Review Article

Degradable Green Polymers, Green Nanopolymers and Green Nanocomposites Derived from Natural Systems: Statistics and Headways

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Abstract

Nowadays, actively researching and developing degradable green materials are efficient means to move towards the future advanced technologies and industries. In this article, we review the state of the art in important aspects of degradable green polymers especially green nanopolymers from natural sources and derived nanomaterials. Consequently, the fundamentals, cataloguing and properties of degradable green polymers or green nanopolymers obtained from natural resources have been presented. Green nanopolymers and derivative green nanocomposites are natural degradable materials. In this article, we also deliver numerous technological applications of the degradable green nanopolymers and derived materials such as transient electronics, film/coating and membrane/packaging, environmental protection and sustainability, and biomedical applications. The resulting green nanocomposites have been found effective to resolve current ecological issues. Moreover, the challenges and future of the natural degradable green nanopolymers and green nanocomposites have been investigated. However, the research and advancement of technical degradable materials with industrial and commercial applications yet have a long way to go.

Keywords: degradable polymer; nanopolymer; green; nanocomposite; electronics; packaging; biomedical

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1 Introduction

Degradable polymer or green polymer is the material which may experience backbone cleavage [1]. Degradation of naturally derived polymers may occur by main chain cleavage through microorganisms and aerobic/anaerobic routes [2]. The most appropriate mechanism for the degradation of these polymers is the hydrolytic decomposition of backbone [3]. As a result, the chemical and physical characteristics of the polymers may deteriorate. Nanopolymers are actually nanostructured polymers with polymeric chain dimensions in nanometric range [4]. Green nanocomposites derived from natural degradable green nanopolymers have benefits of low cost, lightweight, sustainability and eco-friendliness [5]. Green nanocomposites have been designed using green nanopolymers and nanocarbon or inorganic nanoparticles [6]–[9]. In this regard, green synthesis approaches have also been used to prepare the green nanoparticle [10]-[15]. However, the green nanomaterials need to be synthesised in a controlled manner to further increase the final properties and end uses. Green nanocomposites derived from natural degradable green nanopolymers have been applied in the transient electronics, films, coatings, membranes, packaging, environmental/sustainability extents, and biomedical applications such as drug delivery, tissue engineering, and biodevices [16]-[19].

In short, in this review, we comprehensively describe fundamentals, types, features and methodological applications of the natural green nanopolymers and derivative green nanocomposites. The natural green nanopolymer based green nanocomposites have gained important modern research stance owing to exceptional properties. However, numerous challenges need to be overcome for applications of these materials in the industrial sectors. In this innovative review, green polymer, green nanopolymer and green nanocomposites have been surveyed for physical properties and applications. Numerous green and sustainable materials have been studied for green polymers and nanocomposites. Incidentally, indispensable features and the significance of green nanocomposites have been considered. In this context, some previous literature reports have been found on the green nanopolymers and green nanocomposites [20], however, the reported literature is not in an updated form to depict the current state of these materials. Moreover, there is hardly any focused review seen on green polymer and nanocomposites. Nevertheless, future developments in the field of green nanocomposites are not possible for related researchers before getting prior knowledge of recent literature. Accordingly, in this review, we cover significant literature found in the last five years. To the best of our knowledge, such a specific recent review on green nanocomposites has not been published in literature before with well-arranged recent literature and an outline.

2 Degradable Green Polymers From Natural Systems

Degradable green polymers can be defined as the materials with backbone undergoing chemical/physical degradation upon exposure to microorganisms, aerobic/anaerobic conditions, heat, light or other external environmental factors [21]. Degradable green polymers can be referred to as synthetic or natural, depending upon the origin [22]. The degradable polymers can be further classified into various types. Figure 1 demonstrates an overview of the cataloguing of degradable polymers.

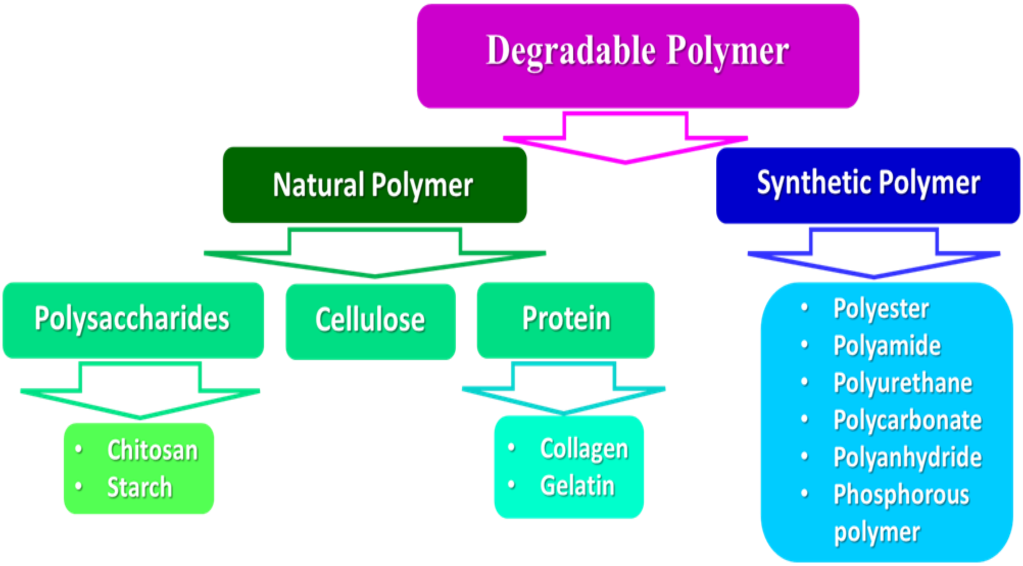

Figure 1: Cataloguing of degradable polymers

Table 1 details the main categories of degradable green polymers obtained from natural resources, types, degradation mechanism, and application areas. The degradable polymers derived from natural sources include polymers such as the polysaccharides [23], proteins [24], fats [25], and natural polymer blends. The degradable polymers from natural sources may undergo degradation through feasible mechanisms such as hydrolysis, enzymatic action, microbial action and environmental factors [26]–[28]. Natural degradable polymers (green) have been applied in important applications such as drug delivery [29], tissue engineering [30], pharmaceuticals [31], biomedical-related devices [32], packaging [33], agronomy [34], and eco-electronics [35]. Nevertheless, the degradation processes of the degradable polymers obtained from natural sources and the degradable polymers formed using non-natural sources are much different. The degradable polymers and naturally degradable polymers form separate categories of materials depending upon the degradation routes and degradation end products.

