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Recent advances in biopolymeric antioxidant films and coatings for preservation of nutritional quality of minimally processed fruits and vegetables

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ABSTRACT

Minimally processed F&V while being as fresh as the intact product, are characterized by an accelerated produce decay which affects its nutritional value during shelf-life. In this sense, food processing needs to further evolve in terms of better preservation of nutritional properties. Active packaging technology has shown positive and promising results to maintain safety and sensory properties of minimally processed F&V. This review aims to present the recent research results regarding biopolymeric antioxidant film and coating for preservation of nutritional quality of minimally processed F&V. The mechanism by which nutritional losses (around 5–30 % loss of ascorbic acid and phenolic compounds) occur from oxidation reactions in F&V and natural antioxidant have been discussed. Furthermore, regulatory aspects related to antioxidant packaging have been also reported. Biopolymers based antioxidant film and coating have been vastly used to pack F&V product. Chitosan, gelatin, casein and alginate were found to be more effective as packaging materials (both as coating and as film) to preserve the nutritional and sensory quality of F&V product. Furthermore, plant extracts (green tea and Aloe vera), essential oils (lemon grass), plant oil compounds (eugenol and citral) and phenolics (thymol) as a component of active film or coating systems have shown promising results in preserving the quality of fresh produce. The collected findings will be useful to accurately design an innovative active film or coating for nutritional quality preservation of minimally processed fresh fruits and vegetables.

1. Introduction

Fruit and Vegetables (F&V) are an important source for providing humans with essential nutrients and bioactive compounds, including vitamins, organic acid, carotenoids, minerals, fiber, and polyphenols. Minimally processed F&V has been developed to offer convenience and functionality to consumers while ensuring food safety and quality. However, although conventional food processing methods have been demonstrated to preserve the freshness of the products, the shelf-life of minimally processed F&V is still limited. Moreover, food processing needs to further evolve in terms of better preservation of nutritional properties while ensuring safety, tasty and sustainable food. One of the main causes of nutritional losses is oxidation. Besides loss of essential nutritive elements, other negative effects of oxidation include enzymatic browning and production of off-flavors (Garcia & Barrett, 2002; Rickman, Barrett, & Bruhn, 2007).

As the phenomenon of oxidation has great economic impact, the food industry is actively looking for new technologies (i.e., direct addition of antioxidants into food products or development of suitable packaging material) to minimize the influence of oxidation. Oxidation reactions can be controlled by reducing the oxygen content by modifying the environment, however, this is only partial because F&V need oxygen to avoid anaerobic conditions that in turn limits the shelf-life of the product (Bhardwaj, Alam, & Talwar, 2019; Ghidelli & Pérez-Gago, 2018; Ribeiro-Santos, Andrade, de Melo, & Sanches-Silva, 2017; Sanches-Silva et al., 2014). Active antioxidant packaging is a novel alternative to these techniques, which restricts the oxidation through sustained release of antioxidants or incorporation in the packaging of an oxygen or radical scavengers. Several antioxidants have been authorized by European Union, under Directive 2006/52/EC (European

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Commission, 2006). Although natural antioxidants (i.e., caffeic acid, catechin, quercetin, gallic acid, curcumin, and α-tocopherol) incorporated into food packaging have been reported in literature, still many of the antioxidants are artificial, such as propyl gallate (PG), butylated hydroxytoluene (BHT) and butylated hydroxyanisole (BHA) and their use is mainly limited to processed food products (Sanches-Silva et al., 2014). Antioxidants can be used for incorporation into food packaging and are released by a controlled diffusion mechanism (Mastromatteo, Mastromatteo, Conte, & Del Nobile, 2010). Thus, antioxidant active packaging may hinder polyphenol and vitamin oxidation to maintain nutritive and sensory attributes of fresh F&V (Pereira & Abreu, 2018).

While most of the literature available has focused on the effect of antioxidant packaging on lipid oxidation, mainly for animal origin food, still there is limited information about the impact of antioxidant packaging on the quality of fresh and minimally processed F&V during shelf-life. Moreover, environmental requirement has led researchers to work on new sustainable biobased or biodegradable materials as suitable alternatives to petrochemical based conventional packaging materials (Qamar, Asgher, Bilal, & Iqbal, 2020). In the past decade, biodegradable materials such as biopolymers (i.e., proteins, polysaccharides and lipids) have gotten the attention of researchers due to their unique properties to preserve the nutritional and sensory quality of F&V by using antioxidants in food contact edible coatings/or films (Hanani, Roos, & Kerry, 2014; Kadzińska, Janowicz, Kalisz, Bryś, & Lenart, 2019; Khalil et al., 2018).

Thus, this review was aimed to give an overview about the impact of antioxidant packaging on preservation of quality of minimally processed F&V during shelf-life. Recent advances on active film or coating based on biopolymer and natural antioxidant have been presented, describing the types of antioxidant packaging and the strategy to design active packaging systems. Furthermore, regulatory aspects related to antioxidant based active packaging have also been explored.

2. Loss of nutritional quality of minimally processed F&V

Fruit and Vegetables (F&V) are key elements of a healthy and balanced diet providing humans with essential nutrients and bioactive compounds. Before minimally processed F&V are consumed, they have to undergo various handling, storage, and processing steps which induce substantial nutritional losses. Furthermore, minimal processing

