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REVIEW

Research progress on antimicrobial materials for food packaging

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ABSTRACT
Microbial contamination is an utmost cause of food spoilage. Antimicrobial agents are used to treat microbial diseases in food. They also serve food packaging industry, as they are used for the formation of antimicrobial packaging films which maintain the structure, texture, color, and nutrition value of food. Due to ever growing population, the demands of food are also rising. There is need to stop food wastage and to prevent spoilage. Most of the food is spoiled during harvesting, transportation, and distribution. This is a serious problem to overcome. Adding antibacterial agents is the most convenient way to reduce food spoilage and contamination. To support the characteristics and properties of antibacterial materials, different modifications are performed in the field of food packaging, which is one of the most demanding techniques for food preservation. This review will summarize the research about antimicrobial agents, with an emphasis on recent findings, to highlight the importance of new developments in this field. Concepts of antimicrobial packaging with a focus on antibiotics and antibacterial agents are discussed briefly in this review, along with the different types of food packaging and applications of antimicrobial packaging. Synthetic and natural antimicrobial materials are described. In summary, this article will explain the importance of antibacterial agents and their use in food packaging industry. Furthermore, readers will get good information about natural antibacterial polymers which were extensively used in past few decades. Subsequently, different innovations should be done to control food spoilage and wastage to ensure food safety. Food packaging is a sole element that helps to provide safe and secure food for all.

KEYWORDS
Antimicrobial agents; food packaging; food safety; polymers

Introduction

Food packaging is an important factor for food safety. Food safety is of the major factors affecting public health and the well-being of society. A possible solution for food borne illnesses is good antimicrobial packaging (Yousefi et al. 2019). Packaging is a highly significant feature for increasing the shelf-life of various foods. It helps not only to decrease the physical harms but also the contamination and microbial influence. Moreover, packaging also helps to improve the acceptance of food in market level. Invasion of microorganisms is one of the main causes of food spoilage and deterioration (Huang et al. 2019). It not only reduces the acceptance of food but also aids in the spread of foodborne diseases. Foodborne diseases are mainly due to such deadly pathogens. A number of people reported every day for food poisoning resulting in number of deaths every year (Al-Tayyar, Youssef, and Al-Hindi 2020).

Basically, the main function of food packaging is to extract the useful material from external environment and to utilize them for food safety. Also, it helps to protect the food from external harsh environment, odor, gasses, dust, moisture, irradiations, light, and microorganisms. Contamination can occur at any place even right after harvesting (Ju et al. 2019). Various stages from harvesting to the display in market, food can get in contact with various elements and there are chances of contamination at every point. In such conditions, a good packaging acts as an obstacle against contaminations. The material used in such packaging materials is very important. Antibacterial agents are of the most important features of food packaging. Various bioactive compounds, essential oils, polyphenols, and polymers are used for this purpose (Moustafa et al. 2019). The traditional methods of food packaging include plastics and nonrenewable resources of fossils fuels which are not recyclable and disposable (Gan and Chow 2018). Such nonbiodegradable packaging materials are responsible for high waste and also dangerous for marine life. So, there is a great need for natural and biodegradable material to be replaced with plastics. For this purpose, various food grade materials are used. These materials are antimicrobial in nature and also biodegradable and thus cause no harm to the environment. Two major sources of antimicrobial agents are inorganic and organic antimicrobial agents (Ritchie and Roser 2018). Various classes of antimicrobial agents are mentioned in Figure 1.

Biopolymers can be used to replace such plastics. They are biodegradable and environmentally friendly. These

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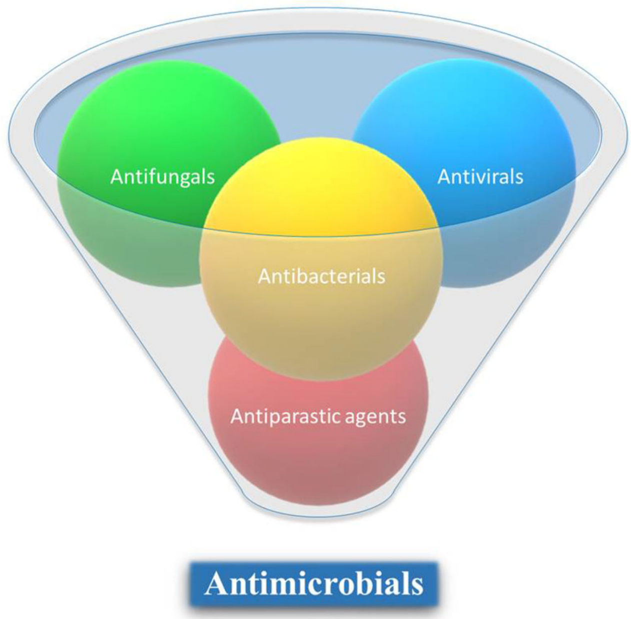

Figure 1. Different classes of antimicrobials.

polymers are very important for food packaging as they are complete compostable and generally recognized as safe. They involve enzymatic and hydrolytic cleavage of bonds in the polymers. Biodegradability is a very important event which occurs via the chemical decomposition of the action of enzymes associated with living organisms (Han et al. 2020). The abilities and action against microorganisms are decided by the abilities of antimicrobial agents itself. These abilities depend on their natural and synthetic sources and the type of source. Natural sources are preferred because they are safe and biodegradable (Topuz and Uyar 2020). However, with increasing food production and demand for packaging, natural antimicrobial materials tend to be insufficient. Thus, organic, or inorganic synthetic antimicrobial agents have been developed. The main antibacterial components in food packaging are synthetic organic materials, which mainly contain ethylene diamine tetra acetic acid (EDTA), fungicides, parabens, and other chemicals (Huang et al. 2019).

Antibacterial and active packaging, on the other hand, are currently considered as very effective with concerning biology and control the activity of bacteria and fungi, also natural polymers are used in several studies as they provide good food protection. Films made of natural polymers provide great moisture barrier to fruits and vegetables and are not harmful to food and human health. Biologically, they are effective as they can reduce fruit respiration and transpiration rates (Wang et al. 2019). Films fabricated with natural-based materials are beneficial and protective for animals, making them capable to suppress the lack or deficiency of any elements, which increases its value in food packaging applications. The use of composite films prepared from natural materials is not only safe and environmentally friendly, but also enables full utilization of the properties of the raw materials by compensating for any shortcomings of the individual components. The meticulous selection of raw resources endows the composite films with distinct features, such as antibacterial properties and biodegradability, making them effective for food packaging applications. Moreover, the use of composite films for food packaging applications helps reduce the adverse effects of using plastics (Lan, He, and Liu 2018). This review will describe the importance of antimicrobial agents, their use and sources along with their applications in packaging industry.

