



## REVIEW ARTICLE

## Reviewing the Use of Nanocomposite Polymers in Food Packaging

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**ABSTRACT:** Food packaging plays a significant and fundamental role in ensuring food safety, thus protecting against the infiltration of foreign substances. Consequently, it serves to prevent spoilage and nutrient loss and extends the shelf life of food products. One noteworthy application of nanotechnology in the realm of food is its utilization in food packaging, specifically in the form of nanocomposites. Nanoparticles present in nanocomposites possess a larger specific surface area compared to their micro-sized counterparts, thereby enhancing the interaction between the matrix and nanoparticles and resulting in the development of unique barrier properties. Nanocomposites can be classified as a system that not only passively safeguards food against environmental factors, but also imparts favorable attributes to the packaging system, ultimately enhancing the stability of food in various ways. For instance, nanocomposites have the potential to confer active and intelligent properties to food packaging systems, such as antimicrobial characteristics, the ability to inhibit oxygen, stabilization of enzymes, and serving as nanosensors that detect the level of exposure to detrimental factors like inappropriate temperature and gases produced by bacterial metabolism. Meanwhile, bionanocomposites have garnered significant attention due to the utilization of biodegradable polymers as the matrix of nanocomposites. Among the prominent nanoparticles employed in food packaging are nanoclay, nano silver, silica, titanium dioxide nanoparticles, carbon nanotubes, and cellulose nanofibers. This article endeavors to investigate the key nanoparticles utilized in nanocomposites for food packaging, as well as their respective applications.

### INTRODUCTION

Food packaging is a vital way to ensure their safety. Petroleum polymer films are widely used in packaging industries due to their easy formability, inexpensive price, lightweight, high chemical resistance, good printability as well as the easier production process. However, due to the very low rate of degradation in the environment and lack of collection infrastructure, this type of packaging has its limitations related to disposal, reuse, and recycling in numerous countries. Given the growing population and the need to preserve resources for future generations, in recent years researchers have been seeking new solutions for food packaging. Nanotechnology is one of the most important options [1,

2]. Nanotechnology encompasses the explication, formation, or control of structures, contrivances, or substances, wherein there is a minimum of one dimension (or constituents with a minimum of one dimension) of approximately 1-100 nm in magnitude [3]. There is differing public opinion on the use of nanotechnologies in the food industry. The public at large is most often opposed to the direct use of nanotechnology in food, while the use of this technology in non-food applications is generally acceptable [4]. Over the last two decades, however, interest in nanocomposites for food packaging has increased, and a strong upward trend occurred from 2010 to 2019 [5, 6].

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Before defining the nanocomposite, the composite should first be defined. Composite materials are created by combining two or more components with different properties [7]. Composite materials have two phases: the matrix, which holds the reinforcement materials in place, and the reinforcing or filling phase, which adds unique properties to the matrix. When reinforcement is a nanomaterial, it's called a nanocomposite [8]. On a matrix basis, nanocomposites are divided into three categories: metallic matrix, ceramic matrix, and polymer matrix. In subjects associated with food packaging, nanocomposites with polymer matrix are used, while ceramic and metallic composites are not convenient. For this reason, in most food packaging literature and the current text, nanocomposites are considered polymer nanocomposites [9]. Organic nanofillers include polymer nanofibers, natural fibers, and cellulose nanofibers [10]. The most important mineral nanofillers are clay and laminated silicates, metals such as gold, silver and metal oxides ( $\text{SiO}_2$  and  $\text{TiO}_2$ ) and carbon nanotubes. Nevertheless, nanopolymer fibers, such as starch, are limited in their use in food packaging because of the unstable composites they form [11]. Nanofillers have special geometric shapes such as nanotubes, nanofibers, nanoplates, nanoparticles, and nanocrystals [12, 13].

