Employing Nanosilver, Nanocopper, and Nanoclays in Food Packaging Production: A Systematic Review

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Abstract: Over the past decade, there has been an increasing demand for “ready-to-cook” and “ready-to-eat” foods, encouraging food producers, food suppliers, and food scientists to package foods with minimal processing and loss of nutrients during food processing. Following the increasing trend in the customer’s demands for minimally processed foodstuffs, this underscores the importance of promising interests toward industrial applications of novel and practical approaches in food. Along with substantial progress in the emergence of “nanoscience”, which has turned into the call of the century, the efficacy of conventional packaging has faded away. Accordingly, there is a wide range of new types of packaging, including electronic packaging machines, flexible packaging, sterile packaging, metal containers, aluminum foil, and flexographic printing. Hence, it has been demonstrated that these novel approaches can economically improve food safety and quality, decrease the microbial load of foodborne pathogens, and reduce food spoilage. This review study provides a comprehensive overview of the most common chemical or natural nanocomposites used in food packaging that can extend food shelf life, safety and quality. Finally, we discuss applying materials in the production of active and intelligent food packaging nanocomposite, synthesis of nanomaterial, and their effects on human health.

Keywords: food; packaging; nanocomposites; shelf-life; nanomaterial

1. Introduction

Nanotechnology is a new field dedicating key advances to food packaging for consumers. Thanks to this promising technology, many of the current challenges related to conventional packaging in food security and food sustainability issues can be addressed. The concept of nanotechnology was first introduced by Richard Feynman in 1959 [1]. Nanotechnology has provided the ability to work on a scale of about 1 to 100 nanometers to understand nanostructures, devices, and systems [2]. In financial terms, the global food packaging market size was USD 304.98 billion in 2019 and is projected to reach USD 463.65 billion by 2027, exhibiting a CAGR of 5.9% during the forecast period [3]. According to statistics published on the StatNano website, 166,000 articles in nanotechnology were registered on the Web of Sciences database in 2018, accounting for 10% of the total articles indexed in the database. In recent years, China, the United States, and India were the first, second and third largest countries publishing nanotechnology-based research, respectively. In 2013, Paulraj Kannan and Jong-Whan Rhim reviewed various synthetic and natural biopolymers to develop nanocomposites by mixing with nanosized materials, such as layered silicate nanoclays, nanosized metal, and metallic oxides. In this study, the antibacterial properties of nanocomposites were also carefully investigated. They believed that the novel nanocomposite packaging materials with antimicrobial functions possessed a high potential to be applied in active food packaging [4]. In 2017, Ahari and his colleagues...
reported a study on nanopackaging covers and their functional mechanisms. In addition, in this study, the toxicity and release of the nanoparticles (NPs) in the packaging covers were investigated. They focused on the usage of nanoparticle synthetic covers like nanoclay and nanosilver and their antibacterial properties that manifest a higher consistency of the covers through increasing the adhesion [5].

In this review study, we first introduce the nanomaterials frequently used in food packaging. We then focus on inorganic NPs (including metals, metal oxides, clays, and derivatives) with their considerable features, providing efficient active and smart food packaging in food industries.

2. Methods
2.1. Search Strategy

Data for this review were collected through a deep literature review of the most recent studies conducted on preparation methods, migration, and antimicrobial aspects of NPs in the food packaging industry. An advanced search was measured out in PubMed databases, using the following keywords in the title and abstract fields: nanocomposite OR nanocomposites, OR bionanocomposite OR bionanocomposites AND food packaging, nanoparticle OR nanoparticles, AND food packaging, nanosilver OR nano silver AND food packaging, nanoclay OR nano clay OR nanoclays AND food packaging, and nanocopper OR nano copper OR copper oxide AND food packaging. A total of 71 entries were obtained and screened according to several inclusion and exclusion criteria defined based on the purpose of this present study.

2.2. Study Selection

The obtained articles were selected based on the following eligibility criteria according to our search strategy (Figure 1): (1) only articles published in the recent 10 years were considered; (2) only English articles were selected (studies in other languages were excluded); (3) only original research articles were chosen (review articles were removed); (4) only full text and peer-reviewed journal articles were considered; (5) only research articles directly associated with NPs for food packaging were selected. In the end, 19 articles were included in the literature review part of this study [6–24].

![Flowchart for the selection process of the included articles.](image-url)
3. Results

To provide a comparative study on the preparation methods, migration and antimicrobial effects of different NPs employing for food packaging, we focused on three main NPs, including nanosilver, nanocopper, and nanoclays, as the most favorite and applicable NPs for food packaging. By searching literature using the defined keywords, 19 articles were identified as eligible studies for providing a systematic review and meeting our inclusion/exclusion criteria on industrial applications and significant properties of nanosilver, nanocopper, and nanoclays particles used in food packaging (Table 1). As illustrated in Table 1, the desired articles were compared given nanoparticle type, the employed method for preparation of NPs or nanocomposites, nanoparticle migration into foodstuff, and antimicrobial effects of the desired NPs.

| Type of Bio-Nanocomposite | Food Product | Antimicrobial Results | References |
|---------------------------|--------------|-----------------------|------------|
| PLA/ZnO:Cu/Ag             | Food simulant: (distilled water)—simulant A, ethanol 10% (v/v) in aqueous solution—simulant B and acetic acid 3% (w/v) in aqueous solution—simulant C | Good mechanical, thermal and barrier properties to ultraviolet light, water vapor, oxygen and carbon dioxide; antibacterial activity and low migration of NPs into food simulants. | [26] |
| Starch/halloysite/nisin   | Soft cheese  | Improved mechanical properties with halloysite addition; antimicrobial activity against *Listeria monocytogenes* (*L. monocytogenes*), *Clostridium perfringens*, and *Staphylococcus aureus* (*S. aureus*) | [27] |
| Low-density polyethylene (LDPE)/Ag, ZnO | Orange juice | A significant decrease of the yeast and mold counts without disrupting juice relevant quality attributes; significant reduction in the growth rate of *Lactobacillus plantarum* | [28] |
| PE/Ag, titanium oxide (TiO₂), kaolin | Chinese jujube | Firmer, heavier, less decay, less browning, slower ripening, decrease in senescence and climacteric evolution | [29] |
| Cellulose absorber/Cu     | Melon and pineapple juices | Excellent antifungal activity, reducing spoilage-related yeasts and molds | [30] |
| Absorbent pad/Ag          | Poultry meat  | Confirmed antimicrobial effect against *Escherichia coli* (*E. coli*) and *S. aureus* | [31] |
| Allyl isothiocyanate, nisin/ZnO absorber/Ag | Liquid egg albumen Kiwi and melon juices | Effective inactivation of *Salmonella* Reduced counts of total viable microorganisms, yeasts and molds | [32] |
| PE/Ag                     | Apple juice   | High bactericide capacity against *Alicyclobacillus acidoterrestris* | [33] |
| Pectin/nanohybrid-layered double-hydroxide salicylate | Fresh apricots | Improved elongation at a breakpoint for pectin; improved water vapor barrier properties; extended shelf life | [34] |
| Corn starch/talc NPs      | Tomatoes      | Reduction in water vapor and oxygen permeability | [35] |
| Gelatin/ethanolic extract from coconut husk/Cloisite Na + nanoclay | Meat powder  | Lower lipid oxidation products; Increased moisture barrier properties; extended shelf life | [36] |
| Type of Bio-Nanocomposite | Food Product | Antimicrobial Results | References |
|---------------------------|--------------|-----------------------|------------|
| Nanozinc oxide–neem oil–chitosan | Carrots | Good inhibition effect as antibacterial activity against *E. coli* | [37] |
| Poly(glycidyl methacrylate vinyl ferrocene)/graphene oxide/iron oxide NPs, and poly(glycidylmethacrylate-vinylferrocene)/MWCNT | Fish | Fish meat freshness control | [38] |
| Alginate/nano-Ag | Shiitake mushroom | Spoilage reduction, improvement of sensory attributes, lower weight loss | [39] |
| Sodium alginate solution containing Ag NPs | Carrot and pear | Antibacterial film effective against the test strains, *E. coli* ATCC 8739 and *S. aureus* ATCC 6538 | [40] |
| Calcium alginate film loaded with ZnO NPs | Ready-to-eat poultry meat | Shelf life increase of carrots and pears; Reduction of microbial load of *Salmonella typhimurium* and *E. coli* | [41] |
| Nanoemulsion coating of chitosan and mandarin essential oil | Green beans | Reduction in the population of *L. monocytogenes* | [42] |
| Chitosan/poly(vinyl alcohol)/TiO2 NPs | Soft white cheese | Antibacterial activity against Gram-positive (*S. aureus*), Gram-negative (*Pseudomonas aeruginosa* (*P. aeruginosa*), *E. coli*) bacteria and fungi (*Candida albicans*); extension of shelf life | [43] |
| Whey protein isolate (WPI)/montmorillonite (MMT) | Food models (water, 3% acetic acid, 15% ethanol, olive oil) | Increased tensile strength of the WPI film and swelling of the WPI film | [44] |
| Soy protein isolate–MMT | Fresh fruits and vegetables | Improvement in mechanical properties, thermal stability, and water vapor permeability with the addition of MMT | [45] |
| Ag NPs-based cellulose absorbent pads | Fresh-cut melons | Retarding the senescence rate of the fresh-cut melons; reduction in antibacterial activity | [46] |
| Edible coating of polyvinyl pyrrolidone (PVP) containing Ag NPs | Asparagus | Inhibiting the growth of psychrotropic microorganisms; lowering weight loss, greener color, and tender textures | [47] |
| Nano-ZnO-coated poly(vinyl chloride) | Fresh-cuts of “Fuji” apples | Preservation of quality indicators, such as ascorbic acid and polyphenol content; Lower counts of typical altering microorganisms | [48] |
| Thyme essential oil (TEO) and MMT-based sweet potato starch films | Baby spinach leaves | Incorporation of TEO in the film reduced the population of *E. coli* and *Salmonella Typhi* (*S. Typhi*) on fresh baby spinach leaves | [49] |
| LDPE/Ag, TiO2, kaolin | Strawberries | Decelerated decay rate | [50] |
| LDPE/Ag2O | Apple slices | Decreased microbial spoilage, delayed browning and weight loss | [51] |
3.1. Nanomaterials in Food Packaging: Definition, Characteristics, and Types

