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Review

Clay Minerals and Biopolymers in Film Design: Overview of Properties and Applications

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Abstract: Research to replace petroleum-based plastics has been quite challenging. Currently, there is a lot of interest in biopolymers as an alternative. However, biopolymers do not have suitable mechanical properties when in film form, which limits their applications. To resolve this issue, clay minerals are being incorporated as a strategy. Clay minerals offer the films good barrier, thermal, rheological, optical, and mechanical properties. They can also work with other additives to promote antioxidant and antimicrobial activity. This brief review focuses on incorporating clay minerals with other nanofillers and bioactives to improve their physical, chemical, and functional characteristics. The synergy of these materials gives the films exceptional properties and makes them suitable for applications such as food coatings, packaging materials, dressings, and bandages for treating skin wounds.

Keywords: clay minerals; biopolymer; film formation; nanofillers; food packaging; wound dressing

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1. Introduction

Clays such as bentonite, laponite, and kaolin, as well as clay minerals like montmorillonite, kaolinite, halloysite, sepiolite, and palygorskite, are commonly utilized as reinforcing agents in polymer nanocomposite materials. These minerals have intrinsic characteristics like high surface area, ion exchange capacity, versatile surface chemistry, and good dispersibility in polymeric matrices. Additionally, they are non-toxic and cost-effective, making them ideal for various technological applications [1–3].

Clay minerals can interact with biopolymers through different mechanisms (intercalation, exfoliation, or interactions on the external surface), resulting in materials with significantly altered properties. These materials, known as bionanocomposites, generally exhibit unique consistency and thermal, structural, mechanical, and optical properties. They can also be biocompatible and biodegradable, making them suitable for applications in areas such as sensors, catalysis, textiles, electronics, and bioplastics [4,5].

The potential use of biopolymers such as starch, gum, alginate, chitosan, gelatin, pectin, protein, and others as alternatives to conventional food packaging materials that use synthetic polymers has garnered significant interest due to the increasing popularity of sustainability and environmental protection [6]. In addition to their environmentally friendly attributes, biopolymers also help to prevent the growth of microorganisms in food, thus extending its shelf life. Biopolymers are also utilized in the field of regenerative therapy for skin dressings. Their biodegradability and compatibility with superficial skin tissues make them helpful in regenerating or healing tissues damaged by injuries, with the goal of tissue repair and maintaining anatomical integrity [7].

Among biopolymers, alginate, starch, and chitosan are the most commonly used. Alginate is a polysaccharide obtained from the intracellular matrix of brown algae. It has low toxicity, biocompatibility, and biodegradability, making it suitable for application in drug delivery and dressings [8]. Starch is a polysaccharide obtained from different botanical sources, such as cereals, tubers, or roots. It is widely used in food packaging due to its non-toxicity, biodegradability, and low cost. Additionally, it is obtained from renewable sources and is highly abundant in nature. Starch granules are generally sensitive to moisture, and to overcome this problem, they are mixed with other materials to form films [9]. Chitosan is a biopolymer obtained from chitin, which is found in the exoskeleton of mollusks and crustaceans. These polysaccharides have crystallinity, hydrophobicity, high nitrogen content, high charge density, high viscosity, and the ability to form films. They are suitable for various applications such as biomaterials, tissue engineering, drug delivery, cosmetics, and dressings, as well as applications in agriculture, nutrition, and the food industry [10]. Gelatin, obtained by denaturing collagen, is also a widely used biopolymer for film formation. These natural polymers are utilized in studies developing films for application in food packaging as they are edible, biodegradable, and compostable. However, gelatin-derived films have limited barrier and mechanical properties, which can be improved with the addition of nanofillers [11]. Pectin, obtained from the skin or pulp of different fruits, has been used in the production of edible films in the food industry. However, the films have poor mechanical, thermal, barrier, and water resistance properties. Studies have revealed that interaction with organic or inorganic materials can overcome these limitations [12].

Nanocomposites made by combining biopolymers with inorganic nanofillers are gaining importance in the literature [13]. When the macromolecule matrix is functionalized with these materials, they exhibit improved physical, chemical, and biological characteristics, making them promising materials for new research in eco-friendly food packaging, biomedical applications, and other fields. The following studies explored the development of biopolymer films with various additives: Heydari et al. [14] manufactured a film containing starch, bentonite, and vitamin B2. They observed thermal stability, good mechanical properties, and controlled vitamin release, making the films suitable for applications in food, pharmaceuticals, or other areas. Aguirre-Loredo et al. [15] prepared a starch and chitosan film containing different concentrations of montmorillonite. The authors concluded that there are strong interactions between the clay mineral and the biopolymer blend, leading to improved physical–chemical properties. In this case, the homogeneous distribution of montmorillonite prevented the formation of aggregates on the surface. Thermal stability and gelatinization of the films were also observed, leading to improvements in mechanical properties. Das et al. [16] developed a protein film reinforced with montmorillonite and debittered kinnow peel powder. The authors observed a decrease in moisture content, solubility, and swelling ability, as well as increased resistance, permeability, water vapor transmission rate, and antimicrobial activity.

The following overview explores the use of different nanofillers, particularly clay minerals, in various biopolymer matrices. The focus is on the interesting effects of combining these valuable materials to create functional films. This work aims to compile relevant studies that utilize clay minerals mixed with other active agents in biopolymer films. It starts with a succinct introduction to the characteristics of bionanocomposites, clay minerals, and biopolymers as well as the main film synthesis methods. The major clay minerals used in these studies are highlighted, as well as the primary nanofillers combined with these clay minerals and the properties they affect. Finally, this work emphasizes the latest achievements in using various biopolymer matrices functionalized with clay minerals and their applications, particularly in packaging, improving the quality and shelf life of foods, and in biomedical uses such as wound healing, including wound dressings for burns or exuding wounds.

2. Different Methods of Manufacturing Biofilms

Different preparation methods can be used to develop films based on biopolymers. The manufacturing method and possible post-treatment can alter the surface roughness, rigidity, and functionalities of the film.

The casting solution method is the most commonly found in the literature [17–20]. This method involves gelatinizing the polymer and is easy to operate and affordable. To create the films, a uniform precursor solution containing chitosan, alginate, protein, pectin, gum, cellulose, or starch is typically mixed with glycerol, which acts as a plasticizing agent. The mixture is then carefully poured into Petri dishes and left to dry at room temperature until the solvents evaporate and the films are formed.

Coating is another common method for preparing biopolymer-based films for covering different food products [21–23]. This method involves coating food products with a protective solution to study their ability to resist natural decomposition. The protective solution is applied to the food covering and allowed to dry at room temperature, forming a dry film. The food is then observed for any physical or chemical changes preserved by the protective film.

Electrospinning is a technique that is used to create ultra-thin membranes from various materials, such as polymers, semiconductors, ceramics, and composites. This technique involves the ejection of a charged straight jet of a polymeric solution, where an electric field is applied. The solvent is evaporated before the jet reaches the collector, and the nanofibers are ultimately deposited in the collector [24]. Clay mineral and biopolymer electrospun films improve the nanofilm's resistance, biocompatibility, biodegradability, and drug release, making them suitable for wound therapy [25].

Blown extrusion was also a method found in the literature for manufacturing films based on starch, chitosan, or gelatin [26,27]. This processing method is cost-effective, continuous, and energy-efficient. It can result in the complete plasticization of films and the production of a uniform and bubble-free material. The method uses minimal amounts of water and plasticizers, relying on the thermoplastic properties of biopolymers.

Electrospinning is an inexpensive technique that creates flexible films with high surface area and porosity, making them suitable for various applications, mainly for wound healing. However, to produce good films, several parameters such as the viscosity of the polymer solution, solvent choice, voltage, jet flow, etc., must be carefully controlled, bringing challenges. The extrusion process is continuous, allowing for high production rates, and is well suited for large-scale use. This makes it very valuable in food packaging production. On the other hand, casting and coating methods are typically employed on a laboratory scale due to their low cost, ease of preparation, and minimal energy consumption without toxic products. However, these methods may have limitations, such as long production times and restricted film sizes. Nonetheless, they are appealing because they enable the direct application of films to various surfaces and foods, making them useful in wound dressing and food packaging management applications.

3. Relevant Characterizations and Properties

Biofilms exhibit varying characteristics, which can be investigated through various structural, morphological, and physical–chemical approaches. The most commonly used techniques for such investigations include X-ray diffraction, vibrational spectroscopy, thermal and morphological characterizations, swelling experiments, and water vapor transmission rate measurements, as well as the assessment of mechanical and biological properties. A brief description of these analyses is provided below.

X-ray diffraction (XRD) is a non-destructive analytical technique that is widely used for characterizing different materials. It helps in identifying the crystalline structure, phases, degree of crystallinity, and size of crystallite of a material [28,29]. For instance, Jin et al. [30] conducted a study to evaluate the crystal structure of a film made by combining soy protein isolate, alginate, silver nanoparticles, and aminoclay. In the XRD patterns, the authors observed the exfoliation of the aminoclay, the peak corresponding to silver, and the changes in the molecular conformation of the protein after interaction with the aminoclay. Naidu et al. [31] created a film made of xylan–alginate and incorporated nanofillers like halloysite and bentonite. They analyzed the film using X-ray diffraction (XRD) and found that the polymeric material was intercalated in the bentonite, causing an increase in the interlayer spacing from 4.38 to 4.52 Å. Similarly, in films reinforced with halloysite, the interlayer spacing of the clay mineral increased from 7.03 to 7.21 Å after interacting with the polymer matrix.