### ***Three categories of nanopolymers:***

#### ***Advanced nanopolymers (polymer nanocomposites)***

Polymer nanocomposites have better polymer-filler interactions than conventional composites. The even distribution of nanofillers within the polymer matrix results in a larger interface between the matrix and filler. This limits the movement of the matrix and improves mechanical, thermal, and barrier properties [14]. In addition, by reducing the mobility around each nanofiller, an interphase network is created in the

composite which enhances the resistance through twisting, reduces the permeability, and plays an important role in enhancing the properties of the nanocomposite. For a constant filling content, reducing the particle size augments the number of filling particles and brings them closer together. As a result, adjacent particulate interface layers overlap and further reduce permeability [15]. Dispersed and uniform nanomaterials lower permeability by increasing resistance through twisting. As mentioned above, most food packaging materials today are virtually non-degradable, which has become a serious environmental problem worldwide. In an endeavor to heighten the longevity of food and mitigate packaging waste, a significant initiative has been to fabricate comestible and degradable films through the utilization of novel bio-sourced substances, specifically biopolymers [16]. Despite the potential of biopolymers to replace fuel-based materials, their utilization in food packaging has been hindered by various issues related to their physical properties and processing defects. These issues include inadequate thermal resistance, a strong affinity for water, minimal obstacles to progress, properties that are incompatible with gases and vapors, and a shortened lifespan due to chemical and physical deterioration. Additionally, there are numerous challenges associated with their high hydrophilicity. The field of nanotechnology offers a promising approach to enhancing the physical and barrier properties of polymeric materials. This presents significant opportunities for overcoming the aforementioned limitations [17]. Common polymers in food packaging include LDPE, HDPE, PET, PP, PA, PHA, PHB, and PLA, which are produced synthetically or by microorganisms and bacterial fermentation [1, 2, 18]. Figure 1 summarizes the composition and properties of polymer nanocomposites in food packaging.

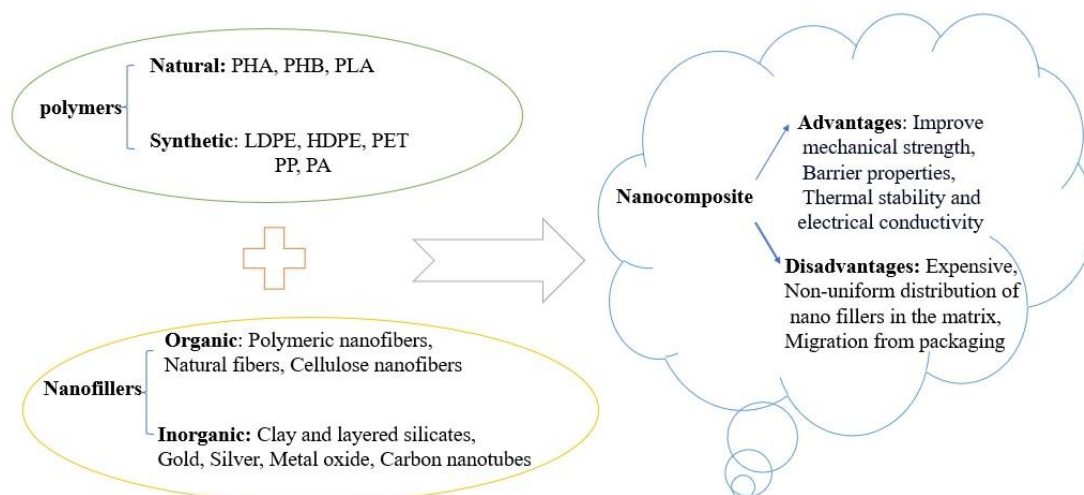


Figure 1. A summary of the composition and properties of polymer nanocomposites

### *The most important nanofillers used in food packaging*

#### *nanocomposites*

#### *Nano clays*

Clay nanoparticles are among the nanoparticles which have been extensively investigated. The reason for this attention is their inexpensiveness, easy access, performance, and good acceptance process [19]. Clay can be used as a nanolayer with a thickness of around 1 nanometer. These particles make the plastic lightweight, strong, and heat resistant and they are a barrier against the passage of gases. The clay used is usually montmorillonite (MMT), which is often considered a commercial nanomaterial [20]. MMT is a clay mineral that has a layered structure. As a result, the double tetrahedral layer of silica is filled with an octahedral layer of alumina (aluminum oxide) at the center. MMT is used as a filler and absorbent and is also present within the structure of ceramics, but its most common application is related to nano usages [21]. Nanoclays are thin plates that limit gas transmission in polymers. Nanoclays have key features that are vital for creating nanocomposites, such as separating layers, distributing nanoclays evenly in the polymer matrix, and improving the interaction between the polymer matrix and nanoparticles [22]. Polymer and nano-clay are used in food packaging for items like meat, cheese, cereals, juices, dairy, and drinks [20].