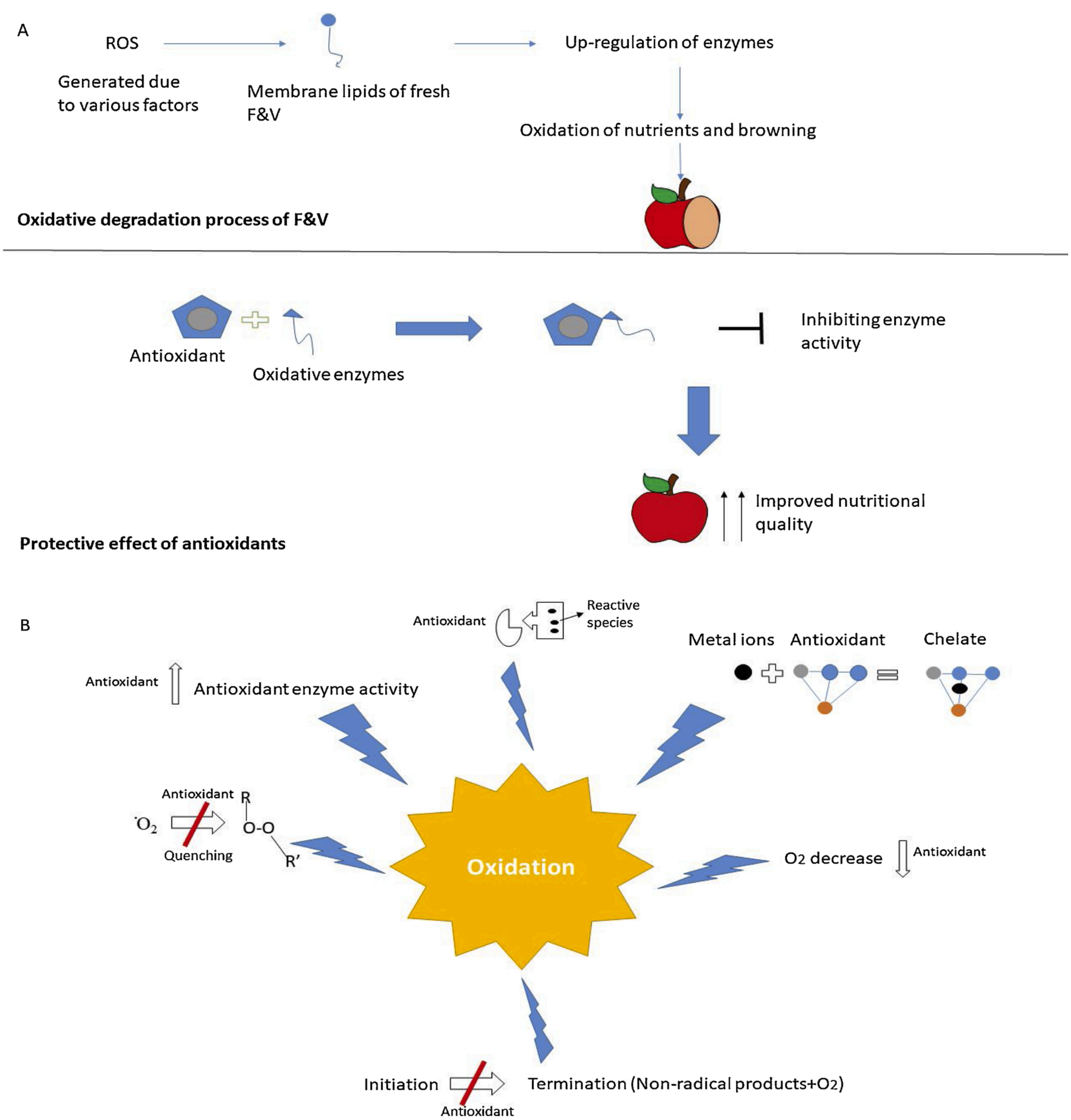

Fig. 1. (A) Mechanism of nutritional losses in F&V and protective action of antioxidants (B) mechanism of action of antioxidants.

operations can affect the content of those bioactive compounds that are susceptible to degradation when exposed to oxygen or light. Generally, oxidation in fresh F&V starts with the production of reactive oxygen species (ROS) as a result of various processes (i.e., respiration, photosynthesis and oxidative burst) occurring within different cellular organelles at various cellular locations inducing decay of the product and loss of nutritional quality (Del Río & López-Huertas, 2016; Valenzuela et al., 2017). These generated ROS can then react with membrane lipids of the fresh F&V, leading to membrane degradation due to peroxidation (Fig. 1A).

Degradation can be promoted also by the activity of oxidative enzymes such as ascorbate oxidase, polyphenol oxidase, cytochrome oxidase and peroxidase. The vitamin C content of sliced and bruised fruits and vegetables may diminish rapidly depending on handling, processing, and storage conditions used. Ascorbic acid can get oxidized and degraded easily when exposed to solar light, oxygen, oxidative enzymes and in the presence of metallic catalysts (Fe^{+3}, Ag^{+}, and Cu^{+2}). Ascorbic acid is often considered to be equivalent to vitamin C content; however, dehydroascorbic acid (DHAA), the oxidized form of ascorbic acid, also has vitamin C activity (Perera, 2007). Further oxidation of DHAA converts it to 2, 3-diketogulonic acid, which is devoid of biological activity and is a useful indicator of oxidative deterioration in minimally processed F&V (Perera, 2007). Dokhanieh, Aghdam, and Sarcheshmeh (2016) reported that freshly cut pomegranate arils suffer from reduced nutritional quality and low marketability when stored at refrigeration temperature due to the fact that ROS can reduce membrane integrity and disrupt compartmentalization between cells, as a result polyphenol peroxidase comes in contact with phenolic substrates produced by phenylalanine ammonia-lyase enzyme, which leads to the production of brown polymers, ultimately reducing nutritional quality due to oxidation of phenolics and fatty acids. Galani, Mankad et al. (2017), Galani, Patel, Patel, & Talati (2017) observed the influence of refrigeration storage on the vitamin C, total phenolics and anthocyanin contents of different F&V. The authors reported a significant decrease in the content of vitamin C for cabbage (25 mg/100 g fw), spinach leaves (~22 mg/100 g fw), tomato (15 mg/100 g fw; with a significant loss of 71.8 %), potato (~12 mg/100 g fw) and apple (~15 mg/100 g fw) after 15 days of storage due to alteration in expression of genes and activity of oxidative enzymes during storage. Furthermore, this enzyme activity and gene expression have been positively correlated with low temperature (Galani, Mankad et al., 2017, 2017b). SHI, Bassa, Gabriel, and Francis (1992) reported the degradation of anthocyanin pigment in sweet potatoes due to enzyme systems (peroxidases, or polyphenol oxidases) present in the tissues. A decrease in provitamin-A precursor (α-carotene, ß-carotene, and ß-cryptoxanthin) values was observed during frozen storage of a variety of minimally processed F&V (mango, kiwi, papaya, and tomato) due to the activation of polyphenol oxidase, catalase and lipoxygenase enzymes in the presence of oxygen (Perera, 2007). Another important group of bioactive compounds that are lost during minimal processing of F&V are polyphenols which are easily oxidized by polyphenol oxidases into quinones (Rahman, 2007). Bunea et al. (2008) observed a decrease (20 %) in total phenolic compounds at different storage temperatures (−18 °C and 4 °C) due to enzymatic degradation of the spinach. Similarly, Preczenhak, Tessmer, Berno, de Abreu Vieira, and Kluge (2018) reported significant losses (almost 1/5th of the initial content) of bioactive compounds (i.e., betacyanin, betanin and phenolic compounds) during the initial stages of minimal processing of red beets due to increased polyphenol oxidase content (303.3–461.58 microkatals/kg) because of oxidative stress. Although quantitative data on nutritional quality loss is well reported, however, information about mechanisms involved and kinetics is not systematically reported. Nevertheless, this information is important to accurately design and predict the requirements of an antioxidant packaging able to maintain and preserve the quality of F&V.

3. Types of antioxidants

Antioxidants can be classified as hydrophilic (water soluble i.e., glutathione, uric acid, ascorbic acid and lipoic acid) and lipophilic (lipid soluble i.e., ubiquinol and carotene) based on their solubility in a solvent (Mishra & Bisht, 2011; Radenkovs & Feldmane, 2017). Generally, lipophilic antioxidants inhibition lipid peroxidation and protect cell membrane, while hydrophilic antioxidants react with blood plasma oxidants (Grażyna, Hanna, Adam, & Magdalena, 2017). Based on the origin of antioxidants, they can be classified either as synthetic or natural. Synthetic antioxidants are chemically synthesized since they do not exist in nature and can be classified on the basis of their mechanism of action into two major groups, primary and secondary antioxidants (Ehsani, Solouk, & Mardafkan, 2018). Primary antioxidants can convert peroxyl radicals by donating a hydrogen atom into stable compounds through rapid reaction; while secondary antioxidants yield products devoid of any biological activity after reacting with hydro peroxides. Moreover, secondary antioxidants can inhibit oxidation by absorbing UV radiation, decomposing hydro peroxides, inhibiting enzymes, binding with the metal ions that are able to catalyze oxidative process (Hidalgo & Zamora, 2017; Mishra & Bisht, 2011). Antioxidants of synthetic nature, such as BHT, PG, and BHA, are commonly used in food products. Nonetheless, recent studies have indicated towards minute carcinogenic effects of these antioxidants in animals at elevated concentration (Aziz & Karboune, 2018; Brewer, 2011). These findings combined with consumer demand for natural additives have encouraged the researchers to explore natural alternatives. Natural antioxidants can not only act as a preventive medicine against various human diseases but can also extend the shelf-life of fresh food products (Aziz & Karboune, 2018).

