



# The emerging role of metallic nanoparticles in food

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## Abstract

Nanotechnology is widely used in biomedical applications, engineering sciences, and food technology. The application of nanocompounds play a pivotal role in food protection, preservation, and increasing its shelf life. The changing lifestyle, use of pesticides, and biological and/or chemical contaminants present in food directly affect its quality. Metallic nanoparticles (MNPs) are useful to develop products with antimicrobial activity and with the potential to improve shelf life of food and food products. Due to the prevention of microbial growth, MNPs have attracted the attention of researchers. Biopolymers/polymers can be easily combined with different MNPs which act as a vehicle not only for one type of particles but also as a hybrid system that allows a combination of natural compounds with metallic nanocompounds. However, there is a need for risk evaluation to use nanoparticles in food packaging. In this review, we aim to discuss how MNPs incorporated into polymers/biopolymers matrices can be used for food preservation, considering the quality and safety, which are desirable in food technology.

**Keywords** Metallic nanoparticles · Food applications · Packaging · Polymers · Films · Hydrogels · Sensors · Toxicity

## Introduction

Nanotechnology is a wide platform in biomedical applications, engineering sciences, food, and agriculture. In this context, nanomaterials play a crucial role in protection and preservation of food and also in development of sensors (Santos et al. 2007, 2011, 2014; Singh and Sahareen, 2017; Pandit et al. 2017; Suryavanshi et al. 2017; Ahmed et al. 2018; Bhardwaj et al. 2020). The frequent use of pesticides and biological/chemical contaminants present in food directly affects the quality of nutrition in food. The packing materials are important for the protection of food products because they prevent deterioration of food products owing to physico-

chemical or biological factors and maintain the overall quality during storage and handling (Youssef et al. 2015). Metallic nanoparticles (MNPs) are useful to develop antimicrobial products with potential to enhance the shelf life of food by prevention of microbial growth and hence, they have attracted tremendous attention. They interact with different microbial cells, kill them, and have potential to inhibit biofilms formation (Rai et al. 2009, 2017). These characteristics have made them important in food technology applications. In this relation, MNP-based films, hydrogels, and sensors are emerging as a tool in food science. Various studies have shown application of MNPs in food packaging, which is an intelligent system for food preservation. Moreover, MNP-based sensors are used to detect food contaminants particularly microbes (Yang et al. 2017; Sarwar et al. 2018).

Polymer nanocomposites are considered as major technological break-through for many engineering applications. Nanoparticles incorporated into polymers can enhance chemical and electrical properties (Rossi et al. 2017; Wrona et al. 2017). The MNPs are also useful for enhancing conductivity of sensors (Malekzad et al. 2017). Biopolymers and polymers can be easily combined with different MNPs, which act as a vehicle not only for one type of particles but also act as a hybrid system that allows the combination of natural compounds with MNPs (Vasile 2018).

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It is well known that nanotechnology plays an important role in food industries, particularly in the processing and packaging of food and food products. In this context, various studies have been performed and some of the good reviews are also published. However, there is no review available in the current year which specifically focuses on nanoparticles incorporated nano products like nanofilms, hydrogel, nanosensors, and application of hybrid nanomaterials. This will enable scientists to develop newer technology. Therefore, we believe that the present review is timely and hence considering the necessity of these aspects, various above-mentioned topics have been incorporated. In addition, some other important aspects like toxicological concerns and regulations for human biosafety have also been discussed.

### Silver nanoparticles in films/hydrogels as novel antimicrobials

Silver nanoparticles (AgNPs) have already demonstrated its potent antimicrobial activities (Rai et al. 2009; Rai et al. 2012; Siddiqi et al. 2018; Shanmuganathan et al. 2019) and therefore, they can be used in food packaging materials. In this context, Incoronato et al. (2011) evaluated the efficacy of an antimicrobial packaging system containing active AgNPs on the quality deterioration of Fior di Latte cheese. The authors used three different concentrations of silver montmorillonite embedded in agar. It was found that the active packaging system significantly enhanced the shelf life of Fior di Latte cheese. The authors claimed that the active packaging system developed in their study could be used to increase the shelf life of Fior di Latte. In another study, Wang and Rhim (2015) prepared ternary blend agar/alginate/collagen hydrogel films with AgNPs and grape-fruit seed extract. The hydrogel film thus developed was highly transparent and showed activity against food-borne Gram-positive (*Listeria monocytogenes*) and Gram-negative (*Escherichia coli*) bacteria.

Ahmed et al. (2018) reported that the utilization of polylactic acid (PLA) biopolymers incorporated with MNPs can enhance the physico-mechanical properties of PLA natural constitution, promoting more flexibility and better mechanical structures. The increase in barrier properties was modified by the addition of cinnamon oil and MNPs. The antimicrobial efficacy against *Salmonella typhimurium*, *Campylobacter jejuni*, and *L. monocytogenes* inoculated in chicken samples was evaluated and it was found that the films loaded with Ag-CuNPs and 50% CEO demonstrated significant activity against the bacteria. The study reinforces the importance of combined natural bioactives and MNPs in films to obtain biomaterials with biological effectiveness. In fact, the polymer blend is influenced not only by the natural constitution but also due to ligands composition. In a similar study by Li et al. (2017a, b), PLA was impregnated with AgNPs and titanium dioxide that showed no effect of nanoparticles on the

glass transition parameters but they caused an increase in the cold crystallization with effective PLA nano-blend films to antimicrobial activity against *E. coli* and *L. monocytogenes*. Cao et al. (2018) developed integrated polypropylene nanocomposite with AgNPs and evaluated antimicrobial effectiveness against *E. coli* and *S. aureus* as a model. The authors believe that in this process, reactive oxygen species (ROS) enrolled as polypropylenes are extensively used for food packaging applications by improving mechanical and biological properties against food contamination. However, they found a limiting factor that the migration in food directly raises a question of biosafety. Further, the study showed that migration is directly related to temperature, time, and constitution. The food simulant property for food packing application was in the following order: acid food > oily food > alcoholic food > water food. The total Ag migration values ranged between 1.8 and 24.5  $\mu\text{g}/\text{cm}^2$ .