Nanomaterials, or NPs, are characterized by having a size smaller than 100 nm at least in one dimension and exhibit unique physical and chemical properties, such as flexibility, flame resistance, recycling and antimicrobial properties. These aforementioned properties make them worthy alternative materials in broad areas of technology, including electronics, energy management, information technology, pharmaceutical industries, medicine, and the food industry [52]. It has been reported that usage of nanomaterial production is continuously increasing 25% per year in different industries [53].

In recent decades, it has been identified that nanomaterials exhibit the potential to be applied in the food industry to provide numerous purposes, including improved food packaging, the prolonged shelf life of food materials, higher food safety, and better food quality [54]. In other words, nanomaterials have made a revolutionary era in the food packaging industries by conferring three main advantages over conventional packaging systems, including improved packaging, active packaging, and smart packaging [55]. Nanocomposites with antimicrobial properties could offer active packaging through active materials that contact packaged food, preventing the excessive growth of microorganisms that existed on the food surface [55,56].

Nanomaterial, including metals and metal oxides, such as silver (Ag), copper (Cu), Gold (Au), iron oxide (Fe$_3$O$_4$), copper oxide (CuO), TiO$_2$, zinc oxide (ZnO), magnesium oxide (MgO), and calcium oxide (CaO) have been demonstrated as the most well-known nanomaterial with antibacterial effects [57]. In fact, the main reason for the effectiveness of nanoscale materials as antimicrobial agents goes back to the possibility provided at the nano-level for these materials to turn into different shapes, such as spherical, triangular, rod, octahedral, sheet, shell and flower-shaped. This variation in the geometry of nanomaterials is especially advantageous for antimicrobial applications since the antimicrobial functions of nanomaterials are directly in proportion to the surface area available for interaction with biological compounds [57,58]. Hence, antimicrobial nanomaterials, also known as “nanoantibiotics”, are considered one of the most promising approaches to preventing or controlling microbial growth and infections. Although the mechanisms underlying antimicrobial actions of nanomaterials are still yet to be explored, it has been suggested that nanomaterials inhibit the microbial growth and biofilm formation through mainly two lethal mechanisms, including: (1) cell membrane damage via the disruption of membrane integrity and potential and the depolarization caused by binding of nanomaterials to the cell membrane; (2) the induction of severe oxidative stress due to production of oxygen-free radicals, leading to vigorous DNA damages and mutations and finally cell death [59–61].

For antimicrobial purposes, many metal oxides are used as photocatalysts, i.e., they can be irradiated with ultraviolet radiation to synthesize highly reactive oxygen species (ROS), which potentially exhibit antimicrobial activity, and is applied for food packaging to reduce biological risks and increase healthiness for human beings [62,63].

The application of nanomaterials is not limited to antimicrobial food packaging. However, NPs and nanocomposites are actively utilized in food packaging industries to provide a barrier from harsh environmental conditions (extreme thermal or mechanical shocks), extend food quality, and prolong shelf life [55,64].

Regarding this aim, a wide array of studies have shown that the incorporation of NPs—or generally nanofillers extracted from organic or inorganic sources (mainly polysaccharide or protein with animal or plant-based with strong reinforcing effects)—into packaging materials like various polymers used in food packaging industries, can produce polymeric nanocomposites with high barrier properties against gases, water vapors and, etc., offering qualified food with prolonged shelf life [65–68].
Previous studies have reported that nanocomposites reinforced with different nanofillers, such as silica NPs, clay, chitin, chitosan, nanocellulose, and metallic NPs (like Ag, Cu, Zn, and Ti) could be incorporated into food polymeric matrices and intensify the mechanical and barrier properties of packaging materials [69–72]. Scientists and researchers are working on the process of incorporating nanocellulose into food packaging. Nanocellulose has at least one dimension equal to or less than 100 nm and can be divided into cellulose nanofibers and cellulose nanocrystals produced mechanically and chemically, respectively [73]. Cellulose nanocrystals are coated on solid surfaces in multiple layers. This continues until a sufficient thickness exhibits the required mechanical, gas barrier, and wet resistance properties [74].

Furthermore, hybrid nanocomposites have been effectively incorporated into the polymers for food packaging [75]. Trifol et al. (2016) reported that the combination of poly-lactic acid (PLA), nanoclay, and nanocellulose led to a hybrid nanocomposite offering up to a 90% reduction in oxygen transmission rate (OTR) and also a 76% reduction in the water vapor transmission rate (WVTR) [76]. Additionally, Khorasani et al. in 2019 fabricated a colored biodegradable nanocomposite film composed of a dye and clay (DC NPs) embedded into PLA, which provided excellent barrier properties, including high thermomechanical resistance, also superior light, and mass transport barrier for packaging materials. Subsequently, they concluded that these features make nanocomposite films promising candidates for food packaging applications [77].

As another ability worth mentioning is that nanomaterials can act as biosensors in food packaging, enabling “intelligent” packaging systems involving highly sensitive nanomaterials to detect foodborne pathogens in foodstuffs or identify alterations in environmental factors affecting storage conditions of food products [78–80]. It should not be underestimated that nanomaterials could sense chemicals, toxins, and pathogens in food products. Numerous studies report various nanomaterials, including NPs, nanofibers, and even thin films, to have potential functions as nanobiosensors in food packaging [81]. These nanobiosensors could be placed in food packaging materials to sense alterations in external and internal conditions of food components [79]. Among different nanomaterials, NPs, due to having several distinctive physical and chemical features are eligible candidates for developing bionanosensors and generally biological detection systems [82,83]. Accordingly, A. A. Anvar et al. (2018) developed a molecularly imprinted polymer (MIP) by forming covalent bonds among methacrylic acid (MAA) monomers and hydrogen bonds between MAA and antibodies. Their developed nanobiosensor exhibited a remarkable capability to detect S. Typhi in polluted waters. They found that the MIP as a nanobiosensor is sensitive enough to detect S. Typhi antigens at concentrations of $10^{-1}$–$10^{9}$ [84].