Types of antibacterial agents

Inorganic antibacterial agents

There are several inorganic compounds which possess certain antibacterial activities. Besides, nanoparticles and nanotechnology exclusively relate to inorganic antimicrobial agents. Some of these inorganic nanoparticles are used in the industry in a very larger scale. One of the most important features of antibacterial inorganic compounds is that they show long life and high stability under certain conditions like heat, yet they have some shortcomings as they exhibit weak mold-resistant activity; thus, the large dosage is needed when professionally used in an industrial scale. Inorganic antimicrobial agents are not only used in the food packaging industry but also in different appliances like microwaves, and refrigerators to kill the microbes around the surfaces, thus providing an ultimate feature to the machinery. The purpose of this section is to present several inorganic substances and their activity (Hoseinnejad, Jafari, and Katouzian 2018).

Nanotechnologies are rising very fast and getting great popularity to support the benefits of the preservation of foods (He and Hwang 2016). The augmentation of disposable materials with nanofillers enables the improvement of their characteristics and expands the range of applications. Due to the lack of barrier properties (e.g., to oxygen, light, or water vapor) or the poor mechanical properties of these products, when compared with ordinary materials, due to the use of biodegradable materials, for instance, nanoparticles of coper oxide, zinc oxide, and silver have been used successfully to obtain antimicrobial nanopackaging using nanotechnology (Espitia et al. 2013; Makwana, Choudhary, and Kohli 2015; Mellinas et al. 2016).

Silver ions

Silver ions are one of the most frequently used materials for antimicrobial protection. They are used in food packaging, and even in the lining of refrigerators and microwave ovens, owing to their antibacterial protection property, and because they are excellent corrosion protection materials. The antimicrobial activity of silver ions is recognized since ancient times (Jung et al. 2008; Koizhaiganova, Yaşa, and Gülümser 2015; Mirzaee, Vaezi, and Palizdar 2016; Saugo et al. 2015; Silver, Schottel, and Weiss 2001; Tsezos, Remoudaki, and Angelatou 1995). There are several chemical forms of silver known to date. Some of the most abundantly and extensively used form are silver nanoparticles (AgNPs) (Sun,

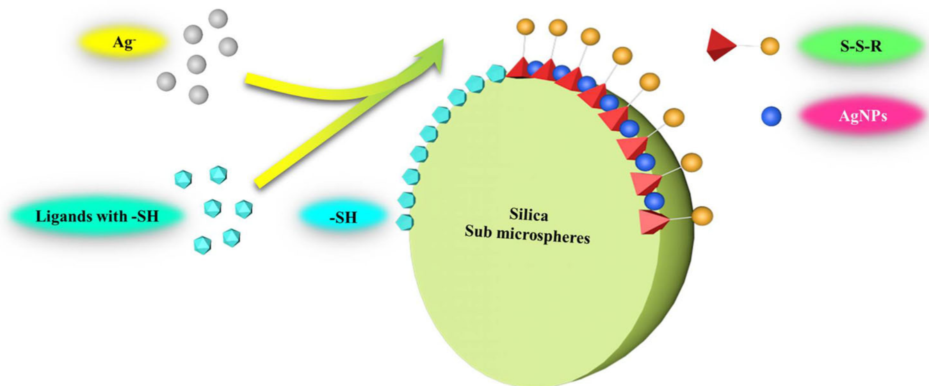

Figure 2. Contribution of AgNPs with organic ligands to promote antibacterial effect.

Sheng, and Liu 2013), silver sulfadiazine, and novel composite materials carrying silver.

Silver is usually used in both, ionic procedure, and silver nanoparticles as antibacterial mediators. Several studies have formerly reported on the antibacterial activity of silver nanoparticles and the effect of size, responsiveness, temperature, or ionic strength on its antibacterial properties. Those factors are all-embracing to polymer–silver nanocomposites. For instance, polyurethane/silver nanocomposites with improved silver ion release using multifunctional invertible polyesters. One study assessed the antibacterial activity of Ag–polyaniline (Ag–PANI) nanocomposites against *Bacillus subtilis* (Triebel et al. 2011).

In addition, numerous studies suggest that metallic AgNPs may bind to the surface of the cell membrane triggering cell membrane permeability and cellular respiration alterations. It is well recognized that Ag ions and Ag-based compounds are remarkably toxic to microorganisms, showing robust biocidal effects on 12 species of bacteria including *Escherichia coli*. The antibacterial exploitation of Ag^+ ions is presently explained by three arguments: (1) the amalgamation of AgNPs recurrently requires the contribution of organic ligands, (2) the mechanism of organic ligands influences the antibacterial action of the AgNPs. Based on an original in situ synthesis approach, silica-supported AgNPs with bare surfaces were created and further functionalized with several organic ligands (-OH, -Ph, -SH, and -COOH), and (3) the antibacterial activity of the bare AgNPs and AgNPs with various ligands were inspected. Though all specimens revealed different antibacterial abilities against *E. coli*, the minimum inhibitory concentration (MIC) of AgNPs functionalized with -COOH was the lowest and as low as 8–16 $\mu g\ mL^{-1}$, indicating the best antibacterial ability among the four ligands. Furthermore, the cytotoxicity test of bare AgNPs and AgNPs with ligands proved that the AgNPs produced no significant damage to human cells, demonstrating their potential for antibacterial applications. Their complete study is mentioned in Figure 2 (Xue et al. 2019).

Zinc oxide

Zinc is considered as a ubiquitous trace metal and an essential element for higher metalloenzymes. ZnO found in different shapes carries various functions. It is reported that zinc oxide particles occurring in various shapes, such as mulberry, lamellar, and flower-shaped, showed 90%, 60%, and 50% inhibition activity against *Candida albicans*, because ZnO could quickly spread and adsorb onto the bacterial cell surface, affecting the bacterial cell membrane permeability, causing leakage, and ultimately leading to bacterial cell death. Furthermore, the small size of ZnO can facilitate penetration of the particle through the bacterial cell wall, making ZnO readily available for gene expression inhibition, which consequently impedes the reproduction of bacteria (Mijnendonckx et al. 2013). Different assemblies of ZnO, namely spherical and nanorods, and different components, e.g., mushroom component and cauliflower component, are found to show antimicrobial activity; in particular, the cauliflower component has the best bactericidal activity (Singh, Barick, and Bahadur 2013).