The packaging by the intelligent nanosystem allows to control oxygen limits, microorganism proliferation, and add sensors to evaluate food security. In this context, Olmos et al. (2018) evaluated the efficacy of low-density polyethylene/Ag (LDPE/Ag) nanocomposites in food preservation against biofilm-forming *E. coli*. The results showed that the thermal properties or the surface properties of polyethylene were not altered after incorporation of AgNPs. Moreover, LDPE/Ag nanocomposites also demonstrated efficacy against *E. coli* and its biofilm. Therefore, the LDPE/Ag nanocomposites can be used as storage materials or general-purpose containers. Recently, Bahrami et al. (2019) developed an interesting antibacterial film for food packaging. They used biocomposite film which consists of tragacanth/hydroxypropylmethylcellulose/ beeswax impregnated with AgNPs. The authors evaluated activity of this film against Gram-positive (*Bacillus cereus* ATCC-1247, *S. aureus* ATCC-25923, *Streptococcus pneumoniae* ATCC-49615, and *L. monocytogenes* ATCC-7644) and Gram-negative (*E. coli* ATCC-8739, *S. typhimorum* ATCC-14028, *Pseudomonas aeruginosa* ATCC-9027, and *Klebsiella pneumoniae* ATCC-10031) bacteria and reported that both Gram-positive and Gram-negative bacteria demonstrated inhibitory effect in a dose-dependent manner. Considering these facts, the authors claimed that this new nanocomposite film can be used in food packaging.

Hybrid materials such as PVA/nanocellulose/Ag nanocomposite films can also be used for packaging application to protect against methicilline-resistant *S. aureus* (MRSA) and *E. coli* (DH5-alpha). The maximum inhibition zone ( $14 \pm 0.70$  mm) was observed at 0.5 g AgNPs with 12 wt% nanocellulose against *E. coli* (DH5-alpha), whereas the maximum zone of inhibition against MRSA was found to be  $13.6 \pm 0.68$  (Sarwar et al. 2018).

In another study, Shankar et al. (2018) prepared hybrid nanomaterials by one-pot synthesis of AgNPs, copper oxide nanoparticles (CuONPs), and zinc oxide nanoparticles

(ZnONPs) during regeneration of cellulose from cotton linter (CL) and microcrystalline cellulose (MCC) which showed potential antibacterial activity against *E. coli* and *L. monocytogenes*. This provides evidence that the hybrid material with a variety of MNPs is also an innovative approach in the treatment of food-borne microbes for safety and preservation. In a recent study, Dairi et al. (2019) developed nanobiocomposite films by incorporating AgNPs/gelatin-MMT and thymol. The AgNPs were biogenically synthesized by using *Curcuma longa*. The film demonstrated antioxidant, antibacterial, and antifungal activities. These novel films can be used for enhancing the shelf life of fruits. Moreover, Kadam et al. (2019) developed chitosan-based nanocomposite films impregnated with biologically synthesized AgNPs by using extract of *Nigella sativa* plant. Interestingly, the film demonstrated pH-dependent sustained release of AgNPs. Moreover, the authors evaluated efficacy of these nanocomposite films against two Gram-positive (*B. subtilis* and *S. aureus*) and two Gram-negative bacteria (*E. coli* and *P. aeruginosa*), which showed a remarkable antibacterial activity. Further, the authors reported dose-dependent inhibition of bacterial growth. Similarly, Vishnuvarthanan and Rajeswari (2019) also prepared nanocomposites of carrageenan/AgNPs/Laponite and coated on the oxygen plasma surface-modified polypropylene film. AgNPs were synthesized by extract of *Digitalis purpurea*. Further, they assessed adhesion, mechanical barrier, and antimicrobial properties of nanocomposites, which were significantly increased. *E. coli* and *S. aureus* demonstrated higher sensitivity to these nanocomposites.

### Copper nanoparticle-impregnated films for food packaging

Ebrahimiasl and Rajabpour (2015) reported remarkable antimicrobial activity of biodegradable hydroxypropyl methylcellulose (HPMC) matrix impregnated with copper nanoparticles (CuNPs). The film showed mechanical and water vapor barrier properties due to incorporation of CuNPs. Antimicrobial efficacy of these films was tested against *S. aureus*, *S. epidermidis*, *B. cereus*, *E. coli*, *E. faecalis*, *Salmonella* sp., and *P. aeruginosa* at different concentrations of CuNPs. The results demonstrated the potential bactericidal activity of nanocomposite films against all tested bacteria. Therefore, the authors proposed that CuNP-impregnated films can be used in the packaging and preservation of food products like meat thereby avoiding microbial growth. Moreover, Tamayo et al. (2016) studied copper-polymer nanocomposite films which have already demonstrated antimicrobial activities. These nanocomposites can be used against *S. aureus*, *E. coli*, *S. cerevisiae*, *Streptococcus* spp., and *Pseudomonas* spp., etc.

