

Oxide-Based Nanocomposites for Food Packaging

Application: A Review

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ABSTRACT

Silver nanoparticles and/or nanoclay [particularly montmorillonite] are used in the majority of nanotechnology applications for food packaging. Other nanomaterials, on the other hand, can also be integrated into packaging. Metal oxide nanoparticles have been added to petroleum-based and biopolymers to produce nanocomposites with improved mechanical, barrier, antioxidants and antimicrobial properties. Nanoparticles migration from packaging, on the other hand, is a source of concern due to their potential toxicity in the human body and the environment. The purpose of this article therefore, was to review the available literature on the utilization of metal oxide-based nanoparticles to produce nanocomposites for food packaging application. Advantages of incorporating metal oxide-based nanoparticles into polymers, as well as migration of these nanomaterials from packaging into foods are discussed. Incorporation of metal oxide nanoparticles into polymers allows for the production of nanocomposites with increased mechanical strength, water and oxygen barrier properties, and can also confer other additional functional properties, such as antioxidant, antimicrobial activity and light-blocking properties. According to migration studies, only a small quantity of nanomaterial migrates from packaging into food simulants or foods, implying that consumer exposure to these nanomaterials and the health concerns associated with them are low. Nonetheless, there is a scarcity of information on the migration of nanomaterials from packaging into actual foods, and more research is desperately needed in this area. This manuscript is useful in the food industries as it indicates the applicability and potential of the oxide-based nanocomposites as a promising approach for use in food packaging applications.

Keywords: Food packaging, Metal oxide, Migration, Nanoparticles

INTRODUCTION

Food packaging plays a vital role in food safety, quality, and shelf-life extension by protecting food products from physical,

environmental, chemical, and microbial hazards during storage and transportation (Youssef & El-Sayed, 2018; Hoseinnejad et al., 2018). In the international market,



packaging also simplifies product end-use suitability and communication at the consumer level. New packaging materials have been developed because of advances in materials science and a need for nutritious food with few additives that stay fresh for longer. Multilayered packaging has improved “barrier properties” and allowed new product lines to be developed like retorted foods; yet this packaging is expensive to manufacture and difficult in recycling (Bumbudsanpharoke & Ko, 2015; Duncan, 2011). As a result, developing monolayer packaging having improved properties, transparency, lightweight, biodegradability or ease of recycling, is required, and nanotechnology can assist in this endeavour (Chaudhry et al., 2008; Garcia et al., 2018).

Nanotechnology is a multi-disciplinary discipline in which materials with at least one dimension between 1–100 nanometers are used (Bumbudsanpharoke & Ko, 2015; Mihindukulasuriya & Lim, 2014). Nanomaterials have a larger surface area than their bulk equivalents due to their small size, resulting in increased reactivity and properties that differ from macroscale level materials (Uskokovi, 2007). Nanomaterials and polymers can thus be mixed to produce nanocomposites having improved barrier, mechanical and thermal properties. As nanoparticles are incorporated into the polymer matrix, they act as reinforcement, and their lesser water and gases permeability causes a diffusion path that is longer and more convoluted, resulting in improved barrier properties in comparison to neat polymer. In addition, bonds formation between the nanoparticles and polymer reduces the sites number that may interact with water molecules in the polymer chain (Duncan, 2011; Mihindukulasuriya & Lim, 2014). A nanomaterial's high aspect ratio and homogenous dispersion in a polymer matrix alters the molecular mobility and relaxation of polymer chains, therefore enhancing

mechanical and thermal resistance (Bumbudsanpharoke et al., 2015). Furthermore, the increased strength of nanocomposites may be due to the bond between polymer chains and nanoparticles (Zhou et al., 2009; Garcia et al., 2018).

Using nanotechnology involving metal oxide nanoparticles and either petroleum-based or biobased biopolymers, “active packaging” & “intelligent packaging” can be developed. Food is protected by active packaging, via mechanisms activated by internal or external factors, whereas “intelligent packaging” offers info on the food's real status (Chaudhry et al., 2008; Ding et al., 2020; Mihindukulasuriya & Lim, 2014). Direct contact between metal oxide nanoparticles and microbial cells and/or the release of antimicrobials like metal ions (Zn^{2+} , Ag^+ , Cu^{2+} , Ti^{4+} , etc.) and reactive oxygen species are required for active packaging to demonstrate antimicrobial activity (Garcia et al., 2018; Jafarzadeh et al., 2020); nevertheless, the release of antimicrobials must be regulated, and nanoparticles migration is undesired owing to the unknown outcome on the human body (Llorens et al., 2012; Duncan, 2011).

Some of the nanomaterials that can be included into packaging are nanosilver and other metals plus their oxides, “nanocellulose”, “essential oil nano-emulsions” and “nanoclays” (Bumbud-sanpharoke & Ko, 2015; Eleftheriadou et al., 2017; Llorens et al., 2012; Mihindukulasuriya & Lim, 2014). Despite the fact that the number of articles on novel forms of nanocomposites is growing, the scientific literature is still predominated by nanosilver and nanoclay applications (Bumbudsanpharoke & Ko, 2015; Mihindukulasuriya & Lim, 2014).

Metal oxide nanoparticles that are also utilized in food packaging include Aluminum oxide, Silica, Titanium dioxide and Zinc oxide. These nanomaterials are commonly utilized as photocatalysts having

antimicrobial plus ethylene-scavenging properties, and they can as well improve the nanocomposites' "tensile strength", barrier properties to UV & gas (Bumbudsanpharoke & Ko, 2015; Llorens et al., 2012). These properties would be beneficial for packaging fresh foods that lose water readily and are susceptible to spoilage by microbes and enzymes (Llorens et al., 2012; Garcia et al., 2018). However, the various features of nanomaterials that make them superior to their bulk equivalents may also lead to a variety of toxicological properties, raising consumer concerns. Thus, to assure the safety of nanocomposite packaging, nanomaterials migration from packaging into foods must be identified (Störmer et al., 2017; Garcia et al., 2018).

This review article therefore focuses on metal oxides as nanomaterials for the purpose of food packaging. Although there are some reviews on uses of nanomaterials in food packaging, their scopes is different from the current review. The focus of previous studies was on the general perspective for nanomaterials in packaging (Bumbudsanpharoke et al., 2015; Mihin-dukulasuriya & Lim, 2014), market situation (Bumbudsanpharoke & Ko, 2015), and regulations (Bumbudsanpharoke & Ko, 2015; Wyser et al., 2016). To the best of our knowledge, there are lack of reviews focusing on nano-metal oxides in packaging. Therefore, the purpose of this article was to summarize the advantages of integrating nano-metal oxides into polymer-based food packaging, and to provide information on how such nanomaterials may migrate into packaged foods. Metal oxides like Aluminum oxide (AlO_x), Copper oxide (CuO), Magnesium oxide (MgO), Silicon dioxide (or Silica, SiO_2), Titanium dioxide (TiO_2) and Zinc oxide (ZnO) are discussed. The information presented here is expected to complement earlier publications and aid in drawing conclusions about the safety of

nanocomposites used in food packaging. The current search was conducted among PubMed, ScienceDirect, Embase and Scopus databases up to 2021 to retrieve the related articles. The following keywords were included "nanocomposite" OR "nanomaterial" OR "nano" OR "nano-food packaging" OR "nano-packaging" OR "Metal oxide" "AND "Packaging" OR "food packaging" OR "food AND "migration". The reference of retrieved articles was checked for additional articles. Full text articles in the English language were included. Review articles, correspondence, thesis, letter to the editor, and conference were excluded.