Vibrational spectroscopy is a useful tool to investigate short-range alterations in materials. It can help identify the functional groups of materials and the presence of new species after chemical or physical modifications. There are many techniques available, but the most common ones are infrared spectroscopy using traditional transmission methods like KBr-pellet or multi-techniques, Fourier-transform infrared spectroscopy (FTIR), and modern reflectance techniques like diffuse reflectance (DRIFT) and attenuated total reflectance (ATR) [32]. These techniques can be used to characterize raw materials, their properties, and the interaction mechanisms of other compounds. Bourakadi et al. [17] conducted a study where they developed chitosan films that were functionalized with organo-montmorillonite. The authors analyzed the films using FTIR and found the presence of thiabendazole, alginate, and montmorillonite as bioactive agents. They also observed that these agents were dispersed in the chitosan film through intermolecular interactions.

Thermal analysis (TGA, DSC, and DTA) is a method used to assess the thermal stability of biofilms and their modified forms. It also allows for the determination of the organic content in the films [33]. By measuring the mass loss, it is possible to determine the success of the interactions between organic and inorganic molecules, complementing results obtained by XRD and vibrational spectroscopy [34]. The study focuses on the relationship between a sample's properties and its temperature as the sample is heated or cooled in a controlled manner. Differential Scanning Calorimetry (DSC) is used to determine the transition temperature, heat capacity transferred, specific heat capacity, reaction temperature, reaction enthalpy, and examination of thermal history. Differential Thermal Analysis (DTA) is used to determine the transition temperature and reaction temperature, and Thermogravimetric Analysis (TGA) is used to determine dehydration, oxidation, pyrolysis, evaporation, sublimation, etc. According to research conducted by Ding et al. [35], an alginate film that contains 4% palygorskite was evaluated for its thermal properties. The TGA results revealed that the films containing clay mineral were more stable due to the strong interactions between the alginate matrix and the palygorskite reinforcement. The DSC results showed that these interactions influenced the T_g (glass transition temperature) of the material, where the presence of palygorskite fillers increased the T_g temperature of the film compared to the alginate-only film.

Scanning electron microscopy (SEM) is a valuable tool for analyzing the shape and morphological structure of materials. When studying biofilms, these images can reveal if there are any contaminants, air pockets, fractures, uneven surfaces, or boundaries between the matrix and other substances [31,33]. Additionally, microscopic techniques can provide insight into the dispersion of clay filler within the polymer matrix, indicating adhesion quality and the presence of aggregates. In addition to generating images, microscopes often use Energy Dispersive Spectroscopy (EDS) to detect chemical elements on the sample's surface. In the studies of Nozari et al. [36], based on the SEM image results of chitosan/alginate and chitosan/bentonite films, it was observed that the films containing sodium alginate had rough and wrinkled surfaces. The surface roughness of the films increased with the increase in the alginate concentration. On the other hand, the surfaces of the bentonite films were smooth and uniform and had no cracks. This could be due to a strong affinity for hydrogen bonds between the bentonite and chitosan structures. Even with different clay concentrations, the films did not show any phase separation of the components.

The degree of swelling in biopolymer-based films can be measured by direct contact with a solvent, usually water. As soon as solvent molecules diffuse into the pore system, the material swells rapidly and becomes highly permeable. The porosity of the material determines the mobility of the solvent within the particles [37]. The degree of swelling is expressed as SD%, which is calculated using the formula $SD\% = ((m_1 - m_0)/m_0)) \times 100\%$. Here, m_0 is the mass of the dry material, while m_1 is the mass of the swollen material. Das et al. [16] investigated the swelling properties of a film made by combining soybean meal protein, montmorillonite, and debittered kinnow peel powder. The authors found that adding montmorillonite significantly reduced the swelling of the biofilm, indicating strong interactions between the hydroxyl groups of the clay mineral structure and the amine groups of the protein matrix. The reduction in swelling capacity was directly proportional to the concentration of montmorillonite added. These results demonstrate the creation of a water-resistant material, which is an important factor for protecting food products.

The water vapor transmission rate (WVTR) is the standard method used to measure a film's ability to resist moisture transmission. Under specific conditions of temperature and relative humidity, water vapor permeates through the film. Permeability is the rate of water vapor transmission per unit area through a film of known thickness [31,33]. These factors can influence the properties of the film. According to Shankar et al. [38], the inclusion of halloysite that has been functionalized with sodium hydroxide and ZnO nanoparticles enhanced the barrier characteristics of the alginate biofilm. The study reported that films containing functionalized halloysite showed a decrease in water vapor permeability and an increase in hydrophobicity, when compared to pure alginate films.

When examining films for their mechanical properties, tensile strength (α), elongation (ε), and Young's modulus (E) are the three most important parameters to consider [39,40]. It is important to assess the mechanical properties of films to ensure their reliability during manufacturing, storage, and application. Tensile strength at break is the maximum stress that the film can withstand before breaking when stretched. Elongation break (%) indicates the film's ability to withstand deformation without breaking. Young's modulus provides information about the stiffness and flexibility of the film, which correlates with its chemical composition [41]. In a study conducted by Giannakas et al. [42], interesting findings were obtained regarding the mechanical properties of chitosan/montmorillonite and chitosan/glycerol/montmorillonite films. The researchers observed that the addition of clay minerals to chitosan films increased stiffness and resistance but decreased elongation. However, in films containing glycerol, the addition of clay minerals resulted in a decrease in stiffness and resistance but an increase in elongation. These results led the authors to conclude that the distribution of water and glycerol in the system results in better plasticization of the films. The interactions between montmorillonite and the polymer matrix lead to the formation of an extensible and resistant film.

The effectiveness of biofilms against the growth of bacteria and fungi, as well as their cytotoxicity, is an essential factor to consider when studying films for application in food packaging and skin wound dressing. Treating wounds is a major challenge for public health due to the risk of infections. As a result, researchers are working on creating dressings that are non-toxic, provide mechanical protection, are easy to apply, and have therapeutic properties [43]. Similarly, food safety is a major concern in the food industry. Therefore, it is crucial to develop protective materials for different food products to eliminate or reduce microorganisms that cause food spoilage and infections in consumers [44]. Kanmani et al. [45] demonstrated that the antibacterial activity against *Listeria monocytogenes* of gelatin/AgNP/clay films increased significantly with the addition of the organoclay Cloisite 30B. According to Ambrogi et al. [46], films that are composed of montmorillonite/chitosan/chlorhexidine have exhibited promising properties against a variety of microorganisms, including *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Pseudomonas aeruginosa*, and *Candida albicans*. The researchers also noted that the presence of montmorillonite in the films reduced the cytotoxicity associated with biofilms.

The antioxidant property is a chemical characteristic of compounds, particularly phenolic compounds, which can inhibit the harmful effects of free radicals. In the food industry and biochemistry, this property, along with antimicrobial, anti-inflammatory, and immunomodulatory abilities, enhances the functionality of films and underscores the importance of incorporating these antioxidant compounds. Various studies have already shown the antioxidant potential of different compounds in forming films for use in food preservation and wound dressings [47–51].

The addition of clay minerals and other nanofillers influences the physical and chemical characteristics of materials. In the food industry, it is important for materials to have resistance, flexibility, optical transparency, and water and gas permeability. The concentration of nanofillers, particularly clay minerals, should enhance the tensile strength without affecting the optical transparency of the films. When adding clay minerals and metallic nanoparticles, it is essential to achieve antimicrobial activity without causing agglomeration, which could compromise the mechanical properties. For wound dressing applications, films must protect the wound, prevent dehydration, and promote healing by being flexible, porous, waterproof, and capable of releasing bioactive compounds. Therefore, it is crucial to produce films that strike a balance between their components to achieve the best possible characteristics.

Incorporating clays or clay minerals in biopolymer-based films can modify their properties. When biofilm structures are combined with inorganic compounds, it affects their structure, morphology, optical properties, and therapeutic effect and improves their barrier properties, thermal stability, and resistance. Different clays and clay minerals have already been successfully used—as nanofillers—in films for various applications. Figure 1 illustrates the different properties that are influenced by the interaction of clays and clay minerals with biopolymer-based films.

