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*Review*

# State-of-the-Art Nanoclay Reinforcement in Green Polymeric Nanocomposite: From Design to New Opportunities

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**Abstract:** Nanoclays are layered aluminosilicate nanostructures. Depending upon the chemical composition and microscopic structure, various nanoclay types have been discovered such as montmorillonite, bentonite, kaolinite, halloysite nanoclay, etc. Nanoclays have been organically modified to develop compatibility with polymers. Polymer/nanoclay nanocomposites have prompted significant breakthroughs in the field of nanocomposite technology. Green nanocomposites form an important class of nanomaterials using naturally derived degradable materials as matrix/nanofiller. This review essentially deliberates the fundamentals and effect of nanoclay reinforcements in the green polymer matrices. Naturally derived polymers such as cellulose, starch, natural rubber, poly(lactic acid), etc. have been employed in these nanocomposites. Green polymer/nanoclay nanocomposites have been fabricated using various feasible fabrication approaches such as the solution route, melt processing, in situ polymerization, and others. The significance of the structure-property relationships in these nanomaterials, essential to attain the desired features, has been presented. Green polymer/nanoclay nanocomposites are light weight, inexpensiveness, ecofriendly, have a low cost, and enhanced indispensable physical properties. Consequently, the green polymer/nanoclay nanocomposites have found applications towards sustainability uses, packaging, membranes, and biomedical (tissue engineering, drug delivery, wound healing) sectors. However, thorough research efforts are desirable to extend the utility of the green polymer/nanoclay nanocomposites in future technological sectors.

**Keywords:** nanocomposite; green; polymer; nanoclay; sustainability; packaging; biomedical

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**Citation:** Kausar, A.; Ahmad, I.; Maaza, M.; Eisa, M.H. State-of-the-Art Nanoclay Reinforcement in Green Polymeric Nanocomposite: From Design to New Opportunities. *Minerals* **2022**, *12*, 1495. https://doi.org/10.3390/min12121495

Academic Editor: Janos Kristof

Received: 3 November 2022
Accepted: 21 November 2022
Published: 23 November 2022



## 1. Introduction

Nanoclays are important layered aluminosilicates [1–3]. The platelet size usually ranges from 10 nm to 100 nm. Nanoclays occur naturally; however, can be formed through synthetic routes. In nanoclays, the individual nanolayers are made up of $SiO_4{}^{4-}$ tetrahedra or $[AlO_3(OH)_3]^6$ octahedra. The aluminosilicate-layered nanosheets are often stacked together via van der Waals forces. The layered nanosheets have entrapped ions between the platelets. The nanoclay-based nanostructures show notable structural, morphological, thermal, mechanical, and barrier properties. Owing to their unique nano-scale structure, the nanoclays have been used as excellent nanofillers in polymeric matrices to form the nanomaterials [4,5]. To progress the compatibility of the nanoclays (inorganic) with the polymers (organic), the layered silicates are usually modified via alkyl cations to form the organoclays [6]. The polymer/nanoclay nanocomposites own the mechanical robustness, heat stability, and flame resistance characteristics. Consequently, these nanocomposites have been applied in wide-ranging industrial applications including the aerospace, automotive, construction, packaging, electronics, etc. Ecofriendly and biodegradable polymers and nanoclays combinations have been recognized to develop the green nanocomposites [7]. Thus, the remarkable green polymer/nanoclay nanocomposites have been designed using the naturally derived polymers [8,9]. Subsequently, the green polymer/nanoclay nanocomposites have been explored in numerous sustainable materials and applied sectors [10–12]. Consequently, the polymer/nanoclay nanocomposites have been designed using several facile intercalation and exfoliation routes [13]. The nanoclay nanofillers including montmorillonite, cloisite, laptonite, sepiolite, halloysite nanotube, etc. have been filled in cellulose [14–16], starch or polysaccharides [17–19], poly(lactic acid) [20–22], silk [23,24], natural rubber [25–27], and other matrices. The green polymer/nanoclay nanocomposites have superior physicochemical, biocompatibility, and hydrophilicity properties. The versatile applications of green polymer/nanoclay nanocomposites have been observed in the fields of drug delivery, tissue engineering, packaging, and strength applications [28,29].

This review states the fundamentals, design, essential features, and potential applications of green polymer/nanoclay nanocomposites. The advanced developments and solicitations of the high-performance green polymer/nanoclay nanocomposites have rendered these materials important for future advancements in the related industries. Thus, this is an all-inclusive, pioneering, and up-to-date overview on green polymer/nanoclay nanocomposites portraying indispensable aspects from synthesis/essential features to technical potentials. To the best of our knowledge, green polymer/nanoclay nanocomposites have not been assessed comprehensively in the literature before with such specific review outline, recent literature reports, and technical analysis. Therefore, this review article is undoubtedly a ground-breaking contribution in the field of green nanomaterials aiming at advanced industrial applications. Subsequently, forthcoming evolutions in the field of green polymer/nanoclay nanocomposites are not conceivable for researchers before obtaining prior knowledge of the accumulated literature and investigations regarding these nanomaterials.

## 2. Nanoclay

In the nanocomposite field, research has focused on the use of inorganic or organic nanofillers for polymeric matrices [30]. Nanoclay was discovered as a unique inorganic nanofiller for the hybrid polymer nanocomposite [31]. Out of a large class of hydrous aluminum phyllosilicate minerals, commonly used nanoclays include montmorillonite, bentonite, kaolinite, mica, and many others [32]. The typical structure of a nanoclay consist of two-dimensional silica tetrahedral nanosheets attached to the alumina octahedral nanosheets (Figure 1). In nanoclays, a layered silicate sheet has a thickness of ~0.7 nm, whereas the double layer has a thickness of ~1 nm. The interlayer spacing between the nanoclay platelets has quite weak van der Waals interactions [33]. The layered silicates are hydrophilic in nature, so the interaction of the nanoclay with the polymer chains is generally difficult [34]. Therefore, to improve the organophillicity of nanoclays, the interlayer spacing is usually modified through exchangeable ions, organic molecules, or surfactants (Figure 2). As the nanoclay nanosheet surface is negatively charged due to the inbuilt silicate structure, the interlayer spacing can easily compensate an exchangeable cation layer ($Na^+$, $K^+$, $Mg^{2+}$, and $Ca^{2+}$, etc.). Accordingly, the charge exchange capacity of the layered silicates has been studied [35]. Due to surface charge on the layered silicate nanosheets, the nanoclays have shown an electrical conductivity of 25–100 $mSm^{-1}$ [36,37]. The structure of nanoclay can be well interpreted on the basis of differences and relative advantageous properties, compared with aluminum silicate, i.e., derived from aluminum oxide and silicon dioxide (Table 1). In nanoclays, the substitution of organic cations (aliphatic chain attached to -$NH_3^+$) between the nanoclay galleries can increase the interlayer spacing for better compatibility with the polymeric chains [38,39]. Synthetic polymers such as epoxy, polystyrene, polyamide, polyurethane, etc., have been used as matrices for nanoclay nanofillers [40]. The ensuing polymer/nanoclay nanocomposites have high tensile, non-flammability, thermal, electrical, and barrier properties [41–43].