In addition, zinc oxide-nanoparticles (ZnO-NPs) not only show good bactericidal effect toward both Gram types of bacteria, but also carry enough biocompatibility. ZnO is widely known as a heat-stable oxide, antimicrobial, and anticancer agents, and ZnO-NPs show excellent properties against bacterial growth. Although the exact bactericidal mechanism needs to be further clarified (Dimapilis et al. 2018), it is known that ZnO causes membrane permeability loss, cell membrane rupture, cell wall damage, and leakage of cellular materials (Padmavathy and Vijayaraghavan 2008; Tiwari et al. 2018); the production of reactive oxygen species (ROS), i.e., H_2O_2 and O_2, is the most accepted mechanism and considered as the main component of antibacterial activity by many previous researchers given the direct contact of ZnO particles with the cell membrane or their internalization. Such properties of ZnO nanostructures make them an excellent material for utilization in relevant biomedical fields (Li et al. 2017, 2018), e.g., for osteointegration and osteoblast progress along with infection prevention (Li et al. 2017; Liu et al. 2010; Park et al. 2010).

Copper oxide

Copper is a well-known element found as ions or salts in food. Copper oxide (CuO) is the purest form of copper and is utilized widely for many valuable purposes. It is relatively cheap, highly soluble, and is stable with polarized liquids and polymers, possessing highly stable chemical and physical properties and other potential attributes, such as superconductivity under high temperature, biofilm control, correlation of electrons, and spin dynamics. CuO nanoparticles are being investigated owing to their potential against the microbial activity. Several research articles reported that CuO nanoparticles in suspension could act against several bacteria like methicillin-resistant *Staphylococcus aureus* (MRSA) and *E. coli*. Moreover, different types of nanoparticles containing CuO have shown to acquire potential abilities, for example, the association of silver nanoparticles with CuO showed the ability to kill microbes due to optimum release of ions. Similarly, when copper oxide was embedded in porous elastomeric polyurethane films, the material also displayed some potential antibacterial application. There has been less research on the oxide formation reaction on copper and its influence on the antimicrobial efficiency. Some available literature indicates that the oxidation of copper is initiated with the formation of a Cu_2O layer, and subsequently, as the temperature rises to 200 °C, this Cu_2O layer reacts with oxygen, thus forming a layer of CuO on the external surface. When available in the form of nanoparticles, copper oxides can be utilized for coatings on the packaging material owing to its antimicrobial behavior (Ahmad et al. 2012).

Organic antimicrobial agents

Organic antimicrobial agents are specific to some biomolecules. This characteristic makes their application wider to many fields, such as medical treatment, health care, and food packaging films. Organic agents are further divided into two section synthetic and natural antimicrobial agents. Although both are organic in nature, their division is done according to their origin as described below individually.

Synthetic antibacterial agents

Vanillin. Vanillin, belongs to the group of aromatic hydrocarbons, is known for its flavor and fragrance (Bezerra, Soares, and de Sousa 2016), and is reported to have various bioactive properties like neuroprotection and inhibition of neuraminidase activity and inflammation (Khan et al. 2017). Vanillin possesses the Generally Recognized as Safe (GRAS) status and has wide industrial application as an intermediate to produce various herbicides and drugs. In the food industry, it performs an antioxidative role to improve food storage quality. It was reported to protect proteins and lipids from oxidation and peroxidation in mitochondria of rat liver. Besides, its inhibitory role against microbes (e.g., bacteria, molds, and yeast) shows its potential as a food preservative. Some previous research reported the antibacterial activity of vanillin derivatives against *E. coli*, *S. aureus*, and *Pseudomonas aeruginosa,* as well as fungal inhibition. A previous study on the effects of vanillin against *Cronobacter sakazakii* and *Salmonella enterica* demonstrated that vanillin combined with caprylic acid exhibits remarkable bactericidal activity (Kim and Rhee 2016), while the chitosan film with vanillin significantly inhibited *E. coli* growth (Stroescu et al. 2015).

Quaternary ammonium salts. Quaternary ammonium salts (QAS) belong to a beneficial group of compounds. They have widespread utilization in industry attributed to the ease in obtaining the intended material through functionalization of their heterocyclic nitrogen atom. They are amphiphilic, and their detergent-like property, i.e., the electrostatic attraction between the positive charge of QAS head and the negative charge of the bacterial cell membrane, results in severe damage to bacteria and causes leakage and lysis that ultimately leads to bacterial death. This characteristic makes them effective against a range of bacteria like *Enterococcus faecium*, *S. aureus*, *Klebsiella pneumonia*, *Acinetobacter baumannii*, *P. aeruginosa*, and Enterobacter species. QAS have a vast utilization in the pharmaceutical industry owing to their protective role against a wide variety of microorganism like bacteria, fungi, and viruses (Duque-Benítez et al. 2016). So far, QAS have been prepared by polymerization of, for instance, quaternate chitosan, ammonium bromides, and *N*-methyl-N-R-N, N-bis, which are being utilized against several microbes, e.g., *Botryosphaeria ribis*, *Physalospora piricola*, *Glomerella cingulate*, and *Clematis mandshurica*. This broad spectrum of antibacterial properties of QAS has attracted technologists to study the methods and designs for their use as a protective coating to prevent a variety of food materials (Moore et al. 2018).

Phenols. Phenols are used as biopreservatives in the vast field of the food industry. Several phenolic compounds were found from the usual bases that have an antimicrobial effect against a plethora of microorganisms, owing to the existence of phenolic mixtures in the extracts. They were extensively studied in previous researches for the preservation of highly perishable foods. Previous studies stated that the uppermost antimicrobial action in phenolic compounds, such as carvacrol, thymol, and eugenol, results from the acidic arrangement of the hydroxyl group. Carvacrol and thymol are considered extremely effective against bacterial infections of the upper respiratory tract. Phenolic extracts from different plants like olive fruits show specific characteristics. The extract from olive oil is an excellent source of phytochemicals as it is less toxic, cheap, and frequently available. Thus, the use of specific phenolic compounds from different natural sources is an interesting opportunity to develop a food packaging material without any synthetic additives, also their biological activities makes the packaging film more special

Isothiazolinone. Isothiazolinone is considered a powerful biocide that is used as preservatives in a wide range of food packaging industry. Based on the possible uses, biocides were classified into four categories: preservatives, pest control, general/disinfectant biocide products, and other biocide products. Isothiazolinone and 3-iodo-2-propynyl butyl carbamate (IPBC) were described as biocidal preservatives and disinfectants that were used across the world to prevent mildew growth and bacterial infection in many consumable

Table 1. Natural antimicrobial agents.