NANOSCALE METAL OXIDES IN FOOD PACKAGING

The most recently explored materials in food packaging are metal oxides (MOs) & mixed metal oxides (MMOs). The use of silver-NPs and/or nanoclay (particularly montmorillonite (MMT)) in Nanotechnology has received a lot of attention. MOs and MMOs, on the other hand, have structural and morphological features that make them promising nanoparticles in a typical composite profile. It's still challenging to synthesize suitable MMOs with the right physicochemical properties that account for the strong interaction with the polymer matrix (Atkins et al., 2006 Jafarzadeh et al., 2020).

When the metal has an oxidation number of I, II, or III, M^+ and M^{2+} ions (M_xO : $x = 1, 2, 3$, etc.) form oxides, and are categorized as: i) MO having a rock salt structure. ii) M_2O having an antifluorite or rutile structure. (iii) M_2O_3 oxides (Atkins et al., 2006; Jafarzadeh et al., 2020).

The metal oxides are formed by defects that differ from structure to structure. The size of the defects is related to the metal oxides type, as well as the changes in partial pressure of oxygen above a metal oxide, both of which cause the lattice parameter and



equilibrium composition to fluctuate continuously. Antimicrobial activity is found in a variety of d-block MOs, many of which are n-type semi-conducting MOs, such as CuO, ZnO, TiO₂, MgO, and others. Their activity may be due to minute differences in stoichiometry and O-atom defects (Jafarzadeh et al., 2020).

Metal oxide nanoparticles have been mixed into either biobased or petroleum-based polymers, to make nanocomposites with improved properties and inherent antimicrobial properties. On the other hand, nanoparticles migration from packaging films is a cause for concern due to their possible toxicity in the environment and the human body as well. In packaging, several nanoscale metal oxides and structural-based nanoparticles (such as clay) are utilized to enhance their antimicrobial activities, barrier plus mechanical properties and thermal stability. This section will cover recent breakthroughs in metal oxides-reinforced polymeric nanocomposites for food packaging, such as Aluminium (Al), Copper (Cu), Magnesium (Mg), Silicon (Si), Titanium (Ti) & Zinc (Zn) oxides (Jafarzadeh et al., 2020).

OXIDES USED IN NANOCOMPOSITES FOR FOOD PACKAGING

Aluminum Oxide (AlO_x)

For long, Aluminum has been utilized in metallized films to safeguard the packaged foods from ultraviolet irradiation and oxygen. These films, nonetheless, are opaque, thick, non-microwavable and recycling is arduous. Aluminum oxide-based coatings, which are light, transparent, and microwavable, are a viable alternative to such films (Struller et al., 2014). Physical vapour deposition or chemical vapour deposition processes are used to produce aluminum oxide thin films, which are utilized as microelectronics coatings (Cibert et al., 2008). Furthermore, atomic layer deposition

has been used to produce Al₂O₃ thin films, which have superb barrier properties and have been advocated for use in food packaging, specifically biodegradable packaging (Hirvikorpi et al., 2010).

Störmer et al., (2017) describe silicon oxide and aluminium oxide coatings as "nanocoatings" or "ultrathin films" because they may produce stable layers that are uniformly scattered and have a thickness of 10–100 nanometers. Some researchers suggest that such films should be classified as "nanostructures" rather than "nanomaterials" under European regulations because they are different from principal nanoparticles and form strong bonds with polymers (Bott et al., 2014; Vähä-Nissi et al., 2015).

Coating board paper with PLA-Al₂O₃ and PLA film double-coated with alginate-chitosan layer and Al₂O₃ layer significantly improved their aroma, oxygen, and water barrier properties (Hirvikorpi et al., 2011; Hirvikorpi et al., 2010). The water vapour transmission rate of PLA double-coated was 25 g/m²/day, which was significantly better than pure PLA (53 g/m²/day) and much nearer to the permissible value for dry food packaging (0–10 g/m²/day) (Hirvikorpi et al., 2011). It has also been reported that coating polyethylene terephthalate with aluminium oxide improves the composite films barrier properties, suggesting that it could be a viable substitute to the currently used metallized retortable packaging; results of biaxially oriented polypropylene coating, on the other hand, were inconsistent (Struller et al., 2014), indicating that further study into AlO_x nanocomposites is required (Garcia et al., 2018).

COPPER OXIDE-BASED NANO MATERIALS

The two primary categories of materials in food packaging are copper nanoparticles (CuNPs) and copper oxide

nanoparticles (CuONPs). CuONP, on the other hand, is one of the most investigated metal oxides in industrial food packaging, because of its antimicrobial activity, which inhibits viruses, fungi and bacterial growth (Kuswandi & Moradi, 2019). Because of their large surface area, CuONPs interact with the cell membrane. Sonochemical (Ávila-López et al., 2019), microwave (Karunakaran et al., 2013), autocombustion (Kamble & Mote, 2019), electrochemical (Jadhav et al., 2011), thermal decomposition (Ibrahim et al., 2018), and other methods can all be used to produce nanosized CuO.

In bioorganisms, copper is a key element for metabolism and transport of electron. Copper levels above a certain threshold can inhibit bacterial cell development. CuO's mechanisms for limiting cell growth can be split into 4 categories:

I. Enzymes inactivation II. Essential ions exchange III. Generation of H₂O₂ free radicals, attacking protein functional groups IV. Breaking the plasma membrane's integrity (Jafarzadeh et al., 2020).

Cu²⁺-incorporated poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHB-co-3HV) (Castro et al., 2018), agar (Roy & Rhim, 2019), cellulose (Muthulakshmi et al., 2019) and chitosan (Almasi et al., 2018), have all been investigated for antimicrobial activity in packaging systems. CuONPs have been shown to have antibacterial activity against Gram-positive (G⁺) & Gram-negative (G⁻) bacteria in practically all biopackaging systems. However, in some polymeric systems, such as CuO & Chitosan nanofibers, a synergistic effect is observed (Almasi et al., 2018).

It is well known that the antimicrobial activity of MOs such as CuO is governed by their structure, size, morphology, surface area and oxidation states variation. Furthermore, doping/ coupling CuO with other active materials (such as MOs and M)

may have an impact on the ultimate properties of packaging films. Cu/CuONPs, CuONPs/AgNPs, and other nanohybrid systems can provide improved optical, Oxygen/water barrier properties and antimicrobial, with biocidal activity against G⁺ve /-ve bacteria (Jafarzadeh et al., 2020).

Copper is attractive for active packaging because it shows antimicrobial properties that are broad-spectrum. Copper ions work by readily receiving and giving electrons, demonstrating a strong redox potential and the ability to destroy microbial cells components, killing them (Nan et al., 2008; Garcia et al., 2018). The incorporation of nano-CuO into LDPE produced an antibacterial nanocomposite, which was evaluated for cheese packaging, and resulted in 4.21 log CFU/g of coliforms reduction after 30 days of refrigerated storage (Beigmohammadi et al., 2016). Nano-CuO/ZnO was added to chitosan, which resulted in nanocomposite films with improved antimicrobial activity, decrease in moisture permeability, improved quality attributes of guava fruits, extending its shelf life by 1 week (Kalia et al., 2021). In addition, CuO incorporated into Chitosan killed more than 99% bacteria (Raghavendra et al., 2017). Nanocomposite was prepared by incorporating nano-CuO into Agar resulting in increased UV-light barrier property and potent antibacterial activity against *E. coli* and *L. monocytogenes* (Roy and Rhim 2019).

