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REVIEW

A review of silver nanoparticles in food packaging technologies: Regulation, methods, properties, migration, and future challenges

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Abstract

The development of antimicrobial food packaging is needed for food preservation and quality maintenance. Silver nanoparticles (AgNPs) have been widely used as an antimicrobial agent in food packaging technologies. However, the risks associated with their potential migration into foods are a major concern. This paper comprehensively reviews the use of AgNPs in food packaging technologies. The application of AgNPs in food packaging technologies has been regulated by the United States Food and Drug Administration and the European Food Safety Authority. The addition of AgNPs into food packaging can improve their barrier, mechanical, and antibacterial properties, as well as maintain the quality of foods. Migration of AgNPs from food packaging into foods is still a concern as it has implications for human health associated with their toxicity properties. A study on the toxicological properties of AgNPs released from food packaging needs to be carried out intensively to ensure their safety before being widely implemented. Moreover, comprehensive economic evaluation to implement AgNPs in food packaging is needed as such a study is missing in the literature.

KEYWORDS

antibacterial properties, food packaging, silver nanoparticles

1 | INTRODUCTION

Currently, silver nanoparticles (AgNPs) have earned enormous popularity in scientific literature due to their exclusive and desirable antibacterial properties against a wide range of bacteria.[1–5] Antibacterial action of AgNPs can be explained by two roles, which are Ag^0 and Ag^+ species.[6,7] Several studies have proven that AgNPs can be accumulated in the membrane of bacteria. As a result, the integrity of the membrane of bacteria can be decreased, causing cellular death. It is also possible that the mechanism can occur via the generation of reactive oxygen species.[8–11] Alternatively, the antibacterial properties of AgNPs can be facilitated via Ag^+ species. In this mechanism, Ag^+ species have a role as an antibacterial agent, and the nanoparticle becomes a reservoir.

The fabrication of antimicrobial composite packaging films has been recently increased, particularly in the food industry because of the demands of both consumers and food processors for safe and high-quality foods.[12] It is noted that food packaging with antibacterial properties has the capability of releasing active biocide substances for the improvement of food quality, extension of shelf life, and prevention of spoilage.[13–20] This can be achieved using organic materials or by adding inorganic materials into food packaging, the latter of which is currently becoming more popular. By employing organic materials, it can be achieved using organic acids and enzymes.[21–23] Alternatively, it can also be achieved by using inorganics such as metal nanoparticles or metal oxides.[24–26]

The use of inorganic materials in food packaging technology seems to be more promising compared to organic materials because of the following reasons. It has been proven that organic materials have several shortcomings, such as instability at high temperatures. Therefore, the use of nanoparticles can diminish the problem as they have the ability to withstand harsher processing conditions.[27] In addition, limitations of the use of organic materials can be associated with their weak mechanical properties and sensitivity to moisture. Another benefit of the use of inorganic materials in food packaging is also related to low effects on the sensory attributes in food.[28] In comparison, organic compounds, such as essential oils or plant extracts, that are commonly used in food packaging technology may strongly modify organoleptic properties of food products because of their strong odor and flavor.[29]

Among existing inorganic nanomaterials, AgNPs have been intensively used in food packaging technology because of their antibacterial properties. Food packaging with AgNPs has been tested for various foods such as fresh fruits, fresh meats, and consumer products. Hence, concern regarding the risks associated with the potential migration of AgNPs or Ag species into food becomes a central issue. This leads food safety authorities to regulate the use of AgNPs in food packaging.

Although numerous review papers discussing the application of nanotechnology in food packaging technology have been reported in literature, review papers specifically focused on the application of AgNPs in food packaging technologies are hard to find.[30–33] As a result of the aforementioned necessity, this paper comprehensively reviews the use of AgNPs in food packaging technologies. This review can be a good strategy for designing advanced nanotechnology for future packaging technology, particularly from the point of view of chemistry.

2 | REGULATION

Due to the intensive use of AgNPs for food packaging technologies, there is heightened concerns about the potential risks related to the release of AgNPs from the packaging into foods. This calls for a proper set of regulations from food safety authorities. In fact, regulation of the use of AgNPs in food packaging has been issued by food safety authorities in the European Union (EU) and the United States.

For example, without authorization, the use of AgNPs in food packaging and food supplements is not allowed as recommended by the European Food Safety Authority (EFSA).[34] In addition, EFSA has established an allowable limit of Ag migration from packaging, 0.05 mg/L in the water and 0.05 mg/kg in food. Moreover, a document published in 2011 by the EFSA enforced that manufactures need to perform series analyses, such as in vitro genotoxicity, absorption, distribution, metabolism, and excretion tests, before their products are made available in the market.

Alternatively, the United States Food and Drug Administration (USFDA) has also published a document (in 2014) presenting guidelines to food container manufacturers to investigate the toxicity of fabricated food containers functionalized with nanomaterials. Because the use of AgNPs in plastic food containers has not been tested using USFDA standardization, the United States Environmental Protection Agency (USEPA) has prohibited the availability of plastic food containers with AgNPs in the market.

In Asia, the Korea Ministry of Food and Drug Safety (MFDS) has carried out several projects regarding safety analysis of nanomaterials in food and food packaging technology. Hence, the government is planning to establish guidelines and safety regulations regarding nanosafety in food and food packaging within a few years.[35] Although the Nanotechnology Directorate under the Malaysian government has conducted several projects on the application of nanotechnology in agriculture and food sectors, there are currently no specific regulations or guidelines for the risk assessment of nanotechnology in these sectors.[34] In addition, no safety assessments or regulations are also found for the application of nanomaterials in the food and agriculture sectors in Indonesia.