Natural antimicrobial agents	Packaging material	Foods	Microorganisms	References
Casein phosphopeptides (CPPS)	Gelatin			(Khedri et al. 2020)
Chitosan		Beef	mesophilic aerobic bacteria (TMAB), *Stapylococcus aureus*, lactic acid bacteria (LAB)	(Duran and Kahve 2020)
Nano-organo-montmorillonite	PLA/starch	mango		(Lima et al. 2019)
D-lemone	PVA	mango		(Lan, Li et al. 2020)
Apple polyphenols/AgNPs	Sodium alginate	Strawberry	*Escherichia coli, Staphylococcus aureus*	(Lan, Wang et al. 2020)
Blood orange peel pectin	Gelatin	Cheese		(Jridi et al. 2020)
Bacteriocins from LAB		Meat	*Lactococcus*, *Enterococcus*, *Pediococcus*, and *Lactobacillus*	(da Costa et al. 2019)
Pomegranate peel extract	PEO/ chitosan	Meat	*E. coli*	(Surendhiran et al. 2020)
Copaiba oil	PEC		*S. aureus* and *E. coli*	(Norcino et al. 2020)
Nisin		Fresh cut iceberg lettuce	*Listeria monocytogens*	(McManamon et al. 2019)
ZnO/clove essential oil	PLA/PEG/PCL	Scrambled egg	*Staphylococcus aureus* and *Escherichia coli*	(Ahmed et al. 2019)
Eryngium planum extract/ Bacterial cellulose		Strawberries	Gram positive, Gram negative	(Shahbazi, Shavisi, and Karami 2013)
Pistacia Atlantica gum/ silica nanoparticles	Polypropylene	Milk	*Staphylococcus aureus*, *Salmonella enterica*, *Escherichia coli*, and *Listeria monocytogenes*	(Ellahi et al. 2020)

products including skin care and hair products, cosmetics, fabric softeners, and cleaning products (Liu et al. 2019). Numerous isothiazolinone derivatives, such as methyl chloro isothiazolinone (MCI), octylisothiazolinone (OIT), benzisothiazolinone (BIT), and methylisothiazolinone (MI), have been used broadly as preservatives in a plethora of products. These compounds have the ability to diffuse in the cell membrane of bacteria, and also the cell wall of fungi, and show their action mechanism in the intracellular media. These compounds contain N-S bonds, the electron deficient sulfur part of this group usually react with the nucleophilic groups of the cellular components, for example, the thiols from the cysteines of protein active sites blocking the activity of enzymes and ultimately causing cellular death (Aerts et al. 2014; Hosteing et al. 2014; Silva et al. 2020). Extensive use and mishandling of isothiazolinone derivatives result in severe problems to the worker and the consumer. It was reported that so far 9% of the patients, accounting for 1% of the total population, was referred for patch tests and was positive for allergic diseases possibly spread through the extensive use of MI/MCI in the 1980s (Aerts et al. 2017).

Natural antimicrobial agents

Natural antimicrobial agents are one of the most important sources for antimicrobial packaging. They are natural and not harmful to health. Table 1 describes the sources of some antimicrobial agents from animal and plant origins along with the brief description of natural antimicrobial agents with the packaging material in which they are commonly incorporated and the foods for which these packaging materials are also made, the microorganism against which they are active.

When food is contaminated as a result of the presence of different microorganisms such as fungi and bacteria, the quality and freshness of food are compromised. This results in the wastage of food and thus affects the whole environment and human health. Perhaps the most effective solution is an antimicrobial food packaging, so that food is protected against harmful and hazardous microbes (Pisoschi et al. 2018; Sung et al. 2013). There are some natural antimicrobial agents described in the subsequent sections.

Essential oils. Essential oils (EOs) are individual volatile compounds. They originate from plant sources, and they are specifically obtained from aromatic plants. These plants have not only several biological activities but also have specific antimicrobial effects. EOs are considered as a Generally Recognized as Safe (GRAS) antimicrobial agent because they are natural extracts of plants, and currently, different studies and research are ongoing to use them as safe food additives. They are also authorized by the Food and Drug Administration (FDA).

Now, the food industry is using useful approaches, which includes the use of several EOs as a natural antimicrobial agent in polymeric materials, to fully exploit the benefits and advantages of each material and simultaneously overcome the disadvantages and drawbacks of each component. Encapsulation of small compounds in particles is also one of the best techniques these days (Beyki et al. 2014; Biddeci et al. 2016; Wen et al. 2016). Some researchers have tried to encapsulate EOs by focusing on their use or to load them directly in polymeric films. However, the incorporation of EOs might be disadvantageous as it has strong flavor and smell, which limited its wide application. However, the antibacterial activity of essential oils can never be denied. Table 2 shows some examples of antimicrobial agents obtained from essential oils along with their main constituents. Table 3 describes some examples of essential oils which were encapsulated or incorporated in the packaging films for the preservation of food as they are a good source of antimicrobial agents.

Table 2. Antimicrobial activity of some EOs against bacteria and fungi.

Essential oil	Antimicrobial action	References
Eucalyptus radiata	*E. coli, P. aeruginosa, K. pneumoniae, Acinetobacter baumannii,* and *Salmonella Typhimurium.*	(Luís et al. 2016)
Cinnamomum osmophloeum	*P. aeruginosa, E. coli, Enterococcus faecalis, Staphylococcus epidermidis, S. aureus, Salmonella sp., K. pneumoniae,* and *Vibrio parahemolyticus.*	(Chang, Chen, and Chang 2001)
Ocimum basilicum L.	*E. coli, S. aureus, B. subtilis,* pathogenic fungi and *Pasteurella multocida, A. niger, Mucor mucedo, Fusarium solani, Botryodiplodia theobromae, Rhizopus solani.*	(Hussain et al. 2008)
Mentha rotundifolia L.	*Salmonella typhimurium, E. coli, S. aureus, Aspergillus niger, Bacillus cereus,* and *C. albicans.*	(Riahi et al. 2013)
Thymus vulgaris L	*Pantoa sp.* and *E. coli.*	(Ribeiro-Santos, Andrade, and Sanches-Silva 2017)

Table 3. Examples of antimicrobial food packaging containing EOs or their main constituents.

