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Promising Polymer Composites for Food Packaging Applications: A Study

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Abstract

The simplest definition of a polymer is a useful chemical made of many repeating units. A polymer can be a three dimensional network (think of the repeating units linked together left and right, front and back, up and down) or two-dimensional network (think of the repeating units linked together left, right, up, and down in a sheet) or a one-dimensional network (think of the repeating units linked left and right in a chain). Each repeating unit is the "-mer" or basic unit with "poly-mer" meaning many repeating units. Repeating units are often made of carbon and hydrogen and sometimes oxygen, nitrogen, sulfur, chlorine, fluorine, phosphorous, and silicon. To make the chain, many links or "-mers" are chemically hooked or polymerized together. Linking countless strips of construction paper together to make paper garlands or hooking together hundreds of paper clips to form chains, or stringing beads helps visualize polymers. Food packaging plays a vital role in preserving food throughout the distribution chain. A package provides protection, resistance, and special physical, chemical, or biological needs. In recent years, the development of novel food packaging (modified atmosphere & active packaging) has increased the shelf life of foods. Moreover, their safety and quality has increased parallely.Nanotechnology offers several advantages to food packaging industry. For example, barrier resistance, functional performance increase with nanotechnology.A recent trend is to use nanocomposites materials for food-packaging applications. Nanoclay is one the most popular material used in food packaging industry. Small plate-shaped particles of clay, 1 nm thick and 1,000 nm in diameter. These tiny partciles are embedded into the plastics normally used for packaging, such as polypropylene and polyethylene.

Keywords:Polymer,Nanoclay,Nanotechnology,Nanomaterial,Food Packaging,Nanosensors

Introduction

Intelligent packagings are able to communicate with the consumers and give information about the product condition through the food chain. These packagings can monitor, trace, or record outer or inner changes that are occurring in the product or its environment . By applying reactive components in the form of nanoparticles and making so-called nanosensors, the packaging will be able to respond to environmental changes, such as temperature, the presence of oxygen, deteriorated products, and microbial contamination . Through the incorporation of nanosensors into food packaging, detection of certain chemical compounds, pathogens, and toxins in food would be possible. This also fulfills the need for inexact expiration dates, which, in many cases, are not suitable for the products due to false estimation of product condition during storage. So they provide real-time status of the food’s freshness . These sensors have many advantages over the costly and time-consuming conventional detection methods, such as High-performance liquid chromatography (HPLC). This includes their speed, high-throughput detection, ease of operation, cost effectiveness, and decreased power requirements

Nanomaterial Migration into Food Matrix

One of the critical issues in food packaging is migration. Migration is the unintentional transfer of packaging materials into the food. This problem may influence the food’s safety and, subsequently, consumers’ health (54. It also can cause undesirable organoleptic changes in the food that is in contact with the packaging. For example, migration of TiO2 into lipid matrix results in rancidity . Nevertheless, in some active packaging, nanomaterials are intended to be released deliberately. Performing migration tests under controlled conditions is essential for evaluating the possibility of the nanomaterials’ incorporation into food packaging and also assessing the associated safety problems. The analysis condition depends on the type of food matrix or food simulant being tested . Different factors impact the migration of nanomaterials into foodstuffs. These factors can be ascribed to the nanoparticles’ properties (e.g., concentration, particle size, molecular weight, solubility, and diffusivity in the polymer), environmental conditions (temperature, mechanical stress), food condition (pH value, composition), packaging characteristics (polymer structure and viscosity), and contact time . For instance, higher temperatures and lower pH values increase the solubility of metal nanomaterials in aqueous solution, leading to an increase in their migration . Low molecular weight polymers that have more free volume accelerate the migration rate and nanoparticles’ diffusivity . There also is an inverse relationship between the migration rate of a system and the size of the nanoparticles . If the nature of the food is compatible with the type of packaging, the food itself may be absorbed into the polymer matrix, enlarging the gaps between the polymer chains, thereby increasing the migration rate . For example, fats have high affinity for polyethylene (PE) and polypropylene (PP), so they may be absorbed by the packaging and cause an increase in plastic mobility and higher migration rates . The preservation method of foods also is important and can affect nanoparticles’ migration. It has been reported that microwave heating causes structural modification of the packaging, thereby speeding up the migration of silver ions . High surface-to-volume ratios in nanomaterials induce an active surface chemistry followed by probably unwanted chemical reactions. This would be troublesome if their presence in the packaging enhances the formation of unforeseen reaction by-products during processing . Nanoparticles also can impact the migration of other packaging constituents, either speeding up or slowing down their migration. To perform migration analysis, the best approach is to use actual food matrices. However, it would be very tedious and difficult to directly estimate the amount of migration into real food matrices due to complex composition of foodstuffs. As a solution to this problem, using natural food stimulants has become an alternative method to measure the specific and overall migration of different packaging substances into foods . The amount and rate of mass transport in the selected food stimulants should be similar to those that occur in the food matrix .