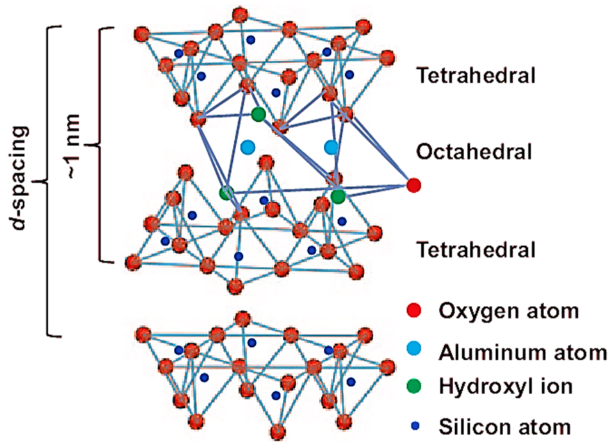


**Figure 1.** Structure of nanoclay (montmorillonite) [44]. Reproduced with permission from Springer.

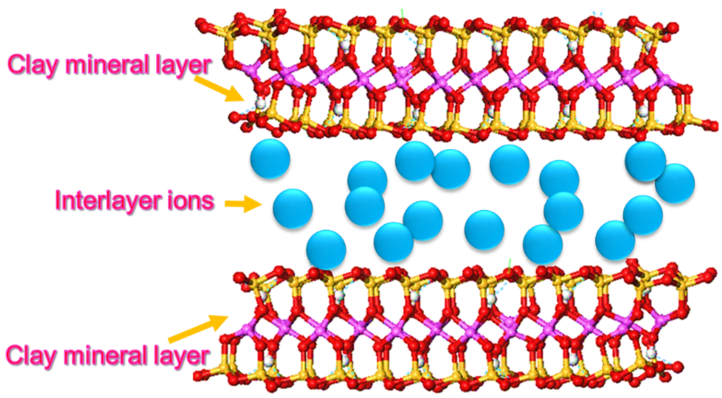


**Figure 2.** Entrapment of ions in layered nanosilicate nanostructure.

**Table 1.** Comparative properties of nanoclays and alumina silicate particles.

| Properties | Nanoclay | Alumina Silicate |
|---|---|---|
| Structure | Platelets or layered | Particles |
| Cation exchange capability | High due to layered structure | No cation exchange ability |
| Plastic behavior | Upon wetting | No plasticity |
| Swelling behavior | High on wetting | No swelling |
| Permeability | Low | High |
| Catalytic abilities | High | Low |
| Dimensional stability | Low | High |
| Dielectric/thermal insulation | Low | High |
| Resistance to thermal impacts | Low | High |
| Chemical stability | Low | High |
| Heat resistant | Low | 1300 °C |
| Application in metal plating | No | Yes |

## 3. Polymer/Nanoclay Nanocomposite

The nanoclay nanofillers have been reinforced in the polymeric matrices through various physical and chemical approaches [45]. However, developing fine interactions between the matrix and clay nanosheets have been found challenging to achieve a compatible polymer/nanoclay nanostructure [46]. In this regard, the interlayer spacing between the nanoclay platelets was improved through the intercalation of surfactant/functional molecules. Consequently, cationic or anionic nanoclay modification processes have been used [13,47]. Figure 3 demonstrates the intercalation of polymer chains between the nanoclay galleries and subsequent dispersibility to form the tactoids, intercalated, or exfoliated structures. For comparison purposes, a polypropylene matrix has been studied with different types of nanoclays to see the intercalation or exfoliation effects (Table 2). Depending upon the nanoclay type, intercalation or exfoliation was facilitated by the melt-blended polypropylene nanocomposites.

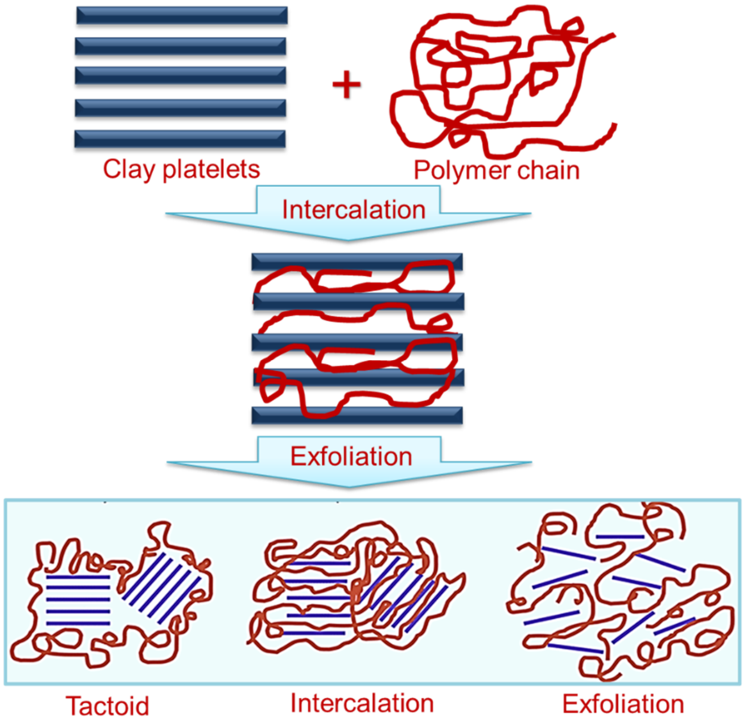


**Figure 3.** Formation of polymer/layered silicate nanostructure via intercalation and exfoliation.

Various facile methods have been adopted to form the polymer/nanoclay nanocomposites [48]. The most commonly adopted techniques involve the solution mixing (nanoclay and polymer dispersion in solvent), melt mixing (polymer/nanoclay direct melting and mixing at high temperature), and in situ polymerization (in situ monomer and nanoclay interaction) [49,50]. In these approaches, homogeneous nanoclay dispersion in the polymer matrices has been investigated.

**Table 2.** Polypropylene/nanoclay nanocomposite prepared by melt blending method.

| Polymer | Nanoclay | Polymer/Nanoclay Structure | Ref |
|---|---|---|---|
| Polypropylene | Montmorillonite | Exfoliated and intercalated | [51,52] |
| Polypropylene | Kaolinite | Intercalated | [53,54] |
| Polypropylene | Cloisite | Exfoliated | [55,56] |
| Polypropylene | Vinyl clay | Exfoliated | [57] |
| Polypropylene | Sepiolite | Exfoliated or intercalated | [58,59] |

Table 3 depicts a brief recent survey on some high-performance synthetic polymer/nanoclay nanocomposites. The polymeric matrices such as polystyrene [60–62], polyethylene [63–66], polypropylene [67–69], polyamide [70–72], poly(methyl methacrylate) [73–76], polyaniline [77,78], polypyrrole [79], etc., have been reinforced with nanoclays such as montmorillonite, bentonite, halloysite nanoclay, kaolinite, cloisite, and several other minerals. The inclusion of nanoclays in polymeric matrices was found to enhance the heat stability, flame defiance, mechanical properties (tensile strength, tensile modulus, toughness, etc.), barrier properties, and self-healing features of the nanocomposites. Wide ranging applications of the polymer/nanoclay nanocomposites have been observed from automotives/construction to electronics to biomedical areas [80–82].