#### *Cellulose nonreinforcements*

Cellulose nonreinforcements (CNRs) are intriguing materials for low-cost, light, and highly resistant nanocomposites. Cellulose chains, synthesized in organisms (mostly plants), form long nanofibers (2-20 nm in diameter, micrometer in length) stabilized by hydrogen bonds. Study by Azerdo et al. (2009) found that adding 10% cellulose nanofibers to mango puree films reduced water vapor permeability. CNR interactions with mango polysaccharides can prevent water vapor and improve tensile strength of the matrix despite minor length increase. At CNR concentrations above 10%, studies showed impact on polymer tensile properties [23]. Similarly, upon increasing the concentration of CNRs in chitosan-containing matrices, water vapor permeability (WVP) declined and the water vapor barrier improved. In addition, the overall tensile properties of the layers increased, but they did not increase in length [24]. Furthermore, Lima add et al. (2004) dealt with study of the effects of CNRs on starch containing matrix. They concluded that the CNR reduces water permeability in starch and lowers the degradation of starch (delays the conversion of starch to glucose) [25].

#### *Carbon nanotubes*

Carbon nanotubes (CNTs) are small cylinder structures made of carbon atoms that can be either single-walled (SWCNTs) or multi-walled (MWCNTs) connected by

van der Waals force. They are commonly used in PET, PA, and PVA matrices. Carbon nanotubes enhance the mechanical properties of polymer matrices in food packaging, increasing tensile strength, thermal stability, and exerting powerful antimicrobial effects by piercing microbial cells, causing irreversible damage [26].

### Silver nanoparticles

The size of silver particle is within 5-56 nm. These particles have high purity, good dispersion, and a quasi-spherical shape. The use of silver nanoparticles as a filler in food packaging containers makes the packaging impermeable to oxygen, moisture, and microorganisms, which can prevent the growth of bacteria and molds inside it and enhance the shelf life of the product while leaving its appearance and physical characteristic intact [27]. Silver nanoparticles have strong antimicrobial properties that efficiently destroy microorganisms. Although the antimicrobial mechanism of silver nanoparticles is not fully understood, two common

mechanisms are the catalytic production of active oxygen by silver, typically in nano silver composites on TiO<sub>2</sub> or SiO<sub>2</sub> nanocomposite bases. This leads to the formation of reactive oxygen species (ROS), small, unstable, and highly reactive molecules. These molecules, increase molecule reactions and cause damage to protein cell membranes and DNA (seen in Figure 2). The second mechanism is direct damage of silver nanoparticles to cell membranes. The increased positive charge of silver in nano dimensions connects with negatively charged microorganisms, resulting in the destruction of their cell membranes. Some physical features of nanomaterials, like their shape, surface characteristics, and chemical composition, can have toxic effects. For instance, silver nanoparticles can destroy helpful bacteria in food and the body due to their strong antimicrobial properties. For instance, silver nanoparticles harm proteins, membranes, and DNA of helpful bacteria by generating reactive oxygen species (ROS) [28, 29].