3 | CURRENT METHODS AND MATERIALS

Currently, several polymers have been explored in the food packaging industry because they demonstrate excellent durability. They include hydroxypropyl methyl cellulose (HPMC), polyvinyl alcohol (PVA), polyvinyl chloride (PVC), polyethylene (PE), polylactic acid (PLA), and poly (3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) as listed in Table 1.[36–66] Several methods, such as solvent-casting and molding methods, can be used for the preparation of films attached with AgNPs. Among these, the

TABLE 1 Packaging materials with AgNPs applied in food packaging technologies

Food packaging material	Main finding	Reference
PLA biocomposite films with nanoclay and AgNPs	The films were highly transparent with enhanced water barrier and strong biocidal properties	[36]
AgNPs-loaded CS lactate films	The composite films had antimicrobial properties against *Escherichia coli*	[37]
AgNPs-loaded CS-starch based films	The addition of AgNPs led to a slight improvement of the tensile and oxygen gas barrier properties and antimicrobial activity against *E. coli*, *Staphylococcus aureus*, and *Bacillus cereus*.	[38]
HPMC with AgNPs	Tensile strength of HPMC/AgNPs films can be improved (51.0 ± 0.9 MPa) compared to neat HPMC films (28.3 ± 1.0 MPa). Water vapor permeability of HPMC/AgNPs films can be decreased. The improved films had antibacterial properties against *E. coli* and *S. aureus*	[39]
Films based on PLA with nanocrystalline cellulose (CNC) and AgNPs	The composite films had antibacterial activity against *S. aureus* and *E. coli*	[40]
LDPE films with AgNPs	The nanocomposite of silver/LDPE showed high antimicrobial and bactericidal activity against *S. aureus*	[41]
PLA-nano-biocomposites with AgNPs	The combination of cellulose and AgNPs can increase the barrier properties	[42]
Agar/AgNP films	The composite films showed an improvement of water vapor, gas barrier, and mechanical properties. The composite films with higher concentration of AgNPs exhibited antibacterial activity against foodborne Gram-positive (*Listeria monocytogenes*) and Gram-negative (*E. coli* O157: H7) bacteria	[43]
PLA nano-biocomposite films with cellulose and AgNPs	The combination of cellulose and AgNPs increased the barrier properties	[44]
HPMC nanocomposites with bacterial cellulose nanocrystals and AgNPs	AgNPs formed complexes with the hydroxyl groups of both HPMC and BCNC and made the nanocomposite less hydrophilic	[45]
Gelatin-based active nanocomposite films with AgNPs and nanoclay	The nanocomposite films, especially for gelatin/AgNPs/clay, exhibited strong antibacterial activity against foodborne pathogens	[46]
Gelatin/AgNPs nanocomposite films	The gelatin/AgNPs composite films had strong antimicrobial activity against both Gram-positive and Gram-negative foodborne pathogenic bacteria	[47]
Pullulan films with essential oils and AgNPs	Edible films made from pullulan incorporated with essential oils and AgNPs can maintain the quality of processed meat and poultry products	[48]
LDPE nanocomposite films with AgNPs	The silver/LDPE nanocomposite films showed high antimicrobial activity	[49]
Carboxymethyl cellulose films with AgNPs	The carboxymethylcellulose film embedded with AgNPs showed the best antimicrobial effect against gram-positive (*Enterococcus faecalis*) and gram-negative (*E. coli*) bacteria	[50]
CS nanocomposite films with AgNPs and AuNPs	The proposed films had antibacterial properties against Gram-positive (*S. aureus*) and Gram-negative bacteria (*Pseudomonas aerugenosa*) and antifungal properties against *Aspergillus niger* and yeast (*Candida albicans*)	[51]
Nanostructured starch-based films with AgNPs	Barrier properties of the nanostructured starch films were improved, and the films presented antimicrobial activity against *S. aureus*, *E. coli*, and *C. albicans*	[52]
Agar/AgNPs nanocomposite films	Agar/AgNPs composite films showed strong antimicrobial activity against *E. coli* and *L. monocytogenes*	[53]
Starch-PVA-based films with AgNPs	Starch-PVA-based films embedded with AgNPs exhibited remarkable antibacterial activity against *Listeria innocua* and *E. coli* and antifungal activity against *A. niger* and *Penicillium expansum*	[54]
Agar and banana (A/B) composite films reinforced with AgNPs (A/B/AgNPs)	The A/B/AgNPs composite film exhibited distinctive antimicrobial activity against foodborne pathogenic bacteria, *E. coli* and *L. monocytogenes*, with stronger antibacterial activity against Gram-negative bacteria than Gram-positive bacteria	[55]
PLA/functionalized cellulose nanocrystals formates (CNFs) films with AgNPs	The PLA and PLA/CNF films have no inhibition zone against *E.* and *S. aureus*, but the inhibition zones for *E.* coli and *S. aureus* were observed for PLA/CNF with AgNPs	[56]

(Continues)

TABLE 1 (Continued)

Food packaging material	Main finding	Reference
Gelatin film reinforced with Ag-cu NPs exhibited strong antimicrobial activity	Gelatin film reinforced with Ag-Cu NPs exhibited strong antimicrobial activity and could be used as active packaging material	[57]
CS/gelatin composite packaging film with AgNPs	Mechanical properties and water vapor transmission rate of CS-gelatin films can be improved due to addition of AgNPs, while optical and oxygen permeability properties remained unchanged. These films had the ability to inhibit growth of *E. coli*, *Pseudomonas aeruginosa*, and *B. cereus*	[58]
Nanocomposite films based on CS-PVA and AgNPs	CS-AgNPs have no cytotoxicity effect, which suggests the possibility of CS-AgNPs use in food packaging applications	[59]
AgNPs-silk/PVA film	The prepared AgNPs–silk/PVA film showed excellent mechanical performance and stability, as well as good antibacterial activity against both Gram-negative and Gram-positive bacteria	[60]
PLA/ZnO:Cu/Ag bionanocomposite films	PLA/ZnO:Cu/Ag bionanocomposites were promising materials for food packaging application	[61]
PVC films with AgNPs	The composites with AgNPs exhibited better antimicrobial and antifungal activities compared to without AgNPs	[62]
LDPE/AgNPs films	The proposed films were effective to inhibit *E. coli*	[63]
PVC-based films with AgNPs	An improvement in the mechanical strength, antibacterial, and antioxidant properties of the films was observed	[64]
PE films incorporated with AgNPs	The improved films exhibited antimicrobial properties against all tested microorganisms	[65]
CS/AgNPs/purple corn extract (PCE) films	CS/AgNPs/purple corn extract (PCE) films showed antioxidant and antimicrobial properties	[66]
PHBV films with AgNPs	The addition of AgNPs can lead to a significant reduction in the water vapor and oxygen permeability of the films the neat matrix. Moreover, the developed nanocomposites films showed a strong and prolonged antibacterial activity against all tested bacteria	[67]

solvent-casting method is the most popular approach because it is relatively easy to prepare. Illustrations for the fabrication of films with AgNPs can be seen in Figure 1.[12,68–71] In general, the process is as follows. A solution containing AgNPs and film matrix was prepared and is then heated combined with stirring. Next, the solution was poured into a leveled cast. For instance, a leveled glass plate (24 cm × 30 cm) or petri dish (11 cm × 11 cm) was used.[12,68,70] This was then followed by drying the poured solution before further analysis. The drying process can be carried out at room temperature either for 24 or 48 hr as proposed in the previous studies.[12,68,70] Preparation and materials for the fabrication of food packaging with AgNPs are comprehensively presented in the following discussion.