Various applications of nanomaterial in food packaging industries are summarized in Figure 2.

Therefore, food packaging is considered one of the most applicable areas for utilizing electroless nickel plating (ENP), which actively and intelligently revolutionized traditional food packaging. Various nanoscale materials have been introduced to food packaging industries as functional additive materials, including Ag NPs, Cu NPs, Au NPs, TiO$_2$, ZnO, nanoclays, carbon nanotubes, cellulose nanowhiskers, and starch nanocrystals. These functional additive materials offer unique capabilities to the host packaging due to their different structures and properties [85]. Numerous reviews have discussed different types of nanomaterials, compared their properties and considered their eligibility for being utilized in food packaging. Recently, R. Sharma et al. also reviewed bionanocomposites, their antimicrobial properties and their application in food packaging industries. These are summarized in Table 1 [25].
reduction in the water vapor transmission rate (WVTR) \[76\]. Additionally, Khorasani et al. in 2019 fabricated a colored biodegradable nanocomposite film composed of a dye and clay (DCNPs) embedded into PLA, which provided excellent barrier properties, including high thermomechanical resistance, also superior light, and mass transport barrier for packaging materials. Subsequently, they concluded that these features make nanocomposite films promising candidate(s) for food packaging applications\[77\].

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A systematic review of eligible studies published during the last decade and related to the applications of silver, copper, and clay NPs in food packaging, is provided in Table 2. Special focus is placed on the migration and antimicrobial properties of the NPs of interest. Several of the most applicable nanomaterials in the food packaging industry are discussed in the following sections.

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**Figure 2.** The main applications of nanomaterials in the food industry.
Table 2. Literature review of the application of nanosilver, nanocopper, and nanoclay particles in food packaging, focusing on their migration and antimicrobial properties.

| Nanomaterial | Nanoparticle Size (nm) | Preparation Method | Analytical Techniques | Migration Test Conditions | Migration Results | Antimicrobial Activity Assay | Antimicrobial Activity Results | References |
|--------------|------------------------|---------------------|-----------------------|---------------------------|-------------------|------------------------------|-----------------------------|------------|
| AgCl-TiO$_2$ NPs (ATNPs) | 6–7 | Low-temperature-based one-pot sol–gel method | SEM | Disc diffusion assay for measuring anti-quorum-sensing activity of ATNPs (100–500 µg/mL) | Increase in inhibition of QS in Chromobacterium violaceum | [6] |
| Carboxymethyl cellulose nanobiocomposites containing metal NPs (ZnO, CuO, and Ag) | ZnO: 20–30 CuO: 35 Ag: <50 | Solution-casting method | SEM, EDXA, WVP, UV-vis, and tensile tests | Agar diffusion method | Ag and ZnO: high antibacterial activity against S. aureus and E. coli CuO: antibacterial activity against E. coli | [7] |
| Nanosilver-polyethylene composite | 7 | Nanosilver-polyethylene composite film was purchased (Anson Nanotechnology Co., Ltd., Zhuhai, China) | ICP-MS | Ag migration in 3% aqueous acetic acid: 1.70%, 3.0% and 5.6% at 20, 40 and 70 °C. Ag migration in 95% aqueous ethanol: 0.24%, 0.23% and 0.22% at 20, 40 and 70 °C | Japanese industrial standard (JIS) method | Intense antimicrobial properties against E. coli and S. aureus by polyethylene nanocomposites containing AgNPs | [8] |
| Low-density polyethylene (LDPE) film containing AgNPs | Not mentioned by the authors | Extrusion method | SEM, XRD, TGA, FTIR, DSC | Japanese industrial standard (JIS) method | Intense antimicrobial properties against E. coli and S. aureus by polyethylene nanocomposites containing AgNPs | [9] |
| PVA nanocomposite films containing nanocellulose (NC) and AgNPs | Not mentioned by the authors | Solution-casting method | SEM, EDX, FTIR, TGA, XRD, MRC, WVTR | Disc diffusion method | Maximum inhibition of E. coli was at 0.5 g AgNPs with 12 wt % NC. Maximum inhibition of S. aureus was at 0.3 g Ag NPs for 16 wt % NC. The highest antibacterial activity was observed for E. coli | [10] |
Table 2. Cont.

