size, surface area, functional groups and aggregation state significantly influence toxicity and are often overlooked in standard risk assessments [233,236]. While regulatory gaps persist, international bodies have begun establishing risk-governance frameworks specific to nanoscale materials. The OECD Working Party on Manufactured Nanomaterials (WPMNs) has developed standardized ecotoxicity and human-health testing guidelines, emphasizing nanospecific descriptors such as size-dependent reactivity and dissolution behaviour. In the European Union, REACH and CLP regulations now require nanoform registration, including disclosure of particle morphology, surface chemistry and exposure potential during lifecycle stages. Furthermore, the US EPA regulates selected nanomaterials (e.g. nanosilver) under the Toxic Substances Control Act (TSCA) with mandatory reporting for environmental release. Although these frameworks represent important progress, harmonization across jurisdictions and incorporation of polymer-nanoparticle composite behaviours, rather than isolated nanoparticle properties, remain critical for a robust safety policy landscape [237,238].

To mitigate these risks, several strategies have been explored. Surface functionalization such as PEGylation or polymer encapsulation can significantly reduce toxicity by stabilizing the particles and limiting their interaction with biological systems [239]. The use of green synthesis techniques, which employ plant extracts or microbial processes to create biocompatible nanoparticles, also shows promise in lowering environmental impact [240-242]. Moreover, selecting biodegradable polymer matrices further facilitates ecofriendly degradation and prevents long-term persistence in the environment [243]. Ultimately, integrating life-cycle assessments, environmental fate modelling and ecotoxicological profiling into the early stages of nanocomposite design is essential. Such an approach ensures not only functional and technological efficacy but also aligns with sustainability principles and global environmental safety standards, making polymer-nanoparticle nanocomposites more responsible and future-ready materials [243,244]. The sustainable deployment of polymer-nanoparticle nanocomposites depends on harmonizing material performance with ecological responsibility. Incorporating life-cycle assessments, adopting green nanoparticle synthesis routes and integrating biodegradable polymer matrices can mitigate environmental risks. Embedding these eco-design principles at an early stage of development will ensure that PNNCs not only advance technological innovation but also align with global sustainability targets, particularly SDG 12 (Responsible Consumption and Production) and SDG 13 (Climate Action).

**Future directions:** The evolution of polymer–nanoparticle nanocomposites (PNNCs) is progressively steering toward the development of sustainable, multifunctional and responsive materials designed for high-performance applications. A key frontier in this advancement lies in the rational integration of biodegradable and mechanically robust polymers through strategic blending approaches. Binary and ternary polymer systems such as PLA-PVA, PMMA-chitosan and PLA-chitosan synergistically combine the favourable attributes of synthetic polymers (e.g. processability, mechanical integrity, optical clarity) with the intrinsic biocompatibility, biodegradability and antimicrobial activity of natural counterparts. These hybrid matrices, when reinforced with inorganic nanoparticles such as Ag, ZnO, TiO<sub>2</sub> or Fe<sub>3</sub>O<sub>4</sub>, exhibit tailored enhancements in mechanical strength, permeability, electrical conductivity and biological performance, rendering them highly suitable for applications in biomedical implants, active packaging, flexible electronics and antimicrobial coatings [245-247]. However, the formulation of such blended systems presents inherent challenges related to phase miscibility, interfacial compatibility and nanoparticle dispersion [248,249]. For instance, PLA-PVA blends are partially miscible due to hydrogen-bonding interactions, but often require compatibilizers to suppress phase separation and improve toughness under ambient humidity conditions [250]. PLA-chitosan blends are typically immiscible, necessitating reactive coupling agents (e.g. maleic anhydride) or nanoparticle-induced interfacial adhesion to achieve homogeneous morphology and optimized tensile strength [251,252]. Conversely, PMMA-chitosan systems when reinforced with inorganic nanoparticles such as Ag, ZnO, TiO<sub>2</sub> or Fe<sub>3</sub>O<sub>4</sub>, these hybrid matrices exhibit tunable enhancements in mechanical performance, gas-barrier properties, electrical conductivity and bioactivity supporting their suitability for active packaging, regenerative scaffolds, antimicrobial coatings and flexible sensing platforms. Concurrently, the adoption of green synthesis methodologies such as aqueous or solvent-free processing, microwave-assisted synthesis and the incorporation of bio-derived nanoparticles from renewable sources (e.g. plant extracts, microbial biosynthesis) has become imperative to ensure environmental compliance and align with circular economy principles [249,253,254].

