Vessa Kola

Plant extracts as additives in biodegradable films and coatings in active food packaging: Effects and applications

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Thesis for Master´s Degree in Biotechnology

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Plant extracts as additives in biodegradable films and coatings in active food packaging: Effects and applications

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Declaro ser o autor deste trabalho, que é original e inédito. Autores e trabalhos consultados estão devidamente citados no texto e constam da listagem de referências incluída.

"I declare that I am the author of this work that is original and unpublished. Authors and works consulted are properly cited in the text and included in the list of references."

Vessa Kola

“It is better to make a mistake
with full force of your being
than to carefully avoid mistakes
with a trembling spirit.”

Socrates

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Vessa Kola

Abbreviations and acronyms

ABTS – 2,2´- Azino-bis(3-ethylbenzthiazoline-6-sulfonic acid)

BHA – Butylated hydroxyanisole

BHT – Butylated hydroxytoluene

CH – Chitosan

CH-BEE – Chitosan black eggplant extract

CH-PEE – Chitosan purple eggplant extract

CIE – Commission internationale de l'éclairage

GFF – *Gloiopeltis furcate* funoran

CFU – Colony-forming unit

GSA – Gelatin-sodium alginate films

GRAS – Generally Recognised as Safe

d – days

DNA – Deoxyribonucleic Acid

DPPH – 2,2-diphenyl-1-picrylhydrazyl

EB – Elongation Break

EFSA – European Food Safety Authority

E.g. – Exempli gratia

EPA – Eastern Psychological Association

Etc – Et cetera

FDA – Food and Drug Administration

FRAP – Ferric-Reducing Antioxidant Power

g – Grams

GAE – Gallic Acid Equivalent

HDPE – High-Density Polyethylene

kg – Kilograms

LDPE – Low-Density Polyethylene

I.e. – Id est “that is”

µg – Microgram

mm – Millimetres

ml – Millilitres

nm – Nanometres

OP – Oxygen Permeability

PGA – Poly (glycolic acid)

PFP – *Pseuderanthemum palatiferum* freeze-dried powder

pH – Power of hydrogen

PHA – Polyhydroxyalkonates

PHB – Poly(hydroxybutyrate)

PHBV – Poly(hydroxybutyrate-co-valerate)

PLA – Poly (lactic acid)

PLGA – Poly (lactide-co-glycolide)

PSE – Peanut skin extract

PVB – Polyvinyl butyral

PVC – Polyvinyl chloride

ROS – Reactive oxygen species

RP – Reducing Power

RT – Room temperature

SCELE – *Sonneratia caseolaris* (L.) Engl. leaf extract

Se – Selenium

SEM – Scanning Electron Microscopy

TFP – *Tremella fuciformis* polysaccharide

TBARS – Thiobarbituric acid reactive substances

TPC – Total phenolic content

TPS – Thermoplastic starch films

TS – Tensile Strength

TVB-N – Total volatile basic nitrogen

TVC – Total viable count

UV – Ultra-violet

v/v – Volume per volume

WVP – Water vapour permeability

wt% – Weight percent

YM – Yerba mate

YOPE – Yellow onion peel extract

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Abstract

Background: Petroleum-based polymers are widely used and known that they cause serious environmental problems due to their non-biodegradability. To this end, researchers have been focusing on the development of ecological packaging materials from natural resources. However, the films produced from natural biopolymers have fewer desirable properties than synthetic polymers; Subsequently, active packaging has arrived. Research has been conducted to develop biodegradable films/coatings based on wide range of ingredients, which may affect the properties of those materials.

Scope and approach: Plant extracts represent an interesting ingredient for biodegradable food packaging. The scope of this review is to present the latest ideas on how plant extracts impact properties like physical, mechanical, barrier, functional, structural, antioxidant, antimicrobial and the biodegradability of the films. Moreover, it has been introduced the interaction of plant extracts with the food product, which may prevent or reduce deterioration and improve the quality of the packaged product during its lifetime.

Key findings and conclusion: Recent studies focus on the identification of extracts from various plant sources such as leaves, fruits, pomace, seeds, etc. for use in biodegradable polymer. Generally, most of the plant extracts increase the thickness of the biopolymer, reduce the transparency, decrease water vapour and oxygen permeability, enhance water moisture and solubility, and have adverse effects on the mechanical properties. Additionally, in most cases natural extracts can boost the antioxidant and antimicrobial properties and alters the biodegradation rate, contact angle (θ), and viscosity of the films and coatings. Furthermore, improvement of shelf life of packaged food, like meat, and meat products, fruits, and vegetables with the incorporation of plant extracts has been observed. Results are promising further research on the effect of plant extracts on sensorial properties and application to many other food products may encourage practical application.

Keywords: Plant extracts; Biodegradable films; Active packaging; Film´s properties; Antioxidant; Antimicrobial

Resumo

Contexto: Polímeros à base de petróleo são usados em grande escala e sabe-se que causam problemas ambientais sérios devido a não serem biodegradáveis. Com base nisto, investigadores têm-se focado no desenvolvimento de materiais para embalagens ecológicas a partir de recursos naturais. Contudo, as películas produzidas de biopolímeros naturais possuem normalmente propriedades desejáveis menores quando comparadas com as obtidas de polímeros sintéticos; mais recentemente apareceram as embalagens ativas. Diferentes estudos têm sido conduzidos para desenvolver películas/revestimentos biodegradáveis, com base numa ampla variedade de ingredientes, que podem influenciar as propriedades destes materiais.

Âmbito e abordagem: Extratos vegetais representam ingredientes interessantes para embalagens biodegradaveis usadas em alimentos ou em produtos alimentares. O Âmbito desta monografia é apresentar as mais recentes ideias do modo como os extratos de plantas afetam diferentes propriedades das películas, tais como físicas, mecânicas, de barreira, funcionais, estruturais, antioxidantes, antimicrobianas e ainda a sua biodegradabilidade. Além disso, foi introduzida a interação entre os extratos de plantas com o produto alimentar, que pode prevenir ou reduzir a deterioração e melhorar a qualidade do produto embalado aumentando o seutempo de vida.

Principais conclusões: Estudos recentes focam-se na identificação de extratos vegetais de diferentes fontes como, folhas, frutas, sementes, bagaço, etc. para serem usados em polímeros biodegradáveis. Geralmente, a maioria dos extratos vegetais aumentam a espessura do biopolímero, reduzem a transparência, reduzir a permeabilidade ao vapor de água e ao oxigénio, elevam a humidade e a solubilidade, e têm diversos efeitos nas propriedades mecânicas. Adicionalmente, na maioria dos casos os extratos naturais podem aumentar as propriedades antioxidante e antimicrobiana e alterar a taxa de biodegradabilidade, ângulo de contato (θ), e viscosidade das películas e revestimentos. Com a incorporação dos extratos vegetais, verifica-se ainda, um aumento do tempo de prateleira, do alimento embalado, como carne, e produtos derivados de carne, frutas e legumes. Os resultados são promissores e o estudo do efeito dos extratos de plantas nas propriedades sensoriais e sua aplicação em muitos outros produtos alimentares, poderá estimular num futuro próximo a sua aplicação prática.

Palavras-chave: Extratos vegetais; Películas biodegradáveis, Embalagens ativas; Propriedades de películas; Antioxidante; Antimicrobiano

1. Introduction

Food packaging plays a crucial role in food preservation throughout the distribution chain. The main aim of packaging materials is to protect food from contaminants and to prolong its shelf life (Popovic´, Lazic´, Hromiš, Šuput, & Bulut, 2018). Traditional food packaging is a passive, inert barrier. The objective of this type of packaging is to make the traditional materials that come into contact with food as inert as possible: there should be a minimum interaction between the food and the packaging (Skinner, 2015).

Most of the traditional materials that are used in food packaging are petroleum-derived polymers, which are non-biodegradable and represent a serious environmental problem. This can explain the search of natural polymers by the researchers, like biodegradable films and coatings which have increased significantly, mainly due to interest in minimizing the ecological impact caused by the use of synthetic packaging materials (Mir, Dar, Wani, & Shah, 2018; Qureshi et al., 2020). Edible films are thin layers of edible materials, which once formed can be placed on - or between - food components, whereas an edible coating is formed as a coating on a food product. The most common materials for the formulation of edible/biodegradable films and coatings are polysaccharides, proteins and lipids, and the combination of these allows for producing blends of improved characteristics (Atarés & Chiralt, 2016).

However, films produced from natural polymers have less desirable properties than synthetic polymers; subsequently, studies have been carried out to improve the characteristics of biodegradable films, by using active food packaging, and make them competitive and applicable in the market (Martins, Romani, Martins, & Filipini, 2019). As synthetic functional food ingredients exist health and ecological concerns, natural ingredients have become an ecologically important alternative of synthetic ingredients, and thereupon become popular subjects of interest in numerous food research studies (S. Wang, Marcone, Barbut, & Lim, 2012).

Plant extracts have received great attention as they contain high concentrations of phenolic components that possess strong antioxidant and antimicrobial activities. Films formed by incorporating plant extracts into polymers usually resulted in modified physicochemical, mechanical, barrier, antioxidant and antimicrobial properties compared to films made of individual components and have been used for a wide variety of functions in polymers (Mir et al., 2018).

Accordingly, this review focuses on the current technology of plant extracts and their influence on properties of biodegradable films and coatings for food packaging. Properties like, physical, mechanical, barrier, optical, functional, structural, antioxidant, antimicrobial, and biodegradability of the films have been analysed. Moreover, application of active films/coatings containing plant extracts in food products has been reviewed as well.

2. Literature review

2.1. Synthetic polymers

Polymer is derived from the Greek words' poly meaning "many" and meros meaning "parts" (Carraher, Hess, & Sperling, 1987). Synthetic polymers are considered to be indispensable to modern sciences and technology (Bhargawa, 2015). Over years, many synthetic products and materials were made to fulfil human needs (Vinod, Sanjay, Suchart, & Jyotishkumar, 2020). They have become a major component of our modern world with their wide range of applications in a variety of fields such as packaging, agriculture, food, consumer products, medical appliances, building materials, industry, aerospace materials for their durability, low cost and resistance to degradation (Pan, Farmahini-Farahani, O'Hearn, Xiao, & Ocampo, 2016).

The list of synthetic polymers includes neoprene, nylon, PVC, polystyrene, polyethylene, polypropylene, polyacrylonitrile, PVB, silicone and many more (Bhargawa, 2015). More than 40% of the plastics are used for packaging and almost half of them are used for food packaging in the form of films, sheets, bottles, cups, tubs and trays etc. (Hosseini & Gómez-Guillén, 2018). Above all, polyethylene is the most used petroleum-based polymer in packaging application. It is very difficult for polyethylene or other types of petroleum-based polymer to be biodegraded after disposing in land or coast, hence creating varying levels of contamination (Zhong, Godwin, Jin, & Xiao, 2020).

Generally, the natural degradation of plastics begins with photodegradation, which leads to thermo-oxidative degradation. Whereas, burning synthetic plastics releases harmful gases into the atmosphere, such as CO, SO_2, NO, N_2O, NO_x, HF, and HCl while burying plastics in the ground can also cause environmental pollution (Jafarzadeh et al., 2020).

Generation of synthetic polymer waste is increasing at a disturbing rate. Studies have uncovered that less than 10% of the generated synthetic plastics are recycled (Vinod et al., 2020). However, the Food and Drug Administration (FDA) of the United States has raised many concerns regarding recycled plastics, particularly for re-use of these materials in food- contact articles. These concerns include (Imam, Glenn, & Chiellini, 2012):

- the contaminants from the post-consumer material that may appear in the final food-contact product made from the recycled material,
- incorporation of recycled post- consumer material that is not regulated for food-contact into food-contact packaging and
- assimilation of adjuvants/additives in the recycled plastic not approved for food-contact use.

Thereupon, people are becoming increasingly concerned about these environmental issues that arise due to the excessive usage of plastics which are non-biodegradable materials, and are starting to search for alternative materials for food packaging (Hanani, Yee, & Nor-Khaizura, 2019).

2.2. Biopolymers

In order to solve this problem, researchers have focused on using biodegradable materials, such as polysaccharides, proteins or lipids, for food packaging purposes (F. Liu et al., 2020; Mostafavi, Kadkhodaee, Emadzadeh, & Koocheki, 2016) which allow the packaging materials to be sustainable, and compostable completely. Therefore, biopolymers are a recent growth for the substitution for synthetic polymers in the concern of environmental awareness (Vinod et al., 2020; Zhong et al., 2020).

The biodegradable biopolymers can be classified in three main categories (Fig. 2.1) based on their origin (Garavand, Rouhi, Razavi, Cacciotti, & Mohammadi, 2017; Vinod et al., 2020) as follows:

- Biopolymers originated from agricultural resources, including polysaccharides (e.g. starch), ligno-cellulosic products (e.g. cellulose and its derivatives), proteins (e.g. whey and collagen), lipids (e.g. bee wax) and free fatty acids;
- Biopolymers achieved by means of microbial fermentation, such as pullulan and polyhydroxyalkanoates.
- Biotechnology products, chemically synthesized biopolymers using monomers attained by natural raw materials such as poly (lactic acid).

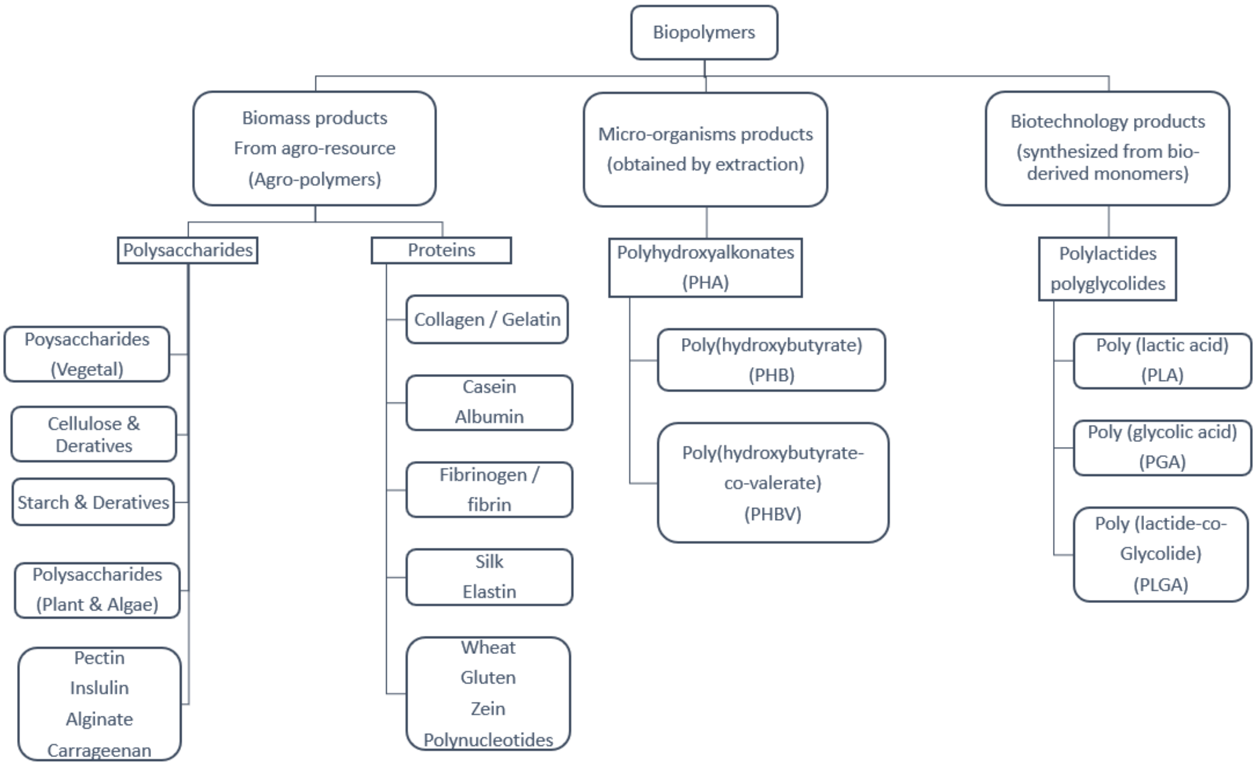


Fig. 2.1 – Classification of biopolymers (Source: Vinod et al., 2020).

The main advantages of biopolymers are biocompatibility and biodegradability. The degradation of biopolymer depends on various factors like composition, environment, type of polymer and chemical bonds between them. The degradation process is classified as below (Vinod et al., 2020):

- Biodegradable: Degradation happens due to the presence of microorganisms.
- Hydro-biodegradable: Degradation occurs in the presence of microorganisms and water.
- Photo-degradable: Delinking between molecules in the presence of light.
- Bioerodable: Degradation due to erosion by natural abrasion.
- Compostable: Degradation occurs due to bacterial action that improves soil condition.

Biopolymers can be decomposed upon disposal in bioactive medium by microorganisms like bacteria, fungi, algae, or by marine water to CO_2, CH_4, H_2O, biomass and other natural substances, which are relevant to potential benefits for greenhouse gas balances and other environmental impacts. For example biodegradability in biopolymers can be achieved by aerobic or an-aerobic fermentation in the presence of microorganisms which secrete extracellular enzymes to fragment the polymer chains in the packaging material to small molecular weight decay products (Fig. 2.2) (Moustafa, Youssef, Darwish, & Abou-Kandil, 2019; Zhong et al., 2020).

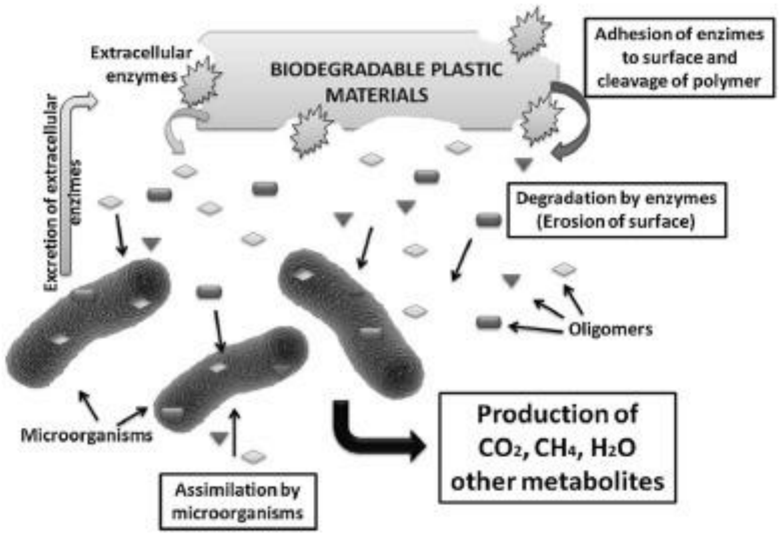


Fig. 2.2 – Biodegradability mechanism in biopolymers (Source: Moustafa et al., 2019).

Over the advantages there are also certain limitations. In general, films produced from natural polymers have less desirable properties than synthetic polymers; as a result, studies have been carried out to improve the properties of biodegradable films by incorporating bioactive compounds which not only provide antioxidant and antimicrobial properties, but also improve for instance the mechanical and barrier properties of the films and they are indispensable in the food packaging sector (Martins et al., 2019; Beikzadeh, Khezerlou, Jafari, Pilevar, & Mortazavian, 2020; Garavand et al., 2017).

Consequently, active food packaging has emerged, to enhance the properties of biodegradable films so they are competitive with synthetic polymers and can be applied in the market. This type of packaging is designed to interact with the packaged food in order to increase the food shelf-life, maintaining or improving the foods organoleptic and nutritional properties (Andrade et al., 2018).

2.3. Active packaging

The globalization phenomenon has brought the possibility to access to countless foods from every part of the globe, so the importance of preserving their original organoleptic properties is increasing. Food industry was forced to develop new ways and technologies to satisfy the consumers' demands. In line with this, smart packaging has emerged (Ribeiro-Santos, Andrade, Melo, & Sanches-Silva, 2017). Recently, smart packaging systems (active and intelligent) based on biopolymers and natural extracts, as it surpasses the original function of traditional food packaging have been attracting increasingly by the food industry (Hanani, Yee, & Nor-khaizura, 2019; Jaróz, Kulawik, Guzik, & Duda, 2019).

Active food packaging is a type of packaging that has an active function to enhance the food shelf-life or to reach some desirable properties, thus is not only providing a barrier from the outside environment. While intelligent food packaging is a packaging that monitors food conditions inside packaging and gives information regarding its quality and safety (Kuswandi & Jumina, 2020).

