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Journal of
Earth & Environmental Waste Management

Review Article

Review on Bio-Nanocomposite Polymers and its Application on Food Packaging Processes

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Received: 2023 Nov 02 **Accepted:** 2023 Nov 21 **Published:** 2023 Nov 25

Abstract

Bio-nano composites are nano composites including a naturally occurring polymer in mix with an inorganic moiety, and appearing no less than one measurement on the nanometer scale. This review presents the most commonly methods of bio-nanoparticles preparation in food packaging, the significant changes they cause in the properties of packaging material, and the commercially available nano-based materials. Those materials are ecofrindly and biodegradable advantages over petroleum products. Bio nanocomposite has good mechanical, barrier, optical, thermal, and other functional properties. It contains especial biomaterials and nano particles. The basic Synthesizing techniques of bio nano composites are in situ polymerization, solution casting, and melt processing. Spectroscopic techniques including Fourier transform infrared spectroscopy, X-ray diffraction, Raman spectroscopy, scanning electron microscopy, transmission electron microscopy, are the most used to characterize the functional properties and the surface structure of bio nanocomposite materials. Bio-nano composites are a smart food packaging whereby they can perceive property of the packaged food such as microbial contamination and uses some mechanism to register and convey information about the quality or safety of the food. Bio-nanocomposite materials for food packaging are important not only to reduce environmental problem but also to improve the functions of the food packaging materials.

Keywords: Bio-Nanocomposite, Biopolymer, Food Packaging, Nanocomposite

1. Introduction

Bio-nanocomposites consist of a biopolymer framework reinforced with particles [nanoparticles] with a ratio of at least the nanometer range [1-100 nm] [1]. Bio-nanocomposites have become the epitome for assigning nanocomposites that contain naturally occurring polymers [biopolymers] mixed with minerals and appear to be above nanometer-scale measurements [2]. Nanocomposites containing normally occurring polymers [biopolymers] mixed with inorganic nano entries represent another class of material called BNC [bacterial nano cellulose] [3].

Bio-nanocomposite has become a potential and sustainable material for use in new and powerful applications. These nanocomposite materials are lightweight, environmentally friendly and replace traditional non-biodegradable petroleum-based plastics [4]. These nano systems are incorporated into a polysaccharide or protein matrix called a "nanocomposite." It is defined as a combination of two or more materials, forming a mixture that enhances the properties of components in which at least one is present on the nanometer scale [5, 6].

The development of nanocomposite has allowed edible coatings to be used as "temporal distribution systems" that release active substances from a matricidal film to the food, to improve conservation [7]. Due to growing environmental awareness, nanomaterials from natural resources (bio-nanomaterials) are being used more and more [8]. The use of synthetic non-biodegradable polymer has caused environmental damage [9]. This raises interest in the development of new renewable and biodegradable matrices called biopolymer or bioplastic [10].

Polymers increasingly used in composite manufacturing to replace non-biodegradable polymers include starch, cellulose, poly (lactic acid), poly (hydroxybutyrate), pectin, chitosan, collagen, and hydroxyl appetite [9]. Because biopolymers are derived from renewable resources, they have many advantages over petroleum-based polymers used in packaging, agriculture, medical and other applications [11]. Advantages of bio-nanocomposites over traditional composites are reduced weight, increased flexibility, greater mold ability, reduced cost, sound insulation and renewable nature [12].

Petrochemical polymers, often referred to as plastics, are widely used polymers in food and beverage packaging due to their high performance and low cost [13]. The unique properties of food packaging are low cost, excellent physical properties [density, molecular weight], mechanical properties [tensile strength], and permeation properties [O_2, CO_2] shown in fig. 1 [5]. The only problem with synthetic polymers is that they are not biodegradable, do not decompose easily and remain as waste for a very long time [6]. Commonly used plastics such as polyethylene and polypropylene take years to biodegrade and are not suitable for food packaging applications where the plastic is used only for a short time and then discarded. In addition, these plastics are usually contaminated with food ingredients and other biological substances [5, 14].