The majority of natural antioxidant compounds are phenolic compounds (with flavonoids, phenolic acids and tocopherols being the most important) and are those agents that play an essential role in restricting autoxidation by impeding the formation and propagation of free radicles by following mechanisms: (1) metal ion chelation, (2) reducing local oxygen content, (3) interrupting the chain reaction for auto-oxidation, (4) hindering the production of peroxides by capturing $\dot{O}_2$ radical, (5) activating the enzymes having antioxidant potential, and (6) capturing radicals that stimulate peroxidation (Fig. 1B) (Aziz & Karboune, 2018; Papuc, Goran, Predescu, Nicorescu, & Stefan, 2017). The antioxidants which can interrupt the free radical chain reaction are the most effective ones. They usually contain one or more aromatic rings (usually phenolic) with one or more than one O—H groups and during oxidation reactions have the ability to donate $\dot{H}$ to the free radical produced to become radical themselves (Brewer, 2011; Yoo, Lee, Lee, Moon, & Lee, 2008). Flavonoids, via two mechanisms display their antioxidant potential, a) by chelating metals ions and b) by scavenging free radicals, on the other hand, phenolic acids trap free radicals (Brewer, 2011). Phenolic antioxidants have the ability to hinder the oxidation of vitamins and other nutrients in F&V and can be used for their preservation. Natural antioxidants can be found in almost all the fruits and vegetables. Table 1 shows the most common natural antioxidants and their typical sources. Generally, antioxidants can prevent oxidation in fresh F&V by reducing oxidation promoting enzyme, due to the hydrogen bonding of hydroxyl groups present in antioxidant compounds with the oxidation promoting enzymes, reducing the risk of oxidative stress and preserving their nutritional quality (Chaemsanit, Matan, & Matan, 2018; Murmu & Mishra, 2018).

Plant extracts (blueberry, strawberry, cranberry, tea, grape seed, etc.), spices (ginger, garlic, nutmeg, black pepper, clove, cinnamon, etc.), herbs (sage, oregano, rosemary, basil, marjoram, etc.) have been reported to contain antioxidant components (Yanishlieva-Maslarova & Heinonen, 2001). The bioactive antioxidative components of spices and herbs can be concentrated as resins, essential oils or extracts (Brewer, 2011). On the other hand, type of solvent and extraction method are two of the parameters on which antioxidant activity of essential oils depends (Aziz & Karboune, 2018; Tongnuanchan & Benjakul, 2014).

Table 1
Some natural antioxidant compounds and their sources.

Compound	Nature based on solubility	Sources
Lycopene	Hydrophobic	Papaya, guava, red grape skin, tomatoes, melon, watermelon, and pink grapefruit
Anthocyanins	Hydrophilic	Cherries, berries, grapes and fruit derived beverages
Ascorbic acid	Hydrophilic	Citrus fruits, tomatoes and some vegetables
Beta-carotene	Hydrophobic	Papayas, spinach, apricot, parsley, kale, tomatoes, potatoes, carrots and red paprika etc.
Tocopherols	Hydrophobic	Cooking oils (sunflower, olive and safflower oils), broccoli, cereal grains, hazelnuts, almonds, and cauliflower
Co Q10 (ubiquinone)	Hydrophobic	Nuts, fruits, some vegetables, fish, meat and some oils
Polyphenols	Hydrophilic/ hydrophobic	Purple/red hued vegetables and fruits such as blackberries, blueberries, concord grapes etc., green tea, coffee, wine, nuts and olive oil
Flavonoids	Mostly hydrophobic	Olive oil, onion, broccoli, kale, apples, cherries, red wine, tea, grape fruit, fruit skins, tomato skin, soya beans, legumes, lemons, thyme, parsley, red pepper, and oranges

Furthermore, the antioxidant capacity of the extract is due to the presence of volatile oils (i.e., thymol and menthol, etc.,) flavonoids (i.e., catechin and quercetin), and phenolic acids (caffeic, protocatechuic, rosmarinic, and gallic acids) as active components.

4. Biopolymers

Nowadays, researchers have shifted their attention towards biopolymers derived from agro-livestock resources for the development of packaging materials as an alternative to plastic-based packaging derived from petro-chemical origin which poses a public health threat due to its non-biodegradable nature and disposal problems (Mangaraj, Yadav, Bal, Dash, & Mahanti, 2019). The term biopolymer means “the natural polymers that are produced by the living organisms” (Adeyeye et al., 2019). The biopolymers commonly used for the development of packaging materials are edible in (Adeyeye et al., 2019) nature i.e., protein, polysaccharides and lipids (Krasniewska & Gniewosz, 2012). A wide range of proteins (i.e., gelatin, corn zein, soy protein, pea protein, wheat gluten, casein and sunflower), polysaccharides (i.e., starch, gum, pectin, chitosan) and lipids (i.e., bees wax, oils, and fatty acids) have been used to develop bio-based packaging systems (Porta, Sabbah, & Di Pierro, 2020). These natural polymers have the ability to effectively form good coating/film with cohesive structure and are able to form protective layer around the food product (Krasniewska & Gniewosz, 2012). Protein-based packaging materials generally possess better mechanical, structural and barrier properties (strong oxygen barrier) as compared to other biopolymer-based materials due to chain-to-chain interactions (hydrophobic, covalent and hydrogen bonding) (Hanani et al., 2014). The films and coatings obtained from biopolymers can not only maintain the quality of food products but can also extend the shelf-life of the produce by controlling transfer of moisture and gases, preventing loss of essential compounds and reducing microbial contamination, furthermore these biomaterials can also serve as a carrier of bioactive compounds i.e., antioxidants and antimicrobials thereby maintaining the sensory and nutritional quality of the packaged product (Otoni et al., 2017).

5. Active antioxidant packaging

Active packaging is a system in which constituents are included intentionally to perform two basic functions, a) scavenging or releasing substances from or into the packaged food or b) maintaining the freshness of the packaged food or the environment surrounding the food product to improve its shelf-life (Hanani, Yee, & Nor-Khaizura, 2019).

Antioxidant packaging works by scavenging and releasing mechanisms; for instance, by absorbing undesirable compounds i.e., molecules/ions, radical oxidative species and oxygen or by releasing antioxidants from package headspace into the food products. Scavengers are those compounds which can trap, change and bind with any substance (i.e., having tendency to initiate or help in progression of oxidation reactions). The packaging material should be developed in a way that it gives permit to oxidative compounds to the position where scavengers are present (i.e., because scavengers are not released into food products) (Gómez-Estaca, López-de-Dicastillo, Hernández-Muñoz, Catalá, & Gavara, 2014). In the case of releasing system, an antioxidant is entrapped in the package material and can be released to the food by direct contact between packaging material and food, when the antioxidant is non-volatile compound or by releasing of the volatile compound in the headspace and successive solubilization in the food. One of the main advantages of incorporating antioxidants into packaging rather directly into food products as additives is that antioxidants are released at a fixed controlled rate from the active substance, which serves as a reservoir of antioxidants, making up for their constant use during storage (Mastromatteo et al., 2010).