Enhancement of nanocomposites of chitosan (CS)/gelatin (GL) with AgNPs for food packaging application was prepared using the solution-casting method.[72] The prepared film was tested for the extension of the shelf life of red grapes. The study observed that the fruits wrapped with plastic film and CS-GL composite film cannot maintain their quality. The wrapped grapes showed obvious mildew appearance, containing several moldy spots, and sticky juice also leaked to the surface. For a comparison, the grapes wrapped using the CS-GL composite film with AgNPs were still fresh without putridity, and the fruit surface maintained smoothness without any leakage of the juice.

PVC containing AgNPs was prepared for food packaging using the solvent-casting method.[62] The composite films were evaluated for the extension of the shelf life of bread samples. The study found that the bread samples packed using the proposed films with AgNPs (1%) showed a total absence of microorganisms in the bread surface compared to the other evaluated films. Alternatively, furcellaran–gelatin films with AgNPs prepared using the casting method for extension of mini kiwi shelf life was investigated.[73] The results from storage test indicated that the modified films with AgNPs were useful for the improvement of mini kiwi shelf life. AgNPs/PVA/bacterial nanocellulose (AgNPs/PVA/BNC) films have been an option for food packaging.[74] Their study observed that the films were able to inhibit the growth of bacteria *Escherichia coli* on raw beef. Interestingly,

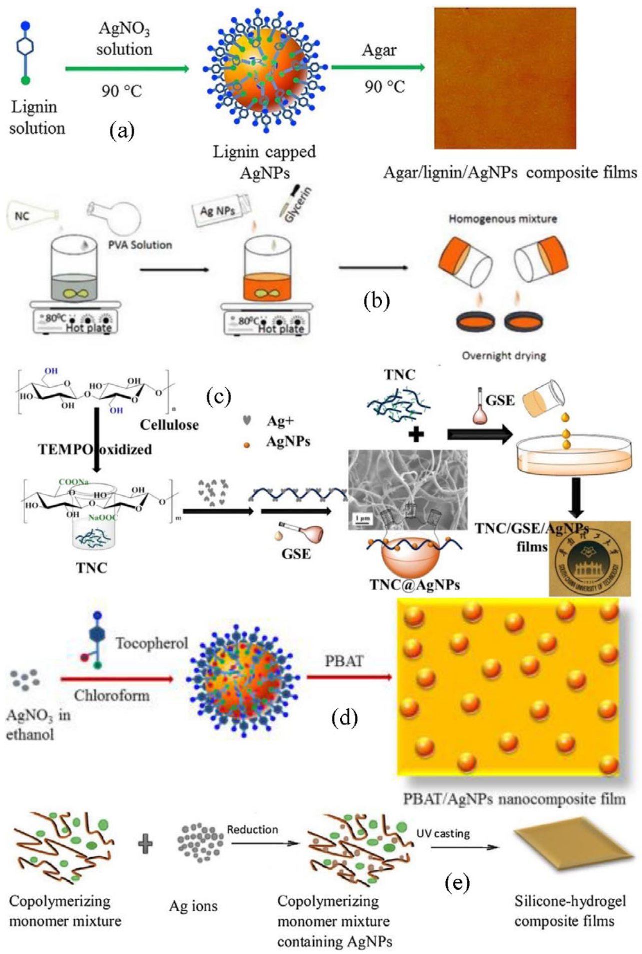

FIGURE 1 Schematic diagram for the preparation of (a) agar/lignin composite films with AgNPs,[68] (b) PVA/nanocellulose/AgNPs nanocomposite films,[69] (c) TEMPO-oxidized nanocellulose with grape seed extract and AgNPs (TNC/GSE/AgNPs),[70] (d) PBAT/silver nanoparticles composite films,[12] and (e) silicone–hydrogel composite films (SiHCFs) with AgNPs.[71] All figures are adapted with permission

AgNPs/PVA/BNC film prepared via the ultraviolet (UV) method showed higher antimicrobial activity than the reduction method. The hyperbranched polyamide-amine/AgNPs composite (AgNPs@HPAMAM) film was proposed for antibacterial food packaging.[75] The proposed composite film had the capacity as a food packing material for the storage of cherry tomatoes. It was also reported that the cherry tomatoes wrapped by AgNPs@HPAMAM-embedded cellulose films still maintained their freshness across 9 days.

By using a different polymer, PE films coated with CS incorporated with liposomes (Lip)-Laurel essential oil (LEO)-AgNPs (PC-Lip/LEO/AgNPs) were prepared for pork preservation.[76] The conducted storage study indicated that the films could keep the quality of pork at 4°C and extend the storage period to up to 15 days. PVA/AgNPs/montmorillonite (PAGM) film was used for the preservation of chicken sausage samples.[77] The PAGM exhibited the capacity to inhibit bacterial growth on the chicken sausage samples. As a comparison, the chicken sausage samples unwrapped with PAGM showed more bacterial growth. Ecoflex/silica-carbon-AgNPs nanocomposite film fabricated via the 3D printing technique was applied for food packaging.[78] It was suggested that the proposed film was powerful for food packaging application.

CS incorporated with AgNPs and 2% laponite (CL2/LAP@AgNPs) film was investigated for the storage of litchis.[79] The litchis wrapped with CL2/LAP@AgNPs films still maintained their freshness for up to 7 days when stored at a constant temperature of 25°C and a humidity chamber of 75%. In contrast, when the fruit samples were not wrapped with the proposed film, they decayed and grew mycete. Another study proposed films produced from PE, AgNPs, clay, and titanium dioxide nanoparticles for the preservation of fresh chicken.[80] This study found that the films were capable of inhibiting *Staphylococcus aureus* (PTCC 1889) and *E. coli* (PTCC 1399).

PE film functionalized with AgNPs was also evaluated and was found to have the capacity to maintain the quality of fresh bottom mushrooms.[81] Alternatively, films of low-density polyethylene (LDPE) with AgNPs were fabricated to maintain the quality of carrots.[82] The study found that the addition of AgNPs did not affect the pH and firmness values of packaged carrots. Interestingly, no AgNPs migration was observed in the packaged carrots.