In active packaging the package, packaging environment and the product interact positively (Ribeiro-Santos et al., 2017). The basic concept is integrating specific substances in the food packaging (Kuswandi & Jumina, 2020) in order not only at ensuring food safety and extending shelf-life, but also reducing the negative impact on the environment (Ahmed et al., 2017; Jaróz et al., 2019).

It can be divided into active-scavenging systems (absorbers) removing undesired compounds from the food or its environment and active-releasing systems (emitters) adding compounds into the headspace or to the packaged food (Yildirim & Röcker, 2018) as Fig. 2.3 shows (Ahmed et al., 2017). For example the first one acts as a barrier against undesirable substances such as O_2, H_2O, CO_2; the other ones have active components, which are able to give several properties such as antioxidant or antimicrobial activity (Raspo, Gomez, & Andreatta, 2018).

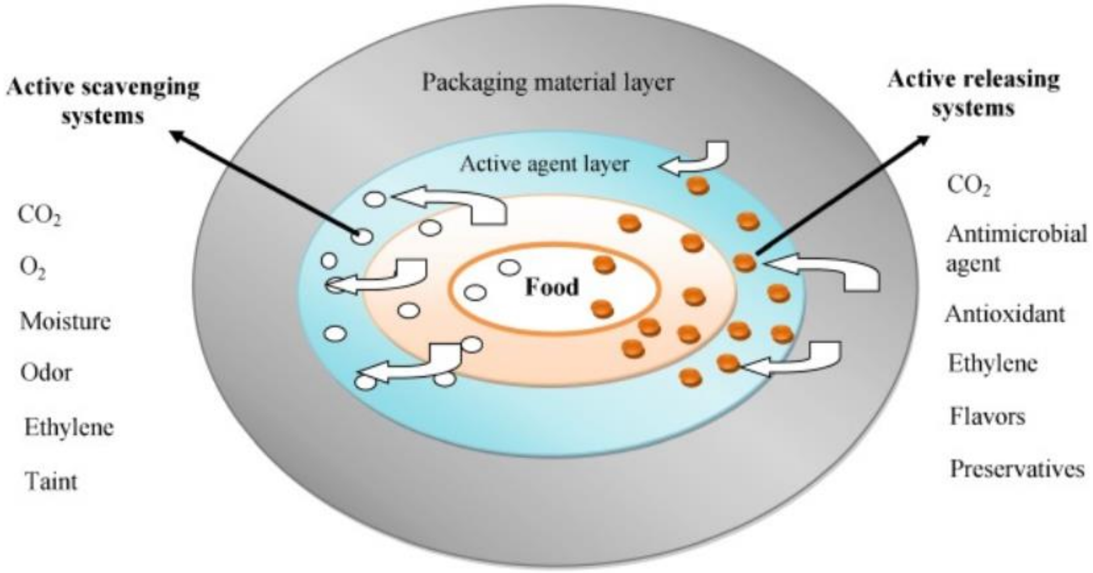


Fig. 2.3 – Active scavenging and releasing systems in food (Source: Ahmed et al., 2017).

The type of absorbers and releasers depends on the physical form of active packaging systems, and they can be in the form of sachet, label, or film/coating (Bodbodak & Rafiee, 2016).

However, the use of sachets is not fully accepted around the world. The consumer can mistakenly perceive the independent sachet to be a gift or a condiment, rather than part of the packaging system with a technological function (this is particularly a problem in foods designed for children). Furthermore, the food industry must add the sachet to the product in the packaging line, which adds complexity to the system as well as additional cost. Equally, this is not a universal solution, as the dimensions of the sachet need to be optimized for each package and food. For these reasons, the sachet is not the preferred active packaging option (Nerín, 2010).

On the other hand, films and coatings are widely used. Films, are defined as a dried thin layer of biopolymer separately added, formed beforehand and after applied on the product like wrapper or among the layers thereof, it can be used as covers, packaging, or layer separation. Whereas the coatings are films formed in the product, whose base is applied directly on the surface thereof, where occurs the drying and thus, the coating formation (Kapetanakou & Skandamis, 2016; Ribeiro-Santos, Andrade, Melo, & Sanches-Silva, 2017).

The ultimate functionality of edible films/coatings is related to their bioactivity (such as antioxidant, antimicrobial, and anti-browning) and their functional (such as their ability to serve as a barrier to water vapor, oxygen, carbon dioxide, and UV–vis light), their mechanical (such as tensile stress and elongation at break) and physical (such as opacity and colour) properties (Silvia-Weiss, 2013).

Among active packaging technologies, materials that release active substances to preserve food are particularly important. Many forms of this special type of packaging involve the use of films of polymeric materials that act as carriers for different active compounds (Suppakul, 2011). One issue in food preservation is the use of synthetic additives, which are associated with different adverse effects on human health. Thus, natural substances such as plant extracts, are important due to their benefits for food preservation as well as in human health, in addition to being natural products (Martins et al., 2019). So, there is much interest among food manufacturers in natural extracts, to act as replacements for synthetic antioxidants currently used (Suppakul, 2011). Different parts of plants and fruits from many agricultural crops have been found to be a viable source as fillers for composite industries. In addition, some agricultural waste available in nature can also be used as a source of raw material for renewable materials (Arroyo et al., 2019).

2.4. Natural plant extracts as antioxidant and antimicrobial agents

In recent years, there has been a global trend toward the use of natural phytochemicals present in natural resources – such as vegetables, fruits, oilseeds and herbs – as antioxidants and functional ingredients (Carvalho, Cavaco, & Brodelius, 2011). Plant extracts are gaining a wide interest in the food industry for their potential as antioxidants and antimicrobials since they are also Generally Recognized as Safe (GRAS) (Nikmaram et al., 2018).

2.4.1. Antioxidant activities of plant extracts

Antioxidants significantly delay or prevent oxidation of oxidizable substrates when present at lower concentration than the substrate (Kasote, Katyare, Hegde, & Bae, 2015). In addition, antioxidant actuality is associated with decreased DNA damages and lipid peroxidation, which level the immune performance and reduces virulent metamorphosis of the cells (Farzaneh & Carvalho, 2015).

Two basic classifications are applied to antioxidants: synthetic and natural. Synthetic antioxidants are phenolic compounds containing various degrees of alkyl substitutes, whereas the natural antioxidants can be phenolic compounds such as quinone and lactone. Synthetic antioxidants such as butylated hydroxy anisole (BHA) and butylated hydroxy toluene (BHT) are widely used in the food industry due to their high antioxidant capacity and outstanding performance in the retardation of oxidation reactions (Cruz et al., 2019). However, their application is linked with potential adverse health effects, such as carcinogenic risk (Jiao, Quek, Gu, Guo, & Liu, 2020). Consequently, natural plant antioxidants have gained attention (Nikmaram et al., 2018).

Most of natural antioxidants are obtained through plant extraction with phenolic compounds identified as their main active constituents, fruits, vegetables and agriculture by products that are rich in polyphenols are usually recognised as potential sources of natural antioxidants (Jiao et al., 2020).

Natural antioxidants can be classified into two classes:

1. Enzymatic antioxidants
2. Non-enzymatic antioxidants

Enzymes, low molecular weight molecules and enzyme cofactors are produced endogenously. When endogenous antioxidants cannot ensure a rigorous control and a complete protection of the organism against oxidation reactions, exogenous antioxidants will be needed. These types of antioxidants, as nutritional supplements or pharmaceutical products contain active compounds of antioxidant activity. Many non-enzymatic antioxidants are obtained from dietary sources which include different groups of chemical classes. Polyphenols are the largest class, as can be seen in Fig. 2.4 (Maqsoudlou, Assadpour, Mohebodini, & Jafari, 2020).

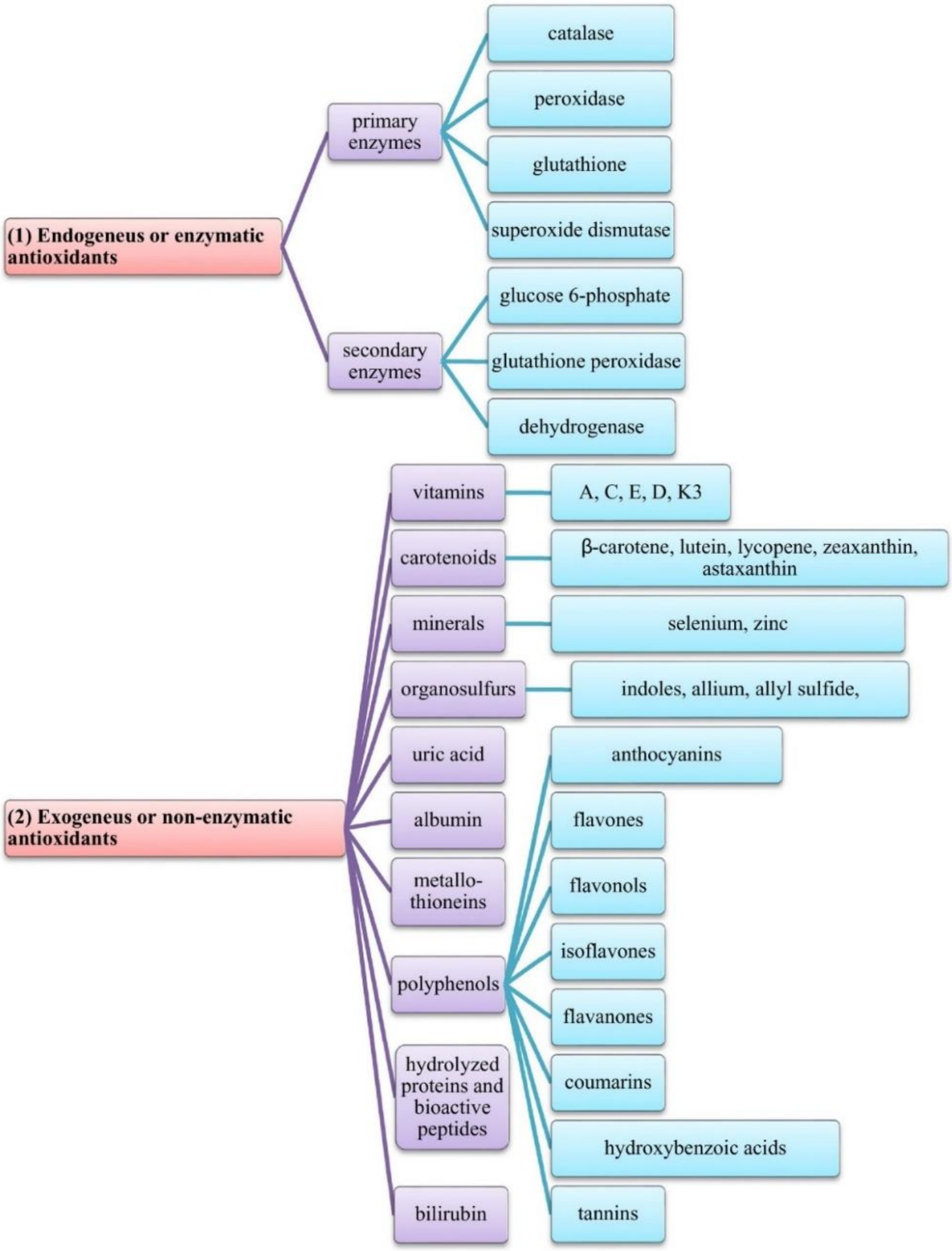


Fig. 2.4 – Classification of natural antioxidants (Source: Maqsoudlou et al., 2020).

Polyphenolic compounds are a large group of secondary metabolites, which contain at least one aromatic ring with at least one hydroxyl group bonded on it, widely distributed in plants, which can be divided according to the number of rings and to their functional groups bound in the structure, and thus we have: phenolic acids, flavonoids, anthocyanins, stilbenes, and lignans, coumarins, tannins (Radulescu, Claudia Buruleanu, Antonia Georgescu, & Dulama, 2019). Flavonoids, perhaps the most important single group of phenolics in foods, comprise a group of over 4000 aromatic plant compounds, which include anthocyanins, proanthocyanidins, flavonols and catechins. Phenolic acids, on the other hand, include hydroxycinnamic acids (*e.g.* caffeic or ferulic acid conjugates, sinapic acid) and hydroxybenzoic acids (*e.g.* benzoic, gentisic or *p*-anisic acids) (Carvalho, Cavaco, Carvalho, & Duque, 2010).

Anthocyanins are water soluble pigments in plants which contribute to the brilliant colours of blue, red, and mauve in flowers, fruits, and leaves. It has reported that anthocyanins exhibit antioxidant activity (Teng et al., 2017). Changes in anthocyanin activity have been linked to the variation of the metal ions and co-existing colourless compounds such as co-pigments, flavones, and flavonols (Nikmaram et al., 2018).

Carotenoids are lipid soluble plant pigments, which are widely spread in nature. These compounds can be classified into two categories according to their chemical structure: carotenes and xanthophylls, α-, β-Carotene and lycopene are examples of carotenes, *i.e.* non oxygenated carotenoids. Lutein, zeaxanthin, astaxanthin and canthaxanthin are examples of xanthophylls i.e. oxygenated derivatives of carotenes (D. Ribeiro, Freitas, Silva, Carvalho, & Fernandes, 2018; Soares, Carvalho, Azevedo, & Fidalgo, 2019). Consequently, are efficient antioxidants for scavenging singlet molecular oxygen and peroxyl radicals. They are considered as part of the antioxidants defence system in the human organism (Nikmaram et al., 2018).

Minerals (*e.g.* selenium, copper, manganese, and zinc) and vitamins (vitamin E, C, and A) present in plants are well known natural antioxidants. Selenium (Se) is considered the mineral that is most specifically associated with the antioxidant function. There are at least 35 antioxidant Se-based proteins including selenoprotein P, five glutathione peroxidases and three thioredoxin reductases. Tocopherols (Vitamin E) acts as a membrane-bound antioxidant, trapping lipid peroxyl free radicals produced from unsaturated fatty acids under conditions of “oxidative stress.” Ascorbic acid (Vitamin C) eliminates peroxyl radicals and easily gives up electrons to provide stability to reactive species such as reactive oxygen species (Nikmaram et al., 2018).

Together with the non-enzymatic components of the antioxidant machinery, the enzymatic components provide a complex and multifaceted protective mechanism to maintain ROS homeostasis in order to both avoid oxidative-induced damages in plant cells and support plant development (Soares et al., 2019).

2.4.2. Antimicrobial activities of plant extract

Consumers preference for natural food additives and their concern about carcinogenicity of synthetic food preservatives like nitrates, benzoates, sulphites, sorbates and formaldehyde, has encouraged the food industry to consider natural alternatives (Olszewska, Gędas, & Simões, 2020; Pisoschi et al., 2018).

Natural plant extracts can assist in maintaining the appearance, taste, and quality of food without any negative impact on the colour, odour, and taste profiles. Besides the antioxidant properties, a large variety of extracts have effective antimicrobial properties as well (Zanetti et al., 2018). The functions of herbal plant extracts and essential oil activities are mainly on the structures and cellular membrane. It has been shown that the permeability of the cell septum or membrane are impacted by bioactive compounds available in infusions, leading the intracellular enclosures outside of the cell, that might be chased by the infraction in electron transfer system, enzyme activity and nutrient adsorption capability of the cell (Farzaneh & Carvalho, 2015).

Pathogenic microorganisms have been a danger to the human race since its genesis, being a major cause of human morbidity and mortality (Górniak, Bartoszewski, & Króliczewski, 2019). The bioactive compounds show different mechanisms of action according to the group of microorganisms (Baptista, Horita, & Sant'Ana, 2020). Phenolic compounds such as simple phenols, quinones, tannins, coumarins, flavones, terpenoids and alkaloids exhibit antimicrobial activity (Castro-Rosas et al., 2017). They act by the alteration of microbial cell permeability, therefore allowing the loss of biomolecules from the inside of cells. Phenolics interfere with cell membrane functionality as they interact with membrane proteins inducing their structure and function alteration. Affected membranes functions are related to electron transport, nutrient uptake, synthesis of proteins and nucleic acids and enzyme activity (Pisoschi et al., 2018).

The structural diversity of plant derived compounds is immense, and the impact of antimicrobial action they produce against microorganisms depends on their (Gyawali & Ibrahim, 2014):

- structural configuration. The hydroxyl (-OH) groups in phenolic compounds are thought to cause inhibitory action as these groups interact with the cell membrane of bacteria to disrupt membrane structures and cause leakage of cellular components

- position of the -OH group. The structural of thymol is along with of carvacrol; however, difference in antimicrobial effectiveness between thymol and carvacrol against Gram-positive and Gram-negative bacteria was observed when tested in agar medium. This difference has attributed to the -OH group located at the *meta* position in thymol compared to the *ortho* position in carvacrol
- number of double bonds. Among citronellol, geraniol, and nerol, citronellol was found to be less effective due to the presence of only one double bond whereas geraniol and nerol with two double bonds, showed higher antimicrobial activity against tested bacteria (*Bacillus. cereus*, *Escherichia coli*, *Staphylococcus. aureus*) and yeast (*Candida albicans*).

There are reports suggesting that flavonoids are important antimicrobials in plant life. To arrest the spread of pathogens, plants possess an innate immunity that involves different layers of defence responses and some of these defences include biosynthesis of flavonoids (Górniak et al., 2019).

Phenolic acids are present in a wide variety of plants mainly in seeds, fruit, peels, and leaves. It has been reported that caffeic acid exerts effects against viruses, bacteria, and fungi, while eugenol has been classified as bacteriostatic. The mechanism of action for low-molecular-weight phenolic acids is described by the diffusion of the non-dissociated acid across the membrane, causing the acidification of the cytoplasm and occasionally, cell death. It should be noted that several factors (*e.g.* pH, ring substitutions, and the saturation of the side chain) play a vital role in the activity of cinnamic acids (Nikmaram et al., 2018).

The antimicrobial activities of tannins have long been recognised and include inhibition of extracellular microbial enzymes, deprivation of the substrates required for microbial growth, direct action on microbial metabolism through inhibition of oxidative phosphorylation, metal ions deprivation or formation of complexes with the cell membrane of bacteria causing morphological changes of the cell wall and increasing membrane permeability (Huang, Liu, Zhao, Hu, & Wang, 2018).

Alkaloids with a great structural diversity are widely used in the formulation of important antibacterial drugs including metronidazole and quinolones. The antibacterial mechanism of action has been evaluated among several classes such as indolizidine, isoquinoline, quinolone, agelasine and polyamine (Nikmaram et al., 2018).

In general, the addition of plant extracts to biodegradable packaging films not only acts as antioxidants and antimicrobials but also modifies the properties of packaging material, so enhances the material´s shelf life, durability and could enhance their overall application. Upon the techno-functional modification of biopolymer material with plant extracts, the resultant films were found to have improved properties and having the potential application in food packaging (Mir et al., 2018).

3. Study approach

The approach used in this dissertation was firstly to review existing literature of plant extracts and their use in active food packaging and to provide a complete summary how plant extracts affect the properties of biodegradable films and shelf life of food products. Relevant studies published in the English language were searched from Web of Science, Science direct, Pub Med, Google Scholar for all combinations of keywords from lists A, B, C, D and E:

- (A): “plant extracts”, “natural extracts”, “natural additives” “herbs”, “spices”, “by-products”
- (B): “biopolymer”, “natural polymer”, “biodegradable polymers”, “biocompatible polymers”, “polysaccharides”, “proteins”, “lipids”
- (C): “food packaging”, “active packaging”, “active food packaging”, “smart packaging”, “active films” “biodegradable films”, “biodegradable coatings”
- (D): “thickness”, “colour”, “optical properties”, “transparency”, “mechanical properties”, “tensile strength”, “elongation break”, “barrier properties”, “water vapor permeability”, “antioxidant”, “antimicrobial”, “biodegradability” “contact angle”, “wettability”, “viscosity”
- (E):“food application” “food products”, “meat”, “seafood”, “fish”, “vegetables”, “fruits”

The keyword search was applied not only to title and abstract but also to the content of the article. Moreover, this study was focused primarily on the most recent scientific papers from the years 2016-2020.