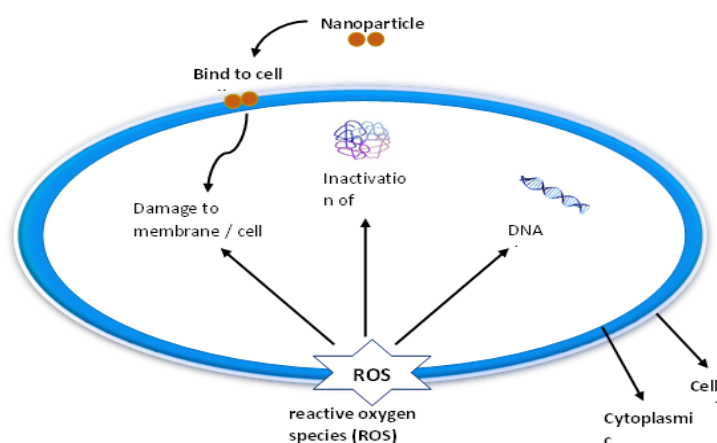


Figure 2. Antimicrobial activity of silver nanoparticles.

### Silica nanoparticles (SiO<sub>2</sub>)

Silicon, a pseudo metal, is the preeminent solid electron element on earth and is extensively found as silica (SiO<sub>2</sub>) and silicates. SiO<sub>2</sub> nanoparticles are efficaciously utilized for augmenting the mechanical strength and thermal stability of plastics, formulating non-stick coating for glasses, bottles, and bags, and ameliorating the water and gas barrier properties in food packaging [30]. Coating polymers constitutes a recently introduced technique aimed at engineering SiO<sub>2</sub> laden nanocomposites. Kim and colleagues (2012) proficiently produced a coating

derived from polylactic acid (PLA) incorporating SiO<sub>2</sub> nanoparticles utilizing the sol-gel technique. Remarkably, this coating yields biodegradable packaging with a 70% enhancement in gas barrier properties in comparison to pure PLA films albeit maintaining its transparency [31]. Packaging with LDPE-SiO<sub>2</sub> has been shown to prolong the freshness of chilled white shrimp for up to eight days, indicating potential for antimicrobial and enzyme inhibition properties in polymer matrices. The integration of SiO<sub>2</sub>/TiO<sub>2</sub>/Ag nanoparticles into

LDPE allows for the development of a versatile nanocomposite that controls CO<sub>2</sub> and O<sub>2</sub> levels, eliminates ethylene, inhibits microbial growth, and extends mushroom storage.[32].

### **Titanium dioxide nanoparticles (TiO<sub>2</sub>)**

Titanium dioxide (TiO<sub>2</sub>) is a frequently employed pigment in a plethora of commodities, including cosmetics and food items, due to its vividly radiant white hue [33]. The Sol-gel technique is frequently employed in the fabrication of this particular type of nanoparticle. The most important feature of TiO<sub>2</sub> nanoparticles is their photocatalytic antibacterial properties, which generate reactive oxygen species (ROS) after exposure to UV light [34]. Using TiO<sub>2</sub> in photocatalysis is highly advantageous for disinfecting water and food-contact

surfaces. It also holds promise for developing active packaging with antimicrobial properties. Additionally, combining TiO<sub>2</sub> with polymers creates UV and oxygen blocking nanocomposites, which protect packaged food and prolong the lifespan of the material. For instance, light exposure degrades carotenoids easily, but this blocking effect prevents degradation [35]. Farhoodi et al. (2017) reported that the photocatalytic activity of TiO<sub>2</sub> has ethylene removal effects, which can be useful for delaying fruit ripening. Also, this nanoparticle can prevent the migration of plastic additives such as ethylene glycol (raw materials in the production of polyester fibers) from packaging to food [36]. Table 1 presents examples of other nanofillers used in the food packaging industry along with the polymer matrix and their applications.

**Table 1.** Some nanofillers used in the food packaging industry

Nanofillers	Polymer matrix	Properties	Application	References
MMT <sup>1</sup>	Cellulose acetate	Mechanical	Packaging with higher mechanical resistance than petroleum plastics	[39]
MMT	PCL <sup>2</sup>	Mechanical	As biodegradable polymer nanocomposites for food packaging	[40]
Cellulose nanocrystals	PLA <sup>3</sup>	Absorbent	As an oxygen absorber in food packaging	[41]
Cellulose nanocrystals	PLA	Barrier and antimicrobial	Antibacterial activity in food packaging	[42]
Modified clay (with organic compounds)	LDPE, HDPE <sup>4</sup>	Barrier	Decrease in permeability to oxygen in polymer with increasing clay concentration	[43]
Starch nanocrystals	Potato starch	Mechanical	As biodegradable edible films for food packaging	[44]
clay with(zno) <sup>5</sup>	Pea starch	Mechanical	The fields of medicine, agriculture, medicine as well as food packaging	[45]
Cellulose nanofibers	Chitosan	Barrier	Reducing moisture permeability in food packaging	[46]