Table 1: Degradable natural polymers or green polymer

Polymer	**Illustration**	**Degradation mechanism**	**Application**
Polysaccharides	Starch Chitin Chitosan Cellulose	Hydrolysis Enzyme action Microbial action Environmental factors	Drug delivery Tissue engineering Biomedical appliances Packaging Pharmaceutical Eco-electronics Agronomy Others
Proteins	Animal sources Gluten Soy protein		
Fats	Lipids Triglycerides		
Natural polymer blends	Starch/cellulose/ chitin		

3 Degradable vs Biodegradable Polymer (Green Polymer)

With regard to sustainability, there are a myriad of related terms and terminologies [36]–[38]. The terms degradable polymer and biodegradable/naturally degradable polymer are frequently considered similar. There is always a curiosity to know the difference between degradable and biodegradable polymer [39]. Degradation refers to the decomposition process that usually involves the breakdown of polymer chains upon exposure to heat, light, moisture and other environmental factors [40]. The result of polymer degradation can be the environmentally harmful microscopic particles [41]. The polymer degradation has therefore been found to create more environmental pollution, rather than solving it. On the other hand, the biodegradable polymeric materials (or degradable polymers from natural sources) are either derived from natural sources or possess backbone materials which can be degraded naturally through bacteria, fungi, algae or other microorganisms [42]–[44]. The biodegradation process of the naturally derived materials results in the natural conditions leading to eco-friendly biomass with time [45], [46]. The biodegradable polymers therefore do not demand any non-natural processing route or chemical additives for their degradation [47]. Although the two terms biodegradable and degradable belong to degradable polymers, there are many differences in their mode of degradation and resulting by-products (Table 2).

Table 2: Difference between the degradable vs biodegradable polymer

Property	**Degradable polymer**	**Biodegradable polymer**
Origin source	Mostly synthetic	Natural sources
Degradation route	Requires external heat, light, moisture, chemicals, etc to degrade No microorganisms/oxygen needed for degradation	Microorganisms/oxygen

Property	Degradable polymer	Biodegradable polymer
Degradation by-products	Environmentally toxic microparticles	Biomass; enriches earth/atmosphere
Degradation time	Degrades over years or decades	Degrades in days to few weeks
Degradation course	Harmful to environment	Eco-friendly

4 Degradable Green Nanopolymers From Natural Sources: Origin, Types and Significance

4.1 Nanopolymers

Nanopolymers are actually a unique class of polymers, which can also be termed as the nanostructured polymers [48]. The nanostructural polymers have polymer chains with dimensions in the nanometric range [49]. A simple example of an inorganic nanopolymer is silica nanospheres [50]. The synthesis and properties of nanopolymers have been focused on in the literature [51]. The nanopolymers may have varying macromolecular architecture depending upon the nature of polymers and micro-level chain arrangement. The nanopolymers may be of different types such as the straight chain polymers, rod-like polymers, hyperbranched macromolecules, and polymer brushes [52], [53]. Various radical mechanisms have been proposed to form the nanopolymers using the functional monomers [54]. The nanostructure of polymers may affect the intrinsic macromolecular properties.

4.2 Natural Green Nanopolymers

Usually, small molecules such as monomers have been used to form the polymeric backbone. Natural green polymers are the substances which are obtained naturally [55]. Like other polymers, natural polymers are formed through condensation polymerisation or addition polymerisation. Natural green polymers are expansively found in nature and derived from natural sources [56], [57]. These polymers are usually extracted from natural sources using chemical, physical or mechanical routes. Naturally derivative green polymers are biocompatible. The polymeric network in these macromolecules characteristically have up to 99% water [58]. The fabrication, properties and applications of polymer nanostructures derived from natural resources have gained attention. The applications of natural nanopolymers have been found in technical fields such as eco-friendly materials, environmental remediation, and the biomedical sector [59]. A few important green nanopolymers include nanocellulose, nanostarch, nanochitin, nanochitosan, nanosilk, etc. (Figure 2).

Nanocellulose is a unique nanoderivative of cellulose. Nanocellulose has a unique combination of the cellulose chains on the nanoscale [60], [61]. Nanocellulose is a degradable and biocompatible green nanopolymer. The supramolecular cellulose structure has expanded research interest having nanocellulose fibres [62]. The nanocellulose has been extracted from wood, plants and bacteria through an in-situ process to obtain nanosised cellulose networks, nanofibres and nanostructures [63]. The nanocellulose nanostructures have been used in the biomedical devices such as bioimplants [64].

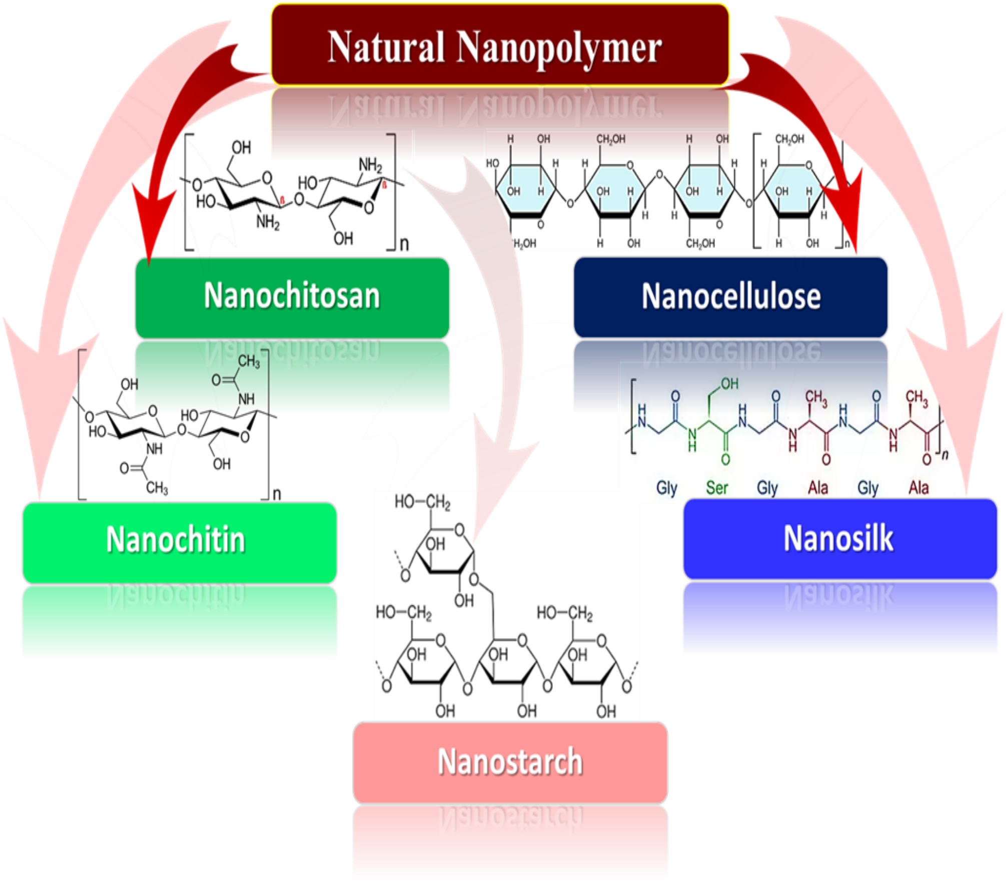

Figure 2: Nanopolymers from natural sources

Nanostarch is a nanopolymer of carbohydrate [65]. Nanostarch has a semicrystalline nanogranular nanostructure. Nanostarch is a unique green nanopolymer with unique, renewable, physiochemical and biodegradable properties [66]. However, nanostarch chains have a tendency to agglomerate, which hinders their processability. Consequently, a functional or modified form of nanostarch has been developed [67]. Functional nanostarch has better dispersion, hydrophobicity and stability, relative to the non-modified nanostarch [68]. The chemical functionalisation routes have been used to alter the nanostarch nanostructure [69]. Technological applications of nanostarch have been observed in the sustainable and beverage industries [70].