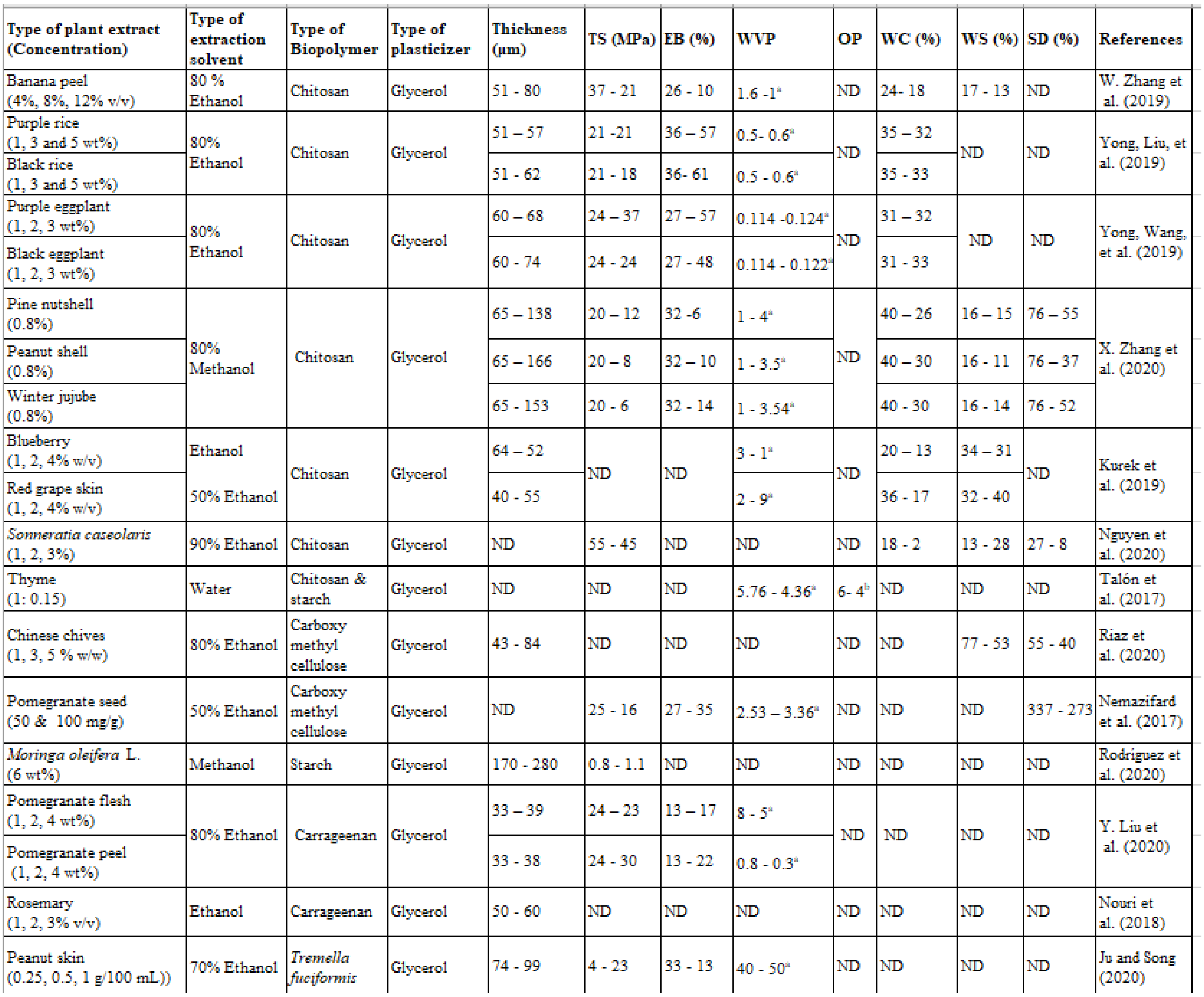
4. Data analysis

4.1. Effects of plant extracts on the properties of the films

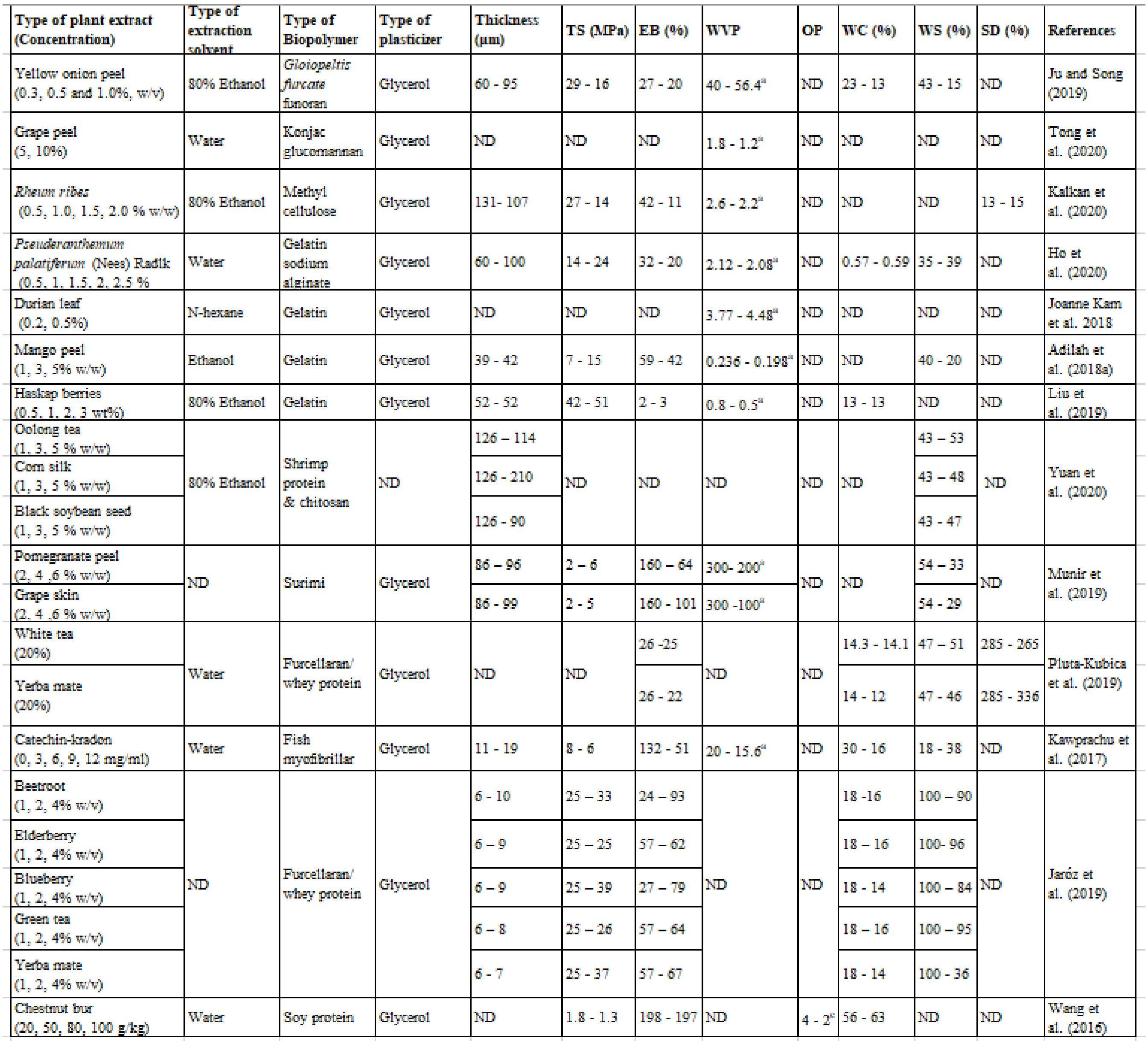
In the presence of plant extracts, properties of biopolymeric films can be changed as a result of interactions between biopolymer and polyphenolic compounds (Staroszczyk et al., 2020). Furthermore, the extent of changes in film properties depends on the type and concentration of extract. The physical or chemical interactions between biopolymers and extracts depends on the nature, chemical characteristics and concentration of biopolymer and extract, as well as on the structural properties of the active components including stereochemistry and conformational flexibility (Mir et al., 2018).

Table 4.1 shows a representative selection of recent studies (2016-2020), giving an overview of some of the tests which are usually carried out when dealing with the effect of plant extract addition on the properties of biodegradable films.

Table 4.1 – Effect of plant extracts on physical, mechanical, barrier and functional properties of the film.



Type of plant extract (Concentration)	Type of extraction solvent	Type of Biopolymer	Type of plasticizer	Thickness (µm)	TS (MPa)	EB (%)	WVP	OP	WC (%)	WS (%)	SD (%)	References
Banana peel (4%, 8%, 12% v/v)	80 % Ethanol	Chitosan	Glycerol	51 - 80	37 - 21	26 - 10	1.6 -1[a]	ND	24- 18	17 - 13	ND	W. Zhang et al. (2019)
Purple rice (1, 3 and 5 wt%)	80% Ethanol	Chitosan	Glycerol	51 – 57	21 -21	36 – 57	0.5- 0.6[a]	ND	35 – 32	ND	ND	Yong, Liu, et al. (2019)
Black rice (1, 3 and 5 wt%)				51 - 62	21 - 18	36- 61	0.5 - 0.6[a]		35 - 33			
Purple eggplant (1, 2, 3 wt%)	80% Ethanol	Chitosan	Glycerol	60 – 68	24 – 37	27 – 57	0.114 -0.124[a]	ND	31 – 32	ND	ND	Yong, Wang, et al. (2019)
Black eggplant (1, 2, 3 wt%)				60 - 74	24 - 24	27 - 48	0.114 - 0.122[a]		31 - 33			
Pine nutshell (0.8%)	80% Methanol	Chitosan	Glycerol	65 – 138	20 – 12	32 -6	1 - 4[a]	ND	40 – 26	16 – 15	76 – 55	X. Zhang et al. (2020)
Peanut shell (0.8%)				65 – 166	20 – 8	32 – 10	1 - 3.5[a]		40 – 30	16 - 11	76 – 37	
Winter jujube (0.8%)				65 - 153	20 - 6	32 - 14	1 - 3.54[a]		40 - 30	16 - 14	76 - 52	
Blueberry (1, 2, 4% w/v)	Ethanol	Chitosan	Glycerol	64 – 52	ND	ND	3 - 1[a]	ND	20 – 13	34 – 31	ND	Kurek et al. (2019)
Red grape skin (1, 2, 4% w/v)	50% Ethanol			40 - 55			2 - 9[a]		36 - 17	32 - 40		
Sonneratia caseolaris (1, 2, 3%)	90% Ethanol	Chitosan	Glycerol	ND	55 - 45	ND	ND	ND	18 - 2	13 - 28	27 - 8	Nguyen et al. (2020)
Thyme (1: 0.15)	Water	Chitosan & starch	Glycerol	ND	ND	ND	5.76 - 4.36[a]	6- 4[b]	ND	ND	ND	Talón et al. (2017)
Chinese chives (1, 3, 5 % w/w)	80% Ethanol	Carboxy methyl cellulose	Glycerol	43 - 84	ND	ND	ND	ND	ND	77 - 53	55 - 40	Riaz et al. (2020)
Pomegranate seed (50 & 100 mg/g)	50% Ethanol	Carboxy methyl cellulose	Glycerol	ND	25 - 16	27 - 35	2.53 – 3.36[a]	ND	ND	ND	337 - 273	Nemazifard et al. (2017)
Moringa oleifera L. (6 wt%)	Methanol	Starch	Glycerol	170 - 280	0.8 - 1.1	ND	ND	ND	ND	ND	ND	Rodriguez et al. (2020)
Pomegranate flesh (1, 2, 4 wt%)	80% Ethanol	Carrageenan	Glycerol	33 – 39	24 – 23	13 – 17	8 - 5[a]	ND	ND	ND	ND	Y. Liu et al. (2020)
Pomegranate peel (1, 2, 4 wt%)				33 - 38	24 - 30	13 - 22	0.8 - 0.3[a]					
Rosemary (1, 2, 3% v/v)	Ethanol	Carrageenan	Glycerol	50 - 60	ND	ND	ND	ND	ND	ND	ND	Nouri et al. (2018)
Peanut skin (0.25, 0.5, 1 g/100 mL))	70% Ethanol	*Tremella fuciformis*	Glycerol	74 - 99	4 - 23	33 - 13	40 - 50[a]	ND	ND	ND	ND	Ju and Song (2020)



Type of plant extract (Concentration)	Type of extraction solvent	Type of Biopolymer	Type of plasticizer	Thickness (µm)	TS (MPa)	EB (%)	WVP	OP	WC (%)	WS (%)	SD (%)	References
Yellow onion peel (0.3, 0.5 and 1.0%, w/v)	80% Ethanol	*Gloiopeltis furcate* funoran	Glycerol	60 - 95	29 - 16	27 - 20	40 - 56.4^{a}	ND	23 - 13	43 - 15	ND	Ju and Song (2019)
Grape peel (5, 10%)	Water	Konjac glucomannan	Glycerol	ND	ND	ND	1.8 - 1.2^{a}	ND	ND	ND	ND	Tong et al. (2020)
Rheum ribes (0.5, 1.0, 1.5, 2.0 % w/w)	80% Ethanol	Methyl cellulose	Glycerol	131- 107	27 - 14	42 - 11	2.6 - 2.2^{a}	ND	ND	ND	13 - 15	Kalkan et al. (2020)
Pseuderanthemum palatiferum (Nees) Radlk (0.5, 1, 1.5, 2, 2.5 %	Water	Gelatin sodium alginate	Glycerol	60 - 100	14 - 24	32 - 20	2.12 - 2.08^{a}	ND	0.57 - 0.59	35 - 39	ND	Ho et al. (2020)
Durian leaf (0.2, 0.5%)	N-hexane	Gelatin	Glycerol	ND	ND	ND	3.77 - 4.48^{a}	ND	ND	ND	ND	Joanne Kam et al. 2018
Mango peel (1, 3, 5% w/w)	Ethanol	Gelatin	Glycerol	39 - 42	7 - 15	59 - 42	0.236 - 0.198^{a}	ND	ND	40 - 20	ND	Adilah et al. (2018a)
Haskap berries (0.5, 1, 2, 3 wt%)	80% Ethanol	Gelatin	Glycerol	52 - 52	42 - 51	2 - 3	0.8 - 0.5^{a}	ND	13 - 13	ND	ND	Liu et al. (2019)
Oolong tea (1, 3, 5 % w/w)	80% Ethanol	Shrimp protein & chitosan	ND	126 – 114	ND	ND	ND	ND	ND	43 – 53	ND	Yuan et al. (2020)
Corn silk (1, 3, 5 % w/w)				126 - 210						43 – 48		
Black soybean seed (1, 3, 5 % w/w)				126 - 90						43 - 47		
Pomegranate peel (2, 4 ,6 % w/w)	ND	Surimi	Glycerol	86 – 96	2 – 6	160 – 64	300- 200^{a}	ND	ND	54 – 33	ND	Munir et al. (2019)
Grape skin (2, 4 ,6 % w/w)				86 - 99	2 - 5	160 - 101	300 -100^{a}			54 - 29		
White tea (20%)	Water	Furcellaran/ whey protein	Glycerol	ND	ND	26 -25	ND	ND	14.3 - 14.1	47 – 51	285 - 265	Pluta-Kubica et al. (2019)
Yerba mate (20%)						26 - 22			14 - 12	47 - 46	285 - 336	
Catechin-kradon (0, 3, 6, 9, 12 mg/ml)	Water	Fish myofibrillar	Glycerol	11 - 19	8 - 6	132 - 51	20 - 15.6^{a}	ND	30 - 16	18 - 38	ND	Kawprachu et al. (2017)
Beetroot (1, 2, 4% w/v)	ND	Furcellaran/ whey protein	Glycerol	6 - 10	25 – 33	24 – 93	ND	ND	18 -16	100 – 90	ND	Jaróz et al. (2019)
Elderberry (1, 2, 4% w/v)				6 – 9	25 – 25	57 – 62			18 – 16	100- 96		
Blueberry (1, 2, 4% w/v)				6 – 9	25 – 39	27 – 79			18 - 14	100 – 84		
Green tea (1, 2, 4% w/v)				6 – 8	25 – 26	57 – 64			18 – 16	100 – 95		
Yerba mate (1, 2, 4% w/v)				6 - 7	25 - 37	57 - 67			18 - 14	100 - 36		
Chestnut bur (20, 50, 80, 100 g/kg)	Water	Soy protein	Glycerol	ND	1.8 - 1.3	198 - 197	ND	4 - 2^{c}	56 - 63	ND	ND	Wang et al. (2016)

ND: Not detected
^{a}: (10^{-10} g/(m·s·Pa))
^{b}: (cc^{-m}/(h·m^{2}·KPa))
^{c}: (10^{-25} m^{3} m^{-1} s^{-1} Pa^{-1})

The interactions of plant extracts could influence the properties of the biodegradable films such as thickness, optical, functional, mechanical, barrier, microstructural, rheological, antioxidant, and antimicrobial and the biodegradation rate.

4.1.1. Thickness

Film thickness is an important parameter that directly affects mechanical strength, water vapor permeability, light transmittance, and opacity of films (Yong, Liu, et al., 2019). Plant extracts with their drying conditions and their preparation methods influence the thickness of the biodegradable films and it significantly affects the properties and the shelf life during packaging (Mir et al., 2018).

Most of the plant extracts showed an increase on film thickness. For example, chitosan-based film thickness significantly increased with the incorporation of the plant-derived extracts (pine nutshell, peanut shell, and winter jujube leaf) (X. Zhang, Lian, Shi, Meng, & Peng, 2020). Another study from Yuan et al. (2020) reported the same effect from oolong tea extract, corn silk extract and black soybean seed coat extract on the film which was combined with shrimp shell wastes protein and chitosan. The reason for the thicker film that was produced might be due to a higher solid content introduced into the film matrix and the interruption of the organized structure of the film matrix by extract droplets. Same explanation was adopted also from Riaz et al. (2020) who observed that the incorporation of Chinese chives root extract increases the thickness of carboxymethylcellulose films. Along with the previous studies Ju and Song (2019) observed that the addition of yellow onion peel extract increases the thickness of *Gloiopeltis furcate* funoran films from 0.060 to 0.095 mm with the increase in the extract concentration.

As revealed by the results of W. Zhang, Li, and Jiang (2019), the thickness of the chitosan film exhibited an increase with the incorporation of different concentration of banana peel extract. The same results were observed from Ju and Song (2020) who investigated the effect of peanut skin extract on the *Tremella fuciformis* polysaccharides films and showed that as the extract increases the thickness of the film increases as well. For example, the control film had thickness 0.074 mm and the film with 1 g/100 ml had 0.099 mm. Moreover, the addition of *Pseuderanthemum palatiferum* (Nees) Radlk. extract into gelatin sodium alginate films resulted in a growth in film thickness, which ranged from 0.05 to 0.11 mm as Ho et al. (2020) reported.

Pomegranate extract is widely used in many studies, for instance Y. Liu et al. (2020) investigated the effect of pomegranate peel extract and pomegranate flesh extract into κ-carrageenan films. The thickness of both active films was proportional to the content of extract. In another study, pomegranate peel and grape seed extract showed increase in the thickness of surimi based edible films. Thickness of the films ranged from 0.086 to 0.099 mm. As the amount of the extract concentration increases also increases the thickness of the films. In general films containing grape skin extract had higher thickness than those with pomegranate peel extract. The source and concentration of the plant extracts had a significant effect on the thickness of the obtained films (Munir, Hu, Liu, & Xiong, 2019).

Yong, Wang, et al. (2019) studied the effect of black and purple eggplant extract on chitosan films. The incorporation of both extracts increased the thickness of the films. However, active films with black eggplant extract were significantly thicker than with purple eggplant extract films at the same extract incorporation. The differences in film thickness between the films could be related to different anthocyanin compositions and contents in the extracts.

The effect of rosemary extracts on the thickness of the carrageenan film were studied from Nouri, Tavakkoli Yaraki, Ghorbanpour, and Wang (2018). The thickness increased as the concentration of rosemary extract increased up to 2% v/v and then the thickness decreased with further increasing extract concentration. The lowest film thickness at 3% rosemary extract may be due to the strong molecular interaction between the functional groups of biopolymer and rosemary extract, decreased OH available groups, as well as reducing moisture absorption.