The antioxidant activity of the active film depends on different aspects: the type of antioxidant compound, the concentration of the antioxidant incorporated into the film and the release kinetics of the active compound from the packaging (Sanches-Silva et al., 2014). A nonlinear behavior between the active compound concentration and the antioxidant activity has been reported in literature (Bentayeb, Vera, Rubio, & Nerin, 2009). This could be justified by the interaction of the active compound with the film matrix or by a loss of the active compound during the film realization. The release from active packaging system is influenced by three parameters: the diffusivity of the active compound in the packaging material (D_i), the affinity of the active compound between packaging and the food product, described as partition coefficient (K_{pf}), and diffusivity of the active compound in the food product or the mass transfer coefficient (k) (Martinez-Lopez, Peyron, Gontard, & Mauricio-Iglesias, 2015; Vilas, Mauricio-Iglesias, & García, 2020; Vilela et al., 2018). The effectiveness of an antioxidant compound after release can be determined by its solubility attributes, and thus antioxidant type should be picked as a function of type of food we intend to pack (Cotabarren et al., 2019). Moreover, apolar antioxidants are unsuitable for low lipid content containing foods (i.e., F&V). However, the phenomenon of “antioxidant paradox” should always be taken into consideration. The antioxidant paradox is usually referred to the observation that antioxidants can work in more than one way anticipated in complex food systems, since antioxidants can influence oxidation kinetics in multiple ways (Decker et al., 2017). For instance, polar antioxidants i.e., catechin are effective in oil-in-water emulsions even though they are hydrophilic in nature (Zhou & Elias, 2012). It has been reported that hydrophobic free radical scavengers (FRSV) are less efficacious antioxidants as compared to hydrophilic FRSV in mass oils, on the other hand, hydrophilic FRSV are less effectual in emulsified oil, due to the reason that non-polar and polar FRSV have the tendency to condense in the oil phase of emulsions and oil-air interface of bulk oils respectively where oxidation was widespread (Aguirre-Joya et al., 2018; Decker, 1998).

5.1. Form of antioxidant packaging

Generally, antioxidant-based systems are produced by using three different methodologies i.e., non-package bound (independent antioxidant devices) and package bound (antioxidant packaging materials and coatings):

i) Non-package bound system: This system was the first packaging system that was introduced in the market and involves the inclusion of an independent device i.e., sachet, label, membranes or pad containing the agent apart from the food product to a passive packaging system. These independent devices contain oxygen scavengers (Baldino, Cardea, & Reverchon, 2017; Fang, Zhao, Warner, & Johnson, 2017) (Fig. 2). Fine powders of ferrous and iron oxide are the most prevalent oxygen absorbers, however, sulphites, ligands, catechols, polyphenols, ascorbic acid and some enzymes have also been used and reported in the literature (Baldino et al., 2017; Fang et al., 2017; Gómez-Estaca et al., 2014). To inhibit the early action of oxygen scavengers, specialized mechanisms can be devised to initiate absorbing reaction. For instance, to initiate removal of oxygen, humid conditions are necessary for iron-based scavengers (Lopez-Rubio et al., 2004). Extensive reviews have already been published on oxygen scavenging and its applications in food packaging (Brody, Bugusu, Han, Sand, & McHugh, 2008; Dey & Neogi, 2019; Gómez-Estaca et al., 2014; Rooney, 2005). Although, F&V need oxygen for their respiration, however, many oxygen sensitive fresh-cut or peeled F&V products i.e., potato products, peeled garlic, and fresh-cut lettuce are prone to oxidative browning caused by polyphenol oxidation, thus a low oxygen concentration (<1 %) is required for such type of food products, furthermore, the oxygen concentration should not be below the tolerable limit as it will lead to the development of anaerobiosis and malodorous compounds thus these oxygen scavengers are only suitable for oxygen sensitive food products and non-respiring products (such as meat) (Charles, Sanchez, & Gontard, 2003; Singh, Gaikwad, & Lee, 2019).

ii) Package bound system: active antioxidant agent is incorporated within the containers in which product is packed or into the packaging film walls, employing its activity by absorbing undesirable agents/compounds around the product from the headspace, or by releasing antioxidant compounds into the headspace surrounding the food products or directly to the food (Fang et al., 2017). The development procedure of antioxidant packaging material mainly depends on the traits of the antioxidant (i.e., mechanism of action and heat tolerance) and the polymer typology. If the mechanism of action of a material is based on the principal of migration of antioxidant compounds into the food product, the antioxidant compounds released should not only adhere to concerned regulations in terms of their maximum allowable limit but should also be permitted as food additives (Gómez-Estaca et al., 2014). When manufacturing an antioxidant packaging material, the antioxidant compounds or the reactive components involved are closely mixed with the polymer, either

a) by polymer melting, inclusion and blending of the antioxidant agent into the melt by using extrusion technology. This technology is favored since most of the standard packaging is either partly or entirely manufactured by extrusion technologies (Gómez-Estaca et al., 2014). However, a censorious drawback that should be kept in mind while using extrusion technology is the degradation of bioactive compounds by means of elevated temperatures during processing.
b) by immobilizing the antioxidant agent on the surface of the film (Fang et al., 2017) (Fig. 2); the only drawback of this technology is the activity of antioxidant is limited only to the food contact surface (Wu, Deng, Luo, & Deng, 2019).
c) by dissolving both (agent and polymer) into an appropriate solvent followed by applying solution to a substrate by coating technology. Casting is most frequently used manufacturing process for the development of antioxidant film, still, coating technology is not a standard procedure, thus, standardization of formulation conditions is highly required. Even so, polymeric dispersions and solutions are utilized in the manufacturing of coating on the surface of film through traditional printing technologies. Furthermore, the formulated film should be able to realize the following requirements: i) the validity of coating material for direct contact with food and good clinginess to the substrate of the film, ii) the packaging needs of food products especially related to their functionality, and iii) maintaining an effective antioxidant activity by regulating the release of antioxidant agent (Gómez-Estaca et al., 2014). Attachment majorly depends on the compatibility between the coating polymers and the substrate and can be promoted by chemical methods i.e., primers, physical methods (i.e., radiation and corona discharge) or a combination of both prior to coating procedure. The coating technology has several advantages i.e., sustained release of antioxidants and no requirement for expensive equipment;