| Nanomaterial                                                                 | Nanoparticle Size (nm) | Preparation Method                                                                 | Analytical Techniques          | Migration Test Conditions | Migration Results                  | Antimicrobial Activity Assay | Antimicrobial Activity Results | References |
|------------------------------------------------------------------------------|------------------------|----------------------------------------------------------------------------------|--------------------------------|---------------------------|-----------------------------------|-------------------------------|--------------------------------|-------------|
| Carrageenan/AgNPs/laponite nanocomposite                                     | Not mentioned by the authors | Green synthesis method from the *Digitalis purpurea* plant                 | SEM, XRD, FTIR, WVTR            |                           |                                   | Agar disk diffusion method       | Strong antimicrobial activity against the *E. coli* and *S. aureus* | [11]        |
| AgNPs                                                                        | 20                     | FreshLonger™ plastic storage bags contained AgNPs, and produced by Sharpener Image Corporation (San Francisco, CA, USA) were purchased | SEM, ICP-MS, AAS, EDS           | EU regulation 10/2011    | No Ag migration into chicken meat | Total microbial count (CFU)   | Bags containing AgNPs had no effect on levels of the three types of spoilage bacteria, including TMC, Enterobacteriaceae and *Pseudomonas* spp. | [12]        |
| Cu NPs (CuNPs) embedded in polylactic acid (CuNPs-PLA)                      | Not mentioned by the authors | Pulsed-laser ablation                                                          | TEM, UV-vis, XPS, ETAAS         |                           | Cu release from the nanocomposites into an aqueous contact solution was carried out by atomic absorption analyses 24 h after putting coating in saline solution | Release kinetics of Cu from CuNPs-C-PLA nanocomposite was described according to first-order kinetic profile | Cell load of *P. aeruginosa* in CuNPs-C-PLA sample was lower (6.0 log CFU/mL) than the blank PLA sample (7.4 Log CFU/mL) | [13]        |
| Biopolymeric films (SA-CNW-CuNPs) containing cellulose, sodium alginate and CuO NPs | Not mentioned by the authors | Antibacterial polymeric film (APF) were prepared by using the three types of the components such as CuONPs, SA and CNW at different ratios | SEM, FTIR, EDS, XRD             |                           | Disc diffusion method                 | CNW (0.5%)-SA (3%)-CuO NPs (5 mM) films showed a higher zone of inhibition against *S. aureus*, *E. coli*, *Salmonella* spp., *C. albicans* and *Trichoderma* spp. | [14]        |
| Carbohydrate-based composite films incorporated with CuONPs                 | Not mentioned by the authors | Solvent-casting method                                                          | SEM, FTIR, XRD, UV-vis, WVVP    |                           | Total microbial count (CFU)       | Nanocomposite films with CuONPs exhibited strong antibacterial activity against foodborne pathogenic bacteria, *L. monocytogenes*, and *E. coli*. | [15]        |
| Nanomaterial                                                                 | Nanoparticle Size (nm) | Preparation Method | Analytical Techniques | Migration Test Conditions | Migration Results | Antimicrobial Activity Assay | Antimicrobial Activity Results                                                                 | References |
|------------------------------------------------------------------------------|------------------------|--------------------|-----------------------|---------------------------|-------------------|----------------------------|---------------------------------------------------------------------------------------------|------------|
| Cu/HPMC bionanocomposites (hydroxypropyl methylcellulose (HPMC) matrix incorporated with CuNPs) | 30                     | Chemical-reduction method | SEM, XRD, WVP         |                           | Disk diffusion method | CuNPs in HPMC suspension had high antibacterial activity against Gram-positive bacteria but not against Gram-negative bacteria | [16]       |
| Poly (ε-caprolactone)-based nanocomposites containing hydroxytyrosol (HT) and cloisite30B (nanoclay) | Not mentioned by the authors | Melt-blending       | DSC, TGA, SEM, TEM, XRD | Evaluation of the release rate of HT from the film for samples immersed in methanol. The extracts were analyzed by UV-vis spectroscopy | Presence of the nanoclay produced a decrease in the HT release from films | Organoclay and ZnONPs could significantly hinder the release of styrene monomers (SM) from nanocomposites to food simulants | [17]       |
| Polystyrene nanocomposites containing organoclay and ZnONPs                 | 20                     | Melt-mixing         | AFM, GC-FID            | 5 °C for 10 days, 40 °C for 24 h, 100 °C for 30 min |                      |                                            | Organoclay and ZnONPs could significantly hinder the release of styrene monomers (SM) from nanocomposites to food simulants | [18]       |
| Nanoclay-loaded low-density polyethylene (LDPE) composite                    | 2–8                    | Spherilene technology | XRD, TGA, CL, FTIR, AFM, SEM, TEM, ICP-OES | Ultraviolet (UV) irradiation and low concentration of ozone at 40 °C |                      | The presence of nanoclay accelerated in degradation of the LDPE and enhanced the release of clay particles | [19]       |
| LDPE nanocomposite                                                          | Variable               | Debbie Meyer BreadBags and Aisaika Everfresh bag as two models of nanoclay food containers were studied | ICP-MS, SEM-EDX        | EU regulation 10/2011 40 °C for 10 days and 70 °C for 2 h |                | Migration value of nanoclay from LDPE into acetic acid 3% for 10 days at 40 °C was higher than ethanol 10% | [20]       |
| Nanomaterial | Nanoparticle Size (nm) | Preparation Method | Analytical Techniques | Migration Test Conditions | Migration Results | Antimicrobial Activity Assay | Antimicrobial Activity Results | References |
|--------------|------------------------|---------------------|-----------------------|--------------------------|-------------------|-----------------------------|-----------------------------|------------|
| Polylactic acid (PLA) biocomposites containing Ag-based nanoclay | 20 | Solution-casting method | WAXS, TEM, DSC, WVP | The norm UNE-ENV 13130-1 | The migration levels of Ag was found within the specific migration levels referenced by the European Food Safety Agency (EFSA) | Total microbial count (CFU) | Ag-based nanoclay showed strong antimicrobial activity against Gram-negative Salmonella spp. | [21] |
| PLA/clay nanocomposite | Not mentioned by the authors | Blown film co-extrusion | TEM, DSC, XRD, WVTR, SEM | EU regulation 10/2011 | Lactic acid migration values in 50% ethanol remained well below the former generic specific migration limit of 60 mg/kg food (10 mg/dm$^2$) of the reg. EU 10/2011 | | | [22] |
| Nanocomposites of polypropylene (PP) + Clay | Not mentioned by the authors | Extrusion method | TGA, FTIR, WAXD, ICP-MS | European Standard UNE-EN 1186 | Migration of the PP-Clay was 0.0 ± 0.4 and 2.0 ± 0.4 mg/dm$^2$, with no significant differences, compared to the controls; migration rate was below those established by the EU regulation | | | [23] |
| Cassava starch films incorporated with cinnamon essential oil and sodium bentonite clay NPs | Not mentioned by the authors | Solution-casting method | Film thickness, WVP, TS | MIC | Cassava starch films incorporated with sodium bentonite and cinnamon oil showed significant antibacterial potential against E. coli, Salmonella typhimurium and Staphylococcus aureus. | | | [24] |
3.2. Inorganic and Metal Oxide Nanomaterials

3.2.1. Silver-Based Nanoparticles

Silver NPs (Ag NPs) are incorporated into thermoplastic packaging materials, such as polyethylene (PE), polypropylene (PP), polystyrene (PS) and nylon [86,87]. Researchers have found out that colloidal Ag particles can exhibit excellent antimicrobial activity against different types of bacteria like *E. coli* and *S. aureus* [88]. Further, Ag NPs can be incorporated into biopolymers, such as alginate, chitosan, and starch, which depict a high antimicrobial activity towards Gram-positive and Gram-negative bacteria [89]. Previous studies have found out that cellulose-based absorbents used as carriers for Ag NPs could significantly reduce the total aerobic bacteria counts while showed less effect on lactic acid bacteria and *Pseudomonas* spp. Ag NPs carried by cellulose-based absorbents can also cause a significant decrease in the population of these bacteria belonged to the Enterobacteriaceae family rather than control samples [90]. In this case, Ahari et al. (2018) showed that polyethylene films containing 5% Ag NPs exhibited high antimicrobial activity against *E. coli* and *S. aureus* and could be considered as an ideal nano-coating to extend food shelf lives [91]. Moreover, it has been interrogated that nanostructured starch-based films containing clay and Ag NPs, can majorly improve the mechanical and gas barrier properties and provide good antimicrobial activity due to complexation between Ag NPs and a large number of hydroxyl groups [92,93].

3.2.2. Copper-Based Nanoparticles

Copper NPs (CuO NPs) are useful in various fields, including food processing, biomedical equipment and devices, agriculture, and water treatments [94,95]. It has been identified that Cu NPs have indicated more antibacterial effects compared to Ag NPs [96]. Cu-based composites can be synthesized from both synthetic and natural biopolymers, such as cellulose, starch, and chitosan [97]. There are numerous studies that have demonstrated that Cu and Cu NPs could be incorporated in biopolymers and matrices. Regarding this, Mirhosseini et al. (2017) reported that Cu NPs, in combination with nisin, exhibit significant antibacterial effects against foodborne pathogens, such as *E. coli* and *S. aureus* [98]. Apart from antibacterial properties, nanocomposites incorporated with Cu NPs depict antifungal properties, as well. However, Cu is not generally used in food packaging industries as there are some concerns about its toxicity and ability for the acceleration of biochemical deterioration with foods through chemical oxidation.

3.2.3. Titanium Dioxide-Based Nanoparticles

Among metal oxide nanoparticles, TiO\(_2\) is an important photocatalytic semiconductor nanomaterial, which denotes strong properties, safety, and long-term physiochemical stability [99]. It shows self-cleaning and self-disinfecting properties and can inactivate the growth of a wide spectrum of microorganisms [100–102]. Many kinds of microorganisms can be killed by TiO\(_2\) upon illumination of light. Hydroxyl radicals and ROS generated on an illuminated TiO\(_2\) surface can oxidize the polyunsaturated phospholipid components of bacterial cell membranes [103]. Therefore, TiO\(_2\) NPs can be incorporated into materials using for food packaging to enhance their antimicrobial properties. TiO\(_2\) incorporated in packaging materials also possesses activities against foodborne microorganisms as well as odor, staining, deterioration, and allergens [103]. In one study performed by Ahari et al. (2015), the antifungal effects of titanium dioxide-based nanosilver packaging materials were compared to that of conventional polyethylene packaging in commonly consumed breads. They demonstrated that breads packaged with 10% titanium dioxide-based nanosilver coatings showed the lowest rate of fungal growth comparing to these samples coated with polyethylene, suggesting the employed nano-coating would be able to increase the shelf life of consuming breads and improve the food security [104].
3.2.4. Zinc Oxide-Based Nanoparticles

ZnO NPs can be incorporated into different materials, such as low-density polyethylene (LDPE), polypropylene (PP), polyurethane (PU), paper, and chitosan. Among metal oxides, ZnO NPs are considered as safe materials for human beings than other metal oxides [105]. Results of some studies have revealed that glass incorporated with ZnO NPs depicted antimicrobial activity against \textit{E. coli} and \textit{S. aureus}, while incorporated PU films had antimicrobial activity against \textit{E. coli} and \textit{B. subtilis}. Further ZnO nanorods deposited onto glass surface showed antifungal activity against \textit{Candida albicans}. These can be incorporated into biopolymers, such as chitosan in food packaging, which is slightly less effective than Ag NPs incorporated into nanocomposites. However, researchers have reported that 99.92% of viable bacteria of \textit{E. coli} was inactivated after 24 h incubation period. In addition, these types of NPs enhanced antibacterial effect against \textit{B. subtilis}, \textit{Enterococcus faecalis}, \textit{P. aeruginosa}, \textit{S. Typhimurium} and \textit{S. aureus}. The mechanism behind these antimicrobial effects is deeply rooted in the disruption of bacterial cell membranes by Zn ions and oxidative stress because of photocatalytic production of ROS [106].

