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Overall Review the Current Tend and Difficulties of Antimicrobial compounds in Composite Food Packaging Applications

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Abstract. Food waste/spoilage caused by microbial cell has recently emerged as a major food insecurity and environmental concern. Additionally, food spoilage contributes to the economic crisis and healthy problems. As a result, an active packaging system is still required to keep the food safe and to protect its quality from foreign contaminants. The purpose of this review was to summarize the current solutions and difficulties of antimicrobial compounds in composite food packaging applications. Specifically, the extrusion and antimicrobial coating methods for incorporating antimicrobial compounds into packaging systems and their optimum processing parameters for common polymer composites were revealed. The common inorganic and organic antimicrobial substances/compounds with their quantities adding to the packaging system and their antimicrobial activity (reduction, partially deactivation and completely deactivation) were presented. The difficulties in creating a package with antimicrobial properties concerning issues of migration of antimicrobial additives from the package to the food product, accumulation of antimicrobial additives in the food product, as well as their processing temperature were elaborated. Therefore, this review work contributes to open up the entire scientific knowledge on antimicrobial compounds used in polymer composite materials for food packaging application and helps to develop important results for large scale operations.

Keywords: antimicrobial compounds, microorganisms, composites, food spoilage, food packaging, challenges.

Introduction

According to an FAO report, food contamination by microorganisms is the world's bottleneck of produced food. The primary problem in food factories is food waste due to microbial contamination. Based on the FAO report, more than 1.3 million metric tons of edible human foods are wasted because of traditional ways of harvesting, storage, and transportation practices, as well as market and consumer waste globally [1,2].The migration of harmful materials and permeability of the foreign materials to the food containers are the current issue related with food contamination have grown globally. The physical, chemical, and biological methods of system can make food decay [3,4]. The fresh foods can be out of date, and drop shelf-life due to microbial spoilage and putrefaction. Hence, around 45; 35; 30 and 20% of fruit and vegetables, fish, cereals, dairy and meat products lost yearly respectively. These mostly wasted by living organisms (microorganisms). Additionally, plastic waste released worldwide exceeds 400 million tons per year and their non-sustainability and non-recyclability are the current issues. Hence, predicts the production rate of plastic waste is expected to enlarge fourfold in 2050 [5, 6]. Next to food contamination by microorganisms and negative impact of convectional food packaging materials, the consumers pertain. Excellent shelf-life and excellent properties of packaging materials are the main goal and concern of the moment food factories [7].

The combination of nano particles and polymer is known as nano-composite, and it is a promising material for food packaging. The food packaging system is categorized into three parts: primary (which coats and communicates with the food), secondary (which covers the primary packaging system), and tertiary (which is the outer covering used for bulk handling, distribution, and further storage) [8]. The best packaging system characterized by good thermal, surface, mechanical, low barrier, green, suitable optical and excellent antimicrobial properties [9,10] represented as Figure 1. In this paper only the antimicrobial materials in food application system are discussed.

The goal of this review was to reveal information about antimicrobial materials, techniques for using in packaging materials, percentile reduction of microorganism activity, and difficulties in creating antimicrobial packaging material properties.