Nanochitin is a derived green nanopolymer from chitin [71], [72]. It is a degradable plus biocompatible nanopolymer. Initially, it was extracted by heating chitin in an alkaline medium. The resulting nanochitin was soluble in organic acid. [73]. Later, nanochitin has been obtained from other natural sources such as crustaceans, arthropods, molluscs, fungi and algae. The nanochitin has been obtained using various methods such as mechanical breakdown, acid hydrolysis, oxidation and electrospinning [74]. The nanochitin consists of nanofibre of ~2–5 nm diameter and ~300 nm in length [75]. Moreover, the nanochitin has remarkable dimensional, mechanical, optical, biological and biodegradability characteristics. In nanochitin, the nanofibres have been found associated by inter- and intramolecular hydrogen bonding.

Nanochitosan is a nanoform of chitosan, which is a carbohydrates polymer [76]. This green nanopolymer also has biocompatibility and degradability properties. Chitosan is an abundant natural polymer in microorganisms such as fungi, yeast, microalgae, insects, arthropods, crustaceans, molluscs and shrimps [77]. Chitosan has been formed by the alkaline acetylation of chitin (degree of deacetylation 60–100%). It has a linear semicrystalline structure [78]. The structural difference between chitosan and chitin is the presence of the N-acetylamine group in the backbone in chitosan [79]. Applications of nanochitosan have been investigated for use in beverages, eco-friendly materials, antimicrobial materials, tissue engineering, etc. [80]–[82].

Nanosilk is a biocompatible green nanopolymer formed from natural silk [83]. Nanosilk nanofibres have a high surface area, strength-to-density ratio and strain hardening. The common method to form nanosilk is the dissolution of silk fibroin in 85 wt.% phosphoric acid and coagulation in acetone [84]. Nanosilk has been prepared from the silk fibres by using pickering emulsion [85]. Nanosilk has been used in the biomedical sectors such as wound healing and antimicrobial applications [86], [87]. Various natural polymers have hence been derived from natural sources using simple extraction routes. Nevertheless, these simple polymers have found significant applications in technical fields.

5 Degradable Natural Green Nanopolymers to Green Nanocomposites

Polymers derived from natural sources or natural polymers have an eco-friendly nature. These polymers have fine processability, biodegradability and renewability characteristics [88]. In the nanocomposite technology, green nanopolymers derived from natural sources have been employed including nanochitosan, nanochitin, nanostarch, and several others [89]. This section explains various green nanocomposites based on natural source derived green nanopolymers. The application of different degradable green nanopolymers as polymeric matrix in the formation of the green nanocomposites has been discussed [90].

Nanochitin based green nanocomposites have been reported [91]. Barandiaran et al. [92] developed nanochitin and nanochitin nanocomposite using the enzymatic action. The nanochitin based green nanocomposite was formed using the graphene oxide and reduced graphene oxide nanofiller. Figure 3 shows the enzymatic action for the degradation of complex chitin structure into small nanochitin nanofibres.

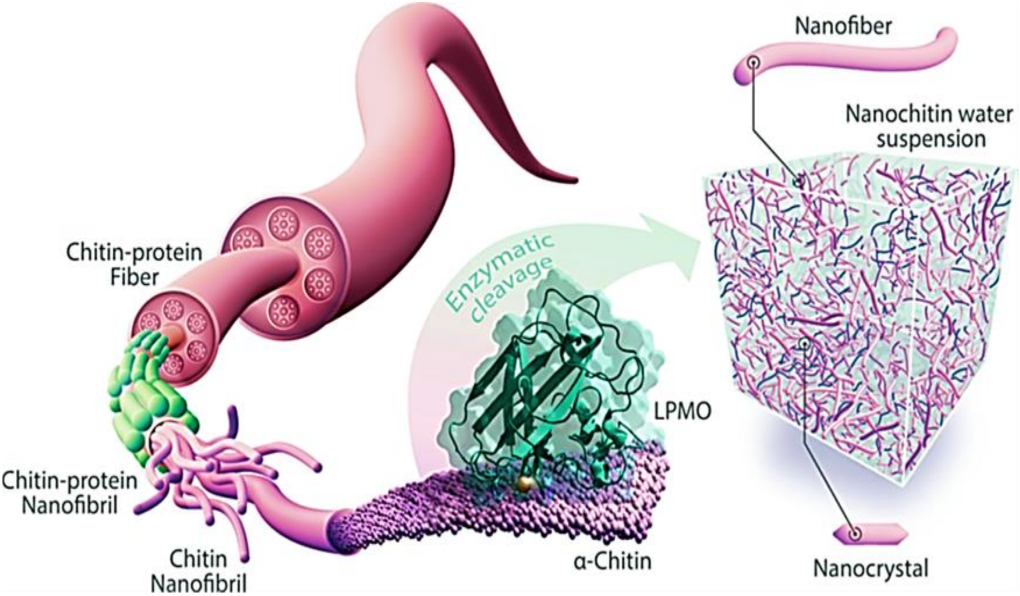

Figure 3: Illustration of α-chitin enzymatic oxidative cleavage
The hierarchical structure of chitin allows deconstruction into nanoscale particles. To obtain chitin nanoparticles enzymatic oxidative cleavage was performed in natural α-chitin. Lytic polysaccharide monooxygenases (LPMO) enzyme affinity to chitin fibres catalyses fibre degradations into smaller fibres. Chitin nanocrystals (CNCh) are small crystalline particles with lengths between 100 and 500 nm and diameters between 3 and 40 nm. (Reproduced from [92]with permission from Nature.)

The resulting enzymatic crystalline nanochitin produced (EnCNCh) was modified to obtain enzymatic crystalline nanochitin/reduced graphene oxide (EnCNCh/rGO). The EnCNCh and EnCNCh/rGO have been used to develop the hybrid conductive bioink. The EnCNCh based hybrid films with graphene oxide are shown in Figure 4. The electrical conductivity of the EnCNCh/rGO nanocomposites was measured and found to enhance with the nanofiller contents. The three-dimensional printing of EnCNCh with 5% nanofiller formed stable honeycomb such as scaffold. EnCNCh with 5% nanofiller was used to form printable conductive ink. The bioink was applied to dye the HEK293T cells. The nucleus and cytoplasm of the bioink dyed cells were studied using the fluorescence microscopy. The EnCNCh nanocomposites with graphene based nanofillers were successfully applied in bioprinting, biomedicine and tissue engineering.