Apart from increase, other effects of plant extract addition on thickness of edible films have been found in literature. *Moring oleifera* extract did not affect the film thickness of papaya based edible films (Rodríguez, Sibaja, Espitia, & Otoni 2020). Moreover, Liu et al. (2019) found that fish gelatin films with haskap berries extract (0.5 to 3 wt%) showed no significant difference as well, which was because of the low content of the extract in the film. Since the hydroxyl groups of polyphenols in extract formed intermolecular hydrogen bonds with the amino/hydroxyl groups in fish gelatin, haskap berries extract was uniformly distributed in the space among fish gelatin film matrix. Adilah, Jamilah, Noranizan, and Hanani (2018) used mango peel extract in different concentrations 1, 3 and 5% and incorporated into fish gelatin films as well. The addition of 1% of the extract into gelatin film did not give significant difference on film thickness. However, incorporation of 3-5% of mango peel extract into the films showed significant increased compared to control films. An increase in film thickness could be due to the conformational changes of gelatin chain by the addition of the extract. In addition, denser films were observed because of greater distribution of polyphenols in the polymeric film matrix.

Experimental results on film thickness of control chitosan and carboxymethylcellulose films and those with different concentrations of blueberry and red grape skin pomace extracts were analysed from Kurek et al. (2019). No significant differences in carboxymethylcellulose film thickness occurred after the addition of different extracts, while chitosan films with different extracts tended to have lower thickness compared to the control films. However, carboxymethylcellulose films were thinner than chitosan films indicating different packing of polymer chains.

On the other hand, the decrease of the film thickness with the incorporation of plant extracts it is not very common, however, Kalkan, Otağ, and Engin (2020) reported that with the addition of ethanolic *Rheum ribes* extract into methylcellulose films, showed a reduction in film thicknesses in comparison to the control is remarkable, from 131.8 µm to 107.5 µm.

4.1.2. Optical properties

The visual aspect of packaging is related to its colour and transparency; they are important attributes that affect product acceptance by consumers and have an unexpected effect in relation to the protection of food against the light permeability. In evaluation of packaging colour, parameters related to the influence of polymer compounds on luminosity, colour, and opacity are generally analysed. In general, polymers with greater transparency and less difference in colour are more desirable in the market because they provide the consumer a realistic view of the product to be purchased. On the other hand, some food products that that are light sensitive require a more opaque package that offers protection against light, which can cause oxidation reactions (Martins et al., 2019).

4.1.2.1. Colour

The apparent colour of the film can affect the appearance of packaged food and the colour is a major indicator of appearance and consumer acceptance of packaging (Ju & Song, 2020; F. Liu et al., 2020; Yong, Liu, et al., 2019). Plant extracts commonly modify the colour properties of polymers. The literature showed that the addition of plant extracts alters the original colour of developed films to some extent, the magnitude of which is dependent on the origin of plant extracts and concentration. Plant extracts are a potent source of polyphenols which leads to different types of interactions with biopolymers and exhibit changes in colour properties (Mir et al., 2018).

Most of the studies analysed the colour properties using CIE colour space L*, a*, and b* coordinates which represent lightness, red-green, and yellow-blue, respectively (Dawson & Acton, 2018).

X. Zhang et al. (2020) reported that the surface brightness (L*) of the control chitosan film was higher, and the L* value of the composite film with three plant methanolic extracts from pine nutshell, peanut shell and winter jujube leaf was lower and the colour was darker. The yellowness (b*) increased significantly with the addition of plant extracts. The composite film of pine nutshell and peanut shell indicated the high a* (red), while the winter jujube leaf film appeared the low a* (green) as Fig. 4.1 shows. The colour differences were mainly caused by the colour of plant extracts itself. Same results were obtained also from W. Zhang et al. (2019) who studied the effect of banana peel extract on the colour of chitosan films as well. Lutein which is one of the most abundant components in ripe banana peel gave the yellow colour to the active films.

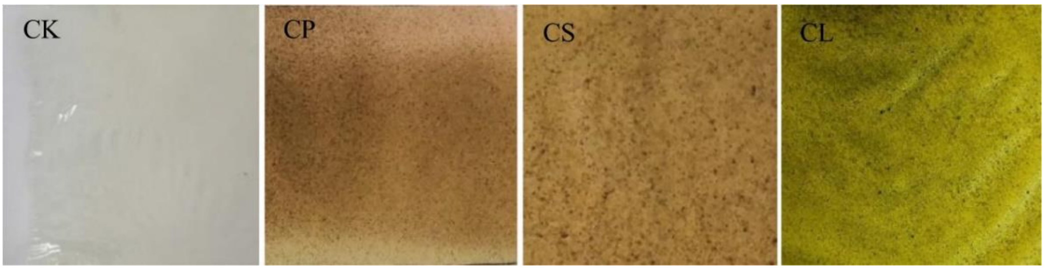


Fig. 4.1 – Appearance colour of chitosan composite films with three plant extracts. Pine nutshell (CP), peanut shell (CS) and winter jujube leaf (CL) (Source: X. Zhang et al., 2020).

The results from Yuan et al. (2020) suggested that films with shrimp shell waste protein and chitosan turn darker with increasing concentration (1 to 5%) of oolong tea extract, corn silk extract and black soybean seed coat extract. Darker films could be produced with the incorporation of phenolic compounds, which possessed an advantage to package foods that were sensitive to light. Except of oolong tea extract that showed lower a* values the other two extracts showed higher a* values comparing with the control. Regarding the b* values, corn silk extract in all concentrations had higher values from the control and the other films. The addition of different phenolic compounds contributed to intensification of the colour of hydrophilic films, which was related to their concentration. Moreover, these results were consistent with visual observations. Hence, these colour difference could be attributed to the different concentration of extracts used in the preparation of the films.

The incorporation of a peanut extract in *Tremella fuciformis* polysaccharides films was studied by Ju and Song (2020). The control film displayed a transparent and slightly yellow colour. However, as the concentration of the extract increased, the lightness of the active films decreased, on the other side a* and b* values increased. These changes may be due to the presence of an original pigment in peanut skin extract which affect the colour. Darker became also the potato starch films with the addition of rice straw extract (Menzel, González-Martínez, Vilaplana, Diretto, & Chiralt, 2020).

The higher b* values, were found when yerba mate was incorporated into furcellaran/whey protein films. The green intensity is highlighted by negative values of a* parameter. The clearest shade of green was seen in the films with yerba mate, which can be explained by the greenish colour of the extract due to the presence of natural pigments found in the leaves of *Ilex paraguariensis.* The addition of yerba mate to the film caused an increase in darkness of the active film. The phenolic components of this extract may have caused diffusion and refraction of light, which resulted in darker films (Pluta-Kubica, Jamróz, Kawecka, Juszczak, & Krzyściak, 2019).

The incorporation of green tea extract and grape seed extract into alginate lipid-based films was studied by Fabra, Falcó, Randazzo, Sánchez, and López-Rubio, (2018). The colour parameters (lightness -L*-, chrome -C*- and hue -h*-) evidence that the incorporation of natural extracts, either green tea extract or grape seed extract, led to darker (lower L*) and more reddish (lower h*) edible films, being this effect more obvious for grape seed extract. In general, the addition of green tea extract gave rise to a more saturated colour in the films (higher C*), whereas grape seed extract provided less vivid films (lower C*).

The *Gloiopeltis furcate* funoran films containing various concentrations (0.3, 0.5 and 1%) of yellow onion peel extract were analysed from Ju and Song (2019). The L* value of the films sharply decreased from 87.97 to 29.44 with an increase in the extract concentration. The redness of the films increased owing to the colour of yellow onion peel extract in the films. The b* value of the films was the highest in the film containing 0.3% extract and decreased when the concentration of yellow onion peel extract was above 0.3% as Fig. 4.2 shows. These colour changes in the films may be due to the natural reddish-brown pigment in natural extract.

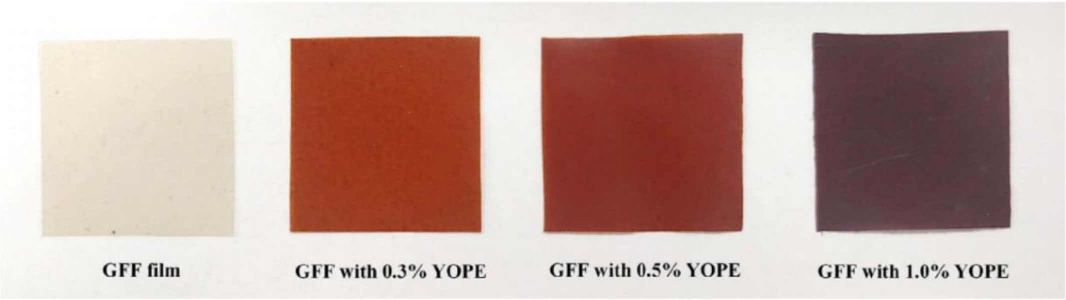


Fig. 4.2 – The photographs of the *Gloiopeltis furcate* funoran (GFF) films with yellow onion peel extract (YOPE) in different concentrations (Source: Ju & Song 2019).

The incorporation of pomegranate peel extract and grape seed extract affected the colour properties of surimi edible films. Control films showed the highest L*, lowest a* and b* value and lowest ΔE*. Films incorporated with both extracts showed the lowest L* values, which decreased in correlation with increasing concentrations of extracts. Lower a* values were observed in films including pomegranate peel extract. On the other hand, films containing grape seed extract had higher a* values, compared to the other films. Highest b* and ΔE* values were obtained from films incorporated with both extracts (Munir et al., 2019).

Farhan and Hani (2020) studied the effect of semi-refined κ-carrageenan films incorporated with water germinated fenugreek seed extracts. In general, with increasing extract concentration from 1 to 25% in the films, L* value significantly decreased from

91.51–66.87, respectively. This result indicates that the films were darker compared to the control film (93.47). In contrast, b* value significantly increased (indicator towards the yellowness) from 16.60–77.57, as the concentration of the extract increased from 1 to 25%, respectively. However, the a* value of films decreased (indicator towards the greenness) with addition of 1 % and 5 % of the extract but increased when higher amounts of the extract (10–25 %) were added. These properties make the active films shielded from the light, enabling them to provide increased protection to packaged foods against discoloration, off-flavour and nutrient losses caused by light-induced oxidation.

The addition of a chestnut bur extract in polymeric matrices based on soy protein was studied by Wang, Hu, Ma, and Wang (2016). As expected, the strong brownish colour of the extract significantly affected the film's colour by a significant increase in redness (a*) and yellowness (b*) and a reduction in the lightness (L*), as the extract level increased.

Colour parameters of fish gelatin films in combination with mango peel extract in different concentrations (1, 3 and 5%) are presented from Adilah et al. (2018). Obtained results showed that the addition of extract significantly affected the colour of the film surface. Incorporation of mango peel extract from 1 to 5% showed significant decrease of L* and a* values. Especially, addition of 1% extract significantly reduced film lightness from 89.07 to 87.88. Increasing mango peel extract concentration from 3 to 5% also has lowered the film lightness but no significant difference was observed. Meanwhile, extract addition has increased the b* value of gelatin film significantly compared to control film. Variations of films colour are most likely from natural pigments of coloured compounds present within the extract.

Yang, Lee, Won, and Song, (2016) studied the optical properties of protein films (extracted from distiller dried grains with solubles) containing tea extracts (green, oolong and black). The lightness L* and yellowness b* of the films containing tea extracts decreased, but the redness a* increased, comparing with the control films. The reason might be explained by the fact that films were affected by the addition of oolong tea extract and black tea extract containing theaflavins which have brown colour and they are generated by polyphenol oxidation.

4.1.2.2. Transparency

The food is easily oxidative under the exposure of visible light and induces to the colourless, loss of nutrients and off flavours. Hence, the barrier property of the films to visible light is one of the key parameters to evaluate the protective effects of food packaging materials (Nguyen et al., 2020). For use as packaging materials, transparency of films is required to meet consumers' desire to see food through packaging. Plant extracts commonly provide opacity to polymers and as a result, films containing extracts are less transparent than films without extracts. The addition of plant extracts in films provides an adequate barrier to light, which is important for preventing the degradation of light-sensitive components. These results revealed the beneficial effects of adding antioxidant rich plant extracts to packaging films (Mir et al., 2018).

For instance, the UV visible light transmittance of chitosan film was remarkably reduced by the incorporation of increased concentration of purple and black rice extract. The potent UV visible light barrier ability of chitosan films with the natural extract was because the abundant aromatic rings in the structures of polyphenols could absorb UV–vis radiation. Notably, chitosan films with black rice extract had stronger UV visible light barrier ability than with purple rice extract at the same concentrations levels which was because black rice extract had higher total phenolic content than purple rice extract (Yong, Liu, et al., 2019). Moreover, Kaewprachu, Rungraeng, Osako, and Rawdkuen (2017) suggested that fish myofibrillar protein films incorporated with catechin-Kradon extract was more sufficient in blocking UV and visible light transmission than the control and LDPE films. Therefore, the active films with the natural extract can be beneficial for food packaging applications, especially high lipid foods, in order to retard lipid oxidation.

Yuan et al. (2020) prepared active films from shrimp shell waste protein mixed with chitosan containing oolong tea, corn silk and black soybean seed extract. At all wavelengths tested (200-800 nm), light transmission level of all films was lower than that of the control film. The decrease in light transmission might be attributed to the addition of polyphenols compounds in the natural extracts, and those films could effectively restrain lipid oxidation induced by UV light in food system.

In general, the greater “transparency value” represents the lower transparency of film. For example, Campa-Siqueiros et al. (2020) showed highest transparency values in those agar films with the highest concentration of hydroalcoholic garlic extract. The bioactive compounds in the natural extracts (antioxidants and pigments) can produce light brown colour in films because these compounds are unstable to light, O_2, pH, temperature, etc.

Moreover, the transparency of the surimi edible films incorporated with the pomegranate peel and grape seed extracts was less as compared to control film and decreased with increasing concentrations of extracts. This study indicated that all phenolic compounds of pomegranate peel extract and grape seed extract efficiently prevent transmission of UV and visible light by protein films (Munir et al., 2019). In another study, Menzel et al. (2019) showed the same results, meaning as the concentration of the sunflower hulls increases into potato starch-based films, the transparency of the films decreases and become more coloured.

The increased concentration of *Sonneratia caseolaris* (L.) Engl. leaf extract was the main factor reducing transparency of chitosan films, meaning that the active films have good light barrier property to protect the inside product of packaging materials against visible light. On the other hand, the films are also enough transparent to satisfy consumers demand (Nguyen et al., 2020). Moreover, same results were obtained from Ho et al. (2020), the incorporation of *Pseuderanthemum palatiferum* (Nees) Radlk. extract into gelatin-sodium alginate decreased the light transmittance with an increase in extract concentration (from 0 to 2.5%).

Optical properties of the pure cassava and its rosemary extract composite films were determined by measuring percent transmittance of light at 600 nm (Piñeros-Hernandez, Medina-Jaramillo, López-Córdoba, & Goyanes, 2017). All films were clear enough to be used as see-through packaging. The film with the highest concentration of rosemary extract showed a significant decrease in film transparency.

Wang et al. (2016) suggested that chestnut bur extract, at higher amounts (80 to 100 g/kg), contributed to limiting the light transmission of soy protein isolate films at visible range. This decrease in UV light transmission can be beneficial for food preservations.

As shown in a Fig. 4.3 the colours of chitosan films incorporation with purple eggplant extract and black eggplant extract were significantly different from that of chitosan film. Control film was pale yellow and transparent, whereas active films were blue. Moreover, the colours of both films deepened when purple eggplant extract and black eggplant contents increased from 1 to 3 wt%. Notably, films with black eggplant extract were darker at the same extract incorporation levels (Yong, Wang, et al., 2019).

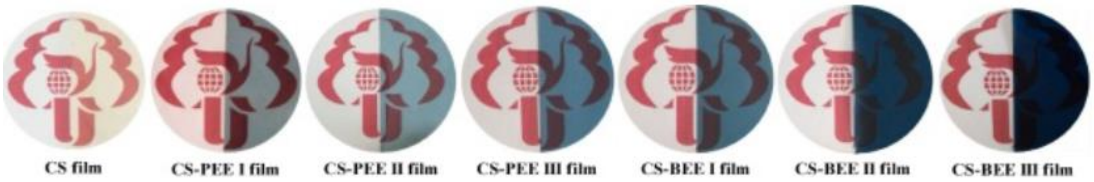


Fig. 4.3 – Physical appearances of chitosan (CS), chitosan purple eggplant extract (CS-PEE) and chitosan black eggplant extract (CS-BEE) films (Source: Yong, Wang, et al., 2019).

Active alginate films containing green tea extract and grape seed extract were investigated from Fabra et al. (2018), and they found out that the absorbance of the extracts contributed to lowering the transmittance (being greater in those containing grape seed extract), indicating that these films were more opaque and heterogeneous than their counterparts prepared without the extract. These effects can be explained by the light selective absorption of polyphenols present in the extracts at low wavelengths that imparts a reddish colour to the films, thus decreasing the hue and transmittance values at low wavelengths.

Apart from “transparency value”, many researchers measure the opacity of the films. As an example, the addition of 1% of the mango peel extract did not show any significant different of opacity compared to control film (gelatin). However, adding mango peel extract concentration from 3 to 5% showed significant increase of opacity from 0.57 to 1.11, respectively. This suggested that opaqueness of gelatin films was contributed by the extract compound added within the gelatin films (Adilah et al., 2018). Moreover, Riaz et al. (2020) prepared active films from carboxymethylcellulose containing Chinese chives root extract. As the concentration of the extract increases the films become opaquer, this might happen due to intermolecular bonding produces by the carboxymethyl cellulose and extract.

4.1.3. Mechanical properties

The mechanical properties of packaging films are associated with the behaviour of the material against forces applied on the surface (Martins et al., 2019). Packaging materials must withstand mechanical stress due to transportation, storage, and consumers themselves. This can lead to diminished containment protection. Since protection is the first and most important purpose of the packaging material, determination of its mechanical properties is indispensable (Schmid, & Müller, 2019). The mechanical properties such as elongation break, tensile strength and elastic modulus, of the films are very important as a determining factor of the packaging material (Ju & Song, 2020; Nouri et al., 2018a; H. Wang et al., 2016). Tensile strength and elongation break represent the film´s resistance to elongation and its stretching capacity, respectively, whereas elastic modulus is a measure of the stiffness of films (Cano, Andres, Chiralt, & González-Martinez, 2020).

These attributes may vary depending on the origin of the polymer, method of manufacture, thickness and exposure to agents that promote changes in polymer structure, such as heat, moisture, light, and others. In addition to these factors, incorporation of active and antimicrobial compounds in materials often has the capacity to influence the mechanical properties (Martins et al., 2019).

The studies published on the effect of plant extracts in the mechanical properties of edible films have revealed diverse effects. Many works have reported an increase of tensile strength and elongation break with the addition of natural extracts. For example, with the incorporation of haskap berries extract on fish gelatin films, the tensile strength and elongation break of the films increased as the concentration of the extract increased (J. Liu et al., 2019). The films were more compact and resistant to apply tensile strength because of the intermolecular hydrogen bonds formed between the extract and biopolymer. Moreover, polyphenols can act as plasticizers and enhance the flexibility of the active films. Similar results were obtained from Yong, Liu, et al. (2019) with the incorporation of black and purple rice extract into chitosan films. However, when the content of extract exceeded 1wt%, the tensile strength of chitosan films with both extracts gradually decreased, because the agglomeration of extract disrupted the homogeneity and compactness of film network.

Increase of tensile strength and elongation break was obtained also with the addition of pomegranate peel and flesh extract into κ-carrageenan films. Due to the presence of abundant hydroxyl groups, pomegranate flesh and peel extract could form hydrogen bonds with κ-carrageenan chains as well as glycerol, thereby enhancing the compactness of κ-carrageenan-based films (Y. Liu et al., 2020). Improved of mechanical properties occurred also when *Centella asiatica* extract was added to bovine-based gelatin film. The phenolic compounds underwent hydrophobic interactions with hydrophobic region of gelatin. This might have occurred because the phenolic compounds contain many hydrophobic groups (Rasid, Nazmi, Isa, & Sarbon, 2018).

On the contrary, some works reported adverse results on the mechanical properties with the addition of natural extracts. A study from X. Zhang et al. (2020) reported decrease of tensile strength and elongation break when natural plant extracts (pine nutshell, peanut shell and winter jujube leaf) were added to chitosan films. In three plant powder, all of the antioxidant components such as 5,7-dihydroxychromone, luteolin, quercetin, apigenin and catechin contained a lot of benzene ring structure which could insert the chitosan film matrix and caused the decrease of tensile properties. Moreover, the tensile strength of *Gloiopeltis furcate* funoran film incorporated with yellow onion peel extract decreases as the concentration of the extract increases. The elongation break values of film containing 0.3, 0.5 and 1% yellow peel onion extract were 35.7, 28.50 and 20.49% respectively. These results suggest that the incorporation of extract to the biopolymer film results in structural changes among funoran polymers through interactions between funoran and extract molecules (Ju & Song, 2019).