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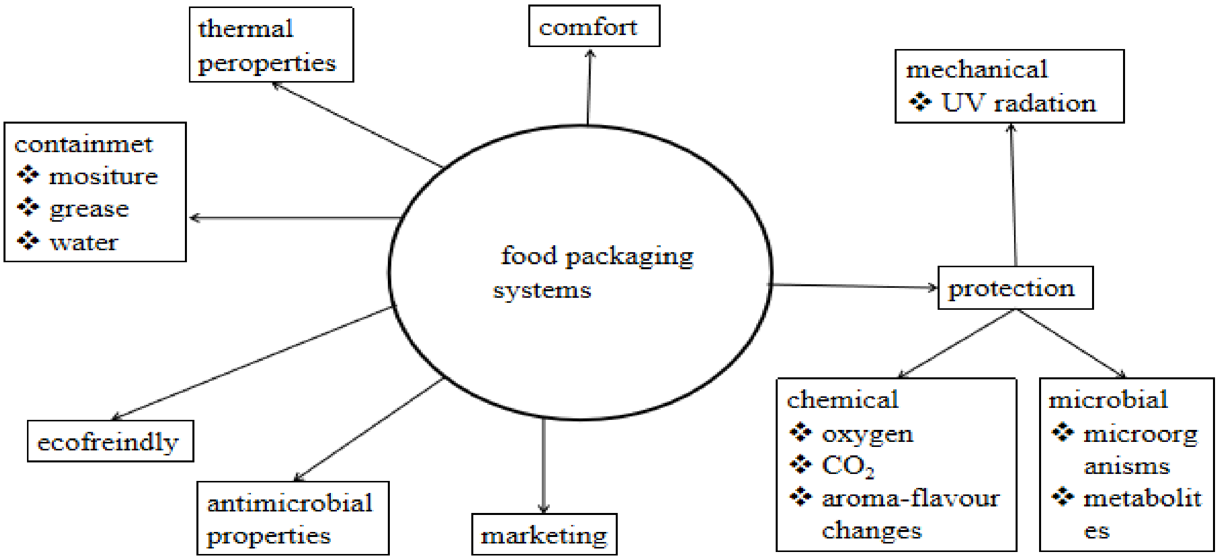

Figure 1. Over all purpose and properties of food packaging systems

2. Common Antimicrobial which is used for preparation of packaging materials

The use of antimicrobial compounds in food packaging systems is gaining popularity due to health concerns and government regulations. The common antimicrobial substance/compounds can be sorted into inorganic and organic matters according the Figure 2. The literature reported a wide range of antimicrobial agents such as nano particles of metallic elements [11], metallic oxides [12], clay [13,14], essential oils such as lemon oil, rosemary oil, sun flower oil, lemongrass, carvacrol oil, bergamot oil, metha pipertia L.oil, Mentha villosa Huds oil, eucalyptus globulus oil, cinnamon oil, and Acid compounds such as acetic acid, ascorbic acid, citric acid, lactic acid, glacial acetic acid [15], natural agents [16–18], biopolymers [19, 20], enzymes [21–23], synthetic antimicrobial agents [24]. Antimicrobial substances/compounds have been studied for their ability to inhibit microbial growth in foods, including organic/natural (essential oils, CO_2, organic acids, antibiotics, and so on) and inorganic (particularly Silver, Zinc metal/oxide nano particles) but their commercial availability remains limited [25, 26]. The silver (Ag) metal/oxide antimicrobial is used commercially as an antimicrobial agent in food packaging applications in the United States and Japan [27]. The use of silver (Ag) metal/oxide as an antimicrobial agent in food packaging solutions is expected to increase in the European Union (E.U.) used as additives in food surface.

Antimicrobial substances/compounds extracted from natural resources are drawn to the meat, bread, and pastry industries. The meat industry has a strong preference for natural extract antimicrobial agents derived from plants (cloves, ginger, rosemary, thyme, garlic, cinnamon, and so on) [28, 29] as well as natural antimicrobial compounds derived from fungal cells (nisin, pediocin and various bacteriocins). Furthermore, technological advancements in the meat industry have been used to improve organoleptic properties and packaging performance by blocking/completely inhibiting microbial activities in the food packaging system [30, 31]. The use of synthetic antimicrobial compounds in the packaging of pastry and bread [32, 33] as well as vegetables and fruits [34–36] has been reported in the literature.

Natural/inorganic antimicrobial compounds in the literature have elucidated the antimicrobial activities of common food spoiling microorganisms. The antimicrobial packaging systems have been tested for *Salmonella enterica, E. coli, S. aureus*, and other pathogens [37].As a result, the yeast and molds fungal strains have also been developed [38].