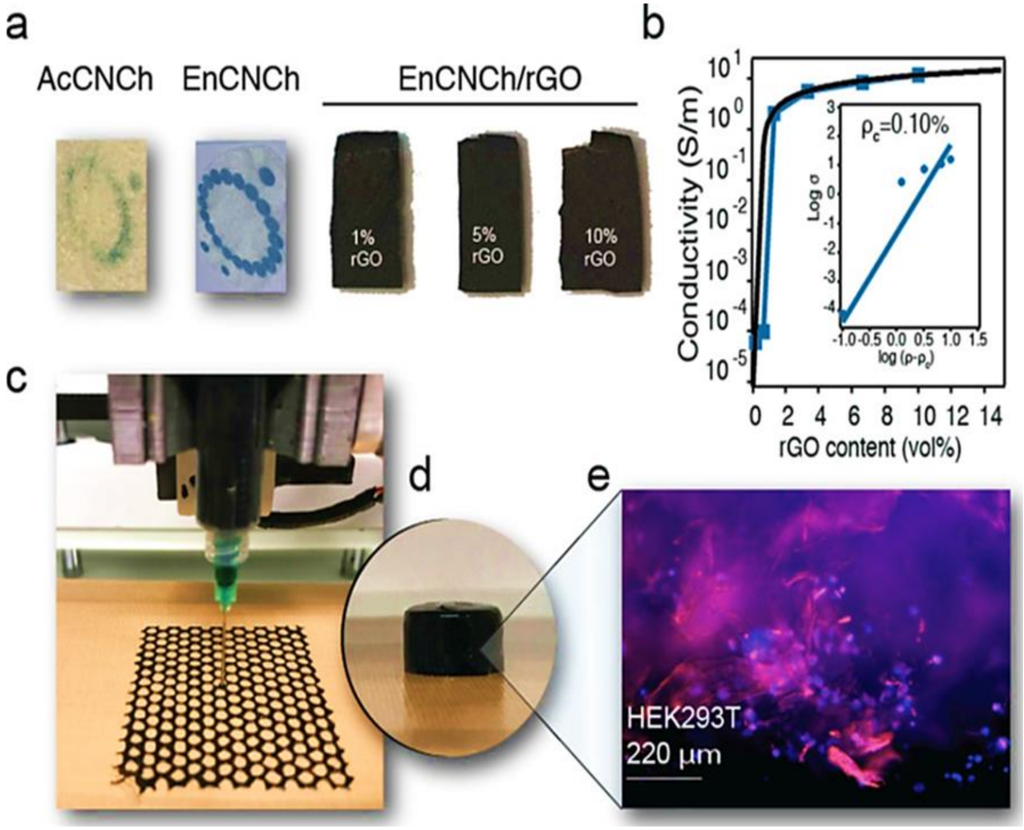

Figure 4: EnCNCh/rGO films fabrication: **(a)** Detail of CNCh films from left to right: AcCNCh film, EnCNCh film, EnCNCh film 1%wt. rGO, 5%wt. rGO and 10 wt. rGO
(b) Conductivity of EnCNCh for different reduced graphene contents: 0.3, 1, 2, 5, 10 and 15 wt.% rGO
(c) Image of 3D EnCNCh + 5% GO 3D printing of a stable honeycomblike scaffold
(d) Picture of a EnCNCh + 5% GO 3D printed bottom shape scaffold where HEK293 cells were grown
(e) Confocal microscopy image at X10 of HEK293T-stained nuclei with DAPI and orange plasma seeded and grown on the hybrid material
GO = graphene oxide; rGO = reduced graphene oxide; AcCNCh = acid treated crystalline nanochitin; EnCNCh = enzymatic crystalline nanochitin;
EnCNCh/rGO = enzymatic crystalline nanochitin/reduced graphene oxide;
DAPI = 4′,6’-diamidino-2-phenylindole) is a blue-fluorescent DNA stain (Reproduced from [92] with permission from Nature.)

Sahraee et al. [93] fabricated the nanochitin/zinc oxide nanoparticle based nanomaterial. The oxygen permeability of the nanochitin based nanocomposite was reduced to 29.645(cm^3µm.m^{-2} dkPa), relative to neat polymer film (39.262(cm^3µm.m^{-2} dkPa)). The nanomaterial was found useful in green packaging application. Li et al. [94] developed polyacrylamide and chitin nanowhisker-based nanocomposites. The nanomaterial was obtained in the form of a hydrogel with physical or chemical crosslinking. The nanocomposite hydrogel had tensile strength and compressive strength of 321.9 ± 8.2 kPa and 6.95 ± 0.20 MPa, respectively. Owing to high elasticity and mechanical properties, the nanomaterials have been used for the eco-friendly electronics. Nanochitin-based green nanocomposites have hence been successfully fabricated and employed in the advanced bioprinting, biomedical scaffolds, eco-friendly packaging, and eco-electronics applications.

Similar to nanochitin, nanochitosan has also been applied as a significant green nanopolymer in the green nanocomposites [95], [96]. Lee et al. [97] used the cryomilling technique to form the green nanochitosan/graphene nanocomposites. The cryomilled chitosan/graphene nanocomposites had higher mechanical and thermal profiles, relative to neat nanochitosan. The reason was fine graphene nanoparticle dispersion in the nanochitosan matrix causing a better reinforcing effect. Cobos et al. [98] adopted the in situ and solution-casting methods to develop the nanochitosan/graphene nanocomposites. The glycerol was used as the plasticiser. The green nanocomposites revealed superior substantially and improved mechanical and electrical properties due to fine graphene dispersion and interactions with the nanochitosan. The green nanomaterials were recommended for the sustainable and eco-electronics applications.

Another important category of green nanocomposites has been developed using the nanocellulose [99]. The nanocellulose-derived green nanocomposites have potential as renewable smart materials for technological purposes. Inclusion of graphene to green nanocomposites has further broadened their scope for multi-functional applications [100]. The formation of high performance green nanocellulose/graphene nanocomposite relies on the nanofiller dispersion, functionality and interaction with the nanopolymer. Malho et al. [101] extracted nanofibrillar nanocellulose for developing with multi-layered graphene nanocomposite. Inclusion of 1.25 wt.% graphene content in the nanofibrillar nanocellulose resulted in high ultimate strength and Young’s modulus of 351 MPa and 16.9 GPa, respectively. The mechanically strong green nanocomposites have found potential for strength applications. Song et al. [102] formed the nanocellulose-based microcrystalline cellulose nanofibre. The nanocellulose derivative was used to produce the microcrystalline cellulose nanofibre/graphene oxide (MSF-g-COOH/GO). The nanocomposite has hydrogen bonding interactions between the nanocellulose derivative and graphene oxide. The physical crosslinking initiated an interesting stimuli responsive effect. The MSF-g-COOH/GO nanocomposite depicted remarkable water-induced shape memory effect. Figure 5 shows the mechanism of shape memory MSF-g-COOH/GO nanocomposite. The nanomaterial presented a water-induced shape memory effect.