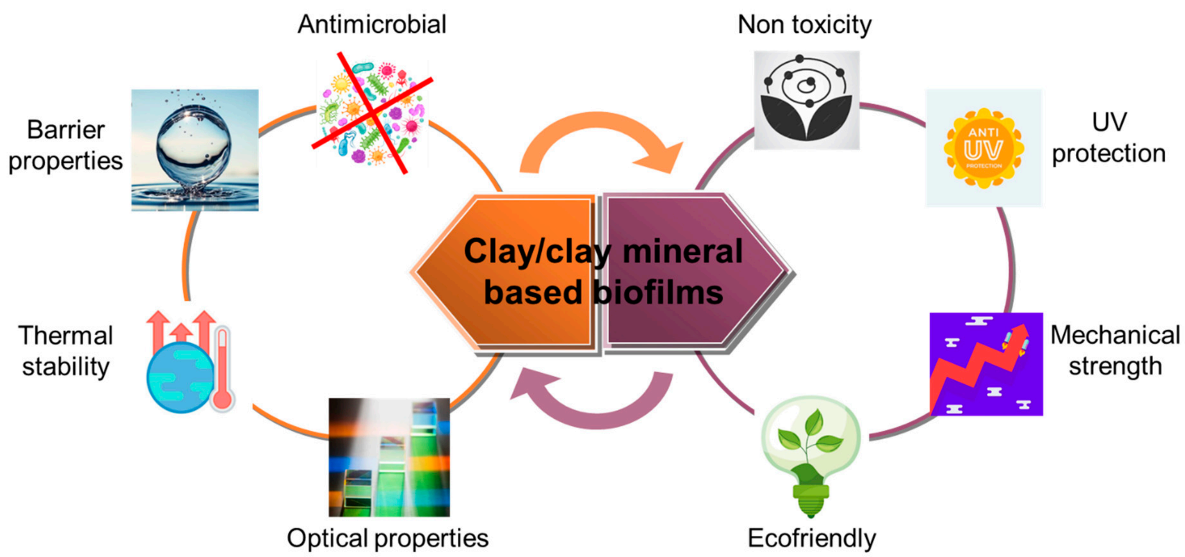

Figure 1. Properties of films obtained by combining clays and clay minerals with biopolymers.

4. Main Clays and Clay Minerals Used in the Composition of Biofilms

Bionanocomposites are nanometric materials consisting of biopolymer matrices with a dispersed phase of the inorganic solids [52]. The properties of these biocomposites can be improved by incorporating clay minerals. Clay minerals are one of the most favorable nanofillers as they help to control the properties of the nanocomposite films. To obtain well-structured films, it is important to have a homogeneous distribution of clay mineral nanoparticles in the biopolymer matrix. The dispersion of clay minerals in a biopolymer matrix can be labeled depending on the degree of separation of the nanoparticles as phase separated, intercalated, or exfoliated [53]. Phase separation does not influence the layered structure of the clay mineral. Intercalation produces a material with a well-ordered morphology, with the biopolymer between the layered spacing but with the clay mineral structure intact. On the other hand, exfoliation completely separates the layer structure resulting in greater dispersion. Figure 2 depicts the biopolymer–clay mineral nanocomposites.

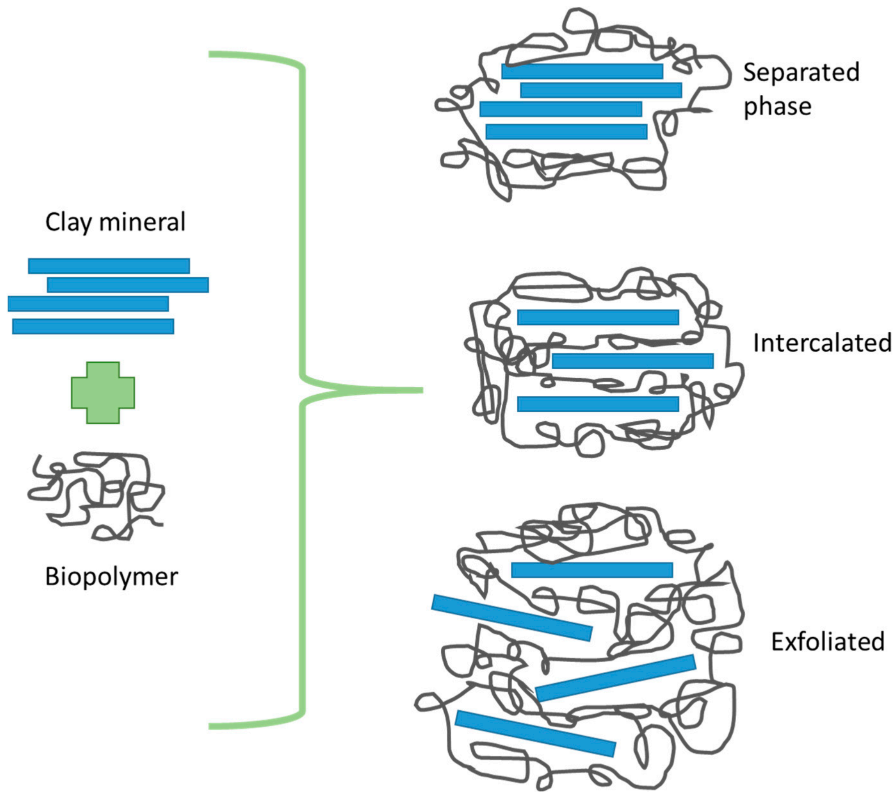

Figure 2. Different structures of nanocomposites obtained by the interaction between clay minerals and biopolymers.

Clay minerals possess several interesting properties such as ionic exchange capacity, high surface area, adsorption capacity, and great surface reactivity. Due to their unique physicochemical properties and structural characteristics, clay minerals are highly versatile, abundant, biocompatible, and affordable in cost, making them suitable for use in various areas including biotechnology, environmental science, food industry, and biomedicine [54–56]. In particular, bionanocomposite formation has received significant attention in using clay minerals. Different clay minerals have been used to prepare biopolymer-based films. In this study, we will discuss the characteristics of the most commonly used ones.

Clays and clay minerals are groups of minerals that can have varying chemical compositions, structures, and modes of occurrence. They belong to the family of minerals found in the Earth's crust and are formed from source rocks under different conditions. These minerals are abundant and possess remarkable properties [29]. Clays are natural materials that have fine grains of less than 2 or 4 μm with nanometric dimensions in their layered structure (layer and interlayer space). When moistened, they become plastic and harden when heated. Clays are classified based on their origin, major clay mineral constituent, and properties. Clay minerals are hydrated and plastic phyllosilicates. They can be natural or synthetic and do not have a defined grain size. Clay minerals are classified into two types, planar and non-planar. Planar phyllosilicate species are further classified based on their interlayer material, layer design, and octahedral character. Non-planar clay minerals are classified based on their modular component and configuration [29].

The basic crystalline units of clay minerals are divided into two types: tetrahedral (T) and octahedral (O). Tetrahedral units consist of Si^{4+}, Al^{3+}, or Fe^{3+} species surrounded by four oxygen atoms. Three oxygen atoms of each tetrahedron can be shared by three neighboring tetrahedra to form a continuous and hexagonal arrangement horizontally, known as the tetrahedral sheet. Similarly, octahedral units contain Al^{3+}, Mg^{2+}, Fe^{3+}, or Fe^{2+} species surrounded by six oxygen atoms. They can be linked together horizontally by sharing their edges to form the octahedral sheet. The combination of tetrahedral and octahedral sheets connected by shared oxygen atoms forms clay mineral structures, which can occur in two layer combinations, 1:1 (T-O) or 2:1 (T-O-T), as shown in Figure 3. Type 1:1 has an octahedral sheet bonded to a tetrahedral sheet. The 2:1 arrangement consists of an octahedral sheet sandwiched between two tetrahedral sheets. Isomorphic substitutions in tetrahedral or octahedral sites promote the formation of a negative layer charge, which is compensated by the presence of different cations in the interlayered spacing [52,57–59].

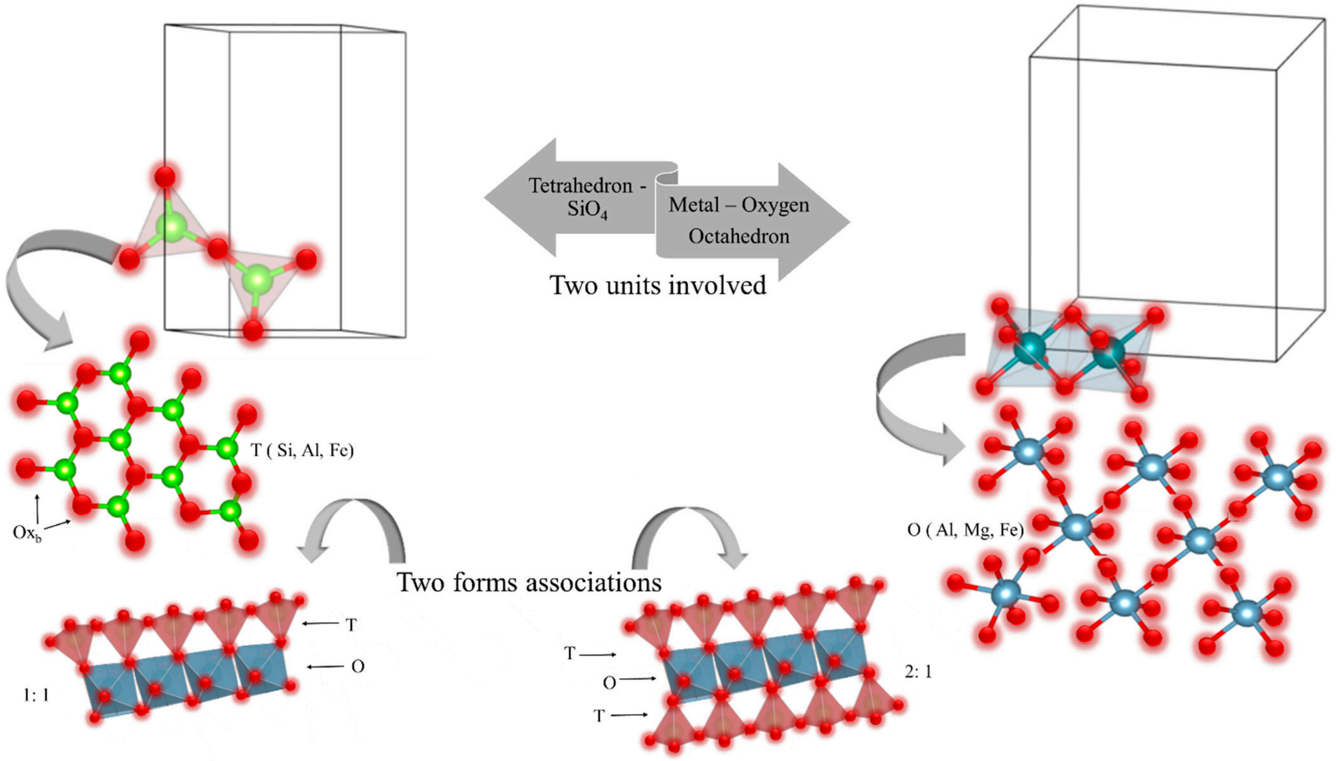

Figure 3. Clay mineral structures formed by tetrahedral and octahedral sheets combining in 1:1 (TO) and 2:1 (TOT) arrangements, shown using Vesta [60]. These sheets are made up of silicon tetrahedra and aluminum octahedra.