Table 1 lists the selective antimicrobial compounds used in food packaging systems. Among them, nisin coated with ally isothiocyanate can completely inhibit *Salmonella* microbial activity, while dihydroxylated coumarins coated with methanolic extract can completely inhibit *T. mentagrophytes* and *R. solani* microbial activities, respectively. Chitosan coated with Lauric acid in starch film and Ethylene in co-polymer films, respectively, reduce the microbial activity of *E. coli* and *L.monocytogenes*.

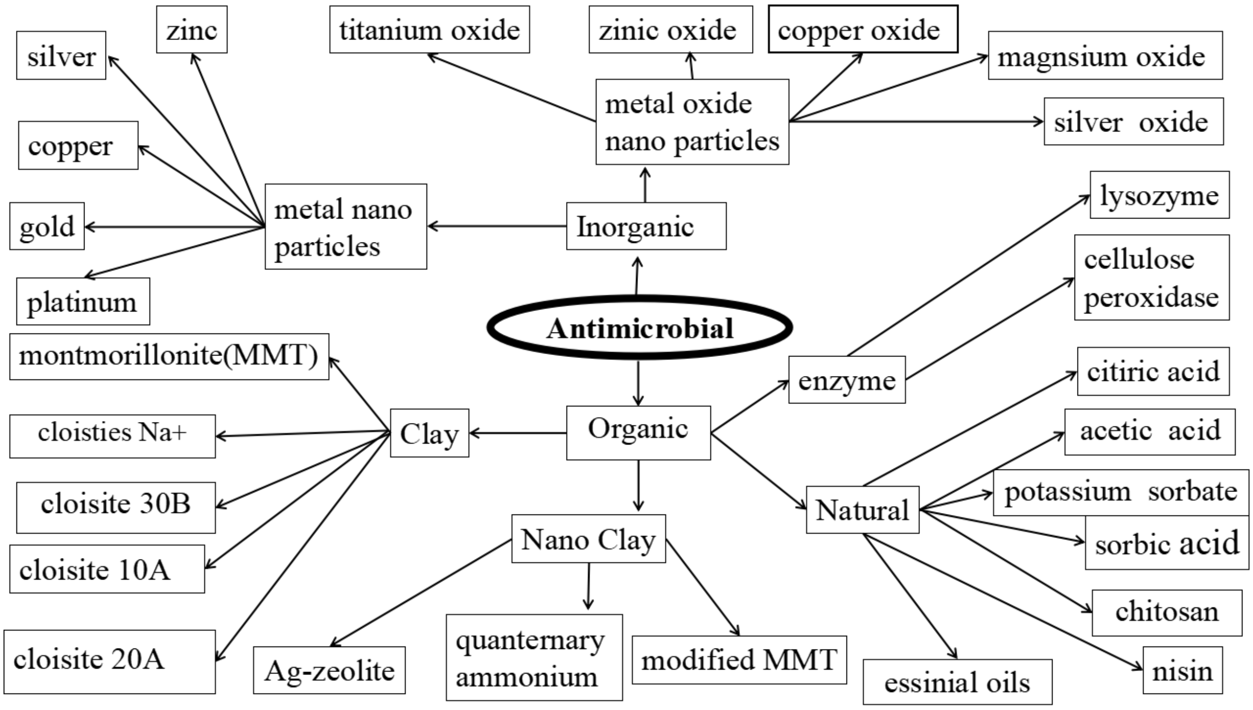

Figure 2. Type of antimicrobial substances/compounds used in food packaging system [39, 40]

Table 1.