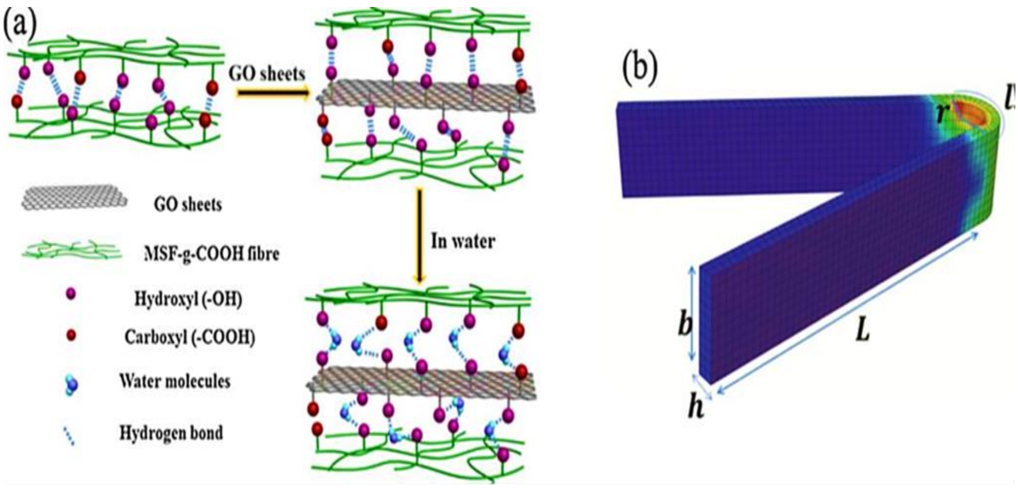

Figure 5: (a) Schematic of water-induced shape memory effect mechanism of MSF-g COOH/GO nanocomposite paper
(b) schematic of simulation dynamic processes of bending
MSF-g-COOH/GO = microcrystalline cellulose nanofibre/graphene oxide
(Reproduced from [102] with permission from Elsevier.)

Figure 6 depicts the shape recovery ratio as a function of immersion time for 0.5 wt.% nanofiller loaded MSF-g-COOH/GO nanocomposite. The shape recovery effect was found to improve with the immersion time. The shape recovery ratio was ~95% after 14 s immersion time. Moreover, the 0.5 wt.% MSF-g-COOH revealed an increment in tensile strength and Young's modulus from 139 to 184 MPa and 5.77 to 8.54 GPa, respectively, relative to neat MSF-g-COOH. The simulation dynamic-based water-induced model was studied for better understanding of the water induced shape memory behaviour of the green nanocomposites.

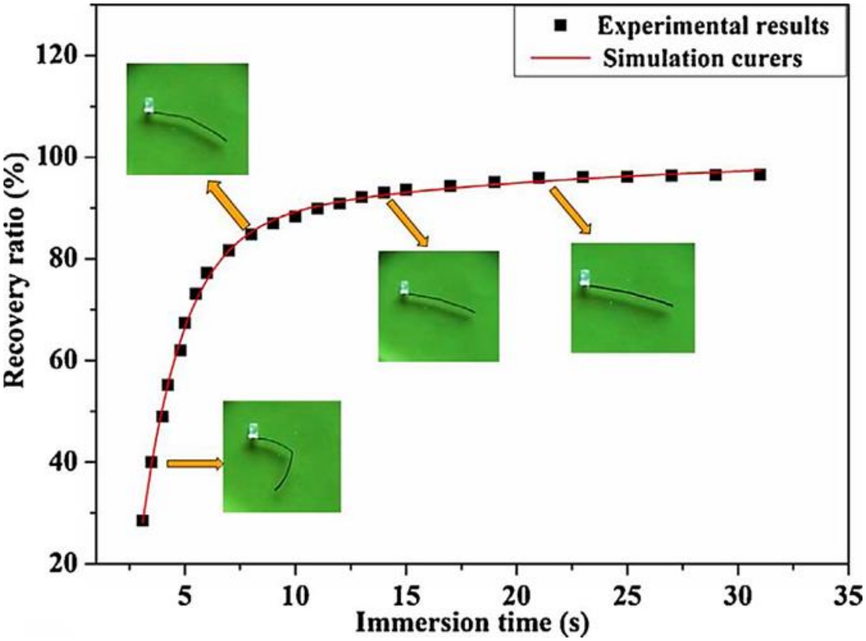

Figure 6: Shape memory behaviour of 0.5 wt.% MSF-g-COOH/GO nanocomposite paper immersed in water and corresponding simulation curve
MSF-g-COOH/GO = microcrystalline cellulose nanofibre/graphene oxide
(Reproduced from [102] with permission from Elsevier.)

Kabriri et al. [103] developed the green nanocrystalline cellulose acetate/graphene oxide nanocomposite using the facile solution route. Figure 7 shows the formation of the hydrogen bonding between the nanocellulose derivative and graphene oxide due to structural compatibility. Inclusion of 0.8 wt.% graphene oxide enhanced the tensile strength by 62% (157.49 MPa), relative to the neat nanocellulose. The increase in the mechanical properties of the nanomaterial was obviously due to better nanofiller dispersion because of matrix-nanofiller compatibility. Moreover, the nanocomposite had fine barrier properties to be employed in the green packaging application.

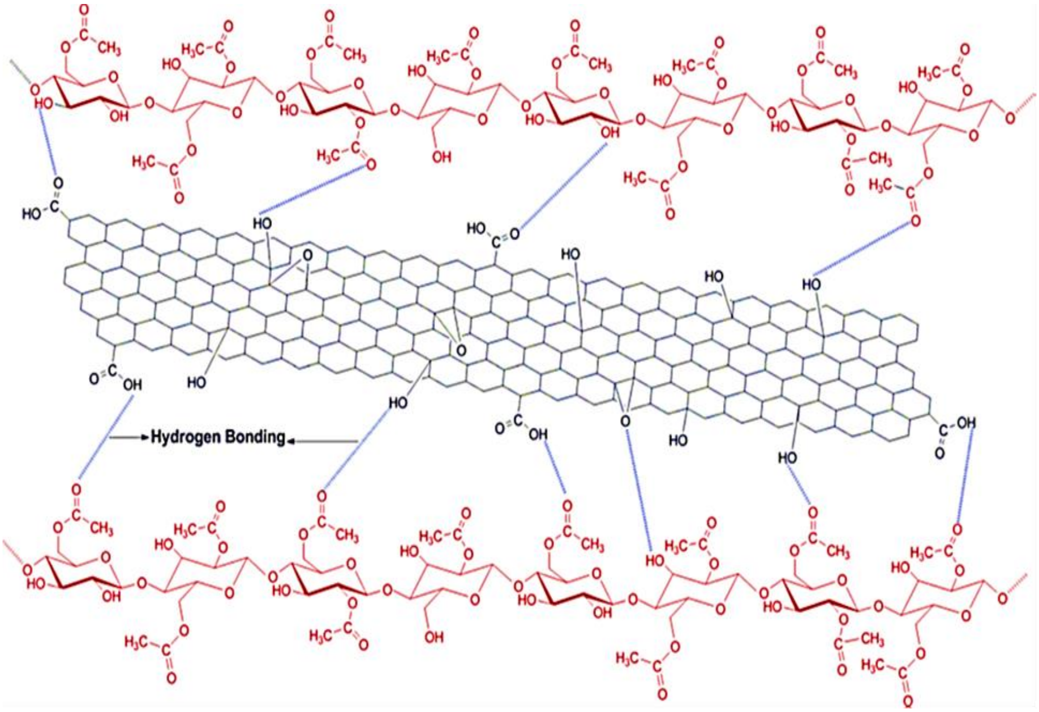

Figure 7: Schematic representation of NCCA/GO (nanocrystalline cellulose acetate/graphene oxide) nanocomposite (Reproduced from [103] with permission from Springer.)