Montmorillonite is the main component of bentonite clay. This clay mineral has a 2:1 structure that is formed by two tetrahedral sheets of silicon and an octahedral sheet of either aluminum or magnesium. Montmorillonite is the most commonly used clay mineral in studies related to biofilms, such as those applied to active packaging [49], edible packaging [61], biodegradable films [62], and wound healing [63]. The abundance, affordability, and rich intercalation chemistry make this clay mineral highly attractive for industrial and research purposes. Giannakas et al. [49] developed chitosan films that were functionalized with either montmorillonite or organo-montmorillonite that was modified with thyme essential oil. The structural analysis showed that there were good interactions between chitosan and clay minerals. The mechanical tests indicated an increase in the rigidity and strength of the films but a slight decrease in elongation when the clay mineral was combined with the essential oil. In addition, the mechanical properties of biofilms improved when montmorillonite or organo-montmorillonite was added to PVA. Water and oxygen permeability measurements were also improved when clay minerals, organo-clay minerals, and polyvinyl alcohol were added. Organo-montmorillonite films showed better antioxidant and antibacterial activities than chitosan and chitosan/montmorillonite films. These films hold great potential as active packaging for extending the shelf life of foods. Rostami and Esfahani [61] developed a smart edible nanocomposite using mucilage obtained from the seeds of Melissa officinalis, montmorillonite (MMT), and curcumin. The concentration of clay mineral nanoparticles in the composite had a significant impact on various physical properties of the films. The thickness, moisture content, water solubility, and vapor permeability of the films decreased, whereas the tensile strength and elongation at break increased with an increase in clay mineral concentration. This phenomenon can be attributed to the strong hydrogen bonds between the montmorillonite structure and the biopolymer. Additionally, the films containing clay minerals and curcumin exhibited excellent antimicrobial activity and were sensitive to the pH of the environment. They are ideal for monitoring packaging for spoilage reactions in food. The research conducted by de Souza et al. [62] demonstrated the creation of biodegradable films using cornstarch, carvacrol essential oil, and montmorillonite. The findings suggested that the presence of clay minerals improved the thermal stability of the films. The authors observed that montmorillonite slowed down the decomposition of starch and reduced the volatility of the essential oil, which can be explained by the strong interaction between the organic structures and the clay mineral. XRD analysis revealed that the biopolymer was intercalated within the interlayer spacing of montmorillonite, and the films formed were amorphous in nature. FTIR analysis showed hydrogen bond interactions between the biopolymer and montmorillonite. Antibacterial assays demonstrated excellent activity against *E. coli* colony growth. Naseri-Nosar et al. [63] developed a PLA film that has chitosan modified with curcumin and montmorillonite, which can be used to promote wound healing. The structural results of the film showed that there were electrostatic interactions between the montmorillonite, chitosan, and PLA microfibers. The wettability assessment demonstrated that the presence of montmorillonite increased the contact angle. Additionally, the incorporation of montmorillonite and chitosan increased the vapor permeability of the film, but the changes were not significant compared to uncoated microfibers. The MTT assays showed that the films did not present cytotoxicity. In vivo tests demonstrated high wound closure due to the synergistic effect between montmorillonite, chitosan, and curcumin. Therefore, the films are suitable for use in wound dressings.

Laponite is a type of synthetic clay that contains layers of magnesium–lithium silicate. It belongs to the group of trioctahedral smectites and has structural similarity to natural hectorite [64]. The mineral's structure is composed of silicon tetrahedra and magnesium or lithium octahedra, which form 2:1 layers. These layers contain mostly hydrated cations of sodium and lithium in the interlayer spacing [65]. Laponite is a synthetic clay that is cost-effective for large-scale production, with controlled crystallinity, grain size, and low impurities, making it highly suitable for pharmaceutical applications. The study of Wu et al. [66] focused on creating biofilms using a casting solution made from chitosan/laponite/AgNPs. The XRD results showed that the laponite structure modified with silver nanoparticles was completely exfoliated after being incorporated into the chitosan film. The micrographs also revealed a more uniform surface as compared to pure chitosan. Furthermore, FTIR analysis indicated that there were electrostatic interactions and hydrogen bonds between the laponite and the polymer matrix. The authors found that the tensile strain and strength increased with the degree of laponite incorporated into the film but then decreased due to the accumulation of clay minerals that affected the internal structure of the film, reducing its mechanical resistance. The addition of laponite and AgNPs made the film more dense and compact, reducing solubility and swelling. Moisture was also reduced as the hydrogen bonds between the structures limited them from forming hydrophilic bonds with water. Additionally, the padding of the film structure limited the diffusion of water vapor through the film. The presence of laponite and Ag decreased oxygen permeability as interactions and pore filling made it difficult for oxygen to pass through the film structure. These results indicated that the barrier properties improved with the addition of silver-modified laponite. The biofilms also showed effective antimicrobial activity against *S. aureus*, *E. coli*, *A. niger*, and *P. citrinum*, making them effective in protecting lychees. Pineda-Álvarez et al. [67] created new films using biopolymers gelatin and lecithin and incorporated laponite nanoparticles modified with the drugs maltodextrin and sodium ascorbate. According to the study, when the laponite and drugs were gradually added, the thickness of the films increased. The introduction of laponite had a positive impact on the tensile strength of the films but decreased the Young's modulus. The addition of drugs resulted in the increased elasticity of the films. Additionally, laponite enhanced resistance. The films had an ideal transmittance of less than 10%, allowing for slight light passage due to the presence of laponite nanovoids. The combination with drugs made the films more homogeneous and suitable as a light barrier. In bioadhesion tests, moistening the films resulted in positive results as the degree of swelling favored interactions between biofilms and the skin. This made the dressing efficiently attached to the wound but easy to remove without causing damage to the injured tissue. DSC tests revealed that laponite platelets affected the glass transition and melting temperature of the films, increasing the thermal stability of the films. Scanning electron and atomic force microscopy showed a reduction in roughness with the presence of clay, increasing the homogeneity of the films. The dressings were suitable for the controlled release of drugs and ideal for application in wound healing.

Kaolinite is the primary component of kaolin clay. Its structure consists of silicate sheets linked with aluminum hydroxide sheets, forming alternating layers of the 1:1 type [68]. Kaolinite is a fascinating clay mineral for use in bionanocomposites, primarily due to its abundance, high mechanical stability, good surface chemistry, and ability to interact with different biopolymers. Kwasniewska et al. [69] developed potato starch films that were functionalized using kaolin clay. The results of the study showed that the concentration of kaolin had a significant impact on the mechanical properties of the biofilms. The tensile strength, Young's modulus, and Poisson's ratio of the films decreased as the concentration of clay increased. The X-ray diffractograms of the films revealed that they had a semi-crystalline structure and that the ordered structure of kaolin was not significantly affected after being incorporated into the polymeric matrix. The AFM images showed that the roughness of the films increased with the concentration of clay. The hydrophobicity of the films varied according to the concentration of clay, which is likely due to the different interfacial interactions between kaolin and the biopolymer structure. The calorimetric data indicated that the melting temperature of the films decreased with increasing clay concentration, while the polymer degradation temperature increased. This increase can accelerate the biodegradability of the film. Kaolin incorporation also affected the water vapor permeability of the films. The film that contained 15% clay had a higher barrier value when compared to the pure starch film. The authors associated these results with the incorporation of layered clay structures arranged parallel to the surface of the polymeric matrix, which likely hindered vapor diffusion, decreasing permeability. Tabassum et al. [70] recently developed a biodegradable film using kaolin, PVA, and potato starch. The authors experimented with different compositions to determine the optimal formulation for the film's production. The study found that the presence of kaolin had a significant impact on the film's tensile properties. A higher concentration of clay increased the film's tensile strength, whereas a lower concentration of PVA decreased the film's elongation break. The interaction between PVA and kaolin played a crucial role in determining the material's resistance, while the amounts of PVA and starch were decisive in achieving the right balance between resistance and film elongation. The wettability results showed that an increase in kaolin concentration led to a decrease in water adsorption. This is because kaolin increases the film's hydrophobicity due to its interaction with biopolymers, reducing its ability to interact with water. Moisture content also revealed that there is a limit to the presence of kaolin. This parameter decreased up to a certain amount of clay. The dispersion of kaolin in the polymer matrix also reduces wettability, swelling, and moisture uptake due to a decrease in water adsorption. However, the strong interactions between the compounds make the film compact and stable. The study also found that the increase in kaolin concentration favors the degradability of the film, with an optimal formulation reaching up to 48% biodegradability. The authors attributed the results as mainly due to intermolecular interactions between the inorganic structure and the biopolymers. The study also observed the high thermal stability of the films with the incorporation of kaolin through thermogravimetric analyses and good interfacial adhesion through SEM micrographs.