Natural antimicrobial compounds and antimicrobial activity

Antimicrobial agent	Coated with	Antimicrobial activity	Type of microorganism	Reference
Chitosan	Lauric acid in starch film	reduction	Subtilis E. coli	[41]
	Ethylene in co-polymer films	2–5 log reduction	E. coli, L.monocytogenes	[42]
		Reduction	Negative and positive bacteria	[42]
Nisin	Ally isothiocyanate	Completely inactive	Salmonella	[43]
	Poly lactic acid	Completely inactive	Listeria monocytogene	[44]
	Polyethylene films	3 to 7 log Reduction	Listeria monocytogene	[45]
essential oils of mustard and cinnamon		inhibit spoilage	Aspergillus flavus, Endomyces fibuliger,	[46]
methanolic extract of pomogranate peel		inhibition of 10–25 mm	S. aureus	[47]
grape seed extract		inhibition	E. coli	[48]
methanolic extracts dihydroxylated coumarins		100% inhibition	T. mentagrophytes and R. solani	[49]
olive leaf extract		Inhibition	Candida albicans	[50]

3. Methods for incorporate antimicrobial compounds and difficulties in packaging systems

Antimicrobial packaging materials are critical for food preservation and safety because they prevent spoilage caused by fungal and bacterial microorganisms [51, 52]. This packaging material has the advantage of increasing shelf-life, dimension microbe growth phases, and protecting foods and preserving their original quality, taste, and protection for an extended period of time [53–55]. This promising packaging method is used in meat, fruits, dairy products, and vegetables [56–58]. Antimicrobial compounds can be mixed into two parts in food packaging systems. The first stage involves direct contact with the food surface (such as foils in this application), and the second involves antimicrobial agents blended into packaging systems (here there is no direct contact the antimicrobial compounds with food) [59–61]. Essential oils, nano metal oxides, chitosan, and nisin, among other antimicrobial agents, can be blended/coated with films or carpeted the surface of food, when the film being ediblE. The movement of the agents to the food in this case is classified as partial or complete migration to the food [62, 63].Casting and extrusion techniques are the most common routes for antimicrobial substance incorporation among the various types of antimicrobial packaging methods.

3.1 Antimicrobial coating methods

Organic and inorganic compounds can be mixed in the coating route for the synthesis of active packaging in food sectors. Among the inorganic nano metal oxides that can be coated to the surface of the food or material covalent or hydrogen bonding interaction are titanium oxide, copper oxide, zinc oxide, silver oxide, magnesium oxides, and nano encapsulation. These nano particles (NPs) depend such as material type [64,65], particle size, shape [66]functionality, hydrophilic-hydrophobic properties and usage concentration [67]. According to the table 2, the application of coating technique has been reported for varies investigation. The PPE/PEE coated at temperature of 75°Cwith rosemary oil, garlic oil, allylisothiocyanate, and trans-cinnamaldehyde showed antimicrobial activity against of *E. coli*, *SaL. typhimurium*, *E. sakazaki*, *B. cereus* [68]. Carboxymethyl cellulose (CMC), agar, carrageenan, coated with ZnO NPs revealed its *E. coli* and *L. monocytogenes* activities [69] and agar film coated natamycin reported antimicrobial assay against *aspergillosis niger* and *Saccharomyces cerevisiae* in strawberries [70]However, the coating technology faces the following challenges. The main limitations are the change in surface structure of antimicrobial substances, particle aggregation, volatile substances losing their antimicrobial properties during drying, and chemical compatibility with solvents and polymers. The antimicrobial compounds distribution in the package system, indirect contact with the food, and non-aggressive thermal treatment, and it can be good for the synthesis of active packaging systems. Temperature labile antimicrobial substances can be blended through coating routes with little loss of activity in active packaging materials. Other methods of mixing inorganic nano metals in packaging systems, such as chemical/physical deposition, are more priceless and require smart processing equipment. Metal oxides such as Ag_2O, ZnO, TiO_2, MgO, and CuO are the most effective surface modification, fictionalization, and deposition agents [71].

