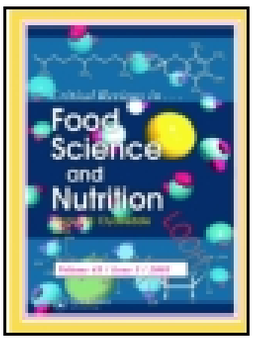

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REVIEW

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Tea polyphenols (TP): a promising natural additive for the manufacture of multifunctional active food packaging films

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ABSTRACT

As a bioactive extract from tea leaves, tea polyphenols (TP) are safe and natural. Its excellent antioxidant and antibacterial properties are increasingly regarded as a good additive for improving degradable food packaging film properties. This article comprehensively reviewed the functional properties of active films containing TP developed recently. The effects of TP addition to enhancing active food packaging films' performance, including thickness, water sensitivity, barrier properties, color, mechanical properties, antioxidant, antibacterial, and intelligent discoloration properties, were discussed. Besides, the practical applications in food preservation of active films containing TP are also discussed. This work concluded that the addition of TP could impart antioxidant and antibacterial properties to active packaging films and act as a crosslinking agent to improve other physical and chemical properties of the film, such as mechanical and barrier properties. However, the effect of TP on specific properties of the active packaging film is complex, and the appropriate TP concentration needs to be selected according to the type of film matrix and the interaction between the components. Notably, the addition of TP improved the efficiency of the active packaging film in food preservation applications, which accelerates the process of replacing the traditional plastic-based food packaging with active packaging film.

KEYWORDS

Film properties; food packaging; property enhancement; tea polyphenols

Introduction

The concept of degradable food packaging film originates from concerns about the environment, the increasing burden of plastic waste disposal, food industry waste, and demand for natural, nutritious and healthy food of consumers (Umaraw et al. 2020). Active food packaging film mainly refers to biodegradable packaging composed of bio-based resource polymers with food packaging and preservation properties, such as mechanical and barrier properties (Carina et al. 2021; de Carvalho and Junior 2020). The

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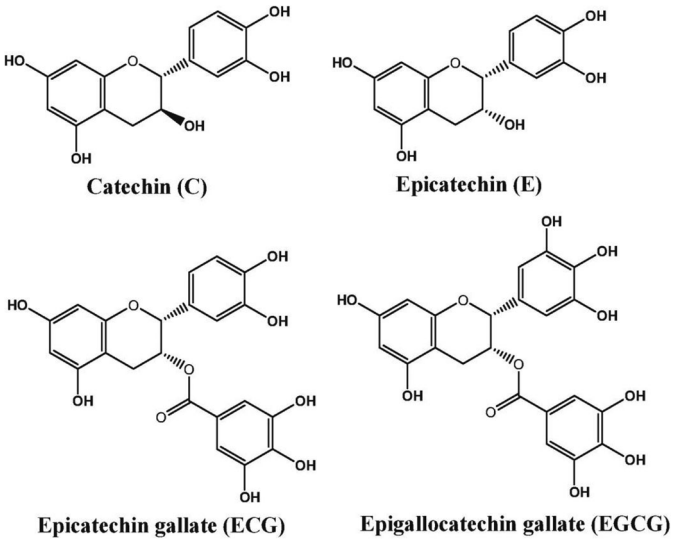

Figure 1. Chemical structures of catechins (Chanphai et al. 2018).

active film could be produced by creating a stand-alone film from the film-forming solution (Zhang et al. 2020d; Luo et al. 2015). The active packaging film could reduce the pollution caused by the mass production of plastics to the global environment, and at the same time, reduce the dependence on petroleum resources (Jeya et al. 2020). In recent years, more and more scientists have devoted themselves to developing active food packaging films formed by the polymerization of biological macromolecules and applying them to food preservation (Umaraw et al. 2020; Carina et al. 2021; Kumar, Mukherjee, and Dutta 2020; Zhu et al. 2019). Compared with traditional petroleum-based packaging materials, biopolymer-based materials have several advantages. However, petroleum-based polymers are cheaper and have better physical strength and gas barrier properties than biopolymer films, so biopolymers are not always better suited for food packaging applications (Jeya et al. 2020).

Recently, biomacromolecules used to develop active food packaging films are mainly polysaccharides, proteins, and lipids. Except for chitosan derived from the shells of crab, most of the polysaccharide substrates that make up degradable food packaging films are derived from natural plants, such as starch (de Souza et al. 2020), cellulose (Roy and Rhim 2020), alginate (Chen et al. 2021), agar (Mostafavi and Zaeim 2020), carrageenan (Zhang et al. 2020c), pectin (Priyadarshi, Kim, and Rhim 2021) and gum arabic (Xu et al. 2019), etc.; Common protein substrates of active films are also natural animal and plant sources, such as zein (Xiao et al. 2020), gelatin (Theerawitayaart et al. 2020), whey protein (Seydim, Sarikus-Tutal, and Sogut 2020) and casein (Mo et al. 2021). Some lipid biomacromolecules, such as beeswax and carnauba wax, cannot form films by themselves, but they can also form films with other biopolymers such as pullulan and gelatin (Omar-Aziz et al. 2020; Zhang, Simpson, and Dumont 2018). However, active food packaging films polymerized by these pure biological macromolecules often could not meet the requirements of the actual application of food packaging and storage. The main limitation is that most of these biological macromolecules themselves not have sufficient antioxidant and antibacterial properties (Zhang et al. 2020d). However, most meat, fish, and derived products lose their commercial value due to oxidation and microbial infestation during storage (Umaraw et al. 2020). In addition, fresh fruits and vegetables, after harvest, will lose water through vigorous respiration resulting in their quality deterioration (Zhang et al. 2021a, Zhang et al. 2021b). Therefore, more and more research is devoted to developing composite active food packaging films with additives (Priyadarshi, Kim, and Rhim 2021; Kumar, Mukherjee, and Dutta 2020). Considering the human body's health, some natural-derived additives with excellent antioxidant and antibacterial properties are the primary candidates (Zhang and Jiang 2020).

Plant polyphenols have a variety of beneficial functional properties, and in addition to the nutritional properties for human health, they also have excellent antibacterial and antioxidant properties (Marquardt and Watson 2014; Li et al. 2019). Polyphenols are found in fruits and vegetables, green and black tea, red wine, coffee, chocolate, and extra virgin olive oil (Visioli et al. 2011). Due to its safety, most plant polyphenols are considered safe and could be directly used as additives in the food industry (Zhang, Li, and Jiang 2020b). Numerous studies in recent years have been devoted to incorporating plant polyphenols into degradable food packaging films to enhance the antioxidant and antibacterial properties of active films to improve food preservation ability (Chen et al. 2018; Biao et al. 2019). Among the many plant polyphenols, tea polyphenols (TP) are natural polyphenols obtained from the tea (*Camellia sinensis*). Its beneficial functional properties, including antioxidant, antibacterial, anti-inflammatory, anti-tumor, and anti-cancer functions, have been widely used in the food and pharmaceutical industry (Liang et al. 2017). In recent years, TP has become a hot issue in preparing degradable food packaging films (Chen et al. 2018; Biao et al. 2019). As an excellent additive, TP has shown great potential in improving the performance of a variety of active films, including alginate (Biao et al. 2019), gelatin (Kuai et al. 2020), chitosan (Zhang and Jiang 2020), pectin-konjac glucomannan (Lei et al. 2019), and starch (Feng et al. 2018), etc.