The molding method can also be used for the preparation of films with AgNPs. For instance, the development of bioactive LDPE films blended with cinnamon essential oil (CEO) and Ag-CuNPs was prepared using the compression molding method.[83] The proposed food packaging has the capacity to completely inhibit *Salmonella typhimurium* and *Campylobacter jejuni* present in the chicken samples for 21 days.

As an alternative polymer, poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) films containing AgNPs were inspected as active food packaging material.[67] The films were fabricated through the melt blending process with subsequent compression molding into films using a hot-plate hydraulic press. Mechanical properties, water vapor permeability (WVP), oxygen transmission rate (OTR) measurements, and antimicrobial activities were inspected. The water vapor and oxygen permeability of the films can be successfully reduced compared to the neat matrix. The proposed films had the capacity to inhibit all tested bacteria.

Nanofibers (PVA–AgNPs) were prepared via the electrospinning method.[84] It can be achieved by incorporating the synthesized AgNPs for the preservation of lemon and strawberry. The study observed the yeast growth in the unwrapped lemon and strawberry. In contrast, the nanofiber with AgNPs had the capacity to maintain their quality for up to 10 days of storage. Cellulosic paper sheets impregnated with AgNPs can also be used for the preparation of packaging materials.[85] The proposed packaging was tested for the extension of shelf life of fresh-cut cabbage and tomato. It is noted from the study that the shelf life of the packaged vegetables can be improved compared to those without AgNPs.

Moreover, fabrication of films with AgNPs for packaging of some foods is summarized in Table 2.[48,62,72–92] It is noted that the proposed films have been tested for the packaging some fruits such as kiwi, litchis, lemon, strawberry, and grape. As a comprehensive overview, their performance is shown in Figure 2.[72,79,84] It is noted that the presented figures are obtained from the previous works and adapted with permission. In addition, they have also been tested for some meat such as pork, beef, and chicken.

4 | IMPROVED PROPERTIES

4.1 | Water vapor permeability

Incorporation of AgNPs into the HPMC composite matrix for the fabrication of food packaging films can decrease WVP up to 0.48 g mm/K Pa h m^2 compared to without AgNPs (0.80 g mm/K Pa h m^2).[39] Another study also confirmed that the addition of AgNPs into agar composite films can decrease the WVP up to 1.47×10^{-9} g m/m^2 Pa s compared to control composites without AgNPs, which is 1.97×10^{-9} g m/m^2 Pa s.[43] Agar/AgNPs composite films were found to have the WVP of 1.38×10^{-9} g m/m^2 Pa s, which is lower compared to without AgNPs, which is 1.52×10^{-9} g m/m^2 Pa s.[93]

Another study also reported similar findings. For instance, gelatin-based nanocomposite films incorporated with AgNPs can decrease the WVP from 3.43 to 2.89×10^{-9} g m/m^2 Pa s.[46] Incorporation of AgNPs into gelatin nanocomposite films can decrease the WVP from 3.02 to 2.92×10^{-9} g m/m^2 Pa s.[47] Decrease in the WVP of composite films prepared from banana powder and AgNPs up to 1.36×10^{-9} g m/m^2 Pa s can also be achieved compared to without the addition of AgNPs (2.32×10^{-9} g m/m^2 Pa s).[94]

This is also consistent with another study which found that the WVP of composite films prepared from tragacanth, HPMC, and beeswax reinforced with AgNPs gradually declined to 2.16×10^{-13} g m/m^2 Pa s, which was substantially lower compared to values calculated for composites control (4.57×10^{-13} g m/m^2 Pa s).[95] CS nanocomposite films incorporated with AgNPs were observed to decrease the WVP property ranging from 0.210 to 0.178 g mm/kPa h m^2.[96]

In general, a decrease in the WVP of composites films with AgNPs can be correlated to the distribution of AgNPs as a discontinuous phase in the polymer matrix. This can affect the diffusion of water molecules through the films.[39,47] In addition, increase in tortuosity of the polymeric matrix is also a major factor affecting the improved WVP property of composite films functionalized with AgNPs.[95]

4.2 | Antibacterial properties

One of the most important properties of food packaging modified with AgNPs is the improvement of antibacterial capability. The properties are useful for maintaining food quality and freshness during the storage. AgNPs have been the most significant antibacterial agent reported in literature. Therefore, several studies have tested the effectiveness of AgNPs attached in food packaging to prevent food pathogens, mainly bacteria. A study on the investigation of antimicrobial activity of agar/AgNPs films was inspected and compared with neat agar films against *Listeria monocytogenes* and *E. coli*.[43] It was found that an increase in Ag content can improve the antibacterial properties of the proposed films against all tested bacteria.

Gelatin nanocomposite films with AgNPs were tested against food-borne pathogenic microorganisms, such as *E. coli*, *L. monocytogenes*, *S. typhimurium*, *S. aureus*, and *Bacillus cereus*.[47] The study found that *Salmonella typhimurium* was observed to be more susceptible to AgNPs in the gelatin films, followed by *B. cereus* and *S. aureus*. Interestingly, the study also found that *E. coli* and *L. monocytogenes* were less susceptible to the AgNPs in the proposed films.

Antibacterial activities of nanocomposite films prepared from CS-PVA and AgNPs were evaluated against *E. coli*, *Salmonella typhimurium*, *Salmonella enterica*, *Pseudomonas aeruginosa*, *Klebsiella pneumonia*, *B. cereus*, *S. aureus*, and *Micrococcus luteus*.[59] The study demonstrated that the modified composite films showed the largest inhibition zone against *B. cereus* and *P. aeruginosa* for the Gram-positive and five Gram-negative bacteria, respectively.