Daniyal et al. [104] produced the green hexadecyltrimethylammonium bromide nanocrystalline cellulose-graphene oxide (CTA-NCC/GO) nanocomposite. The CTA-NCC/GO nanocomposite was tested for the binding affinity towards Ni^{2+} (0.01–0.1 ppm). Surface plasmon resonance analysis predicted high binding affinity constant of $1.620 \times 10^{3} M^{-1}$. Figure 8 shows the detection accuracy of CTA-NCC/GO nanocomposite towards the Ni^{2} ion in the range of 0.01–0.5 ppm. The detection accuracy pointed towards the efficient use of nanocellulose based nanomaterials in toxic ion sensing application. The nanocellulose and nanocarbon nanocomposites therefore have important applications in the eco-packaging, shape-memory materials, and biotechnological industries [105].

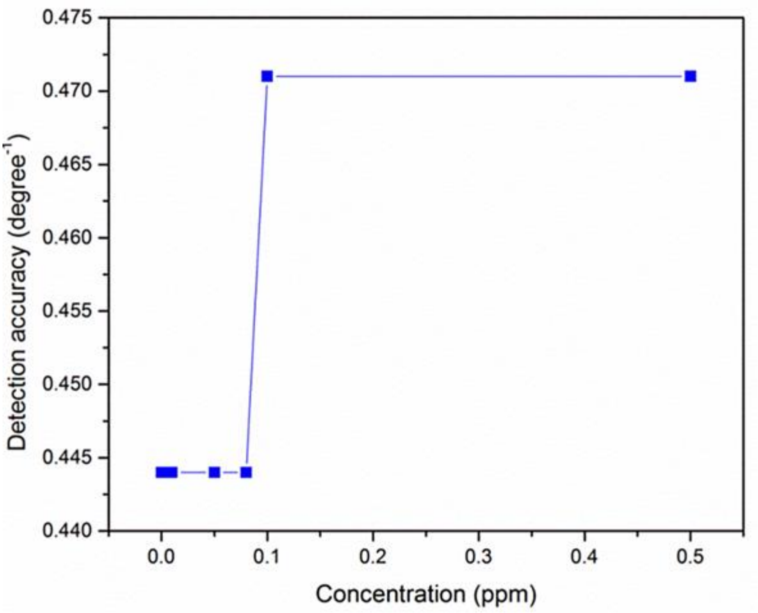

Figure 8: Detection accuracy for CTA-NCC/GO thin film in Ni^{2+} detection CTA-NCC/GO = hexadecyltrimethylammonium bromide nanocrystalline cellulose-graphene oxide nanocomposite (Reproduced from [104] with permission from Elsevier.)

Nanostarch-based green nanocomposites have also been reported [106]. Nanostarch nanocomposites have been used owing to their fine biocompatibility and biodegradability properties. Xu et al. [107] prepared the green nanostarch nanoparticles of ∼130 nm using the reversed phase titration method. The green nanostarch nanoparticles were used as nano-reinforcement in the rubber-grade ethylene-glycidyl methacrylate-vinyl acetate matrix. The rubber-grade ethylene-glycidyl methacrylate-vinyl acetate/nanostarch nanoparticles formed a cross-linked nanocomposite nanostructure. The nanocomposites have high strength and biomedical applications. Nasiri et al. [108] reported the eco-friendly nanostarch/silver nanocomposites and studied their catalytic activity. The catalyst nanoparticles were magnetically separable and recyclable. Sindhu et al. [109] studied the nanostarch based antibacterial packaging materials. Chen et al. [110] prepared the green nanostarch/cellulose nanocrystal nanocomposite. The nanostarch/cellulose nanocrystal nanocomposite with 6% starch content improved the tensile strength by 238%. The nanomaterial was used for the formation of green plant capsules. Poonguzhali et al. [111] fabricated the green nanostarch/chitosan/poly(vinyl pyrrolidone) derived nanocomposite. The nanomaterial had hydrophobic and bacterial antiadhesion properties. The green nanostarch/chitosan/poly(vinyl pyrrolidone) was applied for antibacterial wound healing. The nanostarch based nanomaterials have been used in antibacterial packaging, wound healing, and eco-friendly applications. However, more focused future efforts are needed to reveal full potential of the green nanostarch based nanocomposite.

Nanosilk usually occurs in the form of one-dimensional self-assembled silk fibroin or nanofibres [112]. The green nanosilk nanopolymer is used in drug delivery/tissue engineering [113], nanomedicine [114], biosensors [115], nanoelectronics, etc. [116]. These silk and nanocarbon nanocomposites have hierarchical nanostructures and tunable physical properties. Wang et al. [117] designed green nanocomposite based on the silk fibroin and graphene nanosheet. Inclusion of 0.5 wt.% graphene content revealed a high Young's modulus of 1.65 GPa, which is 5.8 times higher than the neat green nanopolymer. The reason seems to be the high surface area of nanosilk nanofibres and interactions with the nanofiller. The green nanosilk-based nanocomposites depict efficient drug storage/release and tissue engineering uses [118]. Balu et al. [119] formed the silk fibroin/graphene oxide nanocomposite hydrogel using photochemical cross-linking technique. The silk fibroin/graphene oxide nanocomposite hydrogel had a high Young's modulus of ∼8 MPa, which was much higher than the neat polymer (∼1.5 MPa). Figure 9 illustrates the silk fibroin/graphene oxide nanocomposite hydrogel.

The eco and biocompatible nanocomposite hydrogel has been found useful for tissue engineering application. Wang et al. [120] also reported the green silk fibroin/graphene oxide nanocomposite membranes. The silk fibroin and graphene oxide were linked through interfacial interactions. The nanofiller inclusion considerably enhanced the ultimate strength and Young's modulus to 41% and 75%, respectively, relative to neat nanopolymer. The nanocomposite was found useful in tissue engineering. The nanosilk-based green nanocomposites have been applied in the tissue engineering, drug deliveries, and other biomedical applications.