Essential oil	Food packaging	Food	References
Oregano	Milk protein	Whole beef muscles	(Ribeiro-Santos, Andrade, and Sanches-Silva 2017)
–	Whey protein isolate (WPI)	Fresh beef	(Zinoviadou, Koutsoumanis, and Biliaderis 2009)
–	Cellulosic resin	Pizza	(Botre et al. 2010)
Linalool or methyl chavicol	Low-density polyethylene (LDPE)	Cheddar cheese	(Ribeiro-Santos, Andrade, and Sanches-Silva 2017)
Clove	Gelatin	Fish	
Rosemary	Cellulose acetate	Chicken breast cuts	(Melo et al. 2012)
Cinnamon, clove	Cassava starch	bakery	(Souza et al. 2013)

Lysozymes. Lysozymes are found in many natural systems. They are basically small and stable lytic enzymes whose sequences and dimensional structures were completely analyzed. Lysozymes have very good potential in the field of food packaging because of their some unique characteristics such as they show good stability over wide range of pH environment and also in various temperature environment. Because of its poor working against Gram-negative bacteria, it has downmarket value in food packaging industry. Lysozyme is less effective toward Gram-negative bacteria as compared with other antimicrobial agents because of the presence of the protective layer of lipopolysaccharides (LPS) on the cell wall of bacteria. To overcome this drawback, a number of studies performed and tested in past few decades. Besides, it shows great work toward Gram-positive bacteria. Gram-positive bacteria have a membrane consisting of 90% peptidoglycan, in which the hydrolyzation of the β-1-4 glyosidic bond between *N*-acetyl glucosamine and *N*-acetyl muramic acid takes place; therefore, Gram-positive bacteria are very vulnerable to lysozyme. Some researchers studied the several methods of antimicrobial effect of various lysozymes to Gram-negative bacteria. This experiment includes the denaturation of enzyme, modification in lysozymes by the attachment of other compounds, and also the use of membrane-permeabilizing agents with the lysozymes (Park, Daeschel, and Zhao 2004).

Chitosan. Chitosan is a natural polymer mainly obtained from scales of fish and made from chitin, which is the main component of fungi cell walls. Chitosan exerts great antimicrobial activities on a series of foodborne microorganisms, and it is also more efficient against Gram-negative bacteria than against Gram-positive bacteria. The use of chitosan in the preservation of food was limited because of its poor solubility at neutral and alkaline pH. However, this problem has been resolved by using derivatives of *N*-alkylated disaccharide chitosan that act efficiently against *E. coli* and *S. aureus*. To develop disposable antibacterial composite membranes for food packaging applications, chitosan was used in combination with *S*-nitroso-*N*-acetyl DE penicillamine (SNAP) and nanocellulose. The antimicrobial area evaluation of SNAP-assimilated chitosan membranes showed an efficient zone of restriction in different bacterial strains (*S. aureus*, *Listeria monocytogenes*, and *Enterococcus faecalis*) (Sundaram et al. 2016). However, chitosan is very difficult to be electrospun. To overcome this difficulty, chitosan is electrospun in combination with other supporting polymers. For example, a group of researchers recently performed electrospinning of chitosan and poly-lysine and showed some effective antibacterial activity, and suggested it could be easily used as active food packaging material (Figure 3) (Lin, Xue, Duraiarasan, and Haiying 2018).

Bacteriocins. Antimicrobial agents produced by bacterial microbes, known as bacteriocins, have slowly received attention among other natural antimicrobial agents extracted from plants, EOs, and animals. Bacteriocins (Table 1) are gaining popularity because of their ability to resist high temperatures and highly acidic environments. They are byproducts that are metabolic-like antimicrobial peptides formed by the defense system of almost every type of bacteria; bacteria of one strain inhibit the growth of other strains adjacent due to this naturally occurring activity. *Lactococcus lactis* bacteria produce nisin which is commonly present in milk. Nisin is the first isolated bacteriocin, and is approved by the FDA and is considered a GRAS antimicrobial agent, and thus does not cause any harm when employed for preservation of food. The only bacteriocin that is approved in more than 50 countries is nisin. It shows enhanced activity against Gram-positive bacteria and spore-forming bacteria, and the common targets of this bactericide are generally Gram-positive bacteria, which are foodborne pathogens or bacteria known for spoilage like *Micrococcus luteus*, *S. aureus*, and *Bacillus cereus*. For example, it is known that nisin

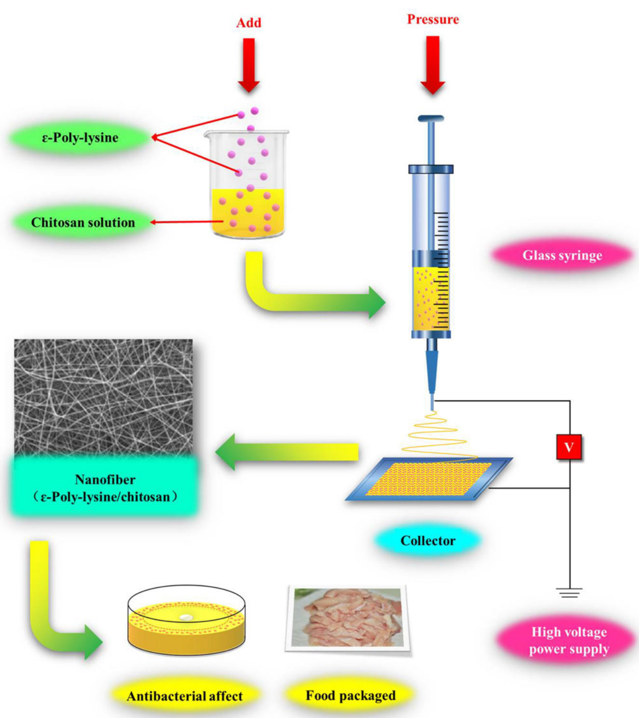

Figure 3. Nanofiber formation of chitosan and polylysine.

can prevent the outgrowth of spores of *Clostridium botulinum* in cheese, and that it inhibits yeast and mold growth in table olives, when used with natamycin. Bacteriocins have a general cationic character, which generally explains the generation of pores around the cellular membranes and enables simple incorporation with Gram-positive bacterial strains that provide elevated membrane levels of anionic lipids (Pisoschi et al. 2018).

Types of antimicrobial packaging

The different types of antimicrobial packaging are as follows: (a) Encapsulated antimicrobial packaging, (b) Addition of antimicrobial sachets to packaging material, (c) Direct coating of antimicrobials, (d) Absorptive antimicrobial packaging, and (e) Volatile antimicrobial packaging.

Encapsulated antimicrobial packaging

Encapsulation is a technique which results in the embedding of one particle which is active to another particle which ultimately controls the release of that active component. Currently, there are different encapsulation procedures, but it is difficult to label one as universal. The bioavailability of active compounds of interest in the target site is favored by encapsulation, as these compounds can be released locally from the carrier, which is managed by different durations of time that are not dependent on the molecular mass of the bioactive compound, and by a suitable carrier that provides targeted delivery, which may be allocated by encapsulation, improving food safety since it enhances the safety of biologically active compounds (Zhou, Liao, and Teng 2014). Nanotechnology and nanoencapsulation technology are also an efficient and practical alternative to overcome problems related to bioactivity and degradation of active molecules, and to retain and release those active molecules at the area of interest by different processes, mostly by diffusion. Nowadays, nanoparticles are obtained typically in the form of powder, and exhibit some characteristics that differ from those of large particles having the same chemical composition (Assis et al. 2012).