Similarly, Varaprasad et al. (2017) prepared metal-oxide polymer nanocomposite films using biodegradable poly- $\epsilon$ -caprolactone, disposed of poly(ethylene terephthalate) oil bottles monomer and zinc oxide-copper oxide nanoparticles. The metal-oxide polymer nanocomposite films demonstrated excellent mechanical properties. The authors claimed that this type of film can be used in domestic packaging.

In another study, chitosan (CS) and endogenous Cu nanoclusters (NCs) capped with protein were used (Li et al. 2017a, b). The chitosan/soy protein nanocomposite film improved the tensile strength and elongation of composite films by 118.78% and 74.93%, respectively. The authors claimed that owing to the improved characteristics, these films can be used in food packaging. Arfat et al. (2017a, b) developed silver-copper alloy nanoparticle nanocomposite films based on guar gum. The authors evaluated the influence of Ag-Cu nanoparticles after loading on guar gum and found the improved mechanical strength, and UV light and oxygen barrier efficacy. Moreover, the film thus prepared demonstrated antibacterial activity against *L. monocytogenes* and *S. typhimurium*. The Ag-Cu nanoparticles were more effective against Gram-negative than the Gram-positive bacteria.

Ahmed et al. (2016) developed plasticized PLA-based nanocomposite films impregnated with polyethylene glycol (PEG) and two nanoparticles including silver-copper alloy and zinc oxide with the size range of 50–100 nm. The film showed significant tensile strength as compared with the combination of plasticized PLA and PEG. Further, Ahmed et al. (2018) prepared plasticized PLA composite films by using bimetallic nanoparticles of silver-copper (Ag-Cu) and essential oil of cinnamon. The efficacy of the film was evaluated on *S. typhimurium*, *C. jejuni*, and *L. monocytogenes* inoculated chicken. The growth of these pathogens was remarkably reduced to 3.87, 2.59, and 2.42 log CFU/g when 50% cinnamon essential oil was used in packaging.

Copper-based nanocomposites have been reported to have potential antibacterial activity against a wide range of Gram-positive and Gram-negative bacteria. Several possible mechanisms have been proposed for the mode of action of copper-based nanocomposites which are explained in detail by many researchers including Tamayo et al. (2016). Based on all the studies, it can be said that copper-polymer nanocomposites mainly target the bacterial cell through three different steps which mainly include (I) release of copper ions, (II) release of CuNPs from nanocomposites, and (III) inhibition of biofilms. In the first phenomenon, the copper ions interact with cell wall (e.g., amines and carboxyl groups in N-acetylglucosamine and N-acetylmuramic acid in the peptidoglycan layer) and various other biomolecules present in the membrane proteins (e.g., sulfhydryl groups). These interactions destabilize the membrane potential and subsequently disintegrate the bacterial cell wall and membrane. In addition, copper ions released also reported to bind and interact with

DNA to disintegrate its helical structures, moreover, ROS generated due to released copper ions lead to bacterial death. In the second phenomenon (i.e., release of CuNPs from nanocomposites), thus released CuNPs bind to the surface of bacterial cells through different adherent forces like electrostatic forces and molecular interactions. Further, these nanoparticles penetrate the cell through the outer cell wall or membrane by endocytosis and direct diffusion. Later, they target the bacterium by similar mechanisms proposed for released copper ions. Similarly, CuNPs inhibit biofilm formation (Applerot et al. 2012; Tamayo et al. 2016). Figure 1 represents the schematic illustration of the antibacterial effects of copper-polymer nanocomposites.

### Gold nanoparticles as sensors in food technology

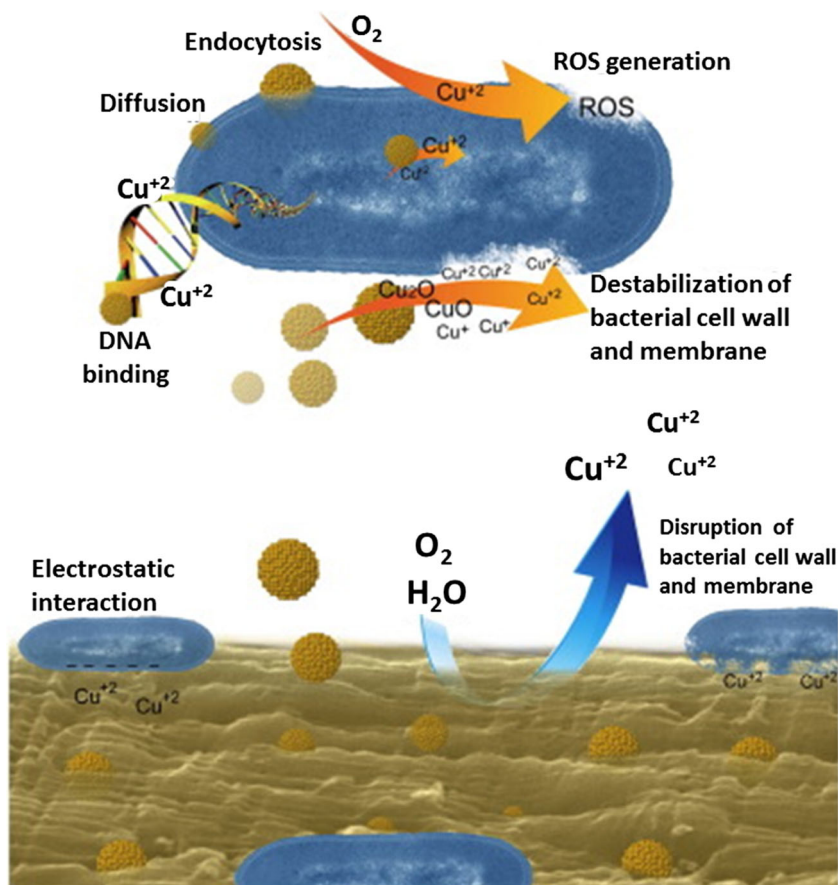
The presence of contaminants in food is a major problem observed by food manufacturer and it is mainly due to the use of toxic chemicals during processing or because of chemical compounds present in packaging materials. Bisphenol A is an example of contaminants present in food that passes through packaging material to food. Yang et al. (2017) developed a surface-enhanced Raman spectroscopy (SERS) system based in gold nanoparticles (AuNPs). In this connection, AuNPs were used to potentialize modification of substrate with the