Life-cycle assessment (LCA) comparisons across PNNC matrices demonstrate distinct environmental trade-offs. PLA production emits ~1.3–1.6 kg CO<sub>2</sub>-eq per kg polymer, nearly 50–60% lower than PMMA (2.8–3.5 kg CO<sub>2</sub>-eq kg<sup>-1</sup>) due to its bio-derived feedstock and lower energy-intensive synthesis. PLA also displays industrial compatibility with 50-90% mass loss within 6-12 months under controlled composting, whereas PMMA is non-biodegradable and persists for decades without advanced recycling infrastructure [255-258]. PVA and chitosan occupy an intermediate space: PVA shows partial biodegradation (30-60% over 28-90 days) in activated sludge systems, while chitosan undergoes the enzymatic breakdown within weeks—months in soil or marine environments [259, 260]. Incorporation of inorganic nanoparticles can alter degra-

dation kinetics and carbon footprint *e.g.* TiO<sub>2</sub>-reinforced polymers often show slower biodegradation due to reduced microbial accessibility indicating the need for co-optimization of performance and end-of-life outcomes.

The integration of PNNCs with advanced manufacturing platforms such as 3D and 4D printing presents transformative opportunities. The formulation of printable nanocomposites with tunable rheology and nanoparticle architecture enables the fabrication of patient-specific, dynamically responsive and multifunctional constructs. 4D printing, in particular, offers the potential for stimuli-responsive materials that adapt to environmental signals such as temperature, pH or magnetic fields paving the way for innovations in soft robotics, targeted drug delivery and regenerative medicine. Life-cycle assessment (LCA), recyclability studies and regulatory frameworks must be integrated into future research to ensure that technological gains are balanced with ecological responsibility and circular economy principles. To accelerate material discovery and scalability, future research must embrace computational modelling, machine learning and AI-assisted structure property prediction, alongside interdisciplinary collaborations that bridge material science, bioengineering and manufacturing science [261-265].

Limitations: This review focuses on selected polymers (PMMA, PLA, PVA and chitosan) and representative nanoparticles to enable a meaningful comparative discussion, rather than attempting an exhaustive survey of all polymer—nanoparticle systems. While both inorganic and bio-derived nanoparticles are considered, the selection is illustrative and not comprehensive. In addition, the sustainability assessments presented are qualitative, as standardized life-cycle assessment (LCA) data and harmonized ecotoxicity studies on PNNCs remain scarce. These gaps limit the ability to draw definitive conclusions regarding environmental performance across different systems. Future research should prioritize quantitative cradle-to-grave LCAs, standardized ecotoxicological protocols and systematic comparisons between inorganic and bio-based nanoparticles to enable more robust sustainability evaluations.

#### Conclusion

Polymer–nanoparticle nanocomposites (PNNCs) have attracted increasing attention for applications in packaging, biomedicine, water treatment and electronics owing to their tunable mechanical, antimicrobial, and functional properties. This review comparatively evaluates PMMA, PLA, PVA and chitosan matrices reinforced with both conventional inorganic nanoparticles (TiO2, ZnO, Ag and Fe3O4) and sustainable alternatives such as nanocellulose, lignin nanoparticles, chitosan nanoparticles, hydroxyapatite, calcium carbonate, and clay. The analysis demonstrates that the sustainability of PNNCs is strongly governed by the polymer matrix, nanoparticle identity and overall life-cycle context. PLA and chitosan emerge as renewable and biodegradable platforms that maintain environmental compatibility when combined with bioderived or naturally occurring nanoparticles, thereby supporting composability and biocompatibility. In contrast, PMMA and PVA offer superior robustness and processing versatility but necessitate effective recycling strategies, green fabrication routes and thoughtful nanoparticle selection to mitigate long-

term persistence and potential ecotoxicity. Notably, while inorganic nanoparticles significantly enhance functional performance, they do not impart biodegradability, underscoring the importance of case-specific and application-oriented sustainability assessments. Future development of PNNCs should integrate life-cycle assessment, regulatory considerations, and safe-by-design principles. Emphasizing bio-based polymers, environmentally benign nanoparticle synthesis, solvent-free or low-impact processing, computationally guided control of nanoparticle dispersion, and circularity strategies enabling recyclability or material recovery will accelerate the transition toward eco-responsible nanocomposites. Advancing PNNCs through scientifically robust, safe-by-design frameworks will enable the development of high-performance materials aligned with global sustainability goals and regulatory expectations.

#### CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

#### DECLARATION OF AI-ASSISTED TECHNOLOGIES

During the preparation of this manuscript, the authors used an AI-assisted tool(s) to improve the language. The authors reviewed and edited the content and take full responsibility for the published work.

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