Halloysite is a type of clay mineral that belongs to the kaolinite group. It has a 1:1 dioctahedral structure and is represented by the chemical formula $Al_2Si_2O_5(OH)_4\ nH_2O$. These minerals can have different shapes such as tubular, spheroidal, or platy-like particles, with the tubular shape being the most common [71]. Halloysite has a lower ion exchange capacity and smaller surface area compared to other clay minerals. However, this clay mineral possesses active internal and external surfaces, increasing its ability to interact with other compounds. Kurczewska et al. [19] conducted studies to prepare two types of films based on pectin or alginate functionalized with halloysite, which was modified with salicylic acid. The aim was to test the films for drug release in food packaging applications. The authors observed that pectin films released the drug very rapidly in a solution containing 20% v/v ethanol, while alginate films showed a prolonged release of salicylic acid. The results showed that the mixed morphology (nanotubes and nanoplatelets) of halloysite promoted sustained release, which was more effective than the tubular morphology that resulted in the rapid release of the drug. When tested in a solution containing 50% v/v ethanol, the films showed a more controlled release of the drug, indicating that they are more suitable materials for food packaging with lipophilic properties. The antibacterial activities of the films against different bacteria showed interesting trends. In general, alginate-based films showed better results against *E. coli*, *S. aureus*, *P. aeruginosa*, and *Salmonella Typhimurium*. The results against *P. auruginosa* were noteworthy, as pectin films containing halloysite with mixed morphology exhibited activity, but films containing tubular halloysite did not show inhibition of pathogen growth. Once again, the morphology of the halloysite influenced the results. Da Silva et al. [72] developed a cassava starch film that was functionalized with silver sulfadiazine-loaded halloysite. The biocomposites showed a more homogeneous surface compared to the pure starch film, indicating a uniform dispersion of solid nanoparticles in the film. FTIR analysis showed that the drug was complexed in the halloysite structure, and the displacement of the starch bands indicated good interaction between the halloysite and the polymeric matrix through hydrogen bonds. The XRD results showed an amorphous and semi-crystalline characteristic of the biocomposites, indicating interaction between halloysite and starch without the formation of a crystalline phase of the drug. The incorporation of silver sulfadiazine-loaded halloysite increased the water vapor permeability of the film by approximately 20%, which favors the use of semi-occlusive dressings for faster healing. The addition of modified halloysite increased the film's resistance and reduced its flexibility, indicating the reinforcing role of halloysite in the biopolymer. Antibacterial assays showed that the film had potential against *E. coli*, *P. aeruginosa*, and *S. aureus*, attributed to the release of silver and sulfadiazine ions, which are potent bacteriostatic agents. The authors concluded that the films are highly suitable for use in wound healing for dry wounds, as they are efficient against bacterial growth and have semi-occlusive characteristics.

Palygorskite is a non-planar phyllosilicate with a fibrous morphology of elongated crystals. This modular clay mineral has a crystalline structure of regularly arranged tunnels/channels that contain 2:1 layers [73]. Palygorskite is a widely used clay mineral due to its abundance and rich surface chemistry. It contains different active sites capable of chemical and physical modifications, improving its morphological characteristics, porosity, and structure for various applications. Ding et al. developed a film made of alginate, polyvinyl alcohol, and palygorskite, which was strengthened with a silanized layer through a vapor deposition–surface polycondensation reaction using methyl trichlorosilane (MTCS). The XRD and FTIR analyses confirmed that there were interactions between the polymer film and the palygorskite nanorods and also showed that the silanized surface did not alter the structure of the biofilms. From thermal analyses, the authors observed an increase in residual mass, indicating that the hydrogen bonds between the clay mineral and the blending film improved their thermal stability. The silanized surface promoted increased thermal stability due to the covalent bonds between the polymer film and the silane groups of the coating. The uniform dispersion of the biopolymers and palygorskite, as well as the interactions between them, created a stable structure that had improved tensile strength with the addition of 4% clay mineral. It is important to note that an excess concentration of palygorskite leads to the agglomeration of the nanoparticles and the formation of structural defects that lead to the film fracturing when requested. Lower amounts of clay minerals lead to a slight reduction in the elongation break and flexibility of the film. The incorporation of palygorskite in the film increases hydrophobicity, probably due to the bonds between the hydroxyl groups of the palygorskite with the hydroxyl and carboxyl groups of the polymer structure, reducing interaction with water molecules. The synergistic effect between the presence of palygorskite and surface silanization increases moisture uptake. The films also showed high chemical stability in contact with different organic solvents.

Sepiolite is a hydrated magnesium-rich aluminosilicate clay mineral with 2:1 layer structures, and it has fiber- and rod-like morphologies. Sepiolite and palygorskite are two types of clay minerals that have different diameters and lengths. Sepiolite has a typical diameter ranging from 10 to 100 nm, and its lengths range from 2 to 10 µm. On the other hand, palygorskite has diameters of 20–70 nm and lengths of 0.5–5 µm [74]. Sepiolite features a fibrous structure with intracrystalline tunnels, providing high surface area and reactivity, making it highly advantageous for industrial applications. Alves et al. conducted a study to prepare cellulose nanofiber films that were functionalized with sepiolite using two synthesis methods: solvent casting and filtration with subsequent hot pressing. The introduction of sepiolite caused a reduction in the transparency of the films since sepiolite is an opaque and colored solid. However, this characteristic was found to be beneficial for the UV protection properties of the films. It was also observed that the synthesis method influenced the properties of the films. Films obtained by filtration and pressing had less roughness and better mechanical properties compared to the films prepared by solvent casting. The addition of 10% sepiolite improved the films' tensile strength and Young's modulus. The results indicated that the strong interactions and uniform dispersion of sepiolite improved the mechanical properties of the biopolymeric films. SEM images showed that there was no segregation between the sepiolite and the cellulose fibers. Additionally, the films prepared by filtration had a more ordered structure and lower surface roughness. The authors also observed that the water vapor transmission rate (WVTR) and water vapor permeability (WVP) showed a similar trend, where the incorporation of 20% sepiolite generated a slight increase in these characteristics. The authors concluded that the addition of 10 to 20% sepiolite favored the interactions between the components, consequently improving the properties of the films. These properties make the films suitable for use in food packaging. Finally, Alcântara et al. [18] developed films based on different polysaccharides such as hydroxypropylmethylcellulose, carboxymethylcellulose, alginate, pectin, and xanthan functionalized with palygorskite and sepiolite (unmodified and modified with zein). The films showed good interactions between the silanol groups of clay minerals and the hydroxyl and carboxyl groups of the polysaccharides. The addition of organo-clay minerals resulted in improved mechanical properties, thermal stability, better stability in water, reduced absorption of water and gases, and an improved barrier to UV light. These bionanocomposites have potential applications in bioplastics for food packaging.

Based on the results presented above, it is undeniable that incorporating clay minerals into biopolymer films brings significant benefits. When used in food packaging, the addition of clay minerals enhances the functionality of the films. As reinforcing agents, they increase barrier properties, improve mechanical and thermal properties, and promote the stability and durability of the films [75]. When applied to wound dressings, they promote flexibility, wrapping, sealing, and impermeability. Clay minerals can also prevent infections caused by environmental bacteria and prevent losses of active dressing components [76].

5. Nanofillers as Active Compounds

Besides clay minerals, various other nanofillers can improve the performance of films. These include organic substances such as lipids, plant extracts, polyphenols, essential oils, and inorganic nanoparticles like silver nanoparticles, iron, zinc, and copper. Carbon-based nanostructures such as graphene, nanotubes, and carbon dots can also be added as functional agents to enhance the mechanical, thermal, chemical, physical, and biological properties of the films. By combining clay minerals and biopolymers, biofilms can be developed using various nanofillers, which are considered promising alternatives as active compounds (as shown in Figure 4).

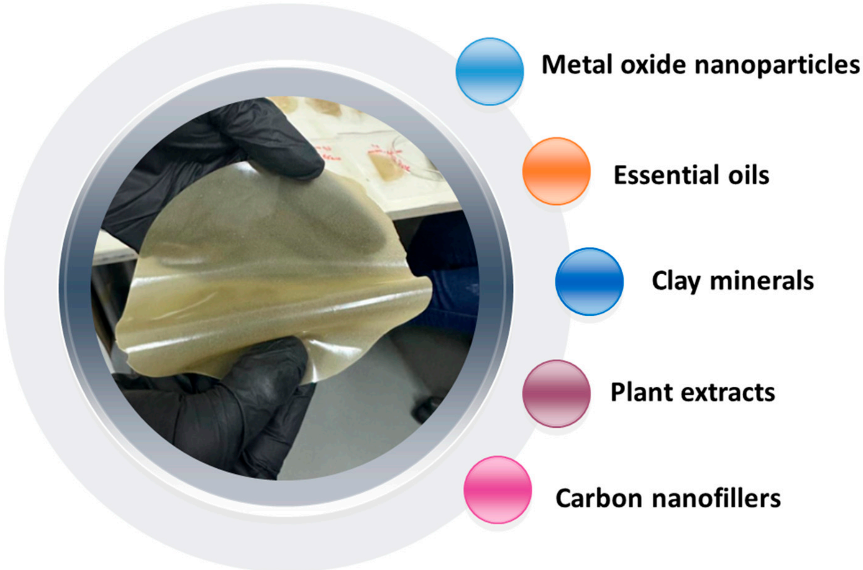

Figure 4. Some nanofillers as interesting alternatives for developing biopolymer-based films.