Magnesium Oxide (MgO)

Magnesium oxide (MgO) is a colorless naturally occurring, crystalline mineral having a high melting point and because of its large-scale production, it is used in a variety of industries. MgO is a material with a poor electrical conductivity and a high thermal conductivity. In recent years, research has focused on MgO's excellent antimicrobial activity and high stability when compared to organic

antimicrobial agents. Its application as a multifunctional solid material is due to this combination of properties. Numerous construction materials in food packaging has been replaced with MgONP-based packaging in the United States and Europe [Food additive approved by the European Union (E number 530)], because it is thermally stable, recyclable, gas impermeable and flexible, and possessing antimicrobial activity as well as Hydrothermal, laser ablation, microemulsion, microwave-assisted, sol-gel, ultrasound-assisted and wet chemical reaction processes can be used to synthesize MgONPs (Mirtalebi et al., 2019).

MgONPs are incorporated into biodegradable polymers including polylactic acid (PLA), Polycaprolactone (PCL), and polyhydroxybutyrate (PHB) to produce nanocomposite with enhanced characteristics and antimicrobial activities. Pure PLA degrades gradually in the environment to CO₂, CH₄ and H₂O. Industrial-scale nanocomposites with enhanced barrier, mechanical, thermal, optical and antibacterial/microbial properties can be formed by incorporating MgONPs into the aforementioned polymers. For instance, Swaroop and Shukla (2019) found that nanocomposite films of PLA/MgONP comprising 2 wt.% MgO enhanced plasticity and tensile strength by more than 146 % and 22 %, respectively. When compared with the sample containing 1wt.% MgONPs, the oxygen and water vapor barrier properties of this nanocomposite was also improved by about 65 % and 57 %, respectively. They also mentioned the film's antimicrobial properties, stating that after 24 hours of treatment in the presence of 1wt.% MgONPs, *E. coli* bacteria (about 44 %) were killed. Zhang et al., (2020) demonstrated that ternary nanocomposites prepared with the combination of Poly(butylene adipate-co-terephthalate) (PBAT) and MgO/Ag showed improved mechanical and barrier properties,

with composite films having 3 wt% NPs content exhibiting the optimal properties. In addition, the ternary composite film inhibition rates against *E. coli* and *S. aureus* exceed 90 %. Nanocomposites prepared by combining Starch-albumin with MgO increases film thickness, decreased water vapor permeability, increased antioxidant activity, antimicrobial properties (Hosseini et al., 2021). Silva et al., (2017) observed that packaging made of chitosan and nano-MgO exhibited remarkable improvement in thermal stability, UV shielding and moisture barrier properties. Wang et al., (2020a) reported improved thermal stability, better UV shielding performance, as well as water-insolubility compared to pure CMCS, exhibited excellent antimicrobial activity against *L. monocytogenes* and *S. baltica* by combining Chitosan-CMCS with nano-MgO.

Silica (SiO₂)

Silicon is a metalloid that is found in the forms of silica (SiO₂) and silicates and is the most plentiful solid element on Earth. Nonstick coatings for jars, bottles and bags are made with SiO₂ as the main raw ingredient in numerous food industries (Bumbudsanpharoke et al., 2015; Liu et al., 2018). SiO₂ is a promoter (a compound that boosts catalytic activity), like other MO. SiO₂-NPs' activities are influenced by factors such as high surface area, low thermal conductivity, average particle size, low level of toxicity, supreme insulation, biocompatibility and stability. Silica can be used in food packaging films as an antimicrobial agent or as a strengthening agent once it has been modified with organic matter. Examples of modified SiO₂ are Tetraethoxysilane, 3-isocyanatopropyltriethoxysilane, and aminopropyltriethoxysilane. When compared to pure polymers, polymer/SiO₂ nanocomposites have been demonstrated to have improved water and gas barrier properties, in addition

to better mechanical strength. Incorporating nano-SiO₂ into plastics increased the mechanical strength and thermal stability of the plastics (Hasan et al., 2006). In comparison with the pure polymer, EVOH-SiO₂ nanocomposites containing 5 wt.% nano-SiO₂ had significantly improved water plus gas barrier properties in addition to improved mechanical strength (Liu et al., 2010). Modifying nano-SiO₂ (0.09 wt.%) with ethylene/vinyl acetate (EVA) and blending with polypropylene, produced nanocomposites with an increase in tensile strength and a decrease in gas permeability, as well as adsorbed a smaller amount of ink solvent as compared to the pure polymer, which could be beneficial for laminated food packaging because for printing, the layer of polypropylene is used (Li et al., 2016).

Another way to make SiO₂-based nanocomposites is to coat the polymers. The sol-gel procedure was used to make a polylactic acid-based coating with nano-SiO₂ utilizing tetraethoxysilane as a precursor and 3-isocyanato-propyl-triethoxysilane as a coupling agent, respectively, the coating can be applied to PLA films to produce biodegradable packaging with gas barrier properties that are up to 70% better than pure PLA films while yet being transparent (Bang & Kim, 2012). Zhao et al., (2020) revealed that packaging made of nano-SiO_x-coated PLA decreased oxygen and water vapor transmission rates by 48.45% and 28.53%, respectively, approximately 100 nm SiO_x layer could prevent diethylene glycol dibenzoate from migrating and diffusing into food simulants.

There are only a few investigations on SiO₂ nanocomposite applications. Nonetheless, Luo et al., (2015) reported on the use of LDPE-SiO₂ packaging for chilled white shrimp, which increased shelf life and kept the shrimp fresh for eight days, implying that this nanomaterial could be combined with polymers to manufacture “active packaging”

with antimicrobial as well as enzyme-inhibitory properties.

According to Donglu et al., (2016), combining SiO₂/TiO₂/Ag nanoparticles with LDPE produced a multifunctional nanocomposite capable of regulating the levels carbon dioxide and oxygen, scavenging ethylene and inhibiting microbial growth, thereby prolonging the packaged mushroom's shelf life, implying that using a combination of nanomaterials to develop nanopackaging can be a good strategy (Garcia et al., 2018).

SiO₂-nanoparticles are utilized separately as a reinforcing agent in the most widely published studies in food packaging, and they have antimicrobial properties when combined with organic and inorganic materials. For instance, ethylene scavengers, carbon dioxide and oxygen regulators plus microbial growth inhibitors have been described using a mixture of SiO₂/Ag, SiO₂/TiO₂/Ag, SiO₂-Al₂O₃, CoFe₂O₄/SiO₂/Ag, and silica-carbon-silver (SiO₂/C/Ag) nanosystems mixed into polymer matrices (Jafarzadeh et al., 2020). Thus combining nanomaterials with SiO₂ can be an effective technique for developing nanopackaging. The oxygen/water barrier effect of SiO₂ nanoparticles incorporated into polymer matrix is depicted in Figure 1.