3.2.5. Silicon Dioxide-Based Nanoparticles

Silicon dioxide or silica NPs (nSiO2) have been reported to improve the mechanical properties and/or barrier of several polymer matrices. Wu, Zhang, Rong, and Friedrich (2002) observed that the addition of nSiO\textsubscript{2} to a PP matrix improves the tensile properties of the material not only in strength and modulus but also in tensile length [107]. Xiong et al. (2008) also reported the addition of nSiO\textsubscript{2} to the starch matrix. They also observed that nSiO\textsubscript{2} reduces water uptake by starch [108]. Vladimirov et al. (2006) combined nSiO\textsubscript{2} into an isotactic polypropylene (iPP) matrix using polypropylene bonded maleic anhydride (PP-g-MA) as an adapter [109]. nSiO\textsubscript{2} enhances the iPP storage module, strengthens the material, and improves the oxygen barrier of the matrix. Zhang et al. (2007) fabricated polyvinyl alcohol (PVOH) nanocomposites with nSiO\textsubscript{2} by radical copolymerization of vinyl silica and vinyl acetate NPs [110].

3.2.6. Nano Clays

Nanoclays, which are NPs essentially composed of layered mineral silicates stacked together, are currently considered as the most commercially applicable nanomaterials all around the world [111]. They have been known as cost-effective nanofillers to reinforce polymer nanocomposites and to improve the mechanical, thermal, and barrier properties of polymeric nanocomposites for food packaging [112,113]. Currently, there are various clay minerals, which are categorized into four major classes, regarding their structure and chemical properties as well as sources, including chlorite, MMT/smectite, illite, and kaolinite.

3.2.7. Montmorillonite Nanomaterials (MMT)

Currently, the incorporation of filler particles into polymers, including MMT) and kaolinite nanoclay, carbon in the formation of nanotubes, and graphene nanosheets are addressed [114]. MMTs are known as the most widely used clays in the preparation of nanocomposites. These clays are composed of nanometer-magnesium aluminum silicate platelets. Their dimensions reach one nanotic and 100 to 500 nm in diameter, leading to the formation of large platelets [115]. The clay structure is composed of hundreds of layers. Platelets collected in droplets vary from 8 to 10 \( \mu \text{m} \) in diameter. The effect of nanoclays on polymer properties is mainly due to their different surface area by volume since the interactions of the polymer and the filler are controlled by the interface forces. The clay particles must be dispersed as separate platelets and evenly within the polymer [116]. Stretched nanoclays are involved in improving the impermeability properties of polymeric materials. When nanoclays are dispersed in polymers, they form a maze structure. Noble gases show a troublesome path, greatly reducing their penetration rate [117]. Traditional
composite structures contain large amounts of filler (approximately 60% by volume), but nanocomposites have significant variations in very low charge properties (<2% volume).

3.2.8. Organic Polymer-Based Nanomaterials

Over the past decades, as has been reviewed earlier, polymers have been replaced with conventional materials (such as metals, ceramics, and paper) in food packaging. Due to worthy features of polymers, such as performance, lightness, ease of processing, and low cost, using synthetic polymers in food packaging industries has been increased [118,119]. These polymers provide all kinds of mechanical, chemical and microbial protection for the food packaging materials.

According to the origin of raw materials and production processes, polymers can be broadly classified into two categories as follows: Natural biopolymers and Synthetic polymers.

4. Natural Biopolymers

Biopolymers have potentially attracted considerable attention as an alternative to conventional plastic packaging materials like plant-based carbohydrates, such as starch, cellulose, chitosan, alginate, agar, and animal proteins, as well as plant-based proteins like soybean protein, corn zein, wheat gluten, gelatin, collagen, whey protein, casein and, etc. [15,120].

Most reports on the formation and properties of biopolymers films focus on their use as edible films (being incorporated in the form of biodegradable films) [121–124]. Biopolymers exhibit relatively weak mechanical and barrier properties, which are currently limited to industrial usages. Particular challenges are developing moisture barrier properties due to the hydrophobic nature of biopolymers. However, biopolymer-based packaging may prevail with nanocomposite technology. The formation of nanocomposites has been proven to improve the mechanical, thermal, and barrier properties of polymers [15].

Using the most appropriate polymers can create an effective barrier against water, gases, and greases. These over-thick plastic crystals create a nanometer winding path structure that is inherently resistant to penetration. There are detailed reports have been published by several researchers on biopolymer nanocomposites [125–127]. Starch has been widely studied as a substance of choice in food packaging due to its environmental friendliness and widespread availability [128,129].

Polymers have been proposed to improve starch resistance [130,131]. Accordingly, there is a remarkable interest in starch mixtures with nanoclays in food packaging. Clay starch is the most widely used degradable nanocomposite for various applications, including food packaging [132,133]. There are significant advances in increasing the mechanical properties of both Young’s modulus and tensile strength with the addition of MMT clay. It has been reported that the impermeability properties of MMT clay are of major importance in a bottle and food packaging [134]. Cyrus et al. (2008) reported that the effective diffusion coefficients for nanocomposites were lower for starch. This suggests that in addition to the ability in water absorbance of the starch films, it is probably reduced due to the tortuous structure formed by the peeling clay of MMT [134]. Recently, sodium starch/ZnO-carboxymethylcellulose nanocomposites have been prepared by using ZnO NPs stabilized by carboxymethylcellulose [135].

4.1. Synthetic Polymers

Industrial usage of synthetic polymers due to mechanical, chemical, and microbial protection is of great prominence in food packaging. Polyethylene (low-density or high-density), polypropylene, polystyrene, polyvinyl chloride (PVC), nylon, polyethylene terephthalate are enumerated as the most frequently utilized polymers in food packaging [136]. Biodegradable polymers, such as polylactic acid (PLA), polyglycolic acid (PGA), poly-ε-caprolactone (PCL), polybutylene succinate (PBS), and polyvinyl alcohol (PVA) are synthetic polymers derived from petroleum resources. These polymers could be processed
through conventional plastics processing methods and used in three main areas, including agriculture, medicine, and food packaging [137].

In the following, some of the marked natural and synthetic polymers in food packaging are discussed.

4.2. Polylactic Acid (PLA)

As properties like being easy to handle in making monomers, having less environmentally burden, sustainability, biocompatibility, and biodegradability are considered as the most important ones for nanomaterials used in nanocomposites for food packaging purposes, it has been demonstrated that PLA-based nanocomposites are one of the acceptable types of polymers used in food packaging. In spite of the fact that there are several merits for the usage of PLA in food packaging, there are also several drawbacks as having no cost-effectiveness and low performance due to gas barrier properties. Results of several studies depicted that adding plasticizers or chemically modifications of PLA-based nanocomposites can effectively hamper these shortcomings and improve barrier properties of PLA-based nanocomposites used for food packaging [137].