There are some unique structures called nanocapsules, which consist of a chemically synthesized polymeric membrane surrounding a core. The active components can be mixed in the polymeric membrane or in the core. Nanocapsules are a thousand times smaller than bacteria, and it is interesting to note this specificity of nanocapsules (Assis et al. 2012). In recent decades, rapid advances were seen in nanotechnology, and these have warranted the generation of different techniques to release active compounds, which are advanced in the different processes, in the biological environment (Donsì, Sessa, and Ferrari 2010). To fabricate active polyethylene films, Wrona et al. (2017) successfully encapsulated inorganic capsules loaded with tea extract by extrusion technology. Over time, for food safety the food industry has undergone daily to avail to the increasing purchases of natural foods that need unique properties for packaging. Active packaging, edible coatings, smart packaging, biodegradable packaging, among others are new technologies of packaging materials (Ghaani et al. 2016; Kapetanakou and Skandamis 2016). Antimicrobial compounds can be assimilated during the preparation of the packaging to exert the function of additives (Zanetti et al. 2018).

Some studies recently reported the encapsulation of silver nanoparticles in silica filler in a polyethylene film to promote the antibacterial and antimicrobial activities of the material (Pavoski et al. 2019). In this study, the authors indicated that silver is used as both silver ions and stabilized nanoparticles. However, silver nanoparticles are very difficult to distribute homogenously in the polymers because frequently the metal starts oxidizing, and the particles also agglomerate. To solve this problem, the authors encapsulated silver into inert fillers, i.e., silica, by the sol–gel method, which further promoted the encapsulation process (Figure 4). The antimicrobial effect was outstanding. However, the antimicrobial activities mainly depended on the particle size. The polymeric composites were prepared by fusing or by using other techniques (e.g., mixing the polymer solution or by direct mixing). Alternatively, the synthesis may be conducted in situ, where the monomer polymerization occurs in the presence of nanoparticles. This also tends to improve the properties of nanocomposites owing to good dispersion of the nanofillers (Pavoski et al. 2019).

Addition of antimicrobial sachets to packaging material

Antimicrobial sachets are some packages imbibed with antimicrobial compounds. It is an effective technique to make antimicrobial packaging. There are several methods to

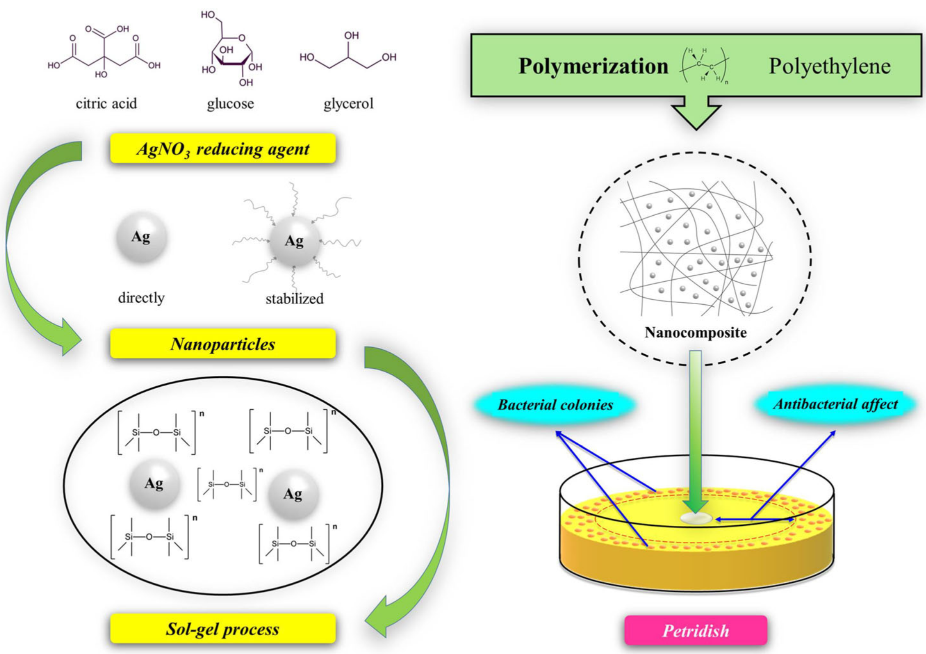

Figure 4. Sol–gel process and polymerization.

prepare antimicrobial packaging including the direct incorporation of a sachet-immobilized active compound in the polymer matrix, which is easy and helpful. The antimicrobial actions of such systems support their main positive roles as well as the antioxidant activity (Gómez-Estaca et al. 2014). At the commercial level, pouches or sachets and pads still play an important role in food preservation owing to their low cost and high performance. The use of sachets was expanded recently through the interaction, incorporation, and subsequent release of volatile organic compounds with very prominent antimicrobial activities against foodborne microorganisms. Active compounds then combine and work with sachets, including chlorine dioxide, ethanol, and different types of plant extracted EOs and their common and prominent active components. Absorbent pads are used to enhance the efficiency of absorbent material inside the packaging (Lee et al. 2015; Mellinas et al. 2016).

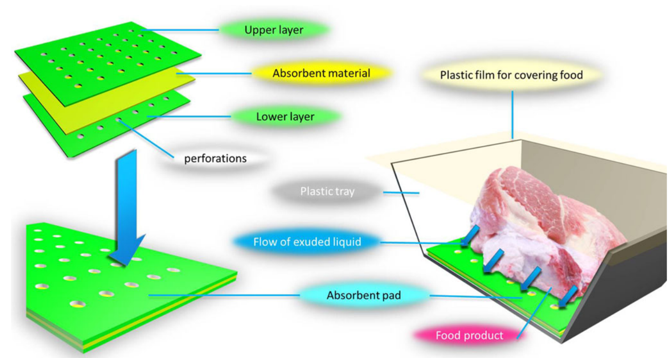

Figure 5. Layers of absorptive pads.

Types of emitting sachets

There are two approaches for the making of antimicrobial sachets for food packaging: (1) sachets that produce antimicrobial compounds in situ and release the compound and (2) sachets that act as carriers that carry and release antimicrobials. Figure 5 describes the arrangement of antibacterial sachets.