Nanocomposites in Active Packaging

Some nanofillers, such as silver, zinc oxide, and magnesium oxide, have antimicrobial or antioxidant activities. Incorporation of these nanofillers in polymer or biopolymer matrices leads to an inhibiting or retarding effect on the growth of microorganisms, thereby reducing food spoilage . The main goal of active packaging systems is extending the product's shelf life. They can also be designed to improve food quality and safety and finally result in less food waste . Anti-microbial nanocomposite films are worthwhile because of their anti-microbial properties caused by natural anti-microbial agents and because of their suitable structural integrity, which results from the barrier properties created by the nanocomposite matrix. Nanoscale materials have a higher surface-to-volume ratio than their microscale counterparts, and, therefore, they are able to attach to a vast number of biological molecules, which enhances their efficiency . Previous reports have identified potential applications of nanocomposites as growth inhibitors ,bacteriocides , and antibiotic carriers . In addition to the application of nanocomposites as packaging materials, they also can be used as delivery systems by helping the migration of functional additives, such as minerals, probiotics, and vitamins into the food . These controlled-released packagings are another example of a nanomaterial application in active packaging. Many anti-microbial nanocomposites used for food packaging are made from silver, which has an intense toxicity to a large variety of microorganisms . Authors have suggested different mechanisms for the anti-microbial activity of silver nanoparticles, such as increasing cell permeability through the attachment to the cell surface and making pits in the membranes , damaging DNA followed by nanoparticle penetration inside bacterial cell , and the release of $Ag+$. Active nanocomposites are advanced alternatives to conventional active plastic technologies or active sachets for the extension of the quality and safety of packaged food products. The term 'active nanocomposite' generally refers to a plastic composite (i.e., a polymer blend) that contains an active, nanostructured material that confers an activity on the plastic matrix . At least one of the dimensions of the active nanostructured material must be less than 100 nm in size. Nanoclays can be used as carriers for the active agent. The efficacy of the active agent is enhanced because it is highly dispersed in the polymeric matrix and, hence, exposed more efficiently to the substance on which it is required to act.

Important Nanoparticles

To date, many nanoparticles have been identified as fillers for making polymer nanocomposites to improve their packaging performances Table . Among them, clays and silicates have attracted significant attention due to their layered structures. This is because of they are abundant, inexpensive, easy to process, and provide considerable enhancements . There are three main polymer-clay morphologies, i.e., tactoid (or phase separated), intercalated, and exfoliated. In the tactoid structure, which usually occurs in microcomposites, the polymer chains and the clay gallery are immiscible because they have poor affinity for each other. Nanocomposite structures do not display this morphology . In ideal polymer-clay nanocomposites, high affinity would exist between the polymer and clay, leading to exfoliated structures in which polymer chains penetrate into the interlayer space of the clay, making single sheets. If the clay shows a moderate affinity for the polymer, the results would be intercalated structures . In the literature, there are reports of other particle fillers being used, including silver, zinc oxide, titanium dioxide, carbon nanotubes, graphenenanoplates, copper, and copper oxides. It is reported that graphenenanoplates (GNPs) are able to form nanocomposites with improved heat resistance and barrier properties, making them a great option for food-packaging applications. Carbon nanotubes (CNTs) are another type of carbon-based nanoparticles that have good electrical and mechanical characteristics. However, their use has been hindered, mainly due to their high cost and the difficulty in processing dispersions. Copper, copper oxide, titanium dioxide, zinc oxide, and silver have been used mainly for their anti-microbial properties.