Kalkan et al. (2020) evaluated not only the tensile strength and elongation break but also Young´s Modulus values of the methylcellulose edible films with *Rheum ribes* extract. The addition of extract decreased the resistance of the edible methylcellulose films. The addition of antimicrobial agent increased film fragility. At the same time, the film's elongation at break, tensile strength, durability, and elasticity values have decreased.

The addition of banana peel extract contributed to the changes to the mechanical properties of chitosan films. It is noteworthy that as the concentration of banana peel extract increased, the opposite occurred, with the significantly lower tensile strength and elongation break at 8% and 12% of banana peel extract (W. Zhang et al., 2019).

Nevertheless, some other studies report increase of tensile strength but at the same time decrease of elongation break. As the concentration of the peanut skin extract increases the tensile strength of the polysaccharides films also increased. In contrast, the addition of the peanut skin extract was found to decrease the elongation break of the films. These findings are explained by the interactions between biopolymer functional groups and peanut skin extract (Ju & Song, 2020). Additionally, Ho et al. (2020) prepared films from gelatin sodium alginate containing *Pseuderanthemum palatiferum* (Nees) Radlk. extract. Tensile strength and Young modulus increased with the increase in extract concentration. On the other hand, elongation break significantly diminished with the growth of the extract amount used in active films. Pomegranate peel extract and grape seed extract in the surimi based edible films had the same results. The interactions between surimi proteins and phenolic compounds may have also reduced the effects of plasticizer in the film, consequently leading to greater rigidity of films incorporated with extracts (Munir et al., 2019).

Decrease also in tensile strength has been reported as well. Campa-Siqueiros et al. (2020) investigated agar film in combination with hydroalcoholic garlic extract and the results showed that the incorporation of the extract (0.5 μg/ mL) produced a remarkable decrease in tensile strength. Therefore, at this concentration, the hydroalcoholic does not interact with the agar chains and reduces the stress transfer in the agar chains. Pomegranate seed ethanolic extract caused also decrease in tensile strength and increase in elongation at break of the cellulose films as Nemazifard, Kavoosi, Marzban, and Ezedi (2017) observed. This phenomenon could be related to replacement of original hydrogen bonds between cellulose chains with new hydrogen bonds between cellulose and pomegranate seed extract, which caused an increase in the segmental mobility of cellulose chains. The increasing flexibility of the film samples by the addition of the extract, as described above, could be related to the formation of new hydrogen bonds between extract and cellulose chain which resulted in a loss compact structure of cellulose matrix and more sliding effect of cellulose chains against each other.

However, the addition of some plant extracts can show non-significant effect on the tensile properties of the film. The addition of chestnut bur extract to soy protein isolate film it had no effect on the tensile strength and elongation break of the films. These results indicate that excessive chestnut bur extract may lead to poor dispersion in the protein film and impede the intermolecular interactions, lowered the tensile strength. Therefore, at concentrations above a certain limit, chestnut bur extract contribute to a reduction in the tensile strength (H. Wang et al., 2016).

4.1.4. Barrier properties

The ability of packaging films or coatings to protect packaged food from negative environmental factors can be determined by barrier property characterization. Barrier properties of polymers are well known and are important to achieve the desired properties for packaging of food. Water vapor, oxygen, or other gases as well as fluids or flavourings can have a significant impact on the product shelf life when coming into contact with the packaged goods. Depending on the product, high or low barrier properties for certain substances are required (Schmid & Müller, 2019). The addition of compounds to the polymer can alter the structure of films and packaging, promoting better properties against moisture and providing greater protection to the stored product (Martins et al., 2019).

4.1.4.1. Water vapour permeability

One of the main functions of food packaging films is to restrict moisture transfer between food and the surroundings. The determination of the barrier properties of polymer is important to estimate and predict the shelf life of the packaged product. Water vapour permeability is a vital barrier parameter reflecting the ability of film against water vapor (Yong, Liu, et al., 2019; Yong, Wang, et al., 2019). Low water vapour permeability means less water vapor transmission between food and surrounding atmosphere, which is desirable for prolonging shelf life (Tong et al., 2020).

Plant extracts significantly affect the barrier properties of polymer-based films. Significant decreases in water barrier properties have been reported when plant extracts were added to various biopolymer films. The permeability of a film depends on its chemical structure and morphology, the nature of permeant and temperature of the environment. The water vapour permeability of the films containing plant extracts has been great interest (Mir et al., 2018) cause moisture plays a key role during preservation to extend the shelf life and quality of packed foods (Munir et al., 2019).

The water vapour transfer processes in films depend on the hydrophilic-hydrophobic ratio of the film constituents (Atarés & Chiralt, 2016). The incorporation of pomegranate peel and grape seed extract resulted in decrease in water vapour permeability coefficient of surimi based edible films. It is suggested that the lower water vapour permeability of films contained phenolic compounds is directly related with higher levels of hydroxyl groups (OH) and cross-linking between proteins and phenolic compounds (Munir et al., 2019). Moreover, similar reduction found from Fabra et al. (2018) who studied the effect of green tea and grape seed extract as well into alginate-lipid films. This effect can be ascribed to the presence of polyphenols which can form hydrogen bonds and hydrophobic interactions with the polar groups in the lipids, thus, limit the amount of free OH groups which could interact with water.

Chitosan is a widely used biopolymer in many studies. The addition of banana peel extract, black and purple rice extract and black and purple eggplant extract into chitosan films decreased the water vapor permeability of the films. This may be due to the interactions of hydroxyl and carboxyl groups of the extract with the hydroxyl and amino groups in chitosan, or because of the low content of extract which could be well distributed in the film (Yong, Liu, et al., 2019; Yong, Wang, et al., 2019; W. Zhang et al., 2019). Talón et al. (2017) evaluated the barrier properties of chitosan- starch film enriched with thyme extract. The addition of thyme extracts significantly decreased the water vapour permeability values of the films.

The water vapour permeability of fish gelatin film was affected when haskap berries extract was added. With the increase of the extract concentration the water vapour permeability decreases, which happens because, the incorporation of the extract into fish gelatin film produced more compact network (J. Liu et al., 2019). Y. Liu et al. (2020) reported as well the reduction of water vapor permeability values of κ-carrageenan-pomegranate flesh and peel extract films, indicating the water vapor barrier property of κ-carrageenan film was enhanced by the incorporation of the extract. A study from Tong et al. (2020) studied the effect of the incorporation of grape peel extract into konjac glucomannan based film. The results showed lower values of water vapour permeability when the extract was added, which may be due to some free grape peel extract blocked the intrinsic hydrogen bonds in biopolymer matrix prolonged tortuous pathways of the water vapor diffusion.

Aside from the reduction of water vapour permeability with the addition of some plant extracts, some other studies had the opposite results. X. Zhang et al. (2020) reported that water vapour permeability of composite films with methanolic plant extracts from pine nutshell, peanut shell and winter jujube leaf was higher than that in the chitosan control film. This might be attributed to the good apparent porosity of seed shell compared with other material. Moreover, Ju and Song (2020) studied the effect of

Tremella fuciformis polysaccharides film in combination with peanut skin extract and showed that as the concentration of the extract increases the water vapour permeability of the film increases as well, which is due to allowing cracks and pores that produced more heterogeneity in the film matrix. In different study, the incorporation of pomegranate seed ethanolic extract into the cellulose films caused a significant increase in their water vapour permeability as well. Pomegranate seed extract interacts with hydroxyl groups of the polymer chain and competitively weakens chain-to-chain hydrogen binding in polymer structure. Moreover, the original hydrogen binding between polymer chains replaced with the new hydrogen bonds between hydroxyl groups of polymer and extract. This event increases the distance between polymer chain and introduced twisting pore in the polymer matrix. It results to increased water vapor transmission across the film samples (Nemazifard et al., 2017). Ju and Song (2019) showed that the water vapour permeability increased with an increase in the yellow onion peel extract concentration in the *Gloiopeltis furcate* funoran films. This increase in water vapour permeability of the biopolymer films with the extract could be attributed to the pores formed during the preparation of the films containing yellow onion peel extract.

Other than increase or decrease of water vapour permeability with the addition of plant extracts, some studies reported no significant effect. For instance, durian leaf extract (0.2 and 0.5%) showed no significant difference when it was incorporated into gelatin-based films. The results showed that the active films with both concentrations did not show any significant difference with the control film. This can be explained from the usage level of leaf extract at 0.2 and 0.5 % which was incorporated into the biopolymer (Joanne Kam et al., 2018).

4.1.4.2. Oxygen permeability

Gas barriers are also important parameters to be evaluated in the production of packaging, according to the needs of each stored product (Martins et al., 2019). Many decomposition reactions in food and other products are oxygen related. Fat rancidity, microbial growth, enzymatic browning, and vitamin loss are just some examples. Hence, many products require oxygen-protective packaging (Schmid & Müller, 2019). Gas barrier properties of films incorporating with plant extracts, particularly those of CO_2 and O_2 are very important and therefore becoming of great interest (S. Wang et al., 2012). For fresh fruit and vegetable products, however, oxygen and carbon dioxide are essential for respiration during storage and demand moderate barrier packaging (Schmid & Müller, 2019).

The oxygen permeability of edible films is measured much more rarely than water vapour permeability. This property is greatly affected by relative humidity and temperature at which film equilibrates since diffusion depending properties are dependent on molecular mobility, which increases when the film moisture content or temperature increase (Atarés & Chiralt, 2016).

In general plant extracts, decrease the oxygen permeability of the films. Like, the incorporation of coffee and rice husks extracts caused a significant decrease of oxygen permeability of corn starch films. The reduction was depended on the amount of extract solids, and can be associated with the oxygen scavenging effect of the compounds with antioxidant capacity (Collazo-Bigliardi, Ortega-Toro, & Chiralt, 2019). Moreover, Menzel, González-Martínez, Vilaplana, Diretto, and Chiralt (2020) studied the effect of rice straw waste extract on oxygen permeability of starch films as well. The results showed that the antioxidant extract seemed to limit the mass transport of oxygen molecules through the films. Moreover, starch films with the highest amount of extract showed the greatest improvement of oxygen barrier properties compared to control film.

Chitosan films containing propolis and spirulina extracts were developed by Siripatrawan and Vitchayakitti (2016) and Balti et al. (2017), respectively. The film´s oxygen permeability decreased with increasing propolis/spirulina concentration, which might be due to interaction between chitosan polymer matrix and phenolic compounds of the extracts. The same reduction with the increase of extract concentration happened when licorice residue extract was added into soy protein films. This could be rely to the rough structure of the composite films, with an increased tortuosity factor for oxygen transfer, raising the oxygen barrier properties of active films (Han, Yu, & Wang, 2018).

Moreover, Menzel et al. (2019) reported that the incorporation of sunflower hulls extract on starch films increases the oxygen barrier capacity as the concentration of phenolic extract increase. This improvement of the oxygen barrier capacity could link to the decrease of glycerol content in the film and hence the formation of a more tightly packed network structure with reduced molecular mobility.

4.1.5. Functional properties

4.1.5.1. Water content

Water content affects the capacity of water-resistant properties, as well as their subsequent use. Food packaging films should maintain certain moisture levels within the packaged products. Moisture content reflects the ability of packaging films to absorb moisture from relatively high humid environment (Yong et al., 2019).

The decreasing trends of the water content was observed when chitosan film was incorporated with banana peel extract, plant-derived extracts (pine nutshell, peanut shell and winter jujube leaf), and *Sonneratia Caseolaris* (L) Engl. fresh leaves extract (Nguyen et al., 2020; W. Zhang et al., 2019; X. Zhang et al., 2020). The decrease of water content in films was mainly due to the interaction among chitosan and plant extracts, thus the interaction between chitosan and water was restricted.

Rosemary and pomegranate peel and flesh extract into κ-carrageenan films were analysed from Y. Liu et al. (2020) and Nouri, Tavakkoli Yaraki, Ghorbanpour, and Wang, (2018) respectively. Both results showed that with the addition of the extracts the water content of carrageenan films decreased. The phenomenon may be associated with hydrophobic nature of extract and intermolecular interactions of hydrogen bonding between hydroxyl groups of carrageenan and plant extract, leading to reduced available hydroxyl groups for interaction with water molecules.

The developed fish myofibrillar protein films´ moisture content was affected by the addition of catechin-Kradon extract. The moisture content of the experimental films gradually decreased with increasing concentrations of the extract. This might be because interactions were induced between protein and the phenolic compounds present in catechin-Kradon extract (Kaewprachu, Rungraeng, et al., 2017). Moreover, Ju and Song (2019) studied the effect of yellow onion peel extract in *Gloiopeltis furcate* funoran films. The moisture content of the films decreases from 23.03% to 13.39%, as the concentration of the extract increases. These results can be explained due to the polyphenol compound contents in yellow onion peel extract.

Some plant extracts though, increased the water content of the films. For example, soy protein films were greatly influenced by the addition of chestnut bur extract, as Wang et al. (2016) reported. The active film with the natural extract had significantly higher moisture content than the control films. These results were attributed to the hydrophilic nature of chestnut bur extract, which led to an increase in the affinity of the films for water. Another study showed that the incorporation of black eggplant extract significantly increased the moisture content of chitosan films, which might be related with the hydrophilic nature of anthocyanins in black eggplant extract. However, when purple eggplant was added to chitosan films no significant difference in water content was noticed (Yong, Wang, et al., 2019).

No significantly difference was observed also when *Pseuderanthemum palatiferum* (Nees) Radlk. was incorporated into gelatin sodium alginate-based films. The extract may possess the sole property of having both hydrophilic and hydrophobic constituents that can balance the hygroscopic properties, causing no effect on the moisture content of the films (Ho et al., 2020). Moreover, the addition of haskap berries extract into fish gelatin films did not show as well a significant change in the moisture content of the films. On one hand, the presence of hydrophilic compounds in the extract could increase the affinity of the films toward water molecules. On the other hand, the interactions between polyphenols and gelatin could decrease the availability of the hydrophilic groups in the films, thereby reducing the affinity of the films toward water molecules (J. Liu et al., 2019).

4.1.5.2. Solubility

Water solubility is an important property of bio-based films that gives indication of the film’s affinity to water. It is characterized as the resistance or tolerance of the polymer to water or moisture. This property is totally related to the chemical structure and hydrophobicity of the surface of the material (Martins et al., 2019). This property becomes significant when films come in contact with high moisture food products during storage (Kurek et al., 2019; Yuan et al., 2020). Highly soluble films can be used as edible coatings, whereas insoluble films can provide a protective function for products with high humidity (Pluta-Kubica et al., 2019).

In most cases, natural polymers present high solubility; in this context, some strategies such as the addition of compounds may be used to control film solubility. Lower solubility leads to greater stability and less interaction with humidity (Martins et al., 2019).

Munir et al. (2019) observed that water solubility was lower in surimi edible films incorporated with different concentration (2%, 4% and 6%) of pomegranate peel extract and grape seed extract as compared to film without extract. It is assumed that the lower solubility is attributed by the hydrophobic nature of the phenolic compounds contained in extracts.

Moreover, increasing concentration of mango peel extract and pomegranate peel extract lowered the solubility of gelatin films as it was respectively reported from Adilah, Jamilah, Noranizan, and Hanani, (2018) and Hanani, Yee, and Nor-Khaizura, (2019). The decrease could be due to high protein-polyphenols interaction during which it formed stronger film network structure. Riaz et al. (2020) reported also that the formation of strong bonding between carboxymethylcellulose matrix and polyphenols compounds from Chinese chives root extract decreased the water solubility of the films.

Furthermore, Ju and Song (2019) studied the effect of yellow onion peel extract in *Gloiopeltis furcate* funoran films. The water solubility of the films decreases as the concentration of the extract increased. Jaróz, Kulawik, Guzik, and Duda (2019) reported that furcellaran films are 100% water soluble, and the addition of beet root, elderberry, blueberry, green tea, and yerba mate extracts only slightly reduced the solubility. Therefore, those films they cannot be used in food products with a high hydrophilic character.

In other studies, the addition of plant extracts resulted increase of the solubility of the active films. Ho et al. (2020) showed that the incorporation of *Pseuderanthemum palatiferum* (Nees) Radlk. extract increased the water solubility from 35 to 39 %, in gelatin sodium alginate films. The increase could be related to a poorer cross-linking level through disulphide bridges relative to control films, owing to interferences of polyphenols with protein-protein interactions preventing aggregation. Increase in fish myofibrillar protein film solubility was also noted as the level of catechin-Kradon extract concentrations increased. Although, the interactions of protein-polyphenol were expected to reduce the films' solubility, it seems that the film solubility is the measurement of the soluble substances that contain in the film. So, the soluble substances could escape from the film during immersion in distilled water. Catechin-Kradon could be released into distilled water due to their hydrophilic nature (Kaewprachu, Rungraeng, et al., 2017).

The water solubility of chitosan and chitosan/protein films increased gradually by the addition of *Sonneratia caseolaris* (L.) Engl. leaf extract and three natural extracts, oolong tea, corn silk, black soybean seed coat as Nguyen et al. (2020) and Yuan et al. (2020) reported respectively. However, when the oolong tea, corn silk, black soybean seed coat extracts reached a certain high concentration, the solubility of the extract in the film forming solution was limited.

4.1.5.3. Swelling

Swelling degree is an instrumental parameter related to degree of cross-linkage occurred in polymer network and for this reason affects to water resistance of film; the lower the swelling degree of polymeric films is, the higher the water resistance of film is. The reduction of water adsorption through films is normally preferred in packaging application. Moreover, it was reported that the swelling degree strongly depend on the number and nature of intermolecular interactions in polymer chains (Nguyen et al., 2020).

Plant extracts are affecting the functional properties of biodegradable packaging films (Mir et al., 2018). Chinese chives root extract was incorporated into carboxymethylcellulose based film which significantly decreased the swelling degree of the films as the concentration of the extract increases. The existence of hydrophilic groups such as carboxylic groups in the carboxymethylcellulose molecule is mainly responsible for the swelling behaviour of the intrinsic (Riaz, Lagnika, Luo, Nie, et al., 2020). Nemazifard et al. (2017) evaluated the swelling capacity of carboxymethylcellulose, hydroxyethylcellulose, hydroxypropylmethylcellulose, and methylcellulose films incorporated with ethanol extract of pomegranate seed. The results showed that the natural extract could reduce the swelling capacity of the films. The hydroxyl groups of cellulose can interact with the extract through hydrogen bonding, increasing the interaction between cellulose and pomegranate extract. This event saturates the cellulose network with extract, preventing the water molecules from interacting with cellulose and decreasing swelling. Similar decreased of swelling degree occurred when *Sonneratia caseolaris* (L) Engl. fresh leaves extract added into chitosan films.

Apart from the reduction some extracts can cause increase of swelling degree. Pluta-Kubica et al. (2019) suggest that yerba mate extract tends to increase the values of furcellaran-whey protein isolate films. However, there were no significant differences between control films and films that are incorporated with white tea.

4.1.5.4 Contact angle (θ)

The films must be designed considering some surface properties in order to satisfy adequate adherence and thickness on food surface. With an adequate spread of the film solution on the food surface, it is possible to avoid the inter-spaces between food and film and avoid thicker film zones that can produce an anaerobic condition that would conduce to the food deterioration. This affinity is fundamental in the coating design, considering that the effective spreading of a coating solution on a fruits skin is greatly influenced by the wettability of the surface by coating solutions. The main parameter used to characterize the wetting of a surface is the equilibrium contact angle (θ) (Ramírez, Gallegos, Ihl, & Bifani, 2012).

The average contact angle (θ) is basically defined as the angle between the solid-liquid surface (γ_{sl}) and a tangent drawn on the drop surface (γ_{lv}), which passes through the triple point of atmosphere-liquid-solid. The average contact angle value gives information about wettability of the surfaces. A large contact angle (or small cos θ) refers a hydrophobic surface while a small contact angle (or large cos θ) corresponds to a hydrophilic surface. The quantitative definition of the relative terms “hydrophobic” and “hydrophilic” surfaces has been done exhibiting a water contact angle $\theta > 65^{\circ}$ and $\theta < 65$, respectively (Kalkan et al., 2020).

Methylcellulose films incorporated with *Rheum ribes* extract exhibited hydrophilic character when compared to the other surfaces. As the amount of the natural extract increased, the average contact angle values of the films decreased. In other words, their hydrophilic properties were improved. Control film specimens showed the highest contact angle (45.10º) in all film surfaces, whereas the active films added with 2% of extract had the lowest contact angle (10.28º), and this can be attributed to *Rheum ribes* extract that could not interact effectively with the active methylcellulose sites and the hydrophilic groups are not embedded in the film surface (Kalkan et al., 2020).