Sachets that generate and release antimicrobials. Ma (2012) developed a system to generate ally isothiocyanate (AITC), which is a vapor, via sinigrin–tyrosinase reaction, and to release it in situ. The production of ClO_2 in situ is another example. In this case, ClO_2 is not a stable gas, and given the risk of explosion, its compression and storage are not possible; it must be generated in situ. Industrially, generation of chlorine dioxide gas within permeable sachets is achieved by mixing two dry precursors (sodium chlorite and an acid), according to the reaction described below (Otoni et al. 2016)

$$5NaClO_2 + 4H^+ \rightarrow 4ClO_2 + 4Na^+ + NaCl + 2H_2O.$$

Sachets that carry and release antimicrobials. The procedure to obtain sachets that carry, and release antimicrobials consists of three steps: (a) incorporation of the antimicrobial agent into the carrier, (b) addition of carrier into the sachets, and (c) sealing the sachets. Researchers prepared very effective sachets by adding liquid AITC into a porous high-density polyethylene carrier resin. The polymer was heat-sealed and then placed inside a non-woven sachet material fabric to form the antimicrobial sachet. Adsorption/incorporation of antimicrobial compounds in a carrier material that manages their release into a sealed packaging system is the principle of the carrier/releasing sachets. In order to prevent premature leakage of the compound from the carrier, the sachet material must withstand. That sachet is placed inside the food stuffs within an isolated system separated from the external atmosphere by the packaging that meets the requirements of specific permeability, which

Table 4. Materials present in antimicrobial sachets and their active components.

Sachets Material	Active compounds	References
Low-density polyethylene	Ethanol	(Hempel, Papkovsky, and Kerry 2013)
Linear low-density polyethylene	Allyl isothiocyanate	(Ma, 2012)
Nonwoven fabric	Allyl isothiocyanate	(Otoni et al. 2014)
	Oregano essential oil	(Passarinho et al. 2014)
	lemongrass essential oil	(Espitia et al. 2012)
	cinnamon essential oil	(Espitia et al. 2011)
Paper/Ethylene-vinyl acetate copolymer laminate	Ethanol	(Day, 2008)
Paper/Polypropylene laminate	Allyl isothiocyanate	(Sekiyama et al. 1995)
Polystyrene tray + barrier film	Chlorine dioxide	(Ellis et al. 2006)
Polystyrene tray + Ethylene-vinyl acetate film	Garlic essential oil	(Ayala-Zavala and González-Aguilar 2010)

depend upon each application, to redesign the gas composition inside the free space of the packaging.

External packaging materials

Depending on the sachet purpose, various needs must be observed for the sachets to be effective in the free space of the food packaging. For this purpose, packaging that has scavengers of oxygen, for example, is needed to allow permeability of oxygen not higher than 20 mL; otherwise, it will result in saturation of the soaking material and return of the oxygen concentration to the atmospheric level (Cruz, Camilloto, and dos Santos Pires 2012). Antimicrobial sachets are composed of varieties of antimicrobial material inside. Table 4 describes some materials which are present in the antimicrobial agents along with the main active component in these materials which mainly acts as bacteria again.

Some antimicrobial compounds have a strong and foul taste and smell, and thus the acceptance food packaging may be impaired. AITC has a very pungent odor that was undetected in the nylon lamination and in different polyethylene bags after one month of storage (Otoni et al. 2016). AITC may also diffuse from the internal packaging to the outer environment due to this behavior. It may also stop or decrease the growth activity of microbes, and does not affect the consumer acceptance at the moment of opening the packaging (Otoni et al. 2016).

Direct coating of antimicrobials on the polymer film

The direct coating is also another convenient method. Thus far, we have achieved the release of antimicrobial agents that are edible and established coating on the surface of food in order to stop the growth of microbes. These also prove to be a hurdle in the presence of moisture and oxygen. We hypothesize that the production of cost-effective and low-waste packaging that protects the food is the reason for the general acceptance of edible coatings. Researchers investigated vegetative materials having hydrocolloid coatings and reported the production of organized ways of selecting edible coatings to increase the shelf life and the quality of freshly harvested fruits and vegetables. There are different categories of components of edible films and coatings, but three of them are very common: lipids, hydrocolloids, and composites. Polysaccharides and proteins, such as alginate, starch, chitosan, cellulose derivatives, and agar, are included in the hydrocolloid category. Acylglycerols, waxes, and fatty acids are included in the lipid group, and materials having both hydrocolloids and lipids are composites (Severino et al. 2015).

Polysaccharide-based films and coatings

Polysaccharides are permeable to O_2 and CO_2 thus, they delay the growth and respiration mechanisms of many fruits and vegetables by reducing the availability of O_2. Polysaccharides are harmless and widely available. Researchers have studied the formation of films and the characteristics of different polysaccharide materials (Severino et al. 2015). Coatings and hydrophilic films, e.g., based on polysaccharides provide good retardation to CO_2 and O_2 under some specific conditions, but are poor barriers to water vapors. Hence, by stopping the supply of O_2, these materials delay the ripening of vegetables and fruits. The reduced water vapor barrier property allows for the movement of water vapor through the film, thus, preventing water accumulation and condensation that can be a potential source of microbial damage in horticultural commodities (Malhotra, Keshwani, and Kharkwal 2015).

Absorptive antimicrobial packaging

Freshly cut or minimally processed foods like meat, poultry, vegetables, and fruits are characterized by the formation of exudates during the storage period. The liquid exuded from the food products may harm the sensorial properties of the packaged foods as well as the microbial quantity in these foods (de Azeredo 2013). The absorbent pads are thus used to overcome this problem and show exclusive results. They are generally used at the bottom of plastic trays or containers to accumulate the exuded liquids from food products during the storage period. Food products placed in this packaging system include fresh meat, poultry, and fruits, among others. According to one study, absorbent pads must meet five specific requirements in order to preserve the integrity and quality of packaged food product successfully:

a. The position of the absorbent pad must be maintained, and in most packaging systems it should be placed horizontally. The absorbent pad must have a different structure having no exuded liquid.
b. The absorbent pad must retain the liquid inside until the packaging is opened, without spilling the liquids in the packaging. Moreover, the pad should be detectable

physically and visually. Thus, the weight of the packaged food should also be maintained.

c. The absorption pads should provide sensorial packaging of food products and should provide a visual presentation, and its use should be affordable concerning the cost of food.
d. The absorbent pad should contribute to the increase of the shelf life of the packaged products and the inhibition of microbial growth.
e. The absorbent pad has different resistances to allow its use at high-speed automation, when possible and where appropriate (Otoni et al. 2016).

Absorbent pad architecture

Commonly, absorbent pads are divided into three layers. The layering structure is defined according to the capillary action mechanism. These layers—upper, middle, and lower layer—are made of an absorbent material. In few cases, the upper and lower layers consist of an impermeable thermoplastic material or are covered with a cellulose-like material; in particular, this different type of cellulosic material can be in direct contact with the food products having adsorbent pad.