**Table 3.** Recent survey on synthetic polymer/clay nanocomposite.

| Matrix | Nanoclay | Properties | Ref |
|---|---|---|---|
| Polystyrene | Montmorillonite; Bentonite | • Thermal stability<br>• Non-flammability<br>• Mechanical properties<br>• Barrier properties<br>• Morphology variation<br>• Barrier properties<br>• Low water permeability<br>• Low gas permeability | [60–62] |
| Polyethylene (low and high density) | Montmorillonite | | [63–66] |
| Polypropylene | Montmorillonite | | [67–69] |
| Polyamide | Montmorillonite | | [70–72] |
| Poly(methyl methacrylate) | Montmorillonite; halloysite nanoclay | | [73–76] |
| Polyaniline | Montmorillonite; kaolinite; bentonite | | [77,78] |
| Polypyrrole | Montmorillonite | | [79] |

## 4. Green Polymer/Nanoclay Nanocomposite

Green polymers (natural or synthetic) such as cellulose, starch, polycarbonate, poly(lactic acid), etc., have gained research interest for the development of ecological nanocomposites [83]. Green synthesis procedures have also been adopted to form ecopolymers and green nanocomposites [84,85]. Green polymers and nanomaterials have been applied in coatings [86], membranes [87], adhesives [88], drug delivery [89], and other biomedical applications [90,91]. In these nanocomposites, green nanofillers have been utilized with polymers to attain the desired biodegradability. Chitosan is usually employed as both a green matrix and a nanofiller [92,93]. Similarly, lignin has been used as a green nanofiller as well as a matrix [94,95]. Green nanofillers have been reinforced in numerous natural and synthetic polymers such as poly(lactic acid), poly(ethylene oxide), poly(vinyl alcohol), poly(lactic acid), and many others [96,97]. Nanoclays have been identified as important green nanofillers for polymeric matrices [98,99]. Among nanoclays, montmorillonite is a well-known ecological nanofiller [100–102]. Nanoclays, along with the degradable natural/synthetic polymers, have been effectively used to develop the green systems [103,104]. In green polymeric nanocomposites, carbon nanofillers such as graphene, graphene oxide, and carbon nanotube have also been reinforced [105–107]. The degradable green polymer/nanoclay nanocomposites have fine biodegradability, and antimicrobial effects, and thermal, mechanical, and electrical characteristics. A few general relative properties and applications of green polymeric nanocomposites with different nanoclays are given in Table 4.

**Table 4.** Property/application of various nanoclay types with green polymers.

| Montmorillonite | Cloisite | Halloysite Nanotube | Application of Nanoclays with Green Polymers |
|---|---|---|---|
| Thermal stability | Heat resistance | Thermal stability | Montmorillonite and halloysite nanoclay in cellulose and starch matrices for electrical, magnetic, and optical devices |
| Mechanical strength | Mechanical stability | Strength properties | Montmorillonite and halloysite nanoclay based green nanomaterials for drug delivery/tissue engineering |
| Gas permeability | Dimensional stability | Flame retardance | Cellulose and starch with montmorillonite for antimicrobials |
| Barrier properties | Rheological properties | Anticorrosion coatings | Cloisite in natural rubber for civil structures |
| Wastewater treatment | Thickener in lubrication oils | Wastewater treatment | Montmorillonite and halloysite nanoclay in cellulose and starch for packaging |

### 4.1. Cellulose/Nanoclay Nanocomposite

Cellulose has been obtained by chemical or enzymatic treatment of raw cellulose sources [108,109]. Cellulose is a natural degradable polymer; however, it has limitations of water vapor permeability and hydrophilicity [108]. Consequently, nanocomposites of cellulose with montmorillonite, kaolinite, and other nanoclays have been reported [110]. The cellulose-based nanocomposites have fine dispersion, mechanical robustness, gas barrier, and flame retardance characteristics [111]. The compatibility between the matrix and nanofiller in the nanocomposite has been analyzed. Barbi et al. [112] designed the bacterial cellulose and ceramic nanoclay-based green nanocomposites. The hydrophilic porous membranes were obtained having fine biomedical potential. Ming et al. [113] designed the green nanocomposite based on the nanofibrillated cellulose and montmorillonite nanoclay. The synthesis, transparency, and flame retardancy of the nanocomposites were investigated. Figure 4 shows the synthesis of nanofibrillated cellulose/nanoclay involving the formation of polymer suspension and nanoclay dispersion. The nanocomposites were fabricated through mechanical stirring and ultra-sonication approaches. Figure 5 displays the sample burning of neat nanofibrillated cellulose film and nanocomposite film with 50 wt.% clay nanoplatelet.

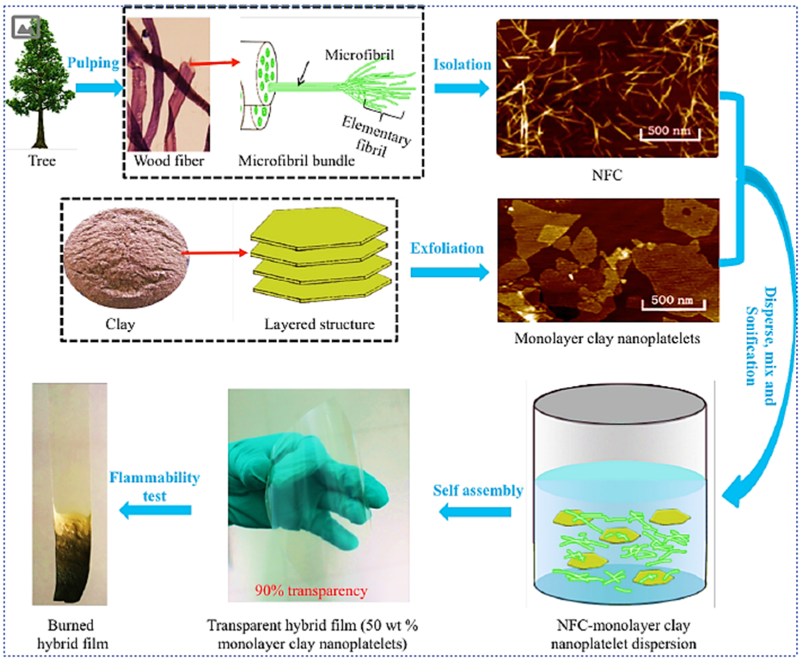


**Figure 4.** Schematic of transparent nanofibrillated cellulose (NFC) monolayer/nanoclay nanoplatelet-based hybrid films with self-extinguishing behavior (90% transparency at 600 nm). One-dimensional

nanofibrillated cellulose was extracted from wood pulp through mechanical and chemical treatments and mixed with uniformly dispersed nanoclay nanoplatelets in water. The nanofibrillated cellulose dispersed monolayer clay nanoplatelets suspension was blended with 0.5 wt.% clay nanoplatelet suspension using mechanical stirring and ultra-sonication methods. Water evaporation induced self-assembly was used to prepare a highly transparent NFC-monolayer clay nanoplatelet hybrid film with superb self-extinguishing effect [113]. Reproduced with permission from ACS.