Incorporating TP into active food packaging films could be used as an antibacterial agent and antioxidant and as a crosslinking agent to improve the performance of films (Lei et al. 2019). But as far as we know, there is no work summarizing the application of TP in active food packaging films. Therefore, the present review summarized the effects of TP as an additive on the performance of active food packaging films in recent years and introduced the current trends and challenges in applying TP for active packaging films. In addition, the practical application of TP composite active packaging films in food preservation is also discussed.

Tp chemistry

In addition to direct drinking water, tea and coffee are the main beverages globally, and tea refers to the different types of tea obtained from the leaves of the tea tree by various

processing methods (Puligundla et al. 2017). The most common are green tea and black tea. Green tea is a non-fermented dry tea, while black tea is a tea that has undergone a complete fermentation process (Khan and Mukhtar 2007). All types of tea contain TP, but because green tea has not been fermented, the content of tea polyphenols in green tea is the highest (Khan and Mukhtar 2007). In contrast, most polyphenols in black tea are converted into theaflavins, thearubigens, and other active substances during the fermentation process (Puligundla et al. 2017). The polyphenols contained in green tea account for 30% of the weight of fresh tea leaves, mainly composed of phenolic acids and flavonoids, and phenolic acids are mainly quinic and gallic acids and four catechins, including epicatechin (EC), epicatechin-3-gallate (ECG), epigallocatechin (EGC), and epigallocatechin-3-gallate (EGCG) (Khan and Mukhtar 2007). Catechins are the main components of green tea polyphenols, accounting for 80% of green tea polyphenols. EGCG is the most abundant among the four catechins and accounts for about 65% of the total catechins (Chanphai et al. 2018). As shown in Figure 1, since both rings contain three phenolic hydroxyl groups, they have a more potent antioxidant capacity and biological activity (Khan and Mukhtar 2007). The health benefits of TP to the human body have been widely recognized, including its antioxidant, antibacterial, anti-inflammatory, anti-tumor, anti-obesity, etc. The excellent antioxidant activity is the root of most functional properties (Puligundla et al. 2017).

The effect of TP on the properties of active films

The purpose of the "active packaging" is to extend the shelf-life of the food and the maintenance or even improvement of its quality. The Regulation 1935/2004 on materials and articles intended to come into contact with food already contains general provisions on active and intelligent packaging safety. It sets the EFSA's safety evaluation process (Dainelli et al. 2008). Compared to traditional packaging, active packaging poses new challenges to the evaluation of its safety. The migration of substances from the packaging to food is the leading risk. Although there is no relevant legislation on TP's food contact compliance aspects, it seems that the safety of TP as a natural additive from edible sources does not need to be considered (Marquardt and Watson 2014). In recent years, due to the compatibility with most biomacromolecule polymers, TP has been regarded as an excellent candidate additive for improving the performance of active films. Its film matrix mainly determines the performance of active packaging films. The addition of TP not only gives it excellent antioxidant and antibacterial properties, but it can also form an interaction force with most biomacromolecule polymers, thereby improving other physical and chemical properties of the active film (Zhang and Jiang 2020). The present review was conducted between February 2021 and April 2021. The sources of the published data are academic journals from electronic databases (ScienceDirect, Scopus, SpringerLink, Web of Science, Wiley Online Library) and chapters of scientific books. The keywords used in the research were "tea polyphenol", "tea polyphenol active film", "tea polyphenol active coating", "epigallocatechin-3-gallate film", and "tea polyphenol food preservation". The collected studies were grouped by topics for easier handling, excluding irrelevant ones and those older than 10. According to different sources, the TP involved in the present work mainly includes commercially purchased TP, green tea extract, black tea extract, and EGCG. Table 1 summarizes the representative literature for recently published tea polyphenol-reinforced degradable food packaging films and TP's effect on physical and chemical properties. The amount of TP added is expressed as a percentage of the dry basis of the polymer.

Thickness

The thickness is a critical parameter for affecting other important properties of active films, such as mechanical properties and barrier properties. Previous studies have shown that the addition of plant extracts and nanocellulose increased the active film's thickness, which is mainly due to the higher solid content in the additives, disrupting the original crystal structure of the film matrix, thereby increasing the spatial distance of the film substrate (Mir et al. 2018; Zhang and Jiang 2020). Since the addition of TP could increase the solid content of active films, adding TP could increase the thickness of the most active film (Table 1). Still, it seems to depend on the additional amount of TP.

In a previous study, chitosan was used to make a composite film by blending it with green tea extracts (Peng, Wu, and Li 2013). The concentration of green tea extracts added had a significant influence on the thickness of the composite film. When the added amount of green tea extracts was from 25 to 100 wt%, the chitosan film's thickness increased from 96.2 to 131.8 μm. The results showed that the increase in thickness of chitosan caused by the addition of green tea extracts is dose-dependent. Besides, in another study, the same concentration of black tea extract also showed a similar trend (Peng et al. 2020). When the concentration of black tea extracts was added from 25 to 100%, the chitosan film's thickness increased from 98.4 to 131.0 μm. Similarly, a recent study prepared a green tea extracts/chitosan composite film, adding 25% green tea extracts resulting in an increase in thickness of chitosan from 71.6 to 94.2 μm (Peng et al. 2020). In another study, when 66% TP was added, the calcium alginate hydrogels film's thickness increased from 32.5 to 46.2 μm (Biao et al. 2019).

Incorporating large doses of TP (more than 20%) could significantly increase the active film's thickness and present a dose-dependent effect. However, containing smaller amounts of TP does not seem to cause an increase in the thickness of the active film. Jamróz et al. (2019) found that the addition of green tea extracts not increased the thickness of furcellaran-gelatin films with concentration from 5–20%. When the concentration of black tea extracts added was 5%, the composite film's thickness was not significantly different from the control; however, when the concentration of black tea extracts increased to 10% and 20%, the thickness of the

Table 1. Effect of TP on the performance of biopolymer-based active food packaging films.