Regarding the oxygen barrier performance, the addition of SiO₂-NPs reduces the oxygen permeability for two main reasons: i) according to the “tortuosity path” concept, the impermeable filler represents a physical barrier to the permeant transfer across the material's thickness, thus reducing the diffusion coefficient (Silvestre et al., 2011); ii) according to the “interface effect” concept, fillers with high surface area promote local changes in the polymer matrix properties at the interfacial regions (Duncan, 2011). In particular, the higher the extent of the cooperative forces at the filler/polymer

interface, the lower the segmental mobility of the molecules (Rovera et al., 2020).

Titanium Dioxide (TiO₂)

Owing to its bright white coloration, titanium dioxide (TiO₂) is extensively used as a pigment in a variety of products, such as Foods, Nutraceuticals, Supplements, as well as Cosmetics (Weir et al., 2012; Guseva et al., 2020). TiO₂ nanoparticles are promising candidates for reinforcing polymer matrixes in food packaging applications because of their low cost, nontoxicity, and biocompatibility (Lotfi et al., 2019; Mohammadi et al., 2015; Segura et al., 2018; Venkatesan & Rajeswari, 2017; Xie & Hung, 2018; Cao et al., 2020). The synthesis of TiO₂ nanoparticles can be done in three different forms: anatase, brookite or rutile, each with distinct characteristics and reactivities based on their band gap (Llorens et al., 2012). To synthesize these nanoparticles, the sol–gel methodology is commonly utilized (Ibrahim & Sreekantan, 2011; Bodaghi et al., 2013). Carré et al., (2014) and Dalrymple et al., (2010) revealed that TiO₂ nanoparticles had photocatalytic antibacterial characteristics, generating reactive oxygen species (ROS) and killing bacteria by peroxidizing lipids in the cell membrane and oxidizing proteins and DNA after being exposed to UV light. The anatase form, in particular, has low toxicity, high photocatalytic efficiency and can become active after exposure to near-UV light; it is inexpensive as well (Carré et al., 2014). The highest antimicrobial effect was achieved when exposed to UV-A light (Chorianopoulos et al., 2011).

Because of its photocatalytic activity, TiO₂ could be used to disinfect food contact surfaces and decontaminate water (Llorens et al., 2012). Furthermore, this property could be beneficial in the development of antimicrobial active packaging (Huang et al., 2018). Various polymeric matrices, including

conventional plastics and biopolymers, can be used to produce such packaging. Xie and Hung (2018) prepared a UV-A activated TiO₂ embedded biodegradable polymer films. They found that 2 hours of UV-A light illumination at a light intensity of 1.30 ± 0.15 mW/cm² reduced bactericidal activity by 1.69 log CFU/mL. Nanocomposites produced from Guar gum-TiO₂ showed UV-light and oxygen barrier properties, antimicrobial activity against *L. monocytogenes*, *Salmonella enterica* sv, *typhimurium* (Arfat et al., 2017). Salarbashi et al., (2018a) also confirmed that soybean polysaccharides-TiO₂ film displayed antimicrobial activity against *S. aureus*, *Pseudomonas aeruginosa*, *E. coli*. Wheat gluten/nanocellulose-TiO₂ film had antimicrobial activity against *Saccharomyces cerevisiae*, *E. coli*, and *S. aureus* (El-Wakil et al., 2015). Pectin-TiO₂ nanocomposites exhibited antimicrobial activity against *E. coli* (Nešić et al., 2018). In addition, soybean polysaccharides incorporated with TiO₂ nanoparticles showed decrease in moisture content and water-vapor permeability, increased tensile strength and antimicrobial activity (Salarbashi et al., 2018b). TiO₂ nanoparticles incorporated into edible films of whey protein had antimicrobial effects (AlizadehSani et al., 2017), and good moisture barrier and mechanical properties (Zhou et al., 2009). 2–5 wt% TiO₂ nanoparticles mixed with ethylene-vinyl alcohol (EVOH) produced self-sterilizing nanocomposites that were effective against both Gram-positive as well as Gram-negative bacteria (Cerrada et al., 2008).

Some of the developed TiO₂ nanocomposites have been tested as food packaging. In in vitro trials and as real packaging for chopped lettuce, *E. coli* growth was inhibited using oriented–polypropylene (PP) films coated with TiO₂ nanoparticles, following exposure to black light (Chawengkijwanich & Hayata, 2008). LDPE

films containing TiO₂ nanoparticles inactivated yeasts and *Pseudomonas* spp., in vitro, as well as when used as packaging for pears, and the impact was increased by UV-A light exposure (Bodaghi et al., 2013). A similar result was demonstrated for chitosan-TiO₂ films, which exhibited antimicrobial activity against *Escherichia coli*, *Staphylococcus aureus*, *Candida albicans*, and *Aspergillus niger* with 100% sterilization in 12 h, it also possessed UV-blocking properties, decreasing light transmittance in visible light region of the composite film and extended the shelf life of red grapes (Zhang et al., 2017). Nanocomposites with improved oxygen barrier properties were produced when TiO₂ (4 wt%) was integrated into LDPE films, and its usage as packaging reduced the rancidity incidence in almonds by 78 percent in comparison to the control (Nasiri et al., 2012). According to Li et al., (2017a) PLA-TiO₂ nanocomposites showed antimicrobial activity, cottage cheese freshness preservation, and a 25-day extension of the cheese shelf life. Other foods like short-ripened cheese, strawberries, shrimp and mangoes have been demonstrated to minimize deterioration and retain their quality using similar nanocomposite films (Gumiero et al., 2013; Li et al., 2017; Luo et al., 2015; Chi et al., 2019). Furthermore, TiO₂'s photocatalytic activity exhibits ethylene-removing properties (Bodaghi et al., 2015; Wang et al., 2010; Bohmer-Maas et al., 2020), Li et al., (2017b), which could be effective in delaying fruit ripening. Incorporating TiO₂ into polymers also produces nanocomposites that blocks oxygen and UV-light (Guo et al., 2014; Krehula et al., 2017; Achachlouei and Zahedi, 2018; Ahmadi et al., 2019; Goudarzi et al., 2017; Yousefi et al., 2019). UV light blocking is desirable because it protects packaged foods while also delaying the ageing and degradation of nanocomposites. Díaz-Visurraga et al., (2010) combined TiO₂

nanotubes (0.05 % to 0.1 % w/v) into chitosan to develop UV-blocking nanocomposite films that were effective against *Salmonella enterica*, *Escherichia coli*. and *Staphylococcus aureus*.

Active packaging with enhanced properties can be developed, when TiO₂ is used in a synergistic way, for instance, HDPE-TiO₂-marigold extract nanocomposite was reported to maintain soybean oil stability via the antioxidant effect of the carotenoid component; as carotenoids degrade easily when exposed to light, their effectiveness was preserved by the UV-blocking effect of TiO₂ (Colín-Chávez et al., 2014). A similar finding was reported by Vejdani et al., (2017) for gelatin-agar-TiO₂ films, which delayed fish oil oxidation. According to a study, TiO₂-based nanocomposites could prevent plastic additives such as ethylene glycol from migrating into food from packaging, besides improved mechanical as well as antibacterial properties (Farhoodi et al., 2017).