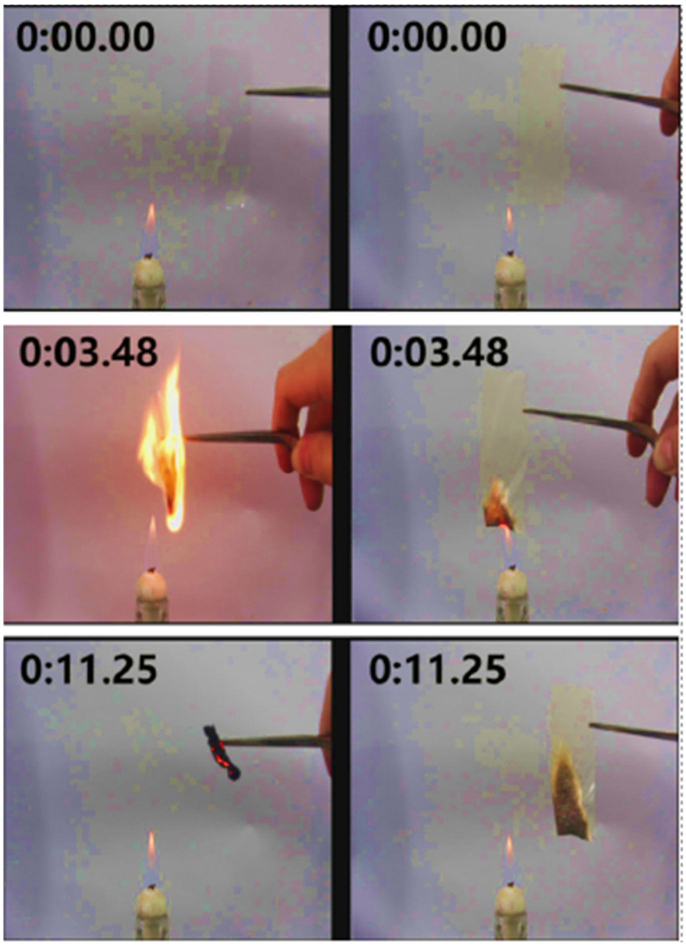


**Figure 5.** Time-lapse flammability of (left) pure nanofibrillated cellulose film and (right) transparent hybrid film with 50 wt.% monolayer clay nanoplatelet, respectively [113]. Reproduced with permission from ACS.

The neat nanofibrillated cellulose film showed an extensive burning for 8 s, and smoldering continues for 32 s. However, the nanocomposite sample was not vigorously burnt and revealed self-extinguishing behavior. The nanofibrillated cellulose/nanoclay films have high transparency (≥90) and superb self-extinguishing behavior, due to the effect of nanoclay nanofiller. Ferfera-Harrar et al. [114] developed green nanocomposites based on various types of organo-modified montmorillonite such as gelatin-modified montmorillonite or chitosan-modified montmorillonite. Triethyl citrate was used as an eco-plasticizer. In microscopic studies, the plasticized nanocomposites displayed well intercalated and exfoliated structure. The 5 wt.% nanoclay content was found to be the optimum to increase the thermal stability of the nanocomposite. In addition to the thermal and flame resistance features, the cellulose/nanoclay nanocomposites have been used for electronic applications [115]. Cellulose paper was filled with nanoclay nanofiller to develop flexible and green electronics [116]. A cellulose matrix with 34 wt.% nanoclay content had improved the young's modulus by 1.5 times compared to the neat cellulose. Moreover, the thermal stability of the cellulose/nanoclay nanocomposite paper was improved from 290 (neat cellulose) to 310 °C. The cellulose/nanoclay nanocomposite paper revealed flexible and easy-to-process electronics, as compared to the conventional rigid circuits.

### 4.2. Starch/Nanoclay Nanocomposite

Starch belongs to the polysaccharide group having low cost, biodegradability, potential availability, and attractive physical properties [117]. Starch is commonly found in wheat, rice, corn, potato, etc. The structure of starch has intense intermolecular hydrogen bonding between its chains, which may affect its solubility [118]. Starch has important applications in packaging, drug delivery, and tissue engineering [119]. Green nanocomposites have been designed using a starch matrix and nanoclay nanofiller via a solvent casting method [120]. Pristine starch usually has poor mechanical properties. In this regard, plasticizers have been used to improve the physical features of starch. Plasticizers are usually low molecular weight compounds, which are added to improve the flexibility and processibility of the starch-based materials [121,122]. Mansour et al. [123] prepared the nanocomposite derived from non-granular maize starch and sodium montmorillonite nanoclay. The nanocomposite was plasticized with the glycerol plasticizer. The addition of 10–20 wt.% nanoclay content and 20 wt.% glycerol plasticizer improved the mechanical properties of the nanocomposite. The effect of the plasticizer was observed in fine processing and dispersion of the nanocomposites. Pandey et al. [124] fabricated starch/nanoclay nanocomposites. The glycerol particles were suggested to act as the plasticizer. Starch plasticization improved the polymer chain mobility and diffusion into the nanoclay galleries. In effect, the thermal and mechanical properties were enhanced. Almasi and co-workers [125] designed the green nanocomposites based on the citric acid modified starch matrix and montmorillonite nanoclay nanofiller. Inclusion of 7 wt.% nanoclay improved the ultimate tensile strength of the nanocomposite by threefold, compared with the neat citric acid modified starch. Accordingly, starch-based green nanocomposites have been mostly developed using the modified nanoclays and plasticizers. The use of additional low molecular weight particles and functional nanofiller has been considered essential to support the homogeneous nanocomposite formation with superior physical properties.