Polymer	TP (wt% based on dry polymer)	Positive effects	Ref.
Chitosan	Black tea extract/25%	Increased thickness, and antioxidant activity; Decreased water solubility, water content, mechanical property and water vapor permeability.	Peng, Wu, and Li (2013)
Chitosan	Green tea extract/25%	Increased thickness, opacity, and antioxidant activity; Decreased water content, mechanical property and water vapor permeability.	Peng, Wu, and Li (2013)
Furcellaran/ gelatin	Green tea extract/10%	Increased TS, antioxidant and antimicrobial activities; Decreased water solubility.	Jamróz et al. (2019)
Chitosan	Black tea extract/3%	Increased opacity, water solubility, antioxidant and antibacterial activities.	Ashrafi, Jokar, and Nafchi (2018)
PLA/ chitosan	Green tea extract/10%	Increased water vapor permeability, heat-seal strength, and food preservation ability; Decreased mechanical properties.	Ye et al. (2018)
Gelatin/ sodium alginate	Green tea extract/0.4–2.0%	Increased b^* value, opacity, TS, and antioxidant; Decreased water vapor permeability, EB and L^* value.	Dou et al. (2018)
Gelatin	Green tea extract/0.7%	Increased TS, antioxidant and thermal properties; Decreased water vapor permeability, EB and water content.	Wu et al. (2013)
Pectin/konjac glucomannan	Green tea extract/2%	Increased TS, antioxidant and antimicrobial activity; Decreased water vapor permeability, EB and water content.	Lei et al. (2019)
Pomelo peel flours	Green tea extract/10 %	Increased thickness, TS, antioxidant and antimicrobial properties; Decreased water vapor permeability, EB and water content.	Wu et al. (2019)
Alginate hydrogels	Green tea extract/66 %	Increased thickness, b^* value, opacity, EB, TS, antioxidant, water vapor permeability and anti-inflammatory properties.	Biao et al. (2019)
Starch	Green tea extract/5 %	Increased antioxidant and antimicrobial properties; Decreased water vapor permeability and mechanical properties.	Feng et al. (2018)
Pectin-chitosan	Green tea extract/2.5 %	Increased EB, antioxidant, water vapor permeability and food preservation ability; Decreased TS and water content	Gao et al. (2019)
Chitosan	Green tea extract/25 %	Increased thickness, b* value, and antioxidant property; Decreased water vapor permeability and mechanical property.	Peng et al. (2020)
Curdlan/chitosan	Green tea extract/ 0.6–3.0 %	Increased antioxidant and food preservation ability; Decreased water vapor permeability and mechanical property.	Zhou et al. (2019)
Poly(vinyl alcohol)/clay	Green tea extract/ 0.5–4.0 %	Increased TS, b^* values, opacity, and antioxidant property; Decreased water vapor permeability, EB and gas permeability.	Chenwei et al. (2018)
Poly(vinyl alcohol)	Green tea extract/10 %	Increased TS, UV light barrier, antioxidant, and antibacterial properties; Decreased EB.	Zhang and Shen (2020)
Poly(vinyl alcohol)	Green tea extract/2 %	Increased b^* value, opacity, TS, and antioxidant property; Decreased water vapor permeability, EB and gas permeability.	Chen et al. (2018)
Poly(vinyl alcohol)	Green tea extract/10 %	Increased TS, opacity, and antimicrobial property; Decreased gas permeability.	Liu et al. (2019)
Myofibrillar protein	Green tea extract/5 %	Increased TS, moisture content and antioxidant activity; Decreased water solubility, EB and water vapor permeability.	Nie et al. (2015)
Agar	Green tea extract/50 %	Increased water solubility, antioxidant and antimicrobial properties; Decreased mechanical property.	Giménez et al. (2013)
Ethylene Vinyl Alcohol Copolymer	Green tea extract/5 %	Increased b^* value and antioxidant.	López de Dicastillo et al. (2011)
Gelatin	Green tea extract/3.3 %	Increased opacity and antioxidant activity; Decreased water vapor permeability and EB.	Li et al. (2014)
Poly(3-hydroxybutyrate-co-3-hydroxyvalerate)	Green tea extract/ 5 %	Increased EB.	Xiang et al. (2013)
Distiller dried grains with solubles	Green tea extract/5 %	Increased EB, antioxidant, and food preservation abilities.	Yang et al. (2016)
Corn Fiber Protein	Green tea extract/20 %	Increased TS, opacity, water solubility antioxidant and antimicrobial activities; Decreased water vapor permeability and EB.	Yang et al. (2015)
Chitosan/polyvinyl alcohol	Green tea extract/2 %	Increased TS, EB, and antioxidant activity; Decreased water vapor permeability and gas permeability.	Zhu et al. (2018)
Gelatin	Green tea extract/2 %	Increased opacity, antimicrobial and antioxidant activity.	Maroufi, Ghorbani, and Tabibiazar (2020)
Corn distarch phosphate/ carboxymethyl cellulose	Green tea extract/1 %	Increased b^* value, EB and antimicrobial activity; Decreased water vapor permeability and TS.	Shao et al. (2020)
Gelatin	EGCG / 5.71 %	Increased b^* value, TS and antioxidant activity; Decreased water content.	Nilsuwan et al. (2019)
Konjac glucomannan/ carboxymethyl chitosan	EGCG / 15 %	Increased b^* value, TS, antioxidant, and antimicrobial activities; Decreased water content, EB and water vapor permeability.	Sun et al. (2020)
PLA/fish gelatin	EGCG / 9 %	Increased b^* value, antioxidant and antimicrobial activities; Decreased water content.	Nilsuwan et al. (2020)
Sodium alginate and carboxymethyl cellulose	EGCG / 30 %	Increased b^* value, TS, and antioxidant activity; Decreased EB.	Ruan et al. (2019)
Gelatin	EGCG / 5.71 %	Increased b^* value, TS, and antioxidant activity.	Nilsuwan, Benjakul, and Prodpran (2018)
Chitosan	EGCG / 15 %	Increased b^* value, antitumor, and antioxidant activity; Decreased mechanical property.	Wang et al. (2018)

composite film increased slightly. The results showed that the addition of a smaller concentration of green tea extract does not increase the thickness of the composite film, which is mainly because green tea extract contains more water-soluble polyphenols, and the 10% content of black tea extract increased the thickness of the film mainly due to its higher solid content (Jamróz et al. 2020).

Similarly, no increase in the thickness of the composite film was observed in the silver carp skin gelatin films with green tea extract blended with 0.3 and 0.7 wt% and the poly(vinyl alcohol)/clay nanocomposite film with blended green tea extract blended with 4 wt% (Wu et al. 2013; Chenwei et al. 2018). However, most active films containing 5–20 wt% TP also showed increased thickness and exhibited dose-dependent. Lei et al. (2019) reported that the addition of TP with content from 1–5% resulting in an increase in thickness of pectin-konjac glucomannan film from 48.3 to 71.2 μm, when 1 wt% TP was added, the thickness of the pectin-konjac glucomannan film increased from 48.3 to 53.1 μm. Consistent with the above report, when 0–5 wt% TP was added to the pectin-chitosan film, the film's thickness showed a concentration-dependent increase trend (Gao et al. 2019). Therefore, the addition of TP usually increased the thickness of active films but sometimes does not affect the thickness of active films, depending on the concentration of TP added and the type of film matrix.