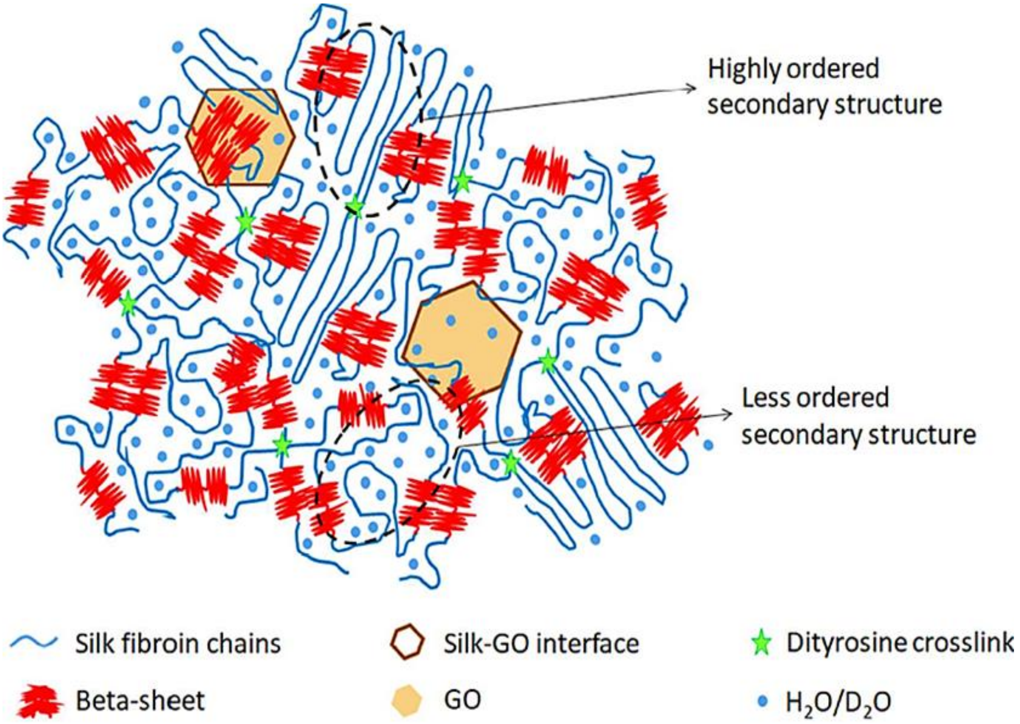

Figure 9: Schematic of silk fibroin/graphene oxide nanocomposite hydrogel nanostructure
Three structural domains: crystalline (containing β-sheet), mesophase (ordered secondary structure) and amorphous (containing random coil) are 4 nm, 6 nm and 30 nm, respectively (Reproduced from [119] with permission from ACS.)

6 Application Areas of Degradable Natural Green Polymers or Green Nanopolymers and Green Nanocomposite

Figure 10 shows possible application areas of the green nanopolymer-based nanocomposite, as discussed in this section. Various application areas have been observed for these nanomaterials, depending upon the green nanopolymer structure, type of nanoparticles, matrix-nanofiller interactions, and final nanocomposite properties. The application areas are vast; ranging from the electronics to biomedical fields.

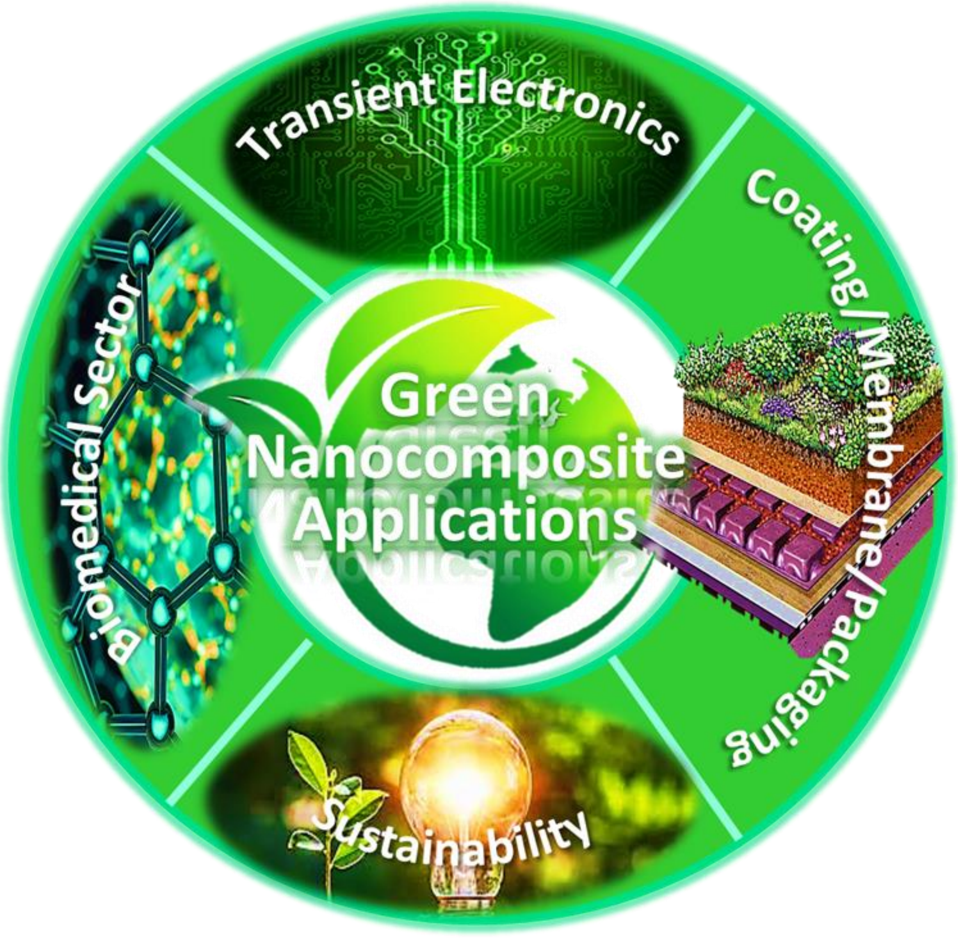

Figure 10: Applications of green nanopolymer-based nanocomposites

6.1 Transient Electronics

Electronic and plastic waste is a major cause of environmental pollution [121]. Technology has been shifted towards the use of the transient electronics [122]. The transient devices have environmental compatibility properties. Moreover, transient electronics have been designed with chemical and biological degradation. The degradation of the transient electronics results in numerous non-toxic by-products [123]. The transient electronics offer opportunities to green replacement of the electronics in the microelectronic industry, eco-devices, environmental sector, and biomedical relevance [124]–[127]. The nanocellulose nanomaterials base transient electronics have been reported [128]. Nanocellulose-based materials have several structural and physicochemical characteristics. These materials have fine thermal, mechanical and biocompatibility properties.