Table

Types and characteristics of nanofillers

Type of Nanofiller	**Morphology/Structure**	**Typical dimensions***	**Density**
Montmorillonite (MMT)	Platelets	D: 100–500 nm, T:~ 1nm ^{a}	~ 2.6 g cm^{-3}
Layered double hydroxide (LDH)	Platelets with distinct hexagonal shape	D: 50-few hundred nm, T: ~0.5 nm^{a}	1.5 g cm^{-3}
Carbon nanotubes (CNT)	Tubular, seamless cylinder of graphene sheet	L: up to tens of μm, D: ~ 1/5/20 nm^{b}	1.3–1.5/1.5/1.8–2.0 g cm^{-3b}
Cellulose nanowhiskers (CNW)	Rod-like	L: 100 nm-few μm, D: 2–10 nm	~ 1.6 g cm^{-3}
Microfibrillated cellulose (MFC)	Highly fibrous network consisting of bundles of nanofibrils	L: few μm, D: 10–100 nm	~ 1.6 g cm^{-3}
Bacterial cellulose (BS)	Ribbon-shaped nanofibers^{c} forming a highly fibrous network	L: few μm, W: 50–150 nm, T: 5–10 nm	~ 1.6 g cm^{-3}
Chitin whiskers (CHW)	Slender rods	L: 50–650 nm^{d}, D: 4–80 nm^{d}	~ 1.5 g cm^{-3}
Starch nanocrystals (SNC)	Platelet-like Nanoparticles; often forming few μm size aggregates	L: 20–150 nm^{d}, W: 15–30 nm, T 6–8 nm	~ 1.55 g cm^{-3}

L: length; w: width; T: thickness; D: diameter;

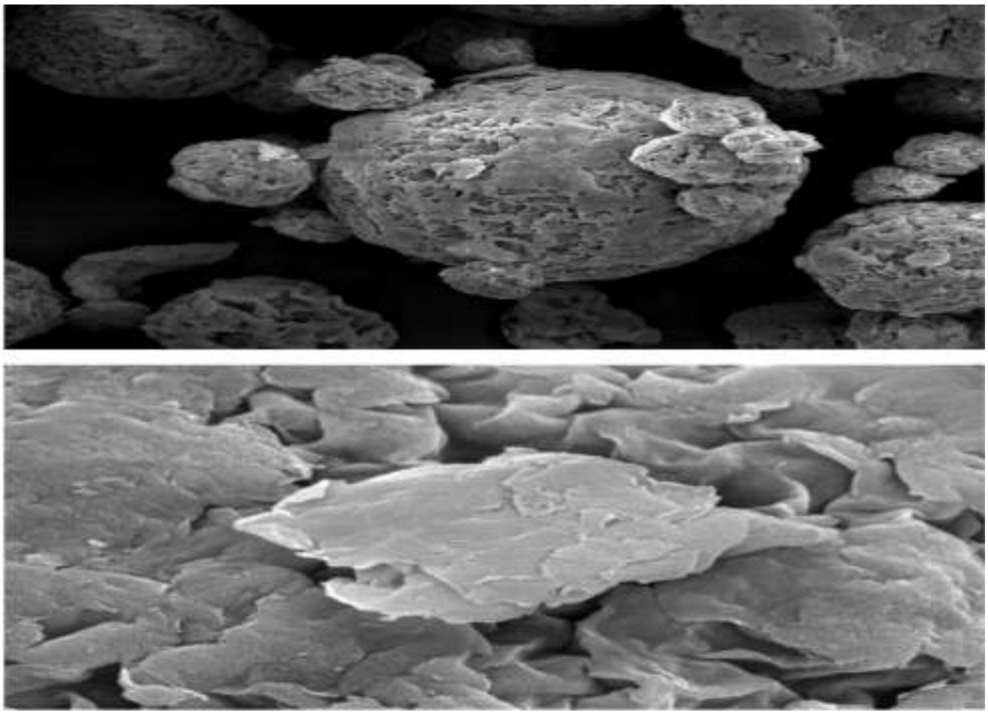
thickness of a single platelet;
for single-, double- and multi-walled CNTs, respectively;
aggregates of individual nanofibrils;
depending on raw material’s origin