Color

As a food packaging film, color is crucial to consumers' choice. The color index of the active film is usually expressed by the L^* value (lightness), a^* value ('-green' to '+red') and b^* value ('-blue' to '+yellow') (Zhang et al. 2020d). According to the results of many studies, the addition of TP caused a significant change in the color of active films. Specifically, it reduced the brightness of active films, increased the yellowness and redness of active films, that is, reduced the L^* value, and increased the a^* and b^* values of active films (Table 1). The effect of TP on the color of the film is mainly due to the colored substances contained therein, such as carotenoids, theaflavins, and thearubigens, etc. (Jamróz et al. 2019).

Dou et al. (2018) studied the effect of TP on gelatin-sodium alginate edible films, which significantly affects the L^*, a^*, and b^* color parameters. The L^* value of films decreased from 94.06 to 92.61, but the a^* value increased from 0.13 to 1.15, and the b^* values increased from 2.90 to 5.16, with the TP concentrations increased from 0 to 2.5 wt%. The results represented that gelatin-sodium alginate films slightly became darker and more reddish. However, due to the small concentration of TP, the color change of active films seems to be visually insignificant. Similarly, a previous study showed that chitosan films without green tea extract were lighter (higher L^* value). The L^* values of the films decreased from 87.50 to 65.70, but the a^* values increased from 1.11 to 3.99, and the b^* values increased from 1.37 to 40.02, with the green tea extract contents increased from 0 to 20 wt% (Siripatrawan and Harte 2010). It is worth noting that since the main components of green tea extract and black tea extract are different, their effects on the color of active films are also different. The furcellaran-gelatin composite film added with the same concentration of green tea extract and black tea extract showed different color changes (Jamróz et al. 2019). With the increase of TP concentration from 5–20 wt%, although the L^* value of all composite films decreased significantly, the appearance of the composite film contained the green tea extract gradually turned yellow, accompanied by a significant increase in the b^* value, while the appearance of the composite film contained the black tea extract gradually turned red, accompanied by a significant increase in the a^* value (Figure 2). When the concentration of green tea extract is from 5–10%, there is no significant difference in the a^* value of the composite film, but the b^* value is significantly increased (Jamróz et al. 2019). This is mainly because that green tea mainly contains carotenoids and catechins, etc., which are yellow substances, while that black tea mainly contains thearubicin, which is red substances. Therefore, compared to the a^* value, green tea extract significantly affects the b* value of active films, while black tea extract significantly affects the a^* value. As the main component of green tea, the addition of EGCG also increased the b^* value in gelatin and konjac glucomannan/carboxymethyl chitosan films, which is consistent with the above results (Nilsuwan et al. 2019; Sun et al. 2020).

Transparency

The transparency of active food packaging films mainly involves two aspects of practicability. First, maintaining good transparency could directly satisfy consumers' visual observation of the product. Secondly, the film's transparency also determines its light barrier and UV-barrier properties, which are essential for the shelf life of certain commodities (Zhang et al. 2020d). For some light-sensitive foods, higher light barrier properties can reduce the oxidation and degradation of nutrients caused by light, thereby extending its shelf life (Mir et al. 2018). Generally, the light transmittance of active films is determined according to the film's light transmittance at 200–800 nm (Zhang et al. 2020d). Specifically, many studies define the active film's absorbance value ratio at 600 nm to the thickness as opacity (Zhang et al. 2020d). Several studies have shown that the addition of TP could increase the opacity of active films and enhance the light barrier properties, especially ultraviolet light (Table 1).

The optical properties of the pure gelatin-sodium alginate and its TP composite films were determined by measuring transmittance (200–800 nm) and absorption at 660 nm (visible light region) (Dou et al. 2018). When the added amount of TP was from 0.4 to 2%, the transmittance of the gelatin-sodium alginate film at 200–800 nm was all reduced. The transmittance of the films at 280 nm decreased from 59.02% to 2.22% with the addition of TP, and the transmittance at 600 nm reduced from 90.29% to 88.33% only. The results showed that the addition of TP could significantly enhance the UV light barrier performance of the active film. This is

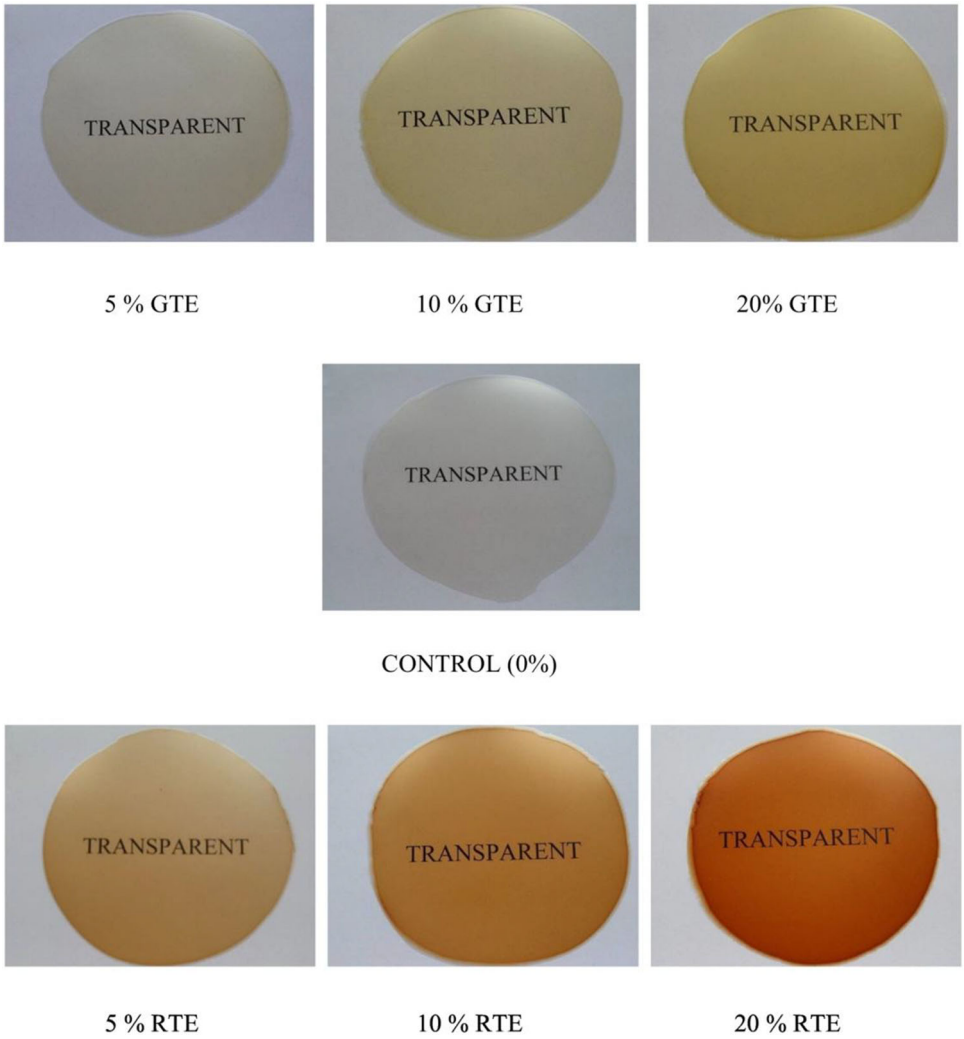

Figure 2. Macroscopic appearance of furcellaran-gelatin films with green or black tea extracts in different concentrations. GTE:green tea extracts; RTE:black tea extracts (Jamróz et al. 2019).

mainly due to that TP contains a large number of phenolic substances, and the benzene ring structure has a strong absorption peak at 280 nm, which could enhance the $n \rightarrow \pi^*$ transition in the UV range, thereby improving the light barrier performance of active films (Dou et al. 2018).