The addition of rosemary extract into cassava starch films increased contact angle θ from 37º to 51º. This behaviour can be explained on the basis of hydrophobicity of the main bioactive compounds isolated in aqueous rosemary extracts (rosmarinic and carnosic acid) (Piñeros-Hernandez et al., 2017).

Rambabu, Bharath, Banat, Show, & Cocoletzi, (2019) incorporated mango leaf extract into chitosan films. Results indicated reduced hydrophilic nature of active films with increase in extract content. Significantly increase in the contact angles for active films is attributed to the inclusion of polyphenolic compounds contained in the mango leaf extract into the chitosan network causing decreased wettability.

The addition of pecan nutshell and hazelnut skin extract in different concentrations into octenyl succinate starch gradually increased the contact angle of the films. This could be related to the interaction between the hydroxyl groups of the phenolic compounds and starch, whereby the amount of free hydroxyl groups, which could interact with the water, is limited. In reaction, the hydrophobicity of the films increased. For this reason, the solubility of the films with the increase of pecan nutshell and hazelnut skin extract concentration decreased, while their hydrophobicity increased (Leon-Bejarano, Durmus, Ovando-Martínez, & Simsek, 2020).

(Kaya et al., 2018) evaluated the contact angle of chitosan films after the addition of methanol extracts of stem, leaf, and seed obtained from *Pistacia terebinthus* which are rich in phenolic compounds. Water wettability of the film surface is associated with the final angle of the water drop. In order to check the effect of the stem, seed and leaf extract on film wettability, the contact angle values were calculated for chitosan control, chitosan-stem, chitosan-seed and chitosan-leaf films as 86.3 ± 1.43°, 82.1 ± 1.83° and 91.35 ± 3.05° and 61.8 ± 5.54°, respectively. Chitosan-leaf extract film showed hydrophilic feature as its contact angle observed at 61.8º. However, chitosan-leaf extract film had lower water solubility rate.

4.1.6. Microstructural properties

The properties of polymer microstructures are essential for good performance in the production of packaging. Through evaluation of these attributes, it is possible to observe the surface and the sectional area, as well as to verify the homogeneity, roughness, crystallinity, and interactions between the multilayers. It is also possible to check the presence of defects, and the micro-holes ruptures which can lead to the loss of mechanical and barrier properties. More than that, it is even possible to identify conformations and molecular structures added to films (Martins et al., 2019).

The qualitative observation of the components arrangement into the film structure is often accomplished using Scanning Electron Microscopy (SEM) or Transmission Electron Microscopy (Atarés & Chiralt, 2016). Scanning electron microscope images provide information on the surface homogeneity of films. A homogeneous film surface is considered to be an indicator of structural integrity and the mechanical properties and water vapour permeability mechanisms of such films are expected to be good (Kalkan et al., 2020).

Plant extracts significantly affect the structural properties of films. The microstructure of films is influenced by the arrangement of components in the film forming a dispersion (Mir et al., 2018). Pomegranate seed and *Rheum ribes* extracts incorporated into cellulose and methylcellulose films respectively, affected the structural properties of the films as Kalkan et al. (2020) and Nemazifard et al. (2017) reported respectively. Control films had smooth and continuous surfaces without holes, friable areas, or bubbles. However, the addition of the natural extract influenced the surface of the film samples.

Nguyen et al. (2020) evaluated the microstructure of chitosan films incorporated *Sonneratia caseolaris* (L.) Engl. leaf extract. The control film presented plane, smooth, homogenous and compact surface without fracture as it is been observed in Fig. 4.4 whereas with the addition of *Sonneratia caseolaris* (L.) Engl. leaf extract, the surface of films became rough and less compact. This may be presumably caused by the aggregation of extract compounds.

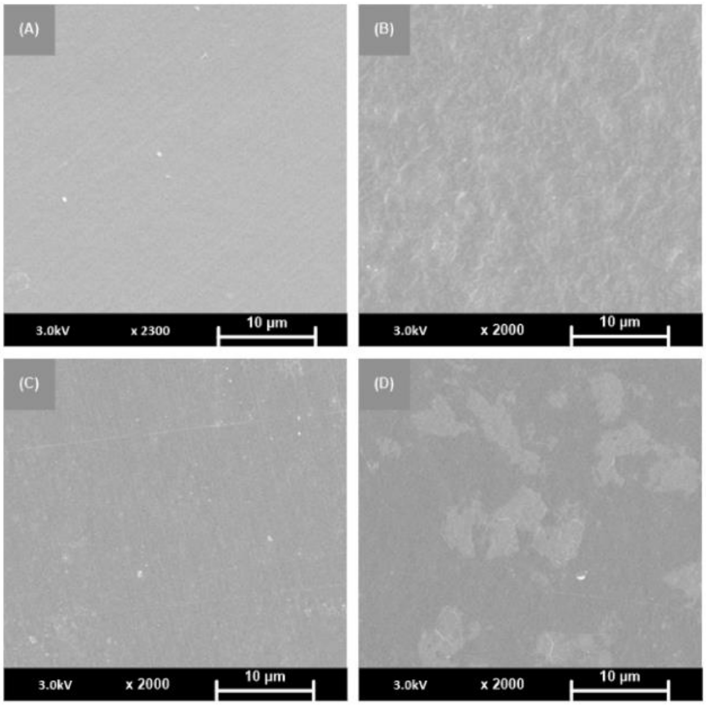


Fig. 4.4 – SEM of control film (A), CH-SCELE-1 % (B), CH-SCELE-2 % (C) and CH-SCELE-3 % (D). CH: chitosan, SCELE: *Sonneratia caseolaris* (L.) Engl. leaf extract (Source: Nguyen et al., 2020).

Yuan et al. (2020) and W. Zhang et al. (2019) also focused on the evaluation of the microstructure of chitosan films incorporated with three extracts (from oolong tea, corn silk and black soybean seed coat) and banana peel extract, respectively. The surface of the control film was smooth and homogeneous with no brittle areas or bubbles. Regardless of the content, the micrographs films added with black soybean seed coat extract had similar characteristics with the control film. While the films incorporated with oolong tea and corn silk extract appeared to consist of coarse texture, which was indicative of a ducile film structure. This can be attributed to the low solubility of the extract in the film-forming solution when the extract reached a certain high concentration. In the case of banana peel extract, several small white spots were observed on the surface of the chitosan films with 8 and 12% extract, which leads to more porous films.

Cano, Andres, Chiralt, and González-Martinez (2020) used three tannins from white peel grape, red peel grape and oak bark to obtain active film based on proteins (caseinate and gelatin). Pure caseinate and gelatin films presented a compact, homogeneous, and continuous structure. The incorporation of tannins led to some differences at microstructural level (i.e., more cracks were observed). Piñeros-Hernandez et al. (2017) reported that cassava control film had a homogeneous structure (without pores and cracks) as well, but when rosemary extract was incorporated into the starch film some cracks were observed on the fractured surfaces.

Despite of the changes that some plant extracts showed on the surface of the films, some others did not affect this property. Sunflower hulls and rice straw waste extract were integrated into starch films. All films showed an homogeneous structure without any large cracks, pores or phase separations (Menzel et al., 2019, 2020). Ho et al. (2020) did not notice any difference as well when *Pseuderanthemum palatiferum* (Nees) Radlk. extract was incorporated into gelatin-sodium alginate films. Scanning electron micrographs of gelatin-sodium alginate incorporated with natural extract showed that all the films displayed a plane and unbroken surface without bubbles (Fig. 4.5). Therefore, the addition of the natural extract into film forming solution did not show negative effects on the film´s appearance.

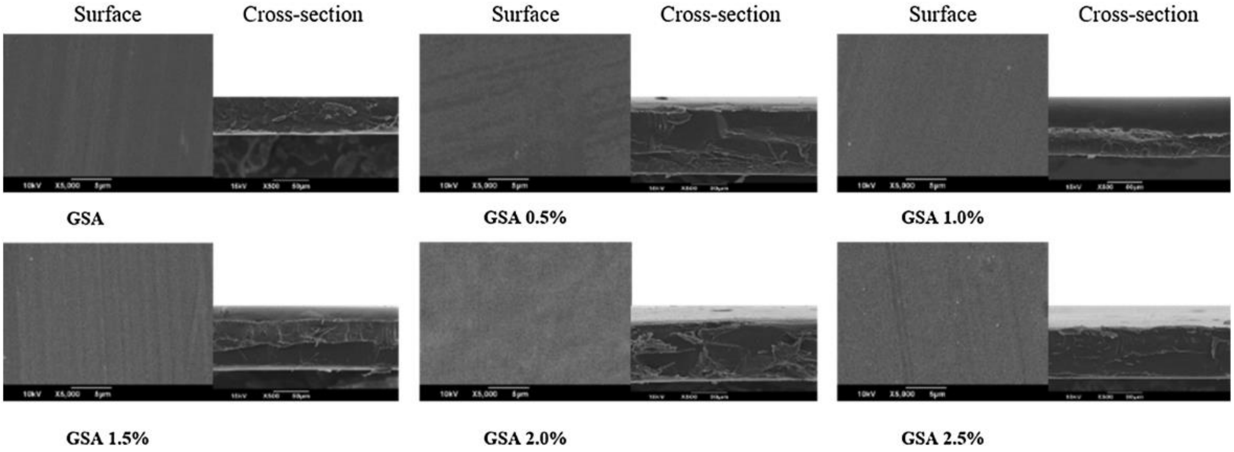


Fig. 4.5 – Surface and cross-section images of GSA based films using Scanning electron microscopy (SEM). GSA (gelatin-sodium alginate films); GSA-PFP 0.5 %, GSA- PFP 1.0 %, GSA-PFP 1.5 %, GSA-PFP 2.0 %, GSA-PFP 2.5 % (gelatin-alginate films with 0.5 %, 1.0 %, 1.5 %, 2.0 %, and 2.5 % of *P. palatiferum* freeze-dried powder obtained from subcritical water extraction, respectively) (Source: Ho et al., 2020).

The cross-sectional microstructures of gelatin films with haskap berries extract were investigated from J. Liu et al. (2019). The cross-sections of control and active films were smooth and uniform, indicating the filmogenic components (fish gelatin, haskap berries extract and glycerol) were homogeneously mixed and well compatible with each other. The dense inner structures of gelatin films with the natural extract were benefitted to improve their mechanical and barrier properties.

4.1.7 Rheological behaviour of film forming solution

To make a good film or a level coating on a solid surface, the viscosities of film forming solutions must be suitable to prevent sagging by gravity effects and to allow capillary levelling. A high viscosity or a gel type structure of film forming solution would make it difficult to eliminate air bubbles, thus hindering the casting of thin layers. Understanding the rheological properties of film forming solutions is important for the casting process of pre-formed films, or for applying coating in the liquid phase directly onto the surfaces of food products by dipping, brushing or spraying (Radusin et al., 2019).

The addition of pitanga leaf extract into cassava/starch affected the apparent viscosity of the film. The additives tended to slightly decrease the apparent viscosity of solutions indicating decrease in viscous nature of the polymer solutions, probably due to weakening of interactions and lesser structural compactness (Sirisha Nallan Chakravartula et al., 2020).

The starch-chitosan film forming solution appeared a pseudoplastic behaviour with the increase of shear rate. When incorporating peanut shell and ski extracts in starch-chitosan film forming solution, the apparent viscosity gradually decreased with the increase of concentration at the high shear rate. Films with peanut shell extract showed lower viscosity behaviour compared with peanut skin films and control film. The viscosity reflected the molecular interaction existing in film forming solutions. The stronger the molecular interaction, the higher the apparent viscosity was. Peanut shell and skin contained a large number of polyphenols and flavonoids, which could disturb the interaction between starch and chitosan and caused the low viscosity at high shear rate. Moreover, the composition of peanut skin extracts was different with peanut shell extracts which could result in the difference in viscosity (Meng et al., 2020).

The apparent viscosity of all the bionanocomposite film sample solutions decreased as the shear rate increase, indicating the pseudoplastic properties or the shear thinning region of these film forming solution. Moreover, the viscosity of the film forming solution decreased along with the incorporation of grape peel extract, which can be attributed to the plasticization of the extract. Grape peel extract contains small molecules with a large number of hydroxyl groups, which can easily penetrate into the network of konjac glucomannan matrix and form hydrogen bonds with konjac glucomannan (KGM) and carboxylation cellulose nanocrystal (C-CNC) molecules. Therefore, the hydrogen bonds between KGM and/or C-CNC were destroyed, thereby leading to the decreased viscosity (Tong et al., 2020).

Liang and Wang (2018) studied the effect of cortex *Phellodendron* extract concentration on the viscosity of the soy protein film forming solution. According to molecular chain theory, there was more entanglements between soy protein and cortex *Phellodendron* extract molecules at higher concentration of extract. As a result, the movement of molecules in mixed solution was more difficult. On the other hand, the average molecular weight of the components was higher with the addition of cortex *Phellodendron* extract. In general, the solution containing cortex *Phellodendron* extract had a higher viscosity than the control films at a higher-shear rate. This result may be ralted to the formation of new hydrogen bonds between biopolymer and extract, which was beneficial for enhancing the tensile strength of the film.

The effects of a natural extract from purple onion peel extract on the rheological properties of the film-forming solutions from *Artemisia sphaerocephala* Krasch. Gum were investigated from. The rheological results showed that active films with the extract formed a weak gel since the viscosity decreased as the shear rate increased and that the purple onion peel extract interacted with the polymer molecules through hydrogen bonds altering their network (T. Liang, Sun, Cao, Li, & Wang, 2018).

4.1.8. Antioxidant properties

Free radicals can lead to food spoilage and nutritional loss. Thus, antioxidant ability is important for active food packaging (Yong, Wang, et al., 2019) which is a very promising technique for extending food product shelf life (Yuan et al., 2020). One potential function of active packaging films is that the active compound can be released by migration directly on the surface of the food, while different foods have a significant effect on the release of the active compound of the film. Therefore, different food simulants are often used to investigate the migration and release of active compounds of the film (W. Zhang et al., 2019). The antioxidant character of polyphenols is associated with their ability to act as free radical scavengers, to inhibit lipoxygenase enzyme activity and to chelate metals (Collazo Bigliardi et al., 2019).

The use of plant extracts is an interesting ingredient for biodegradable food packaging materials, primarly due to its natural origin since consumers are trying to avoid synthetic additives and its potent source of antioxidants gained from polyphenolic and other compounds. Such type of package could be used to inhibit or reduce oxidative degradation of inside food which is one of the major reasons of quality deterioration . Antioxidant packaging is a major category of active packaging and very promising technique for improving the shelf life of food product. Furthermore, enriching films with antioxidants allows nutritional and aesthetic quality aspects to be extended without affecting the integrity of the packaged product, thus improve the self-life eof the product (Mir et al., 2018). Table 4.2 shows some recent studies dealing with the effect of plant extract addition on the *in vitro* antioxidant properties of edible films and coatings.

The antioxidant capacity of edible films is normally correlated with its available phenolic compound, which can be quantified usually using the Folin–Ciocalteau method, 2,2-diphenyl-1-picrylhydrazyl (DPPH) radical was one of the earliest synthetic radicals used to test the activity of phenolic antioxidants, ferric-reducing antioxidant power (FRAP) assay measures directly the ability of antioxidants to reduce a ferric tripyridyltriazine complex to the ferrous complex at low pH, ABTS (2,20-azinobis(3-ethylbenzothiazoline- 6-sulfonic acid) diammonium salt), a colourless compound which is initially oxidized to ABTS radical cation, a blue chromophore (Atarés & Chiralt, 2016) are some of the methods that are widely used from researchers to evaluate the antioxidant activity of the films.

Table 4.2 – Effect of plant extract addition on the antioxidant activity of biodegradable films (*in vitro* tests).

Type of plant extract	**Type of biopolymer**	**Method of measurement**	**Effects**	**References**
Black and purple eggplant	Chitosan	DPPH	Chitosan films with black eggplant showed higher antioxidant activity.	Yong et al. (2019)
Pine nutshell, peanut shell winter jujube	Chitosan	DPPH	The addition of natural plant extract significantly improved the DPPH radical scavenging activity of films.	X. Zhang et al. (2020)
Banana peel	Chitosan	TPC, DPPH, FRAP, ABTS, RP	All assays indicated improvement of the antioxidant capacity with the addition of banana peel extract.	W. Zhang et al. (2019)
Pomegranate peel and flesh	Carrageenan	DPPH	The DPPH radical scavenging activity was proportional to the content of the extract.	Y. Liu et al. (2020)
Fenugreek seed	Carrageenan	TPC, DPPH	The film with the highest extract concentration had the highest TPC and DPPH.	Farhan and Hani (2020)
Pomegranate seed	Cellulose	DPPH, ABTS	The extract resulted an increase of film´s antioxidant activity.	Nemazifard et al. (2017)
Rheum ribes	Methyl-cellulose	DPPH	*Rheum ribes* extract imparted excellent antioxidant activities to the film.	Kalkan et al. (2020)

Type of plant extract	**Type of biopolymer**	**Method of measurement**	**Effects**	**References**
Peanut skin	*Tremella fuciformis*	TPC, DPPH	The highest TPC revealed high DPPH and ABTS values.	Ju and Song (2020)
Oolong tea, corn silk, black soybean	Shrimp shell waste protein and chitosan	DPPH	After the addition of the extract the antioxidant activity increased.	Yuan et al. (2020)
Duria leaf	Gelatin	DPPH, FRAP	The active films showed low values in ferric reducing power.	Joanne Kam et al. (2018)
Centella asiatica	Gelatin	TPC, DPPH	The increasing amount of the extract increased TPC and DPPH.	Rasid et al. (2018)
Mango peel	Gelatin	TPC	TPC increased with the use of plant extract from 3 to 5%.	Adilah et al. (2018a)
Pseuderanthemum palatiferum (Nees) Radlk.	Gelatin sodium alginate	TPC, DPPH, FRAP	The increase in antioxidant activity (DPPH & FRAP) was parallel with TPC.	Ho et al. (2020)
Yerba mate and white tea	Furcellaran/ whey protein	DPPH	The incorporation of yerba mate and white tea resulted in increase of the antioxidant activity.	Pluta-Kubica et al. (2019)

In a study from Adilah et al. (2018) the TPC of mango peel extract and fish gelatin films was measured. There was no significant difference of TPC between gelatin film with 1% mango peel extract and the control. However, incorporation of the extract from 3 to 5% has significantly increased the TPC of film. The increasing values of TPC in active films may be affected by the amount of phenolic composition present within the extract. Ju and Song (2020) used this method to test the release of phenolic compounds over storage of *Tremella fuciformis* polysaccharides-peanut skin extract. The antioxidant capacity of natural plant extracts is mostly derived from phenolic compounds and peanut skin extract contains rich polyphenol content, such as p-coumaric acid, epicatechin, quercetin and resveratrol. Two more assays, apart from TPC were used in this study, DPPH and ABTS which showed that polysaccharides film containing 1.0 g/100 mL peanut skin extract had the highest ABTS (92.85%) and DPPH (84.92%) radical scavenging activities mainly due to the highest TPC (83.66 ± 0.57 mg GAE/g).

The total phenolic content of semi-refined κ-carrageenan films containing water germinated fenugreek seed extract was higher than the control film. The film with the highest extract concentration (25%) had the highest TPC (38.68 mg GAE/ g film), and the highest antioxidant activity (67%) indicating a good ability of the film network to carry the bioactive compounds present in the plant extract, and then to release these compounds in an aqueous medium (during dissolving film samples in water). Consequently, antioxidants-rich carrageenan films containing extract from germinated fenugreek seed can be exploited to extend the quality and shelf life of packaged food products, especially lipid-containing foods by delaying or preventing the oxidation process. (Farhan & Hani, 2020).

The increasing amount of *Centella asiatica* added (from 5 to 25%) into bovine-based gelatin film increased total phenolic content and scavenging activity against DPPH radical. The phenolic compounds inside the natural extract interact with the amino acids of gelatin compound and contribute to the antioxidant activity of the film (Rasid et al., 2018).