help of aggregation agent able to improve the detection sensitivity of BPA residue in milk. The study showed a good linear relationship ( $R^2 = 0.990$ ) between the intensity of SERS signal and the logarithm of BPA concentration in the range. The recovery of BPA residue is reported between 89.5 and 100.2% with relative standard deviation ranging from 4.6 to 2.7%. The application of AuNPs as sensors is the most promising approach owing to their interesting properties like reactivity, selectivity, and specificity which makes them efficient and sensible against food contaminants.

Shi et al. (2013) in a study explored the optical properties of AuNPs in thin films which allowed to obtain semiconductor fluorescent material. The authors prepared gelatin-SiNP composite thin films and successfully incorporated AuNPs. This composite film is important being portable, flexible, transparent, and free-standing with multiple applications. Fan et al. (2020) developed an electrochemical sensitive sensor to determine the  $\text{Ca}^{2+}$  concentration in meat. Oxygen plasma-treated graphene was used with AuNPs. This electrochemical method was used for the detection of pork. The authors believed that this method can be applied for the detection of  $\text{Ca}^{2+}$  in meat samples.

Bozkurt et al. (2018) developed SERS-based biosensor for the detection of *E. coli* through a sandwich immunoassay. This SERS-based biosensor was prepared by coating spherical

**Fig. 1** Schematic illustration of antibacterial effects of copper-polymer nanocomposites [Reprinted with permission from Tamayo et al. (2016), copyright permission from Elsevier]

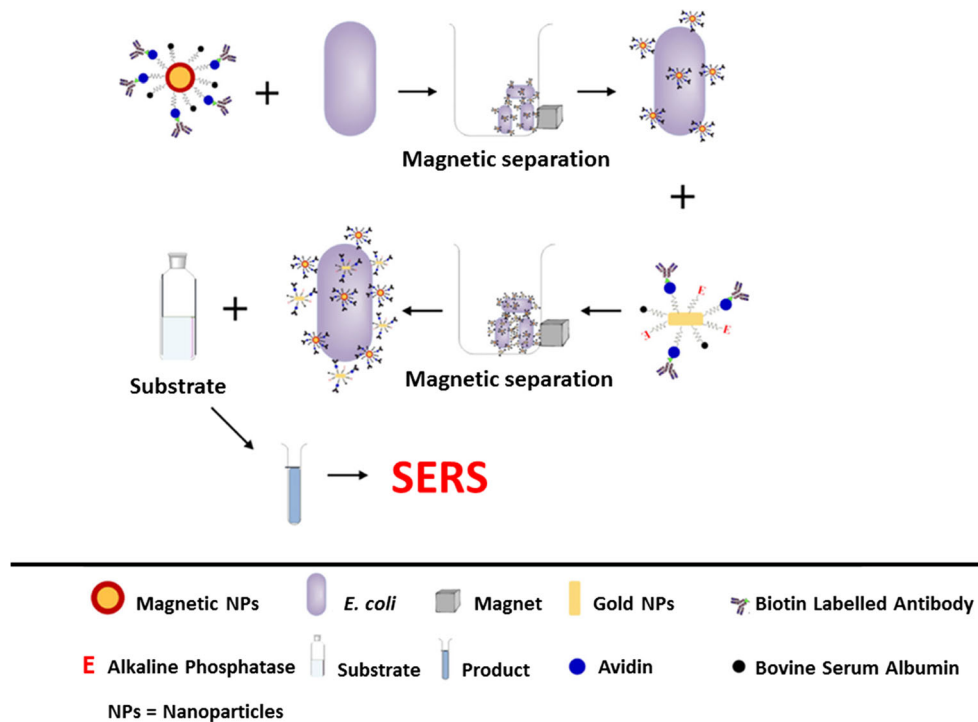


AuNPs on magnetic nanoparticles (MNPs) to form AuNP-coated MNPs complex (MNPs-Au) followed by its modification using *E. coli* antibody for immunomagnetic separation. On the other hand, rod-shaped AuNPs were labeled with alkaline phosphatase (ALP) and this complex was also modified by *E. coli* antibody. Initially, MNPs-Au complex modified by *E. coli* antibody was added to the medium containing *E. coli* and subsequently, *E. coli* were captured on this complex and separated by applying an external magnetic field. Further, another complex already prepared, i.e., ALP labeled and *E. coli* antibody-modified rod-shaped AuNPs, was added to previously captured and separated *E. coli* so as to form the sandwich immunoassay. Finally, the specific substrate for ALP enzyme, i.e., BCIP (5-bromo-4-chloro-3-indolyl phosphate), was added which leads to enzymatic hydrolysis of the substrate, the AuNPs present in the sensor enhanced the SERS signal specific to BCIP, the intensity of SERS signal depends on the concentration of *E. coli* present. Figure 2 represents the schematic illustration of different steps involved in the detection of *E. coli* using active sandwich immunoassay.