Alginate is an anionic polysaccharide found in brown algae, which can be easily cross-linked through ionic crosslinks between the carboxylic acid moieties and divalent cations such as $Ca^{2+}$, $Sr^{2+}$, $Zn^{2+}$, or $Ba^{2+}$ [126,127]. This green polymer has also been filled with nanoclay nanofillers to form nanocomposites. Consequently, the alginate polymer was filled with laponite nanoclay to form three-dimensional printing ink [128]. The nanocomposite was found to be useful for drug delivery applications. The 3D printed architectures of alginate/nanoclay nanocomposites have also been designed [129]. The nanocomposite was found to be useful for the removal of toxic metal ions from water. Furthermore, a halloysite nanotube [130] (a form of nanoclay) has also been used to form the nanocomposite with starch or polysaccharide matrices. The halloysite nanotube is a unique type of one dimensional nanofiller. The biodegradable polysaccharides have weak stability in processing, poor barrier properties, and high susceptibility to environmental variations. The inclusion of a halloysite nanotube in biodegradable polysaccharides has been found to improve the mechanical properties, and decreased the permeability to water vapor/oxygen, lowered water solubility and water adsorption capacity, and so renders a potential for food packaging application. Functionalized halloysite nanotube such as polydopamine-coated halloysite nanotube can be a good option for improving the mechanical and physical properties of biopolymer films and composites [126,131]. Makaremi et al. [132] designed pectin a heteropolysaccharide/halloysite nanotube for food packaging applications. The inclusion of a halloysite nanotube in a pectin heteropolysaccharide matrix produced antimicrobial activity in nanocomposite films towards the Gram-positive and Gram-negative bacterial strains. Hence, the nanocomposites were found to be useful for food packing material. Bertolino et al. [133] produced a polysaccharide/halloysite nanotube aimed at drug delivery applications. For this purpose, the solvent casting, lyophilization, and cryoscopic methods have been used. The physiochemical, morphological, and mesoscopic properties have been investigated targeting drug transport applications.

### *4.3. Poly(Lactic Acid)/Nanoclay Nanocomposite*

Poly(lactic acid) has been employed as an important biodegradable green matrix for nanoclays [134,135]. Darie et al. [136] prepared poly(lactic acid)/Cloisite nanoclay nanocomposite through solution intercalation methods. The nanocomposites were tested and verified for antimicrobial activity versus Gram-positive and Gram-negative bacterial strains. Grigora et al. [137] proposed a poly(lactic acid)/montmorillonite nanoclay nanocomposites via the solution method. The inclusion of 1–4 wt.% nanoclay enhanced the hydrophilicity of the nanocomposites due to the intrinsic hydrophilic nature of reinforced nanofiller. Bai and co-workers [138] processed the poly(lactic acid)/montmorillonite nanocomposite through the laser sintering method. Figure 6 illustrates an enhancement in the flexural modulus using three different laser powers (15, 16, and 17 W).

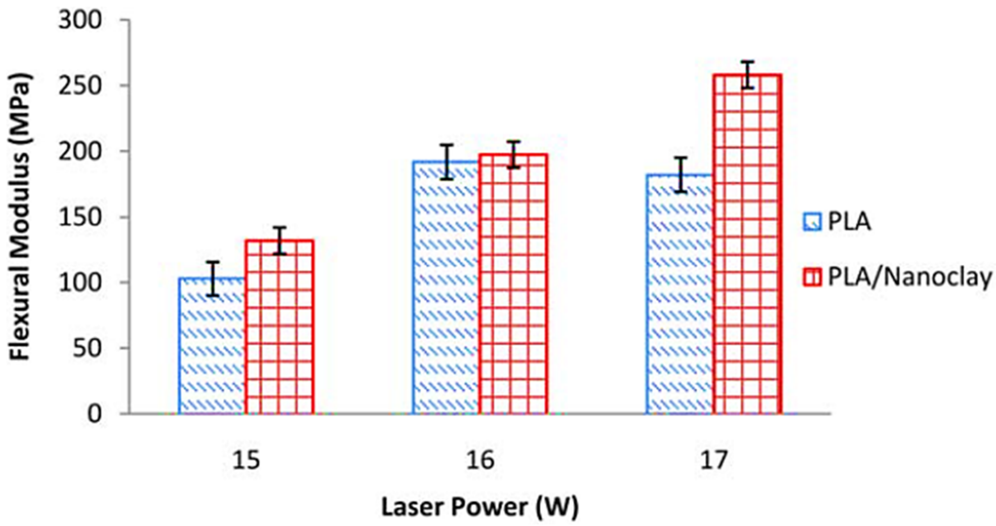


**Figure 6.** Variation in flexural modulus vs, laser power for poly(lactic acid) and poly(lactic acid)/montmorillonite nanoclay nanocomposite (powder bed temperature 60 °C) [138]. Reproduced with permission from Wiley.

The poly(lactic acid)/montmorillonite nanocomposite presented development in the flexural modulus (41%), relative to the neat poly(lactic acid) [139]. In addition, polycaprolactone has been used as a relevant biodegradable matrix for nanoclay nanofillers [140–142]. The polycaprolactone/montmorillonite nanocomposites revealed the antimicrobial and toxic ion removal properties [143,144].

### *4.4. Natural Rubber/Nanoclay Nanocomposite*

The nanoclay and silica nanoparticles have been successfully reinforced in the natural rubber matrix to develop green nanocomposites [145,146]. The nanofiller dispersion, polymer–nanofiller interactions, and morphological and mechanical profiles have been investigated. The exfoliation of nanoclay nanoplatelets in the natural rubber matrix caused nano-reinforcement effects [147,148]. Perra et al. [149] formed an organoclay based on the cetyl trimethyl ammonium bromide-modified montmorillonite. The organoclay was reinforced in a natural rubber matrix, and dispersion and morphological properties were studied. The strength and hardness of the natural rubber were found to improve with the increasing nanoclay contents. Siririttikrai et al. [150] developed natural rubber/nanoclay nanocomposite-based compounds and vulcanizates. The nanoclay nanoparticle coagulation was observed in the rubber matrix. The viscosity of the nanocomposite, with varying nanoclay contents, was determined. An increase in viscosity was observed due to the nanoclay aggregation effect. George et al. [151] prepared natural rubber and organomodified Cloisite nanoclay-derived nanomaterial using the melt blending method. The natural rubber/Cloisite nanocomposite revealed high tensile strength and modulus with the inclusion of 5 phr nanofiller content. The enhancement in mechanical properties was observed due to better nanoclays dispersion and exfoliation in the natural rubber matrix. Sookyung et al. [152] prepared the sodium montmorillonite nanoclay and converted to an organoclay using octadecylamine as a modifying agent. The resulting organoclay was dispersed in the natural rubber matrix. The organoclays with greater *d*-spacing resulted in a higher degree of dispersion in the natural rubber matrix. Consequently, the mechanical and thermal stabilities of natural rubber/organoclay nanocomposites were enhanced. In almost all the natural rubber-based systems, organoclays revealed better dispersion, exfoliation, and interactions with the elastomeric matrix, subsequently increasing the final material properties.