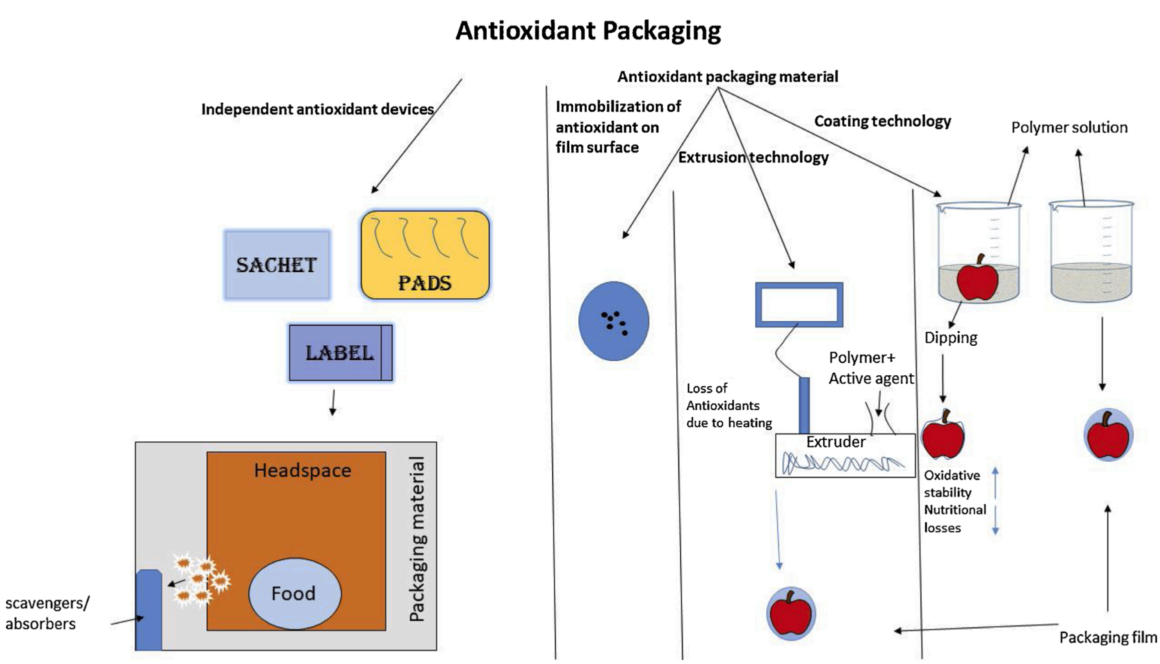

Fig. 2. Manufacturing of different antioxidant packaging systems.

however, its time consuming, labor intensive and can't be used on industrial scale unlike extrusion technology which can be used for bulk production (Gómez-Estaca, Gavara, Catalá, & Hernández-Muñoz, 2016; Hanani et al., 2014; Suhag, Kumar, Petkoska, & Upadhyay, 2020).

iii) edible coating: active biopolymer solution can be applied as coating materials for direct application on the food product by keeping in view their respiration requirements. It can function as barrier against gases or vapor or as carrier of active substances, such as antioxidant. The use of edible films and coatings as carrier of active substances has been suggested as a promising application of active food packaging (Han, 2003). The release of the active compound depends by the chemical interactions between the active substance and the film-forming materials and between the former and the environmental condition (Han, 2005). Moreover, the quantity of active compound carrier by the coating depends also on the final thickness of the coating on food that in turn depends upon the rheological properties of the film forming solution (Avena-Bustillos, Cisneros-Zevallos, Krochta, & Saltveit, 1993).

5.2. Literature review analysis

The quantity of literature published in ScienceDirect database (https://www.sciencedirect.com/) on antioxidant packaging and on different methods used for the manufacturing of active packaging over the last decade (2011–2020) is shown in Fig. 3. The advanced search string of ScienceDirect database was applied by using keyword

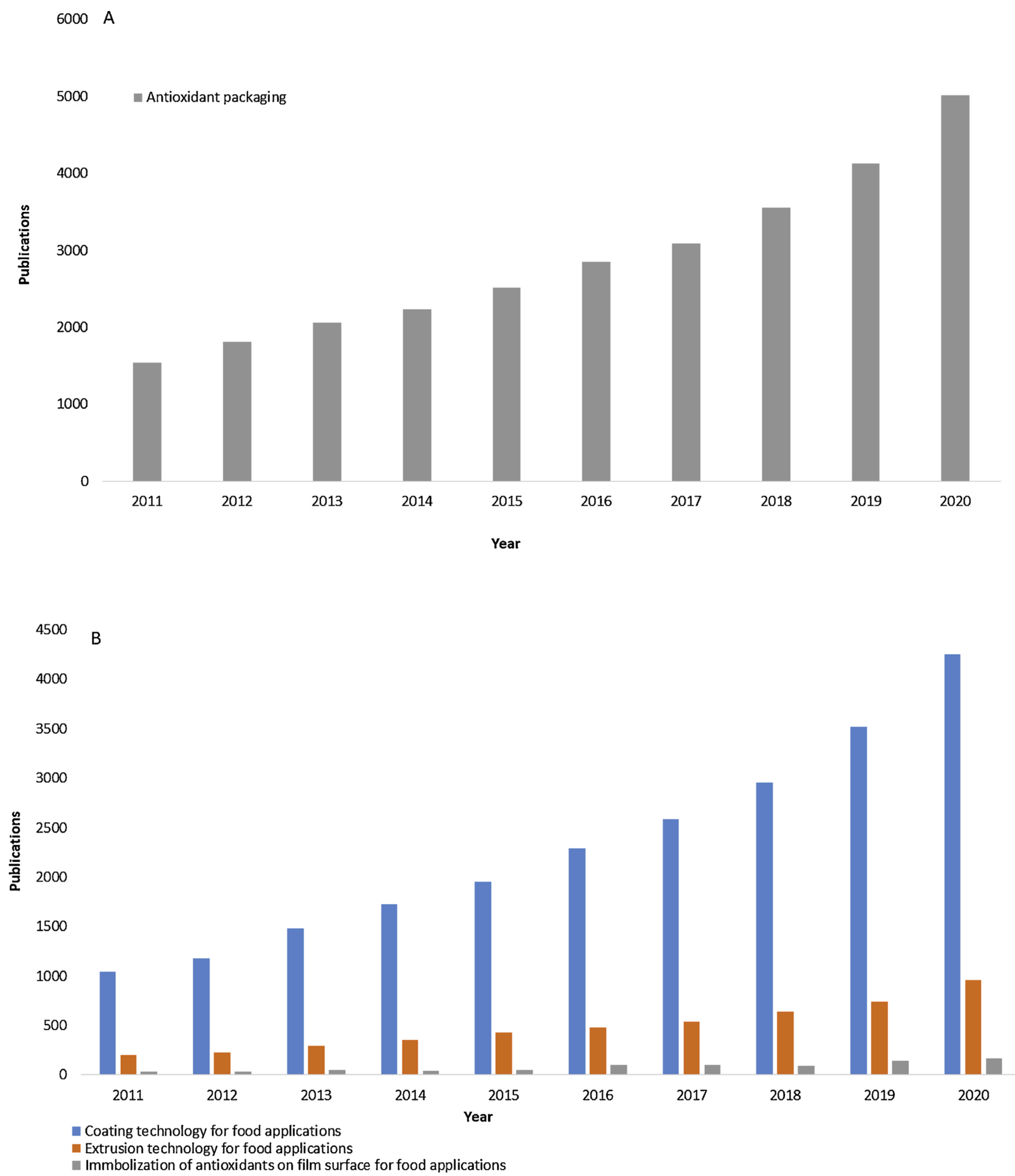

Fig. 3. Number of publications indexed by ScienceDirect based on advanced search related to use of "antioxidant packaging" (A) or specific manufacture technology (B) in the keywords and title of the publication.

"antioxidant packaging" to estimate the number of published research (research papers, short communications and patents) having this search term in their title, abstract or keywords (data retrieved on 01-20-2021). The research on antioxidant packaging is exponentially growing to meet the needs of food and packaging industries. The data indicate that the studies on antioxidant packaging has reached maturity. Similarly, to highlight which type of technology was used mostly in past decade to develop active packaging for food applications, the keywords "coating technology; food applications (for coating technology), extrusion technology; food applications (for extrusion technology), immobilization of antioxidants; film surface; food applications (for immobilization of antioxidants on film surface technology) in the search string (data retrieved on 01-20-2021). It was observed that coating technology was extensively used for the development of active packaging for food applications as compared to extrusion or immobilization of antioxidants on film surface due to its advantages over other two technologies (Fig. 3B). Additionally, its advantageous to study different methodologies for the development of active packaging forms as the technology/methods can have an impact on the nutritional quality index of F&V.