4.3. Polyhydroxyalkanoates (PHAs)

Bioplastics like polyhydroxyalkanoates (PHAs) are considered a potent nominee for food packaging. The ideal features of PHAs comprise hydrophobicity, biodegradability, nonlinear optical activity, thermoplasticity, piezoelectricity, nontoxicity, and impermeability to water and/or gases [138]. In research, PHA-based film incorporated with clove essential oil was prepared for active food packaging purposes. No microbial growth was reported on the film surface [139]. Migration is a key issue in food packaging since monomers or additives employed in PHA preparation might conceivably migrate into the food matrix. Incorporation of PHA with other biomaterials has been considered as a way of improving the vital properties while keeping biodegradability [140]. Furthermore, interest in promoting the main properties via fabrication of PHA-based nanocomposites by addition of organic or inorganic NPs, such as MMT, layered double hydroxides, cellulose nanowhiskers, multi-walled CNT, has rapidly increased [141]. It was also approved that PHA-based films can include metallic NP, such as AgNPs, CuNPs, and ZnONPs, to own more antibacterial, improved barrier and mechanical properties for food packaging purposes.

4.4. Polyamine Polyacid (PPA)

Polyamine polyacid (PPA) is a material with stable compatibility properties, biocompatible, biodegradable with good mechanical properties and optical properties. Lactic acid, a PLA monomer, may easily be produced by the fermentation of carbohydrate raw materials. Thus, PLA has more disposal options and is environmentally heavier to produce than natural oil-based plastic. However, the large-scale use of PLA as a packaging material is still very low in performance and cost compared to other polymers [142,143]. Nanotechnology has the potential to improve the PLA polymer properties and expand applications of composites [156,143]. Amorphous and chemically modified kaolinite PLA nanocomposites were investigated by Cabedo et al. 2006. They observed a good interaction between the polymer and the clay, which resulted in an increase of about 50% in the oxygen permeability. The study also includes the addition of plasticizers to overcome the inherent fragility of PLA. The nanocomposite is a combination of PLA and laminated montmorillonite silicate resulting in nanocomposites with barrier properties suitable for food packaging applications [144].

4.5. Starch Nanocrystals

Native starch grains can be considered due to the longtime of hydrolysis at low temperatures of gelatinization when amorphous areas are hydrolyzed. This property leads to the separation of crystalline lamellae, which are more resistant to hydrolysis. Crystalline starch particles show platelet morphology with thicknesses of 6–8 nm [145].
Kristo and Biliaderis (2007) reported that the addition of starch nanocrystals (SNCs) highly increases the tensile strength and modulus of pullulan films but decreases their length. Experimentally, the water vapor permeability of pullulan films is reduced by adding 20% or more of SNC [146]. According to Chen et al. (2008), the addition of SNC to PVOH led to a slight improvement in the tensile strength and tensile strength of PVOH by adding SNC up to 10 wt. PVOH composite with SNC indicates that SNC disperses more uniformly and creates stronger interactions with PVOH than native starch grains [147].

4.6. Chitin/chitosan Nanoparticles

Zhang et al. (2004) and Rujiravanit et al. (2005) prepared chitin whiskers using hydrolyzed chitin acid [148,149]. Medium dimensions of whiskers obtained by Lou et al. (2004) were 500 nm (length) and 50 nm (diameter), and those obtained by Rujiravanit et al. (2005) were 417 nm (length) and 33 nm (diameter). Lu et al. (2004) added chitin whiskers to isolated soy protein thermoplastics (SPI) and reported that whiskers not only enhance the tensile properties of the matrix but also they greatly increase their water resistance. Chitosan NPs can be formed using ionic gels, containing positively charged amino groups of the electrostatic form of chitosan and interact with crosslinker polyanion, such as Tripolyphosphate. De Mora et al. (2009) prepared chitosan-tripolyphosphate (CS-TPP) NPs and combined them into a hydroxypropyl methylcellulose (HPMC) film [150]. In addition, using CS-TPP NPs significantly improved mechanical and movie barrier features. This group of researchers attributed some of the properties of CS-TPP NPs to the discontinuity of NPs in the HPMC matrix.

4.7. Protein Nanocomposites

The ability to use different proteins to form films has been used in industrial applications for a long time. Accordingly, animal-derived proteins used in commercial applications to form films are mainly casein, whey protein, collagen, egg whites, and myofibrillar fish [151,152]. Plant-based proteins industrially used to form films include soy protein, saddle (corn protein), and wheat gluten [153]. Compared to polysaccharide films, protein-film-based nanocomposites have acceptable advantages such as better oxygen barrier properties, less water vapor permeability due to their polarity, more linear (non-ring) nature and structure, and lower free volume. However, serious concerns remain about their performance in food packaging, including their high modulus, high water absorption, and high gas permeability. Researchers have made meticulous efforts to improve the various properties of proteins used in nanocomposite-based technology. As a prime instance, whey protein has received great attention as an edible film and coating material, and there are several studies conducted on the investigation of whey protein as an edible film. Sothornvit and Krochta (2005) reported an acceptable efficacy and oxygen barrier properties in clear whey-protein films [154].

Zhou et al. (2009) added TiO$_2$ to a nanocomposite with antimicrobial properties. They demonstrated the potentials of TiO$_2$–whey nanocomposites used as food packaging materials in terms of food grade and biodegradability [155]. Similar studies have aimed at investigating interactions in the ZnO–whey-protein nanocomposite [156].

Because of it is thermoplastic properties and potential as a degradable plastic, soy proteins have been studied by many researchers. However, due to its poor response to moisture and high strength in several experimental studies, its degradability has not been effectively exploited [157]. Soy protein is similarly combined with starch to overcome fragility with plasticizers. However, using plasticizers entails disadvantages like a further reduction in barrier properties. Soy protein nanocomposite films showed a reduced water vapor permeability and improved modular strength, and elastic tensile strength than their filler-free counterparts [158,159].
In their comparative experimental study, Din and Yu observed an increase in tensile strength without and with ultrasonic treatment in soy protein nanocomposites of 23% and 47%, respectively. Chen and others investigated the interaction of soy protein and the properties of the electrostatic interaction between soy protein (positive charge) and MMT layers as a hydrogen bond between –NH and the Si–O groups, which are the mechanisms behind interactions in the protein/MMT system. Such mechanisms increase the mechanical strength of the nanocomposites.

Corn zein is a relatively hydrophobic protein known to make films in corn kernels [159,160]. Corn zein is used in the food industry as a coating agent and as a degradable decomposer. Although Corn zein products are less sensitive to water compared to other biopolymers, they still show high water vapor permeability and low tensile strength compared to commodity polymers. The formation of Corn zein-based nanocomposites using kaolin nanoclays intensifies barrier coatings and other useful properties when used on paper and cardboard in food packaging. They are expected to replace fluorocarbons in extruded polymer barrier coatings [160].

5. Effects of Migration of Nanoparticles in Foods on Human Health Situation

Despite the various types of nanocomposites and their effectiveness mentioned above, there are concerns over the serious risks of NPs for consumers. It has been reported that potential migration and exposure with NPs from food packaging into food and beverage can affect the immune system [161]. However, experimental data on migration are not currently much available, and many food packaging types containing NPs are already available in commercial use in some countries. A pioneering study was undertaken by a group led by Maurizio Avella from Italy in 2005 [132]. Nanocomposite films were obtained by homogeneously dispersing functionalized layered MMT NPs in different thermoplastic starches, such as neat potato starch and a mixture of potato starch with biodegradable polyester, via polymer melt processing techniques. The conformity of the nanocomposites with actual regulations and European directives on biodegradable materials was verified by migration tests. The vegetable samples—including lettuce and spinach—were cut into small samples and packed into bags for the contact tests. Subsequently, the closed bags were placed in an electrical furnace and heated to 40 °C for 10 days. After this process, every bag was slowly cooled to the ambient temperature and submitted to the subsequent characterization. The vegetable samples were digested by heating in a furnace at 105 °C to a constant weight, then carbonized in a muffle at 550 °C for a few hours. Finally, they were cooled in a dry atmosphere for 40 min. The ash obtained was dissolved in hydrochloride acid solution, put in a hot water bath and finally filtered. The filtrate was analyzed by atomic absorption equipment to measure the release of Si, Fe and Mg ions. The analyzed results demonstrated an insignificant trend in the levels of Fe and Mg ions in packaged vegetables, but a consistent increase in the amount of Si, which is the main component of the MMT NPs, was observed [162]. Migration-based studies on NPs in food contact materials and food packaging are relatively scarce. Incidentally, difficulties in the characterization of NPs used in nanocomposites and inaccessibility of these analytic techniques (including atomic absorption, inductively coupled plasma-based emission or mass spectroscopy, and X-ray diffraction) for qualitative and quantitative analysis on the migration of NPs in nanocomposites are responsible for this limited research [8,163].