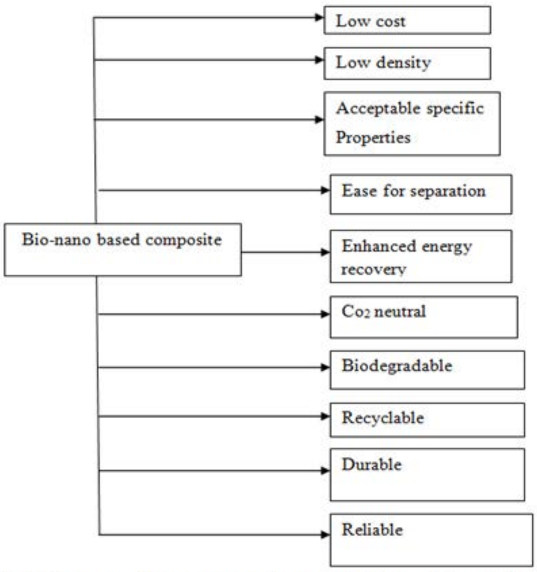

Figure 1: Advantages of Bio-Nanocomposite Materials

1.1. Properties of Bio-Nanocomposite Materials

For packaging applications, the performance characteristics of bio-nanocomposites, such as mechanical, barrier, optical, thermal, biodegradability, and other functional properties, needs to be evaluated [15, 16]. The properties of bio-nanocomposites are closely related to their structure. Significant improvements in properties have been seen with various biopolymer-clay nanocomposites [17]. This is mainly due to the high interface between the clay and the polymer matrix and the proper dispersion of the inserted or exfoliated nano clay [15].

Several studies have been reported on improving the mechanical properties, barrier properties, functional properties, water solubility, thermal stability and other properties of natural polymer-based bio-nanocomposite materials incorporating nanoparticles [18]. In addition, the combination of antibacterial / antioxidant compounds such as essential oils and natural active ingredients with nano clay in chitosan films provides acceptable structural integrity and barrier properties [17, 18]. This finding is due to the fact that nanocomposites do not have the combined effect of nanoclay and antibacterial / antioxidant compounds [19].

1.2. Mechanical Properties

The formation of nanocomposites with organic clay shows that even with low filler filling (<5 wt %), the mechanical properties of various biopolymers are significantly improved [20]. Mechanical properties depend not only on the morphology and dimensions of starting materials such as nanoparticles and polymer matrices, but also on the content of fillers and the formation of infiltrating whiskers networks [21, 19]. Addition of nanoparticles such as nano clay, starch, and cellulose nano fibrils to the polymer matrix can reduce the density of biopolymers [20].

According to it was reported that the tensile strength and modulus of increased monotonically when the filler content increased to 8% and then leveled off. On the other hand, the tensile stress decreased as the filler load increased, except for the 8 % load [22, 23]. The improvement in the mechanical properties of polymer nanocomposites may be due to the high stiffness and aspect ratio of the nano clay, as well as the good affinity of the interfacial interaction between the polymer matrix and the dispersed nano clay from table 1.

Table 1: The Density and Mechanical of the PLA and PHB Biopolymer Nano Composites.

Formulations	Density (g/cm^2)	Tensile strengh (MPa)	Tensile modulus (MPa)	Flexure strength (MPa)	Flexure modulus (MPa)	Load impact strength (kJ/m^2)
Neat PHB	1.22^{a} ±2	8.5^{a} ± 1	583.7^{a} ± 100	12^{e}± 3	1440^{a} ± 250	2.4^{bc}± 0.5
PHB+0.5%CF	1.17^{a} ±1	9.1^{b} ± 2	698.8^{a} ±150	19.3^{c}±5	1977.1^{c} ± 400	2.5^{c}
PHB+2% CNF	1.2^{a} ±0.1	6.4^{a}±01	538.7^{a} ± 100	9.7^{e}± 2	1099^{b} ± 200	1.8^{bc}± 0.5
PHB+4% CNF	1.5^{b}±0.2	16_{d} ± 2	1436.7^{b}± 200	27.4^{d}± 3	1711.8^{c}± 200	2.1^{bc} ± 1
Neat PLA	1.2^{a}± 0.2	42.5^{a}±5	1203.0^{a}± 400	41.6^{a}± 5	3267.7^{a} ± 250	4.4^{a} ± 2
PLA+0.5%CF	1.1^{ab}±01	44.9^{a}±5	1265.0^{a}± 100	44.3^{a}±10	2499.3^{b}± 200	5.1^{ab}± 3
PLA+2%CNF	1.2^{bc}±01	39^{a} ± 5	1308.0^{a}±1500	42.3^{a}± 3	2669.5^{b}± 150	4.3^{a}± 2
PLA+4% CNF	1.18^{b}±0.1	38^{a}± 10	1333.0^{a}± 100	90.9^{c} ± 5	2983.7^{b} ± 200	4.8^{a}± 2

Values with different superscripts along the same columns are significant ($p < 0.05$), as per Tukey test.