Absorption pads are extensively used in food packaging and possess significant advantages. There are small pores made on the surface of the upper and lower layers, and one of these layers must have absorption ability, so this complete structure improves the capillary action. Before, there were few complications observed in pads placed inside the package. In these cases, the pads were not working correctly as they were not absorbing the liquid or moisture released by the food inside the package. Thus, these pads were made with special precautions, for example, specific perforations with proper distance were made so that they would not release their material and could absorb the liquid and moisture from the food properly. There are a variety of materials used for the layers of the absorbent pads so that it works efficiently and get a good impression in the absorbent pads (Lloret, Picouet, and Fernández 2012).

Volatile antibacterial packaging

Volatile compounds are a vital component in volatile antimicrobial packaging. They have low-molecular weight and can easily convert into vapor at normal atmospheric pressure. Volatile organic compounds (VOCs) generally have low- to medium solubility in water and also have a unique smell. There are two major types of VOCs depending upon their source of origin. First, in VOCs obtained from plants (aldehydes, alcohols, essential oils) and VOCs obtained from microorganisms (fungi, yeast, bacteria).

Currently, an increasing number of fungi is shown to produce different VOCs of different molecular sizes and specific numbers, and types. There are variable amounts of VOCs, according to most studies performed with modern analytical equipment. This vapor-based mixture may carry alcohols, aldehydes, aromatics, esters, acids, ketones, heterocycles, thiols, terpenes, and others. Furthermore, this sophisticated cocktail is constant for a limited period, and varies with temperature, environmental variables, and substrates for each species. Typically, VOCs represent a multiplex mixture of lipophilic compounds having low-molecular weight, which are obtained from different biosynthetic pathways. Recently, a term—"Volatile"—was proposed to explain their multiplexity plants having different phenotypes have fruits and leaves that contain significant components of VOCs, which are released into the environment, soil, and the roots.

During recent studies, the antimicrobial activity of microbial volatile organic compounds (MVOCs) and VOCs was extensively studied and tested. If we work to create different programs and strategies to obtain VOCs from different plants, then the disease spread, and disease outbreaks can be controlled. Sometimes, the antimicrobial activity is seen in several pathogen–host interactions. They are extremely attractive because of their microbial activity and extremely toxic at low concentrations (Sivakumar and Bautista-Baños 2014).

Challenges in the development of food packaging

Despite all the above applications of antibacterial agents, there are some challenges and limitations which should be discussed and overcome. One of the main reasons is generally the insufficient compatibility between the polymer base matrix and antimicrobial agents as sometimes they are not adequately incorporated in the polymer matrix. Another reason is may be the irregular migration of antimicrobial agents to target microorganism and the rising concern of innovative food packaging also leads to the limitations in the progress of antimicrobial food packaging (Mlalila et al. 2018). Some limitations are discussed in the following sections.

Action mechanism of antimicrobial agents

There is no exact practical study which shows the exact mechanism of action of antimicrobial agents. However, we have a theory which shows that there are specific antimicrobial agents for the specific species of microbes such as bacteria, virus, and others are effective for yeasts and fungi. It is suggested that there is a specific action of mechanism for antimicrobial agents according to which they involve in the destruction of cytoplasmic membrane of cells thus change the structure and morphology of microbial cell walls. They mainly promote plasmolysis and disintegrates the cell wall. Due to this action, the cell ion gradients balance is dissipated, and the leakages of intracellular constituents occurs which are proteinaceous and thus inhibit the process of respiration. All this progression profoundly affects the functioning of ATPase enzyme, this enzyme is responsible for the function of transportation, mainly nutrients transportation across the microbial cytoplasmic membrane and these nutrients are key element for the proper synthesis of structural component and energy production in the microorganisms (Kuuliala et al. 2015).

Cost of production

The number of antimicrobial agents used in the lab is in a way more affordable because they were applied only in lab-scale. However, when the same agents are applied on the industrial scale, they are ten times more in number to be useful as the original one. The antimicrobial agents are expensive in industrial scale thus are very inconvenient to produce on an industrial level (Kim and Rhee 2016; Kuuliala et al. 2015; Moon and Rhee 2016).

It was also observed that if we buy any antimicrobial agents for specific food it cannot provide the same results for another type of food, that's why it is also costly to buy several antimicrobial agents for several types of food. Besides, many natural antimicrobial agents are quite expensive to use as compared to synthetic ones. Also, they take too much time to diffuse in the polymer matrix and have long production cycle and limited availability which makes them incompatible while using (Rezaei et al. 2016).

Foul odors

Natural antimicrobial agents like EOs possess a high intensity of off-flavors. Some examples are linalool, carvacrol, clove oil, thymol, ginger/garlic oil, basil, and cinnamaldehydes. They show high antibacterial properties but have an awful strong smell and flavor which inhibits the original flavor of food and it is one of the most critical challenges which the food industry is facing these days. Along with this off-flavor, EOs also carries a intense color. About 85%–99% EOs consists of phenolic and hydrophilic volatile terpenoids which contains a generation of intense reddish color to the films and have sharp flavors which limits their applications in the food packaging industry constituents (Bhullar et al. 2015).

Conditions during processing

There are so many antimicrobial agents which are not able to bear the harsh condition during the processing of antibacterial packaging like high temperature. Many promising agents like enzymes and some natural essential oils denature at elevated temperature and high pressure. On the other hand, various bacteriocins and enzymes are not stable under the processing of packaging material.

Many governing bodies universally are not welcoming the active and intelligent food packaging due to absence of rheostat frameworks. All these limitations should be kept in mind so that we should not only plan the antibacterial packaging on lab-scale but also make it possible to imply on the industrial level because our focus is not only to make new and innovative antibacterial packaging but also the packaging which is cost-effective and compatible to apply on an industrial level so that food security is improved (Mlalila et al. 2018).

Conclusion

As the world population increases, the demand for food is also increasing. To overcome this direct proportional relationship between human population and food, innovations aimed at increasing the production of food are in focus. Results showed that antimicrobials are the central part of the packaging for long ago. Different innovative approaches should be implemented to ensure food safety and to control food spoilage and wastage of food by improving the packaging material. Currently, food packaging is the only element that helps to provide safe and secure food for all. Besides, there are some other technologies which are also very useful in the food packaging industry. Electric discharges of high voltage, high hydrostatic pressure, high light intensity, ionization radiation, the addition of bacteriocins, and ultrasonication with high heat and pressure are some of the techniques from the list of new technologies that are very useful. Nevertheless, we must focus on the most compatible and practically possible technique which can be implemented on industrial scale. The focus of the researches now is on the methods of food delivery, which are free from additives, are of higher quality, but have natural constituents and higher nutritional value which is also the main helping point to fulfill the problem of food safety. This review will provide information about antimicrobial polymer materials and some important facts of these materials. So that, researchers will get their hands on a valuable information.

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