3.2 Extrusion technologies

Extrusion is the most common method for incorporating natural extracts or inorganic nano particles into the film surface for packaging applications [72]. Furthermore, this route blends the natural extracts (bio-active compounds) before entering the stage of the processed polymer's melting temperature (inside the extrusion) to ensure uniform distribution in the film. Nonetheless, it faces three challenges: homogeneous dispersion of inorganic metal nano particles in the polymer matrix, thermal degradation of the bio-active elements, and dimension change in their active sitE. The mechanical and barrier properties of the active packaging materials, in particular, had a significant impact on the aggregation (nonuniform dispersion) of the nano particles. Because of the different affinity of the matrix/film(polymer) and the additive agents, this difficulty affects the antimicrobial activities of the packaging (organic or inorganic).

According to table 2, a substantial amount of literature has been published on the incorporation of antimicrobial agents via extrusion. At 160–190 °C, the LLDPE material was extruded with grapefruit seed extract. However, the antimicrobial activity was rendered ineffective [73]. The surface coating is preferably for the natural agents (bio-active compounds), despite of its low temperatures and simple technique, poor adhesion is the main limitation to develop the active packaging systems using the coating techniques [74]. Furthermore, films have been investigated reported from chitosan/essential oil-coated PP [75], cinnamaldehyde, garlic oil and rosemary oil-coated PP/LDPE [68], oregano essential oil and citral-coated PP/EVOH [76], chitosan-coated plastic [77], thyme and oregano-coated LDPE. Interestingly, the active packaging materials synthesized through extrusion route shows more effective against *E. coli*, *Salmonella typhimurium*, and *L. monocytogenes*.

Additionally, the LDPE incorporated with garlic oil extruded with a temperature above 100°C and the antiviral and antimicrobial activities have been decreased [78]. At this point, the authored reported garlic oil and trans-cinnamaldehyde have excellent antimicrobial activity than allyl isothiocyanate [68]. According to current research, rosemary oleoresin is the least effective naturally derived antimicrobial compound against *L. innocua* and *E. coli* in an agar medium when coated onto LDPE/polyamide films compared to trans-cinnamaldehyde, thymol, and carvocrol [79].

Zinc oxide nanoparticles' antimicrobial activities in food packaging applications have been reported in the literaturE. It contained agar, carrageenan, and CMC polymers. It had less of an effect when combined with CMC than when combined with agar and carrageenan. The Gram-positive bacteria has been inhibited by the metal nano particles (example zinc oxide), but it can't surprise for the negative bacteria. This activity of ZnO NPs was also observed in CA films [80]. Gram-positive *Staphylococcus aureus* bacteria were more inhibited/had lower activity than Gram-negative bacteria (*E. coli*, *Citrobacter freundii*, and *Klebsiella pneumonia*). The ZnO mixed with sage starch was also more effective against Gram positive (*S. aureus*) bacteria than Gram negative bacteria (*E. coli*) [81]. This happened due to the different cell walls of them. Gram-positive bacteria have a single thick cell wall (referred to as multi-layers of glycopeptide), whereas Gram-negative bacteria have a complex cell wall (thin glycopeptide layer) that is protected by an outer membrane [74,82]. Furthermore, the pores found on the cell wall of Gram-positive bacteria allow zinc oxide to penetrate, causing leakage into the intracellular part and cell death [83]. However, the Gram-negative bacteria's outer cell membrane protects the zinc oxide from penetration and attachment [80]. When compared to other inorganic compounds (especially metal oxides), zinc oxide nano particles have a higher antimicrobial activity (against *E. coli*, *Bacillus atrophaeus*, and *Salmonella aureus*) [84]. When compared to silver oxide, zinc oxide is less expensive and less toxic to humans and animals, making it appealing for food packaging applications (AgNPs) [40].The percentage of agents (antimicrobial materials) determined the affinity of the selective microorganisms to be deactivated, partially inhibited, or completely inhibited. In the strawberry packaging application, a 5wt percent sliver nano particle (Nano-Ag) blend by solvent evaporation performed welL. TiO_2 (0–20 wt.%) and – CD–thymol (0–5 wt.%) incorporated with PLA via solvent casting, hot-pres processing, and injection process demonstrated that the produced active nanocomposite packaging films have applications in *E. coli* and *Alternaria alternata*, respectively.