Nanocomposites as Degradable Improved Packaging

Currently, there is significant interest in developing different packaging materials because of the increasing demand for foods with minimum processing and longer shelf life. Obviously, such new packaging materials must have excellent barrier properties to prevent the migration of oxygen, carbon dioxide, water vapor, and flavor compounds. This will have a major influence on the shelf life of fresh and processed foods. In the case of using biopolymers, lowering water vapor permeability is especially important due to their hydrophilic nature. However, by the aid of nanocomposite technology, this inherent defect of biopolymer-based packaging materials can be conquered. In many cases, it has been reported that the barrier properties can be improved by about 50% compared to the properties of the neat polymer.This is because of the creation of a maze structure results in a tortuous path for gases and other molecules, thereby reducing their permeation rate. Studies have indicated that incorporating nanocomposites in food packaging materials results in better mechanical and thermal behavior of the packaging. For instance, researchers have reported that engineered nanocomposites of biopolymer-layered silicate have noticeably enhanced physical properties, such as higher tensile strength, enhanced thermal stability, and better gas barrier properties. By using nanocomposites, the food packaging can better tolerate thermal stress of food processing, shipping, and storage. Although traditional composite structures may have large amounts of filler (approximately 60% by volume), nanocomposites usually represent significant changes in properties at quite low loads, i.e., less than 2% volume. It is also suggested that, for achieving the best effects of the final nanocomposites, around 5 wt% of fillers is generally desirable . There is adequate evidence proving other benefits of nanocomposite food packaging, e.g., improved stability of sensory properties, such as flavor, better maintenance of color and texture, increased product stability through the food chain, and less spoilage. Biopolymers can be derived from plant materials as well as animal and microbial products, such as polyhydroxybutyrate. Among these biopolymers, starch, cellulose, and their derivatives , as well as proteins, have been used extensively by many researchers to make bio-based nanocomposites. The most-reported biodegradable nanocomposite is starch-clay, which has been investigated for several applications including food packaging.