As an alternative study, composite films fabricated from tragacanth, HPMC, and beeswax reinforced with AgNPs were found to have antibacterial properties against Gram-positive and Gram-negative bacteria.[95] The study observed that the highest percentage of AgNPs

TABLE 2 Composite films with AgNPs tested for the preservation of several foods

Materials	Food test	Testing duration	Reference
Pullulan films with essential oils and AgNPs	Raw turkey breast, raw beef (top round), and ready-to-eat turkey breast	21 days	[48]
PVC with AgNPs	Breads	15 days	[62]
CS and gelatin (GL) with AgNPs (CS-GL-AGNPs)	Red grape	25 days	[72]
Furcellaran–gelatin films with AgNPs	Mini kiwi	8 days	[73]
AgNPs/PVA/bacterial nanocellulose (AgNPs/PVA/BNC) films	Beef	14 days	[74]
Hyperbranched polyamide-amine/AgNPs composite film	Cherry tomatoes	9 days	[75]
PE films coated with CS incorporated with liposomes (lip)-Laurel essential oil (LEO)-AgNPs (PC-lip/LEO/AgNPs)	Pork	15 days	[76]
PVA/AgNPs/montmorillonite (PAGM) film	Chicken sausage	4 days	[77]
Ecoflex/silica-carbon-AgNPs nanocomposite film	Chicken thigh	28 days	[78]
CL2/LAP@AgNPs film	Litchis	7 days	[79]
PE with Ag, clay, and titanium dioxide nanoparticles	Chicken	5 days	[80]
PE film functionalized with AgNPs	Mushrooms	21 days	[81]
Films of LDPE with AgNPs	Carrots	10 days	[82]
LLDPE films mixed with blended with cinnamon essential oil (CEO) and Ag-CuNPs	Chicken	21 days	[83]
Nanofibers PVA/AgNPs	*Citrus limon* (lemon) and *Fragaria ananassa* (strawberry)	10 days	[84]
Cellulosic paper sheets impregnated with AgNPs	Cabbage and tomato	7 days	[85]
Sodium alginate films with AgNPs	Carrot and pear	10 days	[86]
Agar/alginate/collagen (A/A/C) with AgNPs	Potatoes	10 days	[87]
PVC nanocomposite films with AgNPs	Chicken breast fillets	9 days	[88]
PE polymer composite films with AgNPs	Iranian saffron	180 days	[89]
LDPE films with AgNPs	Pistachio	8 days	[90]
Commercial films coated with AgNPs	Turkey meat	12 days	[91]
PE-AgNPs films	Walnuts, hazelnuts, almonds, and pistachios	365 days	[92]

added into the polymer matrix showed the greater inhibition zone diameters against all tested bacteria. In general, the antibacterial capability of the films with AgNPs was stronger to Gram-negative bacteria compared to the Gram-positive bacteria.

The different antimicrobial activity of AgNPs against Gram-positive and Gram-negative bacteria can be explained from the point of view of the structure and thickness of the bacteria cell wall.[53,55] It is established that Gram-positive bacteria have multiple layers and thicker peptidoglycan ranging from 20 to 80 nm compared to the Gram-negative bacteria, which is only in the range of 7–8 nm. This makes the penetration of AgNPs into the cytoplasmic membrane difficult. By providing a thin peptidoglycan layer, AgNPs penetrate the bacteria easily and cause death.[47]

The mechanism of antibacterial properties of AgNPs has been well established.[97–104] In general, the possible mechanisms are as follows. It was widely acceptable that the positive charge of AgNPs may interact with phosphorus or sulfur, having a negative charge present in proteins and nucleic acids by electrostatic force.[105] This interaction can cause the deformation of bacterial cell walls and membranes, possibly leading to cell death. Alternatively, AgNPs have the capacity to produce reactive oxygen species (ROS) and free-radical species such as hydrogen peroxide, superoxide anion, hydroxyl radical, hypochlorous acid, and singlet oxygen, which can enhance the

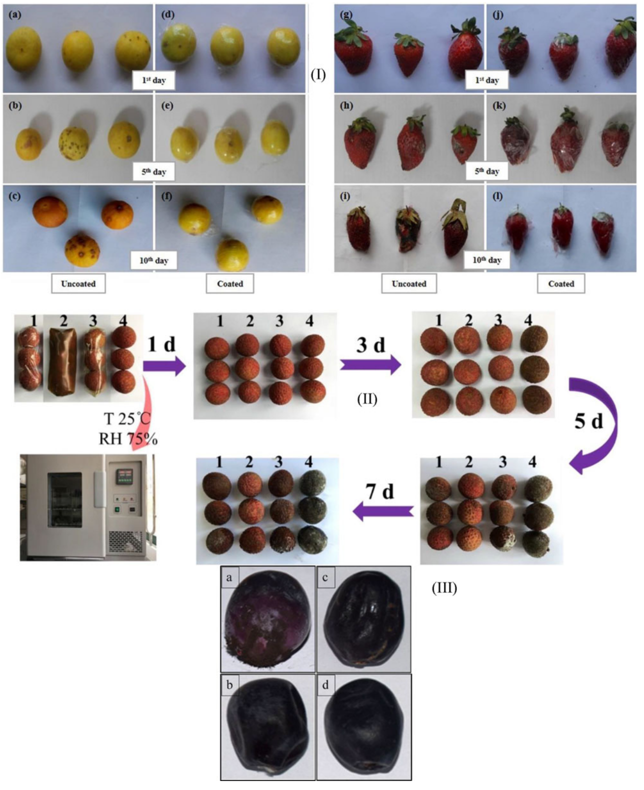

FIGURE 2 Storage study of (I) lemons and strawberries using AgPVA nanofibers as packaging. It is noted that (a–c) and (d–f) refer to uncoated and coated lemons, respectively, and (g–i) and (j–l) refer to uncoated and coated strawberries, respectively.[84] Storage study of (II) litchis using CS-based films as packaging. From left to right, (1) commercial PE cling wrap; (2) CL2/LAP@AgNPs films; (3) pure CS films; and (4) without wrapping tested in constant temperature and humidity chamber of 25°C and 75%, respectively.[79] Storage study of (III) red grapes wrapped with (a) plastic, (b) CS-GL film, (c) CS-GL-AgNPs (0.05%) film, and (d) CS-GL-AgNPs (0.1%) film after 14 days storage at 37°C.[72] All figures are adapted with permission

oxidative stress in bacterial cells.[106] In addition, this can enhance the permeability properties of the membrane of bacteria and can trigger cell death.