### *4.5. Silk/Nanoclay Nanocomposite*

Silk is a natural protein fiber, which is made up of fibroin [153]. The protein fiber of silk is naturally derived from insect larvae. Silk protein has been reported to form high performance nanocomposites with nanoclay nanofillers [154]. Devi et al. [155] developed biodegradable nanocomposites based on a silk matrix and nanoclay nanofiller. The inclusion of 2 phr nanoclay content considerably enhanced the tensile strength/modulus and flexural strength/modulus of the silk/nanoclay nanocomposites, relative to the pristine silk protein. The enhancement in the mechanical properties was observed due to the reinforcing effect of the two-dimensional nanoplatelets and better nanoclay dispersion in the silk matrix. Doblhofer et al. [156] prepared the spider silk protein and layered silicate sodium hectorite-based nanomaterials using the all-aqueous process. The inclusion of nanoclay nanoplatelets offered a fine barrier towards the permeating oxygen molecules and water vapors, through the formation of tortuous paths in the silk matrix. The biocompatible spider silk/hectorite nanoclay nanocomposites have been used in packaging applications due to fine oxygen and water vapor barrier properties. In another attempt, Chen et al. [157] designed silk peptide/chitosan films filled with two types of nanoclays, i.e., montmorillonite and sepiolite, through solution casting route. The montmorillonite nanoclay revealed better reinforcing effects to enhance the mechanical properties of the nanocomposite, due to better nanofiller dispersion. On the other hand, sepiolite nanoclay was less effective in improving the mechanical properties of the nanocomposite, owing to poor dispersion in the matrix. Zhang et al. [158] proposed the silk fibroin/laponite nanoclay nanocomposite-based enzymatically crosslinked hydrogel. The nanocomposite hydrogel was found to be effective in improving the thermal and chemical stability properties. It has been observed that the well oriented self-assembled spider silk proteins on the nanoclay surface produced high barrier properties towards oxygen molecules and water vapors. Moreover, the silk fibroin/Laponite nanoclay nanocomposite was effectively applied in tissue engineering applications, especially to repair hyaline cartilage tissues. The silk fibroin/laponite nanoclay nanocomposite hydrogel was biocompatible and was capable of inducing osteogenic and chondrogenic differentiation of bone marrow stem cells. Kadumudi et al. [159] reinforced the silk fibroins with the laponite nanoclay to form flexible and electroactive thin films. The silk/laponite nanocomposites were water insoluble and thermally and chemically stable. The nanocomposites were used to form highly flexible and ecofriendly wearable motion sensitive sensors. It has been observed that the two-dimensional nanoclay nanoplatelets caused increased folding of the polypeptide chains, so resulted in more ordered and crystalline configurations. The green sensors revealed accurate and fast response time during bending and stretching states. Hence, the silk/nanoclay nanocomposites revealed fine physical properties and applications related to strength, packaging, tissue engineering, and flexible sensor fields.

## 5. Significance of Green Polymer/Nanoclay Nanocomposite

### *5.1. Sustainability*

Nanoclays have been frequently adopted as ecofriendly nanomaterials in the green nanocomposites (Figure 7). Nanoclays own extensive sustainable applications in environmental and ecological friendly areas [160]. The use of the nanoclay-based green nanocomposite in sustainability applications has promoted the economic conditions of the industries and countries [49]. Consequently, the industries employing sustainable nanomaterials play an important part in uplifting the economic conditions of the countries [161]. In construction materials, nanoclays have been introduced to enhance the intrinsic strength and stability properties [162,163]. In the construction industry, nanoclay-based materials have been used to substitute the cement and also to develop the high-performance concrete [164]. Thus, nanoclay-based construction technologies have paved ways towards sustainable engineering applications [165]. The sustainable construction structures have low porosity and permeability, and high durability, compressive/flexural strength, heat resistance, and resistance to chemicals attack [166].

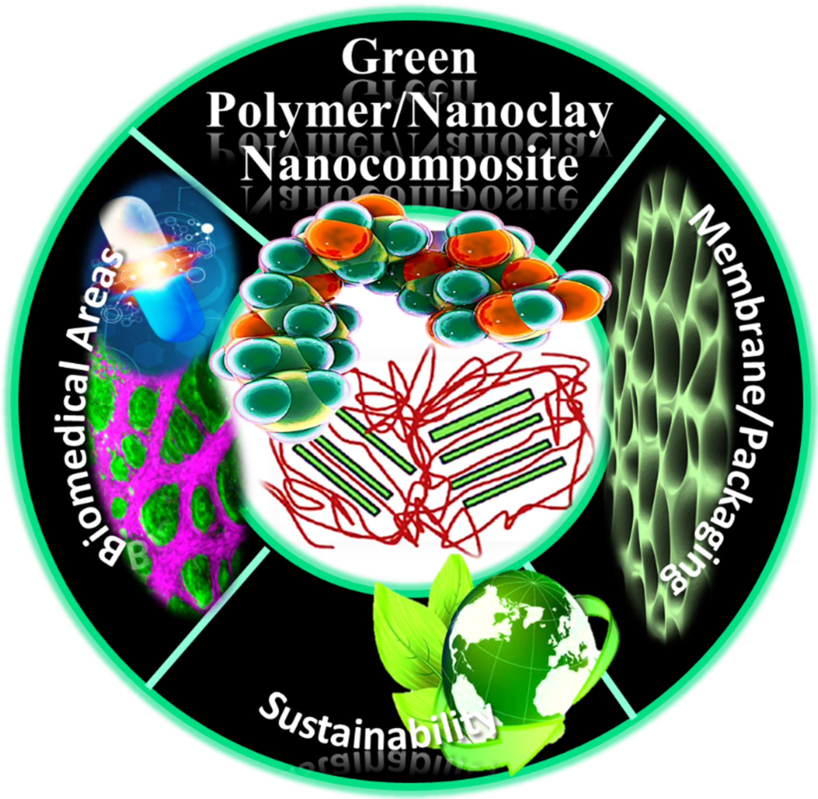


**Figure 7.** Applications of green polymer/nanoclay nanocomposite.

An essential use of nanoclay-derived materials has been observed in the remediation of environmental contaminants [167–169]. Here, the sustainable nanoclay-based nanomaterials act as a biodegradation promoter. The organically modified nanoclays and polymer/nanoclay nanocomposites have improved biocompatibility with living organisms, as compared to the non-modified nanoclays [170]. Therefore, the nanoclay-derived green nanomaterials have been beneficially employed in natural systems avoiding any harmful effects for ecosystem recovery. Accordingly, the use of nanoclays in the agriculture sector has grown enormously. The organoclay-derived nanomaterials have been found to be promising as rheological modifiers, gas absorbents, and drug delivery carriers [171–173]. Hence, the nanoclay-based nanomaterials have been used to produce sustainable, regenerative, and reusable environmentally compatible materials.