Green sensors have been designed using several green polymers and nanocomposite [129]. A nanochitin green nanocomposite-based sensor has been developed to detect toxic gases. Jing et al. [130] developed transient sensor of the green poly(acrylic acid)/nanochitin nanocomposite. The transient sensor had a high self-healing efficiency of 97%. The poly(acrylic acid)/nanochitin nanomaterial was used to form the self-powered pressure sensor. Moreover, the transient sensor was capable of lightweight sustainable wearable devices. Cellulose-based transient electronics have also been developed [131]. Zheng et al. [132] designed the green cellulose nanofibre/polyacrylic acid/graphene nanocomposite based transient wearable sensor. The green nanocomposite was formed using the in-situ free radical polymerisation. The green cellulose nanofibre/polyacrylic acid/graphene nanocomposite has a stretchability of ~850%, compression strength of 2.54 MPa, and electrical conductivity of ~2.5 Sm^{-1}. The transient sensor revealed a high healing efficiency of 96.7% for wearable electronics.

The transient supercapacitors have also been developed [133]–[135]. The ultra-thin supercapacitor was proposed by Migliorini et al. [136]. The transient supercapacitor had power of 120 kW kg^{-1} and may operate above 1.5 V. The transient batteries have also been constructed using the green nanomaterials [137]–[139]. In this regard, a cellulose-based transient lithium-ion battery has been reported by Chen et al. [140]. The ionic conductivity and bulk resistance of 0.54 $mScm^{-1}$ and 4.45 Ω, respectively, have been observed for the transient lithium-ion battery. The green electronics have hence been frequently adopted owing to the sustainability and renewability features. The transient electronics can be applied in the biomedical devices, environmental devices, sensors, and electronics-related energy devices [141]. The transient electronics have several advantages over the traditional non-degradable electronics. High performance green nanocomposites possess enhanced features for transient electronics; however, there are several challenges and opportunities for the complete replacement of electronics with transient electronics.

6.2 Film/Coating, Packaging and Membranes

Nanopolymer-based green nanomaterials have been applied in films/coatings [142]–[144]. Various combinations have been reported for the multi-layer films/coatings-derived packaging materials [145]. The green nanopolymer derived from natural sources have been used to form the compatible packaging applications [146], [147]. The packaging application of the green polymeric nanocomposites has been studied for the beverages and eco-electronics industry [148]. The green nanocomposite membranes have been developed from the nanocellulose, nanochitin and nanochitosan nanomaterials [149]–[151]. Liu et al. [152] fabricated the degradable membranes using the nanopolymer, graphene oxide, and silver nanoparticles. The antibacterial action of the membranes was studied against the gram-negative Escherichia coli and gram-positive Staphylococcus aureus. Nguyen et al. [153] formed the food packaging membranes of the nanocellulose and nanochitin through a layer-by-layer deposition route.

The oxygen and water vapor barrier properties were examined for the packaging membrane. Owing to crystalline nanostructure of nanocellulose and nanochitin, the oxygen transmission rate was reduced from 11.18 to 13.10 $ccm^{-2}day^{-1}$ and water vapor transport was decreased from 2.43 to 2.13 $gm^{-2}day^{-1}$. Moreover, the membranes have been tested for the antibacterial action. The green nanopolymers derived from natural sources have been used to form the film, coating, packaging and membranes with eco-friendly properties [154]–[156]. These green membranes have been designed to attain high performance barrier properties, robustness and antibacterial applications. This field also needs comprehensive research efforts to employ green nanopolymer-based membranes.

6.3 Solution to White Pollution and Sustainability

Currently, using biodegradable materials is an effective method to resolve white pollution [157]. In other words, white pollution is solid waste from different kinds of plastic product [158], [159]. These plastic products are usually made up of polymers. The plastic waste is hard to decompose and if degraded results in environmentally toxic by-products. The term sustainability means fulfilling the needs without compromising or polluting the surrounding environment or ecosystem [160]–[162]. Plastics have been produced from the renewable or fossil sources by using sustainability methods [163], [164]. The green plastic derived from renewable sources may reduce the greenhouse emissions. Nowadays, new technologies are shifting towards the use of nanopolymers derived from natural sources to replace the traditional commercial plastics. Incidentally, nanocellulose, nanochitin, nanochitosan and nanostarch have gained attention for green, sustainable and advanced functional materials [165].

6.4 Biomedical Applications

Green nanopolymer-based green nanocomposites have been applied in various biomedical applications [166], [167]. For application in drug delivery, nanomaterial degradation under hydrolytic conditions has been found indispensable [168], [169]. The synthesis, compatibility, hydrophobicity, degradation, in-situ erosion, etc. have been studied for drug delivery applications [170], [171]. Shariatinia et al. [172] prepared the chitosan-based green drug delivery systems using molecular dynamics simulations. The green chitosan/graphene nanosheet nanocomposite has been used for delivery of anticancer drug cyclophosphamide. The green nanocomposite has diffusion coefficient of $0.0553 \times 10^{-5} cm^2/s$. Figueroa et al. [173] prepared green chitosan functional graphene oxide for drug delivery. The nanomaterial has fine biocompatibility and non-toxicity properties. The therapeutic activity of the phytomedicine was studied using the chitosan functional graphene oxide nanocomposite.

The natural green polymer-derived materials and nanomaterials have also been applied in the tissue engineering assembles [174]. The green biomaterials have been used to recover the injured tissues and to regenerate the original organ. In this regard, the hydrolytically degradable polyester-based materials have been employed [175].

Collagen-based green nanomaterials have also been used in the tissue engineering [176]. The polycaprolactone/gelatin material also formed an effective green combination for the tissue engineering [177]. Accordingly, the natural polymer-derived green nanomaterials have been applied in the tissue engineering scaffolds of the bone, cartilage, skin, skeletal muscles, and regenerative medicine [178]–[180]. The green nanopolymer-based green nanocomposites have been found attractive for drug delivery, bone repair, tissue engineering, and delicate scaffolds for biomedical applications.

7 Challenges, Future and Summary

Conventionally, the polymeric materials used in industries have led to massive environmental issues [181]. To resolve this problem, polymers from renewable resources have been adopted. Natural materials such as chitin, chitosan, cellulose and starch have been used to form green nanopolymers and green nanocomposite. Green polymers are also known as eco-friendly materials. Green polymers have been found ideal for the next-generation lightweight, low-cost, sustainable and eco-friendly nanocomposites [182]. Consequently, numerous green polymers obtained from natural sources have been reinforced with nanofillers to form the nanocomposite. Green nanocomposites are used in electronics, membranes, packaging, environmental resolution, sustainability, and biomedical sectors. In addition to the technical application of green nanopolymers and green nanocomposites, it is essential that the challenges and future prospects be understood. Researchers are attracted to nanopolymer-based green nanocomposites owing to their biocompatibility, biodegradability and sustainability properties [183], [184]. However, there are several hinderances in the use the natural nanopolymers and derived materials. The complex extraction methods of nanopolymers from natural sources may have high cost and purification implications. It is important to understand the degradation mechanism and lifetime of green nanocomposites.

In the article, we present fundamentals of degradable green polymers, green nanopolymers from natural sources and their versatile applications to highlight the significance of these exceptional materials. Various categories of green nanopolymers derived from natural sources and derivative green nanocomposites have been discussed systematically. The current review will help researchers to attain clean environments and to avoid ecologically harmful polymeric materials.

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