The food is usually contaminated by different pathogenic microbes, and therefore their timely detection is very important. Globally, these microbes are a major threat to human health. Such microbes generally include bacteria, fungi, viruses, and other parasites. Among these, fungi secretes various mycotoxins in food, which are of great importance because they cause severe health problems in humans (Richard 2007). Mycotoxins are toxic secondary metabolites produced by a variety of fungi which mainly include various members of

the genus *Aspergillus*, *Fusarium*, and *Penicillium* (Goyal et al. 2017). Mateo et al. (2018) reported that about 25% of the world's food crops get contaminated every year due to mycotoxins, resulting in the huge industrial and agricultural loss. Among the various mycotoxins, aflatoxin is considered as most potent, strong teratogen, mutagen, and carcinogenic. Aflatoxins are low molecular weight toxic compounds generally produced by different species of *Aspergillus* such as *A. flavus* and *A. parasiticus*. These mycotoxins are commonly found in various food and feed stuffs including cereals, oil-seeds, spices, coffee beans, and nuts (Omrani et al. 2016; Negash 2018). Therefore, considering the major health issues caused due to mycotoxins, there is a growing interest among the food technologists to develop the most effective and sensitive approach for the detection of food-borne microbes and their toxins. In this context, various attempts have been made mostly focusing on the development of approaches based on nanotechnology. Pissuwan et al. (2019) reviewed the role of various AuNPs based assays for the detection of food-borne microbes. In another recent study, Bhardwaj et al. (2020) developed surface plasmon resonance (SPR)-based nanosensor (chip) by the incorporation of AuNPs for the detection of aflatoxin in food. The nanobiosensor was developed through three different steps which include (i) Step-I: synthesis of AuNPs and ligand exchange process from CTAB to lipoic acid surface modification, (ii) Step-II: self-assembled monolayer (SAM) of cystamine and lipoic acid on Au sensor chips surface, and (iii) Step-III: in situ activation of SAM-modified chip surface, grafting of protein-A, and immobilization of

**Fig. 2** Schematic illustration of different steps involved in detection of *E. coli* using ALP-labeled SERS active sandwich immunoassay [Reprinted with permission from Bozkurt et al. (2018), copyright permission from Elsevier]



anti-AFB1 antibodies followed by interaction with AFB1 antigen. Figure 3 represents the schematic illustration of the different steps involved in the biosensor active surface modification. The obtained results revealed that thus developed nanosensors are potentially effective and most sensitive in the detection of aflatoxin.

### Hybrid materials in food technology

Hybrid metal-polymer matrices show distinct properties, such as large surface areas, ordered crystalline structures, highly regularized pores, and structure functionalities making them a new class of materials for diverse applications (Zhang et al. 2014). The use of potential MNPs combined with natural compounds is also an approach that needs to be explored. Krishna et al. (2016) and Yang et al. (2017) reported a sensor composed of graphene oxide-nickel nanoparticle biopolymer films that aim to detect glucose concentration. The sensor can be used not only for glucose detection in body fluids, but it can also be used in a food application system, due to biocompatible materials utilized, the low toxicity Ni and low-cost technology.

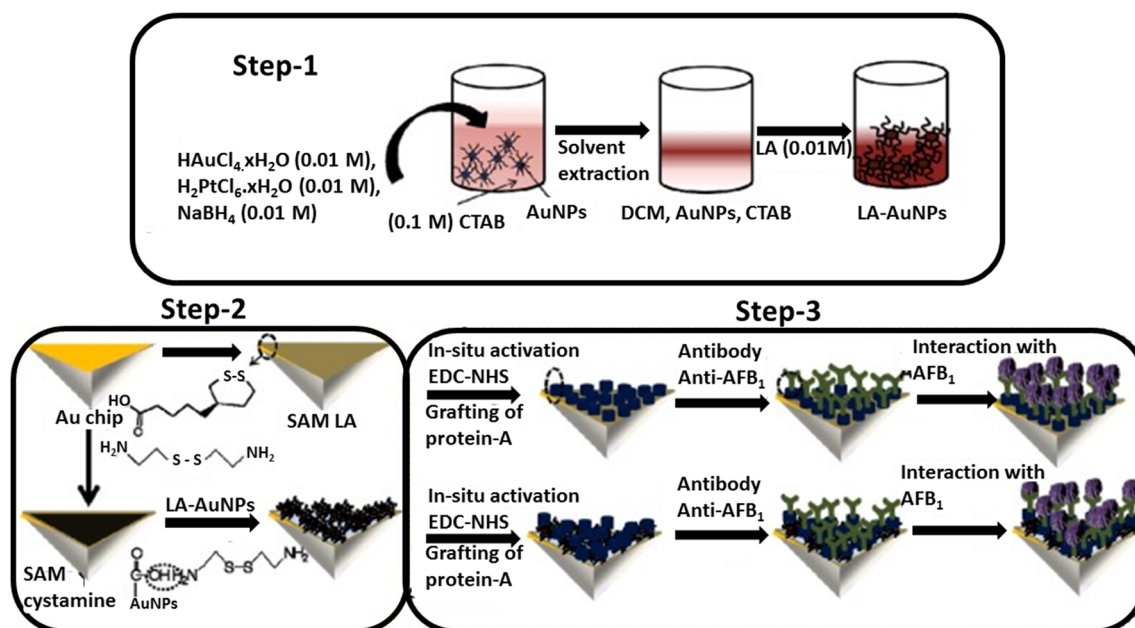
Oun and Rhim (2017) studied the natural properties of chitin combined with ZnO/AgNPs after their incorporation in carboxymethyl cellulose. Hybrid material originated films exhibited strong antibacterial activity against both Gram-positive and Gram-negative bacteria. As discussed in this review, the combination of nanocomposites exhibits better physico-chemical properties of the films when they are