Note: PHB: Polyhydroxybutyrate, CNF: Cellulose nano fiber, PLA: Polylactic acid NF: Norfloxacin

According to reported that polymer/CNT [carbo nanotube] nanocomposite, increasing the CNT loading can also enhance tensile strength and Young's modulus provided that uniform CNT dispersion is achieved. That the tensile strength of PCL [Polcaprolactone]/MWCNT [multiwall carbon nanotube] nanocomposite at 0.5wt % MWCNT loading was increased by 42% when compared to that of neat polymer [3, 22, 24]. However, these values were reduced for higher MWCNT loadings due to poor dispersion, arising from the formation.

1.2. Barrier Properties

Nanocomposites can improve barrier properties by providing versatile chemical functions [25]. MVTR [moisture vapor transmission rate], on the other hand, is the amount of water vapor that permeates a particular region at a particular time under specific humidity and temperature conditions and is expressed in g/(m2.day) [26]. Water vapor permeability [WVP -g.mm/ (m2.day)] is the product of presence and thickness (mm) of the sample.

According to reported that such reduction in gas permeability of nano composites strongly depends on the type of clay [i.e., compatibility between the clay and polymer matrix], aspect ratio of clay platelets, and structure of the nanocomposite.

In general, the best gas barrier properties would be obtained in polymer nanocomposite with fully exfoliated clay minerals with large aspect ratios shown in table 2. The increased gas barrier properties of nanocomposite materials are believed to be due to the presence of a regular dispersed silicate layer with a high aspect ratio that is impermeable to water molecules in the polymer matrix [27]. The enhanced gas barrier properties of nanocomposite make them attractive and useful in food packaging applications.

Table 2: Same Reported Data of Addition of ZnO NPs on the Barrier Properties Biopolymer Composites [26].

ZnO NPs Based Composite	Oxygen Transmission Rate /Oxygen Permeability (PO^2)	Water Vapor Permeability (WVP)	Carbon Dioxide Permeability (PCO2)
LLDPE films reinforced with ZnO NPs	OTR decreased by 23.2% for 10 wt. % ZnO incorporation	NA	NA
PLA/ZnO bio composite	For 1 wt. % ZnO incorporation, oxygen permeability (PO2) decreased by 18%. Then there is no further decrease for addition up to 5 wt.%	For 1 wt. % ZnO incorporation, water vapor permeability (WVP) increased by 16%. Then there is no change for higher ZnO content	For 1 wt. % ZnO incorporation, carbon dioxide (CO2) permeability decreased by about 17%. Then there is no further decrease for higher ZnO content.
ZnO/PHB bio-nanocomposite	PO^2 decreased by about 53% at 5 wt.% ZnO NPs loading	WVP decreased by up to 38% at 5 wt.% ZnO NPs loading	NA
PLA-ZnO nanocomposite films	NA	WVP decreased on increasing ZnO NP concentration from 1 to 3 wt.%	NA

Note: LLDPE: Linear low-density polyethylene, NPs: Nanoparticles, PLA: Polylactic acid, WVP: Water vapor permeability, NA: Note acceptable

1.3. Biodegradation Properties

Polymer biodegradation can occur alone or in combination by any of the mechanisms such as hydrolysis, enzymatically catalyzed hydrolysis, solubilization, ionization, or microbial degradation [28-30]. Biodegradation of polymers generally occurs in two different steps: depolymerization and mineralization. Bio-nano composite packaging materials after disposal, it may deteriorate in the environment in a short time [30]. In general, the biodegradability of biopolymer films is known to be significantly improved after formation of nanocomposite with nano clays.

1.4. Antimicrobial Properties

One of the most promising active packaging systems, as antibacterial packaging materials help improve food safety and shelf life by destroying or inhibiting spoilage and pathogenic microorganisms that contaminate food has long been recognized as [31, 32]. Typically, antibacterial packaging films are made by binding antibacterial materials to the polymer surface or blending them during the polymer treatment step [33].

Nanocomposite antibacterial systems are particularly effective due to the high surface area-to-volume ratio and enhanced surface reactivity of nano-sized antibacterial agents, which can inactivate more microorganisms compared to larger counterparts increase [33, 34]. Nanoparticles or nanocomposite materials have been studied for antibacterial activity as growth inhibitors, antibacterial agents, antibacterial carriers, and antibacterial packaging films [35, 36]. One of the most studied nanocomposites used in antibacterial food packaging is incorporated into biopolymer films such as chitosan and starch that exhibit strong antibacterial activity against both Gram-positive and Gram-negative bacteria [34].