The migration of NPs from packaging to packed food raised a public health concern and had been corroborated by animal oral administration and in vitro cell experiments. In this case, there is another study done by Ahari et al. (2021) investigated the migration of Ag and Cu NPs from food coatings to food ingredients, cytotoxicity, and their hazardous effects on the health situation. They reported that the possibility of migration of Ag and Cu-based NPs is more in acidic environments. Entrance of such ionic NPs to cellular microenvironments and damage host cells through changes in mitochondrial functions, adverse effects by ROS production, and induction of chronic disorders [163]. In general, two types of mechanisms can be adopted to explain the toxicity effects of migration of...
NPs-based nanocomposites from food packaging into food context on humans. One is that the toxicity procedure is independent of the NPs, and could be realized by generating ROS within the cells [164]. Another one is the existence of a strong relationship between toxicity procedures and the chemical composition of NPs. For example, the crystallization and recrystallization of some metal or metal oxide NPs modify the secondary or tertiary conformation of the proteins. While other types of NPs, such as the metal alloy, or single-wall carbon nanotube (SWNT) or MWNT, can result in direct or indirect gene toxicity [165]. Another reason related to migration goes back to the reductive trend in the viscosity of nanoparticle/polymer composites, which is accompanied by an incremental trend in the migration rate of NPs into foodstuffs [166,167].

Some toxicities are dependent on the chemical composition of the NPs. For example, results from an experimental study indicate that the accumulation of insoluble NPs in the human body may be responsible for the compromised functions of the gastrointestinal (GI) system and the emergence of chronic autoimmune disorders like inflammatory bowel disease (IBD) and Crohn’s disease (CD). Although little is known about whether NPs are linked to the initiation of IBD and CD, it seems that the NPs may act as adjuvant triggers for exacerbation of these mentioned diseases. For instance, if some insoluble NPs (such as TiO$_2$, ZnO, and SiO$_2$) are absorbed by GI and pass through the intestinal tracts, they can contact and adsorb calcium ions and even lipopolysaccharides. The resulting triple structures (NPs–calcium–lipopolysaccharides conjugates) can activate both peripheral blood mononuclear cells and intestinal phagocytes, which are usually resistant to stimulation [168]. Metal or metal oxide NPs, such as Fe/Pt alloy, Co/Cr alloy, ZnO, SiO$_2$, TiO$_2$, SWNT, and MWNT represent direct and indirect genotoxicity. For TiO$_2$ and CB, it was reported that particles with a mean size of \( \sim 20 \) nm induced DNA damage, while larger particles (\( \sim 200 \) nm) showed no genotoxic effect [169].

As has been reviewed earlier, nanosilver-based nanocomposites can be easily oxidized and turned into ionic structures. Additionally, they have the potentials for reformation in chemically reductive situations. Hence, to solve the problems related to the possibility of migration of NPs into the food context, it seems that encapsulation can be a solution for refraining from detrimental effects on the health situation of human beings [170]. Additionally, results of another study demonstrated migration of Ag from Ag NPs into milk products through inductively coupled plasma–atomic emission spectrometry. Their results showed that treatment of Ag NPs with the diblock copolymer and polystyrene–block–polyethylene oxide could significantly reduce the migration rate of Ag ions in Ag NPs [171].

6. Environmentally Friendly Nano Packaging in Food

Today, most of the materials used for food packaging are virtually non-degradable and represent serious global environmental issues. Edible and biodegradable films represent a major approach to increasing shelf life and quality of food while reducing packaging waste and even loss of food [172]. However, the use of edible and degradable polymers is limited due to problems related to performance (such as fragility, weak gas or moisture barrier), processing (such as low-temperature heat distortion), and cost. For example, starch has attracted considerable attention as a degradable decomposer. The thermoplastic polymer has poor performance due to its sensitivity to water and limited mechanical properties with high fragility related to the anarchic growth of amylose [173].

7. Improvement in Barrier Properties of Polymers by Nanoparticles in Food Packaging

Increasing the path length or blocking the passage of oxygen gas in the polymer by various particles is a new approach that has become one of the most important and effective pathways. There are several strategies for making improved barrier properties in polymers (like bioplastics) used for food packaging due to refraining from the migration of oxygen, carbon dioxide, anhydride, aroma, flavor, and water vapor to the ingredients of food, eventuating to make them as the most sustainable biopolymers [68,174,175]. Results
of several studies proposed nanoscale size fillers like MMT and kaolinite clay. Protein, starch, cellulose, PLA-based nanocomposites are the ones that were separately reviewed earlier and compared, guaranteeing barrier performances and physical packaging integrity. Taken together, it seems that industrial usage of polymer blends, multilayer systems, natural biopolymers with chemical modifications, coatings, and composites can improve the features mentioned above [68,174,175].

Preserving food properties like taste, color, flavor, texture, and nutritional value provides proper mechanical strength, barrier properties, and antimicrobial features for packaging compounds. NPs showed their potential in improving the barrier properties of novel food packaging systems. Size, morphology, and type of nanoparticles affect their potential in improving the barrier properties [176]. The shape of the particles in the change in permeability of the composite relative to the background is a function of the volume fraction of the particles. In fact, the data volume fraction is the main input in most permeability prediction models, showing the sub-relationship of how the weight fraction is converted to volume [177]. The following equation shows how the weight loss is converted to volume:

$$\varnothing = \frac{w}{\rho_p + \frac{1-w}{\rho_m}}$$

(1)

where $\rho_p$ and $\rho_m$ are the density of the particle and the polymer, and $w$ is the weight percentage of the particle in the polymer. Spherical and cylindrical NPs mostly showed different effects on barrier properties compared with plate NPs. To increase the distribution of particles in the polymer field, the chemical surface improvement of particles is usually used, and the most important goal is to bring closer the polarity of the polymer and nanoparticles [178]. In general, according to the polymer background, various carbonyl, hydroxyl, etc., groups can improve the surface of particles. For example, in spherical particles, such as $\text{SiO}_2$, silyl or silane, functional groups can improve the particle surface. One of the most important reasons for the decrease in permeability by these particles may be the increase of curvature in the passage of oxygen-permeable gas, germination in the polymer matrix and the formation of an adsorbent crystal in the condenser [179]. In this regard, it seems that nanoclay, due to its plate form, may illustrate more improvement in barrier properties comparing with Cu and Ag NPs.