To highlight the influence of antioxidant packaging on the quality of fresh and minimally processed F&V product in the last decade (2011–2020), Preferred Reporting Item for Systematic Reviews and Meta-Analytic (PRISMA) method was used by selecting ScienceDirect database (Bush, Stevenson, & Lane, 2019). All the articles passed the selection process were reviewed and summarized based on the objectives, year of publication and research results. The inclusion criteria included: a) research on "antioxidant packaging" b) its "application on fresh and minimally processed F&V", c) published in the form of research article, short communication and or patent. The exclusion criteria involved: a) research on antioxidant packaging for their application on other food products or without any application, b) conference abstracts, review articles, and book chapters. The search process started by reviewing the titles and abstracts of entire search results and comparing them with established criteria. The database search in all keywords yielded 1431 search results, out of which 372 articles (research article and short communication) were obtained. Furthermore, references of shortlisted articles were also checked for any missing references and manual search was also conducted. Based on the criteria, only 15 articles were selected for discussing the application of antioxidant packaging on fresh and minimally processed F&V (Fig. 4) (data retrieved on 01-30-2021).

5.2.1. *Overview on antioxidant packaging and its application on F&V product*

A very recent trend in active antioxidant packaging is the reduction in the use of synthetic additives and their replacement to natural antioxidants in the past decade (Fang et al., 2017; Sanches-Silva et al., 2014). The utility of antioxidant active food packaging for fresh fruit to preserve deterioration of food quality, change in odour, color and taste are widely reported between 2011–2020 (Riva, Opara, & Fawole, 2020; Sciences, Nagar, Sciences, & Nagar, 2017). Zhang, Liu, Sun, Wang, and Li (2020)) reported the effect of chitosan/zein film enriched with 50 % of α-tocopherol on packaged mushroom (*Agaricus bisporus*) stored at 4 °C for 12 days; the results showed the relatively lower weight loss (<12.5 %), relative leakage rate, browning index (1.62-fold reduction than control), respiration rate, polyphenol oxidase (1- fold lower than control), peroxidase activity (4.96 folds lower than control) and malondialdehyde (MDA) content of samples packed with chitosan film and chitosan/zein film as compared to control samples. Rossi Marquez et al. (2017) showed the effect of pectin/protein films, prepared in the presence of transglutaminase, on the preservation of fresh cut apple, carrots and potatoes properties. The film was able to totally prevent the weight loss of potato and carrot samples during storage. The cross-linked blended film prevented microbial growth in all samples analyzed, and preserved phenolic and carotenoid content in carrots.

Murmu and Mishra (2018) prepared arabic gum, sodium caseinate coatings with cinnamon and lemon grass oil to improve oxidative stability of guava. It was reported that the uncoated guava samples exhibited roughly two times higher polyphenol peroxidase activity (i.e., 5.8 unit/mg protein) after seven days of storage as compared to coated samples. Coating created a modified environment around guava samples which reduces oxygen availability, restricting the rapid rise in polyphenol oxidase. Furthermore, Arrieta et al. (2020) prepared fruit extract (*Cucumis metuliferus*) loaded acetate cellulose coatings to coat corona treated LDPE films to develop bilayer packaging system. The active packaging system demonstrated the ability to delay the oxidation process of freshly cut apples as compared to control even after 3 days of storage. This could be due the fact that the respiration rates in fresh and minimally processed fruits and vegetables can be slowed down by means of active antioxidant package, thus maintaining and enhancing the quality of the produce (Sharma, Shehin, Kaur, & Vyas, 2019). Contrarily, some studies have reported an undesirable influence of antioxidant additives/extracts on F&V packed in the active packaging. For instance, Shemesh, Krepker, Nitzan, Vaxman, and Segal (2016) developed polyamide films with carvacrol (2–4 %) for preserving tomatoes. Authors reported that when the concentration of carvacrol was increased above 2 % in the packaging formulation it enhanced the fruit decay in both trials (58–100 %) due to tissue sensitivity and phytotoxicity. Similarly, Hashemi and Raeisi (2018) studied the influence of apricot gum coatings containing *Satureja intermedia* extract (1−3 %) on wild almond kernels. Authors reported an increase in peroxide (1.89–2.4 meq kg oil^{-1}) and free fatty acid values (~1.25−1.65 %) for kernels

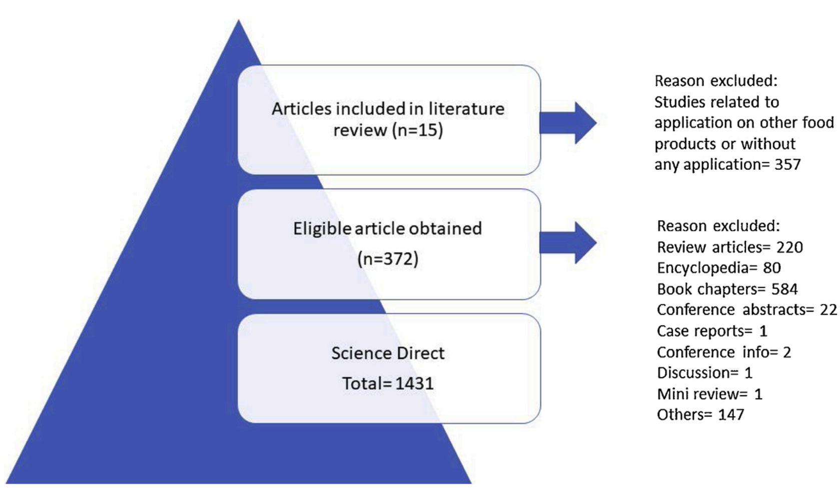

Fig. 4. PRISMA diagram for literature review.

stored in coatings containing extract throughout the storage period although less than the control. An increase in free fatty acids was due to chemical hydrolysis and intermediate enzyme activity. Table 2 also shows some studies to further highlight the influence of active antioxidant packaging materials on F&V. Thus, it can be concluded from the above literature review that the application of antioxidant package to preserve the nutritional quality of F&V is a phenomenon which still needs to be explored to achieve advancements in this field.

5.3. Methodologies used to evaluate the antioxidant capacity of an active film

There are many methods to measure the efficacy of antioxidant compound. Based on multiple aspects of antioxidants and inhibited autoxidation kinetic models, several antioxidant capacity assays were utilized. Generally, there are two different pathways through which antioxidants neutralize free radicals, depending on the possibility for side reactions and the kinetics: Single Electron Transfer (SET) and Hydrogen Atom Transfer (HAT) (Huang, Ou, & Prior, 2005). Procedures under SET can determine the efficacy of a possible antioxidant agent by transferring an electron to reduce carbonyl, metals and radical compounds. Potential for ionization and dehydronation of the reactive functional groups are used as basis for SET methods. One of the most important SET-based methods is Ferric Reducing Antioxidant Power (FRAP) assay, which estimates the reduction of 2, 4, 6-tripyridyl-s-triazine (TPTZ) into a colored compound (Guo et al., 2003). On the other hand, methods based on HAT estimate the capacity of an active substance to donate hydrogen for quenching free radicals and determination of reactivity is done by bond dissociation energy of potential antioxidant's hydrogen giving group (Apak, Özyürek, Güçlü, & Çapanoglu, 2016). It has been reported in literature that there are two important HAT mechanism utilizing methods namely: Total Radical-Trapping Antioxidant Parameter (TRAP) and Oxygen Radical Absorbance Capacity (ORAC). Additionally, some procedures use both SET and HAT mechanisms. These types of methods are quite important because both mechanisms can simultaneously occur in the food samples (Gómez-Estaca et al., 2014). Generally, 2,2-diphenyl-1-picrylhydrazyl (DPPH) and 2,2'-azino-bis (3-ethylbenzothiazoline-6-sulfonic acid) (ABTS) assays usually come under the umbrella of SET reactions. However, these two indicator radicals may be neutralized either by radical quenching via hydrogen atom transfer or reduction through electron transfer (Adilah, Jamilah, & Hanani, 2018). The DPPH and ABTS tests are routinely used for the assessment of free radical scavenging potential of an antioxidant molecule and considered as one of the standard and easy colorimetric methods for the evaluation of antioxidant properties of polyphenols into different matrix. The DPPH radical appears purple in color and can be evaluated spectrophotometrically at a wavelength of 517 nm, in methanol. DPPH accepting a hydrogen (H) atom from the scavenger molecule changes the color from purple to yellow with a consequent decrease in absorbance at 515 nm (Sharma &

Table 2
Influence of active antioxidant packaging materials on F&V.