1. Montmorillonite / 2. Polycaprolactone / 3. Polylactic acid / 4. Low density polyethylene, High density polyethylene / 5. Zinc oxide

### **Zinc oxide (ZnO)**

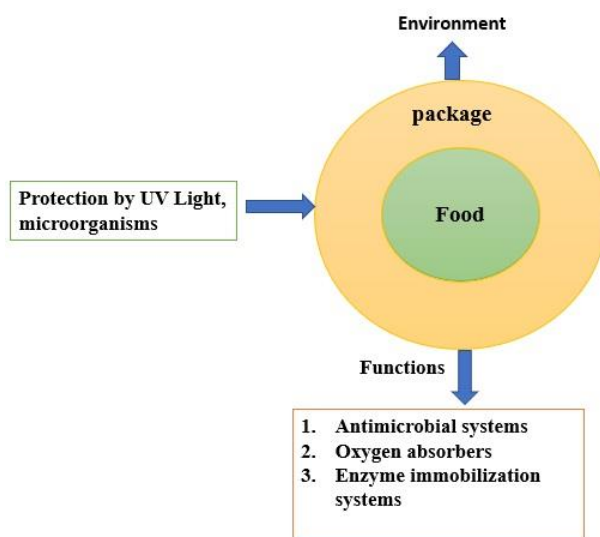
Zinc oxide (ZnO) nanofillers are commonly used in food packaging applications. They are known for their antimicrobial properties against both gram-positive and gram-negative bacteria. Zinc oxide nanofillers can be incorporated into both synthetic and natural polymers to enhance the mechanical and barrier properties of the packaging materials [37]. The addition of zinc oxide nanofillers can also provide UV resistance, moisture resistance, and antioxidant properties to the packaging material. However, there is a need for further research on the toxicity and migration of zinc oxide nanofillers from the packaging into the food. Studies have shown that

only a small quantity of nanomaterials migrates from the packaging into food simulants or actual foods, indicating low consumer exposure and health concerns [38]. Additionally, ZnO nanocomposites have been shown to preserve the quality and extend the shelf life of food products, such as grapes, by inhibiting the growth of pathogenic microorganisms. However, it is important to consider the long-term effects of repeated contact with food simulants on the properties and functionality of ZnO nanocomposite films to ensure their safety for food packaging applications [39, 40].

### Active nanocomposites

The EU Commission regulations include active ingredients that enhance the shelf life, preserve, or improve packaged food. These ingredients release or absorb substances into the food or its surroundings. Conventional food packaging acts as a barrier, protecting the food [6]. Active food packaging is a system that not

only acts as a barrier but also interacts positively with food. This can involve emitting desired substances like antimicrobial or antioxidant agents, or removing harmful components like oxygen or water vapor. The interactions enhance food stability [41]. Figure 3 shows the results of using active packaging to extend the shelf life of packed food products.



**Figure 3.** The performance of active packaging to improve the shelf life of packaged food

### Antimicrobial systems

Antimicrobial packaging for food is popular due to its ability to control harmful microorganisms on food surfaces. Nano materials have a larger surface-to-volume ratio than micro materials. Thus, nanocomposites are very efficient in these systems since they can contain many copies of molecules and microbial cells. Nanoscale materials act as growth inhibitors or lethal agents for antimicrobial activity. Meanwhile, silver nanoparticles, titanium dioxide, chitosan, and carbon nanotubes have more antimicrobial activity, which will be investigated below.