Zinc Oxide (ZnO)

ZnO is a white powder that is extensively used in sunscreens as well as in the Food industry (Espitia et al., 2012). Due to their inexpensive cost, low toxicity, in addition to UV-blocking properties, ZnO nanoparticles have an advantage over Ag nanoparticles and they can also be utilized as an antimicrobial material (Chaudhry et al., 2008; Llorens et al., 2012). Wurtzite, zinc blende, and rock salt are the three crystal structures of ZnO. In comparison to the other structures, the wurtzite structure is more thermodynamically stable. Various physicochemical and biological methods are used to synthesize ZnONPs (Kumar et al., 2019). Mechanochemical processing (MCP) and physical vapour synthesis (PVS) are used in industrial production of ZnONPs, while wet chemistry methods such as coprecipitation (Ubani & Ibrahim, 2019),



microwave (Salah et al., 2019), thermal decomposition (Alp et al., 2018), sol-gel (Delice et al., 2019; Khan et al., 2016), and hydrothermal (Kumaresan et al., 2017) methods can also be used. Nano-ZnO, like TiO₂, has an antimicrobial effect due to a photocatalytic reaction, but unlike TiO₂, it also produces Zn²⁺ ions, which damage and kill bacterial cells (Bumbudsanpharoke & Ko, 2015), and UV exposure does not trigger the antimicrobial activity (Espitia et al., 2012).

ZnO, like TiO₂, has been integrated into or coated onto a variety of synthetic and biological polymers to produce antimicrobial nanocomposites. The growth of both *Staphylococcus aureus* and *Escherichia coli* were inhibited using polyvinyl chloride (PVC) films coated with ZnO; however they were more effective against *S. aureus* (Li et al., 2009). Furthermore, nanocomposites containing nano-ZnO have better mechanical and oxygen barrier properties (Lepot et al., 2011). With the incorporation of nano-ZnO (0.5 wt%) into HDPE and modification by the coupling agent KH550, nanocomposite films with increased mechanical, barrier and UV-blocking properties, in addition to antimicrobial activity, especially against *Staphylococcus aureus* was produced (Li & Li, 2010). According to Ghozali et al., (2020) LDPE-ZnO nanocomposites was effective by showing a widest inhibition zone against *E. coli*. In addition, a study (Applerot et al., 2009) reported the production of antimicrobial glass via coating it with ZnO nanoparticles, which resulted in 89 % decrease in *E. coli* counts.

Biodegradable packaging development is appealing since it would result in generation of less waste as well as environmental pollution while lowering reliance on oil; nevertheless, bioplastics have high water permeability, which is a disadvantage. As a result, nanocomposites with enhanced properties have been produced

with the incorporation of nano-ZnO into bioplastics. By incorporation of ZnO up to 3% into the polymer, multifunctional PLA-ZnO nanocomposites were produced, yielding a biodegradable packaging film with antibacterial effects against both G⁺/- strains and improved water barrier properties, UV light blocking ability (Pantani et al., 2013). Coating of ZnO (0.5 wt%) to PLA coating layer was effective in inactivating *E. coli* and *S. aureus*, with *E. coli* been more susceptible, showing 3.14 log reduction (Zhang et al., 2017). The effectiveness of PLA/ZnO nanocomposites showed increase in the hydrophobicity, good UV-light barrier properties and excellent antibacterial activity against *S. aureus* and *E. coli* (Kim et al., 2019). Poly (butylene adipate-co-terephthalate) (PBAT) combined with nano-ZnO yielded another biodegradable nanocomposites with improved mechanical, thermal as well as antimicrobial properties (Venkatesan & Rajeswari, 2017). Nanocomposites prepared by combining poly(ε-caprolactone) (PCL) with nano-ZnO, (PCL/ZnO5%) had optimal antibacterial activity (Pina et al., 2020). Qiu et al., (2019) in their work, documented that nanocomposites developed by incorporating nano-ZnO into chitosan significantly improve mechanical and antibacterial properties. Nanocomposites produced with the combination of nano-ZnO with PBAT, showed high antibacterial activity against *S. aureus*, *P. aeruginosa* and *B. subtilis* strains (Seray et al., 2020). Starch-kefir combined with ZnO yielded nanocomposites that exhibited increased Tensile strength; decreased in WVP by about 16%, decrease in moisture absorption of the films (Shahabi-Ghahfarrokhi and Babaei-Ghazvini 2019). Nanocomposites of Zein-ZnO increased the hydrophobicity and reduced up to 3 times, the amount of water uptake of the composite films compared to pure zein, UV-blocking

properties, and antibacterial activity (Schmitz et al., 2020).

Food packaging tests have been carried out on some of the produced ZnO nanocomposites. Emamifar et al., (2011) observed that LDPE-ZnO-Ag nanocomposites were successful at inhibiting *Lactobacillus plantarum* growth in orange juice, while Li et al., (2011) discovered that nano-ZnO-coated PVC packaging reduced decay and prevented bacteria and fungi in cut apples. ZnO nanoparticles incorporated into a poly-lactic acid (PLA) matrix resulted in nanocomposite films that inhibits microbial growth and preserved the organoleptic qualities of fresh-cut apple at 4° C, extending the shelf life by 2 weeks (Li et al., 2017). El-Sayed et al., (2020) demonstrated that coating Ras cheese with chitosan/guar gum/roselle extract-ZnO protects their surface for around three months from yeasts, molds and other bacteria growth as compared to uncoated cheese. Heydari-Majd et al., (2019) in their work, documented the effectiveness of nanocomposites of PLA-ZnO-(*Zataria multiflora* Boiss. essential oil and *Menthe piperita* essential oil) for enhancing antibacterial and antioxidant activities and extends shelf life of fillets by 8 days. Chitosan– cellulose acetate phthalate film loaded with 5% (w/w) nano-ZnO exhibited optimal tensile strength, significantly lower the water vapour and oxygen transmission rates, excellent UV shielding ability, antimicrobial properties and extended the shelf life of black grape fruits up to 9 days (Indumathi et al., 2019). Furthermore, *Salmonella* in egg whites was inactivated after 3 weeks of storage in glass jars coated with a mixture of PLA, allyl isothiocyanate, ZnO nanoparticles and nisin (Jin & Gurtler, 2011). In addition, in a study, ZnO nanocomposites were found to be effective against yeasts and molds that cause bread spoilage (Noshirvani et al., 2017). Negahdari et al., (2021) revealed the

effectiveness of PLA-(added *Origanum majorana* essential Oil)-ZnO nanocomposites, (1.5% *O. majorana* EO and 1% ZnO NPs) extended meat shelf life and lead to the most favorable sensory properties of ground meat. Chitosan-ZnO nanocomposites exhibited excellent antimicrobial activity in raw meat by complete inhibition of microbial growth on the sixth day of storage at 4 °C (Rahman et al., 2017). Coating of pomegranate with the combination of Nano-ZnO + CMC extended its storage life by decreasing total yeast + mold during 12 days of storage while total mesophilic bacteria decreased during 6 days of storage (Saba and Amini 2017). Polyurethane/chitosan with nano-ZnO composite films showed enhanced antibacterial properties, barrier properties, and extended the shelf life of carrot pieces up to 9 days (Sarojini et al., 2019). The chemical composition, surface functional chemical groups, crystalline structure, specific surface area and morphology of ZnONPs can all be used to their advantage. ZnONPs possess a broad spectrum of antimicrobial activity and a low proclivity to induce resistance (Reyes-Torres et al., 2019). Although the specific mechanism of most NP reactions/interactions is still not known, the antimicrobial activity of nanoparticles can be predicted based on electrostatic interaction, the release of antimicrobial ions, and reactive oxygen species (ROS) formation (Figure 2).