Type of packaging	Antioxidant agent	Food product type	Major results	References
Alginate based coating	Ascorbic/citric acid	Fresh-cut Kent mangoes	Ascorbic acid content for samples was found to be two times lower in control (<15 mg/100 g f.w.) due to presence of oxygen as compared to active antioxidant package (>30 mg/100 g f.w.) at the end of storage period after 12 days	Robles-Sánchez, Rojas-Graü, Odriozola-Serrano, González-Aguilar, and Martin-Belloso (2013)
Alginate based coating	Lemongrass essential oil	Fresh-cut pineapple	Pineapple samples packed inedible coatings with lemongrass oil displayed L values (>45) higher than the control samples (~40) due to presence of ascorbic and citric acids as potential antioxidants.	Azarakhsh, Osman, Ghazali, Tan, and Adzahan (2014)
Alginate based coating	Essential oil compounds (eugenol and citral)	*Arbutus unedo fruit*	Coating materials containing0.15 % citral+0.1 % eugenol displayed best results in maintaining sensory and nutritional attributes of fruit samples by showing 970 μM TE/100 g antioxidant activity and lightness values (L*) of 37.39 as compared to control (546 μM TE/100 g and 32.44 respectively).	Guerreiro, Gago, Faleiro, Miguel, and Antunes (2015)
Soy-protein film/ coating	Ferulic acid	Fresh-cut apples	The packaging formulation at 7 pH with 4 g/l ferulic acid concentration demonstrated potential in extending shelf-life of apple samples by keeping browning index below 13 % as compared to uncoated samples (43.4 %) at the end of storage period.	Alves, Gonçalves, and Rocha (2017), Apak et al. (2016)
Papaya film	*Moringa* leaf extract + ascorbic acid	Minimally processed pears	Least browning observed in pear samples packed in papaya films containing ascorbic acid and *moringa* extract.	Rodríguez, Sibaja, Espitia, and Otoni (2020)
Polyamide film	Carvacrol	Cherry tomatoes, lychee, and grapes	A significant reduction in decay (50 %) was observed for tomatoes, followed by grapes (25 %), and lychee (17 %) at 2 % carvacrol concentration. However, any further increase in concentration of carvacrol led to an increase in decay percentage of tomatoes due to phytotoxic effect.	Shemesh et al. (2016)
Fruit and vegetable residue film	Mixture of orange, lettuce, passion fruit, mint, taro, spinach, carrot, rocket, watermelon, courgetti and cucumber residue flour	Minimally processed carrots	Packaging decreased weight loss and whiteness index of carrot samples.	Fai et al. (2016)
Multilayer coating (Chitosan and pectin)	Trans-cinnamaldehyde	Fresh-cut cantaloup	The coating effectively delayed post-harvest ripening process. Furthermore, a reduction in vitamin C content was observed from 0.39 to 0.224 mg/mL.	Martiñon, Moreira, Castell-Perez, and Gomes (2014)
Chitosan coating	Acetic acid	Peeled prickly pear fruit (white/red)	Chitosan coating with 1−2.5 % acetic acid did not significantly affected the phenolic content of white pear samples. The coating with 2.5 % acetic acid concentration reduced the overall acceptability of fruit (3.7−4.6).	Ochoa-Velasco and Guerrero-Beltrán (2014)

Bhat, 2009). On the other hand, in ABTS assay, the antioxidant activity of the active film is rapidly measured by generating ABTS radical cation by reacting a stock solution of ABTS with potassium persulfate, with absorbance adjusted between 0.7−0.01 at 734 nm. The drop in absorbance is typically measured after reaction time (from minutes to hours) (Schaich, Tian, & Xie, 2015; Yang, Lee, Won, & Song, 2016). One of the critical aspect in these methods is the contact time of the active compound with the radical. Cuvelier and Berset (1995) focalized the attention on the interaction kinetics of polyphenols in contact with DPPH radical in which authors classified single polyphenols under the categories of fast (<30 min), intermediate (30 min to 1 h) and slow (>1 h), as function of the time needed to reach the steady state when in contact with DPPH radical. If the contact time with the DPPH radicals is not respected, the antioxidant molecules will not able to express all the antioxidant activity in the reaction and will remain unused. However, most of the methods mentioned above, are global and non-specific, therefore it is essential to use several methods in parallel. DPPH and ABTS assays are mostly commonly used for estimating the antioxidant potential of active agents loaded in a packaging material by using only the UV–vis spectrophotometer (Mehmood, Sadiq, & Khan, 2020). Two kind of approaches to perform the test are reported in literature: (i) the active compound previously extracted from the active film into distilled water or organic solvents at room temperature and then the antioxidant capacity of the extract was evaluated by one of the above mentioned methods (Jaramillo, González Seligra, Goyanes, Bernal, & Famá, 2015; Dashipour et al., 2015; Joanne Kam et al., 2018); (ii) the film was immersed directly into a DPPH and/or ABTS solution to test its antioxidant capacity (Busolo & Lagaron, 2015; Caetano, Hessel, Tondo, Flôres, & Cladera-Olivera, 2017; Echegoyen & Nerín, 2015; Hromiš et al., 2014; Licciardello, Wittenauer, Saengerlaub, Reinelt, & Stramm, 2015). In the first case, the antioxidant capacity of the film depends on the extraction conditions before the contact with the DPPH radical, such as temperature and contact time, whereas in the second case, the result depends on the contact time of the film with the radicals. In the latter case, it also important to consider the stability of the radical, which for DPPH is almost 8 h and for ABTS is over 48 h (at 21 °C with minimum exposure to light and air). Recently, a novel approach was developed based on indicatory ABTS-gel (ABTS solution in agarose) to estimate antioxidant capacity of the antioxidant film; whose results were correlated with spectroscopic ABTS method for the film (Kusznierewicz, Staroszczyk, Malinowska-Pańczyk, Parchem, & Bartoszek, 2020). Unfortunately, this type of information is not always properly reported in the paper and when reported there is a missing of standard condition (Table 3). The substantial differences were in sample preparation, extraction method (solvent and temperature, etc.), selection of endpoints and expression of results. Additionally, one major issue in ABTS assay is that the kinetic pattern and antioxidant reaction rate is totally ignored. This can be tackled by measuring the absorbance continuously throughout the reaction from which reaction kinetics can be estimated. On the other hand, reaction response rate does not always show linear behavior; the reaction becomes impeded at higher antioxidant concentration due to increase in size of phenols and structural complexity (Schaich et al., 2015). For all the above, to compare results is a tricky task. Standardization procedures are highly required.