Nanoclays in Food Packaging Materials

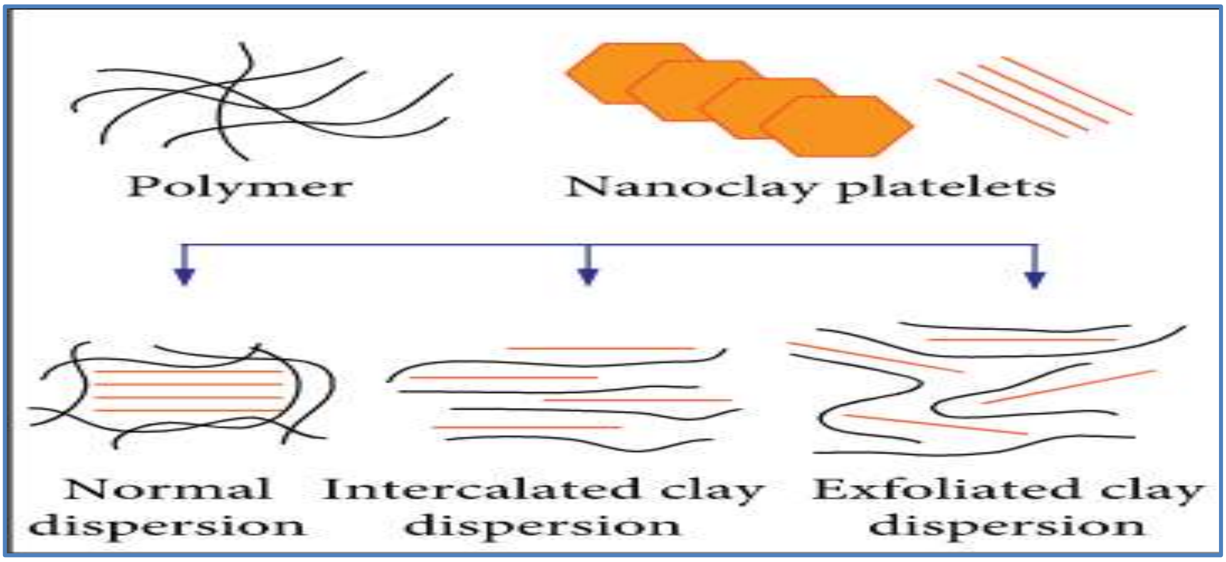
Nanoclays possess a characteristic platelet form, flaky soft structure, low specific gravity, and high aspect ratio with nanoscalethickness . Different types of nanoclays are incorporated into the polymers to improve their characteristics. Among these nanoclays, montmorillonite (MMT, MMT-Na^+) and organophilic MMT (organic modified MMT, OMMT) have gained most attention in the packaging area, from both academic and industrial researchers, because they possess a high surface area with a fairly large aspect ratio (50–1000) and good compatibility with most of the organic thermoplastics . Figure illustrates the flake-like particles of different types of nanoclays. The agglomerated form is observed in a powder format before being processed with a polymer.



SEM micrographs of different types of nanoclays with 3000x and 25000x magnification. (a) Bentonite, (b) Cloisite® 30B, and (c) Nanocor® I.44P.

In nature, MMT is the determinative component in bentonites, which has a hydrophilic surface. It is miscible with only hydrophilic polymers, i.e., poly vinyl alcohol and polylactic acid. However, most of the food packaging materials are from petroleum-based polymers, such as polyethylene and polypropylene that are hydrophobic in nature. To improve compatibility with an organophilic host matrix, the hydrophilic silicate surface (typically Na$^+$, K$^+$, or Ca$^+$) of a nanoclay must be chemically modified with organic cations (i.e., ammonium salt) through ion-exchange reactions to yield an organophilic surface, which exhibits lower surface energy and higher affinity with the polymer . Moreover, the basal spacing of an organically modified clay is expanded because of the bulkiness of alkyl ammonium, thereby improving the penetration level of the polymer chain .Table summarizes various types of commercially available MMTs and OMMTs frequently used for the preparation of food packaging materials.

Nanoclays were first used for food packaging to improve the mechanical and barrier properties of the packaging . The incorporation of an inorganic nanoclay into an organic polymer matrix yields a hybrid material called polymer nanocomposite. By using a lower content of nanoclay, a nanocomposite exhibits lower weight and superior properties than conventional microcomposites. The well-dispersed layered silicates of nanoclays and the confinement of the polymeric matrix at a nanometer level lead to a new class of structural materials . In principle, a nanocomposite can be prepared via four different techniques: solution intercalation, in situ intercalative polymerization, in situ direct synthesis, and melt intercalation. However, the latter technique has gained relatively more interest from industrial and academic researchers because of its cost effectiveness, simplicity, feasibility, and environmentally benign process . In this process, a nanoclay is blended with a polymer by shear force, at a temperature above the softening point of the polymer. The shear force from the blending process, produced by using either the twin screw of an extruder machine or the mixing blades of a Brabender mixer, alters the orientation and dispersion of the nanoclay in the host polymer . The organization of the nanoclay platelets in the polymer matrix is considered to be most crucial for reinforcement. As displayed in Figure , a delaminated or intercalated nanocomposite is obtained when the polymer is located between the interlayers of the stacked nanoclay platelets, while an exfoliated nanocomposite is obtained when the individual nanoclay platelets are well separated and randomly dispersed throughout the host matrix . Figure illustrates the images obtained from a scanning electron microscope. The formation of sheet-like layered structures of nanoclay platelets from the cross-section of an LDPE nanocomposite film with 10 wt% Nanocor® I.44P loading was observed. The parallel orientation of the nanoclay to the film surface was initiated by the shear force produced during the extrusion and film-forming processes.