### *5.2. Membranes/Packaging*

Owing to their low cost and light weight, synthetic polymers have been recurrently applied in membranes and packaging industries [174–176]. However, the resulting environmental pollution is a major drawback of using these non-degradable synthetic polymers [177]. To resolve this issue, naturally derived polymers or biodegradable polymers have been adopted for packaging applications [178]. The naturally derived polymers have fine decomposable behavior, and so are ideal for packaging applications [179]. Several degradable natural and synthetic polymers have been applied to form ecofriendly membranes or packaging. The cellulose, starch, poly(lactic acid), poly(vinyl alcohol), etc., have been successfully applied in the food industry and electronics industry related to packaging materials [180–182]. Such membranes or packaging reveal facile decomposition and recyclability. Garusinghe et al. [183] developed the nanocellulose and montmorillonite nanoclay-derived membranes. The nanocellulose/montmorillonite nanocomposite membranes were tested for the water vapor permeability (WVP) and enhanced nanocomposite tortuosity. Figure 8 presents a mechanism for montmorillonite dispersion in the matrix at a high loading level. The high nanoclay contents decreased the tortuous paths in the nanocomposites and so improved the barrier properties. On the other hand, high-pressure homogenization enhanced the homogeneous dispersion and reduced the nanoclay stacking in nanocellulose/montmorillonite, so increasing the tortuous pathways. However, the nanoclay nanoplatelets have a tendency to restack/aggregate due to electrostatic or van der Waals interactions [184,185]. The WVP of pristine nanocellulose/montmorillonite nanocomposite nanosheets, and the nanocomposite membranes prepared at high-pressure homogenization and sonication steps were measured (Figure 9). For pristine nanocomposite and membrane prepared with high-pressure homogenization, the WVP was reduced with the nanoclay loading and reached a minimum value (6.3–13.3 g·µm/m$^2$·day·kPa) at 16.7–23.1 wt.% loading. However, the WVP was increased pointedly with the nanoclay loading for the nanocomposite prepared via sonication. In the nanocomposites processed by sonication, enhanced WVP was suggestive of better homogenization of cellulose/nanoclay. However, for packaging application, the nanocomposites with low WVP were preferred. Thus, efficient recyclable nanocellulose/montmorillonite nanocomposites-based packaging materials have been designed.

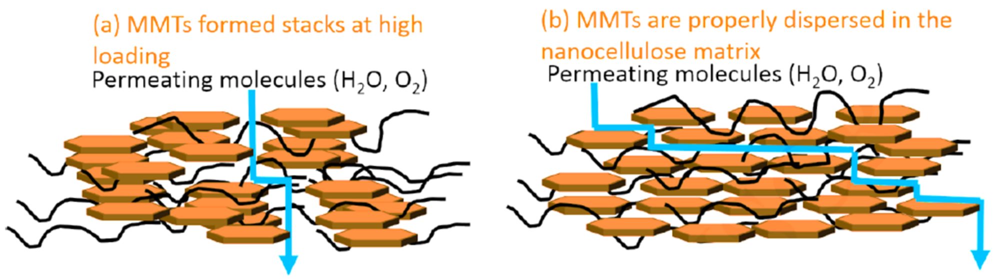


**Figure 8.** Mechanism of MMT stacking and arrangement of stacks in nanocellulose network. (**a**) MMT stack formation at high loading level which decrease the tortuous path; (**b**) MMT stacks broken down using high-pressure homogenization of nanocellulose/MMT suspension which increase the tortuous path [183]. MMT = montmorillonite. Reproduced with permission from Elsevier.

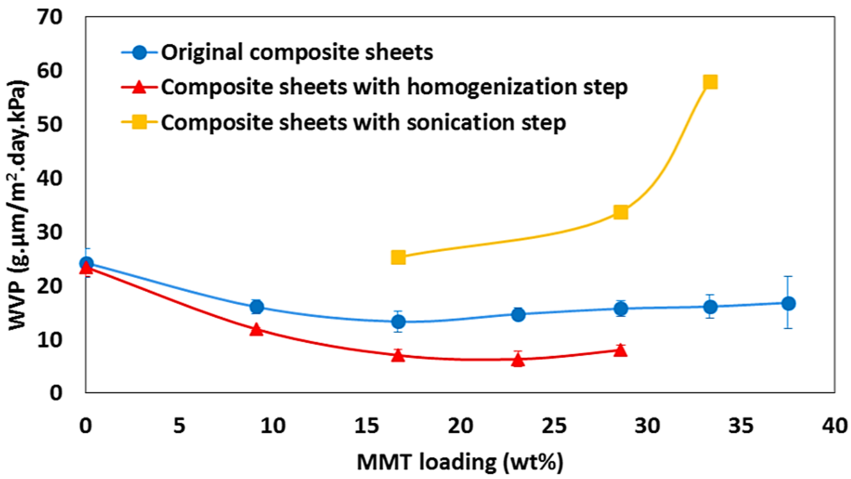


**Figure 9.** WVP of nanocellulose/MMT nanocomposites. The pristine nanocomposite sheets; nanocomposite sheets with high pressure homogenization step, and sonication step [183]. WVP = water vapor permeability; MMT = montmorillonite. Reproduced with permission from Elsevier.

Aulin and co-workers [186] reported the nanocellulose nanofiber and vermiculite nanoclay nanoplatelet-based nanocomposite for packaging membranes. High-pressure homogenization was used to form the nanocellulose nanofiber/vermiculite nanoclay nanomaterials. The nanocomposite membranes revealed high strength and modulus of 257 MPa and 17.3 GPa, respectively. Scanning electron microscopy analysis showed the consistent spreading of nacre like nanoplatelet layers in the nanocellulose matrix. The oxygen permeability of nanocellulose nanofiber/vermiculite nanoclay nanocomposite (0.07 $cm^3 \mu m \cdot m^{-2} d^{-1} kPa^{-1}$ at 50% relative humidity) membrane was found to be better than the commercial cellulose packaging material. The packaging material was proposed for the electronics and barrier coatings for the large laminations. Farmahini-Farahan et al. [187] also designed the cellulose/nanoclay-based packaging materials. Sodium montmorillonite was used as nanofiller in the nanocomposite membranes. Due to the superior barrier effect of the nanoclay, the low water vapor transmission rate was observed, i.e., 43 $g/m^2$/day. Souza et al. [188] prepared the thermoplastic starch carvacrol and essential oil-modified montmorillonite-based packaging. Due to the crystallinity of the nanofiller and fine interactions between the organomodified nanoclay and starch matrix, a strong antimicrobial effect was observed against *E. coli* bacteria. Thus, the research on naturally derived polymer and nanoclay-based packaging films depicted a potential towards moisture, gases, microbials, etc. and can be utilized in the electronics, food/beverages, and other advanced packaging industries.

### *5.3. Biomedical Relevance*

Nanoclays have been used in numerous biomedical applications due to their high surface-to-volume ratio and biocompatibility effects [189]. Moreover, nanoclays may develop interactions with living systems due to heterogeneous charge distribution [190]. Tissue engineering is a speedily mounting field in regenerative medicine [191]. Recently, the three dimensional scaffolds based on nanomaterials have gained research interest in the field of tissue engineering [192]. Such three-dimensional scaffolds have been found to be effective for tissue reformation [193]. Natural polymer-derived nanostructured materials have gained substantial consideration due to feasible cell attachment and proliferation in the biological environment. Nouro et al. [194] fabricated the poly($\epsilon$-caprolactone)/nanoclay nanocomposite nanofiber through an electrospinning technique. The presence of dispersed nanoclay nanoparticles generated homogeneous nanofiber structure. The nanoclay nanoparticles facilitated the cell adhesion and bioactivity of the nanofibers to fibroblasts cells for tissue regeneration. Moreover, the poly($\epsilon$-caprolactone)/nanoclay nanocomposite nanofiber possess fine wettability and degradability properties. In this way, the nanoclay-based nanocomposites acted as active candidates for tissue engineering.