MECHANICAL PROPERTIES

Deformability, TS, elongation at break (EAB), and elastic modulus (EM) are important mechanical characteristics of food packaging since these materials must maintain their integrity during storage, transport, and handling. The maximum stress that the film can withstand while being stretched or pulled before failing or breaking is known as TS and EM, which indicates the flexibility and intrinsic stiffness of the films,



respectively (Jafarzadeh, 2017). The mechanical properties of biopolymer films depend both on their composition and on the environmental conditions. For example, the addition of plasticizers causes a higher mobility of polymer chains, which leads to expanded elongation and diminished TS of the plasticized films. Embedding of different additives, such as cross-linking materials or lipids, can improve film strength and extensibility (Vieira *et al.*, 2011). Moreover, humidity and moisture of the environment influences the mechanical properties of polymer/biopolymer films. For example, hydrophilic films absorb humidity more promptly at higher moisture levels, consequently enhancing the plasticizing impact of water, which subsequently decreases the TS and increases the extendibility of the films. In addition, the contact between polymer/biopolymer packaging materials and packaged product can likewise influence the functioning of packaging films.

In recent years, the incorporation of NPs has turned into a well-known way to improve the properties of different films, since the utilization of NPs typically gives them improved mechanical properties (Cho & Rhee, 2002). This result is due to the increased surface interaction between the matrix and NPs with a high surface area, as well as the hydrogen bond formation between them. EAB has a reverse relation to TS in most cases, and YM is directly related to TS. Furthermore, the mechanical properties of films are closely associated with the density and distribution of the intra- and intermolecular interactions between polymer chains in the film matrix.

OPTICAL PROPERTIES

The optical properties (color, transparency, and UV transmission/absorbance) of packaging materials are extremely significant since

maintaining food quality depends greatly on their protection against UV and light. Also, to prevent lipid oxidation, retain food nutrients, and prevent the discoloration of food, the packaging must protect the content against UV light. Furthermore, the consumers' acceptance of food packaging is influenced by the color of the packaging, and it is one of the most important properties of films which could affect their application (Jafarzadeh *et al.*, 2017; Jafarzadeh and Jafari, 2020).

The apparent color properties of neat semolina films showed that they are totally transparent and colorless; however, the addition of ZnO-nr significantly decreased the L* and a* values, meaning an increased number of NPs, resulting in reduced transmissions from films (Jafarzadeh *et al.*, 2017; Jafarzadeh and Jafari, 2020). Arfat *et al.*, (2017) reported that the addition of Ag-Cu NPs into fish gelatin films significantly influenced their color by reducing the lightness (L*), while the a* and b* values significantly increased. These results were consistent with those for agar nanocomposite films incorporated with nano-silver, as reported by Rhim *et al.*, (2013). Moreover, the control gelatin film remained highly transparent while the addition of nano-clay was found to decrease the transparency of films. It has been reported that nanocomposite films with nano-silver had a brown surface color with reduced transparency (Kanmani and Rhim 2014b). Zolfi *et al.*, (2014) also reported that the transparency of nanocomposite kefir-WPI films significantly decreased with the addition of TiO₂, and this decrease was dependent on the level of NPs.

The light transmission and absorption spectra of nanocomposite films is determined by UV-vis spectroscopy. Three zones have been identified for the UV region, namely, UVA (320–400 nm), UVB (280–320 nm), and UVC (180–280 nm). The nanocomposite films incorporating ZnOnr showed a clear

absorption peak, but with the addition of ZnO-nr displayed greater absorption peaks at UVA (Jafarzadeh *et al.*, 2017). Also, Kanmani and Rhim (2014a) and Rouhi *et al.*, (2013) reported a lower UV transmission by the incorporation of Ag-clay and ZnO-nr into gelatin films.

THERMAL PROPERTIES

It has been demonstrated that thermal analysis methods can be used to define suitable processing conditions, applications, and polymer chain structures. The thermal profiles of polymers can be investigated by thermogravimetric analysis (TGA), derivative thermogravimetry (DTG), differential thermal analysis (DTA), and differential scanning calorimetry (DSC) (Gabbott, 2008). TGA describes the relation between the weight change and temperature. The amount of mass decreased versus temperature, or time, in a controlled atmosphere can provide information about thermal and oxidative stabilities of materials. Based on TGA thermogram, the composition of materials can also be identified. Using TGA, the mass loss/mass gain due to decomposition, oxidation, or loss of volatiles can be examined. It is a useful technique for measuring the polymeric materials like thermoplastics, films, fibers, etc. In industries, TGA measurements can be used to select materials for end-use applications, either by product performance or/and product quality.

DSC is a thermal technique to obtain a wealth of information about materials, including polymers, and organic-inorganic composites/hybrids. The energy changes during continuous heating and cooling can be obtained from DSC measurements. This enables the scientists to find the transition temperatures, like glass transition temperature (T_g), melting temperature (T_m), and crystallization temperature (T_c). In addition, this quantitative thermal analyzer can provide

detailed information regarding the degree of crystallinity. Melting is an endothermic process, that is, the sample absorbs energy. Integrating the peak area gives the heat of fusion (ΔH_f). Crystallization of the polymer, which is a process of partial alignment of molecular chains, occurs upon cooling, mechanical stretching, or solvent evaporation. Crystallization can affect optomechanical, thermomechanical, and chemical properties of the polymers (Billmeyer, 2007).

THE EFFECT OF OXIDE-BASED NANOPARTICLES ON DIFFERENT PROPERTIES OF FOOD PACKAGING MATERIALS

In order to protect the foods from moisture, oxygen, pathogenic microorganisms, dust, light, and a variety of other harmful or dangerous materials, the packaging must be inert, cheap to produce, light weight, easy to dispose of or reuse, able to withstand extreme conditions all through processing or filling, impervious to a variety of environmental storage and shipping conditions as well as resistant to physical and mechanical abuse. A substantial number of commercial food packaging materials are made of non-biodegradable materials, which pollute the environment and utilize petroleum derivatives in their manufacture. Bio-nanocomposites for food packaging are not only safer and extend the shelf life of food, but they are also more environmentally friendly because they eliminate the need for plastics as packaging materials. However, present biodegradable films have weak mechanical and barrier properties, which must be enhanced before they can completely replace traditional packaging and, as a result, contribute to global waste reduction. As a result, the primary objective for nanopackaging is to extend the shelf life of food packaging by improving barrier properties to reduce transfer of gas and moisture, UV light



exposure, and enhance the mechanical, thermal, and antimicrobial properties (Jafarzadeh *et al.*, 2020). Figure 3 depicts the most important requirements for food packaging materials.