(a) Scheme showing states of dispersion of nanoclay platelets in a polymer matrix, with permission from the Hindawi Publishing Corporation). (b) Cross-sectional scanning electron microscopy (SEM) images of an LDPE nanocomposite film with 10 wt% Nanocor® I.44P produced by melt intercalation.

Polymers for Nanoclay Composites in Food Packaging

The use of petroleum-based polymers is ubiquitous in food packaging. They are superior materials in terms of their costs, chemical inertness, mechanical strength, weight, and processability. Various polymers are used for different types of food and beverages. The market is dominated by PET and PS for glossy, rigid, and transparent containers; high-density polyethylene (HDPE) for milk bottles and bags; linear low-density polyethylene (LLDPE), low-density polyethylene (LDPE), and polypropylene (PP) for translucent bottles and flexible bags; expanded PS foam for trays; and polyvinyl chloride for overwrapping .

Packaging materials for fast-moving consumer goods, especially food and beverages, have been a major concern for the global environment because of the waste disposal problem . A replacement for conventional plastics from petroleum-based polymers is in high demand because they are produced from nonrenewable resources and are unassimilated by the ecosystems upon disposal. The search for bio-based and biodegradable polymers from renewable carbon resources is being vigorously conducted since the 1970s. The term “bio-based polymers” are either naturally synthesized polymeric materials from plants and animals or entirely polymerized materials with high molecular weight produced by chemical and/or biological methods from renewable resources . However, not all bio-based polymers are biodegradable and vice versa. In general, bio-based polymers are preferred as raw materials over petroleum-based ones, such as polylactide (PLA), polyhydroxyalkanoates (PHA), and starch thermoplastics. Whereas, biodegradable polymers such as polycaprolactone (PCL) and poly (butylene adipate-co-terephthalate) (PBAT) are synthesized from either petrochemical feedstocks or bio-based resources and are biodegradable at the end of their lifecycle . Starch, sugar, cellulose, protein, vegetable oil, lignin, and chitosan are considered as the main renewable resources for bio-based and biodegradable polymers.

Conclusion

In food packaging, the use of biodegradable or natural polymers is restricted because of their poor barrier and mechanical properties. By incorporation of even low percentages of nanofillers, such as clay, into these biopolymers, a considerable improvement in general performance can be achieved. This includes mechanical, thermal, and barrier properties. Moreover, nanoparticles could impart as their active or intelligent properties to food packaging so that they can preserve the food against external factors and increase the food’s stability through antimicrobial properties and/or responding to environmental changes. In spite of several advantages of nanomaterials, their use in food packaging may cause safety problems to human health since they exhibit different physicochemical properties from their macro-scale chemical counterparts. For studying the effect of nanoparticles on human health, more research is needed and identification, characterization, and quantification of the nanoparticles are prerequisite steps. Migration possibilities and safety control of the nano-packaged foods also are crucial. Unfortunately, present legislation does not distinguish nanotechnology-produced materials from those made by ordinary manufacturing approaches.

The organophilicnanoclays not only provide better dispersion in polymers but also offer additional advanced features for packaging materials. Several studies proposed various promising novel functions of nanoclays in food packaging as an antimicrobial agent, control and release for active ingredients, colorimetric indicator template, and additive partitioning, which expand the application area of nanoclays. The lack of legislation for nanomaterials with respect to consumer and environmental safety appears as a restriction in the use of clay nanocomposite films in food packaging. However, the available migration studies revealed that the diffused levels of aluminum and silicon in such packaging are within the limitations of the current regulations.

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