4.3 | Mechanical properties

Several composite properties, such as tensile strength (TS) and elongation at break (EB), are important in food packaging application. These properties show behaviors related to their ability to maintain the integrity of food packaging against various environmental stress factors. The TS of composite films prepared from gelatin and AgNPs was reduced up to 46.12 ± 0.97 MPa compared to neat films, which is 56.55 ± 1.75 MPa, depending on the nanoparticle filler concentration.[107] However, the addition of AgNPs into the composite films can improve the EB up to 47.23 ± 3.68% compared to neat film (37.99 ± 4.02%). The TS of the neat agar film was 45.8 ± 2.9 MPa, which was decreased to 35.1 ± 2.0 MPa after adding AgNPs.[53] However, the EB of the neat film was 13.9 ± 3.4%, which can be improved after adding AgNPs up to 21.9 ± 1.7%. Decrease in the TS of composites after adding AgNPs can be explained as follows. This is possibly because the addition of AgNPs in the composite matrix can decrease the continuity and cohesion of the polymer networks compared to the control composite films.[107]

In another study, contradictory findings were found compared to the above-mentioned studies. The addition of AgNPs into pectin films can improve their TS of 25.2 ± 3.3 MPa compared to neat pectin film, which is only 23.3 ± 4.4 MPa.[108] However, their EB was reduced to 19.0 ± 3.6% compared to neat pectin film (21.6 ± 6.9%). An alternative study also observed similar findings when AgNPs were incorporated into poly(butylene adipate-co-terephthalate) (PBAT) film.[12] The TS of the neat PBAT film was 4.1 ± 1.6 MPa, which can be improved after adding AgNPs, which is 7.8 ± 0.8 MPa. In addition, EB of the neat film (126.7 ± 1.6%) was higher compared to that with AgNPs added (121.6 ± 3.6%).

Another study also found that the TS of the carrageenan/clay nanocomposite films filled with AgNPs can be improved (64.6 ± 20.3–68.9 ± 11.7) compared to without AgNPs (56.5 ± 20.1 MPa).[109] A similar finding was also reported when the CS nanocomposite films were incorporated with AgNPs by improving the TS from 21.07 ± 1.64 (neat CS) to 24.35 ± 1.96 MPa (modified with AgNPs).[110] Neat agar and agar/lignin composite films had TS of 40.3 ± 4.0 and 44.1 ± 3.6 MPa, respectively, which can be improved after adding AgNPs up to 49.7 ± 3.3 MPa, depending on the AgNPs concentration, while EB was decreased to 16.2 ± 3.4% from 19.4 ± 4.2% (neat agar).[68] An increase in the mechanical strength of the nanocomposite films is partly due to the physical attraction between the filler and polymer matrix.[109] A previous study reported that the van der Waals interaction between the hydroxyl groups of a biopolymer and the positive charge of AgNPs was the main force in nanocomposite films.[111] In addition, it may also be explained that the increase of contact area between composite matrix and nanoparticles filler can enhance the mechanical strength of nanocomposite films.[109]

The above studies exhibited that, in some cases, the addition of AgNPs into composite matrixes can improve their TS. However, other studies reported contradictory findings. It is noted that the mechanical properties of composite films with AgNPs depend highly on the type of matrix, composition of composites, and the preparation method. In addition, physiochemical parameters during the experiment can also affect the mechanical properties.

Commonly, a composite with higher TS is recommended for a variety of packaging products. When they are used in the packaging of heavier products, packaging with higher TS is preferred to ensure a better seal. In addition, a stretch composite film with higher TS has the potential to reduce shipping damages and increase load stabilization.

4.4 | Freshness indicators

Freshness indicators can be evaluated using several signs, such as a reduction of pH or an increase in the carbon dioxide, or it can be analyzed through the formation of biogenic amines. Microbial growth within the package can be used as a sign about the microbial quality of the product. Alternatively, carbon dioxide produced in meat products can also be used as a sign of food spoilage. In general, common indicators of quality loss of some fresh-cut foods have been discussed in a previous report.[112] For instance, browning, decaying, juice leakage, and softening are common indicators used for describing the quality loss of some fresh-cut fruits.

It is noted that pH values can be used as an indicator to evaluate the quality of foods. It indicates the effect of microbial secretion–proteolytic enzymes or it can also be through the decomposition of muscle protein into alkaline compounds.[113] Evaluation of the freshness of pork meat was performed by investigating pH values over a period (up to 21 days) after the meat was wrapped with PC-Lip/LEO/AgNPs as the improved packaging, as shown in Figure 3.[76] At 15 days, sensory analysis of pork meat stored at 4°C exhibited pH of 6.5 and 7.11 using the improved packaging and control, respectively. It is interesting to note that the pork wrapped with the improved packaging was under a limit regulated by the Chinese hygienic standard for fresh meat of livestock, which is <6.7.

In the case of meat, total volatile basic nitrogen (TVB-N) values were used as one of the important indicators to indicate the degree of decomposition of the meat.[114] For instance, pork meat wrapped with PC-Lip/LEO/AgNPs was also inspected in terms of TVB-N values.[76] By following the allowable limit regulated by the Chinese food safety standard for fresh pork (<15 mg/100 g, GB 2707–2016), their study reported that the wrapped pork can be of good quality for up to 15 days.

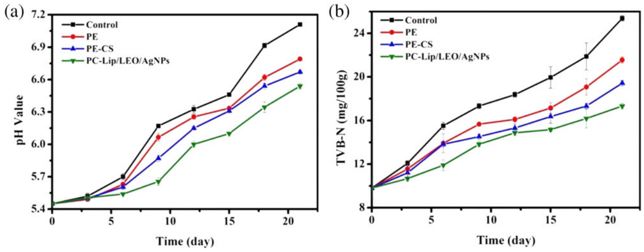

FIGURE 3 Physicochemical properties in terms of (a) pH value and (b) total volatile basic nitrogen (TVB-N) of pork meat wrapped with PC-Lip/LEO/AgNPs films in comparison with other packagings.[76] All figures are adapted with permission

Vitamin C content can also be used as a quality indicator for fruits. Vitamin C and titratable acid content of grapes stored in AgNPs-containing PVA film were compared with that of unpacked grapes and grapes stored in neat PVA films.[115] The study found that the vitamin C content in the fresh fruit (before starting packaging) was 24.96 mg/100 g. In addition, the vitamin C content for the fruit was 18.61, 19.50, and 21.55 mg/100 g for the unpacked, packed with neat film, and packed with AgNPs-film, respectively. It is noted that the highest Vitamin C content was found for fruits packed with AgNPs-PVA film compared to others.

Nutritional status of vegetables in terms of total antioxidant, protein, phenol, and flavonoid contents wrapped with food packaging with AgNPs was also evaluated.[85] The study observed that there was no significant change in the nutritional status of the vegetables after storage for 7 days, but in the food packaging without AgNPs, a significant loss was recorded. A similar finding was also observed for the moisture content of the vegetables, which was found to have no significant loss when stored in food packaging with AgNPs.