8. Discussion

In recent years, following the growing world population, much attention has been paid to increase the production of foodstuffs, including agricultural and livestock products, sufficient to meet global nutritional demands. Apart from the necessity to increase the production of food products, the issue of providing suitable conditions for long-term storage and preservation of food products, increasing food shelf-lives, and preventing food spoilage through inhibition on the growth of bacteria, fungi, or any other pathogens is of particular importance and emphasize on the key role of the food packaging industry. In the last decades, nano-biotechnology science has revolutionized, so the food packaging systems that numerous nanomaterials are currently being incorporated in food packaging materials to enhance food shelf life, preservation of some food features like freshness, taste, and color as well as to provide antimicrobial effects for packaging materials against a wide variety of pathogenic microorganisms [180,181]. The applicable nanomaterials in food packaging include NPs, such as metals and their oxide forms like nanosilver, nanocopper, nanogold, $\text{TiO}_2$, $\text{ZnO}$, and magnesium dioxide, nanoclays, which are composed of layered mineral silicates and classified into several classes, such as montmorillonite, bentonite, kaolinite, hectorite, and halloysite, and also biopolymers like starch, cellulose, chitosan, plant and animal proteins, etc. However, biopolymers as food packaging materials showed drawbacks, such as low strength, poor mechanical, thermal, and barrier features [126]. To improve the properties of the biopolymers, much research has been conducted to reinforce biopolymers with different types of NPs playing roles as nanosized fillers, introducing bionanocomposite concept this means a multiphase material composed of two or more
compounds, including biopolymers and one or two nanofillers [136]. The use of nanofillers, such as Ag, titanium dioxide, clays and silicates for reinforcement of the biopolymers could improve not only the chemical and physical properties of biopolymers but also allocate some privileged features to nanocomposites, such as antimicrobial, antioxidative and biosensing abilities [182,183]. It is not surprising that NPs exhibiting a variety of intrinsic physicochemical features possess numerous functions. Antimicrobial activity is one of the most remarkable properties identified in NPs. Hence, it has been proposed that these antimicrobial nanomaterials can be considered promising alternatives to common antibiotics [184]. As mentioned earlier, the inhibitory effects of NPs on the growth of different microorganisms rely on two major mechanisms, including (1) disruption of cell membrane integrity and potential and (2) production of free radicals such as ROS, which can lead to induction of oxidative stress. It has been revealed that the physical properties of NPs, like size, shape, and chemical modifications and combination with other NPs, influence their antimicrobial activity [185]. Jonghoon Choi et al. (2014) examined the antimicrobial effects of the different sizes of Ag NPs on *Methylobacterium* spp. They prepared Ag NPs with controlled sizes by chemical reduction of Ag cations and found that the smaller Ag NPs around 10 nm showed more antibacterial activity than 100 nm particles [186]. In another study, Yamamoto (2001) prepared ZnO samples in a size range of 0.1 to 0.8 µm and tested the samples’ antimicrobial activities against *S. aureus* and *E. coli* by assessing changes in electrical conductivity with bacterial growth. His research indicated that the antibacterial activity of ZnO particles significantly increased along with reducing particle size [187]. On the other hand, numerous studies demonstrated that however smaller NPs manifest higher antimicrobial effects against common pathogenic bacteria, they also show a higher rate of migration into foodstuffs than the larger NPs, making their application in food packaging more challenging [170]. Yolanda Echegoyen and Cristina Nerín (2013) examined the migration of nanosilver particles from three commercial nanosilver plastic food containers into food simulants (3% acetic acid and 50% ethanol. They found that the migration rate of small Ag NPs aggregates and some other particles bigger than 200 nm was significantly higher than larger Ag NPs [188]. The result of these studies implicates that using NPs in optimal size and concentrations is necessary for obtaining NPs with high antibacterial activity and low migration rate in food packages, ensuring long shelf life and safety of foodstuffs. Regarding this point, Ahari et al. (2019) could prepare Ag NPs in optimum size and concentration for use in food packaging. They prepared Ag NPs with a size less than 50 nm and examined NPs concentrations of 1000, 2000, 3000, 4000, 5000 and 6000 ppm to obtain the proper concentration in which NPs show high antimicrobial activity and low migration rate. They observed a remarkable decrease in growth of *S. aureus*, *E. coli*, *Aspergillus flavus* and *Penicillium* strains in 5000 and 6000 ppm nanosilver packages. Moreover, their migration tests showed that the migration rate of nanosilver particles with the studied concentrations into their samples (Sturgeon caviar) was completely negligible [189]. As another important note, many studies have reported that as all metallic NPs cannot kill pathogenic microorganisms, the combination of different metallic NPs could be effective against pathogens. One study performed by Bankier et al. (2019) showed that using three metallic NPs, tungsten carbide (WC), Ag and Cu, in combination together exhibited significantly higher antimicrobial effects against *S. aureus* and *Pseudomonas aeruginosa* compared to using them separately [190]. In another study by Ahari et al. (2018), the antimicrobial effects of several nanocomposites of polyethylene containing Ag, clay, and TiO₂ NPs, which were prepared by melt-mixing and sol–gel methods, were compared. Their results showed that the nanocomposites containing clay and titanium dioxide both lacked antimicrobial activity on *E. coli* or *S. aureus*, while it was observed that the nanocomposites containing both 5% Ag and 5% TiO₂ NPs possess the highest antimicrobial functions and could be considered as a proper cover for foodstuffs [91].
Additionally, recent studies have reported that as preparation methods of NPs or nanocomposites affect their physical features such as size, the type of method employed may also indirectly be effective on the antimicrobial potentials of NPs. Regarding this finding, Ahari et al. (2018) showed that the preparation of low-density polyethylene/Ag/titanium dioxide nanocomposites using sol–gel and melt-mixing methods could effectively increase their antimicrobial activity against pathogens such as *E. coli*, *S. aureus*, *C. albicans*, and *A. niger*. They also found that the efficiency of antimicrobial activity of NPs prepared through the melt-mixing method was highly better than the sol–gel method, which was concluded by measurement of the inhibition zone caused by nanocomposites. Thus, they suggested that low percentages of NPs produced by the melt-mixing method could impressively increase food shelf life, so they are highly recommended for packaging of expensive food products [191]. As a result, to achieve suitable and safe nanomaterials applicable in packaging materials to increase the food shelf lives, it is critical that all the described parameters related to nanomaterials—including the selection of appropriate NPs or biopolymers for applying alone or in combination—are used in the validated preparation methods, including applying NPs in the right size and concentration.

9. Conclusions

As there is a direct relationship between food quality and the health situation of consumers, there is an increasing demand for the prolonged shelf life of food in storage and processing—especially “ready-to-eat” foods. According to increasing ecological and environmental concerns on conventional food packaging, it seems that now is the exact era for applications of modern sciences in nanotechnology for food packaging purposes. Hence, it seems necessary to understand the different materials used in this field with the biological, chemical, mechanical, and antimicrobial properties of these materials used in food packaging. It was demonstrated that the results of such studies could provide comprehensive views on the quality of material, especially barrier properties. By using polymers with high barriers for gases, flavor, aroma, and water vapor, the migration of material into food ingredients is reduced, and food quality is increased by prolonged shelf life. Subsequently, such results can facilitate food transfer/food distribution and reduce the risk factors for long-term food storage. Additionally, the usage of optimal methods in the production of NPs and better monitoring of food may reduce concerns about the quality of food packaged by consumers.

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Abbreviations

| Silver        | Ag     |
|---------------|--------|
| Copper        | Cu     |
| Gold          | Au     |
| Iron oxide    | (Fe₃O₄) |
| Copper oxide  | CuO    |
| Titanium oxide| TiO₂    |
Zinc oxide  ZnO  
Magnesium oxide  MgO  
Calcium oxide  CaO  
Reactive oxygen species  ROS  
Water vapor transmission rate  WVTR  
Molecularly imprinted polymer  MIP  
Starch nanocrystals  SNCs  
Methacrylic acid  MAA  
Electroless nickel plating  ENP  
Nanoparticles  NPs  
Silicon dioxide or silica  nSiO₂  
Chitosan-tripolyphosphate  CS-TPP  
Carbon nanotubes  CNTs  
Polyvinyl pyrrolidone  PVP  
Thyme essential oil  TEO  
Salmonella Typhi  S. Typhi  
Escherichia coli  E. coli  
Staphylococcus aureus  S. aureus  
Listeria monocytogenes  L. monocytogenes  
Crohn’s disease  CD  
Pseudomonas aeruginosa  P. aeruginosa  
Montmorillonite  MMT  
Total microbial count  CFU  
Low-density polyethylene  LDPE  
Polypropylene  PP  
Polyurethane  PU  
Polyvinyl chloride  PVC  
Polyactic acid  PLA  
-ε-Caprolactone  PCL  
Polybutylene succinate  PBS  
And polyvinyl alcohol  PVA  
Polyamine polyacid  PPA  
Polyhydroxyalkanoates  PHAs  

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