incorporated with nanoparticles. The increased thermal stability by the incorporation of ZnO-AgNPs showed the high UV barrier property. Similarly, the tensile strength and elastic modulus of the composite film were also increased, whereas elongation at break was found to be decreased.

Yaşa et al. (2012) reported antimicrobial activity of nanosized Ag-doped TiO<sub>2</sub> colloids against *E. coli*, *S. aureus*, *B. subtilis*, *S. typhimurium*, and *C. albicans*. Although the study was not focused on food applications, the microorganisms tested show an excellent prognostic in the use of this hybrid in food application process. In chitosan films, the combination of AgNPs and AuNPs showed promising antimicrobial activity against bacteria (*S. aureus*, *P. aeruginosa*), fungi (*A. niger*), and yeast (*C. albicans*) (Youssef et al. 2014). Later, in another study, Youssef et al. (2015) combined bionanocomposite based on chitosan/poly (vinyl alcohol)/titanium nanoparticles (TiO<sub>2</sub>NPs) (CS/PVA/TiO<sub>2</sub> nanocomposite) which were used as packaging materials for soft white cheese. Chemical and antimicrobial properties were also evaluated showing an increase in storage time and reduced bacteria, yeast, and mold counts compared with the control.

### Other bionanocomposites in food technology

Bionanocomposites are considered as one of the most important class of hybrid materials, which are mainly composed of biopolymers and inorganic solids having at least one dimension on the nanometer scale. The discovery of such biodegradable materials is found to be promising gift to present and



**Fig. 3** Schematic representation of the different steps involved in the active surface modification of nanobiosensor. Step-1: synthesis of AuNPs and ligand exchange process from CTAB to lipoic acid surface modification. Step-2: self-assembled monolayer of cystamine and lipoic acid on Au sensor chips surface. Step-3: in situ activation of SAM-

modified chip surface, grafting of protein-A, and immobilization of anti-AFB1 antibodies followed by interaction with AFB1 antigen (Reprinted with permission from Bhardwaj et al. (2020), copyright permission from Elsevier)

future generations. To date, various synthetic and natural polymers have been successfully used in the development of bionanocomposite materials. Among the various polymers, natural polymers such as starch, cellulose acetate, chitosan, and different proteins are considered as environment friendly and hence preferably used in the past few years for a wide range of application including food industry. For example, optically transparent plasticized PLA-based bionanocomposite films have been utilized for packaging in the food industry (Mohanty and Swain 2017; Arora et al. 2018).

The combination of starch and clay is considered as the most promising biodegradable nanocomposites having potential applications in food packaging because these bionanocomposites exhibit excellent barrier properties and hence essentially required for any ideal food packaging material (Cyras et al. 2008). In another study, Yu et al. (2009) developed a starch/ZnO-carboxymethylcellulose sodium nanocomposite using ZnONPs stabilized by carboxymethylcellulose sodium (CMC) as the filler in glycerol-plasticized pea starch. These films also found to have promising applications in food industry. Similarly, De Moura et al. (2009) synthesized a bionanocomposite material by incorporating chitosan-tripolyphosphate (CS-TPP) nanoparticles into hydroxypropyl methylcellulose (HPMC) films. It was observed that incorporation of CS-TPP nanoparticles into HPMC films significantly improved its mechanical and barrier properties. Thus, the authors claimed its applications in food industry as a packaging material. Zare et al. (2019) synthesized zinc oxide-silver nanocomposites (ZnO-Ag NCs) using leaf extract of *Thymus vulgaris* as a stabilizer and a reducing agent through biomimetic hydrothermal decomposition reaction. Later, this synthesized nanocomposite was incorporated into poly(3-hydroxybutyrate-co-3-hydroxyvalerate)-chitosan (PHBV-CS) as an active agent in a highly precise ratio to develop the novel degradable biopolymer nanocomposite. Further, this bionanocomposite was evaluated for its enhanced mechanical properties and antimicrobial activity to improve the shelf life of poultry items. These findings showed the application of ZnO-Ag NCs as a nano-active agent in the food packaging by controlling its spoilage. Moreover, these bionanocomposites demonstrated potential antimicrobial activity that offers effective and novel replacement of traditional petrochemical-based polymers conventionally used for packaging of poultry items. Moreover, some reports suggested that incorporation of ZnONPs to PHBV enhances antimicrobial activity of ZnONPs, which can be used to increase the shelf life of food. Abdalkarim et al. (2017) demonstrated that PHBV/ZnO nanocomposites possess potential antibacterial activity against *E. coli* and *S. aureus*. Further, it was also observed that incorporation of ZnO (ZnONPs) not only positively affects barrier and hydrophilicity of the polymer blend coatings but also improves the tensile strength, Young's modulus of PHBV and increases its maximum decomposition temperature. As far as