In the drug delivery application, the polymeric scaffolds impregnated with drugs have been studied [195]. In polymer-based scaffolds, the role of solubility, permeability, systemic circulation, and pharmacological response (upon certain concentration) of the drugs have been investigated [196,197]. Ferrández-Rives and co-workers [198] formed electrospun mats of poly(vinyl alcohol) hydrogel for drug delivery. The efficiency of nanoclay encapsulation in poly(vinyl alcohol) nanocomposite hydrogel has been studied. The absorption of bovine serum albumin on the nanocomposite hydrogel was considered. The inclusion of nanoclay enhanced the drug entrapment and release behavior in the nanocomposite. Hsu et al. [199] prepared the polycaprolactone, poly-DL-lactic acid, and Laponite (disk-shape) nano-clay based nanocomposite for drug delivery. The drug administration was performed for the treatment of type-2 diabetes to control the body weight and blood glucose level. The drug was used in the gastrointestinal tract to limit food absorption. Accordingly, the biocompatible polymer/nanoclay nanocomposites have good potential for drug delivery applications.

Wound healing application has also demanded the use of nanoclay-based nanomaterials due to the lowering of pain, infection and scarring effects [200–202]. Bibi et al. [203] designed the biodegradable poly(vinyl alcohol)/nanoclay nanocomposite to deliver the Penicillin drug to wounds. The nanomaterial had fine antimicrobial effectiveness. Asthana et al. [204] also prepared the poly(vinyl alcohol)/nanoclay nanocomposite through the solution method. The nanocomposite had low microbial growth and high microbiological stability. The material was applied using Aloe vera gel to the wounded skin areas. In addition to montmorillonite, the kaolinite nanoclay has also been used with the poly(vinyl alcohol) matrix to enhance the chemical stability and to lower the infection rate in wound healing applications [205].

For a better judgement purpose, essential green polymer/nanoclay systems for technical applications (sustainability, packaging, drug delivery, tissue engineering, wound healing, and antibacterial purposes) are portrayed in Table 5. The data listed show that comprehensive efforts are still desirable to further expand the implication of green polymer/nanoclay systems in various fields.

**Table 5.** Specifications of green nanocomposite systems for technical fields.

| Polymer | Nanofiller | Processing | Property/Application | Ref |
|---|---|---|---|---|
| Wood | Montmorillonite | Solution/melt method | Sustainable construction materials | [206] |
| Cellulose, starch, poly(lactic acid) | Montmorillonite | Solution/melt method | Packaging | [180–182] |
| Nanocellulose | Montmorillonite 16.7–23.1 wt.% | Solution method | Low water vapor permeability 6.3–13.3 g·µm/m$^2$·day·kPa; packaging | [183] |
| Nanocellulose nanofiber | Vermiculite nanoclay | Solution method | Oxygen permeability 0.07 cm$^3$µm·m$^{-2}$d$^{-1}$kPa$^{-1}$ at 50% relative humidity; packaging | [186] |
| Cellulose | Montmorillonite | Solution method | Low water vapor transmission rate 43 g/m$^2$/day; packaging | [187] |
| Starch | Montmorillonite | Solution method | Antimicrobial effect against *E. coli* bacterial strain | [188] |
| Poly(ϵ-caprolactone) | Montmorillonite | Electrospinning | Tissue engineering; better cell adhesion and bioactivity | [194] |
| Poly-(DL-lactic acid) | Laponite | Solution method | Drug delivery; Type-2 diabetes curation | [199] |
| Poly(vinyl alcohol) | Montmorillonite | Solution method | Wound healing; penicillin drug to wounds | [203] |
| Poly(vinyl alcohol) | Montmorillonite | Solution method | Wound healing; high microbiological stability | [204] |

## 6. Encounters, Future and Summary

Nanoclays have been recognized as widely utilized nanofillers for advanced polymeric nanocomposites. In green polymer/nanoclay nanocomposites, degradable polymers and nanoclays (eco nanofiller) have been utilized. The property enhancement has been observed even at a very low nanofiller loading level. Polymer/nanoclay nanocomposites have been prepared through facile processing methods such as solution and melt mixing techniques to accomplish high-performance materials. During nanocomposite processing and final uses, several challenges have been observed and need to be resolved [207,208]. First of all, only a few natural polymers have been reinforced with this green nanofiller. Nanoclay nanoplatelet dispersion, intercalation, and exfoliation strategies need to be improved to attain the homogeneously dispersed nanomaterials [209]. Especially the conversion of nanoclay to nanoclay-based salts and the subsequent formation of the organomodified nanoclay need to be scrutinized. Through effective organophilic ion treatments, the pristine hydrophobic nanoclays have been modified to form the alkyl chain intercalated hydrophilic nanoclay. Organophilic and hydrophilic nanoclays have been found to be compatible with the polymers. The matrix-nanofiller interactions have facilitated the microscopic properties of the green nanocomposites [210]. Moreover, the resulting chemical, mechanical, electrical, non-flammability, and barrier properties have been enhanced [211]. By overcoming the challenges, applications of green polymer/nanoclay nanocomposites can be extended to eco-friendly automobiles, aerospace, construction, electronics, and environmental relevance [212–216]. However, up till now the green polymer/nanoclay nanocomposites have found uses in sustainable materials, eco packaging and membranes, and biomedical areas such as tissue engineering, drug delivery, wound healing, etc. The conventional polymeric materials used in the above-mentioned industries may cause huge environmental issues [217]. The future of sustainable nanoclay nanomaterials rely on the biodegradability of these materials [218]. By adopting sustainability strategies, polymers have been developed from the renewable resources. The green polymers obtained from the renewable sources may lessen the greenhouse emissions. These days, new techniques have been focused to substitute old-fashioned commercial plastics with natural degradable polymers. Researchers need to emphasize the green nanocomposites having better biocompatibility, biodegradability, and sustainability properties.

Briefly, this article offers foundations for the selected green nanofiller, i.e., nanoclay, degradable polymers from natural sources, and their multipurpose applications. The main purpose of this review is to highlight the applications of green polymer/nanoclay nanomaterials. Green polymer/nanoclay nanocomposites have been examined in the various reported categories. The present article aims to assist scientists and researchers to formulate ways to be more ecofriendly to the environment through adopting the innovative green nanomaterials.

**Author Contributions:** Conceptualization, A.K.; data curation, A.K.; writing of original draft preparation, A.K.; Review and editing, A.K., I.A., M.M. and M.H.E. All authors have read and agreed to the published version of the manuscript.

**Funding:** This research received no external funding.

**Data Availability Statement:** Not applicable.

**Conflicts of Interest:** The authors declare no conflict of interest.

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