Table 2.

Development techniques, antimicrobial compounds and applications

Polymer	Antimicrobial agent	Percentage (%)	method	Temperature (°C)	application	references
LDPE	garlic oil	2,4,6,8w/w	extrusion	170		[78]
PLA	Waste Orange Peels Extracts	0.25–2.00% wt	hot pressing/extrusion	175, 100 bar	yellowish color increasing with addition level and anunacceptable browning at the 2% dosage	[85]
PET and HDPE	TiO_2 NPs		extrusion	260	milk	[86]
PLA	green tea extract (GTE)		extrusion		antioxidant activity	[87]
polypropylene polyethylene	rosemary oil, allyl isothiocyanate, trans-cinnamaldehyde, garlic oil	0.6–1.2%, v/v	coat	75	E. coli, SaL. typhimurium, E. sakazaki, B. cereus	[68]
PLA	α-tocopherol		extrusion	193	inhibitor of lipid oxidation of whole milk powder	[88]
HDPE LDPE	catechin, quercetin and tea extract		extrusion		improved their thermal resistance	[72]
CMCagar, carrageenan	ZnO NPs		solution casting	80	E. coli and L. monocytogenes	[69]
agar film	natamycin	0%, 0.33%, 0.66%, 0.99%, 1.33%, w/w	solvent casting	100	Aspergillus niger and Saccharomyces cerevisiae	[70]
PLA	Nano-Ag	5wt%	Solvent evaporation		Strawberry	[89]
	TiO_2	0–20 wt.%	solvent casting and hot-press processing		E. coli	
	β-CD–thymol	0–5 wt.%	injection process		Alternaria alternata	
cellulose	chitosan		solution casting			
PHB	bacterial cellulose nanofibers		melt compounding technique		Gram-positive bacteria	
cellulosic paper	chitosan		dip-coating			

Advantages and challenges of antimicrobial compounds incorporated in composite food packaging systems

TiO_2, monolaurin, clove leaf oil, enterocin, and pomegranate peel extract, when combined with chitosan/PVA, cellulose/chitosan, starch, agar, and zein, demonstrated promising results in cheese product packaging applications. Natural extracts of mentha pipetia and bunium percicum garllic acid incorporated with PLA/NC have been used in Protected food (ground beef) applications. According to table 3, natural extract antimicrobial materials are more popular than inorganic (metal and metal oxide nano particles) antimicrobial materials in food packaging applications due to their safety, low cost, non-thermal processing requirements, and edibility.

The advantage of the bio-based nano-composite packaging materials in synthesis of food packaging materials has been reported in the Figure 3.

Table 3.

Type of food, packaging material, and antimicrobial compounds [90]

Protected food type	Polymer	Antimicrobial material
Cheese	Chitosan/PVA	TiO_2
	Cellulose/Chitosan	Monolaurin
	Starch	Clove leaf oil
	Agar	Enterocin
	Zein	Pomegranate peel extract
Peanuts(roasted)	Banana flour	Garlic essential oil
Salami	Whey protein	Cinnamoum cassia, Rosmarinus officinalis oils
Ham	Chitosan/starch	Gallic acid
Rainbow trout fillet	Chitosan	Grape seed extract
Cucumber		Limonene
Tomato		TiO_2 nano particles
Poultry		Ginger oil
Strew berries	Chitosan/CMC, PLA, Gelatin	Chitosan/citric acid, AgNPs, mentha spicata oil, butylated hydroxyanisole
Shrimps	Chitosan	Carvacrol
	Gelatin	ZnO/clove oil
Chicken		Thyme oil
	Chitosan	Acerola residue extract
	Pullulan	Nisin
Crap fillets	Alginate/CMC	ZNO/Ziziphora clinopodioides oil
Ground beef	PLA/NC	Mentha pipetia, Bunium percicum garllic acid
Salmon	PLA	Glycerol monolaurate
Fish		Thymol
Iceberg lettuce	Cellulose	Clove and oregano oils
Ostrich meat	Kefiran/polyurethane	Zataria multiflora oil