It is noted that the freshness indicators are commonly used to make the quality of a packaged food visible. These indicators show the changes in food quality because of microbial growth or chemical changes of food products. The above discussions showed that the incorporation of AgNPs into food packaging can improve or maintain the quality of foods. It was hypothesized that the release of AgNPs from composite films has antibacterial activity.[76] Therefore, it can act by blocking the entry of oxygen and inhibiting bacterial growth.

5 | MIGRATION OF AGNPS

5.1 | Suitable methods

Currently, several methods, such as chromatographic techniques, filtration, and inductively coupled plasma-mass spectrometry (ICP-MS), have been used for the detection of AgNPs in environmental samples. Each technique shows its own advantages and limitations. A combination of these methods is commonly proposed as the information provided by each method is usually complementary. Among these techniques, ICP-MS has the potential for the detection and analysis of metals by having higher sensitivity. However, only using ICP-MS, it is hard to analyze the differentiation of elemental metal and ions.[116] In the current case, AgNPs migration from food containers can be possible through either silver ions and AgNPs or more complex forms.

Currently, single-particle (sp) ICP-MS (sp-ICP-MS) is an advanced technique that has the capacity for the quantification of elemental metal and ions. However, the technique has limitations, such as inability to analyze complex samples, particularly for nanoparticles having broad size distribution.[116,117] It has been well known that AgNPs can be transformed into more complex forms (ions +1 or + 2) depending on the physiochemical conditions. Hence, an inaccurate estimation can probably be obtained using this technique.

5.2 | Concentration

The investigation of AgNPs migration from food packaging generally follows a regulation by the EU (Commission Reg No: 10/2011/EU). As an example, the procedure for the investigation of AgNPs migration from composite films was proposed as shown in Figure 4.[78] Migration of silver from a food container made from PE nanocomposites tested to boneless chicken breasts was comprehensively evaluated.[118] Their study observed that the migration of AgNPs ranged from 3 to 5 μg/dm^2. Migration of AgNPs from various commercial plastic food containers was also investigated using ICP-MS.[119] It was observed that 3.0–3.4 μg/dm^2 of AgNPs can migrate from the food containers. Total Ag migration from Fresherlonger bags in stimulants of ethanol (EtOH) 50% vol/vol and acetic acid (HAc) 3% vol/vol were 0.2 and 0.4 μg/dm^2, respectively. For the kinetic go green (a brand of the food container) in stimulants of EtOH and HAc 3% vol/vol, the corresponding values were 0.9 and 3.1 μg/dm^2, respectively. For the Oso fresh in stimulants of EtOH and HAc 3% vol/vol, they were 0.7 and 1.0 μg/dm^2, respectively.

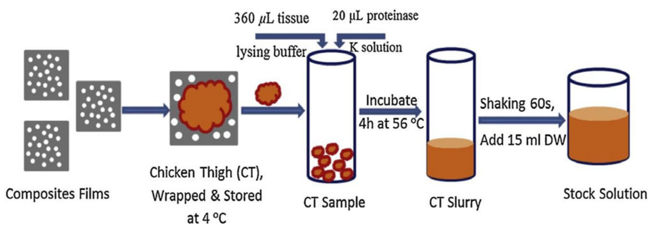

FIGURE 4 Schematic diagram for the investigation of AgNPs migration from composite films.[78] The figure is adapted with permission

Two plastic food containers, a baby feeding bottle and a food box, made of polycarbonate and PP, respectively, were investigated in terms of AgNPs migration.[120] Migration of AgNPs from the baby bottle was up to 0.01, 0.06, and 0.003 μg/dm^2 in the stimulants of water, 3% acetic acid, and 10% ethanol, respectively. In addition, migration of AgNPs from the baby bottle was up to 0.4, 1.8, and 0.42 μg/dm^2 in the stimulants of water, 3% acetic acid, and 10% ethanol, respectively. Migration of AgNPs from AgNPs/PVA/BNC films via a reduction and UV method was inspected.[74] The study confirmed that almost no silver release can be found in the film prepared by the reduction method. However, about 28.4 μg/dm^2 Ag release can be detected in the film prepared by UV method.

Several studies have proven that AgNPs have different toxicological properties depending on their size, shape, and environmental conditions. The release of AgNPs from food packaging into foods could have implications for human health. A study was conducted to evaluate the toxicity of cellulose nanofibril (CNF)/AgNPs composite as an active food packaging on human colon (FHC; CRL-1831) cells.[121] The study employed MTT and WST-8 cell proliferation assays for the determination of the cell viability of colon cells. The cells were tested at concentrations ranging from 50 to 1,000 μg/ml of CNF/AgNPs composites. Interestingly, the study found that no significant decrease in the number of viable cells was observed. In general, the study showed that the proposed packaging did not show significant toxic effect on human colon cells within 24 hr.

For further clarification, the study assessed the cell morphology and transport of AgNPs using transmission electron microscopy (TEM). It was found that AgNPs can be observed in the endosomes of the cells, which proved the uptake of AgNPs released from the composites into the cells. The penetration of AgNPs into cell membranes can be explained via the endosomal mechanisms.[122] In general, the study proved that the AgNPs were absorbed by the human colon cells but without significant toxicity effects. No significant toxicity effects of AgNPs to the human colon cells was probably due to the limited treatment time (24 hr). A previous report suggested that the toxicity of cellulose/AgNP composites can be more significant after 72 hr.[123] Hence, evaluation of CNF/AgNPs composites toxicity to the human colon cells with a longer treatment time (more than 72 hr) must be carried out.

Several studies have established the toxicity effects to human cells. For instance, AgNPs can be agglomerated in the cytoplasm and nuclei of human hepatoma cells and can induce intracellular oxidative stress.[124] In addition, blue-gray hyperpigmentation of the skin, commonly called agyria, can be caused by the oral exposure to high levels of AgNPs.[125] Decrease in cell viability of human lung epithelial A549 cells can also be observed after exposure to AgNPs.[126] After exposure to AgNPs, the nanoparticles were detected inside the perinuclear region of human mesenchymal stem cells associated with the endolysosomal cell compartment.[127]

5.3 | Parametric effects

Migration of AgNPs from composite films with AgNPs increased gradually with the extension of time. AgNPs can migrate from LDPE films with AgNPs via 3% acetic acid ranging from 12 to 17 μg/ml at 70°C.[128] Similar findings were also observed when distilled water and 50% ethanol were used as food simulant models, which consistently increase with the increase of time. Migration of AgNPs from nanocomposite films developed from AgNPs and an LDPE under three food simulants (3% acetic acid, 50% ethanol, and distilled water) was studied.[129] The study observed that the highest migration concentration can be found in 3% acetic acid followed by distilled water and was the least in 50% ethanol.