5.1. Metal Oxide Nanoparticles

There are several methods for creating metallic nanoparticles, including precipitation, co-precipitation, hydrothermal, microwave, ultrasound, sol–gel, combustion, and more [77–81]. The morphology of the nanoparticles can be controlled by adjusting the synthesis parameters, resulting in shapes such as prisms, rods, flowers, spheres, flakes, and others. These nanoparticles possess excellent optical, dielectric, catalytic, thermal, and mechanical properties and are used in a variety of applications, including electronics, photocatalysis, batteries, sensors, and biological applications. While various types of nanoparticles, such as TiO_2, ZnO, CuO, CeO_2, and Fe_3O_4, have been successfully incorporated into biopolymer-based films, there have been few studies that have looked at the addition of clay minerals and metal nanoparticles simultaneously. Table 1 shows the different nanoparticles that have been used in conjunction with clay minerals for biopolymeric film formation, along with their main properties.

Table 1. Biopolymer-based films functionalized by metal nanoparticles and clay minerals.

Biopolymer	Clay/Clay Mineral	Metal Nanoparticle	Key Findings	Ref.
Chitosan/alginate	Bentonite	ZnO	Flexible and transparent polymeric films; strong activity against *S. aureus* and *P. aeruginosa*; and epithelium regeneration in histological studies from in vivo wound healing.	[36]
Alginate	Halloysite	ZnO	Great mechanical resistance, UV and water vapor barrier, and hydrophobicity and impressive activity against *E. coli* and *L. monocytogenes*.	[38]
Chitosan	Nanoclay	ZnO	Biodegradable and effective film in preserving the quality of sweet cherries and excellent mechanical and barrier properties.	[82]

Table 1. *Cont.*

Biopolymer	Clay/Clay Mineral	Metal Nanoparticle	Key Findings	Ref.
Chitosan/PVA	Montmorillonite and halloysite	ZnO	Great mechanical resistance and oxygen and water vapor barrier; great effect against *E. coli*, *S. aureus*, *S. enterica*, and *L. monocytogenes*; and extension of food shelf life.	[83]
Chitosan	Montmorillonite	ZnO	High mechanical response and great antimicrobial activity against *S. aureus* and *E. coli*.	[84]
Chitosan	Kaolinite	ZnO/MnO_2	The synergistic effect between chitosan and nanoparticles obtained better results in biocompatibility and antibacterial activity against *S. aureus* and *E. coli*.	[85]
Chitosan	Palygorskite	ZnO	Good transparency and remarkable tensile strength and excellent antibacterial activity against *S. aureus* and *E. coli*.	[86]
Corn starch/gelatin/ bacterial nanocellulose	Halloysite	ZnO	Improved thermal stability and citocompability and antibacterial activity against *S. aureus* and *E. coli*.	[87]
Chitosan	Palygorskite and montmorillonite	Ag Nps	The clay mineral mixture achieved better water resistance and mechanical, antioxidant, and antibacterial responses; the antibacterial effect against *E. coli* and *S. aureus* reached 100%.	[47]
Chitosan	Laponite	Ag NPs	Low cytotoxicity; good mechanical properties; inhibition of the growth of *S. aureus*, *E. coli*, *A. niger*, and *P. citrinum*; and extends the storage time of food.	[66]
Chitosan	Bentonite	Fe_3O_4	Increased concentration of clay minerals decreases the release of curcumin; high cytotoxicity against human breast cancer cell lines (MCF-7 cells); and excellent activity against the growth of *E. coli* and *S. aureus*.	[88]
Hydroxyethyl cellulose	Bentonite	Fe_3O_4	Intercalation of polymer chains into clay spacing; reinforcement of polymer film due high density of the hydrogen-bonding network; and antifungal activity against *C. albicans*.	[89]

Zinc nanostructures are extensively used in cosmetic and sunscreen products due to their UV absorption properties. They also possess biomedical features such as anticancer, antifungal, drug delivery, antibacterial, and anti-inflammatory properties. Additionally, they aid in wound healing and bioimaging [90]. ZnO incorporated into biofilms improves thermal, mechanical, and barrier properties and offers stable, photocatalytic, antimicrobial, and antioxidant characteristics, making it safe for use [82].

Silver nanoparticles have been the subject of extensive research due to their remarkable chemical and physical stability. This stability is due to the quantum excitations of the conducting electrons within the particles. Ag NPs have numerous applications, including in biosensors and photocatalysis. Additionally, they are widely used as antimicrobial agents and in various fields of biomedicine [91]. They are used in food packaging as active agents and released to improve the quality of food, extend shelf life, and prevent spoilage [92]. Ag nanoparticles have been extensively investigated for their effectiveness against resistant bacteria. They can cause physical or oxidative damage to pathogen cells when used in wound dressing applications [93].

Magnetite (Fe_3O_4) is a type of nanomaterial that has been extensively studied in technological fields due to its ferromagnetic, electronic, and catalytic properties. In the field of biomedicine, it has gained significant attention due to its low toxicity, physical and chemical properties, and high saturation magnetization values. Additionally, scientists have extensively researched its anticancer and antibacterial effects [94]. When added to polymeric films, it enhances the mechanical properties, biocompatibility, and antibacterial potential [95].

Metallic nanoparticles are a type of nanostructure that are currently in high demand. These nanoparticles are highly preferred in films due to their affordability, large surface area, and small size. They are known for their exceptional ability to prevent the growth of various types of bacteria and fungi. Additionally, they possess excellent thermal and chemical stability, are biocompatible, and can reinforce various film materials. Because of these properties, metallic nanoparticles are ideal for use in the food and biomedical areas.

5.2. Essential Oils

Essential oils (EOs) are organic compounds that are obtained as byproducts from various parts of medicinal, herbal, and edible plants. These oils are volatile and hydrophobic in nature, known for their active biological properties such as antibacterial, antifungal, antiviral, and insecticidal properties. The increasing trend towards natural products replacing synthetic agents has boosted the popularity of essential oils. They offer greater flexibility, thermal properties, and safety in biofilms [96]. However, they are highly susceptible to degradation, volatilization, and oxidation due to external factors like light, temperature, and oxygen. To prevent this, organic or inorganic matrices can be used to encapsulate these molecules, thereby protecting them from deterioration [3,97].

Incorporating essential oils directly into polymeric films is not an effective approach due to the low stability of the oil molecules. Combining the oil load with various nanoparticles, particularly nanoclays, can lead to better chemical and mechanical properties, as well as enhanced antioxidant and antimicrobial activities. Moreover, the compounds work synergistically to achieve a controlled release of bioactives [98,99]. Some works on functional films based on biopolymers using clay minerals and essential oils are showcased in Table 2.

Table 2. Biopolymer-based films functionalized by essential oils and clay minerals

Biopolymer	Clay/Clay Mineral	Essential Oil	Application	Ref.
Alginate	Montmorillonite	Clove, coriander, caraway, marjoram, cinnamon, and cumin	Active packaging	[44]
Chitosan	Montmorillonite	Thyme	Active packaging	[49]
Chitosan	Montmorillonite	Thyme	Shelf-life prolongation of sweet cherry	[82]
Chitosan	Montmorillonite	Rosmarinus officinalis	Active bio-based films	[100]
Chitosan	Montmorillonite	Rosmarinus officinalis	Shelf-life extension of poultry meat	[101]
Chitosan	Montmorillonite	Ginger	Food packaging	[102]
Chitosan	Montmorillonite	Ginger	Chilled beef preservation	[50]
Starch	Montmorillonite	Carvacrol	Antimicrobial packaging material	[103]
Cellulose	Montmorillonite	*Origanum vulgare*	Active films	[104]
Gelatin	Montmorillonite	Black pepper	Active food packaging materials	[105]
Alginate	Montmorillonite	Lemon	Active packaging	[106]
Zein	Bentonite	Zataria multiflora Boiss	Bioactive packaging	[107]
Cassava starch	Bentonite	Cinnamon	Edible and biodegradable film	[108]
Levan	Bentonite	Calendula, citronella, lemon, tamanu, and peppermint	Biodegradable and antimicrobial active food packaging	[109]
Chitosan	Halloysite	Clove	Antioxidant and antimicrobial food packaging	[48]

5.3. Plant Extracts

Concentrated solutions obtained from plant materials (roots, stems, leaves, fruits, and seeds) are called plant extracts. The combination of natural polymer films and plant extracts as bio-based actives is a promising approach to improving food quality and safety in the food industry. Plant extracts have high levels of phenolic compounds, which possess strong antioxidant properties, as well as coloring and flavor particulars. Incorporating plant extracts and clay minerals into polymer films enhances their physicochemical, barrier, and mechanical properties and their antimicrobial and antioxidant effects [110,111]. This results in the development of nanocomposites that offer better functionality and performance.