ZnONPs-based bionanocomposites are concerned, in addition to above-mentioned PHBV, various other biopolymers like chitosan, poly(3-hydroxybutyrate) (PHB), poly(butylene adipate-co-terephthalate) (PBAT), low-density polyethylene (LDPE), semolina flour, and bovine skin gelatin type-B (BSG) have been used for its incorporation. For instance, Abbas et al. (2019) reviewed the role of ZnONPs bionanocomposites films incorporated with all such biopolymers. The authors proposed that incorporation of ZnONPs in the biopolymer films make them resistant to fire, lightweight, stronger in thermal and mechanical performance, and less permeable to moisture and gases. Hence, due to these improved properties, such bionanocomposite films can be effectively used for food packaging.

Recently, Kaseem et al. (2019) reviewed the recent advances in application of PLA/TiO<sub>2</sub> composites for various fields including food preservation and packaging. In this context, Garcia et al. (2018) reported that addition of TiO<sub>2</sub>NPs into the PLA matrix was highly effective against a wide range of bacterial strains and hence authors suggested the use of these nanocomposites instead of AgNPs. Actually, the migration phenomenon of nanoparticles is considered as a critical factor to evaluate the safety and relevance of the PLA/TiO<sub>2</sub> composites (Girdthep et al. 2014). Therefore, Li et al. (2018) studied migration phenomenon of TiO<sub>2</sub>NPs and AgNPs from PLA/TiO<sub>2</sub> and PLA/TiO<sub>2</sub> + Ag composite films. The results obtained showed that the amounts of TiO<sub>2</sub>NPs and AgNPs migrated above-mentioned PLA composite films to cheese specimens were much lower than the migration limit proposed by European Food Safety Agency for food contact materials. Considering all these advantages, it has been confirmed that PLA/TiO<sub>2</sub> nanocomposites can be safely used as antimicrobial food packaging films.

Recently, Zubair and Ullah (2019) reviewed the role of various protein-based bionanocomposites in food industry and proposed that such bionanocomposites can be promisingly used for various purposes including food packaging. Kanmani and Rhim (2014) developed gelatin-based active nanocomposite films incorporating AgNPs using a solution casting method. The incorporation of AgNPs into the gelatin-based nanocomposite film promisingly enhanced its antimicrobial activity including. Further, it was observed that thus developed bionanocomposite showed potential antibacterial properties against both Gram-negative and Gram-positive food-borne pathogens. In another study, Shankar et al. (2015) prepared the gelatin/ZnONP nanocomposite films and after evaluation of their antibacterial efficacy it was confirmed that these films considerably more active against Gram-positive food-borne pathogenic bacteria than Gram-negative. Similarly, TiO<sub>2</sub>NPs at different concentrations (1, 3, and 5 wt%) were used to develop biodegradable whey protein isolate (WPI) films and evaluated their physical and mechanical properties. The results obtained revealed that

increase in concentration of TiO<sub>2</sub> from 1 to 5% showed a decrease in water barrier permeability as well as water vapor transport rate of films. These properties are considered as important for food packaging materials and hence such films can be effectively used in food packaging to increase its self-life (Zolfi et al. 2014). Arfat et al. (2016) developed effective antimicrobial nanocomposite film using fish protein isolate and fish skin gelatin by the addition of ZnONPs and used an efficient food packaging to prevent the growth of pathogen and spoilage bacteria in foods.

Besides all these bionanocomposites, there are few other bionanocomposites also developed by the incorporation of nanoparticles in different polymers. Due to improved antimicrobial and other properties like moisture and gas barrier, these bionanocomposites have been promisingly used in food industries for various purposes.

### Toxicity and biosafety concerns

The applications of important MNPs and other various nanomaterials in food are already described in this review. But the toxicity and biosafety of these materials is a major issue when nanoparticles are in materials especially those in contact with food. The polymers commonly used as films, hydrogels, and sensors are most of the time composed of biopolymers or materials that are ecofriendly and biocompatible for human use. However, nanomaterials like MNPs are described with potential toxicity, depending on exposure factors, time of exposure, and contact. To date, toxicological effects of different nanomaterials have been studied by both in vitro (using various cell lines) and in vivo (using experimental models like zebrafish, mice, etc.) approaches.

From the available reports, it is clear that nanomaterials exert toxicological effects in living organisms, as well as in environmental ecosystems, which is commonly referred as ecotoxicity. The flora and fauna present in the ecosystem helps to maintain the ecological balance and hence, there is ecological connectivity with such living organisms and ecosystem. But, it was found that nanomaterials through their toxicological effects may disturb this ecological balance. Therefore, evaluation of toxicological effects of nanomaterials and the understanding of their interaction with different taxa present in the ecosystem are essentially required. In this context, a number of studies have been performed demonstrating the negative effects of nanomaterials in various trophic levels. Bour et al. (2015) demonstrated the toxicity of cerium dioxide nanoparticles (CeO<sub>2</sub>NPs) in aquatic environments on living systems from different trophic levels which include four species (i.e., one is diatoms *Nitzschia palea*, second is the sediment-dwelling invertebrate *Chironomus riparius*, and the third and fourth are amphibian larvae *Xenopus laevis* and *Pleurodeles waltl*). It was found that CeO<sub>2</sub>NPs inhibit the growth of all above-tested species.