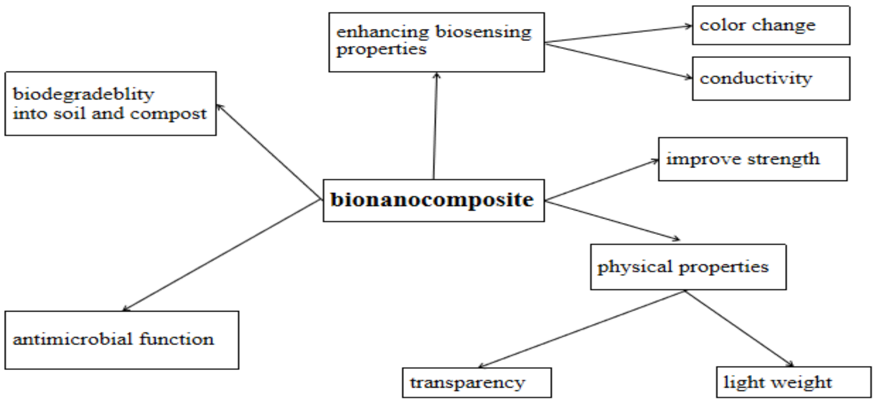

Figure 3. Bio-based nano-composites have an advantage in the synthesis of food packaging systems

The combination of nano particles and polymer is known as nano-composite, and it is a promising material for food packaging [91, 92]. According to some published research, nano particles may migrate from packaging materials (bio nanocomposites) to foods [40].If the nano particle concentration is high, these migration particles may cause rancidity (oxidation of food). Despite the fact that Zn, Ag^{+} ions, and clay have been transferred from chitosan, polypropylene(pp), and potato starch-based nano-based composites [39, 92], their migration to food is negligible and classified below the quantification limit. However, due to the high concentration transfer of nano particles from food packaging composites, the critical concern at this stage is that these nano particles can migrate into the main parts of the human body (brain, liver, fetus, and spleen), production cost, nano particles migrate to foods and have an environmental concern. The nano particles have a high surface area, which causes a high reaction with heavy metals, implying contamination of the soil and water bodies [40]. Therefore, it is a major challenge to prevent the migration of nano particles from bio nanocomposite materials to food mass when considering smart food packaging.

The most promising antimicrobial agents have eco-friendliness, safety, and lower scented human risk to consumers by reducing food contamination by microorganisms. When compared to conventional composites, the extraction process of bio-active compounds used in food packaging as antimicrobial agents from natural resources is expensive and limited in raw material availability.

Conclusion

Antimicrobial compounds combined with food packaging systems have the potential to keep fresh food for a long timE. The current art of questioning in the food sectors and research areas is to design active packaging system for the purpose of shelf-life and safety of food. Currently, the cost of natural antimicrobial compounds and safety of metallic/oxide nano particles, as well as regulatory concerns, are limiting the production and synthesis of antimicrobial food packaging systems. Because of the low energy intensive, non-thermal requirement, and the agents do not migrate into the food, the coating technique of incorporating antimicrobial compounds into the food packaging composite has received more attention than the extrusion method, and it can be good for developing active packaging materials. Incorporating natural extracts (especially essential oils) with antimicrobial properties into food packaging systems holds more promise than inorganic antimicrobial (such as metal/metal oxide nano particles). When compared to silver oxide, the zinc oxide nano particle is less expensive and less toxic to humans and animals, making it appealing for food packaging services. Researchers and academics can improve coating and extrusion techniques to incorporate natural antimicrobial compounds in polymer film to create an active composite food packaging system and further investigate the health effects of metal nano particles.

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Conflict of interest

The authors declare no conflict of interest.

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