Similarly, a recent study incorporated TP into polyvinyl alcohol (PVA) in different percentages, and prepared PVA/TP composites containing only 5% TP showed excellent anti-UV properties even after washing with water. The presented UV protection factor (UPF) value is still greater than that of the typical values reported in Australia and New Zealand (AS/NZS 4399-1996) that are used to evaluate the anti-UV properties of materials (Zhang and Shen 2020). Therefore, it is an excellent additive to improve the UV light barrier properties of active films due to TP's nature. This is a necessary packaging performance for some foods easily degraded by UV light, such as lipid.

Water solubility and moisture content

Water sensitivity determines the application range of active food packaging films. Since most film substrates are hydrophilic, for some foods with high moisture content, the active packaging film will disintegrate by itself due to hydrophilicity, limiting its application in food packaging (Zhang et al. 2020d). Generally speaking, the water solubility index is the mass ratio of the active film that can be dissolved in water at room temperature, and the moisture content can also reflect the water sensitivity of the film to a certain extent. According to previous research, the addition of TP could reduce the moisture content of the active film and increase water solubility. The moisture content decreased from 28.91 to 10.78% with green tea extracts into chitosan films, but the water solubility has risen from 15.42 to 75.03% (Peng, Wu, and Li 2013). The addition of TP leads to the decrease of the moisture content of the chitosan film mainly because chitosan itself is hydrophilic, and there are some water molecules in its matrix. The addition of TP breaks the original combination of water molecules and chitosan in the matrix, forming the new hydrogen bond interaction with chitosan, thereby reducing the number of water molecules that initially existed in the matrix. The addition of TP leads to the increase of the water solubility of the chitosan film mainly because the original linkage between chitosan molecules is not easy to be released. Still, the high water solubility of TP could cause the chitosan molecules bound to it also to be

released, thereby increasing its water solubility (Aljawish et al. 2016). The same concentration of black tea extract (2 wt%) also reduced the moisture content of the chitosan film to the same degree as green tea extract but only increased the water solubility to 31.65%. The main ingredient is theaflavins, which is different from green tea extract (Peng, Wu, and Li 2013). A recent study is also in line with the above results; the addition of 30% EGCG significantly increased the water solubility of the chitosan film and reduced the moisture content (Wang et al. 2018).

However, the effect of TP on the water solubility and moisture content of different active films is not consistent. Some studies have obtained the opposite results from the above studies. The addition of green tea extract also significantly increased the sliver carp myofibrillar protein-based film's moisture content but reduced the water solubility. The decrease in the composite film was attributed to the phenol-mediated aggregation of protein molecules (Nie et al. 2015). In another study, in the furcellaran-gelatin film added with 5–20% TP, the water content has increased significantly, while the water solubility has decreased considerably (Jamróz et al. 2019). Therefore, in addition to the concentration of TP, the effect of the addition of TP on water solubility and moisture content of active films is mainly related to the type of matrix. In general, the addition of TP seems to increase the water solubility of the polysaccharide-based film but reduces the water solubility of the protein-based film. But this conclusion is not stable because the properties of each film matrix are different; it mainly depends on the interaction between TP and film matrix, film matrix and water, TP and water.

Water barrier properties

The exchange of water vapor between the inside of the food and the outside environment can easily cause water loss, which leads to a decline in the quality of the product. For some dry foods, the entry of external moisture can lead to the occurrence of corruption. Therefore, the water vapor barrier performance is the main indicator for measuring food packaging films' application ability. The higher water vapor barrier performance can effectively extend the shelf life of the packaged food (Zhang, Li, and Jiang 2020b). At present, most studies on active food packaging films use water vapor permeability (WVP) to represent their water barrier properties, and the measurement method is based on the gravimetric method (ASTM E96-95) (Atarés and Chiralt 2016). The process of water vapor passing through the active film will be affected by many factors. The first is the film matrix arrangement, which directly determines the path length through which water vapor passes through the film. Second, the film matrix's hydrophilicity and other components determine the residence time when water molecules pass through the film. Besides, the humidity and temperature in the environment can also affect the water vapor transmission rate (Zhang et al. 2020d). The addition of TP could affect the water barrier performance of the active film, and most studies have shown that the addition of TP could improve the water barrier performance of the active packaging film and significantly reduced the water vapor transmission rate (Table 1).

Peng, Wu, and Li (2013) prepared an antioxidant active chitosan film functionalized with green tea extract and black tea extract. The incorporation of TP significantly enhanced its water vapor barrier properties. By incorporating 25–100% green tea extract in the chitosan film, the WVP of the chitosan film was reduced from 13.39×10^{-11} g m^{-1} s^{-1} Pa^{-1} to 5.07×10^{-11} g m^{-1} s^{-1} Pa^{-1} (reduction by 62%), and the incorporating 25–100 wt% black tea extract reduced from 13.39×10^{-11} g m^{-1} s^{-1} Pa^{-1} to 5.82×10^{-11} g m^{-1} s^{-1} Pa^{-1} (reduction by 56%). In another study, a smaller amount of TP also resulted in the enhancement of the water vapor barrier properties of active films. WVP of gelatin-sodium alginate films with TP (0.8–2.0 wt%) significantly decreased compared to gelatin-sodium alginate films ($p \leq 0.05$), which ranged from 0.228 ± 0.017 to 0.138 ± 0.006 g·mm/m^2·h·kPa (reduction by 39%) (Dou et al. 2018).