Silver exhibits potent toxicity towards a diverse array of microorganisms and possesses low volatility and exhibits remarkable stability at high temperatures. Undoubtedly, the prevalent nanocomposites employed as antimicrobial films for the purpose of food packaging are founded on nanoparticles of silver. The nanoparticles exhibit antimicrobial activity through various mechanisms. Firstly, they adhere to the cell surface and decompose lipopolysaccharides, thereby forming holes in membranes. Consequently, a substantial escalation in

permeability is observed. Secondly, the said nanoparticles successfully infiltrate the bacterial cell and inflict DNA damage. Lastly, they release antimicrobial  $\text{Ag}^+$  ions by dissolving silver nanoparticles in the cell membrane. Additionally, they produce active oxygen, which was previously discussed [29, 42].

Titanium dioxide ( $\text{TiO}_2$ ) is used as a photocatalytic disinfectant for surface coating. It is activated by UV light, producing active oxygen that destroys bacteria through lipid peroxidation and oxidation of proteins and DNA [43]. Metal doping of  $\text{TiO}_2$  with silver and copper enhances visible light absorption and photocatalytic activity under UV radiation [44]. Doping  $\text{TiO}_2$  nanoparticles with silver significantly improves bacterial photocatalytic inactivation. Chang et al. found successful antimicrobial activity of  $\text{TiO}_2/\text{Ag}^+$  nanocomposite on PVC matrix [45]. Chitosan nanoparticles kill bacteria by interacting with their cell membrane, increasing permeability and causing rupture and leakage. Chitosan nanoparticles have two antimicrobial functions: chelating agents control trace metals in fungal cells, and they

penetrate the cell wall and membrane to bind to DNA, inhibiting RNA synthesis. Additionally, carbon nanotubes also possess antimicrobial properties. Carbon nanotubes have been shown to eliminate E-coli bacteria, possibly by piercing the cells and causing permanent damage [46]. On the other hand, studies suggest that carbon nanotubes can harm the skin and lungs of workers handling them during processing. These effects can impact the health of consumers, but not those who simply use food packaged with carbon nanotubes [47].

### ***Oxygen absorbers***

Oxygen (O<sub>2</sub>) participates in food spoilage reactions in different ways. Direct oxidation reactions are known to result in the occurrence of browning reactions and unpleasant flavors, whereas indirect reactions involving O<sub>2</sub> are typically associated with food spoilage caused by aerobic microorganisms. The most successful oxygen scavenger is TiO<sub>2</sub> polymer nanocomposite. This nanocomposite can be used as a packaging film for various oxygen-sensitive food products. Nonetheless, due to the photocatalytic mechanisms employed by TiO<sub>2</sub>, its principal limitation resides in the requisite of UV light [48].

### ***Enzyme demobilization systems***

Enzymes have many uses in the food industry, but their sensitivity to processing conditions and inhibitors can restrict direct use in foods. Enzyme demobilization involves attaching an enzyme to inactive matter to enhance stability against pH and temperature fluctuations, as well as resistance to denaturation-causing agents. It has been effective for packaging applications. Adding lactase enzymes to packaging materials enhances food value and addresses enzyme deficiencies. Nano-scale enzyme immobilization systems surpass conventional samples in performance due to increased contact surface and mass transfer rate. Enzyme immobilization is often done using nanoclays, which have strong protein absorption abilities and work well as enzyme carriers [49, 50].

### ***Smart nanocomposites***

Estimating food expiration dates considers anticipated

distribution and storage conditions, which often differ from actual maintenance conditions. For example, refrigerated food exposed to higher temperatures may spoil due to packaging vulnerabilities like micropores or sealing defects, allowing oxygen exposure. An intelligent food packaging system can understand food attributes record and transmit information about the quality and safety of the food. Nano sensors are particles integrated into the packaging. Nanosensors in food packaging detect environmental changes, pathogens, pollutants, and freshness status in real-time. Here, we present three instances where nanocomposites have been used as intelligent packaging systems [51, 52].