Yong, Wang, et al. (2019) formulated chitosan films incorporated with black and purple eggplant extract and used DPPH test. They found that pure chitosan films showed some antioxidant capacity, and this increased with increasing extract concentration. Moreover, chitosan films with black eggplant extract showed higher antioxidant ability than chitosan purple eggplant extract films at the same extract incorporation levels, which was because black eggplant extract had higher anthocyanin content than purple eggplant extract. X. Zhang et al. (2020) used the same method and biopolymer and reported that plant extracts from pine nutshell, peanut shell and winter jujube leaf significantly improved the antioxidant capacity of chitosan films.

Yuan et al. (2020) used DPPH assay and showed that after adding of three natural extracts from oolong tea, corn silk and black soybean seed coat it could enhance the antioxidant property of film compared with control film (shrimp shell waste protein combined with chitosan). The phenolic compounds in those plants could be responsible for its antioxidant activity. Kalkan et al. (2020) used this method as well to test the antioxidant capacity of methylcellulose films with *Rheum ribes* extract and found that the control film showed a low antioxidant activity and the DPPH scavenging activity of the films significantly increased with an increase in extract concentrations. Pluta-Kubica et al. (2019) reported that furcellaran/whey protein films showed almost no antioxidant activity when they were tested. The average radical scavenging effects of films with yerba mate and white tea extracts were significantly higher. This can be attributed to the antioxidant activity of the extracts, which are both known to contain polyphenols. The addition of pomegranate peel or pomegranate flesh extract remarkably elevated the DPPH radical scavenging activity of carrageenan film. Additionally, the DPPH radical scavenging activity of composite films was proportional to the content of extract (Y. Liu et al., 2020).

Pure carboxymethylcellulose films showed a very low antioxidant activity. However, when pomegranate seed extract was added to the films, it increased the antioxidant activity. It has been reported that the antioxidant activity of pomegranate peel extract is related to the diverse phenolic compounds in it (Nemazifard et al., 2017).

FRAP was used from Joanne Kam et al. (2018) to test the antioxidant capacity of gelatin based films after adding durian leaf extract. The active films showed low values in ferric reducing power due to the absence of any significant differences when compared to the control. This could be attributed to low extraction yield from the films during FRAP assays. In addition, it could also be due to less efficiency in antioxidant compound release when the fractions of the leaf extracts were incorporated into the film sample. The controlled release of an active compound from a matrix is very dependent on its diffusion capability. The introduction of a hydrophobic bioactive compound within a gelatin-based medium in this study might have altered the release kinetics. Moreover, DPPH test was measured and showed that films with 0.5 % of extract had a significantly different DPPH scavenging activity from the control film.

Ho et al. (2020) studied the effect of *Pseuderanthemum palatiferum* (Nees) Radlk. extract on antioxidant properties of gelatin sodium alginate films using FRAP, DPPH and TPC assays. The antioxidant of active films by FRAP assay showed a similar result with DPPH assay, as the concentration of extract increased, the antioxidant capacity increased as well. The increase in antioxidant activity was parallel with TPC. This investigation revealed that the antioxidant activity is not only contributed by phenolic compounds but also by gelatin and sodium alginate. *P. palatiferum* leaves contained phenolic and other compounds, such as protein, saponin, total sugar, and phytosterol which can also contribute to the antioxidant activity of gelatin sodium alginate-based films.

W. Zhang et al. (2019) showed that the incorporation of banana peel extract could improve the antioxidant capacity of the film significantly when compared to the chitosan film as TPC, ABTS, DPPH, FRAP and RP methods revealed. Despite phenolic substances being a major contributor to the antioxidant capacity of banana peel extract, other antioxidant active substances are found to be present in the natural extract as well, including carotenoids, biogenic amines and ascorbic acid. The antioxidant activities increased with the concentration of the natural extract.

4.1.9. Antimicrobial properties

Foodborne pathogens are one of the major factors affecting food safety and human health. Therefore, it is of great significance to develop antimicrobial active packaging films, to protect it from microbial growth and preserve it for an extended period of time (F. Liu et al., 2020; Riaz, Lagnika, Luo, Dai, et al., 2020). The incorporation of plant extracts into polymer films will create antimicrobial properties of packaging material. These plant extracts inhibit or reduce the growth of pathogens and spoilage micro-organisms in packaged foods and enhanced shelf life (Mir et al., 2018). The antimicrobial nature of polyphenols is associated with their capacity to inhibit extracellular microbial enzymes, to destabilise the cytoplasmic membrane and to provoke a deficit of the substrates required for microbial growth (Collazo-Bigliardi et al., 2019). Table 4.3 shows some recent studies dealing with the effect of plant extract addition on antimicrobial activity of edible films and coatings.

Table 4.3 – Effect of plant extracts addition on the antimicrobial activity of biodegradable films (*in vitro* tests).

Type of plant extract	**Type of biopolymer**	**Microorganisms tested**	**Comments**	**References**
Sonneratia caseolaris (L.) Engl. leaf	Chitosan	*S. aureus,* *P. auruginosa*	The strongest antibacterial activity of chitosan film with incorporated 1 % and 3 % natural extract was found against *Pseudomonas aeruginosa* during 12 h exposure.	Nguyen et al. (2020)
Rosemary	κ-carrageenan /nanoclay	*E. coli,* *P. auruginosa,* *B. cereus,* *S. aureus*	The antibacterial activity of rosemary comes from the existence of some phenols.	Nouri et al. (2018b)
Pomegranate peel and flesh	κ-carrageenan	*E. coli, Salmonella,* *S. aureus,* *L. monocytogenes*	Due to pomegranate peel extract had a higher total phenol content the films presented a higher antimicrobial activity.	Y. Liu et al. (2020)
Chinese chives	Carboxy-methyl-cellulose	*B. cereus,* *S. aureus,* *E. coli,* *S. typhimurium*	Phenolic acids, sulfur-containing compounds, flavonoids and allicn, which are present in the Chinese chives extracts are responsible for the antimicrobial activity.	Riaz et al. (2020)
Rheum ribes	Methyl-cellulose	*B. cereus, E. coli,* *S. aureus,* *L. monocytogenes,* *S. typhimurium,* *K. pneumonia,* *P. vulgaris*	As the concentration of *Rheum ribes* extracts increased, the inhibition zone increased significantly at all the concentrations.	Kalkan et al. (2020)
Rosemary	Whey protein	*L. monocytogenes,* *S. aureus*	Despite the minor incorporated quantity of the rosemary extract was able to inhibit *S. aureus* and *L. monocytogenes*.	Andrade et al. (2018)
Catechin-Kradon	Fish myofibrillar protein	*V. parahaemolyticus,* *S. aureus,* *S. typhimurium,* *L. monocytogenes*	The active films exhibited antimicrobial activity against only *V. parahaemolyticus*.	Kaewprachu et al. (2017)

Type of plant extract	Type of biopolymer	Microorganisms tested	Comments	References
Rowanberry, blue-berried honeysuckle, and chokeberry	Fish gelatin	*E. coli*, *P. fluorescens*, *S. aureus*, *L. innocua*,	As the extract concentration increased the antimicrobial properties increased as well.	Staroszczyk et al. (2020)

Some of the plant extracts have strong antimicrobials properties because of their high percentage of phenolic compounds such as catechin, epicatechin, epigallocatechin, carvacrol, thymol and eugenol (Mir et al., 2018). Nouri et al. (2018) reported that the κ-carrageenan/nanoclay matrix containing rosemary extract showed a clear antibacterial activity against Gram-negative (*Escherichia coli* and *Pseudomonas auruginosa*) and Gram-positive (*Bacillus cereus* and *Staphylococcus aureus*) bacteria and the inhibitory was increased as the rosemary extract concentration increased. This antibacterial efficiency of rosemary extract comes from the existence of some phenol diterpenes, such as carnosic acid, carnosol, rosmanol, isopropanol, and rosmarinic acid. Rosemary extract was investigated also from Andrade, Ribeiro-Santos, Costa Bonito, Saraiva, and Sanches-Silva (2018) who reported that the whey protein films containing rosemary extracts presented an excellent growth inhibition against *Listeria monocytogenes* and *Staphylococcus aureus*.

Y. Liu et al. (2020) reported the antimicrobial activity of κ-carrageenan films incorporated with pomegranate peel and flesh extract. The carrageenan control film did not show any inhibitory activity against four foodborne pathogens (*Escherichia coli*, *Salmonella*, *Staphylococcus aureus* and *Listeria monocytogenes*). However, both active films with their extracts exhibited certain antibacterial activity against foodborne pathogens. Due to pomegranate peel extract had a higher total phenol content the films presented a higher antimicrobial activity than with the pomegranate flesh extract, at the same extract incorporation level. Gram-negative bacteria (*E. coli* and *Salmonella*) were more sensitive to the composite films than Gram-positive bacteria (*S. aureus* and *L. monocytogenes*), which was because of the difference in the cell wall structures between Gram-positive and Gram-negative bacteria.

The antimicrobial properties of fish myofibrillar protein films contained different concentrations of catechin-Kradon against selected microorganisms, including *Vibrio parahaemolyticus*, *Staphylococcus aureus*, *Salmonella Typhimurium* and *Listeria monocytogenes*, are presented by Kaewprachu et al. (2017). The control film did not show any antimicrobial effect against all the tested microorganisms. The active films exhibited antimicrobial activity against only *V. parahaemolyticus*.

Staroszczyk et al. (2020) reported that fish gelatin films containing pomace aqueous extracts from rowanberry, blue-berried honeysuckle, and chokeberry in amount of 1.2 mL per film (6%) did not possess antimicrobial properties against *Escherichia coli*, *Pseudomonas fluorescens*, *Staphylococcus aureus*, and *Listeria innocua*, while strong antimicrobial properties against each of them were observed when the extract volume was increased.

Two Gram-positive (*Bacillus cereus* and *Staphylococcus aureus*) and two Gram-negative (*Escherichia coli* and *Salmonella typhimurium*) bacteria were used to assess the antimicrobial activity of carboxymethylcellulose films incorporated with Chinese chives root extract. The control film (without extract) did not show any antimicrobial activity, whereas the active film exhibited inhibitory effects on both Gram-positive and Gram-negative bacteria and zones of inhibition increased with the increasing concentration of the extract to the control film. There are certain compounds present in the chives root like phenolic acids, sulfur-containing compounds (diallyl sulfides), flavonoids and allicn, which are responsible for the antimicrobial activity (Riaz, Lagnika, Luo, Nie, et al., 2020).

Rheum ribes extracts have been proven to be antimicrobial against various microorganisms. The antimicrobial activity of the *Rheum ribes* extracts on methylcellulose films was studied from Kalkan et al. (2020). The results showed that the films containing extract were effective against *Bacillus cereus*, *Escherichia coli*, *Staphylococcus aureus*, *Listeria monocytogenes*, *Salmonella typhimurium*, *Klebsiella pneumonia*, and *Proteus vulgaris*. As the concentration of *Rheum ribes* extracts increased, the inhibition zone increased significantly at all the concentrations.

Edible packaging films based on chitosan and *Sonneratia caseolaris* (L.) Engl. leaf extract was evaluated from Nguyen et al. (2020). The antibacterial activity of chitosan-based films with the addition of natural extract was stronger against Gram negative bacteria (*Pseudomonas aeruginosa*) than Gram positive bacteria (*Staphylococcus* *aureus*), and it was mainly attributed to extract flavonoids, i.e. luteolin and luteolin 7-o-β-glucoside.

4.1.10. Biodegradability

The term “biodegradable” materials is used to describe those materials which can be degraded by the enzymatic action of living organisms, such as bacteria, yeasts, fungi and the ultimate end-products of the degradation process, these being CO_2, H_2O, and biomass under aerobic conditions and hydrocarbons, methane and biomass under anaerobic conditions (Medina Jaramillo, Gutiérrez, Goyanes, Bernal, & Famá, 2016). Polymer biodegradation is a natural complex phenomenon, which is difficult to be fully simulated in the laboratory environment due to several parameters related to the entire biogeochemical process. Therefore, the current situation offers limited flexibility in methodologies for testing polymer biodegradation (Zhong et al., 2020). It is known that packaging must decompose by at least 90% by biological action in a period of 6 months to be considered biodegradable (Martins et al., 2019). Natural polyphenols affect the activity of the microorganisms present in the composting environment (Riaz, Lagnika, Luo, Dai, et al., 2020).

Medina-Jaramillo, Ochoa-Yepes, Bernal, and Famá (2017) and Medina Jaramillo et al. (2016) evaluated cassava starch films with green tea, and basil and yerba mate extract (Fig. 4.6), respectively. The results showed that the addition of the natural extracts decreased the biodegradation time of cassava starch films to 12 days in vegetal compost.

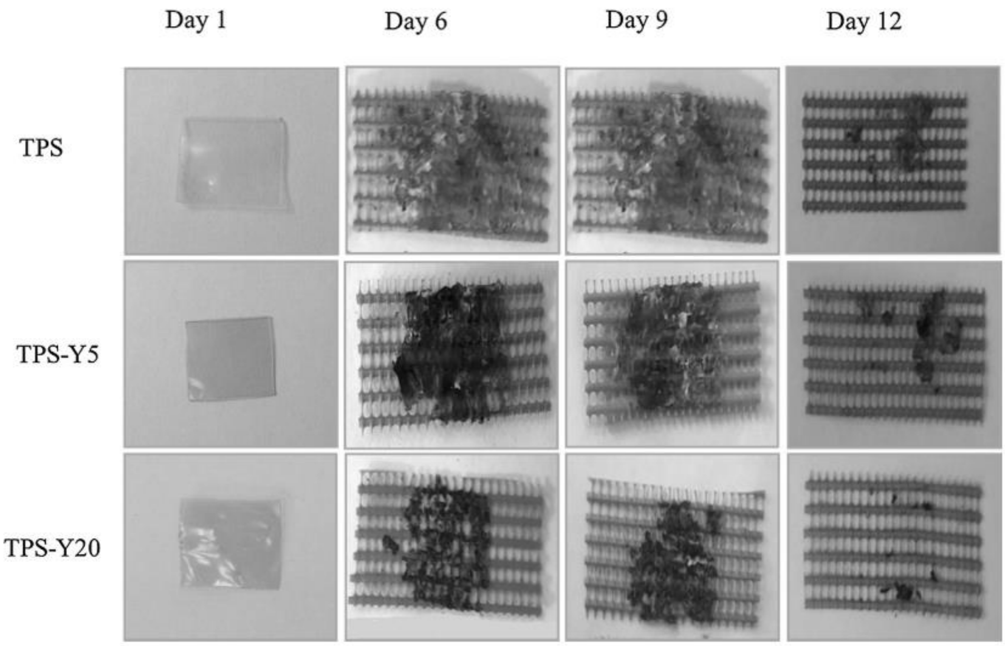


Fig. 4.6 – Biodegradability of thermoplastic starch films (TPS) with yerba mate (YM (5 and 20%)) extract in vegetable compost (Source: Medina Jaramillo et al., 2016).

Riaz, Lagnika, Luo, Nie, et al. (2020) evaluated the effect of Chinese chives root extract on biodegradation test of carboxymethyl cellulose film. The weight loss of control film and active film was increased as the time soil dumbing was increased to three weeks. Carboxymethylcellulose film with the highest concentration of the extract (5% w/w) exhibited the highest weight loss 58,14% after three weeks, whereas, for control film, it was 32.66%. The improvement of biodegradability is due to the formation of new polymeric materials and reduced the degradation burdens on the environment.

In some cases, the biodegradation of active films can be retarded with the addition of plant extracts. Piñeros-Hernandez et al. (2017) studied the effect of rosemary extract on disintegration of cassava films. The decomposition of the control films occurred almost entirely after 14 days of assay. For this time, the integrity of the rosemary extract-containing films (5, 10 and 10%) was better preserved, indicating that the biodegradation was retarded by the presence of the extract. Moreover, in another study, the octenyl succinate starch films showed a higher percentage of film biodegradability compared to the films with the incorporation of pecan nutshell extract or hazelnut skin extract. After 146 days in soil, the biodegradation value of the control film was around 81%, while the values for the films with both natural extracts were decreasing with the increase of the extract concentration. The biodegradation values did not exceed 71%. The decrease in the biodegradability of active films could be due to their low solubility (Leon-Bejarano et al., 2020).

Apart of the positives or negatives effects, plant extracts can show non-significant difference in the biodegradability rate. For example, biodegradation of the *Tremella fuciformis* polysaccharides films with various concentrations (0, 0.25, 0.5, and 1.0 g/100 mL) of peanut skin extract was evaluated for 50 d. During the first 10 d, there was no noticeable change in terms of biodegradability for all polysaccharide films. However, after 15 and 20 d, all films were degraded, and after 50 d, all the films were nearly degraded regardless of natural extract content (Fig. 4.7). Temperature, moisture, enzymatic activity of microorganisms, and soil composition are the main factors that determine the biodegradation rate. In this study, the incorporation of peanut skin extract did not affect the biodegradation rate of *Tremella fuciformis* polysaccharides films as figure shows (Ju & Song, 2020).

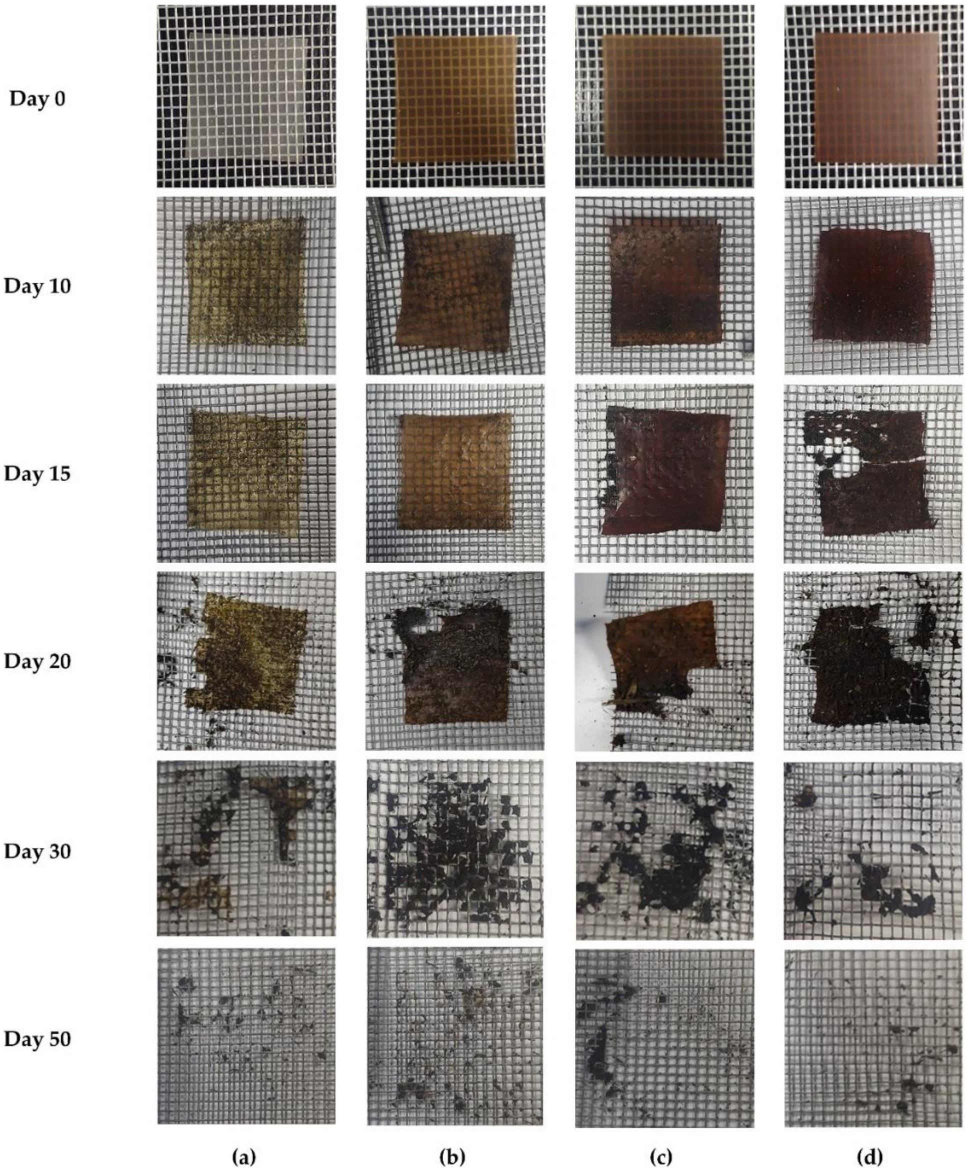


Fig. 4.7 – Biodegradability of the TFP films containing PSE. (a) TFP film without PSE, (b) TFP film with 0.25 g/100 mL PSE, (c) TFP film with 0.5 g/100 mL PSE, (d) TFP film with 1.0 g/100 mL PSE (TFP: *Tremella fuciformis* polysaccharide, PSE: peanut skin extract) (Source: Ju & Song, 2020).