Similarly, the enhancement of water barrier properties caused by the addition of TP has also been observed in gelatin (Wu et al. 2013), pectin-konjac glucomannan (Lei et al. 2019), pomelo peel flours (Wu et al. 2019), and starch (Feng et al. 2018) active films. Also, Sun et al. (2020) observed that the addition of EGCG also improved the water barrier properties of the konjac glucomannan/carboxymethyl chitosan film and significantly reduced the WVP. The increase in water barrier properties of active films caused by the addition of TP is mainly due to the formation of hydrogen bond interactions between TP and the film matrix, which makes the film have a denser network structure and increases the barrier to water vapor transmission. Besides, due to TP's hydrophilicity, the time for water vapor to pass through the film can be increased by absorbing water vapor (Sun et al. 2020). However, it has been reported that the addition of TP could lead to the weakening of the water barrier properties of the active film. A recent report showed that the addition of 2.5–15% TP significantly increased the WVP of the pectin-chitosan film and reduced its water barrier properties (Gao et al. 2019). This may be because although the incorporation of TP forms a new hydrogen bond with the film matrix, the newly formed hydrogen bond is not as strong as the interaction between the original film matrix and does not form a denser network structure than the original one, making it easier for water vapor to pass through the film. In some studies, although the addition of an appropriate TP concentration can lead to a decrease in WVP of active films, the addition of excessively high content (above the optimum concentration) of TP could also cause an increase in the WVP of active films (Table 1). This is mainly because excessive TP will form agglomerations in the film matrix to create voids, which will lead to the passage of water molecules (Lei et al. 2019).

Mechanical properties

The mechanical properties of the active packaging film determine its stress range during application. Tensile strength (TS) and elongation at break (EB) represent the

mechanical properties of the active packaging film. The tensile strength mainly means the maximum pressure that the film can withstand, and the elongation at break represents the maximum deformation range of the film (Zhang et al. 2020d). The ideal active food packaging film should have sufficient mechanical properties under specific application scenarios to avoid failure. The mechanical properties of an active film mainly depend on the internal structure and interaction forces of the film matrix (Atarés and Chiralt 2016). Most of the published studies on the properties of active films containing TP have focused on the effect of TP on the mechanical properties of active films (Table 1).

First, the effect of TP on the mechanical properties of the active film seems to be directly related to its concentration. Siripatrawan and Harte (2010) explored the effect of incorporating different concentrations of green tea extract on the mechanical properties of chitosan films. The results showed TS and EB did not significantly change when green tea extract concentration increased from 0 to 5%. Still, they significantly increased from 25.13 ± 1.91 to 27.55 ± 3.46 MPa and from 54.76 ± 3.14 to 60.73 ± 3.37%, respectively when green tea extract concentration increased from 5 to 20%. However, in another study, the addition of 25% green tea extract and black tea extract significantly reduced the TS and EB of the chitosan film. The addition of a larger dose (50 and 100%) of green tea extract and black tea extract also weakened the mechanical properties of chitosan films, but there was no significant difference from 25% (Peng, Wu, and Li 2013).

Similarly, a decrease in mechanical properties was observed in the chitosan film incorporated with 25% TP and 15% EGCG (Peng et al. 2020; Wang et al. 2018). The contradictory effect of TP on the mechanical properties of chitosan film is mainly due to the concentration effect of TP. When the TP amount is 0–5%, TP could be uniformly dispersed in the long chitosan chain (Peng et al. 2020). The hydrogen bond interaction between chitosan molecules makes the network structure closer, improving the chitosan film's mechanical properties. When the TP concentration exceeds 25%, TP will aggregate in the chitosan long chain, which will disrupt the original arrangement of the chitosan chain and weaken the mechanical properties (Peng, Wu, and Li 2013).

Similarly, other polysaccharide-based films have also been observed that the effect of TP addition on mechanical properties is mainly related to its concentration. Recent research has developed an active blending film based on pectin and konjac glucomannan and used TP as additives to improve its mechanical properties (Lei et al. 2019). The results showed that as the concentration of TP increased from 1–5%, the TS of the film first increased and then decreased, reaching the maximum when the TP concentration is 2%, which is significantly higher than the pectin-konjac glucomannan film. The TS was smaller than the pectin-konjac glucomannan film when the TP concentration was 5%. At the same time, the EB decreased continuously during this period. This is mainly because the addition of less than 2% TP could form hydrogen bonds with pectin and konjac glucomannan, which arranges the composite film to more compact, which can be proved by the FT-IR spectra results. The decrease in TS caused by the addition of more than 2% of TP is due to the appearance of the discontinuous structure of the film caused by excessive TP, which is shown as cross-sectional holes and discontinuous structure on the microstructure of the film (Lei et al. 2019). The decrease of EB is due to the crosslinking effect caused by the addition of a low concentration of TP, which reduces the flexibility of the film, or the addition of a high concentration of TP directly leads to the destruction of the film structure (Benbettaïeb, Karbowiak, and Debeaufort 2019).

Secondly, the effect of TP on active films is also directly related to the type of film substrate. For the alginate hydrogels film, a high TP concentration increased the TS of the alginate hydrogels film. It is worth noting that the addition of 66% TP increased the TS by 126%, and the addition of 133% TP increased the TS by 75% (Biao et al. 2019). This is mainly because the mechanical properties of the alginate hydrogels film itself are too weak. Although aggregation occurs with the addition of a high concentration (133%) of TP, the newly formed gel network structure formed by the added TP in the film matrix is stronger than the force between the original alginate film, thus increasing the TS of the alginate film (Biao et al. 2019).

On the contrary, for some active films with strong mechanical properties, the addition of TP will destroy their original network structure, thereby weakening their mechanical properties (Zhou et al. 2019). A study has shown that when a series of TP concentrations (0–3%) are incorporated into the curdlan/chitosan blending film, the mechanical properties of the film have been significantly reduced, including the decline of TS and EB (Zhou et al. 2019). This is mainly due to the combination of the original curdlan and chitosan, resulting in a dense and orderly network structure. The addition of TP disturbed the original orderly structure, resulting in a decrease in mechanical properties. The effect of TP on the mechanical properties of protein-based active films seems to be similar to the trend mentioned above. In the optimum addition range, the TP could increase the TS and reduced the EB of protein-based active films. A previous study developed an active film from silver carp skin gelatin incorporated with green tea extract. Regarding the physical properties, 0.3 and 0.7% green tea extract incorporated into gelatin films revealed significantly higher TS and lower TB than gelatin film. In addition to forming hydrogen bonds with proteins, TP could also form hydrophobic forces with hydrophobic parts of proteins (Wu et al. 2013). But TP can also cause protein aggregation, which may have a negative effect on the mechanical properties of the protein film (Nie et al. 2015). Therefore, the addition of TP has a complicated effect on the active film, which is mainly related to the added concentration and the film matrix.

Antioxidant and antimicrobial properties

Corruption caused by oxidation and microbial contamination is the main problem of food deterioration and

degradation (Huang et al. 2017). Therefore, the active film with antioxidant and antibacterial properties can be more conducive to extending the shelf life of food. However, the most common biomacromolecule polymers used to form active films do not have sufficient advantages in antioxidant and antibacterial activities (Zhang et al. 2020d). As a natural active substance with strong antioxidant capacity, TP also has excellent antibacterial properties (Liang et al. 2017). It is an excellent candidate for enhancing the antioxidant and antibacterial properties of active food packaging films. Most studies use TP as an additive to increase the antioxidant and antibacterial properties of the film (Table 1).