Cheikh et al. [112] conducted a study where they added myrtle berry extract and sepiolite to alginate films. The study aimed to investigate the mechanical, physicochemical, and oxidizing properties of the film. The authors found that the addition of sepiolite enhanced the thermal stability, mechanical strength, and barrier properties of the biohybrid film. The improved properties were due to the strong interactions between sepiolite, polyphenol, and the polymeric matrix in the film. Moreover, the combination of myrtle extract with sepiolite increased the oxidizing activity of the film. Mouzahim et al. [51] conducted a study where they produced a chitosan film that contained Ficus carica leaf extract and kaolinite. The purpose of this film was to preserve apples. The study concluded that the addition of the clay mineral and plant extract greatly impacted the characteristics of the film. The film was found to be biodegradable, possess high antioxidant properties, and have good tensile strength, as well as UV and water vapor barriers. Additionally, the film provided protection for the fruit by reducing weight loss, maintaining moisture levels, decreasing the browning index, and preserving the total phenolic content. Li et al. [47] conducted studies where they developed a film using Ag NPs, montmorillonite, palygorskite, and curcumin incorporated into chitosan films. The results showed that the film exhibited excellent resistance and barrier properties. Additionally, the film demonstrated exceptional antioxidant and antibacterial activity against the colony growth of *S. aureus* and *E. coli*. Huang et al. [25] developed a film using chitosan, curcumin, polycaprolactone, and montmorillonite. The authors observed that the clay mineral and polyphenol enhance the strength of the film and can disinfect the skin. The tests conducted by the authors against *E. coli* showed that it is effective in disinfection. Additionally, the biocompatibility of the film makes it a viable option for use as a wound dressing. Toro-Márquez et al. [113] developed pH-sensitive films by using corn starch that is functionalized with montmorillonite and Jamaica (Hibiscus sabdariffa) flower extract. The authors reported that the exfoliation of the clay mineral favored high interaction between the compounds, promoting excellent barrier, mechanical, and thermal properties. Nouri and their co-authors [114] developed a film that is based on k-carrageenan/nanoclay which is functionalized with rosemary extract. In their study, the authors examined the impact of the combination of nanoclay and plant extract on mechanical, barrier, and thermal properties and antibacterial potential. The study results showed that the film has increased elongation break and tensile strength. Additionally, it protects against UV light and reduces water vapor permeability and visible light transmission. Moreover, the film has a high level of antibacterial activity, with almost 100% inhibition against bacteria such as *B. cereus*, *E. coli*, *P. aeruginosa*, and *S. aureus*. Rammak et al. [115] incorporated kaolin and calico plant extract as additives in starch films. The researchers found that the resulting biofilms exhibited high mechanical strength, hydrophobicity, and thermal stability. The addition of the clay and calico extract resulted in good interaction and improved functionality of the starch film. The film was also sensitive to different media with varying pH levels, making it a promising option for smart packaging. Recently, according to studies conducted by Sayah et al. [116], a gelatin film was developed and functionalized with illite, smectite, and chamomile extract to create a potentially active film. The combination of clays and plant extract affected the properties of the biofilm, resulting in better barrier and mechanical properties. Additionally, the film exhibited a remarkable antioxidant property.

Plant extracts are nutrient-rich compounds and offer several benefits when integrated into films. They promote antimicrobial and antioxidant activity and can affect the mechanical properties of the films, such as flexibility. When combined with clay minerals, they enhance the physical, chemical, and biological properties of the films, making them suitable for use in various applications. Their use in food packaging is particularly noteworthy because they can help preserve the shelf life and nutritional value of the food. However, there are not many studies that have utilized the combination of clay minerals and plant extracts at the same time to form new biofilms, making this an area that needs further exploration in the literature.

5.4. Carbon Nanofillers

Carbon nanostructures are carbon materials that are produced in a controlled manner and have a size of less than 100 nm. These structures have a diverse size and shape and include a wide variety of carbon allotropes such as nanoparticles, carbon dots, fullerene, graphene sheets, nanowires, nanotubes, and multi-walled carbon nanotubes [117]. Carbon-based nanofillers are exciting structures to incorporate into polymeric films to enhance their functionality, mechanical and barrier properties, and antimicrobial capacity. This makes them ideal for use in food packaging and wound dressings [75,118]. However, a disadvantage is the tendency for agglomeration and uneven distribution of the polymer matrix. On the other hand, one of the major advantages of these films is their chemical, physical, and thermal stability. Mao and colleagues [119] conducted a study where they added amino-functionalized carbon dots and layered clay in an alginate matrix. The results indicated the presence of hydrogen bonds between the N-H groups of the N-carbon dots and OH groups of the alginate. SEM images showed that there was interfacial compatibility between the clay, carbon structure, and alginate. The addition of these materials increased the tensile modulus and UV barrier while reducing the gas barrier. Furthermore, the films showed excellent antioxidant and antibacterial activity compared to the alginate-only film. Carbon-based structures have been used in certain studies to create polymeric films that are useful as wound dressings and food coverings [120–123]. However, like other bioactive compounds, they are not commonly combined with clay minerals. Therefore, exploring the use of biopolymer films that are functionalized with various clay minerals and carbon-based structures could be a promising area of study.

In short, bionanocomposite films are commonly used in food protection and wound healing. However, these films have some drawbacks such as low thermal stability, poor mechanical properties, and permeability. When functional nanofillers are added to these materials, they can improve these characteristics and enhance their potential applications. However, in accordance with Perera et al. [1], more comprehensive investigations are necessary to evaluate the toxicity and migration of components, predominantly clay minerals, as safety is a crucial factor for the clinical or food industrial utilization of these products.

6. Main Applications of Biofilms Based on Clay Minerals and Biopolymers

Functional films are materials created by blending an organic polymer matrix with organic or inorganic solids, resulting in nanocomposites with improved properties. As the demand for natural and sustainable products increases, scientists are exploring using natural biopolymers to develop biofilms. Among the various additives that can be used, clay minerals are particularly noteworthy due to their numerous surface active sites, rheological properties, small size, stability, and gelatinization capacity. The high interfacial compatibility and good dispersion of clay mineral nanoparticles in various biopolymer matrices make these materials incredibly versatile. As a result, these films can be used in two main applications, food packaging/covering and wound dressing/healing, as shown in Figure 5.

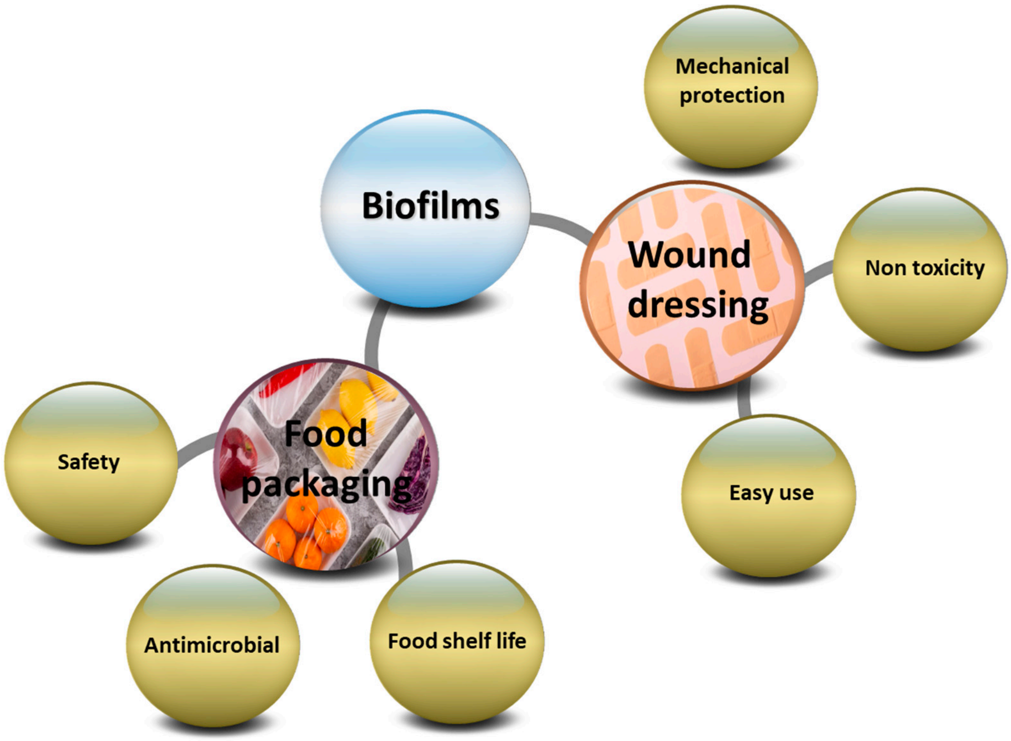

Figure 5. Main characteristics of using biofilms based on clay minerals in food packaging and wound dressing.