OXIDE NANOPARTICLE MIGRATION FROM PACKAGING

When nanomaterials are utilized in foods as additives, they are characterized as free; when they are incorporated into food contact materials, they are classified as embedded. This distinction is important when nanomaterials' oral exposure and toxicological risk is been estimated, because in the latter instance, nanomaterials must migrate to the packaging's surface, and then interact with the food (EASAC & JRC, 2011; Störmer *et al.*, 2017). Diffusion, dissolution, and abrasion of the packaging surface can promote nanomaterial migration into food, which is a matter for concern because it may have severe health repercussions (Wyser *et al.*, 2016). Nanoparticles have a large surface area-to-volume ratio due to their small size, and therefore may have different physicochemical properties compared to bulk-sized materials (Chaudhry *et al.*, 2008). Moreover, their charge, composition, and surface morphology, as well as the chemistry of the nanomaterial itself, may affect their toxicology (Wyser *et al.*, 2016). As a result, the toxicological characteristics of nanomaterials must be determined, which necessitates physicochemical characterization in addition to *in vitro* plus *in vivo* experiments (Oberdörster *et al.*, 2005). It's also crucial to figure out whether and how much nanoparticles in packaging migrate when they come into touch with food, as well as the impact of consuming them on the gastrointestinal system and other organs. Nonetheless, the existing data is inadequate, and further investigation is urgently required in this area.

Furthermore, nanomaterials release from packaging is a source of worry because these materials may contaminate the environment and be consumed by humans in turn. The migration of nanomaterial into non-food matrices is complicated, with some materials forming aggregates and others dissolving into ions or binding with organic matter. However, little research has been done on the environmental destiny of nanomaterials from packaging (Karimi *et al.*, 2018). A study (Part *et al.*, 2018) examined the likely destiny of nanomaterials (mostly silver & titanium dioxide) in garbage, indicating that while waste degrading mechanisms are unaffected, microbial communities may be altered. However, there is a significant knowledge deficit in this area.

The majority of research on nanomaterial migration from food packaging to foods has focused on silver nanoparticles (Bumbudsanpharoke & Ko, 2015; Störmer *et al.*, 2017), with only a few investigations available on oxide nanoparticles migration. The “European Commission (EC)” has enacted rules that describe the criteria under which migration tests have to be carried out (EC, 2007, 2011b). Depending on the type of food product in contact with the packaging, food simulants such as water, ethanol (10%–50% v/v), acetic acid (3% v/v), and vegetable oil can be used. As a result, such simulants have been used in most migration studies, and research on nanoparticles migration into real foods is scarce.

Even though the amounts of nanomaterial migrated have been reported to be small (Bumbudsanpharoke and Ko, 2015; Echegoyen and Nern, 2013; Farhoodi *et al.*, 2014), the migration rate of “nanosilver” and “nanoclay” components (Al and Si) increases with increasing temperature and time.

Migration studies dealing with oxide nanoparticles have been documented by some researchers. Lin *et al.*, (2014) investigated the influence of 30 and 100 nm

particle size on Ti migration into food simulants (acetic acid and ethanol), from LDPE-TiO₂ nanocomposites. As a result, they noticed that raising the exposure temperature caused the nanomaterial to migrate from the packaging, into the acetic acid in particular; after exposure of 8 hours to 3 percent acetic acid at 100 °C, the maximum amount of migrated Ti was 12.1 g/ kg. In one more investigation (Lian *et al.*, 2016), high hydrostatic pressure was used to compact and stabilize polyvinyl alcohol-chitosan-TiO₂ nanocomposites; migration tests showed that only a small amount of Ti ($3.87 \times 10^{-3}\%$ for film treated at 200 MPa) migrated in olive oil after eleven hours, but none in acetic acid, water or ethanol. The toxicity of nano-TiO₂ has been found to be minimal; to demonstrate tissue damage and bioaccumulation in mice, a substantial dose of 5 g/kg administered orally at once was required (Wang *et al.*, 2007).

Alternate to petroleum-based plastics, bioplastics have been promoted as a better-for-the-environment; nevertheless, they have drawbacks like poor gas and water barrier properties. It is therefore advantageous to add nanoparticles to bioplastics to enhance mechanical as well as barrier properties; however nanomaterial migration into packaged foods is still a concern. The quantity of Ti migrated from PLA-based nanocomposites was examined (Li *et al.*, 2018; Li *et al.*, 2017), suggesting that the quantity of nanomaterial migrated was low and within the safe range, as in synthetic composites. Furthermore, Li *et al.*, (2018) compared the amount of Titanium that migrated from Titanium dioxide into a liquid simulant as well as the real product (cheese), and found that the amount of Titanium migrated to the food was significantly lower than that in the simulant, implying that food simulants migration testing may overestimate the real exposure, especially when dealing with solid foods.

The migration of silver and zinc from LDPE-Ag-ZnO nanocomposites into orange juice was investigated by Emamifar *et al.*, (2011), after 112 days of refrigerated storage, they found that both migrated in low amounts of 0.15 and 0.68 g/L, respectively, remaining within the range regarded safe for consumers. Panea *et al.*, (2014) used an aqueous simulant to test the migration of silver and zinc from an LDPE-Ag-ZnO nanocomposite for packaging chicken for 10 days; only a minuscule quantity of silver (0.01 mg/kg) as well as zinc (2.44 mg/kg) migrated, both of which were below the European legislation established limit for zinc (5 mg/kg). It is worth noting that ZnO is GRAS for food applications and has a low toxicity in bulk form (Espitia *et al.*, 2012). Furthermore, the “European Food Safety Authority (EFSA)” stated that ZnO nanoparticles don’t migrate from polyolefins and unplasticized polymers; consequently, the release of soluble ionic Zn is the primary concern when evaluating the safety of nano-ZnO in food contact materials (EC, 2016).

According to the European Food Safety Authority (EC, 2016), when incorporated into polymers, silanated synthetic amorphous SiO₂ nanoparticles don’t migrate. Similarly, a research found that only (0.23 mg/kg) of CuO migrated from an LDPE-based nanocomposite (Beigmohammadi *et al.*, 2016), which is also lower than the “European legislation’s” limit for Cu release from plastics (5 mg/kg), though it is worth noting that the nanoform hasn’t been ratified (EC, 2011b). At 0.5 mg/dm² concentrations, equivalent to 0.9 mg/dm² Al₂O₃, aluminium has been observed to migrate from an Al₂O₃-coated polymer to acetic acid, implying that to prevent migration, an extra polymer layer on top of such coatings may be required (Vähä-Nissi *et al.*, 2015). Only nanoparticles with diameters less than 3.5 nm can migrate, according to a migration model based on TiN-LDPE



nanocomposites, whereas most nanoparticles in the form of composites clumps with sizes greater than 100 nm in practice; as a result, nanoparticles integrated into plastics are immobilized, and there's virtually no migration risk into foods (Bott *et al.*, 2014; Störmer *et al.*, 2017).

To sum up, nanoparticles integrated into polymers tend to agglomerate and remain firmly ingrained in the polymeric matrix, and hence don't migrate, according to studies on nanoparticle migration. This seems to be especially true for non-ionizing materials such as Ti. Materials such as Zn, conversely, can still migrate as soluble ions, and while ion migration is essential for antibacterial actions, the migrated levels from active packaging must be kept below the safe limits. When nanoparticles immobilized on the surface of packaging come into contact with the microorganisms, antimicrobial effects can be produced as well. However, as long as the polymer is intact and the nanoparticles are adequately incorporated, there appears to be little chance of nanoparticle migration.