TTIs or indicators show a food's freshness to consumers by tracking its temperature history, which is important when stored in less than ideal conditions. For example, in freezing a product, a Time Temperature Indicator (TTI) can determine if the product has been properly exposed to temperature and time limits. There are three types of TTIs: critical temperature indicators, partial temperature history indicators, and full temperature history indicators. Critical temperature indicators indicate if the reference temperature has been reached. Partial temperature history indicators use the temperature-time history only when the temperature exceeds a preset value. Complete temperature indicators offer a continuous record of temperature changes over time. These markers are typically revealed through color changes caused by temperature-responsive chemical reactions. Gold nanoparticles are used in cold food packaging to indicate temperature changes. When the temperature is above freezing, the system turns red. However, if the nanoparticles freeze, they clump together and the red color disappears. This serves as a visual indicator for temperature changes in the packaging [53, 54].

Detecting gases from food spoilage is a common concern. Microorganisms produce gases through their metabolism in response to spoilage. Gas nanosensors detect gases in packaging. The nanosensors are made of metal oxide on conductive polymer matrices and can detect microorganisms based on gas emitted by them. Conductive polymers have conductive particles in an insulating polymer matrix. Examples include polyaniline, polyethylene, and polypropylene, which are commonly used [55]. There is high interest in developing non-toxic

O<sub>2</sub> sensors to detect oxygen presence in vacuum packaging systems. Lee et al. study this field. In 2005, they introduced a color-changing UV-activated O<sub>2</sub> sensor. This sensor uses TiO<sub>2</sub> nanoparticles to induce photosensitivity while reducing methylene blue (MB) with triethanolamine in a polymer medium under UV light. UV radiation induces the sensor to lose color, indicating no oxygen presence. In contrast, exposure to oxygen causes the sensor to turn vibrant blue. The color change directly relates to the level of atmospheric oxygen interaction [56-58].

### ***Bionanocomposites***

The term 'nanocomposite' refers to composites where at least one constituent is at the nano scale. In polymer nanocomposites, there is greater interaction between the polymer matrix and filler compared to normal composites. The even dispersion of nanoparticles in the polymer matrix improves contact surface area and enhances mechanical, thermal, and barrier properties. Bionanocomposites are biodegradable materials with biopolymer matrix and nanoparticle filler. In addition to improving properties and having biodegradability, key examples include: (1) starch and derivatives - natural biopolymers with elasticity and low protection after use as packaging, but enhanced when combined with nanoparticles; (2) polyesters - synthetic biopolymers made of polylactic acid and polyhydroxy butyrate biological monomers. Biopolymers have become popular in packaging due to their ability to act as a shell and mold around food. However, they are fragile and lack resistance against gas entry. To address this, nanoclays are used as fillers [59-61].

### **CONCLUSIONS**

Nanocomposites serve the dual purpose of safeguarding food and augmenting packaging through their inherent attributes of stability and active functionalities such as antimicrobial properties, oxygen hindrance, enzyme stabilization, and temperature monitoring. Bionanocomposites strive to augment the utilization of biodegradable polymers by incorporating nanoreinforcements, thereby rendering them more efficacious and viable in a market predominantly

characterized by non-degradable substances. Apprehensions surrounding the utilization of nanotechnology in food contact, particularly pertaining to safety, do exist. Limited evidence suggests that nanostructures originating from packaging materials possess the potential to migrate into food due to their minute size. Consequently, research has been undertaken to assess the risks associated with minuscule particles, some of which exhibit activity within the realm of human physiology and food. Thus far, it has been discovered that silver nanoparticles employed in food packaging do migrate from the packaging to the actual food. The migration of TiO<sub>2</sub> nanoparticles is perilous owing to their heightened toxicity. Additionally, concerns have been raised regarding the transference of carbon nanotubes from the packaging to consumable products. Currently, there is a dearth of conclusive evidence to substantiate the movement of these two distinct nanomaterials. Enthusiasts of nanotechnology strive to extensively augment nanocomposites. Provided those novel technologies are adequately supervised, they hold immense potential for fostering product innovation and contributing to the well-being of individuals.

### ***Conflict of interest***

The authors hereby assert that there exists no conflict of interests.

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