6.1. Food Packaging

Food packaging is the process of preparing food for transportation, distribution, storage, and consumption in a safe and cost-effective manner. It plays a crucial role in modern society as it enables the safe and efficient handling and commercial distribution of food products. Without food packaging, it would be challenging to distribute food safely and efficiently to the end consumer [124]. This industry area is rapidly growing as it is becoming increasingly popular among consumers globally. However, it faces the challenge of transitioning from synthetic materials derived from petroleum to natural components. Furthermore, there is a push to incorporate active biocomponents in producing both active and smart packaging [125]. Clay minerals are fascinating natural nanofillers to use in food packaging films. They enhance various characteristics of the film, such as its ability to block gases, humidity, water, and odors. Additionally, they provide protection against abrasion and cracks, improve transparency, mechanical strength, and puncture resistance, and contribute to thermal stability. They are also neutral to fats, grease, and oil [126,127]. Different clay minerals have already been used in studies on biopolymer films to preserve several food types.

Wu et al. [66] developed films using chitosan and laponite with silver nanoparticles. They tested the preservation of lychee for up to 10 days. The films exhibited strong antibacterial activity against *S. aureus*, *E. coli*, *A. niger*, and *P. citrinum* and showed lower toxicity compared to pure chitosan film. Storage studies revealed the deterioration of uncoated fruit and fruit covered with chitosan films, while fruits covered with chitosan/Ag/laponite films were effectively preserved. Souza et al. [101] developed a film using chitosan, montmorillonite, and rosemary essential oil to study its effectiveness in preserving poultry meat for up to 15 days. The researchers found that the covered samples exhibited lower lipid oxidation and inhibited microbial growth compared to uncovered samples. Moreover, the film demonstrated good oxygen permeability, which helped preserve phenolic compounds and extend the shelf life of the food. Studies by Mouzahim et al. [51] utilized chitosan film, kaolinite, and Ficus carica leaf extracts to explore the preservation of apple pieces over 24 h. The researchers observed that the samples covered with the films exhibited reduced weight loss, browning index, and total phenolic content. Additionally, the covered films showed less oxidation compared to the uncovered apple pieces. Barikloo and Ahmadi [128] assessed the impact of temperature and storage duration on the physical–chemical attributes of strawberries. They used chitosan films functionalized with polyolefins, clay, and silica nanoparticles as coverings for the strawberries. The researchers observed that after 10 days of storage at 4 °C, the strawberries exhibited different physical and chemical properties such as elasticity modulus, pH, firmness, weight loss, and soluble solids, as well as levels of oxygen and carbon dioxide, compared to storage at 25 °C. Rangaraj et al. [129] produced a film using Ag/sepiolite combined with a mixture of gelatin and date waste extract. The authors noted that the presence of Ag/sepiolite enhanced the antioxidant and antibacterial properties, as well as the slow release of active compounds and silver nanoparticles in food simulants. Visual observation of a piece of apple, both covered and uncovered with the film, revealed that the uncovered fruit oxidized, while the covered fruit remained preserved after 5 h at room temperature. Perera et al. [130] conducted studies to develop alginate/gelatin films using bentonite as a reinforcing agent. These films were used to preserve cheese for up to 45 days, and the results showed high storage stability. The films increased the shelf life of the cheese, inhibited the growth of *E. coli*, *L. monocytogenes*, and *P. expansum*, and maintained the pH, weight, and visual appearance of the food.

6.2. Wound Dressing

Dressings are used as a clinical treatment for skin wounds, particularly exudative wounds. Conventional dressings cover wounds to protect them from the external environment but do not contain active compounds that directly aid the healing process. Smart dressings have been developed to facilitate healing by triggering steps in the natural healing process, which accelerates wound closure [131]. The incorporation of bioactive compounds is a strategy for enhancing these dressings. Clay minerals are widely used to improve important properties of these materials, such as mechanical protection, toxicity reduction, and ease of handling. These modern films are designed to have antioxidant and antimicrobial activity to aid in wound treatment. Furthermore, due to their physiological inertness, biocompatibility, mixed surface charge, chemical stability, adsorption capacity, and presence of surface active sites such as hydroxyl groups and silanol groups, clay minerals have healing promotion and hemostasis characteristics, making them highly suitable for incorporation into films based on biopolymers for wound management [76].

Ghadiri et al. [132] developed a laponite/mafenide/alginate-based film to explore its potential for treating burn wounds. The findings were highly positive, as the films exhibited a slow degradation rate, high water absorption capacity, and balanced water vapor permeability. The laponite-containing films demonstrated high drug release at the wound site, strong antimicrobial activity, good fibroblast proliferation, and no cytotoxicity. These results indicate that the films are suitable for treating exuding wounds. Dutta and Devi [133] developed a chitosan/sepiolite film and studied its properties for use as a wound dressing material. The results showed strong antibacterial activity against *B. subtilis* and *E. coli*. Films containing sepiolite exhibited hemostatic properties, promoting rapid blood clotting and thrombus formation. Furthermore, the films are non-hemolytic and non-cytotoxic, making them promising for use in wound-care products. In a separate study, Devi and Dutta [134] created a series of films using chitosan and bentonite. Based on their findings, the authors concluded that the optimized presence of bentonite increased the mechanical strength, bending resistance, water absorption capacity, and porosity of the films. Additionally, the nanobiocomposite films exhibited antibacterial activity against both Gram-positive (*Bacillus subtilis*) and Gram-negative (*Escherichia coli*) bacteria when compared with chitosan alone. Hemocompatibility tests showed that the nanocomposite films with bentonite nanofillers were highly compatible with blood. Therefore, chitosan/bentonite films possess several qualities suitable for use in exudative wound dressings. Zhang et al. [135] developed chitosan/glycyrrhizic acid/ZnO/palygorskite films for treating skin wounds. The films exhibited light transmittance, barrier properties (including water content, water solubility, and swelling degree), improved mechanics, and thermal stability. They also showed hemocompatibility and antibacterial activity against pathogenic bacteria like *E. coli* and *S. aureus* and drug-resistant bacteria like *ESBL—E. coli* and *MRSA*. As a result, these films can be utilized for skin wound healing applications. Jaberifard et al. [87] developed a starch/gelatin film functionalized with ZnO, halloysite, and nanocellulose. The films exhibited thermal and mechanical stability and cytocompatibility, inhibiting bacterial growth against *Escherichia coli* and *Staphylococcus aureus*. They also had a stimulating effect on the migration and differentiation of fibroblast cells, accelerating healing through in vitro scratch wound assays. Recently, Jaberifard et al. [136] developed a xanthan gum/soy protein film that was functionalized with halloysite and propolis. The film demonstrated impressive antibacterial properties against *E. coli* and *S. aureus*, as well as antioxidant properties. Additionally, it showed improved mechanical properties, hydrophilicity, cell adhesion, cytocompatibility, and great interactions between the organic and inorganic components, enhancing the functionalities of the film.

Biopolymers are popular and biocompatible materials, but their mechanical properties need improvement to enhance their interaction with other bioactive molecules. To enhance their biological properties (antimicrobial and antioxidant activities), biopolymers are often combined with other polymers such as alginate, starch, PVA, PCL, PLA, chitosan, gelatin, and pectin. However, the resulting materials often lack sufficient mechanical properties for use in physical and chemical processes. To improve both the biological and mechanical properties, as well as bioactivity, various bioactive inorganic materials can be developed with different formulations. The incorporation of clay minerals can be an interesting alternative to enhance the resistance properties, barrier features, and antimicrobial potential of biofilms [36,39,87,136–140].

7. Conclusions and Outlooks

Films made from biopolymers show great promise for use in food coatings, packaging, and healing skin wounds. The addition of clay minerals has a positive impact on physical and chemical properties, such as resistance, elongation, gas and water permeability, humidity, thermal properties, morphology, transparency, and UV blocking. When clay minerals are combined with other organic or inorganic compounds, they create a synergistic effect that enhances these characteristics and the bioactivity of the films, improving antioxidant and antimicrobial properties, cell adhesion, and biocompatibility. The alteration in these properties is mostly because of the molecular interactions among the clay minerals, the additives added at the same time, and the polymeric matrix. Additionally, the ability to include natural elements in films makes these materials more sustainable and appealing to consumers, as it can enhance safety by substituting synthetic compounds. Films based on biopolymers functionalized with clay minerals are highly promising for clinical and industrial use.

The potential combinations of biopolymer-based films functionalized with various clay minerals and combined with different components have not been thoroughly explored in the literature. By incorporating metallic nanoparticles, essential oils, plant extracts, and carbon nanostructures into different types of clay minerals, it is possible to create films with enhanced properties. This opens up opportunities for developing new films and suggests areas for future research. The research and development of films for use in clinical and industrial applications is expected to continue in order to discover new materials and functionalities. It is important to emphasize the social and environmental responsibility of these materials and to always seek materials that are environmentally friendly, such as biodegradable or compostable materials.

The use of nanoparticles, such as clay minerals or metallic nanoparticles, on a nanometric scale has been found to enhance the mechanical and antimicrobial properties of films. However, there is a concern that these nanoparticles may be released from the films and come into direct contact with the human body. Therefore, it is important to consider the biosafety of the films for both biomedical and food industry applications. It is essential to conduct thorough tests for extended contact, migration, toxicity, and biocompatibility to enhance the reliability of the films. Since these materials are meant for human consumption, it is important to conduct a risk assessment. This includes identifying the potential risks, characterizing the dangers of exposure to humans, and employing methodologies to better assess the safety of these materials in order to avoid harm to human health.

Marketing these materials can be challenging for several reasons. Firstly, large-scale production needs to consider material costs and productivity. Since most of the films were developed on a laboratory scale, there is a long way to go before they can be mass-commercialized. Therefore, developing these materials on a pilot scale should make the process more appealing from a commercial standpoint.

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