The antioxidant activity of most active films is related to the phenolic substances contained in them; therefore, the main indicator used to evaluate active films' antioxidant activity is the total phenol content (Atarés and Chiralt 2016). Polyphenols in TP will lose part of it due to oxidative degradation during the film formation process after being added to the film. The added amount of TP cannot be used directly to represent the active film's total phenol content (Zhang, Li, and Jiang 2020b). Also, the antioxidant activity of active films could be measured by methods such as 2,2-diphenyl-1-picrylhydrazyl (DPPH), 2,2′-Azinobis-(3-ethylbenzthiazoline-6-sulphonate) (ABTS), original trolox equivalent antioxidant capacity (TEAC) and ferric reducing antioxidant power (FRAP), etc. (Zhang, Li, and Jiang 2020b). However, each method has some limitations, and multiple methods are usually used to determine the antioxidant activity of the active film. Since tea polyphenols are converted into theaflavins, thearubigens, and other active substances during the fermentation of black tea, the active film's antioxidant activity containing black tea extract is lower than that of the active film containing green tea extract (Liang et al. 2017). Peng, Wu, and Li (2013) studied the antioxidant activity of chitosan film containing black tea extract and green tea extract at the same concentration. The results showed that the chitosan film containing green tea extract had higher antioxidant activity than the active film containing black tea extract in all aqueous solutions (0%, 20%, 75%, and 95% ethanol). For example, the equilibration time (i.e., the time reaching the highest scavenging activity) of chitosan film containing 2 wt% green tea extract in distilled water was 2 min, while chitosan film containing 2 wt% black tea extract was 6 min. However, in another study, there was no significant difference between active films contained black tea extract and green tea extract with the same concentration in the DPPH and ABTS antioxidant test, although the total phenol content of the active film incorporated with green tea extract was significantly higher than the active film contained black tea extract (Jamróz et al. 2019). The spectrum of phenols in black tea is very different from that in green tea (Liang et al. 2017). Most studies have used green tea polyphenols as additives to improve the antioxidant properties of active films (Table 1). A previous study found that incorporating TP (0.4–2.0 wt%) increased the antioxidant activity of the gelatin-sodium alginate film in a dose-dependent manner, including total phenol content, DPPH, and ABTS. It is worth noting that although the control film does not have TP, it also showed slight antioxidant activity. This is mainly due to some amino acid residues on gelatin with a slight antioxidant capacity (Dou et al. 2018). In addition, the gelatin-sodium alginate film containing 2% and 1.6% TP has no significant difference in the DPPH test results. In comparison, the total phenol content and ABTS of the 2% TP film are significantly higher than 1.6% TP film, and this showed that the DPPH method has some deviation, so it is necessary to use a variety of tests to determine the antioxidant activity of the active film (Dou et al. 2018). At present, a variety of antioxidant active films containing TP have been successfully developed, including pectin-konjac glucomannan (Lei et al. 2019), pomelo peel flours (Wu et al. 2019), starch (Feng et al. 2018), curdlan/chitosan (Zhou et al. 2019) and poly(vinyl alcohol)/clay (Chenwei et al. 2018), etc.

TP has always been considered as a source of natural antibacterial agents and has antibacterial activity against a variety of foodborne pathogenic bacteria, including *Staphylococcus aureus, Escherichia coli, Helicobacter pylori, Legionella pneumophila, Mycobacterium tuberculosis, Salmonella, P. aeruginosa, L. monocytogenes*, and *Vibrio cholera* (Bansal et al. 2013). There are many hypotheses about the antibacterial mechanism of TP. An earlier study pointed out that EGCG in TP could directly bind to bacterial peptidoglycan to precipitate it, leading to the destruction of the cell wall structure (Shimamura, Zhao, and Hu 2007). In another study, TP could combine with reactive oxygen species inside bacteria to generate hydrogen peroxide under the action of superoxide dismutase, destroying its protein and nucleic acid (Arakawa et al. 2004). Using TP as an additive to develop an active food packaging film can increase the active film's antibacterial properties, thereby reducing the microbial contamination of the packaged food, extending its shelf life, and securing food safety (Dehghani, Hosseini, and Regenstein 2018). The methods used to determine the antibacterial properties of active films mainly include disk and well diffusion assays, agar dilution, broth dilution, viable cell counts, vapor-phase technique, and optical density-based methods (Abdollahzadeh, Nematollahi, and Hosseini 2021).

Jamróz et al. (2019) used the agar diffusion method to study the antimicrobial activity of the furcellaran-gelatin film containing green tea extract and black tea extract against *Staphylococcus aureus*, *Escherichia coli*, *Candida albicans,* and *Hanseniaspora uvarum*. The results showed that the control film did not show any antimicrobial activity against all bacteria and fungi. All films containing TP did not show antimicrobial effect against fungi, which may be due to the low TP concentration (0–20 wt%) of active film not has antimicrobial activity against fungi. The film containing 10 and 20% green tea extract showed antibacterial activity against two kinds of bacteria. Green tea extract at the same concentration had higher antibacterial activity against *Staphylococcus aureus*. However, it was only observed in the film containing 20% black tea extract that it had antibacterial activity against *Staphylococcus aureus*, and other films containing black tea extract not showed any antibacterial activity against both types of bacteria, which is

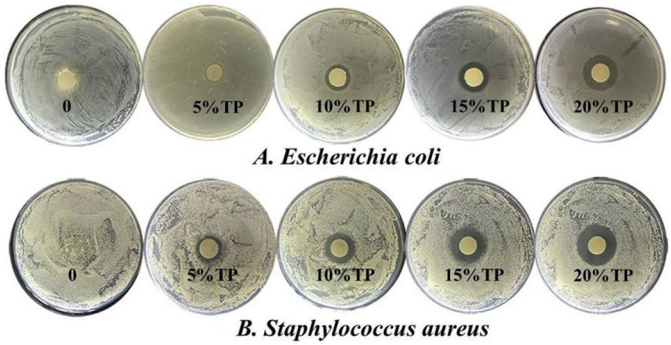

Figure 3. Inhibitory zones of pomelo peel flours based films without and with TP against test microorganisms: *A-Escherichia coli*, *B- Staphylococcus aureus* (Wu et al. 2019).

related to the content of catechins (Friedman 2007). A recent study has shown that the antibacterial activity of green tea extract is significantly higher than other tea extracts, including flowery broken orange pekoe, broken orange pekoe, and red dust (Nibir et al. 2017). It has also been observed in the tapioca starch/decolorized Hsian-Tsao leaf gum coating mixed with green tea extract that its antibacterial activity against gram-positive microorganisms is significantly greater than that of gram-negative microorganisms. This may be due to the additional lipophobic outer membrane on the surface of the Gram-negative bacteria, which is not easy to penetrate by TP (Chiu and Lai 2010). In another study, an antibacterial food packaging film was developed by mixing different concentrations (5, 10, 15, and 20%) of TP into pomelo peel powder (Wu et al. 2019). The results showed that as TP concentration increased, the antibacterial activity of the active film against *Escherichia coli* and *Staphylococcus* continued to increase. The active film's antibacterial activity against *Staphylococcus aureus* was greater than *Escherichia coli* (Figure 3). Similar results were also observed in the starch film containing TP (Feng et al. 2018). Therefore, using TP as an additive can develop an active food packaging film with excellent antibacterial ability, and its antibacterial activity against Gram-positive bacteria is greater than that of Gram-negative bacteria.