CONCLUSION AND FURTHER REMARKS

Nanocomposites made of metal oxides are shown to have improved mechanical, barrier, as well as antimicrobial properties. TiO₂ and ZnO in particular have been demonstrated to be effective against a variety of bacterial strains and, due to the former's non-ionization and latter's lower toxicity, could be used instead of Ag nanoparticles. The incorporation of metal oxide nanoparticles into biodegradable polymers allows for the production of nanocomposites with improved mechanical strength, water and oxygen barrier properties, and other additional functional properties which include antimicrobial and antioxidants activities and light-blocking properties,

making them viable petroleum-based plastics replacements.

According to migration studies, only a small quantity of nanomaterial migrates from packaging into food simulants or foods, implying that consumer exposure to these nanomaterials and the health concerns associated with them are low. Furthermore, if nanoparticles are merely utilized to enhance "mechanical and barrier properties", the migration risk can further be reduced by incorporating a functional barrier between the food and the nanocomposite. Nonetheless, a dearth of human research, and a thorough characterization of the nanomaterials properties and toxicology raises doubt, and questions regarding nanotechnology's application persist.

Even though they have exhibited beneficial properties, most oxide-based nanocomposites are still in the development stage. One of the challenges that need to be addressed right now is the lack of validated methods for determining nanomaterial migration from packaging. Because regulators are taking a careful approach, this signifies that nanoparticles migration values should be below the detection boundaries in practice. Nonetheless, safety concerns will persist till satisfactory answers are found to these questions: "how much nanomaterial migrates into real foods from packaging"? "In what form is it ions or nanoparticles"? As well as "how toxic it is" thus stopping the mass commercialization of a technology that has proven enormous potential in the laboratory. As a result, academics, industry, and governments are being urged to prioritize this effort.

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Table 1. The properties and activities of various SiO₂/polymer nanocomposites

Nano-material	Method of preparation	Polymer Matrix	Properties/activity	Ref.
SiO _x	Plasma enhanced chemical vapor deposition (PECVD)	PLA (SiO _x /PLA-coated films).	Decreased oxygen and water vapor transmission rates by 48.45% and 28.53%, respectively, approximately 100 nm SiO _x layer could prevent diethylene glycol dibenzoate from migrating and diffusing into food simulants.	Zhao <i>et al.</i> , (2020)
SiO ₂	Casting method	Whey protein isolate/pullulan	Tensile strength increased, improved barrier properties of the films, water vapor permeability decreased.	Hassannia-Kolae <i>et al.</i> , (2016)
SiO ₂	Extruded film	Polypropylene (surface treated with ethylene vinyl acetate)	The permeability of O ₂ as well as water vapour was decreased by 28 percent and 23.8 percent, respectively. Tensile strength increased by up to 30%.	Li <i>et al.</i> , (2016)
SiO ₂	Extruded film	LDPE (titanate crosslinking)	Improved gas barrier properties, resulting in 33% increase in packaged shrimp shelf life.	Luo, Xu, <i>et al.</i> , (2015)
SiO ₂	Cast film	PLA	The permeability of O ₂ and water vapour was lowered by 50%.	Bang & Kim, (2012)
SiO ₂	Blown film	EVOH	Water permeability coefficient decreased by 54%; mechanical strength increased by 50% and heat resistance increased.	Liu <i>et al.</i> , (2010)
Ag + TiO ₂ + SiO ₂	Blown film	LDPE	The packing kept the mushrooms' nutrient content and quality for 2 weeks by lowering the respiration rate and scavenging ethylene.	Donglu <i>et al.</i> , (2016)

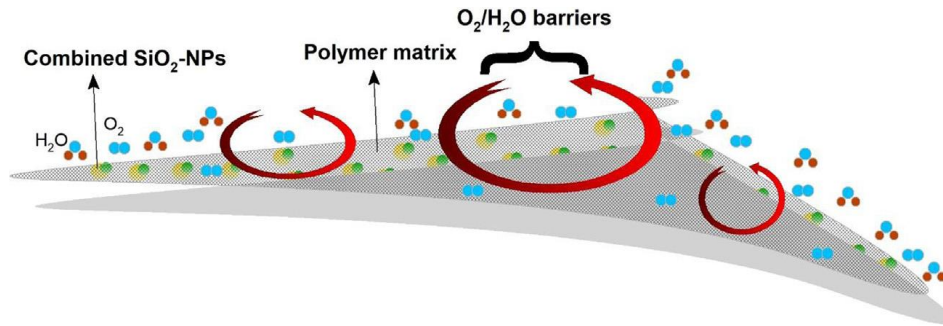


Figure 1. SiO₂ nanoparticles incorporated into polymer matrix provide an O₂/H₂O barrier effect. Adopted from Jafarzadeh *et al.*, (2020)

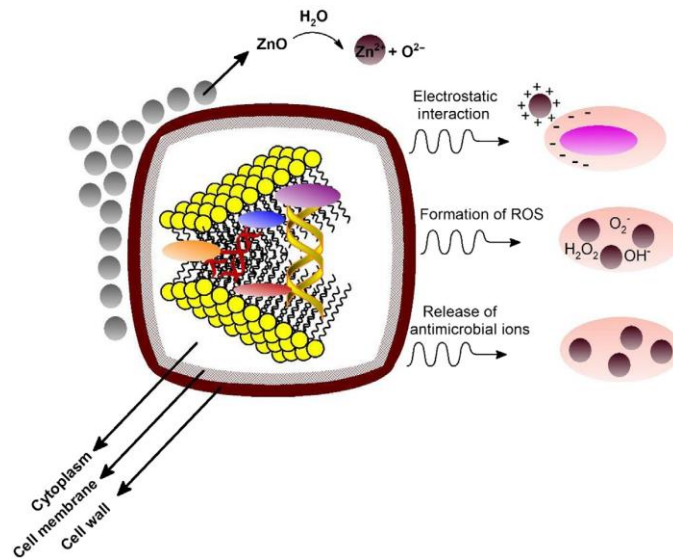


Figure 2. Mechanisms of ZnO nanoparticles antimicrobial activities (Jafarzadeh *et al.*, 2020)

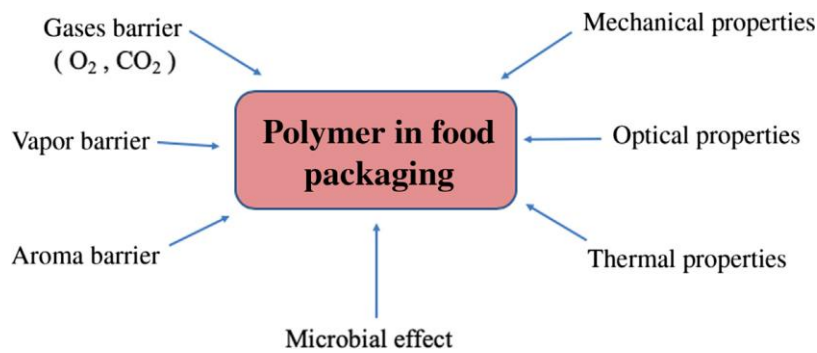


Figure 3. General requirements for polymers in food packaging (Jafarzadeh *et al.*, (2020).