Ferreira Nogueira, Matta Fakhouri, and de Oliveira (2019) reported also that the incorporation of blackberry particles into arrowroot starch films did not affect the biodegradation rate of the films for 38 days, showing that biodegradability is characteristic of chemical composition of both arrowroot starch and blackberry powder.

4.2. Application of active food packaging

The main function of food packaging is to maintain the quality and safety of food products during storage and conveyance, and to increase the shelf life of food products by preventing unfavourable factors or conditions like spoilage, chemical contaminants, oxygen, moisture, light, external force etc (Hosseini & Gómez-Guillén, 2018).

In recent years, many research studies have focused on the application of active food packaging on a broad array of real food systems with the aim of studying their antioxidant and antibacterial effect (Atarés & Chiralt, 2016). This type of technology has the objective of their interaction with the food product, in the sense of modifying or maintaining parameters to improve the quality of the product packaged during its shelf life (J. S. Ribeiro et al., 2019). Nowadays, consumers prefer convenience food that is of good quality, having a long shelf life, with minimum preservatives and no synthetic additives (Kanatt, 2020), therefore, plant extracts are widely used. The actual capability of edible films and coatings to improve the shelf life of food products is presented in Table 4.4.

Table 4.4 – Reports about the application of edible films and coatings with plant extracts on food products.

Type of plant extract (concentation)	Type of biopolymer	Type of application	Food system	Storage	Effects	References
Green tea, black tea, oolong tea (0.5%)	Grain protein	Film	Pork	10 d at 4 °C	Inhibition of lipid oxidation.	Yang et al. (2016)
Green tea (0.5 and 1%)	Collagen	Coating	Sausages	14 d at 4 °C	Inhibition of lipid oxidation.	Shokraneh et al. (2017)
Green tea (5 %)	Potato starch	Film	Beef	10 d at 4 °C	Inhibition of lipid oxidation.	u Nisa et al. (2015)
Caesalpinia decapetala (0.3, 0.7 and 1%) *Caesalpnia spinosa* (0.07, 0.1 and 0.2%)	Gelatin	Film	Beef patties	12 d at 4 °C 13 d at 4 °C	Inhibition of TBARS.	Gallego et al. (2016)

Type of extract (concentation)	**Type of biopolymer**	**Type of application**	**Food system**	**Storage**	**Effects**	**References**
Asparagus racemosus (1 and 2%)	Calcium alginate	Film	Chevon sausages	21 d at 4 °C	Antimicrobial properties and lipid oxidative stability.	Noor et al. (2018)
Terminalia arjuna (0.5 and 1%)	Calcium alginate	Film	Chevon sausages	21 d at 4 °C	Antimicrobial properties and lipid oxidative stability.	Kalem et al. (2018)
Clove and cinnamon (10 and 20 mg/mL*)*	Tamarind seed starch	Film	Mutton	21 d at 4 °C	Shelf life increased by 3 weeks.	Chandra Mohan et al. (2017)
Germinated fenugreek seeds (10, 15, 20%)	Semi-refined-κ-carrageenan	Film	Chicken breast	7 d at 5 °C	Microbiological shelf-life extension at least 6 days.	Farhan and Hani (2020)
Red seaweed (0.5, 1 and 1.5%)	Microalgal exopolysaccharides	Coating	Shrimps	8 d at 4 °C	Reduced TVB-N, TMA and TBARS.	Balti et al. (2020)
Citrus wilsonii (0.2 mg/mL)	Calcium alginate	Coating	Shrimps	6 d at 4 °C	Lower TVC, pH and TVB-N.	X. Liu et al. (2016)
Grape seed (0.5 and 1%)	Carboxy-methyl-cellulose	Coating	Trout fillets	20 d at 4 °C	Antioxidant and antimicrobial activities.	Raeisi et al. (2015)
Catechin-Kradon (0.9%)	Myofibrillar protein	Film	Bluefin tuna	10 d at 4 °C	Lipid oxidation, microbial activity.	Kaewprachu et al. (2017)
Pomegranate peel (0.361 g/mL)	Chitosan/ locust bean gum	Coating	Oranges	5 d at 26 °C	Antimicrobial activity (*Penicillium digitatum*).	Kharchoufi et al. (2018)

Type of extract (concentation)	**Type of biopolymer**	**Type of application**	**Food system**	**Storage**	**Effects**	**References**
Green tea, black tea, and *Aloe vera* (5 and 10%)	Gelatin	Coating	Oranges	17 d at 4 °C	Antimicrobial activity and reduced weight loss, ascorbic acid degradation, and colour darkening.	Radi et al. (2017)
Pomegranate peel (1%)	Chitosan	Coating	Guava	20 d at 4 °C	Reduced respiration rate.	Nair et al. (2018)
Banana peel (4%)	Chitosan	Coating	Apples	35 d at 25 °C	Reduced respiration rate and weight loss.	W. Zhang et al. (2019)
Pink peper (6%)	Starch/ protein	Coating	Apples	12 d at 4 °C	Inhibition of enzymatic browning.	Romani et al. (2018)
Moringa oleifera (0.5 and 1%)	Carboxy-methyl-cellulose	Coating	Avocado	21 d at 5.5 °C	Reduced respiration and ripping rate, antimicrobial activity.	Tesfay and Magwaza (2017)
Lotus leaf (0.2%)	Sodium alginate, konjac glucomannan and starch	Coating	Goji fruit	9 d at RT[a]	Reduced weight loss, decay rate and malondialdehyde.	Fan et al. (2019)
Sonneratia caseolaris (L.) Engl. Leaf (1%)	Chitosan	Coating	Banana	4 d at RT[a]	Better surface, delay the decay of the fruit.	Nguyen et al. (2020)

[a]: Room temperature

d: Days

Meat, including poultry and fish (meats and products) is the first-choice source of animal protein for many people all over the world. Oxidation of lipid and protein components during meats and products storage is the major cause for their quality changes, often followed by formation of rancid flavour (Popovic´ et al., 2018).

Yang, Lee, Won, and Song (2016) studied the lipid oxidation of pork meat wrapped with a film from grain protein containing 0.5% tea extracts during 10 days of storage. The results indicated that the lipid oxidation rates of pork meat wrapped with the active films containing tea extract were slower than the control film. Green tea extract founded to be the most desirable than oolong tea and black tea extract, due to low increase of TBARS value. The TBARS value are in good agreement with the antioxidant activity, meaning the 0.5% green tea extract had the highest antioxidant activity and the slowest lipid oxidation during storage. Similar results found from Shokraneh, Ariaii, Rasouli Ghahrodi, Hasannia, and Sabbaghpour (2017) when green tea extract was incorporated into collagen-based edible coating to reduce the oxidative degradation of sausages during two weeks of storage at 4 ^{o}C. In another study, lipid oxidation of fresh beef decreased as well from the incorporation of green tea extract into potato starch-based film (u Nisa et al., 2015). Gallego, Gordon, Segovia, and Almajano Pablos (2016) studied beef patties which were packaged into gelatin films with *Caesalpinia decapetala* and *Caesalpnia spinosa* extracts. The results showed that the active films with *Caesalpnia spinosa* was the most effective antioxidant for the beef patties, inhibiting the formation of TBARS more effectively than the synthetic antioxidant BHA over the course of 12 days.

Chevon sausages (goat meat) were packaged into calcium alginate edible films with *Terminalia arjuna* and *Asparagus racemosus* extracts and investigated from Kalem, Bhat, Kumar, Noor, and Desai, (2018) and Noor, Bhat, Kumar, and Mudiyanselage (2018) respectively. Both studies showed that with the incorporation of natural extracts antimicrobial properties were gained and lipid oxidative stability was improved.

Clove and cinnamon extracts were incorporated into tamarind seed starch and applied into mutton meat. Results showed that the shelf life of the meat increased by 1 week at storage temperature of 10 ^{o}C and three weeks at storage temperature of 4 ^{o}C (Chandra Mohan et al., 2017).

Chicken breast that were packed in HDPE and control semi-refined-κ-carrageenan matrix films showed that the total bacterial counts on the surface of fresh chicken breasts reached unacceptable levels after day 1 of cold storage. Whereas, active semi-refined-κ-carrageenan films containing 10, 15 and 20 % water extract of germinated fenugreek seeds had a significant controlling effect on the total bacterial counts on the surface of fresh chicken breast meat during 7 days of cold storage, resulting in a microbiological shelf-life extension of at least 6 days of fresh chicken breast meat (Farhan & Hani, 2020).

Moreover, it has been reported that the spoilage of fish muscle is a combination of different spoilage mechanisms including lipid oxidation, microbial and endogenous enzymes activities as well as enzymatic browning (Raeisi, Tajik, Aliakbarlu, Mirhosseini, & Hosseini, 2015). An active coating based on a combination of microalgal exopolysaccharides film and red seaweed extract was prepared and applied on shrimps. The experiment was carried on for 8 days at 4 °C. Shrimp coating with the mixture as it was described effectively extended the shelf life of shrimp by reducing TVB-N, TMA and TBARS values and retained all sensory attributes while keeping bacterial levels relatively low, especially psychrotrophic bacteria (Balti et al., 2020). In another study shrimps were packaged as well over six days of storage at 4 °C from X. Liu et al. (2016) with alginate-calcium film and *Citrus wilsonii* extract. The results showed that the total viable count of the treated shrimps was more than 10 times lower than the negative untreated control. The lower rate of the increase in pH and total volatile base nitrogen was also observed in shrimps treated with the extract compared to the control group of shrimps (see Fig. 4.8).

Fig. 4.8 – Appearance of shrimps. A: fresh shrimps; B: shrimps after storage, coated with alginate-calcium based film containing *Citrus wilsonii* extract; C: untreated control shrimps after storage (Source: X. Liu et al. 2016).

Trout fillets were packed with carboxymethylcellulose coating and grape seed extract (0.5–1%). The higher concentration of the extract resulted in stronger antimicrobial and antioxidant activities (TVC, LAB and *Pseudomonas* spp.) (Raeisi et al., 2015).

Kaewprachu, Osako, Benjakul, Suthiluk, and Rawdkuen (2017) investigated and determined the changes in various qualities of Bluefin tuna slices that were wrapped with fish myofibrillar protein films and catechin-Kradon extract and stored in a refrigerator (4 °C) for 10 days. The results showed that fish myofibrillar protein film incorporated with catechin-Kradon extract could suppress the lipid oxidation, the formation of metmyoglobin, as well as the microbial growth in tuna slices more effectively than the control film, and LDPE.

Fruits and vegetables vary in their respiration rates, ethylene production and sensitivity to ethylene, storage temperature and relative humidity which accordingly affect packaging requirements and their shelf life (Ayhan, 2019).

Active packaging was applied on oranges from Kharchoufi et al. (2018) (Fig. 4.9) and Radi, Firouzi, Akhavan, and Amiri (2017) using pomegranate peel extract in chitosan/locust bean gum films and green/black tea and *Aloe vera* in gelatin coating, respectively. Pomegranate peel extract showed inhibition growth of *P. digitatum* and *Aloe vera* and green tea extracts showed lower microbial population because of their antimicrobial compounds.

Fig. 4.9 – Visual effect of the application of chitosan (CH) and locust bean gum (LBG) coatings, incorporating 0.361 g dry water pomegranate peel extract (WPPE)/mL and 108 cells/mL *W. anomalus*, on oranges artificially inoculated with *Penicillium digitatum* after incubation at 26 °C for 5 days (Source: Kharchoufi et al. 2018).

Chitosan films with pomegranate peel was used in guava fruits and chitosan films with banana peel extract was used in apples (Nair, Saxena, & Kaur, 2018; W. Zhang et al., 2019). Chitosan with the extract coatings invariably significantly reduced the respiration rate of the fruits in both situations. The impact of pomegranate peel extract may be due to its antimicrobial and lipophilic properties which enhanced the barrier properties of the coating and restricted the gas diffusion (Nair et al., 2018).

Romani, Hernández, and Martins (2018) showed that the blend used as coating (15/85 of starch/protein added of 6% (v/w) pink pepper phenolic compounds) in fresh-cut apples for 12 days achieved a better conservation, especially in terms of inhibition of enzymatic browning.

The most efficient mass loss control observed in the avocado samples which were coated with a combination of moringa extract and carboxymethylcellulose films. Untreated control fruit continued losing water into the cold room atmosphere. Moreover, this combination had lower rates of respiration, and higher values of firmness (Tesfay & Magwaza, 2017). Moringa leaf extract in carboxymethylcellulose -based coatings reduced the rate of ripening and inhibited the growth of postharvest pathogens.

Formulation of coating was obtained by sodium alginate, konjac glucomannan and starch with lotus leaf extract which was studied on the quality of fresh goji fruit during postharvest storage at ambient temperature. Goji fruit coated with the natural extract incorporated coating had significantly lower weight loss, decay rate and malondialdehyde content than the blank control (uncoated) and the positive control (fumigated with 1-methylcyclopropene) after 9 d of storage (Fan et al., 2019).

Chitosan coating with *Sonneratia caseolaris* (L.) Engl. Leaf extract was used to in banana fruits. The appearance of bananas stored for 0 day and 4 days at room temperature were observed in Fig. 4.10. The surface of bananas coated with chitosan and natural extract was better than of bananas with chitosan film and without coating. This demonstrates that the incorporated plant extract into chitosan may delay the decay of bananas (Nguyen et al., 2020).

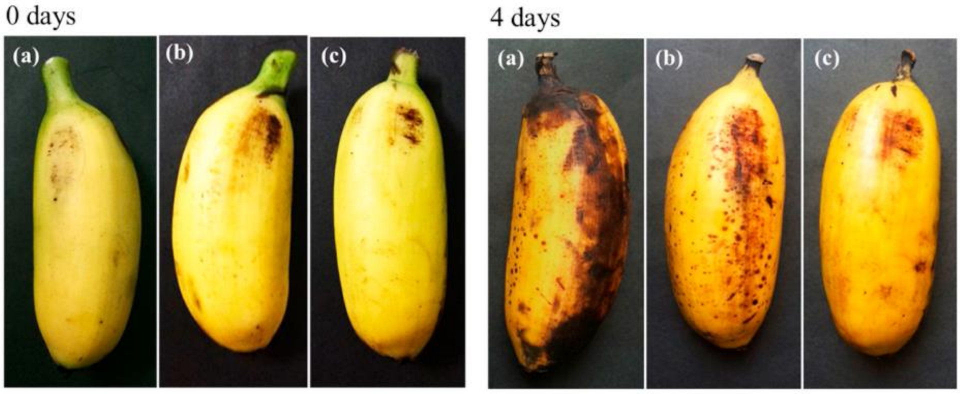


Fig. 4.10 – The photos of uncoated banana (a), coated banana using chitosan film (b) and chitosan-*Sonneratia caseolaris* (L.) Engl. leaf extract (1%) (c) (Source: Nguyen et al., 2020).

5. Commercialization

Although active packaging research has been ongoing since the 1990s, globally few of these technologies currently on the market may be considered commercially significant (Werner et al., 2017). Taking research from laboratory to market is a formidable task and often the challenges met limit product innovation. To this end, the primary challenges lay around development of a technologically and economically viable solution that is consistent with consumer demands. The developmental work involves (Padmanabhan, Cruz-Romero, Kerry, & Morris, 2018):

- understanding of technology, understanding the application area(s) for that technology,
- proving the effectiveness of the product based on the developed technology to investors, manufacturers, customers, and any regulatory bodies,
- developing practical methods that facilitate scaling the technology and associated production processes for large volume manufacturing,
- consideration of costs (both production and capital),
- consideration of social impacts and benefits the technology will have, and,
- finally, but most critically, the economic benefits from the technology/application.

Werner et al. (2017) described in the Fig. 5.1 the major hurdles to commercialization of next generation active packaging technology.

Fig. 5.1 – Hurdles to commercialization of active packaging technologies. Academic and industrial researchers must collaborate to overcome hurdles (e.g. consumer acceptance, environmental sustainability, regulatory approval, raw materials manufacturers, package converters, and food manufacturers) and promote commercial translation of active packaging technologies (Source: Werner et al., 2017).

The cost and consumer acceptance are the key factors that will determine the commercialization potential of novel antimicrobial packaging technologies. In order to achieve a reasonable cost, the process of manufacturing antimicrobial/antioxidants films should be simple and integratable with the existing processes and manufacturing facilities. Further, the films should be chemically stable for long-term usage and storage. The current multimillion-dollar manufacturing setups in place, high processing throughputs and demands, however, make industries play safe than going for such possibly positively disruptive changes. When it comes to regulation, there are many hurdles for surface engineering technologies to address. They are those put down by FDA, EFSA, EPA, and so on; a systemic solution should address all these aspects and should be competent in cost aspects as well (Padmanabhan et al., 2018). These efforts will lead to the increasing availability within a global marketplace of next generation packaging technologies in support of ongoing efforts to improve the safety, quality, and sustainability of our food supply (Werner et al., 2017).

Today, there are a number of companies around the world which have produced and built equipment and facilities for the production of biodegradable materials applicable in food industries. Table 5.1 lists some of the commercially available bio-based and biodegradable polymers used in food industries.

Table 5.1 – Commercially available biodegradable biopolymers in food industry

Commercial name	**Company**	**Base polymer**	**Application**
Mater-Bi	Novamont	Starch blends	Packaging materials
Ingeo	Nature Works	PLA	Packaging materials
Cozeen	Flo	Corn prolamine/ guar gum	Packaging materials
Nutrasave^{TM}	AGELESS OMAC	CMC	Packaging materials
Ecofoam	Ecofoam Nature	Starch	Packaging materials
Luminy	Total Corbion	PLA	Packaging materials
BioEnvelop	NuitriCorp	Cellulose derivative/starch	Packaging materials
Nodax	Danimer Scientific	PHA	Packaging materials
Ecoflex	BASF	PBAT	Packaging materials

6. Conclusion and future trends

Packaging is an important factor in the food industry and is dominated by petroleum-derived polymers (Mir et al., 2018). Synthetic plastics are the wonder material of today´s world, and life without them is unimaginable. Unfortunately, some useful qualities that they have, are overshadowed by their steady contribution to litter worldwide and its negative consequences for the environment (Imam et al., 2012). Therefore, the amount of research involving the production and characterization of biodegradable films has increased substantially, mainly due to interest in minimizing the ecological impact caused by the use of synthetic packaging. However, biodegradable films/coatings usually prepared from polysaccharides, proteins, and lipids or from their combination do not have the preferable properties. Consequently, blending those biopolymers with additives is one of the most effective methods to have a new material with desired properties.

In the recent years, the addition of plant extracts into biodegradable films and coatings for active food packaging has been extensively studied since consumer´s demand for more natural and environmentally friendly products has increased. Plant extracts have proved to have some impact on the biopolymer´s film properties though interactions with the active groups. For instance, the literature showed that usually the addition of plant extracts increases the thickness, alters the original colour and reduces the transparency of developed films. Moreover, studies have revealed diverse effects in the mechanical (tensile strength and elongation break) and barrier (water vapour permeability) properties of edible films with the addition of natural extracts. In addition, water content, solubility and swelling degree, contact angle (θ), and viscocity of the films and coatings have been modified as well. Likewise, plant extracts significantly affected the structural properties and the biodegradation rate of the films.

Additionally, being concerned about the deterioration and the short life of food products, plant extracts can provide the films and coatings with antioxidant and/or antimicrobial properties, depending both on their composition and the interactions with the polymer matrix, thus they can contribute to preserving and extending the shelf life of packaged food. Most of the studies observed positive effects on the characteristics of products like meat, fish, vegetables, and fruits after the addition of plant extracts by controlling the quality deterioration (microbial growth inhibition, antioxidant activity and preservation of sensory properties).

Besides the promising results obtained from the literature, there is a lot to achieve, in relation to product quality, shelf life and consumer acceptance of extract-based films on food products. The plant based biodegradable films are lacking in the market because there are still obstacles in using sustainable polymers instead of synthetic ones. For this reason more detailed studies must be done to improve the properties of the films and coatings for the industrial application of food packaging, since the improvement of some characteristics of the material is, in general, associated with a decrease in others (Martins et al., 2019). In addition, there is a great variety of natural plant sources with bioactive properties that remain to be studied and used for the development of active packaging or biopolymer-based edible films for preserving and adding value to foods (Mir et al., 2018).

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