Food packaging application of active films containing TP

At present, active films containing TP have achieved excellent results in food applications and can effectively enhance active films' application efficiency in food preservation (Table 1). The application of active films in food preservation can be directly used as food packaging film and as an edible coating form. The edible coating is a microscopic form of active films. A thin layer with a small thickness is directly formed on the food's surface as a fresh-keeping method. The edible coating is produced by directly immersing the food into the film-forming solution or spraying the film-forming solution to the food's surface. In contrast, active films can create a stand-alone film from the film-forming solution (Zhang et al. 2020d).

The discoloration of pH-sensitive (halochromic) smart packaging films could detect the freshness of food while preserving the food quality, which has attracted great interest from many scientists recently (Alizadeh-Sani et al. 2020). Some active substances in TP also have pH-sensitive properties, such as chlorophyll, catechins, and theaflavins, etc. (Chaturvedula and Prakash 2011). Jamróz et al. (2019) developed a intelligent and active furcellaran-gelatin films containing green tea extract and black tea extract. The intelligent film showed different color properties in different pH solutions (pH 3.0 and pH 12.0). Specifically, in an acidic solution with pH 3.0, all films are white, while in an alkaline solution with pH 12.0, all films turn orange. This property could provide the active film with the ability to monitor the freshness of fish in real-time. Due to the active film's high water solubility, direct contact with food will cause partial disintegration of the active film. Therefore, the active film can be placed in a closed packaging bag of fish products. Among them, the putrefaction bacteria will produce a large amount of ammonia and volatile amines, which will cause the pH of the air inside the packaging bag to drop, which will cause the discoloration of the active film (Jairath et al. 2015). The film's discoloration effect in different pH solutions was also observed in the starch film containing green tea extract, mainly attributed to chlorophyll's effect in the green tea extract (Medina-Jaramillo et al. 2017). The chlorogenic acid contained in TP can be converted into chlorogenic acid quinine under different pH environments. The reaction system can turn green and black color, which may also be the reason for the intelligent performance of active films containing TP (Jeszka-Skowron, Krawczyk, and Zgoła-Grześkowiak 2015; Wildermuth, Young, and Were 2016). TP has other substances that are sensitive to pHs, such as EGCG, which is a stable form of phenol in an acidic environment and can be oxidized to phenolic ions and quinones in an alkaline environment that changes color (Arakawa et al. 2004). In a recent study, the addition of EGCG gave the hydroxypropyl methylcellulose film intelligent sterilization and discolored performance, which could detect the growth of bacteria through the change of pH and release H_2O_2 to kill bacteria. It has great potential for detecting food bacteria's growth and maintaining food safety (Huang et al. 2021).

Due to excellent antioxidant and antibacterial properties, chitosan film containing TP showed great potential in extending the shelf life of beef samples (Ashrafi, Jokar, and Nafchi 2018). Compared with unpackaged and pure chitosan film-packaged beef samples, chitosan film containing TP extended the shelf life of beef samples from 3 and 4 days to 6 days, accompanied by fewer bacterial colonies and a degree of lipid oxidation. Besides, the beef sample's pH value decreased with the increase of the TP concentration in the chitosan film, which may be related to the synergistic antibacterial effect of TP and chitosan and lactic acid formation by lactic acid bacteria (Ashrafi, Jokar, and Nafchi 2018). Similarly, the application of pectin-chitosan, curdlan-chitosan, and distiller dried grains active films containing TP also significantly extended the fresh meat shelf life (Gao et al. 2019; Zhou et al. 2019; Yang et al. 2016). Fresh fruits are still undergoing vigorous respiration in postharvest storage,

accompanied by water loss and quality deterioration (Zhang and Jiang 2019; Zhang et al. 2020a). The active packaging film/coating has excellent barrier properties, slowing down the water loss and gas exchange of fresh produce, thereby maintaining its quality during storage. Compared with unpackaged and polylactic acid/chitosan-packaged cherry fruits, the cherry fruits packaged with polylactic acid/chitosan film containing TP showed lower decay rate and weight loss, maintaining higher firmness and ascorbic acid content during postharvest storage (Ye et al. 2018). The oil storage process is easily degraded under the influence of light and oxygen, leading to deterioration (Mohammadi et al. 2016). Since the addition of TP could improve the light and oxygen barrier performance of active films, it could alleviate the quality degradation of oily foods during storage. Wu et al. (2019) prepared a bioactive edible packaging film based on pomelo peel flours incorporating TP and used it as an edible packaging film to store oil. Compared with the unpackaged and pomelo peel flours film-packaged oil samples, the results showed that the pomelo peel flours film containing TP packaged oil has a lower peroxide value in the storage period. And the pomelo peel flours film containing TP film has an excellent ability to prevent oil penetration.

The addition of TP enhanced the application efficiency of actives films or edible coatings in food preservation can be attributed to the following facts: TP could directly endow actives films or edible coatings with excellent antioxidant and antibacterial activities, delaying the occurrence of food spoilage; the crosslinking of TP and the film matrix could improve the barrier properties of actives films or edible coatings, resulting in slowing down the water loss and gas exchange of the food; TP could give actives films or edible coatings the UV barrier properties and delay the oxidation of food caused by UV light; some phenolic substances in TP could be used as a pH-sensitive indicator, giving the film intelligent discoloration performance.

Conclusions

Due to its excellent antioxidant and antibacterial activities and good biocompatibility, TP has become an excellent choice of additives to improve active food packaging films' performance. This work concluded that the addition of TP could impart antioxidant and antibacterial properties to active packaging films and act as a crosslinking agent to improve other physical and chemical properties of the film, such as mechanical properties and barrier properties. However, the effect of TP on certain properties of the active film is complex. For example, the effect of TP on the water vapor barrier properties and mechanical properties of active films is related to the concentration of TP and closely related to the components of the film matrix and the interaction between the components. The application of TP could also endow the active film with intelligent pH-sensitive discoloration performance, monitoring the freshness of the food in real-time while preserving the food. Notably, the addition of TP improved the efficiency of the active film in food preservation applications, which accelerates the process of replacing the traditional plastic-based food packaging with active packaging film.

Disclosure

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