Table 3
Methodologies used to evaluate the antioxidant activity.

Biopolymer films obtained by casting	Extraction of active compound	Contact time with radical	Results	References
Gelatine /durian leaf waste (200 mg/mL)	Film into ethanol (no time indicated)	30 min at rT	I% varies from 0.025 to 0.687 mg/ml TE per 100 mg of film	Joanne Kam et al. (2018)
Corn Starch /*Bunium persicum*, and *Zataria multiflora* oils	Film into distilled water until dissolving	30 min at rT	80 % and 71 % in highest concentration for ZMEO and BPEO,	Aminzare et al. (2017)
Novel basil-seed gum/oregano essential oil	Film into distilled water until dissolving	30 min at rT	0.14–0.71 g/kg (DPPH)	Hashemi and Mousavi Khaneghah (2017)
Potato starch /green tea extract	Film into distilled water; 10 % ethanol; 95 % ethanol (no time indicated)	30 min at 37 °C	>95 %	u Nisa et al. (2015)
Carboxymethyl cellulose /*Zataria multiflora* 1−3%(v/v)	Film into water until dissolving	60 min at rT	I% varies from 38 % to 80 %	Dashipour et al. (2015)
Soybean/ *Zataria Multiflora* and *Mentha pulegium* oils	Film into distilled water until dissolving	60 min at rT	IC 50 = 4188.60 ± 21.73 mg/l Zataria M and EC50 = 8.86 ± 0.09 mg/ml Mentha p.	Salarbashi et al. (2014)
Cassava starch/ oregano essential oil (2 % w/v) and pumpkin residue extract (3 %w/v)	Direct contact of film with DPPH radicals	45 min at 23 °C	58.40 %	Caetano et al. (2017)
Chitosan/ oregano/ caraway oil	Direct contact of film with DPPH radicals	until 24 h at rT	increase of AA prolonged the time of contact with DPPH;	Hromiš et al. (2014)
Polyethylene/ Resveratrol (0.1; 0.5 and 1 %) film by extrusion	Direct contact of film with DPPH radicals	24 h at rT	>87 %	Busolo and Lagaron (2015)
Paraffin/paper/ cinnamon oil	Direct contact of film with DPPH radicals	Over the time(50 h) at rT	46.22 ug/mL for eugenol;3606 ug/ mL for cinnamaldehyde; 3480 ug/mL for Limonene	Echegoyen and Nerín (2015)

rT=room temperature, AA= antioxidant activity and I%= (percentage inhibition).

6. Regulatory aspects of antioxidant packaging

Active packaging materials can be presented on the commercial scale if they efficiently and effectively serve the motive for their use as well as fulfill the requirements dictated in the legislation. As per European Union (EU) regulation No. 1935/2004, active packaging systems, which are designed for exposure with the food and can change the organoleptic properties and composition of the food products, provided that these changes are in congruence with the food safety regulations on food and the active agents released into the food products are allowed to be incorporated into food. Furthermore, the transition of food product resulting from active packaging forms should not misguide the consumers by hiding any indications of food spoilage (Wyrwa & Barska, 2017).

There is a clear fundamental difference in food contact material regulations between United States (US) and EU. The US approach focuses more on the fact that "concentration of a compound is responsible for making the compound a poison" so toxicological evaluation is not required for the food contact materials. Whereas, EU approach is focused on the fact that "for all substances provision of toxicological data is mandatory regardless of expected level of exposure" (Restuccia et al., 2010). So active and intelligent packaging is not subject to any specific regulatory provisions in the US [except premarket clearance of the food additives under the section 201 under the Food, Drug, and Cosmetic Act of the Food and Drug Administration (FDA)]. As per article 3 and 4 of EU regulation 1935/2004, all articles and materials coming into contact with the food products should be safe; furthermore, active packaging materials or articles must be inert enough to prevent the release of their components into the food products in quantities that might cause decay of the food products, which may cause unacceptable changes in the food composition or which could lead to harmful effects on the human well-being. Additionally, if the active materials or articles may lead to any changes in food product quality and/or properties these changes must be in-line with food regulations stipulated. Another EU regulation that deals with active and intelligent packaging materials and articles that are intended to come into contact with food products is 450/2009. This regulation regulates issues related to security of applications of the materials utilized in active packaging, furthermore, it deals with marketing of these materials along with provision of the list for permitted articles (European Commission, 2009). Active edible coatings and films should follow all the regulations related to food ingredients/additives, since these packaging materials are an essential part of the edible part of the food products (Galus, Arik Kibar, Gniewosz, & Kraśniewska, 2020). Critical analysis of literature revealed potential health and safety issues related to the use of edible packaging films due to the all film forming components i.e., film forming materials and functional compounds (especially synthetic antioxidants) (Druchta & Jonhston, 1997; Pavli, Tassou, Nychas, & Chorianopoulos, 2018). Therefore, all packaging material components used during the formulation of films/coatings should be within limitations stipulated by the FDA of the US and should be generally recognized as safe (GRAS). Furthermore, according to the US regulations, if a polymeric material is not currently registered as GRAS, but the manufacturer can demonstrate and prove its safety, the manufacturer can either file a GRAS affirmation petition to the FDA or can directly proceed to the market without FDA's approval. On the other hand, EU regulations demand appropriate labelling of food additives (antioxidants, antimicrobials, colorants, and even coatings for food products) with their E-number and specific name (Robin & Sankhla, 2013). Another critical aspect within the regulatory domain of active packaging is allergic reaction and toxicity. Several biopolymers of agro-livestock origin i.e., soy, wheat gluten, whey and casein used in active packaging formulations can cause allergic reaction in some consumers, furthermore, higher doses of natural compounds (i.e., essential oils) may pose oral toxicity threat to the consumers (Cheftel, 2005). Thus, it is essential to find a right balance of active ingredients to avoid toxic effects and clearly mentioning on label about the potential allergen and its quantity no matter how small the amount is being used in the package.

7. Conclusion and perspectives

This review highlights the possible mechanism by which nutritional losses (around 5–30 % loss of ascorbic acid and phenolic compounds) in F&V occur from oxidation reactions. This information will be useful for food businesses to accurately design an innovative active packaging material to preserve the nutritional quality of the fresh foods. Coating technology has been vastly studied (3–7 times more published material as compared to other technologies) as preservation technology for fresh and minimally processed F&V due to its advantages over other technologies, while little work has been published on the application of antioxidant films as packaging materials for preserving the nutritional quality of F&V. It was further observed that protein and polysaccharide-based biopolymers have been vastly used as packaging materials to pack F&V, however, chitosan, gelatin, casein and alginate were found to be more effective as packaging materials (both as coating and as film) to preserve the nutritional and sensory quality of F&V. Furthermore, plant extracts (i.e., green tea and Aloe vera), essential oils (lemon grass), plant oil compounds (eugenol and citral) and phenolics (i.e., thymol) as a component of active packaging systems have shown promising results in preserving the quality of fresh produce. Safety, toxicity and allergenicity are critical regulation issues for active coatings application on food. Critical analysis of recent patents of antioxidant packaging revealed that much of work has been done on preserving different food products except fresh and minimally processed F&V, thus this sector needs to be given attention for development and optimization of active antioxidant package to ensure food safety and high nutritional quality.

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