1.5. Components of Bio-Nanocomposites

Biomaterial: It naturally comes from plants, animals and microorganisms. It mainly contained cellulose, lignin, hemicellulose, chitosan, PLA (polylactic acid), starch, chitin and PHA [Polyhydroxyalkanoates] [27, 37, 38].

Nanoparticles: They are nano-sized [39]. Nanoparticles are present in spheres, tubes and plates [40]. It can evolve through large particles to form small particles with micro or nano-sized dimensions in fig. 2. Including; layered silicates, nanotubes, spherical particles, etc. [41].

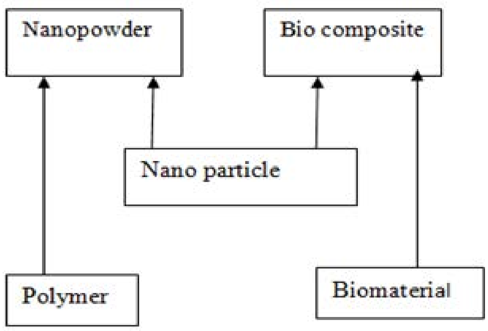

Figure 2: Components of Nanocomposite and Bio- Nanocomposite

1.6. Preparation Methods of Bio-Nanocomposites Materials

In situ polymerization method: In this process, the nanoparticles are premixed with a liquid monomer or monomer solution in fig.3. Polymerization is then initiated by heat, radiation, or the appropriate initiator. In contrast to melt intercalation, layered silicates are mixed with the monomers before the polymerization takes place in-situ [42, 43]. Due to the low viscosity of the monomer [compared to the melt viscosity], it is much easier to evenly mix the particles in the monomer using a high shear mixer [44]. In addition, its low viscosity and high diffusivity increase the rate of diffusion of the monomer into the middle layer region [45]. It is also possible to control the morphology of nanocomposites by combining reaction conditions and clay surface modifications [1]. For most thermosetting polymers, in-situ polymerization is the only viable method for producing nanocomposites.

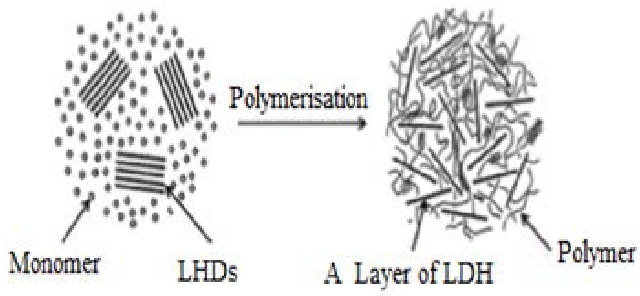

Figure 3: Schematics depiction of Solution Blending Method

1.7. Solution Blending Method

In solution mixing (based on a polymer or solvent system in which the prepolymer is soluble), a solvent or solvent mixture is used to disperse the nanoparticles and dissolve the polymer matrix [46]. Depending on the interaction of the solvent with the nanoparticles, the nanoparticles aggregates can dissolve in a good solvent due to the weak van der Waals forces that stack the layers on top of each other [47, 48]. Solution intercalation is based on a solvent system in which the polymer is soluble and the layered silicates are swelling and dispersible [49]. To produce nanocomposites based on other polymers, the polymer and layered silicate are usually dissolved / swollen separately in the solvent, then allowing solvent molecules to enter the interstices of the layered silicate [50].

Intercalation is done through the polymer chains that are being exchanged shown in fig.4. The polymer chain can then be adsorbed to the nanoparticles. However, when the solvent is removed, the nanoparticles tend to react again. Another disadvantage of this method is that it requires a large amount of solvent [51]. This will increase the cost of the product. The type of polymer that can be used to synthesize the nanocomposite ultimately depends on the choice of the appropriate solvent, limiting the applicability of this method [52]. An attractive route for producing base nanocomposites and layered silicate nanocomposites. Inorganic phyllosilicates are known to separate in water to form colloidal particles.

1.8. Melt Processing

Instead of using solvent as the medium, nanoparticles can be directly mixed with a molten polymer [53]. This process eliminates the use of solvent and is compatible with industrial polymer extrusion and blending processes [54, 55]. It offers an economically attractive route in fabricating polymer nanocomposites. A wide variety of polymer/clay nanocomposites have been prepared via this route, i.e., nylon 6, polystyrene, and polypropylene [56]. Melt intercalation offers a "simple" way of preparing nanocomposites. However, care has to be taken to "fine-tune" the layered silicates surface chemistry to increase the silicate compatibility with the polymer matrix [57, 58]. Many studies have shown that the polar interactions of polymer and clay surface play a critical role in achieving particle delamination/dispersion [55]. For no polar polymers, e.g., PP, a polar compatibilizer such as maleic anhydride–modified PP (polyethylene) (PP-MA) is commonly added to improve the compatibility of PP and clay and thus the clay nanoparticle dispersion [59].