Further, the authors observed that in aquatic environment, CeO<sub>2</sub>NPs tend to aggregate and sediment quickly. These findings suggested that, in the case of water contamination with such nanoparticles, exposure not only affects organisms from the water column but it also harms the benthic organisms. These organisms are of prime importance from an ecological point of view, as they represent the base of the trophic chain.

Apart from this, Ramachandran et al. (2018) used zebrafish as animal model and exposed to AgNPs and AuNPs to evaluate damages and compared both for toxicity when they are used in the same conditions. It was observed that the treatment of AgNPs enhances the secretion of alanine aminotransferase and aspartate aminotransferase enzymes in the cell compared with AuNPs. The enhanced secretion of these enzymes leads to the increase in the ROS generation level and thus caused more damages than AuNPs through oxidative stress and immunotoxicity. Moreover, the formation of micronuclei and nuclear abnormalities was observed in the cells treated with AgNPs, which was not observed in cells treated with AuNPs. From these observations, it can be concluded that AuNPs are relatively safe and less toxic bionanomaterials for possible applications. Similarly, Rajput et al. (2018) demonstrated the toxicological concerns of copper oxide nanoparticles in barley (*Hordeum sativum distichum*) which is considered as one of the most important staple food crops. The study involves the evaluation of effects of these nanoparticles on stomatal aperture and root morphology, metaxylem size, and changes in cellular organelles (plastids, mitochondria), as well as in plastoglobules, starch granules, protoplasm, and membranes. All the above-mentioned studies confirmed that nanomaterials exert harmful effects in all kinds of living systems present in the environment such as plants, aquatic organisms, terrestrial animals, and human beings.

### Regulations for human safety

For assessment of risk concerning the use of nanoparticles in food packaging, the degradation of nanomaterial in the environment for human safety is imperative. Kotsilkov et al. (2018) evaluated the release of graphene carbon nanotubes from poly(lactic) nanocomposite films. Regarding polymer film, the release of nanocompounds and the impact of contaminations are a real concern. The observed limit for migration was estimated at around 0.028–0.053 mg/cm<sup>2</sup> and 0.006–0.011 mg/cm<sup>2</sup> for PLA films and nanoparticle migration, respectively. Both are below the EU regulation overall limit that was 0.10 mg/cm<sup>2</sup>.

Dimitrijevic et al. (2015) reported that the effect of nanomaterial on the human body depends on its property. The time of nanoparticles' circulation increases in the case of hydrophilic and positively charged nanoparticles. Although the exact mechanism is not known, it is believed



that the toxicity is based on oxidative stress. Gallochio et al. (2015) reported that the impact of nanomaterials in food science is very important since early stages until the uptake by living organisms. The authors summarized the principle rules regarding use of nanomaterials in food. If nanomaterials are used as primary ingredients (e.g., nanoemulsions), they fall within the scope of “Novel Food” Regulation (258/97) as “foods and food ingredients with a new or intentionally modified primary molecular structure” and they are subjected to a risk assessment procedure before market approval. If they are used as food additives, a different procedure is applied (Reg 1333/2008) and they are expected to be inserted in the EU register before use (Gallochio et al. 2015). It is noteworthy to mention that Switzerland seems to be the unique country where nano-specific provisions have been incorporated in existing legislation, while in other countries nanomaterials are regulated more implicitly by mainly building on guidance for industry (Amenta et al. 2015).

Rainieri et al. (2018) emphasized on metallic safety of commercial food products describing “The European Food Safety Authority (EFSA) Scientific Opinion entitled Guidance for risk assessment of engineered nanomaterials which recommends examination of the following conditions: i) the state in which the nanomaterial was produced; ii) the state in which the nanomaterial is used or is present in the food and feed; iii) the state in which the nanomaterial is present in the toxicological studies, and iv) the state of the nanomaterials in biological fluids and tissues.”

## Conclusions

The applications and advantages of MNPs and nanocompounds in food preservation and treating contamination are well recognized. The versatile nature of materials, combinations, and hybridization makes them more attractable. The materials that combine nanocompounds with polymers and natural compounds are promising sources that need to be explored. The MNPs such as AgNPs, AuNPs, and CuNPs have already demonstrated a wide antimicrobial activity against pathogenic bacteria, fungi, and yeasts. In addition, the physical properties such as high reactivity, specificity, and selectivity make them an attractive combination with other materials to provide large applicability. There is a greater need of sensors in food technology in order to constantly monitor process, compounds, and nutrients. The risk assessments, biosafety, and regulations concerning these compounds are still a challenge and warrants extensive research. The lack of uniformity in worldwide regulations makes them potentially dangerous when one takes an account of body exposure or the environment impacts.

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## Compliance with ethical standards

**Ethics approval** This article does not contain any studies with human participants or animals performed by any of the authors.

**Conflict of interest** The authors declare that they have no competing interests.

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