Temperature also affects the migration behaviors of AgNPs from composite films. In general, an increase in temperature can increase the migration of AgNPs from food packaging. For instance, concentrations of AgNPs in the distilled water were 7.81, 9.63, and 12.87 μg/ml when inspected at different temperatures of 25, 40, and 70°C, respectively.[129] Consistent findings can also be seen when 50% ethanol was used as the food simulant. Concentrations of AgNPs in the 50% ethanol ranged from 0.19 to 0.43 μg/ml at different temperatures ranging from 25 to 70°C, respectively.[129] Increase in temperature can also increase the migration of AgNPs from a nanocomposite film by 11.75, 14.72, and 17.50 μg/ml at different temperatures of 25, 40, and 70°C, respectively.[129]

Consistent findings were also reported when the migration of AgNPs from AgNPs-PE composite film was inspected in 3% (wt/vol) aqueous acetic acid or 95% (vol/vol) aqueous ethanol.[130] The study found that the maximum migration ratios for 3% (wt/vol) aqueous acetic acid ranged from 1 to 5.6% and 0.22 to 0.24% for 95% (vol/vol) aqueous ethanol, depending on the experimental temperature. In general, the migration ratio of AgNPs from AgNPs-PE composite film was higher in 3% (wt/vol) aqueous acetic compared to 95% (vol/vol) aqueous ethanol.

5.4 | Mathematical modeling

Several mathematical models have been proposed and evaluated for the description of migration of AgNPs from food packaging. For instance, migration of AgNPs from food packaging was simulated using the Williams–Landel–Ferry model.[118] The model has successfully predicted levels of AgNPs in the laboratory migration test experiment.

Migration kinetics of AgNPs from starch-PVA films embedding AgNPs were modeled using the Peleg equation.[54] Their study reported that the model was a close fit for all experimental points for all food simulants, which are simulant A (ethanol 10% (vol/vol)), simulant B (acetic acid (3% (wt/vol)), simulant C (ethanol 20% (vol/vol)), and simulant D2 (oleic acid as a vegetal oil).

Alternatively, migration behaviors of AgNPs from LDPE film were evaluated using three mathematical models, which are the Korsmeyer-Peppas, the Higuchi, and the Elovich.[131] In general, all employed models performed well with experimental data by providing the determination coefficient (R^2) exceeding .90. Moreover, the Korsmeyer-Peppas model was the best because it demonstrated R^2 exceeding .95 for all experimental data via food simulants. It is noted that the model is generally proposed for the description of drug release from composite matrix. The model is developed based on the assumption of Fickian and non-Fickian diffusion approaches.[132]

5.5 | Proposed mechanism

In general, the mechanism of AgNPs migration from food packaging can be explained as follows. First, food simulant is diffused into composite matrix. This process can cause the oxidation of AgNPs, leading to release Ag^+.[128,133] A study observed that the AgNPs were not directly exposed by simulants, but simulants must penetrate into the matrix for the dissolution of AgNPs.[131]

Thermodynamic study of migration of AgNPs from AgNPs/LDPE found that both enthalpy change (ΔH^o) and entropy change (ΔS^o) were positive.[131] This indicates that AgNPs release from the composite films cannot occur endothermically, but it can be possible entropically. Positive values of the Gibbs free energy difference (ΔG^o) demonstrated that the release of Ag from the AgNP/LDPE was not possible spontaneously. It is noted that ΔG^o in 4% acetic acid was found to be lower compared to that in distilled water.[131] In addition, activation energies' (E_a) values were 32 and 14 kJ mol^{-1} in the distilled water and 4 wt/vol% acetic acid, respectively.[131] This suggests that the E_a of AgNPs migration was higher in distilled water. Consequently, lesser energy is needed to release Ag in 4% acetic acid compared to in the distilled water.

6 | FUTURE CHALLENGES

Currently, the fresh-cut fruit industry has rapidly increased. This leads to the need for an improvement of packaging to maintain the shelf life of the food. In the future, nanotechnology advancement is needed not only to improve shelf life of the food but also to improve the quality and nutritional value of fresh-cut products. In addition, it is noted that the application of AgNPs in food packaging technologies is still in an early stage of development. Therefore, intensive study for the investigation of the potential toxicity and risk assessment of AgNPs to food products must be conducted in the future.

Migration of AgNPs from food packaging into foods is a concern as AgNPs are commonly associated with their toxicity. Application of binders may be an alternative for permanent attachment of AgNPs in food packaging. For instance, AgNP-embedded cellulose film with HPAMAM as a binder was evaluated in terms of AgNPs migration compared to without the binder.[75] The study confirmed that the proposed film can release AgNPs up to 30.9 $\pm$ 1.5%. In contrast, the migration of AgNPs from the film with HPAMAM as a binder can be significantly reduced by up to 4 times (8.2 $\pm$ 0.3%,) lower than that from without the binder.

Another challenge associated with the development of food packaging with AgNPs is cost-effectiveness. It is noted that the incorporation of AgNPs in packaging systems can increase the packaging cost. It was recommended that the total packaging cost should be 10% of the product cost.[16] Hence, a proper cost–benefit analyses is needed to implement AgNPs in food packaging. In addition, public acceptance for new nanotechnologies is probably poor as this is highly dependent on demographic and marketplace.

7 | CONCLUSIONS

This review has highlighted the current state of knowledge concerning the application of AgNPs for the enhancement of food packaging technologies. This paper has shown that the USFDA and the EFSA have established a regulation for the use of AgNPs in food packaging. Numerous polymers combined with AgNPs for food packaging technology have been explored and are found to enhance the properties (antibacterial, mechanical, and barrier properties) of packaging and improve food quality, extend shelf life, and prevent the spoilage. Although the application of AgNPs in food packaging technology has several advantages, their migration into food must be carefully considered because of their toxicity properties. The potential toxicity and risk analyses of AgNPs released from food packaging need more research to ensure their safety before being widely implemented. A study on the evaluation of economic analyses to implement AgNPs in food packaging needs to be carried out as such a study has not been found in the literature.

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