1.9. Characterizations of Bio- Nanocomposites Materials

Spectroscopic techniques including Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), Raman spectroscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), atomic force microscopy (AFM) have been used in different studies to understand the mechanism of enhancement of functional properties [1, 19, 45, 60].

1.10. Application of Bio-nanocomposite

Food packing application: The most commonly used food packaging material is associated with environmental issues as they are non-degradable in nature [61]. The numbers of attempts are made for developing the eco-friendly degradable biopolymers as ideal food packaging in table 3. The biopolymers developed are not commercialized as they have poor mechanical strength and resistance properties [62]. Thus, to enhance the following faults in the reinforcing material are added which resulted in the composites formation. Bio-nanotechnology offers advanced expectation from food packaging that will provide the longer shelf-life, secure packaging with improve traceability of food quality [11, 63]. They are significant due to their nanoscale dispersion with size less than 1,000nm [61].

According to the bio-nanocomposite can be an active food packaging whereby the food packaging can interact with food in some ways by releasing beneficial compounds such as antimicrobial agent, antioxidant agent, or by eliminating some unfavorable elements such as oxygen or water vapor. The bio-nanocomposite can also be a smart food packaging whereby it can perceive property of the packaged food such as microbial contamination or expiry date and uses some mechanism to register and convey information about the quality or safety of the food [18, 64-66]. Improved shelf life and lower packaging costs are the reasons why nanocomposites are being pursued for consumer packaging [67]. Nanocomposites have already led to several innovations with potential applications in the food packaging sector [30].

Table 3: Potential Applications of Nanocomposites in the Food Packaging [65].

Product Feature	Application
Improved packaging performance (mechanical, thermal, barrier properties)	Shelf-life extension, down-gauging of film, reduction in package waste
Thermal stability	Heat resistance, dimensional stability
Optical property	See-through packaging, ultraviolet-screening packaging
Biodegradation	Enhanced biodegradation, environmentally friendly packaging
Active packaging	Shelf-life extension, oxygen scavenger, antimicrobial packaging
Intelligent packaging	Interaction with the environment, self-cleaning, self-healing, indication of deterioration
Delivery and controlled release	Nutraceuticals, bioactive compounds
Monitoring product conditions	Time-Temperature Integrator (TTI), freshness indicator, leakage indicator, gas detector
Nano sensor	Indication of food quality, sensing and signaling microbiological and biochemical changes
Nano coating	Surface reinforcement of base packaging material
Antimicrobial	Active antimicrobial and antifungal surfaces
Information on product	Radiofrequency identification (RFID), nano barcode, product authenticity

1.11. Medical, Electronic and Sensor Applications

Reinforcing biopolymers with nano fillers improves not only mechanical, thermal, and barrier properties of bio-nanocomposites but also their electromagnetic shield, electrical, optical, and magnetic properties [68, 69].

These impressive features are related to increasing environmental concerns to replace conventional composites with bio-nanocomposites, particularly in the manufacture of electric products [70, 71]. Furthermore, transparent and flexible bio-nanocomposites reinforced with nano cellulose are used in various applications, ranging from displays, organic light emitting diode, solar cells to roll-to-roll technology [72]. The continuous deposition of various functional components is required in order to make electronic deposition device [71]. Metal wiring and active gas barrier films are good examples of functional components implemented in roll-toroll technology [73]. Natural abundance, adaptability, and environmental friendliness make bio-nanocomposites a suitable candidate for a wide range of medical applications including, tissue engineering, drug delivery, gene therapy and cosmetic applications [16, 74-80].

2. Conclusion

The development of bio-nano composite materials for food packaging is important not only to reduce environmental problems but also to improve the function of food packaging materials. Bio-nanocomposite materials are a good consideration from an end-user safety perspective. Bio-nanoparticles are used to develop enhanced packaging, active packaging, and smart packaging that help maintain food quality and traceability throughout the supply chain. It promises to develop food packaging with improved properties that help extend the shelf life of food.

Acknowledgments

The authors would like to thank to the research group of Arba Minch University (Ethiopia) and the Department of Chemistry for supporting the writing of this review and the research that drives the author's work in this area.

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